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VOLUME 2

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PREFACE

This is volume II of a history of the research work of the many chemists who worked with me in the University of Chicago Metallurgical Laboratory during World War II. The work of these groups, which were affiliated under my direction in a unit which became known as "Chemistry Section C-I" as part of a broader Chemistry Division, was concerned with the development of chemical procedures for the extraction of plutonium, for the purification of plutonium and, in the later phases, for research on the isotopes of other heavy elements including other transuranium elements. Volume I, "History of Met Lab Section C-I, April, 1942 to April, 1943," was published as LBL special report PUB 112 in February, 1977.

This history is written on the basis of some 60 categories of information, including a wide range of Met Lab Progress Reports, notes on meetings, an almost complete file of all the laboratory notebooks, personnel records, patent files, travel vouchers, organization charts, records of pile and cyclotron bombardments, health monitoring records, administrative bulletins, etc. Unfortunately, I did not keep a diary, but we were able to locate hundreds of notes I had taken to cover meetings, telephone calls, etc.; these were often written in a style that amounted to a code (in order to protect the secret nature of the information), but I found it possible to decipher them. This was one of the most important categories of information, as was the file of meeting notes (such note taking was not inaugurated until 1943) which had been issued as informal reports and which usually included names of the attendees. In addition Elwin Covey kindly placed at my disposal his diary, which covered the period until his departure for Clinton Laboratories in August, 1943. Another welcome source was a very brief diary, actually an intermittent daily notation or a few words in a small pocket-size appointment book, kept by my wife Helen; this made it possible to include a large number of entries concerning our social life during most of the period 1942-1946 when we were in Chicago.

The style is such that the entry for each day is written as though it was entered in a diary on the basis of information available to me at the end of the day. There is more reliance on quotations from letters, written or received, than would be usual for an actual diary, which is justified on the basis that this kind of information comes closest to emulating a diary. An exception to this style of diary imitation is the footnotes that are included to give additional background material; these go beyond the activities of Chemistry Section C-I, covering meetings that I did not attend, and hence often include information that I could not have had on those entry dates.

Essentially all of the events, and the dates on which they are recorded, are based on the numerous categories of documentation; only a very small portion is based on memory alone, and even then it is usually associated with related information based on documentation. Chicago

newspapers for this period, available in libraries, and weather records were used to embellish the narrative with some additional information on current events.

To help me in this writing task I wrote and talked to many who participated with me in the Met Lab experience to seek their recollections to augment the documented record. I am very grateful for this help. Since photography was not encouraged, the supply of illustrations is limited. I am indebted to Bernie Donlan and Al and Kay Florin for their photographs taken at a Christmas party in December, 1943, and for pictures of the occupants of Room 12. I appreciate receiving the photographs of New Chem, Chem Annex, and Winston Manning and me taken by Howard Lange. In addition, Donald Stewart supplied photographs of Site A and Site B while Ray Stoughton located early pictures of Oak Ridge.

I am indebted to Joseph Katz, Carol Flaumenhaft, Sydney Gaarder, Bernard Saunders, Donald Stewart, John Farmakes, and Peter Seaborg for help in gathering and preparing material and to Margie Hollander, Kathleen Van Der Haeghen, and Helen Seaborg for much help in putting this volume into publishable form.

MAY 1943

Saturday, May 1, 1943

In making the rounds of the laboratories today, I found a number of interesting experiments in progress. One of them, started by Burris Cunningham yesterday, involves the measurement of the adsorption of plutonium, at high concentrations, by silica gel from 10% UNH solutions. Cunningham's object is to test whether the adsorption-elution method for the separation of plutonium from uranium and fission products, which has been developed at tracer concentrations of plutonium, will work at the concentration of plutonium that will be produced in the piles at Site W. He finds at a ratio of concentration of uranium to plutonium of about 890:1, which corresponds to a higher concentration of plutonium than will be found at Site W, adsorption of 66% of the plutonium in two equilibrium experiments. This is very encouraging. In another experiment, Dan Miller is starting to test the Sodium Uranyl Acetate Process, developed at Berkeley for the separation of plutonium from uranium and fission products. He will run through several oxidation-reduction cycles with tracer concentrations of plutonium in order to determine the yield of plutonium and the degree of its decontamination from fission products. Then he will test the flow sheet for the process recommended by the Berkeley group.

Perlman attended a conference on the Project Handbook along with Coryell, Creutz, Fussler, W. C. Kay, Lane, Leverett, Morrison, Mulliken, Wilson, and Wollan. The contributions of our section to Chapter III, "Nuclear Physics and Chemistry: The Fission Products," and Chapter VII, "Instrumentation and Technique," were discussed.

In a memorandum to us today, Whitaker asks for a written description of the methods we propose to use in the further purification of the plutonium that our group at Clinton Laboratories will receive from the separations plant there.

Helen has been helping me since before we left Berkeley a year ago on the next edition of the "Table of Isotopes" that I am preparing for publication in Reviews of Modern Physics. She went to Crerar Library (86 E. Randolph) today to abstract a number of articles, after which she visited the Stage Lounge in downtown Chicago.

Sunday, May 2, 1943

I held a meeting of the Council of my section at 10:30 a.m. in my office. Attending were Perlman, Willard, Kirk, Cunningham, Brown, Orlemann, English, and Thompson. We discussed the following topics:

Extraction methods. A point was brought up that when the plutonium precipitate is comparable in quantity with that of the carrier, a separation might occur due to differences in the rate of precipitation. It was suggested that in the oxidized solution a part of the carrier be added before reduction takes place in order to eliminate the separation as much as possible.

Wet Fluoride Process. The progress in the wet fluoride work was reviewed, and the following tests are in process or yet to be made. (1) Tests on pure plutonium with special emphasis on carrying from zirconium solution. (2) Runs on the semiworks scale embodying some of the later changes in the method. (3) The effect of Site W concentrations of fission products on the decontamination. (4) Wet fluoride method to be tried in the presence of aluminum. (5) Further work on oxidation, reduction, and carrying power in the presence of zirconium.

Bismuth Phosphate Process. A search is being made for a new reducing agent in the Bismuth Phosphate Process, and it was suggested that a combination of H_2O_2 and Fe^{+2} might work. Microchemical tests are being carried out, and these indicate that the carrying is good at ratios of Bi:Pu of 100:1.

Acetate method. Miller is being shifted full-time to this work and is starting out to test the carrying power of reprecipitation and the effect of aluminum. Noyes and Bray have pointed out in their book on qualitative chemical analysis that certain ratios of acetic acid and acetate ion will not precipitate aluminum. This ratio seems to be about 2.

Adsorption process. Tests are being started on batch processes using silica gel. It is visualized that the operating plant might consist of a series of tanks through which the solution would progressively pass until all of the plutonium is removed. It might be possible in this way to effect a 20- to 50-fold concentration after which the acetate method might be added for decontamination.

Control work. The 5,000-gauss magnet seems to increase the maximum beta background permissible in the counting of alpha particles by a factor of at least ten. English is going to meet at periodic intervals with Struther's men to discuss control work problems.

Purification. Orlemann states that the Project will probably buy a new optical spectrograph. With respect to the progress being made along the analytical lines in Boyd's section, Wayne Marshall has been assigned to the spectrographic work full-time; and another man is being sought. On the general analytical side, Potratz, plus three other men, are being transferred full-time to purification work. It was decided that reporting on the analytical work will be done as follows: (1) methods in Boyd's reports and (2) applications in my reports. Microchemical analytical development will have to be done largely in our section. Dempster and Nier have promised to try tests on the mass spectrograph. Carbon and oxygen analysis should have priority because of the refractory problem.

5/2/43 (cont.)

After the meeting Helen and I and Al and Wilma Ghiorso hurried to the Loop where we had lunch with Wally Reynolds, Don Cooksey, Bob Thornton, and the Arthur Snells in the Walnut Room of the Bismarck Hotel. This offered a welcome chance to reminisce about the Berkeley Radiation Laboratory where Wilma worked as Cooksey's secretary and Helen as Ernest Lawrence's secretary. Reynolds, Cooksey, and Thornton were on their way to Oak Ridge for a visit in connection with the work on the electromagnetic process for enriching U^{235} going on in the Y-12 area. They were in Chicago on a lay-over between trains.

We returned home in the late afternoon, then joined my UCLA friends, the Baumbachs, for dinner.

Monday, May 3, 1943

I sent a letter to Oppenheimer in Berkeley telling him I have learned that Blanchard, a member of Latimer's group, has been offered a position at Site Y. I explained that we have been very much interested in having Blanchard come to Chicago but decided Latimer needed him more. I pointed out the difficulty in making plans for our men if we don't have any idea when we may lose them.

In a memorandum to Stearns I suggested that the salary scale for academic employees be increased to avoid losing men we badly need. As examples of men we may not be able to get because we have offered less than our competitors, I cited the cases of Hindman, Blanchard, and Morris.

Helen's volunteer work with the YWCA took her to the Metro Y (59 E. Monroe) today.

The steel strike which started Saturday has been halted for two weeks, during which time negotiations will take place.

Tuesday, May 4, 1943

In the last couple of days Miller has run through a cycle on the Berkeley Sodium Uranyl Acetate Process on the tracer scale, and he finds very good recovery of plutonium.

In accordance with my request of April 13 Creutz is sending to Hamilton in Berkeley two uranium discs cut from a large ingot and also two of Hamilton's probes to which uranium metal sheets have been silver-soldered. With regard to preparing similar targets of thorium, Creutz indicates in a letter sent to Hamilton today that he has not yet been able to obtain any thorium metal.

This is the war news summary as it appeared on the front page of the newspaper this morning: African Headquarters - Yanks capture Mateur in 14 mile advance. Chungking - Chinese repel 3 Japanese attacks on town. London - Reds kill 900 men in Kuban fighting.

Wednesday, May 5, 1943

I sent a memorandum to Greenewalt asking if he could arrange to have sent to us from the du Pont Experimental Station some equipment used by Balthis in his experiments on the hot-wire technique for the preparation of certain compounds and metals.

I wrote Latimer in Berkeley suggesting ways of replenishing the file of CC and CN reports which Art Wahl has taken with him to Site Y.

S. K. Allison was appointed today as Director of the Metallurgical Laboratory by Compton, who also appointed H. D. Smyth as Associate Director (with principal responsibility for coordinating the P-9, heavy water pile, program). Compton will continue to serve as Project Director, coordinating the scientific activities of the Project as a whole, including the Metallurgical Laboratory at Chicago, the Clinton Laboratories, and associated contracts. Hilberry is to continue as Compton's aide, serving as Assistant Project Director. Compton is trying to obtain the services of Robert Brode of the Physics Department at Berkeley as an Associate Director for the Metallurgical Laboratory, with particular responsibility for coordinating the graphite unit program.

In a memorandum to Greenewalt, Allison summarized the present goal as to the necessary purity of plutonium-239 to be delivered from Hanford as follows: (1) It should be produced in the form of some compound, the exact composition of which is at present undetermined, but should be at least 99% pure. (2) In particular, the elements lithium, beryllium, and boron should not be present to more than 0.01% by weight. (3) The gamma-ray activity of a 250-gram batch should not be greater than that of 20 millicuries of radium.

I attended an evening meeting of the Purification and Metal Production groups of Section C-I. Others present were Baumbach, Boyd, Brown, Cefola, Cunningham, Davidson, English, Mark Fred, Jaffey, Katz, Katzin, Kirk, Kohman, Magel, Orlemann, Patton, Potratz, Rosenfels, Clifford Smith, Stoughton, and Willard. The general analytical problem was reviewed, and the uncertainty of the impurity limits at present pointed out. Kohman is to keep in touch with Roberts's study of α,n yields. Mark Fred outlined the pyroelectric concentration method used in the analysis of uranium spectrographically. Using 100 mg samples, he gave the following estimates of detectable impurities in ppm: Be, .01; B, .03; Li, 0.1; Na, 1; Mg, 10; Al, 10; Si, 5; P, 10; K, 10; Fe, 5; Ca, 10. Potratz discussed chemical methods for the detection of iron, aluminum, chlorine, and sulfur. Orlemann

mentioned several possibilities, none too good, for oxygen and carbon analyses. A method which is to be tried is to treat the metal sample with Br_2, volatilize the bromide which can be easily collected and leave the oxide behind. The oxide is then determined by counting the alphas from Pu^{239}.

Six U.S. ships have routed twelve Japanese ships in a battle in the Arctic according to today's newspaper.

Present at the regular Wednesday morning Laboratory Council Policy Meeting in Room 209, Eckhart Hall, this morning were Allison, Compton, C. M. Cooper, Doan, Fermi, Franck, Hilberry, Jeffries, Miles, Smyth, Spedding, Stearns, Stone, Whitaker, and Wigner. Compton opened the meeting, as he often does, with his "State of the Nation" message. In this he summarized as the critical problems that require solution the following: (1) determination of the conditions for a controlled, self-sustained fission reaction; (2) provision of adequate amounts of primary materials in sufficient quantities and proper purity; (3) developing methods for operating the self-sustaining nuclear chain reaction at large power, with emphasis on the water-cooled graphite configuration; (4) extraction and purification of plutonium, indicating that methods of separation from uranium and fission products are available, but purification to the desired stage is unsolved but hopeful; (5) maintenance of safety and health of operators and public. Among the routine matters discussed was a suggestion by Doan to purchase photostat equipment since the use of the equipment in the Oriental Institute requires three days a week; it was agreed to purchase such equipment. There was also discussion and agreement to expand the Ryerson shop facility. Then in line with Compton's request that a Division Director give a report each week at the Laboratory Council Policy Meeting, Cooper reported on the status and progress of the research and development program in the Technical Division. These reports are to be given by the Division Directors to the Council at the Wednesday meeting following the weekly Monday Laboratory Council Information Meeting at which the Division's program is presented.

Thursday, May 6, 1943

Harrison Brown sent a memo, in my name, to Stearns giving the educational and personal background of K. A. Kraus in whom we are interested.

In a memorandum in my name, Willard informed Stearns of the desired starting dates for Gordon Johnson of the University of Wisconsin and Jonathan Dixon of Haverford College.

Perlman wrote, in my name, to Whitaker in response to his request of May 1, giving information on our plans for isolating plutonium from the Clinton Laboratories' separation plant. He asked that our chemists there receive the concentrate in batches that contain not more than

10 millicuries of gamma fission product activity nor more than 100 millicuries of 93^{239}. He indicated the method by which the plutonium will be isolated is not yet finally determined -- the only method thoroughly tried is successive wet fluoride cycles, which is time-consuming. The plutonium will probably be isolated as the peroxide and should then be at least 90% pure with the principal impurity being lanthanum.

Compton received a memo today from Greenewalt dated May 4, 1943, in which Greenewalt points out that a decision between the Wet Fluoride and Bismuth Phosphate Processes should be made by June 1. He suggests that Sutton's Separation Processes Section and W. Q. Smith's Chemical Engineering Section make tests of these processes before that date in order that sufficient information will be available to make the decision.

I attended a meeting in Franck's office to discuss Chicago information centers; others present were Boyd, Burton, Coryell, Dew, Franck, Mulliken, Potratz, Sugarman, and Willard. The meeting was concerned with setting up a central Chemistry Library for classified reports. In addition each Section Chief will have a report file; I indicated a need for reports in the categories CN, CC, CE, CP, CT, and CN-CF.

Helen went to the Crerar Library to abstract articles for the "Table of Isotopes."

This evening Kirk was the speaker at the Research Assistants seminar with a continuation of his discussion of uranium and plutonium metal production. He described in some detail the various methods of producing uranium metal and methods of measuring its properties and then related this to our proposed experiments on the production of plutonium metal.

Friday, May 7, 1943

During the last week Cunningham has conducted experiments to measure the adsorption of plutonium by silica gel from 10% UNH solutions at high concentrations of plutonium. At a concentration ratio of uranium to plutonium of about 800:1, which is somewhat lower than that which will exist in neutron-bombarded uranium from the piles at Site W, he finds in two equilibrium experiments an adsorption of plutonium of 61% and 64%. Repeating the experiments at higher ratios of uranium to plutonium, namely about 100,000 to 1, in two equilibrium experiments, he finds adsorption of plutonium to the extent of 79% and 81%.

Vance Cooper has tested the carrying of tracer amounts of plutonium in the extraction step of the Bismuth Phosphate Process. He has tried to increase the carrying by reducing the concentration of H_2SO_4 and so keeping the uranium in solution by adding sodium

sulfate. He finds that he can reduce the H_2SO_4 from the standard 1 N to 0.5 N by adding 2 N Na_2SO_4 without getting a precipitate of uranyl phosphate. Under these conditions he finds that bismuth phosphate carries 99% of the tracer plutonium.

I received a letter from Joe Hamilton in Berkeley letting me know he is sending, along with the assay results, a sample of unseparated fission products that has been freed from uranium. He said the uranium metal has not arrived as yet but promised me an adequate bombardment when it does.

A listing of Metallurgical Project personnel as of today shows the following organization of the Chemistry Division:

G. T. Seaborg (Chief of Section I)

J. E. Willard (Associate Section Chief)

S. G. Thompson (Group Leader)
H. S. Brown (Group Leader)
R. W. Stoughton (Group Leader)

I. Perlman (Associate Section Chief)

B. B. Cunningham (Group Leader)
E. F. Orlemann (Group Leader)
S. G. English (Group Leader)

P. L. Kirk (Associate Section Chief)

M. Burton (Chief of Section II)

W. M. Garrison (Group Leader)
C. V. Cannon (Group Leader)
T. J. Neubert (Group Leader)

C. D. Coryell (Chief of Section III)

N. Elliott (Group Leader)
N. Sugarman (Group Leader)
W. Rubinson (Group Leader)
A. Turkevich (Group Leader)

G. Boyd (Chief of Section IV)

H. A. Potratz (Associate Section Chief)

R. Curtis (Group Leader)
M. S. Fred (Group Leader)
R. W. Spence (Group Leader)
R. I. Martens (Group Leader)
F. Huke (Group Leader)

The organization of the Technical Division under C. M. Cooper showed Development Engineering--Section Chief, Leverett; Group leaders, Lyon, Abbott, and Johnson. Chemical Engineering--Section Chief, W. Q. Smith; Group leaders, Apple, Peery, and M. D. Peterson.

Separation Processes--Section Chief, Sutton; Group leaders, Balthis, Olsen, and J. H. Peterson. Metallurgy--Section Chief, Chipman. Technological Group--Group leaders, Creutz and Greninger. Coatings - Corrosion - Erosion Studies--Group leaders, Howe and Foote. Control and Instrumentation--Section Chief, Wilson; Group leaders, Brill, Jesse, Overbeck, and Kanne.

Helen and I had the Kirks, Allisons, and Willards to dinner in our apartment this evening. This gave us the opportunity to repay each of them for the hospitality shown us by including us in dinner parties in their homes. It was a congenial group, and everyone had a chance to get better acquainted.

Saturday, May 8, 1943

Today Cunningham got back to the plutonium concentrates prepared by Cefola from the material given him by the chemical engineers resulting from their extractions from neutron-bombarded UNH of the third St. Louis bombardment which ended January 23 (Chicago III). Cunningham made a preliminary assay of one purified fraction of this material on April 29. Now he has assayed all the fractions and finds a total of nearly 350 micrograms of plutonium. He will attempt to isolate as much as possible of this in pure form.

Covey is in St. Louis today to make arrangements for the fifth neutron bombardment of uranium at the Washington University cyclotron (Chicago V).

Arthur C. Bond left Section C-I and the Met Lab to take a teaching position in the Chemistry Department of the University of Chicago on the basis of an urgent request from Professor Schlesinger to Hilberry. This will be a severe loss to us for he is a good man, and I worked hard to get him to join us.

Harrison Brown issued a memorandum in my name giving the results of UF_6 corrosion tests done by Urey's group in Columbia on aluminum, beryllium, uranium, and bismuth. Only bismuth gives poor results.

The Technical Division semiworks operation (Chemical Engineering Section) completed two bismuth phosphate runs (semiworks runs No. 14 and No. 15), each consisting of an extraction step and one decontamination cycle. The decontamination factors are smaller than expected by a factor of ten.

Helen had lunch with Wilma, and in the evening she and I played cards with the Ghiorsos.

Sunday, May 9, 1943

I held a meeting of the Council of my section at 10:30 a.m. in my office. Attending were Perlman, Willard, Kirk, Cunningham, Brown, Orlemann, English, and Thompson. I mentioned the new program on heavy-water pile work which includes the UF_6 system. A good deal of this work, especially the corrosion tests, will be done at Columbia; but any special chemistry will be done in our section. Reports were made on the following:

Wet Fluoride. A run has been made in which the Site W concentrations of fission product elements were added, and no difference was observed as compared with runs made in which only tracer amounts of the fission products were present. Work is still continuing to determine the best oxidizing conditions in the zirconium decontamination cycle (which consists of dissolving the mixed lanthanum-plutonium fluoride precipitate in a zirconium solution, followed by an oxidation-reduction cycle). The carrying power for plutonium from zirconium solutions which has not yet been adequately tested is also being investigated. Greenewalt has requested the semiworks group of W. Q. Smith's section to carry through three runs by the Wet Fluoride Process and three by the Phosphate Process as soon as possible (preparatory to the June 1 decision), principally to test decontamination.

Phosphate method. Reduction by SO_2 has given some trouble in the semiworks equipment by causing sulfur to precipitate. A search is being made for an alternate reducing agent. It is possible that high concentrations of fission products under Site W conditions might result in a zirconium phosphate precipitate. It was suggested that the addition of a small amount of HF might complex the zirconium to a sufficient degree to prevent the precipitation.

Adsorption method. Cunningham has shown good adsorption of plutonium by silica gel in batch tests at four times Site W concentrations. Under the standard conditions about 65% of the plutonium was adsorbed as compared with 80% when tracer amounts of plutonium are present. Fries is working on high temperature adsorption and finds that only about 0.1% of gamma fission product activity adsorbs at room temperature if a previous treatment has been made at about 90°C.

Dry fluoride work. Corrosion tests are being carried out at present at Wilmington which, when finished, will be discussed in terms of what other tests need to be made in view of recent modifications in the method. Cobaltic fluoride has been tried as a solid fluorinating agent for plutonium with negative results.

Control work. The question of corrections for uranium alpha particles in precipitates seems to be a rather complex one, and several people are either working on it directly or are gathering data from their experiments that will be of aid in evaluating its causes. English reported that the best instrument to count alpha particles over the beta backgrounds that will be encountered will probably be one that

combines a magnetic field with moderately high resolving power of the electronic circuit used to amplify the alpha particle pulses from the ionization chamber. Certain troublesome features of the high resolving power instrument could thus be eliminated.

Concentration methods. A possible program was outlined for developing the peroxide, iodate, and acetate concentration methods for use at Sites X and W. The recommendation was made that one or two men start working on the concentration problem as soon as possible.

Metal production. The difficulty in remelting small samples of powdered plutonium metal is undoubtedly caused by the oxide coating. A search is being made to find a flux for allowing the metal to melt through the coat. Work is also proceeding on the mounting of small samples of metal in lucite for metallographic examination.

The Soviets have retaken Kursk, a victory approaching the Russian triumph at Stalingrad.

Monday, May 10, 1943

Thompson participated in a Bismuth Phosphate Process conference with Apple, Balthis, Chambers, Peery, M. D. Peterson, W. Q. Smith, and Sutton. Semiworks runs C-14 and C-15 were discussed. Reasons for the low decontamination factors (50-100 rather than 500-2,000) were considered, and it was concluded there should be thorough cleaning of tanks and lines before the next run. It was agreed that Thompson will continue his study of reducing agents--he has been investigating ferrous iron and hydrogen peroxide.

Werner has been checking the carrying of plutonium by bismuth phosphate at Site W concentrations of plutonium from solutions that do not have any uranium or sulfate present; this corresponds to subsequent steps following the extraction step in the Bismuth Phosphate Process. In varying the concentration of bismuth he finds that at least two milligrams per milliliter is required in order to get adequate carrying of the plutonium. Today, however, he had some rather disappointing results. He found under the conditions of his experiment the carrying of plutonium is in the neighborhood of only 60%.

Cefola tested today the by-product precipitation step of the Wet Fluoride Process at concentrations of plutonium that will exist in the Site W chemical extraction plant. Plutonium was oxidized with the peroxydisulfate-silver ion combination with zirconium and lanthanum present in this solution. When HF was added to precipitate lanthanum fluoride, it was found that only 1% of the total plutonium was present in the precipitate indicating that the oxidation and by-product precipitation are satisfactory in the Wet Fluoride Process at the concentrations that will prevail at Site W.

Helen went to the West Side Y (101 S. Ashland) in the morning and later to the Girl Reserve meeting held at the 63rd Street YWCA.

The Soviet troops have entered the suburbs of Rostov.

Tuesday, May 11, 1943

Covey returned at 7:45 a.m. from St. Louis where he has been arranging the fifth cyclotron neutron bombardment (Chicago V).

Perlman has tested the use of uranium peroxide as a carrier to isolate the plutonium to come from the chemical extraction plant at Site X following the possible use of the Wet Fluoride Process. Using the concentrations of plutonium that would be found at that extraction plant after the operation of the Wet Fluoride Process, he finds that uranyl peroxide carries about 75% of the plutonium in one precipitation and that most of the remaining can be recovered by subsequent precipitations of uranium peroxide by adding more uranyl ion to the solution.

Helen worked at the Metro Y today.

A Stockholm correspondent reported that the Germans are evacuating Kharkov. Russian forces are reportedly only 2.2 miles away.

Wednesday, May 12, 1943

I felt ill today and stayed at home. However, Helen and I worked on the "Table of Isotopes."

My office received a telegram addressed to me from Gofman asking for the date of the next Laboratory Council Information Meeting on Chemistry.

Cunningham completed today the isolation of the plutonium in the procedure that he started on Saturday. He put the bulk of the material (about 350 micrograms) through another lanthanum fluoride oxidation-reduction cycle, then precipitated the combined material as plutonium peroxide and gave it to English and Ghiorso for additional counting of gamma rays and detection of any gamma rays from alpha particle bremsstrahlung. They made measurements on the gamma rays emitted from this plutonium sample. Their results are in essential agreement with their measurements of April 23, that is, about one gamma ray per 10,000 alpha particles and about three or four electrons per 10,000 alpha particles. They also find that the yield of gamma rays is the same whether the alpha particles are stopped by cellophane or lead, indicating the absence of any bremsstrahlung effect.

Today Werner continued his experiments on the carrying of plutonium at Site W concentrations with bismuth phosphate in the absence of uranium and sulfate, with the hope of improving on the disappointing

results that he found on Monday. He finds that by varying the conditions, adding the reagents in concentrated solutions and then diluting, that the bismuth phosphate carries plutonium to the extent of about 96%. This is much more encouraging.

Present at the Laboratory Council Policy Meeting this morning were Allison, Compton, C. M. Cooper, Doan, Fermi, Franck, Hilberry, Miles, Smyth, Spedding, Stearns, Stone, Whitaker, and Colonel Matthias who has responsibility for the construction of the facilities at Site W for the Manhattan District. Later Wigner and Urey joined the group. Compton opened the meeting by indicating the need for essentially doubling the space for the chemistry work here in Chicago and read his draft memorandum to Major A. V. Peterson, the local representative of the Manhattan District, making a recommendation for such additional space. He said that the present space in New Chemistry at Ingleside houses about 140 staff and the addition being recommended would bring the capacity up to 250. The annex to New Chemistry that is recommended would about double the space. He also said that he is making a similar request for additional facilities at Argonne to take care of the heavy water experimental work, again about doubling the laboratory space.

Compton then called on Colonel Franklin T. Matthias of Groves's staff to bring the group up to date about Site W in southeastern Washington, where construction started six weeks ago on March 22. Matthias presented a sketch of the site (see Figure 1) showing the proposed location of the piles, the chemical separations plants, and the town — all located near the Columbia River, the water-cooled piles essentially adjacent to the river at six-mile intervals along the south bank and the four chemical separations plants in two areas some distance from the river and south of Gable Mountain. Hanford, shown on the map, is a construction camp and was a small town before the hundred or so inhabitants were evacuated. The permanent town to be constructed for personnel is at a village 23 miles down the Columbia River called Richland. Compton then called on Stone to give his report on the program of the Health Division. Stone raised the question as to the responsibility of his division at Site W, and Compton indicated that his responsibility is only to keep the people at Site W informed. There was then a discussion of the recovery of fission products for use as a weapon. Cooper noted that both the Wet Fluoride and Bismuth Phosphate Processes would recover the fission products in relatively high concentration. Compton said that he would like to see this considered and studied further and will appoint a coordinator to do this.

Thursday, May 13, 1943

I received a letter from Hamilton saying that the uranium metal shipped by Creutz arrived on May 11, and he will put some in a good place near the cyclotron's beryllium target for me. He has some carrier-free

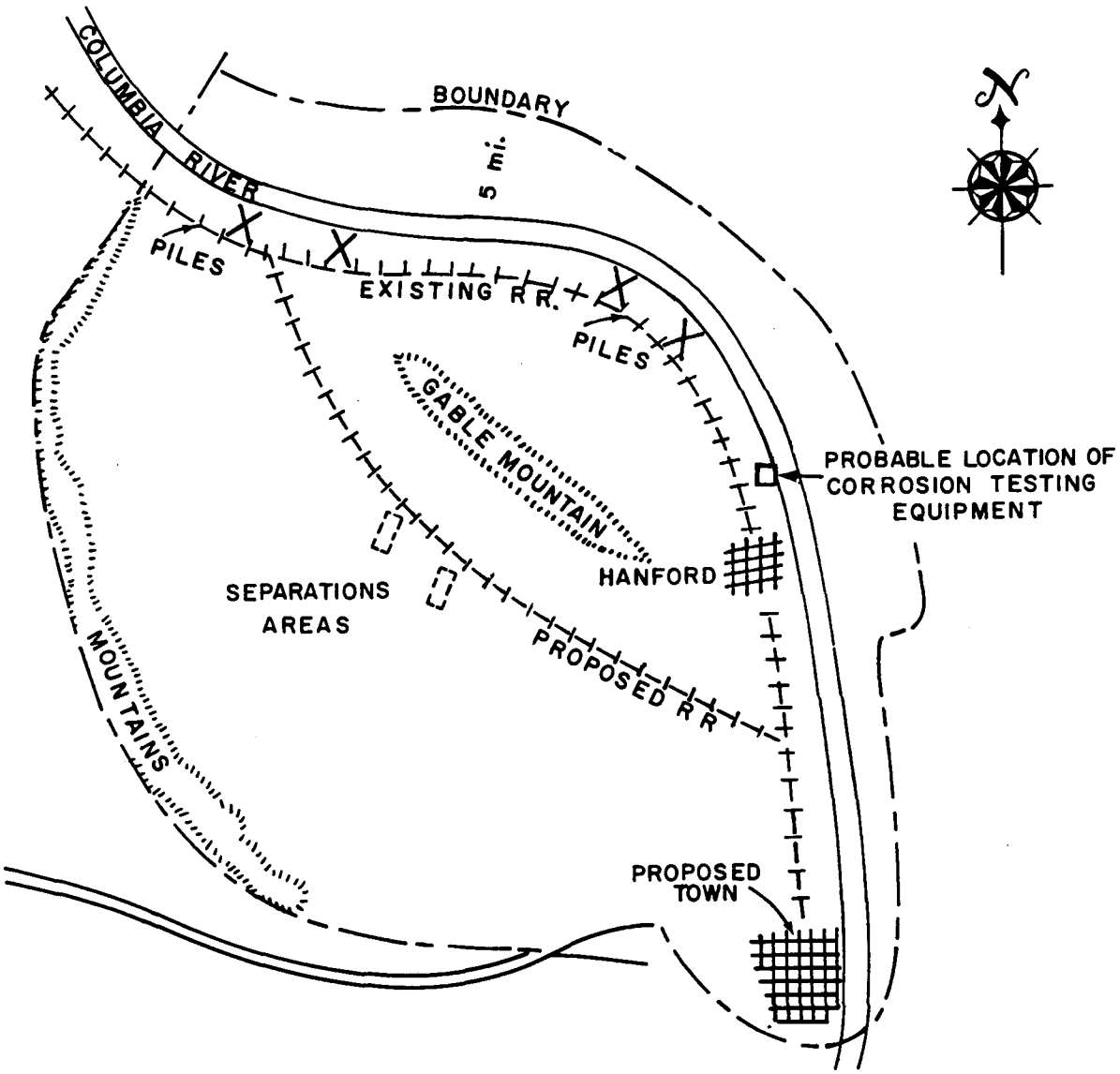

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Figure 1. Sketch of plans for Site W. May 1943.

radioruthenium if I need it. He asked if we could make up a counter circuit with a scale of either 16 or 32 to replace Tibbetts' counters which are giving lots of trouble.

I wired Gofman that the next meeting of the Laboratory Council for Chemistry Information will be May 24.

Helen went to the Crerar Library today to get more references for the "Table of Isotopes" which we worked on this evening.

The regular meeting of the Chemistry Section C-I Research Assistants was held in my office this evening.

In a nation-wide radio address at the dinner of the White House Correspondents Association, President Roosevelt promised decisive blows at Tokyo, "constant and unrelenting pressure" on Germany and Italy with an air offensive about to begin in Tunisia.

Friday, May 14, 1943

Today Cunningham resumed work on the recovery of plutonium and neptunium from the 40 pounds of neutron-bombarded UNH in the fourth large St. Louis bombardment (Chicago IV); work on this recovery was suspended on April 26 in order to do some other experiments.

The fifth large neutron bombardment of UNH at the St. Louis cyclotron (Chicago V) started today. A total of 500 pounds of UNH is involved this time. A neutron bombardment of eighty pounds of uranium metal is also commencing. This is to be used by Cunningham as a source of 93^{237}.

In the Technical Division, Sutton's complete section is now concentrating on the Bismuth Phosphate Process with most of the effort being expended on the process details before the three decontamination cycle cross-comparison runs between the Bismuth Phosphate and Wet Fluoride Processes are made in the semiworks.

Helen had an appointment with Miss Young about the Girl Reserve program.

In the evening Helen and I again worked on the "Table of Isotopes." Stan Thompson stopped to see us later.

Saturday, May 15, 1943

C. Blanchard, R. B. Duffield, J. W. Gofman, and I are issuing our report, "Test for Slow Neutron Fission of U^{234}," with the report number CF-673. This describes work carried on at Berkeley that I initiated before I left there in April of last year. The U^{234}, a sample weighing

13.2 micrograms, was prepared by Fontana and co-workers by our method of extracting uranium-free UX_1 from uranium and allowing the UX_1 to decay to U^{234}. The slow neutron-induced fission rate in this U^{234} sample was measured with Ra-Be neutrons, in comparison with that of a U^{235} sample of known weight. Results indicate that U^{234} does not undergo fission with slow neutrons. The ratio of the measured fission cross section of U^{234} to that of U^{235} is 0.002 ± 0.001, and this small effect could be accounted for by only 4 micrograms of natural uranium impurity in the U^{234} sample.

Dan Miller has completed his test of two cycles of the Sodium Uranyl Acetate Process and has found a good yield of plutonium and good decontamination from fission products in agreement with Berkeley results.

In the Technical Division the results of bismuth phosphate semi-works run No. 14 (extraction and one decontamination cycle) show a product yield of 83%. Fission activity calculations on run No. 15 (extraction and one decontamination cycle) show a gamma decontamination factor of 108 as compared with 81 for the No. 14 run. A third bismuth phosphate run, No. 16 (extraction and two decontamination cycles), was carried out during the week using a combination of $K_2Cr_2O_7$ with ceric ion for the oxidation steps and H_2O_2 for reduction steps. The gamma decontamination factor was 95 at the end of the first decontamination cycle, but the activity level was too low for accurate measurement at the end of the second cycle.

Bert Goldschmidt is in town. He paid us a social call at the New Chemistry Building today; it is the first time I have seen him since his visit the first week in February. So far, the Montreal Laboratory has spent about $100,000 on equipment and supplies. Besides the plutonium and fission products that we gave him in February, the Montreal Laboratory has on hand three tons of uranium oxide, ten tons of high-quality graphite, and 180 liters of heavy water that was smuggled out of Norway in 1940. The Laboratory now has over a hundred members on its staff, including the heavy-water team from Cambridge. Outstanding Canadians there are George C. Laurence, formerly of the Division of Physics and Electrical Engineering, National Research Council at Ottawa; B. Weldon Sargent, who comes from the Physics Department of Queen's College, Kingston, and like Laurence, studied under Lord Rutherford at Cavendish Laboratory; and George M. Volkoff from the Department of Physics, University of British Columbia. I knew Volkoff in Berkeley, where he got his Ph.D. in 1940 under Oppenheimer in theoretical physics.

Considerable gloom prevails at the Montreal Laboratory since Conant sent a memorandum to the British and the Canadians in early January, cutting off the flow of information from the United States. It reads in part as follows:

"This memorandum sets forth the general rules and regulations for interchange of information with the British and Canadians. It derives from the basic principle that interchange on design and construction of new weapons and equipment is to be carried out only to the

extent that the recipient of the information is in a position to take advantage of this information in this war. Since neither the Canadians nor the British Governments are in a position to produce elements '49' and '25,' our interchange has been correspondingly restricted by orders from the top.

"1. Electromagnetic Method--No further information to be given to the British or Canadians.

"2. Diffusion Process--All interchange, including basic scientific information, to be under the control of General L. R. Groves, who will draw up specific regulations on this matter to cover both the industrial companies concerned and all others working on this subject.

"3. Production of Heavy Water--No further information to pass to the British or Canadians.

"4. Use of Heavy Water in Chain Reaction--Inasmuch as the Canadian-British group will be working on the scientific branches of this subject in Montreal, it is provided that there be complete interchange of scientific information concerning chain reactions between the group at Montreal and the group at Chicago. The Chicago group, however, is not to give any information on either properties or production of element '49,' including methods of extraction or design of power plant.

"The Canadian scientific group is to work closely with the du Pont engineers in order to provide the latter with basic scientific information. Engineering design is to be given to the Canadian group by the du Ponts only insofar as is necessary for their carrying out basic scientific work.

"5. Fast Neutron Reaction and all matters concerned with 'bomb' and use of material--No further information to be given to the British or Canadians.

"6. Manufacturing details of UF_6, metal, etc.--Under the jurisdiction of General Groves. In general, no information to be transmitted."

Another reason for the doldrums and rankled feelings is that the Montreal Laboratory was expecting to get raw materials from the U.S., such as heavy water, purified uranium oxide, and uranium metal; but now the supply has been cut off. Goldschmidt says they will go it alone until, hopefully, the situation improves.

Helen and I had dinner at the Perlmans' (5638 Kenwood) this evening. Goldschmidt was there, and we had a congenial evening of reminiscing.

The Nazis lost Rostov and Voroshilovgrad, an important industrial center 85 miles northwest of Rostov.

Sunday, May 16, 1943

I held a meeting of the Council of my section in my office at 10:30 a.m. Attending were Perlman, Willard, Kirk, Cunningham, Brown, Orlemann, English, and Thompson. I mentioned that W. Q. Smith has been asked by the du Pont Company to set limits of plutonium yields to be expected through operation of the Wet Fluoride Process. In terms of total losses through three decontamination cycles, he gave a range of 16-55% which seems like a reasonable guess. Next Wednesday there will be a meeting with Greenewalt for comparing the Wet Fluoride and Phosphate Processes. This meeting is a preliminary to the meeting to be held June 1, following which a decision will be made as to which process should be used in the Site W plant. The following topics were discussed:

Present status of Wet Fluoride Method. A real problem exists in finding the proper reducing agent in (1) the presence of uranium and HF, and (2) the absence of uranium but in the presence of zirconium. Oxidation seems to work satisfactorily in solutions from 1-1.5 N HNO_3 in which the zirconium to lanthanum ratio is about two. A run was made using Site W concentrations of fission products in the phosphate-modified Wet Fluoride Process. In one decontamination cycle plus the extraction cycle, a factor of 5×10^5 in the reduction of beta activity was achieved. Work is proceeding on metathesis to the hydroxide as a step for placing lanthanum and plutonium fluorides in solution.

Present status of the Phosphate Method. The problem of finding a good reducing agent is the most difficult step in this process. If H_2O_2 is present in excess, there seems to be an inhibition in the carrying of the product by bismuth phosphate, possibly caused by a reaction between the phosphate and the peroxide. It is planned to attempt reductions under conditions in which most of the oxidizing agent is reduced with hydrogen peroxide, while the remainder is reduced with ferrous ion or SO_2. An attempt has been made to use hypophosphite as a reducing agent, and a very remarkable time curve was obtained. The "percent reduction" as a function of time climbs to a rather sharp peak after which it falls rapidly. This brings up the possibility that plutonium is reduced in time to an oxidation state less than +4 which is not very fluoride insoluble (carrying by a lanthanum fluoride precipitate was used to determine the degree of reduction). A discussion was then held as to whether or not some effort should be diverted to the investigation of lower oxidation states of plutonium in the hope of finding compounds of interesting properties or even means of producing the metal.

Control work. The uranium correction seems to be the most serious problem confronting this work at this time. It may be as high as 30-40 alpha counts per minute in a given sample of plutonium.

Adsorption. The work on high temperature adsorption is still proceeding with the result that fairly good decontamination seems to be possible by use of alternate hot and cold adsorptions. Batch adsorption using silica gel is receiving considerable attention especially in view of the interest shown by the engineering group for such a method.

5/16/43 (cont.)

Helen and I took a long walk this afternoon, stopping at both the Baumbachs' and Ghiorsos' before returning home.

Russian troops are in Kharkov today, while the third day of a tank battle continues in North Africa following an offensive by Rommel.

Monday, May 17, 1943

Weekly meetings are now being held on Monday with attendance by our chemists concerned with separation work and by the chemical engineers. These meetings are devoted to a discussion of the Bismuth Phosphate Process for which there is a stepped-up program this month in order to obtain information prior to the June 1 deadline. Attending the meeting today were Thompson and Willard from our Chemistry Section C-I and from the Technical Division, Sutton, W. Q. Smith, M. D. Peterson, Peery, Balthis, Apple, and Elliott. The results of semiworks runs Nos. 14-17 on the Bismuth Phosphate Process were presented. They have found that the oxidation with dichromate plus ceric ion is satisfactory and also have found excellent reduction in lab tests with the new hydrogen peroxide-ferrous ion combination. They plan to use 10 pounds of neutron-bombarded UNH in run No. 18 and 40 pounds of neutron-bombarded UNH from Chicago IV, the bombardment that ended April 12, in run No. 19.

H. S. Brown and O. F. Hill are visiting the du Pont Company Experimental Station in Wilmington to discuss the dry process corrosion studies being carried out there under H. L. Maxwell.

Willard lectured today to the Site X trainees on methods for extracting plutonium from neutron-irradiated uranium.

Kohman was asked by Burton today to provide data on ion accelerators for the Project Handbook.

Helen went to the West Side Y in the morning and attended the Girl Reserve meeting on 63rd Street in the afternoon.

Helen, Al Ghiorso, and I went to a dinner for Miss Woodsmall. After dinner Helen and I saw the movie, "Cabin in the Sky," featuring Ethel Waters, at the downtown Apollo Theater.

Tuesday, May 18, 1943

Since last Friday, Cunningham has continued his recovery program for plutonium and neptunium from the 40 pounds of neutron-bombarded UNH from the fourth large St. Louis neutron bombardment (Chicago IV). He has put the material through a cold bromate oxidation process designed to separate the neptunium from the plutonium. In the neptunium fraction he finds a recovery of 81% based on the 93^{239} tracer present; he set this aside for later recovery of the 93^{237}. His assay shows the presence of about 230 micrograms in the plutonium fraction. He plans to combine

this with the approximately 350 micrograms purified last Wednesday from Cefola's material.

During the last couple of days Werner has continued his experiments on the carrying of plutonium by bismuth phosphate at Site W concentrations in the absence of uranium and sulfate ions. He finds that by adding the bismuth last and very slowly, allowing the precipitation to take place at temperatures as high as 95°C, that the carrying in a number of experiments is consistently around 95% and in some experiments as high as 98-99%. This is good news indeed.

General Groves is in Chicago for a meeting with Compton on the potential military value of fission products. It was agreed that research on this possibility should not be allowed to interfere with development of a separation process for plutonium.

Using neutron-bombarded UNH from the large St. Louis bombardment that ended on April 12 (Chicago IV), Miller has run through two complete cycles of the Berkeley Sodium Uranyl Acetate Process using the Berkeley flowsheet. He obtained a yield of plutonium of about 95% and a decontamination factor for beta active fission products of a few times 10^4. The decontamination factor was probably better than this because much of the beta activity remaining may have been due to 93^{239}. He left by train at 4:30 p.m. on a visit to Notre Dame to use the van de Graaff accelerator for studies of radiation damage on the chemicals in the various steps of the Sodium Uranyl Acetate Process.

At the meeting of our extraction group in my office this evening the discussion was devoted to a consideration of the present status of the Wet Fluoride and the Bismuth Phosphate Processes in anticipation of the impending need to make a decision between the two soon after June 1.

Our army has been pushed back 22 miles in Rommel's drive in Tunisia.

Wednesday, May 19, 1943

Perlman sent me a memo outlining a proposed program to study the methods to be used in isolating a relatively pure plutonium compound from the Site X material that will come out of the chemical extraction plant. He believes that the peroxide method and the iodate method look most promising: adsorption or dry methods could not be made usable in the near enough future. The acetate method is a possibility, but it involves an oxidation-reduction cycle that could lead to losses. For the peroxide method he recommends a program of investigations of the carrying power of uranium peroxide for plutonium and of the dissolving of lanthanum and plutonium fluorides. For the iodate method he suggests all steps in the procedure be checked in view of the lack of information.

5/19/43 (cont.)

I attended the scheduled meeting today at 2:30 p.m. in Room 209, Eckhart Hall, to bring Greenewalt up to date on separation studies. Besides Greenewalt those present were Charles Cooper, Squires, Sutton, W. Q. Smith, Miles, Collins, Willard, Thompson, and Apple. Most of the discussion revolved around the Bismuth Phosphate Process and the prospects for having enough information by June 1 to make a decision between this process and the Wet Fluoride Process. I summarized the status saying that dichromate ion seems to be working out well as the oxidizing agent, that perhaps a mixture of hydrogen peroxide with a little ferrous ion might be satisfactory as reducing agent, that carrying at Site W concentrations of plutonium (that is, at ratios of the order of 100:1 of bismuth to plutonium) seems to be satisfactory as demonstrated by the ultramicrochemical work provided certain precautions are taken with respect to the rate of precipitation, and so forth. Greenewalt indicated that he would like to see a semiworks run on the Bismuth Phosphate Process through two decontamination cycles, and Sutton and Smith said that this will be done. I indicated that there are a number of experiments that need to be made within the next two weeks in order to get ready for the June 1 meeting with Greenewalt and his people.

I asked Hilberry by memo to procure a set of six radium-beryllium sources ranging from 0.1 microcurie to 10 millicuries in strength; details are to be worked out by Kohman.

Allison wrote Oppenheimer that Groves and Greenewalt do not want him (Allison) to make any plans for moving to Site Y at the present time; therefore Oppenheimer should go ahead and fill the Associate Director job at Site Y with someone else as soon as possible.

In response to Garner's inquiry Cefola wired him at Site Y giving the source of supply of a micromuffle furnace.

John Willard sent a letter, over my name, to Gordon Johnson at the University of Wisconsin suggesting he start working at the Met Lab as soon as possible in order to enhance his chances of securing a draft deferment.

Wilma spent the afternoon with Helen today.

Our troops are having more trouble in Africa; they yielded three additional towns to Rommel.

Present at the Laboratory Council Policy Meeting this morning were Compton, C. M. Cooper, Doan, Fermi, Franck, Hilberry, Miles, Smyth, Spedding, Stearns, Stone, and Wigner. Greenewalt was also present: this gave the group an opportunity to exchange ideas and information with him. In his "State of the Nation" message Compton indicated that construction at Site W is now under way; and, therefore, within the next month or two most of the features to be built into the piles and the chemical separations

plants will have to be decided upon. He said that the decisions concerning Site X in these matters have mostly been made already. Compton said that there was a meeting with Groves yesterday regarding the utilization of fission products and reminded the group that Conant heads a committee to look actively into the potential military uses of these. Franck and Cooper indicated that they do not believe that a concentrated effort on fission product activity and purification methods should be made at this time because the present separation methods already will recover large fractions of these fission products in concentrated form. Compton and Greenewalt seemed to agree that General Groves is of this opinion also.

Compton then called on Fermi to give his report on the research program of the Physics Division. Fermi indicated that the three main problems under investigation by his division are those in connection with the water-cooled piles for Site W at Hanford, Washington, those in connection with a heavy-water pile, and the general problems in the field of nuclear physics that might be expected to have a bearing on our objective. He covered these problems and indicated that a good deal of emphasis will be put on the third; as an example of this, there is the problem of the properties of neutron emission from Pu^{239}.

Greenewalt raised the question of whether there shouldn't be a larger group of theoretical physicists with du Pont in Wilmington because the present group of John Wheeler and Kay Way is not sufficient to answer all their theoretical questions. There was some discussion of this, including the possibility of asking Weinberg to join the du Pont group; but no conclusion was reached after it was pointed out that Weinberg is also very much needed in Chicago. Greenewalt said that he would like to have the matter of "the manual on atomic power" cleared up. It was agreed that this information should be made available without interfering with the general problem of obtaining information for du Pont.

At the end of the meeting Charles Cooper presented in more detail the report he and Franck authored on the recovery and use of fission products that was requested by Compton at the last meeting. They pointed out that the Wet Fluoride and Bismuth Phosphate Processes, as well as various modifications of the adsorption process, lead to good recovery and concentration of the fission products and summarized the weight of material with which the fission product gamma emitters would be associated. It is clear that about 10^6 curies of fission product gamma radiation can be made available in a volume of the order of one cubic foot; and, according to the calculations of Friedman and Turkevich, this is sufficient to develop 100 r per day over an area of the order of one square mile. It should be possible to transport and dissipate such an amount of fission products by air. It was recommended that less emphasis be placed on further

fission product purification on the basis that the recovery processes seem to have solved the problem sufficiently for the present evaluation of their use.

Thursday, May 20, 1943

Miller returned from his trip to Notre Dame at 2:00 a.m. today. He finds that there are no adverse effects from radiation on the Sodium Uranyl Acetate Process.

Today Werner tested the carrying of plutonium at Site W concentrations with bismuth phosphate as the carrier from solutions in which the plutonium is oxidized; this is to test the feasibility of the oxidation-reduction cycle in the Bismuth Phosphate Process. As expected, and hoped for, the by-product bismuth phosphate from the solution of oxidized plutonium carries less than 1% of the plutonium.

Perlman and I today sent a summary of the present status of the work in our section to Franck as follows:

"1. Extraction methods. Certain details in the wet fluoride process such as the choice of the best reducing agent in the decontamination cycles are being worked upon. Several such tests yet remain in order to make it a finished process. A modification in the procedure whereby zirconium phosphate is removed in the oxidized solution of the extraction cycle shows promise of producing excellent decontamination. A factor of more than 10^5 in the extraction cycle and one decontamination cycle might be expected. Studies are being carried out in which the fission elements are present at high concentrations; and, thus far, no differences in decontamination have been observed.

"The bismuth phosphate method is also receiving considerable attention with the point of view of ironing out details of the best oxidizing and reducing agents and methods of complexing the uranium to prevent its precipitation. Microchemical tests are being carried out to determine the behavior of the phosphate method under the concentrations of plutonium to be expected in the Site W material. These various experiments on the phosphate as well as the fluoride extraction methods are of imminent importance because of the short period of time remaining before operation at Site X begins and before a choice must be made as to the method to be primarily designed into the Site W plant.

"In the adsorption tests being carried out with silica gel on the ultramicroscale it has been found that the capacity of the gel appears satisfactory for the highest concentrations of plutonium anticipated. Work is proceeding on the use of silica gel in batch extractions.

"2. Purification. Vacuum sublimation of uranium and plutonium halides is being pursued as a possible means of purification from light impurities and to obtain vapor pressure data that will be needed for

any type of volatilization process. In the case of uranium chloride vapor pressure determinations checked published values satisfactorily.

"Work is continuing on fluorination as a possible purification step from impurities other than fluorine.

"An upper limit on the gamma radiation from plutonium has been set at 1 gamma ray per 10^4 alpha particles. The especially decontaminated sample used had undergone a gamma-ray decontamination factor of more than 10^8 below that present in the bombarded material. No detectable gamma-ray excitation by the alpha particles being stopped in lead was in evidence.

"An analytical program has been outlined as far as is possible at this time to take care of the needs of the purification program and work has been started in Mr. Boyd's section. Anticipated analytical tools include conventional chemical analysis, ultramicrochemistry, spectrography, polarography, mass spectrography, and physical shot-gun methods.

"3. Metal production. Metal is being prepared on the microgram scale with the problem of obtaining a solid pellet still standing in the way of a density determination. The search for a suitable refractory and flux to allow remelting is being pushed. Procedures for metallographic examination of microgram amounts of metal are being developed.

"4. Plant control work. Instruments are being developed to allow the determination of alpha radiation over high beta backgrounds. The steps being taken that are effective are (1) the increase in resolving power of the circuit by altering the time constants, (2) introduction of a magnetic field, (3) use of beryllium electrodes. The methods of preparing samples from the various solutions in the extraction procedure for alpha, beta, and gamma counting are also receiving considerable attention with the primary goal the design of control methods for Site X. Site W control work will involve micro methods and tentative plans have been made for design and procurement of equipment.

"5. U^{233} production. A small group has been assembled and are now working on the many problems involved in the isolation of U^{233} through its protactinium parent. Special emphasis is being given to the design of methods to be used at Site X.

"6. UF_6 pile. Cyclotron bombarded UF_6 is being used to test volatility separations of uranium, plutonium, and fission products."

"Chemical Research--Special Chemistry of 94--Analytical, Report for Month Ending May 15, 1943" (CK-659) is being issued. It explains that a joint analytical unit is being organized as a common responsibility of the sections of G. E. Boyd and G. T. Seaborg. In the absence of the necessary amounts of plutonium, uranium will be used as a substitute for preliminary investigations of methods of analyses. A review of existing methods of analysis for light impurities in uranium shows that they are reasonably adequate with the exception of oxygen, carbon, fluorine, aluminum, and possibly lithium. However,

the methods use 100 mg or more of material; and, as 1 mg of plutonium has been decided upon as the sample size, the major portion of the analytical work will be devoted to development of methods which meet this requirement.

"Chemical Research--Special Chemistry of Plutonium, Report for Month Ending May 15, 1943" (CN and CF-647) is being issued with the following information. Cunningham and Clifford Smith, using their vacuum sublimation apparatus and starting with the metal form, find it possible to prepare and sublime the chlorides and bromides of both uranium and plutonium. In no case, however, is the compound completely sublimed, due presumably to the presence of oxides or possibly the fluoride in the case of the plutonium. By a slight modification of the sublimation apparatus it has been found possible to determine the vapor pressure of uranium halides, the value obtained agreeing in order of magnitude with the published value for UCl_4 of 2×10^{-4} mm Hg at 330°C. English and Ghiorso report on the gamma radiation from plutonium, measured using a sample that has been separated from fission product activity to a greater extent than has been previously attained. These measurements now set the nuclear gamma radiation at less than one gamma ray for each 10,000 alpha particles emitted instead of the 1:1,000 relationship previously reported. The plutonium sample was prepared by Cefola and Cunningham.

With my approval, Edrey Smith, my secretary, asked Mr. Stearns if she could be permitted to work 44 rather than 48 hours per week (taking Saturday afternoons off) because of the strain of working a full six days with a long commuting distance and the lack of time for personal errands. She has to travel via two streetcars and a train in each direction to and from work in the commute from her home in the Beverly Hills section (9514 Damen Avenue) southwest of the Metallurgical Laboratory.

The regular weekly seminar of Research Assistants was held in my office this evening.

Friday, May 21, 1943

James Clark Hindman from the Scripps Institute of Oceanography started work today in Section C-I. He was recruited for us by Kirk, who had him as a student in an undergraduate microchemistry course at Berkeley. We are lucky to get him because the Berkeley group also made him an offer and at a higher salary. Here at the Met Lab his salary will be $3600 per year. We plan to have him work with Cunningham on our ultramicrochemistry program; Kirk introduced him immediately to Cunningham, Werner, Cefola, Patton, Rosenfels, and Baumbach.

I sent a memorandum to Charles Cooper summarizing Brown's and Hill's corrosion conference of May 17 at the du Pont Experimental Station.

5/21/43 (cont.)

In a memorandum to Stearns I said that our situation in regard to men with experience in fluorine work is becoming very critical, and I gave him the names of the following men Harrison Brown has learned about: Walter Koski (Hercules Powder Company), Harry C. Anderson (Johns Hopkins University), and Roger Sweet (Johns Hopkins University).

Anticipating a favorable reaction from Stearns, Harrison Brown sent a letter, over my name, to Harry Anderson, Department of Chemistry, Johns Hopkins University, telling him he will be contacted by Stearns concerning possible employment with us at the Met Lab.

"Chemical Division Research Program" by J. Franck (CN-651) is being issued and describes the main tasks of the Division with manpower and priorities indicated. For my section the tasks are identified as follows. Extraction methods: wet fluoride--A priority, 5-1/2 men; bismuth phosphate--A priority, 6 men; acetate--B priority, 1 man; dry fluoride--B priority, 3 men; adsorption process--B priority, 4-1/2 men. Purification: vacuum sublimation--A priority, 3 men; fluorination--A priority, 2 men; measurement of gamma radiation from plutonium--A priority, 1 man; analytical technique development--A priority, 3 men. Plutonium metal preparation--A priority, 5 men. Plant control work: instrument development, A priority, 3 men; sample preparation--B priority, 2 men. U^{233} production--B/C priority, 4 men. UF_6 pile--A priority, 1 man. Total men: 44.

Saturday, May 22, 1943

General Groves is in Chicago today.

Today Arthur Compton sent a memorandum to Major A. V. Peterson, the Manhattan District representative who is supervising operations at Chicago for General Groves, regarding the urgent and immediate need for more space for the Chemistry Division. He points out that the 20,000 square feet at the New Chemistry Laboratory on Ingleside Avenue is housing most of the 142 people now in the Chemistry Division. He estimates that the Chemistry personnel will have to increase to about 250 and that an annex to the New Chemistry Building will be required in order to accomplish this. The target date for completion of the annex is September 1. At about that time also there will be some alleviation of the problem because about 50 chemists will move to the Clinton Laboratories at Site X. He points out that the number of chemists now at work and the new ones requested to meet urgent justified demands exceeds by about 50 our present accommodations. This emergency has arisen because the chemical problems raised by the chemical engineers in order to specify the details of the chemical extraction plants are requiring more attention than has been anticipated and also because the date of transfer of chemists to Site X has been postponed.

Werner carried out experiments today using his new technique of adding the bismuth very slowly and varying the final concentration of the bismuth. He again finds that concentrations of bismuth of about 2 mg per milliliter are required in order to get good carrying. At this

concentration plutonium carrying of 96% is observed. These experiments again were performed in the absence of uranium and sulfate ions.

Helen and I had dinner at the Ghiorsos', a more special occasion than usual because today is Wilma's birthday.

Sunday, May 23, 1943

I held a meeting of the Council of my section at 10:30 a.m. in my office. Attending were Perlman, Willard, Kirk, Cunningham, Brown, Orlemann, English, and Thompson. One of the purposes of this meeting was to compare the merits of the wet fluoride and phosphate methods. I pointed out that we should attempt to evaluate the extraction processes ourselves before the June 1 meeting, after which it will be necessary to make a choice of processes so that all groups can concentrate and work out the details as soon as possible. The primary purpose of choosing a process is so that Site W design work can be started immediately. I also mentioned that it is planned, at Site W, to complete approximately one extraction and one decontamination cycle with heavy shielding after which further decontamination and concentration can be carried on outside of the so-called "canyon" where less shielding will be necessary. In the initial decontamination of the product before the concentration step, it is not considered absolutely necessary to concentrate the product. Finally it is assumed that one gallon can be arbitrarily chosen as the bulk to which it will be necessary to concentrate.

We then discussed the two processes in some detail. In comparing the processes it is necessary to assume that both processes are operable. The work being done by the microchemists at high ratios of plutonium to carrier is considered to be of greatest importance. Cefola is checking the behavior of plutonium hydroxide formed by metathesis in order to be sure plutonium hydroxide will dissolve in acid when the hydroxide is present as a solid phase after a metathesis reaction. This should be tested before a semiworks run is made using the metathesis step. The carrying power of lanthanum fluoride for plutonium at a ratio of 5:1 is being tested by Cefola. In the phosphate method the two major points being tested at "W" concentrations are the solubility of phosphate in 10 N HNO_3, and the precipitation of plutonium with bismuth phosphate in the extraction step. The engineers consider the acetate method unsatisfactory as an extraction process because of the large initial precipitate and the amount of material to be handled in waste disposal.

A comparison of the phosphate and fluoride methods was then made on an item-by-item basis. The Bismuth Phosphate Process was considered the better from the standpoints of ease of operation, less corrosion, adaptability to filtration, and the possibility of using alternate decontamination materials. The Wet Fluoride Process was considered the better from the standpoints of adaptability to centrifugation, disposal of waste material, effect of radiation (possibly), and concentration. There appeared to be no overall advantage to either process.

5/23/43 (cont.)

In the afternoon Wilma and Al came up to our apartment to play cards. Helen and I spent the evening working on the "Table of Isotopes."

Monday, May 24, 1943

Ermon Eastman is visiting the Met Lab from Berkeley in order to participate in the Laboratory Council Information Meeting on Chemistry.

James I. Watters, who has been an instructor at Cornell University, Ithaca, New York, started work in Section C-I today. Watters received his Ph.D. from the University of Minnesota in 1941.

I attended the Laboratory Council Information Meeting for chemistry held this morning at 10:00 a.m. in Room 209, Eckhart Hall. The meeting for chemistry is usually scheduled for the fourth Monday of the month. Present today were Boyd, Allison, C. M. Cooper, Hilberry, Whitaker, Hughes, Stone, Spedding, Johns, Mulliken, Compton, Cannon, Garrison, Neubert, Coryell, Miles, Zachariasen, Wigner, Doan, and Willard; Eastman joined us after the meeting started. Boyd was the first speaker, presenting the analytical program of his section including the work of Potratz, associated with uranium production, plutonium purification, corrosion and water analysis, and the coating program. Boyd also presented the results of his section on the adsorption process indicating that he believes that they now have a practical process. Then Garrison, speaking for Burton's section, summarized their results on the effect of radiation on a number of materials (water, plastics) important to the Project. There were also reports by Cannon on the effects of radiation on solutions involved in the Bismuth Phosphate Process and by Neubert on the effects of radiation on graphite. Spedding gave a report on the metal production work at Ames, followed by Johns of his group who told about the use of alloys for the separation of fission products in a completely metallurgical process. Eastman reported on the Berkeley program, including the studies on decontamination by the Sodium Uranyl Acetate Process, the work of Rollefson and Dodgen on the grating spectrograph to look for plutonium lines (none yet identified), the work of Brewer on the thermodynamic properties of materials important in the preparation of plutonium metal, their work on uranium as a stand-in for the production of plutonium metal, and their work on sulfide refractories for use in this production.

Willard and I then made the presentation for our Section C-I. He reviewed the main efforts under way in the Wet Fluoride and Bismuth Phosphate Processes, mentioning that aluminum (used to encapsulate the uranium) will have to be removed in the fluoride process but not in the Bismuth Phosphate Process. There is no indication that Site W concentrations of fission products or plutonium have an adverse effect on either process. Compton reminded the group of the June 1 deadline for the choice between the two processes. Willard indicated they both still look about equally good. He also covered our work on the dry fluoride and adsorption methods. I emphasized our work on the

concentration step following the operation of the extraction and decontamination cycles and on the purification step following that, indicating that precipitation of the peroxide or the iodate of plutonium from acid solution looks good. I did indicate, however, that volatility methods using PuF_6, $PuBr_4$, or $PuCl_4$ should be considered in connection perhaps with vacuum distillation. I also reported on the practice experiments that we have made with uranium as a substitute for plutonium in the metal production step and on our joint program with Boyd's section, involving Potratz and Orlemann, on the analysis for light element impurities. I reported on English's and Ghiorso's work on setting limits for gamma rays, that is, less than one gamma ray per 10,000 alpha particles in Pu^{239}. I indicated that a number of plutonium metal samples in the microgram range have been prepared by Kirk and co-workers by the reduction of PuF_4 with sodium, but we have been having trouble with oxidation when attempts are made to remelt these samples.

Finally Coryell reported on the studies by his section on the behavior of individual fission products in the Bismuth Phosphate Process. He also reported on the joint efforts between his and our sections on the improvement of the production of plutonium at the St. Louis cyclotron by including the bombardment of metal as well as UNH, indicating that it should be possible to triple yields although at lower concentrations than we have been achieving. He also discussed his data on gamma-ray energy determinations of fission products.

Because it is hoped to operate the Wet Fluoride Process with a 20% UNH solution in the extraction step, Fries tried experiments today to see whether the carrying of tracer amounts of plutonium by lanthanum fluoride would be satisfactory at such higher concentrations of UNH. He finds that carrying of plutonium is essentially 100% from such solutions and that the precipitation of uranyl fluoride at these concentrations of UNH can be prevented by the presence of about 1 N HNO_3. Higher concentrations of HNO_3 lead to the precipitation of uranyl fluoride.

A combination semiworks and laboratory run consisting of three decontamination cycles has now been completed on the Wet Fluoride Process. Thirty pounds of the neutron-bombarded UNH from Chicago III, the St. Louis bombardment that ended January 25, was put through the Wet Fluoride Process in the semiworks by W. Q. Smith's section using dichromate as the oxidizing agent at 75°C and arsenite ion as the reducing agent at room temperature in the extraction step. Peroxydisulfate plus silver ion was used as the oxidizing agent and sulfur dioxide as the reducing agent in a subsequent decontamination cycle. This was then followed by two additional oxidation-reduction cycles in the laboratory by Knox. The overall yield of Pu^{239} was about 60% and decontamination from fission product beta activity was 1.4×10^6 and from fission product gamma activity 4.3×10^5. The semiworks portion of this run corresponds to semiworks run No. 13.

5/24/43 (cont.)

Miller returned to the University of Notre Dame for experiments on the effect of radiation on the Bismuth Phosphate Process, leaving Chicago by train at 2:00 p.m.

I received a letter from Rollefson in Berkeley indicating that the source of calcium and magnesium in the plutonium samples we sent him has been found to be the paraffin on the corks used to stopper the sample vials. This clears up what has been a puzzling mystery.

Helen went to the Newcomers Luncheon Girl Reserve meeting.

Helen and I had dinner with Ermon Eastman at Gus' Good Food Restaurant (Steak House), 420 N. Dearborn, and then saw the movie "Desperados" at the Roosevelt Theater.

The U.S. troops were finally able to stop the Axis drive in central Tunisia.

Tuesday, May 25, 1943

I was home ill today.

Today Werner measured the solubility of plutonous phosphate in various HNO_3-H_3PO_4 mixtures. At a concentration of H_3PO_4 of 1.15 M he finds the following solubilities: in 0.1 M HNO_3, 5 mg per liter; in 1 M HNO_3, 50 mg per liter; in 5 M HNO_3, 5 gm per liter.

Brown, Hill, and Bohlmann have tested the feasibility of the distillation method for isolating the plutonium formed in a UF_6 pile using three pounds of UF_6 bombarded for 150,000 microampere-hours with neutrons from the St. Louis cyclotron (sample F-6-12). They find that, even with these larger amounts, the UF_6 can be quantitatively distilled from its brass container tank leaving the plutonium that was recovered as a residue in the tank after the distillation.

John Willard sent a memorandum in my name to Stearns asking that the Met Lab hire Bernard Brody, who is about to graduate from the University of Wisconsin.

In a memo to Allison, Franck relayed my special request for the services of a draftsman for several weeks to prepare a big isotope chart for Site X.

"Chemical Research--Production and Extraction of 94, Report for Month Ending May 20, 1943" (CN-653) is being issued. The following topics were covered: Adsorption methods--Beaton, Cunningham, Fries, La Chapelle, Sheft, and Turk. Experiments conducted at concentrations of plutonium greater than those which will occur in the extraction step at Site W indicate that the capacity of Hypersil silica gel is adequate. Quantitative elution of adsorbed plutonium from a 1,200 gram silica gel column is obtained with one liter of 4.5 N HNO_3; a decontamination

factor of about 25 is obtained in the column operation. Batch tests show high adsorption of fission products from uranyl nitrate solutions at the boiling point by Hypersil, with very low adsorption in subsequent treatment at room temperature.

Bismuth Phosphate Process--V. R. Cooper has studied the effect of reducing the acidity and varying the sulfate concentration in the extraction step and finds that at 0.5 N H_2SO_4 and 2 N Na_2SO_4 the precipitation of uranyl phosphate is satisfactorily inhibited. Carrying of tracer amounts of plutonium by bismuth phosphate under these conditions is 99%.

Wet Fluoride Method--James. Studies to improve decontamination with respect to strontium show that strontium could not be quantitatively removed by co-precipitation with $BaSO_4$, CaF_2, or LaF_3 in the pre-extraction step but can be carried with the LaF_3 by-product precipitate in the decontamination cycle. A modified extraction cycle is proposed which increases removal of strontium by a factor of two.

Rommel received the first defeat in his Tunisian drive. There is a report that Churchill has been suffering from pneumonia.

Wednesday, May 26, 1943

I was home ill in the morning but Stan Thompson came to our apartment for lunch to help keep me abreast of developments.

Today Cunningham finished isolating plutonium from the combined samples of about 350 micrograms and about 230 micrograms. These values are estimates from rather rough alpha counting of aliquots indicating that perhaps about 550 micrograms were present in his combined batch of plutonium when he started the procedure. He has put the plutonium through additional lanthanum fluoride oxidation-reduction cycles. As a final purification step the plutonium was precipitated as the peroxide. He made an assay today of the purity of the final solution by weighing plutonium oxide prepared from an aliquot portion; he finds a purity of the plutonium of 90% based on the value of 155,000 disintegrations per minute per microgram and on a counting efficiency on the inside alpha chamber of 47%. In his final purified solution he has a total of about 450 micrograms of Pu^{239}. This is in a total volume of 225 microliters, hence corresponding to a concentration of two micrograms of plutonium per microliter. He is distributing some of this material as follows: 5 microliters each to Patton and Werner for work on the carrying of plutonium by bismuth phosphate; 5 microliters to Cefola for work on the Wet Fluoride Method; 5 microliters to himself for a specific activity determination of the Pu^{239}; another 5 microliters to himself for work on the carrying of plutonium by bismuth phosphate; 15 microliters to Hamaker at Berkeley for general use.

I received a wire from Hamaker in Berkeley asking for information on the method of purification and degree of purity of the 25-microgram plutonium sample being sent to Berkeley by Cunningham.

Allied armies are blasting Rommel's rear guard in Kasserine Pass.

At the regular Laboratory Council Policy Meeting this morning in Room 209, Eckhart Hall, the group consisted of Allison, Compton, C. M. Cooper, Doan, Fermi, Franck, Hilberry, Spedding, Stearns, Stone, Whitaker, and Wigner. Fermi reported on neutron shielding experiments that have been made at the Argonne pile, which led Compton to ask Allison to be sure that this information gets to du Pont. He then asked Whitaker to make a report on the status of Site X. Whitaker indicated that the semiworks end of the engineering building will probably be ready for occupancy as early as late in June. Compton indicated that he has received word from the Manhattan District to go ahead with the annex to the New Chemistry Building. He emphasized that the space situation will be very difficult in the interim before the annex is completed. He said that the authorization for buildings at Argonne for the heavy water work has not yet been received. Also, the location of the proposed heavy water pile at Argonne is still under discussion.

Compton then called on Franck to make his report summarizing the work of the Chemistry Division. Franck described the work of our section, that is Section C-I, including work on the Wet Fluoride Method, the Bismuth Phosphate Process, volatility methods, adsorption methods, purification, and the search for gamma rays from plutonium. He then described the work of Burton's section, Section C-II, including the effects of radiation on a number of solids and solutions of importance to the Project. He described the work of Coryell's section, Section C-III, on fission products, including their work on the analysis of fission products in various steps of the Wet Fluoride and Bismuth Phosphate Processes. He covered the work of Boyd's section, Section C-IV, including their work on analytical chemistry in connection with the purification and metal production program and their work on the adsorption method, and finally the work of the California group under Latimer and Eastman, including their experiments on the electrolytic reduction of uranium, purification experiments on plutonium, decontamination by Sodium Uranyl Acetate Process, ultramicrochemical studies of the chemical behavior of plutonium, and experiments on the oxidation of plutonium with sodium dichromate.

Following this, Spedding made a report on the program at Ames covering metal production and their analytical control work. Spedding indicated that by June 1 his group will have made a total of 200,000 pounds of uranium metal at a cost of about $4 per pound exclusive of the cost of the uranium salt.

Compton pointed out that Site Y is asking to borrow for two weeks 200 micrograms of plutonium to be delivered soon after the first of June. They would like to have 500 micrograms if this should be possible. Compton asked Fermi to ascertain from Site Y the details of when they will want the 200 micrograms of plutonium and the form in which they would like to have it. Compton reminded the group that the meeting with Greenewalt leading to a decision between the Wet Fluoride and the Bismuth Phosphate Processes will be held on June 1 and requested that there be a special Laboratory Council Policy Meeting on Monday of next week, at which a summary of the two processes and a decision on the proposed recommendations to the du Pont Company should be made. Wigner called attention to some articles in the German scientific literature on the diffusion length of neutrons and their adsorption indicating that they were running neck and neck with us in 1941 and 1942 in their investigation of these subjects. There was also some discussion of using thorium in the pile shield in order to produce some uranium-233 as a second line of defense should the use of plutonium-239 prove not to be feasible.

Thursday, May 27, 1943

During the last couple of days Miller has tested the Bismuth Phosphate Process under the influence of electron radiation using the two-megavolt electrostatic generator at Notre Dame University. This is to simulate the radiation conditions in the actual chemical extraction plant at Site W. He arrived back in Chicago at 1:00 a.m. today and reported that he finds normal yields in going through a complete bismuth phosphate cycle on the tracer scale, indicating no adverse effect of the radiation field.

Today Franck discussed with me the request that has come from Site Y asking whether we might lend them 200 micrograms of plutonium for fourteen days in order to make some critical measurements there. They have in mind measuring ν, the number of neutrons produced per fission in Pu^{239}. After our discussion he sent a letter to Compton saying that we can lend the 200 micrograms as requested. He pointed out that we have about 400 micrograms of pure plutonium compound isolated just this week. To lend half of this for fourteen days would slow down the work going on here on metal production and purification. He pointed out that we can supply it as a pure compound of plutonium, either as a solid such as plutonium peroxide, or as plutonium nitrate or chloride in solution.

During most of this month La Chapelle has been testing a number of reducing agents as potential replacements for SO_2 in the Bismuth Phosphate Process since the latter has occasionally proved unsatisfactory in stainless steel. He has tested hydrogen peroxide, ferrous ion, phosphorous acid, hypophosphorous acid, hydroxlamine,

and hydrazine. None has been found to be entirely satisfactory. They either interfere with the carrying of plutonium by bismuth phosphate or act too slowly in the reduction process. Hydrazine gives nearly quantitative reduction in 10 minutes at 75°C in glass vessels but gives erratic results in steel vessels, suggesting incomplete carrying of plutonium by bismuth phosphate.

Brown and Hill have made experiments to test the possible use of the fluoride volatility method to remove the light element beryllium that might be present as a possible impurity in plutonium. Their experiments indicate that the plutonium might be purified from large quantities of beryllium by fluorination, thus volatilizing the higher fluoride of the plutonium away from the beryllium. On the other hand they find they can remove small quantities of beryllium by keeping plutonium in the reduced state by hydrofluorination and then with vigorous treatment with HF at 600°C volatilize the beryllium as beryllium fluoride away from the plutonium. They use beryllium-7 as tracer and find that 90% of the beryllium can be removed by hydrofluorination at 600°C for about an hour.

Later in the day, Franck called a meeting of Chemistry Division Section Chiefs to go over the list of laboratory equipment being ordered for Clinton Laboratories. It was concluded there are no unnecessary duplications of orders among the sections.

Cunningham mailed the 25-microgram sample of plutonium to Hamaker in Berkeley. It was in nitric acid solution, sealed in a glass tube.

Allison asked Fermi and Wigner to bring Franck up to date on the present situation as regards purity requirements for 49.

Doan authorized the purchase of four Coca Cola machines, one of which is to be installed in the New Chemistry Building.

Helen and Wilma Ghiorso had lunch in Jackson Park. During the evening the Baumbachs dropped in to see us.

Friday, May 28, 1943

Cunningham sent a letter over my name to Hamaker in Berkeley notifying him the 25-microgram plutonium sample has been mailed and giving the method of preparation and known impurities.

Helen had lunch at Wilma's; afterward they spent some time in Jackson Park.

Saturday, May 29, 1943

Conant is at the Met Lab today to discuss the possible military uses of fission projects. He has been assigned by Groves to head a committee to look into this.

Fries finds that arsenite ion is not too good a reducing agent at room temperature for use in the Wet Fluoride Process, but the addition of a little hydrazine together with arsenite leads to complete reduction at room temperature in about an hour.

It has been qualitatively observed by Cunningham that, if HF is added to a fairly concentrated solution of plutonium ion, no precipitate forms if no other metallic ions are present. Today Cefola, working on the ultramicroscale, added HF to a concentration of 1 M to a solution that contained 200 mg per liter of plutonium and was 0.2 M in KCl. Analysis of aliquots taken from the supernatant indicate a solubility of plutonium of some 7-10 mg per liter. This probably indicates an upper limit to the solubility because equilibrium may not have been attained.

Run No. 19 on the Bismuth Phosphate Process has been successfully completed in a joint experiment between W. Q. Smith's Chemical Engineering Section and Sutton's Separation Processes Section in the Technical Division. This run utilized 40 pounds of the neutron-bombarded UNH from Chicago IV. The first two cycles were carried out by Smith's section in the semiworks and then about 15% of this was carried through another cycle, using a four-liter scale, by Sutton's Separation Processes Section. The oxidizing agent was the dichromate-ceric +4 combination at 75°C and the reducing combination was hydrogen peroxide plus Fe^{++} at 75°C. An overall yield of Pu^{239} of 72% and a decontamination factor for beta fission activity of 2.2×10^6 was achieved. This is very encouraging and relevant information for the meeting on June 1.

Since semiworks runs Nos. 14-19 were performed on the Bismuth Phosphate Process, it hasn't been possible to run a complete additional test on the Wet Fluoride Process. However, semiworks run No. 20 has been completed for the extraction step and one complete decontamination cycle for the Wet Fluoride Process. Oxidation was performed with dichromate ion at 75°C. Precipitation from oxidized solution prior to the extraction step was made of barium sulfate and zirconium phosphate. Reduction was made with arsenite ion at room temperature and then the plutonium removed on lanthanum fluoride in the extraction step. This was then dissolved with the help of zirconyl ion, oxidation performed with dichromate ion, reduction with arsenite ion, in the decontamination cycle. An overall yield of 90% was obtained and decontamination factors of about 1,500 obtained for both beta and gamma activity, indicating successful operation of the Wet Fluoride Process.

I recommended to Stearns today a raise in salary for Edrey Smith of $10 per month in view of her competence and good work since she came with us seven months ago.

5/29/43

Helen and I went to the movies at the Tower Theater on 63rd Street and saw "Desert Victory" and "Hello Frisco, Hello."

Sunday, May 30, 1943

I held a meeting of the Council of my section at 10:30 a.m. in my office. Attending were Willard, Orlemann, Cunningham, Perlman, Thompson, English, and Brown. I pointed out that most of the trouble in both the Wet Fluoride and Bismuth Phosphate Processes is due to the presence of uranium. In the Wet Fluoride Process the reduction of plutonium in the presence of uranium and HF is difficult to obtain without getting some UF_4 formed, while in the phosphate method the carrying power of bismuth phosphate for plutonium in the presence of uranium is not entirely satisfactory. It is now evident that we have placed too much emphasis on decontamination in the extraction step. Further, since centrifugation appears to be a satisfactory method of separating precipitates, it is not necessary to have crystalline carrier precipitates that can be handled by filtration. Therefore, carriers such as uranous phosphate or thorium phosphate for extraction, making a great volume reduction possible, should be considered. Such compounds as the oxalates of U^{+4} and Th^{+4}, and the arsenates, pyrophosphates, and hypophosphates should also be investigated. These carriers would be followed by decontamination carriers such as uranyl peroxide, thorium peroxide, uranous oxalate, thorium oxalate, or the acetate method for decontamination steps. I mentioned that the use of 20% $UN \cdot 6H_2O$ in the extraction step of the Wet Fluoride Process offers possibilities, but it should not be recommended as affecting the choice of processes yet.

Willard suggested that precipitation of peroxide in the presence of HF is possible, but it may require the presence of steel and high temperatures. Thompson suggested trying a procedure in which zirconium phosphate and barium sulfate are removed from an oxidized uranium solution and reduction is carried out in the absence of HF.

I mentioned that the Bismuth Phosphate Process should give very little trouble at Site X but that difficulties with carrying power might show up at Site W. I suggested the carrying power can be tested in ordinary laboratory experiments at Site W concentrations by using thorium to simulate plutonium. Possibly this can also be done in the semiworks. Work on the concentration step for both processes should be done, and Werner is scheduled to do this with one man to help. With regard to the microchemical work, one more good microchemist is needed. I questioned whether we have emphasized sufficiently the advantages and possibilities of decontaminating with plutonium itself using no carrier. If concentration of the product were carried out first, decontamination could then be carried out on a small scale. If thorium phosphate or uranous phosphate were the precipitates obtained in the extraction step, such a concentration might be practicable. The major problem in extraction, followed by concentration and subsequent decontamination from concentrated solution, would be the heating of the solutions by radiation, and I suggested that the magnitude of such heating effects be calculated.

With regard to the two present leading extraction and decontamination methods, Willard said that major considerations in the choice of the process would be flowrates, temperatures required, analytical work, and composition of material required in the plant. These factors are difficult for us to evaluate and it will have to be done by the engineers. I suggested that we consider the use of aluminum phosphate glass for control work at Site X if the fluoride method is used. We should run laboratory tests as soon as possible and make recommendations as to whether it is satisfactory. Cunningham suggested more work on dissolving $Pu(OH)_4$ (formed by metathesis) in concentrated acid. Cefola is working on the problem.

After our meeting Brown and I met with Kurt Kraus of Tulane University who came to town to talk with us. We are interested in having him join our chemistry section.

This afternoon Helen and I went with the Ghiorsos to a baseball game in Comiskey Park to see Satchel Paige who pitched very well before an enthusiastic crowd. The four of us played cards in the evening.

Monday, May 31, 1943

I attended a Policy Meeting of the Laboratory Council this morning at 10:00 a.m. in Room 209, Eckhart Hall. Present also were Allison, Compton, Charles Cooper, Fermi, Franck, Jeffries, Miles, W. Q. Smith, Spedding, Stone, Sutton, and Whitaker. The purpose of the meeting was to prepare for the meeting tomorrow with Greenewalt and the du Pont people--the meeting at which information will be presented to enable du Pont to make a choice shortly thereafter between the Wet Fluoride and the Bismuth Phosphate Processes. Franck reiterated that he hopes work on alternate processes will, nevertheless, continue in the Chemistry Division. Cooper then called on Smith to present a summary of the information available on each of the processes. Smith said because of rapid developments in both processes one cannot be sure that either is better than the other. He believes that 80% recovery of Pu^{239} is probable in either process.

Cooper emphasized that in considering a choice it must be emphasized that our information on either process is less complete than would be considered essential in a peace-time industrial development. He pointed out that the two processes are very similar, the principal difference being that either centrifuges or filters appear to be feasible for the Bismuth Phosphate Process. He said it is the specific recommendation of the Technical Division that the design of the chemical extraction plant at Site W be made to fit both processes with only minor changes required in shifting from one to the other and that sufficient flexibility be provided so that desirable process changes can readily be incorporated. He emphasized that both processes appear at present to be nearly equally favorable and the decision will have to be based largely upon factors

of engineering operation and procurement. He recommended that the choice be left to du Pont since these are du Pont responsibilities. Cooper's motion was carried without dissent. Cooper made the point that following the choice the Technical Division will expect to concentrate its effort solely upon the process chosen. I made the point that even after the choice of the process, considerable laboratory work will have to be carried on in order to back up the semiworks experiments.

Franck emphasized again that six separation processes which look promising have been developed in the Chemistry Division, and it is only because these two separation processes were developed far enough to justify extensive work on the semiworks scale that they could be the only ones considered as eligible before the deadline of June 1. He indicated that the Chemistry Division approves the decision that the du Pont Company make the selection from one of the two methods but went on to say that the Chemistry Division is not convinced that one of these two methods is necessarily the best one and that further studies of the other processes might show that they offer greater advantages for the operation of the chemical extraction plant.

We have been holding weekly and more recently bi-weekly meetings on the adsorption process, but because of the emphasis being placed on the Wet Fluoride and the Bismuth Phosphate Processes the meeting was not held two weeks ago; today a meeting was held under the leadership of J. O. Maloney of the Technical Division who has been coordinating the semiworks aspects of adsorption processes. I attended the meeting along with Franck, Maloney, W. Q. Smith, Sutton, Beaton, Olson, Boyd, Swartout, Adamson, Schubert, Russell, Ketelle, Willard, Squires, and Collins. Beaton presented our work on the silica gel process, pointing out that the adsorption of plutonium on silica gel in column experiments is substantially lower than would be estimated from batch tests. Collins indicated that the du Pont corrosion specialists have decided that brick- and platinum-clad vessels will probably be necessary for elution from the organic resins (IR-resins). Tests by Boyd's group indicate substantial losses by adsorption of plutonium by several acid-proof bricks that they have under investigation. All in all there seem to be problems with the adsorption processes that make them appear less attractive than either the Wet Fluoride or the Bismuth Phosphate Processes.

Patton has been investigating the carrying of plutonium by bismuth phosphate at the high concentrations of plutonium that would exist in the extraction process at Site W. To my immense consternation he has been getting erratic results indicating that the process might not be reliable. In view of the impending decision tomorrow between the Wet Fluoride and the Bismuth Phosphate Processes at the meeting to be attended by Greenewalt, I asked Cunningham to drop everything and make some crucial tests which he has done during the last couple of days. He has tested the carrying of plutonium at ratios of bismuth

to plutonium of 100:1 under conditions roughly those that would exist in the extraction step at Site W. To my relief he has found plutonium carried to the extent of 90% or greater, and in fact with some variations in the conditions which could be made applicable to Site W he has found carrying as high as 99%. In order to be safe we need to repeat these tests and also check the carrying of plutonium at Site W concentrations in the subsequent steps after the separation from the initial uranyl nitrate solution containing sulfate, that is, the carrying of plutonium at Site W concentrations by bismuth phosphate in the absence of uranium and sulfate ions. We will have Miller conduct these additional experiments. I feel confident that it will be possible to find conditions under which plutonium will be carried efficiently; the experiments that Werner has performed seem to indicate this.

I received a letter addressed to Harrison Brown and me from Robert A. Lavender, Adviser on Patent Matters, OSRD, Washington. He has considered the question of priority of invention of the dry fluoride method for the separation of element 94 from uranium and its fission products. His conclusion is that while Urey was the first to conceive the method and the first to disclose it to others, Brown and I were the first to independently conceive and reduce the process to practice and therefore are considered the first inventors. He believes that the process must be claimed in an application in our names, if a valid patent is to be obtained and the interests of the Government protected.

"Technological Research--Chemical Engineering Report for Month Ending May 31, 1943" (CN-698) is being issued. Results are presented of six bismuth phosphate semiworks runs (14 through 19). No serious operating difficulties were experienced, and product yields and decontamination factors were found satisfactory. There were also two semiworks wet fluoride runs during the month--numbers 13 and 20, with satisfactory results.

Compton is issuing the Met Lab Progress Report (MUC-NH-537) for May 1943 which indicates the major effort during the month in connection with product production has been to obtain information necessary for the design of the Hanford graphite-moderated production unit. In product recovery work the primary effort has been in completing the investigations necessary to bring both the Wet Fluoride and Bismuth Phosphate Processes to a point at which a choice between them can be made by the operating company. Total expense for the month of May was $250,818.07. Total personnel on the Project on May 31 is 593.

The U^{233} production and extraction group now consists of Katzin, Stoughton, Schulze, and Steahly. They have been working the last month or so on the development of methods for separating the U^{233} that will be produced by neutron bombardment of thorium in the pile at Site X. Since the concentration of U^{233} in the neutron-bombarded thorium will not exceed about one part in 10^7, it is necessary to isolate the U^{233} in the form of its parent, the Pa^{233} intermediate. They have developed methods for precipitating Pa^{233} from the dissolved neutron-bombarded thorium on manganese dioxide and also on thorium iodate in a partial

precipitation of thorium. The decision has been made to bombard the thorium in the pile at Site X in the form of thorium carbonate because this compound dissolves quite readily in nitric acid and should contain no elements other than thorium which have high neutron-capture cross sections.

Helen spent the afternoon with Wilma. In the evening she and I took a long walk to help relieve the pressure brought on in anticipation of the meeting tomorrow.

Tuesday, June 1, 1943

Robert L. Butenhoff, Gordon L. Johnson, and Humbert Morris started work in Section C-I today. Butenhoff has just received his B.S. in chemistry from North Central College in Naperville, Illinois; Johnson comes from the University of Wisconsin, where he, also, has just graduated with a B.S. degree in chemistry; and Morris has completed his Ph.D. work at Northwestern University under P. W. Selwood.

I attended the meeting in the room next to W. Q. Smith's office in Ryerson Laboratory, called by Greenewalt to help make a final choice between the Bismuth Phosphate and Wet Fluoride Processes for the separation and decontamination of plutonium. Others present at the meeting were Acken, Apple, Balthis, Collins, Charles Cooper, Coryell, Greager, Hughes, Miles, W. Q. Smith, Squires, Sutton, Thompson, and Willard. The selection of the process to be used is considered to be of such importance that Smith's secretary, Betty Campbell, took extensive shorthand notes in order that the various factors could be thoroughly studied by those who will make the decision.

A summary of the available yield and decontamination factors obtained by Sutton's, Smith's, and my sections was presented, pointing out the limited amount of data available on the phosphate modification of the Fluoride Process because of its newness.

Willard pointed out that the results from our group represent what we believe to be obtainable on day in and day out operations in the laboratory. Such results have not been consistent, however, due to the scouting nature of the work. Frequently as soon as good results were obtained, a new angle was investigated, often with initial poor results. On this basis the estimated decontamination factor for each process is 10^7. In the Bismuth Phosphate Process a factor of 10^7 has been demonstrated in an extraction plus three decontamination cycles; in the regular Wet Fluoride Process a factor of 2×10^6 has been demonstrated in an extraction (oxidation-reduction) cycle and three decontamination cycles.

Sutton reported on the work from the development laboratory. He pointed out that the data he presented were in the form of averages of all pertinent runs, and maximum values obtained at each point. The overall values were obtained by multiplication of the respective unit values. On this basis the maximum values obtained were a beta decontamination of 1.5×10^7 and a yield of 93% for $BiPO_4$ process.

Apple and W. Q. Smith discussed yield and decontamination results obtained in semiworks operations. The values reported represent only one run on each process. It was pointed out that the yield has recently been improved on the Fluoride Process to approximately 98±% per cycle whereas yield on the Phosphate Process has not risen above 85±% per cycle.

The loss on the Phosphate Process is believed due to corrosion of 18-8 stainless steel equipment in the oxidizing solution which results in a product reduction. Such loss has not occurred in glass containers.

The concentration of plutonium in the carrier at the end of the decontamination cycles and the factors affecting this concentration were next discussed. The lowest safe ratio of carrier to product determines this in each case. The calculations are based on a 2 to 1 ratio of lanthanum to plutonium, and an 80 to 1 ratio of bismuth to plutonium. I pointed out that this lanthanum ratio may not be high enough to insure complete carrying of product but that 5 to 1 has proven safe. Thus, for the Fluoride Process, the grams of plutonium per pound of solids should be 70 instead of 160.

I then discussed the relative carrying power of bismuth phosphate and lanthanum fluoride for plutonium. Neither compound has been observed to carry oxidized plutonium. However, on the basis of present data, the acidity and composition of the uranium solution must be controlled much more critically to insure satisfactory carrying of reduced plutonium with bismuth phosphate than with lanthanum fluoride. Furthermore, at a bismuth/plutonium ratio of 100/1 the bismuth must be added in three portions to obtain quantitative carrying in the presence of uranium. I also pointed out that there are more factors which are questionable from a chemical point of view in the Phosphate Process than in the Fluoride Process. These are mainly questions which must be investigated on a micro-chemical scale. The carrying power of bismuth phosphate is not so questionable in the absence of uranium. In these steps it may be possible to reduce the ratio of bismuth to plutonium below 100 to 1, but this has not yet been adequately checked.

Continuing with other information obtained by the microchemists, I said that on standing, plutonous hydroxide becomes insoluble in dilute nitric acid, but that it is soluble in concentrated nitric acid. Its solubility in concentrated acid after standing for a day or so has not been checked. Likewise the solubility of plutonous phosphate has not been checked after standing for such a period.

W. Q. Smith, in discussing the number of cells required for the two processes, pointed out that the Fluoride Process requires more cells because the first steps of this process are performed in 10% uranyl nitrate solution while the phosphate process employs a 20% nitrate solution. There is a good possibility that the fluoride process can also be worked in a 20% nitrate solution, but the data are not conclusive.

The mechanical dependability of the two processes is judged to be the same if centrifuges are used in both.

Collins, a metallurgist, reported on the relative corrosiveness of the materials handled in the two processes. Under the operating conditions specified, corrosion rates are the same for the two processes. However, if in fabrication of the equipment the stainless steel vessels are not properly heat treated, the corrosion in the Fluoride Process might be so critical that a weld would fail in a period of one week, even

at 35°C. Collins further estimated that it was possible to have 5% of the welds improperly heat treated. The steam transfer jets are particularly vulnerable spots in the Fluoride Process, but it was pointed out that these could be lined with a noble metal or made from plastic if necessary.

Because of the low operating temperature of the Fluoride Process the operating temperatures can be exceeded more readily, resulting in excessive corrosion.

The total weight of chemicals (including caustic to neutralize the wastes) used in the Phosphate Process is considerably greater than that used in the Fluoride Process. In the Phosphate Process this might be reduced by lowering the ratio of carrier to product; in the Fluoride Process it might be reduced by increasing the concentration of the uranyl nitrate solution.

There seems to be little question of the availability of any of the rare chemicals. However, the complete picture on the availability of raw materials for the bismuth salt is not known. The quantity of bismuth indicated is large compared with the pre-war consumption of bismuth in this country. If developments show that the bismuth to plutonium ratio can be reduced, the bismuth consumption can then be reduced.

On the basis of control the two processes are essentially equal.

In discussing the chances of obtaining poorer results in the plant than obtained in the development work, C. M. Cooper reported that Burton has not found any adverse effect of radiation on either process. The effect of high concentrations of fission products on the process chemistry was discussed with the conclusion that there is no large effect on either process. Large quantities of aluminum interfere with carrying in the Fluoride Process, necessitating removal of most (80-90%) of the coating material before solution of the uranium. Aluminum has only a small, if any, effect on the phosphate process when using tracer quantities of plutonium.

I reported that I believe the weakest point in the chemistry of the Fluoride Process is the possibility of reducing uranium to the insoluble UF_4 in the reduction step. However, based on the experience in the semiworks, which should be reasonably close to plant conditions, no trouble is expected from this.

In considering the possibilities of improvement in the two processes, I said there is an excellent possibility of using 20% uranyl nitrate in the Fluoride Process. There are good prospects of improving oxidation and reduction procedures and of improving decontamination factors by making additional precipitations in both processes. The Phosphate Process might be improved mechanically if it develops that filters may be used instead of centrifuges. Also there is a possibility of decreasing the amount of bismuth carrier in the Phosphate Process.

The operational difficulties which were mentioned for the two processes are difficulties in keeping bismuth phosphate slurries in suspension, since a portion of the crystals settle very rapidly, and difficulties in washing the lanthanum hydroxide precipitate in the Fluoride Process.

In case difficulty should be encountered in decontaminating specific elements it is possible to switch from either process to another decontamination procedure, although neither process shows appreciably more flexibility on this basis. Neither process as presently operated separates neptunium from plutonium.

A general discussion followed summarizing the outstanding differences in the two processes. I stated that from a chemical point of view the Fluoride Process is definitely safer, principally because lanthanum fluoride is a better carrier for plutonium than is bismuth phosphate. Furthermore the Bismuth Phosphate Process would require considerably more work on a microchemical scale to establish safe operating conditions than would the Fluoride Process, necessitating the training of additional chemists in micro-chemical techniques.

Greenewalt, in comparing the two processes, pointed to the chemical questions as the principal weakness of the Phosphate Process and the question of obtaining properly heat treated stainless steel (to withstand the fluoride solutions) as the principal weakness of the Fluoride Process. He stated that the difficulties from a chemical point of view would result at worst in a very considerable loss in yield, while failure of the equipment would result in total loss of yield. He therefore gave a slight edge to the Bismuth Phosphate Process.

C. M. Cooper, in considering the two weaknesses, offered the suggestion that the safest choice would be the Fluoride Process since the question of obtaining satisfactorily heat treated equipment would be adequately answered from the experience obtained at Clinton Laboratories, while the question of the chemical feasibility at Hanford concentration of materials could only be answered from experience with these concentrations. Thus, the principal weakness of the Fluoride Process would be evaluated in time to change, if necessary, to the Phosphate Process long before the start-up of the Hanford plant.

Greenewalt disagreed with the adequacy of time for changing processes. He then indicated that he did not wish to choose the process to be used until the views of the operating division had been obtained. An early decision on the question was promised.

Cefola has conducted an experiment in which he metathesized PuF_4 to $Pu(OH)_4$ in order to test whether the metathesis is complete and whether the hydroxide will dissolve. He finds the metathesis to the hydroxide is complete and that even after standing overnight, the hydroxide dissolves in concentrated HNO_3.

Harrison Brown sent a letter, over my signature, to Lt. Col. Crenshaw of the Manhattan Engineer District in New York, asking that delivery of 150 pounds of UF_6 be expedited; no further runs can be made until it is received.

I attended the regular Tuesday evening meeting of the extraction group of our section. Others present were Apple, Beaton, Brown, V. R. Cooper, Cunningham, Dreher, English, Fries, Ghiorso, Hindman, Jaffey, Katzin, Orlemann, Peery, Perlman, Clifford Smith, Steahly, Thompson, Watters, and Willard. Katzin and Steahly discussed the choice of a thorium compound for bombardment in the pile at Site X. Present plans call for the use of thorium carbonate which will be compressed into cans, with about 80 pounds to be irradiated at one time. As the amount of U^{233} to be produced is small, it is planned to separate the parent Pa^{233} from the thorium in order to obtain the daughter U^{233} free from traces of ordinary uranium that may have been present in the original thorium. Experiments now in progress show that MnO_2 carries 90% of the Pa^{233} with only 1% of the thorium. The Pa^{233} concentrates in the precipitate when thorium is partially precipitated as the iodate. This method of concentration might work better at higher levels of Pa^{233} concentration than does the MnO_2 method. It is estimated that 4 or 5 mg of U^{233} may be formed in 80 pounds of thorium carbonate by 10 days' bombardment in the pile at Site X. Brown discussed current experiments he and Bohlmann are performing at Argonne to determine the feasibility of pumping UF_6. It is planned eventually to circulate UF_6 through a pipe inserted in the pile.

Helen attended a Girl Reserve meeting today. Helen Lane dropped by later to visit with her, and Wilma spent the evening with her.

Wednesday, June 2, 1943

H. P. Zvolner from the Sangamon Ordnance Plant near Springfield, Illinois, started work in Section C-I today. Zvolner received a B.S. in chemistry from the Lewis Institute of Technology in 1937 and has attended Northwestern University. Another new employee is John R. Dam, who has a B.A. in chemistry from Kalamazoo College in Kalamazoo, Michigan. He will work in Thompson's group as a Research Assistant.

Today Cunningham sent me a memorandum indicating that our commitment to lend 200 micrograms of plutonium to Site Y will leave us in short supply. He reminds me that of our recently available stock 30 micrograms have been consumed in an intensive study of extraction processes at Site W concentrations, 50 micrograms have been consigned to Berkeley for their program of investigation (including 30 micrograms for Hamaker and 20 micrograms for Rollefson); and he emphasizes that many of the planned experiments on purification and metal production will require the order of 10 micrograms per experiment. He suggests that the present St. Louis bombardment (Chicago V started on May 14) of 500 pounds of UNH be terminated upon reaching 100,000 microampere-hours around June 15; then provision should be made for ether extraction and recovery of plutonium by the lanthanum fluoride oxidation-reduction procedure from about 400 of the 500 pounds of uranyl nitrate at the earliest possible date.

6/2/43 (cont.)

Helen had lunch in Jackson Park with Wilma, discussed the Girl Reserve program with Charlotte Pearson at the 63rd St. YWCA, and later had a coke at home with Wilma.

War was pushed from the front page today by the headline "Canned Milk Rationed Now!!"

Thursday, June 3, 1943

Dreher has studied the combination of H_2O_2 and Fe^{+2} to reduce oxidized plutonium rapidly. H_2O_2 alone seems to interfere with the carrying of plutonium by bismuth phosphate. The idea of the combination is that the H_2O_2 reduces the dichromate oxidizing agent almost instantly and then a small amount of Fe^{+2} is added to reduce any remaining peroxide and to reduce the plutonium. He has tested this in a number of experiments and finds uniformly that 98% of the plutonium in tracer amounts is carried by bismuth phosphate after reduction by this method.

I conducted the Research Assistants seminar in my office this evening speaking the whole time myself. I gave an overview of the history of nuclear science beginning with the discovery of radioactivity with emphasis on the advances in the last ten years and especially on advances in connection with the Metallurgical Laboratory mission.

Helen went to a card party at Mary Curtis' while I was attending the seminar.

Again war news was relegated to back pages of the Chicago Tribune with domestic items clogging the front page--mine strike, income tax bill, and a cut in the beef ration.

Friday, June 4, 1943

I wrote to R. E. Lauterbach, transmitting a letter of recommendation in response to his request for support of his application for a Naval Reserve Commission. I expressed regret that he has decided to join the Navy rather than come with us.

Perlman sent a memo to Lane giving the text of the material on the chemistry of plutonium for inclusion in the Project Handbook.

In a memo to Stearns, Thompson recommended F. M. Dean for a position of Research Associate with us.

Today Cunningham gave Hindman and Cefola some of his plutonium stock solution which contains 2 micrograms of plutonium per microliter. Hindman received 5 microliters for work on the specific activity of plutonium while Cefola got another 5 microliters for work on the Wet Fluoride Process.

Helen went to Camp Sagawan with the Girl Reserves and is spending the night there.

Saturday, June 5, 1943

Today Cunningham put his remaining stock of plutonium through another peroxide precipitation in order to purify it further. The procedure resulted in a solution volume of 225 microliters now containing 1.34 micrograms per microliter which he plans to distribute, starting immediately in the coming weeks as follows: 4.5 microliters to Hindman for work on the specific activity of plutonium; 15 microliters to go to Rollefson at Berkeley to work on the spectroscopy of plutonium; 5 microliters to Werner to work on methods of concentrating plutonium after its extraction in the chemical separation plant; 15 microliters to Patton for oxidation experiments; another 23 microliters to Hindman to work on plutonium metal production; and 5 microliters to Cefola for further work on the Wet Fluoride Process. This will leave barely enough to lend to Site Y the 200 micrograms of plutonium that they have requested.

During the last few weeks Cefola has tested the carrying power of lanthanum fluoride for plutonium at high ratios of plutonium to lanthanum in the presence of zirconium, as a test of the operability of the Wet Fluoride Process at Site W concentrations. He finds a carrying power of more than 90% when the ratio of plutonium to lanthanum is 1:5, even in the absence of potassium ion. The carrying power when the ratio of plutonium to lanthanum is 1:1 is 75% or less either in the presence or absence of zirconium or potassium ion; in this case, the carrying power is improved by the presence of 0.1 M potassium ion in the solution. This seems to indicate that the carrying of plutonium by lanthanum fluoride in the absence of other metallic ions is caused by the formation of a double salt of plutonium and lanthanum fluoride.

Recently Knox and James have carried out oxidation-reduction cycles on the Wet Fluoride Process in order to determine factors for decontamination from fission products. They have carried out three cycles in a number of experiments and find decontamination factors from beta activity between 10^5 and 10^6. They have investigated a number of oxidizing and reducing agents and the use of by-product precipitates. In the procedure in which the by-product is precipitated from oxidized solution in the first cycle, they find the precipitation of zirconium phosphate is effective. They also find that it is effective to precipitate lanthanum fluoride repeatedly from the oxidized solution in the second cycle.

During the week the Technical Division made a phosphate-modified wet fluoride run (No. 21) in the semiworks in which by-products and product were precipitated from a 20% UNH solution. Both yield and decontamination factors decreased slightly from the previous run using 10% UNH solution.

Charles Cooper sent a memo to Compton, Allison, and Whitaker asking for a firm decision as to whether the Chemical Engineering Group should be established at Clinton by early July or whether the move should be postponed in the interest of securing additional semiworks data using the Chicago facilities. (He made reference to the choice by du Pont of the Bismuth Phosphate Process.)

6/5/43 (cont.)

Perlman attended a conference on the Project Handbook. It was noted that Willard is helping with Part A and English is helping with Part E of Chapter VIII.

Helen is still at Camp Sagawan with the Girl Reserves.

Sunday, June 6, 1943

I held a meeting of the Council of my section at 10:30 a.m. in my office with Perlman, Willard, Cunningham, Brown, Thompson, and English. I mentioned that two meetings are scheduled for discussing the Bismuth Phosphate Method since it is certain that this method will be chosen for development work. Some of the questions that we should develop in the meantime would be the amount of labor that could be placed on the problem. We should also find out whether oxidation and reduction would be done in the same tank since this might affect the oxidizing and reducing agents used. I asked that a detailed program of work on the phosphate method be drawn up by next week's meeting. Some experimental results obtained recently with hypophosphorous acid and hydrazine indicate the possibility of lower oxidation states than +4 for plutonium. Plutonium, by analogy with uranium, might have +2 and +3 oxidation states which would be more stable than the corresponding states of uranium. It would be presumed that the +3 state would be fluoride insoluble and that the +2 state might be sulfate insoluble. Both lower states would probably be phosphate soluble. If such possibilities are facts, it would explain some of the anomalous results obtained with these reducing agents. La Chapelle and James are scheduled to do more work on this problem. Brown has experiments in mind to attempt to get Pu^{+3} using a dry treatment with hydrogen. I suggested there is a good possibility that such a lower oxidation state will be found. I listed on the board some of the purification methods under consideration and asked for other suggestions. The methods listed were as follows:

1. Precipitation reactions
   a. Peroxide
   b. Iodate
   c. Oxalate
   d. Sodium plutonyl acetate
   e. Ammonium plutonyl phosphate
2. Volatility methods
   a. Use of equilibrium--$PuF_4 + F_2 \rightleftharpoons PuF_x$ (removing volatile fluorides with HF and volatile PuF_x with F_2)
   b. Fractional distillation of PuF_x
   c. Fractional distillation of $PuCl_4$
   d. Fractional distillation of $PuBr_4$
3. Vacuum distillation methods with $PuBr_4$ and $PuCl_4$

4. Solvent partition methods

   a. Diethyl ether with plutonyl nitrate
   b. Use of PuI_4 complexes and organic solvents

5. Pyroelectric volatilization (concentration) methods (use of Ga_2O_3)

Boron is difficult to remove by the peroxide method according to Clifford Smith's experiments. Some work is being done with thorium oxalate precipitated in 0.5 N acid. There do not seem to be any advantages in using the iodate method. There is a question as to how much boron is present in the H_2O_2 reagent. Arrangements have been made to obtain an analysis of it. H_2O_2 does pick up boron from glass. Nitric acid picks up sodium from glass. The oxalate of U^{+4} has been found to be fairly insoluble. Cunningham said that we know fairly well now what substances can be removed from plutonium by the peroxide method. He said that phosphate is difficult to remove from the product by the peroxide method and that it appears to be removed readily only above about 100 ppm. Chloride is readily removed, but sulfate may behave about the same as phosphate. The allowable limit on the presence of phosphate is about 700 ppm. It would be desirable to reduce the phosphate to about 0.01 of this if possible. Our present program involves starting with pure materials and carrying them through the entire process. We have no effective means for testing for F^- as yet, but it is planned to save samples and test them together when we get a method for determining F^-.

Cunningham mentioned that he thought we might reach levels of impurities which could not be removed by further reprecipitation. Willard did not believe this too likely because such effects had not shown up in adsorption work with silica gel. Furthermore, Thompson said we have not observed similar effects in our decontamination studies with fission products; Cunningham, however, mentioned results that have been obtained by Coryell on the carrying of barium by lanthanum fluoride that show that the carrying increases as the amount of barium decreases. The ether extraction method applied to purification is attractive, but it does present a problem because of the need for using a strong oxidizing agent during the extraction. It might be possible to make hydrated plutonyl nitrate first and then dissolve it in the ether. There is an actual compound formation with the ether in the case of uranyl nitrate. Volatile plutonium halide compounds should make it possible to remove oxygen provided plutonium doesn't form oxyhalides. Brown said it would certainly appear that we would have to use the dry method to remove oxygen. I thought that more consideration should be given to recrystallization with something like sulfate. Cunningham said the trouble with plutonium nitrate might be that it would be too soluble in ether. Perlman thought that dry methods would be best for purification if they must be used to remove oxygen. Cunningham suggested that, since the final product would need to be pure only with respect to certain elements, we might be able to remove the dangerous ones by adding large amounts of impurities which are not dangerous. For example, rubidium might be used to help remove sodium. I mentioned that we should try to settle on one method soon so that we can study the reagents. We could probably do some investigation with thorium as a stand-in for plutonium.

6/6/43 (cont.)

I indicated that the ether extraction, peroxide precipitation, and volatility methods are most promising at this time. Plutonium chloride is volatile at a temperature of about 250°C while the fluoride is volatile at about 350°C. English asked about the possibilities of cathode reduction for purification. I thought the reduction potential of plutonium would be too high. It is hoped that metal production will produce further purification since it is possible to remove boron from uranium metal. For example, at Westinghouse the raw material before electrolysis is much richer in boron than the final metal. Perlman suggested that we find out how difficult plutonium bromide is to handle. Cunningham said that plutonium peroxide is insoluble in 2 N HCl to the extent of about 35 mg/l in 5% H_2O_2. These tests were in the absence of bismuth. If this method is satisfactory, it would be very good for the concentration process. It was found that when such solutions are allowed to stand, more precipitate comes down after several days. Possibly the oxide is formed under such conditions or there are two forms of plutonium peroxide, one of which is difficult to dissolve. It has been found that the amount of plutonium precipitating as the peroxide is fairly independent of the amount of uranium used as a carrier.

I came home exhausted and ill after the Council meeting, the condition in which Helen found me in when she returned from Camp Sagawan this evening on our first wedding anniversary.

Monday, June 7, 1943

I am home ill today, but I am keeping in touch with the laboratory.

There was a Laboratory Council Technology Information Meeting at which W. Q. Smith and C. M. Cooper discussed the choice of separations processes. Cooper was reported as stating, "Last Tuesday all our information on the Wet Fluoride and Bismuth Phosphate Processes was presented to du Pont representatives, and choice of the Bismuth Phosphate Process was made." However, Smith, who favors the Fluoride Process, does not agree with Collins's assumption that the Fluoride Process will fail because of corrosion difficulties.

The word spread today that, as a result of our conference last Tuesday, there was a meeting in Wilmington between Greenewalt and the du Pont manufacturing people in which it was decided that the Bismuth Phosphate Process would be the process to be put into operation at Site W. The comparison between the two processes was reduced essentially to a consideration of the greater inherent corrosion hazard for the Wet Fluoride Process as against the smaller margin of carrying power for the Bismuth Phosphate Process. On this basis it was decided that design should proceed on the Bismuth Phosphate Process but that sufficient capacity should be provided to accommodate the Fluoride Process as a possible alternative in case of necessity. It was also decided that the Metallurgical Laboratory would be asked to concentrate attention on further development of the Bismuth Phosphate Process with the objective of working out a completely operable process by August 1.

6/7/43 (cont.)

We learned today that du Pont has decided that the Clinton Laboratories also will use the Bismuth Phosphate Process, the same process essentially that will be used at Site W.

A bismuth phosphate flowsheet was issued by the Technical Division for the extraction and first decontamination cycle. In the extraction step the flowsheet calls for the addition of ferrous ion reductant and lanthanum and cerium holdback carriers with the Bi^{+3} carrier added in three equal "strikes." The decontamination cycle uses ceric ion plus dichromate for oxidation and hydrogen peroxide plus ferrous ion for reduction. Again, three strikes are used in the bismuth phosphate precipitation to carry plutonium.

Stan Thompson dropped by for lunch to discuss our viewpoints for the meeting on the Bismuth Phosphate Process that will be held in my office this afternoon.

The 3:00 p.m. meeting in my office to plan the research program on the Bismuth Phosphate Process for the immediate future was attended by Miles, Coryell, Willard, Burton, Perlman, McBride, Rubinson, Thompson, W. Q. Smith, and Sutton. It was agreed that my section would carry out the following studies between now and August 1, a deadline we hope to meet with a process that is completely operable for Site W:

Microchemical: (a) Further check on solubility of $Pu(IV)PO_4$ in 1 N HNO_3 plus all other chemicals present in the actual process. (b) Determination of carrying power of bismuth phosphate at higher concentrations of plutonium, 0.025 g/l (control), 0.05 g/l, 0.10 g/l, and 0.20 g/l. (c) Determination of effect of H_3PO_4 concentration on carrying power. (d) Effect of precipitation conditions on carrying power. (e) Check separate steps, oxidation, reduction, etc. (f) Check effect of sulfate concentration and sulfate/acid ratio in extraction step on carrying power.

Process Scouting: (a) Oxidation procedure--development of two or three possibilities. (Elimination of complexing of phosphate approach.) (b) Development of alternate reduction procedure. (c) Reduction procedure for small amount of oxidized plutonium following dissolving cycle. (d) Reprecipitation and milking. (e) Yield and decontamination at Site W concentration of fission products.

Concentration Procedures: We will investigate methods of concentrating the plutonium after the extraction and decontamination steps.

It was also agreed that Sutton's section will work on process development, Smith's section on semiworks studies, Coryell's on specific decontamination of fission products, and Burton's on the effects of radiation. The status will be reviewed on July 1 to decide whether the process is in sufficiently good shape at that time to start reproducibility runs in the laboratory and semiworks.

Wilma Ghiorso stopped at our apartment to see Helen.

Tuesday, June 8, 1943

I am still home ill today. Iz Perlman dropped by for lunch, and we went over laboratory business.

Beaton was visited by W. H. Gabeler of the Davison Chemical Corporation in Baltimore to discuss, on an unclassified basis, the properties and manufacture of silica gel, which is being studied as a possible adsorbent for plutonium. Davison is one of the largest manufacturers of silica gel in the country, and Gabeler was recommended by the du Pont Company as an expert in this field.

Perlman sent a memo to Dr. Stone indicating additional possible hazards to personnel in the experimental work to be carried out by our section at Clinton Laboratories. The major new hazards anticipated are radiation from neptunium-239 during the isolation of the plutonium and from protactinium-233 during its separation from pile-irradiated thorium and the possibility of burns from fluorine gas and fluorides.

We had the Perlmans, Merlin Petersons, and Katzins over to dinner.

Wednesday, June 9, 1943

Stan Thompson came by for lunch at our apartment to bring me up-to-date on laboratory matters; however, at 2:30 p.m. I felt well enough to go to the laboratory for a short time.

Willard asked Stearns to make an offer of employment to Bernard Brody of the University of Wisconsin.

Milton Burton, Chief of the Radiation Chemistry Section, wrote to Sheline in Berkeley suggesting that he do microradiation experiments on the Bismuth Phosphate Process rather than the Sodium Uranyl Acetate Process because, during the last two days, it has been decided to standardize on the phosphate process.

Compton sent a letter to Segrè in Berkeley saying that the Manhattan District patent office recommends that signatures as inventors in Case S-1/61 (the basic plutonium production patents) be made by him (Segrè), Kennedy, and me. He further indicated to Segrè, who has been concerned about the propriety of this procedure, that both Fermi and Robert Christy are in complete agreement with this proposal.

We have been requested to provide Spedding with 20 pounds of thorium nitrate for experiments in the chemistry of thorium at Ames.

Wilma Ghiorso spent the afternoon with Helen.

Again the soft coal contract negotiations take the top headline. The talks are not going well.

There was a Laboratory Council Policy Meeting at 10:00 a.m. in Room 209, Eckhart Hall. Present were Allison, Compton, C. M. Cooper, Doan, Fermi, Franck, Hilberry, Smyth, Spedding, Stone, Whitaker, and Wigner. Cooper presented the distribution of effort of the six sections of his division. In answer to Compton's questions it was indicated that by July 1 the uranium metal in the Argonne pile will have accumulated 300 kilowatt hours, corresponding to about 1/10 the plutonium concentration of the St. Louis neutron-bombarded uranium nitrate. Compton reiterated that we are responsible for supplying plutonium to Site Y and that therefore we must carry out whatever studies are required toward that end. Compton mentioned that this project is one of the few war researches increasing in importance at the present time and the Metallurgical Project is still expected to produce the bulk of fissionable material for war use before other processes can do so. Our instructions are solely to produce plutonium; there is no effort at present on power production.

Thursday, June 10, 1943

I am again staying at home.

Magel is in Milwaukee today to contact the Globe Union Company about fabricating some special refractories.

Allison wrote to Greenewalt today summarizing for him the results of the conference held in my office on Monday. He informed Greenewalt that weekly meetings on the Bismuth Phosphate Process will be held and that he will attend and chair these meetings himself. The meetings will be attended in addition by Burton, C. M. Cooper, Coryell, Franck, Miles, W. Q. Smith, Squires, Sutton, and me. The purpose of the meetings will be to keep the work going and on schedule in order to make the August 1 deadline of having a completely operable process ready for Site W.

Franck informed Eastman in Berkeley of the decision to use the Bismuth Phosphate Process and stated that consequently efforts are being made to complete the research on the process in a hurry. He asked Eastman if he would be willing to have his colleagues coordinate their work with ours in Chicago and, in particular, to do specific fission product decontamination studies on the Bismuth Phosphate Process. He also asked if Sheline could do separations studies using micromethods under radiation conditions duplicating those expected at Site W; he suggested Sheline attend the next chemical conference on Monday, June 28.

La Chapelle, Dreher, and Thompson have studied a number of reducing agents for plutonium in the Bismuth Phosphate Method, including hydrogen peroxide, ferrous ion, phosphorous acid, hypophosphorous acid, hydroxlamine, and hydrazine. No single one of these is found to be completely satisfactory.

I received a letter from Hamaker in Berkeley saying that the ampoule in which Cunningham sent them the sample of plutonium has a hole in the bottom and that he has been unable to find more than 5-6 micrograms of

the 30 micrograms he was to receive. He is of the opinion that there has been no leakage of material in transit and wonders if for some reason the full amount was not sent.

I recommended to Stearns raises to $200 per month for the following: James, Knox, Jarrett, La Chapelle, Scott, Crawford, Dallas, Nathalie Baumbach, Schulze, and Sheft. I also recommended raising Koshland from $200 to $220, Kohman from $250 to $275, and Covey from $160 to $180.

Helen went to see Goddard's House; later Wilma came up to have coffee with her.

The final organized Japanese resistance on Attu Island has collapsed with a mass suicide by the Japanese survivors left on the island.

Friday, June 11, 1943

I am still at home but keeping in touch with the laboratory.

The du Pont Company forwarded the bismuth phosphate flowsheet to its Design Division today, which indicates they mean business; design work must begin immediately in order to meet the schedule.

Maloney of the Technical Division sent a memo on ether extraction to C. M. Cooper about his visit to M. R. Fenske at Pennsylvania State College on June 9. Maloney is impressed with Fenske's staff and laboratory facilities for doing solvent extraction work and is of the opinion that we should consider his facilities before embarking on our own program.

My office sent a memorandum to Doan asking if he knows of any beryllium powder that can be made available to us. It is needed by Kohman for his work in connection with the preparation of radium-beryllium neutron sources by an outside fabricator.

Wilma came up from their apartment below ours to see Helen, enthusiastic about a house she has found and into which she and Al will probably move. They are tired of apartment living.

The Soviets have turned back a Nazi lunge at Rostov.

Saturday, June 12, 1943

I am still weak and at home but keeping in touch with the laboratory.

Marie Garre began working as a secretary in Section C-I.

Franck has been asked by Compton to go to Berkeley to confer with Latimer and Oppenheimer with regard to the distribution of the research effort of Latimer's group between Metallurgical Laboratory and Site Y problems.

6/12/43 (cont.)

During this week the Separation Processes Section of the Technical Division directed its main attention to oxidation procedures, effect of Site W concentrations of inactive fission products, waste disposal, filtration, and recrystallization connected with the Bismuth Phosphate Process and terminated its work on the sodium uranyl acetate, adsorption, and ether extraction processes. The semiworks made run No. 24 and began run No. 25 to obtain centrifuge data on the Bismuth Phosphate Process.

In a meeting in St. Louis today it was agreed that Professor Hughes will be released from Washington University to join Oppenheimer's group at Site Y; and his place as the official investigator on the cyclotron project will be taken by Professor Rodewald, also of Washington University. Fulbright will continue to be in technical charge of the cyclotron. Present at the meeting were A. L. Hughes, Chancellor George R. Throop, and C. W. Rodewald, with H. T. Wensel representing the Manhattan District.

General Doolittle declared today that the air conquest of the island of Pantelleria (in the Mediterranean between Tunisia and Sicily) "is definitely a landmark in the history of military aviation."

Sunday, June 13, 1943

There was a meeting of the Council of my section in my office at 10:30 a.m., attended by Perlman, Kirk, Cunningham, Orlemann, Willard, English, Brown, and Thompson. (I was at home ill.) Kirk discussed the present status of the work being done on plutonium metal production, the most difficult problem at present being to get a compact form of plutonium metal for density measurements. One method being considered is to heat the sample in a covered magnesium crucible and melt the magnesium. Beryllia is better than thoria as crucible material. Producing the metal by treating UI_4 and PuO_2 with barium was also mentioned. In his discussion of plutonium purification problems Cunningham presented data which give the results of studies with thorium as a stand-in using oxalate and peroxide precipitations. Iodate precipitation of thorium has been tried and found to be not so effective as hydrogen peroxide. Orlemann discussed the analytical work in the purification program. The spectrographic methods are complicated by the presence of uranium. The colorimetric method for aluminum is nearly sensitive enough but a great many ions interfere. Work is being continued on the determination of carbon by absorbing the carbon dioxide produced by combustion in barium hydroxide and measuring the conductivity change. With regard to electrolyte methods for reduction to plutonium metal, Kirk said the operations would have to be carried out in a molten bath in which case the purity problem is difficult and it is impossible to get a solid chunk. The best hope is a reduction method in which a pure reducing agent is used in a good refractory. Orlemann suggested asking the King Company to prepare about 50 pounds of pure barium for use as a reducing agent.

Monday, June 14, 1943

I still feel too weak to go to work but as usual am keeping in touch.

Jonathan S. Dixon started work in Section C-I. He comes from Haverford College where he has recently graduated with an M.S. degree in chemistry. Dwight C. Lincoln, who recently received an M.S. degree from the Illinois Institute of Technology, also arrived today. These men will work with Thompson.

A wire was sent in my name to Hamaker in Berkeley to the effect that we are at a loss to explain the mishap which occurred to the plutonium sample we sent him and that we are sending an additional twenty micrograms. We asked that he return the packing for inspection. In the meantime Cunningham is sending him 15 microliters of his stock solution (1.34 micrograms per microliter).

The neutron bombardment (Chicago V) of 500 pounds of UNH at St. Louis was terminated today, in line with Cunningham's request for more plutonium at the earliest possible date and in view of the impending consumption of our supply and the planned loan of 200 micrograms to Site Y.

At the Chemistry Division Seminar this evening Dave Ritter in Sutton's section suggested to Thompson that BiO, bismuth suboxide, prepared by ignition of basic bismuth oxalate in the absence of air, should be a good reducing agent for plutonium. It was decided they would cooperate in evaluating the material.

Two large formations of American heavy bombers blasted Nazi U-boat bases at Bremen and Kiel today in the greatest air battles yet waged by the 8th U.S. Air Force.

There was a Laboratory Council Policy Meeting at 12:30 p.m., attended by Allison, Compton, C. M. Cooper, Fermi, Hilberry, Miles, Spedding, Stone, and Wigner. The purpose of the meeting was to select someone to act as the Met Lab representative at the du Pont organization. Compton proposed sending Herbert L. Anderson, who would report to Chicago by telephone on a daily basis. It was agreed that Compton should take this up with du Pont.

Cooper of the Technical Division reported on the trip W. Q. Smith and he took to Clinton Labs on June 10 and 11 to appraise the possibilities of an early transfer of chemical engineers. His conclusion is that the semiworks group should remain in Chicago until the Bismuth Phosphate consistency runs are completed on August 15.

Tuesday, June 15, 1943

I am still at home today.

Bernard B. Brody, who recently received his B.S. degree from the University of Wisconsin, started work as a Research Assistant with Orlemann today.

My office sent a memo to R. L. Doan telling him that 50 grams of beryllium powder have been received by Graves from the General Electric Company and probably will be suitable for making neutron sources. I therefore suggested he place the order for the radium sources with Boris Pregel of the Canadian Radium and Uranium Corporation in New York City. Kohman will go to New York to supervise the fabrication of the radium-beryllium neutron sources.

Another Italian outpost in the Mediterranean fell today--the island of Ampedusa.

There was a Laboratory Council Policy Meeting at 9:30 a.m. in Room 209, Eckhart Hall, attended by Allison, Compton, C. M. Cooper, Doan, Fermi, Greenewalt, Hilberry, Miles, Spedding, Stearns, and Stone. Cooper reported that semiworks facilities at Clinton Labs cannot be ready until early August. Compton told of the change in responsible personnel at St. Louis. Franck is now visiting Berkeley to set up a division of effort between work for the Met Lab and Site Y satisfactory to the people at both places. Compton also reported his suggestion to Conant that within six months we should be giving emphasis to other aspects such as power production. Conant agreed in principle but questioned the date. In a discussion with Compton and Allison, Fermi indicated that probabilities are that U^{233} will be less useful than Pu^{239}. Five hundred micrograms of U^{233} would be enough for neutron multiplication tests. There were discussions of the program of the Health Division and the heavy water pile program.

Wednesday, June 16, 1943

I am still at home today; I have a low-grade fever--probably the cause of my weakness.

James C. Kroner and Norman H. Nachtrieb started work with us today. Nachtrieb comes from Pittsburgh Plate Glass in Barberton, Ohio and has a Ph.D. from the University of Chicago. Kroner, whose home is Mahomet, Illinois, has recently received a B.S. in chemistry from the University of Illinois.

My office received a letter from Hamaker with further information about the leaking ampoule of plutonium solution that we sent him. He has found activity on some of the packing equivalent to at least 6 micrograms of plutonium, thus indicating the ampoule did leak in transit. The packing is being returned to us.

The material from the fifth St. Louis bombardment (Chicago V) arrived at the laboratory today.

Baumbach, Rosenfels, and Kirk, during the last month or so, have been performing a number of experiments on the reduction of uranium under varying conditions using uranium as a stand-in for plutonium. The aim is to produce metallic plutonium in the form of discrete pieces of solid

material rather than as a fine powder. This is needed for the determination of the density of the metallic plutonium as well as for the study of other chemical, physical, and metallographic properties. They have constructed a number of microfurnaces and microcrucibles out of refractory material, making a lot of progress in the design and building of these critical items that will be needed in the metal production program. They have fabricated crucibles and covers out of thorium dioxide and beryllium oxide suitable for conducting reduction reactions on the microgram scale with reductants such as magnesium, calcium, and barium. After reduction the excess reductant metal is distilled off, leaving solid chunks of uranium metal. The experiments are conducted in very high vacuum. They have had best success with magnesium in reducing uranium tetrafluoride and have been able to obtain spheres of uranium metal weighing about 100 micrograms. Attempts to reduce UO_2 with magnesium metal have not been successful.

Having developed the method, today they made an attempt to reduce PuF_4 with magnesium using this system. Two beryllium oxide crucibles were charged, each with about one microgram of PuF_4, pieces of magnesium turnings were placed on top of the PuF_4, covers placed on the crucibles, the system evacuated, and the crucibles raised to high temperatures, approximately 1,500°C. This led to the apparent production of plutonium metal in the form of a black powder that was not melted. In another similar experiment a total of about 5-8 micrograms of plutonium in the form of PuF_4 was placed in the bottom of a beryllium oxide crucible, magnesium put on top, cover placed on the crucible; and this was raised to a temperature of about 1,500°C under high vacuum. Upon examination of the contents of the crucible after the experiment, it seemed possible that plutonium metal had been formed because the product melted into bright, white, silvery looking spheres. One of these spheres has been given to Hindman for determination of its density by measuring its diameter and then weighing it. Other spheres are being mounted by Rosenfels for metallographic examination.

Today news reports in Europe are all favorable for the Allies--the RAF ripped the Ruhr in their fourth successive raid; the Yanks pasted Rabaul (Japanese stronghold in the Pacific); the Soviets repulsed new Nazi attacks in the Orel area.

Thursday, June 17, 1943

I am at home ill again but, as usual, keeping in touch.

Hindman has started to work on ultra-microchemical problems and at Cunningham's suggestion has made an attempt to determine the specific alpha activity of Pu^{239}. He attempted to convert the oxide to the fluoride and measure the weight change, but this has not been successful because of the attack on the platinum weighing pans by hydrogen fluoride. Today he attempted to make a determination of the density of the microgram piece of what might be plutonium metal furnished by Kirk yesterday. He finds that it weighs almost exactly one microgram, and his measurements under the microscope give a diameter of 62 microns. If the particle is spherical,

this corresponds to a density of about 8 gm per cc; so low a value indicates that the metal has voids in it and probably is contaminated with oxide or fluoride and that it may not actually be spherical in shape.

On the front page today is a full column article about the marriage of Charlie Chaplin (54) to Oona O'Neill (18), daughter of the famed playwright Eugene O'Neill.

Friday, June 18, 1943

I still feel enervated and again stayed home.

The regular weekly meeting chaired by Allison, of the principal chemists in the Chemistry and Technical Division involved in the program to study the important remaining uncertainties in the Bismuth Phosphate Process, was held today. Present were Allison, Burton, C. M. Cooper, Coryell, Miles, W. Q. Smith, Squires, Sutton, Perlman, and Thompson (representing me).

Today Baumbach reduced UO_2 to uranium metal with barium using the same techniques as he has been using unsuccessfully with magnesium.

Rosenfels mounted in lucite the "metallic" looking plutonium globule that was used by Hindman yesterday to try to measure its density. He ground and polished it and found that it contains several fissures caused by incomplete merging of smaller molten globules. He also mounted two smaller pieces of plutonium metal in lucite, and these were polished and showed little of this fissure effect; one had a single fissure and the other had none. Examination of the plutonium "metal" under these conditions shows that it is white and reflective, equalling or surpassing platinum in these respects; it is also suggestive of platinum in its hardness (i.e., softness). Although no quantitative estimate of hardness could be made, the use of a steel needle showed that the metal could be readily distorted and deformed. In fact, in removing it from the bottom of the crucible, it smeared in the fashion of a typical soft metal. Original and polished surfaces of the metal do not show any apparent discoloration or loss of brightness after standing exposed to air. Treatment with 1 N HCl produces effervescence very similar to that obtained with uranium metal.

Experiments by Knox show that as much as 60% of tracer amounts of plutonium may be oxidized in the 60% HNO_3 that is used to dissolve the uranium metal from the pile preparatory to the extraction step in the Bismuth Phosphate Process--the step after which the plutonium must be in reduced form in order to be carried by the bismuth phosphate.

Vance Cooper completed today experiments on the use of Ce^{+4} as an alternative to dichromate ion as an oxidizing agent in the Bismuth Phosphate Process. He finds that Ce^{+4} is satisfactory in the presence of 25-12 stainless steel under the following conditions: 0.02 M Ce^{+4}, 5 N HNO_3, at 75°C for one hour.

The U.S. downed 77 Japanese planes in a raid by the Japanese on Guadalcanal Island.

Saturday, June 19, 1943

I am still at home today.

The sixth St. Louis cyclotron neutron bombardment of uranium (Chicago VI) started today.

During the last month Clifford Smith and Cunningham have performed purification experiments with thorium and uranyl uranium as stand-ins for plutonium. These have consisted of adding a number of the impurities that must be removed in the purification process and then adding reagents to precipitate insoluble compounds such as peroxide, oxalate, iodate, sometimes precipitating one compound and then dissolving it and then precipitating another. They find that they can remove a number of the light element impurities by this technique; but it will, of course, have to be tested with plutonium itself. The use of hydrogen peroxide to precipitate the insoluble peroxide seems to give the best result.

Today Baumbach apparently reduced plutonium dioxide to plutonium metal with magnesium. The oxide was placed as usual in a beryllia crucible, magnesium placed on top; and in a high vacuum the covered crucible was heated to about 1,500°C. Examination of the crucible showed that each flake of the oxide had formed a bright metal sphere resembling platinum in appearance. Thus, contrary to the inability of magnesium to reduce uranium oxide, magnesium apparently can reduce the oxide of plutonium to the metal.

James and Dixon completed a series of experiments today aimed at testing the suitability of bismuthate ion as an alternative to dichromate ion as an oxidizing agent in the Bismuth Phosphate Process. They find that bismuthate can be made to work satisfactorily under the following conditions: 0.01 M $NaBiO_3$, 5 M HNO_3, when the solution is heated at 50°C for one hour. The oxidation is slower in freshly pickled steel containers than in steel that has been pretreated with a solution of the oxidizing agent.

The U^{233} production and extraction group, consisting of Katzin, Stoughton, Schulze, and Steahly, has found that Pa^{233} can be volatilized by hydrofluorination of neutron-bombarded thorium.

Sunday, June 20, 1943

There was a meeting of the Council of my section at 10:30 a.m. in my office, attended by Perlman, Orlemann, Kirk, Cunningham, Willard, Brown, Thompson, and English. (I was again not present.) Willard summarized the present status of extraction work for the Bismuth Phosphate Process. Sodium bismuthate appears promising as an oxidizing agent,

as does ceric ion. Considerable work is being done on metathesis in which bismuth phosphate is treated with KOH to produce $Bi(OH)_3$ which in turn can be dissolved in a very small volume of nitric acid. With regard to the work of La Chapelle on lower reduced states of plutonium, nothing has been definitely established. The falling off of carrying of tracer amounts of plutonium reduced by hypophosphite as shown by LaF_3 precipitation now appears to be an artifact due to thick precipitates formed by some reaction of fluoride with hypophosphite. Further experiments are planned using a Jones reductor and also dissolving bombarded uranium metal in hydrochloric acid. Cefola is investigating what ratio of lanthanum to plutonium is required for complete precipitation of plutonium by the fluoride method. Knox's experiments on the percent of plutonium oxidized when uranium metal is dissolved in 60% nitric acid have shown approximately 60% of the plutonium to be in the oxidized state. Experiments are planned to confirm this by testing carrying by sodium uranyl acetate; there is also the possibility that the plutonium is present in a state lower than the +4 state. There are indications that the carrying of plutonium is affected markedly by the rate of bismuth phosphate precipitation, time of digestion, and agitation. Perlman mentioned the Manhattan District is interested in obtaining 10-15 curies of polonium which can be manufactured by the irradiation of bismuth in the pile. Kirk summarized the status of metal production. Magnesium will reduce PuO_2 while it does not reduce UO_2. One chunk of plutonium that has been prepared weighs about 1 microgram; it appears to be a soft metal, similar to platinum. At least 5 micrograms are needed for density measurements.

Helen insisted, over my protests, upon calling Dr. Robert Stone about my illness. Dr. Leon Jacobson came to our apartment to see me and then put me in the University of Chicago Billings Hospital. The doctors have diagnosed my illness as an upper respiratory infection, with some pulmonary complication associated with fever and fatigue. Helen went with me to the hospital. When she finally returned home, she felt lonely and visited the Ghiorsos downstairs, playing cards with them during the late evening.

Monday, June 21, 1943

I am in Billings Hospital, but not too sick to keep in touch with the laboratory, which I intend to do during my stay here. Helen visited me in the afternoon.

Sydney R. Gaarder, Frank A. Getz, and Joseph Halperin started work in Section C-I today; all three come from the University of Chicago where they have just received B.S. degrees in chemistry.

We have asked Miller to learn ultramicrotechniques in order to make tests of the carrying power of bismuth phosphate at Site W concentrations of plutonium. He has made preparations for the last few weeks and performed his first experiment today at a ratio of plutonium to bismuth of 1:100. Working with 20% UNH and the conditions suggested for the extraction step, he finds erratic carrying--his highest result is a little more than 90%.

A status report sent today to Greenewalt by Miles, his Met Lab representative, included the following information on the Bismuth Phosphate Process: "Tests of Al and Hg at W concentrations indicate no effect. Three new oxidizing agents have been uncovered. The most attractive, sodium bismuthate, has given almost complete oxidation in one oxidation cycle with active material in the laboratory, yield 98%. Will be run in semiworks. Microchemical work checking extraction and will check oxidation as soon as established. Work started on concentration methods, will precipitate Pu with H_2O_2 in presence of high concentration of La."

Today Baumbach reduced with magnesium about 8 micrograms of PuF_4, distributed in two beryllium oxide crucibles. In one of the crucibles containing a relatively large thin flake of PuF_4, about a dozen small spheres of bright plutonium metal could be seen after the reduction. The other crucible was broken in detaching the cover so the whole contents, including the magnesium, were dumped into another crucible and remelted; the pieces of plutonium formed were mostly black cinders with a few bright spheres at the edges.

Knox repeated today the experiment that he performed on Friday by testing the oxidation of plutonium by 60% HNO_3 in the absence of uranium. He finds in this case that 98% of the plutonium remains in the reduced state.

Helen had dinner with the Ghiorsos tonight.

Moscow announced the destruction of 276 Nazi planes this week in raids in the Orel area.

Tuesday, June 22, 1943

Perlman left by train last night at 11:55 for Knoxville to inspect the Clinton Laboratories.

Floyd M. Dean started work in Section C-I as a Junior Chemist coming from Armour and Company, Chicago. Dean graduated from the University of Washington in 1925.

Franck received from Doan a copy of comments made by R. D. Smith, a Safety Inspector for the du Pont Company, concerning conditions he found in the New Chemistry Building. Among his criticisms are: excessive congestion and a lack of proper housekeeping, aisles blocked with equipmetn, extension cords across aisles and in contact with steam radiators, compressed gas cylinders not secured in place.

Franck reported to Compton that on the basis of his trip to California he has concluded that the work which Oppenheimer wants done by the California group (separation of plutonium from uranium metal bombarded in the cyclotron and the separation of polonium from bismuth) will not interfere with the progress of the work that is to be done for us. According to Latimer and Eastman, Oppenheimer's needs will require only a small part of the manpower available.

Hilberry sent a memo to the Area Engineer urging that the annex to the New Chemistry Building occupy all the remaining area on Ingleside Avenue in order to provide sufficient working space for the staff.

The crucible in which the plutonium remelt was attempted yesterday was loaded again with magnesium metal and reheated to cause melting of the cinders of plutonium; the plutonium remelted into shiny spheres. It appears that the present method of making plutonium preparations work well in principle, but it has certain mechanical disadvantages--(1) the crucibles are difficult to load and must have the PuF_4 (produced by Brown and Hill by the hydrofluorination of plutonium dioxide) or the PuO_2 in one location so that large spheres are formed; (2) the prepared plutonium spheres are difficult to remove from the bottom of the crucibles so that they can be used for measurements; (3) the cover must be forced on tightly (which may break the crucible) so that it will not be blown off by the vaporization of the magnesium.

Helen visited Wilma and Al Ghiorso in the evening, playing cards until late.

War headlines have been removed from the front page today, this time by reports of race riots in Detroit; U.S. troops were moved in.

Wednesday, June 23, 1943

Helen spent the afternoon with me at Billings Hospital.

Cunningham has completed the isolation of 93^{237} from the neptunium fraction that he set aside on May 18. He has obtained a disappointing yield of only about a microgram of 93^{237}, and the reason for the small yield is not clear. He is setting the sample aside for possible combination with larger samples later on.

Continuing his experiments along the lines of those he performed on Friday and Monday, today Knox tested the oxidation of plutonium with 1 N HNO_3 using tracer amounts of plutonium in the absence of uranium for four hours at boiling temperatures. He finds that 99% of the plutonium remains in the reduced state. Thus it is clear that further work is necessary to obtain an understanding of the oxidation that occurs in the case where uranium is present when uranium metal is dissolved in 60% HNO_3 in the dissolving step. In all of these tests the criterion for plutonium being in the oxidized state is its non-carrying by lanthanum fluoride.

Spof English sent a letter, over my name, to Joe Hamilton in Berkeley telling him that the circuit Hamilton sent us has been received and the wiring defect corrected; it will be returned soon. We are also sending through V. C. Wilson, of the Project electronics group, a scale-of-64 Columbia-type scaler for him to keep if he wishes.

Ed Orlemann sent a letter, over my name, to the King Laboratories in Syracuse, New York, regarding their indicated willingness to produce barium of high purity on a cost-plus basis. We indicated that a decision on our purity requirements and quantity needed would be made within the next few weeks.

Helen again had dinner at the Ghiorsos' and then went to a show with them.

A four-month truce has been declared in the coal mine strike.

There was a Laboratory Council Policy Meeting at 10:00 a.m. in Room 209, Eckhart Hall. Present were Allison, Cantril, Compton, C. M. Cooper, Doan, Fermi, Franck, Hilberry, Jeffries, Miles, Nichols, A. V. Peterson, Smyth, Stearns, and Whitaker. Fermi reviewed the past month's work and the future plans for the physics program. Compton reported on his visit to Clinton Laboratories last week. The semiworks end of the chemistry building there is expected to be ready by August 1. Pile operation may start early in September. Compton also announced that the St. Louis program is now under F. W. Bubb, who takes the place of A. L. Hughes as representative of Washington University. In regard to Site Y, Charles Thomas, Director of Research at Monsanto, is being contacted in the hope he will be Chemical Coordinator at Site Y. He will be in Chicago Friday and Saturday. Compton stated that such a coordinator could also help us in the planning of our own work on purification. Compton also announced that Richard Tolman, Vice Chairman of the National Defense Research Committee, has agreed to take on close supervision of all uranium projects with especial reference to Site Y. Compton further observed that the processes for production of U^{235} are going ahead in such a way that limited amounts will probably be available before Pu^{239} will be, but it is still believed that kg/day quantities will lag behind Pu^{239} production.

Thursday, June 24, 1943

I am getting a lot of rest here in Billings Hospital, but I still have a little fever. Helen spent the afternoon with me.

Dreher has completed a series of experiments in which thorium, with UX_1 as tracer, has been used to simulate plutonium in the extraction step of the Bismuth Phosphate Process, i.e., precipitation of thorium phosphate from 20% UNH, with flowsheet concentrations of H_3PO_4, H_2SO_4, HNO_3. This has the advantage, if thorium is indeed a good stand-in for plutonium, of making it possible to carry out experiments on the ordinary laboratory scale using an abundant relatively inactive material in lieu of experiments on the ultramicroscale with scarce and highly radioactive plutonium. With the thorium at the concentration at which plutonium will exist in the Site W process, he finds carrying no better than about 90%; the method has the disadvantage that we can't be sure that thorium acts as a true stand-in for plutonium.

Today Hill fluorinated about 3 micrograms of plutonium metal furnished to him by Kirk. The metal was placed in a copper reaction vessel and fluorinated for three-quarters of an hour at 480°C, resulting in the volatilization of more than 99.7% of the plutonium. The collection of the volatile plutonium fluoride was very much the same as that found

in previous work with smaller quantities. A deposit of cupric fluoride around the spot in which the metal was placed indicates that, as in the case of uranium metal, plutonium metal fluorinates very vigorously.

Allison authorized Stearns to appoint A. C. G. Mitchell of Indiana University as a consultant to the Metallurgical Project to permit him to be used on small projects such as the determination of the energy of gamma rays by the recoil-coincidence method.

"Chemical Research--Production and Extraction of Plutonium, Report for Month Ending June 21, 1943" (CN-728) is being issued. The following items are covered.

Wet Fluoride Process--Clifford, Fries, James, Knox, Koshland, Shepard, and Thompson. Oxidation studies show the following: (1) Oxidation of plutonium by 0.3 N $K_2Cr_2O_7$ in 3 N HNO_3 is complete in 30 minutes at 98°C but incomplete in three hours at 80°C. (2) Co^{+3} ions oxidizes plutonium rapidly, but its usefulness is limited by its instability and the difficulty of preparation. (3) Co^{+2} ion at a concentration of 0.01 M or greater catalyzes the oxidation of plutonium by $K_2Cr_2O_7$. (4) Nearly quantitative oxidation of plutonium may be obtained in 1-1.5 N HNO_3 by 0.1 N $K_2Cr_2O_7$ at 75°C in one hour in the presence or absence of steel. Carrying power studies show that the carrying of plutonium by preformed LaF_3 is reduced by the presence of zirconyl ion in solution. Reduction studies indicate that when dichromate is used as the oxidizing agent: (a) satisfactory reduction of plutonium with arsenite is obtained in three hours at room temperature; (b) hydrazine shows promise of being an effective and rapid reducing agent for plutonium but has not been extensively studied as yet; (c) hydrogen peroxide (to reduce the dichromate) followed by Fe^{+2} (to reduce the plutonium) produces 97% reduction of the plutonium in two hours at room temperature. (5) When Ag^{+2} - $S_2O_8^{-2}$ is used as the oxidizing agent, Fe^{+2} may be used as reducing agent. Ninety-seven percent recovery of plutonium has been obtained in such a cycle. Knox and James studied reduction and carrying power in the extraction cycle and find that reduction of dichromate and plutonium by sodium arsenite in the presence of steel is complete in less than one hour. Decontamination studies were carried out by Knox and James which tested the following modifications of the Wet Fluoride Process: (1) precipitation of zirconium phosphate from the first oxidized solution before precipitation of LaF_3; (2) precipitation of cerium and thorium fluorides with LaF_3 from the first oxidized solution; (3) the use of strontium holdback carrier during the LaF_3-PuF_4 precipitation in the first cycle; (4) repeated precipitation of LaF_3 from the oxidized solution in the second cycle; (5) precipitation of $BaSO_4$ with LaF_3 from the oxidized solution in the second cycle; (6) reprecipitation of the LaF_3-PuF_4 precipitate to eliminate occluded fission activity. The first and fourth of the modifications result in a significant improvement of the decontamination per cycle. Studies by Fries of extraction with LaF_3 from 20% and 30% UNH solutions give results similar to those obtained with the standard 10% UNH solution, through an extraction cycle and two decontamination cycles.

Bismuth Phosphate Process--La Chapelle, Dreher, and Thompson. A number of new reducing agents have been tested by La Chapelle as replacements for SO_2 since the SO_2 has occasionally proved unsatisfactory in steel. (1) Hydrogen peroxide reduces plutonium in 30 minutes at 75°C but interferes with the subsequent carrying of plutonium by bismuth phosphate. (2) Ferrous ion reduces plutonium in 30 minutes at 75°C but, at the usual phosphate concentrations, interferes with the subsequent carrying of plutonium by bismuth phosphate. (3) Phosphorous acid acts too slowly. (4) Hypophosphorous acid reduces plutonium, but gives unpredictable precipitates during analytical procedures and reduces bismuth ion to bismuth metal. (5) Hydroxylamine acts slowly at 75°C. (6) A 0.02 M hydrazine solution has given nearly quantitative reduction of plutonium in 10 minutes at 75°C in glass vessels. Erratic results, however, have been obtained, particularly in steel containers in which the supposedly reduced plutonium is not carried quantitatively by bismuth phosphate. These are being investigated further. (7) A satisfactory reducing procedure found by Dreher is to use H_2O_2 to rapidly reduce the dichromate, followed by a small amount of Fe^{+2} to reduce the plutonium.

Microchemical tests at high plutonium concentrations on the Bismuth Phosphate and Wet Fluoride Processes--Cunningham, Cefola, Patton, and Werner. In experiments using plutonium concentrations equivalent to those which will occur in the process solutions from the Site W piles, the carrying power of bismuth phosphate for plutonium has been consistently poorer than that of lanthanum fluoride. The results indicate that, within the range of conditions tested, the recovery of plutonium per precipitation will be 90% or below, resulting in a product yield of 70% or below for one extraction step and two decontamination cycles. The carrying power of bismuth phosphate for plutonium is quite sensitive to the concentrations of reagents in solution and to the technique of conducting the precipitation. The results further indicate that: (1) completeness of carrying of plutonium by bismuth phosphate is favored by addition of the Bi^{+3} in several portions, addition of the Bi^{+3} from 10 N HNO_3 and possibly by the presence of 2 N Na_2SO_4 (Patton and Cunningham); (2) the presence of uranyl ion favors completeness of carrying of plutonium by bismuth phosphate if the concentration of $UO_2(NO_3)_2 \cdot 6H_2O$ is in excess of 10% (Patton and Cunningham); (3) bismuth phosphate may be precipitated from solutions of oxidized plutonium at Site W concentrations without carrying more than 1% of the plutonium (Werner); (4) the carrying power of bismuth phosphate for plutonium in the absence of uranyl ion falls off rapidly at concentrations of Bi^{+3} below 1 mg/ml (Werner); (5) the solubility of plutonous phosphate in 0.15 M H_3PO_4 is ~ 5 mg/l in 0.1 M HNO_3 ~ 50 mg/l in 1 M HNO_3, and ~ 5 g/l in 5 M HNO_3 (Werner). Cefola tested a number of steps in the Wet Fluoride Process at Site W concentrations of plutonium. He finds that oxidation of plutonium by either Ag^{+2} - $S_2O_8^{-2}$ or by dichromate proceeds nearly quantitatively at the high plutonium concentrations of Site W and that LaF_3 may be precipitated from the solution without carrying the oxidized plutonium. Reduced plutonium may be precipitated to the extent of greater than 95% in the presence of 2.5 mg Zr/cc with a 5:1 weight ratio of La:Pu but not with a 1:1 ratio. Attempts to precipitate PuF_4 in the absence of both K^+ and LaF_3 indicate that it has a solubility of greater than 67 mg/l. In the presence of K^+ ion, however, the solubility is about 10 mg/l.

Adsorption methods for the extraction and decontamination of plutonium--Beaton, Fries, Sheft, and Turk. The treatment of a 10% $UO_2(NO_3)_2 \cdot 6H_2O$ solution of plutonium and fission activity with silica gel at 100°C to adsorb fission activity, followed by treatment with fresh silica gel at room temperature to adsorb plutonium which is subsequently eluted with acid gives a decontamination factor of ~100. This value is only about 10-fold better than that obtained with a single room temperature adsorption and elution. Adsorption of plutonium by silica gel from 10% uranyl nitrate solutions does not appear to be seriously affected by the presence of inactive fission product elements at the concentrations expected at Site W. A sixth cycle completed with a 1,200 gm column of silica gel shows 100% removal of plutonium during the first 8 liters of throughput. Preliminary tests have been made on the adsorption of plutonium and fission activity by a series of adsorbents, including carbonaceous, synthetic resin, and alumino-silicate types.

Sodium uranyl acetate method. Miller finds results comparable with those obtained by the Berkeley group on plutonium recovery (95%) and decontamination (greater than 10^5) in two cycles. Experiments show that the presence of Al^{+3} and Hg^{+2} in the concentrations anticipated in the uranium solutions from the pile probably will not interfere with the process.

Volatility methods--Brown, Hill, and Bohlmann. Experiments conducted on UF_4 and PuF_4 samples of comparable size suggest that PuF_4 is considerably less volatile at 600°C than is UF_4. When a mixture of UF_4, PuF_4, and CoF_3 was heated in a stream of He at 500°C for 0.5 hour, all the uranium (as UF_6) and none of the plutonium volatilized, indicating that the higher fluoride of plutonium forms less readily than UF_6 when CoF_3 is used as a fluorinating agent and suggesting that PuF_6 is a stronger fluorinating agent than CoF_3.

Perlman returned to Chicago at 3:00 p.m. from his inspection trip to Clinton Laboratories.

Helen had Wilma to our apartment to lunch and had dinner at the Thompsons'.

American bombers flew 2,000 miles from Australia to bomb Macassar, Dutch Celebes.

Friday, June 25, 1943

I am still in Billings Hospital, where Helen spent the afternoon with me. Dr. Cantril thinks I should take a four-week vacation when my fever goes down; I said I may take a two-week vacation later on. I was visited by Charles A. Thomas of the Monsanto Chemical Company (and his right-hand man, Carroll Hochwalt). He was with me when Helen came in with Wilma Ghiorso, who stayed just a few minutes. Thomas has been appointed by Groves and Conant to coordinate the plutonium purification and metal production work under way at the various sites to eliminate the light element impurities which undergo the α,n reaction.

A request is being made that Magel and Dallas transfer to the Technical Division.

General Groves is in Chicago today.

During much of the month La Chapelle has followed up on the strange results he obtained last month in which the carrying of plutonium by bismuth phosphate seems to be incomplete when the plutonium is reduced with a powerful reducing agent like hydrazine. He has performed numerous experiments with powerful reducing agents like hydrazine and hypophosphite and continues to find such diminished carrying by bismuth phosphate. This may be caused by the reduction of plutonium to an oxidation state below +4 that is not so well carried, or there may be some complex ion formation with the reductant that leads to reduced carrying.

The report of the Berkeley group, "Chemical Research--Production and Extraction of 94. Report for Month Ending June 20, 1943" (CN-723) is being issued and includes the following.

Ultramicrochemical Investigations--Craig, Hamaker, and Sheline. An investigation has been made of the nature of the precipitate which appears when $Pu(NO_3)_4$ and $Na_2Cr_2O_7$ are mixed in acid solution. Specific activity determinations on the precipitate indicate that it is a plutonium basic chromate. The crystal structures of sodium uranyl acetate and sodium plutonyl acetate were studied and show that the plutonium compound crystallizes as regular tetrahedra just as the uranium compound does. Both crystalline substances are found to be isotropic (photomicrographs of both are included in the report). Since the formation of tetrahedral crystals with Na^+ and Ac^- ions is quite characteristic of uranyl ion, the formation of similar crystals with plutonium (oxidized) lends weight to the idea that the oxidation number of plutonium (oxidized) is +6 and the compound is actually $NaPuO_2Ac_3$.

Decontamination Studies in the Sodium Uranyl Acetate Process--Beaufait, Connick, Crandall, Gofman, Pimentel, and Stein. Recoveries of plutonium have remained consistently lower than expected, and it is believed the loss is occurring during the first precipitation-filtration operation; the cause of the loss is now being investigated. Separation studies with inactive fission product elements added at W concentrations show an acid-insoluble residue upon dissolving the acetate precipitates; it may be columbium which could be troublesome. Work is continuing on the determination of individual fission element decontamination factors.

"Chemical Research--Special Chemistry of Plutonium. Report for Month Ending June 21, 1943" (CK-736) is being issued which includes the following.

Fluorine method for purification from beryllium--Brown and Hill. Samples of Be^7 tracer along with different amounts of inactive beryllium were treated with anhydrous HF and with F_2. Results indicate that if the beryllium is present in extremely small quantity, HF treatment at 600°-700°C is effective in removing a large fraction of it.

New techniques and studies of fluxes and reducing agents in metal preparation--Baumbach, Rosenfels, and Kirk. A study has been made of various means of reducing micro-quantities of uranium fluoride at high temperatures and of remelting uranium metal in the presence of fluxes in order to obtain a product which has been melted and compacted into solid metal, rather than aggregates of powder. Reheating or reducing with metallic magnesium or with barium and remelting in the presence of certain fluxes appear to be fairly satisfactory, yielding, in some cases, well over half of the metal in a single piece and giving no finely divided powders.

Precipitation reactions as a means of purification--Clifford Smith and Cunningham. Using uranium or thorium to simulate plutonium, Smith and Cunningham have performed precipitation reactions from solutions containing added impurities and the degree of purification was determined spectrographically. When boron, beryllium, iron, potassium, lithium, magnesium, sodium, and phosphorus are added to thorium in amounts of 10^4 ppm each, and the thorium is first precipitated as the oxalate and reprecipitated as the peroxide, beryllium, iron, potassium, magnesium, and sodium are collectively below the tolerance levels. Purification from boron below the tolerance level is not achieved, probably because the reactions have been carried out in Pyrex vessels; lithium purification cannot be evaluated because of analytical limitations, while phosphorus apparently does not separate from the thorium in these particular precipitations.

Tolerable concentrations of light element impurities in the final product. Kohman has compared the previous tolerable concentration limits calculated by Seaborg and Perlman with results based on experimentally determined neutron yields from the α,n reactions of polonium alpha particles recently made by J. Roberts. The results are tabulated in terms of maximum tolerance levels of the light elements in parts per million of Pu^{239}.

Brown reported that, on the basis of the June 23 monthly fluorine meeting at the Hooker Electrochemical Company, there appears to be no problem of supply of fluorine if a dry separations process is chosen for operation at Hanford.

The regular weekly meeting, chaired by Allison, of the principal chemists involved in the program to study the important remaining uncertainties in the Bismuth Phosphate Process was held today.

Helen ate dinner with the Ghiorsos, and then they went to a movie.

Saturday, June 26, 1943

Helen spent the afternoon with me in Billings Hospital.

Morris and Lincoln have been working on an alternative to dissolving the precipitate of bismuth and plutonium phosphate in 10 N HNO_3. They have evolved a process in which the phosphates are converted to the hydroxide by stirring with KOH and the precipitate may then be dissolved in as little as 1/8 of the HNO_3 otherwise required. The smaller amount of acid allows

greater freedom in choice of oxidation conditions and may make it possible to reduce volumes from one decontamination cycle to another. Potassium hydroxide seems to be preferable to sodium hydroxide, and the presence of a small amount of lanthanum during the metathesis seems to be necessary to prevent the loss of plutonium from the bismuth hydroxide precipitate.

The Technical Division semiworks made runs No. 27 and No. 28 during the week to obtain information on the use of sodium bismuthate as an oxidizing agent. Incidental to these, extraction carrying in the two runs was only 70-75% and 45%, respectively. This is a surprise and indicates a problem that must be solved.

C. M. Cooper sent a memorandum to Allison summarizing monthly progress in the Technical Division. He indicates that following the decision early in June by the du Pont Company to employ the Bismuth Phosphate Process, intensive development work has been shifted entirely to this process. Several process questions remained to be settled, but it is anticipated that early in the coming month it will be possible to start a series of successive runs intended to develop the degree of consistency which can be expected in actual plant operation.

"Chemical Research--U^{233} Production and Extraction. Report for Month Ending June 21, 1943" (CC-739) is being issued and includes the following report by Katzin, Schulze, Steahly, and Stoughton. The precipitation of Pa^{233} with MnO_2 from a solution of thorium nitrate has been studied in some detail and found satisfactory up to 36 gm $Th(NO_3)_4 \cdot 4H_2O$ per 100 cc solution. The concentration of Pa^{233} with respect to the manganese dioxide carrier and the thorium precipitated with the carrier appears to be accomplished quite well by either a series of MnO_2 cycles or thorium iodate cycles. Volatility methods for both the separation of Pa^{233} from the original thorium and for its separation from MnO_2 are now under investigation. After a study of its physical characteristics, it has been decided that thorium carbonate will be the thorium compound used in the pile at Site X.

Mrs. Compton came by to see Helen in the evening, with flowers and cookies for me.

Within three hours after President Roosevelt vetoed the Smith-Connally anti-strike bill today, Congress passed the measure over his veto, and it became law.

Sunday, June 27, 1943

In Billings Hospital. Helen spent the afternoon with me.

There was a meeting of the Council of my section in my office at 10:30 a.m., attended by Kirk, Willard, Cunningham, Orlemann, Brown, Thompson, and English. The following topics were covered.

Purification work. Orlemann said one of the new men will be assigned to make a study of ether extraction methods for purification. Dean will study organic reagents for purification using thorium as a stand-in. The problem of purifying various reagents has been discussed with Boyd and Potratz, and it is agreed that someone should be placed in charge of this. The space problem is serious, and it may be possible to make arrangements with Burton to share some of his space. We may be able to get some space in Kent for our purification work. Brown discussed the problem of collecting plutonium by the dry methods. He feels we have solved the problem and that with higher plutonium concentrations there will be no difficulties. He plans to try experiments with Site W concentrations of plutonium in UF_4. There is no definite data on the volatilization of the lower plutonium fluoride as yet. It is probably less volatile than UF_4. Katz will attempt to convert PuO_2 to the bromide as a connecting link between the peroxide concentration method and the metal production step. Watters is still working on the determination of carbon. Cunningham mentioned that Rollefson has checked us on the purity of the plutonium samples we supplied him. He estimates 2.5% as the maximum impurity, and we calculate that 2.5% would be a maximum. Clifford Smith is setting up to get some vapor pressure measurements on plutonium bromide. A fraction of a microgram of plutonium can be volatilized at 270°C in 15 minutes as the volatile bromide. Fairly consistent purification with respect to lithium and beryllium should be obtained. For purification a combination of the oxalate and peroxide methods for precipitation seem to give the best results. He plans to prepare pure MnO_2 and find out if it will absorb impurities present after oxalate precipitation. Hindman is determining the weight change when PuO_2 is converted to PuF_4. Patton is working on the oxidized state of plutonium. The Berkeley group has strong evidence for the +6 state as against Cunningham's work showing a +5 state. The discrepancy may be due to difficulty in removing traces of dichromate from the glassware at Berkeley. Similarly in Cunningham's work, traces of a reducing agent might be causing trouble. Patton finds that reduction of dichromate by HNO_3 occurs except when the HNO_3 is treated by distillation in the presence of dichromate. The microchemists are planning to measure solubilities of organic compounds prepared by Dean.

Metal production. Most of our present problems are mechanical according to Kirk. None of the crucible coatings now available is satisfactory. We are being held up somewhat on the job of determining the density because of lack of material. It was suggested that we should study several reducing agents for plutonium to find out how electropositive the metal is.

Bismuth Phosphate Method. Weekly meetings on Friday are now being held by Allison in order to follow the work being done on the method, and it appears now that the only remaining problem that is serious is the carrying power in the semiworks. These semiworks results are not so good as those obtained in similar conditions in the laboratory. The last semiworks run gave only a 45% yield, and this run involved the use of Na_2SO_4 to compensate for higher acidities produced in the metal dissolving step. Miller is working on the carrying power at high plutonium concentrations, and his results indicate that we can expect a yield of only about 90% for each precipitation in the extraction and decontamination steps. We are planning to do some

work on the composition of bismuth phosphate using radiophosphorous and radiobismuth. More men are needed on the microchemical work at the present time, and Cunningham thinks Cefola will be available in about one week.

Monday, June 28, 1943

In Billings Hospital. Helen visited in the afternoon.

Miller has continued to obtain erratic results in the carrying of plutonium by bismuth phosphate in the extraction step at Site W concentrations. Today he finished an experiment in which he finds about 88% carrying, better than he obtained in a number of experiments last week. but still not very good.

Willard received a letter from J. J. Howland, Jr., at the University of Rochester, who explains that there has been a delay in scheduling his final examination; he will notify us as soon as he knows definitely when he can leave to join us.

In a memo to Whitaker English discussed the present status of counting equipment for the control laboratory and other counting rooms at Site X.

There was a Laboratory Council Information Meeting on Chemistry attended by Allison, Boyd, Burton, Cohn, Compton, Connick, C. M. Cooper, Coryell, Doan, Franck, Garrison, Hilberry, Johns, Miles, Orlemann, Potratz, Rollefson, Spedding, Turkevich, and Willard. Rollefson discussed his attempts to get a plutonium spectrum using his facilities in Berkeley. With the 20 microgram plutonium sample furnished by Cunningham, he has obtained 176 lines, mostly attributed to plutonium; it looks as if the spectrum is simpler than that of uranium. Willard discussed extraction and decontamination work on the Bismuth Phosphate Process; he mentioned that up to 60% of the plutonium is oxidized in the dissolving of uranium metal but can be reduced with 0.02 M Fe^{++}. Yield of plutonium at each step is 90% to 70% in one extraction plus two decontamination steps. Consideration is being given to dissolving the bismuth phosphate precipitate by metathesis with NaOH. On oxidation studies of plutonium, tests are being made on use of $NaBiO_3$, or Ce^{+4}, or metathesis followed by $K_2Cr_2O_7$. Willard also mentioned that Kirk has produced pieces of plutonium metal up to one microgram in weight in thoria and beryllia microcrucibles. Orlemann discussed purification work, mentioning the analytical work on Be^7, carbon, and oxygen, the precipitation studies using Th^{+4} as substitute for Pu^{+4}, involving oxalate, peroxide, and iodate precipitates. He also mentioned that work on organic precipitates is under way. Solvent extraction methods are being studied. Volatility methods have been studied using fluorides, and chlorides and bromides will be studied.

Cunningham, Werner, Miller, Dreher, Patton, and Thompson held a meeting to discuss a program for working out the best method of carrying high concentrations of plutonium with bismuth phosphate. It was decided that three programs should be carried on simultaneously: (1) Microchemical experiments in which Cunningham will personally assist Miller in determining

the difficulties which have been encountered in Miller's work, after which Miller will study effects of agitation and increasing the phosphoric acid concentration. He will also do experiments to determine whether the chemistry of plutonium and thorium are sufficiently similar to justify continuation of Dreher's work with thorium to simulate plutonium. (2) Tracer studies will be made of the effect of high concentrations of fission products, effects of Na_2SO_4 to compensate for the nitric acid remaining from the dissolving step, and the effect of Fe^{++}. Phosphoric acid concentration and acidity variations also will be studied. (3) Experimental work using thorium at Site W concentrations will be continued, since this work has given some clues as to the factors which influence carrying power. It was further agreed that, as soon as Cefola is finished with his present work, he will assist Miller on bismuth phosphate work.

"Technological Research--Separation Process Section. Report for Month Ending June 26, 1943" (CN-758) is being issued. It is reported that activities during the month have been largely concentrated on the Bismuth Phosphate Process. Intensive study has been made of the oxidation procedure, and it has been found that the use of sodium bismuthate as oxidizing agent appears to have eliminated an earlier yield difficulty. Ceric ion also shows a great deal of promise. Preliminary evidence has been obtained that the addition of inactive fission products in Site W concentrations has no adverse effect on the beta decontamination achieved by three decontamination cycles. Both beta and gamma decontamination are appreciably poorer, however, in the first and second decontamination cycles. Also studied during the period were adsorption and ether solvent extraction processes.

"Technology--Chemical Engineering, Report for Month Ending June 28, 1943" (CN-756) was issued and mentions that the Bismuth Phosphate Process has been selected as the process to be used at Site W. Chemical engineering investigations have proceeded far enough to permit specification and design of the major part of the equipment and plant. Details of the process have not yet been sufficiently stabilized to warrant reproducibility runs in the semiworks.

Today is the Comptons' wedding anniversary.

In the Mediterranean our bombers have raided Messina for the third time in two days.

Tuesday, June 29, 1943

In Billings Hospital. Helen dropped in to see me in the evening.

There was a conference on adsorption processes in Franck's office. Beaton reported on the work of Fries, Sheft, and Turk and his own work on Hypersil silica gel and other absorbents (synthetic resin ion exchangers obtained from the Chemical Process Company of San Francisco, "basic resins" from du Pont, "Catex," synthetic zeolite, and "Catex-284"). Adsorption

from complexed plutonium solutions was also covered. Boyd presented information on experiments with Amberlite IR-1 and Zeo-Karb H.

Tonight there was the regular Tuesday evening meeting of the Extraction Group of my section attended by Apple, Brown, V. R. Cooper, Cunningham, Davidson, Dean, Dreher, English, Ghiorso, Hindman, Katz, Morris, Peery, Potratz, Clifford Smith, Steahly, Stoughton, and Thompson. The meeting consisted of a review of current work on extraction methods. Thompson discussed the Bismuth Phosphate Process. Beaton reviewed the situation relative to the irreproducibility of results with silica gel and presented the more recent and more favorable results on the use of tannic acid in connection with the adsorption on Catex-284. Stoughton outlined the work now being done on the effect of thorium to iodate ratio and total iodate concentration on the carrying of protactinium by the partial precipitation of thorium as the iodate. English cited data compiled by Jarrett on the counting loss versus counting speed on the Geiger counters. These data indicate a loss of about 8% at 10,000 counts per minute.

Helen met Alice Thompson downtown and, when she returned home, stopped by the Comptons'.

War news today is of U.S. bombing raids in the Athens area in Greece and against Japanese bases in the Pacific.

Wednesday, June 30, 1943

I came home from Billings Hospital, Vance Cooper bringing Helen and me in his car. My release from the hospital was the result of a successful stratagem. I was running a low-grade fever and knew the hospital would not release me until my temperature was normal. By using my ingenuity I managed to keep the thermometer from going above 98.6°F and in this way fooled the nurse.

Gaarder finds that the presence of 2.5 N Na_2SO_4 prevents the precipitation of uranyl phosphate from 20% UNH, 0.36 M H_3PO_4, 0.5 N H_2SO_4 solution--a UNH concentration suitable for the extraction step in the Bismuth Phosphate Process.

Today Katz attempted to produce plutonium hydride using a small (approximately 0.2 microgram) button of metallic plutonium supplied by Kirk. The sample of plutonium was heated in a high vacuum (10^{-5} mm) to 500°C; and then hydrogen, furnished by the decomposition of uranium hydride, was admitted at a pressure of 630 mm. The temperature was allowed to fall slowly to room temperature over a period of four hours, after which the hydrogen was pumped off and air admitted to the system. Examination of the metallic button of plutonium, if it was plutonium, indicates that no visible change has occurred. This is quite surprising and casts some doubt on the identification of the button as plutonium metal.

6/30/43 (cont.)

B. Brody sent to Orlemann a memo summarizing the Project work to date on the solvent partition method for purifying plutonium. Solvents mentioned were ethyl ether, carbon tetrachloride, ethyl acetate, chloroform, ethylene dichloride, methyl ethyl ketone-10% xylene, ethyl bromide, nitromethane, 1-nitropropane, and 2-nitropropane.

The Laboratory Council Policy Meeting today was attended by Allison, Cantril, Compton, C. M. Cooper, Doan, Franck, Greenewalt, Hilberry, Miles, Spedding, Stearns, and Whitaker. Franck presented information on the chemistry program as will be given in his monthly report MUC-JF-58. Total scientific manpower of my section was given as 54 persons. Compton reported there is great activity in evidence at Site W. On information exchange between sites, he mentioned that Chicago does not receive from Site Y information on military uses, and Site Y will not receive details of production piles. Both groups interchange basic scientific information. Compton also said the New Chemistry Building Annex has been approved and the cell for the Van de Graaff machine is already built just south of the present New Chemistry Building in an opening between that building and the proposed Annex. Chicago has been authorized to go ahead with experimental P-9 (heavy water) piles, both heterogeneous and homogeneous, to be built at Argonne.

Thursday, July 1, 1943

At home convalescing. The Baumbachs visited me during the early part of the evening, and Stan Thompson came by later.

I received a friendly note from Joyce Stearns. He wrote, "Dr. Cantril has sent me a memo in which he recommends a vacation of four weeks for you. I want to supplement his request that you take such a vacation. We need you in good shape for a long period of time a darn sight more than we do for a short period at the present time. You have worked hard on this project and will have to do so again. This is not a request--it is an order, one of the few ever issued by this office. Go off on this vacation and don't let's hear from you for four weeks."

Manfred Lindner started work today as a Research Assistant. He received his bachelor's degree in chemistry from Northwestern University in 1940 and has been working at the S.A.M. Laboratory at Columbia University. Lindner is being assigned to the Bismuth Phosphate Process.

Franck sent a memo to William J. Vatter, the budget officer, stating that Willard is authorized to sign for me during my absence from the Laboratory.

Allied land, naval, and air forces occupied the Tobriand and Woodlark groups, between New Guinea and the Solomons, and are attacking the neighboring New Georgia Islands.

Friday, July 2, 1943

At home convalescing.

Today Katz reacted with oxygen the metallic plutonium sample that he treated with hydrogen on Wednesday. In a previously evacuated system he passed dried tank oxygen over the metal for one-hour intervals at 100°C, 200°C, 300°C, 500°C, and 800°C. No change in appearance was observed until 500°C was reached when a slight tarnish was observed. At 800°C the surface of the button appeared to be covered with a dark brown coating. This resistance to oxidation is quite surprising and may cast doubt on the identification of the sample as metallic plutonium.

Also today Baumbach reduced CeF_4 to cerium metal with magnesium as a reductant and LaF_3 to lanthanum metal with magnesium as a reductant using beryllium oxide crucibles and his usual technique.

"Monthly Report of the Activities of the Chemical Division" (MUC-JF-58), J. Franck, Director, was issued. For Section C-I, activities are reported as follows:

I. Extraction and Decontamination of Plutonium

a. Wet Fluoride Process: The effectiveness of various oxidizing and reducing agents has been studied. Carrying power of lanthanum fluoride for plutonium under different conditions has been determined. Oxidation with dichromate and reduction with arsenite show the best results. Twenty percent UNH gives about the same results as the usual 10% solution. A number of steps in the process have been successfully tested at W concentrations. (Manpower: 8 before selection of $BiPO_4$, 1 after; Priority B.)

b. Bismuth Phosphate Process: Hydrogen peroxide in combination with ferrous ion seems the best reducing agent in presence of steel. Microchemical tests at W concentrations of plutonium show poorer carrying than at lower concentrations. Solubility of plutonous phosphate is 5 mg/liter in 0.1 N HNO_3, 50 mg/liter in 1 N HNO_3. (Manpower: 7 before selection of $BiPO_4$, 17 after; Priority A.)

c. Adsorption qualities of silica gel are variable within the same batch. Adsorption of plutonium is not changed with changes in fission product concentration. New adsorbents are being studied. (Manpower: 3; Priority B.)

d. Experiments on the Sodium Uranyl Acetate Method confirm Berkeley results and show aluminum and mercury do not interfere. (Manpower: 2; Priority B.)

e. Microchemical tests show PuF_4 is much less volatile than UF_4. PuF_6 is not formed by the use of CoF_3 as fluorinating agent. (Manpower: 1-1/2; Priority B.)

II. Purification of Plutonium and Metal Production

a. Radioactive Be^7 for tracer use has been isolated. Treatment of tracer amounts of beryllium with HF at 600°C removes 90% of the beryllium in 1-1/2 hours, whereas larger amounts (3 mg) show no losses when fluorinated at 500°C for 3/4 hour.

b. Purification by precipitation methods has been studied using uranium and thorium as stand-ins. Boron, beryllium, iron, potassium, lithium, magnesium, sodium, and phosphorus were added to thorium in amounts of 10^4 ppm each and the thorium was precipitated first as the oxalate, then as the peroxide. Purification determined by Mark Fred--beryllium, iron, potassium, magnesium, and sodium were removed to below the tolerance level.

c. Tolerable concentrations of light elements have been recalculated.

d. Metal production has been studied with microquantities of uranium fluoride. Barium and magnesium as reducers give good results. (Manpower on all of Purification and Metal Production: 11 before selection of $BiPO_4$, 19 after; Priority A.)

III. Instrumentation and Physical Measurements (Manpower: 8; Priority A)

IV. Work on UF_6 for heavy water plant (Manpower: 1-1/2; Priority C)

V. Rollefson's work in Berkeley on spectroscopic analysis of plutonium also is reported. Using 20 micrograms of plutonium, he has observed about 170 lines, ranging from 1,800 to 8,800 angstroms. The excitation is done by means of a 15,000 volt condensed spark between si_ `r electrodes.

The regular weekly conference on the Bismuth Phosphate Process was held this afternoon with Allison presiding. Among those present, besides Allison, were Willard, Greager, and Coryell. Semiworks flowsheet changes recommended in the extraction step include doubling the concentration of Fe^{++} (to 0.04 M) in the reduction step, halving the nitric acid concentration (to 0.2 M), and doubling the H_3PO_4 (to 0.8 M) in the product precipitation step, with the Bi^{+3} added in three strikes. A semiworks run with 0.2 N HNO_3 gave 85% carrying compared with 45-50% carrying at 0.4 N HNO_3. The decontamination cycles flowsheet revisions include oxidation with sodium bismuthate in place of dichromate and ceric ions and reduction with a higher concentration of Fe^{++} instead of the combination of H_2O_2 and Fe^{++}.

Complete occupation of Rendova Island in the Central Solomons by our forces was announced today.

Saturday, July 3, 1943

At home convalescing.

During much of the last month Miller, under Cunningham's tutelage, has been learning the ultramicro technique in order to make an independent check of the carrying of plutonium by bismuth phosphate at Site W conditions. During the last week he has been getting discouraging results which we had hoped were caused by deficiencies in his technique. To our relief today he performed an experiment under the conditions of the extraction step at Site W and obtained yields of 98%.

Today Baumbach reduced ThF_4, prepared by Schulze, with magnesium metal by carrying out the reaction at about 2000°C, made necessary by the high melting point of thorium metal which is about 1845°C.

"Metallurgical Laboratory Progress Report for June 1943" (MUC-NH-651) is being issued. Included is a summary of work on separations processes, purification, and metal production as follows. The report states that work on the Wet Fluoride Process has been largely cleaned up and emphasis transferred to the Bismuth Phosphate Process. Difficulties have appeared with the carrying of plutonium by bismuth phosphate at Site W concentration. Work has continued on the study of oxidizing and reducing agents in the presence of stainless steel and on the effect of operation variables on their efficiency. This program will continue at full strength for some considerable time. Work on alternative methods has continued, particularly on the adsorption method, during the month. This work is to be continued on low priority. Further basic studies in the purification program have been carried on during the month, particularly on volatilization and precipitation methods. Work has been started on metal production

by development techniques using microquantities of uranium. Total expense for the month of June was $362,532. Total personnel on the Project on June 30 is 743 (325 academic).

The Technical Division Weekly Report states that, as agreed yesterday, run No. 30 is being carried out in the semiworks using the following conditions for extraction: 20% UNH, 0.2 N HNO_3, 1.0 N H_2SO_4, 0.8 M H_3PO_4, W concentrations of fission products, La^{+3} and Ce^{+3} holdback carriers, and three-step precipitation of 2.5 g/liter Bi^{+3}. In the decontamination cycles the oxidant is $NaBiO_3$ and the reductant is Fe^{++}.

Both the U. S. and Australia made gains in New Guinea.

Sunday, July 4, 1943

Today is Independence Day. I am still at home convalescing, so Helen and I did not go anywhere to celebrate. Because members of the Council of my section had plans of their own, we did not hold the customary Sunday morning meeting.

Monday, July 5, 1943

At home convalescing.

"Chemical Research--Production and Extraction of 94, Report for Period Ending July 1, 1943" (CN-778) is being issued with the following content. Bismuth Phosphate Process for the Extraction and Decontamination of Plutonium. Dixon, James, V. Cooper, Shepard, and Thompson report that both ceric ion and sodium bismuthate have proved in laboratory tests to be satisfactory oxidizing agents for plutonium. Control Analyses. English and Crawford survey the control analysis problems related to the use of the Bismuth Phosphate Method at Sites X and W. A table shows the amount of uranium beta and gamma activity, associated with samples containing 3,000 plutonium alpha disintegrations per minute from different steps in the process. They have developed an ionization chamber with a permanent magnet to bend out the beta rays; using this chamber alpha counts can be made on samples containing up to 1.3×10^9 beta dis/min. Uranium Correction in the Lanthanum Fluoride Method of Analysis. Beaton, Fries, and Turk find the amount of uranium carried by lanthanum fluoride precipitates in the fluoride method of analysis for plutonium may be reduced by dissolving the lanthanum fluoride in zirconyl nitrate and reprecipitating it with more HF. Struthers finds that digesting a LaF_3 precipitate in a uranyl solution at 50°C may increase the uranium correction several-fold.

Allied forces continue to expand their gains in the new southwest Pacific offensive.

Tuesday, July 6, 1943

Still at home.

During the last couple of weeks Davidson and Katz have been trying to study the properties of plutonium hexafluoride in the pure form. They mounted about 0.2 microgram of Pu^{239} as the oxide in a small nickel boat, treated it with HF at 550°C for four hours, and then inserted it in a small nickel microreactor that they have built. A slow stream of fluorine was passed through the apparatus, and the temperature of the reactor gradually raised to 650°C. At the conclusion of the experiment they found that 30% of the plutonium had not left the nickel boat, and the rest of it was in the nickel reactor vessel. They believe that most of the plutonium collected on the surface of the nickel at temperatures between 300°C and 500°C.

Harrison Brown sent the following letter, over my name, to Whitaker at Site X: "We have included in our plans for research at X a rather intensive study of the possibilities of the dry fluoride process for extracting and decontaminating plutonium. Plans, which have been under way for many months, include both laboratory studies and engineering studies. The laboratory work will be carried out under the direction of H. S. Brown who in turn will be responsible to I. Perlman. The engineering studies will be carried out in collaboration with the engineering group of W. Q. Smith.

"It is planned to install in the semiworks area of the 706 building two rather large fluorine cells. These will operate at 750 amps each and will generate enough fluorine to process as much as 200 lbs. of uranium metal per day if necessary. A fraction of the output from these cells will be stored in tanks for use in the laboratory studies. The initial scale of the engineering studies will be of the order of 5-25 lbs. of uranium metal per day. The construction of the large fluorine cells is already well under way, and it is hoped to have them in operation by the first of September. A large DC motor generator set will be required to operate the cells. This has already been purchased and is at present in our laboratory in Chicago. It is now only necessary to ship the generator to X and install it. Adequate power provisions have been installed for its operation. The personnel working on this particular problem will consist of Brown, three men working on small-scale laboratory experiments, and approximately four or five men working on the engineering problems involved. Of these men one or more men will be assigned from the engineering group of W. Q. Smith.

"So far as we can determine at the present time all major points concerning the installations at X for the development of this process have been adequately taken care of."

In order to facilitate the setting up of a comprehensive research program for Site X, Lyle B. Borst compiled a 27-page document entitled "Research Program at the Clinton Laboratories." It is divided into nine parts: (1) Operation and interpretation of the X pile; (2) Design, construction, and operation of the W pile; (3) A. Operation of the X

separation plant. B. Design, construction, and operation of the W separation plant; (4) Concentration of product; (5) Study of physical problems; (6) Study of fission products; (7) Study of new separation processes; (8) Study of the effect of radiations and products on biological tissue, and (9) Study of the health of employees.

Slightly abridged quotes from the above topics that are of greatest concern to me, namely, the separation plants for Site X and Site W (3A and 3B), the concentration of product (4), and the study of new separation processes (7), are as follows:

SEPARATIONS PLANT

(3) A. Operation of X Separation Plant.

1. Dissolver performance. The dissolving cycle and the exact nature of the dissolving operations are difficult to estimate on the basis of semiworks data. Studies of dissolver performance should be made in the early stages of plant operation to determine the optimum method of solution and the time required. Most of these data may be obtained on inert material before active material is handled. The results of this study will be important in determining the operating schedule and dissolver design for W.

2. Loss of iodine in solution step. Preliminary laboratory experiments indicate that an appreciable fraction of the 8-day iodine will be lost during the solution of the metal and will be carried with the off-gases out of the stack. The radiations from I^{131} are much more energetic than those from xenon. A determination of this loss under the conditions of plant operation is of great importance from the point of view of personnel safety. The problem will be much more severe at W, and results from X operation will influence W design. Measurements will be taken of the activity in the off-gases at the base of the stack. An attempt is to be made to recover active xenon from these off-gases in a "xenon factory." It is improbable that the iodine will follow the xenon through this purification system. Measurements through this process may be used to find conditions under which the iodine deposits and the efficiency of its removal. Data may be obtained on this problem for semiworks operation which may be used to get an estimate of conditions for plant operation.

3. Performance of steam jets. As the chemical process is now set up, neither pumps nor valves will be used in contact with active material. Transfer from one cell to another will be done by steam siphons. The requirements imposed upon this equipment are far more severe than normal. The pressure head against which the jets operate will vary; yet the discharge, which goes to a centrifuge, must be constant. The change to the Bismuth Phosphate Process will greatly relieve corrosion conditions. Performance tests have already been made in the laboratory, and it is believed that specially designed jets will be satisfactory for plant operation. They are being used at present in the Chicago semiworks, and better data will be available before the start-up of the plant. Operating performance of these siphons is, nevertheless, of great importance for W design.

4. Centrifuge performance. An extended period of plant operation will be necessary to check the adequacy of centrifuge design.

5. Equipment cleaning. Preliminary work has indicated that some difficulty may be experienced in washing equipment free of radioactive material. Since this problem will be much worse at W, special attention should be given to it at X.

6. Reduction of 49 by radiation. The possibility that 49 in an oxidized state may be reduced in the presence of large radiation densities is apparent under the conditions at W. This will constitute a loss of product in the separations process. The seriousness of this effect should be determined under plant conditions at X as well as under laboratory conditions with the radiation densities expected at W.

7. Flowsheet for individual fission products. The course followed by individual fission products through the separation and decontamination processes is of importance to determine the intensity of radiations at any particular stage. Most of the present work on decontamination has been based on overall beta counting.

It will be important to know the nature and quantity of the various gamma emitters at different stages of the process. For the purpose of X plant operation decontamination followed by gamma-ray analysis is probably adequate, but to obtain a complete picture it is highly desirable to know the course of each individual element.

8. Semiworks production. In order to obtain a few milligrams of product at the earliest possible date, it is probable that the semiworks will operate on active pile material as soon as this is available. About 15 lbs. of metal will be processed each day until the separation plant has completely processed two or three batches.

(3) B. Design, Construction, and Operation of the W Separations Plant.

1. Radiation failure of materials of construction. All construction materials for the present equipment, metals and alloys, gaskets, and electrical insulators, have been selected on the basis of laboratory tests for corrosion resistance and structural stability when subjected to radiations. It will be important to carry out an analogous program of plant tests at Site X to evaluate these materials under actual operation conditions.

2. Effect on process of W concentrations of fission products. The effect of W concentrations of fission products has been evaluated on a microscale. It will be important for W design to know whether unexpected effects occur on a macro or semiworks scale. If the engineers desire, they can probably obtain W radiation concentrations by processing newly discharged metal.

3. Effect on process of W concentrations of 93 and 94. It is not anticipated that W concentrations of 93 and 94 will affect the process, but this should probably be checked on a laboratory scale. A small sample can be artifically enriched to the proper concentration of 94 and processed at the proper time after shutdown to give an equivalent concentration of 93. To do this on a semiworks scale would require about a week's output

of product from the separations plant. It is unlikely that we would be permitted to do such an experiment in the first few months of plant operation.

4. Bismuth phosphate filters. The du Pont Company would greatly prefer to use filters instead of centrifuges for the Bismuth Phosphate Process at W. They wish performance tests made on such equipment. This will require the investigation of filtering materials which may be done either at Chicago or by the chemical engineering group at X. Equipment must be designed and procured to replace one or more centrifuges in the separations plant. It could then be installed and its operating performance determined.

(4) Concentration of Product

The product discharged from the separations plant or semiworks will be decontaminated to a point safe for laboratory manipulations. Further concentration will be carried out in the chemistry building. The chemical methods used will probably be a combination of wet fluoride and peroxide precipitation methods. In a few steps 94^{239} may be concentrated to a point satisfactory for shipment to Chicago. The purity shipped is expected to be 90% element 94 as a compound such as peroxide, fluoride, or nitrate. The remaining 10% will consist of uranium, lanthanum, and bismuth. A considerable portion of this group's time will be devoted to chemical trouble shooting in the semiworks and in the separations plant.

(7) Study of New Separation Processes

1. Dry $HF-F_2$ Method. Laboratory and semi-scale equipment will be built at Site X to study the use of hydrogen fluoride and fluorine in the separation of product. Anhydrous hydrogen fluoride will be obtained in cylinders at the site. A fluorine generator is being installed capable of producing a few pounds of fluorine per day.

2. Adsorption studies. A group in the chemistry building will devote its attention to the development of an adsorption method for the separation of 49. It will be working on a scale of a few pounds of metal per batch with active pile material. Studies will be made of the effect of radiation on adsorbents under the conditions to be expected at W operation.

3. Solvent extraction studies. Preliminary work is being done in Chicago headed toward developing a method of product separation based on solvent extraction. The most satisfactory solvent at the present time is ether, but a further survey of the field will be made. When this work has progressed far enough, the semiworks equipment will be constructed at X to test the process on 10-pound batches of active metal. The stability of the system at radiation levels to be expected at W will be further investigated by the photo chemical group.

4. Recovery of metal from wastes. In the production unit at W it will be highly desirable to recover the uranium from waste materials. Studies of this recovery probably will be made at Chicago and X after solving more urgent problems.

7/6/43 (cont.)

Covey has been on a two weeks' vacation in Canada. Today he returned to the Lab and was just in time to participate in the ether extraction of the first and second 50-pound batches of UNH from the Chicago V neutron irradiation completed June 14.

The evening meeting of the Purification Group of my section was attended by Boyd, Brown, Cefola, Cunningham, Davidson, Dean, Fred, Fries, Ghiorso, Hindman, Jaffey, Katz, Katzin, Kirk, Nachtrieb, Orlemann, Patton, Potratz, Rosenfels, Clifford Smith, Stoughton, and Willard. Katz described work on the hydrogenation and oxidation of plutonium metal. At 500°C in an oxygen atmosphere, there was some tarnishing; and at 800°C, the material changed to a black speck. Potratz described the extraction of aqueous solutions of reduced plutonium with cupferron into a chloroform layer. This method will be particularly useful for the concentration of aluminum impurities. Nachtrieb outlined various possible materials for use as a matrix in the spectrographic determination of refractory light impurities. Potratz discussed the possibilities for fluorine analysis. A potentiometric method involving the Fe^{++}, Fe^{+++} couple; the conversion of fluoride to BF_3 and spectrographic determination of boron; and a fluorometric method involving the AlF_6^- complex are all under consideration. Fred reported that the pyroelectric excitation method is not suitable as a purification method. Orlemann described the work of Kroner on purification of hydrogen peroxide by the formation of the urea complex. Brody's work on ether extraction is apparently complicated by reduction of oxidized plutonium by something in the ether. Patton and Dean are collaborating on inorganic precipitants.

The meeting continued with Patton reporting that the plutonium salt of m-nitrobenzoic acid has a solubility of 4 mg/l in aqueous solution and 2 mg/l in chloroform. He said phenylarsonic acid gives a precipitate in acetic acid and picrolinic acid gives a crystalline precipitate in acetic acid solution. Cunningham and Smith described work on the purification of thorium (as stand-in for plutonium) by precipitation reactions. Phosphate impurities are removed by oxalate precipitations, and other troublesome light elements are effectively removed by fractional precipitation of the thorium hydroxide. Boron and sodium are troublesome when working in glass. Davidson described unsuccessful experiments on the fluorination of plutonium in a micro system. All the plutonium is found in the nickel reactor and does not reach the platinum collector. Consideration of density as a function of atomic number suggests the possibility of a density greater than 22 for plutonium metal.

A big sea battle is raging off the Solomons. In addition, the Germans have opened a long-expected offensive in the Soviet Union.

Wednesday, July 7, 1943

At home convalescing.

Harrison Brown sent a memo to my office listing 16 distinct sections that he visualizes for the complete dry fluoride research program at Chicago until August 1 and at Chicago and Site X after August 1.

The Nazis made initial gains in their new offensive in the Belgorod sector. In the naval battle in the Pacific, six Japanese ships were probably sunk and four damaged.

There was a Laboratory Council Policy Meeting held at 10:00 a.m., attended by Allison, Compton, C. M. Cooper, Doan, Fermi, Hilberry, Hughes, Jeffries, W. Johnson, Miles, Rapier, Smyth, Spedding, Stearns, Stone, Whitaker, and Wigner. Cooper presented the Technical Division report giving the distribution of effort of the 167 scientific and technical personnel in the Division. Whitaker mentioned it would probably be September 1 before the Clinton pile starts up and that he would like to have some definite plans and schedules on the materials that are to go into the pile. This brought up the subject of U^{233} production. Allison said that Newson is planning to put in enough thorium to make 1 milligram per day at 1,000 kw, and Compton remarked that Site Y has requested 2 milligrams in addition to what is needed here at Chicago. Compton then requested Allison to prepare a memo in collaboration with Fermi on the amounts of U^{233} required. Compton announced that Doan will take over the duties of research coordinator at X. He gave his personal guess that the first Site W reactor will start up around April 1. Demineralized water will not be used in the first pile unit there. Some 5,000 persons are already at work on construction.

Thursday, July 8, 1943

At home convalescing.

General Groves is in Chicago.

Jerome J. Howland, Jr., who just received his Ph.D. degree from the University of Rochester, started work in Section C-I as a Junior Chemist. Willard was his senior research advisor at the University of Wisconsin in 1939-40. One reason why Howland accepted Willard's invitation to join us is that Professor Albert Noyes told him that if he "wants to work on a long-shot project, then the Chicago group has the approach with the best chance of success."

Harrison Brown wrote, over my name, to Dr. R. Rosen of the Kellex Corporation in New York City giving at his request estimates of the gamma radiation due to UX (the mixture of U^{238} decay products Th^{234} and Pa^{234}) that would be found in Rosen's UF_6 extraction unit. Brown's conclusion was that Rosen should take precautions against the UX radiation.

My section issued a summary, consisting of 18 pages and 23 tables, of the Bismuth Phosphate Method for the extraction and decontamination of plutonium, which includes summaries of our experimental results at

each step in the process. It will be distributed at the weekly Bismuth Phosphate progress meeting tomorrow.

The Research Assistants in my section had their regular Thursday evening meeting. Brown gave a talk on hex production.

American troops have landed at two points on New Georgia Island, but in Europe more Russian towns have fallen in the new Nazi drive.

Friday, July 9, 1943

At home convalescing.

Cefola has performed experiments to measure the possible formation of double salts between PuF_4 and LaF_3. In experiments in which plutonium is precipitated with amounts of lanthanum fluoride insufficient to carry all of the plutonium, the lanthanum and plutonium precipitate in a constant stoichiometric ratio of 2:1, suggesting that carrying occurs by formation of the double salt $PuF_4 \cdot 2LaF_3$.

The weekly Bismuth Phosphate progress meeting this afternoon was attended by Apple, Balthis, Brown, C. M. Cooper, Coryell, Franck, Greager, W. Johnson, Kay, Leader, Miles, Nylen, Olsen, Peery, W. Q. Smith, Squires, Sutton, Thompson, and Willard. Willard reported on 14 separation runs during the week by the microchemists employing an improved technique. Plutonium recoveries were consistently high (97-98%) under varying conditions. Sutton presented data from intermediate (liter) scale runs indicating yields in the range of 90-98% for five runs under varying conditions. Smith reported on semiworks run No. 31 in which product recovery in the extraction step was only 50% in a three-step precipitation. A further precipitate from the centrifuged liquor raised this to 90%. Decontamination results by all groups are now satisfactory. It was the consensus that the separations step is the present major uncertainty and that effort should be concentrated on it so that, by the time of the meeting next week (July 16), we can decide upon conditions to be employed in the semiworks consistency runs.

Today's reports indicate that the U. S. troops are still gaining in the Pacific but that the Nazis have driven a new wedge into Soviet lines in the Belgorod sector.

Saturday, July 10, 1943

At home convalescing.

Warren C. Johnson started work today as the Associate Director of the Chemical Division and will be Franck's Deputy. Johnson comes to us from the University of Chicago where he is an associate professor of chemistry. He received his Ph.D. from Brown University in 1925.

Willard wrote to his friend, Winston Manning, who is at the Carnegie Institution of Washington, Division of Plant Biology, Stanford University, that he is pleased to learn from Stearns that Manning has accepted a position with us and invited him to stay at the Willards' while Manning is house-hunting.

"Capture and Fission in Metal, Nitrate, and Oxide" by A. Turkevich, T. P. Kohman, A. H. Jaffey, C. D. Coryell, D. W. Engelkemeir, N. Elliott, T. B. Novey, H. S. Brown, and N. R. Davidson (CN-771) is being issued. The 20-page report covers measurements made at the St. Louis cyclotron to determine which configuration and combination of uranium metal, oxide, and nitrate give the highest yield of 94^{239}. It is concluded that a 5-inch layer of metal around the target does not decrease the yield of 49 obtained in the nitrate mass and, in addition, provides a large amount of product in the metal itself.

We have sent to Los Alamos the 200 microgram sample of Pu^{239} that they requested for use in measuring the value of ν for Pu^{239}.

Today's headline in the paper is two inches high: Allies Invade Sicily!

Sunday, July 11, 1943

There was a meeting of the Council of my section at 10:30 a.m. in my office. Present were Willard, Kirk, Cunningham, Brown, English, and Thompson. Willard mentioned that Doan will be the coordinator of research at Site X. W. C. Munnecke, who was vice president of Marshall Field and has been an administrator in the Army program, will take Doan's place as Associate Director of the Met Lab. Joe Hamilton is visiting us from Berkeley and will obtain some radiophosphorous for us. Cunningham suggested that arrangements be made to obtain the 70 pounds of uranium metal that was placed very close to the target of the St. Louis cyclotron in the present bombardment. He said this material should be very rich in 93^{237}, and we should arrange to have the engineers dissolve it for us. The fluoride precipitate from the present ether-extracted solution of 400 pounds of active nitrate (from the fifth St. Louis bombardment which ended June 14) has settled, and we plan to siphon off the supernatant. The LaF_3 will be metathesized and dissolved in zirconium. The 200-microgram sample of plutonium which has been sent by plane to Site Y will be returned to us as soon as their measurements have been made. We now have approximately 100 micrograms of plutonium on hand for immediate use, of which Kirk has 50 micrograms. By this time the engineers should have an additional approximately 90 micrograms to be concentrated.

At the present time the manpower situation is very critical; and, when the move to Site X takes place, we will be severely short-handed at Chicago. We will try to contact some more men immediately, and it should be remembered that there will be plenty of space after the move. Magel is moving to Site B where he will be in Chipman's group.

Cunningham said that we must have a purification laboratory very soon that will be large enough for six men. This is necessary because we must keep all sodium and sodium salts away from this area. Brown suggested the idea of moving Covey's men into the shop and converting Covey's room into a laboratory. English has been trying to get a glassblower for Site X. When the New Chemistry Building Annex is completed, "Osty," the glassblower, will move; and we will have the space (Room 3) vacated by him. We will also get three rooms in the new Annex and about two more in this building. The Annex is scheduled to be finished about October 20 (see Figure 2). Discussion of the best arrangement of the present space was made. It was felt that Orlemann would have to have more space. Some of his men could occupy space in Room 5 as well as Room 6.

Vance Cooper drove Helen and me to the railroad station where we took the evening train for a three weeks' vacation in New Mexico. We are headed for Valley Ranch, near Santa Fe, recommended to us by friends as a good spot for a rest and vacation. The fact that this will put us near Site Y, within reach of some of our friends there, also has contributed to our choice of destination.

Monday, July 12, 1943

Helen and I are enroute to Lamy, New Mexico.

French T. Hagemann received a Ph.D. degree from the University of Washington in Seattle this year and started working in Section C-I today. Katherine Harvey began work as a laboratory assistant for English today.

Joseph Hamilton is visiting the Metallurgical Laboratory from Berkeley to discuss possible military applications of fission products.

Stan Thompson sent a letter over my name to Connick in Berkeley notifying him that the stainless steel beaker that he requested has been sent to him, along with the neutron-bombarded Cb-Mo-U alloy prepared by Magel. Thompson mentioned that the conditions for the final semiworks operation of the phosphate method have not been established as yet--the engineers have not decided whether to use Ce^{+4} or $NaBiO_3$ as oxidizing agent. He added that conditions that were established as giving good results in the laboratory for the extraction step do not work well in the semiworks. "Results," he wrote, "using these same conditions in microchemical work at 100:1 ratios gave very good results; so a lot of worrying is being done about why the semiworks doesn't check. They do, however, get good results in the decontamination cycles."

Doan ordered from Ruhoff in the Manhattan District Office in New York 600 pounds of UNH, 200 pounds to be shipped to Fulbright in St. Louis for our neutron bombardments and 400 pounds to be shipped to Jaffey at the Met Lab.

There was a meeting to discuss the ether extraction process attended by Willard, Adamson, Boyd, Hughes, Kay, Olsen, Peery, Russell, and Maloney.

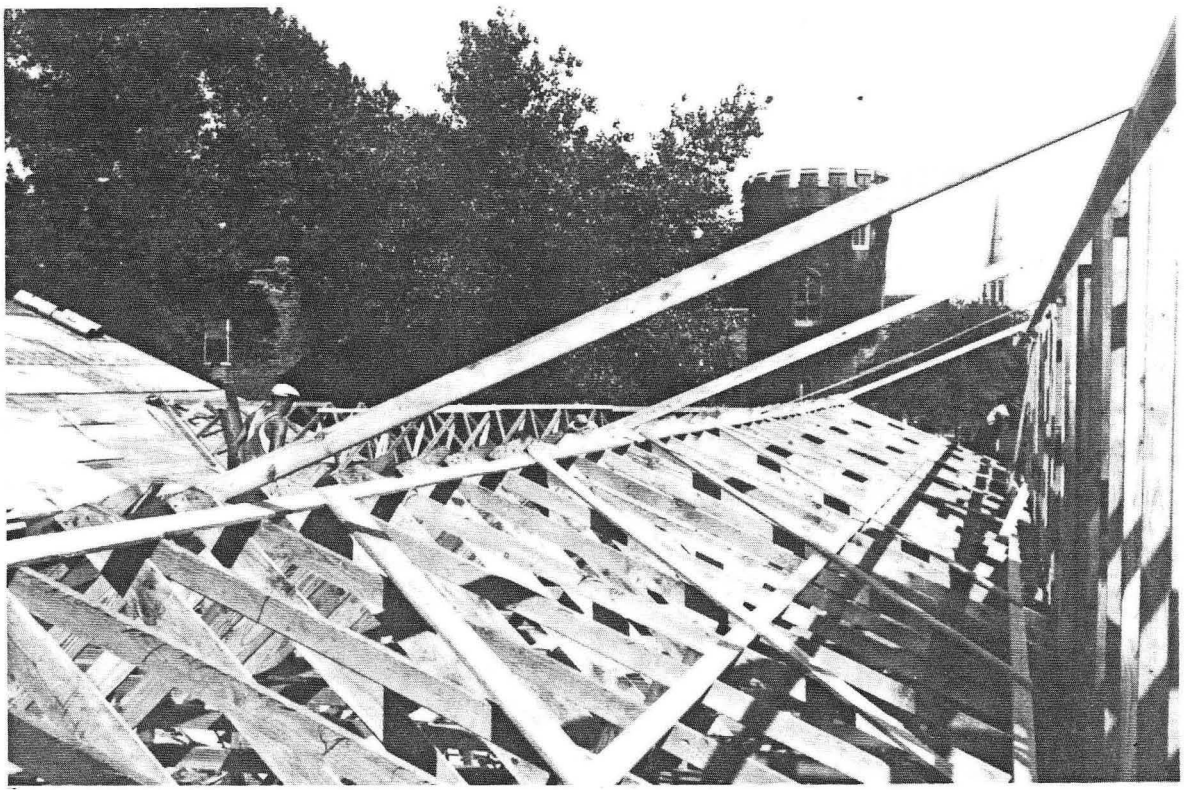

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Figure 2. New Chemistry Building Annex under construction. July 1943.

It was agreed that there are two major problems which should be investigated as soon as possible: (1) effect of radiation on ether-water-nitrate mixtures; (2) development of a suitable oxidation procedure for plutonium.

Tuesday, July 13, 1943

Helen and I arrived in Lamy, New Mexico, where we were met by a driver who took us to the Valley Ranch at Valley Ranch, New Mexico, elevation 7,200 feet, about 25 miles from Santa Fe.

John Willard answered the inquiry of Benjamin C. Bradshaw of Pembroke, Kentucky, about possible employment. Willard wrote that we would very much like to hear from him further if he is interested in laboratory research in inorganic chemistry, as his record suggests he is well qualified to fill an opening that we will have in the near future. He noted that although there is no time-clock punching, most of our staff works more than the prescribed 48 hours a week.

Franck asked Doan to send five pounds of unbombarded uranium metal to Gofman in Berkeley for use in practicing dissolution of uranium metal in preparation for working up the uranium metal target material being bombarded with neutrons using the Berkeley cyclotron. Franck then wrote to Connick and Gofman in Berkeley telling them of the forthcoming shipment of uranium metal to them and suggesting that they read the Technological Group's reports on metal dissolution. Franck mentioned he would not be present at the July 26 Laboratory Council Information Meeting on Chemistry because he wants to take a vacation the latter part of the month, but he wanted to know who would be coming from Berkeley and whether they will want to talk at both the morning and evening meetings.

Covey will interview some applicants for his job while I am on vacation. Because of his experience I have asked him to go to Site X next month for the duration and help set up our new laboratories.

The Extraction Group of my section met this evening. It was noted that for the first time neutron-bombarded metal is being used to test the process. Each step of the Bismuth Phosphate Process was discussed--solution step, reduction after dissolving, extraction step, metathesis as an aid in dissolving the bismuth phosphate, the oxidation step in the decontamination cycle, the by-product precipitation and subsequent reduction, and the precipitation of bismuth phosphate.

Wednesday, July 14, 1943

Helen and I are at the Valley Ranch. The food so far has not been very appetizing.

During the last couple of weeks Dreher, Vance Cooper, and Gaarder have tested the conditions under which uranyl phosphate might precipitate

in the extraction step of the Bismuth Phosphate Process in order to establish conditions to prevent this. They have established the concentrations of H_2SO_4, HNO_3, and H_3PO_4 that meet the requirement of non-precipitation of uranyl phosphate from 20% UNH solution at 75°C--conditions applicable to the extraction step.

Hamilton, who is still visiting from Berkeley, met with Warren Johnson and C. M. Cooper to discuss the technical aspects of the possible military applications of fission products.

There was a Laboratory Council Policy Meeting at 10:00 a.m. in Room 209, Eckhart Hall, attended by Allison, Compton, C. M. Cooper, Doan, Fermi, Franck, Greenewalt, Hilberry, Hughes, Miles, A. V. Peterson, Smyth, Spedding, Stearns, Stone, Whitaker, and, later, Wigner. Stone presented a report on the work in the Health Division for June and July. Greenewalt summarized the status of activities at Site W: Drawings on the separations processes already have been sent to the field. Separation and two decontamination cycles will be carried out in the canyons (heavily-shielded concrete structures) and the third cycle and bulk reduction will take place in less heavily-shielded facilities. The first pile is scheduled to operate May 1, to be followed by piles 2 and 3 at one-month intervals. Next, Allison summarized the situation at Site X during the time of his visit last week. Some 24,000 men are working on all projects there. Fermi followed with a report on Site Y. He said that the laboratory there now has four neutron sources--a cyclotron, two van de Graaff generators, and one D-D source. All are ready or nearly ready. A comparison of U^{235} and Pu^{239} made there indicates that Pu^{239} may not be as much better as had been anticipated. He said, however, that people at Site Y appear to be putting their hopes on Pu^{239}.

Compton read a letter to the Council. It was from President Roosevelt to General Groves, dated June 29, emphasizing the importance of our program. Compton suggested that the letter be read generally to members of the Project; and, after some discussion about whether or not to hold a large meeting, it was decided to read the letter at each Division meeting. Roosevelt wrote as follows:

"My dear General Groves:

"I have recently reviewed with Dr. Bush the highly important and secret program of research, development and manufacture with which you are familiar. I was very glad to hear of the excellent work which is being done in a number of places in this country under your immediate supervision and the general direction of the Committee of which Dr. Bush is Chairman. The successful solution of the problem is of the utmost importance to the national safety, and I am confident that the work will be completed in as short a time as possible as the result of the wholehearted cooperation of all concerned.

"I am writing to you as the one who has charge of all the development and manufacturing aspects of this work. I know that there are several groups of scientists working under your direction on various phases of the program. The fact that the outcome of their labors is of such great signficance to the nation requires that this project be even more drastically guarded than other highly secret war developments. As you know, I have therefore given directions that every precaution be taken to insure the security of your project. I am sure the scientists are fully aware of the reasons why their endeavors must be circumscribed by very special restrictions. Nevertheless, I wish you would express to them my deep appreciation of their willingness to undertake the tasks which lie before them in spite of the possible dangers and the personal sacrifices involved. In particular, I should be glad to have you communicate the contents of this letter to the leaders of each important group. I am sure we can rely on the continued wholehearted and unselfish labors of those now engaged. Whatever the enemy may be planning, American Science will be equal to the challenge. With this thought in mind, I send this note of confidence and appreciation.

"Very sincerely yours,

"Franklin D. Roosevelt"

Across the letter Roosevelt had scrawled "Secret," although all White House communications are considered to be secret. It was his way of emphasizing the special secrecy of this Project.

Thursday, July 15, 1943

Helen and I are at the Valley Ranch. This is a very quiet place--almost too quiet.

During the last few weeks Clifford Smith, Kroner, and Cunningham have continued the program of using thorium as a stand-in for plutonium in precipitation reactions as a means of purification. They have performed many experiments involving the precipitation of the peroxide and the iodate of thorium, some including the use of redistilled water, quartz rather than pyrex vessels, waxed vessels, and many other precautions in order to insure purity. They have concluded that purification of the water in nitric acid doesn't result in a marked increase in purity of the final ThO_2, whose content of impurities was determined spectroscopically; that waxed apparatus is about as satisfactory as the more difficult-to-obtain quartz apparatus; and that the following purification scheme is the most effective--one oxalate precipitation followed by two or more peroxide precipitations, then a fractional hydroxide precipitation of about 10% of the thorium which is discarded, then precipitation of the remainder of the thorium as oxalate, washing the final precipitate at least five times. Similar experiments using thorium as a stand-in and organic precipitants have been performed by Dean and Orlemann. They believe that the use of metanitrobenzoic acid and 8-hydroxyquinoline offer promise.

Knox has continued his tests of the oxidation state of plutonium following the solution of neutron-bombarded uranium metal in 60% UNH. He has attempted to check for the oxidation state by precipitating the plutonium with sodium uranyl acetate that carries the oxidized form of plutonium in contrast to the use of lanthanum fluoride, as in his previous experiments, that carries the reduced state of plutonium. In experiments in the last few days he has obtained small amounts of carrying by the sodium uranyl acetate, but today he found carrying corresponding to about 50% of the plutonium present in the oxidized state.

Zvolner has been testing the reduction of oxidized plutonium resulting from the use of 60% nitric acid in the metal dissolving step in the Bismuth Phosphate Process. The plutonium must be in the reduced state in order to be carried by the bismuth phosphate in the extraction step. He finds that ferrous ion is satisfactory for this, but a number of other reducing agents have not proved satisfactory under the conditions thus far tested.

Herbert H. Anderson, who has been working on an NDRC project under Professor Schumb at M.I.T., started work in Section C-I today. Anderson received an A.B. from Harvard in 1934 and a Ph.D. from M.I.T. in 1937; he is a specialist in fluorine chemistry.

A six-pound cylinder of unbombarded uranium metal was shipped by express to Gofman in Berkeley in accordance with Franck's July 13 request to Doan.

The weekly Thursday evening Research Assistants seminar at 6:45 p.m. is now being held in Room 209, Eckhart Hall. This evening a lecture on the structure and chemical characteristics of plutonium was given by Cunningham.

At the meeting of Research Associates the speakers were Wollan of the Division of Health and Bellas of the Engineering Development Section.

Friday, July 16, 1943

Helen and I are still at the Valley Ranch.

During the last couple of weeks Miller has completed an exhaustive test of the carrying of plutonium by bismuth phosphate at Site W concentrations. The tests were made by precipitation of bismuth phosphate from 20% UNH solution using 2.5 milligrams of bismuth per cm^3, 0.4 M H_3PO_4, 0.02 M Fe^{+2}, and inactive fission products added to correspond to those that would be present in one ton of uranium which had been in the Hanford pile for 100 days at full power. The solution was kept at 75°C for three hours. He tested a number of variations of H_2SO_4 and Na_2SO_4 concentrations as well as HNO_3 concentration and varied the method of precipitation from one precipitation up to three precipitations of bismuth phosphate. To our relief he uniformly finds very excellent

carrying of plutonium, 98% or better; and it appears that a single precipitation produces as good carrying as a three-shot precipitation.

The weekly Bismuth Phosphate progress meeting was held today. Data were presented which confirm that in the metal dissolving step considerable oxidation of plutonium occurs, as evidenced by precipitating sodium uranyl acetate (which carries the +6 oxidation state) from the solution of the metal. In the extraction step it has been shown that carrying by bismuth phosphate is improved by three hours digestion time instead of one hour.

Saturday, July 17, 1943

Helen and I are at the Valley Ranch. We rode into Santa Fe and visited the museum, "Palace of Governors." Because of our dissatisfaction with the Valley Ranch--the food is poor and it is a dull place--we made reservations for a room at LaFonda Hotel there for a stay to start next Wednesday.

Doan was appointed Research Director for Site X by Compton and will be relieved of his responsibilities as Chief Administrative Officer of the Metallurgical Laboratory at the earliest possible date.

Perlman sent a memorandum to Donald Hughes, Jr., Special Assistant to Compton, commenting on a letter from Conant to Compton regarding the relative amounts of gamma activity associated with the "23" system as compared with the "49" system. Perlman gave the results of computations on gamma activity associated with 250 grams of product (Pu^{239} or U^{233}); these show that, at moderate intervals of time after pile shutdown (6 days to 60 days), the gamma activity of the two systems is comparable; at a shutdown time of one year, the U^{233} system is considerably more favorable with a fission product activity of 200 curies for U^{233} versus 10^4 curies for Pu^{239}. At the earlier time periods there is a considerable contribution from the 27-day Pa^{233} in the "23" system.

W. Q. Smith recommended that L. C. Peery be placed in charge of a trouble-shooting group at Clinton with responsibility for maintaining contact with the Chemistry and Process Development Sections at Chicago.

The Separation Processes Section reported (Technical Division Memo 101) on the status of their intensive study, begun last week, to locate the cause of the low yield in the extraction step in the semiworks. Samples removed at different points in the semiworks procedure were processed further in the laboratory, and this gave good yields. It is concluded that the factors involved in the yield differences occur during the digestion step. Possibilities being explored are differences in (1) agitation, (2) heating rate to reach digestion temperature, (3) maintenance of digestion temperature, (4) pick-up of impurities during digestion, and (5) changes in state of oxidation of some impurities during digestion. In the Chemical Engineering Section the semiworks runs Nos. 33 through 36 were completed. Yields of Pu^{239} in the extraction step range from 20% (run No. 33) to 57% (run No. 35) indicating the persistence of unsolved problems.

7/17/43 (cont.)

War news in the paper here shows Allied gains on all fronts. In the Pacific bombers ripped Maunda air base. The Soviets made new gains in their Orel offensive. Allies continued to hurl the Nazis back in Sicily.

Sunday, July 18, 1943

Helen and I are at the Valley Ranch. I certainly am getting a lot of rest here, but the boredom is very frustrating.

Monday, July 19, 1943

Helen and I are still at the Valley Ranch.

Francis Donlan, whom Covey interviewed and hired as his replacement, is starting work today in Section C-I.

Joe Katz has built a generator that produces HBr by the action of bromine on tetraline (tetrahydronaphthalene). Today he treated a small sample of carrier-free Pu^{239}, in the form of PuO_2, on a platinum foil with HBr from this source. He finds that a stream of HBr at 875 - 910°C for a half hour removes the plutonium from the platinum plate. This may suggest the production of a volatile plutonium bromide. This source of HBr, however, is erratic and in order to obtain a quantitative volatility curve, it will be necessary to have a purer and more reproducible source of HBr.

Miller has tested, on the ultramicroscale, the carrying of the plutonium at Site W concentration by bismuth phosphate in the first step of the decontamination cycle, that is, the first precipitation of reduced plutonium after the extraction step from the UNH solution. Under the flowsheet conditions using Fe^{+2} ion as the reducing agent, he finds in a number of experiments uniformly good carrying of the order of 97%.

In a letter dated July 16, 1943, Oppenheimer wrote Compton from Site Y using a letterhead with the address "Los Alamos, Santa Fe, New Mexico, P.O. Box 1663." He was reporting exciting news on the results of measurements using the 200-microgram Pu^{239} sample that we lent them. He described their preliminary result for ν, the average number of neutrons emitted in fission, for Pu^{239}. He said in part, "The method we have used is proton recoils with a Li(p,n) source. If we assume that the thermal fission cross section of 49 is 1.83 times that of 25, the result of our measurement may be stated as follows: $\nu(49)$ is 1.20 ± 0.06 times that of 25. This error is the standard rather than the probable error of the observations alone. It includes no provision for the possibility that the figure 1.83 may be in error or that the chemical analysis of the sample may be wrong. On this latter point we are fairly sure of our ground. The limits of error set by Wahl are about 2 percent, but as you know, surprises can always be present in work on this scale.

"We propose to remeasure the ratio of $\nu(49)$ to $\nu(25)$ by at least two other essentially independent methods and are confident that we can

complete this within the time that the material has been assigned to us. You will, I know, apply all the usual appropriate scientific precautions to a result which is only two days old. Nevertheless, I think that we can assure you that the chances are insignificant that the neutron emission of 49 is substantially worse than that of 25."

There was an emergency meeting of members of the Extraction Group of Section C-I to discuss one of last week's semiworks runs (No. 33) on the bismuth phosphate extraction step which was carried out under the following conditions: 0.2 M Fe^{+2}, 0.2 N HNO_3, 1.0 N H_2SO_4, 2.5 mg/cc Bi - 1 shot, Ce^{+3} and La^{+3} holdback carriers, 0.8 M H_3PO_4, and Site W concentrations of fission products for a pile operating at 2,800 kw per ton. The digestion period was four hours at 75°C. Thorough discussion attempted to explain the small yield of only 20%, with inconclusive results.

Tuesday, July 20, 1943

Helen and I are at the Valley Ranch, but we are looking forward to leaving here tomorrow.

Cunningham and Ghiorso have initiated a project to attempt to define the specific activity of plutonium more accurately by determining the counter geometry with more precision. The approach decided upon is the preparation of counting plates having a known weight of uranium thinly and uniformly spread over them. Because the specific activity of uranium is supposed to be accurately known, the fraction of the theoretical number of alpha particles emitted (which are registered by the counter) could be used to define the counter geometry. As the first step, a solution of Mallinckrodt UNH was standardized by igniting duplicate aliquots to U_3O_8 and weighing.

Winston M. Manning, Willard's friend, started working in Section C-I today.

"Chemical Research--Special Chemistry of Plutonium, Report for Month Ending July 17, 1943" (CK-806) is being issued with the following content. Baumbach, Rosenfels, and Kirk describe their work on the preparation of plutonium and other metals. Two samples of plutonium fluoride (prepared by Brown) and one of plutonium oxide (prepared by Hindman) have been reduced with magnesium in a vacuum at 1550-1750°C. Both were readily reduced to minute spherical globules of metal which were bright white and quite soft. The largest globule was weighed by Hindman and found to be one microgram. As a test of the technique, thorium, lanthanum, and cerium metals were prepared and agreed in appearance with their descriptions found in the literature. Katz and Davidson report their work on reactions of plutonium metal with hydrogen and oxygen. Since the reaction of hydrogen with massive uranium is a good means of powdering the metal and rendering it accessible to further chemical reactions, it was considered of interest to determine whether the same process could be applied to plutonium. Treatment of a 0.2-microgram sample of metallic plutonium with hydrogen at 500°C showed no visible change. The effect of oxygen

was next investigated at various temperatures up to 800°C, and the sample appeared resistant to oxidation also. In evaluating these results, the possibility that the sample is actually a plutonium-magnesium alloy must be kept in mind. Katz and Davidson also report on their studies on the use of the higher plutonium fluoride in a purification process. They have attempted to prepare the volatile higher fluoride of plutonium from the lower fluoride in an all-glass high vacuum apparatus using cobaltic fluoride and argentic fluoride. No volatilization of plutonium was observed. In order to minimize surface effects, plutonium lower fluoride was treated with fluorine in a semi-micro apparatus employing a nickel reactor and a platinum capillary. All the plutonium remained in the nickel reactor.

Clifford Smith, Kroner, and Cunningham describe precipitation reactions as a means of purification. Using thorium as a substitute for plutonium, they have made further studies of (a) the effect of using quartz apparatus and specially purified reagents on the purity of the final ThO_2; (b) partial precipitation as a means of purification; (c) techniques for removal of phosphorous and boron. The general conclusions are that purification of the water and nitric acid do not markedly increase the purity of the final ThO_2, that waxed apparatus is as good as quartz apparatus, and that the most satisfactory purification cycle found to date is one oxalate precipitation followed by two peroxide precipitations and a partial (10%) hydroxide precipitation that is discarded. Sodium and magnesium are still the cause of some difficulty. Preliminary studies on the use of organic reagents in the purification of plutonium are summarized by Dean and Orlemann. Using thorium as a substitute for plutonium, they have made precipitation tests with hexamethylenetetramine, 8-hydroxyquinoline, and m-nitrobenzoic acid. The tests show a promising decrease in impurities. Tests of the solubility of various organic thorium compounds indicate that picrolonic acid and 8-hydroxyquinoline may be of value in solvent extraction methods for purifying plutonium. Bond and Katz describe an analytical procedure for U^{232} tracer; an analytical procedure for isolating small amounts of uranium which depends on the carrying power of barium carbonate has been evolved for possible use in purification work using uranium fortified with U^{232}.

Compton requested that Allison transfer an experienced cyclotron operator to Washington University until September 15 in order to insure continued production of 49 from the St. Louis cyclotron. He suggested V. Alexander Nedzel, a Research Assistant, as a possibility.

W. C. Munnecke replaced Doan as Chief Administrative Officer of the Met Lab; this is in addition to his present responsibilities as Associate Project Director for Administration.

Orlemann sent a letter over my name to the King Laboratories in Syracuse, New York, placing an order for 10-20 pounds of pure barium, to be prepared on a cost-plus basis, and to be investigated as a reductant to produce pure plutonium metal as free as possible from light element impurities.

7/20/43 (cont.)

The regular evening meeting of the Purification Group of my section was attended by Baumbach, Boyd, Brown, Cefola, Cunningham, Davidson, English, Fred, Fries, Ghiorso, Hagemann, Hindman, Howland, Jaffey, Katz, Katzin, Kirk, Kohman, Manning, Nachtrieb, Orlemann, Patton, Perlman, Potratz, Rosenfels, Stoughton, Watters, and Willard. Kirk reported successful practice reductions of thorium, lanthanum, and cerium salts. A five-microgram plutonium sample as the nitrate was evaporated in a lucite-coated beryllia crucible and ignited to remove the lucite. The sample was reduced and remelted by successive magnesium treatments and gave one quite large metal lump that, however, did not contain all the plutonium. Rosenfels described an improved technique for mounting samples in lucite. Cunningham reported that Patton has been studying the ideal conditions for precipitating thorium with organic reagents. English exhorted the group against contamination in the counting rooms. The magnets and magnetizer for the high background counters at X are almost ready for use. Nachtrieb reported encouraging results in the detection of trace elements using the impurities with no carrier. Orlemann reported that Watters can determine carbon to less than 0.05 microgram. Brody found that a bismuthate-oxidized solution of plutonium is stable for several days in 3 N HNO_3, 1 N HCl, after centrifuging off the bismuthate. When shaken with ether about 50% of the product is extracted. Dixon has extracted reduced plutonium into chloroform, in the presence of acetylacetone. Patton stated plutonium cupferride is reddish brown. Potratz said that plutonium is well extracted into a chloroform phase with cupferron, whereas potential light impurities such as lithium, beryllium, iron, sodium, magnesium, aluminum are not so extracted. Orlemann claimed that the work of Kroner indicates that a silver iodide precipitation from 3 N HNO_3 removes iron, phosphorus, and magnesium impurities. The meeting adjourned early.

Top headlines today tell of the bombing of Rome and the Soviets' capture of 130 more towns near Orel.

Wednesday, July 21, 1943

Helen and I moved from Valley Ranch to the LaFonda Hotel in Santa Fe.

Madeline Lenhard began working in Section C-I today as a clerk for Ghiorso.

Continuing the work of accurately determining the alpha counter geometry as a preliminary to measuring the specific activity of plutonium, Cunningham prepared four counting standards using the calibrated UNH solution. Counting of the standard samples was begun by Ghiorso.

The Council of my section changed the time of its meeting from Sunday morning to Wednesday evening. Tonight's meeting was attended by Perlman, Kirk, English, Willard, Cunningham, Orlemann, Thompson, and Brown. Perlman indicated the pile at Site X should start up about September 15. He asked Thompson to consider a program for carrying on

extraction research at Site X so that as little time as possible will be lost when the move occurs. Brown mentioned that there are now 33 milligrams of plutonium in the Argonne pile, 6 milligrams of which are in the center of the pile. This activity corresponds approximately to the activity of the last metal that was bombarded with neutrons at St. Louis. English indicated there will be nine Geiger counters and 15 electroscopes available at Site X, as well as one FP54 electrometer setup which can measure activities as high as 50 millicuries. The first magnetic alpha counter should be ready for testing in a few days; it will be capable of counting alpha particles over a background of 1.3×10^9 betas but has the disadvantages of having only a 7% geometry and of being very sensitive to the location of the sample. Cunningham mentioned there is no space for any more men to do microchemical work here. There are only 30 micrograms of plutonium on hand at the present time, but it is estimated that 350 micrograms will soon be ready as a result of working up the material from the fifth St. Louis cyclotron bombardment (Chicago V); the product from the last ether extraction has been carried through two fluoride cycles so far by Hindman. The 80 pounds of hot metal from St. Louis for the 93^{237} extraction is here, and we are arranging to have the engineers dissolve it. N. Baumbach is working on routine recovery of plutonium from waste solutions and is succeeding in effecting a good augmentation of our supply in this manner.

Patton's work on organic precipitants was discussed--the cupferron complex is the only organic plutonium compound found so far which is appreciably soluble in chloroform. Howland is occupied with learning our techniques of measuring the pH of microliter solutions; these will be applicable to the measurement of the hydrolysis of Pu^{+4}, that, in the form of $Pu(ClO_4)_4$, has been found to hydrolyze in weak acid solutions. Clifford Smith is measuring vapor pressures of the chlorides and bromides of uranium, which necessitates the preparation of complicated high vacuum apparatus. Orlemann described the method of determining oxygen that involves reacting CCl_4 with UO_2 and measuring the phosgene produced. Considerable work is being done on the determination of chlorine. Hagemann is working on the determination of oxygen, and Katz is studying conversion of the oxide to the bromide. PuO_2, when heated to 1600°-1700°C in vacuum, gives a shiny black material which is evidence of a lower oxidation state of plutonium. Brown said that 800 pounds of UF_6 is now circulating through the equipment at Argonne at the same rate necessary in a larger pile.

Thompson discussed the discrepancy between the results in the semiworks and in the laboratory. The lower yields in the semiworks may be due to combinations of such differences as the size of equipment, agitation, use of 25-12 steel equipment, and method of adding reagents. A program has been set up for the semiworks to eliminate such variables one by one. The subject of coprecipitation was discussed, and it was agreed that considerable work should be done on the carrying by bismuth phosphate which Thompson believes is not an adsorption phenomenon. A program has been undertaken to examine crystals prepared in a variety of ways--possibly radiographs will be prepared. James is doing some work on testing bismuth phosphate in accordance with some of the rules laid out by Otto Hahn. Koshland is doing some work on the composition of bismuth phosphate, while Miller is attempting to carry plutonium at very low ratios of Bi:Pu.

There was a Laboratory Council Policy Meeting at 10:00 a.m., attended by Allison, Compton, C. M. Cooper, Fermi, Franck, Hilberry, Hughes, W. C. Johnson, Miles, Smyth, Spedding, Stearns, Stone, and Wigner. Fermi reported on progress during the past month in the Physics Division; Cooper reported that the concentration of plutonium in the Argonne pile metal is nearly equivalent to the present metal from St. Louis. By September 1 two tons of uranium will have been removed from the Argonne pile and will have had several weeks cooling, according to Fermi. Compton reviewed an organization plan he is preparing for Chicago and related laboratories, under which the Laboratory Council will become the "Project Council" with essentially the same membership. Also, Compton proposed, and it was agreed, that Council meetings will occur on Tuesday and Wednesday, rather than on Monday and Wednesday.

Thursday, July 22, 1943

Helen and I are now staying at the LaFonda Hotel in Santa Fe.

Cunningham and Ghiorso completed counting the uranium samples and computed the specific activity of the uranium, using the mass of uranium known to be present in each sample. The results indicated a counter geometry that exceeds the maximum possible 50% in two of the three samples. It was concluded that contamination of the uranium with alpha activity has occurred.

John Willard requested by memo to Stearns that an offer of a position be made to B. C. Bradshaw.

Compton sent a letter to Conant based on the information Perlman furnished in his memorandum of last Saturday to Hughes, concerning the relative ease of manufacture of "23" as compared with "49." Compton points out that a cooling period of two years would have to elapse to take full advantage of the possibility of processing 23 without the need for radiation precautions but agrees with Conant that in any case the absence of a whole series of fission products would make processing the "23" easier than the "49." Conant will be visiting the Met Lab next week.

"Chemical Research--Production and Extraction of Plutonium, Report for Month Ending July 17, 1943" (CN-813) is being issued, including the following reports. Bismuth Phosphate Method. Using neutron-bombarded uranium metal, Knox has conducted experiments to determine the amount of oxidation which may occur in the dissolving step and finds as much as 60% is in the oxidized state. Zvolner has investigated methods of reducing this oxidized plutonium and finds ferrous ion satisfactory. Experiments with hydroxylamine, phenylhydrazine, formic acid, and sodium nitrite show none is a satisfactory reducing agent. Prevention of precipitation of uranyl phosphate (Dreher, Cooper, and Gaarder). Conditions of sulfate and phosphoric acid concentrations are established that prevent an appreciable amount of uranyl phosphate from precipitating from 20% UNH solution

at 75°C in three hours or more. Carrying of plutonium by bismuth phosphate. Because of poor yields in the semiworks, the effect on carrying of different variables has been studied. Dreher, Dixon, and Dam have conducted tracer experiments that show uniformly good carrying over a wide range of conditions, both with and without 0.02 M Fe^{+2} and Site W concentrations of fission product elements present. Miller has conducted microchemical experiments using Site W concentrations of fission product elements at Bi:Pu ratios of 100:1 and finds 96% or better carrying. Dreher, Gaarder, and Miller have tested the extent to which the analogy between thorium and plutonium is valid by carrying out duplicate microchemical experiments on the carrying of thorium and plutonium by bismuth phosphate plus identical beaker scale experiments with thorium. Results indicate comparable carrying with both macro and microscale experiments and that plutonium is carried more completely than thorium. Morris and Lincoln have studied the metathesis of bismuth phosphate as a means of reducing the amount of acid required for dissolution and thereby have achieved a volume reduction. Satisfactory conditions are found using KOH plus a small amount of lanthanum to prevent loss of plutonium. G. Johnson has studied neutralization of the acid after the oxidation step in the decontamination cycle, as an alternative to dilution. The results suggest that NaH_2PO_4, Na_3PO_4, or $(NH_4)_2HPO_4$ may be used without causing any appreciable amount of plutonium to be lost in the by-product precipitate. V. Cooper and Halperin find that Fe^{+2} in concentrations from 0.0025 M to 0.02 M will reduce more than 95% of the oxidized plutonium in a decontamination cycle that employs Ce^{+4} as the oxidizing agent; the reason such low concentrations are effective is that the Ce^{+4} is precipitated quantitatively in the by-product bismuth phosphate precipitate before the reducing agent is added. Dam has studied disposal of the waste uranium solution from the extraction step and finds that the desired alkaline solution could be formed by treatment with Na_2O_2 and Na_2CO_3 or with H_2O_2, NaOH, and Na_2CO_3.

Volatility methods for the extraction and decontamination of plutonium (Brown, Hill, Bohlmann, and Hubble). A dry fluoride cycle incorporating the hydride formation step has been carried out on neutron-bombarded uranium metal and found to work quite well. Three micrograms of plutonium have been fluorinated and found to volatilize and condense in a similar fashion to the tracer quantities investigated earlier. An experiment has been conducted with the Chicago cyclotron to test the existence of a Szilard-Chalmers effect that would separate U^{239} from the bulk of the uranium; very little, if any, concentration of the U^{239} is found. Steel cylinders are demonstrated to be satisfactory for tanking fluorine under pressure in place of the copper, nickel, and Monel cylinders used in the past.

Adsorption methods of extraction and decontamination of plutonium (Beaton, La Chapelle, Sheft, and Turk). Tannic acid, $(NH_4)_2C_2O_4$, and KSCN have been tested as complexing agents to reduce the adsorption of fission products on the resin Catex-284. Tannic acid has been found to be the most promising and has been further investigated by studying its stability under intense radiation and the effect of varying concentrations on relative adsorption of plutonium and fission products. Attempts have also been made to reduce plutonium adsorption by Catex-284 without

affecting its high adsorption of fission activity by the addition of aluminum, lanthanum, and thorium ions and by oxidizing the plutonium with Ag^+ and $K_2S_2O_8$. Experiments have also been carried out which show that silica gel does not adsorb cerium, yttrium, or strontium--the elements eluted with the plutonium from a Catex-284 column.

Lanthanum fluoride separation process. Cefola has carried out microchemical experiments in which plutonium was precipitated with different amounts of lanthanum, insufficient to precipitate all of the plutonium. The experiments indicate that the lanthanum and plutonium precipitate in a constant stoichiometric ratio of 2:1, suggesting the existence of a double salt of the type $PuF_4 \cdot 2LaF_3$.

"Summary of TNX Requests for Work by Metallurgical Laboratory, July 22, 1943" is being issued as follows:

I. HEALTH

Request: Danger of contamination of river water from pile. Status. To establish positive experimental proof of negative radioactive effects on salmon have plan to establish program in cooperation with local research organization, with egg-bearing salmon and fingerlings. Hope to have Army set up program. Will be reviewed week of July 26.

Request: Define danger from neutrino. Status: Letter Allison May 6 states danger unknown. As far as now known there is no danger. Wigner has outlined present knowledge of limits of detectability of the neutrino, CP-742. Will be tested at Clinton.

II. SEPARATION PROCESSES

Request: Method for cleaning radioactive ppt. and other materials from equipment. Status: Work will be postponed until the process is stabilized.

Request: Effect of aluminum and mercury in Bismuth Phosphate Process. Status: Tests at W concentration indicate no effect. Item to be kept open until process more definite.

Request: Specifications for product to be delivered by du Pont. Status: Exact composition undetermined, 99% pure, less than 0.01% lithium, beryllium, boron. Less than 20 millicuries of γ rays per 250 grams. Will have to be revised as more information develops.

Request: Performance of stacks for gas dilution. Status: Data now being analyzed to give formula for combined vertical and horizontal flow with cold gas. In about two weeks expect to study mixing of heated gases.

Request: Development and demonstration by August 1, if possible, of a satisfactory Bismuth Phosphate Process including: (1) microchemical demonstration at W concentration, (2) concentration of the 1% plutonous phosphate in bismuth phosphate to necessary purity, (3) a method of waste disposal. Status: A decision as to what process will be used in a demonstration of reproducibility has again been postponed and may be made July 23. Very low yields have been obtained in the extraction step

carried out in the semiworks. The present procedure gives good yields on microchemical and laboratory scales. Immediate program directed toward establishing cause of difference and obtaining semiworks yields equal to laboratory. Concentration work is under way. No important further work on waste disposal in immediate plans.

The regular Thursday evening meeting of Research Assistants took place in Room 209, Eckhart Hall. Perlman gave an interesting report on Site X--organization, jobs, and problems. He also reviewed the plutonium research going on there and here in Chicago.

Friday, July 23, 1943

Helen and I are at the LaFonda Hotel in Santa Fe. The bustle of Santa Fe is more relaxing for me than the quietude of Valley Ranch.

Cunningham and Ghiorso began seeking the source of possible alpha contamination that could account for the abnormally high counts on their uranium standards. The sample preparations were repeated using a new pipette and running blanks on the reagents used in spreading the uranium on the counting discs. Results still show a specific activity several percent higher than the value reported in the literature by Kovarick and Adams.

A Chemical Engineering Section status report on the Bismuth Phosphate Process is being issued. Seven hot St. Louis slugs (uranium sealed in aluminum cans), it said, have been run in a scale model dissolver without difficulty. Data were presented on semiworks runs No. 36, No. 37, and No. 38. These runs gave plutonium yields in the extraction step of 12%, 40%, and 84%, respectively. The last run (No. 38) is very encouraging and may indicate the semiworks problem may be on the road to solution; this run returned to conditions of extraction used for neutron-bombarded UNH for which better results have been attained.

Last February Compton persuaded Wigner to take a four-week leave of absence, rather than accept his resignation. Wigner has been discontented for some time with du Pont's role in the Project. Today, Compton wrote Wigner a four-page letter on the necessity for cooperation with du Pont. It is a strong letter, but whether it dispels Wigner's growing dissatisfaction is hard to say. Wigner and his associates may feel that Compton is blind to the real reasons for their lack of enthusiasm.

Perlman sent a memo to Doan transmitting information on the functions of our section at Site X, along with the present state of progress of these various activities. Perlman will be in charge of our program there. Five groups were identified:

I. Extraction Research (Knox, Koshland, Turk, Miller, V. R. Cooper, Dreher, Morris, G. Johnson, Dean)

II. Plutonium Isolation (Werner, Fries, Dam)

III. Physical Measurements (English, Jarrett, Butenhoff, Borkowski, East)

IV. U^{233} (Stoughton, Katzin, Steahly, Schulze)

V. Fluorine (Brown, Hill, Bohlmann, Lindner, Anderson, Hubble)

The weekly Bismuth Phosphate progress meeting was attended by representatives of the Chemical and Technical Divisions. We presented data which show that plutonium can be reduced quantitatively by 1.5 M formic acid in 40% UNH and 1.0 N HNO_3 in one hour at 80°-90°C. Results were given of experiments on the carrying of plutonium by bismuth phosphate which show (1) with sufficiently long times of digestion at 70°C very little falling off of the carrying in the presence of 20% UNH occurs at low Bi:Pu ratios when the amount of bismuth is reduced from 2.5 mg/cc to 0.5 mg/cc; (2) Fe^{++} or Fe^{+3} reduces the carrying of plutonium by bismuth phosphate in the presence of 20% UNH if the H_3PO_4 is 0.4 M rather than 0.8 M; (3) solid preformed bismuth phosphate crystals rapidly incorporate tracer amounts of plutonium at room temperature.

"Monthly Report on the Activities of the Chemical Division" (MUC-JF-64) is being issued by the Director, James Franck. The work of Section C-I was summarized as follows:

I. Extraction and Decontamination of Plutonium. Bismuth Phosphate Process (10 men, A priority): Ceric ion and sodium bismuthate are satisfactory oxidizing agents for plutonium. Control analyses of plutonium in process solutions can be made in the presence of high beta activity using a newly developed counter which makes use of a magnet to deflect the beta rays from the ionization chamber. Ferrous ion has been found to be the best reducing agent for plutonium after metal dissolution. Metathesis of the bismuth phosphate permits a significant volume reduction in dissolving the precipitate. Volatility methods (4 men, B priority): A dry fluoride cycle incorporating a hydride formation step has worked well. Attempts to separate U^{239} from the bulk of UF_6 by the Szilard-Chalmers method were not successful. Adsorption methods (5 men, B priority): The organic resin Catex-284 has been further studied; tannic acid reduces the adsorption capacity for some fission products without disturbing plutonium adsorption. Lanthanum Fluoride extraction (1 man, B priority): Microchemical tests indicate the formation of a double salt of the type $PuF_4 \cdot 2LaF_3$.

II. Purification and Chemistry of Plutonium. Reduction of the fluoride and oxide of plutonium by magnesium produces a soft metal resembling platinum (3 men, A priority). At 500°C hydrogen does not give a powdered product as in the case of uranium; the effect of oxygen is slight up to 500°C (1 man, B+A priority). When the lower fluoride of plutonium is heated with CoF_3 and with AgF_2, no volatile higher fluoride is obtained (3 men, A priority). Satisfactory methods have been found for the purification of thorium from added phosphorus involving oxalate, peroxide, and hydroxide precipitations (3 men, A priority). Preliminary studies were made on the use of organic reagents for purification, using thorium as a substitute for plutonium (1 man, B priority). An improved analytical procedure for small amounts of uranium has been

developed (1 man, B priority). Problems pertaining to the Wet Fluoride Process (2 men, B priority), concentration work (2 men, A priority), and U^{233} work (4 men, A priority) are being continued. The instrument group consists of 8 men (A priority). In addition, no report is available for three men associated with the purification program.

Saturday, July 24, 1943

Helen and I are at the LaFonda Hotel in Santa Fe.

Still seeking to eliminate any possible alpha contamination in measuring the specific activity of uranium, Cunningham made up a new uranyl nitrate solution using new equipment. Four samples were prepared and counted by Ghiorso. The specific activity is still high compared with the literature value.

Franck is taking a four-week vacation and will return around August 23. Warren C. Johnson, as Associate Chief of the Chemical Division, will carry on in his place. Franck and Johnson are very much concerned about the possibility of one of the reactors at Site X or Site W catching fire and polluting the air with radioactive debris. So today a letter was sent to Stone in the Health Division summing up the hazards and what steps should be taken to protect the immediate neighborhood against such a catastrophe. This is what they wrote:

"The undersigned believe it worthwhile to reconsider whether the health protective regulations for X and W are sufficient to include cases, in which by accident, sabotage, or attack, a pile catches on fire. Once started the quenching of such a conflagration is a difficult task, and if not successful a tremendous cloud will be formed containing uranium oxide, etc., and especially all the accumulated radioactive products. Under unfavorable weather conditions, steady wind with a velocity below 5 miles per hour, such a cloud would travel many miles without losing its dangerous character by dilution with unpolluted air....

"Consequently, we believe that protective measures should be considered not only for the immediate neighborhood of the pile but also for regions as far as 30 miles away if they lie in zones supposed to be dangerous because of meteorological conditions and the profile of the landscape.

"As already mentioned by one of us in earlier discussions, we believe that the protection of the immediate neighborhood should include the following safety measures:

"Two sheds about 3/4 of a mile long and crossing each other at about right angles at a point very near to the pile should be built. The sheds should have some windows and at suitable distances, doors. A light wooden construction sufficient to prevent a quick exchange of the air with the outside would serve the purpose. Everyone in the immediate neighborhood of the piles should have a combination gas and dust-mask, and should wear overalls....

"For villages or towns at distances of 20-30 miles, the protective measures may consist of a telephone message to the proper authorities or to an air-raid center.... Since danger at a greater distance occurs only if the wind velocity is very small, there will be ample time to warn everybody before the cloud arrives. It is not proposed to warn the population unless it is necessary since the probability of danger is so small."

The Technical Division Weekly Report (Technical Division Memo 105) is being issued. Sutton's Separation Processes Section continued investigating the causes of yield difficulties in the semiworks. Most likely possibilities still appear to be differences in agitation, impurity pickup, or change of oxidation state of some impurity. Semiworks runs No. 37 and No. 38, carried out in Smith's Chemical Engineering Section, were reported. Run No. 37 duplicated run No. 36 except for increased agitation, more rapid heating, and adding most of the H_3PO_4 cold. The yield was 40% compared with 30% for run No. 36. Run No. 38 was an attempt to return to conditions that have previously given good yields when neutron-bombarded uranyl nitrate rather than metal was used as the starting material--18-8 stainless steel was used, and the neutralized solution was aerated several hours to remove dissolved gases. The yield was 86%, the best yet obtained with dissolved metal.

This evening Helen and I had cocktails at the LaFonda Hotel and then dinner at a nearby restaurant with Robert and Kitty Oppenheimer, Joe and Adrienne Kennedy, Charles A. Thomas, and Carroll Hochwalt. Thomas and Hochwalt were visiting Site Y, and the group came down to Santa Fe for the occasion.

Headlines here in the newspaper indicate that the U. S. Army has trapped 45,000 Axis troops in seizing Palermo.

Sunday, July 25, 1943

Helen and I are at the LaFonda Hotel in Santa Fe.

My secretary, Edrey Smith, started her vacation today.

Gofman and Brewer arrived in Chicago from Berkeley to participate in the Monday morning Laboratory Council Information Meeting for chemistry and the evening Chemistry Division meeting.

Monday, July 26, 1943

Helen and I are at the LaFonda Hotel in Santa Fe.

The Chicago Area Office of the Manhattan District approved a change in salary status for Allison, Burton, Hilberry, Ohlinger, Whitaker, and me. My salary has been increased from $5535 per year to $6600, and I am now classified as a Senior Chemist.

John Willard wrote to Professor Tom Jones at Haverford College telling him that his former student, Jonathan Dixon, is doing very well with us and asking if there is any possibility of Jones joining the Met Lab. Willard told Jones he could not elaborate on the nature of the work, but that it is extremely interesting, highly educational, and in a field in which Jones has experience.

"Chemical Research--Production and Extraction of 94, Report for the Month Ending July 20, 1943" (CN-808) was issued, covering the work of the Berkeley group. In experiments supporting the investigation of the use of $Cb_2O_5 \cdot XH_2O$ as the basis for a plutonium extraction process, Hamaker and Sheline determined the carrying power at a Cb:Pu ratio of 45:1 to be above 90%. Pimentel determined the decontamination factor for Cs^{135} in one decontamination cycle of the Sodium Uranyl Acetate Process to be 3000 ± 1600.

There was a Laboratory Council Information Meeting on chemistry at 9:30 a.m., attended by Allison, Boyd, Brewer, Burton, Chipman, Compton, Conant, C. M. Cooper, Coryell, Davies, Doan, Fermi, Garrison, Gofman, Hilberry, Hughes, W. Johnson, Miles, Mulliken, Perlman, Potratz, Rubinson, Stearns, Stone, Sugarman, Wigner, Willard, Smyth later, Spedding later, and Johns even later. The presentation of the work of our Chemistry Section C-I was as follows. Willard discussed reduction methods for plutonium, waste treatment studies, the work of English, et al., on control analysis procedures and the new alpha counter with magnetic deflector. Also cited was Cefola's work indicating possibility of a double salt of lanthanum and plutonium fluorides and the work on adsorption processes using Catex-284. Perlman discussed the plans to obtain U^{233} from the Clinton pile for the study of physical properties of its production process. Willard discussed the discrepancies between semiworks and laboratory results with the Bismuth Phosphate Process and indicated we are now trying to get fundamental information on how $BiPO_4$ and LaF_3 carry--there is evidence that plutonium can get into a $BiPO_4$ crystal already precipitated, indicating a mixed crystal rather than surface adsorption. He also presented the latest thinking on purification methods worked out using thorium to simulate plutonium: an oxalate precipitation, then two peroxide precipitations, followed by precipitating 10% of the thorium as the hydroxide which then is discarded. In connection with the Berkeley program, Gofman described the Cb_2O_5 process for the extraction of plutonium.

In his "Summary of Monthly Progress -- Technical Division" sent to Allison, Cooper summarizes the chemical process development as follows: "On liter scale the Bismuth Phosphate Process chosen by du Pont for W has been further tested with gratifying results. On a semiworks scale results have, however, been poor with the cause not yet definitely established. Recent results have contributed considerably to our understanding and a satisfactory solution is confidently expected in the near future."

A summary of manpower for the Chemical Division shows that my section increased from 50 to 53 persons over the past month. Distribution of manpower was Bismuth Phosphate Process, 9; Wet Fluoride Process, 2;

Adsorption Process, 4; dry methods, 5; concentration, 3; alternate methods, 2; U^{233}, 4; instrument group, 8; purification, 12; and plutonium metal production, 4.

The usual evening seminar of the Chemistry Division was held. Out-of-town visitors included Gofman and Leo Brewer, who spoke about the Berkeley program, and Conant from Washington.

Mussolini has resigned, and Badoglio has become premier in Italy.

Tuesday, July 27, 1943

Helen and I are at the LaFonda Hotel in Santa Fe.

Cunningham thinks that the apparently high specific activity values of the uranium samples might be caused by the fact that UO_2, rather than U_3O_8, was being weighed in the measurements last week. Accordingly, he prepared three more samples using a uranium solution made up by starting with a weighed amount of pure uranium metal obtained from Brown. The samples have now been turned over to Ghiorso for counting.

Norman Hilberry, the Assistant Project Director here at the Lab, sent a letter to Charles A. Thomas. He wrote:

"At Major Peterson's request, I am presenting this outline of the chemistry program of the Metallurgical Project. As here presented, it covers the work in progress at the Metallurgical Laboratory at Chicago, at Iowa State College at Ames, and in the Chemistry Department at the University of California. The program at Clinton will follow the same outline and will be coordinated with the work at the other three institutions as soon as operations start about the first of September.

"The work in chemistry can be broken down into two major classifications which, of course, necessarily overlap. The first of these consists of the fundamental research investigations necessary in providing basic data on the chemical properties of primary materials, of the various product materials, of by-product substances, and of various closely related elements. It also includes investigations into the basic chemical processes involved in our operations. The second and, at the present moment, the much larger classification includes those studies directly concerned with product recovery research and development.

"I. FUNDAMENTAL RESEARCH INVESTIGATIONS

"These we have tentatively broken down into four general classifications including general chemistry, the chemistry of products and by-products, nuclear chemistry, and radiation (photo) chemistry. Due to the pressure upon the Laboratory to answer specific questions in connection with proposed processes, this work is at the present under-emphasized. The personnel involved include approximately 30 men at Chicago and 11 men at Ames. It is planned to increase the emphasis on this phase of the work as rapidly as the present pressure will permit.

"II. PRODUCT RECOVERY RESEARCH AND DEVELOPMENT STUDIES

"A. Separation and Decontamination--The work under this heading has been carried forward both by the Chemistry Division and by the Technical Division with the fundamental chemistry studies largely in the former and those of process and semiworks investigation in the latter.

"Work on the solution of jacketed slugs and the associated chemical problems has been carried through the semiworks stage using the proposed industrial dissolver. Both the basic chemical questions and the technical questions seem to be in hand.

"Three 'wet' chemical separation and decontamination procedures have been developed. Of these both the wet fluoride and the bismuth phosphate have been carried through flowsheet and semiworks stages. While the fundamental chemistry of the wet fluoride process is somewhat better in hand than that of the bismuth phosphate, certain corrosion features of the former have led to the selection of the latter as the process to be pushed for industrial use both at X and at W. The major chemical effort of the Laboratory both in the Chemical and Technical Divisions is now engaged in perfecting this process. A minor amount of work is being done on the wet fluoride process to bring it to completion and further work, also of relatively minor emphasis, is being done on the uranyl acetate procedure to bring it to competion. The equipment for both X and W has been designed with sufficient flexibility to make possible the use not only of the bismuth phosphate but also of the wet fluoride or the uranyl acetate method or some combination of the three of them in case this should seem advisable from actual experience in the use of the bismuth phosphate process.

"In addition to the wet methods, there are certain alternative procedures, particularly for separation process, which are being developed in case the high radiation densities present in the separation stage should interfere in some as yet unforeseen fashion with the operation of the wet separation methods. These include the adsorption method, the dry fluoride method, the solvent extraction method, and the metallurgical method. Of these, the adsorption method is already in the semiworks stage; the dry fluoride method is nearing readiness for semiworks studies; the solvent extraction process is in the process chemistry stage; and considerable ground work has already been laid for possible metallurgical methods.

"There are approximately 50 men working on these problems at Chicago, 20 men at the University of California, and 7 men at Ames.

"B. Concentration, Purification, and Product Metallurgy--Studies are actively under way on methods of purification of the final product. On the analytical procedures necessary for determining tracer quantities of impurities and upon the metallurgy of the product considerable progress has already been made with each of these studies indicating that no impossible obstacles are to be anticipated.

"There are 22 men engaged on this work at Chicago, 16 at the University of California, and 8 at Ames.

"C. Waste Disposal--Problems of waste disposal are shared by the Chemistry Division and the Technical Division with certain studies also being contributed by the Physics and Health Divisions. This work has progressed through the flowsheet and semiworks stage and appears to be in hand. Two outstanding problems are the recovery of the processed uranium in the form of metal and the possible recovery of fission products. Only rudimentary studies have been made in each case.

"There are 5 men in the Technical Division and about two men in the Chemistry Division engaged on these investigations at Chicago.

"D. Instrumentation--This work is again divided between the Chemistry, Technical, and Health Divisions. Instruments have been designed and constructed for most of the major needs. Further development work is progressing on certain instruments.

"There are 8 men in the Chemistry Division and perhaps 4 men in the Technical Division concentrating their efforts on instruments for purely chemical work.

"E. Protection (Health)--This work has involved both the Health and Physics Divisions and appears to be well in hand. It has involved specifications of the shielding required for safe operation, the development of suitable instruments for determining the health hazards associated with the chemical processes and for establishing clinical health routines for the maintenance of the health of the personnel.

"F. Materials (Construction and Process)--The Laboratory is still involved in the problems concerned with the production of primary materials of the necessary purity and for the analyses of such materials. It is also involved in determination of the radiation stability of the materials to be used either for construction purposes or as process ingredients. This phase of the chemistry work is associated primarily with the product production research and development studies rather than with the recovery program. It involves the time of approximately 35 men. The materials investigations for the product recovery research and development studies are concerned with the chemical and physical properties of the materials used, the production and purity, particularly in the case of reagents for the purification program, the radiation stability of such materials, and all of the associated corrosion problems with the enhancement of the last due to radiation.

"There are 11 men at present engaged on this work.

"G. Recovery of Activated Materials--A certain amount of laboratory routine is involved in the actual recovery of product and by-products from the cyclotron-bombarded primary material. This work is largely covered in the above experimental program since the recovery is at present incidental to the experimental work being carried on.

"H. Operations Studies--Particularly in the case of the semiworks investigations, considerable emphasis is being placed on actual performance of the process, of the equipment, of the instruments, and on process control procedures. The men involved are in general those carrying through the actual experimental work.

"I hope that this outline may give you what you wish in terms of an overall picture and of the approximate emphasis which we are placing on the various phases of the work. As the work of the Laboratory progresses we expect, there will be a considerable strengthening both of the fundamental research investigations and a shift from the separations and decontamination to the purification and product metallurgy studies. If you have further questions, I shall be glad to do what I can to obtain the information which you may desire."

At the Laboratory Council Information Meeting on chemistry yesterday it became apparent that the word "hex" has two different meanings, depending on which group is using it. Allison issued a memorandum today saying that we should try to avoid confusion and use the terms "hex," or "process gas," to refer to uranium hexafluoride and the symbol UNH to mean uranyl nitrate hexahydrate.

Today there are reports in the press of an Italian peace bid.

Wednesday, July 28, 1943

Helen and I are at the LaFonda Hotel in Santa Fe.

W. Q. Smith provided Whitaker with extraction and decontamination flowsheets for the recovery of plutonium at Clinton, indicating that work on the plutonium extraction process is continuing and the flowsheets will be revised when the data warrant it.

There was a Laboratory Council Policy Meeting at 9:30 a.m., attended by Allison, Compton, Conant, C. M. Cooper, Doan, Fermi, Greenewalt, Hilberry, Hughes, W. C. Johnson, Miles, Smyth, Spedding, Stearns, Stone, Vernon, Wigner, Hough, and Kilpatrick of Columbia, Thomas and Hochwalt later. Smyth summarized the P-9 (heavy water) pile program and progress to date. He listed several reasons for constructing a heavy-water pile: (1) possible failure of graphite pile, (2) 23 production, (3) greater efficiency, (4) power production, (5) small size, (6) production of radioactive materials, and (7) use as an experimental tool. After a discussion of the type of heavy-water pile to be built (e.g., homogeneous pile, heterogeneous pile, homogeneous slurry pile), Compton asked that each member of the Council be prepared by next meeting to express his opinion on the recommendations for construction of a heavy-water pile. Johnson presented his summary of the work and the manpower distribution in the Chemistry Division indicating that Section C-I will have 53 men next month.

Thursday, July 29, 1943

Helen and I are still at the LaFonda Hotel in Santa Fe, but we are scheduled to leave for home tomorrow.

7/29/43 (cont.)

Ghiorso completed counting Cunningham's uranium samples. The values for specific activity are slightly lower than the last three samples, but Cunningham thinks this may be caused by the presence of oxide in the uranium metal used to prepare the solution. In any event, his conclusion is that it would appear that the Kovarick and Adams value for the specific activity of uranium is low by about 4½%.

In a memo to J. A. Lane, Perlman gave computations showing that a temperature rise of 195°C can be expected in the thorium carbonate cylinders to be placed in the Clinton pile, assuming the pile operating level is a maximum of 40 kw per ton.

W. Q. Smith sent Greenewalt a report, including flowsheets, covering the concentration step of the Bismuth Phosphate Process. He indicated modifications can be expected as research and development continues.

Sutton, Section Chief for Process Development in the Technical Division, sent M. C. Leverett a memorandum on research study projects concerned with the Bismuth Phosphate Process. "On the basis of a critical comparison between the Wet B and $BiPO_4$ process for separation of product," he said, "the du Pont Company chose the latter for use at Hanford. The choice was largely governed by the lesser corrosion attending the use of the $BiPO_4$ process; all other properties appeared essentially equal in both processes. The comparison was based upon the best data available at that time (June 1, 1943) but was far from complete on either process. Consequently it is of the utmost importance that much additional data be obtained on the process chosen, with particular emphasis on those factors which are directly related to operation at both X and W." He then commented on the following steps, the study of which he considers of primary importance: A. extraction step, B. decontamination step, C. concentration, and D. process reproducibility.

Coryell discussed fission products at the meeting of Research Assistants this evening.

There are reports of Germans battling Italians in Italy and of calls for peace from the Italian people.

Friday, July 30, 1943

Helen and I met Robert Wilson and John DeWire (physicist from Cornell University) of Los Alamos before dawn in the Blue Ribbon restaurant in Santa Fe. Bob returned the 200-microgram sample of plutonium-239, which he was guarding with his personal Winchester 32 deer-hunting rifle. We sent the sample to Los Alamos earlier this month so that measurements could be made of the secondary neutron emission from the fission reaction (Wilson obtained positive results but noted that Wahl had experienced difficulty in dissolving the sample in order to prepare it in the form Wilson required). With the sample in my suitcase, Helen and I took a bus from Santa Fe to Lamy, where we boarded a morning Santa Fe train for Chicago.

7/30/43 (cont.)

The regular weekly meeting on the Bismuth Phosphate Process was attended as usual by representatives of the Chemical and Technical Divisions. Today, my section presented data on the reduction of plutonium by formic acid as a function of time; the effect of Ni^{+2}, Fe^{+2}, Fe^{+3}, and NO on carrying of plutonium by bismuth phosphate; the effect of interchanging semiworks and laboratory precipitates and supernatants; the carrying of thorium (as a stand-in for plutonium) by bismuth phosphate.

Perlman lectured on radiochemistry at 7:45 p.m. to the du Pont people who missed the other lectures on this topic.

The paper we bought before catching our train indicates that General Eisenhower has given Italy the Allies' proposed peace terms but that the Italians are trying to "bargain" for less drastic terms.

Saturday, July 31, 1943

Helen and I are enroute by train to Chicago.

Jack D. East, who has been assistant manager at the Southeastern Optical Company of Knoxville, Tennessee, started working as a technician for English.

The Technical Division weekly report (Technical Division Memo No. 108) was issued. The Separation Processes Section reports that there is evidence that agitation in the semiworks runs is not adequate--sampling indicates that 1/2 to 2/3 of the $BiPO_4$ settles to the bottom portion of the tank during agitation. The semiworks in the Chemical Engineering Section carried out extraction cycle runs 39, 40, 41, 42, and 43 using St. Louis neutron-bombarded uranium metal. Both 18-8 vs 25-12 stainless steel tanks and low pressure vs high pressure steam heating have been studied, using conditions which previously gave good yields using neutron-bombarded UNH (no added Fe^{+2}, no HNO_3, 1 N H_2SO_4, 0.8 M H_3PO_4, 2.5 g Bi^{+3}/liter. Hot Strike). In general, the best and most consistent yields (80-99%) were obtained in the three runs carried out in the 18-8 equipment. It is apparent that the semiworks problem is being solved.

Sunday, August 1, 1943

Helen and I arrived in Chicago this morning. We were met by Iz Perlman who drove us to Alexander's Restaurant (on 63rd Street) for breakfast and then home.

Monday, August 2, 1943

I returned to work for the first time since Thursday, June 10, but came home early as I still tire easily.

Edrey Smith also returned to work today at my request, interrupting her vacation for two or three weeks until we clear away some of the office work that has piled up during my absence.

Casimer J. Borkowski started working in Section C-I today as a Junior Chemist. Borkowski received a B.S. in chemistry from the University of Chicago in 1940 and has been employed by The Best Foods, Inc. He will spend a month with the instrument group before transferring to Clinton.

Among the correspondence awaiting my return was a July 23 letter from Friedlander at the University of Idaho. He said that he has been very busy with arrivals--his new daughter at home, and the Army specialized training units that he teaches at the University. He wondered if I had heard through channels that he is now a full-fledged U.S. citizen. He was called to Oakland for his final hearing the early part of this month and, while he was in the Bay Area, went to Berkeley to visit Latimer and his associates on the Project. They seemed keen to have Friedlander join them once he is cleared by Army intelligence. He is interested in returning to Berkeley for personal reasons but is also curious about the situation here and so asked whether or not I wish to make him an offer.

Another pleasant letter awaiting me was from Bertrand Goldschmidt, who wrote on July 20 from Montreal. He said that he would not like July 20 to pass without sending me a few lines in remembrance of exactly a year ago when he joined my team here at the Met Lab. He wrote that he realizes now, when "unfortunate circumstances make the distance between our respective labs nearly infinite, how happy I was in your group and also how interesting and useful was my training under your direction. Here in Montreal," he continued, "we are having a rather cool summer as a happy reward for our winter sufferings. The lab is entirely finished and the work is now progressing rather nicely (though not with the typical Seaborg rapidity)." He completed his letter by saying that he misses all of us and then added a postscript: "What about Sicily--it is grand news." Goldschmidt was referring to the July 10 invasion of Sicily by the Allied forces.

Harrison Brown and I signed the patent petition today for the "Chlorine Process for Separation." We submitted the abstract for this to our attorney in February.

I learned that Cefola is taking two weeks of leave for an operation.

At the evening meeting of the Chemistry Division, Harrison Brown presented information on the dry fluoride process studies planned for Site X. With two 750-ampere cells, he said, it will be possible to process 150 pounds of metallic uranium per day if desired.

The Americans advanced nine miles and seized nine towns in Sicily, according to today's newspapers. The Nazis also are in retreat in the Orel area in the Soviet Union.

Tuesday, August 3, 1943

At home, still recuperating.

Harrison Brown received a letter from Kurt Kraus of the Department of Chemistry, Tulane University, discussing his possible availability for work at the Met Lab beginning in November. Kraus said he would be unable to come at this time because his department believes that one of the men on the staff will be sent to a camp for conscientious objectors, and they are reluctant to let Kraus go because of the increased teaching load caused by the Navy Training Program that commenced there July 1.

In a memorandum to Major A. V. Peterson of the Manhattan District Area Office Compton proposes that the Met Lab construct exponential P-9 (heavy water) piles of the heterogeneous type, exponential piles of the homogeneous type, and a Fermi (low-power) chain-reacting P-9 pile of the heterogeneous type at Argonne Forest. The low-power pile would provide neutron densities intermediate between those available at the Clinton Laboratories and Hanford. In addition, operating experience with the pile would supply information needed in the design of a plutonium-producing pile. Attached to the memorandum is a table showing estimates of various kinds of space needed and the new construction that will be required.

"Separation and Recovery of Plutonium Concentration" (MUC-WQS-75) was issued. This detailed description of the proposed concentration steps following the Bismuth Phosphate Process at Hanford was prepared by the Technical Division. The dissolved plutonium is oxidized, the bismuth separated by precipitation as the phosphate, a by-product precipitation of lanthanum fluoride made, the plutonium reduced and separated by co-precipitation on lanthanum fluoride, then dissolved after metathesis with hydroxide in a small amount of nitric acid.

At the Laboratory Council Information Meeting today, it was the time for the Technical Division to make its monthly report. Those reporting were Bellas, Chipman, Creutz, Johnson, Monk, W. Q. Smith, Sutton, and Wilson. Spedding asked what yields are to be expected in the bismuth phosphate extraction step. Sutton replied 90-95% at present, but 95-100% is possible in the future. Smith remarked that all the trouble is encountered in the extraction step, with subsequent cycles giving full yield.

Sutton revealed that the trouble is caused by the presence of iron which interferes with the carrying by bismuth phosphate. In reply to Compton's question as to the source of the iron, Sutton said it is added to reduce plutonium prior to the precipitation of bismuth phosphate in the extraction step. Hilberry mentioned that ether extraction might be used as an alternative method for the recovery of uranium, and Charles Cooper replied that such a process is being developed.

Doan sent Whitaker a list of 48 names of Chemistry Division employees who will transfer to Clinton Laboratories this year and promised to send corresponding lists soon for the Technical, Health, and Physics Divisions.

Name	Tentative Transfer Date
Anderson, H. H.	September 20
Ballou, Nathan	September 1
Bohlmann, E. G.	August 30
Borkowski, C.	September 13
Boyle, J.	August 23
Brady, E. L.	September 13
Brown, H. S.	August 30
Burgus, W. H.	September 1
Butenhoff, R.	September 8
Cannon, C. V.	August 16
Cooper, V. R.	September 6
Coryell, C. D.	September 13
Covey, E. H.	August 30
Dam, J. R.	September 20
Davies, T. H.	September 13
Day, R. A.	August 23
Dean, F. M.	September 20
Dreher, J. L.	September 20
East, J.	September 6
Elliott, Norman	September 1
English, S. G.	August 30
Fries, B. A.	September 20
Gest, H.	September 13
Glendenin, L. E.	September 13
Hill, O. F.	September 13
Hubble, H. H.	September 13
Hume, D. N.	September 13
Jarrett, A. A.	September 6
Jenks, G.	August 23
Katzin, L. I.	September 13
Knight, J. D.	September 1
Knox, W. J.	August 16
Koshland, D. E.	September 13
Lindner, M.	September 20
Martens, R. I.	September 1
McClinton, L. T.	August 23
Miller, D. R.	September 20
Morris, H.	September 20
Novey, T. B.	September 13

Perlman, I.	August 16
Pomerance, Herbert	September 1
Schover, D.	September 1
Schulze, P. A.	September 6
Shapiro, E.	August 16
Steahly, F. L.	September 20
Stoughton, R. W.	September 6
Waters, Dewey	September 1
Werner, L. B.	August 30

In a memorandum to Charles Cooper, Doan recommends that he form a committee to develop information on alternative separation and decontamination processes. He estimates that it will take six to eight weeks to complete the investigation and proposes that personnel from the Technical Division be assigned to work on it full time. His directions are as follows:

1. List all alternative processes known or believed to be capable of turning out a satisfactorily decontaminated product. Frequently a combination of processes will be required to meet this end, e.g., ether extraction--lanthanum fluoride or bismuth phosphate or sodium acetate, etc.

2. For each process considered, prepare a flowsheet on which is indicated the operations to be performed and the conditions employed.

3. Where equipment of an unusual nature is required, indicate by a sketch and description an arrangement with rough dimensions where possible.

4. Based upon the flowsheets and other sources, prepare a table in which the various process considerations are summarized under at least the following headings. (Here he listed 16 items, an example being: uranium recovery; degree of decontamination; form in which recovered; volume as stored.)

From both the Pacific and Europe come favorable reports for the Allies. U. S. Troops are within three-fourths of a mile of Munda; naval and air drives have opened on Italy; and the Germans say the Soviets have fought their way into Orel.

Wednesday, August 4, 1943

Still at home, recuperating.

Magel sent me a list of the apparatus taken by him to Site B (see Figure 3) near the campus when he and Dallas transferred from my section to J. Chipman's section. He stated that replacement will be made for all items needed by us and asked that I sign internal transfer forms.

XBB 779-8848

Figure 3. Site B. 1943.

The weekly evening meeting of the Council of Section C-I was held with Brown, Cunningham, English, Kirk, Orlemann, Perlman, and Thompson present. Kirk described the difficulties being encountered in making plutonium metal because the plutonium wets the inner surface of the beryllium oxide crucibles and soaks into the walls. This occurs whether barium or magnesium is used as the reducing agent. Attempts are being made to fabricate crucibles that do not give trouble with wetting--we are trying to obtain thoria for this purpose. Perlman announced that Laboratory Council Information meetings are now being held on Tuesdays and each Division will be called upon to report every fourth Tuesday. Thomas, formerly chief chemist at Monsanto, is coordinator of the purification program and will be present at these meetings. He is an adviser to General Groves. Lum, one of his assistants who is getting some background on our work here, says that a purification program at Site Y has not been set up yet. Monsanto may work on the purification problem possibly in a manner similar to the way that du Pont does on the extraction work at Chicago.

Orlemann presented data at the Council meeting showing the sensitivity of some of our present analytical methods as compared with the purity limits; there appears to be a sufficiently sensitive method available for each of the potential impurities. Cunningham discussed the work of his group on purification: Clifford Smith's experiments on vapor pressure are being held up because of the necessity for constructing a high vacuum apparatus; Hagemann is working on methods for determining oxygen; Brody on ether extraction; and Dixon on precipitation methods. Orlemann would like to try recrystallization procedures using such ions as sulfate for purification. Orlemann said he has contacted Westinghouse about a "Precipitron" for purifying and removing dust from the rooms in which the purification work is being done. The estimated size of the equipment needed may be so large that it cannot be installed here. The present status of extraction work was discussed, and it was mentioned that iron seems to have a considerable effect on the carrying power of bismuth phosphate. It was also mentioned that the semiworks is doing experiments with a paddle agitator which seem to result in much better carrying than when a "Lightning" mixer is used.

This afternoon Helen went to a punch party at Betty Compton's.

The front page news today indicates that the Axis line in Sicily has cracked at two points.

The usual Laboratory Council Policy Meeting was held at 10:00 a.m., attended by Compton, C. M. Cooper, Doan, Hilberry, Hughes, Jeffries, W. C. Johnson, A. V. Peterson, Spedding, Stearns, Stone, Vernon, Whitaker, and Wigner. Compton announced that Colonel Nichols is now the District Engineer for the Manhattan District, replacing Colonel Marshall who was called to other work overseas. He also stated that Conant and Tolman are acting as scientific eyes and ears for General Groves. The P-9 Reviewing Committee will be here next week. Finally, Compton explained that Charles A. Thomas is chief chemist in the plutonium purification program and chemical adviser to

Groves. He is the coordinator of purification chemistry for the entire Plutonium Project. Cooper described the progress in the Technical Division program, including data showing the distribution of effort in the Division during July and the plans for August. There was a discussion of the heavy water pile program.

Thursday, August 5, 1943

I returned to work.

The "Metallurgical Laboratory Progress Report for July 1943" (MUC-NH-652) is being issued with the following content. In response to a request by du Pont, the Physics Division has started an experimental determination of the overall temperature coefficient to be expected for the W lattice. This is being determined by inserting a thermally insulated section of the W lattice in the removable core of the Argonne pile, making provision for raising its temperature, and then determining the effect of this temperature change on the reproduction factor of the Argonne pile. An interesting development during the month was the use of thermocouples as neutron density detectors. The theoretical group presented a report on the design of a possible operating pile (P-9) using heavy water as a moderator.

In the plutonium recovery studies, according to the Progress Report, the emphasis has been strongly on the Bismuth Phosphate Process. Semiworks results indicate poor carrying of plutonium in the extraction step. It appears that this is due to the type of agitation used during digestion of the bismuth phosphate precipitate, and it is believed that a resolution of the difficulty is at hand. In the purification and basic plutonium chemistry investigations, the outstanding development of the month has been the production of a compact globule of what appeared to be plutonium metal of a few micrograms in weight. The Site B Laboratory, which is nearby in the University of Chicago neighborhood, is practically complete and about 75 percent of the space is already in active use. In the metallurgical group, the principal work during the month has been the development of a bonding procedure for the aluminum coating of Site W slugs of uranium. The optical section has constructed an overall viewer for use in a shield and design and development work continued on periscopic devices.

The total Laboratory expenses for the month of July are approximately $454,000, and personnel total 810--343 academic and 467 non-academic.

"Chemical Research--Production and Extraction of Plutonium, Report for Period Ending August 1, 1943" (CN-849) was issued. The topics and authors are as follows. Oxidation state of plutonium after the metal dissolving step--Knox. It is confirmed by dissolving neutron-bombarded uranium metal that part of the plutonium is present in the oxidized state, as evidenced by carrying with sodium uranyl acetate.

Reduction by formic acid of the plutonium oxidized in the metal dissolving step--V. Cooper, Gaarder, Halperin, Knox. Further experiments with formic acid show that plutonium can be reduced quantitatively by 1.5 M formic acid in 40% UNH and 1 N HNO_3 in one hour at 80-90°C. The effect of nickel on the carrying power of bismuth phosphate for plutonium--V. Cooper. In the amounts expected in the extraction step due to corrosion of stainless steel equipment, nickel does not appear to have an adverse effect on carrying. Effect of ferrous and ferric ions on the carrying of plutonium by bismuth phosphate--Dreher, Dean, Kroner. Since Fe^{+2} can be used to reduce the plutonium oxidized in the metal dissolving step, experiments have been conducted to determine its effect on carrying. It is found that carrying is inhibited at low concentrations of H_3PO_4 and Bi^{+3}. Similar studies by Dreher on the effect of formic acid on carrying show there is no adverse effect. Carrying of plutonium by bismuth phosphate at Site W concentrations--Miller. In microchemical experiments with plutonium at 25 mg/liter, no appreciable differences in carrying are observed with "single-shot" additions of plutonium as compared with "three-shot" precipitations in either the extraction or decontamination steps. Carrying of thorium by bismuth phosphate--Gaarder. Further experiments using thorium to simulate high concentrations of plutonium (25 mg/l) indicate that a more extensive correlation between plutonium and thorium must be established before further experiments with thorium are justified.

Wigner wrote a three-page reply to Compton's letter of July 23, in which Compton stressed the importance of complete cooperation with du Pont. Wigner stated he believes there has been a misunderstanding, that he was referring only to the heavy water P-9 pile program when he advocated bringing in another contractor rather than du Pont. He assured Compton that, "It will remain our earnest desire to help to the best of our abilities in the solution of any further problems which may be brought to our attention by du Pont."

An organization chart for the Metallurgical Project was issued. It shows Compton as Director, with Associate Directors Hilberry (Research), Munnecke (Administration), and Stone (Health). As for the Metallurgical Laboratory, the Director is Allison, with Smyth as Associate Director. Another chart issued simultaneously gives the organization for Clinton Laboratories. It is shown in Figure 4.

Helen stopped by to see Alice Thompson and Wilma Ghiorso during the day. While we were vacationing in July, the Ghiorsos moved into a picturesque old house at 1219 E. 54th St.

Today the newspaper indicates that the key base of Orel has been evacuated by the German army.

Friday, August 6, 1943

Willard is visiting the University of Wisconsin to interview students about possible employment with us.

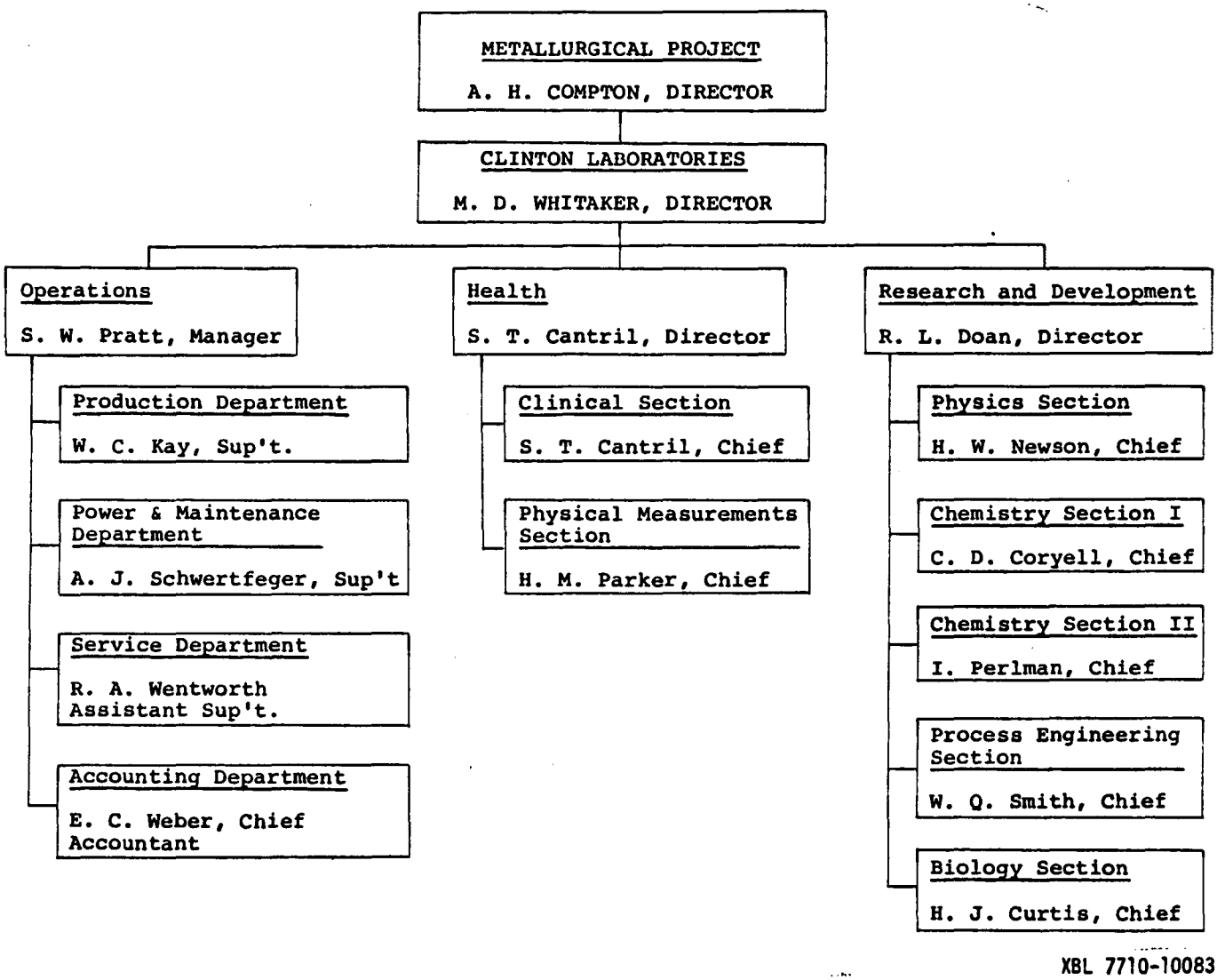

Figure 4. Clinton Laboratories Organizational Chart. August 1943.

8/6/43 (cont.)

Orlemann received a reply from A. J. King of the King Laboratories who indicates they can produce ten to twenty pounds of high purity barium at a price somewhere near $50 per pound. Orlemann needs the barium for investigation as a reductant in experiments on the production of plutonium metal; it has the advantage that there is no limit on its presence as an impurity in the final product because it is too heavy to produce neutrons from reaction with plutonium alpha particles.

The regular meeting on the Bismuth Phosphate Process was attended by Apple, Bellas, Brown, Chambers, Coryell, Frankenberry, Warren Garrison, Graves, Greager, Kay, Maloney, Peery, Perlman, J. H. Peterson, M. D. Peterson, Rubinson, W. Q. Smith, Squires, Struthers, Sutton, Thompson, and Vaughan. It was decided that formic acid, rather than ferrous ion, will be used for the time being as the reducing agent in the extraction step. The amount and type of agitation appears to influence semiworks yields in the extraction step, and the paddle agitator appears to be much better than the "Lightning" mixer for this purpose--further investigations will be made. Preformed bismuth phosphate precipitates do not look promising. Ceric ion does not appear to be too corrosive; a choice will eventually be made between it, bismuthate, and dichromate. For the future it is agreed that Section C-I will conduct the following:

1. Experiments on carrying and crystal structure of bismuth phosphate.
2. Determination of lowest Bi/Pu ratio for good carrying.
3. Effect of pyrophosphates (formed in phosphoric acid) on the process.
4. Investigation of other reducing agents including U^{+4}.
5. Effect of fission product element concentrations at levels higher than those at Site W.

Helen went looking for a new, larger apartment for us without much luck. She then stopped by Alice Thompson's.

U. S. troops are in control in Munda in the South Pacific according to the news today.

Saturday, August 7, 1943

"Chemical Research--U^{233} Production and Extraction, Report for Month Ending August 7, 1943" (CC-887) was issued. Stoughton reported it is now planned that the thorium will be irradiated in the Site X pile in the form of the carbonate, with the extraction to be accomplished by dissolving the thorium carbonate in nitric acid and removing the intermediate Pa^{233} from this solution with manganese dioxide precipitation. The Pa^{233} will then be allowed to stand in a suitable medium until a large portion of it has decayed to U^{233} that will then be extracted and purified. Experiments demonstrate that MnO_2 carries about 90% of the protactinium and less than one percent of tracer quantities of uranium.

8/7/43 (cont.)

The latter finding is important from the standpoint of separation of the Pa^{233} from any natural uranium impurity in the thorium.

The Technical Division Separation Processes Section (Memo 114) reiterates its findings that it appears a difference in type of agitation is responsible for a large portion of the previous discrepancies between laboratory and semiworks extraction yields in the Bismuth Phosphate Process; furthermore, laboratory work indicates that agitation is more critical when Fe^{+2} is present in the solution. The semiworks reports on runs 45, 46, and 47 are identical except for the type of agitation used. Good yields (91% and 95%) are obtained with a flat paddle agitator at slow speed, while the high speed propeller-type agitator gives a yield of only 11%.

Warren Johnson informed Doan that my section will need 25 pounds of irradiated metal from the Argonne pile.

Helen and I had dinner at the home of Spofford and Frodin English (6104 South Woodlawn).

The Soviets are nearing Kharkov in their summer campaign against the Germans.

Sunday, August 8, 1943

Helen and I took the elevated train to Evanston, where we spent the afternoon with Foster and Ann York and with whom we then had dinner. Foster plays a lot of golf. He convinced me that playing golf will help me recuperate from my illness.

Monday, August 9, 1943

The core section of the Argonne pile, which consists of 448 uranium "eggs" weighing 6.5 pounds each, is now being dismantled after 1,000 kilowatt-hours of operation. Of the 300 eggs in the center section--the only ones that contain appreciable amounts of plutonium--24 are being left at Argonne for Spedding and 100 are for disposition at the Met Lab in Chicago. The balance of the eggs from the center section, plus those from the stringers now being emptied, constitute about 2,000 pounds and will be shipped to Clinton Laboratories. There, the engineers will use half for their experiments, while Perlman's group will try to extract plutonium from the other half.

Covey drove the Met Lab station wagon to Argonne Laboratory to pick up the 100 plutonium-rich uranium eggs and deliver them to the West Stands. A portion will go to the semiworks laboratory in the cyclotron building for use by the men in W. Q. Smith's Chemical Engineering Section, and the rest will remain in storage at the West Stands until needed. When Covey reported back to me, he said that Argonne resembles

a concentration camp with its triple fencing and what appears to be more armed guards than workers.

Gofman wrote us from Berkeley asking about the activity of the uranium eggs being discharged. Perlman answered for me saying that we have analyzed one egg, the history of which is not entirely clear, but we assume it to be representative of the other eggs in the core. The 6.5-pound egg that was analyzed yielded 3/4 of a microgram of plutonium per pound -- about the same as for the uranium bombarded with neutrons for 100,000 microampere-hours at the St. Louis cyclotron. Perlman also mentioned the plan for the ton of uranium eggs being shipped to Clinton. He suggested that Hamilton go ahead with his bombardment of uranium in the Berkeley cyclotron as planned, since there might be something radically wrong with our analysis or the egg that we analyzed might be unrepresentative of the ton or so of uranium. Furthermore, it will be of value, no doubt, to have some uranium containing as much 93^{237} as possible; and this should be in uranium around the Berkeley cyclotron.

The British have captured two Nazi strongholds in Sicily.

Tuesday, August 10, 1943

Benjamin C. Bradshaw, who has been on the faculty of Lambuth College, Jackson, Tennessee, started work in Section C-I. He received his Ph.D. in physical chemistry from Harvard in 1932.

Yesterday afternoon and evening Davidson performed volatility experiments with approximately 0.01 microgram of PuF_4 in vacuo. He found appreciable volatility at 700°C but noted that after each partial volatilization he had to heat the sample with HF in order to observe further volatilization.

The chemical engineers began a run in the semiworks to extract some plutonium for us from neutron-bombarded uranium from the Argonne Laboratory using the Lanthanum Fluoride Process.

"Chemical Research--Special Chemistry of Plutonium, Report for Month Ending August 7, 1943" (CK-888) is being issued. The topics and authors are as follows. Technical studies on the production of plutonium metal--Kirk, Rosenfels, Baumbach, and Zvolner. Serious difficulties have been encountered in preparing microgram quantities of plutonium metal. Attempts to reduce plutonium oxide with magnesium or barium in beryllium oxide crucibles results in the wetting of the crucibles and the penetration of the plutonium into its walls. This behavior is sharply different from uranium, indicating the latter is not an adequate "stand-in" for plutonium. Attempts are being made to produce more satisfactory crucibles. An effort to decompose plutonium dioxide by means of heat and vacuum alone resulted in conversion from a yellow-brown color to shiny black, accompanied by shrinkage, suggesting the possibility of a suboxide. A successful technique for mounting microgram quantities of metal in lucite has been developed. Ether extraction of plutonium as a method for purification--

Brody and Orlemann. Experiments show that about 50% of plutonium oxidized with sodium bismuthate can be extracted into the ether phase from a 5 N HNO_3 solution indicating that if satisfactory separation from impurities can be effected, a continuous extractor might be a feasible means of purification. Uranyl nitrate hexahydrate also has been tested as a salting-out agent on the assumption it would have an effect similar to the effect plutonium would have on itself. The fraction of plutonium entering the ether layer was considerably less than the fraction of the uranium entering the ether layer. Possible error in the Kovarik and Adams value for the specific activity of uranium--Cunningham, Ghiorso, and Jaffey. An attempt has been made to determine more accurately alpha counter geometry by using known weights of uranium, the specific activity of which has been determined very precisely by Kovarik and Adams. Counting measurements give values consistently 2-5% higher than expected, even assuming counter geometry to be as high as 50%. It is believed the results indicate an appreciable error in the Kovarik and Adams value.

Reports today indicate that the Soviets are within twelve miles of Kharkov.

Wednesday, August 11, 1943

Willard sent letters to Fred Schuler and Robert Goeckermann, undergraduate students at the University of Wisconsin, telling them of our interest in having them join the Met Lab. He said that he would ask Stearns, our Personnel Director, to send each one an application form. Willard also wrote to his friend Tom Jones at Haverford College in Philadelphia saying he would like very much to talk to him about joining the Met Lab before he makes any other commitments.

Helen and I had dinner at the home of Merlin and Marian Peterson (5811 Dorchester).

Winston Churchill is in Quebec to confer with Mackenzie King, Prime Minister of Canada. Later Churchill will met with Roosevelt.

The regular Wednesday Laboratory Council Policy Meeting was held, starting at 9:30 a.m. and attended by Allison, Compton, C. M. Cooper, Doan, Hilberry, Hughes, W. Johnson, Captain McKinley, A. V. Peterson, Smyth, Spedding, Stearns, Stone, Whitaker, and Wigner. The latest word from Site Y, Compton said, indicates our product will be useful; he was referring to Bob Wilson's observation of secondary neutrons produced in the neutron fission of Pu^{239} as demonstrated with the 200-microgram sample we lent them. He announced that the P-9 (heavy water) Reviewing Committee (Chairman, W. K. Lewis) is here today and will stay for the balance of the week. Smyth presented the P-9 Committee's conclusion how the heavy water pile can best be used. "A. A heterogeneous heavy water moderated pile can be built which can serve both as a prototype for later units to be built on the same pattern and as an experimental and testing unit. In order to serve both purposes, this pile should be designed

for 40,000 kw but run at about 5,000 kw at X. It is probable that such a plant could be ready in less than a year. B. With 50 tons of P-9 a slurry pile probably can be built which will be capable of producing 600 gm/day of 49. It is not probable that such a plant could be ready in less than 1-1/2 years." The plans for the P-9 pile were then discussed by the Council. Compton then went on to say that it is the obligation of each Division Director to see that Site W plans are carefully reviewed and criticisms and suggestions returned to du Pont. A discussion followed as to how to ensure their adequate review. This was the week for review of the Health's Division's program, which was done by Stone. The meeting adjourned at 1:30 p.m.

Thursday, August 12, 1943

Willard wrote to Professor V. W. Meloche at the University of Wisconsin, Madison, informing him that he had been able to see Malm, Manion, Schuler, and Schultz on his visit to the University last Friday and that we have initiated correspondence with Malm, Manion, Webers, Schuler, Christensen, Ames, Dunn, Alexander, Lipton, Goeckermann, Newell, Spector, Lardinois, Huebner, Wilfong, Koppel, Watt, and Tiedke. He also asked for the current addresses of Alvin J. Dirksen, Paul Bente, and Carl Zulke.

Willard also wrote to George W. Watt at the University of Texas saying Watt has been recommended to us as a man who is expert in inorganic chemistry and asking if he would be at all interested in joining our staff, possibly on a leave-of-absence basis.

"Chemical Research--Production and Extraction of Plutonium, Report for Month Ending August 7, 1943" (CN-894) is being issued. The topics and authors are as follows. Volatility methods for the extraction and decontamination of plutonium--Brown, Hill, Bohlmann, Lindner, Anderson, and Hubble. The effect of flow rate and temperature on deposition of PuF_6 in copper tubes has been studied by Hill and Lindner who find that at temperatures below 400°C deposition occurs in a fraction of a second, while at temperatures of about 425°C the PuF_6 is carried through the copper tubing with relatively little deposition. A system for pumping liquid UF_6 has been constructed by Bohlmann and Hubble, and flow rates of 10 feet per second have been attained. Hill has conducted experiments on the volatility of U^{239} in UF_6 bombarded with slow neutrons in the Chicago cyclotron and finds preliminary evidence for a Szilard-Chalmers effect. Hill also has studied the isolation of plutonium from columbic oxide (as obtained in a method being developed at Berkeley) and finds that hydrofluorination followed by fluorination is successful. He also finds that polonium possesses a volatile fluoride. Anderson has prepared volatile fluorides of tellurium and columbium and measured the vapor pressure-temperature curve for TeF_6. Hubble has designed and built a monel Sylphon bellows pump for gaseous fluorine and liquid UF_6. Adsorption methods--Beaton, Dam, La Chapelle, Lincoln, Sheft, and Turk. Amberlite IR-1 and Catex-284 columns have been tested with citric acid-complexed solutions of neutron-bombarded UNH

and show a gamma decontamination of 25-fold. Preliminary efforts have been made to couple the adsorption-extraction cycle to a precipitation process by subjecting the elutriant to a bismuth phosphate precipitation, a lanthanum fluoride cycle, and a sodium-uranyl acetate cycle, respectively. Of these, the sodium-uranyl acetate appears the most promising. An investigation indicates that the use of bismuth phosphate as a selective adsorbent is impractical because of high adsorption of by-product activity and the difficulty of eluting the adsorbed product. Cb_2O_5 powder has been found to be an excellent adsorbent for tracer amounts of plutonium in the absence of uranium; however, neither the adsorption nor elution has as yet been sufficiently selective to afford appreciable decontamination.

I held the regular evening meeting of the Council of Section C-I in my office attended by Perlman, Brown, Cunningham, Willard, and Thompson. I pointed out there will be three groups under Perlman at Site X--English, in charge of electronics and extraction; Brown, in charge of dry methods and also concentration; Stoughton, in charge of the U^{233} problem. There is now great pressure on the plutonium metal production work, and Manning will be transferred from the extraction group to work with Kirk. It is evident that all the groups of Section C-I must have more men. Cunningham will take over Stoughton's U^{233} sample. It is now planned that the pile at Site X will start up on September 25 and by the first of November we should begin to get some production of plutonium from Site X. The concentration phase of our problem needs more attention as we are having trouble with the fluoride method for this purpose. English mentioned a need for work to be done on the mounting of samples for the magnetic counter. He requested Koshland's services, but it was agreed that Koshland was already overworked. I suggested one of Koshland's men might do the job with supervision. The space situation will ease somewhat with the move to Site X, and it is planned to transfer the extraction crew completely to Room 4; so Orlemann can take over the room now occupied by James and Koshland (Room 5). Possibly Kirk and Cunningham can take over Room 1. The latest report on the annex to the New Chemistry Building is that it will be finished on October 20.

At the evening meeting of the chemistry Research Assistants held in Room 209, Eckhart Hall, Katz discussed purification methods.

Roosevelt and Churchill will meet in Canada soon.

Friday, August 13, 1943

Allison asked me to examine drawings of the architectural features prepared by du Pont for Site W buildings 221 and 222 at the chemical extraction plant (200 Area) and to communicate my suggestions and criticisms to Franck, our Division Director. Willard, Thompson, and I have the responsibility in my section of reviewing all blueprints for the 200 Area. Others in my section who may look at them without written permission are Perlman, English, Cunningham, Orlemann, and Brown.

8/13/43 (cont.)

There was a meeting of those from my section who are going to Site X to make plans for the research program.

Compton recommended to W. K. Lewis, Chairman of the P-9 Reviewing Committee, that a low-power (100-250 kw) heavy water pile be approved for construction at Argonne by February 1944. He estimated a cost of $500,000, which includes the building and services, but excludes the heavy water and uranium.

I attended the regular Friday meeting on the Bismuth Phosphate Process with Acken, Borst, Brown, Collins, Davies, Frankenberry, Greenewalt, Leader, Maloney, Peery, J. H. Peterson, M. D. Peterson, W. Q. Smith, Squires, Struthers, Sutton, Thompson, Vaughan, and Willard. My section presented the results of the following studies: Successful reduction of plutonium by U^{+4} following metal dissolving step; optimization of different variables on the carrying of tracer amounts of plutonium by bismuth phosphate (method of addition of Bi^{+3}, agitation, H_3PO_4 concentration, temperature of digestion, $NaNO_3$ and silica concentration); optimization of carrying of plutonium by previously formed bismuth phosphate precipitates; carrying of plutonium by a bismuth phosphate precipitate formed by diluting to 20% UNH from more concentrated solutions; unsatisfactory carrying of thorium by bismuth phosphate. The Chemical Engineering Section of the Technical Division reported on the results of semiworks runs 48, 49, and 50 which all give better than 80% yields in the extraction step with a paddle agitator. Semiworks run 50 is the first using neutron-irradiated metal from the Argonne site. The principal decision made was that reproducibility runs will now begin, taking run 50 as the standard.

Today we received news that the Soviets have reached the suburbs of Kharkov.

Saturday, August 14, 1943

Blanchard, Gofman, and I issued a report (CF-907) "Further Search for Spontaneous Fission in U^{234}." The abstract reads as follows: "We have made a search for spontaneous fission of U^{234} using a sample weighing 13 micrograms. The result, zero counts in 347 hours, indicates that the spontaneous fission rate corresponds to a half-life for this process of greater than some 5×10^{14} years. In fact, this result leads to about a 50% probability that the half-life is greater than 10^{15} years." The sample of U^{234} was isolated by Fontana and co-workers by extracting the intermediate UX_1 from large quantities of UNH (the process we worked out early last year at Berkeley).

C. M. Cooper sent to Whitaker a proposal prepared by W. Q. Smith on development work at Clinton Laboratories on chemical processes recommending that development work be carried out on the Lanthanum Fluoride Process as well as on the Bismuth Phosphate Process because of difficulties of a fundamental nature that have developed on a tracer scale in the latter process. Smith has always favored the Lanthanum Fluoride Process and is

not yet ready to give up on it. He also proposes that a new group be formed under Donald S. Webster to make semiworks studies of the Dry Fluoride Process.

The Technical Division's Weekly Report (Memo 115) states that semiworks runs 48, 49, and 50 on the Bismuth Phosphate Process were made during the week. Yields in the extraction step were 81%, 85%, and 89% respectively. In all cases paddle-type agitations were used. Run 50 used uranium for the first time from the Argonne pile. The uranium was from the center of the pile and contained about one microgram of plutonium per pound of uranium.

General Groves is in Chicago today.

Today was the last day of work at the Met Lab for Harrison Brown, Spof English, Bill Knox, and Iz Perlman, who are transferring to the Clinton Laboratories. Knox and Perlman will report to L. Riordan at the Clinton Labs employment office in Knoxville on August 16. Brown and English are taking two-week vacations and will report there on August 30.

Following the advice of Foster York, this afternoon I played golf with Stan Thompson at Jackson Park--the first time since my arrival in Chicago. Helen, meanwhile, stopped by Alice Thompson's.

Randazzo, historic city of the kings of Lombardy in Sicily, has fallen to U. S. troops.

Sunday, August 15, 1943

As a result of playing golf yesterday, last night I had my first good night's sleep in months.

Stan Thompson and I went out to play golf again today at Jackson Park but had trouble getting on the course because of the crowd of players. Helen, Alice Thompson, and daughter Ruthie met us at the golf course.

Monday, August 16, 1943

The sixth St. Louis cyclotron neutron bombardment of uranium (Chicago VI) ended today with 100,317 microampere-hours. It started on June 19.

"Monthly Report on the Activities of the Chemical Division; James Franck, Director" (MUC-WCJ-58) is being issued. The work of my section is reported as follows.

I. Extraction and Decontamination of Plutonium

a. Bismuth Phosphate Process. A number of factors that might affect the carrying of plutonium by bismuth phosphate have been reinvestigated on the laboratory scale. From these data it appears that the

conditions now used in the semiworks process are well chosen and quite adequate on the basis of present information. The factors studied are the effect of temperature; digestion time; agitation; H_3PO_4 concentration; presence of NO, NO_2, and SiO_2; effect of Fe^{+2} and Fe^{+3}. Some study has been given to the use of preformed bismuth phosphate precipitates in the carrying of plutonium. (10 men, A priority).

b. Volatility Methods of Extraction. The higher fluoride of plutonium deposits rapidly in copper tubes. A pump has been constructed for handling gaseous F_2 and liquid UF_6. It has been found that the latter may be pumped through a 25-foot Monel system at a rate of 10 feet per second. Preliminary evidence has been obtained for a Szilard-Chalmers effect when UF_6 is radiated with fast neutrons. Volatile fluorides of tellurium, columbium, and polonium have been prepared. A dry volatility method has been worked out for isolating plutonium when present with Cb_2O_5. (5 men, B priority).

c. Adsorption Methods. Citric acid is found to be more satisfactory than tannic acid in the formation of complexes with zirconium and columbium and permits the separation of plutonium by ion exchange resins, such as Catex-284 and Amberlite. Columns of these two resins show a gamma decontamination of 25-fold and in the elutriant a 750-fold factor for zirconium and columbium and a 300-fold decontamination of ruthenium. A preliminary effort has been made to couple the adsorption cycle with a precipitation process by treating the nearly gamma-free elutriant with a bismuth phosphate extraction step, a lanthanum fluoride, and a sodium uranyl acetate cycle, respectively. Of these, the sodium uranyl acetate appears to be the most promising, resulting in overall decontamination factors (for adsorption plus one cycle of sodium uranyl acetate) of 1.7×10^3 for beta and 2.6×10^4 for gamma decontamination. Cb_2O_5 is found to be an excellent adsorbent for plutonium, in the absence of uranium, in tracer solution having a pH of about 3.5. (5 men, B priority)

d. Physical Measurements. Since an error in the specific activity of uranium would result in a corresponding change in fission cross sections, the measurement has been repeated using two samples of $UO_3(NO_2)_2 \cdot 6H_2O$ and one of uranium metal. A minimum value of 1,548 alpha particles/min/mg of $U_I + U_{II} + U^{235}$ has been obtained, compared with a value of 1,486 previously reported by Kovarik and Adams. Work is proceeding on the construction of high efficiency BF_3 counters and on two types of ionization chambers for measuring alpha-particle ranges. (2-1/2 men, A priority)

e. U^{233} Production and Extraction. MnO_2 is found to carry less than 1% of tracer quantities of uranium from HNO_3 solutions. The amount of protactinium carried by this precipitate up to weight ratios of protactinium to MnO_2 of 5×10^{-3} is about 90%. The carrying of tracer amounts of Pa^{233} by MnO_2 from 36% $Th(NO_3)_4 \cdot 4H_2O$ or 23% $Al(NO_3)_3 \cdot 9H_2O$ solutions is found to be greater than 90%. Thorium carbonate, when compressed into a biscuit of 1.1" diameter by 1" under a pressure of 15 tons, attains a density of 2.6 gm/cc. (4 men, A priority)

f. Physical Measurements Group. Counters now being assembled permit counting of alpha particles with a geometrical efficiency of about 7% in the presence of 10^9 beta dis/min. Work is continuing on the determination of resolving times of Geiger counters. (5 men, A priority)

II. Purification of Plutonium

a. Analytical Program. Studies are being continued on the determination of carbon in uranium metal by conversion to CO_2 and subsequent measurement of conductivity in aqueous solution. At present, 0.05 microgram of carbon can be satisfactorily determined, and carbon contents of 150 ppm can be easily measured in 1 mg of samples of uranium. Work has been started on the determination of Cl_2 using the familiar o-tolidine procedure. The bromide volatility method for determination of oxygen is also under investigation. (3 men, A priority)

b. Volatility Methods for Purifying Plutonium. The isolation of small amounts of the higher fluoride of plutonium is essential so that its properties may be studied in relation to the purification process. Previous methods that were used to collect the fluoride have been abandoned in favor of a platinum receiver that permits determination of volatility and stability without removal from the apparatus. Experiments with PuF_4 in a vacuum of 10^{-4} mm of Hg have shown that 20% of a 0.1 microgram sample is volatilized in one hour at 700°C. Present work indicates that PuO_2 is completely volatilized, probably as $PuBr_4$, at about 600°C in an atmosphere of HBr. (3 men, A priority)

c. Solvent Extraction Methods. Oxidized plutonium nitrate (tracer amounts) in 5 M HNO_3 is removed to the extent of 20-30% by ether in one hour. The presence of $KMnO_4$ and $K_2Cr_2O_7$ does not increase the amount of extraction. No appreciable amount of reduced plutonium is extracted from acid solution by ether. When an extraction medium of 10% acetyl acetone in chloroform is used, 20% of the reduced plutonium is removed at pH 2 and 94% at pH 4. (2 men, A priority)

d. Physical Measurements. Since an error in the specific activity of uranium would result in a corresponding change in fission cross sections, the measurement has been repeated using two samples of $UO_2(NO_3)_2 \cdot 6H_2O$ and one of uranium metal. A minimum value of 1,548 alpha particles/min/value of 1,486 previously reported by Kovarik and Adams. Work is proceeding on the construction of high efficiency BF_3 counters and on two types of ionization chambers for measuring alpha-particle ranges. (2-1/2 men, A priority)

e. Metal Production. Studies are being continued on the reduction of plutonous salts, the construction of suitable crucibles for this purpose, and on methods of mounting microgram quantities of the metal. (4 men, A priority)

f. Problems in Progress not Covered in Reports. (1) Servicing of instruments and research on special counting problems (2 men, A priority). (2) Spectrographic analysis (1 man, B priority). (3) Recovery of plutonium from residues and extracted material (2 men, A priority). (4) Precipitation reactions for purification (1 man, A priority). (5) Microchemical work for extraction group (1 man, A priority).

Helen and I went to a show in the evening.

The U. S. Army pushed ahead fourteen miles in Sicily while the RAF bombed Milan for the third time in four nights.

Tuesday, August 17, 1943

I attended the monthly Laboratory Council Information Meeting on Chemistry at 9:30 a.m. in Room 202, Eckhart Hall. Others present were Allison, Boyd, Burton, Chipman, Coryell, Doan, Fermi, Foster, Johns, Johnson, Leverett, Lum, Magel, Mulliken, Orlemann, Spedding, Stone, Sugarman, Thomas, W. W. Watson, Wigner, Willard, and later Whitaker and Miles. After Coryell, Boyd, and Burton presented a review of their programs, Willard spoke on extraction and decontamination methods indicating that semiworks results are now approaching laboratory results in yields. He mentioned that studies are now being made on the crystal structure of bismuth phosphate, carrying by externally preformed bismuth phosphate, carrying at low ratios of Bi:Pu, ratio of plutonium precipitated to carrier precipitated. We are putting more emphasis on the Wet Fluoride Process again. On volatility methods, further experiments on fluorination of $UF_4 + PuF_4$ have been made. Brown's group finds that $(Cb_2O_5 + Pu) + HF$ yields CbF_5, leaving plutonium behind. On adsorption methods, citric acid is found to be more satisfactory than tannic acid in complexing zirconium and columbium. Willard also discussed concentration of plutonium following decontamination, U^{233} work, and counting developments. Orlemann discussed the purification step and metal production. He mentioned that Kirk has difficulties when attempting reduction to the metal because the plutonium goes into the walls of the BeO crucibles. He also discussed Cunningham's and Ghiorso's attempt to calibrate counter geometry using the specific activity of uranium as reported in the literature by Kovarik and Adams. Our findings would indicate a counter geometry of 52% which is impossible as 50% is the absolute maximum. I suggested that these new results of Cunningham's could affect the values of fission cross sections. Later in the meeting Fermi asked when the density of plutonium will be known, to which I replied that for some time we have hoped to have results in a few days.

I also attended a meeting at 1:30 p.m. in Room 259, Ryerson Laboratory, on future plans for process development of the Wet Fluoride Process. Also present were Apple, Greager, K. N. Millet, Peery, J. H. Peterson, M. D. Peterson, W. Q. Smith, Squires, Struthers, Sutton, Thompson, and Willard. An outline of the Wet Fluoride Process as it existed June 1 was presented, and it was agreed that future work will include the following:

1. Precipitation of plutonium from a reduced solution of uranium metal without first going through an oxidation step.

2. Elimination of a zirconium phosphate-barium phosphate precipitation from oxidized solution as the first step in the process.

3. Attempt extraction from 20% UNH solution.

4. Study of formic acid, U^{+4}, H_2O_2, As_2O_3, N_2H_4, and Fe^{+2} as reducing agents and $NaBiO_3$ and Ce^{+4} as oxidizing agents.

5. Further work on the coupling procedure involving metathesis and the use of zirconium.

The question of corrosion was discussed, and Smith claimed that the fluoride method is no more corrosive than the Bismuth Phosphate Process. Squires expressed some doubt; he feels that corrosion is still a problem in the Lanthanum Fluoride Process. I asked for a definite written statement as to the actual status of the corrosion situation in the Lanthanum Fluoride Process, because I feel there has been some change in viewpoint here.

The regular evening meeting of the Purification Group of Section C-I was attended by Baumbach, Cefola, Cunningham, Davidson, Fries, Ghiorso, Hagemann, Hindman, Howland, Katz, Katzin, Kirk, Kohman, Manning, Orlemann, Patton, Clifford Smith, Watters, and Willard. Baumbach reported that plutonium metal seems to wet beryllia, zirconia, and thoria crucibles and to disappear into the pores of the crucible. Attempts are being made to glaze crucibles, and the possibility of obtaining a crucible from a natural or synthetic jewel will be investigated. Cunningham favors the return to the method of reduction of plutonium fluoride by sodium in quartz. Orlemann summarized the present status of the analytical program. Gallium oxide (Ga_2O_3) is being used as an aid to spectroscopic analysis. The oxygen analysis is being undertaken by the bromine volatilization method and by treatment of metal with carbon tetrachloride to produce phosgene for detection. A fluoride analysis is being attempted by treatment of the sample with sulfuric acid-boric acid mixture to volatilize the fluoride as boron trifluoride. It now appears that ether does not cause reduction of oxidized plutonium. An equal volume of ether will extract 50% of plutonium from nitric acid solution, but before more extensive work is pursued, the behavior of the impurities will be studied. Plutonium acetylacetonate apparently can be sublimed at 150°C in high vacuum. Cunningham stated that Smith's work on the vapor pressure of UBr_4 is now proceeding satisfactorily.

Helen went downtown with Wilma. In the evening Helen and I went to a show in order that I might relax.

Americans are on the move in both Europe and the Pacific. In Sicily the U. S. Air Force is within fourteen miles of Messina, and in the Pacific U. S. troops have captured Vella Levella Island in the Solomons.

Wednesday, August 18, 1943

Our Chicago VI shipment arrived at 11 p.m. last night. Covey and four other fellows had it unloaded and safely behind lead in North Stands by 11:30 p.m.

I wrote to Goldschmidt thanking him for his letter of July 20, written on the anniversary of his first day with us at the Met Lab in 1942. I expressed the hope that things will work out so that some of us will be able to visit him and learn more about the work of his laboratory. "In any case," I said, "there is nothing to prevent you from paying us a social visit down here, as you have done before." I mentioned that Iz Perlman "has already gone down below," but will be back to spend a week with us early next month.

8/18/43 (cont.)

Orlemann sent a letter over my name to Eastman in Berkeley, telling him we have made arrangements to have the King Laboratories in Syracuse prepare barium metal in high purity and that he can have any amount of the initial order of ten pounds he needs.

In a memo to Warren Johnson I complained of the long periods required for completion of jobs in the shops and that consequently work on the Project is being delayed.

Stan Thompson had lunch with Helen and me in our apartment. We then played golf at Jackson Park as a contribution to my recuperative program. Helen met us and accompanied us during our final holes of play.

Roosevelt has gone to Quebec for talks with Churchill. Italy is being shelled as the Allies have finished their drive on Sicily.

The regular Laboratory Council Policy Meeting took place at 9:30 a.m., attended by Allison, Compton, Doan, Fermi, W. C. Johnson, Leverett, Lum (of Monsanto), Miles, Spedding, Stearns, Stone, Thomas (of Monsanto), Watson (of Yale), Whitaker, and Wigner. After bringing up for discussion several points that arose in yesterday's Chemistry Division Information Meeting, Johnson presented a table showing the distribution of personnel and research problems in the four sections of the Chemistry Division and in the California group. He stated that, of the 58 men in my section, 25 will transfer to Site X between August 15 and September 15. Compton commented that we must carry forward the study of the fundamentals of the Bismuth Phosphate Process at Chicago as well as at Clinton. Doan and Whitaker agreed but said that some good men are needed at Clinton too, indicating that the top men are being left here and the younger men are being taken to Clinton. Compton mentioned that the emphasis on U^{233} is reduced at present because it would be impossible to produce military quantities of it before 1947. According to the present research program on U^{233}, we will investigate the nuclear characteristics of U^{233} and then stop, unless Pu^{239} proves unsatisfactory as bomb material. There was some discussion of the P-9 pile program, and then the meeting was adjourned.

Thursday, August 19, 1943

My secretary, Edrey Smith, began the remainder of her vacation.

L. H. Gevantman is spending some time this month in Section C-I studying volatility methods preparatory to transferring to Site X early next month. Gevantman has a B.Ch.E. from Johns Hopkins University.

Orville Hill wrote to Harrison Brown, who is vacationing in Baltimore before starting at Clinton Laboratories on the 30th; he passed on information on the latest fluorination runs he and Lindner have made.

8/19/43 (cont.)

In the continuing search for carriers for plutonium, yesterday and last night James tested $NH_4UO_2PO_4$ and concluded that it appears that both oxidized and reduced tracer are carried completely by $NH_4UO_2PO_4$.

"California Project, Chemical Research--Chemistry of 94, Report for Month Ending August 14, 1943" (CN-867) is being issued. Continuing work on the columbium oxide extraction process is reported by Pimentel and Stein. Hamaker and Sheline describe their ultramicrochemical investigations on the carrying of plutonium by $Cb_2O_5 \cdot XH_2O$ at a ratio of Cb/Pu = 20/1 from 1.7 M uranyl nitrate and find that two extractions remove 87% of the plutonium.

I attended a meeting on the ether extraction process along with Armstrong, Brody, Cunningham, Gilbert, Kay, Maloney, Orlemann, Patton, Russell, Jack Schubert, and Thompson. Russell, of Boyd's section, presented information on the work of his group on the ether extraction process. In one experiment, about 87% of the oxidized plutonium was extracted in a 3/4-inch diameter packed column 3 feet tall when operating continuously. I agreed to have the microchemists investigate whether the oxidized state of plutonium at high concentrations (such as at Site W levels) would cause oxidation of the ether. I also expressed the opinion that, because the plutonium is extracted in tracer amounts, it would probably extract at higher concentrations.

At the Research Associates meeting in Room 251 of Ryerson Laboratory at 7:45 p.m., the speakers were Herbert E. Metcalf from the Patent Division and Philip Morrison from the Physics Division.

The regular evening meeting of the chemistry Research Assistants was conducted in Room 209, Eckhart Hall. Wayne Marshall of Boyd's section gave a talk on spectroscopy.

Helen spent the afternoon on the Midway with Wilma.

The RAF blasted a German plane radio factory on the Baltic Coast near Stettin. A broadcast was sent from London to the rest of Europe to warn occupied Europe of an impending Allied invasion.

Friday, August 20, 1943

Beaton left on a trip to Madison, Wisconsin, to visit the Research Products Corporation and the University of Wisconsin. The purpose of the visit is to obtain samples and information concerning several new types of adsorbents being manufactured by this company.

I had a big argument with Franck, who still strongly opposes the Bismuth Phosphate Process; he believes that the carrying of plutonium by bismuth phosphate is unreliable.

I wired Gofman in response to his request telling him we have no separated neptunium on hand but are sending him a small amount of one-week-old neutron-bombarded UNH. I said I have arranged for a two-day

exposure of a pound of this material at St. Louis to be shipped directly to him the middle of next week.

Jaffey answered Gofman's letter about the specific activity of uranium for me. He cites the several specific activity values given in the literature (e.g., 1,500 dis/min/mg U in Secret Report A-80, Bureau of Standards, by Curtiss, Stockmann, and Brown) and states our confidence in the validity of the higher value (1,550 dis/min/mg) obtained by Cunningham and Ghiorso, on the basis of their better techniques and the reliability of our counters. He points out that one big advantage we have over previous determinations is the thinness of our samples (0.1 mg/cm^2 as compared with 0.75 mg/cm^2 in the case of the NBS determination). He mentions that the work here is coming out in a report in the very near future.

In a memorandum to Stearns, Warren Johnson commented on the matter of a chemistry course in the University of Chicago Chemistry Department proposed for the coming year. It was felt in his discussions with Section Chiefs of the Chemistry Department that such a course would be very desirable but that the men probably wouldn't take it if there is a large tuition fee. Another question that arose during the discussions was the possibility of the men using Project research for thesis material for higher degrees. This problem has come up several times in the N.D.R.C. Some institutions have not permitted the use of research results obtained from governmental projects for a thesis. On the other hand, other institutions have taken the opposite point of view; and, as a matter of fact, the University of Chicago Chemistry Department has already granted several degrees in which the thesis work was entirely that of a defense project supported by the Government. Johnson wrote that perhaps it would be advisable to consider the problem as a whole when attempting to arrive at a decision as to whether a course should be given.

Allison sent a memorandum to me, as well as to Boyd, H. S. Brown, Burton, Coryell, Doan, Franck, W. C. Johnson, Latimer, J. B. Miles, Pratt, W. Q. Smith, Sutton, and Vernon, announcing that C. M. Cooper has set up a committee in the Technical Division to evaluate the various processes now under study for the recovery of plutonium. This action is in response to a memorandum that Doan wrote to Cooper on the third of this month. Attached was a memorandum from Cooper to Maloney requesting that he take charge of the investigation on essentially full-time basis for the next two months or so; he also assigned Gilbert of Balthis' Process Development Group and Armstrong of Peery's Chemical Engineering Group to assist full time. Cooper's memorandum to Maloney also included Doan's list of the evaluations to be made. Allison asks us to cooperate with Maloney's committee. He says the findings of the committee are internal to the Technical Division itself and will in no sense be automatically considered as the findings of the Project as a whole.

Warren Johnson sent Allison the suggestions and criticisms that arose from a review of Site W, 200 Area, blueprints by Vance Cooper, Ghiorso, and men from other chemistry sections.

8/20/43 (cont.)

Compton wrote Conant commenting on a draft report on the military use of radioactive materials that Conant has written for General Groves. Compton expresses the opinion that there is a real likelihood that the enemy can and will attack us with radioactive weapons within the next six months and refers to the recent announcement of a secret weapon to be released by Germany in November as an invitation for us to reveal what we ourselves have along this line. He goes on to say that we should be ready to "reply in kind" and indicates that we can do so before Christmas if this is considered military strategy. Compton suggests the possibility be brought to the attention of the General Staff.

The regular weekly meeting on the Bismuth Phosphate Process was attended by members of the Chemical and Technical Divisions. My section presented the results of (1) successful reduction of plutonium by U^{+4} after the metal dissolving step, (2) the successful use of Ce^{+4} and unsuccessful use of $NaBiO_3$ as oxidizing agents in the presence of 40% UNH, (3) the effect of HF on carrying and decontamination in the extraction step, (4) the effect of columbium at Site W concentrations on extraction and decontamination cycles, (5) the satisfactory carrying of plutonium by bismuth phosphate at Bi/Pu ratios as low as 20. The Chemical Engineering Section presented information on the solution of uranium metal slugs from the Argonne pile, the first three semiworks reproducibility runs (which show very encouraging results), and agitation studies. It was decided that we have now reached the stage where we can reduce the frequency of future meetings to alternate weeks. Biweekly meetings will start after next week's (August 27) meeting.

Wilma dropped by to see Helen.

John Willard had lunch with Helen and me in our apartment, after which John and I played golf in Jackson Park. In the evening Helen and I went to a movie.

Saturday, August 21, 1943

Alan E. Florin, who has held a fellowship at the Institute for Gas Technology, Chicago, started working in Section C-I. He is one of the few persons on the Project, so to speak, who came in off the street. A friend of his studying chemistry here at the University told him of "an important project" where a couple of Nobel Prize winners are trying to "crack the atom with a cyclotron" and suggested that he should check into the possibility of a job before applying for service in the Air Force. He first went to Ryerson, where he was sent to see Art Snell who is in charge of the cyclotron. Since he never had any experience with a cyclotron, he was referred to our section, which resulted in interviews with Orlemann, English, Willard, and me. We like him and think he will be valuable to our work. He will first work as a Research Assistant with Davidson on volatility methods.

Beaton returned from his visit to the Research Products Corporation and the University of Wisconsin. There he met with Kemper Slidell

and M. J. Shoemaker of Research Products Company and with O. A. Hougen of the Chemical Engineering Department of the University and a graduate student named Foster. Samples of the following adsorbents were brought back by Beaton or are soon to be forwarded by Research Products: Alkalex, R. P. Zeolite, titanium zeolite, titanated silica gel, and silica gels.

Luther Peery of the Technical Division transferred to Clinton Laboratories today.

I received a reply from Allison commenting on my complaint of Wednesday about delays in completion of shop orders. Allison wonders if I have visited the shop since returning from my vacation. He points out that there has been a large expansion of shop facilities and that he believes the shop situation is improving rapidly.

Warren Johnson informed me that Spedding is anxious to obtain drawings of the large fluorine generator to be used at Site X as he wants to set up a similar outfit at Ames.

Willard recently asked George W. Watt of the University of Texas whether he would be interested in joining our staff, and Watt replied affirmatively. Today Willard again wrote Watt, expressed his pleasure that Watt is interested in working with us, and suggested that they meet at the Fort Pitt Hotel in Pittsburgh on September 6 or 7 during the American Chemical Society meeting.

Willard sent another letter to Tom Jones at Haverford College, saying he is looking forward to seeing Jones in Chicago on Sunday, August 29, and inviting him to stay overnight in Willard's apartment as Adelaide and the children are in Wisconsin for a few weeks. He also wrote to Alvin J. Dirksen at Niagara Falls, New York, that V. W. Meloche has recommended him for a job with us as he is trained in the field of inorganic chemistry. Willard asked if Dirksen would be interested in the possibility of joining our staff.

The Technical Division Weekly Report states that during the week three semiworks reproducibility runs (numbers 50, 51, 52) of the Bismuth Phosphate Process were completed observing the following conditions:

<u>Extraction</u>

Neutralization of dissolved U metal solution
with HCOOH
1 N in H_2SO_4
W concentration of fission products
0.8 M in H_3PO_4
W type double paddle agitator with 4 baffles
in tank
Hot reverse strike, 2.5 g Bi^{+3}/l, added over
10 minute period (0.25 g Bi/cc in 1 N HNO_3)
Digest precipitate 2 hours at 90°C; cool
Centrifuge, wash precipitate with
1 N HNO_3-0.3 M H_3PO_4 mixture
Wash once with H_2O

Decontamination Cycle

Dissolve precipitate in 10 N HNO_3
Dilute to 5 N
Add $NaBiO_3$ to 0.01 M
Oxidize at 50°C for 1 hour
Dilute to 1 N HNO_3
Heat to 75°C
Add H_3PO_4 to 0.1 M
Digest precipitate 1 hour
Cool; centrifuge
Wash precipitate as before, dissolve and discard
To effluent add Fe^{+2} to make 0.03 M
Digest for 1 hour at 75°C
Add H_3PO_4 to 0.6 M
Add 2.5 g Bi^{+3}/l (0.15 g/cc in 10 N HNO_3)
Digest 2 hours at 75°C
Cool; centrifuge
Wash as before
Dissolve in 10 N HNO_3 and continue with next cycle

Extraction yields for all three runs are between 91% and 94%. Overall yields through two decontamination cycles are 64%, 82%, and 68% respectively. Decontamination from fission product gamma-ray activity, through two decontamination cycles, by a factor of 1,000 to 1,500 is observed.

I played golf with Willard at Jackson Park after which he had dinner with Helen and me since his wife Adelaide and the children are in Wisconsin.

The Soviets are moving and have smashed deeply into the Ukraine to capture Lebedin, ninety miles northwest of Kharkov.

Sunday, August 22, 1943

Helen and I visited the Field Museum of Natural History with the Baumbachs and Drehers; afterward we took them to dinner at Pete's Steak House (165 N. Dearborn).

Monday, August 23, 1943

John Willard wrote to thank Raymond Keller of the Department of Chemistry, University of Michigan, Ann Arbor, for suggesting Jerome Karle as a possible employee and asked about Keller's own availability.

During July while I was gone, Fries and Werner made a study of the precipitation of plutonium peroxide. Their results indicated satisfactory carrying by uranium peroxide of tracer amounts of plutonium, but they obtained inconsistent results and unsatisfactory carrying at higher concentrations of plutonium. They then began an investigation of the carrying of plutonium by Cb_2O_5. Fries expressed concern today that his

results indicating high carrying of plutonium at very low columbium concentrations and very low Cb/Pu ratios might mean merely that the plutonium is hydrolyzing and going out of solution independent of the columbium present.

C. A. Thomas of Monsanto wrote Allison that he was particularly interested in Orlemann's discussion last Tuesday at the Laboratory Council Information Meeting on Chemistry on the experimental determination of oxygen. Specifically, he asked if Warren Johnson would write up in detail the experiments on (a) the bromine volatility method using uranium as a stand-in; and (b) the reaction of the oxygen-containing uranium with carbon tetrachloride to form phosgene and subsequently analyzing the phosgene and calculating the amount of oxygen in the sample therefrom. He also requested an outline of all the work that has been done here on fluorine analysis.

Stan Thompson and I played golf at Jackson Park in the late afternoon.

The Allies pounded vital rail centers near Naples, and the Soviets have seized thirty more villages in their drive on Kharkov.

Tuesday, August 24, 1943

Edrey Smith returned from vacation.

Baumbach attempted reduction of PuO_2 with magnesium at 1,000°C in a thoria crucible in vacuo. The result is a residue of black flaky pieces held in a matrix of MgO.

Howland has spent some time reworking old residues in an attempt to recover more product. Today he began a study of the hydrolysis of $Pu^{(r)}$ in weakly acid solutions. In a first determination today he finds that in 0.01 N HNO_3 the solubility of $Pu^{(r)}$ is 0.022 g/liter.

I sent a memo to Apple in the Technical Division about setting up a semiworks group for U^{233} work. As background information I say that in 1934 Amaldi, D'Agostino, Rasetti, and Segrè published the first report of production of a 24-minute radioactivity from the bombardment of thorium by neutrons. This was later shown by Gofman, Stoughton, and me (Report A-192 and CC-126, published June 11, 1942) to be the first step of the chain:

$$Th^{232}(n,\gamma)Th^{233} \xrightarrow[\beta]{23.5\ m} Pa^{233} \xrightarrow[\beta]{27.4\ d} U^{233} \xrightarrow[\alpha]{1.2 \times 10^5\ y} .$$

Our report stated that since U^{233} has a slow neutron fission cross section at least as great as that of U^{235}, it becomes important to consider it as a substitute for the fissionable material which forms the basis of the present program.

A chemical program on preparation of U^{233} is now under way in my section, the memo continues, with a staff of four under the leadership of Stoughton, which will be continued under Perlman at Site X. The work has progressed to the point where small-scale production at Site X is feasible. The process depends on isolation of Pa^{233} from bombarded thorium carbonate by successive concentrations on manganese dioxide carrier. After the last concentration step, the material will be allowed to decay for a period of one to several half-lives (one to three months). It is then planned to precipitate the remaining Pa^{233} away from the U^{233} and purify the latter by appropriate means.

It should be pointed out, I write, that the reason for this primary isolation of Pa^{233} is the natural uranium content of the thorium used. Especial care must be taken to purify the Pa^{233} from uranium. The uranium impurity in the thorium carbonate is equal to or larger than the U^{233} to be expected as a product under Site X conditions. If we assume full rated power of the Site X pile, a charge of 65 pounds of $ThOCO_3$, irradiated for ten days, should contain about 4.5 mg of Pa^{233}. If 2 mg of this can be recovered in the final concentration step (very possibly an optimistic figure) and two half-lives allowed for decay, 1.5 mg of U^{233} is available. Assuming 1 mg of this can be isolated in pure form, we can see that 65 pounds of $ThOCO_3$ must be irradiated in the pile for 10 days, Pa^{233} isolated, and two half-lives (8 weeks) of decay allowed before it is possible to isolate 1 mg of U^{233}. These are minimum conditions, as the yield is probably optimistic.

I go on to say that inasmuch as present demands of experimental groups seem to be at least one milligram U^{233} a month, a considerable amount of work is entailed. In addition to the isolation of the product from the irradiated material, there is the labor and time of preparing and treating approximately 800 1-inch pellets of carbonate and filling and sealing 200 aluminum cans for each charge. There is also an important research program on volatility methods, improvements on present methods, and development for operation under other conditions, as at Site W. The manipulations of isolation themselves are greatly complicated by the intense radioactivity of Pa^{233} itself (20 curies per milligram) and, therefore, the necessity for remote control operations. Because of these factors, I tell Apple, the chemical group as constituted at present cannot adequately handle the whole assignment unaided. We believe, however, that a group of four or five persons to handle the semiworks portion of the aluminum can preparation and the extraction procedure should be sufficient to do the job.

In a summary to Franck I give the dates of significant events following the discovery of uranium fission. These milestones are as follows:

<u>About February 1939</u>

Bohr points out that slow neutron fission is due to the isotope U^{235}.

<u>April or May 1939</u>

Proof of the emission of secondary neutrons.

May 27, 1940

Discussion by Louis A. Turner of Princeton in a secret report (Report A5 for the Advisory Committee on Uranium), "Atomic Energy from U^{238}," of the possibility of capture of neutrons by U^{238} and the possibility of running the chain reaction for the main purpose of producing 94^{239}.

December 1940

Discovery of 50-year 94^{238}.

January to May 1941

First sample (0.5 microgram) of 94^{239} isolated and demonstration of fission by slow neutrons in Berkeley by Seaborg, Segrè, Kennedy, and Lawrence (Report A-33, May 29, 1941).

The seventh St. Louis cyclotron neutron bombardment of uranium (Chicago VII) began today. It was taken to St. Louis by the same truck that picked up our Chicago VI bombardment and consists of eight 85-pound batches of uranium metal and 600 pounds of UNH. It is scheduled to run for about a month.

This evening the regular meeting of the Extraction Group of my section took place. The 25 men in attendance included Beaton, Bradshaw, V. R. Cooper, Cunningham, Dreher, Fries, Ghiorso, Hagemann, Hindman, Howland, James, Katz, Katzin, Lindner, Manning, Miller, Morris, Orlemann, Turk, Watters, Werner, and Willard. Beaton discussed recent work on adsorption, including complexing of zirconium and columbium with tannic or citric acids, decontaminating the elutriant from an adsorption extraction step by following with various precipitation cycles, adsorption of plutonium and fission activity by bismuth phosphate, use of columbic oxide as an adsorbent, and the work under way on the new adsorbents recently obtained from the Research Products Corporation. Lindner reported on the results of experiments on the collection of the higher volatile fluoride or fluorides of plutonium in copper tubes which show there might possibly be two higher fluorides of plutonium in the gas. Katzin discussed current plans for U^{233} production at Site X. Werner outlined briefly the history of the concentration problem for plutonium produced at Sites X and W, and Fries discussed in more detail the problems relating to concentration of plutonium following the chemical separation process at Site X including the possible use of peroxide, oxalate, iodate, and hypophosphate precipitations.

Wednesday, August 25, 1943

Willard replied to a letter from Ralph Firminhac in Baraboo, Wisconsin, telling him we may have an opening for him on our staff and suggesting a meeting if Firminhac comes to Chicago. Firminhac's name was suggested to Willard by Professor Al Wilds of the University of Wisconsin.

James and Miller have been studying the use of UP_2O_6 as a carrier for plutonium from HCl solutions of $BiPO_4$ for use as a concentration step. Today they noted that the carrying falls off in acid concentrations greater than 6 N and that higher $BiPO_4$ concentrations also seem to decrease carrying. They plan to extend the experiments to test both the effect of the $BiPO_4$ concentration and the carrying at Site X concentrations of plutonium.

I held the evening meeting of the Council of my section with Willard, Orlemann, Kirk, Ghiorso, and V. R. Cooper. The meeting was concerned primarily with non-technical matters. Because of the lack of housing at Site X, the transfer of some of the men from Chicago will be delayed. The allotment of space in the new building annex and the reallotment of space in the present building were discussed (see Figure 5). The tentative plans call for Beaton and Thompson to transfer to the new annex. Kirk will have Rooms 1, 12, and 13. Cunningham will have Rooms 4, 9, 10, and 11, plus sharing Rooms 5 and 6 with Orlemann, who also will have Rooms 2 and 41. Donlan will have Room 3 and Ghiorso Rooms 7, 8, and 16. My office will remain in Room 15. We also reviewed our stock of microchemical equipment; we now have on hand or on order 15 wide field binocular microscopes, 20 mechanical stages, and 29 micromanipulators.

Nazi troops have taken over Copenhagen despite local protests. The Soviets have gained in their Donetz drive and are racing west from Kharkov, which they have captured.

The weekly Laboratory Council Policy Meeting was held at 9:30 a.m. and was attended by Compton, C. M. Cooper, Doan, Fermi, Franck, Greenewalt, Miles, Munnecke, Spedding, Stone, Whitaker, and Wigner.

Fermi presented an outline of the current physics program. There were five headings: (1) W-Pile Design, under which were listed such topics as heating of graphite, behavior of thermocouples in pile, danger coefficient of coatings, delayed neutrons for test of coating failure, pile behavior after rods are introduced. (2) Materials, e.g., testing of metal. (3) Understanding of Process--fine structure of cell, range of fission neutrons, diffusion in channels, and fast neutron contribution. (4) New Processes, such as cross section of D, neutron age in Be, Be lattice in pile, P-9 pile design, and O^{19} radioactivity. (5) General Nuclear Physics, which includes the topics absorption cross sections, age of neutrons from various sources in graphite and water, chemical identification of delayed neutron emitters, neutron reactions of Pb^{208}, samarium isotope responsible for poisoning and gammas from alpha decay; Service--cyclotron and active metal. At the conclusion of Fermi's presentation, Compton asked if any thought is being given to a general physics program that could result in keeping the U.S.A. well in the lead. Wigner replied that attempts are being made in the Theoretical Physics Section, but the practical needs of the Project have prevented a continuing effort. It was Compton's opinion that a group, even if small, should have such work as its primary objective. Fermi

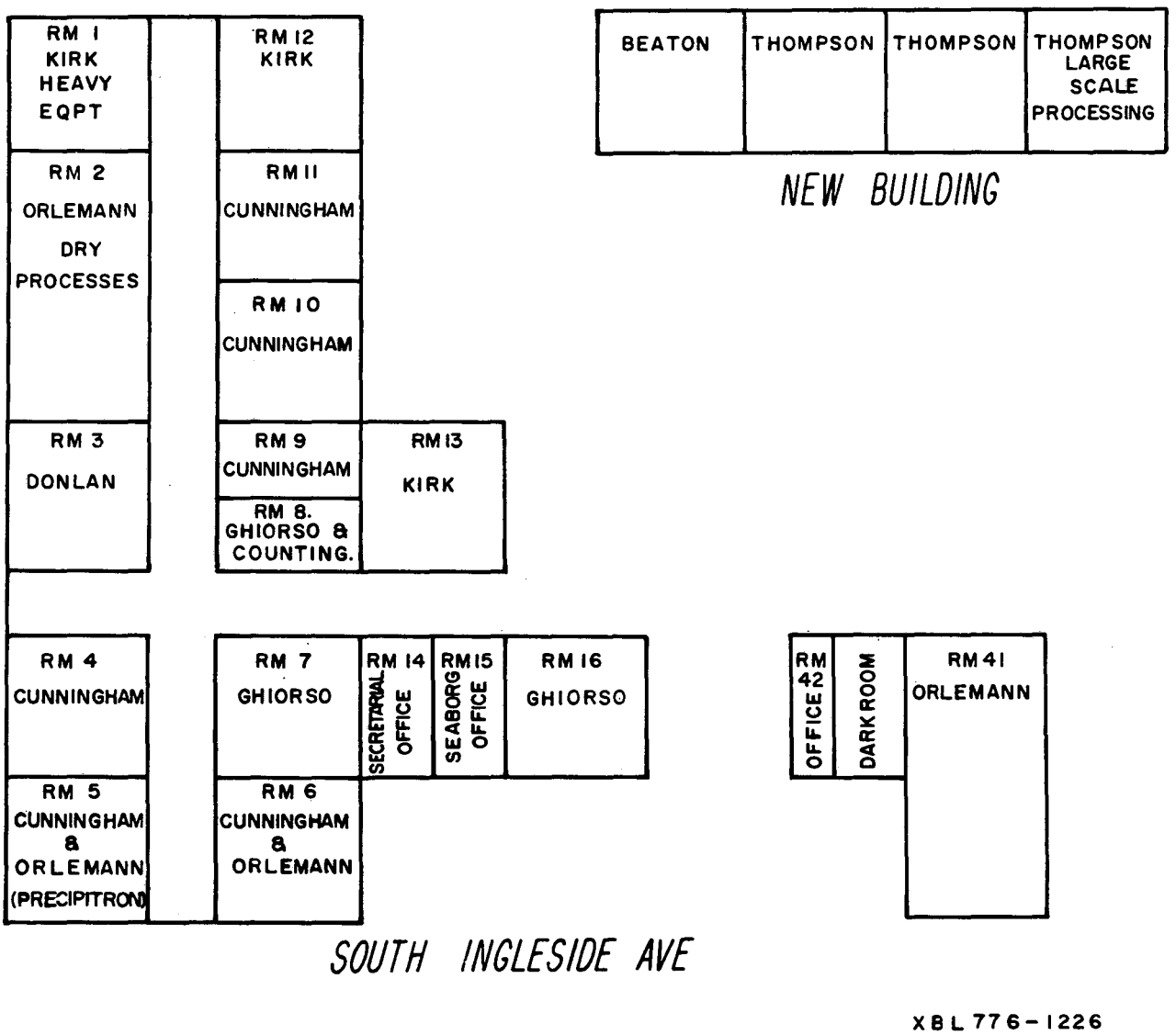

Figure 5. Tentative room assignments, Section C-I, New Chemistry and Chemistry Annex. August 1943.

thought that, if the war is short, we may forego such work; but, if long, then provisions must be made.

Compton then wondered if we are in a position to adequately handle the study of blueprints in connection with the W-pile design. This prompted a heated discussion. Wigner said that, although he has studied all the blueprints, he finds it impossible to obtain from them adequate information to give a clear understanding of a great many points. In addition, no one at the Met Lab has yet enough information to help. It would take the full time of a group of several physicists for weeks or months to do a really proper job. Compton expressed his opinion that the Laboratory is responsible primarily for all questions in which nuclear physics, radioactivity, and radiochemistry are concerned. Matters of structure, strength of materials, and general engineering are du Pont's primary responsibility though, of course, we must call attention to any points that do not look right to us. It is the responsibility of each individual inspecting the blueprints to be as certain as possible that a workable plant is being constructed. When Fermi remarked that a thorough study would necessitate postponing the P-9 pile work because Wigner, Young, and others would be devoting full time to the job, Compton answered that we cannot allow work on alternative processes to interfere with the prompt starting of the Hanford plant. Franck and Greenewalt made some comments and then Whitaker inquired if the blueprints must be reviewed twice, once here and once at Clinton. Compton said that he wants to make sure that the experience at Clinton Laboratories is incorporated into the Hanford design; and, although he is not asking for approval by the Clinton people, he expects a careful review with comments. After a short discussion of cyclotrons and the lack of operators for them, the meeting adjourned with the announcement by Whitaker that the Clinton pile is now not expected to start before October 15, also the completion date for the separations plant.

Thursday, August 26, 1943

Elsy Anderson was added to our secretarial staff today.

I recorded in my notebook the conception that the use of uranous ion precipitated as the oxalate, iodate, fluoride, phosphate, hypophosphate, pyrophosphate, etc., as a carrier for plutonium, should be very useful in the concentration step as well as being useful in the extraction step. I pointed out that the uranous precipitate, containing the carried plutonium, can be dissolved with the aid of an oxidizing agent that oxidizes the uranous uranium to uranyl uranium. From this solution the plutonium can be further concentrated by the precipitation of a second uranous precipitate after the addition of a further smaller amount of uranous ion to the solution, repeating the process until essentially pure plutonium compound is obtained. I also suggested the linking of such a procedure with other carriers, such as sodium uranyl acetate, in a combined concentration procedure.

Baumbach again attempted reduction of 2.5 micrograms of PuO_2 in a ThO_2 crucible with magnesium in vacuo at a temperature of 1,600°C. Black flakes of material of size and shape of the original oxide were formed. They do not react with HCl, and Baumbach concludes they are probably the lower oxide of plutonium.

Cunningham and Werner have started to isolate the 93^{237} from the 80 lbs. of uranium metal bombarded with neutrons at the St. Louis cyclotron from May 14 to July 16, 1943.

The Technical Division issued two detailed reports (totalling 30 pages and three flowsheet charts) of the processes for decontamination (MUC-WQS-95) and waste disposal (MUC-WQS-89) in the Bismuth Phosphate Process as it will be operated at Hanford. The decontamination cycles call for $NaBiO_3$ as the oxidizing agent and Fe^{+2} as the reducing agent.

Friday, August 27, 1943

I was present at the regular Bismuth Phosphate Process meeting attended by representatives of the Technical (Process Chemistry and Chemical Engineering Sections) and Chemistry Divisions. Other present included Leverett, Miles, and Squires. We presented information on the behavior of neptunium in the process. One extraction and one decontamination cycle have been run indicating that the neptunium is partially carried by bismuth phosphate in its reduced state but not in the oxidized state. The status of decontamination was reviewed, and it was agreed that a considerable amount of work is still essential in the decontamination steps before we can be certain of its operation. It was decided that during the next two weeks a concentrated study should be made on the laboratory scale in Sutton's, Coryell's, and my sections. On September 10 a special meeting on the Bismuth Phosphate Process will be held in Chicago and will emphasize the decontamination steps only. Then the next full meeting of the Bismuth Phosphate Group will be held at Clinton Laboratories on September 17.

I received from Howe a summary of the arrangements for canning the pressed thorium carbonate to be irradiated in the Clinton pile. After being filled by Stoughton, the cans are to be sealed by the Aluminum Company of America in New Kensington, Pennsylvania, by welding and spinning down of the ends to simulate the slugs designed for the Clinton pile.

W. Q. Smith sent Whitaker revised extraction, decontamination, and concentration flowsheets for the recovery of plutonium by the Bismuth Phosphate Process at Clinton Laboratories. "These flowsheets," he wrote in the accompanying memo, "were calculated on the basis of carrying out two operations--reduced product precipitation followed by oxidized product precipitation--in each cell. We believe this mode of operation is to be preferred to that of making only one precipitation in each cell since, by increasing the utilization of the equipment, a very considerable increase in flexibility of the plant is obtained."

Saturday, August 28, 1943

Today was the last day at the Met Lab for Ed Bohlmann, Elwin Covey, and Lou Werner of my section, who are transferring to the Clinton Laboratories as of Monday, August 30. Katherine Harvey, English's laboratory assistant, is also transferring at this time. The Chemical Engineering (semiworks) Section of the Technical Division is also going to Clinton now.

In a letter to G. F. Mills of the Chemical Process Company in San Francisco, I apologized for not acknowledging the receipt of resin samples which he sent us in May. I said his samples of R-SH, R-PH_2, and amine resins have been subjected to exchange-adsorption tests but do not appear promising. I indicated we will be pleased to continue testing any other materials he might care to submit.

New Georgia in the Pacific has fallen, and the Japanese have evacuated that island.

Sunday, August 29, 1943

Stan and I played golf at the Evergreen Golf Club (91st Street and Western Avenue), and in the evening Helen and I went to the home of Leonard and Dagmar Dreher (7934 S. Dorchester) for dinner--the Thompsons also were there. After dinner, Stan and I practiced golf shots (putting only) in the living room.

Monday, August 30, 1943

Joe Katz took me out in front of New Chem today to meet his friend and University of Chicago Chemistry Department classmate, Sherman Fried. Fried is a research chemist at Northwestern University. On the basis of Joe's recommendation and my favorable impression, I offered him a job on the spot. Although somewhat flabbergasted at my apparently hasty action, he accepted my offer and told me that he will notify his employer that he is leaving. I am suggesting to Willard that he request that Fried be made a consultant as soon as possible in order to become familiar with our work.

My section issued specifications for the following special reagents wanted in the purification program: copper and graphite electrodes for spectrographic analysis, 30% H_2O_2 solution or urea peroxide, I_2O_5, $H_2C_2O_4 \cdot 2H_2O$, Ga_2O_3 for analytical purposes, and ThO_2 and U_3O_8.

Germany clamped down a military dictatorship on Denmark today, but the Danes succeeded in scuttling most of their fleet to avoid its falling into the hands of the Germans.

There was a special meeting of the Project Council attended by Allison, Compton, C. M. Cooper, Doan, Fermi, Franck, Hilberry, Jeffries, Munnecke, Smyth, Spedding, Stone, W. W. Watson, and Whitaker.

The meeting, which involved morning, afternoon, and evening sessions, considered the Reviewing Committee Report on the P-9 program, organizing for work on the P-9 programs, and allocation of effort on alternate separations processes. Compton announced that the Fermi Argonne heavy water pile is now fully approved. Fermi described the pile as containing 5 tons of D_2O and having a power level in excess of 100 kw. Radiation intensity will be one-tenth that of the piles at Hanford. The pile should be operating by next February. The Reviewing Committee also recommended exponential piles and a U^{235}-enriched pile at Argonne. The latter would be a high radiation level unit especially to study effects of radiation at Hanford radiation levels; it would give a very high level intensity of radiation in a very small space and with a low power output. Compton said that the S-1 Committee will meet September 10, at which time it will be determined if a kilogram of U^{235} is available for use in such a pile.

Franck told the Council that chemistry is really the most critical matter at Clinton and he is especially concerned that the closest possible cooperation between Chicago and Clinton be maintained. In a discussion of alternate process developments, it was agreed that both the Bismuth Phosphate Process and Wet Fluoride Process be tested on a plant scale at Clinton. It was also agreed that at Clinton a group be set up with Boyd and, if possible, Beaton to carry on the Adsorption Process development through operation in the plant, but not to interfere with the development of Bismuth Phosphate and Lanthanum Fluoride Processes. It was further agreed that the possibilities of a Dry Fluoride Process be explored as soon as possible; work was authorized at both Clinton and at Ames. It was decided that for the present the Ether Extraction Process studies should remain at Chicago. Concerning the Acetate Process, it was brought out that the Adsorption Process would automatically require study of the Acetate Process. The Council recommended that California be asked to supply one or more men to help at Clinton on the Adsorption-Acetate Process. Doan suggested obtaining a chemist of considerable stature to supervise the chemistry work for Clinton. He recommended Warren Johnson or some other equally capable man; other names suggested were E. Bright Wilson, Coryell, and Don Yost.

Tuesday, August 31, 1943

Baumbach carried out another reduction of PuF_4 (2 micrograms) with magnesium in a ThO_2 crucible at 1,600°C in vacuo, increasing the reduction time. After the run there was a melted piece of slag near the bottom of the crucible that held several small bright spheres of metal. The piece was counted for alpha particles and gave 2,200 d/m. Evidently plutonium metal was made, but its purity is questionable. The experiment was repeated, using 4 micrograms of PuF_4. One large piece of 2-3 micrograms of plutonium metal was produced, apparently free from slag. Several other very small pieces adhered to the slag.

Willard wrote Nison Hellman of the College of Agriculture, University of Wisconsin, that Hellman's background and experience indicate

we might have a place for him at the Met Lab and telling him he would receive an application blank.

I attended the evening meeting of the Purification Group of my section. I presented an outline of the purification program classifying the reactions under the headings "Precipitation reactions--inorganic," "Precipitation reactions--organic," "Liquid phase distribution--inorganic," "Liquid phase distribution--organic," "Volatilization," and "Fractional distillation of metallic plutonium." I pointed out that the most promising method for plutonium purification appears to be a coupling of peroxide precipitations and a bromine volatilization. Orlemann discussed the analytical program indicating that methods of chemical analysis are particularly needed for the elements carbon, oxygen, fluorine, sulfur and possibly iron and titanium since there is less hope of adapting spectrochemical methods to these elements. Watters' method for carbon is more or less satisfactory; he is at present developing a chloride method based on colorimetry. Another promising analytical method involves the use of the mass spectrograph, such as used by Dempster to detect trace elements in uranium. Marshall and Nachtrieb discussed spectrographic methods and presented a table showing the lower limits of detection. Clifford Smith gave a brief description of the vapor pressure measurements on UBr_4. Patton has been practicing ether extraction methods on the ultramicrochemical scale for the dual purpose of purifying U^{233} and preparing to study the extraction of plutonium by this method. Brody has studied the distribution of tracer plutonium between various nitrate solutions and ether. Howland is studying the hydrolysis of $Pu^{(r)}$ in weakly acid solutions; preliminary indications are that even in 0.03 N HNO_3, $Pu^{(r)}$ is sufficiently hydrolyzed so that its solubility is only of the order of 200 mg/liter. Kirk discussed the metal production program, indicating that the use of magnesium and PuF_4 in thoria crucibles now appears to be the most promising method. Present indications are that plutonium is a fairly volatile metal. Manning is setting up to try atomic hydrogen as a reductant; zinc and manganese have been reduced by this method. Zvolner will study electrochemical methods.

Helen and Wilma took a walk this afternoon.

The Germans have arrested the King and the Cabinet of Denmark and have seized all civil and court functions. On all other fronts the Allies are on the move with bombings in Germany, Italy, and the Pacific.

Wednesday, September 1, 1943

At Clinton Labs the stacking of graphite for the air-cooled pile is beginning today. The job is expected to take about six weeks.

A new clerk, Kathleen Gavin, began working in Section C-I today.

Franck sent a joint letter to Eastman and Latimer in Berkeley, notifying them that the Laboratory Council Information Meetings have been shifted from Monday to Tuesday, and that the next such meeting for the Chemistry Division will be on Tuesday, September 21, followed the next day by a Policy Meeting of the Council. "There has been no change in the Monday evening seminars reserved for scientific contact between the different sections of the Chemistry groups here and related projects," he wrote. "We hope, therefore, that your representatives will plan to attend the Monday evening colloquium, and if they want to, we should like to have them give a talk. In the usual way, they are expected to discuss the highlights of their work. That will mean they will also talk (for about ten minutes) at the Tuesday Information Meeting of the Council."

This afternoon Willard interviewed a chemical engineer, Asher J. Margolis of Chicago, about the possibility of joining our Section C-I. Margolis has an A.B., a B.S., and a Ch.E. (graduate degree) from Columbia University.

A meeting on the Bismuth Phosphate Process is now scheduled for September 17 at Clinton Laboratories. Thompson and I, as well as representatives from the Technical Division, plan to attend. Next week Perlman will return from Clinton to visit us for a few days.

Compton asked Fermi to write down his recollections of the part Fermi played in the early phases of the scientific and technical development that has led to our involvement in the production of plutonium by chain-reacting piles. This is what Fermi had to say:

> "The first news about the discovery of fission was brought to this country early in January 1939 by Professor Bohr who came for a visit to Princeton University. I heard indirectly of that discovery through W. Lamb in January 1939. It occurred to me at that time, that there was a fairly high theoretical probability that neutrons could be emitted in the process of fission in a number adequate to maintain a chain reaction. I expressed this view at a meeting that was held in Washington under the sponsorship of the Carnegie Institution and of George Washington University towards the end of January 1939. At the time, of course, I could not indicate anything more than a theoretical possibility. However, the press somehow got hold of my statements and published a few sensational articles on the subject.
>
> "Soon after, Anderson and I, on one hand, and Szilard and Zinn, on the other hand, started experiments at Columbia University

in order to test whether or not neutrons were actually emitted in the fission process. Both groups concluded that neutrons were actually emitted in rather large numbers. At the insistence of Szilard, we agreed not to rush the publication of these results, but to send letters to the 'Physical Review' with the request that their publication should be delayed until further notice. In the meanwhile, Szilard contacted Joliot who was known to be working on the same subject in Paris and tried to persuade him also to withhold publication of any results that he may have obtained. Joliot did not agree, in part I believe, due to the fact that Roberts of the Department of Terrestrial Magnetism had in the meanwhile announced his discovery of the delayed neutrons. After the publication of Joliot's results, we did not see any reason to further delay the publication of ours.

"Late in February or early in March of 1939, I pointed out to the Navy Department in Washington, the potential implications of the fission phenomena. I explained the situation to a number of men at the Navy Building in Washington in the office of Admiral Hooper. Among those present was Ross Gunn. At this meeting, I indicated the possibilities of a controllable reaction using slow neutrons and of a fast neutron reaction of an explosive character. At the time, of course, our knowledge of the constants involved was so incomplete that the discussion had to be kept in terms of general possibilities.

"In the spring and early summer of 1939, we kept on investigating properties of the neutrons emitted in fission and the most complete results were published by Anderson, Szilard and myself (Phys. Rev. 56, 284, 1939). In this paper, we reached conclusive evidence that the number of neutrons emitted is larger than that of the neutrons absorbed by uranium. Not large enough, however, to maintain a chain reaction with a water moderator.

"In interpreting the experimental results, it occurred to me also at that time, that it was advantageous from the point of view of reducing the resonance loss of neutrons to lump the uranium. This conclusion was not included in the paper, but I mentioned it to Szilard and Anderson. The possibility of using non-hydrogenous moderators occurred quite early to most investigators in the field. It appears reasonable to test first water as the most obvious choice. As second choice in the case that water had to be excluded, graphite appeared the best among the easily available materials.

"So far as I know, this possibility occurred independently to Pegram, Placzek, Szilard and myself. Szilard, Pegram and I exchanged some correspondence on moderation with graphite during the summer. In this correspondence, the desirability to use uranium in a lump form was mentioned. Placzek investigated theoretically some properties of non-hydrogenous moderators.

"During the fall of 1939, Szilard tried to enlist active support on the part of the Government for a research program on chain reactions. He also wrote, in the winter of 1940, a detailed paper on a carbon-uranium chain reacting system which was sent to the 'Physical Review' with instructions to withhold publication.

"Experimental work was in progress at the time at Columbia where Anderson measured the essential characteristics of the resonance bands.

"In the spring of 1940, a few tons of graphite were made available to us by the Briggs Committee, and Anderson and I performed experiments for the determination of the carbon cross section.

"Partly due to the results of these experiments and partly, I believe, due to the worsening of the international outlook, the interest of the Government in the new development grew steadily during the summer and fall of 1940. Under the leadership of Briggs, a separation was made at about that time between the program involving a separation of the uranium isotopes and a program to obtain a chain reaction without isotopic separation. Our group at Columbia was instructed to concentrate efforts on the second possibility. Columbia University was given, successively, two contracts for a total amount of somewhat more than $100,000, and work was started officially under Government sponsorship in November 1940. In March 1941, we obtained the help of the group at Princeton where a program of investigation of the resonance absorption was started with the use of a cyclotron. Anderson and I went to Princeton to perform the first experiments with the group there. The experiments were thereafter continued by the Princeton men. In the summer of 1941, we performed, at Columbia, the first exponential experiments using uranium oxide lumps embedded in graphite.

"During 1941, the possible use of the chain reaction for the production of element 94 was investigated at the University of California. My connections with this development have been the following:

"About Christmas 1940, Segrè was visiting New York and I mentioned to him the desirability of an experimental investigation of the properties of element 94 in view of determining whether or not this element was capable of undergoing fission with slow neutrons. We agreed, at that time, on a plan of investigation that was approved by Lawrence who also happened to be in New York. Work was started soon after in Berkeley by Segrè and Seaborg.

"The possible use of a chain reaction to produce high explosives was recognized very early by all of the workers in this field. The estimates of the amounts of 25 isotope needed for an explosive reaction were, however, considerably in excess of what is now believed to be the correct value. This was due to a wrong value of the cross sections used. For this reason, most of the physicists in the United States did not consider this possibility of immediate practical importance. As far as I know, the emphasis on the development of high explosives and the first approximately correct estimates of the amounts of materials needed came from the British group. I do not believe that the British physicists considered the possibilities of 49, but such possibilities were immediately evident to the American physicists and served to stimulate considerably the interest in the production of this isotope.

"I have tried to put down the facts as I remember them without much reference to the work that is described in the various reports. A résumé of this is contained in a memorandum that I sent to Pegram on June 22, 1942, and of which you have a copy."

The regular evening meeting of the Council of my section was held in my office with Cunningham, Ghiorso, Kirk, Orlemann, Thompson, Willard, and me present. I announced that biweekly meetings are now to be held in cooperation with the newly formed Division of General Physics under W. W. Watson; these meetings will, among other things, concern the design of radiation counters for Site W. The visit of Orlemann to King Laboratories to discuss purification of barium has been approved by Allison. I said we should now give some attention to extraction and decontamination problems associated with the proposed heavy water piles for which the power output per ton of uranium will be 10-50 times greater than that expected at Site W. I suggested that Perlman, Willard, Brown, and Thompson prepare a report on some of the considerations involved in undertaking the problem. The Health Physics Section complains that one of our girl helpers was exposed to excessive amounts of radiation in Covey's room. Cunningham admitted that someone may have been careless about the sample. I recommended that matters such as this should also be mentioned in the individual group meetings. Squires has pointed out there is corrosion difficulty in the lanthanum fluoride crossover for concentration following the Bismuth Phosphate Process, perhaps accentuated by the presence of the As_2O_3 used for reduction. He will be here on Thursday, September 9, to discuss the concentration process. Spedding's proposal to take over the dry fluoride work was not approved by Compton, but he will work in this field at Ames. At the present time Beaton is trying to work out an adsorption process which will give decontamination in addition to extraction.

The extraction and concentration of plutonium from various solutions and residues is proceeding satisfactorily. James, V. R. Cooper, Zvolner, and some of the microchemists are working on this and should be finished with everything but the last ether extracted material and the semiworks solutions this Saturday. We will hold up work on the ether extraction material until we can get some purified lanthanum to work with from Waldo Cohn. The Health Division is interested in getting some concentrated activity from this sample; otherwise we might attempt to work it up by means of some new methods, as suggested by Thompson. Cunningham mentioned that the time required to work up residues is one of our biggest problems. Dixon will handle the ether extraction material because of manpower shortages. I mentioned the extraction group's findings that LaF_3 and CeF_3 can be dissolved by an equivalent amount of Ce^{+4}--this may be the basis for a greatly improved Wet Fluoride Process.

In the metal production work, small chunks of one to two micrograms of plutonium metal are being obtained but removal of slag from them is difficult. We are working on the preparation of larger pieces for density determination. There is evidence that the volatile fluoride of plutonium prepared by fluorinating PuO_2 is actually an oxyfluoride. Equipment for analogous work with Br_2 and HBr is now being set up. Cunningham mentioned that Hindman will also get more evidence on whether an oxyfluoride is formed or not. Brody's promising work on salting out $Pu^{(o)}$ into ether

with NH_4NO_3 was discussed. I mentioned the Site Y success in purifying thorium by extracting the uranium with ether. More work was suggested on the extraction of $Pu^{(o)}$ with other ethers, in addition to the work of Koshland last fall. Thompson mentioned Johnson's finding that the solubility of plutonium phosphate in the extraction step appears to be about 800 mg/l, and that this might account for the sensitivity of plutonium yield in the extraction step. Perhaps the reliability of the method might be improved by reducing the uranium concentration in the extraction step--this could be tried if trouble is encountered at Site X.

Helen and Mary Cooper went downtown this afternoon.

Today's headline, "Russians Gain on 3 Fronts," refers to widely separated areas on the 600-mile Soviet front-- toward Smolensk on the north and toward Mariupol in the south.

Thursday, September 2, 1943

I wrote to the King Laboratories in Syracuse, New York, suggesting that Orlemann visit them on September 8 and 9 to clarify our needs for high purity barium and to determine in what ways we can cooperate with them in solving this problem.

Orville Hill sent a letter to Harrison Brown at Clinton Laboratories bringing him up-to-date on his work. He reports that the Cb_2O_5 method of concentration does not work well in the presence of lanthanum and uranium. He also says that further experiments to see whether there are two volatile fluorides of plutonium resulted in all the activity remaining in the first of the two traps--there seems to be no indication of the formation of two volatile compounds.

At my request Allison wrote Cooksey in Berkeley asking for the plans for the Kirk-Craig type ultramicrobalance. He also asks whether we might borrow the balance already built by the Radiation Laboratory, pending completion of our own balance in the shop.

Last month on the 16th Groves, in a letter to Compton, asked how much of the isotope U^{233} is in existence. Today Compton replied, "I can say with certainty that until material is produced at X, there will be only several micrograms of 23 available; 23 does not occur naturally and to date the total amount, produced by cyclotron bombardment of thorium, is at most 15 micrograms. This 23 is not separated from the thorium as only tracer amounts (hundredths of micrograms) have been used in preliminary development of chemical methods of separation for use at X. A few micrograms could probably be separated but this would be an upper limit."

Helen took a walk with Wilma.

The newspaper today summarizes the war news from many fronts. The Soviets have advanced in the Donetz basin killing or capturing more than 40,000 Nazis. An RAF assault has razed large sections of Berlin. The

U. S. Navy has raided Manus in the Solomon Islands, 1200 miles from Tokyo. The Allies are closing on Salamaua on New Guinea from two sides. And in Italy the U. S. Air Force has raided Pisa.

Friday, September 3, 1943

Manning replied to his friend, Michael J. Wolf in Columbus, Georgia, transmitting employment application forms. Wolf has a Ph.D. in plant physiology and is presently employed as a chemist in the Corps of Engineers at Fort Benning.

Franck sent a memo to Nickson in the Health Division, explaining the safety measures that we are observing to prevent another overexposure to radiation such as occurred to the dishwasher in our section last month.

The Metallurgical Laboratory Progress Report (MUC-NH-653) for August 1943 was issued. The summary of the Chemistry Division's work includes the following. The Separation Group has placed its major emphasis on continued study of the Bismuth Phosphate Process, the data indicating that the semiworks flowsheet is in agreement with the best laboratory experience. Work was started during the month on concentration procedures for Site X and Site W. This program will continue. Work on the purification and basic chemistry of plutonium has been continued, including studies of both volatility and solvent extraction methods. Further work has been done on metal production. These studies will continue. Work has continued at low priority on the Adsorption-Uranyl Acetate Process. With regard to semiworks operations, the report states that product recovery has been satisfactory since adoption of the new agitation system. It also states that the entire semiworks group is being transferred to Clinton. Total expense for the month of August is $421,966. Total personnel on the Project on August 31 is 838 (365 academic).

"Chemical Research--Production and Extraction of 94. Report for Period Ending September 1, 1943" (CN-914) was issued. The following research was covered. Concentration Methods. Fries and Werner reviewed possible methods for separating the decontaminated plutonium from carrier material and its precipitation as a pure compound, under the conditions expected at both Site X and Site W. With regard to methods for changing from bismuth phosphate to lanthanum fluoride carrier as a means of effecting a volume reduction, they note that experiments are now in progress to determine which of three methods is the most desirable: (1) solution of bismuth phosphate and plutonium phosphate in HCl or HNO_3; (2) precipitation of LaF_3 in the final decontamination cycle; (3) metathesis of bismuth phosphate and plutonium phosphate. Concentration by the uranium peroxide-lanthanum fluoride method was studied by Fries and Werner, who carried out experiments on the precipitation of plutonium peroxide in the pure form and with varying amounts of uranium peroxide. The results indicate that uranium peroxide satisfactorily carries tracer quantities of plutonium. Microscale experiments indicate that plutonium forms an insoluble peroxide (solubility 1 mg/liter) at pH 1-2 and that precipitation is not made more complete by co-precipitating with up to 1,000 times

as much uranium. Preliminary studies show that a Cb_2O_5 precipitation might be successfully coupled with a lanthanum precipitation. James and Miller investigated uranous hypophosphate as a carrier for reduced plutonium and find it shows good promise in achieving volume reduction from bismuth phosphate solution; other experiments indicate the method might also be used for an extraction procedure. Dam and Fries investigated the carrying of plutonium by uranous oxalate and find that it, too, shows promise as a method of concentration. Other carriers for concentration investigated by James are $NH_4UO_2PO_4$, $U_3(AsO_4)_4$, $Bi_2(C_2O_4)_3$, $Th(C_2O_4)_2$, and ThP_2O_6. Preliminary Work on New Extraction Methods. Clifford did experiments which indicate that uranous oxalate may be used to carry reduced plutonium from 20% UNH solutions. James and Thompson made a survey of a number of compounds which might be expected to carry plutonium. Bismuth arsenate is very similar to bismuth phosphate as a carrier for plutonium. Bismuth oxalate was also tested with satisfactory results. Other compounds considered were ThP_2O_6, $NH_4UO_2PO_4$, and $U_3(AsO_4)_4$.

The Vance Coopers and Roy Beatons came to dinner at our apartment this evening.

Top war news today states that Allied troops have invaded Italy from Sicily!

Saturday, September 4, 1943

This was the last day at the Met Lab for Jack East, Robert Butenhoff, Orville Hill, and Al Jarrett, who are transferring to the Clinton Laboratories. East and Butenhoff are to report to Knoxville on Monday, September 6; Hill and Jarrett are first taking vacations and will arrive in Knoxville on Monday, September 13, and Monday, September 20, respectively.

I received a copy of a September 3 memo from Stearns to principal staff members about pirating of personnel from other projects. He cited a letter from OSRD stating that no individual engaged elsewhere on an OSRD project shall be approached regarding a change of employment unless and until the approval of his then employer has been obtained.

In a memo to Franck, I urged the creation of a group under A. J. Dempster to work on mass spectrographic methods of plutonium analysis as a means of meeting some of the very difficult analytical problems that arise in connection with the purification program.

Willard indicated to Alvin J. Dirksen in Niagara Falls that we are pleased to learn from his letter of August 30 that he is interested in joining the staff of the Met Lab. Willard enclosed application forms.

Miller is continuing to check variables in the carrying of plutonium by UP_2O_6 for possible use in a concentration process that would be operable in the presence of Fe^{+3}. He dissolves the $BiPO_4$ in a mixture of HNO_3 and HCl. In experiments yesterday and today he found that at constant Fe^{+3} concentrations (0.05 and 0.01 M), increasing the $BiPO_4$ concentration decreases the carrying of the plutonium.

Helen spent the afternoon at Wilma's.

9/4/43 (cont.)

Allied troops under General Montgomery have consolidated their bridgehead and have advanced as much as ten miles into Italy.

Sunday, September 5, 1943

Today I played golf, and later Frances Chilson visited us. In the evening Helen and I went to a party in the apartment shared by Roy and Peg Beaton and John and Ellen Swartout (6850 South Shore Drive).

Willard left by train for Pittsburgh today to attend the American Chemical Society meeting and to interview prospective staff members for Section C-I.

Monday, September 6, 1943

Perlman is visiting from Clinton Labs. We had a long talk about his proposed research program there and started preparing a report on it for Doan.

A new Division of General Physics has been established, effective today. It is headed by William W. Watson, a physics professor from Yale University. Section chiefs selected so far are Dempster, Snell, Wilson, Wollan, and Zachariasen. Zachariasen has set up a section with excellent facilities for x-ray crystallographic investigations. Dempster's section will specialize in mass spectrometry (as hoped for in my memo to Franck on Saturday) as soon as he can find the necessary manpower. Fermi's division is now called the "Nuclear Physics Division."

Helen and I played golf in Jackson Park in the afternoon.

Perlman ate dinner with us in our apartment. Afterwards he and I attended the 7:45 p.m. seminar of the Chemistry Division in Ryerson Laboratory.

Again the front page is filled with war news. The Allies have won forty towns in Italy. Stalino in the Donetz basin is reported in flames as the Soviets are closing in.

Tuesday, September 7, 1943

Today's new employee is Irma Saxton, a secretary, who will handle Willard's recruitment correspondence in addition to other duties.

Orlemann left on his trip to Syracuse to visit the King Laboratories on September 8 and 9 for the discussions concerning the preparation of high purity barium needed as a reducing agent in plutonium metal preparation.

Latimer wired me that he plans to stop in Chicago when returning from Washington and asked that I make a reservation at the Palmer House for the night of September 15, which I did.

Helen and Wilma went to the University of Chicago Billings Hospital to visit Frances Burton, who is very ill.

Perlman again joined us for dinner.

American paratroopers have landed behind Lal in New Guinea to complete the encirclement of 20,000 Japanese in the Lal-Salamaua sector.

Wednesday, September 8, 1943

Willard returned from his trip to Pittsburgh. Among the prospective staff members he interviewed were Lawrence B. Magnusson, George W. Watt, and Donald G. Pye. Today he sent a letter over my name to Magnusson, in care of the American Chemical Society Employment Clearing House, indicating that as a result of their talk on Monday in Pittsburgh we are very much interested in having him join the Met Lab and asking that he send us his home address and the completed application blank. He also wrote to Frederic Schuler and Robert Goeckermann at the University of Wisconsin, saying he will be in Madison on September 11 and would like to talk to them about joining the staff of the Met Lab.

This was the last day at the Met Lab for Elton Turk, who is transferring to Clinton Laboratories. After four days of vacation he is due to report to the Clinton Laboratories office in Knoxville on Tuesday, September 14.

Lyle Jensen who recently received a Ph.D. degree from the University of Washington started work in Section C-I today.

In a letter to R. C. Tolman (Scientific Advisor to Groves and a Vice Chairman of the NDRC) at the National Academy of Sciences in Washington, Compton asked if he could arrange the release of Darrell W. Osborne from his work with the NDRC in order to have him join my section where he is urgently needed.

I sent Doan the write-up he requested; it is 17 pages long and incorporates Perlman's and my ideas concerning a program for Perlman's chemistry section at Clinton Laboratories and a suggested organization for the staff there. A summary of this report (MUC-GTS-215) is as follows:

1. A Bismuth Phosphate and Wet Fluoride Group, under English, to work on process development and do troubleshooting for the extraction plant and semiworks plant (staff: V. R. Cooper, Dean, Dreher, Knox, Koshland, Miller, Morris, Turk).

2. An Instrument and Physical Measurements Group, also under English, to provide and maintain specialized and routine radiation detection instruments (staff: Borkowski, Butenhoff, East, Jarrett).

3. A Concentration Group under Brown, to isolate, in relatively pure form, the plutonium from the carrier material as turned out by the semiworks and extraction plants (staff: Covey, Dam, Fries, Werner, plus one or two more).

4. A Dry Processes Group, also under Brown, to develop a dry process using anhydrous HF and fluorine as an alternate to the wet process methods currently emphasized (staff: Anderson, Bohlmann, Gevantman, Hill, Lindner, plus one or two more).

5. A U^{233} Production Group, under Stoughton, to obtain milligram amounts of pure U^{233} for determining physical constants and to develop extraction methods for removing uranium from thorium and protactinium from thorium and uranium (staff: Katzin, Schulze, Steahly, plus one or two more).

In response to a request from Charles Thomas of the Monsanto Chemical Works in Dayton and coordinator of the plutonium purification program, Franck transmitted to him Orlemann's description of (a) the bromine volatility method for determining oxygen in plutonium metal, (b) the determination of oxygen by converting the oxide to phosgene using CCl_4, and (c) the determination of chlorine by a colorimetric method using o-tolidine. Concerning the bromine volatility method, Orlemann cited the work of Clifford Smith and Cunningham which shows that plutonium metal plus gaseous bromine at 210°C gives about 50% conversion to a volatile bromide.

The regular evening meeting of the Council of Section C-I was attended by V. R. Cooper, Ghiorso, Kirk, Perlman (still here on his visit from Oak Ridge), Thompson, and Willard. Kirk mentioned the medical staff has complained that Hindman is working with too much radiation and that his hands may be affected. Perlman mentioned that no one on the Project has been hurt by radiation yet but that we should be more concerned about injury to the hands than any other possible injury. It was mentioned that Nickson is interested in setting up equipment for checking contamination on hands. Perlman asked that we make it a practice to send him pertinent memoranda on our activities; otherwise his group will have to go half a mile to another building where reports are received. He reported that English has some counters set up and working at Clinton Labs, that the laboratories there are generally good, that the concrete has been poured for the extraction plant, that the three 200-foot stacks are finished, and that W. Q. Smith's group is now operating the semiworks at its new location.

I took the evening off. Helen and I went out to dinner, followed by a movie.

The Laboratory Council Policy group met as usual at 2:00 p.m. in Room 209, Eckhart Hall; it was attended by Allison, Compton, C. M. Cooper, Doan, Fermi, Franck, Hilberry, Hughes, Miles, Spedding, Stone, Watson, Whitaker, and Wigner. Cooper presented the Technical Division Program, which consisted of a long outline showing the distribution of effort in his Division during the month. There was a discussion of better control on the length of Information Meetings; and it was decided that, beginning October 1, there will be combined meetings devoted to Health and Physics during the first week of the month and combined meetings on Chemistry and Technology during the third week of the month. Information Meetings will be on Tuesdays and Policy Meetings on Wednesdays. The change in name from Laboratory Council to Project Council is to be effective October 1.

Thursday, September 9, 1943

I stayed home this morning because of a cold.

One of our secretaries, Marie Garre, terminated today.

English is visiting here from Site X.

Willard wrote Professor Meloche of the University of Wisconsin explaining that he will be in Madison on Saturday and so can interview any qualified students Meloche has in mind, in addition to Ames, Christensen, Goeckermann, Hellman, Malm, Newell, Schuler, and Sedlet, who already know of his visit. Willard also explained that we are exploring with the University of Wisconsin the possibility of having some of the February degree men use their work at the Met Lab to replace their normal last semester's work at the University.

I attended a meeting in the afternoon to discuss various procedures for concentration of plutonium which might be used at Site X and adapted for use at Site W. Other participants were M. Acken, Balthis, English, O. H. Greager, D. Miller, Perlman, D. M. Ritter, Squires, Sutton, and Thompson. I presented a general classification of the concentration procedures which have been investigated:

1. Those involving a change in oxidation number of plutonium, e.g., LaF_3 and $NaUO_2Ac_3$.
2. Those involving a change in oxidation number of the carrier, e.g., U^{+4} salts ($C_2O_4^{-2}$ and $P_2O_6^{-4}$).
3. Those in which the plutonium is alternately carried by two different carriers, e.g., Cb_2O_5-LaF_3 and uranium peroxide-LaF_3.

I mentioned the advantage of procedures falling in classification #2 above is in reducing both volume and amount of carrier without oxidation and reduction of the plutonium. The UP_2O_6 and $U(C_2O_4)_2$ procedures were described. I pointed out some of the differences in the concentration procedures at Site X and Site W: At Site W, there will be a factor of

100 less to be accomplished in concentration, and it will be possible to precipitate a pure plutonium compound earlier than at Site X. At Site W it may be necessary to decontaminate by an additional factor of 100 before concentration; Thompson suggested this could be accomplished by a combination of another bismuth phosphate cycle and the concentration procedure itself, each of which could give a factor of 10. A sodium uranyl acetate cycle might also be used; it provides good concentration in addition to decontamination. Space and equipment necessary for concentration work at Site W were discussed before we adjourned.

Compton has now established two more committees, one to allocate the distribution of activated uranium metal from Argonne and Clinton Labs, composed of Hilberry (Chairman), Allison, and Whitaker; the other committee is to consider a course of instruction that can be given to Met Lab staff members through the University and to formulate a policy regarding the substitution of Project research for thesis research, and this committee is composed of Stearns (Chairman), Warren Johnson, and W. W. Watson.

The U. S. Army is in Naples! But German forces have taken over Milan and other large cities in northern Italy.

Friday, September 10, 1943

Paul R. Fields, who comes from the Tennessee Valley Authority, Alabama Site, started work today. I talked with him and gave him some general reports to read. Fields mentioned that he was eager to leave Alabama because the climate there adversely affected his wife's health; but, like so many other new employees, he was dumbfounded at the scope of the project. Frank A. Getz, who will join the Armed Forces, terminated his employment.

Willard informed Roy Heath at the Wyandotte Chemical Corporation in Michigan that the Met Lab is trying to arrange for his company to grant him a leave of absence to come with us and assured him that, if such a leave of absence can be arranged, Heath will not regret the change. Willard then left on the trip to the University of Wisconsin to interview candidates for employment. Orlemann, on the other hand, returned from his trip to the King Laboratories. There he discussed the company's preparation for the production of high purity barium.

There was a meeting to discuss decontamination in the Bismuth Phosphate Process, arranged by Warren Johnson as a result of the Bismuth Phosphate Process meeting on August 27. Participating in the meeting were members of Coryell's, Sutton's, and my section.

An informal conference on instrument problems of Section C-I was held in Ghiorso's office which I attended along with Crawford, English, Ghiorso, Jaffey, Kohman, and Scott. Among the topics discussed were range chambers, both spherical and differential, neutron counters of two types--the BF_3 pressure ion chamber and the proportional (low and high pressure BF_3), vacuum low geometry alpha counters, magnetic deflection

alpha chambers and magnetizing coil, slow and fast neutron fission counters, beta counting as related to bremsstrahlung effects, resolving power losses and construction factors, freon ion chamber for FP-54, beta and alpha contamination-measuring counters, new instruments (register tester, interval selector, small capacity meters, instantaneous radiation meter, and a constant current charger for FP-54), "statistics chart" and explanatory booklet, standard alpha-chamber development, coincidence counting, Elliott zero-thickness beta counter, and revision of Tibbetts' standard counter.

Wilma visited Helen today, and after work the Orlemanns came to our apartment for dinner.

American and British forces are gaining in their drive on Naples.

Saturday, September 11, 1943

Our New Chemistry Building guard informed me this morning that a man by the name of Lawrence Magnusson was outside waiting to see me. I introduced myself to the visitor, and he said that Willard met him at the A.C.S. meeting in Pittsburg on September 6 and asked him to come by for an interview. I told him that Willard is visiting the University of Wisconsin, but that I would like to talk to him. I believe he assumed that I was some kind of administrative assistant until we had a few minutes discussion about his academic background and I said, "Yes, I think we can use you." Magnusson was a graduate student at Iowa State last year and since then has been working as a control chemist for the Chemical Warfare Service at Huntsville Arsenal in Alabama. I offered him a position as Junior Chemist which he will consider.

This is the last day in Chicago for C. J. Borkowski. His transfer to Clinton is effective Monday.

I received a letter and report from J. A. Collins of TNX, Wilmington, with copies to Sutton, C. M. Cooper, and Miles, in which he states: "On the basis of the following data and discussion, it can only be concluded that the Bismuth Phosphate Process still is less corrosive than the Wet Fluoride Process, and that it has other potential advantages which make it much more attractive than the Wet Fluoride Process." Some of the chemical engineers, especially W. Q. Smith and members of his group, still prefer the Wet Fluoride Process and Collins's report is a kind of refutation to that point of view.

Allison received a letter from Cooksey in Berkeley saying that the drawings I want for the ultramicrobalance will be sent within about a week. He cannot, however, lend us their new balance because it is needed there.

In a memo to Stearns, Manning gave his personal recommendation regarding the qualifications of his friend Michael Wolf, at Fort Benning, Georgia, for work at the Met Lab.

According to the Technical Division's Weekly Report (Memo 122), the Technical Division Separation Process Section has carried out runs in stainless steel equipment to establish the decontamination level of the Bismuth Phosphate Process. The results indicate that the Bismuth Phosphate Process in its present form will give a gamma-ray decontamination factor of 10^4 after two cycles. It is expected that this factor will be improved.

The biweekly meeting on the overall Bismuth Phosphate Method for the extraction, decontamination, and concentration of plutonium was held today. My section presented the results of our experiments. With regard to concentration methods we reported that, as an alternate to the fluoride procedure, the uranous hypophosphate procedure shows good promise. This procedure consists of dissolving bismuth phosphate product precipitate in hydrochloric acid, precipitating UP_2O_6 carrying plutonium, dissolving the UP_2O_6 precipitate and plutonium in warm HNO_3, converting U^{+4} to soluble UO_2^{++}, adding a small amount of U^{+4}, then again precipitating UP_2O_6 and plutonium. Experiments show greater than 90% carrying of plutonium by UP_2O_6. A similar procedure using oxalate instead of hypophosphate, we said, also gives good results. Another concentration procedure tested was the Cb_2O_5-LaF_3 procedure. In the extraction cycle, further work has been done on the use of U^{+4} as a reducing agent for plutonium after the metal dissolving step. We reported also on the use of U^{+4} as a reducing agent in the decontamination cycle and presented the results.

Helen went to Wilma's in the afternoon, and the Baumbachs spent the evening with us.

The Germans have seized Rome and Vatican City. The Soviets have taken Mariupol; the German army is retreating so rapidly there that much materiel is being abandoned.

Sunday, September 12, 1943

Helen and I spent much of today working on the "Table of Isotopes." Several more months of effort will be required.

Monday, September 13, 1943

Willard sent letters to Robert Goeckermann and Frederic Schuler at the University of Wisconsin referring to their meeting last Saturday and informing them that the University may agree to their receiving credit for work done at the Met Lab in lieu of their last semester's work at the University. The matter will be brought up before the University Executive Committee in a few days. If the verdict is favorable, Willard said, we would like very much for them to join us as soon as possible after the end of the current term.

Saturday Fries, using K^+-free plutonium tracer obtained from Brody, tested the carrying of plutonium by $U(C_2O_4)_2$ in the absence of K^+ but in

the presence of Fe^{+3}. Carrying varied from 52% (0.05 M Fe) to 27% (0.1 M Fe). Deciding that the carrying was low because of the absence of K^+, he repeated the experiments today with 0.05 M Fe. His results varied from 95% with 0.1 M K^+ to 36% with 0 K^+. Fries concluded that K^+ is essential for complete carrying of plutonium by $U(C_2O_4)_2$.

The evening seminar of the Chemical Division was held at 7:45 p.m. in Room 251, Ryerson Laboratory.

Just before midnight Stan Thompson and I left from the Englewood Station for Knoxville in order to attend the first meeting at Clinton Laboratories on the Bismuth Phosphate Process, as the main responsibility for carrying on the development of the Bismuth Phosphate Process is being transferred from the Metallurgical Laboratory to the Clinton Laboratories. We are aboard an overnight Pennsylvania train to Cincinnati, where our car will be changed to an L & N train to Knoxville. We plan to spend a day or two of vacation in the area before attending the conference on Friday. This will be our first opportunity to visit the Site X laboratory facilities.

Allied pincers, according to today's newspapers, are closing in on two New Guinea bases -- Salamaua and Lal.

Tuesday, September 14, 1943

Stan Thompson and I arrived in Knoxville at 2:30 p.m. Since the Andrew Johnson and Farragut Hotels were fully occupied, we found a room here at the Whittle Springs Hotel, located on Highways 33 and 71. We played 13 holes of golf at the adjoining Municipal Golf Course--I shot 48 for 9, 76 for 13; Stan 51 for 9, 81 for 13.

In Chicago, Winston Manning had a long-distance telephone conversation at 10:20 a.m. with Wolf at Fort Benning, Georgia, about his employment in Section C-I. Wolf is classified 1A by his draft board, but the War Department Deferment Board in Atlanta is asking that he be deferred on the basis of the work he is now doing. Manning informed Wolf that our personnel people are sending him a letter that could be used to influence either his local board or an appeal board to grant him at least a temporary 2B rating; we cannot proceed with his employment while his draft status is 1A.

"Chemical Research--Production and Extraction of Plutonium, Report for Month Ending September 11, 1943," (CN-931) is being issued. The whole report is concerned solely with the Bismuth Phosphate Process. The work reported is as follows: Halperin and Knox have investigated the use of U^{+4} for reduction of plutonium oxidized in the metal-dissolving step and find it satisfactory under varying conditions of acidity, time, and temperature. Halperin has also studied the reduction of plutonium with Fe^{+2} in 25-12 stainless steel beakers, which he finds to be satisfactory. Cooper, Dean, Dreher, Gaarder, and Kroner have performed further investigations of variables that might affect carrying

of plutonium by bismuth phosphate and find that the carrying of tracer amounts is not appreciably affected by the order or temperature of addition of Bi^{+3} and H_3PO_4, by varying the H_3PO_4 concentration from 0.3 M to 1.6 M, by varying the temperature of digestion from 60°-95°C, or by adding inactive isotopes of the fission products at concentrations equivalent to 7,500 kw/ton pile operation for 100 days. Carrying is not affected by the presence of NO, N_2O, or SiO_2; however, carrying is appreciably affected by the degree of agitation, even in laboratory-scale tests. V. Cooper, Dean, Dixon, Dreher, Gaarder, Halperin, James, G. Johnson, Knox, Kroner, and Miller find that plutonium can be carried quantitatively from UNH solution by a precipitate of bismuth phosphate prepared externally and added to the solution as crystals. A 96% carrying at Site W concentrations of plutonium may be obtained under carefully controlled conditions. Koshland finds that the solubility of bismuth phosphate in 20% UNH solutions increases rapidly with increasing acid concentration. James studied the percentage of plutonium precipitated as a function of bismuth precipitated and finds that the plutonium tends to concentrate in the bismuth phosphate precipitate, in contrast to lanthanum fluoride where the plutonium distribution is the same as the La^{+3} distribution. Dixon has investigated the effect of hydrochloric acid and bismuth on the carrying of plutonium in the lanthanum fluoride analytical method. He finds that in the presence of Bi^{+3} and 1-2 N HCl, a minimum of 2 N HF is required to insure quantitative carrying of plutonium by LaF_3.

Wednesday, September 15, 1943

This morning Stan and I again went to the adjoining Knoxville Municipal Golf Course and played nine holes of golf (I shot 49, Stan shot 50); then we played nine holes more in the afternoon (I shot 48, Stan shot 47).

In Chicago it is the last day of work at the Met Lab for Ray Stoughton and Paul Schulze. They are transferring to Clinton Laboratories and will report to Riordan of the Clinton Laboratories in Knoxville tomorrow.

Latimer is visiting the Met Lab today on his way back to Berkeley from Washington, D.C. He will stay at the Palmer House tonight. I am sorry that I will not be able to see him.

At 8:15 p.m. there will be a general meeting of the Met Lab Association of Scientific Personnel to discuss the proposed constitution for the Association.

Thursday, September 16, 1943

Stan and I played golf again this morning. We then went to downtown Knoxville and took a Site X limousine to the Site, arriving here around noon. After passing through a security fence, we came to the

town, which is called Oak Ridge. The residential area sprawls over the hills and consists of barrack-like dormitories, thousands of trailers, cemesto houses (constructed of panels made of cement and asbestos) and prefab houses. Bulldozers and earth-moving equipment are everywhere, cutting up roads, pulling down trees, and levelling plots for new homes. We went directly to the "Guest House," a hotel near the shopping area in the flats, where we have room reservations.

Stan and I visited Perlman and his group in the afternoon to discuss their progress. Stan also saw Spof English and went over the details of the Bismuth Phosphate Process with him.

In Chicago Report CK-932, "Chemical Research--Special Chemistry of Plutonium, Report for Month Ending September 11, 1943," is being issued. The items covered are the following. Baumbach, Rosenfels, and Kirk report on the following difficulties encountered in attempting the reduction of plutonium to metal: wetting of the crucible by the plutonium metal, volatility of the plutonium metal, interference by the slag with melting of the metal into solid pieces, failure of the slag to melt and separate from the metal, difficulty of removing excess reducing agent, and possible alloying of the plutonium with excess reductant. Present indications are that plutonium fluoride may be successfully reduced with good yield of usable metal by heating it in vacuo with excess magnesium in a thoria crucible at 1,600°C for about one minute. It is noteworthy that the fluoride and oxide of plutonium are not reduced to the metal by zinc, thus giving an approximation of the lower limit of electropositivity of the metal. Davidson has made a preliminary study of the volatilization of the lower fluoride of plutonium by volatilizing about 0.01 microgram of plutonium fluoride at 700°C in vacuo. There is evidence that gases evolving during the degassing of the system convert some of the PuF_4 to a less volatile substance. Brody and Orlemann have studied the ether extraction of oxidized plutonium to determine the value the method may have in the purification of plutonium. In nitric acid solutions, from 1 M to 5 M, the percentage of oxidized plutonium extracted by an ether phase of volume equal to that of the aqueous phase varies from 1% at the lower to 50% at the higher acid concentration. Similar results are also obtained in 1 M HNO_3 solutions with varying concentrations of NH_4NO_3. Katz and Orlemann are studying the reaction of hydrogen bromide with plutonium oxide. The volatilization of the oxide in a stream of HBr has been studied; a static apparatus has been designed to facilitate collection. Dixon and Orlemann have investigated the extraction of buffered solutions of tracer amounts of reduced plutonium with solutions of acetylacetone in chloroform with regard to possible usefulness as a purification method. At a pH of 4.5, tracer amounts of plutonium are 95% extracted by several washes. Ultramicrochemical techniques employed to prepare plutonium acetylacetone give a substance readily soluble in chloroform. Katz has prepared an apparatus for studying plutonium higher fluorides using calcium fluoride tubing in an effort to employ as inert material as possible. Initial experiments indicate that the plutonium higher fluoride condenses to a considerable extent in the calcium fluoride tubes but that the temperature at which condensation takes place is probably lower than in copper tubing. Clifford Smith and

Cunningham have constructed an improved sublimation apparatus capable of operating at higher vacuum and have tested it in the course of extending earlier work on vacuum sublimation, a possible purification technique. Vapor pressure measurements of UBr_4 were made as a means of checking the apparatus, and the values obtained appear consistent with published values.

Friday, September 17, 1943

Thompson and I are at Site X attending a meeting with Clinton Lab personnel on the Bismuth Phosphate Process in the Chemistry Research Building (No. 706A). Other people who came from Chicago to attend the meeting are C. M. Cooper, Franck, G. R. Leader, Maloney, J. H. Peterson, and Sutton. Thompson and I, representing Section C-I, gave the latest experimental findings of our group. In summary, our experiments show that the presence of Fe^{+3} inhibits the carrying of plutonium by UP_2O_6 from HCl solutions of bismuth phosphate. Data on the use of $U(C_2O_4)_2$ in a concentration process indicate that the presence of K^+ ion may be necessary for good carrying of plutonium and that Fe^{+3} also inhibits the carrying of plutonium by $U(C_2O_4)_2$. The results of two decontamination studies show that adding the Bi^{+3} after the H_3PO_4 gives better decontamination but poorer product recovery than when the order of addition of these reagents is reversed. Reduction of plutonium by U^{+4} in the decontamination cycle is not appreciably affected by the use of glass or steel equipment or by the use of a hot or cold strike; one addition of U^{+4} to a concentration of 0.0025 M in 0.1 M H_3PO_4 and 1 N HNO_3 gives 98% reduction of plutonium in 15 minutes at 75°C. Solubility measurements of "$Pu_3(PO_4)_4$" show increasing solubility with increasing UNH concentration. Finally, experiments on the effect of UNH concentration on the carrying of plutonium by bismuth phosphate show a decrease in carrying in solutions greater than 17.5% UNH, using 0.5 mg/cc Bi^{+3}. The Met Lab Technical Division also reported today on their results of larger scale laboratory and semiworks testing of the Bismuth Phosphate Process.

Stan and I then visited the Pile Building (No. 105) and the Chemical Separations Plant (No. 205) where construction is nearly completed. (See Figure 6.)

As requested by G. D. Graves, Allison sent a letter to Greenewalt on the present status of the Bismuth Phosphate Process. He quoted a summary by C. M. Cooper which states that the extraction step has been satisfactorily demonstrated on the semiworks scale (95% yield), but that neither the yields nor the decontamination in the decontamination steps (69% yield, 1,200 decontamination factor) have as yet been demonstrated on a semiworks scale as being adequate to provide a satisfactory process. He went on to say that he believes the process as presently developed can be confidently expected to give satisfactory decontamination at Clinton with yields of decontaminated and concentrated product of at least 50 percent and also that further development work will insure similar results under Hanford conditions.

The "Monthly Report on the Activities of the Chemical Division" at Chicago is being issued today (MUC-JF-91). The activities of my section are summarized as follows:

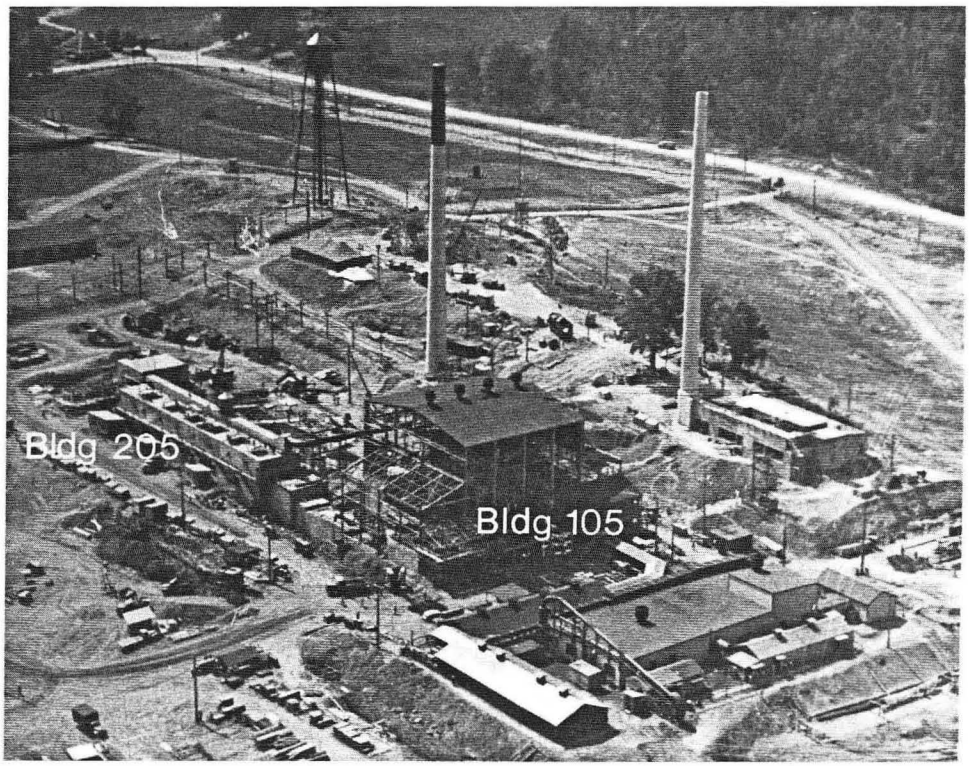

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Figure 6. Chemical Separation Building (No. 205) and accompanying stack; Pile Building (No. 105) and accompanying stack. Site X, September 1943.

I. Extraction and Decontamination of Plutonium

a. Bismuth Phosphate Process. U^{+4} is more satisfactory than Fe^{++} for the reduction of plutonium in the semiworks scale process since the latter inhibits carrying. Carrying of tracer amounts is not appreciably affected, within the limits tested, by temperature and order of addition of Bi^{+3} and H_3PO_4, temperature of digestion, and by addition of inactive isotopes of fission products at concentrations equivalent to 7,500 kw/ton pile operation for 100 days. Also, NO, N_2O, and SiO_2 are without effect; but agitation is an important factor, even in the laboratory scale tests. With vigorous agitation, the carrying reaches a final limiting value at a digestion time of two hours. Plutonium is carried quantitatively (96%) by a preformed bismuth phosphate precipitate when the latter is added to a 20% $UO_2(NO_3)_2 \cdot 6H_2O$ solution containing no H_3PO_4. Quantitative carrying of plutonium can be obtained by adding an amount of H_3PO_4 sufficient to precipitate less than half of the Bi^{+3} to solutions containing tracer amounts of Pu^{+4}. This result is contrary to that found in the lanthanum fluoride precipitation where the fraction of the plutonium carried is equivalent to the fraction of lanthanum precipitated. (5-1/2 men, A priority)

b. New Extraction Methods. Preliminary work has shown that $U_3(AsO_4)_4$ in the presence of SO_4^{-2} and $BiAsO_4$ (without SO_4^{-2}) may be used to carry reduced plutonium from 20% $UO_2(NO_3)_2 \cdot 6H_2O$ solutions. Also, it has been found that oxalate ion will quantitatively reduce $Pu^{(o)}$ by heating for 45 minutes at 80°C. (1 man; B priority)

c. Concentration of Plutonium following Decontamination. Since the concentration of plutonium at Site X will be of the order of 1/1000 that at Site W, it will be necessary to provide for a number of concentration cycles at the former site before obtaining plutonium free from the carrier. A number of processes have been given consideration. The uranyl peroxide process has been found to be unsuitable for low uranium/plutonium ratios, but the Cb_2O_5-LaF_3 method has given satisfactory results. The Wet Fluoride Process is not as rapid as the other processes. Recent evidence indicates that the use of uranous hypophosphate or uranous oxalate may be the most effective method thus far investigated. (4 men; A priority)

II. Purification and Chemistry of Plutonium

a. Metal Production. An improved procedure for preparation of plutonium metal has been found; it depends upon heating plutonium fluoride with magnesium in thoria crucibles at high temperatures for a short time. (5 men; A priority)

b. Volatility of Uranium and Plutonium Halides. A preliminary study has shown that about 0.01 microgram of plutonium is volatilized, probably as the lower fluoride when heated in a vacuum at 700°C for several hours. The volatilization of a plutonium bromide by the reaction of HBr on the oxide has been studied, and apparatus has been designed for handling the material. An improved sublimation apparatus has been tested in the course of extending earlier work on the vacuum sublimation of uranium halides. Vapor pressure measurements of UBr_4 have been made at 250°C and at 300°C as a means of checking the apparatus prior to plutonous bromide measurements. (4 men; A priority)

c. Solvent Extraction Methods. In HNO_3 solutions, from 1-5 M, the percentage of oxidized plutonium extracted by an ether phase of volume equal to that of the aqueous phase varies from 1-50%, respectively. The addition of NH_4NO_3 to the HNO_3 increases the percentage of plutonium extracted. At a pH of 4.5 tracer amounts of plutonium are 95% extracted with several washes of solutions containing acetylacetone in chloroform. If oxalate ion is present, only 1% of the plutonium is extracted. Ultramicrochemical techniques used to prepare plutonium acetylacetonate have shown that a substance is produced which is readily soluble in chloroform and quite volatile at reduced pressures. (2 men; A priority)

III. Problems in Progress Not Covered in Report

a. Spectrographic Analysis (1 man; A priority)

b. Precipitation Reactions for Purification (2 men, A priority)

c. Additional Purification Problems (4 men; A priority)

d. Wet Fluoride Process (2 men; B priority)

e. Adsorption Processes (4 men; B priority)

f. Dry Methods (3 men; B priority)

g. Recovery of Plutonium (3 men; A priority)

h. U^{233} Production and Extraction (3-1/2 men; A priority)

i. Instrument Group (7 men; A priority)

During the coming month, eight men will be transferred to Site X.

Saturday, September 18, 1943

Stan and I attended the continuing meetings on the Bismuth Phosphate Process in the Chemistry Building at Site X. We discussed and agreed upon the dates for future progress meetings at Clinton and Chicago--Charles Cooper will issue a memorandum giving this schedule. In the afternoon we were driven back to the Whittle Springs Hotel in Knoxville. We played some golf on the Knoxville Municipal Golf Course and then spent the night at the Whittle Springs Hotel.

In Chicago it is the last day at the Met Lab for H. H. Anderson and Manfred Lindner, who are transferring to the Clinton Laboratories, effective September 20.

The Separation Process Section of the Technical Division in Chicago reported, in the Technical Division Weekly Report (Memo No. 123), on four extraction plus two decontamination cycle liter-scale runs (one in glass, in my section) on the Bismuth Phosphate Process completed during the week, in accordance with the currently preferred master flowsheet. Overall product yield ranges from 86.5 to 90.7%. Gamma-ray decontamination factors vary from 10^4 to 3.5×10^4.

Sunday, September 19, 1943

Thompson and I caught the 2:00 p.m. train from Knoxville to Cincinnati, where we transferred to a Penn Train for Chicago. We are scheduled to arrive in Chicago tomorrow morning.

Monday, September 20, 1943

Stan Thompson and I arrived back in Chicago at 8:00 a.m.

Helen told me about her activities during my week's absence. Tuesday she had dinner and spent the evening with the Baumbachs. Wednesday afternoon she went downtown with Wilma, had dinner with Wilma and Al at Henrici's (71 W. Randolph), followed by a movie. Thursday Helen had lunch at Mary Cooper's and then spent the afternoon with her. Friday was a full day for Helen. She visited Alice Thompson, had dinner with the Ghiorsos at Isbell's (51st St. [Hyde Park Blvd.] near Blackstone Ave.) followed by a show with Wilma, then back to the Ghiorsos' for ice cream, where Al Jarrett was visiting. On Saturday Frances Chilson took Helen to the Great Lakes football game. They had dinner downtown and went to a movie. Yesterday she again spent the day with Frances Chilson.

Ray D. Frank and Asher J. Margolis joined Section C-I last week during my absence. Frank, who has a Bachelor's Degree in metallurgical engineering from Fenn College in Cleveland, will be working as a Research Assistant in Kirk's group. Margolis, a chemical engineer, will work with Thompson. He comes to us from the Emulsol Corporation in Chicago. Last Saturday Frank Belletire was hired as a technician at $30 per week. He has studied chemistry at Northwestern University and has worked there for Professor W. Evans.

Last Wednesday in a memo to A. V. Peterson, Jaffey detailed the history and composition of our waste uranyl nitrate which amounts to 1,800-1,900 pounds of UNH in about 250 gallons of approximately 50% hexahydrate solution and 275 pounds as solid UNH. This is material that has been subjected to neutron bombardment at the Washington University cyclotron in St. Louis. The solutions are now stored in 5-gallon bottles just outside the North Stands East Laboratory, and the solid is stored in 25-pound bottles in the same place.

Last Saturday Compton was at the Montreal Laboratory in Canada, together with General Groves, Tolman, Smyth, and Wigner, to attend a meeting for the "Exchange of Information with the British." Such an exchange was sanctioned last month on August 19 when Roosevelt and Churchill signed the so-called Quebec Agreement, "Articles of Agreement Governing Collaboration Between the Authorities of the U.S.A. and the U.K. in the Matter of Tube Alloys." The British code word "Tube Alloys," of course, stands for the uranium project. Those representing the British at the Montreal meeting were Chadwick, Halban, Placzek, and Newell; and there were informal discussions between the Americans and Paneth, Auger, and Sir Edward Appleton, as well as other research men.

At this meeting Halban pointed out that they have been concerned with the relative merits of U^{233}, U^{235}, and Pu^{239}. Much of their work has been aimed toward using U^{235} for producing U^{233} or Pu^{239}. Compton mentioned that Wigner has emphasized likewise the possible advantage of transforming U^{235} into U^{233} or Pu^{239} as Halban suggests. Chadwick commented that his "money is on the U^{235}," giving as his chief reason the probable occurrence of spontaneous fission neutrons in the other isotopes. These he anticipated from his experience (with which we Americans are unfamiliar) that neutrons are present with radon in clean nickel tubes to the extent of about 0.001 the number that are present in radon-beryllium mixtures. Compton and the other Americans were skeptical regarding the applicability of this observation to the presence of spontaneous fission neutrons with Pu^{239} but did not discuss the point.

Halban outlined the five main types of chain-reacting piles with which they are concerned. These are:

1. A homogeneous pile (which he calls the "all-wet" pile), using uranium enriched with U^{235} or Pu^{239} to reduce its size. Two alternative forms were suggested: (a) A single reaction chamber surrounded by a reflector and containing about 2-4 kg of Pu^{239} plus 10 to 20 tons of heavy water and a suitable amount of uranium. (b) A central "seed" of 1 kg of Pu^{239} with one ton of heavy water, surrounded by a container with uranium and heavy water. Calculations, evidently rough, indicate about 10 megawatts per ton of heavy water in excess of the critical amount. Advantages claimed for this system are a low heavy water requirement and simple engineering. The Canadians are considering the use of solutions of uranyl sulfate rather than slurries. In discussing this type of plant Wigner described recent American work on the nuclear constants involved. There was essential agreement with the British representatives on all points except the neutron capture cross section for heavy water, for which the Chicago value is 0.003 as compared with the value of 0.012 as used by the British. This accounts for the Chicago estimate of 30 tons as the critical mass using unenriched material, whereas the Montreal estimate is much larger and requires enriched mixtures to become practicable. Some discussion of the experimental bases of the two values followed, each group apparently having greater confidence in its own data.

2. A "boiler" of tubes, perhaps aluminum, filled with "hex" and immersed in heavy water. Halban estimated a requirement of 180 tons of hex immersed in 10 to 20 tons of heavy water, for a 300 megawatt plant. He suggested as a major advantage the possibility of continuous extraction of the Pu^{239} and the by-products. As disadvantages he mentioned the large amount of hex required and the possibility of its solidification in cool parts of the equipment. Groves emphasized the difficulty of securing the necessary amount of hex within a reasonable time for use in this war, and Wigner noted the double corrosion hazard of hex on the inside and heavy water on the outside of the boiler tubes.

3. The use of doubly enriched uranium metal with water, the metal being in the form of thin plates close together between which the water would flow. Halban calculates 7 tons of doubly enriched metal would be required in 1.3 cubic meters of water. Chadwick pointed out

that this could be used in place of the top stages of a diffusion system, to produce plutonium in place of U^{235}, and avoiding the large "hold-up" time inherent in the diffusion method. Compton criticized the process from the point of view of time schedule, since it would mean putting through two different processes (diffusion and plutonium production) in series. He thought eventually this might be an excellent system for making Pu^{239} or U^{233} at the expense of U^{235}.

4. The "hedgehog," a seed of U^{235} or Pu^{239} in heavy water, surrounded by a spherical shell containing uranium rods in water. This would give roughly 5 times as much plutonium in the outside rods as used up in the seed. Wigner noted that this system affords poor means of heat dissipation in the outer shell.

5. A large heterogeneous pile, using small diameter rods to increase the cooling rate, and an excess of uranium. This will give a k lower than the optimum, but a high rate of heat dissipation and a thermally stable, non-explosive unit. At least 10 megawatts per ton of heavy water would be possible.

In the general discussion, in response to a question from Groves, Newell estimated that to build a unit of type 1 would require about 15 months and $5,000,000. Chadwick increased cost estimate to $25,000,000. This time estimate is optimistic, according to Compton, but not unreasonable. Chadwick's cost estimate is good if it does not include site, housing, and other accessories.

In later discussion within the American group alone, the opinion was expressed that, if any of these plans are to be carried through by the British, it should be done by British personnel using British materials because U. S. help given to this program will slow down the U. S. plutonium work designed for use in this war. It was not considered possible that this British program could contribute Pu^{239} for the present war and that encouragement of their work might accordingly result in diversion of effort from more essential jobs.

The research program in hand at Montreal is outlined in the following table:

	Project	Material Required	Help Needed from U.S.A.
a	Measurement of "age"	150 l D_2O	Check purity of D_2O
b	Diffusion length in "poisoned" material	300 l D_2O	Should do at Argonne
c	k by Cambridge method	112 l D_2O	Some Np^{237}
d	" "	250 l D_2O	Loan of 100 l of D_2O
e	(n,2n) reaction with D	-	none
f	"Slowing down length"	500 l D_2O	400 l of D_2O
g	Test of neutron utilization using type IV pile		10 to 20 tons uranium rods for one month
h	Slurry experiments		
i	Radiochemistry experiments		

The Americans agreed among themselves that the purity test for experiment a should be referred to Urey, if needed, at once and that similar tests can be made at Chicago as soon as the equipment now being set up is in operation; that before performing experiment b at Argonne, it would be desirable to have a representative of the Montreal laboratory come to Chicago to discuss the procedure with Fermi; that an effort will be made to secure a suitable sample of Np^{237} at Chicago for use at Montreal; and that for the remaining experiments on the list, the loans of large quantities of heavy water and uranium are not possible now but could be considered about March 1944.

Joe Kennedy is here from Site Y to visit the Met Lab. We will attend the Laboratory Council Chemistry Information Meeting tomorrow, a meeting on the metallurgy of plutonium, and will confer on the decontamination of highly enriched uranium containing fission products. The contamination with fission products will result when Site Y constructs and operates a chain-reacting unit immersed in water and using a fuel of U^{235}-enriched uranium.

Last Thursday Charles Cooper wrote a "Memo to Reading File" that I read today. It said that, since I was absent from Chicago during the week, Miles had sent a copy of my September 10 letter from Collins directly to Franck for his immediate information. Cooper wrote: "The letter indicates that for both Bismuth Phosphate and Lanthanum Fluoride Processes the corrosion to be anticipated is sufficiently low for the process to be feasible. As has been realized for some time the Lanthanum Fluoride solutions are somewhat more corrosive than the Bismuth Phosphate."

I read a memo my office received on Saturday addressed to Franck, W. C. Johnson, Boyd, Coryell, Potratz, and me from Allison concerning the feasibility of decontaminating and reclaiming some highly-enriched uranium that is to be contained in a small chain-reacting pile being built at Site Y. The Army is reluctant to consent to the construction of this small pile if there is any danger that the enriched uranium would be so contaminated with fission products that it could not be used again. Oppenheimer, during a recent visit to Chicago, told Allison that he considers his staff at Site Y too small to take on the task of decontaminating this U^{235}-enriched uranium from fission products and wondered if we can be of help. Allison, therefore asked us to meet and draw up a short statement covering what is known about such a decontamination.

In a memo to me regarding recent progress in the extraction and decontamination of plutonium by adsorption methods, Beaton says that a promising process for extraction and decontamination is the combination of an extraction cycle employing Amberlite IR-1, followed by a decontamination cycle with titanium zeolite which had been subjected to heat treatment. The Research Products Corporation of Madison, he says, could produce this material in quantity if $20,000 were invested in an expansion of their facilities.

Compton wrote to J. A. Burdine, Vice President, University of Texas, urging that the University grant a leave of absence to George W. Watt, whom we want for Section C-I, assuring him that Watt's "specialized training and ability will be utilized here to the utmost of his capacities in fundamental research which is of great importance to the prosecution of the war."

Nickson of the Health Division instructed Reed A. McWilliams, Superintendent of Services, how to keep down the dust in the West Stands pressing room where Katzin will be preparing thorium carbonate pellets off and on for the next several months.

C. M. Cooper is the secretary for the Laboratory Council Meetings. Today he sent a memo to Compton, with copies to members of the Council, guests, and observers, saying that starting with October the regular meetings of the Project Council (which we have been calling the Laboratory Council) will begin and progress reports (Information Meetings) of the various Divisions, together with reports from associated laboratories, will be scheduled as follows: First Tuesday (October 5), Health, Nuclear Physics, and General Physics. Third Tuesday (October 19), Chemistry and Technology. This will require both morning (9:30 to 12:30) and afternoon (2:00 to 5:00) sessions. Project Council Policy Meetings will be held on Wednesday following the Information Meetings.

Cooper also issued a schedule for future progress meetings on the development of the Bismuth Phosphate Process as decided upon at the September 17 meeting at Clinton Laboratories. For the immediate future, two meetings will be held each month as follows: First meeting of the month will be held at Clinton on the Friday following the first Tuesday and will start at 1:00 p.m. The second meeting will be held at Chicago on the Wednesday following the third Tuesday and will start at 2:00 p.m. Corresponding dates for the balance of this year are:

October 8	at Clinton	1:00 p.m.
October 20	at Chicago	2:00 p.m.
November 5	at Clinton	1:00 p.m.
November 17	at Chicago	2:00 p.m.
December 10	at Clinton	1:00 p.m.
December 22	at Chicago	2:00 p.m.

Kennedy had dinner with us in our apartment. Ermon Eastman is here from Berkeley also to attend the meeting; however, he could not come to dinner as he had other plans; so we invited him to eat with us tomorrow night. There was an evening seminar of the Chemical Division at 7:45 in Ryerson Laboratory, Room 251, which the three of us attended.

The Germans have fled Sardinia under Italian attack. Marshal Pietro Badolgio, who took over the Italian government and is in Allied-held Italian territory, gave the order to oust the Germans.

Tuesday, September 21, 1943

This was the last day at the Met Lab for C. D. Coryell, Chief of Chemistry Section C-III, who is transferring to Clinton Laboratories as a Senior Chemist. Another laboratory assistant, Minnie Daniels, was hired today for Section C-I.

Willard transmitted to Watt at the University of Texas a copy of the letter Compton sent to the University asking that Watt be given a leave of absence.

Davidson and Katz performed an experiment last Thursday to test the volatility of the higher fluoride of plutonium using the apparatus they recently built. They produced the higher fluoride by treating plutonium lower fluoride with fluorine at a temperature corresponding to red heat and collecting it on platinum kept at dry ice temperature. When the higher fluoride on platinum was heated to about 230°C, very little of it volatilized. It is not known whether this is due to low volatility or to the difficulty of maintaining the higher plutonium fluoride in that form.

Last week G. Johnson completed a series of experiments on the solubility of plutonous phosphate. To solutions of 1 N H_2SO_4, 0.4 M H_3PO_4 with varying amounts of UNH he added a precipitate of plutonous phosphate which had been prepared and then washed free of H_3PO_4. This was allowed to digest overnight at 75°C. He found the solubility of plutonous phosphate to be 60 mg/l with 0% UNH and increased rapidly from 156 mg/l at 13% UNH to 770 mg/l at 19.6% UNH.

Thompson spoke long distance with English in Oak Ridge giving him some information on extraction conditions for the Bismuth Arsenate Method.

I attended a Laboratory Council Chemistry Information Meeting at 9:30 a.m. (now held on the third Tuesday of the month). Present were Allison, Beaton, Boyd, Chipman, Connick, C. M. Cooper, Coryell, Cunningham, Dean, Eastman, Fermi, Foster, Franck, Fred, Garrison, Johns, W. C. Johnson, Kennedy, Kirk, Magel, Miles, Mulliken, Potratz, Cyril Smith, Spedding, Sugarman, Sutton, Thomas, W. W. Watson, Whitaker, Wigner, Willard, and later Compton, Hilberry, and Smyth. Beaton, Willard, Cunningham, Kirk, and I were scheduled for the latter part of the program; and we presented information on various aspects of our section's program. Beaton described the promising results of an adsorption process using Amberlite IR-1 and titanium zeolite. Willard spoke on separation processes indicating that Zachariasen's work shows the same crystal structure of bismuth phosphate precipitated from strong or weak uranyl acetate solutions. The work to determine whether plutonium phosphate is isomorphous with bismuth phosphate should be completed in a few weeks. Willard also discussed the shift of the Chicago group to scouting and fundamental work, such as, possible initial extraction steps to couple with sodium uranyl acetate decontamination. He mentioned bismuth arsenate as an alternative carrier to bismuth phosphate and discussed the latest experiments on concentration procedures for Site X and Site W involving the use of UP_2O_6 or $U(C_2O_4)_2$. Cunningham reviewed the status of methods of analysis and purification. Kirk discussed plutonium metal production indicating that the best results have

been obtained with Mg + PuF_4 in ThO_2 crucibles. I mentioned the main obstacle to progress in metal production is the lack of 10-20 microgram samples of plutonium. Warren Garrison in Burton's section wondered if plutonium metal could not be prepared by bombarding PuF_4 with electrons and pumping off the fluorine.

I attended a special meeting on the metallurgy of plutonium at 2:30 p.m. in Room 259, Ryerson Laboratory. The meeting was called by Chipman, Chief of the Metallurgy Section, for the benefit of Charles A. Thomas, the Monsanto research director who is coordinating the work on the metallurgy of plutonium at Chicago, Site Y, Ames, Brown University, and Berkeley. Present besides me were Thomas, Chipman, Baumbach, Eastman, Foster, Franck, Hilberry, W. C. Johnson, Kirk, Magel, Rosenfels, Cyril Smith, Spedding, and later Compton, Mulliken, and Kennedy. The meeting was opened by Thomas, who made a few introductory remarks and then turned the meeting over to Chipman. The plan of the program was to call on members of various groups working on the plutonium metallurgical problem and then to have open discussion. The first person to speak was Kirk. He presented the work on plutonium metal preparation, pointing out that those physical properties of most importance to be measured are density, melting point, volatility, hardness, and hydride formation. Because of the lack of plutonium, reductions can be made only at 2-4 week intervals using 3-5 micrograms per reduction. Thomas speculated that perhaps further efforts should be deferred until larger quantities of plutonium are available. Kirk disagreed, believing that a 5-microgram piece of metal would be enough for a rough density determination. In a discussion that then took place between Thomas and me, I estimated that it might be necessary to have 100 micrograms of plutonium in order to provide the 25-50 microgram piece of metal needed for a density measurement, good to 5 percent. In answer to a question from Johnson, I indicated it is exceedingly difficult to predict the density of plutonium from the Periodic Table. Franck asked about the range of alpha particles in plutonium metal, to which I replied that there is no good value yet. There followed a discussion of work on possible hydride formation of plutonium. I indicated that previous work is going to be repeated, and Thomas said such work should be given a very high priority. Cyril Smith said that at Site Y the study of the mechanical properties of plutonium will be postponed until at least one gram of plutonium becomes available. Thomas put the group on record as stating that the minimum immediate requirement of plutonium is 10 milligrams with which to make measurements of density, hydride formation, melting point, etc. Responsibilities assigned to my section are (1) Kirk and his group are to carry out the preparation of microgram quantities of plutonium metal and the study of density, hydride formation, and melting point of the metal, and (2) Manning is to work on the preparation of metallic uranium and plutonium by atomic hydrogen, in parallel with Cyril Smith's work at Site Y. The meeting was adjourned at 6:10 p.m.

I had a migraine headache in the evening, and we had to cancel our dinner for Ermon Eastman.

French commandos have stormed into Corsica, joining local French Corsicans who had begun to fight immediately following the Italian armistice.

Wednesday, September 22, 1943

Willard received a letter of recommendation from M. L. Jackson, Assistant Professor of Soils, University of Wisconsin, for Nison N. Hellman who has just completed his work for the Ph.D. in the Department of Soils. Jackson endorsed Hellman, both as to his professional and personal qualities.

I wrote unofficially to my friend, Fred W. Albaugh, Union Oil Company of California, Wilmington, California, expressing my disappointment at learning from Vance Cooper that he has decided not to come with us. In an effort to persuade him to change his mind, I broke some of the rules under which we are supposed to be operating and told him that the importance and interest of the work we are doing here is so great that it can hardly be described. I said it is my belief that if the war lasts for another year or two, this laboratory will provide the means for an Allied victory. I also said it is my opinion that the discoveries being made here are greater than the discovery of electricity. I asked him to regard my letter as entirely confidential and to destroy it as soon as he has absorbed its contents.

A second letter went to another friend, George G. Manov, who is employed by the Bureau of Standards in Washington, D.C. I told him that I have been here in Chicago for the last year and a half doing interesting work important to the war effort. I asked him if he would like to join us; and, in case he does, I would like to discuss the matter with him.

Last Saturday, in his recruitment efforts, Willard called Magnusson in Rock Island, Illinois, to ascertain if he has come to a decision regarding our offer of a position. Magnusson is making a trip East and will stop to see us again on the 20th or 21st. He then wrote four letters. The first was addressed to Alvin J. Dirksen in Niagara Falls, saying that we have asked the Met Lab Personnel Office to make him a formal offer and assuring him of the importance and interest of the work of the Lab. The second went to Roy Heath at Wyandotte, Michigan, enclosing a copy of a letter Compton has sent to the Wyandotte Chemical Company requesting that Heath be granted a leave of absence. The third letter, sent over my name, went to Fred Meyer in Pittsburgh, Pennsylvania, saying we have asked the Met Lab Personnel Office to make him an offer of a position with us, and assuring him that, should he come with us, he will be able to work to the utmost of his capacity in a job which is extremely urgent to the success of the war effort. The final letter, also over my name, was mailed to Donald G. Pye in Palo Alto, California, referring to his and Willard's meeting in Pittsburgh and expressing the hope that Pye will be able to see us when he is in Chicago this month. Willard also told him our Personnel Office has been asked to offer him a position to take effect at the time Pye receives his Ph.D. degree.

9/22/43 (cont.)

Today Willard again wrote A. J. Dirksen in Niagara Falls, giving him more information on the nature of the work at the Met Lab and the working conditions. He told Dirksen we would like to have him join us as soon as possible and hoped for a favorable reply. He also wrote J. G. Malm in Madison that the Met Lab Personnel Office forwarded him a formal offer of a position yesterday.

I attended a meeting of the Purification and Metal Production Groups of Section C-I in Room 209, Eckhart Hall at 7:45 p.m. We plan now to have our meeting on Wednesday nights, alternating the meetings of the Purification and Metal Production Groups and those of the Extraction Group. Others present were Baumbach, Brody, Cunningham, Davidson, Dixon, Fred, Ghiorso, Hagemann, Jaffey, Jensen, G. Johnson, Katz, Kirk, Kohman, Manning, Marshall, Nachtrieb, Orlemann, Patton, Rosenfels, S. W. Sheel, Clifford Smith, and Willard. Brody discussed his work on the purification of oxidized plutonium by ether extraction. Using a solution 0.02 M in dichromate, 10 M in NH_4NO_3, and 1 M in HNO_3, he extracted 55-60% of the plutonium. I discussed the effect of hydrogen and plutonium in the final nuclear device--the presence of hydrogen, because of its better scattering power for neutrons, would be an advantage if the density of the hydride is not too different from that of the metal. Katz discussed the reaction of massive thorium with hydrogen, which resulted in considerable disintegration. I pointed out the possible utility of this reaction in powdering massive thorium as preliminary to a dry process for obtaining U^{233}. I indicated that with regard to plutonium, the highest priority projects at present are to determine the density of the metal, its behavior with hydrogen, its melting point, and its mechanical strength. Kirk discussed general results. Lithium does reduce uranium fluoride but apparently not plutonium fluoride. This indicates that plutonium is slightly more electropositive than uranium. Results with magnesium reduction, however, indicate that plutonium may be less electropositive than uranium. Zvolner has been successful in electroplating microgram quantities of copper, iron, and zinc and is now working with manganese. Manning described his apparatus for reduction with atomic hydrogen. He has succeeded in reducing zinc oxide to zinc at room temperature. Davidson discussed the preparation and revolatilization of plutonium higher fluoride. Apparatus was described consisting of a nickel strip which could be heated electrically, placed in close proximity to a platinum cold finger. It did not prove to be possible to revolatilize the material collected on the platinum at temperatures up to 250°C. Jaffey discussed range counters, and Kohman described a nitrogen-filled counter. Crawford and Jarrett are checking counters by statistical methods and are writing a booklet. I indicated another experiment of high priority is the investigation of the volatility of plutonium acetyl-acetonate.

Helen went out to lunch with Al and Wilma Ghiorso, after which Al, who is taking some vacation, and Helen went bowling. The Ghiorsos came back to have dinner with us. The Baumbachs dropped in after the meeting.

There was a Laboratory Council Policy Meeting at 9:30 a.m. in Room 209, Eckhart Hall, attended by Allison, Compton, C. M. Cooper, Doan, Eastman, Fermi, Franck, Hilberry, W. C. Johnson, Miles, Mulliken, Munnecke, Nichols, A. V. Peterson, Smyth, Spedding, Spedding, Thomas, W. W. Watson, Whitaker, and Wigner. The preparation and distribution of reports was discussed. Johnson reported on the distribution of personnel and research problems in the Chemistry Division. Because this report lists all the major problems being studied in the Met Lab Chemistry Division, I reproduce it here as follows:

DISTRIBUTION OF PERSONNEL AND PROBLEMS

	Period I (8/17-9/22)	Period II (9/22-10/17)	Priority
Seaborg--Section C-I			
Subject			
Bismuth Phosphate Process	5-1/2	4	A
Wet Fluoride Process	2	2	B
Adsorption Processes	4	5	B
Dry Methods	3	1	B
Concentration	4	4	A
Alternate Methods	1	2	C
Recovery of Plutonium	3	2	A
U^{233} Studies	3-1/2	1	A
Instrument Group	7	6	A
Purification	13	13	A
Metal Production	5	6	A
Total	51	46	

(During the month 8 of these men will transfer to Site X)

Burton--Section C-II			
Subject			
Shielding Materials	2	2	A
Plastics	1/2	1/2	A
Lubricating Oil	1/2	1/2	A
Gas Analysis	1	1	A
Ion-pair Measurements	1	1	D
Radiation Effects in Pure Hydrocarbons	1	1	D
Radiation Corrosion of Aluminum	5	5	A
Basic Studies on Water	1	1	D
P-9 Pile Problems	1	1	B
Radiation of Hex	2	2	C
Resonance Frequency Measurements on Graphite	2-1/2	1-1/2	A
Electrical Resistance of Graphite	2-1/2	1-1/2	A

9/22/43 (cont.)

Adsorption and Pore Volume	1	1/2	A
Thermal Conductivity of Graphite	0	1-1/2	A
Radiation Effect in Metals	0	1	B
Gas Solubilities	1	1/2	A
Bismuth Phosphate Process	1	1	A
Radiation of Formic Acid	1	1	A
Radiation of Ether	1/2	1/2	A
Actinometers	1	2	D
Electrostatic Generator	2	2	A
Total	27-1/2	28	

Coryell--Section C-III

Subject			
Bismuth Phosphate Process	10	5	A
Determination of Absolute Fission Yields	2	1	A
Gross γ-Decay Curves after Short Irradiation	2	2	A
Th Fission Products	2	1	C
Ce-UX Separation	1	1	D
Determination of Fission Activities	1-1/2	1	A
U^{237} Fission Yield	1/2	0	D
Electrostatic β-ray Spectrograph	1	1	B
Methods for Obtaining Large Amounts of Fission Products	3	2	B
Apparatus Maintenance and Development	3	2	A
Total	26	16	

(14 men are now in process of moving to Site X, which accounts for reduction in personnel available for the coming month)

Boyd--Section C-IV

Subject			
Primary Materials			
Metal, Oxide, Fluoride	10	6	A
Graphite	6	10	A
Special Samples	4	4	B
Semiworks Assistance	3	3*	B
Research for Methods	5	6	A
Product Purification			
Spectrochemical Research	4	4	A
Chemical Research	7	8	A

Extraction Methods			
Adsorption Process	7	7*	C
Ether Process	1	4	C
Total	47	52	

(* In process of transferring to Site X)

Thursday, September 23, 1943

Beaton is again visiting the Research Products Company in Madison to discuss the marked differences in adsorption characteristics of two batches of their "Titanium Zeolite No. 2" received by us for testing.

Warren Johnson is being temporarily assigned to Clinton.

I wrote to Darrell Osborne in Arlington, Virginia, transmitting copies of correspondence between Compton and Tolman regarding our need for his services. I emphasized the importance of the role he would have in our project because of his knowledge of thermodynamics. I suggested that, if he can't come with us now, he might come after the six-month interval mentioned earlier by Tolman as necessary to train a replacement for him.

In a letter to Ralph Firminhac of Baraboo, Wisconsin, Willard expressed continued interest in the possibility of his coming with us.

Franck sent a memo to Allison, which has been prepared for him by Willard, regarding the feasibility of decontaminating the highly enriched uranium proposed to be used in a pile at Site Y. He indicates that either ether extraction or the sodium uranyl acetate method would be satisfactory, but points out that it would be necessary to wait two months or more before the 7-day U^{237} produced in the chain-reacting unit would decay to a sufficient extent to allow normal handling of the uranium. Franck further observed that in view of our manpower shortages, the most economical procedure would be to use the enriched uranium without the expectation of recovering it for other uses.

A batch of thorium carbonate pellets was shipped to Stoughton at Clinton Laboratories from New Kensington, Pennsylvania, where it was sent for encasing in aluminum cans. The pellets will be used in the pile to generate U^{233}.

Aunt Minnie Seaborg and her sister are visiting Chicago. They had lunch with Helen and me in our apartment.

We have decided to hold the weekly meetings of the Council of my section on Thursday, instead of Wednesday, evenings. Present in my office this evening were Brown (visiting from Clinton Labs), V. Cooper, Cunningham, Davidson, Dreher, Jaffey, Kirk, and Thompson. I discussed a number of general points. Davidson is starting a new group on

volatility, which includes Katz, Florin, and Bradshaw. Some new men who have arrived recently or will soon join us include Margolis, who will work with Thompson, and Magnusson, scheduled to arrive in about a week, who will work with Cunningham. Malm should be here next Wednesday; he also will work with Thompson. I asked that Kohman, Brown, and Thompson cooperate on a report to cover our present ideas on the problems connected with the extraction of plutonium from the P-9 pile material as a result of the higher concentrations of fission products and plutonium expected in this case. Cunningham will keep records of all plutonium isolated and distributed at Chicago. I also suggested the following group under Cunningham do product recovery and concentration: Magnusson, Belletire, N. S. Baumbach, Dixon, and Zvolner. A rough inventory indicates we have about 200 micrograms on hand in several samples. In addition, a total of 300 micrograms is being worked up, and we should receive in a few weeks from Werner at Clinton Labs about 200 micrograms recovered from semiworks runs. The metal still in the Argonne pile contains about one microgram of plutonium per pound of uranium, and Brown plans a conference with Apple about receiving Argonne-produced material to run in the Clinton extraction plant. A great deal of plutonium is in demand at the present time for work on the solubilities of the sulfate and molybdate which Orlemann is eager to use in purification procedures. Cunningham asked about the drawings for the Kirk-Craig microbalance, and I agreed to contact California again on this matter as soon as possible. Site W will require at least one such microbalance. I questioned Kirk about preparations to be made for measuring vapor pressures of the plutonium fluorides and bromides, to which he replied that the equipment is set up and can be made ready in a day. Magel will measure the vapor pressure of PuO_2 as soon as we can provide him with some material. I asked if anyone is prepared to determine whether a hydride of plutonium is formed--Davidson said that he and Katz are prepared to make the required pure hydrogen. It was agreed that Kirk and Davidson will prepare for cooperative work on the hydride. Kirk mentioned the melting point apparatus has been built and tried out, but he will need about 10 micrograms of plutonium--the temperature measurements will be made with an optical pyrometer using a filament of the metal. Kirk is also working on the hardness determination of the metal--micro equipment for this purpose is on the market. The present (Chicago VII) St. Louis neutron bombardment will arrive here Tuesday night. It consists of eight 85-pound batches of uranium metal and 600 pounds of UNH. Some of the metal will be sent to Eastman in California, some metal and UNH to Clinton Laboratories. Jaffey will start working up the material on Friday, using a crew composed of Gaarder, Halperin, Morris, Lincoln, and Malm. Katz will take notes on the Section C-I purification meetings, and Kroner will take notes on the extraction meetings.

The two big headlines in the morning's newspaper indicate that the Soviet army is within sight of Kiev and that there is a big Allied drive in Guinea.

Friday, September 24, 1943

This was the last day at the Met Lab for Vance Cooper, who is transferring to Clinton Laboratories effective September 27.

In response to my inquiry I received a wire from Charles D. Shane in Berkeley, the Rad Lab's Assistant Director, telling me that Kamen is rushing the drawings of the Kirk-Craig ultramicrobalance to completion and they should be shipped by Monday or Tuesday of next week.

Willard wrote Frederic Schuler in Madison that the University of Wisconsin has approved Schuler's receiving academic credit to fulfill his bachelor's degree requirements for his work at the Met Lab. Willard asked him to report here as soon as he can.

I was one of the addressees to receive a memo from Turkevich and Rubinson transmitting a table of corrections and additions to my isotope chart compiled last year, called the "California Seaborg Chart of Stable and Radioactive Elements." They surveyed Project reports and literature published up to September 15 of this year. Only the fission product region from 31(Ga) to 65(Tb) was covered. The "Table of Isotopes" that Helen and I are now working on will be a complete, up-to-date revision.

Roy Beaton reported on his visit yesterday to the Research Products Company. There he found that the difference in characteristics of the two batches of titanium zeolite is due to different sources of supply for the starting material. Beaton suggested that we provide a grant of about $500 to help finance further experimental work by the company.

The Beatons, Drehers, Baumbachs, Helen, and I went to a farewell dinner at Pete's Steak House for the Vance Coopers. This was followed by a visit to the Downbeat Room on Randolph Street. The Coopers are leaving for Knoxville tomorrow by car. Recently it was decided that Dreher, who also had been scheduled to transfer to Clinton, will remain in Chicago as a group leader.

The Germans have started to burn and dynamite large parts of Naples as they prepare to evacuate. They have scuttled thirty ships in the harbor.

Saturday, September 25, 1943

General Groves is in Chicago for a meeting on P-9 (D_2O) work.

Bernard Fries terminated at the Met Lab today and is transferring to Clinton Laboratories effective September 27. The house at Oak Ridge is not quite ready, so he will live in a dormitory temporarily while his wife and baby stay in Knoxville.

I sent Compton a suggested letter that he might send to Dr. Tolman of the NDRC in a further effort to obtain the services of Darrell Osborne. The letter points out that we are faced with the problem of reducing our product to metal with particular attention to its extreme purification from light elements and that we are seriously in need of a man with Osborne's training and practical experience in thermodynamics. The development of satisfactory metal production methods may be termed a bottleneck in our work. The purification and metal production problem is so difficult that it may well be the chief obstacle to the achievement of the ultimate aim of the Project.

In a letter to Fred Meyer in Pittsburgh, Pennsylvania, Willard expressed pleasure that Meyer will be able to join us and asking to be informed of his arrival date.

In more recruitment efforts Willard also wrote to Ralph Firminhac in Baraboo, Wisconsin, referring to letters which crossed in the mail and enclosing an employment application blank. Willard identified in broad terms the research work that he would be doing in inorganic chemistry. He sent two other letters: one to R. H. Goeckermann in Madison, Wisconsin, expressing disappointment that Goeckermann will not be able to join us at the present time and saying that we shall look forward to having him with us in January; and the other to Dr. J. H. Mathews at the University of Wisconsin, thanking him for the approval of Schuler's academic credit and assuring him that Schuler will be able to make a valuable contribution to our work.

I attended a meeting with Apple, Sutton, Willard, and Brown concerning difficulties experienced at the Clinton Laboratories semiworks with formic acid reduction in the Bismuth Phosphate Process.

Helen and I went to the home of Enrico and Laura Fermi (5537 S. Woodlawn) for an evening with the Met Lab Book Club members.

The Soviets have repossessed Smolensk, which the Nazis evacuated in the face of advancing Soviet troops.

Sunday, September 26, 1943

I played golf. Helen and I had dinner at the Esquire restaurant (876 E. 63rd) with the Ghiorsos and played cards afterwards.

The St. Louis (Chicago VII) cyclotron neutron bombardment of uranium ended at 113,600 microampere-hours. It started August 24.

Monday, September 27, 1943

Frederic W. Schuler, who has been a student at the University of Wisconsin, started work in Section C-I today.

Willard received a memo from Nathan Sugarman, Coryell's Associate Section Chief, asking that his section be given 25 pounds of nitrate from the present St. Louis bombardment. Their stock is depleted considerably because they shared the neutron-bombarded nitrate on hand with the group that moved to Site X.

I notified Doan that Dreher will replace Vance Cooper as the man helping me in regard to transactions and business involving the Clinton Laboratories.

The evening seminar of the Chemical Division was held at 7:45 p.m. in Room 251, Ryerson Laboratory.

The Soviets report that the Germans are fleeing from Kiev. American troops and tanks are gaining in their Naples drive.

Tuesday, September 28, 1943

This is the last day at the Met Lab for Dan Koshland, who will start at Clinton Laboratories Thursday, September 30.

R. A. James outlined for me a proposed method for detection of new heavy isotopes by bombardment of 94^{239} with 12 Mev deuterons. We think that the probable new heavy isotopes produced by such a bombardment would be 95^{240}, 94^{240}, or 95^{239}; 94^{240} would be the easiest to identify by isolating the plutonium fraction by the fluoride method and looking for an unidentified beta emitter or alpha emitter with range different than that of 94^{239}. Element 95 could be sought for in other separation fractions from the fluoride method, e.g., the first by-product precipitate from a solution in which plutonium was oxidized, on the assumption that element 95 would differ from plutonium in oxidation potential or rate of oxidation. Or, if the fluoride of element 95 is soluble, it would be found in the supernatant from the first fluoride precipitation. James adds that, if all or any of these isotopes are short-lived, their daughters would have to be detected.

In a memo to Allison, C. M. Cooper summarized monthly progress in the Technical Division. He stated that the separations step of the Bismuth Phosphate Process is now in reasonably good shape and that during the past month emphasis has been placed upon decontamination with recrystallization of precipitates, which give considerable promise of bringing decontamination factors up to the desired level.

Some time ago we arranged with Hamilton for a supplementary neutron irradiation at the 60-inch cyclotron in order to augment our supply of plutonium and fission products. Today I wired Hamilton that Hilberry and I believe October 10 to 15 is a good time to stop the Berkeley bombardment. I am referring to the irradiation of 1,100 pounds of uranium metal and 300-400 pounds of UNH with the neutrons produced from the bombardment of beryllium with 15 Mev deuterons. I suggest immediate shipment by truck with cooling of activity on the way and redistribution of the

material after it arrives here. I am hoping that the metal that is directly in line with the deuteron beam will have a high concentration of Np^{237}.

During the evening the Chicago VII material from the seventh St. Louis bombardment arrived.

The Research Assistants meeting took place at 7:00 p.m.

American troops are opening a new drive in New Guinea; they are moving toward the Japanese shipping base of Madang.

Wednesday, September 29, 1943

Another Research Assistant started today. He is John G. Malm, who received a bachelor's degree from Wisconsin this year. Willard, who remembers him as a good student, interviewed him at the University of Wisconsin on September 11. After a security talk from Stearns this morning, Malm came to the New Chemistry Building, where Willard described the Project in general and the work being carried out in chemistry and then introduced him to James, with whom he will be working for awhile. He was assigned a small section of a laboratory bench in Room 2, the large room at the north end of the building where Davidson, Hagemann, Katz, and others are doing "dry chemistry" work. Next week, however, he will go to North Stands at Stagg Field and help with the ether extraction of the Chicago VII material, probably the last such extraction to be conducted here at Chicago.

Cummingham has completed the isolation of 93^{237} that he began late in August. See Figure 7. Today he dissolved the hydroxide precipitate in nitric acid, transferred part of the solution to a weighing boat, ignited it, and then weighed it. The oxide was then dissolved in concentrated HNO_3, and the boat reweighed, this gave a weight of 1.47 micrograms for the oxide. The nitrate solution of the oxide was then delivered onto a platinum disc and counted for alpha particles. Two separate determinations gave half-life values for Np^{237} of 9.5×10^6 years and 7.5×10^6 years if one assumes that the compound prepared was Np_2O_3.

I informed Allen J. Holzinger, the Section Payroll Chief, that Frank Belletire of my section is taking an afternoon course at the University on Tuesdays and Thursdays and will make it up by punching out at 5:30 p.m. on those days, if Holzinger approves.

Franck sent a memo to Davidson confirming his appointment as Group Leader of the Volatility Group in my section.

We held our evening meeting of the Extraction Group of Section C-I in Room 209, Eckhart Hall. Thompson reported that the high loss of product in the first decontamination cycle of the Bismuth Phosphate Process may be attributed to the presence of zirconium and columbium which precipitate with the extraction of bismuth phosphate, but do not dissolve in HNO_3--they may adsorb the plutonium. Thompson then outlined the extraction, decontamination, and concentration methods being worked on by his group. Dreher reported that the compound $BiAsO_4$ as a carrier is less soluble than the bismuth

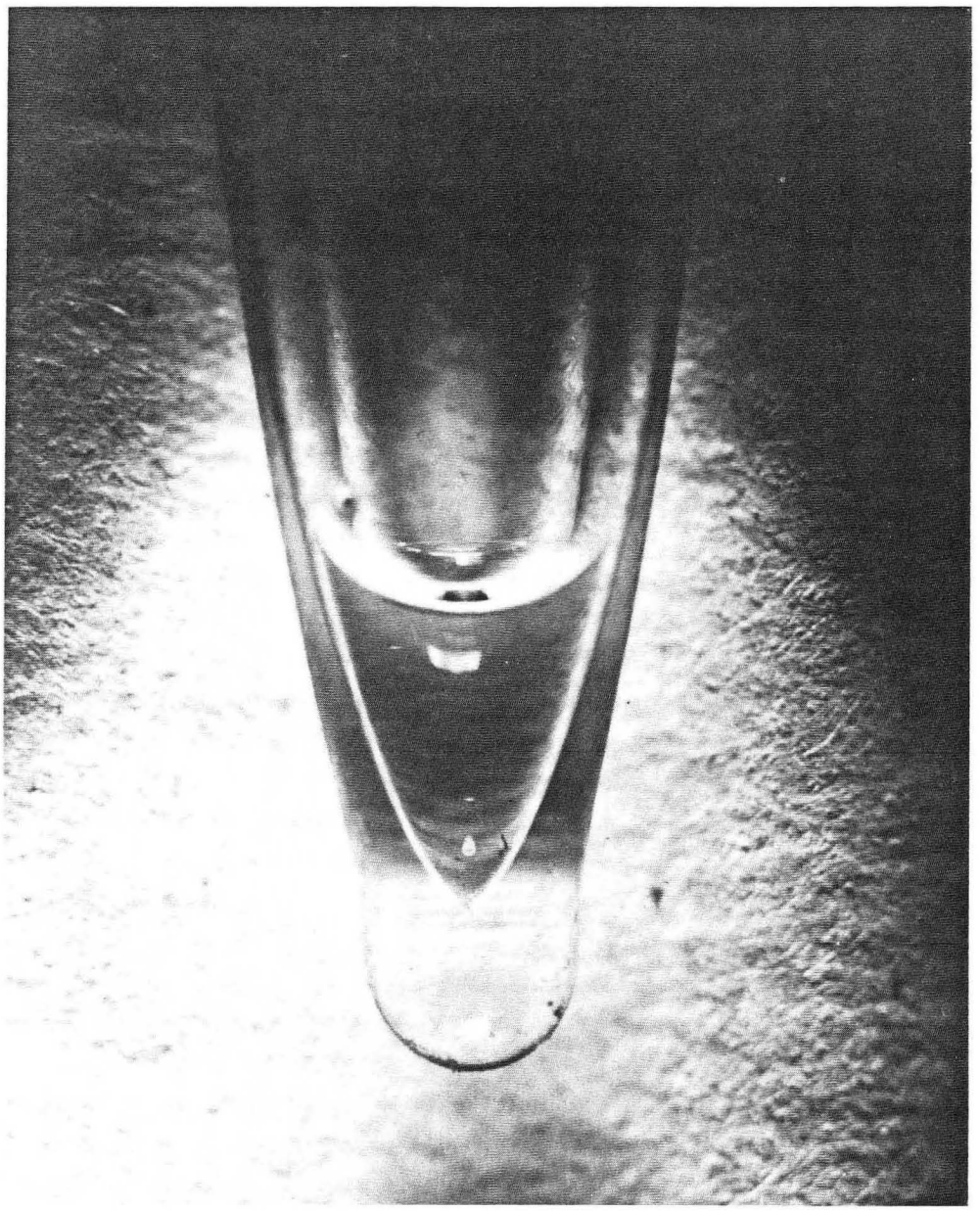

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Figure 7. Neptunium Hydroxide. September 28, 1943.

phosphate and much harder to dissolve. Dam reported on $U(C_2O_4)_2$ method for concentration, indicating that the $U(C_2O_4)_2$ precipitate can be dissolved in K_2CO_3 solution and decomposed by H_2O_2, after which a new $U(C_2O_4)_2$ precipitate can be made. This method probably will give adequate concentration. James discussed alternate methods for extraction. The engineers think $NaUO_2Ac_3$ produces too heavy a precipitate. $U(C_2O_4)_2$ can be used for extraction; further work is under way on this method, as a single precipitate carries 90% of the plutonium. The $U(C_2O_4)_2$ method can be run into the sodium uranyl acetate method by dissolving the $U(C_2O_4)_2$ in HNO_3 and oxidizing with dichromate. The solution is heated to oxidize the product and sodium uranyl acetate precipitated. Beaton discussed his troubles in the adsorption process with the inferior decontamination obtained from a new batch of titanium zeolite.

The Americans destroyed a Japanese convoy at Wewak, New Guinea, on Monday, according to today's newspaper.

Thursday, September 30, 1943

Today Cunningham took the remainder of the neptunium solution not used for the specific activity determinations yesterday, evaporated almost to dryness, dissolved it in water, and then added 30% H_2O_2. On centrifugation a small amount of precipitate was observed in the bottom of the tube, but Cunningham was not certain that this is a peroxide. See Figure 8.

The plans for the Kirk-Craig ultramicrobalance arrived from Berkeley today.

In a letter to Dr. Raymond Keller, Department of Chemistry, University of Michigan, Willard indicated that the Met Lab does not discriminate against women chemists and that if Isabelle L. Karle would like to consider work with the Met Lab, she should forward a statement of her training, interests, etc. Willard also asked Keller if he knows of any especially good bachelor's degree men who might be interested in coming with us.

Willard also wrote to Jacob Sedlet in Madison that we are disappointed that he will not be able to join our staff at the end of the summer semester, but we hope he can come in January.

I held an evening meeting in my office of the Council of my section attended by Cunningham, Davidson, Dreher, Ghiorso, Kirk, Orlemann, Thompson, and Willard. I mentioned that Thomas O'Donnell's shop in Ryerson is starting immediately on the construction of three Kirk-Craig microbalances. I also announced that Warren Johnson is going to Site X to be Chemical Director there and Thorfin Hogness (who is Professor of Chemistry here at the University) will take Johnson's place as Associate Director of the Chemistry Division at Chicago. Donlan has requested another room, but we have no space--perhaps he can place desks in the hall near the fire exit to 56th St. Kohman has suggested revising the Seaborg chart of

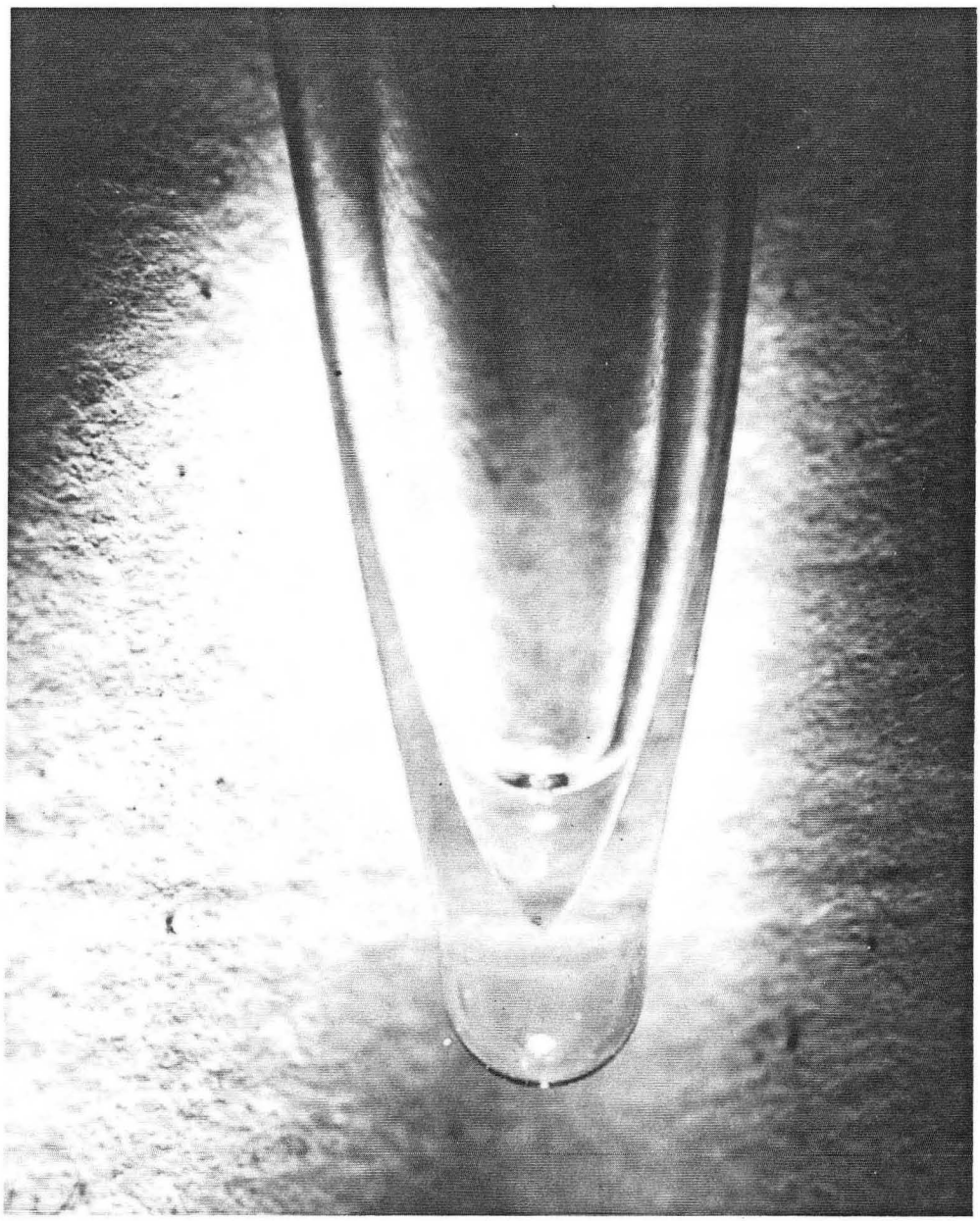

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Figure 8. Neptunium Peroxide. September 30, 1943.

radioactivity. The system he has suggested may be very good but would require more space than the present chart. The main argument for the present chart is that we have become accustomed to it. It may be possible to use cupferron as the basis for the concentration method at Site X, and Thompson will take some there on his next trip. The advantages of this concentration method are that the chloroform solutions can be ashed to very small volumes and that very small amounts of chloroform solutions can be used for the extraction. Cunningham reported that lusteroid tubes have been found to hold up large amounts of plutonium. Zvolner has found 58 micrograms of plutonium on some tubes he is recovering. It was agreed that, since the amount of Pu^{238} on hand is fairly small, this would be a good time to throw away all of it so as not to contaminate our Pu^{239} samples. I suggested Perlman also discard any Pu^{238} that he has on hand. I asked Ghiorso to measure the range of Pu^{239} alpha particles using a sample of about 2,000 counts per minute--such measurements might be used to indicate Pu^{238} contamination in samples. I suggested finding out if a uranous basic acetate precipitation can be carried out from UNH solutions and that this be tested for carrying of plutonium--this would be an excellent extraction step for the sodium uranyl acetate decontamination method. Dixon is preparing about 5 micrograms of plutonium acetonyl acetonate, and Cliff Smith will attempt to determine its volatility. Jensen is working with thorium molybdate which has rather nice properties; carrying for plutonium has not been tested yet as a potential purification procedure. Howland, using 40-50 micrograms of plutonium, has been unable to crystallize out the product in 6 N H_2SO_4 solution. Smith is now ready to make another run on the vapor pressure of plutonous bromide. In view of the space situation the U^{233} work remaining here in Chicago and temporarily under Thompson's supervision will have to be carried out in Davidson's laboratory room (number 2). Ether extraction under Jaffey's direction is starting on the UNH from Chicago VII.

After weeks of searching, Helen has found us a new apartment. This afternoon we moved from our old one on the top floor of 6128 Woodlawn Avenue to the larger apartment on the first floor (left front) at 5418 Woodlawn Avenue. Stan and Alice Thompson have been living in an apartment in this building (it is just across the hall) for some time and helped Helen locate ours. Helen and I had dinner with the Ghiorsos in the house where they are now living at 1219 E. 54th St.

The Allies are close to Naples having driven the Germans in full flight across the coastal plains near Mt. Vesuvius.

Friday, October 1, 1943

Jaffey started processing the material from the Chicago VII St. Louis bombardment under North Stands with a crew composed of Gaarder, Halperin, Morris, and our new man Malm. Fortunately we are having good stiff winds these days which ventilate the area. The material has received a total bombardment of 113,600 microampere-hours of beryllium-deuteron neutrons.

George E. Creek, who received his M.S. from the University of Illinois in 1939, started working in Section C-1. He has been employed by the Pittsburgh Testing Laboratories in Chicago.

This was the last day at the Met Lab for George E. Boyd, Chief, Analytical Section, who is transferring to Clinton Laboratories as a Senior Chemist, effective October 4.

Sherman Fried phoned me from Northwestern to tell me that his boss removed his draft support when Fried notified him that he is terminating. Consequently Fried has been reclassified to 1A status and has had to report for his physical examination. I told him not to worry as I will have the Met Lab people take care of the problem.

In a letter to George W. Watt at the University of Texas, Willard expressed pleasure that Watt will join us by November 1 or earlier. Willard enclosed with the letter my Chemical Reviews article ("Artificial Radioactivity," August, 1940) and the set of notes on the lectures on Nuclear Chemistry that I gave during May-July 1942 but cautioned that while the material is not "classified" he should not intimate to others any ideas which it may give him as to the nature of our work. Willard also wrote to Firminhac in Baraboo, Wisconsin, suggesting that he visit us to discuss job possibilities if he is in Chicago next week.

Lombard Squires met with me and asked me to designate an isolation process for the Hanford extraction plant. I suggested the peroxide method, even though the choice is necessarily premature, and promised to send him a flowsheet.

Helen had lunch out with Wilma. Later she and I had dinner at Schall's Steak House (5475 S. Lake Park) and then visited with the Ghiorsos.

Saturday, October 2, 1943

M. D. Peterson is here from Site X for consultations on separation processes.

A letter arrived from George Manov in reply to mine of September 22, which I find to be of considerable interest. "It was a pleasure to read your letter," he said, "for it's been quite a while since we corresponded.

Perhaps we ought to kid each other--we have both traded the climate of God's country for something less desirable; you, for a blast of wind and I, for a Turkish bath. Still, the war makes changes in all of us." He went on to say that his job with the Bureau of Standards is to assist the Navy with certain specific details concerning their small aircraft carriers, which have been written up in "Time" magazine. He asked if I would write him a little more about the vacancy here in my section so that he will be able to discuss the matter of a possible transfer with his chief. He concluded his letter with the following:

"Your letter brought back a flood of memories, Glenn, of school days--of Gamma Alpha--of the Ph.D. "Exuberance Meeting" the 15 of us held back in the hills the night after we got our degrees, and how we invited Lat who thought he shouldn't come because he was a faculty member, so we sent him a big bottle of Scotch. Yes, and the time you, Dorr Etzler, Leon Jehle, and I went to Yosemite in two model-T Fords, one of which boiled over every half mile; how the second Ford broke a fan blade which promptly punched a huge hole in the radiator. We heard a loud noise and saw a cloud of steam which completely hid the car from view. Too, I remember how we walked, dirty, dusty, with a week's growth of beards on our faces, into the bank at Livermore. The cashier thought we had come to hold up the joint. Yes, I was certainly glad to hear from you. It will be a pleasure to hear from you again later; drop me a line at your convenience."

Last Thursday in his continued efforts to reduce PuF_4 to metal Baumbach molded a small (0.05 inch diameter) inner crucible of thoria. He has attempted to reduce PuF_4 with various mixed reductants such as sodium and barium or strontium and sodium in a double crucible. Today in an experiment with about 6 micrograms of PuF_4 with magnesium and sodium, he produced a cokey glossy product that was spread over the bottom of the crucible. He counted one small (possibly a microgram) fragment and found that the counter was saturated (11,000 c/m). The fragment reacted vigorously with HCl, and Baumbach concludes that this product is probably the most pure metal sample he has yet obtained.

Today we heard most distressing news. Samuel Ruben, who was the co-discoverer of carbon-14 with Martin Kamen in 1940, was fatally gassed with phosgene while working on a gas warfare project in the Chemistry Annex (the Rat House) on the Berkeley Campus; a glass tube containing phosgene broke when it was plunged into liquid air. All of us who knew Sam are terribly upset. He was a graduate student at Berkeley at about the same time as I, working with Professor Willard F. Libby. He left a wife and three children including a three-month old baby daughter.

Helen and I went to the Beatons' new apartment (on E. 78th Street near Lake Michigan) for dinner--the Willards were also there. It was not too happy an occasion because of the news about Sam.

Naples has fallen, and the Allies are pushing on toward Rome. The Soviets are making a push toward Minsk, although they are still some fifty miles away.

Sunday, October 3, 1943

I played golf.

Helen and I ate dinner at Isabell's Restaurant because the gas has not yet been turned on in our new apartment. Afterward we stopped by the Ghiorsos'.

Monday, October 4, 1943

General Groves is in Chicago.

Lawrence B. Magnusson started work in Section C-I as a Junior Chemist under Cunningham. He comes from the Chemical Warfare Service, Huntsville Arsenal, Alabama. Patricia D. Walsh and Lorraine Heller are now working as Research Assistants for Ghiorso in Section C-I. Both come from the University of Chicago where they have been electronics instructors.

Today I signed a patent application based on the ideas expressed in my letter to Oppenheimer of November 3, 1942, that plutonium must be extremely purified from light elements. Our plutonium purification program resulted from the recognition that alpha particles from the plutonium might react with the light elements to produce neutrons and possibly trigger a premature chain reaction.

Compton received from Groves a directive concerning the P-9 work at Chicago in which he states that at a meeting of the Military Policy Committee it has been decided "that our efforts not exceed the construction of a low-powered heavy water pile for use as a general experimental tool and the setting up of a group of scientists assisted by a small number of engineers to study the general problems and to prepare the way in the event that it should become necessary to carry on rapid development." Groves also states that with regard to the detail of our relations with Montreal, the situation is not yet entirely clear. It will be necessary for the time being to clear proposed communications through his office until a definite procedure has been set up.

There was an evening seminar of the Chemical Division at 7:45 p.m. in Room 251, Ryerson Laboratory.

"Chemical Research--Production and Extraction of Plutonium. Report for Month Ending September 30, 1943" (CN-979) is being issued. It states that further development of the Bismuth Phosphate Process has been carried out by V. R. Cooper, Dean, Dreher, Halperin, G. Johnson, Kroner, Miller, Morris, and Thompson. Halperin has studied the use of U^{+4} as a substitute for Fe^{+2} or HCOOH for reduction of plutonium oxidized in the dissolving step and finds that it may be superior. He also finds that it gives nearly quantitative reduction of plutonium in the decontamination cycles. The effect of concentration, temperature, mode of addition of reagents, and types of equipment on reduction have been studied. Cooper, Miller, and Morris have conducted further investigation of variables that might affect

carrying of plutonium by bismuth phosphate. Using unusually small amounts of Bi^{+3} carrier in order to increase the sensitivity of carrying to other variables, they have found that the efficiency of carrying of tracer amounts of plutonium increases with decreasing UNH concentration. Pyrophosphate, which might be present in the H_3PO_4 reagent used, appears to inhibit carrying of plutonium in the presence of Fe^{+2} or stainless steal. Miller has conducted microchemical experiments on the carrying of plutonium with low Bi:Pu ratios from an extraction solution containing 20% UNH, "W" concentrations of fission products, holdback carriers, and 0.02 M Fe^{+2}. He finds 96% carrying at a Bi:Pu ratio of 20:1. At a ratio of 6.4:1, carrying is still as high as 75%. Kroner has investigated the possibility that small amounts of HF might improve carrying and decontamination in the extraction step but finds no beneficial effects. Dreher has studied the possibility of obtaining improved decontamination in the extraction step by first oxidizing the plutonium and making a by-product bismuth phosphate precipitation to carry fission activity without the plutonium, followed by reduction and product precipitation. Such a procedure gives essentially the same decontamination as the usual one-step extraction procedure. He also has investigated oxidizing agents for use in this procedure and finds poor results with 0.02 M $NaBiO_3$, but good results with 0.01 M Ce^{+4}. G. Johnson has done microchemical experiments on the solubility of plutonous phosphate at 75°C, 1 N H_2SO_4, and 0.4 M H_3PO_4 and finds that solubility varies from 60 mg/l with no UNH present to 770 mg/l with 19.6% UNH present. Dreher reinvestigated the behavior of neptunium in the Bismuth Phosphate Process in view of process changes made since the early work and finds that neptunium in the reduced state is partially carried by bismuth phosphate but not at all in the oxidized state. Dreher also has studied the effect of the order of addition of Bi^{+3} and H_3PO_4 on decontamination and product recovery in an extraction and two decontamination cycles. He finds that adding the Bi^{+3} to the H_3PO_4 solution gives better product recovery but poorer decontamination than the reverse order of addition. Morris finds no effect of "W" concentrations of columbium on decontamination and product recovery in extraction and decontamination cycles. Miller has studied the use of the uranous hypophosphate procedure for the concentration of plutonium following decontamination. He has carried out experiments on the effects of a number of variables which show that in the presence of iron the efficiency of carrying by UP_2O_6: (1) is decreased by increasing bismuth phosphate concentration in solution at 0.01-0.02 M $P_2O_6^{-4}$ but not at 0.1 M $P_2O_6^{-4}$; (2) seems to have a maximum value at 0.1 M $P_2O_6^{-4}$; (3) at 0.1 M $P_2O_6^{-4}$ is nearly independent of the Fe^{+3} concentration; (4) falls off rapidly when less than 1 mg of U^{+4}/cc is used in the presence of 0.05 M Fe^{+3}; (5) is not improved by the addition of PO_4^{-3} or CNS^{-1} to complex the iron; (6) is distinctly better when a co-formed rather than a preformed precipitate is used. Miller also has conducted preliminary investigations which indicate that UP_2O_7 is inferior to UP_2O_6 as a carrier for plutonium, whereas $U(IO_3)_4$ appears promising.

The Ghiorsos dropped by to see our new apartment in the evening.

The American Air Force bombed Haiphong and shot down nearly fifty Japanese planes.

Tuesday, October 5, 1943

A new semiworks setup, to replace the one moved with W. Q. Smith's section to Clinton Laboratories, is being assembled in the cyclotron chemical laboratory where Smith's people worked. Today Jaffey and I learned from Charles Cooper that the Chemical Division has permission to use the cyclotron chemical laboratory hood for the next month or six weeks while semiworks equipment is being erected. Cooper said that it will require some program juggling to get our work in, but arrangements can be made although not on demand.

In a letter to Eastman I suggested that Brewer's visit to Chicago be postponed, at least for a month or two. I told him I had heard the news about Ruben and hardly knew what to say. "I still find it very difficult to believe," I wrote. "A number of us who knew Sam so well have felt practically ill for the last few days."

Franck wrote Thomas at Monsanto that Kirk thinks about 0.5 mg of plutonium is needed to prepare 20 micrograms of pure metal in the one lump needed for a density measurement. With luck, he may be able to do it with 0.2 mg, and we will try our best to scrape that amount together as quickly as possible, but it will certainly take several weeks to do so. Franck suggested our chances will be better if Site Y can give us part of their supply from Berkeley. He estimated that making the measurement will require six weeks from the time we get the material together. The object of this is to make a plutonium metal density measurement before much material starts flowing from Site X.

The eighth St. Louis cyclotron neutron bombardment of 775 pounds of UNH (Chicago VIII) started today. The run should take about three weeks to accumulate 100,000 microampere-hours of beryllium-deuteron neutron exposure.

The Metallurgical Laboratory Progress Report for September 1943 (MUC-NH-654) is being issued. The work of Section C-I was summarized as follows.

Again the major effort of the separation group has been on the Bismuth Phosphate Process with particular reference to the development of new reducing agents. Further studies have been made on the carrying of the bismuth phosphate which has been unsatisfactory in semiworks. Studies show that even on a laboratory scale the agitation during digestion is a critical factor, but that with proper agitation carrying is satisfactory. More effort was placed during this month on the concentration procedure necessary for Site X and Site W. This program will continue during the coming month.

In the purification and basic chemistry of product, further work has been done on the production of metal on a microscale. The work on volatilization and solvent extraction methods was pressed during the month. This work will also continue during the coming month.

In the adsorption-uranyl acetate alternative procedure, the work has been largely confined to obtaining the data necessary to put the process on a semiworks basis. This work is continuing.

10/5/43 (cont.)

The report also notes that at Clinton construction of the laboratory facilities has proceeded satisfactorily and the 100 Area (pile area) is well along toward completion. The indications are that it will be ready to be turned over to operations some time in October. The 200 Area (chemical extraction plant) construction has proceeded satisfactorily, and the plant should be turned over to operations some time in November. During the month, the operations staff has been reporting for duty and preparations for start-up are proceeding. Total expense for the month of September was $438,008. Total personnel on the Project on September 30 was 905 (382 academic).

Stan Thompson and I left the Chicago Englewood Station at 11:50 p.m. on the Penn R.R. to Cincinnati, where we will continue on the L&N R.R. to Knoxville, in order to attend the Bismuth Phosphate Process Progress Meeting scheduled for Friday at Clinton Labs. Gordon Leader of Sugarman's group in Section C-III is along, and the three of us are sharing a drawing room.

The top headline in today's paper is from the Pacific, where U.S. ships are intent on thwarting Japanese withdrawal from Kolombangara Island in the Central Solomons.

Wednesday, October 6, 1943

Our train arrived in Knoxville in the afternoon--late. Stan and I then played 12 holes of golf at the Municipal Golf Course. Although I had a migraine headache, I managed to get a postcard off to Helen. We are spending the night at the Whittle Springs Hotel.

There was a Project Council Policy Meeting in Chicago at 9:30 a.m. Present were Allison, Franck, Hilberry, Hughes, Jeffries, Lawrence A. Kimpton (Chief Administrative Officer), Mulliken, S. L. Rapier, Stearns, Stone, W. W. Watson, Whitaker, and Wigner. The Laboratory's safety program was discussed by Rapier. There was a review and discussion of the Health Division's programs, and Watson presented the General Physics Division program under the new plan of reporting. Allison reported on his visits to Site Y with Fermi and told of Site Y's plans to set up a small homogeneous pile with 10-12% enriched uranium nitrate for determination of critical size but not to run for an appreciable time. While there they discouraged the plan to build a 10 kw homogeneous H_2O-moderated pile at Site Y to operate at 10 kw as an intense neutron source. Wigner commented that it would be more logical if we constructed it. When Franck mentioned they could send men here to learn the trade, it was noted that present rules might prohibit this. Franck mentioned my estimate that the Met Lab needs $20,000 worth of platinum. It was agreed this should be ordered.

Thursday, October 7, 1943

In the morning Stan and I played 18 holes of golf at the nearby Knoxville Municipal Golf Course. I shot a 102. After lunch at the

Whittle Springs Hotel we left at 2:00 p.m. for the Clinton Labs, where we checked into the Guest House in Oak Ridge, and then conferred with Perlman and his people during the late afternoon.

I ate dinner with Iz and Lee Perlman at their home at 300 E. Fernhill Lane. (See Figure 9.)

In Chicago there was an evening meeting of Section C-1 Research Assistants at 6:45 in Room 209, Eckhart Hall.

Friday, October 8, 1943

This was the last day at the Met Lab for Humbert Morris, who is transferring to the Clinton Laboratories as a Junior Chemist under English, effective October 11. Morris is single and will therefore live in a dormitory.

At 9:30 a.m. Thompson and I attended the semi-monthly Bismuth Phosphate Process Meeting at Clinton Laboratories. We presented data on the scouting work done since the last meeting. This has been mainly new concentration methods, the most promising being the uranous oxalate procedure which can be used either in HNO_3 or HCl solution. The use of HCl for dissolving bismuth phosphate produces, in a single step, a concentration factor of about 80. We also presented the results of further investigation of the wet fluoride concentration method that has resulted in a new coupling procedure involving the use of Ce^{+4} for dissolving the fluoride precipitate, which makes possible a concentration factor of 50 for each concentration cycle. We reviewed the work on development of alternate oxidation and reduction procedures with Ce^{+4} and oxalic acid. We mentioned our cooperative work with Zachariasen on x-ray crystal studies and the techniques being developed for working with samples of 500 micrograms, which will make it possible to obtain data on the structure of bismuth phosphate crystals containing 2-3% plutonium.

Saturday, October 9, 1943

Thompson and I again spent the day at Clinton Laboratories talking to the people in Perlman's section, the chemical engineers, etc. Around midnight we took a Southern R.R. train from Knoxville to Cincinnati, where we will transfer to the New York Central R.R. to Chicago.

It was the last day at the Met Lab for Dan Miller and John Dam, who are transferring to Clinton Laboratories as of October 11, 1943. Both will live in dormitories. Dam's wife will join him when a house becomes available.

Sunday, October 10, 1943

Stan Thompson and I arrived back in Chicago at 2:30 p.m.

When I returned home, Helen told me what she had done while I was away. On Wednesday she had dinner at the Ghiorsos' and then played pinochle

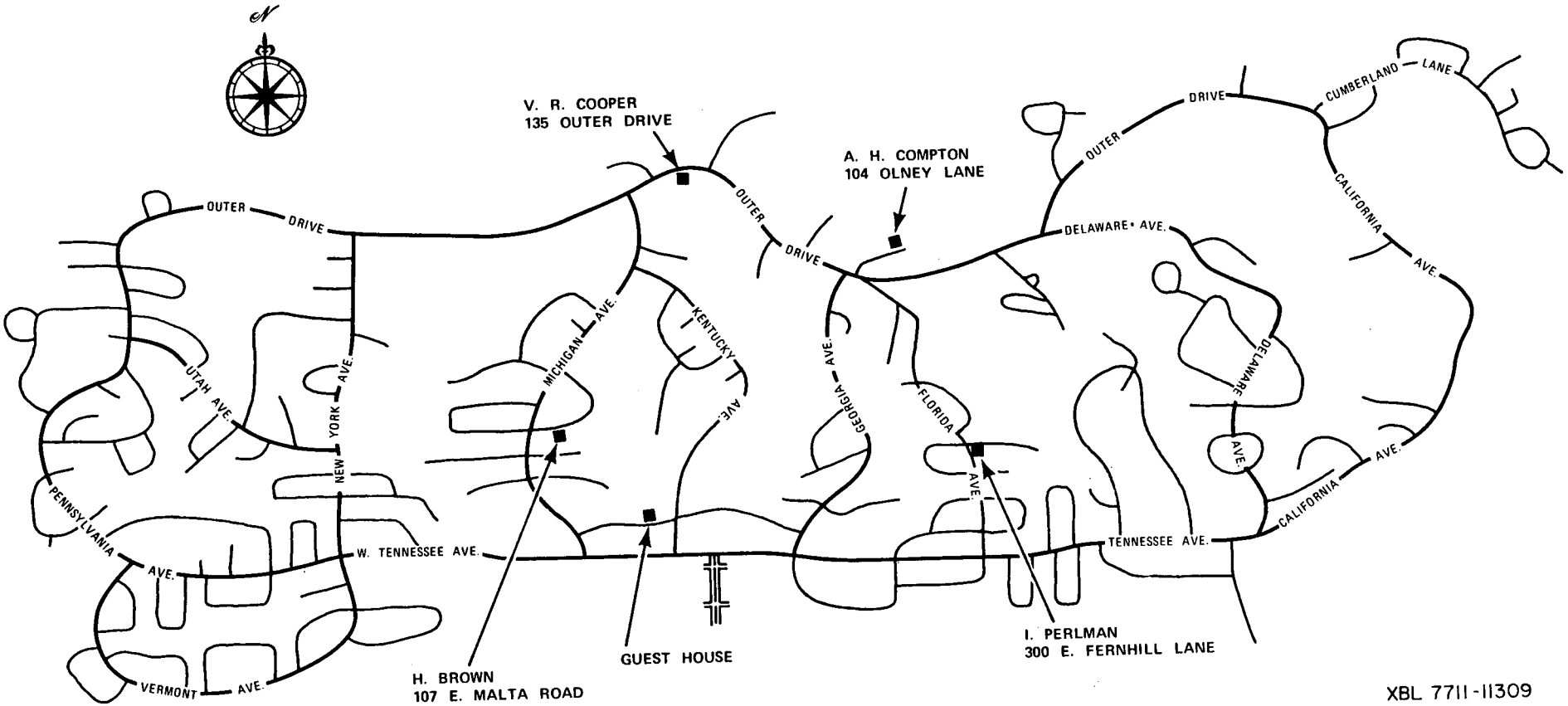

Figure 9. Map of Oak Ridge. 1943-44.

with them. She went shopping with Wilma Thursday, had dinner with Wilma and Al at Isbell's, and then again spent the evening playing pinochle. Yesterday she spent the entire day working on the new "Table of Isotopes."

The Ghiorsos came over to our apartment this evening to hear about the new city of Oak Ridge.

Monday, October 11, 1943

Friday my office received a reply from Fred Albaugh. He wrote that he has changed his mind and decided to accept my offer and that I should direct a request for his leave of absence to D. E. Carr, Manager of Research of Union Oil Company. He further indicated he would be unable to leave the company before December 1 or 15. Albaugh commented on the fact that the collection of UCLA and Standard Oil names which I have on my roster is no small inducement to join up with us. He went on to say, "However, something tells me that this deal is more in the nature of 80 hours per week of work than an alumni reunion dinner dance."

This morning I wired him of my delight in learning that he has decided to come with us. I asked him to return immediately the application blank we are sending him so that a definite offer may be made before he asks for a leave of absence.

I also sent a wire to Hamilton in Berkeley asking that he please try to keep track of our uranium metal close to the target, especially right in front in line with the beam. I am interested in identifying material with the highest content of Np^{237}.

When I talked with Cunningham, I learned that last week he prepared a compound believed to be NpF_3. Interestingly, the air-dried compound appeared almost colorless but turned bright yellow when ignited. Last Friday from specific alpha activity measurements, he calculated the maximum half-life of Np^{237}, assuming the compound to be NpF_3, to be approximately 6×10^6 years.

Last Wednesday Norm Davidson wrote a letter to the Director of Research of the Harshaw Chemical Company in Cleveland, Ohio, asking for information on possible impurities in their anhydrous hydrogen fluoride which we are using. Davidson is particularly interested in water and reducing or oxidizing impurities such as SO_2.

Willard attended, in my absence, the meeting of the Chemistry Division Section Chiefs held on Wednesday. Recruitment of personnel was discussed. It was agreed that we have exhausted our sources for new personnel (acquaintances, etc.). It was felt that the only way to get new personnel is to have a man on the road who interviews people at universities and meetings. Furthermore, this man should have the right to hire qualified people on the spot in order to compete with various companies who, by operating in this fashion, took some good prospects away from us at the Pittsburgh ACS meeting. Franck agreed to approach Stearns to see if the Personnel Office will hire a man or appoint someone already on the staff to travel around the country recruiting people.

Stearns followed up on this on Thursday by writing a memo to Wayne Johnson stating that the need for chemists is very acute at this time and suggesting that Johnson initiate a program to canvass all colleges and universities, the American Chemical Society, and industry for names of prospective candidates. Stearns also mentioned the possible need for a man to do full-time recruiting in the field.

Willard told me that on Thursday he asked Stearns to increase J. A. Crawford's salary from $200 to at least $225 per month in the interest of maintaining the proper equivalents of salary within the Project.

Also on Thursday Franck wrote Doan at Clinton Labs saying that 220 pounds of uranium metal exposed at St. Louis would go to Clinton in a few days for use by the engineers and Perlman's section.

I also learned that on Saturday Willard recommended to Wayne Johnson that Roy Heath be hired for work in our section. He mentioned that he has known Heath rather well since they were first colleagues on the staff of the Chemistry Department at the University of Wisconsin last year and that Heath is an able research worker with extensive experience in the field of inorganic chemistry, in particular with anhydrous HF.

"Chemical Research - Special Chemistry of 94 - Analytical. Report for Period Ending October 9, 1943" (CK-993) is being issued. Besides covering spectrochemical analysis under Mark Fred in Boyd's section, the report describes the work of Hagemann and Orlemann on (1) the bromine volatilization method for determination of oxygen, which is concerned with discovering satisfactory materials in which to carry out bromination and volatilization--glazed porcelain appears to be the most promising; (2) the carbon tetrachloride, phosgene method for determination of oxygen, which has involved preliminary experiments using uranium.

The Technical Division report for week ending October 9, 1943 (Memo No. 129), states that two 5 microgram samples of plutonium oxide were received from Cunningham for vapor pressure measurements. Apparatus for the measurement of vapor pressures of metals and oxides has been redesigned and construction started. Magel and Cefola, who have transferred to the Technical Division, are working on this project.

The evening seminar of the Chemistry Division was held at 7:45 in Room 251, Ryerson Laboratory. The program included discussion by Beaton on adsorption work, Thompson on the concentration methods to be used following the use of the Bismuth Phosphate Process for extraction and decontamination of plutonium, and Howland on our program of investigation of the fundamental chemistry of plutonium.

Vila, former Japanese base on Kolombangara Island, has been occupied by American troops with no last-ditch oppostion.

Tuesday, October 12, 1943

Bert Schoenberg started working in Ghiorso's group. He has been doing research in electronics for the Signal Corps at the University of Chicago. Fred Meyer also began in Section C-I. He comes from the Explosives Research Laboratory in Pittsburgh where he has been a Research Associate and received his Ph.D. from the University of Wisconsin in 1940.

Joe Hamilton called this morning in response to my telegram. He said that he plans to stop our bombardment on the 18th. We reaffirmed our plans to ship both the metal and the UNH to Chicago. The metal will be unloaded, and the nitrate will return to Berkeley; thus giving it a chance to cool before being processed. Joe said the shipment should arrive about October 30.

Saturday my office received a memo from W. W. Watson asking if we would mind sharing the chemical preparation room adjacent to the cyclotron with Snell and Wollan, as they need it to handle samples irradiated in the cyclotron. The people in Sutton's section have a chemical room in the West Stands and have no need for the chemical preparation room. Today I replied saying that I have no objection to Snell and Wollan using the room as we do not plan to be using the room continuously ourselves.

Jensen has been conducting experiments the last few days to determine the solubility of plutonium molybdate. His procedure consists of mixing 2.25 λ 5% ammonium acetate and 0.25 λ $Pu(NO_3)_4$ [1.4 N in HNO_3, conc. = 2 g Pu^{+4}/l] in a capillary tube and then adding ammonium molybdate. Yesterday he added 0.50 λ ammonium molybdate (as opposed to 0.25 λ in previous experiments). The precipitate was then centrifuged, the pH of the supernatant determined to be 2.0, and an aliquot counted; the solubility was determined as approximately 16 mg/l.

Jaffey shipped by express to Eastman a seven-pound uranium egg which has received 320,000 microampere-hours of bombardment with beryllium-deuteron neutrons at the St. Louis cyclotron.

"Chemical Research, Production and Extraction of Plutonium. Report for Month Ending October 9, 1943" (CN-991) was issued. It states that Beaton, Fields, La Chapelle, Lincoln, Sheft, and Turk have conducted experimental studies on adsorption methods. A preliminary investigation of the effects of uranyl ion concentration and flow rate on the adsorption of plutonium by Amberlite IR-1 during the citric acid extraction cycle indicates that 5% UNH and 60 gal/ft^2/hr may represent the optimum conditions for plutonium adsorption. To separate the plutonium from the fission products remaining in the elutriant from the Amberlite IR-1, a series of new ion-exchange adsorbents have been tested. Of these, a titanium zeolite made by briquetting TiO_2 and mica under pressure is the most promising; extensive studies of the heat-treatment of this material are now underway. Bremsstrahlung measurements have been made using pure P^{32} in order to improve the accuracy of measurement of gamma decontamination factors. In connection with the possible tie-in of the adsorption process with wet precipitation processes, oxidation studies have been

made on the Amberlite IR-1 product elutriant, which show that $Ag^{+} + S_2O_8^{-2}$ is effective but $(NH_4)_2S_2O_8$ is not. Cunningham has carried out a chemical isolation of Np^{237} in microgram amounts from neutron-bombarded metal. Separation of neptunium from plutonium was accomplished by means of cold bromate oxidation in which the neptunium, but not the plutonium, is oxidized to a fluoride-soluble state. Two specific activity determinations made on the isolated material, based on the weight of oxide (assumed Np_2O_3) and fluoride (assumed NpF_3), give half-life values of 7.5×10^6 and 6.0×10^6 years, respectively. These values are much higher than the value of 3×10^6 years determined by Wahl and me and indicate possibly that the neptunium has been isolated in a relatively impure state. The hydroxide, oxide, fluoride, and aqueous solutions of the nitrate of neptunium were prepared. Because the purity of the preparation is still doubtful, the conclusions reported are tentative.

Helen had lunch downtown with Wilma and then went to the Crerar Library in the afternoon to work on the "Table of Isotopes." In the evening we played pinochle with the Ghiorsos.

The Soviets have taken forty more villages in their drive on Vitebsk, have entered the outskirts of Gomel, and have penetrated the outskirts of Kiev.

Wednesday, October 13, 1943

Last Thursday while I was away, Beaton called M. J. Shoemaker of Research Products Corporation to discuss the manufacture of titanium zeolite. Beaton urged him to proceed with their efforts to uncover the variables involved in preparing material with reproducible adsorption characteristics and said that we are placing an order for an additional 100 pounds. He then confirmed his phone call by a letter. Today Beaton received a letter from Shoemaker concerning their need for a blanket order with a priority rating for services and materials in order to work effectively on the problem of developing a satisfactory titanium zeolite for use in the adsorption process. Shoemaker advanced the possibility that if the work becomes too burdensome they may have to serve as advisers, with the work being done in Chicago.

Willard wrote to Ralph Firminhac that we are delighted to hear that he has decided to joint the Project. Firminhac will spend a few weeks in Chicago in order to become familiar with the work before reporting to Site X. Willard suggested Firminhac store his household equipment until after his arrival in Oak Ridge.

Today Helen had lunch at Adele Brown's and later had coffee with Wilma.

I attended an evening meeting of the Purification and Metal Production Groups of my section in Room 209, Eckhart Hall, starting at 7:45 p.m. Others present were Bradshaw, Brody, Davidson, Dempster, Dixon, Foster,

Fred, Fried, Ghiorso, Hagemann, G. Johnson, Katz, Katzin, Kirk, Kohman, Manning, Marshall, Nachtrieb, Patton, Potratz, Rosenfels, and Willard. Dempster discussed the application of the mass spectrograph to the analysis of light element impurities. A spark is the excitation source. It appears that this method will detect light impurities at the required levels. In a new mass spectrograph almost ready for operation, the spectra of all light impurities will fall on a single photographic plate using a one-milligram sample. Potratz talked about the development of analytical methods for fluorine, sulfur, and boron. Work on cupferron extraction and other organic reagents is underway, with a view toward making separations prior to spectrographic analysis. Fred discussed a method for determination of oxygen that depends on getting it out of the metal as molecular oxygen by sparking. Brody described further work on purification by ether extraction. Using uranyl nitrate containing added impurities to 500 ppm, he found that the only element which follows through is boron. Jensen described purification of thorium by molybdate precipitation. After two precipitations by ammonium molybdate reagent, boron, lithium, and beryllium are reduced to 2 ppm, sodium and potassium to 100 ppm, magnesium to 40 ppm; but there is no reduction of phosphorous. Plutonium molybdate was precipitated and its solubility measured as 15 mg per liter. Foster discussed some of the recent work at Site B where some luck has been achieved with the preparation of small quantities of metallic uranium by centrifugal reduction. Kirk described the recently developed apparatus for the reduction of plutonium fluoride. The fluoride in effect is reduced by vapors of magnesium and sodium, with the sodium functioning as a getter for whatever oxygen is present. The product so obtained differs from those obtained previously in that it is grayer, harder, and not so shiny. It still reacts with acid, but unlike earlier samples it seems to tarnish on exposure to air for several days. On the basis of counting experiments the substance is substantially all plutonium. It appears to be very porous and unsuitable for density determinations. Manning described the results of experiments on reduction with atomic hydrogen. Uranium oxides and plutonium oxides show no evidence of reduction to metal. Uranium tetrafluoride is reduced with several products being formed. With PuF_4 some change takes place, but quantities are too small to draw any definite conclusions.

The British have been granted war bases in the Azores in an agreement between Great Britain and Portugal.

Thursday, October 14, 1943

Jerome Karle started work in Section C-I. He is a highly recommended physical chemist from the University of Michigan, who received a Ph.D. this year and has been a teaching fellow.

Compton informed me that Conant has said Darrell Osborne cannot be spared because "he is tied up in an important problem in NDRC which has immediate applications to the battlefront and on which we are being heavily pushed." Compton quoted Conant as noting that chemists are now becoming almost as scarce as physicists; furthermore Conant is requesting

that a careful review be made to see if there are any chemists available within the S-1 organization. Compton is asking Stearns to help us look further into the matter.

Willard received a reply from Stearns saying that, effective January 1, 1944, he is recommending that Floyd Dean be promoted to Associate Chemist. However, with regard to John Crawford, Stearns does not wish to consider his case until December because of his merit increase last July 1.

In a letter to Waldo Cohn at Clinton Laboratories I asked for the data I understand he has on the impurities in lanthanum nitrate obtained from the Lindsay Light and Chemical Company. These impurities could affect its use in the concentration process.

Jaffey wrote to Eastman about the October 12 shipment of the bombarded uranium metal egg and asking for the return of the lead shield in which the egg was sent.

Franck suggested to Stone that, as a means of saving time, someone on his staff should come to the New Chemistry Building periodically to take blood counts of the men, rather than having the men visit the medical unit.

The regular evening meeting of the Section C-I Research Assistants took place at 6:45 in Room 209, Eckhart Hall.

Helen had lunch at Wilma's. Later in the afternoon Wilma stopped over for coffee.

I held an evening meeting of the Council of my section in my office attended by Beaton, Cunningham, Davidson, Dreher, Ghiorso, Hagemann, Kirk, Manning, Thompson, and Willard. I summarized the schedule for Project Council meetings on the Bismuth Phosphate Process and on purification that will be held next Monday, Tuesday, and Wednesday. During the course of the meeting we discussed the following matters: Perlman wants a binocular microscope and a #2 centrifuge sent to Site X. Cunningham stated that Dixon is now working over 100-150 micrograms of plutonium for Kirk before working on the product from the last ether extraction of St. Louis material. Kohman has been checking on losses in the alpha counter circuit and finds 2½% loss per 1,000 counts/minute. I mentioned that an arrangement may be made so that men on the project who are interested in working for a Ph.D. at Chicago can do work toward certain of these requirements. The question of appointing Gershbein (experienced in high pressure reactions) was discussed. I raised the question of what work should be done first with 93^{237}. Cunningham recommended solubility measurements of the phosphate, oxalate, and, possibly, hypophosphate. Other suggestions were alpha particle range measurements by Ghiorso, cupferron extraction studies by Dixon, tests to determine if the hypophosphate is formed indicating a +4 ion for neptunium, and adsorption studies and determination as to how much neptunium is carried in the Bismuth Phosphate Process. I described the plutonium peroxide precipitation flowsheet now being designed for bulk reduction and concentration at Site W--the process I

have recommended, somewhat prematurely but of necessity, to Squires so that design of facilities can get started. We discussed possible alternates to the present plan to dissolve the first plutonium peroxide precipitate in 10 N HNO_3, dilute to 1/2 N HNO_3, and reprecipitate as plutonium peroxide; this procedure leads to an undesirable volume increase. Thompson mentioned it might be necessary to dilute only to 2 N HNO_3. Kirk suggested converting the peroxide to the solid nitrate with concentrated HNO_3. I said that in view of the discussion it would be very desirable to have Howland measure the solubility of the peroxide in 2 N HNO_3. Davidson announced that the fluorine production cell has come from MIT and is found to yield a high purity product. He is considering the desirability of studying the revolatilization of the higher fluoride of plutonium at 500°C in the absence of fluorine. He also plans to test the behavior of the fluorides in calcium fluoride tubing and is considering the possible use of BrF as a more active agent than F_2. I suggested also using calcium fluoride tubing for the revolatilization experiment. Davidson stated Katz is working on the bromination apparatus and that the use of bromine looks promising for a purification process. I pointed out that so far as the dry methods are concerned there will be great pressure for purification studies; still there should be some work toward developing alternate methods for extraction and decontamination. Cunningham suggested, and it was agreed, that Kirk should check the fluorination of his forthcoming large sample of plutonium by weighing; this would also serve as a check on the valence of plutonium. I announced that when the 120 microgram plutonium sample isolated the day before yesterday by Perlman's section from the Argonne pile metal is sent up from Site X, a sample of about 30 micrograms of the oxide (PuO_2) will be given to Compton for preservation for posterity; this is the first significant sample isolated from pile material. Mounting in lucite was suggested. I raised the question of future manpower. Kirk will get 4 or 5 men within the next month; the need for them is obvious. Ghiorso's group is full, but more men are needed for Site X. Thompson's group is nearly large enough. Watt will come in about two weeks and will probably start a subgroup on concentration under Thompson. A plutonium recovery group, composed of Mrs. Baumbach, Belletire, and Meyer, is being formed under Cunningham. Similarly, extraction work is being continued as a subgroup in Davidson's group. Cunningham urged that the valence of plutonium in the upper state be definitely determined--40 or 50 micrograms of plutonium will be needed.

Allied planes blasted Rabaul, New Britain, on Tuesday, today's newspaper reports.

Friday, October 15, 1943

Alvin J. Dirksen reported for work in Section C-I. He has been a research chemist at the Oldbury Electrochemical Company in Niagara Falls, New York. Dirksen received his Ph.D. from the University of Wisconsin in 1941.

Craig wrote from Berkeley giving the specifications for some of the components of the quartz ultramicrobalance we are building here.

He suggested I might get some personal help with the quartz fibers from Rebecca Bradford, a former employee of Linus Pauling, whom Kennedy has hired for quartz work and who is now in Berkeley.

I have been very impressed by the performance of Winston Manning. In view of my need for assistance in the administration of Section C-I, I sent a memo today to Franck asking that Manning be appointed as Assistant Section Chief of my section to replace Perlman who has gone to Site X. (See Figure 10.)

The "Monthly Report of the Activities of the Chemical Division," (MUC-JF-103), was issued by Franck's office. It summarizes the work of my section as follows:

I. Extraction, Decontamination, and Concentration. (a) Bismuth Phosphate Process. This work is described in Report CN-979. X-ray studies of bismuth phosphate and bismuth-plutonium phosphate are being made in cooperation with Zachariasen's group. (4-1/2 manpower; A-1 priority) (b) Alternate Processes for Extraction and Decontamination. 1) Adsorption method. Citric acid has been used in the Amberlite IR-1 extraction cycle to complex the gamma active fission species, zirconium, columbium, and ruthenium; an extraction step gamma decontamination factor of 50 was reached. The most favorable conditions for that purpose are the use of 5% $UO_2(NO_3)_2 \cdot 6H_2O$ and a flow rate of 60 gal/ft^2/hr. An adsorption column containing titanium zeolite looks promising to use for decontamination cycles. A high selectivity of plutonium adsorption has been obtained by calcining the zeolite. (4-1/2 manpower; B-1 priority) 2) Bismuth arsenate is very similar to bismuth phosphate as carrier for plutonium. (1/2 manpower; B-2 priority) 3) The use of ceric ion to dissolve lanthanum or cerous fluoride and to oxidize plutonium following the dissolving step has been tested for its effect on decontamination concentration procedures. Plutonium may be carried by cerous fluoride as well as it is carried by lanthanum fluoride. 4) Uranous oxalate shows promise as a carrier to extract plutonium from solutions of pile metal in preparation for the use of the sodium uranyl acetate method for decontamination. (1/2 manpower; B-2 priority) 5) The use of organic solvents for the extraction of organic salts of plutonium form solutions of pile metal is being investigated. (1 manpower; B-2 priority) 6) Dry Methods for Extraction and Decontamination. (1 manpower; B-1 priority) (c) Concentration. (3 manpower; A-1 priority).

II. Isolation of Neptunium, U^{233} production, and plutonium recovery. (a) Neptunium as 93^{237} has been isolated in microgram amounts from neutron bombarded uranium metal. The specific activities of weighed samples of Np_2O_3 and NpF_3 give calculated lifetimes of 7.5×10^4 and 6.0×10^6 years. The latter value is believed to be more correct. These values are much higher than the Wahl and Seaborg value of 3×10^6 years and possibly indicate that the neptunium had been isolated in a relatively impure state. The hydroxide, oxide, fluoride, and aqueous solutions of the nitrate of neptunium were prepared and are described. Preliminary solubility figures for the hydroxide and fluoride were obtained. Because the purity of the preparation is still doubtful, it is emphasized that the

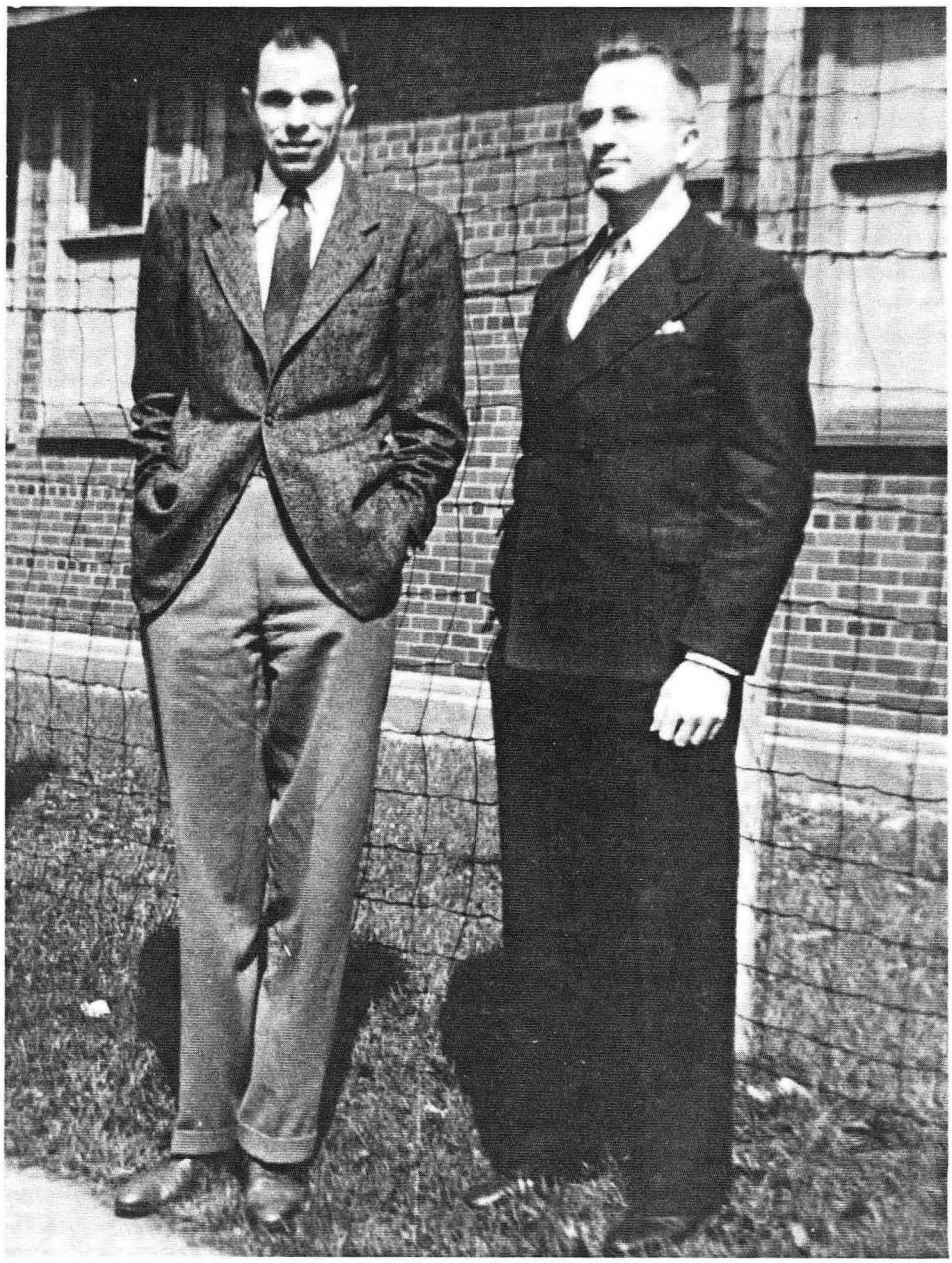

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Figure 10. Glenn T. Seaborg and Winston M. Manning in front of New Chem--Autumn 1943

conclusions in this report are tentative. (1 manpower; A-2 priority) (b) Two hundred aluminum cans have been filled with compressed thorium carbonate, taken to New Kensington for sealing, and shipped to Site X to be placed in the pile for the purpose of U^{233} production. (2 manpower; B-1 priority); (c) A double ether extraction has been made of the September 26th St. Louis bombardment of uranyl nitrate (Chicago VII), and the product solutions are being processed for the recovery of the pure plutonium. (3 manpower; A-1 priority).

III. Metal Production and Purification. Attempts to prepare solid single pieces of ~20 micrograms weight have continued. Plutonium compounds to be reduced are put in a crucible which in turn is placed in a larger crucible containing the reductants. By heating the system, plutonium is brought in an atmosphere of reducing vapors without amounts of oxide and other impurities being introduced by the reductants. Experiments to produce the metal by electrolysis are in progress. Atomic hydrogen as reductant has great advantages since it leaves the product uncontaminated; uranous fluoride is reduced. There is reason to hope that a reduction of plutonous fluoride may be accomplished in the same way. Other Metal Production Work. Thorium metal reacts with molecular hydrogen to form ThH_3 similar to the behavior of uranium. Studies on the volatility of plutonium fluorides continue. Studies of the chemical behavior of plutonium molybdate seem to indicate that molybdate precipitations may be useful in purifying plutonium from boron, bismuth, iron, potassium, lithium, magnesium, sodium. Some new results are being gained on the composition of plutonium peroxide, on the hydrolysis of plutonous plutonium, and the solubility of plutonous sulfate. With uranyl nitrate as stand-in, information is being gathered that will be useful for purification of plutonium by ether extraction. The method for determination of the vapor pressure of plutonium bromide is being improved, and several other apparatuses for volatility measurements are under construction. Preparations are being made for the study of hydrogen uptake by plutonium. Analytical problems in purifications program are being worked on in cooperation with Section C-IV. (18 manpower; A-1 priority).

IV. Instrument Group. (6 manpower; A-1 priority)

Helen went shopping with Wilma today.

U.S. planes flew deeply into Germany to smash the ball-bearing plant at Schweinfurt but at the high cost of sixty bombers.

Saturday, October 16, 1943

Sherman Fried started work in Section C-I today. Fried received his Ph.D. from the University of Chicago last year. After first informing him that we have arranged for his draft deferment, I introduced him to Jonathan Dixon and asked Jon to demonstrate the carrying of plutonium tracer by lanthanum fluoride. Later, as with other recent newcomers, I asked him to sit down and read some chemistry reports. He, like others before him, was mystified by the word "product" and "pile"--some of the reading material didn't make much sense. Fortunately his friend, Joe Katz, came by and sat down and talked to him like a father telling his son the "facts of life."

Manning received a memo from Elmer W. Brugmann, an engineer who is C. M. Cooper's assistant, confirming that Manning is to make a literature survey for the Technical Division on the control of slimes and algae in water systems, an area in which he is an expert. The information is needed for the control of such growths at Site W.

In a memo to Allison, Franck endorsed my request that Manning take Perlman's place as Assistant Section Chief of Section C-I. He recommended that the appointment be made as soon as possible.

"Chemical Research - Special Chemistry of Plutonium. Report for Month Ending October 11, 1943" (CK-987) is being issued. Topics covered are: Baumbach, Rosenfels, and Kirk report that little plutonium has been available for use but that definite progress had been made in improving the quality of the metal prepared. They have tested three new reduction techniques: (1) use of a system by which the sample and reductant are introduced in a preheated furnace; (2) use of a double crucible system; and (3) use of mixed metals as reductants. Technique No. 1 gives little improvement in quality of the product. Technique No. 2 yields metal which is notably free of contamination and is very little oxidized. Technique No. 3 is also helpful--a mixture of two metals, such as barium and sodium which vaporize at different temperatures, provides a "getter" atmosphere which reduces the chance of oxidation of the product. It is concluded that a combination of technique Nos. 2 and 3 is most likely to produce a useful product. Zvolner and Kirk have constructed a micro electrolysis cell capable of electrolyses of a few micrograms of metal from aqueous media and have successfully tested it by preparing copper, iron, zinc, and manganese metals in quantities less than 5 micrograms. Tests of plutonium will be made shortly. Manning has made preliminary tests of the use of atomic hydrogen as a reductant and finds that while oxides of uranium and plutonium are not reduced, uranous fluoride gives reason to hope that reduction of plutonous fluoride may be obtained by this method; it has the outstanding advantage of leaving the product uncontaminated by slag or oxide. Katz and Davidson, using a specimen of thorium metal prepared by powder metallurgy, find that it takes up hydrogen at room temperature to form a substance of composition $ThH_{3.0}$. At 1000°C, practically all the hydrogen is expelled at atmospheric pressure. The treatment with hydrogen produces some disintegration of the metal. (The behavior of thorium metal with hydrogen is of interest as a stand-in for plutonium and also as a possible means of developing a dry process for metallic thorium used in a pile.) Davidson, Katz, and Orlemann have studied the behavior of the higher fluoride of plutonium. They have constructed an apparatus in which the higher fluoride can be volatilized directly onto a cold platinum surface in an atmosphere of fluorine and revolatilized from the platinum at different temperatures. Results with the apparatus indicate that the higher fluoride is not very volatile at 200°C.

Helen and I went shopping in the Loop in the late afternoon.

More than 25,000 coal miners have gone on strike. This is the main headline in today's paper; there is fear this will lead to a general coal strike.

Sunday, October 17, 1943

Helen and I played golf with the Ghiorsos at the Evergreen Golf Club. The Ghiorsos and Baumbachs then had supper at our apartment. Melvin Calvin who is in town from Berkeley spent the night with us.

Monday, October 18, 1943

I have decided to ask our new man Fried to join our metal production group because he seems especially well suited for this, and we need to move this program along as fast as possible.

Iz Perlman and Spof English are visiting the Met Lab from Site X.

At 9:45 a.m. I attended a meeting in C. M. Cooper's office in Eckhart Hall to plan our presentation for the semi-monthly Bismuth Phosphate Process Meeting to be held here Wednesday afternoon.

In a letter to Hamilton in Berkeley, D. J. Hughes indicates that I would like to have use of the Berkeley cyclotron for at most three days per month and that this can easily be worked into Burton's program.

At 2:00 p.m. in Room 209, Eckhart Hall, I attended the first of a series of monthly meetings on plutonium purification and metal production called by C. A. Thomas; the meetings will be held on Monday afternoon of the week when the Laboratory Council Information Meetings of the Chemistry (and Technology) Division and the Chicago meeting on the Bismuth Phosphate Process are held. Others present were Allison, Boyd, Cunningham, Davidson, Dempster, Eastman, Foster, Franck, Hilberry, Hogness, Johns, W. C. Johnson, J. W. Kennedy, Lum, Magel, Manning, Orlemann, Potratz, Cyril Smith, Spedding, Sugarman, Thomas, W. W. Watson, and Willard. Dempster discussed the utility of the mass spectrograph for the analysis of light impurities. The method has sufficient sensitivity and appears especially useful for O_2 and F_2, but quantitative methods need developing. Fred discussed optical spectroscopic methods of analysis. The sensitivity is adequate for all except boron, sodium, lithium, and phosphorus. Air-borne contamination and impurities in the copper electrodes are problems. The present status of the development of chemical methods of analysis for carbon, oxygen, fluorine, sulfur, and chlorine were discussed by Orlemann, Potratz, and Kennedy. The question of standard samples was brought up; and a committee consisting of Kennedy, Orlemann, Potratz, Dempster, and Fred was designated to make recommendations for future action. Purification work was discussed by Davidson and Orlemann. It was mentioned that studies of purification through the use of the volatile plutonium compounds, chloride and bromide, appear to be the most important; the utility of the higher fluoride is still doubtful, as is that of the plutonium acetylacetone. In precipitation and extraction methods, the most promising involve plutonium peroxide or molybdate. Thomas pointed out the need to test a great number of solvents, as the matter is entirely empirical and a simple method of evaluating solvents is most desirable. Orlemann and

I stated that the method of determining whether a plutonium tracer is extractible furnishes just such a method. There was also a discussion of the program for purification of reagents, and I pointed out that the container problem will be met if we are successful in obtaining the $20,000 worth of platinum we have ordered.

The Berkeley bombardment of 1,100 pounds of metal and 300-400 pounds of nitrate with neutrons from 15 Mev deuterons on beryllium was completed today.

The regular evening seminar of the Chemistry Division was held at 7:45 p.m. in Room 251, Ryerson Laboratory. Perlman talked on activities at Site X, and Curtis of Boyd's group spoke on rapid methods of analysis.

Helen has obtained the necessary clearance to work at the Met Lab, where she needs the typing facilities and the use of the library to work on my "Table of Isotopes." She tried to work under an arrangement of no compensation, but the bureaucracy found this to be impossible; so she has consented to work as an hourly employee. Today she worked in the Information Division typing my "Table of Isotopes."

The top war news today comes from the Pacific, where in six air battles 104 Japanese planes were destroyed with only two Allied craft lost. In a small item on today's front page is word that the Nobel Prize winning French author, Romain Rolland, has been interned in a concentration camp.

Tuesday, October 19, 1943

I wired Kamen in Berkeley for the remaining plans for the ultramicrobalance.

At 9:30 a.m. I attended a Project Council Information Meeting on Chemistry in Room 209, Eckhart Hall. Others present were Allison Burton, Boyd, Cannon, Compton, C. M. Cooper, Coryell, Eastman, Elliott, Fermi, Foster, Franck, Garrison, O. H. Greager (Manhattan District), Greenewalt, Hilberry, Hiskey, Hogness, Hughes, Jeffries, Johns, W. C. Johnson, Kennedy, Lum, Manning, Miles, Mulliken, Perlman, Potratz, Cyril Smith, Spedding, Stearns, Sugarman, Sutton, Thomas, W. W. Watson, Whitaker, Wigner, Wilhelm, and Willard. Perlman reviewed the work of his section at Site X on the Bismuth Phosphate Process started at Chicago. He mentioned that they have obtained 120 micrograms of concentrated plutonium from Argonne pile uranium using the Bismuth Phosphate flowsheet. In the matter of physical measurements, the magnetic-deflection alpha counter has proved satisfactory. The U^{233} group is working on handling 10-20 pound lots of thorium, in preparation for processing the $Th(CO_3)_2$ that is to be irradiated in the Site X pile. Kennedy talked about the chemical work at Site Y on analysis, mostly oxygen and fluorine. He also discussed the over-all neutron intensity test, which then led to

a discussion of counters. In answer to a question by Fermi, I indicated that Kohman is working on BF_3 neutron detection chambers. Eastman reported that Gofman and Connick have completed the extraction of one milligram of plutonium for Site Y from 500 pounds of uranium nitrate and 500 pounds of uranium metal bombarded with neutrons from the Berkeley cyclotron. The material was put through three acetate cycles, with an overall yield believed to be more than 85%. The overall soft gamma decontamination factor was about 5,000. About half of the plutonium is being shipped to Site Y. Eastman also reported on their work to develop a process for the separation of Po^{210} from neutron-bombarded bismuth. I reviewed our work on the Bismuth Phosphate Process (CN-991) and the preparation of plutonium metal. I mentioned our plan to have 30 micrograms of the 120 micrograms of Pu^{239} prepared at Site X from Argonne pile material set aside for historical purposes.* We also discussed plans to get a 100-microgram metal sample to Zachariasen for the determination of its structure and density by the x-ray diffraction method. In a discussion of material on hand I estimated there is perhaps a total of 500 micrograms distributed in my section. The largest single lot is the 1-milligram sample just produced at Berkeley. Total plutonium production so far has been 3-4 milligrams.

At 2:00 p.m. in Room 209, Eckhart Hall, I attended the afternoon session of the Project Council Information Meeting concerning the Technology Division's work. Present were Allison, Bellas, Brugmann, C. M. Cooper, Eastman, Fermi, Foster, Franck, Greager, Greenewalt, A. B. Greninger, Hilberry, Hogness, Howe, Huffman, Hughes, Jeffries, Johns, W. C. Johnson, Kircher, Leverett, Magel, Miles, G. S. Monk, Mulliken, Perlman, J. H. Peterson, Cyril Smith, Spedding, Sutton, Thomas, Vernon, W. W. Watson, Whitaker, Wigner, and Wilhelm. The Process Development Section's work during the reporting period was stated to have been directed toward improvement in decontamination in the Bismuth Phosphate Process with some promising leads being developed. A second important line of study has been the further improvement in plutonium yield throughout the process. Attention has also been directed toward the development of concentration procedures for both Sites X and W. The work of the Clinton Process Development Division was summarized. They are now using Site X levels of fission products in tests in the semi-works. In the extraction step the average yield of plutonium is 94%; decontamination steps give 85-86% yield for one cycle. There is little evidence on the bulk reduction step as yet because many batches must be combined to get enough material for a run. Thus far, no mysterious

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*This 30 microgram sample of PuO_2 in a small glass capillary embedded in a lucite disc and housed in a six-inch stainless steel cube is now at the Argonne National Laboratory in Chicago. A picture of this same sample is on display in the Museum of Science and Industry in Chicago in a special exhibit devoted to early nuclear research in the Chicago area. (See Figure 11.)

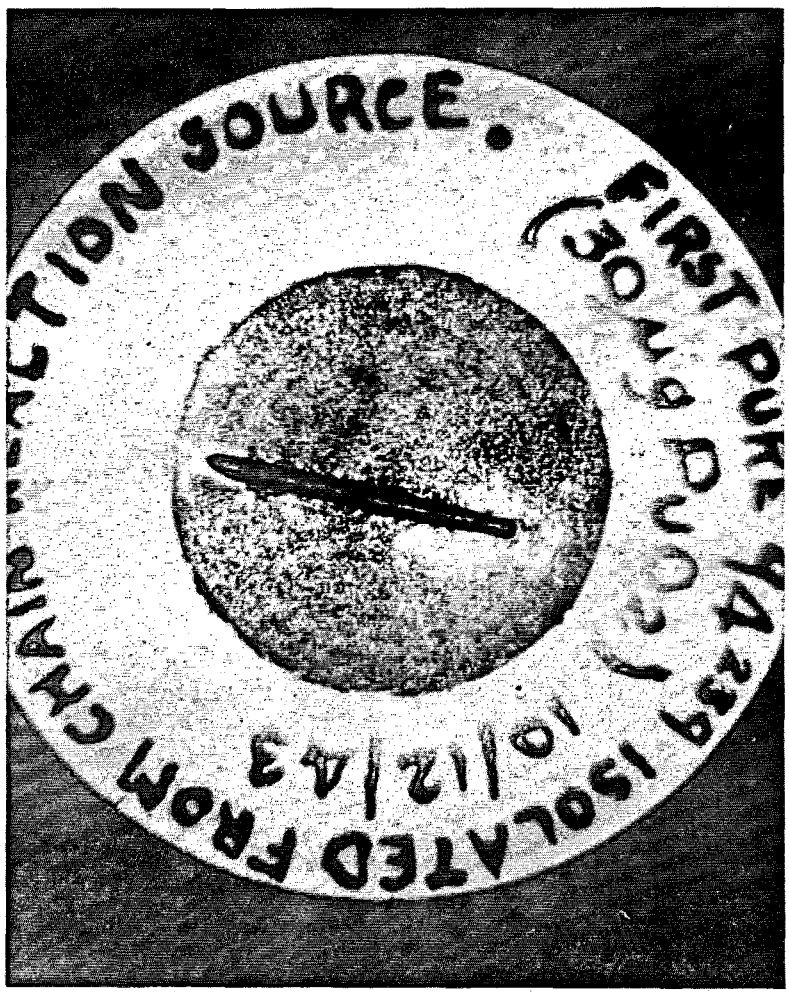

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Figure 11. First pure Pu^{239} isolated from chain reaction source (30 micrograms of PuO_2). October 12, 1943.

reversals of behavior have turned up in going from a laboratory process to a semiworks scale. In a discussion of the availability of heavily irradiated material, Fermi noted that the Argonne pile now has material 2½ times as heavily irradiated as previous material and in a few weeks the factor will be 4. I mentioned that we have a Berkeley bombardment now underway, at perhaps 75,000 micro-ampere hours, which is scheduled to go to Clinton--this material is about five times as strong as the previous Argonne pile shipment.

As a significant step in our plutonium metal production program we have decided to have Kirk and Baumbach try to reduce a "large" sample of PuF_4 to plutonium metal. We decided to gamble 100 micrograms of pure plutonium oxide in the effort in order to produce a reasonable amount of metal. To minimize the hazard of losing the oxide by having it blown out of the boat during hydrofluorination, a complex triply-stacked house-in-house baffle system has been constructed. Florin was given the task of the actual fluorination today. After the fluorination process was completed, Kirk opened the labyrinth and much to his dismay could find no evidence of the plutonium or its oxide. An immediate search for it was launched, but no trace could be found. Florin scraped out and washed the fluorine reactor. He then scraped down the transite walls of the hood into which the reactor was exhausted, took samples, dissolved them and checked them for alpha particle counts but to no avail. I was called in to assess the situation, and my first reaction was to tear down the hood and ship it to Oak Ridge where it could be dissolved in the dissolvers. Kirk and I then agreed that our precious sample had either gone up the flue, or more likely, the plutonium never actually was put into the baffle system due to some confusion or mix-up.

Bert Goldschmidt is in town on a visit from Montreal; he visited us in our apartment this evening.

Cordell Hull, U.S. Secretary of State, and Anthony Eden, Britain's Foreign Secretary, have arrived in Moscow for a conference with Foreign Commissar Molotov. Accompanying Hull is W. Averell Harriman, newly appointed Ambassador from the U.S. to the U.S.S.R.

Wednesday, October 20, 1943

Waldo Cohn of Clinton Labs responded to my October 14 letter regarding impurities in lanthanum nitrate. He says that the material contains about 0.5% cerium and that probably there are many more elements present.

I attended the Bismuth Phosphate Process Progress Meeting at 2:00 p.m. Others present included Perlman and English from Clinton Labs. My section presented information about the search for new reducing agents. We mentioned that the microchemical experiments to test plutonium recovery through a bismuth phosphate extraction step and one decontamination cycle using Site W concentrations of plutonium and fission products have been started. In cooperation with Sutton's and Sugarman's sections, work has been started on the use of by-product scavenger precipitates. Concentration work is

continuing, particularly on improvement of the method which uses coprecipitation of plutonium with rare earth fluorides to reduce the volume after the application of the bismuth phosphate decontamination procedure; this we refer to as the "crossover" cycle. We also reported on the method of concentration in which plutonium is coprecipitated with $U(C_2O_4)_2$, then dissolved in HNO_3-K_2CrO_7 solution followed by coprecipitation of the oxidized plutonium with sodium uranyl acetate.

The evening meeting of the Extraction Group of my section was held in Room 209, Eckhart Hall, at 7:45 p.m. Others present in addition to me were Perlman, Beaton, Brody, Dirksen, Dixon, Dreher, English, Firminhac, Frank, Gaarder, Ghiorso, Hagemann, Halperin, Jaffey, James, Johnson, Katzin, Kohman, Kroner, Lincoln, Malm, Manning, Margolis, Meyer, Orlemann, Schuler, Thompson, and Willard. The presence of Perlman gave us the opportunity to coordinate plans for work at Site X and here at the Met Lab. I summarized the extraction, decontamination, bulk reduction (crossover), and concentration (lanthanum fluoride-reduction cycle) steps in the Bismuth Phosphate Process as presently carried out in the semiworks. Thompson discussed the basis for some of the conditions used and the work now underway. Perlman commented on the experiences with the Process at Site X and indicated that the metathesis of the lanthanum fluoride in the concentration step is a serious problem.

The Allies have destroyed 60 more Japanese planes and have sunk three ships in a raid on Rabaul, Japan's New Britain stronghold in the South Pacific.

The Project Council Policy Meeting about the Chemistry Division Program, at 9:30 a.m. in Room 209, Eckhart Hall, was attended by Allison, Compton, C. M. Cooper, Eastman, Fermi, Franck, Greager, Greenewalt, Hilberry, Hogness, Hughes, Jeffries, W. C. Johnson, Mulliken, A. V. Peterson, Spedding, Thomas, W. W. Watson, Whitaker, Wigner, and Stearns. Franck included a summary of the distribution of manpower on, and the priorities of, the tasks of my section. Thomas stressed the importance of a density determination of plutonium and urged that it take precedence over almost any other phase of the work. After a discussion Compton summarized the situation as follows. Present techniques have not been developed to the point where one can be at all confident of results, even if we have one milligram quantities. Hence, at present, we shall go on with increased emphasis in the development of techniques. At the same time we must look to Clinton for milligram quantities as soon as possible, with the requirements for density determinations to be satisfied with the first milligrams. Jeffries suggested that an alloy of aluminum and plutonium with perhaps 30-50 times by weight of aluminum should permit determination of density with a precision of at least 5%. Eastman presented the California program and indicated that oxidation states and potentials and heats of reactions are highly important data which will need milligram amounts of plutonium. Franck asked if men will go from California to Clinton to work on the acetate process, to which Eastman replied that, if the acetate process should become the mainline, they would

supply, at least, men for consultation. If it is an alternate process of low priority, he questions the use of men in that connection.

Thursday, October 21, 1943

Thorfin Hogness, Professor of Chemistry, University of Chicago, has been appointed Associate Director of the Chemistry Division. His new duties begin today.

There was a meeting of the Executive Committee of the Metallurgical Project attended by Compton, Allison, Hilberry, and Whitaker, and as consultants, Greenewalt, Fermi, A. V. Peterson, and Jeffries. At the meeting it was agreed to recommend the transfer of the Process Development Section under Sutton to the Clinton Laboratories, now called X-10.

I received a telegram from Kamen in Berkeley saying that he is sending several more prints of the ultramicrobalance this week and that they hope to finish the job before a month's time.

At 6:45 p.m. there was an evening meeting of Section C-I Research Assistants in Room 209, Eckhart Hall.

The 7:45 p.m. meeting of the Council of my section was attended by Beaton, Cunningham, Davidson, Dreher, Ghiorso, Kirk, Manning, Orlemann, Thompson, Willard, and me. The reconditioning of Room 5 is proceeding slowly--it will be used as necessary by groups working under Kirk, Cunningham, and Orlemann. The uranium metal and UNH from the Berkeley bombardment is due to arrive October 31; 100 pounds of the hottest metal will be retained for extraction of 93^{237}, and the remaining 1000 pounds will be sent to Site X. Thompson is making plans for handling the 100-pound batch. I mentioned that I plan to ask Fermi to let us have ten pounds of metal from the Argonne pile which has operated at a high level, in order to test for 93^{237}. G. Johnson is to prepare a plutonium-containing bismuth phosphate precipitate for Zachariasen to use in a crystal structure determination, using 15 micrograms of plutonium.

For the time being the recovery group under Cunningham will work in the New Chemistry Building Annex together with the U^{233} and concentration groups. Thompson mentioned that Sutton's group is definitely moving to Site X on November 15, and I said I would look into the possibility of getting some of their space in the West Stands; it would be ideal for Beaton's work. Thompson's group is ready to start setting up a small semiworks plant in the present room near the cyclotron. It is planned to ship the new 15-ton press to Perlman at Site X. The possibility of preparing a probe type of alpha counter to assist in plutonium recovery operations was strongly supported, and I asked Ghiorso to give it a fairly high priority. I also suggested the use of a device for reducing counter backgrounds; essentially this would consist of a glass sleeve extending down over the upper part of the central wire. Cunningham mentioned that about two weeks ago Cliff Smith began keeping a detailed record of the disposition of plutonium. Meyer is now started on the plutonium recovery work.

Plans for future research were discussed. In considering possible distillation processes for purification, I suggested that some thought be given to the possible distillation of the plutonium metal itself, if it melts at a low enough temperature. Bromide distillation and the accompanying need for platinum or porcelainized apparatus was discussed. The large order for platinum is now waiting for Kimpton's approval. Kirk announced that some progress is being made on plutonium metal production. Uranium and thorium will be prepared for Zachariasen's use in testing the x-ray diffraction method for density determination. Kirk also said that a promising preparation of plutonium metal has just been made; it may serve to establish purity and degree of compactness which can be expected by this method. Thompson announced that Johnson has found 98% carrying of plutonium by cerous fluoride at a 9:1 ratio of cerium to plutonium. Cunningham has predicted that cerous fluoride would not carry as well as lanthanum fluoride.

Helen worked on the "Table of Isotopes" at the Crerar Library.

Moscow dispatches say that a large scale German evacuation of the Crimea is underway and the Russians are trying to cut the Germans off.

Friday, October 22, 1943

I attended a 10:00 a.m. meeting in Room 251, Ryerson Laboratory about P-9, the heavy water pile project. Allison explained that the project is insurance against the failure of the graphite piles planned for Site W, and that by July 1, 1944, we must have the data needed for immediate and rapid construction of a 250 to 600 megawatt pile if needed. Zinn will be in charge of the Argonne program of exponential experiments preparatory to this project. A 250 kw heterogeneous system is a strong possibility. The P-9 Section of the Technical Division now meets every morning from 8:30-9:00 in Room 407, Eckhart Hall.

Willard informed Stephanie McAdams in Stearns's Office that we would like to have our new man George Creek transfer to Perlman's section at Site X on November 15 or as soon thereafter as it is possible to provide housing.

Our decision to transfer Creek to Site X led to an interesting incident. As I usually do after we have decided to transfer someone to Site X, I went through the laboratories to find Creek to inform him of our decision and to seek his concurrence. I returned to my office with the man whom I thought to be Creek, preparatory to discussing the matter with him. Fortunately, as we were entering my office, John Willard recognized that the man I had in tow was John Malm and not George Creek. Willard signalled my impending mistake by clearly exclaiming "Hello, John Malm." I caught the message, spent about 30 minutes with Malm inquiring about his work, to his great surprise because he was not used to this much

personal attention from me. I then went out and found Creek and finally arranged to send the right man off to Site X.

Helen again visited the Crerar Library to look up references for the "Table of Isotopes."

The Allied foreign ministers, meeting in Moscow, are nearing the end of the military portion of their discussions.

Saturday, October 23, 1943

The Technical Division Process Development Section reported in Memo. No. 135 that, in carrying out runs during the week according to the Clinton flowsheet, two alkali metatheses and six water washes are not sufficient to solubilize the lanthanum fluoride produce precipitate. They also carried out a bismuth phosphate-lanthanum fluoride crossover cycle in the concentration step at 0.2 N HF instead of 1.0 N HF and attempts are being made to follow the crossover with a lanthanum fluoride concentration cycle. Some difficulties are being encountered in the metathesis step and in handling the small volumes in the second cycle.

C. M. Cooper instructed H. W. Bellas of his Division, now Chief of an Engineering Development Section, to carry through on the installation in one of the handball courts in the West Stands of facilities for dissolving active uranium slugs and for carrying out large-scale chemical extraction experiments with active solutions; this will be essentially a new semiworks setup. This is to be accomplished before the Process Development Section transfers to Clinton Laboratories. Since this equipment is expected to be of principal interest to the Chemistry Division, Cooper asked that we be consulted before proceeding further. Charles E. Kircher, the Assistant Division Chief, will be in charge.

The Du Pont TNX Division in Wilmington reached agreement on the design of Waste Storage Tanks Building 241 for Site W that conforms to suggestions brought up by the review of their blueprints here at the Met Lab.

Kohman transmitted to Greager a tabulation prepared by Turkevich and him giving the latest estimates on the weights of fission products expected in Site W pile material. The calculations were made assuming a capture-to-fission ratio of 0.88 expected in graphite-uranium metal piles. The particular values relate to a pile operation of 100 days, followed by 60 days of cooling. In their table, reproduced here, the weights of the fission products are given in grams for every 100 grams of Np^{239} plus Pu^{239}:

Se	0.1	Sb	0.02
Br	0.06	Te	1.3
Kr	1.5	I	0.7
Rb	1.8	Xe	11.9
Sr	4.3	Cs	11.6

Y	3.0	Ba	4.4
Zr	10.6	La	4.4
Cb	4.9	Ce	11.7
Mo	8.7	Pr	5.2
43	2.9	Nd	11.3
Ru	5.6	61	--
Rh	1.0	Sm	3.3
Pd	0.6	Eu	0.5
Ag	0.1	Gd	0.02

I sent a memo to Mulliken saying that it would be extremely helpful to the physical measurements group in my section if he would ask Site Y for information on the circuits used to amplify alpha particle pulses in their nitrogen-filled alpha counting chambers. I mentioned that the drawing of the N_2 chamber that Kennedy left with us shows the name of C. S. Garner as designer.

The Assistant Personnel Director, W. Wayne Johnson, received a letter from Fred Albaugh in Inglewood, California, in which was enclosed the completed application form for employment. "I trust you will forgive my delay in forwarding it," he wrote, "inasmuch as for the past two weeks I was lost in the Sierra Nevada mountains and hence out of touch with all correspondence. If you indulge in deer hunting, duck shooting, or trout fishing I am sure you will understand."

The Baumbachs and Wilma joined Helen and me for lunch at our apartment, a delightful interlude from the day's schedule at the laboratory.

Russian forces are tightening their vise on German troops left in the Dnieper River elbow between the Dnieper and the Black Sea.

Sunday, October 24, 1943

Helen and I played golf in Jackson Park. Later in the evening we played cards with the Ghiorsos.

Monday, October 25, 1943

I sent a memo to Allison about assistance to the Monsanto Chemical Company for training of an instrument man. I suggested that the best arrangement would be to have the Monsanto trainee come here, but if there are objections to this on security or other grounds, I would be willing to have Scott of Ghiorso's group spend up to six weeks in Dayton to get them started at their polonium laboratory.

As a follow-up to C. M. Cooper's October 23 memo I detailed for Bellas our need for operation of semiworks equipment at Chicago. One job I gave, as an example, is our plan to start work in the next few weeks on

the extraction and concentration of Np^{237} from a hundred pounds (or more) of neutron-bombarded uranium metal (due to arrive from Berkeley on October 31). Other work that we have in mind that would require semi-works equipment include (1) testing of concentration methods where it is necessary to start with large volumes of solution in order to study the method properly; (2) decontamination studies where large initial volumes are needed in order to obtain sufficient radioactivity; (3) testing of alternate extraction and decontamination methods; (4) development of new or revised extraction and decontamination methods in connection with the heavy water piles where product concentrations will be higher than those anticipated for Site W.

Pursuing the matter of setting up a new semiworks, Cooper requested authorization from Allison to set up in handball court No. 9 in the West Stands, metal dissolving and handling facilities capable of handling Clinton slugs with activities as much as ten times that of the most active metal from St. Louis bombardments. (See Figures 12 and 13.) He also proposed organizing a group of about five men in the Engineering Development Section to maintain and operate these facilities.

Maloney of the Technical Division asked Allison if conditions in the precipitation steps of the wet separation processes are such that a chain reaction could occur. He outline the precipitation conditions for four of the processes--bismuth phosphate, lanthanum fluoride, sodium uranyl acetate, and adsorption-sodium uranyl acetate.

The eighth St. Louis cyclotron neutron bombardment of 775 pounds of UNH (Chicago VIII) ended at 100,100 microampere-hours. It started October 5.

The regular evening seminar of the Chemistry Division was held at 7:45 p.m. in Room 251, Ryerson Laboratory.

Helen worked all day at the Met Lab Information Division typing on my "Table of Isotopes."

Today's top war news comes from the Italian front where the Allies have won four more Italian towns.

Tuesday, October 26, 1943

I replied to Allison about the allocation of the West Stands space being vacated by Sutton's section. I suggested we be given Rooms 3 and 5 and that Ghiorso maintain the alpha and beta counters in Room 8 for use by Sugarman's, Kircher's, and my groups.

Franck informed Thomas at Monsanto that Zachariasen, using the x-ray diffraction technique here, has successfully measured the density of uranium with 2 pieces of metal totaling 70 micrograms that was prepared by Kirk starting with 400 micrograms of the fluoride. On the basis of this success, Franck asked if Thomas could induce the people at Site Y

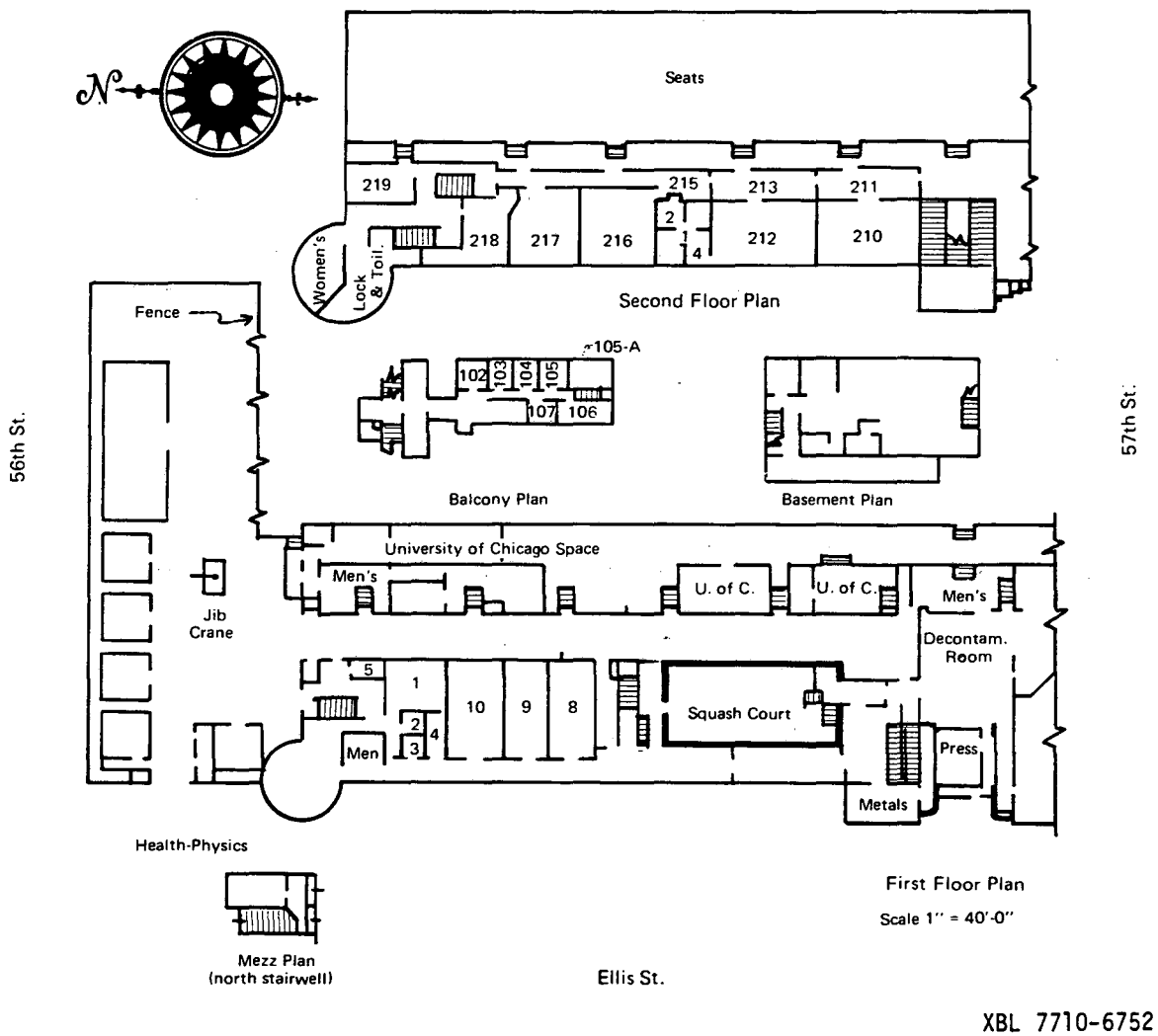

Figure 12. Guide plan -- West Stands.

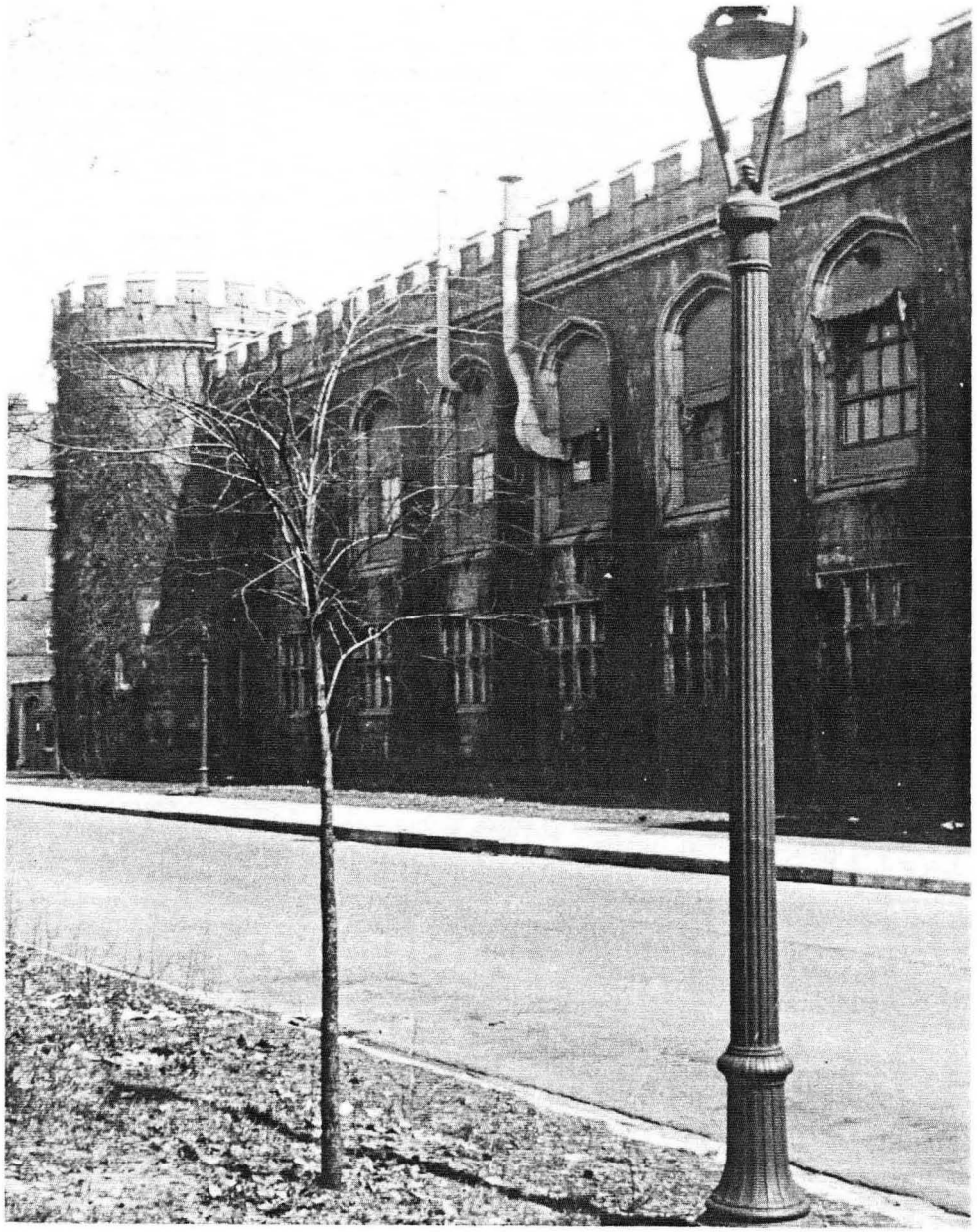

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Figure 13. West Stands. Ellis St. in foreground. 1943.

to send us 500 micrograms of their plutonium for a density determination experiment, which, with luck, could be carried out in a week. He added that he is quite sure the losses will be small.

In the evening Helen and I played cards with the Ghiorsos.

U.S. troops destroyed 123 more Japanese planes during last weekend in more raids on Rabaul.

Wednesday, October 27, 1943

Today's new employee in Section C-I is Kurt A. Kraus, who has been an instructor at Tulane University, New Orleans. He received his Ph.D. from Johns Hopkins in 1941.

In a memo to Franck, Hogness said that Compton called from Clinton and wants a statement on the Chemistry Division's need for 50 additional chemists. He must answer this question for the Army authorities in Washington who cannot understand the need for chemistry expansion and who believe that perhaps there is duplication of work here and at Clinton.

War news from four fronts is favorable to the Allies. Four Japanese attacks were repulsed in New Guinea in the Pacific. Twelve thousand Nazis were slain in the Dnieper trap. The Allies have won ten more towns in Italy. Jugo-Slav partisan fighters have driven the Germans from the steel city of Vares-Mayden, 20 miles north of Sarajevo.

Thursday, October 28, 1943

Katzin wrote to Ray Stoughton in Oak Ridge, detailing the problems being encountered in preparing 1,300 cans of pressed thorium carbonate--a job that will require six to seven weeks.

The regular evening meeting of Section C-I Research Assistants was held at 6:45 p.m. in Room 209, Eckhart Hall.

The evening meeting in my office of the Council of my section was attended by Beaton, Cunningham, Davidson, Dreher, Ghiorso, Kirk, Manning, Orlemann, Thompson, and Willard. Thompson received a cigar from Cunningham as a payoff for his successful experiment on CeF_3 carrying. I mentioned that according to the latest plans, the bombarded metal from Berkeley (100-150 pounds) will be dissolved under the supervision of Kircher's group early next week. The crude crystallized material will be given to Jaffey for ether extraction in the cyclotron lab and sulfur dioxide will be used for reduction of the plutonium. Two ether extractions are planned to remove the bulk of the uranium. The distribution of neptunium will be followed all the way through with Np^{239} tracer in order to recover the Np^{237}. Plutonium will, of course, also be separated and recovered. Jaffey will also ether extract the 775 pounds of St. Louis material (Chicago VIII) while awaiting the material from Kircher. New men here or coming soon and

the groups to which they will be assigned are Kraus (Cunningham), Heath (Davidson), Hellman (Kirk), Watt and Hopkins (Thompson). I raised the problem of metal production. The facilities of the entire section must be placed at the disposal of the metal production group. I reviewed the schedule of plutonium deliveries to Kirk which indicate he will have 375 micrograms soon available, with an additional 125 micrograms to be delivered in 5-10 microgram lots, making a total of 500 micrograms now in sight for delivery. In several weeks there will be another 250 micrograms from the eighth St. Louis bombardment. I estimated that a total of 1,000 micrograms from St. Louis has been delivered to Cunningham; hence, it would appear that not more than 500 micrograms have been lost during the constant use and reworking. To me, this loss is small indeed, considering that this has yielded all of our knowledge concerning plutonium on which the plans for Site X and Site W have been based. I mentioned that Franck has suggested we carry out reduction on a large (300-500 microgram) sample of the fluoride in hopes of producing enough material at one time for x-ray density measurements. Kirk doubted the advisability of this procedure as results with 25 microgram samples are poorer than with 10 microgram samples; furthermore, the present furnace cannot treat samples larger than 100 micrograms. In a discussion of the possibility of using cerous fluoride instead of lanthanum fluoride in plutonium recovery work, mention was made of the alpha counts found in cerium--approximately four alpha counts per minute per milligram of cerium--and we speculated that the counts might be due to naturally-occurring neptunium, an interesting matter to look into if time permits. The topics being worked on by members of Thompson's group were listed as follows:

1. Uranous oxalate - sodium uranyl acetate process (James handling--looks promising)
2. Bismuth arsenate extraction - bismuth phosphate decontamination process (in good shape - Thompson thinks it may be better than the Bismuth Phosphate Process)
3. Zirconium or ceric ion metathesis in concentration process. (James, Malm, Creek)
4. Bismuth phosphate decontamination studies (Thompson, Sutton, and Leader cooperative project--Dean, Gaarder, and Kroner)
5. Cerous fluoride extraction - decontamination process. (Scouting work nearly complete)
6. Solvent partition extraction - decontamination process (Margolis)
7. Adsorption - bismuth phosphate extraction - decontamination process (Thompson and Beaton will cooperate)
8. Ether extraction - bismuth phosphate extraction - decontaminatio process (Meyer, in cooperation with Maloney)

9. Lanthanum phosphate, uranous oxalate and cupferron concentration methods (in the background at present)

10. Uranium hypophosphate for separation of elements 93 and 94 (inactive at present)

11. Concentration of Site W plutonium by peroxide process (Watt and Hopkins will work on this.)

12. Alternate reducing agents for the Bismuth Phosphate Method (peroxide and ferrous ion are being compared)

13. Alternate bismuth phosphate - lanthanum fluoride crossover method

14. Bismuth phosphate extraction and decontamination studies using Site X and Site W concentrations of product and by-products

15. X-ray crystal studies of bismuth phosphate (a sample of bismuth phosphate containing 3% plutonium will be ready for Rose Mooney, a physicist in Zachariasen's group, on Saturday).

Friday, October 29, 1943

Horace H. Hopkins, Jr., started work in our section today. He received a bachelor's degree from Oberlin College this year.

Willard requested of Richmond a salary increase from $31.50 to $37.50 per 40 hour week for my secretary, Edrey Smith, who is now in charge of the section office with three women, and an increase from $28 to $35 for Irma Saxton, Willard's secretary.

Franck sent a memorandum to Oppenheimer apologizing for the trouble caused by his inquiry to Thomas regarding the possibility of obtaining a loan of 500 micrograms of plutonium to make density measurements. "It is not our intention to rob the poor," he said. Franck has now learned that there will be sufficient material on hand for the job, particularly as Zachariasen believes he can work with much smaller quantities than the 100 micrograms of plutonium metal originally specified. The amount of plutonium Franck has in mind is 175 micrograms to be reconditioned from small, scattered units of 5-10 micrograms each and 250 micrograms several weeks from now from our St. Louis bombardment of UNH.

Helen went to the library across the Midway to look up references for me.

In the evening Helen and I went to the Picadilly Theater at 51st and Blackstone to see a double bill: "Let's Face It," with Bob Hope and Betty Hutton and "Forever and a Day," with Ida Lupino and Brian Aherne.

U.S. forces invaded two Japanese islands in the North Solomons.

Saturday, October 30, 1943

I asked Franck if Hilberry could investigate the possibility of obtaining 100 pounds of uranous chloride and other uranous salts from the Harshaw Chemical Company in Cleveland, Ohio. We have a number of needs for these compounds in our work here, and Perlman wants uranium tetrachloride for use at Clinton as a reducing agent.

Two lists of Metallurgical Laboratory Personnel as of October 15, 1943, are being issued, one list (CA-1022) being alphabetical and the other (CA-1015) arranged by organizational unit. The personnel of Section C-I is shown in the following table, while room assignments are shown in Figure 14. Figures 15 and 16 show New Chemistry Building and Chemistry Annex. Room 5 is temporarily unoccupied because of the departure of Halperin and Koshland for Clinton and the remodeling (including the installation of a precipitron) taking place. The glass blowers are still in Room 3, but they will move to the Annex in a week or so. Sometime ago Room 16 was split into two rooms in order to provide Howard Lange, plant curator, with an office. He has been cramped into a space in our storeroom. Recently, however, there has been a change in plans. Room 16A, the smaller part at the south end, is now being used to house the FP-54 electrometer, and Lange is moving into a room in the Annex. Changes have been made in the leadership of Section C-III and Section C-IV of the Chemistry Division. Nathan Sugarman, who was Associate Chief of Section C-III, has now replaced Coryell as Chief, and Herbert A. Potratz is moving up from Associate Chief of Section C-IV to take Boyd's place as Chief. Both Coryell and Boyd are now assigned to work at Clinton Laboratories. A new section, Section C-V, has been added, with Clarence F. Hiskey as the Section Chief. He has five persons under him; they are working on the P-9 (heavy water) pile projects.

Section C-I

Seaborg, Glenn T.	Senior Chemist	Section Chief
Smith, Edrey E.	Secretary to G. T. Seaborg	
Willard, John E.	Senior Chemist	Associate Section Chief
Manning, Winston M.	Chemist	Assistant Section Chief
Anderson, Elsy	Secretary	
Saxton, Irma N.	Secretary	
Donlan, Francis B.	Clerk	
Gavin, Kathleen B.	Clerk	

10/30/43 (cont.)

Group 1

Thompson, Stanley G.	Chemist	Group Leader
Dreher, J. Leonard	Associate Chemist	
Katzin, Leonard I.	Associate Chemist	
Margolis, Asher J.	Associate Engineer	
Creek, George E.	Junior Chemist	
Dean, Floyd M.	Junior Chemist	
Gaarder, Sydney R.	Research Assistant	
Halperin, Joseph	Research Assistant	
James, Ralph A.	Research Assistant	
Johnson, Gordon L.	Research Assistant	
Kroner, James C.	Research Assistant	
Malm, John G.	Research Assistant	
Schuler, Frederic W.	Research Assistant	
Summers, Mildred	Laboratory Assistant	

Group 2

Beaton, Roy H.	du Pont	Group Leader
Fields, Paul R.	Junior Chemist	
La Chapelle, Theodore J.	Research Assistant	
Lincoln, Dwight C.	Research Assistant	
Sheft, Irving	Research Assistant	
Saunders, Addie	Laboratory Assistant	
Young, Robert B.	Laboratory Assistant	

Group 3

Ghiorso, Albert	Associate Chemist	Group Leader
Jaffey, Arthur H.	Associate Chemist	
Kohman, Truman P.	Associate Chemist	
Schoenberg, Bert	Junior Chemist	
Crawford, John A.	Research Assistant	
Heller, Lorraine	Research Assistant	
Scott, Benjamin	Research Assistant	
Walsh, Patricia	Research Assistant	
Lenhard, Madeline	Clerk	

Group 4

Cunningham, Burris B.	Chemist	Group Leader
Patton, Robert L.	Chemist	
Hindman, James C.	Associate Chemist	
Meyer, Fred	Associate Chemist	
Smith, Clifford	Associate Chemist	
Howland, Jerome J.	Junior Chemist	
Magnusson, Lawrence	Junior Chemist	
Baumbach, Nathalie S.	Research Assistant	
Belletire, Frank L.	Technician	

Group 5

Orlemann, Edwin F.	Associate Chemist	Group Leader
Nachtrieb, Norman	Associate Chemist	
Watters, James I.	Associate Chemist	
Dixon, Jonathan S.	Junior Chemist	
Hagemann, French T.	Junior Chemist	

Group 5 (cont.)

Jensen, Lyle H.	Junior Chemist	
Brody, Bernard B.	Research Assistant	
Harding, Reid B.	Laboratory Assistant	

Group 6

Davidson, Norman R.	Associate Chemist	Group Leader
Bradshaw, Benjamin C.	Associate Chemist	
Katz, Joseph J.	Associate Chemist	
Florin, Alan E.	Research Assistant	
Daniels, Minnie	Laboratory Assistant	

Group 7

Kirk, Paul L.	Senior Chemist	Group Leader
Baumbach, Harlan L.	Chemist	
Rosenfels, Richard S.	Chemist	
Dirksen, Alvin J.	Associate Chemist	
Fried, Sherman	Associate Chemist	
Karle, Jerome	Associate Chemist	
Zvolner, Hymen P.	Junior Chemist	
Frank, Ray D.	Research Assistant	
Fulan, Emil H.	Laboratory Assistant	
Jennings, Nellie	Laboratory Assistant	

The Technical Division in its Weekly Report (Memo 137) states that additional runs during the week show the present washing and metathesis procedures in the decontamination step to be satisfactory. They also report that a cooperative program on study of decontamination improvements as drawn up by Thompson, Leader, and Olsen is well underway. The study involves the determination of comparative improvements for a large number of precipitation and holdback carriers.

In the evening the Ghiorsos came to our apartment to play cards.

Cossack horsemen and tanks are chasing German troops across the Steppes, with a big tank battle going into its second day.

Sunday, October 31, 1943

I played golf today.

The Berkeley bombardment arrived today by truck. It consists of 1,100 pounds of uranium metal and 400 pounds of UNH. One hundred pounds of the metal, directly in line with the deuteron beam, will be worked up here to isolate Np^{237}, the remaining 1,000 pounds will go to Clinton Labs for use in separations studies. The UNH will go back to Berkeley; it was included in the truck shipment so it can decay enroute before it is dug out from the shielded container. Joe Hamilton is here to help in the unpacking and in identifying the particular lots of material to be used by us and by Clinton Labs. His wife, Leah, is here with him and they had dinner with Helen and me in our apartment.

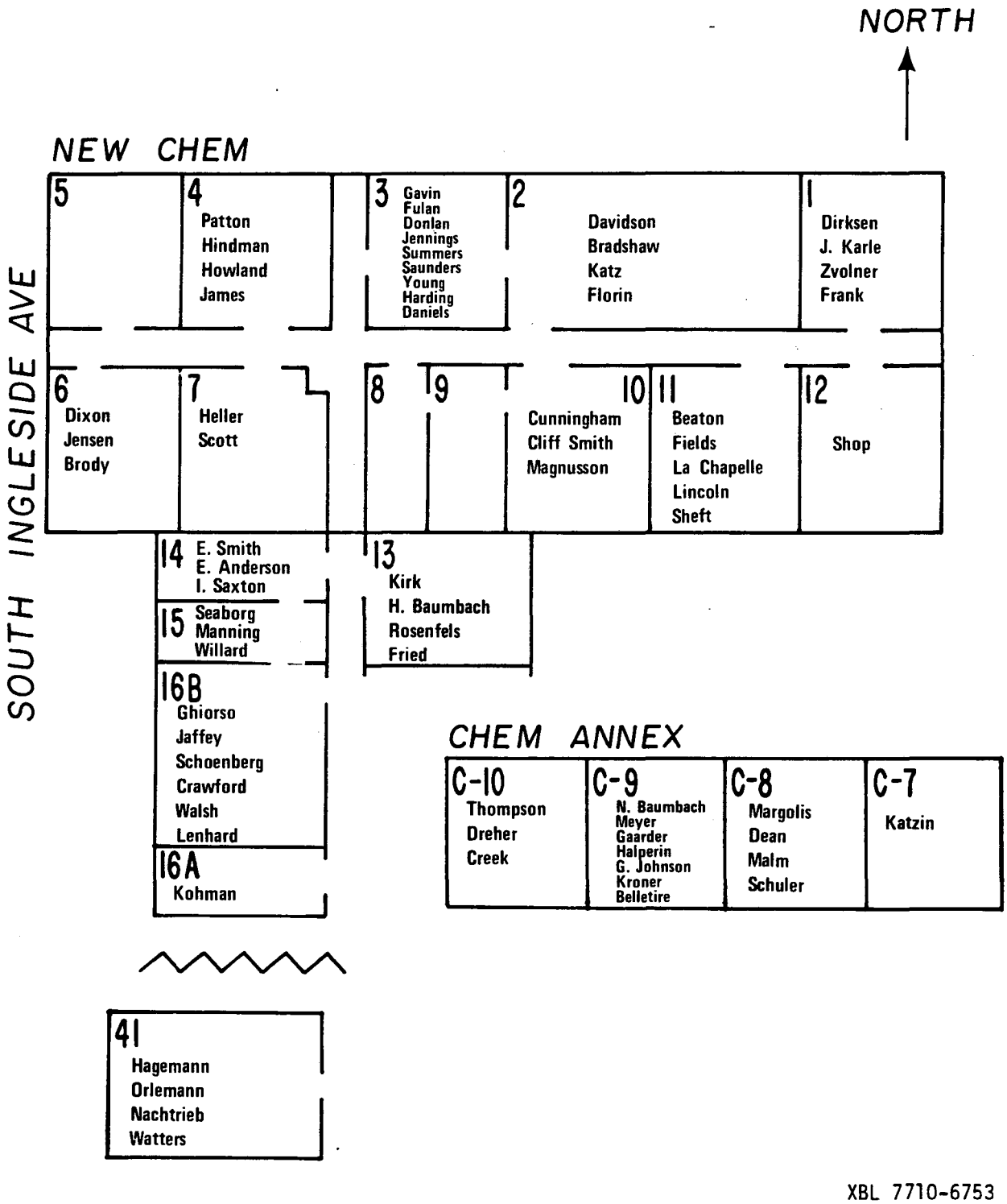

Figure 14. Room assignments. October 1943.

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Figure 15. New Chemistry Building, Autumn 1943. Main Entrance. View is facing southeast from across the street near corner of 56th Street.

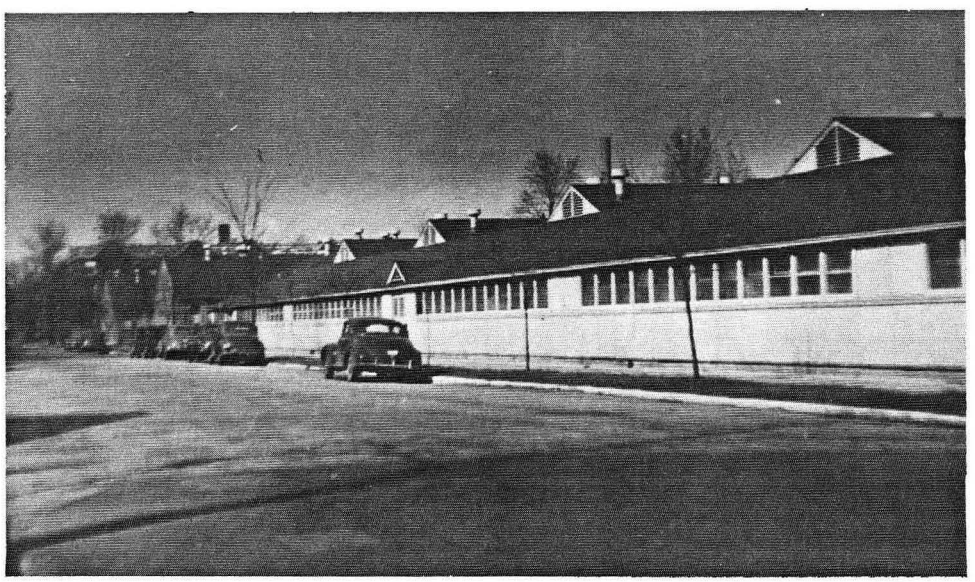

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Figure 16. Chemistry Annex, Autumn 1943. View is facing northeast from across the street near corner of 57th Street.

Monday, November 1, 1943

George W. Watt, on a leave of absence from the University of Texas in Austin where he is an associate professor of chemistry, started working in Section C-I. Watt received his Ph.D. from Ohio State University in 1935. Another new employee today is Roy E. Heath, who was manager of marketing research at the Wyandotte Chemical Corporation in Michigan. He received his Ph.D. in 1940 from Western Reserve in Cleveland, Ohio. His thesis was "Reactions of Anodic Fluorine in Anhydrous HF," which is very apropos to our research here. I have asked him to work in the volatility group on the chemistry of plutonium fluorides. Beatrice Foreman began working in the Kirk-Baumbach group as a technician.

Gordon Johnson began the preparation of a 500-microgram bismuth phosphate sample containing 3% plutonium for x-ray study of its crystal structure by Mrs. Mooney.

I transmitted to Squires a suggested flowsheet to cover the concentration step which follows the bulk reduction (crossover) step at Hanford. I pointed out that the suggested procedure must be considered very tentative since all the steps represent a very great scale-up and it has not yet been possible to determine experimentally a number of the needed constants because of the lack of adequate amounts of plutonium. The flowsheet as outlined provides for the precipitation of plutonous peroxide, without carrier, from the lanthanum-containing solution delivered from the bulk reduction step. Among the estimates used in drawing up the flowsheet are (1) density of 4 for the plutonous peroxide, (2) solubility of plutonous nitrate in 1 N HNO_3 to be either 167.5 g/l or 560 g/l (alternative assumptions for flowsheet variations), (3) solubility of plutonous peroxide to be 300 mg/l at 1 N HNO_3, (4) the formula of plutonous peroxide is PuO_4. I informed Squires that we will have a small group working exclusively on this concentration process within a few weeks. I made the guess that our direct investigations would yield methods whereby the volumes would be even smaller than the more favorable case given in the flowsheet.

The Metallurgical Laboratory Progress Report for October 1943 (MUC-NH-655) was issued. The work of my section is summarized as follows:

The Separation Group have continued their work on the Bismuth Phosphate Process throughout the month. Emphasis was placed on the carrying of product by bismuth phosphate at both "X" and "W" concentrations. Further detail work was done on reducing and oxidizing agents and the effects of operating variables, both on carrying and on decontamination. Work on concentration procedures has continued in an attempt to develop suitable flowsheets for Sites X and W. Some work was done during the month on other possible product elements, such as Np^{239} and U^{239}. In the plutonium metal production and purification work, the program has continued both on volatility methods and on precipitation methods. Some

work was continued during the month at the Metallurgical Laboratory on adsorption, but most of this work has been transferred to Clinton. Total Project expense for the month of October was $530,483. Total personnel on October 31 were 944 (377 academic), a net increase of 39.

Helen took a walk with Wilma in the afternoon and then prepared dinner for Adele Brown and her sister Gloria, who ate with us this evening.

There was an evening seminar of the Chemistry Division at 7:45 p.m. in Room 251, Ryerson Laboratory.

The Russians are closing their trap on the Germans in the Crimea. At the same time the U.S. has shot down 45 more Japanese planes in another raid on Rabaul.

Tuesday, November 2, 1943

The ninth St. Louis cyclotron neutron bombardment of uranium, Chicago IX, started today with about 100,000 microampere-hours scheduled.

James Maloney, who is now the "Information Counselor" in the Technical Division, and I discussed the possibility of using ether extraction as a procedure for removing and concentrating the plutonium remaining in the solution from the last decontamination cycle of the Bismuth Phosphate Process. A counter-current ether extraction column would be used. I expressed my opinion that such a procedure might prove feasible. I also agreed to undertake research on the coupling of the ether extraction method to both the Bismuth Phosphate and Lanthanum Fluoride Processes. Lawrence Myers of Chemistry Section C-IV will operate the extraction column.

Manning sent Brugmann the requested summary of his literature survey of means for controlling algae and slimes in the water systems at Site W. His conclusion is that chlorination is the best single treatment to consider.

Thompson, Kohman, and I, representing Section C-I, attended a Chemistry Division P-9 (Heavy Water) meeting at 4:30 p.m. in Hogness' office (Room 42, New Chemistry). Hogness and Franck were present, as well as Augustine Allen and Arild J. Miller of Section C-II, Sugarman and Anthony Turkevich of Section C-III, Potratz and Mark Fred of Section C-IV, and Hiskey and Maxwell L. Eidendorf of the new Section C-V (concerned with heavy water problems). Hiskey opened the meeting by relating some history of the P-9 project, saying that either Urey or Halban conceived the concept of a slurry pile. Possible materials for use in a slurry pile and possible pile designs were discussed. I suggested we look into the possibility of adding beryllium to the slurry in some form in order to utilize the $Be^9(n,2n)Be^8$ reaction, thereby decreasing the critical size. Hogness asked our section to get out by Wednesday, next week, a summary of how the three proposed slurries (uranium oxide, $Bi_2O_3 \cdot UO_3$, $PbO-UO_3$) fit into the Bismuth Phosphate Process.

11/2/43 (cont.)

Helen and I went to the Frolic Theater on 55th at Ellis Avenue to see Betty Grable, George Montgomery, and Cesar Romero in "Coney Island."

At 11:30 p.m. Stan Thompson and I left Chicago (Englewood Station) by Penn R.R. to Cincinnati, where we again will continue on the L&N R.R. to Knoxville to visit Clinton Laboratories and to attend the Friday progress meeting on the Bismuth Phosphate Process. Compton and Fermi are planning to visit Oak Ridge also. The pile at Clinton Laboratories is now already loaded with uranium slugs and is expected to go critical during the latter part of the week.

Today's paper reports that American ground forces have made a bold invasion on Bougainville Island in the Solomon Islands, threatening Japan's entire position in the southwest Pacific.

Wednesday, November 3, 1943

Stan and I arrived in Knoxville in the afternoon and spent the rest of it playing golf at the Municipal Golf Course.

Thursday, November 4, 1943

Stan and I are now staying at the Guest House in Oak Ridge. We went out to the X-10 area this morning, where there was much excitement. The reactor went critical for the first time at 5:07 a.m. Loading of the 1,248 channels with aluminum-encased uranium slugs started yesterday. Only 31 tons of uranium were required, which filled about half the channels. Whitaker expects to use twice this much uranium in the next month or so to bring the pile up to 1,000 kw, its design capacity. This is the world's third pile to become chain-reacting that we know of, the other two being the Chicago and Argonne piles. It is planned that 5 tons of the irradiated uranium slugs will be removed from the pile November 26. After cooling until December 9, the uranium will be put through the Bismuth Phosphate Process in the Chemical Separations Plant (200 area) followed by the concentration and isolation procedure that hopefully will yield 10-40 milligrams of plutonium by January 15. A run-through of the Chemical Separations Plant will start for the first time around November 24 when one ton of uranium metal from the Argonne pile will be processed for approximately 10 milligrams of plutonium. (See Figure 17 and 18.)

Friday, November 5, 1943

I attended the 1:00 p.m. progress meeting on the Bismuth Phosphate Process at Clinton Laboratories. Thompson and I described our progress in our Section C-I during the last month. Perlman discussed the work of his chemistry section at Clinton Laboratories. Greagor reported on the design, equipment, and operating plans for his semiworks group at Clinton Laboratories. Sutton also reported on process development work of his section in the Technical Division at Chicago. His section has made

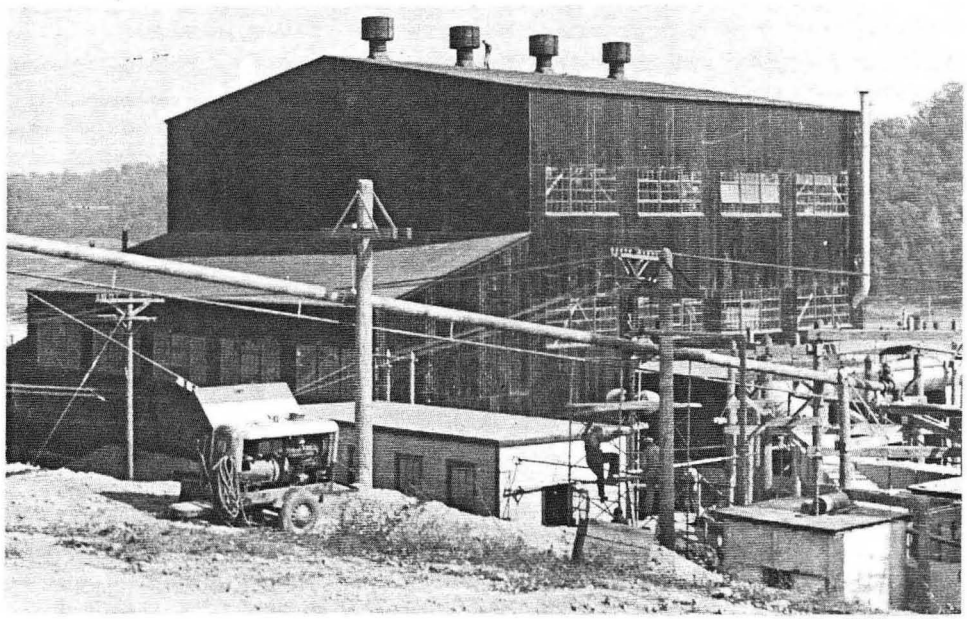

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Figure 17. Pile Building (No. 105), Site X. November 1943.

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Figure 18. Chemical Separations Building (No. 205) and Off-Gas Stack, Site X. November 1943.

special tests of the flowsheet of the Bismuth Phosphate Process as it will be applied to irradiated uranium from the Clinton pile. Satisfactory results have been obtained in the extraction step and the decontamination cycle. The flowsheet conditions for the metathesis step prove to be unsatisfactory, however, since the use of such conditions result in large product losses in the metathesis heels. These losses have been eliminated by changing from NaOH to KOH and doubling the volumes of both alkali and wash water. Greagor gave a detailed report on the changes contemplated in the flowsheet of the Bismuth Phosphate Process for the Clinton Chemical Separations Plant.

We now have a Clinton Steering Committee, consisting of Warren Johnson, Thorfin Hogness, O. C. Greager, Bill Kay, and me, who will meet to coordinate the work on the Bismuth Phosphate Process and related concentration and isolation procedures each month at the time of the Bismuth Phosphate Process Progress Meeting. We met today in Warren Johnson's office to discuss the status of the program and to coordinate the plans of the Chicago and Clinton groups for the coming month so as to avoid duplication and maximize efficiency. We decided that Section C-I will investigate KOH as a substitute for NaOH and the use of Zr^{+4} in the step of dissolving LaF_3, Perlman's section at Clinton will investigate the use of $KOH-K_2CO_3$ for this step, and Greagor at Clinton will purify some lanthanum salt to test its performance vis a vis unpurified lanthanum salt. Hogness asked me to send him a memorandum summarizing the presently contemplated lanthanum fluoride concentration (i.e. crossover) step for use at Clinton and the alternate processes that might eliminate the somewhat lengthy crossover step.

Saturday, November 6, 1943

Stan and I visited the Chemistry Division at Clinton Laboratories to have further discussions with the members of Perlman's section.

In Chicago "Chemical Research - Production and Extraction of Plutonium. Report for Period October 9 - October 31, 1943" (CN-1041) is being issued and summarizes the work in my section. The topics reviewed are: 1. Concentration at Site W by precipitation of plutonium peroxide. Dam, Fries, and Werner. The following constants necessary to set quantitatively the optimum flowsheet conditions are identified for which an intensive investigation is being initiated: (a) solubility of plutonous nitrate in the nitric acid used to dissolve the plutonous peroxide, (b) solubility of plutonous peroxide as a function of acid concentration, (c) liquid holdup per weight of plutonous peroxide. 2. Concentration at Site X using uranous oxalate as carrier. Dam and Fries. A further study has been made of the variables affecting the carrying of plutonium from HCl solutions. Carrying of 95% or better is obtained by the addition of U^{+4} in one or two shots to give a final concentration of either 1 or 2 mg/ml in a solution 5 N in HCl and 0.4 M in $H_2C_2O_4$. Carrying from similar solutions containing 0.1 M Fe^{+3} is unsatisfactory. Experiments using a semiworks solution showed good

carrying by $U(C_2O_4)_2$ from solutions 0.05 M in Fe^{+3}. They find also that traces of K^+ are necessary for good carrying. 3. Concentration by Cb_2O_5. Fries and Werner. Experiments have been conducted on the dissolving of Cb_2O_5 (length of standing has no effect on solubility), carrying of plutonium by $La(OH)_3$ from columbate solution (93% carrying), carrying by $La_2(C_2O_4)_3$ from oxalate-columbate solution (95% carrying), loss of plutonium on washing Cb_2O_5 precipitates (no appreciable loss). In a test of the Cb_2O_5 - LaF_3 concentration scheme, two LaF_3 precipitations, with one intervening Cb_2O_5 precipitation, there results an overall yield of 81% for tracer amounts of plutonium. 4. Use of uranous oxalate in an extraction or crossover step preceding decontamination of plutonium with sodium uranyl acetate. Gaarder, Halperin, James, Koshland, Kroner, Malm, and Thompson. Carrying of plutonium by $U(C_2O_4)_2$ in 10% UNH is found to be greater than 95%, both with tracer and with Site W concentrations of plutonium. Experiments have been conducted that show that a change-over from the Bismuth Phosphate Process, avoiding the use of HCl, can be made by reducing the solution with $H_2C_2O_4$ after a by-product bismuth phosphate precipitate and then adding U^{+4} to give $U(C_2O_4)_2$ which carries the plutonium precipitate. Carrying of plutonium at Site W concentrations is excellent; the presence of 0.03 M Fe^{+3}, however, causes the method to fail. Since the use of dichromate in nitric acid solutions has been developed by the California group for oxidation of plutonium in the Sodium Uranyl Acetate Process, its use as an oxidizing and dissolving agent for $U(C_2O_4)_2$ and plutonium in the crossover to the Sodium Uranyl Acetate Process has been studied and satisfactory conditions found. Flowsheets are worked out, on a one liter scale, for the $U(C_2O_4)_2$ - $NaUO_2Ac_3$ process and for the bismuth phosphate - sodium uranyl acetate. Using the flowsheet procedures, they have achieved plutonium recoveries of 94% and 95% in steel and in glass, respectively, and decontamination factors of 350 are obtained by one extraction-step precipitation with $U(C_2O_4)_2$, followed by a product precipitation with sodium uranyl acetate. 5. Bismuth arsenate method for the extraction of plutonium from UNH solutions. Bradshaw, Dreher, Johnson, and Thompson. Bismuth arsenate has been found to be very similar to bismuth phosphate as a carrier for plutonium in 20% UNH solutions. Experiments have been performed that show that carrying of plutonium is not affected when the HNO_3 concentration is varied from 0.5 N to 1.0 N. Other experiments demonstrate that carrying is reduced in the presence of 0.2 M Fe^{+3}. Microchemical tests at a bismuth/plutonium ratio of 69 show greater than 90% carrying. The solubility of plutonium arsenate in 20% UNH-HNO_3 solution has been determined to be 480 mg/l at 75°C. A bismuth arsenate extraction step followed by two bismuth phosphate decontamination cycles has been run on a liter scale in parallel with a bismuth phosphate extraction step followed by two bismuth phosphate decontamination cycles. Overall decontamination factors are about 10^4 for both cases. Overall product yield is 97.4% when arsenate extraction is used and 89.5% when phosphate extraction is used.

I had stomach sickness (Clinton Fever) and was not able to eat lunch. Stan, I, and others went to Whittle Springs in the afternoon; I was in no mood to play golf, but the others did. Stan and I took the Southern R.R. train from Knoxville to Cincinnati, continuing to Chicago on the New York Central R.R. We shared a drawing room with Bill Rubinson of Coryell's section.

Sunday, November 7, 1943

I and the other members of the Met Lab who went to Oak Ridge for the Bismuth Phosphate Process Meeting arrived in Chicago at 2:30 p.m. on the New York Central R.R. I am feeling somewhat better.

Last week while I was in Oak Ridge Helen spent each day with friends. On Wednesday she had lunch with Wilma Ghiorso at the Thomas Drugstore (NW corner of Blackstone and 53rd), saw Dagmar Dreher in the afternoon, and then had dinner and played cards with the Ghiorsos in the evening. She also entertained Joe Hamilton's wife, Leah, who remained in Chicago all week. Thursday she and Leah went downtown in the afternoon, had dinner at Tiffin's Tea Room above the downtown Stop and Shop Market, and then went to a movie. They spent the next day together as well, which included lunch and dinner. On Saturday, after Leah left, Helen had Alice Thompson and Wilma to lunch. That evening Wilma and Al came over to our apartment for waffles.

Monday, November 8, 1943

The first thing I learned about when I came to the office this morning was the accident that occurred Saturday morning during the early hours. Brody, Frank, and Halperin were conducting the ether extraction on the UNH from the eighth St. Louis bombardment when the 50-liter flask in which the extraction was being run broke. The mishap occurred in the second extraction of the 775 pounds of UNH that had undergone a 100,000 microampere-hour neutron bombardment. Because the amount of salt recrystallized was 80 pounds, it was decided to re-extract it in two batches. In the process the flask broke and its contents flowed into the metal pan beneath, placed there for just such an eventuality. The lead brick shielding and the wooden frame had to be dismantled so the solution could be poured into another flask. Because of the size and unwieldiness of the pan, this was a lengthy process; the personnel were in direct and almost bodily contact with the solution and its containers for most of an hour. Fortunately, very little of the material was lost. The accident is believed to have been caused by the rotating motor shaft knocking a porcelain chip (used in determining density of the solution during recrystallization) against the side of the flask.

In the course of their attempts to produce powdered plutonium metal suitable for study by the x-ray diffraction method, Kirk and Baumbach submitted a sample that turned out to be plutonium dioxide weighing 10 micrograms which they gave to Zachariasen last Wednesday; he succeeded in obtaining a satisfactory x-ray diffraction pattern, enabling him to determine its crystal structure. This is a tremendous accomplishment! This probably means that it is possible, even on a microgram scale, to obtain the structure of numerous plutonium compounds. This would be a tremendous boost to our program. Consider the following results. The oxide has the fluorite structure. There are four molecules in the unit cube which has an edge equal to 5.370 ± 0.005 Å. The density

of the oxide is 11.54 gm/cm^3. For comparison, the edge of the unit cube of thorium oxide and uranium oxide is 5.58 and 5.47 Å, respectively. All of this information was obtained with 10 micrograms!

On Wednesday Katzin sent Stoughton the current status of the pressing and canning of the thorium carbonate. He mentioned that the operation has been interrupted by a couple of emergencies that came up in the lab, but that the work was finished yesterday.

The first meeting of the P-9 Information Committee took place last Wednesday at 2:00 p.m. in Room 209, Eckhart Hall, with Wigner presiding. Others present were Allison, Burton, Chapin, Creutz, Hiskey, Hogness, Huffman, Ohlinger, Sugarman, Szilard, Vernon, Willard, Young, and Zinn. Wigner, speaking for the P-9 Steering Committee, reviewed the general status of the P-9 program and outlined in brief detail the events and directives that have led to the present program. He read the directive from General Groves for the Military Policy Committee that states "that our efforts (should) not exceed the construction of a low-powered heavy water pile for use as a general experimental tool and the setting up of a group of scientists assisted by a small number of engineers to study the general problems and to prepare the way in the event that it should become necessary to carry on rapid development." Wigner said that the application of this directive is interpreted by the Steering Committee as meaning a selection of suitable arrangements for a production pile should it be needed, with the principal effort being concerned with the development of the slurry pile. He mentioned that at the present time there are only two groups working full time on the slurry pile: these are Vernon's and Hiskey's, representing engineering and chemical research.

After Wigner spoke, each of the individuals on the Committee discussed his function as far as the P-9 program is concerned. Willard, speaking for Section C-I, said that our section is making an effort to adapt existing methods to the extraction of plutonium produced in a slurry pile; Hogness asked us to summarize this for him in a memorandum. Allison informed the Committee his function is to serve as interim director of the P-9 Steering Committee until Smyth arrives from Princeton November 9 to take up his duties as Associate Laboratory Director and as director of the P-9 Steering Committee; he plans to spend alternate weeks in Chicago. Zinn stated that he is principally concerned with the immediate construction of the heterogeneous P-9 experimental pile to operate at 250 kw. Concrete for the shield is being poured at the present time. It was decided that the next meeting of the P-9 Information Committee is to take place at Argonne, November 10, and that Zinn will arrange for a bus to leave that day at 11:30 a.m. from Eckhart Hall for Information Committee members.

The Chemistry Division Section Chiefs met last Wednesday at 5:00 p.m. in Franck's office, attended on my behalf by Willard. Security and medical matters were discussed. There was a review of personnel requirements in Chemistry; Willard stated that Section C-I can use 22 additional men (4 on fundamental separations research, 4 on extraction, 5 on concentration, 3 on purification, 2 on reduction of plutonium to metal, 2 on the U^{233} problem, and 2 on recovery of Pu^{239} from residues).

11/8/43 (cont.)

Last Friday my office received a wire addressed to me from Albaugh saying his leave-of-absence negotiations are proceeding favorably and that Don Carr, Research Manager of Union Oil Company, will be in Chicago this week and will probably visit me.

In a memo to Doan last Friday, Hilberry asked that the batch of 100 micrograms of plutonium recently isolated at Clinton be transferred to my section, where it is urgently needed for work of high priority--he has in mind our high priority plutonium metal production program.

There was another piece of good news. On Saturday Baumbach prepared plutonium metal by reduction of 50 micrograms of plutonium tetrafluoride with barium in a thorium oxide crucible. The metal formed was finely divided, coating practically the whole inside of the crucible. A few pieces, perhaps 1-2 micrograms each, were formed and given to Kirk for density measurements.

Today I jotted down all alternate processes for plutonium recovery I can think of that feasibly might be employed on a plant-size scale as follows:

1. Wet fluoride includes LaF_3 and CeF_3 processes and Ce^{+4}, Na_3BiO_3, metathesis, etc., coupling.
2. Sodium uranyl acetate.
3. Dry fluoride.
4. Adsorption - sodium uranyl acetate.
5. Adsorption - bismuth phosphate.
6. Adsorption - wet fluoride.
7. Adsorption (extraction and decontamination).
8. Uranous oxalate - sodium uranyl acetate.
9. $BiAsO_4$ - bismuth phosphate.
10. Ether cycle - bismuth phosphate.
11. Ether cycle - wet fluoride.
12. Ether cycle - sodium uranyl acetate.
13. Ether cycles (extraction and decontamination).
14. Bismuth phosphate - sodium uranyl acetate.

Included in the Technical Division report on work completed during the week of October 30 to November 6 (Memo 147) was the work by Sutton's people on the decontamination and concentration steps in the Bismuth Phosphate Process. They find 3,000 to 9,000-fold gamma decontamination in recent Clinton flowsheet tests involving an extraction, a decontamination cycle, and crossover cycle. Significant decontamination improvement (60-90 fold) has been obtained by the use of columbium oxide-ceric phosphate and zirconium hydroxide-ceric phosphate as by-product precipitation carriers; the work is being done in cooperation with the chemistry division.

"Chemical Research - Special Chemistry of Plutonium, Report for Month Ending November 6, 1943" (CK-1072) is being issued. The contents are as follows: 1. Self-absorption in solid metal alpha emitters. Jaffey has calculated the self-absorption of alpha particles by spherical pieces of plutonium and finds that the fraction absorbed is greater than 70 percent, even for one microgram pieces, indicating that direct counting cannot be used to assay pieces of metal. 2. Precipitation of plutonium molybdate as a means of purification. Jensen, Cunningham, Orlemann. The solubility of plutonium molybdate in the reduced state has been determined on a microchemical scale to be about 16 mg per liter in the pH range from 2 to 4 in the presence of excess molybdate. In its properties it closely resembles thorium molybdate. This is of interest because preliminary experiments on the purification of thorium by molybdate precipitation and recrystallization show a promising degree of separation from several added light element impurities. 3. Use of ether extraction in the purification of plutonium. Brody and Orlemann. Results obtained by shaking equal volumes of the aqueous phase and ether show that ether extraction might be used to purify plutonium (oxidized) nitrate from lithium, sodium, potassium, magnesium, calcium, aluminum, and phosphorus; possibly from beryllium, silicon, and iron; but not from boron. 4. Relative volatilization of plutonium and uranium oxides. Bradshaw and Davidson. At a temperature of 1,350°, a plutonium oxide sample has been volatilized from a platinum filament in an atmosphere of oxygen at a rate of 0.01 microgram per hour, suggesting that plutonium oxide is sufficiently refractory so that the pyroelectric method of analysis for light impurities will be applicable to it.

The evening seminar of the Chemistry Division met as usual at 7:45 p.m. in Room 251, Ryerson Laboratory.

Today's headlines report that London was bombed in the crowded suburban areas last night, causing many casualties.

Tuesday, November 9, 1943

Recently Joe Katz built a better apparatus for the production of HBr in which the HBr is produced by the direct combination of hydrogen and bromine. He has used this apparatus to treat a number of samples of tracer plutonium, in the form of the oxide, with HBr over a range of temperatures for a half hour at each temperature. Last Friday he completed a study in which he finds no volatilization at 300, 400, or 500°C; a little volatilization at 600 and 705°C; and essentially complete volatilization at 804°C. This indicates that a volatile plutonium bromide is produced under these conditions at temperatures near 800°C.

Kirk made a plutonium metal density determination by the capillary technique, using two of the pieces of plutonium that were reduced with barium by Baumbach on Saturday. The pieces were placed in a capillary tube 118 microns in diameter and centrifuged down into dibutylphthalate solution whose meniscus had been previously measured using an ocular micrometer. After centrifuging, the meniscus was found to have moved

41.6 microns, corresponding to a volume displacement of 0.000462 cubic millimeter. The pieces were removed and weighed at 6.73 micrograms. Using this weight and the volume displacement, Kirk calculated the density to be 14.6 grams per cubic centimeter. The larger piece, weighing 4.22 micrograms, was re-run in the capillary system and a density of 14.1 grams per cc calculated. Kirk considers these values low but is uncertain of the reason. These are the most successful experiments to date; however, they are disappointing since it is expected that the density of metallic plutonium should be at least as large as that of metallic uranium (19.0 grams per cc). Such low values would mean that the critical mass of a nuclear weapon made from plutonium would be larger than planned, thus placing a larger burden on the Hanford production plant.

Today Baumbach reduced 35 micrograms of PuF_4 with barium at 1,600°C. The product had many bright metallic surfaces, but almost all the metal was found to wet the crucible and no pure pieces were removable. One fairly good looking piece was removed and used to prepare the hydride. This was done by placing the piece in a capillary system that was then filled with hydrogen gas and sealed with a drop of mercury. When heated to 210°C, the particle disintegrated and increased in volume giving a gray black powder. After successive heatings to an ultimate temperature of 320°C, the resulting change in the position of the mercury in the capillary tube was measured as an indication of the amount of hydrogen taken up by the plutonium. The tube was allowed to stand overnight at room temperature.

I notified John Chipman that Kirk, Baumbach, Rosenfels, Willard, Manning, and I will attend the November 15 conference here on the metallurgy of plutonium, called by Thomas. I included a tentative program of our contributions as follows:

1. Discussion of status of preparation of metal (Kirk)
    a. By metals of first and second group
    b. Atomic hydrogen
    c. Electroysis
2. Discussion of the attempts to determine the density of the metal (Kirk)
    a. Preparation for x-ray diffraction studies
    b. Direct microchemical determination
3. Discussion of hydride formation studies (Baumbach)

H. D. Smyth began work as Associate Laboratory Director of the Met Lab and as director of the P-9 Steering Committee. Smyth was a member of the S-1 Section (The National Defense Research Committee Uranium Section) before it became the S-1 Executive Committee in June 1942. He comes from Princeton University where he is Chairman of the Department of Physics.

As requested during the P-9 Information Committee Meeting of November 3, I sent a memorandum (MUC-GTS-290) to Hogness, with copies to Franck and Hiskey, outlining our present thoughts on extracting plutonium from the uranium of the P-9 slurry production pile, using as nearly as possible the Bismuth Phosphate Process. I suggested that the experimental problems to be solved would include the determination of decontamination factors when the fission products are present in concentrations equivalent to 750 grams of plutonium per ton, the efficiency of carrying of plutonium by bismuth phosphate at higher plutonium concentrations and lower bismuth/plutonium ratios than now anticipated at Site W (planned for 250 grams of plutonium per ton), the possibility of using neutron absorbing elements as carriers or diluents in order to allow processing of larger batches of plutonium without interference from critical mass effects, and the effect of impurities introduced by the pipes of the slurrying system.

In response to Hogness' request last Friday, I summarized (MUC-GTS-289) the status of five concentration methods for possible use in room D at Clinton following the bismuth phosphate extraction step and one bismuth phosphate decontamination cycle. First I reviewed the present concentration (crossover) method, which involves (1) dissolving in nitric acid the bismuth phosphate from the decontamination cycle, (2) oxidation of plutonium, (3) by-product precipitation of the bismuth phosphate, (4) reduction of plutonium, (5) precipitation of lanthanum fluoride, (6) metathesis of lanthanum fluoride with hydroxide, (7) dissolution in nitric acid, (8) oxidation of plutonium, (9) by-product lanthanum fluoride precipitation, (10) reduction of plutonium, (11) product lanthanum fluoride precipitation, (12) repetition of steps 6 to 11, inclusive, before the product is turned over to the chemists. (We call the first five steps of the method the "bulk reduction" cycle or the "crossover" cycle.) The four other concentration methods I reviewed were (a) concentration by dissolving the bismuth phosphate in hydrochloric acid and precipitating lanthanum fluoride, (b) precipitation of lanthanum fluoride from nitric acid solutions of bismuth phosphate, (c) uranous oxalate precipitation from hydrochloric acid solutions of bismuth phosphate, and (d) method involving precipitation of uranous oxalate from nitric acid solutions of bismuth phosphate, all eliminating the more complicated crossover steps.

Helen went downtown in Chicago to shop today. This evening we had dinner with the Baumbachs at the Hyde Park YMCA (on 51st Street between Blackstone and Dorchester Avenues).

The Soviet army is pursuing remnants of twelve German divisions west of Kiev, where they have pushed the Germans.

Wednesday, November 10, 1943

Continuing his plutonium hydriding experiment, Baumbach observed that the position of the mercury in the capillary tube has not changed overnight, corresponding to a volume of 0.385 cu.mm of hydrogen consumed

in the reaction. He dissolved the material in perchloric acid, counted the alpha particles in an aliquot portion, and determined the weight of the plutonium as 2.77 micrograms. From these data he computes the formula of plutonium hydride as $PuH_{2.97}$. This is the first unequivocal proof that plutonium metal has indeed been produced!

Baumbach next carried out two further reductions of PuF_4 in an all thoria double crucible system using magnesium rather than barium as the reductant. The second reduction, using 30-40 micrograms of PuF_4, appeared very successful and yielded two spheres of bright plutonium with no wetting of the crucible. The weights of the spheres were 21.4 and 6.41 micrograms. These were turned over to Kirk, who in the meantime recalibrated the ocular micrometer used to read the shifts in the dibutylphthalate meniscus. Using the new calibration, he recalculated his density determinations of yesterday and obtained revised values of 15.5 for the first run and 15.0 for the second run.

As a member of the P-9 Information Committee I attended the second meeting of the Committee at the Argonne site at 1:45 p.m. I left the university campus by bus from Eckhart Hall at 11:30 a.m. and ate lunch at the site. (See Figure 19.) Others present at the meeting were Allison, Burton, Chapin, Creutz, Hiskey, Hogness, Ohlinger, Smyth, Sugarman, Szilard, Vernon, Wigner, Young, and Zinn. Prior to the meeting Zinn took us around and showed us the Argonne pile. He demonstrated a method for testing for impurities in uranium metal by pulling the uranium into the pile in a testing stringer and showing us the effect on the reproduction factor. The question of meeting times was discussed; and it was decided that the next two meetings of the Information Committee will be on November 17 and November 25, so Smyth can attend them during his scheduled time in Chicago. There was a discussion of the possible adverse effect of O^{18} present in the heavy water as currently produced--Zinn is worried about radioactivity in the system outside the pile due to the reactions O^{18}(n,gamma)O^{19}; $O^{19} \longrightarrow$ beta + gamma; gamma + $H^2 \longrightarrow$ neutron. After the meeting the other members of the Information Committee (except Smyth, Hiskey, Hogness, Wigner, and Zinn, who are also members of the Steering Committee which continued in session), and I went out on the grounds to look over the construction of the P-9 pile building and wander about the area.

I attended the regular meeting of the Purification and Metal Production Groups of my section at 7:45 in Room 209, Eckhart Hall. Others attending were Baumbach, Davidson, Dirksen, Dixon, Frank, Florin, Fred, Ghiorso, Hagemann, Hopkins, Jaffey, Karle, Katz, Katzin, Kohman, Kraus, Magel, Malm, Manning, Margolis, Marshall, Meyer, Nachtrieb, Orlemann, Patton, Potratz, Schuler, Clifford Smith, Watt, Watters, and Zvolner. Orlemann conducted the meeting. Katz discussed his recent experiments on the formation of plutonium bromide and described the apparatus for the production of anhydrous hydrogen bromide as well as the apparatus with which the various materials examined may be treated with hydrogen bromide. Present results indicate that conversion of UO_2 to UBr_4 takes place at a lower temperature than with plutonium bromide. Smith reported on bromination followed by partial sublimation of an alloy of plutonium

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Figure 19. Site A -- Autumn 1943.

and uranium containing 1% plutonium; the results indicate that the vapor pressure of plutonium bromide is approximately that of uranium bromide (UBr_4), perhaps one quarter of it. Baumbach reported that the best method of producing metallic plutonium evolved so far utilizes double thoria crucibles whose contents are heated to 1,600°C in the presence of a metallic reducing agent. His experiments this afternoon using magnesium yielded excellent results in that two large metal buttons were obtained with no wetting of the crucible. These buttons are being used by Kirk this evening for density determinations. Baumbach also reported that a number of samples of plutonium metal and compounds have been submitted to Zachariasen for x-ray density determinations. By this method the density of the sample of plutonium dioxide was determined to be 11.53 gm/cm^3. One sample of "metal" prepared by low temperature reduction of the fluoride by sodium gave a complicated x-ray pattern that indicates the presence of PuF_3. Another sample of barium-reduced material failed to give identifiable lines. Baumbach discussed his success in the formation of plutonium hydride today using his capillary apparatus, which indicates the formula of plutonium hydride to be $PuH_{2.97}$. Watters described a colorimetric method for determining chlorine. Patton discussed recent ether extraction work on a micro-scale for the purpose of determining the stability of oxidized plutonium in an ether phase. It was found that oxidized plutonium is reduced at a rate of less than 1.5% per hour. Nachtrieb, who is working jointly with Boyd's and my sections, reported that, using a high-powered spark, the upper limit of the sensitivity of the spectrographic method has been reached; on a one-milligram sample the necessary limits are just about attained. Ghiorso reported on developments in the electronics group. A portable low geometry alpha counter for detecting alpha emitters that have been spread as contamination in the laboratory is being built. Nitrogen counters are being constructed, and a vacuum low geometry counter is now in operation. Magel discussed vapor pressure work on plutonium at Site B that indicates the vapor pressure of plutonium oxides at 1600° C appears to be 6.4×10^{-6} mm of mercury.

I learned after the meeting that Kirk made two more density runs using the larger pieces he received from Baumbach this afternoon and obtained density values of 15.5 for the larger (21.38 microgram) piece and 15.6 for both pieces together (27.79 micrograms). This work kept him occupied throughout the evening.

Thursday, November 11, 1943

Willard is at Clinton Laboratories to attend an afternoon conference on alternative processes for plutonium extraction. A principal reason for this meeting of high level people is Conant's concern that duplicative and unnecessary research on such alternative processes is being carried out.

Lois Moquin in Berkeley wrote asking for the negatives of the wall chart of isotopes so additional copies can be made for the "up and coming young chemists." She added a personal note that she had just

gone to court that day and been granted an interlocutory decree in her divorce proceedings and felt happier already.

Manning and I attended a Section Chiefs' meeting in Franck's office. Compton is planning to review Met Lab personnel needs with Conant; so we were asked to provide information on our manpower needs to Franck by Saturday morning. It was announced that the medical group will install a bloodcounting set-up in the room across the hall from Franck's office (NC 43). We were also told that uranium and its compounds must not be ordered through ordinary channels, but through the Army.

D. J. Hughes, Assistant to Compton, requested that Major Peterson release 300 pounds of 2-inch uranium cubes to me in order to be delivered to Hamilton in Berkeley. Hamilton will arrange for a neutron bombardment of the cubes at the cyclotron to produce Np^{237} for us.

The evening meeting of the Council of my section in my office at 7:45 was attended by Beaton, Cunningham, Davidson, Dreher, Ghiorso, Kirk, Orlemann, Manning, Thompson, and me. With regard to moving schedules, it was noted that the remodeling of Room 5 is approaching completion and may be ready for occupancy next week. The precipitron for Room 5 is due on Monday. The glass blowers will move to the Annex in about a week. Donlan's group will then move into the present glass blowers' room (number 3). Beaton's group will move to the West Stands about December 1. Four of Thompson's men--Dreher, Dean, Margolis, and either Malm or Creek--also will move to the West Stands then. The construction of the new quartz balance is being held up because the necessary optical parts cannot be secured for many months. Manning will see if the Army office can help. A ton of metal from the present Argonne pile that has operated for 4,300 kw-hours is to be shipped to Clinton in the near future for processing in the Clinton plant.

When we finished the discussion of general matters, we turned our attention to the subject of research: Hamilton will subject about 200 pounds of uranium metal to a bombardment with very fast neutrons with the Berkeley cyclotron as soon as we can get the metal to him. It is hoped to produce considerable Np^{237} by this process. We are still waiting for a dissolver from Clinton in order to process over 200 pounds of uranium metal from the last Berkeley bombardment. There is a possibility that a lithium target will be used in the St. Louis cyclotron near the end of the present (Chicago IX) neutron bombardment in order to step up the production of 93^{237}. I asked Cunningham to prepare a report on the recent plutonium recovery experiments and to describe in particular the use of Ce^{+4} coupling. Cunningham announced that there remains about 200 micrograms of plutonium to be delivered to Kirk in the near future. After that there will be no more forthcoming until material used by Kirk's group is recovered. Katz has called attention to the recent synthesis of sodium thiophosphate. It might be desirable to test this substance to precipitate a carrier for plutonium. The Victor Chemical Company will be consulted concerning sodium thiophosphate. Attention was again called to the "W" concentration flowsheet that has been recently prepared. A summary

of concentration methods for possible use at Clinton has just been prepared. The recent preparation of uranium-plutonium alloys was reported. Baumbach will be able to prepare small quantities of such alloys for use by the other groups. Such material is preferable to neutron-bombarded uranium for such work as distillation for vapor pressure measurements of plutonium. George R. Harrison of M.I.T. will be at the Met Lab on Thursday, November 18, for a meeting to decide whether or not we should go into vacuum spectroscopy. Dempster is continuing work on the mass spectrograph method for the detection and measurement of critical light impurities in the final product, particularly for the determination of oxygen.

Ben Scott in Ghiorso's Instruments Group left today for the polonium laboratory operated by the Monsanto Chemical Company in Dayton, Ohio. He will be there for two weeks to help establish facilities for alpha and beta counting.

The Research Assistants in Section C-I held their meeting as usual at 6:45 p.m. in Room 209, Eckhart Hall.

Helen had chocolate with Wilma in the afternoon and later spent the evening with her.

The U.S. Marines are successfully defeating the Japanese landing force on Bougainville Island, according to today's newspaper.

Friday, November 12, 1943

I received a copy of a memorandum from Creutz to Allison reporting on a meeting Wednesday night between Creutz, Zinn, Young, and Wigner to investigate the necessity of removing O^{18} from heavy water for pile use. It was concluded that, as far as the Argonne experimental pile is concerned, the presence of excess O^{18} will not cause trouble.

I attended a 10:00 a.m. meeting in Room 251, Ryerson Laboratory about Site W blueprints. Squires discussed the facilities in Building 224, "Recovery." I learned that Hanford can run a bismuth phosphate by-product and complete lanthanum fluoride cycle without using any equipment for more than one operation. Squires provided information on the functions and equipment of all six cells in Building 224 where the lanthanum fluoride bulk reduction (crossover) cycle will be carried out.

In a three-page memorandum to Franck I outlined our needs for additional chemists in Section C-I. I pointed out that only the extraction and decontamination steps have gone as far as a semiworks scale, and that we are charged with developing the other three essential steps (concentration, purification, and metal production) to the point where they may be put in operation. I also mentioned our responsibilities for scouting work on the fundamental chemistry of plutonium and on processes

for the P-9 and the U-233 work. I tabulated our additional manpower needs as follows:

Concentration (3)
Purification (2)
Metal production (2)
Extraction, decontamination, and bulk reduction (4)
Extraction and decontamination of plutonium from the P-9 pile (4)
Recovery of plutonium from lab residues (2)

Of the total of 17 men needed, I said, we are in contact with four who have agreed to join us within the next month, and have started preliminary negotiations with four or five other men, some of whom may be available to us.

Hogness, in referring to my November 9 summary of concentration processes, asked Warren Johnson to carry out that part of the program I indicated should be done at Clinton Laboratories.

The "Monthly Report on the Activities of the Chemistry Division" (MUC-JF-110) was issued. This included summaries of the work of all five sections plus the California Projects under Latimer and Eastman. The work of Section C-I was summarized in the following words:

Extraction, decontamination, and concentration scouting work

Bismuth Phosphate Process. Oxalic acid reduction of plutonium is not quantitative, and its use is therefore not recommended. The density of $BiPO_4$ was found to be 5.5. X-ray studies of $BiPO_4$ containing plutonium are being continued with Zachariasen.

Decontamination studies in cooperation with C-III and Process Development Section are being continued. The use of zirconium and columbium compounds used as scavenger precipitates improve effectively the decontamination. (7 men; C-2 and A-1 priority)

Crossover from Bismuth Phosphate to Uranyl Acetate. Crossover to sodium uranyl acetate is possible by the use of uranous oxalate as a precipitation carrier. It carries plutonium > 95% in 10% $UO_2(NO_3)_2 \cdot 6H_2O$ solutions at all concentrations of plutonium. $H_2C_2O_4$ was used with good results as a reducing agent of plutonium and dichromate for oxidation.

Flowsheets have been worked out on a one-liter scale for the $U(C_2O_4)_2$ - $NaUO_2Ac_3$ process and the $BiPO_4$ - $NaUO_2Ac_3$ crossover. With the flowsheet procedures, plutonium recoveries in steel and glass of 95% and decontamination factors of 350 were obtained by one extraction step precipitation with $U(C_2O_4)_2$ followed by a product precipitation with $NaUO_2Ac_3$. (1 man; B-1 priority)

Bismuth Arsenate Extraction. Bismuth arsenate behaves very similarly to bismuth phosphate as carrier for plutonium and even has some advantages: It carries at 1 N HNO_3; (addition of H_2SO_4 may be omitted; it is needed in the bismuth phosphate method to prevent precipitation of uranyl phosphate); the arsenates of zirconium and columbium do not precipitate together with the product. A bismuth arsenate extraction step followed

by two bismuth phosphate decontamination cycles gave 10^4 decontamination just as the normal bismuth phosphate method, but gave an over-all product yield of 97.4% while the parallel run of the normal method gave 89.5%. (2 men; B-1 priority)

Adsorption Process. Studies of titanium zeolite absorbent for decontamination after extraction by an amberlite cycle continued. Beta decontamination of ~110 was obtained with one run through a zeolite column. (5 men; B-2 priority)

Concentration. Current plans for Site W foresee removing of $BiPO_4$ process solutions from canyon after decontamination factor of 10^5 is achieved; then follows a bulk reduction from 3000 gallons to 8 gallons (in the 224 Building) by a combined $BiPO_4$ and LaF_3 cycle. It is hoped that ~ 250 g plutonium can be separated from 2000 g of lanthanum in 8 gallons without carrier by precipitation of plutonous peroxide. More experimental data have to be collected when more plutonium becomes available.

If uranous oxalate is used for plutonium precipitation in HCl solutions of $BiPO_4$, a concentration factor 80 can be obtained. Variables affecting the carrying power have been studied.

Studies on concentration by the Cb_2O_5 - LaF_3 method continued; especially the influence of acidity and alkalinity on this process and washing of the Cb_2O_5 precipitate were studied. The use of Ce^{+4} for dissolving LaF_3 and CeF_3 and for oxidizing of plutonium was studied further. The problem will in the future be investigated at X. Lower concentration of HF in the fluoride process for concentration reduces corrosion of stainless steel.

New carriers being studied for the use of concentrating plutonium and separating 93 from 94 are: thorium molybdate (it carries 94 but not 93); uranous hypophosphate (carries 93 and 94); lanthanum phosphate (carries 94).

Cupferron extraction of plutonium from lanthanum solutions and some details of the LaF_3 method for concentration have been studied. (4 men; A-1 priority)

Purification. Plutonous molybdate has a small solubility in the pH range 2 to 4, but is readily soluble in 2 N HCl or HNO_3 or H_2SO_4. These qualities make purification by repeated recrystallization possible.

It was shown, using uranium as a stand-in for plutonium, that ether extraction might be used to purify plutonium from lithium, sodium, potassium, magnesium, chromium, aluminum, phosphorus, and possibly from bismuth, silicon, and iron, but not from boron.

Plutonium oxide has a much smaller vapor pressure than U_3O_8. That is of importance for the application of gallium oxide method to analyze spectrographically for light impurities.

Plutonyl nitrate is reasonably stable in ether solution (2% reduction per hour).

The vapor pressure of plutonium bromide at 450°C is only slightly smaller than that of uranium bromide.

Studies of reaction of PuO_2 with HBr indicate that plutonium bromide sublimes at 600°C.

Preliminary observations are made on the specific activity and half-life of 94^{239}; the torsion balance used in these experiments has been recalibrated. (14 men; A-1 priority)

Metal Production. The efforts of the entire metal production group are being directed to metal production for density measurements. A single experiment on a three microgram piece indicates that a hydride is produced by heating plutonium metal to 210°C in an H_2 atmosphere.

The self-adsorption of alpha rays even in pieces as small as 1 microgram is ~70%; hence pieces of plutonium cannot be assayed by direct counting. (6 men; A-1 priority)

Instrument Group. (8 men; A-1 priority)

U^{233} Production of Thorium Carbonate for Use at Site X. (1 man; B-2 priority)

Wilma visited Helen in the afternoon, then Jean Zinn and Laura Fermi came by to see Helen and to deliver a book from the new Book Club. In the evening Helen and I went dinner-dancing with the Drehers and Baumbachs.

Today's newspaper reports that the Soviets are 55 miles past Kiev and have recovered one hundred villages.

Saturday, November 13, 1943

In a memo to Allison I pointed out that a large number of different names are being used to describe the processes which occur in the final stages of processing at Hanford, and I proposed that he and Greenewalt give these steps distinctive names. I mentioned that a number of us in the laboratory have been using the term "bulk reduction step" for the lanthanum fluoride cycle that will occur in Building 224, Hanford, and the term "concentration step" for the preparation of 99% plutonium via peroxide precipitation that will occur in Building 231.

Willard returned from the Alternative Processes Conference at Clinton. Also attending were Compton, Conant, C. M. Cooper, Doan, Greager, Greenewalt, W. C. Johnson, Kay, Nichols, A. V. Peterson, Pratt, and Whitaker. At this meeting Greenewalt, after hearing the presentations, stated the following conclusions which he believes to be representative of Franck's and Allison's views:

1. Drop work on Dry Fluoride Process as an extraction-decontamination process.
2. Drop work on the Lanthanum Fluoride Process, except insofar as it will be developed in concentration and crossover procedures.

3. Press work on adsorption as an extraction process because of fear of adverse effects of very high radiation levels on the extraction step of the Bismuth Phosphate Process.

4. Continue sodium uranyl acetate studies as a decontamination process, but on a laboratory scale, either at Clinton or Chicago.

These rather far-reaching conclusions are based on the need to emphasize the Bismuth Phosphate Process and the philosophy that the development of alternate processes should be confined to those differing fundamentally from one another. Cooper, although not mentioning it at the conference, feels that the Lanthanum Fluoride Process should continue to receive attention as a decontamination process, at a higher level of emphasis than the acetate process. It is evident that Greenewalt continues to fear the Lanthanum Fluoride Process because of its corrosion aspects and also reflects Franck's preference for the adsorption approach. I do not feel that these conclusions will have too much of an effect on our Section C-I research program.

"Chemical Research - Production and Extraction of Plutonium. Report for Period Ending November 11, 1943" (CN-1071) is being issued and covers the work of Beaton, Fields, La Chapelle, Lincoln, and Sheft on adsorption methods for the extraction and decontamination of plutonium. The use of titanium zeolite adsorbent to obtain additional decontamination after extraction by a citric acid-Amberlite IR-1 cycle has been further investigated. Titanium zeolites made from Sherwin-Williams TiO_2 and "Aratone" and from Krebs TiO_2 and vermiculite have been tested and calcining treatments for optimum selectivity for plutonium adsorption have been determined. At present, extensive studies are underway on the effect of briquetting pressure on the selective adsorption of plutonium by the calcined zeolite. Batch adsorption tests with Amberlite IR-1 and barium and lanthanum tracer solutions show that for Site W concentrations of barium and lanthanum, these fission products are highly adsorbed. Recent column tests, however, reveal lanthanum in tracer amounts is but slightly adsorbed.

The Technical Division Separation Process Section in Memo 144 reports that work during the week on the decontamination step of the Bismuth Phosphate Process has confirmed previous indications that yield losses in by-product precipitates can be eliminated by the use of very small amounts of HF during the oxidation step. In the concentration step, the use for metathesis of KOH in place of NaOH continues to show promise.

Wilma took a walk with Helen and then came back to our apartment for tea. Later John Gofman, who is here from Berkeley for the Met Lab meetings next week, went out to dinner with Helen and me.

Fifteen Japanese warships have been bombed in the Pacific. This morning's paper reports also that in Europe the Soviets have reached within 15 miles of Zhitomir, important Ukranian rail center.

Sunday, November 14, 1943

Helen and I saw a movie, "Dixie," with Bing Crosby and Dorothy Lamour at the Frolic Theater in the afternoon. Then Betty Seaborg, my first cousin from Ishpeming, Michigan (my birthplace), visited us; she has come to Chicago to attend nursing school. The three of us went to the Museum of Science and Industry and then had dinner at Isbell's.

Monday, November 15, 1943

Allison sent a memo to Greenewalt, quoting my proposal of Saturday about terms used to describe the processing activities to take place in Buildings 224 and 231 at Site W. He suggested that during Squires's next visit a little conference be held with Maloney and me in Allison's office to discuss this matter.

I attended the special meeting with C. A. Thomas on the metallurgy of plutonium at 1:30 p.m. in Room 209, Eckhart Hall. Others present were Baumbach, Chipman, Dempster, F. Foote, Foster, Franck, Gofman, Hogness, D. J. Hughes, Zay Jeffries, Johns, J. W. Kennedy, Lum, Kirk, Magel, Manning, Mooney, Mullikan, Potratz, Rollefson, Rosenfels, C. S. Smith, Spedding, W. W. Watson, Wilhelm, and Zachariasen. Kirk, Baumbach, and Zachariasen reviewed their current work with plutonium metal. Thomas made the observation that the work done in the past month has been very intense and encouraging. Dempster described progress of work with the mass spectrometer; Kennedy spoke on the analysis of plutonium at Site Y; Magel gave the results of vapor pressure measurements of plutonium oxide; Gofman spoke on the Berkeley study of metal-halide systems in order to find the oxidation-reduction potential of plutonium; and Thomas and Lum discussed the availability of and characteristics of rhenium metal. The meeting adjourned at 5:00 p.m.

I went to the evening seminar of the Chemistry Division at 7:45 in Room 251, Ryerson Laboratory, at which Coryell described the hot lab proposed for Site X. This will have sufficient shielding against gamma rays to make it possible to chemically process slugs from 30 days of operation of the Clinton pile at 500 kw after a cooling period of 10 days.

Berlin has announced that the Soviets have broken through the German line near Krivoi Rog, west of the Dnieper River.

Tuesday, November 16, 1943

An all-day Project Council Information Meeting met in Room 209, Eckhart Hall. The conference was divided into two sessions: Chemistry in the morning and Technology in the afternoon. Laboratories at Ames, Berkeley, Chicago, Clinton, and Site Y were represented. The Chemistry Information meeting opened at 9:30 a.m. Others present, in addition to me, were Allen, Allison, Boyd, Burton, Chipman, C. M. Cooper, Coryell,

Fermi, Foster, Franck, Fred, Gofman, Greager, Hilberry, Hogness, Hughes, Jeffries, Johns, W. C. Johnson, Kennedy, Magel, Miles, Mulliken, Potratz, Rollefson, C. S. Smith, Spedding, Stone, Sugarman, Sutton, Thomas, W. W. Watson, Wilhelm, Willard, and later Compton, Stearns and Wigner. Rollefson reported on spectrochemical analysis work at Berkeley that shows that as little as 0.001 microgram of light elements can be detected with the 3-meter grating spectrograph after concentration of the light elements from a uranium sample by removing the uranium through precipitation as the peroxide from acid solution. Gofman reported on decontamination work at Berkeley using Cb_2O_5 and on studies to eliminate the large amounts of $NaNO_3$ now used in the Sodium Uranyl Acetate Process; extraction and two decontamination cycles on the latter process gave good plutonium yield but not so good decontamination from fission products. Warren Johnson reported on the work of Perlman's Section I at Site X where emphasis has been on wet processes, especially on crossover from bismuth phosphate to sodium uranyl acetate. He also said that by this week 80 cans of thorium carbonate will be in the Clinton pile and should yield about 1 mg of U^{233}. Kennedy, reporting on purification work at Site Y, mentioned that microchemical work has been started, continuing Sheline's Berkeley work on the similarity of sodium uranyl acetate and sodium plutonyl acetate crystals. The respective indexes of refraction (Na D line) are 1.4993 and 1.5110. Sodium plutonyl acetate is about four times less soluble in water than its uranium counterpart. Sugarman, Burton, Potratz, and I discussed some recent developments in our sections. With regard to metal production work, I said that we have been concentrating our efforts on preparing powdered plutonium metal for examination by Zachariasen--so far we have reduced 400-500 micrograms without producing wholly suitable samples. We have obtained some solid samples (5-20 microgram size) which, by the microcapillary method, give a lower limit of 15.5 for the density of plutonium. I also mentioned Baumbach's preparation of plutonium hydride.

The 2:00 p.m. session of the Project Council Information Meeting was devoted to Technology. Others present were Allison, Bellas, Brugmann, Chipman, Compton, C. M. Cooper, Fermi, Foote, Foster, Franck, Greager, Hilberry, Greninger, Hogness, Howe, Jeffries, Johns, W. C. Johnson, K. G. Jones, G. E. Kidd, Leverett, R. D. McCrosky, Magel, Monk, Miles, Mulliken, Spedding, Sutton, C. S. Smith, Smyth, Stone, Vernon, W. W. Watson, Wilhelm, and later Hiskey, Kennedy and Thomas. In connection with Magel's discussion on small-scale metal production, I informed the group of Zachariasen's recent determination of plutonium oxide density to be 11.54 gm/cm^3; hence the relative densities are in the order of PuO_2 > UO_2 > ThO_2. Greager reviewed semiworks results at Clinton and indicated that uranium metal solution, extraction, and decontamination steps are satisfactory, with over-all plutonium yields of 80%. The concentration step is not satisfactory and is being actively studied. I asked if any of the troubles are attributable to poor carrying by lanthanum fluoride, to which Greager replied that the trouble is more probably due to leakage of the precipitate in filtration. Sutton indicated that his recent work has been largely directed toward Clinton problems, especially the lanthanum fluoride crossover cycle and concentration steps. I mentioned

that the use of zirconium in nitric acid solution to help dissolve $La(OH)_3$ from metathesized LaF_3 is under active study and will probably be a big help.

Thompson wrote a memo for me on the present status of extraction-decontamination work that summarizes the activities of his group as follows:

1. Process development. Installation of the dissolver in the West Stands will be completed today and dissolving of the Berkeley metal will be started immediately. Margolis, Schuler, Creek, Gaarder, and Halperin will work on this job. Thompson is ready to start work on any alternate process I wish to try. Cunningham has already started testing the reduction method that might use uranous ion (U^{+4}) as reducing agent after dissolving the neutron-bombarded uranium metal from Berkeley. Dean has been following the ether extraction work being carried out in the West Stands by Meyers of Boyd's section. George Creek has just about finished a uranous oxalate-sodium uranyl acetate extraction-decontamination run using Site W concentration of fission products and centrifugation throughout.

2. Scouting work. Following my suggestion that we might be able to detect a lower (than +4) oxidation state of plutonium by carrying with thorium molybdate, James has started some experiments using a Jones Reductor. An experiment is being completed by Kroner to compare decontamination in the Bismuth Phosphate Process when U^{+4}, Fe^{+2}, and oxalic acid are used separately for reduction. Schuler is testing my suggestion about not having dissolved all of the bismuth phosphate with nitric acid during oxidation in a bismuth phosphate cycle. As I had suggested, of the order of 5-10% of the bismuth phosphate is left undissolved. Hopkins has just finished some experiments on the use of uranous molybdate for carrying plutonium and separating it from neptunium. The microchemical experiments by Johnson and Halperin, comparing the Bismuth Phosphate Process through a decontamination cycle using (a) "W" concentrations and (b) "X" concentrations of fission products and plutonium, should be finished this week. Gaarder, after completing the tests of the effect of zirconium on carrying of plutonium by lanthanum fluoride, is now determining the maximum concentration of zirconium with regard to its effect on the oxidation step of the fluoride concentration process.

3. Concentration. Watt has just completed a run involving the Bismuth Phosphate Process through a crossover and metathesis step. He is now planning to make out a large chart showing all the concentration methods and ideas on this subject which we have had to date.

Helen had Wilma over for lunch and tea. In the evening I worked with her on the organization of my new "Table of Isotopes."

Today's reports indicate that U.S. flyers flew across the North Sea to Norway to strike at Germany's essential metal supply and to bomb a power station.

Wednesday, November 17, 1943

I attended a 2:00 p.m. progress meeting on the development of the Bismuth Phosphate Process.

In the evening at 7:45 p.m. I went to the meeting of the Extraction Group of my section in Room 209, Eckhart Hall. Among those present were Thompson, Gaarder, Creek, James, Kroner, Malm, and Schuler. The various types of chain-reacting piles, now in existence or contemplated, were tabulated for the benefit of the uninformed as follows:

Carbon-Uranium

1. Fermi pile at Argonne: about 1 kw — no cooling.
2. X-pile: 500 kw and hope to go to 1,500 kw — air-cooled.
3. W-pile: 250,000 kw — water-cooled.

D_2O-Uranium

1. Pilot plant at Argonne: 250 kw. It will be a heterogeneous pile and water-cooled.
2. W-pile: 600,000 kw — 40 to 100 tons of heavy water. It would use a slurry of a suspended compound of uranium.

It was announced that the Clinton pile has been operating at 500 kw for about a week and 5 tons of uranium metal containing one to two grams of plutonium have been discharged already and are now cooling. The extraction plant at Clinton will start working up the Argonne material which will arrive there soon. Thompson, James, Halperin, Gaarder, Kroner, and Malm are going to Site X to serve as trouble-shooters while the extraction plant is getting under way. After the Argonne metal has been extracted, the 5 tons from the Clinton pile will be "cool" enough to allow it to be run through the extraction plant.

A brief description of the extraction process at Clinton Laboratories was presented as follows: The dissolved pile uranium is brought to Cell 2, and H_2SO_4, H_3PO_4, and Bi^{+3} are added. Bismuth phosphate is precipitated and run through the centrifuge. The supernatant goes to the catch tank and is then piped to the neutralization room. The bismuth phosphate is washed into the dissolving tank and dissolved in 10 N HNO_3. The plutonium is then oxidized with $NaBiO_3$, the diluted solution pumped to the precipitating tank again, and a by-product precipitation of bismuth phosphate is made. After centrifugation the by-product is slurried to the catch tank for dissolving before disposal.

The product solution goes to Cell 3. The plutonium is reduced; bismuth phosphate is precipitated, centrifuged, and sent to a catch tank. It is then redissolved, the plutonium reoxidized, and the by-product is precipitated, as in Cell 2. The product solution goes to Cell 4; the plutonium is reduced, and lanthanum fluoride precipitated. This precipitate is slurried to Room D.

In Room D, two batches are combined in a small 26-inch centrifuge; then, after initial processing by fluoride oxidation-reduction cycles, four of these double batches are combined in a smaller 12-inch centrifuge. The material is then taken to Perlman's group for isolation of the carrier-free plutonium.

The use of one catch tank for two purposes is a poor arrangement, especially if scavengers are used as they may not be soluble and the precipitate might "hold up" on the walls. At Site W, as more equipment becomes available, the same equipment is not to be used for more than one type of operation. Hanford will have four canyons, one for each of three piles, and one for a standby. Sampling is to be made by removing a sample from the top of a cell through use of an air lift.

Extraction and decontamination problems were then reviewed. James reported on a neptunium-plutonium separation procedure using thorium molybdate as carrier. There is good separation with 99% of the plutonium carried and only 2% of the neptunium, but the method is critical as to pH. I mentioned the possibility of a valence state for plutonium lower than +4; one might find a carrier for Pu^{+4} but not for Pu^{+3}--perhaps thorium molybdate--it is also thought that neptunium has a +3 state. In a discussion of reducing agents in the crossover step, I mentioned that arsenite and H_2O_2 have been tried and that some work is being done on $H_2C_2O_4$ and Fe^{+2}. Thompson said that some of Sutton's men are using $H_2C_2O_4$ successfully. It was mentioned that Watt has run through the concentration procedure successfully with 67% recovery with most of the loss occurring in the H_2O_2 treatment.

Helen worked at the Met Lab library on the "Table of Isotopes" in the morning. In the afternoon she went shopping with Wilma and then visited with her while I was attending the Extraction Group meeting.

The Project Council had its Policy Meeting at 9:30 this morning in Room 209, Eckhart Hall, attended by Allison, Chipman, Compton, C. M. Cooper, Doan, Fermi, Franck, Greager, Hilberry, Hogness, Hughes, Jeffries, Warren Johnson, Miles, Captain Murphy, Smyth, Spedding, Stearns, Thomas, W. W. Watson, Whitaker, and Wigner.

The meeting was opened by consideration of the Chemistry Division program as described in the Monthly Report of November 12 (MUC-JF-110) Following a lengthy and very detailed discussion of the immediate usefulness to the Project of several studies noted on the program, Smyth pointed out the impossibility of arriving at decisions on such matters in Council. He feels that the responsibility for making such decisions should in any event lie between the individual Division Director and the Laboratory Director. Spedding believes that the Laboratory Director should come to agreement on program matters with the Division Director and bring up for discussion in Council only those matters on which advice is needed. Hilberry emphasized the impossibility for fair consideration of program matters in Council and advised that decisions should not be made on such a superficial basis; he feels we must depend on the Division Director to keep his program properly oriented with broad policy. Smyth stated that the Council members should, of course, be kept informed on such matters as program but felt that this could be best accomplished by circulation of a good summary. Council members could then take up matters in question directly with the

Laboratory Director who would then seek advice from Council in such matters as seemed necessary. Following general agreement with this proposal Compton indicated that he would like future meetings to proceed in keeping with these new proposals.

In a discussion of the Chemistry programs that followed, it was brought out that the need for microchemists in future studies is not entirely clear--probably after a few months some people can be released to other phases of chemistry. The present St. Louis bombardment is expected to be the last cyclotron bombardment for the production of Pu^{239}--Whitaker indicated 4 milligrams of plutonium will be available from Clinton early in December as a result of processing Argonne material.

Compton gave his "State of the Nation" summation. He stated that the successful operation (better than expected) of the Clinton pile represents another milestone. The improved international military situation emphasizes the need to concentrate all possible effort toward bringing Hanford into production at the earliest possible date. Because of the national scientific manpower shortage, we must re-evaluate our program and transfer all possible effort from alternatives which cannot be useful within a year to those which have a high probability of such usefulness. The effect of this new emphasis on current programs is: (1) The P-9 project. Build the heavy water heterogeneous pile as a radiation tool, but discontinue work toward P-9 production units as they cannot offer a real alternative to present Hanford plans. (2) Dry fluoride work. A process adaptable to Hanford needs cannot be developed within the year. Hence, the work will have to be dropped, although use for purification and other purposes will be considered. (3) Uranium-233 studies. Without a P-9 power plant it is not possible to make enough U^{233} to be useful; therefore, this work should be discontinued for the present.

Thursday, November 18, 1943

At the 8:30 a.m. Section Chiefs' meeting in Franck's office, Hughes, Potratz, Franck, Sugarman, Burton, Hiskey, Manning, Allen, and I were informed by Franck that at yesterday's policy meeting the Project Council decided to abandon U^{233} work and dry fluoride work for extraction and decontamination. The P-9 program and adsorption and scouting work are to come up for discussion later.

I attended a patent meeting on separation processes at 10 a.m. in Room 209, Eckhart Hall. Others present from the Met Lab were Willard, Manning, Boyd, Sutton and J. H. Peterson. Representing the OSRD Washington Office and Chicago Patent Groups were York, Holton, Richtmyer, F. W. Neill, and Moulton. The purpose of the meeting was to review for the patent people all the potential extraction and decontamination methods as well as the concentration processes. I listed 17 possible extraction and decontamination methods as follows:

11/18/43 (cont.)

1. Bismuth Phosphate Process
2. Sodium Uranyl Acetate Process
3. Wet Fluoride Process
4. Adsorption Process
5. Dry Fluoride Process
6. Ether Extraction Process
7. Adsorption-Sodium Uranyl Acetate Combination Process
8. Adsorption-Bismuth Phosphate Combination Process
9. Adsorption-Wet Fluoride Combination Process
10. Ether Extraction-Sodium Uranyl Acetate Combination Process
11. Ether Extraction-Bismuth Phosphate Combination Process
12. Ether Extraction-Wet Fluoride Combination Process
13. Bismuth Phosphate-Sodium Uranyl Acetate Combination Process
14. Uranous Oxalate-Sodium Uranyl Acetate Combination Process
15. Columbium Pentoxide-Sodium Uranyl Acetate Combination Process
16. Bismuth Phosphate-Wet Fluoride Combination Process
17. Bismuth Arsenate-Bismuth Phosphate Combination Process

I also described the concentration procedure that we now prefer for use at Site W and a number of alternate concentration procedures that may be more specifically applicable to Site X.

In the pre-reduction of product after the metal solution step formic acid is now preferred, but uranous salt (U^{+4}) is also good and may become first choice.

In the bismuth phosphate-wet fluoride combination, sodium bismuthate is the preferred oxidizer in the first step. Then ferrous ion or uranous ion will be used for reduction. After this the change is made to lanthanum fluoride where sodium bismuthate or dichromate will be used as an oxidizer and hydrogen peroxide as the reducer. Oxalic acid may be a better reducer than hydrogen peroxide in the wet fluoride cycles. Arsenite as a reducing agent is objectionable because of its corrosiveness.

The following concentration process seems workable in conjunction with the bismuth phosphate extraction-decontamination process. Oxidize, take out a bismuth phosphate by-product precipitate, lanthanum fluoride by-product precipitate, reduce, throw down a lanthanum fluoride product precipitate and then proceed with fluoride cycles. At Site W the concentration of plutonium will be such that plutonium peroxide may be precipitated without carrier after a single lanthanum fluoride precipitation.

A number of alternate concentration processes, especially applicable to Site X, include the following:

1. Oxidize, take out a $BiPO_4$ by-product precipitate, reduce, take a LaF_3 product precipitate, and then go to wet fluoride cycles. This is the procedure now called for on the X flowsheet.

2. Oxidize, take out a $BiPO_4$ by-product precipitate, reduce, take a $U(C_2O_4)_2$ product precipitate, and then go to wet fluoride cycles.

3. Oxidize, take out a $BiPO_4$ by-product precipitate, reduce, take a $U(C_2O_4)_2$ product precipitate, and then go to uranous oxalate cycles.

4. Dissolve the $BiPO_4$ product precipitate in nitric acid, take a LaF_3 product precipitate, and then go to wet fluoride cycles.

5. Dissolve the $BiPO_4$ product precipitate in hydrochloric acid, take a LaF_3 product precipitate, and then go to wet fluoride cycles.

6. Dissolve the $BiPO_4$ product precipitate in hydrochloric acid, take a $U(C_2O_4)_2$ product precipitate, and then go to wet fluoride cycles.

7. Dissolve the $BiPO_4$ product precipitate in hydrochloric acid, take a $U(C_2O_4)_2$ product precipitate, and then go to $U(C_2O_4)_2$ cycles.

8. Oxidize, take a $BiPO_4$ by-product precipitate, reduce, neutralize, take a lanthanum phosphate-hydroxide product precipitate, and then go to wet fluoride cycles.

In the wet fluoride cycles there are a variety of ways of dissolving the precipitates. In later stages of the above processes it is possible to go to the sodium uranyl acetate procedure.

At W concentrations of product, as indicated above, direct precipitation of plutonium as the peroxide or oxalate should be possible after a single bulk reduction step.

I also attended the latter part of a 2:00 p.m. meeting with Harrison of M.I.T. to discuss spectroscopy. Others present were Dempster, Franck, Fred, Hogness, Kennedy, Manning, Nachtrieb, Orlemann, Potratz, Rollefson, and W. W. Watson. The purpose of the conference was to decide whether it would be feasible to undertake vacuum spectrograph work for the determination of oxygen and fluorine in plutonium metal. Oxygen and fluorine lines appear in the ultraviolet, and therefore vacuum spectroscopy is required. Although Harrison regards the mass spectrograph method as more promising, he is willing to undertake development of the vacuum spectrograph as a possible alternate method. He could devote two days a week to the problem at M.I.T., but it will take several months to get any answers.

As usual, there was an evening meeting of Section C-I Research Assistants at 6:45 p.m. in Room 209, Eckhart Hall.

The evening meeting of the Council of my section in my office at 7:45 was attended by Beaton, Cunningham, Davidson, Dreher, Ghiorso, Kirk, Manning, Orlemann, Thompson, and Willard. I discussed the forthcoming directive from Conant that, in view of manpower shortage, attention be concentrated on "mainline" work; I expressed the view that since plans for production of U^{233} have gone this far, it would be very desirable to do sufficient additional work to obtain a small quantity of pure U^{233} for physical measurements. A Christmas party is planned for the afternoon of December 24, preferably just for our section.

In personnel matters, Dorothy Gottlieb, the new secretary, is in room 108 in Chemistry Annex. We now have four secretaries, Edrey E. Smith, Irma N. Saxton, Elsy W. Anderson, and Miss Gottlieb. The need for someone to specialize in quartz work was discussed, and it was decided that Zvolner or Beatrice Foreman, the laboratory technician in Baumbach's group, be asked to learn quartz blowing techniques. In the realm of equipment, the necessary materials for four quartz balances have been ordered. It will probably be desirable to increase the number to five, as one additional balance is needed here and two each are planned for Clinton and Site W. In a discussion of research activities, I mentioned my recent memo about standardizing on names for bulk reduction and concentration cycles. Davidson's suggestion of "concentration" and "isolation" was received with favor; we shall use this nomenclature, and I'll press for its general adoption. I said we should decide more or less arbitrarily on a definite value for the geometry factor of alpha counters since the work of Ghiorso and Cunningham indicated larger values than we have been using. A factor of 49% was agreed upon; this results in a half-life of 22,000 years for Pu^{239} and a value of 149,000 disintegrations per minute per microgram. I announced my forthcoming trip to Washington, D. C. next Sunday and Monday to discuss the NBS findings of variation in alpha activity of different uranium samples that might be caused by the presence of natural plutonium. It was agreed to retest the possible use of uranyl ion to dissolve lanthanum fluoride, following a suggestion I made to Willard and Thompson. Recent carrying experiments at Berkeley suggesting a possible lower oxidation state (+3) of plutonium formed by reduction with hydroxylamine were discussed. Recent x-ray studies by Zachariasen furnish strong evidence for the existence of a +3 Pu (PuF_3) in the dry state. Cunningham has found good evidence of a +5 state for plutonium. It was agreed that knowledge of these valence states is of great practical importance and that this work should have high priority. It was mentioned that Zachariasen's evidence of a +4 state (PuO_2) is almost incontrovertible and that the +6 state has been practically proven by recent work at Berkeley (isomorphism of sodium uranyl acetate and sodium plutonyl acetate).

Gordon Johnson completed the preparation of a plutonium-containing bismuth phosphate precipitate to be used by Mrs. Mooney in an x-ray study of its crystal structure. The plutonium content of the precipitate is 2.4%.

Helen and I had dinner at the Baumbachs' (4910 Blackstone) before the Section C-I Council meeting. Helen stayed to visit and I picked her up after the meeting.

Friday, November 19, 1943

General Groves is in Chicago.

Sutton's Separation Processes Section is moving to Clinton Laboratories and packing began yesterday. Today, the transfer of about two dozen persons started--all of the personnel will be gone within a week. Sutton hopes to begin some laboratory work at Clinton by the 24th.

The Germans report that Berlin was bombed by the British forces last night. The paper today also states that U.S. air forces struck blows at Japanese bases in the North Solomons.

Saturday, November 20, 1943

Arthur LeFeuvre began working as a technician for Ghiorso today.

The chemical engineers and I held a conference in my office at 10:00 a.m. to discuss the immediate work to be done by the Kircher's semiworks group (in Bellas' section) using equipment installed in the West Stands. Present were Bellas, Dreher, Kircher, Maloney, Thompson, and Willard. It was agreed that this group will carry out tasks that cannot be performed as efficiently by the semiworks groups at Site X. The following program was organized:

1. Special extractions, such as the one now in progress for the purpose of obtaining 93^{237} from the recent shipment of Berkeley uranium metal and uranium metal from the Argonne pile (to determine pile yield of 93^{237}).

2. Processing of sufficient volumes of solution to allow a large-scale investigation of concentration procedures and determination of large decontamination factors.

3. Studies of the Wet Fluoride Method as it applies to bulk reduction and concentration at Site X or as a replacement for the Bismuth Phosphate Process.

4. Studies of alternate concentration methods which might be of immediate use at Site X.

5. Tests of a modified Bismuth Phosphate Process using bismuth arsenate in the extraction step.

6. Tests of the $U(C_2O_4)_2$ - $NaUO_2Ac_3$ process.

The semiworks group will work as a service group to us, collaborating closely through Thompson and Dreher with Willard and me.

The Separation Processes Section (under Sutton) made its final section report (Memo No. 146) to the Met Lab.

Thompson, Halperin, Kroner, James, Gaarder, and Malm left Chicago at 11:30 p.m. via Penn R.R. train for an extended trip to Site X in order to be available if needed during the initial operation of the Clinton

extraction plant for extracting plutonium from uranium metal from the Argonne and Clinton piles. They are scheduled to return about December 15.

The Germans have struck a counter blow in the Ukraine and recaptured Zhitomir.

Sunday, November 21, 1943

This morning's headline in the Chicago Sun indicates that the Soviets have crushed the tank thrust made by the Germans west of Kiev.

At 3:25 p.m. I left Chicago by Penn R.R. train for Washington, D.C.

Monday, November 22, 1943

I arrived in Washington in the morning and visited Dr. Curtiss and Dr. Levinos at the National Bureau of Standards in order to discuss their study of an alpha-emitting radioactive impurity that they found associated with certain samples of uranium. They believe this impurity, which amounts to 1.3% of the total activity, to be plutonium. I checked the chemical procedures used to eliminate the impurity, and it seems clear the alpha activity cannot be caused by any isotope of plutonium. The weakest part in their research is that they have been unable to isolate the radioactive impurity itself. I was given a sample of the impurity-containing U_3O_8 to take back to Chicago with me for testing.

In the evening I left for Chicago again by Penn R.R. train.

The evening seminar of the Chemistry Division started at 7:45 in Room 251, Ryerson Laboratory, which, of course, I missed.

Tuesday, November 23, 1943

I arrived in Chicago at 9:00 a.m.

At the lab I was greeted by the announcement that Zvolner's experiments the last few days seem to indicate that he is producing metallic plutonium by electrolysis of aqueous solution. This seems quite unlikely, but he insists that the product reacts as if it were the metal.

In a wire to Albaugh I said that I have heard he has obtained a leave of absence and that I hope he will join us as soon as humanly possible.

I recorded the following conception in my notebook concerning the search for new transuranium elements:

"In addition to the bombardment of 94^{239} with high energy deuterons we intend to bombard it with high energy alpha-particles (for example, 30 Mev) and high energy protons (for example, 10 to 14 Mev), when sufficient amounts are available, in order to investigate the products formed, particularly those in the transuranium region. We may find, for example, the isotope 95^{242} (and its decay products) from the reaction $94^{239}(\alpha,p)95^{242}$ and/or the isotope 95^{241} (and its decay products) from the reaction $94^{239}(\alpha,np)95^{241}$. We may find, for example, the isotope 96^{242} (and its decay products) from the reaction $94^{239}(\alpha,n)96^{242}$ and/or the isotope 96^{241} (and its decay products) from the reaction $94^{239}(\alpha,2n)95^{241}$. With protons we may find, for example, 95^{239} (and its decay products) from the reaction $94^{239}(p,n)95^{239}$ and/or 95^{240} (and its decay products) from the reaction $94^{239}(p,\gamma)95^{240}$."

I sent a memo to Whitaker at Clinton Laboratories about the proposed curtailment of the U^{233} program. I gave the following information which may warrant consideration in deciding upon the rate of deceleration of the work: (1) There is valid reason to speculate that U^{233} may be the product for which the Germans are striving in connection with their large scale production of heavy water. Thus, it is important that we know its fission cross section, spontaneous fission rate, and neutron yield per fission. (2) Teller, who feels strongly that enough U^{233} should be produced and separated to make the basic physical measurements at Site Y, has stated that uranium containing only 10% U^{233} would be adequate for these measurements. With this tolerance for natural uranium it should be possible to extract and separate uranium directly (rather than via its protactinium parent) from the thorium being irradiated in the Clinton pile, a much easier task which would involve only 12 man-months. (3) To stop the U^{233} work before attaining the goal stated above would have a very bad effect on the morale of many of the chemists on the project since it would appear to them as an unreasonable reversal of policy.

Stan Thompson has summarized the work completed last spring by James and La Chapelle on the possibility of an oxidation state of plutonium lower than +4. The work has not been written up before because the results were inconclusive, but Thompson feels the information might be of interest to Gofman and others in Berkeley who are starting to look for such a lower oxidation state. Experiments described by Thompson are (1) reduction by hypophosphite, (2) reduction with hydrazine sulfate, (3) reduction in a Jones Reductor, (4) reduction by Cr^{+2} to stabilize any reduced plutonium, and (6) dissolving of bombarded uranium metal in HCl. After treatment with these reagents, plutonium was generally carried less completely by bismuth phosphate than it is in their absence when it is presumed to be in the +4 oxidation state. This may be evidence, admittedly inconclusive, that the plutonium may have been reduced to a lower, perhaps +3, oxidation state.

I held the usual Thursday meeting of the Council of my section in my office at 7:45 this evening (instead of Thursday) with Davidson, Dreher, Ghiorso, Kirk, Manning, Orlemann, and Willard. I reported on my visit to the National Bureau of Standards in Washington, D. C. In regard to working space, I agreed to consult with Hogness on the possibility of

getting some space for Ghiorso's group in the former storeroom (room 19), made available as a result of the move to the Chemistry Annex. Maloney has asked us to check the effect of various slug coating material (zinc, tin, aluminum, silicon, etc.) on the extraction and decontamination of plutonium, and Dreher will assign a man to this work. Recent work by Katz continues to indicate that plutonium and uranium differ considerably in their behavior during HBr treatment. Bradshaw has constructed an apparatus for two-stage volatilization studies during treatment with $HBr-Br_2$ mixtures. Plutonium can be volatilized from a hot filament carrying the oxide; 60% of the counts can be collected on the first cold finger, but only 1% of this first-collected activity is collected on a second cold finger. We discussed Zvolner's claim that he may have produced plutonium metal by electrolysis of aqueous solution. About 1/2 microgram of plutonium, as the sulfate, was electrolyzed in a solution of rather high acidity. A dark deposit was obtained on the cathode which, when counted, indicates a recovery of about one-third of the plutonium. Karle is trying to reduce UCl_3 to metallic uranium by using atomic hydrogen.

The U.S. has invaded the third Gilbert Island. Reports of the invasion of two other Gilberts arrived during my trip to Washington.

Wednesday, November 24, 1943

I received a summary from Davidson about the present state of work of his volatility group and related efforts. Fluoride experiments are being conducted by Heath (behavior of plutonium fluoride in CaF_2 tubes) and Florin (use of F_2 activated in a discharge of low pressure). Davidson suggests that the most important experiment to do with larger quantities of plutonium when available are the classic long-tube experiments (fluorination in CaF_2 tubes), probably at Oak Ridge. Bromide research is being done by Katz (effect of HBr on tracer quantities of plutonium) and Bradshaw (attempts to revolatilize condensate from a heated plutonous oxide-$HBr-Br_2$ mixture). Davidson thinks that both these experiments will be worth doing at the 10-microgram level, as well as the experiments of Smith and Cunningham on measuring the vapor pressures of halides by the use of plutonium metal, hydriding, powdering, and bromination. He feels that, at present, adequate manpower is being devoted to fluoride research, but more manpower might be used profitably on research on plutonium bromides and on plutonium iodide.

Willard received a letter from Milton Ader, a graduate assistant in chemistry at Oberlin College, expressing interest in a job at the Met Lab. He said he heard about our interest in hiring chemists from Professor Harry N. Holmes of the Severence Chemical Laboratory and noted that he will be in draft status 2B until February 3, 1944. Willard replied immediately, enclosing application blanks and suggesting he seek the personal reaction of Horace Hopkins, formerly of Orberlin, who has recently joined our staff.

Arthur and Betty Compton have moved to Oak Ridge because they feel it is their duty to help keep up morale there. Nevertheless, they will

maintain their present home here at 5637 South Woodlawn. For security's sake, Compton goes by the name of "Holly" in Oak Ridge. The Comptons' elder son, Arthur, is in the Army and their 15-year-old son, John, will be attending the Asheville School for Boys in Asheville, North Carolina. Compton will continue to shuttle back and forth between here and Oak Ridge because of his continuing duties here in Chicago.

In a memo to Compton at Clinton Laboratories Allison discussed a request by Potratz for six milligrams of plutonium for development of analytical techniques by chemical methods; he suggested that an estimate should be made of the needs of the Met Lab as a whole. Allison wondered if Compton has appointed someone as a "czar" of plutonium to act as a central point to whom requests can go.

Luther B. Arnold, who received his Ph.D. in organic chemistry from Harvard in 1933 and has worked for du Pont ever since, is now serving as an Assistant Director in the Chemistry Division to coordinate procurement of pure chemicals for the purification and metal production program. Today I attended a meeting that Arnold called in his office (Room 110, Chem Annex) to discuss the procurement of high purity reagents, refractories, and containers. Others present were Davidson, Fred, Kirk, Manning, Orlemann, Potratz, and Willard. First priority is being assigned to the procurement of pure concentrated H_2SO_4, concentrated HNO_3, 30% H_2O_2, and copper for electrodes. Refractories are considered of the utmost importance, but the status of the work with refractories is such that no list of desired materials can be drawn up. It was concluded that platinum ware, now on order and already available, will be used in meeting immediate needs for containers.

I gave a talk to members of the Technical Division. My subject was "Artificial Radioactivity, Tracer Techniques, and Methods of Separation."

The Gilbert Islands are falling to U.S. troops, and the first American offensive in the central Pacific appears to be successful.

Thursday, November 25, 1943

Willard requested that Wayne Johnson take the necessary steps to hire Mrs. Isabella L. Karle, the wife of Jerome Karle, for our section.

I ran into Leo Szilard today, and I mentioned to him my concern that the capture of neutrons by Pu^{239} to form Pu^{240} might lead to problems in the design of a plutonium weapon if the capture cross section is appreciable and if Pu^{240} decays by spontaneous fission at an appreciable rate. He expressed doubt that this constitutes a problem.

Art Jaffey is attempting to make a determination of the half-life of 94^{238} by measuring its rate of decay; this will be difficult because of its long half-life (estimated to be of the order of 50 years with a probable error of a factor of 2). He has prepared two samples of 94^{238}

(mounted on platinum) with an intensity of about 800 alpha-particle counts per minute each (from uranium bombarded with deuterons in the Berkeley 60-inch cyclotron) and will make the measurements on one of the standard "inside" alpha particle counters. In order to make compensation for slight changes in sensitivity, measurements will be made with two similar samples of 94^{239} whose activity, of course, will remain essentially constant over the several years' period of the proposed measurements.

Katzin wrote to Stoughton telling him that 250 cans of thorium carbonate should go out for welding tomorrow. He asked who is working on ether extraction and what territory of investigation he is covering, so we will know what to work on here. Katzin mentioned that I had agreed he should spend his time in this general area of investigation.

Thompson has recorded for me the present thoughts of his group on new ideas on the following subjects:

1. The use of new carriers for extraction, decontamination, and concentration, viz., uranous oxalate, bismuth arsenate, cerous fluoride, thorium molybdate, uranous hypophosphate.

2. Improvements in the Bismuth Phosphate Process and some alternate processes: (a) use of ceric ion for coupling cycles in the Wet Fluoride Process, (b) modification of the oxidation step in the Phosphate Process, (c) possible improvements in decontamination in the Phosphate Process, (d) use of complexing agents for dissolving bismuth phosphate, (e) effect of high concentrations of columbium from pile metals, (f) use of other reducing agents, (g) use of hydroxylamine as a complexing agent for uranium in the extraction step, (h) solvent extraction methods, (i) use of uranous iodate as a carrier, (j) use of ammonium uranyl phosphate as a carrier, (k) uranous oxalate - sodium uranyl acetate combination method, (l) use of uranous pyrophosphate and thorium pyrophosphate as carriers, (m) uranous phosphate and uranous arsenate as carriers, (n) bismuth antimonate and bismuth pyroantimonate as carriers, (o) bismuth oxalate as carrier, (p) zirconium phosphate as carrier.

3. Other new ideas on neptunium and plutonium separation and the investigation of a possible lower oxidation state of plutonium.

The evening meeting of Section C-I Research Assistants was held at 6:45 in Room 209, Eckhart Hall.

The German counterdrive on the Zhitomir front has been halted by the Soviet army.

Friday, November 26, 1943

At 9:00 a.m. Manning and I went to a personnel conference in Hogness' office about rating procedures and rules for determining salaries. Stearns, Wayne Johnson, Franck, and Hogness were there, along with Burton, Hiskey, Potratz, and Sugarman.

Compton sent Allison a statement of the present objectives of the Met Lab, based on recent discussions in the Project Council and a reevaluation in the light of recent developments. He said there are two major objectives of the Laboratory: (1) To make the Hanford water-cooled pile and its bismuth phosphate separation plant work as well as is possible at the earliest possible moment. (2) To join with Los Alamos and the associated projects in developing the methods for the necessary purification of plutonium and its reduction to metal. Compton then broke these down in terms of Divisional operations, stating that the objectives of the Chemistry Division will be to:

1. Finish studies of concentration methods necessary for Clinton Laboratories to develop complete separation processes for both Clinton and Hanford materials.

2. Do all useful studies on the fundamental chemistry of the wet B (fluoride) and D (phosphate) processes so that the necessary basic information will be available to assist in interpreting difficulties as they arise at Clinton and Hanford.

3. Furnish all required information on the behavior of materials and processes under intense radiation.

4. Determine properties of the by-products and their behavior in the processes.

5. Develop methods of purification of the product and of its reduction to metal in collaboration with Los Alamos and the associated contracts.

6. Develop analytical procedures for the purification program and carry out necessary analyses.

With respect to the objectives of the Nuclear Physics Division, the memorandum stated the following:

1. Expedite in every way possible the completion of the du Pont plans for the water-cooled pile. We want an all-out effort to check and complete their plans with a minimum of change from the present design. A plant operable somewhat below rated capacity but completed in time to be effective can be of value; one operable at rated capacity but which cannot be completed soon enough is useless.

2. Carry through theoretical and experimental investigations which can throw light on the fundamental behavior of the chain reaction and, in particular, of the water-cooled lattice so that as much fundamental information as possible will be available at the Hanford set-up for interpretation of any basic difficulties which may arise. This includes completion of the Argonne P-9 high level unit and its use as a source for high neutron and radiation density investigations.

3. Within the limits of time and facilities dictated by the above program, carry out investigations which may be requested by Los Alamos in connection with their experimental program.

4. In addition to the P-9 high level unit at Argonne, the two exponential experiments designed to be used there should be completed. The remainder of the P-9 program should be closed out as of December 31, 1943.

In a memo to Franck, Allison repeated the objectives of Compton's memorandum, but added a comment that for Section C-I, the U^{233} production and recovery should be severely curtailed and that the present charge of thorium carbonate in the Clinton pile should be left there until the U^{233} that forms can be taken out as element 92 without being merely a small fraction of the material already present as uranium. He questions my contention that as many as three men will be needed during the four months (while the U^{233} is growing) to devise methods for separating uranium from thorium.

Allison expressed the view in a letter to General Groves that much more rapid progress in investigating enemy installations in Europe could be made if a limited number of Met Lab personnel were allowed to have some inkling of what Army Intelligence has discovered there. He suggested that, if we were allowed to learn what was found by the party that raided the Rjukan Hydroelectric Plant (heavy water) in Norway, or what information Professor Bohr has concerning enemy activities in this field, we could draw conclusions which would not be apparent except to experts in the field of our project.

I received a copy of Stoughton's November 24 memo to Warren Johnson at Clinton that gives a proposed program for finishing the U-233 work with a minimum number of man-days. Stoughton proposes a two-step procedure whereby (1) some of the thorium carbonate will be irradiated only long enough to produce 10-100 micrograms of Pa^{233} that will be isolated through Pa^{233} separation, thereby reducing the radiation hazard by a factor of 100. This will provide sufficient material for specific activity measurements. (2) Irradiate another quantity of carbonate to produce one or more mg Pa^{233} that will be allowed to decay for about 2 half-lives; the resulting U^{233} is then to be separated, together with the natural uranium impurities, for physical measurements.

The transfer of Sutton's Separation Processes Section to Clinton Labs was completed today.

The Soviets have started a new drive and gained 28 miles in White Russia, according to today's newspapers.

Saturday, November 27, 1943

Several weeks ago Jaffey and Kohman interviewed Martin H. Studier for a job with Ghiorso, inasmuch as Studier has considerable electronics training with the Army Signal Corps here at the University. When I learned that Studier has a bachelor's degree from Luther College and is within about six months' work of getting a Ph.D. in chemistry, I decided he might be more useful working with Thompson or Cunningham. Strangely enough, he dropped out of sight before all the Army red tape could be processed and so spent the last six weeks in basic training at Miami Beach until we could rescue him. Today he started working for us. He showed up in an Army uniform. This attire is so out of place that many questions are being asked.

Cunningham went through a plutonium separation procedure on a natural uranium sample purported by Curtiss of the National Bureau of Standards to contain a high concentration of an alpha impurity (1.3% of the total alpha activity), believed to be an isotope of plutonium. The results indicate there is no plutonium in the sample.

On my behalf, Compton wrote to Oppenheimer at Site Y, attention Kennedy, asking for a value for the range of alpha particles from U^{235}, which is needed in connection with the preparation of material for the Project Handbook.

Sunday, November 28, 1943

The front page of this morning's Chicago Sun contains items of varying importance. The top headline reads "Third of Berlin Wrecked--'Colossal' Blow to Hit Italy Nazis." In contrast is news of labor discontent both in the U.S. and Great Britain, and the upset of Notre Dame's Irish by the Great Lakes Naval Training Station football team, 19-14, in the last 35 seconds.

Monday, November 29, 1943

I sent Franck a description of the present status of the U^{233} program at Site X, pointing out that we have placed 120 cans of thorium carbonate in the pile, that 60 have been withdrawn for experimental purposes after 4 kw-days of pile operation per average ton of uranium, and that 60 cans still remain in the pile and have received about 120 kw-days of irradiation per ton of uranium. I proposed that these 60 cans, plus an additional 38 cans on hand at Site X, be left in the pile for a few months after it starts up again, thereby producing about 1 ppm of U^{233} to be recovered by the direct method and not via the Pa^{233} intermediate. I suggested that the three men (Stoughton, a research associate, and a research assistant) be asked to work on the separation procedure as their first objective during the time the carbonate is being irradiated and then if they have any time left over, they might direct their efforts to other, mainline, work.

Ghiorso described for English at Clinton Labs the current status of counting instrument developments.

Stearns received a letter from Neal Thurman of du Pont in Wilmington indicating the status of the proposed transfer of John Willard and Paul Kirk to the Hanford Engineer Works organization. Kirk would leave about April 1, 1944, and Willard around June 1, 1944.

On November 4 Latimer wrote me that the University is engaged in a discussion with the U. S. Government (represented by Vannevar Bush, the OSRD Director) over its alleged rights to patents taken out by Wahl, Kennedy, Segrè, and me. Bush contends that the University has no right to the patents and is insisting that the University relinquish (without

compensation) all claims which it may think it has. Latimer asked for my opinion. Today I responded, saying in part: "As you know, there are two basic inventions involved. The first one is in the names of Seaborg, Wahl, and Kennedy, and is essentially a chemical patent, although very broad claims are contemplated on the basis of this chemical work. The early, basic work on this was all done in the Chemistry Department, except for one or two short cyclotron bombardments, without the benefit of any government support. As I understand it, it is for the purpose of including certain very broad claims, which are at present out of the case, that this application must be signed by us again. I haven't heard anything about this for quite some time.

"The second case, based on the somewhat later physical measurements, is in the names of Segrè, Seaborg, and Kennedy, and also includes some broad claims. So far as I know, the basic work for this was done without the benefit of government aid so far as the Physics Department is concerned. This of course is also true with respect to the Chemistry Department.

"As I said above, I favor the re-establishment of the University's rights in this matter if it can be done correctly, as defined by your judgment. Of course, I have not the slightest wish to personally benefit financially from this."

The evening seminar of the Chemistry Division met at 7:45 in Room 251, Ryerson Laboratory. My section was in charge of the program. Manning talked on the Site W concentration flowsheet, Hindman described the preparation of pure plutonium compounds, and Davidson spoke on the volatility of the halides.

The Soviets have won the key rail town of Korosten on the road to Minsk, 45 miles north of Zhitomir.

Tuesday, November 30, 1943

Our most recent employee is Nison N. Hellman, who received a Ph.D. from the University of Wisconsin this year. Hellman will work with Baumbach as an Associate Chemist at $3300 per year.

Today, Beaton and his group (Fields, La Chapelle, Lincoln, and Sheft) moved to the West Stands.

I had an exchange of wires with Gofman concerning the number of the most recent report on James's work on the oxalate method. I informed him the report number is CN-1041 but that more recent work at Site X is being written up by James.

The Metallurgical Laboratory Progress Report (MUC-JF-113) for November 1943 is being issued. The work of my section was summarized as follows:

"Probably the most important single result for the month is the preliminary determination of the density of 49 as not less than 15.5.

In general the work on methods of extraction and decontamination has been directed toward greater reliability, greater yields, and better understanding of the processes. Various improvements of the Bismuth Phosphate Process have been established; others, while promising, have not yet been shown to be definitely prefereable. The main part of the effort is now being devoted to purification and metal production, the work being coordinated by Dr. Thomas with that of other laboratories." It was stated that during the next two months the emphasis will be shifted more to purification and metal production of plutonium and some work will be started to develop the method of plutonium concentration to be used at Site W.

"The total project expense at Chicago and Argonne for the month of November is $539,195, of which $232,343 went for payroll. Total personnel employed as of today is 1,104 (408 academic) a net increase of 60."

The monthly report, General Physics Research--Properties of Plutonium (CK-1096), is being issued by W. W. Watson, the Division Director. The following individual section reports were included: General Physics by Wollan, Instruments by Wilson, Cyclotron by Snell, X-Ray Diffraction Research by Zachariasen, Mass Spectroscopy by Dempster, and Physical Measurements and Metallurgy by Creutz. Zachariasen had this to say:

"During the last month the efforts of this section have almost exclusively been devoted to investigations connected with the problems of determining the density of plutonium by x-ray diffraction. Because of the high priority of this problem and because of lack of duplicate equipment, it was necessary temporarily to halt work on other problems.

1. Graphite (Zachariasen): No results to report.

2. Uranium Fluorides (Zachariasen): No results to report.

3. Bismuth Phosphate (Mooney): No results to report.

4. Plutonium Metal and Plutonium Compounds (Mooney and Zachariasen).

A. Metal. Extensive tests show that we will be able to obtain satisfactory x-ray diffraction patterns of plutonium metal with as little as 25 micrograms provided the sample is in powder form. Dr. Kirk and his group have been trying to prepare a sample of the required type, but so far they have been unsuccessful due to the rapid oxidation of the metal when it is in finely divided form. At the time of the present writing, Dr. Kirk and his associates are trying to prepare a powder sample of the metal by decomposition of the hydride. We have obtained excellent diffraction patterns of uranium using a 23-microgram sample prepared by this method.

B. Plutonium Oxide. Excellent diffraction patterns of plutonium oxide were obtained using a sample weighing only 10 micrograms. The compound is cubic with a = 5.370 ± 0.002 Å. Assuming the composition PuO_2, we obtained a density of 11.54 ± 0.01. The structure is that of fluorite. The following table shows the relationship to ThO_2 and UO_2.

Table 1

	a	density
ThO_2	5.587	9.99
UO_2	5.46	10.94
PuO_2	5.370	11.54

C. Unknown Plutonium Compound. One sample submitted to us allegedly of metal gave good diffraction patterns. The substance proved to be hexagonal.

$$a_1 = 6.12 \text{ Å} \qquad a_3 = 3.75 \text{ Å}$$

By placing two metal atoms in the unit cell at positions ±(1/3 2/3 u) with u = 1/4, good agreement with the observed intensities is obtained. The substance is evidently some compound of plutonium, the identity of which is yet to be determined.

5. Uranium Hydride (Mooney and Zachariasen): In the course of investigations to test the hydride method of preparing powdered metal samples we obtained excellent diffraction patterns of UH_3. In agreement with earlier work (Rundle, CT-609, p. 30) we found the substance to be cubic with 8 molecules per unit cell. Our value for the lattice constant is 6.634 ± 0.002 Å, giving a density of 10.90 ± 0.01. The directly measured value is 10.95 ± 0.1 (Nottorf, CC-1063, p. 2). The atomic arrangement proposed by Rundle (loc. cit.) was confirmed.

6. Unknown Compound of Thorium (Zachariasen): A test sample allegedly of thorium metal prepared in the same manner as the plutonium sample referred to under 4(C) was examined. The substance proved to be hexagonal with dimensions

$$a_1 = 5.99 \text{ Å} \qquad a_3 = 3.81 \text{ Å}$$

There are two thorium atoms per unit cell at positions ±(1/3 2/3 u) with u ≈ 0.19. The substance is definitely a thorium compound and in all probability analogous to the plutonium compound mentioned under 4(C)."

Wednesday, December 1, 1943

Lyle R. Dawson, who was an associate professor and head of the Department of Chemistry at Louisiana Polytechnic Institute, Ruston, Louisiana, started work in Section C-I today. His wife and daughter Venita will join him at the end of January when Venita's school semester is finished. Also Ruth Casler joined Beaton's group as a technician.

The ninth St. Louis cyclotron neutron bombardment of uranium (Chicago IX), which started November 2, ended today at 106,082 microampere-hours.

I attended the evening meeting of the Purification and Metal Production Groups of my section at 7:45 in Room 209, Eckhart Hall. Others present were Baumbach, Brody, Davidson, Dirksen, Dixon, Florin, Fred, Ghiorso, Hagemann, Heath, Jensen, G. Johnson, Karle, Katzin, Kirk, Kohman, Manning, Marshall, Nachtrieb, Orlemann, Potratz, Rosenfels, and Willard. Kirk described attempts to prepare powdered plutonium metal for use by Zachariasen in determining the density of plutonium by x-ray diffraction. The method consists of reduction of PuF_4 with sodium or lithium at as low a temperature as possible; reduction with hydrogen was also attempted. The fairly rapid oxidation of metallic plutonium presents a serious difficulty since superficial oxide films absorb most of the x-rays. Zvolner has succeeded in depositing a few micrograms of metallic uranium from a molten calcium chloride-sodium chloride-UF_4 bath. Plutonium seems to have been deposited as the oxide cathodically from aqueous acid solution. Karle reported on experiments to reduce UF_4 and UCl_3 with atomic hydrogen. The black powdery masses formed do not appear to be metal, and experiments to determine if the substance is uranium hydride will be undertaken. Further work on plutonium is also contemplated. I mentioned that the metal production group is beginning work on hardness testing and the measurement of melting points. Dirksen and Fried described attempts to produce microcrucibles of thoria with a glazed surface to minimize wetting. Heating in a tungsten furnace (Fried) shows promise. Jensen discussed recent experiments in purification by precipitation methods, using thorium as a stand-in for plutonium. A very promising method is recrystallization of $Th(NO_3)_4$ and $ThCl_4$ from acetone or nitromethane; thorium peroxide (Th_2O_7) still rates high. Brody discussed recent experiments involving purification by solvent extraction. Nitromethane shows some promise as a solvent. Brody also conducted a set of experiments in an attempt to determine the oxidation-reduction potential of the plutonous-plutonyl couple by adding either oxidized or reduced plutonium to solutions of mercuric-mercurous ion of known concentration. Orlemann has reservations about the interpretation of the experiments because of the assumptions involved.

The top headline in today's paper reads, "F.D.R. to see Stalin in Iran." Roosevelt, Churchill, and Chiang Kai-Shek have been meeting in Cairo. Reports are that Roosevelt and Churchill are now enroute to meet Stalin.

Thursday, December 2, 1943

Using a sample of plutonium which was prepared by Baumbach (reduction of 40 micrograms of PuF_4 with lithium in a ThO_2 crucible system at 1,000°C) and sealed in an evacuated capillary by Fried, Zachariasen obtained an x-ray diffraction photo which gives lines corresponding to a plutonium density of about 13 gm/cm^3.

I responded to Allison's request for ways to improve the present liaison procedure between here and Site Y. The present procedure, I said, is inadequate for effective collaboration. I proposed that we receive more copies of the Site Y reports and that two or three of us here at Chicago who are intimately tied up with the purification and metal production program have authorization to visit Los Alamos, just as two or three Site Y men have been visiting us on a monthly basis.

In a memo to Allison, Szilard said that he has thought some more about my remark that the capture of neutrons by Pu^{239} might cause problems in the design of a plutonium weapon. In his words, "I thought more about what Seaborg said of the dangers of capture by 239, and also talked to Seaborg about it. I find that I was wrong and he was right, and that this capture might cause trouble."

I attended a meeting at 1:30 p.m. in Room 251, Ryerson Laboratory at which F. A. Otto described the equipment in the hot semiworks to be built at Hanford.

Orlemann and Kirk discussed the preparation of pure uranium halides and their reduction to metal with Charles A. Kraus of Brown University. For extreme purification, Kraus favors the use of ether extraction and volatilization.

I held the evening meeting of the Council of my section in my office at 7:45 p.m., attended by Beaton, Cunningham, Dreher, Ghiorso, Kirk, Manning, Orlemann, and Willard. I mentioned that Los Alamos report LA-30, a review report on the general chemistry of plutonium, has arrived in the New Chem library. I also mentioned that Hogness will be away next week on a trip to Site W and Berkeley. It was announced that the Chicago IX material will arrive tonight from St. Louis. Also, 50 pounds of Argonne metal from the Argonne pile has been received and will be stored until it can be handled in the semiworks. We will also get 50 pounds of metal from the Clinton pile within a month--it should contain approximately 750 micrograms of plutonium.

In a discussion of current research that followed, Cunningham mentioned that impurities from the dissolver used in dissolving the recent shipment of Berkeley neutron-bombarded uranium metal not only interfere with the purification of neptunium and plutonium but also make it difficult to perform accurate assays. It was agreed that the trouble with the dissolver should be cleared up before treating the 85 pounds of bombarded metal included in tonight's incoming shipment of St. Louis material. In connection with a discussion of the ether extraction studies

on U^{233}, I pointed out that the difficulties in dissolving the residue from evaporation of the ether fraction to dryness could be avoided by treating the ether fraction with excess water and boiling off the ether. Mrs. Mooney has now examined x-ray diffraction photographs of the sample of bismuth phosphate crystals containing 2½% plutonium, prepared by G. Johnson. She reports that results are inconclusive, although there is some slight evidence of a displacement of the bismuth phosphate x-ray lines. Johnson will attempt to prepare another sample containing 5 or 6% plutonium. Zachariasen reports a tentative density of 13 gm/cm^3 for the latest plutonium metal sample prepared by the metal group, but it is far from certain that the lines which he used for his interpretation are due to pure plutonium metal.

After Cunningham, Kirk, and others finished, I discussed the possible formation of heavy isotopes in pile operation. It is my contention that the search for U^{236} should be carried out again in view of the very recent news from Site Y that the reaction U^{235} + n shows three sharp resonances. Inasmuch as U^{236} is probably an alpha emitter, the detection procedure should be to get uranium from pile metal and to determine accurately the specific alpha activity. It is also possible that resonance levels similar to those of U^{235} exist in the case of 94^{239}; a rough calculation shows an estimated ratio of 10^{-2} for the ratio $94^{240}/94^{239}$ after 100 days operation in a future Site W pile. If this calculation is correct and if the half-life of 94^{240} were about 200 years, the alpha emission from 94^{240} would equal that from 94^{239}. A shorter half-life would greatly complicate the purification problem, particularly if the half-life were as short as six months, resulting in alpha particles with an energy of 7 Mev or greater which would make α,n reactions possible on O^{16} and N^{14}. Spontaneous fission decay of 94^{240} could also cause problems. If 94^{240} should prove to be a beta emitter, then 95^{240} would be formed that in all probability would be an alpha emitter and hence complicate the purification problem. I suggested it would be a desirable precaution to study the chemistry of element 95 with a view to separating it from plutonium if the need arises. To obtain element 95 for chemical studies, cyclotron-produced deuteron bombardments of plutonium should be tried. Another possibility which must be considered is the occurrence of neutron reactions with 93^{239} in the pile, which could also lead to the production of 94^{240}. I feel that the foregoing discussion constitutes a strong argument for continuation of the U^{233} work as insurance against unfavorable reactions in the pile.

The evening meeting of the Research Assistants of Section C-I was held in Room 209, Eckhart Hall at 6:45.

The neutron-bombarded material from the St. Louis cyclotron (Chicago IX) arrived during the evening. It consists of 600 pounds of UNH and 85 pounds of uranium metal. The metal sample is particularly important since it has been bombarded with high energy neutrons from a lithium target for the last 34,000 microampere-hours in order to increase the Np^{237} content.

Our navy has revealed large casualties in landing operations in the Gilbert Islands. At Tarawa marine losses were described as the heaviest of any operation in the Pacific.

Friday, December 3, 1943

I wired Zene Jasaitis in Los Angeles that I am glad to hear he has been granted a leave of absence and to come to the Met Lab as soon as possible.

I transmitted to Mulliken a table and chart of all the known heaviest induced radioactivities from isotopes of elements thorium through plutonium, together with a complete list of references, that Helen and I have compiled as a separate secret portion of my new "Table of Isotopes." Shown are half-lives, methods of production and decay schemes, and types of radiations and their energies.

In a memo to Mildred Thompson in Personnel, Willard recommends that every effort be made to obtain a grant of an additional year's leave of absence from the Owens-Illinois Glass Company for Clifford Smith, starting January 1, 1944. Smith joined Section C-I last April 10.

D. J. Hughes of Hilberry's office notified Captain J. H. McKinley that we have 1,540 pounds of uranium dioxide that is no longer needed and should be returned to the Manhattan District. The material, which has been subjected to a very slight bombardment, is located in the cyclotron room; Jaffey is to be contacted for instructions concerning its removal.

The Clinton Laboratories progress report for the month of November 1943 (MUC-NH-564) has been issued. Almost 800 persons (of which 160 are in research) are now employed, the November payroll amounting to approximately $193,000. The report states that the 100 Area [pile area] started in operation on November 4 this year with somewhat less delay and difficulty than had been anticipated. During the month 5 tons of metal were removed from the pile for use in the Separations Plant. The 5 tons contain 1.2 grams of plutonium. It is planned to deliver plutonium according to the following schedule during the first three months of 1944:

1+ mg	by January 1
1+ mg additional	by January 6
30 mg total	by January 10
500 mg total	by February 1
2 g total	by March 1
10 g total	by April 1

Hilberry, who wrote the progress report, states that the basic objectives of the research and development program at the Clinton Laboratories are:

1. To obtain fundamental information needed to facilitate the production and isolation of sufficient quantities of 49 for urgent physical studies.

2. To develop the Bismuth Phosphate Process of product recovery to the point where it can be utilized effectively at Hanford. This includes the development of necessary analytical controls.

3. To supply experimental data needed in Hanford design or operation.

4. To carry out properly authorized investigations requested by allied laboratories at other locations.

5. To develop background data essential to the formulation of an adequate health protection program at Hanford and elsewhere.

During the month of November at Clinton Laboratories, most of the previously planned research activities got under way on an effective basis. All of the time of the physicists was devoted to measurements associated with the start-up of the pile and its subsequent operation at a level of a few hundred kilowatts. In the groups concerned with product isolation studies, approximately 75% of the personnel in the Separations Development Division, and 50% in the Chemistry Division, worked on problems directly related to the Bismuth Phosphate Process. The remaining 25% in Separations Development and 20% of the chemists spent their time on alternate processes, principally dry fluoride and adsorption. Roughly 30% of the personnel in the Chemistry Division were concerned with various phases of fission-product studies, effect of pile radiation on construction materials, and the isolation of U^{233}. Most of the work of the Biological Group was of a preparatory nature, although some exposures of experimental animals in the pile were possible during the latter half of the month. Engineering Development efforts were divided about equally between stack gas disposal studies, measurements associated with the pile start-up, and operation and preparations for the Hanford water-tube corrosion experiments.

Plans for December and January at Clinton Laboratories call for dropping all work on the dry fluoride process and on the isolation of U^{233}, using the fifteen men thus released to step-up activities on the process flowsheet (Bismuth Phosphate), and also on adsorption as an alternative extraction method which may prove useful in some part of the mainline process. The Separations Development work on the main process flowsheet has been further strengthened by transfer of Sutton's group from Chicago. The emphasis on process development is gradually shifting from the extraction step, on which most effort has been previously placed, to the decontamination and concentration steps where a number of problems yet remain to be solved. The recent availability of uranium metal with much higher concentrations of plutonium and fission products than previously obtainable greatly increases the effectiveness of research work on decontamination.

Berlin reports that the U.S. has opened a big push in Italy and has made a three and a half mile gain.

Saturday, December 4, 1943

Baumbach summarized for me his discussion with Zachariasen about PuF_4 - UF_4 analogs. Zachariasen mentioned the existence of many x-ray diffraction photos that show there are two crystalline forms of UF_4, one black, the other green. This suggests the analogous two structures may exist for PuF_4, as material that has been reported to be very pure has produced a white fluoride and a black fluoride. A sample of finely divided plutonium metal prepared by slow, low-temperature (1,000°C) reduction of PuF_4 with lithium in a ThO_2 crucible system gives an x-ray spectrum showing ThO_2, LiF, Li_2O, and other x-ray lines, the latter presumably plutonium or a plutonium compound that fits a cubic structure and gives a density value of about 13 gm/cm . A sample of uranium metal prepared exactly the same way shows no ThO_2, but produces the normal structure of uranium metal. Baumbach suggests that the following possibilities be considered in deciding which approach should be used to prepare a satisfactory sample for future work.

1. Reduce PuF_4 to plutonium with barium to obtain a solid piece, followed by successive hydride preparations and decompositions until the sample is satisfactorily powdered.
2. Reduce PuF_4 with lithium at low temperature to form plutonium, LiF, and Li_2O free from ThO_2.
3. Reduce PuF_4 with barium at low temperature, to form plutonium, BaF_2, and BaO.

Dreher sent me a description of possible ways to improve the filterability of the $U(C_2O_4)_2$ carrier precipitate formed in a proposed method of concentration of plutonium at Clinton. These suggestions involve precipitation of $U(C_2O_4)_2$ from a HCl solution of plutonium-containing bismuth phosphate in glass-lined equipment or crocks.

In a memo to Wayne Johnson of Personnel, Watt endorses the qualifications of Fowler R. Yett for a job with the Met Lab. He says that he has known Yett for five years and that Yett completed his work for the B.S. degree in chemical engineering at the University of Texas in June 1943 with one of the best scholastic records made in that department in recent years. One of Yett's references on his personnel record at the University of Texas is Congressman Lyndon B. Johnson who is from Yett's home town of Johnson City, Texas.

Orlemann informed A. J. King of King Laboratories in Syracuse, New York, that the fluoride content of his November 3 crude barium sample was found to be 5,000 ppm.

General Groves is in Chicago. He attended a meeting at which it was decided that the first two milligrams of plutonium extracted from the Clinton Laboratories pile and to be purified there will be turned over to the Met Lab here about January 1. This will be followed by a delivery of another two milligrams to Los Alamos a week later.

Prime Minister Churchill, President Roosevelt, and Premier Stalin have conferred in Teheran and agreed upon military and political plans for the war against Germany.

Sunday, December 5, 1943

The eight-column headline in this morning's Sun reads, "Big 3 Prepare Invasion." At home the Office of Price Administration has raised the ceiling on corn and frozen the price of oats and barley.

Monday, December 6, 1943

I replied to Albaugh in Inglewood, California, that I am certainly glad to hear his leave of absence has been granted and that I am sure his Union Oil Company supervisor, Carr, will never regret the decision to permit him to come with us. I said that we had hoped he could join us before Christmas but that the personal considerations he has listed certainly seem important enough to account for the delay.

In a letter to Gofman in Berkeley, Ghiorso asks for the counting data obtained with our uranium standards using Gofman's counters. He also provides detailed information on the resolving power of our alpha counters, which give a constant resolution for counting rates up to at least 5,000 c/m.

"Chemical Research - Production and Extraction of 94. Report for Period Ending December 1, 1943" (CN-1095), covering the work of my section, is being issued. Information on the Wet Fluoride Method for the extraction and decontamination of plutonium is reported, based on work by Bradshaw, Dean, Gaarder, G. Johnson, Kroner, Koshland, and Thompson. A tabulation of corrosion data on the Wet Fluoride Process, supplied by J. A. Collins of the du Pont Company, indicates that the corrosion rates in 25-12-S-Cb stainless steel are within the limit of safety and that the Wet Fluoride Process could be run in the available equipment. Accordingly, work has been conducted on improvements to the flowsheet. Experiments show that ceric ion will readily dissolve LaF_3 or CeF_3 at room temperature and may have advantages over the use of zirconyl ion or of a metathesis step for this purpose. When ceric ion is used in the dissolving step, it may also serve to oxidize the plutonium. The cerium may then be precipitated in the by-product precipitate. Also, Ce^{+4} is an effective oxidizing agent for plutonium in solutions obtained by dissolving a LaF_3 product precipitate in a zirconyl salt solution, and Fe^{+2} is a satisfactory reductant in such solutions. Cerous fluoride has been found to carry plutonium both at tracer concentrations and at Ce:Pu weight ratios as low as 9:1.

"California Project Chemical Research - Chemistry of 94, Report for the Month Ending December 1, 1943" (CN-1134), is being issued. Hamaker and Sheline report on ultramicrochemical investigations of the carrying of $Pu^{(r)}$ (when present at "W" concentrations) with the by-product precipitate in the Sodium Uranyl Acetate Process. Initial experiments

indicate that 20-50% of the plutonium is carried. More recent work in which a preliminary oxidation of plutonium, carrying with sodium uranyl acetate, and then reduction by hydroxylamine were performed before the by-product precipitation, shows only 9% to be carried. It is suggested that the 20-50% carrying in the first experiments results from some difference in the form of the $Pu^{(r)}$ itself--perhaps a hydrolyzed form. L. J. Beaufait, F. J. Leitz, A. Stein, and F. Stevenson studied the Sodium Uranyl Acetate Process used as a decontamination process only. The cycle with no volume reduction is found to be as efficient as the normal acetate process in regard to both product recovery and fission product decontamination.

At 3:40 p.m. Jaffey held his regular weekly meeting in the New Chemistry Annex Conference Room.

The evening seminar of the Chemistry Division was held at 7:45 in Room 251, Ryerson Laboratory.

Today's reports concern the U.S. troops' storming of key heights on the road to Rome. British troops have captured an important ridge on the southern flank on the road from Mignano to Rome.

Saturday's New York Times quoted the Berlin radio as saying that the German High Command "intends by one fell, drastic stroke to end the unbridled mass murder," and added that "mankind is not far from the point where it can at will blow up half the globe." We all wonder about the status of the German nuclear bomb research.

Tuesday, December 7, 1943

In a summary of the work of the Chemistry Division for the month, Franck states that the most important single result for the month is the preliminary determination of the density of Pu^{239} as not less than 15.5 gm/cm^3. He goes on to say that, in general, the work on methods of extraction and decontamination have been directed toward greater reliability, greater yields, and better understanding of the processes. Various improvements of the Bismuth Phosphate Process have been established; other methods, while promising, have not yet been shown to be definitely preferable. The main part of the effort is now being devoted to purification and metal production, the work being coordinated by Charles A. Thomas with that of other laboratories. During the next two months, he said, emphasis will be shifted still more to purification and metal production of plutonium. Some work will be started to develop the method of concentration of plutonium to be used at Site W.

At a meeting of the administrative staff attended by Granger Cook, L. C. Doak, T. E. Hall, J. C. Hobart, D. K. Keith, L. A. Kimpton, W. C. Munnecke, L. H. Richmond, J. C. Stearns, and W. J. Vatter, it was announced that the Army has given its approval for Met Lab employees to punch out at 4:30 on the afternoon of December 24, 1943, provided that no lunch hour is taken. Decentralized Christmas parties are being organized at the New Chemistry Building, Armory, Ryerson Laboratory,

and Argonne. Kimpton reported that from certain reports originating from Germany it appears that the Germans are working on a program similar to ours and quite possibly are nearer to the solution. He emphasized that the job of the administrative staff is to cooperate with us scientists and to expedite fulfillment of our needs.

Franck sent a memo to Compton concerning recent announcements being made by the Nazi Ministry of Propaganda and the German High Command that they will be able to win the war by the use of secret weapons. Franck is of the opinion that part of what they are saying indicates quite clearly that atomic bombs are in preparation; it is his further deduction that the weapon is not completed. He believes the Germans have a uranium-heavy water pile and have used this to produce uranium-233 from thorium. He goes on to urge that a veiled announcement of our own atomic bomb work be made at the present time for three reasons: (1) the threat of a more devastating retaliatory attack by us may make the Germans think that it would be better to surrender than to attack us with an atomic bomb; (2) if the German weapon should be available before we are ready with ours, Franck thinks they will strike at London and then send an ultimatum to the United States--if such an ultimatum were then to be answered with the promise that we would retaliate in kind, it would already be too late and might be regarded as a bluff; (3) it is important to prepare the opinion of the American public slowly to the possibility of a terrible secret weapon so that if the Germans use it first, the psychological shock will not be too great. Franck also says there seems to be no doubt that German scientists know we are working on an atomic bomb, just as every physicist in the United States knows it and as he knew it before he joined the Project--without anyone having to tell him.

Some of the physicists in Herbert L. Anderson's P-IV Section, Division of Nuclear Physics, have been measuring the radioactive properties of various isotopes produced at Argonne Laboratory. Today Leo Seren sent me a tabulation of newly measured properties of several isotopes (i.e., O^{19}, S^{35}, Fe^{59}, $Ag^{108,110}$, Cd^{115}, W^{185}, and Tl^{206}), that were not listed in my August 1940 review article in Chemical Reviews. The source of the slow neutrons used in the production reactions was the Argonne pile, and the fast neutrons used in the In(n,p) and Sn(n,α) reactions to produce Cd^{115} were from the bombardment of beryllium by deuterons at the St. Louis cyclotron. He says that they are still following the decay of the Cd^{115} from the indium and tin bombarded by fast neutrons.

U.S. carrier task forces smashed anew at the Japanese-held Marshall Islands last Saturday, today's newspaper reports.

Wednesday, December 8, 1943

Hogness began a four-day visit in Berkeley to discuss the research activities under way in the California Project under Latimer.

Reports in the news today indicate that Turkey will join the Allies. After their conference with Stalin, Roosevelt and Churchill met in Cairo with President Inonu of Turkey. There are predictions of a full-scale assault in the Balkans. The conference was held December 4, 5, and 6.

The Project Council assembled at 9:30 a.m. in Room 209, Eckhart Hall for its Policy meeting. It was attended by Allison, Capt. Chapman, Chipman, Compton, C. M. Cooper, Dempster, Doan, Fermi, Franck, Hughes, Jeffries, Warren Johnson, Miles, Mulliken, Munnecke, Capt. Murphy, Smyth, Spedding, Stearns, Stone, Vernon, W. W. Watson, Whitaker, and Wigner. Compton surveyed the history of the Project, beginning with the meeting in Washington on December 6, 1941, when Compton, Urey, and Lawrence were asked to report to Vannevar Bush of OSRD on the feasibility of making an atomic bomb. He summarized the present situation by stating the developmental stages of production are now well in hand, extraction work is not yet complete but is moving satisfactorily, and the production plant is under construction. Compton mentioned that U^{235} also is in experimental production and that Site Y is an efficient, going organization making satisfactory progress. The present objectives of the Metallurgical Project were stated to be the following (roughly in order of importance and urgency).

1. Make Hanford go promptly and effectively.
2. Produce what product we can, as soon as possible.
3. Purify the product and reduce it to metal and hydride (joint task with Site Y).
4. Aid Site Y in solving special problems.
5. Defensive and offensive radiological warfare.
6. View toward the future.

In a discussion of the Met Lab program, Allison said that my section has three men working on the U^{233} problem, and he proposed that the thorium in the Clinton pile be left until appreciable amounts of U^{233} can be extracted, as Oppenheimer and Teller desire. Compton agreed that this program is a Chicago responsibility, whereupon Johnson said the cans should then be left in the pile indefinitely. Whitaker discussed the Clinton Labs production program for the near future that now calls for the production of 98% purity plutonium on the following schedule:

By January 1	1 mg
2-3 weeks later	2 or 3 more mg
January 20	0.5 grams
February 20	2.0 grams

He pointed out that meeting the last two dates will depend upon the success of the chemical plant operation.

Thursday, December 9, 1943

Fowler R. Yett, whom George Watt recommended so highly, from the Institute of Paper Chemistry in Appleton, Wisconsin, started work in Section C-I. When I was introduced to Yett this morning, he calmly put out his hand and in a very serious manner said, "Others may stink, but I am Fowler Yett."

I sent Mulliken a table giving a complete list of all isotopes from K^{40} through U^{238} having natural radioactivities, and showing their atomic and mass numbers, type of radiation, and half-life. This table supplements the "Table of Heaviest Induced Radioactivities" I sent him last Friday and is part of the "Table of Isotopes" that Helen and I have been working on.

Franck sent Eastman the following letter:

"The program for the Council meeting has been changed to some degree. The Policy meeting will be held on Monday, December 20th, at 9:30 a.m.; on Monday afternoon there will be the meeting with C. A. Thomas on purification; Monday evening, the usual seminar of the Chemistry Division and the Technological Division will report.

"Will you kindly let me know who will come from California to the meetings, and if anyone will talk at the Thomas meeting. You understand, of course, that not metal production but purification is on the agenda this time. Will somebody from California talk in our seminar on Monday evening, and if so, for how long? How much time shall we reserve for the report of California in the information meeting Tuesday morning? I may remind you that the time available at the meeting is very limited. Since I have to send the program of this meeting to Dr. Thomas beforehand, I would appreciate a prompt answer from you." Franck also notified Spedding and Thomas of the change of date of the meeting.

In a memorandum to Compton, Smyth suggests plans to be implemented if and when our enemies make first use of the weapons which we are trying to develop. This is a follow-up to meetings that he, Compton, and Fermi held last Saturday and Monday. The memorandum is as follows:

"With reference to the discussion that we had last Saturday with Fermi and again on Monday, I would like to suggest the formation of a committee which should make plans to put into effect if and when our enemies make first use of the weapons which we are trying to develop. This committee should make plans for action within the DSM projects and also for information to be given to the public should the need arise. In considering each of these objectives it must bear in mind the possibility of either radioactive or explosive action.

"DSM Projects Considerations. As typical of the questions which might be considered one may mention the need of altering plans so that radioactive poisons might be produced or of concentrating on the rapid production of 49 or 25, discontinuing production methods that do not seem likely to bear fruit rapidly. It is conceivable that in any

emergency operating crews for the electromagnetic separators should be supplied from the professional personnel of the Metallurgical Laboratory and other parts of the project. It is conceivable that one Hanford pile should be built with early possible simplification instead of the three now planned. Perhaps the whole K-25 project should be scrapped.

"Public Information. In preparation for the possible use of either radioactive or explosive bombs, plans should be drawn up for informing the public as to what protective action could be taken, what the general nature of the problem is, and possibly something about our own state of preparation. Such information bulletins which would presumably be broadcast and published when the need arose should be kept on file and constantly revised in terms of new information.

"Needless to say, the above are the crudest kind of suggestions but perhaps will serve as a reminder of the desirability of discussing this possibility with the General."

The evening meeting of the Research Assistants of my section took place at 6:45 p.m. in Room 209, Eckhart Hall.

The regular evening meeting of the Council of my section at 7:45 in my office was attended by Beaton, Cunningham, Davidson, Dreher, Ghiorso, Kirk, Manning, Orlemann, and Willard. I told the group that J. C. (Jake) Warner, a competent inorganic chemist from Carnegie Institute of Technology in Pittsburgh, will join the Project and be stationed here in Chicago more or less permanently to serve as C. A. Thomas' representative to help the Project-wide coordination of the plutonium purification and metal production program. The problem of the apportionment of plutonium after gram quantities are available was discussed; it was agreed that some mechanism should be set up for planning the distribution of plutonium. The remaining blueprints for the quartz microbalances have not yet been received from Berkeley, and I agreed to wire Kamen immediately. Cunningham reported that the alpha activity of the El Dorado uranium sample I brought back from the Bureau of Standards is 1,513 d/m/mg for 50% geometry, which is about 3% lower than the value of 1,555 obtained earlier for several samples of pure uranium. No detectable amount of Pu^{239} was present in the sample. Cunningham also mentioned Crawford's calculation that it is possible to have 1.3% back reflection of alpha particles from platinum. Plans are underway to check this calculation by comparing samples mounted on quartz and on platinum. We talked about the isolation of Np^{237}, which is proceeding slowly; so starting tomorrow, work on the St. Louis sample (Chicago IX) will be extended to three shifts.

Six Japanese ships, including two cruisers, have been sunk by the U.S. in an attack on the Marshall Islands.

Friday, December 10, 1943

Last month when I visited the Bureau of Standards in Washington, Curtiss and Levinos gave me a sample of uranium oxide (U_3O_8) that contained an alpha-emitting impurity which they conjectured might be plutonium. Today I wrote Curtiss that we have now completed some tests on the sample and show that it is completely devoid of plutonium within the accuracy of our measurements. This accuracy is such that we can say that less than 10^{-4} percent of the alpha count can be ascribed to plutonium, whereas the Bureau's measurement shows that the impurity amounts to 1.3% of the total activity. I told him that we have also made a determination of the specific alpha activity showing it to be 1,520 disintegrations per minute per milligram of uranium (assuming the sample to consist of pure U_3O_8), which checks very well with the Bureau's value of 1,500 d/m/mg plus 1.3%. I asked to hear from Curtiss and Levinos about their alpha analysis of a UNH sample that we recently shipped to them at their request. For comparison, I said that according to our best determination, it measures 1,500 alpha d/m/mg of uranium. I then thanked Curtiss for his hospitality during my recent visit.

Today we are somewhat dismayed to read an editorial in the Chicago Daily News which includes the statement: "The Germans have been monkeying with that isotope of uranium, too, supposed key to unlock the power imprisoned in the atom, we are told. If the Germans are busy with uranium, it is for no good purpose." There is some concern that this might represent a leak of information from our Project and relates to our continued concern that the Germans may be ahead of us in the race to produce a nuclear weapon.

The Soviets have dealt big blows to the German army in the Kiev area.

Saturday, December 11, 1943

It is expected that a milligram or so of plutonium will be available to use here in Chicago within the next three weeks or so as a result of the semiworks operations at Clinton Laboratories. Several more milligrams will be available from the chemical extraction plant operations not long thereafter. Since my section has the responsibility for a number of determinations on which greatly accelerated progress can be made with milligram amounts of plutonium, and which are critical to the success of the Project, I wrote a memorandum to Allison indicating the nature of our needs and, insofar as possible, their priority. I wrote the following:

"(1) Our highest priority problems at present are the determination of the density, melting point, and hardness of plutonium metal. It now appears that we may not be able to obtain final values for these constants until milligram quantities of plutonium are used. It is certain that the answer can be obtained more rapidly with several milligrams than with a single milligram.

"(2) Several weeks ago we were requested by Mr. Squires of the du Pont Company to draw up a flowsheet for the concentration-isolation procedure at Site W. In so doing we were forced to speculate rather broadly as to the solubility and rates of dissolving of certain plutonium compounds. Before final designs can be approved for the concentration building, work needs to be done with quantities of plutonium at least as great as a milligram.

"(3) Although tests of the various steps in the Bismuth Phosphate Process have been carried out at Site W concentrations of plutonium on a microliter scale, it is of the highest priority to test the overall process at Site W concentrations as soon as milligram quantities are available.

"(4) As you know, the work on purification of plutonium from light impurities to date has of necessity been done largely with "stand-ins." Since a number of proposed methods of purification have now been studied in this fashion, it is essential that they should be tested with plutonium at the earliest possible date if there is to be hope that satisfactory purification methods will be available when product is ready from Site W. As a part of the purification program material should also be assigned as soon as possible for the study of the analytical problems involved (which work is going on largely in Potratz's section).

"(5) Further development of our knowledge of the basic chemistry of plutonium is extremely important if we are to be prepared to meet emergencies which may arise in the course of large scale processing of the product. For that reason I believe that as soon as possible milligram quantities should be assigned for the determination of oxidation states, solubilities, etc.

"It is difficult to assign relative priorities to these problems unless it is done on the basis of the time schedule. On this basis I think I should place them in the order listed above. I believe it will be in the interest of the attainment of the aim of the project if you will take such steps as are necessary to have the first amounts of material which are produced at X assigned to these problems."

The Bulgarian capital of Sofia was blasted for an hour and forty-five minutes in a daylight raid today.

Sunday, December 12, 1943

The headline in the Sun today reads, "U.S. Warns Nazi Satellites." Secretary of State Cordell Hull called on Hungary, Rumania, and Bulgaria to get out of the war to avoid suffering and sharing in Germany's defeat.

In the afternoon Beaton, Russell, Kidde, and I met with Major Vance, Dr. Winters, and Colonel John Ruhoff of the Madison Square area of the Manhattan District to discuss the use and limitations of resins in adsorption processes. They are interested in the possible use of resins for the purification of uranium in the Mallinckrodt plant in St. Louis.

This plant now uses the ether extraction process for this purification, but the recent explosion has raised some questions about the safety of this process.

At 11:30 p.m. I left Chicago (Englewood Station) by Penn R.R. train for Cincinnati, where I will continue on the L&N R.R. to Knoxville and then by car to Oak Ridge.

Monday, December 13, 1943

Meetings scheduled for today at the Met Lab are Jaffey's regular weekly meeting in the New Chemistry Annex Conference Room at 3:40 p.m. and the evening seminar of the Chemistry Division that regularly meets at 7:45 in Room 251, Ryerson Laboratory.

I arrived in Knoxville in the afternoon and was met by Stan Thompson and Mary and Vance Cooper. Stan, Vance, and I played nine holes of golf at the Municipal Golf Course (near the Whittle Springs Hotel), while Mary went shopping. We all had a steak dinner in Knoxville, then drove to Coopers' home in Oak Ridge where we had coffee and interesting discussion. I spent the night at the Coopers' in their project-built home at 135 Outer Drive in the village of Oak Ridge.

Tuesday, December 14, 1943

I awoke this morning to find a light snow had fallen during the night. I had breakfast with the Coopers, and Mary Cooper made up lunches for Vance and me to take to the Laboratory. I have decided to forego eating in the Laboratory cafeteria in view of my experience with stomach trouble (the "Clinton Fever") during my visit here last month.

I spent the day in visits to the laboratories of and discussion with Perlman's people. I am especially interested in the assistance being given by Thompson, Kroner, Halperin, James, Gaarder, and Malm from my Chemistry Section C-I on the start-up of the Clinton chemical extraction plant; we are anxious that the Bismuth Phosphate Process operate successfully and are doing everything possible to help. Perlman's people are also helping the chemical engineers and chemical plant operators in every conceivable way. Everyone is quite excited because the pile has operated very well; neutron-irradiated uranium has been discharged and is scheduled to start through the chemical extraction plant next week.

I also am especially interested in the preparations for the concentration and isolation of the plutonium from the final solutions as they are received from the chemical extraction plant. The concentration will be done by Vance Cooper's group with the final isolation by Werner's group. The plan is to use lanthanum fluoride oxidation-reduction cycles for concentration after which the nearly pure plutonium will be precipitated, perhaps as the peroxide or oxalate. The fellows are trying to be

ready for any contingency, but I suspect they will encounter many unexpected impurities and problems that will tax their ingenuity.

I am happy to have dinner alone with Vance and Mary Cooper tonight after my exciting day. I am again spending the night with them.

Wednesday, December 15, 1943

I had breakfast at the Coopers', and Mary again made my lunch to take to the Laboratory.

During the day I attended the joint Met Lab-Clinton Laboratories progress meeting on the development of the Bismuth Phosphate Process and the meeting of the Steering Committee for this Process. We focused our attention on the adaptation of the Process for use at the Hanford plant.

This evening I had dinner with the Comptons in their new home at 104 Olney Lane. Crawford Greenewalt was invited also. I was reminded of the last time Greenewalt and I were together at the Comptons' home in Chicago. That was March 3 of this year when Clinton Laboratories was still in the design stage. Since then a city has risen from the farmlands of the Tennessee countryside, the graphite pile has been built and is operating, we have settled on the Bismuth Phosphate Process for extracting plutonium from the uranium and fission products, and the separations plant is completed and ready to process tons of neutron-irradiated uranium.

Compton informed Greenewalt and me of a directive he has just received from General Groves. Groves asks that the Metallurgical Laboratory halt until further notice all work directed toward the development of a uranium-233 process and that the Clinton Laboratories discontinue all work with the chemistry of U^{233} until April 1 next year, when the position of this problem may be reconsidered. Compton has now implemented this directive by sending a letter to Allison and a memo to Whitaker. In order to make use of the materials already prepared, he further instructs Whitaker to leave the capsules containing thorium in the pile to accumulate U^{233}; also, such additional capsules as may be already prepared should be inserted in the pile if this procedure does not interfere with its operation.

Thompson, Kroner, Halperin, James, Gaarder, and Malm concluded their stay at Oak Ridge which began on November 21. They boarded the night train on their way back to Chicago and are scheduled to arrive there tomorrow afternoon.

I again spent the night at the Vance Coopers'.

Thursday, December 16, 1943

I had breakfast at the Coopers, and Mary again packed a lunch for me. I visited the chemical laboratories to continue my discussions with the people in Perlman's section and to make plans for the coordination of their work with the related work in my Chemistry Section C-I.

"Chemical Research - Special Chemistry Plutonium. Report for Month Ending December 11, 1943" (CK-1145) is being issued in Chicago. The work accomplished during this period is reported as follows. Davidson and co-workers studied the reaction of tracer quantities of plutonous and uranous oxide with anhydrous HBr as part of a continuing investigation of the possibility of using the volatility of plutonous bromide in the purification program. They find that tracer quantities of plutonous oxide are not appreciably volatilized as the bromide in HBr at a temperature below 600°C, whereas uranous oxide volatilizes at 400°C. The hydrogen bromide generator used was constructed with the help of Jaffey and Miller. Florin and Davidson constructed an apparatus for the study of the reactions of fluorine activated at room temperature in a low pressure, water-glow discharge and find it to be a potent fluorinating agent that might be useful in preparing a volatile higher fluoride of plutonium for purification purposes. Efforts were concentrated on attempts to prepare metal for density determination by means of x-ray diffraction because of its two great advantages--(1) it is relatively independent of the presence of inclusions and non-metallic solid phases, and (2) it gives the true density of the metal crystallites to a very considerable degree of accuracy. The final isolation and purification of plutonium nitrate solution for metal production was carried out by Hindman, after which Kirk and Rosenfels converted the solution to oxide. Florin performed the conversion of plutonous oxide to fluoride by treatment with gaseous anhydrous HF at a temperature of 550°C. (Several 100 microgram samples were prepared, and the products had a variety of colors: gray, purple-tinged transparent, and red-brown.) Baumbach, Fried, Kirk, and Rosenfels carried out the production of powdered plutonium metal by metallic reduction using barium, sodium, magnesium, and lithium as reducing agents under various conditions. In all, 30 reductions were carried out with PuF_4 or PuO_2 samples ranging from 5 to 50 micrograms. Rosenfels and Fried developed techniques for mounting microgram samples for x-ray diffraction studies; six samples of plutonium oxide, fluoride, and metal were mounted in lucite and seven samples of plutonium metal, uranium metal, and uranium hydride were mounted in glass capillaries. Mounting in glass proves to be the more satisfactory technique. No satisfactory sample of powdered plutonium metal, required for density determination by the x-ray diffraction method, has been prepared--prevention of oxidation of the metal during the sample mounting procedure presents a serious problem. Kirk, Rosenfels, Fried, and Baumbach carried out determinations of density by a capillary method whereby the volume of weighed solid pieces (4-28 micrograms) of plutonium metal is determined by displacement of a meniscus of butylphthalate in a capillary cone. The density found for the particular samples of metal studied, that were about 90% plutonium, is 15.5 gm/cm^2 (two samples were the product of

barium reduction and two were the product of magnesium reduction). Probable impurities are lanthanum, barium, or magnesium fluorides, thoria or plutonium oxide. It is calculated that if all of the 10% impurity is magnesium fluoride, the true density of the metal would be about 28 gm/cm^3; other impurities would give lower values down to 17 gm/cm^3 if all the impurity is thoria. Baumbach and Fried prepared plutonium hydride from microgram quantities of plutonium metal using a pyrex glass capillary procedure and find that in one quantitative experiment, the data yield a formula of $PuH_{2.97}$. Karle further investigated reduction by atomic hydrogen and obtained evidence that a stream of atomic hydrogen will reduce UF_4 to metal at temperatures of 600-800°C. UCl_3 is found to be more readily reduced than UF_4. Plutonium fluoride was reduced in a single experiment, the temperature being about 700°C for 15 minutes. Kirk and Zvolner made further attempts to electrolyze a few micrograms of uranium metal from aqueous or organic solutions and were partially successful. Similar attempts with plutonium were unsuccessful. Patton carried out microchemical tests on the stability of oxidized plutonium in ether in view of the interest in ether extraction as a possible purification method. At concentrations of 50-100 mg/liter he finds that less than 2% of the plutonium in the ether is reduced per hour.

The Vance Coopers and I had dinner at the home of Harrison and Adele Brown (107 E. Malta Rd.). Then I caught the midnight train at the town of Clinton headed for Cincinnati, where I will continue on the N.Y. Central R.R. to Chicago.

Friday, December 17, 1943

In Chicago "Chemical Research - Production and Extraction of Plutonium. Report for Month Ending December 11, 1943" (CN-1153) is being issued. The following results are reported. Beaton and La Chapelle have conducted preliminary experiments on adsorption-bismuth phosphate coupling that indicate that plutonium tracer quantities may be separated with a yield of more than 95% from the elutriant of an Amberlite IR-1 extraction cycle by precipitation with bismuth phosphate. An investigation is in progress to determine the decontamination factors obtained by bismuth phosphate cycles following such a cycle. Belletire, Cunningham, Hindman, Howland, Jaffey, Kraus, Magnusson, Meyer, Patton, and Clifford Smith completed the isolation of 300 micrograms of plutonium from 700 pounds of UNH which have been subjected to 100,000 microampere-hours of bombardment with neutrons from a Be + 12 Mev deuteron source at the St. Louis cyclotron during the period October 5 to October 25 (Chicago VIII). About 55 man-days of work were involved. After a preliminary cooling period of 8 days, the bulk of the uranium was separated from the plutonium and fission products by two ether extraction cycles under the direction of Jaffey. The resulting aqueous phase had a volume of about 7 liters and consisted of about 60% UNH. It contained much solid matter and required filtration, after which analyses were made that showed 323 micrograms of plutonium in the filtrate and 20 micrograms in the residue. In the

course of the subsequent concentration, using a series of lanthanum fluoride oxidation-reduction cycles, two oxidizing agents (Ce^{+4} and Ag^{+}, $S_2O_8^{+}$), two reducing agents (SO_2 and H_2O_2), and three methods of dissolving LaF_3 (solutions of Ce^{+4} ion, fuming with $HClO_4$, and fuming with H_2SO_4) were used. Near the end of the processing an additional 85 micrograms of plutonium containing about 100 micrograms of La^{+3} (received from Clinton Labs) were added. The final steps were two peroxide precipitations, following which the sample was dissolved in 2 microliters of HNO_3 and delivered to the metal production group. It contained about 210 micrograms of plutonium (additional amounts of plutonium were recovered separately from other fractions).

James and Thompson conducted experiments on the thorium molybdate method of separation of plutonium and neptunium and found that in an acetate solution at pH 0.9-1.5, 95% of tracer amounts of plutonium may be precipitated with thorium molybdate, whereas about 5% of tracer neptunium present is carried (separation of plutonium and neptunium may become necessary at Site X or Site W if the cooling time is shortened, or if Np^{237} is present in undesirable amounts). Thompson, Dean, Dreher, Gaarder, and Kroner worked on decontamination improvement in the Bismuth Phosphate Process by use of scavengers and holdback carriers. (These studies are a cooperative effort by Technical Section T-1 and Chemistry Sections C-I and C-III). The precipitation of columbic oxide, zirconium hydroxide, or ceric phosphate with the by-product precipitate in the first decontamination cycle of the Bismuth Phosphate Method shows a 10-20 fold improvement of decontamination with respect to both beta and gamma species. If any two of these substances are used together, the total improvement is still higher. The use of lanthanum, yttrium, and ruthenium holdback carriers in the product precipitation of the decontamination cycle (following the use of the above scavengers in the by-product precipitate) does not appear to produce any significant improvement in decontamination.

I arrived in Chicago at eight o'clock in the evening; the train was unusually late.

Today's headline news is from the war in the Pacific. U.S. troops have landed at Arawe to gain their first foothold on New Britain Island.

Saturday, December 18, 1943

I learned that last Tuesday the dissolver in the West Stands was charged with uranium metal from the Chicago IX St. Louis bombardment.

On Wednesday, Dreher, Kircher, and Christiansen visited the Belke Manufacturing Company to obtain information on rubber coating materials that could be applied to stainless steel.

On that same day, Davidson wrote to A. F. Voigt at Ames Laboratory asking whom to contact at the Jackson Laboratories regarding their rumored capability of fluorinating UF_4 to UF_6 at the remarkably low temperature of 70°C; the usual temperature is 200°C.

Day before yesterday, on Thursday, Dean, Margolis, and Creek moved to Room 5 in the West Stands. The same day Ghiorso attended a meeting on instrument problems with Bradford, Bradley, Broxon, Girdler, Jesse, Landsverk, Moon, W. W. Watson, Weller, and Wollan.

Dreher sent a summary to Thompson on Thursday of the developments in extraction-decontamination work during Thompson's absence in Oak Ridge as follows. Margolis has done considerable work in the laboratory attempting to develop conditions for dissolving the St. Louis-bombarded metal that will avoid the difficulties experienced with the Berkeley metal dissolved in the West Stands dissolver (insoluble "cruds" in subsequent concentration steps carried out by Cunningham). Dean and La Chapelle are now testing the coupling of the Bismuth Phosphate Process to the adsorption process. Schuler and Margolis have tested the Wet B (Lanthanum Fluoride Process) through the extraction and one decontamination cycle starting with 20% UNH solution and using dichromate as the oxidizing agent and ferrous ion as the reducing agent. Product yield was about 75% and decontamination factors were 80 for beta particles and 30 for gamma radiation. Schuler and Creek have been testing the effect of the proposed "W" coatings on the Bismuth Phosphate Process. Creek has tested the characteristics of the uranous oxalate precipitate with regard to settling and filtration by the "filter stick" technique. G. Johnson has been testing the Bismuth Phosphate Process under "W" conditions with tracer amounts of product on a 200 microliter scale. Dean has tested a process in which plutonium in the oxidized state is extracted into ether, followed by the use of bismuth phosphate oxidation-reduction cycles; the plutonium yields are normal, but accurate decontamination factors could not be determined.

Thursday, also, Clifford Smith, Willard, and Stearns met with H. T. Griswald of the Owens-Illinois Pacific Coast Company about an extension of Smith's leave of absence from the company. It was agreed to reconsider the case further in March 1944.

The Research Assistants of my section met as usual at 6:45 Thursday evening in Room 209, Eckhart Hall.

Also Thursday evening, the Council of my section had its meeting, attended by Cunningham, Davidson, Dreher, Kirk, Manning, Orlemann, and Willard. It was mentioned that as a result of the most recent speed-up, the first Site W pile is now scheduled to go into operation in June, with the whole plant to be in full swing before winter of 1945. Fifty pounds of hot uranium metal will be shipped to Kircher from Clinton. This material may contain several micrograms of Np^{237}. Cunningham pointed out that most of his people have been working almost exclusively on plutonium isolation and recovery for many weeks; he hopes that many will be released for other work soon. Work on the quartz fiber balances is slowly progressing, and it is hoped that the wheels will be available soon. The present status of alternate processes was discussed. The Wet Fluoride Process is now the first alternate, but a great deal of study is needed to test new modifications including operation at low temperature to minimize corrosion. The Technical Division is giving

attention to a continuous ether extraction process that may be useful for purification as well as extraction. The processing of the UNH part of the last St. Louis (Chicago IX) bombardment is nearly completed. Hydrogen peroxide has worked well as a reducing agent for plutonium in the presence of fluoride. Approximately 50 micrograms of plutonium will be available in a few days for research by groups other than the metal production group. It was mentioned that Hagemann will need 10 micrograms for his work. Fluorine problems were discussed by Davidson. Heath's work shows that the behavior of plutonium fluoride in calcium fluoride tubes is very similar to the behavior in copper tubes, indicating that the effect of different surfaces may not be very important. If a calculation is made on the basis of Heath's tracer fluoride volatility experiments, the vapor pressure of the plutonium higher fluoride turns out to be of the order of 0.005 mm of mercury at 500°C, but this calculation is questionable. Davidson raised the question of whether or not an experiment with about 10 micrograms of plutonium should be carried out under conditions similar to that of the tracer experiment. It is possible that other methods of forming a higher fluoride should be tried. It may be desirable to test the fluoride volatility method with microgram quantities of uranium containing sufficient U^{232} to give several thousand alpha counts per minute. If the behavior of uranium under these conditions is like that of plutonium rather than like that of large amounts of uranium, there would be a clear indication that the present method is not adequate for very small quantities of material. The bromide work of Bradshaw was reviewed briefly. Several revolatilization experiments have been carried out with definitely negative results. Kirk described Baumbach's experiments with a hot-wire technique. In several experiments with uranium iodide the uranium did not deposit on the wire (8 mil tungsten wire was used in these experiments). Experiments on formation of refractories were discussed. It is planned to test the degree of contamination of plutonium metal after pure metal is heated in various refractories. The next experiment in the cooperative program with Zachariasen will be a low-temperature reduction of plutonium fluoride with barium in order to see if the x-ray lines that are found are the same as those that were observed in material produced by reduction with lithium.

Yesterday Theocian H. Carter was hired to work as a technician for Orlemann.

I have decided to shift one of the especially able young men to a problem of particular interest to me. This morning Ralph James and I had a general discussion of the problem of research to synthesize and identify new heavy isotopes and elements. One object of these experiments is to look for element 95 and to study its chemistry. A primary object is to search for new isotopes of plutonium (other than Pu^{238} and Pu^{239}). (The presence of such isotopes, if they are produced in the pile, and if they decay at sufficiently high rates by spontaneous fission, or even by alpha particle emission, could seriously affect the purification of Pu^{239} necessary for success of the final device.) We decided to carry out practice bombardments with 10 mg amounts of uranium and thorium in order to test our techniques preparatory to bombarding

plutonium in our search for isotopes such as 94^{240}, 95^{239}, and 95^{240}. Later in the day I held a meeting on this subject in my office, attended by Fulbright from the St. Louis cyclotron, James, and Art Snell. I explained that the first practice experiments will be performed by bombarding thorium and uranium with deuterons to give the following reactions:

$$U^{238}(d,n)93^{239} \xrightarrow[2.3\ d]{\beta^-} 94^{239} \xrightarrow[21{,}000\ yr]{\alpha}$$

$$U^{238}(d,2n)93^{238} \xrightarrow[2.0\ d]{\beta^-} 94^{238} \xrightarrow[50\ yr]{\alpha}$$

$$Th^{232}(d,n)Pa^{233} \xrightarrow[25\ d]{\beta^-} U^{233} \xrightarrow{\alpha}$$

$$Th^{232}(d,2n)Pa^{232} \xrightarrow[1.7\ d]{\beta^-} U^{232} \xrightarrow[30\ yr]{\alpha}$$

I said that the problem in these practice runs is to perfect a technique of bombardment of amounts of 10 mg and less of the oxides (or fluorides or other easily prepared salts) of uranium and thorium so that when this large an amount of another isotope in this region of the periodic table (I have in mind 94^{239}) becomes available, it can be bombarded. After bombardment we will measure the yields of products indicated in the above reactions to determine whether the bombardment was successful. I suggested that Fulbright is to be responsible for getting the targets prepared and the material bombarded. We will then make the necessary chemical separations and measurements to determine the yields. If the subsequent bombardments of 94^{239} lead to the production of isotopes like 95^{239}, 95^{240}, or 94^{240} we might, after establishing their decay properties, look for them in pile material.

Gordon F. Mills, Research Director of the Chemical Process Company in San Francisco, wrote that he will be in Chicago in January and expressed a hope that his security clearance investigation will have proceeded sufficiently by then to permit a thorough discussion of the application of their adsorbents to our problem.

Before I left for Oak Ridge last Sunday, I asked that someone in Dreher's group conduct an experiment to test the suitability of the use of the Bismuth Phosphate Process for further processing of plutonium that has been extracted in the "oxidized" state by ether and then removed from the ether by washing with SO_2 water. Today I reviewed a memo written to me on Monday that gives the results of an experiment performed by Dean. His results indicate that the Bismuth Phosphate Process is readily adaptable for the purpose. A bismuth phosphate precipitate carried 97.5% of the plutonium from the aqueous solution that has been used to wash the plutonium from the ether, and the yield from a subsequent oxidation-reduction cycle was similar to those normally obtained.

12/18/43 (cont.)

Manning and I attended a meeting today in Franck's office to plan our presentations for the Chemistry Information Meeting of the Project Council next Tuesday. I am allotted 10 minutes to speak on our attempts at hydrogen reduction of PuF_4 and perhaps on extraction of plutonium by adsorption, a process being developed for use if the need arises. We also discussed a meeting to be held next Wednesday morning with Greager to determine ways in which separation process development can be accelerated.

Allison sent a memo to Compton referring to General Grove's directive that the U^{233} program should be postponed completely until further notice. Allison admitted that he had been misinformed when he claimed at the Project Council Policy Meeting that three men are working on this problem at the Met Lab, that actually only one man (Katzin) was employed in canning thorium for insertion in the pile. This will now be discontinued. He went on to say that the other chemists who are working on this problem have been moved to the Clinton Laboratories and the decision on their activities should be made by Whitaker and Doan.

All Division Heads received a memo from Allison (MUC-SKA-380) quoting the following directive for operations of the Meb Lab as communicated to him by Compton: "There are two major and two minor objectives for the technical program of the Laboratory. The major objectives are: 1. To make the Hanford water-cooled pile and its bismuth phosphate separation plant work as well as is possible at the earliest possible moment. As far as the Metallurgical Laboratory is concerned, the water-cooled pile is its main responsibility. On the separation plant, the Laboratory needs to carry through further studies of concentration processes, but its major task should be one of obtaining a fundamental understanding of all of the chemistry involved in the separation process. 2. To join with Los Alamos and the associated projects in developing methods for the necessary purification of the project and its reduction to metal.

"Until the work is well in hand, the former of these objectives should have the primary emphasis. The minor objectives are: 1. To undertake certain small scale investigations connected with the offensive use of radioactive materials. This will involve chemical studies for the preparation of these materials in suitable form, biological studies of the effects of such materials, and possible field studies in the actual distribution of such materials. The work will be carried on in conjunction with other branches of the 49 project. A decision as to where the primary responsibility shall be placed has not yet been made, pending further investigation. 2. To aid Los Alamos in carrying through certain physical measurements which can be made more effectively at Chicago than at Los Alamos.

"When broken down into terms of divisional operations, this would mean the following divisional objectives... C. Chemistry 1. Finish studies of concentration methods necessary for Clinton Laboratories to develop complete separation processes for both Clinton and Hanford materials. 2. Do all useful studies on the fundamental chemistry of the B and D processes so that the necessary basic information will be available to assist in interpreting difficulties as they arise at Clinton

and Hanford. 3. Furnish all required information on the behavior of materials and processes under intense radiation. 4. Determine properties of the by-products and their behavior in the processes. 5. Develop methods of purification of the product and its reduction to metal in collaboration with Los Alamos and the associated contracts. 6. Develop analytical procedures for the purification program and carry out necessary analyses."

Today's headlines report the fact that Americans, British, French, and Italians are fighting side by side against the Germans in the mountains of Italy.

Sunday, December 19, 1943

Today's Sun reports that a new Soviet drive is forcing a Nazi retreat. The Germans are retreating before a Soviet offensive that concentrated nearly 300,000 men in a narrow sector, "on the Nevel front in White Russia.

Monday, December 20, 1943

I suggested to James that while Fulbright is in St. Louis working out the techniques for deuteron bombardment of 10-mg samples, we could carry out some neutron bombardments in the Argonne and Clinton piles with small amounts of 94^{239} now available. The idea is to mount 94^{239} samples on counting plates made of a material in which very little radioactivity will be introduced by neutron bombardment (such as beryllium) and irradiate them in the pile. Then, by comparison with identical samples that have not been irradiated, we will attempt to discover whether any alpha-active species are present. I also suggested we isolate some plutonium from uranium metal that has had a long irradiation (several months at highest power) in the Clinton and Argonne piles to look for other isotopes of element 94.

More specifically, the following six experiments are planned.

1. One-day bombardment in the Clinton pile to detect alpha activity with a half-life of the order of a day. Measurements would be made on equipment there as soon after removal from the pile as possible.

2. One-month bombardment in the Clinton pile to detect alpha activity with a period of the order of weeks, months, or a few years. This sample could be sent here for measurements.

3 and 4. Experiments the same as (1) and (2) except in the Argonne pile where the neutron density is higher.

5 and 6. Isolation of plutonium from uranium metal that has had long irradiation (few months at highest power) at Site X (and Argonne in a separate experiment) to see if it has a different specific alpha activity than that which we now consider as correct for 94^{239}. This would test whether or not there is any plutonium alpha activity that has

as its primary origin the capture of neutrons by heavy isotopes in the pile (e.g., 93^{239}) other than 94^{239}. If any of these experiments show new alpha activity, measurements on an alpha range chamber would be valuable for giving confirmatory evidence.

Walter C. Beard who received a B.S. degree from Rollins College, Winter Park, Florida, started work in Section C-I. I plan to have him work on extraction methods for plutonium.

The Clinton Separations Plant started operating today. It is using slugs from the first five tons removed from the pile during the one-week shutdown period which ended December 7.

Franck, Hogness, and I received copies of a memo from Allison authorizing Cooper's Technical Division to proceed with a semiworks program to test the feasibility of the ether extraction counter flow method for recovery of plutonium at Site W. He also authorized the Chemistry Division to carry out incidental work connected with Cooper's project in order to ascertain whether or not a bridge can be formed between the ether extraction and the bismuth phosphate precipitation methods.

William Rubinson of Chemistry Section C-III has reported that Mrs. Mooney of Zachariasen's section has verified by x-ray diffraction measurements that there are two different forms of bismuth phosphate precipitates; the normal hexagonal form, that is acid soluble, and a form with a quite different pattern, that is insoluble in 10 N HNO_3. Additional research on the characteristics of the abnormal form is planned. This observation has caused quite a bit of excitement throughout the Chemistry Division because of the possibility that bismuth phosphate precipitates carrying plutonium in the Bismuth Phosphate Process will convert to the insoluble form leading to a great deal of trouble with the Process. Franck and members of other chemistry sections are much more worried about this than I; there is no evidence from the many laboratory and semiworks experiments that this will pose a problem. Actually, Humbert Morris, of our section, also prepared this new insoluble form of bismuth phosphate, as identified with x-ray diffraction by Mrs. Mooney, last September but did not identify its characteristic insolubility. The insoluble form seems to form somewhat capriciously except under unusual controlled conditions that are not characteristic of the present Bismuth Phosphate Process flowsheet.

At 2:30 p.m. I attended a special meeting in Room 209, Eckhart Hall, with C. A. Thomas on the purification of plutonium. Others present were Allison, Baumbach, Boyd, Chipman, Connick, Davidson, Franck, Dempster, Foote, Fred, Hogness, Jeffries, Jensen, Katz, Kennedy, Kirk, Latimer, Magel, Manning, Orlemann, Potratz, Cyril Smith, Spedding, Warner, W. W. Watson, and Willard. The program was as follows:

<u>Analytical Methods</u>

1. Spectrochemical Developments - M. Fred
2. Mass Spectrograph - A. J. Dempster

3. Chemical Methods and Summary of Analytical Work at Chicago - H. A. Potratz and E. F. Orlemann
4. Summary of Analytical Work at Y - J. Kennedy
5. Analytical Work at Berkeley - R. E. Connick
6. Report on Analytical Standards - H. A. Potratz

Pure Reagents

Procurement and Purification - H. A. Potratz

Purification and Metallurgy

1. Metallurgy at Y - C. Smith
2. Purification by Solvent Extraction - E. F. Orlemann
3. Purification by Precipitation - E. F. Orlemann
4. Volatility Methods for Purification - N. R. Davidson
5. Metallurgy at Ames - F. Spedding
6. Purification in the Metal Production Step - F. Foote and E. F. Orlemann

Orlemann reported on some improvement in chemical methods of analyses for fluorine, sulfur, and phosphorus. In a discussion of the vacuum fusion method for oxygen determination being developed at Site Y, Davidson and I suggested investigating the possibility of trouble from calcium, magnesium, or barium. When Orlemann reviewed work on the purification of plutonium by solvent extraction, he said that about 60% of plutonium can be extracted from the aqueous phase with an equal volume of nitromethane and 70% with 2-ethylbutylcellosolve. These two solvents seem promising, but the behavior of solvents is so specific that it seems advisable to extend the study to many more solvents. Purification by precipitation does not appear as promising as by solvent extraction. In the course of this discussion I referred to an experiment about two months ago that showed the plutonyl state to be fairly stable in ether.

In response to Hogness' statement that the reduced plutonium state sometimes behaves differently in the presence of hydroxylamine, I mentioned that the plutonium is probably reduced from the +4 to the +3 state. In the review of volatility methods for purification, Davidson said that it appears from the data that the slow step in the volatilization of plutonium halide is in the conversion of the oxide to the halide and not in the volatilization of the halide. In a discussion of purification in the metal production step, I suggested that the purification of plutonium metal by direct distillation, after conversion of the crude metal to iodide, followed by thermal decomposition to the metal, appears promising.

The California Project, under Latimer and Eastman as Project Directors, and Connick and Gofman as Group Leaders, issued a report (CN-1170) entitled "Chemical Research--Production and Extraction of 94." The abstract reads as follows:

I. The Extraction of One Milligram of Plutonium from 750 Pounds of Uranium by the Sodium Uranyl Acetate Process. S. Carniglia, R. Connick, O. Cook, H. Crandall, C. Crompton, J. Gofman, J. Hamaker, E. King, M. King, W. McVey, W. Orr, G. Pimentel, W. Reas, N. Reed, A. Stein, G. Sheline, F. Stevenson, A. Stickland, B. Thamer, and L. Zumwalt--A description of the extraction of one milligram of plutonium from 500 pounds of uranium metal and 500 pounds of uranyl nitrate hexahydrate by the sodium uranyl acetate process is given. The estimated overall plutonium yield for three cycles of the $NaUO_2Ac_3$ process is 88%, with a uranium reduction factor of about 2,400. The decontamination factor for gammas (as measured through 2 gms/cm^2 of Pb) averaged 78 for the first cycle, 13.7 for the second cycle, and 7.3 for the third cycle. The overall factor was 7,800 for three cycles.

II. Purification of 380 Micrograms of Plutonium. S. Carniglia, R. Connick, J. Gofman, W. McVey, G. Sheline--The procedure used to purify 380 micrograms of plutonium after three acetate cycles is described. Wet fluoride cycles were used, and an overall decontamination of gammas of ca. 10^7 was achieved, including the decontamination in the acetate cycles. Some properties of a double fluoride of sodium and plutonium and of sodium plutonyl acetate are given.

Brown planned to come from Oak Ridge to attend the Project Council Information Meeting on Chemistry that meets tomorrow morning. Today I received a telegram from him that his train, due in at 7:30 a.m. tomorrow, will arrive five hours late.

I attended the evening seminar of the Chemistry Division at 7:45 p.m. in Room 251, Ryerson Laboratory. Boyd talked about the work of his section on the adsorption process. Connick and T. H. Davies discussed work at Berkeley and Clinton, respectively.

In the drive that the Soviets have started on the Nevel front, they have gained 18 miles in a five-day surge toward the old Polish and Latvian borders.

There was a Project Council Policy Meeting at 9:30 a.m. in Room 209, Eckhart Hall, attended by Allison, Chapman, Chipman, Compton, C. M. Cooper, Dempster, Doan, Fermi, Franck, Greager, Hilberry, Hogness, Hughes, Jeffries, W. C. Johnson, Latimer, Miles, Mulliken, Murphy, Smyth, Spedding, Stearns, Stone, Thomas, Vernon, Warner, W. W. Watson, Whitaker, and Wigner. Compton observed that two years ago today the 49 project started. On December 20, 1941, he wrote a memorandum addressed to Vannevar Bush, James B. Conant, and Lyman J. Briggs outlining a theoretical and experimental program to be centered at the University of Chicago and carried on with the cooperation of the groups at Columbia, Princeton, and Berkeley for the production of 94^{239} for use in a nuclear explosive, through the operation of a chain reaction with natural uranium. Spurred on by Pearl Harbor, he suggested a speeded-up time schedule to obtain knowledge of the conditions for a chain reaction with a schedule for obtaining knowledge of the conditions for a chain reaction by

June 1, 1942, production of a chain reaction by October 1, 1942, a pilot plant for use of the chain reaction to produce 94 by October 1, 1943, and the production of usable quantities of 94 by December 31, 1944.

Returning to the present situation, he said du Pont has advanced the completion date for the first Hanford pile to June 1, 1944, and production on a small scale can be expected during 1944. It had been expected that there would be 12 more months available for process development at Clinton, but this has now been reduced to six months, putting a heavy burden on the Clinton organization. Compton further stated that the Clinton separations plant is due to start on active material today, and milligrams of plutonium can be confidently expected by the first of the year. Microchemical tests at Chicago with Site W concentrations are of great importance. He said we must also press hard in cooperation with Site Y to solve problems of purification and metallurgy of plutonium. In answer to a question by Smyth, Compton replied that the first canyon (chemical extraction plant) unit at Hanford is expected to be ready for tests about June 1 of next year. Vernon reported that the P-9 (heavy-water pile) Section is now largely disbanded. Johnson reported that at Site X the dry fluoride work was terminated during the month, as has been the U^{233} program. Cans of thorium will be left in the pile until April, when some 15 mg of U^{233} will have been produced. Analyses at Site Y indicate that perhaps 2.5 mg of U^{238} also will be present in the neutron-irradiated thorium. Greager reported that four batches of slugs from the Clinton pile were handled during the past week in the semiworks by remote control, and there was about 50% recovery through extraction, one decontamination cycle, crossover, and one concentration step. A 1.2 mg sample of plutonium will be turned over shortly to the chemists. Allison reported on his recent visit to Site Y stating that the present thinking there indicates the possibility that the extreme purity requirements may be later relaxed. For the present, however, all efforts must be made to meet the high purity requirements. The distribution of personnel and problems in my section was reported to be as follows during the past month.

Bismuth phosphate process - 17 men

Alternate Processes - 7 men

Metal Production - 11 men

Purification and Basic Research - 14 men

Concentration - 5 men

Instrument Group - 8 men

Total - 62 men

During the first half of the month 11 men were shifted to the extraction of plutonium from uranium metal and nitrate that was neutron irradiated at the St. Louis cyclotron, and 6 men worked at Clinton Laboratories on the Bismuth Phosphate Process.

Tuesday, December 21, 1943

I received a memo from Orlemann and Brody describing an experiment they have performed for the purpose of establishing the oxidation-reduction potential of plutonium. In the experiment, plutonium in the reduced state was added to one aqueous mercurous-mercuric system and to an identical system in the oxidized state. The air above the liquid was replaced by nitrogen and then the tubes sealed. At the end of one week the amount of reduced plutonium was determined by lanthanum fluoride carrying, and then the total plutonium determined. The two results for the electromotive force E for the plutonium couple in 0.1 M H^+, for $PuO_2^{+2} = Pu^{+4}$, are: greater than or equal to -0.977 and -0.963 volts. These results do not conflict with those reported by Gofman on his work with the bromine-bromide system.

My section prepared a list of information needed preparatory to setting the exact conditions for operation of the Bismuth Phosphate Process at Site W. The list includes problems that will require all types of investigation, including fundamental research, process development, semiworks, and Site X plant operation.

At 9:30 a.m. I attended the Project Council Information Meeting on Chemistry in Room 209, Eckhart Hall. Others present were Allison, Boyd, Burton, Chipman, Compton, Connick, C. M. Cooper, Davies, Dempster, Doan, Fermi, Franck, Garrison, Greager, Hilberry, Hiskey, Hogness, Jeffries, Warren Johnson, Kennedy, Kircher, Latimer, Magel, Mulliken, Murphy, Potratz, Cyril Smith, Smyth, Spedding, Stearns, Stone, Sugarman, Sutton, Thomas, Vernon, Warner, W. W. Watson, Whitaker, and later Brown.

In a discussion of some of the chemical results obtained at Clinton, Johnson mentioned that 40 one-half pound cans of thorium carbonate are in the pile and that 50 more are to be added at the next shutdown. The yield will be 15 mg U^{233} by next April, assuming 500 kw average power. The sample will be 20% contaminated with natural uranium impurity. Johnson also mentioned that the dry fluoride work has been stopped. Sutton stated that they are emphasizing semiworks study of the crossover cycle and of subsequent concentration steps. Greager said the main activity of the semiworks group is to work up material as it comes from the pile, using the Bismuth Phosphate Process. His group will turn over a 1-1/4 mg sample of plutonium to Perlman's section today and will continue operations until he receives a total of 3 mg. At present, yields are about 50%; the losses are probably mainly mechanical losses in various places in equipment although much of the loss has not yet been specifically accounted for. I summarized our work on crossover and concentration steps, our search for better oxidizing and reducing agents, our work on coupling of the adsorption process and Sodium Uranyl Acetate Process to the Bismuth Phosphate Process, and the work on the Bismuth Phosphate Process directed toward both Site X and W. I discussed the essential differences in the concentration problem at Site X and at Site W. I mentioned that purification studies are proceeding along three directions and stated that work on metal reduction is being continued by Cunningham's group. I also mentioned that reduction of PuF_4

with atomic hydrogen has now (without much doubt) been effected and should be useful for making powdered metal. Cyril Smith noted that they have given up trying atomic hydrogen reduction at Site Y.

I received a memo from Thompson discussing proposed work on the Bismuth Phosphate Process as applied to Hanford. In order to obtain maximum assurance that the Bismuth Phosphate Process will be operable at Hanford, Thompson proposes tests on a large scale using "W" concentrations of plutonium and fission products with equipment simulating that to be used at Site W. He also proposes the use of 5 mg of plutonium in a study of the concentration step. For process improvement he gives first priority to the development of alternate oxidizing and reducing agents to give protection against the failure of present reagents and emphasizes the need to develop an alternate concentration process. His memo summarizes many other problems that need investigation and constitutes a good status report and proposed research program.

Someone showed me a copy of the current "Newsweek." After remarking about the Nazi superiority to the Allies in certain kinds of technological development, e.g., rocket-glider-bombs, the article states that "there is the so-called German secret weapon. The propaganda concerning it is not being discounted as usual in Allied capitals, and last week the Nazis unofficially announced that it would be used against Britain within fifteen days." Some of us suspect this announcement might herald the explosion of an atomic bomb!

In a memorandum to all Division and Section Chiefs, Allison warns of the danger that the University of Chicago can become publicly associated with the Clinton Laboratories through the joint approach of our two organizations to companies who supply us with equipment. He cites a case of this type which already has led to one breach of security.

Another memo from Allison (addressed to Franck, Hogness, Szilard, Oppenheimer, and me,) quotes the directives issued by Compton to the Met Lab and Clinton Labs concerning curtailment of the uranium-233 program.

At 2:00 p.m. I attended the Project Council Information Meeting on Technology in Room 209, Eckhart Hall along with Allison, Bellas, Brugman, Chapman, Chipman, C. M. Cooper, Doan, Fermi, Foote, Greager, Greninger, Hiskey, Howe, Jeffries, K. G. Jones, Kidd, Kircher, Latimer, Magel, Mulliken, Seitz, Cyril Smith, Smyth, Spedding, Stone, Sutton, Vernon, Warner, W. W. Watson, Whitaker, and Wigner. Uranium-molybdenum alloys were discussed by Smith. Magel described reduction experiments with uranium on a one-gram scale. Greninger reported on the present status of the uranium slug-jacketing problem; the Al-Si bond appears hopeful.

There was a meeting of the administrative staff at which Munnecke, having recently returned from Washington, reported that the draft deferment situation is becoming difficult and that shortly an order will be issued cancelling deferment for anyone not past his twenty-third birthday, except that such deferment may be granted by the State or National

Director of Selective Service. The mood of the administration here is to stop hiring men who are under age 38. Stearns believes that we should do more toward the training of women to replace men. It was suggested that a survey be made of the male employees under age 23 because wholesale drafting of these employees could cause serious damage to our scientific program.

U.S. forces have captured Arawe air strip on the island of New Britain in New Guinea. There were also gains in Allied campaigns in Italy; U.S. forces have gained two miles in a push toward Cassino.

Wednesday, December 22, 1943

Allison announced by memorandum that Professor Franck has asked to be relieved of his duties as Director of the Division of Chemistry of the Metallurgical Laboratory "because the onerous organizational and administrative responsibilities of the position are endangering his health." Allison stated that Thorfin Hogness has consented to accept this position; so effective January 1, 1944, he will become Division Director. This is good news to me because Hogness has such an excellent chemical background, including a Ph.D. in physical chemistry, obtained in 1921 from the University of California, Berkeley.

There is a good deal of consternation about the recent decision to speed up the Hanford schedule, especially as it concerns the accelerated program of putting the Bismuth Phosphate Process into operation. Today we took advantage of all the people in town and held a meeting from 9:00 a.m. to 12:30 p.m. in Room 209, Eckhart Hall, to discuss and make plans for this speed-up. Among those present were Brown, C. M. Cooper, Davies, Doan, Franck, Greager, Hogness, Warren Johnson, Latimer, Manning, Squires, Sugarman, Sutton, Thompson, Whitaker, Willard, and I. I had prepared a paper, "Chemical Work Needed on Bismuth Phosphate Process Preparatory to Hanford Operations" (MUC-GTS-357), that listed the problems and proposed experiments, under the following outline: I. Metal Dissolving Step, II. Reduction Step Preceding Extraction, III. Extraction Step, IV. Decontamination Steps, V. Bulk Reduction Step, VI. Concentration and Isolation, and VII. General. The discussion was lively and spirited. Greager pointed out that so far all Site X work has been concerned with Site X conditions, but now the Site X work must be allotted partly to Site W problems. He proposed that this allotment should be increased gradually to 90% by June 1. He also believes that a definite increase in process development is needed; Sutton now has 12 men and should have 24. Willard gave detailed information on the manpower assignments in our section, indicating our utilization of manpower. Cooper indicated a need for 12 men in the semiworks group here at the Met Lab. Franck disputed the need for these, asked who they would be and what they would do; Cooper replied that they would be chemical engineers, would work on such things as the concentration procedures which now must have great emphasis. In a discussion of cooling periods at Hanford, the matter of Np^{237} was raised, and I observed that we are not in control of the situation unless we can

separate neptunium (undecayed Np^{239}) from the plutonium during the separation from uranium fission products. I also deplored the lack of systematic work on a complete alternate extraction-decontamination process. Squires stated that emphasis should be put on studies involving Site W concentrations of product and by-products. He indicated that dummy runs should start in the Hanford canyon by the middle of July and mentioned that much testing work can go on at the Site X canyon around June without much sacrifice of production. He emphasized that any alternate process must fit into the plans for the first canyon; the first alternate is the Wet Fluoride Process with the Sodium Uranyl Acetate Process having much lower priority. On the matter of the Hanford canyon construction schedules, Squires indicated that the first canyon will be completed in July; the second, several months later; and the third, six months later. The meeting concluded with much discussion of the philosophy of alternate processes.

As a follow-up to the meeting this morning there was a high-level meeting this afternoon in Franck's office for the purpose of correlating the chemical work of Site X and Chicago, attended by Greagor, Squires, Warren Johnson, Franck, and Hogness. Greager asserted that, in order to carry out the Process Development Section's work at Site X with the shortened schedule, he will need 10 additional highly qualified men. After consulting with me, the group tentatively agreed that I would provide four men to Greager and five would be provided by Perlman. In addition, I would transfer three men to Perlman's section to partially replace those he would lose to Greager and, in turn, I would receive three men from Burton or Spedding. Perlman also would received three Burton-Spedding men. Thus, the net result would be a loss of four men for me and a gain of one man for Perlman. It was the general opinion that all process development (i.e., semiworks and large laboratory scale) work on the Bismuth Phosphate Process should be carried out at Site X in Greagor's section and that process development on concentration be done at Chicago at least for the next few months. With these changes Perlman will not be able to handle his planned scouting work, so some of this will have to be turned over to my section at the Met Lab. Squires also said it will be necessary to place uranium recovery from the Bismuth Phosphate Process at low priority to be considered later after the main problems have been solved.

In a memo to Allison on the probable changes in the research program at Clinton and Chicago resulting from the speed-up in the Hanford program as a whole, C. M. Cooper refers to our meeting this morning and indicates that the following points were not fully appreciated by many of the people present:

1. If the plant is operated with a very short holding-time, the separation of neptunium will be necessary and the problem of disposing of xenon and other radioactive gases becomes of critical importance.

2. The capacity of the separations operation is very much smaller than the designed capacity for dissolving metal. This means that the extraction capacity could probably be considerably increased, perhaps by a factor of three, if a concentration step, such as adsorption or

ether extraction, is inserted in the front end of the system. He emphasizes that no quantitative studies have been made of these possibilities and urges that studies be made of these matters.

I attended the evening meeting of the Purification and Metal Production Groups of my section at 7:45 p.m. in Room 209, Eckhart Hall with Brody, Crawford, Cunningham, Dawson, Dirksen, Florin, Fred, Heath, Hellman, Jaffey, James, Jensen, Katz, Katzin, Manning, Nachtrieb, Orlemann, Patton, Rosenfels, Sheft, Clifford Smith, Watters, Willard, and Zvolner. The following items were considered. Rosenfels discussed the microhardness apparatus and his plans to calibrate it by comparing micro tests of a number of substances with those obtained on a large scale. Zvolner has been attempting electrolysis of one microgran quantities of plutonium from aqueous solutions of five microliters. Results are not reproducible, and experiments are underway on plating the plutonium out on a mercury surface. Dirksen reported that fusion of ThO_2 is not possible with the existing equipment. Nachtrieb reported on a spectroscopic analysis that indicates that a plutonium sample submitted by Cunningham has a purity of 95% or better. Heath summarized work on the plutonium higher fluoride using calcium fluoride tubes that show that revolatilization of the higher fluoride fails at 400°C as well as at 300°C. An experiment with U^{232} tracer shows a uranium distribution very similar to that of plutonium; if this fact is confined, it will make interpretation of the plutonium volatility experiments much more difficult. Cunningham said that his people have been largely concerned with extraction and recovery of plutonium. He reported that Smith and Dixon have volatilized plutonium acetyl acetonate at 175°C and collected the sublimate on an aluminum foil to be used by Crawford for determining the extent of backscattering in alpha counting; it is now thought that the scattering may be multiple and may amount to as much as 1.5%. Various backing materials will be placed under the aluminum foil, and it should be possible to determine the extent of backscattering experimentally. This is a very important experiment and its outcome will affect our half-life for Pu^{239}. I proposed checking these results by the use of the low-geometry counter. In connection with a report by Kohman of recent developments in neutron counting aimed at a detection efficiency of 1%, I explained the importance of neutron counters in the purification program. Jensen and Brody presented their latest work on purification by precipitation reactions and solvent extraction, respectively. I discussed the functions and activities of Warner and Arnold and urged that further requirements for pure reagents be anticipated as much as possible and that requests for reagents be forwarded to Arnold, who has the responsibility of procuring them.

Lt. Col. John R. Ruhoff wrote thanking me for meeting with him, Major Vance, and Dr. Winters on Sunday afternoon, December 12, to discuss the use of resins for the purification of uranium by the adsorption technique.

General George C. Marshall, U.S. Army Chief of Staff, has flown to the New Guinea area to confer with General Douglas MacArthur.

Marshall came from the Cairo and Teheran conferences, stopping for other meetings on the way.

Thursday, December 23, 1943

The first spectographic analysis of a compound of plutonium, the nitrate, prepared by Hindman and Cunningham, shows that the sample has high purity, as Nachtrieb reported last night.

I received a letter from John Dam at Clinton Labs, expressing his extreme unhappiness with the living conditions in Oak Ridge and asking if I would consider rehiring him in Chicago if he should obtain a release from Clinton Laboratories. He considers the work and the working conditions very satisfactory but is very concerned that the delay in completion of his house will prevent his wife from joining him until March. He also indicated that the food in Oak Ridge disagrees with him in every way.

Allison wrote a memo giving the latest schedule of plutonium deliveries to Chicago and Los Alamos from Clinton Laboratories starting next month:

Date	Amount Available	To Site Y	To Met Lab
Jan. 1	2 mg		2 mg
Jan. 8	2 mg	2 mg	
Jan. 15	20 mg		20 mg
Jan. 25	1 gram	1 gram	
Early Feb.	2-1/2 grams	1-1/2 grams	1 gram
March 1	15 grams	14 grams	1 gram

I attended a meeting with Bellas, Kircher, Christiansen, Thompson, Willard, and Dreher to discuss the program for operation of the Chicago semiworks. The long-range program will remain unchanged. It was decided that about 250 pounds of Clinton neutron-irradiated uranium metal per month will be needed for the operations. The metal should have 3,000 kwh/ton and should have an activity roughly 10 times that of the hottest St. Louis bombardments and one-tenth that of the average material put through the Clinton chemical separation plant. A detailed program for the next two weeks was planned. In this program U^{+4} will be used as the reducing agent in work on both the prereduction stage and the decontamination cycle; otherwise, extraction will be in accordance with the present flowsheet. The HCl-uranous oxalate concentration method will be tested. Approximately 100 pounds of Clinton pile metal has been received. Half of this material, which was originally scheduled to be used for 93^{237} recovery, will be held until Cunningham's present 93^{237} work has been completed so that a process for the isolation of 93^{237} can be planned. The other 50 pounds of metal will be used for semiworks runs as soon as two practice runs have been completed on Argonne pile neutron-irradiated uranium metal solution. It was estimated that one week will be required

for the practice runs, and that the following week should complete the runs involving the Clinton metal solution.

Jaffey and Davidson summarized for me, by memo, the experiments of Jaffey pertaining to volatility work during the period, October 1942 to March 1943. This describes preliminary experiments on plutonium tracer in U_3O_8, experiments on volatilization of tracer quantities of fission products and UX_1, halogenation of bismuth phosphate precipitates, chlorination and bromination of uranium samples, halogenation of tracer and "massive" quantities of plutonium, and tracer quantities of neptunium.

Johnson in Personnel wrote to E. J. Mathews, Dean of Admissions of the University of Texas in Austin, at Cunningham's behest. He asked that Roy G. Post and Mathew T. Walling, Jr., two students who will be graduated in February, be allowed to come here immediately thereafter to engage in war research. He quoted George Watt, who joined us from the University of Texas, as saying that the work the men would do here might apply toward an advanced degree at the University of Texas.

The evening meeting of Section C-I Research Assistants was held at 6:45 in Room 209, Eckhart Hall.

I held an evening meeting of the Council of my section at 7:45 in my office, attended by Beaton, Cunningham, Davidson, Dreher, Ghiorso, Kirk, Manning, Thompson, and Willard. I informed the group of the schedule of plutonium deliveries just received from Allison. Of the first two milligrams to come into our section in January, about 1.9 mg will be assigned to the metal production group. The recovery group will attempt to keep the material turning over as rapidly as possible. Dawson will keep complete written records of the material coming into and going out of the recovery group. Plutonium will be issued only to group leaders. I raised the question of holding periodic meetings of individual groups as currently done by Thompson and as has been done by Cunningham until recently. I suggested it would be desirable for all but possibly the smallest groups to hold meetings of this type in order to take advantage of the combined thinking on the various problems in the groups. Early morning might be the best time for such meetings. I indicated that Hogness, the new director of the Met Lab Chemistry Division, might attend a Thursday night meeting of our Section C-I Council, in which case we will give him a comprehensive review of the work going on in our section. On the subject of personnel, Meyer will be shifted to Kirk's group to handle final preparation of material for metal production. Hellman will spend part of his time cooperating with Meyer. They will work in Room 1. S. Katz and Jasaitis (due to arrive early in January) will join Kirk's group the first week in January (S. Katz may work on the electrolysis problem as he has some experience in metallurgy). Anderson probably will join Davidson's group soon. Seven men from Thompson's group will be transferred to Clinton in the near future; two of them, Creek and Dean, have already been scheduled for transfer. On the subject of apparatus, equipment for the quartz microbalances is gradually arriving. The instruments group is actively working on the fission chamber and also on the range chambers. Davidson

suggested that it might be desirable to use a neutron source to search for lost plutonium by means of the induced radioactivity of fission products as a result of neutron bombardments. Experiments in the instrument group to test backscattering of alpha particles were discussed. Ghiorso pointed out that the plan to measure the counting rate of material deposited on a thin aluminum foil with and without a platinum backing would not give quantitative results. He suggested that a comparison of the specific activity of samples mounted on quartz and on platinum discs would give a more quantitative measure. Cunningham stated that Connick in Berkeley has found conditions where SO_2 does not reduce plutonium rapidly, and that his own group has encountered trouble of this sort after a Ce^{+4} oxidation. Tests will be made soon for a +5 oxidation state of plutonium. The Berkeley sample that is being worked up for Np^{237} is now in solution after the first LaF_3 precipitation. It was necessary to carry out a nitrate-carbonate fusion to get the material into solution. Davidson will prepare a small sample of UF_6 rich in U^{235} for the Bureau of Standards.

The temperature dropped to -2°F last night. Predictions are for a high of 15° tonight but with higher temperatures tomorrow, possibly reaching 30°.

Friday, December 24, 1943

At my request, Vernon asked the Manhattan District Area Engineer to obtain a sample of about one pound of uranium nitrate from each of our suppliers. We want this material in order to check some discrepancies in our alpha counting data, which may be caused either by the source or by the processing.

This afternoon the members of Section C-I had a Christmas party in Room A-7 of the New Chemistry Building Annex. Milton Burton, chief of Section C-II, joined us. Someone brought a phonograph, several people brought records, and our secretaries were in much demand as dancing partners. Edrey Smith, my secretary, was a particularly popular choice. Some of the fellows, and Art Jaffey were certainly in this category, displayed special talents in the dancing department. (See Figures 20-32.)

According to the Press Service, President Roosevelt moved tonight to seize the nation's railroads, should such action become necessary to prevent a nationwide strike scheduled for December 30.

Saturday, December 25, 1943

Helen and I spent a quiet Christmas at home.

President Roosevelt announced today that plans for a "gigantic attack upon Germany" were completed at the Teheran conference. General Eisenhower will command the Allied invasion.

Sunday, December 26, 1943

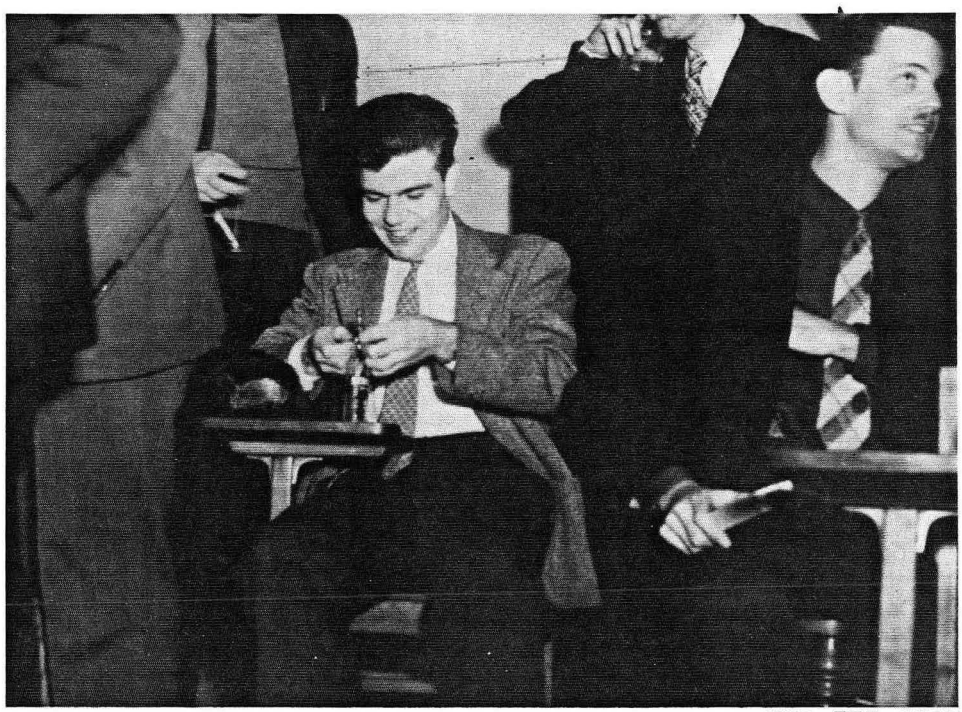
Helen and I went to Wrigley Field and saw the NFL Championship football game between the Chicago Bears and the Washington Redskins, won by the Bears

XBB 777-6698

Figure 20. Section C-I Christmas Party, December 24, 1943. Kay Gavin, Bob Young, Emil Fulan, Al Ghiorso, Elizabeth Wallerstein.

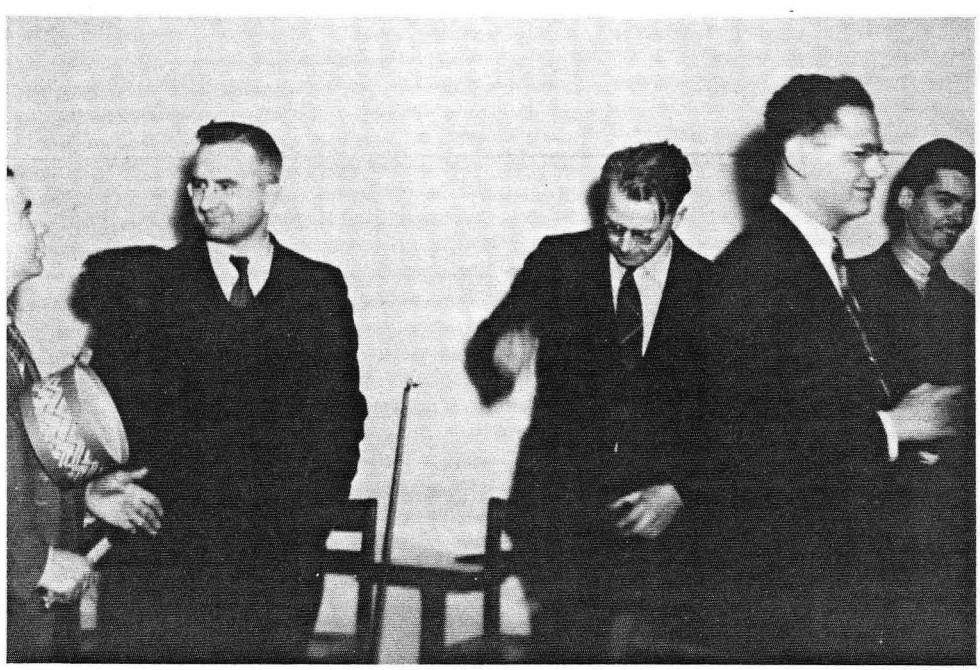
XBB 777-6696

Figure 21. Winston Manning, Seaborg (back), Irving Sheft, Leonard Katzin, Edrey Smith, and others.



XBB 777-6697

Figure 22. Ralph James, Clark Hindman (partly hidden), Lyle Jensen, and others.



XBB 777-6695

Figure 23. Roy Heath, Winston Manning, Bob Patton, Paul Fields, and others.

XBB 763-7227

Figure 24. John Willard, Bob Young, Emil Fulan, Al Ghiorso.

XBB 763-7226

Figure 25. Fowler Yett, Walter Beard, Pat Walsh, Martin Studier (behind Walsh), Art Jaffey, Lorraine Golden, Bert Schoenberg, Truman Kohman, Al Florin, and others.

XBB 761-7435A

Figure 26. Seaborg, Milton Burton, Pat Walsh, Al Kline.

XBB 777-6699

Figure 27. Beatrice Foreman (foreground), Al Ghiorso, Ted La Chapelle?, Dwight Lincoln, Irv Sheft, Martin Studier (center), Asher Margolis, Truman Kohman, Al Florin, Norm Davidson, Ben Scott (partly hidden), Len Dreher (foreground, head), Roy Heath (foreground, back to camera), Joe Katz, and others.

XBB 777-7243

Figure 28. Lorraine Golden, Bert Schoenberg, Edrey Smith, Al Florin, Winston Manning, Roy Beaton, Lyle Dawson, John Willard (mostly cut off) and others.

XBB 777-7241

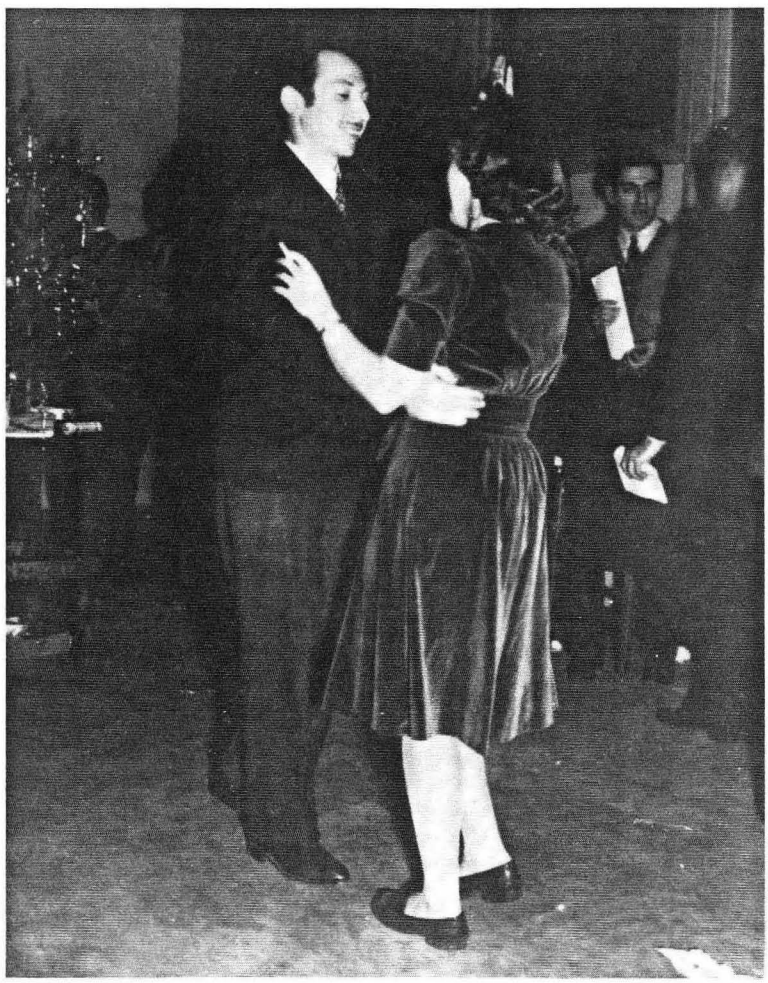
Figure 29. Clark Hindman (partly hidden), Joe Katz, Irma Saxton, Martin Studier (foreground), George Creek, Art Jaffey, Bea Foreman, Bob Young, Mel Freedman (at door), Kay Gavin, Paul Fields, and others.

XBB 777-6242

Figure 30. Harlan Baumbach (partly cut off), Asher Margolis, Norman Davidson, Joe Katz (back), Walter Beard, John Willard, Roy Beaton, Roy Heath (back), Bert Schoenberg (back), Truman Kohman, Leonard Katzin, Bob Patton, Al Florin, Bernard Brody, Lorraine Golden, Dick Rosenfels, Seaborg, and others.

XBB 777-7240

Figure 31. Winston Manning, Al Kline, Beatrice Foreman (back), Truman Kohman (back), Edrey Smith, Pat Walsh, Bob Young (back), Kay Gavin, and others.



XBB 777-6700

Figure 32. Al Ghiorso, Art Jaffey, Pat Walsh, Joe Halperin, Lyle Dawson (back), and others.

41-21. We bought the tickets well in advance and were afraid the weather might be cold, but it turned out to be a very nice day.

"Bitterest Battle in Italy" is the eight-column headline in today's Sun; Germans are fighting for every building. The Japanese are hinting at a second invasion by the Allies on New Britain, but there has been no confirmation by Allied sources.

Monday, December 27, 1943

Cunningham and Florin finished the conversion of 0.24 mg of plutonium to the fluoride by treatment of the oxide with anhydrous HF and turned the sample over to Kirk for metal production.

I wired Albaugh in Inglewood, California, that I am anxious to know how soon he will be coming.

In response to the letter of December 15 from Mills of the Chemical Process Company in San Francisco, I said that he has been cleared with respect to our project and that we will be glad to discuss the application of his adsorbents to our problem, when he is in Chicago next month. I suggested a meeting on the morning of Tuesday, January 11, with Beaton and Willard participating.

Beaton wrote Shoemaker of Research Products Corporation, giving the results of the preliminary testing of various titanium zeolites prepared by the Company. He indicated the best zeolite seems to be one made by briquetting Sherwin-Williams titania and 5% Aratone at 15,000 p.s.i., calcining is at 1,000°C for 45 minutes, and using it at a pH of 3.5.

I have organized a group of chemists in my section who have the special task of recovering plutonium from the various waste solutions to make it available for re-use. In fact the plutonium is being recycled over and over again to get maximum use of our limited supply. I have asked Lyle Dawson to serve as leader of the Recovery Group. With the impending arrival of milligram amounts of plutonium from Clinton Labs at the end of this month, this group will play an increasingly important role. They met this afternoon to make plans for the arrival of the product from Clinton.

I informed Hogness of the seven men we have decided to send to Site X: Dean, Creek, Gaarder, G. Johnson, Kroner, Halperin, and Schuler.

The evening seminar of the Chemistry Division was held at 7:45 p.m. in Room 251, Ryerson Laboratory.

The British have sunk the German battleship Scharnhorst, off North Cape in the Arctic Ocean north of Finland.

Tuesday, December 28, 1943

I received a memo from Hogness saying that a directive has been received from the Laboratory Director to the effect that Beaton is to be transferred from the Met Lab to Clinton in about two weeks. He also sent me a copy of a memo

informing Allison that I have made arrangements for the following seven men, all from Thompson's group, to transfer to Site X: Dean and Creek (the only married men), Gaarder, G. Johnson, Kroner, Halperin, and Schuler.

As a follow-up to the information given at the meeting last Tuesday the administrative staff was informed at today's meeting that after February 1, 1944, all deferments by the Army will be canceled for men not past their 22nd birthday, after which time deferments will be granted only by the State or National Director of Selective Service. A Met Lab survey shows there are 20 scientific staff members who were born during 1921 or later.

The "Monthly Report on the Activities of the Chemical Division, December 20, 1943" (MUC-JF-114) is being issued by Franck. Activities of Section C-I are reported as follows:

	Manpower		
	This Month	Next Month	Priority
I. Separation, Decontamination, and Concentration			
A. Bismuth Phosphate Process			
The isolation of plutonium from 700 pounds of neutron-bombarded nitrate is described in detail as example (CN-1153). The use of columbium and zirconium and cerium as scavengers improves the decontamination 10-20 fold or more if used in combination.	11	0	A-1
Use of lanthanum-yttrium and ruthenium as holdback carriers in product precipitation did not improve decontamination. (Cooperation with C-IV and T-I) (CN-1153).	2	2	A-1
Microchemical tests of each step of the $BiPO_4$ process are made with W concentration of (inert) fission products (CN-1153).	1	1	A-1
The effect of zinc, tin, and silicon (materials proposed for coating) on the yield of $BiPO_4$ process is being tested. Silica and tin may have to be removed before dissolving uranium (CN-1153).	1	1	A-1
S. G. Thompson and five members of his group have worked for one month with Perlman's section at Clinton on the $BiPO_4$ process (No report).	6	0	A-1

	This Month	Next Month	Priority
Recovery of plutonium from residues and separation from carrier (No report).	0	6	A-1
In addition to manpower listed above, work on $BiPO_4$ and on general research on extraction and decontamination (No report).	0	7	A-1
B. Wet Fluoride Process			
Report of corrosion data on wet fluoride process solutions taken from a letter from J. A. Collins (du Pont Company) (CN-1153).	0	0	-
C. Alternates			
Separation of neptunium from plutonium can be achieved by thorium molybdate precipitation (CN-1153).	1	1	A-1
The γ activity remaining in Amberlite IR-1 after an extraction cycle can be reduced to 15% by the use of 1 M H_3PO_4 as an elutriant. Studies continued on titanium zeolite adsorbent for decontamination. (No report) Results on coupling of adsorption extraction to the $BiPO_4$ process look promising (CN-1153).	5	6	B-1
II. Metal Production			
The preparation of plutonium compounds for metal production occupied the work of two men. A review of the procedure is published in CK-1145.	2	2	A-1

	This Month	Next Month	Priority
Finally divided plutonium metal is needed for x-ray studies. Reduction with lithium vapor at lower temperatures looks promising but the difficulty of reoxidization is not overcome (CK-1145).	3	3	A-1
Mounting of samples for x-ray work in lucite needles and glass capillaries gives good distribution of samples for x-ray examination (CK-1145).	1	1	A-1
Formation of plutonium hydride was observed by using the technique tested for uranium and thorium hydrides. According to one quantitative experiment the formula would be $PuH_{2.97}$ (CK-1145).	1	1	A-1
Experiments on reduction of UF_3 and plutonium fluoride by atomic hydrogen give encouraging results (CK-1145).	1	1	A-1
Studies continue on microelectrolytic metal production from aqueous and organic solutions (CK-1145).	1	1	A-1
Other work on metal production not reported: preparation of thorium hydride for x-ray studies and experimentation with refractories (No report).	2	2	A-1
III. <u>Purification</u> (CK-1145).			
Studies on volatilization of plutonium bromide for use in purification work.	2	2	A-1
Fluorine atoms produced by a glow discharge convert UF_4, UCl_4, and uranium oxides to the volatile compound UF_6, that might be useful in preparing volatile higher fluorides of plutonium (CK-1145).	1	1	A-1

	This Month	Next Month	Priority
The reaction of uranium oxides with CCl_4 to form $COCl_2$ was studied as preparation for the corresponding reaction with PuO_2 which may be used for determination of the oxygen content of plutonium. The reactions of UO_2, U_3O_8, and UC_2 and uranium metal with Br_2 are being studied to obtain information as to whether the bromide volatility method may be applicable for O_2 determination in metal (No report).	1	1	A-1
0.1 microgram of chloride can be measured with 10% accuracy by the Conway microdiffusion cells (No report).	1	1	A-1
To determine carbon in uranium, the method was improved which consists of combustion of the sample, followed by the determination of CO_2 by a conductivity method (No report).	1	1	A-1
A number of organic liquids have been tried as solvents for reduced and oxidized plutonium: nitromethane, nitroethane, and 2 ethylbutylcellosolve look promising for purification of plutonium from light elements (No report).	1	1	A-1
Promising precipitants for separation of plutonium from light elements are: molybdate, sulfite, sulfate periodate, fumarate, and picrolonate (No report).	1	1	A-1
Experiments on the volatility of the higher plutonium fluoride continue (No report).	1	1	A-1

	This Month	Next Month	Priority
The stability of plutonyl plutonium in ether was tested. (No report)	½	0	A-1
Other work on purification and basic research on plutonium.	½	0	A-1/ C-1
IV. Concentration			
Ceric ions dissolve LaF_3 and CeF_3 carriers at room temperature. That may be advantageous in concentration work if the use of steel equipment does not interfere. CeF_3 is a very good carrier of plutonium at all concentrations and is easier to purify than LaF_3 (CN-1153).	1	1	A-1
Work on flowsheet for separation of plutonium from carriers is in progress (No report).	3	4	A-1
Instrument group (No report)	8	8	A-1
U^{233} (CK-1145).	1	0	B-3

The army took possession of the railroads last night on orders from President Roosevelt, according to this morning's paper.

Wednesday, December 29, 1943

Donald G. Pye started work in Section C-I. He received the Ph.D. degree this year from Stanford University in Palo Alto, California. I intend to have him work on the Hanford Process for the concentration and isolation of plutonium.

Watt suggested by memo that we adopt the following definition for concentration factor: "In any series of operations in a wet concentration process, the concentration factor is the ratio of the volume of any product-containing solution to that of any succeeding product-containing solution."

I attended an evening meeting of the extraction groups of my section at 7:45 in Room 209, Eckhart Hall. I summarized the status of the extraction plant at Site X, mentioning that diluted material is now being run through the plant and that the plutonium from the crossover

step in the Bismuth Phosphate Process has already been slurried into Room D. The yield was 55%, and a decontamination factor of 300 was attained after one bismuth phosphate cycle. Watt discussed five concentration methods that appear to be workable at Site X, three of which depend upon solution of the bismuth phosphate in HNO_3 and two of which use HCl to dissolve the bismuth phosphate. It was pointed out that while HCl gives a concentration factor of 43 compared to 5 for HNO_3, the engineers dislike methods using HCl because of corrosion. Watt outlined the concentration and isolation method contemplated for Site W--the bismuth phosphate-plutonium precipitate from the "N"th decontamination cycle will be dissolved, the plutonium oxidized, by-product bismuth phosphate and lanthanum fluoride precipitated and removed, the plutonium reduced and co-precipitated with lanthanum fluoride, the precipitate metathesized to the hydroxide, dissolved in 8 gallons of HNO_3, the plutonium precipitated directly as the pure peroxide. In a discussion of possible alternates to the metathesis step, I mentioned there is a question as to whether the peroxide or oxalate can be precipitated quantitatively from bismuth phosphate-hydrochloric acid solution and cited Fries's and Werner's difficulties in work along these lines. Adsorption methods were discussed by Beaton: extraction and decontamination, and the coupling of adsorption and the Bismuth Phosphate Process. Some work on the latter has been completed by Dean and La Chapelle, a process that is proposed for an alternate to the Bismuth Phosphate Process at Site X. It was mentioned that it might be more feasible to couple the adsorption extraction step with some other process such as sodium uranyl acetate or lanthanum fluoride in order to be sure of having a complete alternate process. Dreher reported on the program of the semiworks group; they have 100 pounds of metal from Site X, supposedly of concentration 100 micrograms of plutonium per pound, plan to get HCl-resistant, rubber-lined, equipment to study alternate concentration methods involving solution of bismuth phosphate in HCl. Thompson identified his two major problems as decontamination at Clinton and carrying at Hanford concentrations. At Clinton, a decontamination factor of 5,000 is necessary before the product should be taken to Room D. The quickest way to shorten the process is to have better decontamination, perhaps by the use of scavengers and hold-back carriers. In this connection, I remarked that Cb_2O_5 and $Ce_3(PO_4)_4$ are probably the scavengers that will be incorporated at Site X. I mentioned that seven of the very best men, Halperin, Schuler, Kroner, Gaarder, Johnson, Dean, and Creek, are soon moving to Site X.

In accordance with our discussion last week, Ralph James prepared counting disks of beryllium containing a few thousand counts per minute of 94^{239} that he will hand-carry to Clinton early next month for neutron-irradiation in the pile there. This represents the first two of the six experiments we talked about. The disks are small enough to place in the aluminum capsules that fit into the stringers of the pile and were first surveyed to make sure they were free of alpha and beta contamination. Plutonium was obtained from Cunningham, which he reports is free of carrier, having been precipitated as the peroxide, dissolved, and then reprecipitated. This solution of plutonium was then pipetted onto the beryllium disks. Finally, James made a careful measurement of the alpha counting rate for each of these samples.

I was one of several addressees receiving a memo from Hogness asking for comments on the suitability of three different coating procedures for uranium slugs as to (1) effect on k, (2) ease of solubility in the dissolver stage of extraction, (3) interference of coating materials in any stage of the chemical operations at Site X or Site W, and (4) hazards from any long-lived activities produced in coating materials.

Thursday, December 30, 1943

Coryell wrote to me on Clinton Laboratories stationery, with the address: "P.O. Box 1991, Knoxville 11, Tennessee." He asked if I would send him ten or twenty mimeographed copies of my Chemical Reviews article of August 1940, and thirty or forty sets of the notes covering the lecture series I gave in May, June, and July of 1942, saying, "There have been so many men coming in in the various sections who have to contribute to the process and Project without having much chance of getting any background that I am anxious to carry on a side job as educator to increase their efficiency."

The Research Assistants of my section held their regular evening meeting at 6:45 p.m. in Room 209, Eckhart Hall.

This morning's paper says that the Soviet army has surged to within 48 miles of the pre-war Polish border west of Kiev in a breakthrough along a 110-mile arc.

Friday, December 31, 1943

I told James that as long as he is going to irradiate samples of plutonium in the pile, he might as well try some uranium samples as well. The possibility exists that detectable amounts of U^{236} might be produced by neutron bombardment of the U^{235} that is present in natural uranium. If it is assumed that the capture of neutrons by U^{235} is only 10% as great as the fission cross section and the half-life of U^{236} is 1,000 years or less, but not too short, then its presence could be detected. James agreed and proceeded to prepare uranium samples (approximately 0.1 mg) by evaporating uranyl nitrate solution on platinum and beryllium disks with tetraethylene glycol as a wetting agent and a zapon ring to prevent flowing off the counting disk.

In a memo to Hogness I suggest that Burton have some tests made to determine the effects of radiation on uranous sulfate which is being considered as a reducing agent for the pre-reduction step and the decontamination cycles of the Bismuth Phosphate Process.

I commented, by memo, to Allison on the effect on the Bismuth Phosphate Process of three proposed coating materials for uranium slugs described in Hogness' December 28 memo, i.e., solder bonding, aluminum

plus 12% silicon over chromium electroplate, zinc over nickel electroplate. I indicated that the last named, which avoids the use of tin and silica, seems to be the most desirable from the standpoint of possible effects on the chemical separation process.

Hogness told Hilberry that the requirements of my section for hot slugs from Site X are 250 pounds per month for operation of the semiworks and for other experimental work. The metal should contain 30 to 40 micrograms of plutonium per pound and should be cooled at least two weeks before shipment to Chicago. My estimate is based on the fact that in the normal operation of the semiworks about 30 pounds of uranium will be used in each run, there being about two complete runs each week.

Helen and I ate New Year's Eve dinner with Stan and Alice Thompson in their apartment.

The year ends with all Allied armies making advances. The Soviets, for example, captured more than 300 communities yesterday, making 1,300 captured in seven days. They are plunging forward on a front 115 miles long.

Saturday, January 1, 1944

The beginning of 1944 finds our Project deep in the problems of plutonium production, extraction, and purification. This vast involvement with a secret, synthetic element unheard of not much longer than two years ago and unseen until sixteen months ago in August 1942, would seem incredible to the outside world. Moreover, the means of producing plutonium in copious quantities--the chain-reacting pile--became operational just one year ago. I thought about these matters today when we received our first shipment of plutonium from Clinton Laboratories--1,500 micrograms! It equals almost the total amount of plutonium produced by all previous cyclotron bombardments. It is hard for me to remain nonchalant when I realize that before the end of February, production of plutonium will increase more than a thousandfold and gram quantities will then become available.

Jerome Karle's wife Isabella started work in Section C-I today. L. O. Brockway of the University of Michigan wrote on November 19 that she and her husband had made the best records there of any recent graduates in chemistry. Those who know Mrs. Karle praise her as "brilliant" and having a "most pleasing personality." She expects to get her Ph.D. this year.

Effective today, Hogness becomes Director of the Chemistry Division; Franck has stepped down to Associate Director for health reasons.

We have been making comparisons between the Geiger counters at the Monsanto Chemical Company's polonium laboratory in Dayton, Ohio, at the Los Alamos Laboratory, and at our laboratory with respect to the geometrical efficiency of counting beta particles from a RaD-RaE-Po^{210} source. Today I sent James Lum of Monsanto Ghiorso's comments on the results obtained here by Ben Scott from specific activity measurements on the radio-lead dioxide beta source. These results were compared with the results reported by Monsanto and by Los Alamos. I pointed out that backscattering could contribute significantly to errors in the measurements and that I feel the results obtained at Los Alamos (2800 d/m/mg) are probably the most nearly correct of the three. I mentioned we are looking forward to receiving the polonium samples Lum promised to send me in a telephone conversation earlier this week.

Thompson wrote Iz Perlman at Site X attaching a brief literature survey on lanthanum phosphates made by Watt and Hopkins. Thompson stated there is very little information on the subject and that most of the work was done between 1840 and 1890. He passed along information on work done in Section C-I that indicates that lanthanum phosphate, rather than lanthanum hydroxide, is the precipitate formed when lanthanum is added to reduced nitric acid solutions after bismuth phosphate by-product precipitation and the solution then neutralized with a base. He suggested that decontamination in the Bismuth Phosphate Process could be improved by using a higher phosphate ion concentration for all product precipitations.

I read a copy of Charles Cooper's letter to Vernon which presented requirements for uranium-bearing plutonium from the Clinton pile for chemical studies by our semiworks group and referred to the high gamma dose rates experienced with the recent shipments from Clinton. Cooper suggests that future material of this nature be cooled at least two weeks before shipment and that the radiation levels at the surface of an outer shipping container be no more than 15 mr/hr when containing four slugs. He also requests that the plutonium concentration in future shipments be as much above 30 micrograms per pound as possible. This seems like a reasonable request because slugs from the periphery of the Clinton pile, which have been in place since startup, should have product concentrations exceeding 100 micrograms per pound and should give acceptably low radiation levels after cooling. About 250 pounds of "X" slugs are estimated to be required by the Chicago semiworks per month. This will represent about five or six four-slug containers per week to be trucked from Site X. Cooper estimates that the first of these shipments should reach Chicago within ten days.

Willard received a copy of a letter from W. W. Johnson, Personnel, to Herbert H. Anderson. Anderson has returned to his home in Auburndale, Massachusetts, from Site X after having his clearance questioned. That problem has been cleared up, but a recent telegram from Anderson indicates that he is being inducted into the Armed Services. Johnson requested information from Anderson so that the induction could be halted, or so that arrangements could be made to obtain an immediate release from the Armed Services. Johnson said every effort will be made to secure Anderson's return to the Metallurgical Laboratory.

Helen and I played cards with the Ghiorsos tonight.

To start out the year come reports that the Soviets have captured Zhitomir, that the U.S. has bombed Paris, and that Allies have landed behind the Nazi lines in Italy, near Minturno.

Sunday, January 2, 1944

The top headlines in today's Sun read "Reds Gain 9 More Miles in Drive toward Poland"--"Attack Perils Nazis Rail Links."

Helen and I spent the entire day at home.

Monday, January 3, 1944

Zene V. Jasaitis started working in Section C-I. He is on a year's leave of absence from the Shell Oil Company in Wilmington, California. Zene was born in Lithuania, naturalized here in 1929, and is another of the chemistry majors who attended U.C.L.A. when I did. He is married to a charming girl, Celeste Major, who also majored in chemistry at U.C.L.A. and was one of our most popular fellow students. They are expecting the birth of a child and therefore have decided that Celeste will remain at home during the time Zene will be with us. Because of his extraordinary

skill in working on a small scale--he is an expert amateur repairer and builder of watches--I shall have him work with the plutonium metal production group to measure some of the metal's physical properties, such as hardness, ductility, etc., on the microgram scale. In a letter of recommendation we received a month ago, William G. Young, Chairman of the Department of Chemistry, U.C.L.A., said of him, "We have never had a student...who possessed more ingenuity for experimental manipulation."

Another technician, R. S. Dreyfuss, was hired to work for Donlan.

Baumbach summarized for me the data on the crystal structure and calculated density values for the tetrafluoride, oxide, and metallic forms of thorium, uranium, and plutonium. The data were derived from samples we supplied over the past few months to Zachariasen. The data show that PuF_4 has a density of 9.93 gm/cm^3. The crystal structure of this compound is still in question, however; should it be monoclinic, the estimated density for PuF_4 would be 7.3 gm/cm^3. On the other hand, PuO_2 has a cubic structure and a density value of 11.54 gm/cm^3. The "unknown material" thought to be plutonium metal, made by low temperature, slow reduction of PuF_4 with lithium and barium, gives a calculated density value of 13.01 gm/cm^3. Should the material be PuO_2 or PuO, however, the calculated densities would be 13.4 gm/cm^3 and 13.9 gm/cm^3 respectively. We desperately hope this is not plutonium metal because such a low density would correspond to such a large critical mass for a plutonium weapon that we would need two Hanford production sites to meet our production objectives.

R. S. Stone transmitted a memorandum from Nickson concerning the possibility for excessive gamma radiation exposures to the whole body and hands of those operating the extraction and purification facilities for cyclotron neutron-bombarded material in Room 10 of my section and in the Cyclotron Chemistry Laboratory in the Service Building. The need for improved shielding and handling equipment was stressed in order to keep the whole body exposure to less than 0.1 r per day.

Willard supplied Johnson with information justifying the need to rehire Herbert H. Anderson. Anderson is a specialist in fluorine chemistry. He joined our staff on July 15, 1943, to work with Brown's group and in September was transferred to Site X. Willard mentioned that the type of work in which Anderson is a specialist has been discontinued at Site X but is continuing with a need for additional men here on our section. He pointed out that failure to rehire a man under the circumstances of Anderson's case might have a very undesirable morale effect on other members of the Project.

I received a copy of a reply written by C. M. Cooper to L. B. Borst, in response to an earlier letter from Borst concerning the shipment of active uranium slugs from Site X to Chicago for use by our semiworks group. Borst had commented that the shipping containers used appeared to be unsatisfactory from the standpoint of protection and ease of handling. Cooper's letter points out that the material shipped had several times more activity than had been estimated would be the maximum level at the time shipping containers were designed. At that time 10 micrograms

of plutonium per pound of crude product was considered to be the highest concentration desirable for work within the Chicago semiworks. The material shipped in the twelve shipping containers (four slugs per container) that recently arrived from Site X had about twelve times the design level of radiation at time of shipment. The measured dose-rate at the surface of one of the twelve containers amounts to about 60 mr per hour, or a surface dose of about one-half roentgen per 8-hour day.

Cooper said that for future shipments we would like to keep the level of gamma-ray activity below about one-fourth that of the recent shipment, but the plutonium concentration should be as much above 30 micrograms per pound as possible. He suggested maintaining a stock of materials from the outer periphery of the Clinton pile. The need for small, easily handled shipping containers is defended as opposed to Borst's suggestion for a single, massive container. At the suggestion of Miles Leverett, lugs are to be attached to the sides of the existing containers so that a long rod can be inserted which will permit up to four men to carry the containers if desired.

I presented my suggested reorganization of Section C-I to Hogness. This is in recognition of the broadening responsibilities of Section C-I and features a division into three sub-sections, each under an Assistant Section Chief. In addition Winston Manning, who has proved to have excellent leadership qualities, will join John Willard as a second Associate Section Chief to help me with the overall direction of the activities of Section C-I; Manning will have some special concern with the work of Orlemann's sub-section, and Willard will place emphasis on the work of Thompson's sub-section. I will give special attention to Cunningham's sub-section although I will continue to watch over the work of the entire Section C-I. I now plan to rotate our Wednesday evening Section C-I research meeting among the three sub-sections and to have the three Assistant Section Chiefs assume the responsibility of planning and conducting these meetings.

The proposed organization chart is shown in Figure 33. This reorganization involves the following changes in titles:

W. M. Manning, from Assistant to Associate Section Chief

S. G. Thompson, from Group Leader to Asst. Section Chief

E. F. Orlemann, " " " " " " "

B. B. Cunningham, " " " " " " "

F. W. Albaugh, new appointment, to Group Leader

G. W. Watt, to Group Leader

J. L. Dreher, to Group Leader

L. R. Dawson, to Group Leader

H. L. Baumbach, to Assistant Group Leader*

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*(To assume full group leadership on Kirk's transfer to Section C-IV)

SECTION C-I, CHEMISTRY OF FINAL PRODUCTS

G. T. Seaborg, Section Chief
J. E. Willard and W. M. Manning, Associate Section Chiefs

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Extraction, Decontamination, and Concentration
S. G. Thompson, Assistant Section Chief

Extraction and Decontamination
F. W. Albaugh, Group Leader

Concentration
G. W. Watt, Group Leader

Process Development
(Cooperation with Semiworks)
J. L. Dreher, Group Leader

• • • •

Purification and Metal Production
E. F. Orlemann, Assistant Section Chief

General Methods and Cooperation
in Analysis Development
E. F. Orlemann, Group Leader

Volatility Methods
N. R. Davidson, Group Leader

Metal Production
P. L. Kirk, Group Leader
*H. L. Baumbach**, Assistant Group Leader

• • • •

Basic Chemistry, Recovery, and Instruments
B. B. Cunningham, Assistant Section Chief

Basic Chemistry
B. B. Cunningham, Group Leader

Recovery
L. R. Dawson, Group Leader

Instruments and Physical Measurements
A. Ghiorso, Group Leader

*To assume full responsibility upon Kirk's transfer to Section C-IV

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Figure 33. Proposed organization chart. January 1944.

Although Willard and Manning are now Associate Section Chiefs, Willard will transfer to Site W later this year to serve as leader of the plutonium extraction chemistry group there.

Beaton gave me a summary (MUC-GTS-381) of the current status of our work on the extraction and decontamination of plutonium by adsorption methods. At my direction, Beaton and his group have been investigating adsorption methods that might make possible an all-adsorption process for the extraction and decontamination of plutonium. A small effort has been directed toward investigating the possibility of coupling the proposed Site X adsorption-extraction cycle to the bismuth phosphate decontamination cycle. Beaton reports that: (1) use of H_3PO_4 to elute zirconium and columbium selectively in the Amberlite IR-1 extraction cycle makes possible much higher decontamination factors with a resulting removal from the adsorbed plutonium of normally held-up fission activity; (2) a high decontamination of the Amberlite IR-1 elutriant (containing the plutonium) by a second adsorbent, titanium zeolite, appears to be a practicable possibility; (3) the coupling of the adsorption extraction cycle to the Bismuth Phosphate Process results in a high product yield but may produce lower decontamination factors for the coupled bismuth phosphate cycle than are normally obtained with a regular cycle; (4) a program of closer cooperation with ion-exchange manufacturers, such as Resinous Products and Chemical Co., Research Products Corp., and the Chemical Process Co. has been initiated to determine if other adsorbents exist that are not on the market that may be even better decontamination agents than the titanium zeolite.

The Chemistry Division seminar was held in Room 251, Ryerson Laboratory, at 7:45 p.m.

Today U.S. troops have landed at Suidor on the north coast of New Guinea without opposition. Some troops also landed behind Japanese lines with little resistance.

The Project Council Meeting was held at 9:30 a.m. in Room 209 of Eckhart Hall. In attendance were Allison, Compton, C. M. Cooper, Dempster, Doan, Fermi, Franck, Greager, Hilberry, Hogness, Hughes, Jeffries, Warren Johnson, Mulliken, Newson, Spedding, Stone, Vernon, Warner, W. W. Watson, C. J. Watson, and Wigner. Compton, in his State of the Nation message, indicated that experience with dropping bombs indicates that a number of nuclear weapons should be available to insure success and the progress of the enemy seems to be slow enough so that we can afford to wait until we have these. He announced that the "synthesis step" has now been demonstrated in the production unit at Clinton Laboratories and that we have the chemical separations plant operating smoothly with full-scale equipment and with useful recoveries of the plutonium product. Stone's Health Division report contained information presented by C. J. Watson that physiological changes have been observed upon examining one man who has been exposed to uranium dust from machining operations over a period of several years. However, there is no evidence to date that ingested metal or oxide causes these observable effects. Vernon reported that ventilation equipment will be installed

for the uranium metal machining operations as rapidly as possible. A suggestion was made that Chicago consider setting up laundry facilities for decontamination of clothes, as has been done at Clinton, since more active material is now being handled here. A possible conflict in patent areas between Chicago and du Pont was brought up by Fermi while making the nuclear physics report. The problem centers primarily around blueprints that are stamped as being the property of du Pont. Compton said this matter is being taken up with Roger Williams, and a clarification should be obtained in the near future. Doan from Clinton Laboratories reported that the first shipment of plutonium from the concentration and recovery group in the amount of 1.54 milligrams is now in Chicago and that several more milligrams should arrive in the next few days. The separations plant has turned over some 30 to 40 milligrams to chemists for concentration and purification. Plant yields are running between 49% and 73%, and the measured losses to waste run between 16% and 20%. The decontamination values obtained in plant operations appear to be following that obtained with the semiworks. Site Y is making a strong bid for four Chicago individuals, Nachtrieb of my section, Potratz, Magel, and Keith. Hogness said that Beaton will probably be going to Site X. Allison commented that all these moves make it very necessary to look closely at the personnel needs of the Metallurgical Laboratory. It is expected that Hanford will draw additional people away from the Laboratory. Wigner mentioned that a rumor is circulating around the Laboratory that by June 1 some 90% of the men in the Laboratory will have been released. Compton said that there is no basis for such rumors; it is certainly expected that the Laboratory will continue in operation even after the present emergency is over. Franck inquired whether or not it might be possible for more long-range work to be undertaken. Compton thought this would be possible but could not promise what the extent might be. Compton announced that the next meeting of the Project Council will be held on January 18 and 19 at Clinton Laboratories. This will be the first Project Council meeting to be held at Site X.

Joe Hamilton had dinner with Helen and me at home this evening.

Tuesday, January 4, 1944

Amanda Pauline Stein started work in Section C-I as a Research Assistant in Orlemann's sub-section and group. She comes from Carleton College, Northfield, Minnesota, where she received her B.A. degree. An additional laboratory assistant, Eleese Bowman, was also hired today.

I stressed for Hogness the importance of adequately testing the Hanford process at Clinton Laboratories as this is one of the primary aims of the separations plant at Clinton. In this connection a difficulty is presented by the fact that plutonium and fission product precipitations are often performed at Clinton using the same equipment which allows for contamination by more active material from a preceding step. No provision is made at present for carrying the product through two complete

bismuth phosphate decontamination cycles, such as are planned for the Hanford canyon operation and which should result in a decontamination factor of 10^5. I also point out that it may not be possible to test the use of scavengers with the present Clinton equipment. To overcome these problems, I suggest that equipment similar to that which is now present at Clinton in cells 2, 3 and 4 be installed in cell 6, now empty. This should make it possible to more nearly simulate various proposed Hanford operating conditions in the Clinton plant.

The Metallurgical Laboratory Progress Report for December 1943 (MUC-NH-570) is being issued. It states that in the Nuclear Physics Division the Experimental Sections have continued the construction of the P-9 pile and prepared for the erection of the "W" exponential test pile and the P-9 exponential lattice test pile. The P-9 lattice experiment will be run as soon as the necessary heavy water is obtained. The "W" test exponential pile will yield some preliminary data, but final results will be delayed until slugs in their final can-design are obtained.

The work of my section was summarized as follows: In a test of the Bismuth Phosphate Process one extraction, one product and one by-product precipitation on "W" concentration (including plutonium) gave a yield of ~95% using 0.5 cc of solution. Various reducing agents are being investigated to determine effect on decontamination, crossover cycles, and concentration steps; preliminary results are encouraging. High percentages of plutonium in bismuth phosphate have shown lattice expansions indicating that the plutonium carrying involves incorporation into the crystal lattice. Dissolving rate of plutonous peroxide has been checked and indicates that the data used heretofore for flowsheets are satisfactory.

The use of H_3PO_4 to elute strontium and columbium selectively from Amberlite IR-1 gives a decontamination factor of > 400. Lanthanum phosphate carries tracer amounts of plutonium quantitatively, but lanthanum phosphate plus cupferron does not seem promising. Uranous hypophosphate precipitated from $La(NO_3)_3$ solution has possibilities for carrying plutonium.

Work on the separation of neptunium and plutonium by the thorium molybdate method is being deferred in view of erratic results. The half-life of plutonium is recalculated as 24,000 years, taking into account back scattering of alpha particles; and this may effect the fission cross section value. Considerable work was done, without definite results, in preparing metal for x-ray determinations of density. With thorium as a stand-in, work is progressing on purification by solvent extraction.

Several reagents are regarded as promising for plutonium purification on the basis of work done with thorium; these will be checked as soon as plutonium is available. Work on volatile compounds is progressing but must await plutonium for conclusive results. Apparatus to determine hardness and the melting point of plutonium metal is on hand and will be used as soon as milligrams are available. Work on determination of oxygen and carbon is progressing. Atomic fluorine investigation, atomic hydrogen for reduction, refractory crucibles, Bismuth Phosphate Process precipitates solution in HCl, and determination of oxygen by the bromine method, are all topics receiving some attention.

The report also states "Intensive effort has been placed on the problem of the density of metallic 49, but the problem is not yet completed. Numerous x-ray diagrams have been obtained and interpreted, but it is not certain that any of them results from 49 metal. An unambiguous result is expected as soon as larger quantities of 49 are arriving." In the part covering the work of Section C-III (fission products) the following statement is included, "A 7-day uranium has been found in X pile material amounting to ca. 0.2% product yield." Under the heading "Plans for the Next Two Months" the report states that a conference with Clinton Laboratories has led to agreement that Clinton will work on problems centering around Site X and Chicago on problems centering around Site W. Total Project expense for the month of December is $532,652. Total personnel employed on December 31 is 1188 (422 academic), a net increase of 84.

The Clinton Laboratories Progress Report for the month of December 1943 (MUC-NH-558) was issued. It states that the 200 area (chemical extraction plant) started operations with radioactive material on December 20 and has been processing one batch (about one-third) ton every other day, with 20 mg of product per batch. The 100 area (pile) was shut down from about December 1 to December 7 for completion of construction work. Since that time operating level of the pile has been increased from 450 kw to 750 kw.

According to plans made last week, Ralph James is to visit Clinton Laboratories to neutron-irradiate his uranium and plutonium samples in the pile. He will look for new uranium and plutonium isotopes, produced by the capture of neutrons, that might decay by the emission of long range alpha particles which can be detected in the presence of the alpha particles originally present in the samples. James leaves by train at 11:00 tonight and will return next Monday.

Helen and I had dinner at the Quadrangle Club with Thorfin and Phoebe Hogness. After dinner we spent the evening at their apartment (5758 Blackstone Ave.)

The Soviets have crossed the old Polish border.

Wednesday, January 5, 1944

Albaugh wired me from Wilmington, California, saying that he will arrive in Chicago to start work next Wednesday. Joe Halperin, who was to be transferred from my section to Clinton on January 14, is being inducted into the Army tomorrow.

The "Complete Flow Sheet for Production of Plutonium" (CN-1220) has been issued and identifies the 95 or so principal operations required to go from uranium on to pure plutonium metal.

As I was making the rounds of the laboratory rooms this morning, I was suddenly struck by a disturbing vision. I pictured in my mind

the expanded scale of work with solutions containing plutonium that will soon result from the large quantities of plutonium soon to be received from Clinton Laboratories. I visualized beakers of plutonium solutions throughout the laboratory rooms, and it struck me forcibly for the first time that plutonium handling will now no longer be confined to micro quantities manipulated by specially trained experts. Recalling the health problems incurred by workers in the radium dial painting industry, I realized clearly that similar hazards face those of us working with alpha-particle emitting plutonium-239. I was struck by the fact that despite the great care in planning by the Project medical people, no one has anticipated and made special provision for the wide-scale handling of alpha-active material which presents special hazards of ingestion. It became clear to me that our rather ordinary laboratory hoods are inadequate for this task and that rather extensive rebuilding of our laboratory facilities to emphasize adequate air flow and extraordinarily clean operations will be necessary. I am determined that none of the people for whom I am responsible shall be subjected to any avoidable dangers from handling alpha-active plutonium. I immediately returned to my office and sent the following to Dr. Stone:

"It has occurred to me that the physiological hazards of working with plutonium and its compounds may be very great. Due to its alpha radiation and long life, it may be that the permanent location in the body of even very small amounts, say one milligram or less, may be very harmful. The ingestion of such extraordinarily small amounts as some few tens of micrograms might be unpleasant, if it locates itself in a permanent position. In the handling of the relatively large amounts soon to begin here and at Site Y, there are many conceivable methods by which amounts of this order might be taken in unless the greatest care is exercised.

"In addition to helping to set up safety measures in handling so as to prevent the occurrence of such accidents, I would like to suggest that a program to trace the course of plutonium in the body be initiated as soon as possible. In my opinion such a program should have the very highest priority."

Hogness requested approval from Allison of a reorganization of Section C-I based on the proposed chart I sent him last Monday. Hogness explained that the reorganization appears appropriate in order to better meet Met Lab responsibilities to both Site W and Site Y.

I attended a meeting of Section Chiefs in Franck's office. In addition to myself, those present included Burton, Franck, Garrison, Hiskey, Hogness, Manning, Potratz, Sugarman, and Willard. Hogness presided and explained that he and Franck have decided to exchange positions because of Franck's health concerns. Hogness reviewed the information presented by Compton at last Monday's Project Council Meeting. It is felt the enemy cannot be far along as the total amount of heavy water in Europe is so small as to barely run one pile. I mentioned that this was obtainable from only one source. Sufficient production to produce many devices is required.

The problems of increased hazards because of the contamination of clothing and personnel discussed at a Health Meeting yesterday were mentioned. It was stated that every section supervisor is responsible for his staff with regard to radiation safety. I raised the question of plutonium poisoning from ingestion and pointed out the urgent need for reliable information concerning physiological effects.

I attended the 7:45 p.m. meeting of the Purification and Metal Production Sub-section held in Room 209 of Eckhart Hall. Others present were Baumbach, Beard, Davidson, Dawson, Dirksen, Dixon, Frank, Ghiorso Hagemann, Heath, Hellman, Howland, Jaffey, Jensen, Jasaitis, I. Karle, J. Karle, Katz, Katzin, Kirk, Kohman, Kraus, Manning, Nachtrieb, Orlemann, Patton, Potratz, Pye, Rosenfels, Schuler, Sheft, Willard, Yett, and Zvolner.

I discussed the deliveries of plutonium fron Clinton. On January 1, we received 1.5 mg, and we should receive another milligram soon; 2 mg should be available on January 8 and will be sent to Site Y. On January 15 we should receive 20 mg. It is planned that the first gram quantity of plutonium, which will be available on or about January 25, will be sent to Site Y. Early in February we should receive a 1-gram quantity and following that, we should receive about one gram per month. It appears at present that Clinton will have little difficulty in meeting these deliveries as scheduled. Orlemann presided for the rest of the meeting.

Nachtrieb reported on the concentrations of impurities found in a 50 μg plutonium sample purified in our section and analyzed spectoscopically. Cerium, lanthanum, platinum, silicon, thorium, and tungsten seem to be present in the 1,000 ppm level. The material is 99% pure. Techniques using cupferron extraction are being investigated for the analysis of beryllium and lithium. Potratz reported that the analysis for oxygen, carbon, chlorine, fluorine, boron, sulfur, and phosphorus is still an object of concern at the present time. He described work underway for the analysis of phosphorus, sulfur, fluorine, and boron.

Hagemann covered two methods that might be considered for determining oxygen in plutonium metal. In one method the plutonium is brominated to yield plutonium tetrabromide that can be sublimed away from the rest of the sample leaving a residue consisting of carbides, oxides, and fluorides of plutonium. The amounts of these residue materials can be determined by alpha counting. The second method depends upon the reaction of carbon tetrachloride with the oxide at 200°C to yield phosgene that can be detected by standard methods. This metnod is expected to have a sensitivity of about 0.02 micrograms of oxygen.

Orlemann described a high temperature (1,800°C) fusion method for determining oxygen used at Site Y. Carbon monoxide is evolved when the oxide is heated in a carbon crucible. The carbon analysis method developed by Watters will determine 0.05 microgram of carbon, a method sensitive enough to be useful. Chlorine can be determined in quantities of 0.1 microgram. Interferences caused by bromine have not as yet been entirely solved. Davidson discussed the current interest that exists in plutonium chloride and plutonium bromide. These plutonium halides may be suitable as raw materials for metal production and for use in a purification cycle. Heath discussed experiments on the treatment of

U^{232} tracer with fluorine. It appears that surface effects at the tracer level are very significant and give variable and questionable results.

I talked about the experiments of Clifford Smith and Cunningham on the vapor pressure of plutonium bromide. Results show that the plutonium bromide has a vapor pressure of roughly one-quarter that of UBr_4. Current work within the metal production group was briefly discussed. Rosenfels is working on the hardness of metals, and Fried is doing preliminary work on making wires for the determination of melting points.

I took the occasion of this meeting to emphasize the hazards of handling plutonium and expressed my own concern that small amounts might have very serious effects if ingested. One hundred micrograms might be very unpleasant. I stressed the necessity of recognizing the dangers involved in handling plutonium and the necessity for taking precautions.

At midnight Hogness and I went to Englewood RR station and boarded a Pennsylvania RR train to Cincinnati where we will continue on the L&N RR to Knoxville and Site X.

According to today's papers, the Soviets are surging onward after their initial drive into Poland.

Thursday, January 6, 1944

Hogness and I arrived in Oak Ridge this afternoon. Then Perlman and I got together and spent considerable time talking about our joint problems. In particlar, I mentioned my growing concern for the hazards involved in the handling of plutonium, and we discussed how they can be avoided. I also saw James. He is spending today and tomorrow measuring the alpha-particle activity of his various plutonium and uranium samples. Aluminum Capsule I, containing two plutonium samples and one uranium sample mounted on beryllium disks, will be placed in the pile on Saturday for a one-day irradiation. Capsule II will contain two plutonium samples mounted on beryllium disks and one sample of uranium mounted on platinum This capsule will go into the pile on Sunday for a month's irradiation.

I am staying in the home of the Vance Coopers tonight. At Chicago in my absence, a meeting of the Council of Section C-I is being held in my office this evening, and the meeting of Research Assistants of Section C-I is also being held.

Friday, January 7, 1944

The two-day Planning Conference on the Bismuth Phosphate Process at Clinton Laboratories opened today. Priorities and manpower requirements associated with research problems at Clinton, Hanford, and Chicago are being discussed. Others present besides myself during the morning session are: Doan, Greager, Hogness, Warren Johnson, W. L. Kay, Kirst, Don Smith, and Squires. Squires mentioned several problem areas including

waste disposal, equipment decontamination, and tracing the distribution of fission products through the Bismuth Phosphate Process. The Clinton problem areas described by Greager and Johnson concerned better control methods for metal solution, the solution of bismuth and plutonium phosphates, bismuth phosphate extraction and decontamination, a study of the highest "W" product concentrations expected in the Process, improvements in decontamination, the use of manganese and lanthanum fluoride as scavengers in the Process, the development of a better concentration cycle, hydrochloric acid concentration methods, and the semiworks demonstration of the adsorption process.

I talked about our problem areas in Chicago, which include studying the Bismuth Phosphate Process at "W" concentrations, hydrochloric acid concentration methods, Hanford concentration methods, the Wet Fluoride Method, and the effect of various slug coatings on the process. Squires said that Whitaker received a letter from G. D. Graves, written just yesterday, about the lack of start-up procedures for Hanford when the concentration of plutonium is expected to be low. After discussion, I said that Section C-I, as part of our general study of the concentration and isolation of plutonium at Hanford, will be happy to include the investigation of suitable procedures for operation during this start-up period. Doan asked Johnson to write a formal request to have this work done; Johnson agreed to do so.

This afternoon I met with Greager, Kay, Warren Johnson, and Hogness. We discussed slug coating materials and canning procedures and their probable effect on the product. Two hundred each of aluminum-silicon bonded slugs and of zinc-bonded slugs are to be irradiated in the Clinton pile. Truck shipments of irradiated slugs to Chicago from Clinton are planned for January 11 and 17 with truck returns being made on January 14 and 20, respectively. Two batches of slugs should be available for starting material for our semiworks. One batch will have a product concentration of 88 micrograms per pound and the other 41 micrograms per pound. I was informed that Sinclair will reserve an additional 125 to 150 pounds of the same type of material for us. It was determined that the irradiation of 25 slugs, averaging about 65 pounds total, in the center of pile channel number 764 every week should provide suitable material to meet our future needs. This irradiation program will be started immediately with each batch being given a number, C-1, C-2, etc. Each batch will be cooled for four weeks before shipment. Sinclair will provide us with the irradiation data and other information immediately after each batch of slugs is removed from the pile. After the meeting Hogness and I confirmed these arrangements with Whitaker, Director of Clinton Laboratories.

I returned to the Vance Coopers' to spend the night.

Saturday, January 8, 1944

Ralph James's Capsule I containing the uranium and plutonium samples was put into the Clinton pile (stringer 21) this morning. Capsule II will go into the pile in the same place tomorrow (after Capsule I is removed) for a neutron irradiation of one month.

Kay, Warren Johnson, Hogness, and I met today as a Steering Group. We discussed the distribution of manpower required to effectively work on the problems assigned to Clinton and Chicago. A proposal was made concerning the transfer of personnel to meet manpower requirements. Seven men from my section will be transferred to Clinton as soon as feasible. Four of these men will go directly to the Separations Division at Clinton. The time for the transfers will depend in part upon the reassignment of men from Perlman's group to the Separations Division and upon the procurement of additional personnel elsewhere. Stoughton, Frank Steahly, and Paul Schulze are slated for immediate transfer. One man from Shapiro's group will transfer to Perlman's section. W. H Sullivan of the Ames Laboratory would be requested to come to Clinton as soon as possible. Two men from Burton's section will be shifted to Perlman's section in Clinton. As these transfers are made, Koshland, Turk, and Knox will be shifted from Perlman's section to the Separations Division. It is presently planned that the group within my section doing process work will be increased in number to approximately 20 by March 1. Perlman's section for carrying out process work at Clinton will consist of only 4 men for the remainder of this month but should increase to 6 during February and by March 1, 10 or 12 men will make up the group. It is planned to maintain the number at this point.

We planned that today's group (i.e., Hogness, Greager, Kay, Johnson, and I) will meet here at the Clinton Laboratories again on February 4 and 5 to review the program and to recommend any changes which appear desirable.

The distribution of problem assignments for Clinton and Chicago, together with manpower requirements, may be summarized as follows:

Problem No.	Title	Comments
202-X8C	The Behavior of High Concentrations of Plutonium in the Bismuth Phosphate Process	Essentially completed. Will be reviewed at Clinton by Steering Committee Jan. 15
202-X2C	Tetravalent Uranium as a Reducing Agent	In reference to the decontamination and crossover cycles to be carried out at Chicago. Estimated 1 man for 3 months to complete work.
203-X3C	Use of Ceric Ion as an Oxidant in the Complete Bismuth Phosphate Process	To be carried out at Clinton and Chicago. 1 man for 3 months required.
203-X7C	Search for New Oxidizing Agents--A Survey	To be carried out at Clinton & Chicago. 1 man working in each place for a period of 4 months.

1/8/44 (cont.)

Problem No.	Title	Comments
203-X27C	Oxalate as a Reducing Agent	Problem completed and a report of results has been requested.
203-X40	Decontamination in the Crossover Cycle of the Bismuth Phosphate Process at Clinton	Recommended that the problem be continued until Feb. 15, (present review date Jan. 15) with 2 men engaged in the work and that a report of progress to Jan. 15 be requested.
203-X3C	Use of Ferrous Iron as a Reductant for Plutonium in Fluoride Solutions	Work is practically completed. Report will be requested for Jan. 15, 1944.
211-X8C	Ceric Method of Concentration	It is recommended that this assignment be deferred until a later date.
213-X9C	Isolation of Plutonium from Room D Concentrations	Eight men will be required on a full-time basis for the coming month.
211-X22C	Alternate Methods of Hydroxide Metathesis of LaF_3 in the Concentration Cycles	This problem is completed and a report has been requested.
211-X23C	Uranous Oxalate Method for Concentration of Plutonium	It is recommended that the assignment be deferred until a later date.
211-X24C	Concentration Processes Involving the Use of Hydrochloric Acid	This problem is being carried at Chicago as a W problem and will be followed here if a man is available from the Concentration Group assigned to 213-X9C.
278-X18C	Development of Radio Chemical Analysis Procedures	Problem to be reconsidered in the near future in relation to the overall analysis program now in process of organization at Clinton.

HANFORD PROBLEMS

Title	Comments
Use of Bismuth Phosphate for Concentrating Plutonium from Lanthanum Solutions	Deferred
Solubility of Plutonium Peroxide	Will be carried out at Clinton by Concentration Group as a part of the isolation of product as time permits.
Purification of Plutonium from Lanthanum by Peroxide Precipitation	Assigned to Chicago
Investigation of Plutonium Compounds Suitable for Storage and Shipment	To be carried out at Clinton only as the problem pertains to Clinton by Concentration Group.
Microchemical & Semi-micro Product Yield Studies at Hanford Concentrations	To be carried out at Chicago. 1 man working for 3 months.
Oxidation and Reduction Rate Studies	To be carried out at Chicago at a later time.
Product Yield and Decontamination at Hanford Concentration on 100 cc Scale	To be carried out at Chicago at a later time.
Hanford Coatings	2 men working at Chicago in Dreher's group for 1 month.
Basic Chemistry of Plutonium	To be carried out at Chicago at a later date.
Hanford Concentration Process	To be carried out at Chicago at a later time.
Hanford Decontamination Studies	To be carried out in Chicago. 1 man working 1 month.
Special Fission Product Chemistry at Hanford Concentrations	Deferred
Determination of Necessity for Pre-Reduction	Deferred
Hanford Radiochemical Analytical Problems	To be located at Chicago in a separate group.

Roy Beaton and I caught a train at Knoxville in the afternoon to return to Chicago via Cincinnati. On the train we had a long talk about the adsorption program. He proposes a program involving both Oak Ridge and Chicago with the following specific assignments:

(1) Chicago--General Engineering Section to work on pressure drop with adsorbents of various mesh sizes and viscosity measurements of UNH solutions. Boyd's section to have four men on scouting work. (2) Clinton--Seven men in Boyd's section to test temperature effects, bed depths, flow rates, elution studies, etc. Beaton with six men to carry out semi-works studies of reproducibility and the high spots of Boyd's results; Beaton to also correlate the work of Boyd and the engineering design work of Leverett and five men. Coupling of the adsorption process and the Bismuth Phosphate Process will be studied by Apple and three men. I estimated that this effort would require 30 men on the extraction step alone.

Sunday, January 9, 1944

I arrived back in Chicago early this morning. During my absence Helen had Wilma for lunch on Thursday and was then invited to the Ghiorsos' home for dinner. Friday she had dinner with Peg Beaton--Roy Beaton was also in Oak Ridge. Last night Helen and Peg accompanied the Willards to the Studebaker Theater in downtown Chicago to see Noel Coward's play, "Blithe Spirit," with Clifton Webb, Mildred Natwick, Haila Stoddard, and Peggy Wood.

A delegation of scientists from the Montreal Laboratory in Canada arrived here yesterday at General Groves' invitation. The visitors (H. von Halban, B. Pontecorvo, G. Placzek, P. Auger, G. C. Laurence, G. M. Volkoff, B. Goldschmidt, R. E. Newell, F. A. Paneth, A. Nunn May, A. G. Maddock, D. W. Ginns, and J. Chadwick) will stay through Tuesday to exchange information with us and discuss collaboration between the two laboratories. The last meeting took place September 20 of last year when Compton and Wigner went to Montreal to attend the meeting, "Exchange of Information with the British."

A general meeting with the delegates took place yesterday morning in Room 209, Eckhart Hall. Our representatives included Allison, Compton, Fermi, Franck, Hilberry, Hiskey, Smyth, Vernon, Wigner, Willard, and Wilson. Willard described the meeting to me:

Compton opened the meeting with a word of welcome and asked Allison to say a few words about the P-9, heavy water, program of the Metallurgical Laboratory. Allison mentioned that the large-scale engineering development program of our Laboratory, which was pointed toward the construction and operation of a P-9 plant to produce plutonium in quantities of military importance, was cancelled last October. That portion of the P-9 program remaining consists of measurements of fundamental nuclear properties of the atomic constituents of heavy water, an exponential experiment to determine the critical size of a heterogeneous heavy water-heavy metal system, a similar experiment for a homogeneous system, and the construction of an experimental heavy water pile operating between 10 and 1,000 kw.

General Groves arrived at the meeting somewhat later and spoke about the permissible interchange of information with the Montreal Group.

He said that all scientific information about heavy water that is useful in designing a pile, and all engineering information toward the same end, can be freely exchanged. On the other hand, the chemistry of extraction of plutonium and engineering details of the graphite piles are not to be discussed nor are the time schedules for plutonium production. It was then decided to adjourn the general meeting and reassemble in small groups devoted to discussion on nuclear physics, chemistry, and engineering.

Conversations with the Canadian delegation and the Chemistry Division occurred during the afternoon. Goldschmidt, Maddock, and Paneth met with Willard, Burton, Hiskey, and Franck. The discussions centered primarily around heavy water analysis and standardization. Radiation effects and corrosion problems in high radiation fields were given some attention by Burton. Willard, representing my section and the Separation and Purification Groups, briefly described the basic chemistry of plutonium. The Canadians apparently are very interested in our preliminary work which gives evidence for the +3 valence state for plutonium. No details of the separation and purification processes were disclosed to the Canadians during the meeting.

This morning Goldschmidt, Maddock, and Paneth met with Hiskey to resume discussions. The meeting lasted through the entire morning and a considerable part of the afternoon. Hiskey outlined the preliminary P-9 pile research program of the Chicago group. The chemistry of uranium compounds such as the oxides, fluorides, oxyfluorides, uranates, and oxide hydrates was covered. A considerable portion of the meeting centered around the problems associated with the construction of a heterogeneous heavy water reactor, such as a slurry drift. The topics discussed were: the effect of particle size on the viscous properties of slurries; chemical considerations affecting viscosity of the slurries; effects of radiation on slurries; and the rates of crystal growth and crystal degradation under reactor operation conditions. The desirability of using uranium compounds in which the U^{235} content has been fortified was also pointed out. Possible corrosion and erosion problems were touched upon briefly. Rapid small-scale techniques used by the Chicago group were outlined, and the program involving particle size measurements using an electron microscope technique was described.

A discussion of the nuclear physics programs of the two groups was held from 9:30 to 11 a.m., at which Allison, Fermi, and Wigner were present. After this, Newell, Halban, May, Auger, Allison, and Zinn visited Argonne Laboratory, and the visitors were shown the operation of the graphite pile and the present status of construction on the experimental P-9 pile.

Tonight, Helen and I entertained Goldschmidt in our apartment.

The eight-column headline in today's Sun reads "Tokyo-Berlin Ship Sunk." U.S. and Brazilian planes and surface vessels sank an armed German merchantman in the South Atlantic as it tried to carry war materiel from Japan to Germany. (Brazil is the only South American country in the war.)

Monday, January 10, 1944

Last week, while I was away, the Lab received its second shipment of plutonium from the Clinton Laboratories pile--1.0 milligram.

Cunningham, Davidson, Dreher, Ghiorso, Kirk, Manning, Orlemann, and Willard attended last Thursday's meeting of the Council of Section C-I. They considered the tentative record form to be used in accounting for plutonium materials. Dawson will be given the job to set up and keep the plutonium control records in our section. The work being done on plutonium tetrafluoride was discussed in relation to method of formation, color, analysis, and density determinations. Davidson suggested forming $(NH_4)_2PuF_6$ and heating in the presence of HF to form PuF_4. Willard described Zachariasen's reasoning that PuF_4 should be found to have a density of 7.2 ± 0.2 gm/cm^3, based on its relationship to other fluorides and its position in the ThF_4, UF_4, PuF_4, HfF_4, and ZrF_4 density sequence. Although the x-ray analysis evidence is good concerning the +4 valence of plutonium, there are still questions to be answered--the black fluoride of plutonium being one. The compound PuF_3 has still not been positively identified by Zachariasen. Chemical analytical techniques that might be tried to identify PuF_3 were discussed. Florin, or others, will try to identify plutonium tetrafluoride by determining weight changes when a sample of plutonium dioxide is reacted with anhydrous HF.

Kirk described the planned allocation of our present plutonium supply--500 µg are planned for x-ray studies of the metal; 500 µg for density, hardness, melting point, and other determinations on the metal; and 200 µg for hydrogen reduction work. Actually, nearly all will be made available for x-ray analyses, as Zachariasen and Kirk may determine density (and other physical properties) on the same samples. Samples of at least 200 µg are needed by Zachariasen to give satisfactory x-ray data. Density determinations of plutonium metal are still of highest priority. What is left will be used for studying the possible PuH_3-to-powdered metal conversion and atomic hydrogen reduction to plutonium metal for which Karle has some evidence. Davidson thought it might be better to try the atomic hydrogen reduction procedure on uranium first. Willard mentioned the other plutonium sample allocations--100 µg went to Watt, 50 µg to Davidson, and the remainder is being used by G. Johnson to test the phosphate process at Site W concentrations.

Cunningham reported that weight and counting data obtained from the big plutonium sample received from Clinton Laboratories on New Year's Day give a value of about 131,000 disintegrations per minute per microgram, which compares favorably with previous values for smaller samples produced at Clinton Labs. However, earlier cyclotron-produced samples were 8% higher. The possible presence of Pu^{238} was discussed. Cunningham suggested the possibility that the Clinton pile sample might have contained impurities which did not precipitate with the hydroxide. The recent meeting of Section Chiefs in Franck's office was reviewed with special emphasis on the possible plutonium alpha radiation hazard problem. Orlemann recommended that someone be hired to mop down our rooms at least once a week and Manning agreed to look into this. The wearing

of coveralls, with a twice weekly change, was discussed. Ghiorso reported that a decision on counters for Site W will be made by February 1, and new alpha counters for Section C-I will be coming very soon.

Last Friday Wigner sent Compton a 16-page discussion of the planning for Hanford and the status of his review of the blueprints. He expressed concern that the Metallurgical Laboratory does not have enough to say with regard to the type of pile selected, and he feels that the du Pont Company should have consulted with Laboratory members to a much greater degree. The present Hanford plant is being designed on the basis of plans prepared and submitted by Wigner's group early in January 1943. It had been estimated by Leverett and Fermi that the plant could have been constructed and ready for operation within seven months if given high priorities. Wigner feels that du Pont is proceeding very slowly and that even on their "accelerated" schedule, the construction job will require seventeen months. He believes that this time delay is caused largely by the fact that du Pont wants to do the job completely its own way and has not been willing to work in close cooperation with Laboratory personnel. It is estimated that the first pile will now be completed around July 1, 1944, the second should be completed around August 1, and the third pile should follow about two or three months later. Wigner feels that, in view of the decision made by du Pont without consulting laboratory personnel, the laboratory cannot be responsible either for the speed with which the plants are being erected or for any delay between erection and operation caused by imperfect design.

On Saturday, Bernard M. Winner, from the Allison Division of GMC, Indianapolis, Indiana, arrived as a new employee in Section C-I. Winner graduated from Indiana University with an A.B. degree in chemistry in 1942. Today, Sidney Katz started work in the Kirk-Baumbach group. Katz received his Ph.D. from McGill University in 1937 and engaged in post-doctoral work at the University of London in 1937-38. He has been employed with Goldsmith Bros. Smelting and Refining Company, Chicago. Other new members of our staff today are James R. Gilbreath, Henry R. Hoekstra, and Earl K. Hyde, all men with bachelor's degrees who have been working with Schlesinger on a U.S. Signal Corps project at the University of Chicago. Bernard B. Weissbourd also is starting today. In December Ghiorso asked the General Staff Corps for the services of Weissbourd, who is in the army and has an ideal background--a combination of electronic and chemical experience. Today was Nachtrieb's last day with Section C-I, as he is transferring to Site Y.

The Canadian group which has held discussions with Willard and others for the past two days was given a tour of the third floor of Site B today and was invited to view the laboratory work in progress on heavy water analysis and on slurries. After lunch Maddock and Goldschmidt were granted permission by Compton to read selected reports issued by the Columbia and Chicago slurry groups.

Gordon F. Mills of the Chemical Process Company in San Francisco replied affirmatively in a letter dated January 6 to my suggestion that he meet with Willard, Beaton, and me on Tuesday, January 11, to discuss ion-exchange resins.

I received a response from Stone to my memorandum of January 5 concerning the health hazards of working with plutonium. The text of Stone's memo is as follows: "The matter of the toxicology of plutonium and the way it will be distributed in the body are under consideration by the Health Division. The question of tracer studies to find its distribution in the body have long been planned as a part of Dr. Hamilton's program, although it has never been mentioned in official descriptions of the program. Recent discussions with Dr. Hamilton make it obvious that the studies can be much more accurate and much more quickly done when a few milligrams can be made available to him, rather than when only microgram amounts are available. Informal discussions with Mr. Compton make it apparent that this amount could be made available in a couple of months or more.

"Sufficient quantities to study the chemical toxicology will not be available for some time, and it will probably take quite large amounts to demonstrate the radiological damage that might be caused by the alpha radiation."

I attended the 7:45 p.m. Chemistry Division seminar in Room 251 of Ryerson Hall.

The top headline in today's paper comes from the Soviet front, where the Russian army is hitting the Nazis on a 450-mile front.

Tuesday, January 11, 1944

Two new men started in our section today. They are Milton Ader from Oberlin College and Roy W. Greenlee from Ohio State University. A. L. Henne, Professor of Chemistry at Ohio State says in his recommendation of Greenlee, "Extremely brilliant intelligence; outstanding originality and imagination."

I received a memorandum from Hogness that in the very near future draft boards will allow no deferments whatsoever for men between ages of 18 and 22. Since this will create a very difficult situation for us, he requested that I send him a list of all men under 23 years of age in Section C-I so that some action can be taken.

At 6:00 a.m. James returned from Oak Ridge where he has been since last Wednesday bringing with him the uranium and plutonium (Capsule I) samples which received one-day neutron irradiations in the Clinton pile. His return was delayed because of an unexpected 11-hour stop by the L&N R.R. at Somerset, Kentucky. Prior to submitting the uranium and plutonium samples for neutron irradiation in the Clinton pile, James spent Thursday and Friday counting the samples in order to compare alpha-particle counting rates before and after neutron capture. After 24 hours in the pile, which ran at low power during the day and full power during the night, Capsule I containing these samples was withdrawn at 10:30 Sunday morning, and Capsule II containing similar samples was inserted into the pile for a month's irradiation with neutrons. James measured the alpha activity of

the samples in Capsule I within an hour after removal from the pile. He was disappointed to find no increase in the alpha counting rate due to neutron absorption. Today he made additional measurements on the samples and is now forced to conclude that neutron activation has not perceptibly increased the alpha activity of these particular samples. We hope that the one-month neutron irradiation of samples in Capsule II will produce positive results.

This morning at 11:00 a.m. Halban and Allison conferred on the exchange of reports, visits, and information between the Montreal and Chicago Laboratories. Representatives of the Montreal Group will come again to the Met Lab on the 3rd of next month, remaining until the 5th.

Gordon F. Mills arrived today for his scheduled conference with Beaton and Willard about synthetic resin ion-exchangers for the decontamination of product materials. For security reasons Mills was told only that our problem consists of first separating thorium from 10% $UO_2(NO_3)_2 \cdot 6H_2O$ and then from zirconium, columbium, cerium, yttrium, and strontium "impurities". Three potentially useful resin materials were discussed: "Duolite C-3", a phenol-formaldehyde resin possessing sulfonic exchange groups, and a glycine type resin capable of forming an internal complex with specific ions. This latter material would more correctly be called a chemi-sorption rather than an ion-exchange resin. Mills believes it is possible to make a resin of this type as a specific adsorbent for thorium. Samples of the first two mentioned resins will be sent to us in the very near future for preliminary testing. An effort will be made by Mills, possibly with his own company's funds, to make a resin of the third type; samples will be sent to us as soon as they are available.

Two monthly reports of our section for the month ending January 5 are being issued. The first of these reports (CN-1241) is entitled: "Chemical Research--Special Chemistry of Plutonium" by Hogness, Franck, Seaborg, Willard, Cunningham, and Manning. Cunningham, Ghiorso, and Hindman present their recent results on the determination of the specific activity and half-life for plutonium-239. Through comparisons between measurements made with a recently developed low geometry alpha counter and our so-called "50%" high geometry (inside) alpha counter, the true counting yield for a plutonium sample mounted on platinum is determined to be 52% and not the formerly assumed 47%. This has been essentially confirmed by counting identical weight uranium and plutonium samples, mounted on platinum and on quartz, on the "50%" high geometry alpha counter to test the concept that the effect is caused by small angle backscattering of alpha particles on the supposition that the backscattering would decrease with atomic number. Using this counting yield figure and the most recent determination for the specific counting rate, they obtained a half-life for plutonium-239 of 24,000 years. Preliminary calculations by Crawford indicate that such small angle backscattering of alpha particles should take place.

Kroner, Schuler, and Thompson report on the use of uranous hypophosphate as a concentration method for plutonium in nitric acid solution. Earlier experiments on the carrying of plutonium by uranous

hypophosphate have been extended to show that satisfactory carrying may be obtained from solutions containing as much as 10 mg/cc of La^{+3}. A precipitation procedure is described that could be used to further concentrate plutonium following a bismuth phosphate extraction-decontamination procedure and a lanthanum fluoride concentration step. An additional concentration factor of at least 1,000 may be expected.

Creek, James, and Thompson note that, under given conditions, greater than 95% of tracer amounts of plutonium are carried by lanthanum phosphate from solutions. Such a step would form a possible alternate for the present lanthanum fluoride concentration step following bismuth phosphate extraction and decontamination. They also report that in contrast to earlier work by Potratz, it does not now appear possible to satisfactorily extract plutonium from aqueous solutions of lanthanum phosphate using the chloroform-cupferron extraction method. This would seem to preclude the possibility of coupling this extraction step with a lanthanum phosphate precipitation as a method of concentrating plutonium.

A comparison of the effects on decontamination produced by the use of Fe^{+2}, U^{+4}, and $H_2C_2O_4$ as reducing agents in decontamination cycles of the Bismuth Phosphate Process is reported by Kroner and Thompson.

The second monthly report (CK-1221) is entitled: "Chemical Research--Special Chemistry of Plutonium" by Hogness, Seaborg, Willard, and Manning. Work accomplished by the Purification and Metal Production Groups is covered in this report which has the special security classification of a CK report. Brody and Orlemann report that 15 organic solvents have so far been investigated for their efficiency in tracer plutonium extraction. Several of these solvents show reasonable efficiency for extracting plutonium in the oxidized state from ammonium nitrate-nitric acid solutions. A few show some promise in extracting reduced plutonium. Thorium is being used as a stand-in to study purification that may be effected by solvents which extract 40% or more reduced plutonium. The choice of thorium as a stand-in is based on the similarity in solubility and general characteristics of various known insoluble salts of reduced plutonium and the corresponding thorium salts. In order to prepare plutonium metal of satisfactory purity, the starting materials in the metal production step will have to be freed of most, if not all, the light element impurities. These elements must not be present in the final product. Based on the limited number of experiments run so far, the investigators feel that solvent extraction appears to be the most promising of the methods available for the purification of a plutonium salt.

Jensen and Orlemann discuss the purification of thorium from light element impurities by the use of precipitants. Although it is not expected that the results obtained with thorium will be completely valid for plutonium, they feel that these results will serve as a useful guide in choosing the more promising precipitating agents when larger available quantities of plutonium make this possible. Thirteen promising reagents are listed in the report.

Katz and Davidson describe their studies on plutonium and uranium bromides. Plutonium bromide may be of considerable future importance in both the purification and metal production steps. If its volatility is

sufficiently high, the sublimation of plutonium bromide should afford some measure of purification from light element impurities. Plutonium bromide, or plutonium chloride, would seem to be a good starting material in the final large-scale production of plutonium metal. One microgram of plutonium oxide, spread over a one square millimeter area on platium, was volatilized as plutonium bromide in about one-half hour in a stream of HBr at 700°C.

Heath and Davidson report on volatilization studies of plutonium samples that were placed in calcium fluoride tubes and treated with a stream of fluorine at 500°C. The plutonium volatilizes, but then it deposits just outside the region of fluorination just as it does when copper tubes are used. Experiments with tracer amounts of uranium (U^{232}) give similar results; since uranium hexafluoride in massive quantities is known to be volatile, it appears that reliable conclusions cannot be drawn from such tracer experiments.

Dixon, Cliff Smith, Cunningham, and Orlemann discuss the microchemical preparation of thorium and plutonium acetylacetonates. In Report CK-923 (September 11, 1943), it was tentatively reported that plutonium forms a volatile acetylacetonate. Later experiments carried out to verify the original results have been unsuccessful. Many of the lighter elements form volatile acetylacetonates; it is felt that even if plutonium acetylacetonate could be formed and were volatile, it would have only a limited application to the purification program. However, the work on the preparation and volatilization of plutonium acetylacetonate is continuing in view of its potential application to the preparation of thin films of plutonium. Thorium, in microgram quantities using UX_1 as tracer, is being used as stand-in in the hope that the preparation and volatilization of thorium acetylacetonate will be similar to that of plutonium acetylacetonate. Although repeated past attempts to prepare a volatile thorium acetylacetonate on the 5 to 10 microgram scale using the method of Urbain have failed, a literature search uncovered a method described by Bilitz that, when slightly modified, gives satisfactory results on the 0.5 gram and 10 microgram scales. Preliminary experiments on the 10 microgram scale show that plutonium acetylacetonate exhibits a volatility comparable to that of thorium acetylacetonate.

Baumbach and Fried give the current status of their studies on the production of finely divided plutonium metal. Although the supply of plutonium salt was limited in December, an attempt was made to reduce plutonium tetrafluoride in thoria crucibles, using lithium for one sample and barium for another sample. The samples were submitted to Zachariasen for x-ray analysis of their structure. Lithium fluoride, lithium oxide, thorium oxide, and an unknown substance were found in the sample reduced by means of lithium. If this unknown substance is plutonium metal, the density would be 13.01 gm/cm^3 in contrast to the value of 15.5 gm/cm^3 for material prepared at a higher temperature and measured by the capillary method described in an earlier report (CK-1145). The x-ray pattern produced in the sample reduced with barium was exclusively that of the unknown substance obtained with lithium, except for a trace of PuO_2. It is possible that this "unknown substance" is a compound of lower valent plutonium rather than plutonium metal. The chemical analysis necessary to characterize this substance, which dissolves completely in one normal hydrochloric

acid with vigorous gas evolution, probably must wait until larger quantities are available.

The last item in this report describes studies by Fried and Baumbach on the preparation of uranium and thorium hydrides. Uranium and thorium have been used as "stand-ins" in an attempt to develop an ultramicro method that can be used to determine the structure of the hydride of plutonium by means of x-ray analysis. A sample containing about 100 micrograms of uranium tetrafluoride was reduced to metal with barium and made into the hydride using the apparatus described in CK-1145. A capillary tube containing about 50 micrograms was analyzed by Zachariasen, and the typical pattern for uranium hydride was obtained. Hydrided thorium metal samples were prepared but due to interferences of thorium oxide, no satisfactory x-ray analysis could be made.

I attended the meeting of the Council of Section C-I in my office, held this evening rather than the regular time of Thursday evening. This fits better with the meeting of the Thomas group scheduled for all day Thursday. We had a long agenda to cover, much of it relevant to the Thomas meeting. Also in attendance were Cunningham, Davidson, Dreher, Ghiorso, Kirk, Manning, Orlemann, Thompson, and Willard.

My exchange of memos with Stone concerning plutonium hazards was circulated. Kirk doubts whether plutonium would be eliminated from a person's system under natural conditions. Nickson will be asked to consider at the meeting of the Purification and Metal Production Sub-section next week the setting up of safety rules by the Health Division to guard against these hazards.

Of the next 15 milligrams of plutonium we will obtain from Site X, the Council decided that Cunningham and Ghiorso should be allocated 2 mg; Orlemann and Potratz 2 mg; Davidson 1 mg; Thompson, Johnson, and Watt 1.5 mg; and Kirk 8.5 mg. This distribution information will be given to Dawson. Cunningham pointed out that a large discrepancy still exists between the amount of plutonium that goes into the metal production group and what goes back to recovery. Plutonium loss into the air is a possibility. Davidson suggested a sampling technique in which a thin layer of stopcock grease on a plate, suitable for counting in an alpha counter, be used to collect dust samples and check for plutonium. I asked Ghiorso to check the air and parts of Kirk's room (room 13) with an alpha counter, and I said Kirk's room should have an especially good hood and an exhaust blower. It may be necessary to filter the air that passes out in order to permit recovery of airborn plutonium.

Plans concerning the use of the next plutonium sample allotments were discussed briefly. Davidson stated that their 1 mg will be used for chlorine and bromine experiments. Heath will test out the procedure first using uranium. Thompson said Watt will use 1 mg of plutonium to study precipitation of the peroxide from $La(NO_3)_3$ solution at "W" concentrations. He will then go on with further flowsheet tests. Johnson will run the "mainline" process on a 10 cc scale using twice the "W" fission-product concentration. A sample will also be prepared for Mrs. Mooney in Zachariasen's laboratory. Thompson mentioned that later Watt will try precipitating plutonium fluoride (and oxalate) from an HCl solution of bismuth phosphate.

Kirk said that the recently obtained very black fluoride (which behaves so poorly upon reduction to metal) thought to be PuF_4, may not be PuF_4 at all and the x-ray data obtained by Zachariasen seems to support this. The black compound is very fine, reduces to plutonium metal just like plutonium oxide does, and is not volatile at 1,200°C. A lower temperature and a higher acidity was used previously within a lucite container with ignition to obtain a PuF_4 precipitate that was not black and that behaves better in the reduction procedure. I suggested that someone should go through the notebooks to look for a correlation between results and the experimental parameters and conditions. A plan was outlined that consists of preparing a history of past experiments and investigating the parameters associated with five other studies: (1) the effects of HNO_3, etc. in the treatment of $Pu(NO_3)_4$ to form PuO_2, followed by treatment with HF; (2) fusion of PuF_4 (black); (3) the possible reducing action and other effects of the old ignition procedure in the lucite container; (4) the effects of time and temperature in the "HFing" procedure; and (5) comparative studies using PuF_4 made by the wet method.

Orlemann recommended that all five of these areas of investigation be looked at more or less simultaneously. It was decided that Baumbach will correlate the data, including the data from Davidson's notebook. Kirk thinks all the studies could be done with a total of 0.5 mg of plutonium. The present black fluoride will be dissolved in H_2SO_4 to convert it back to the nitrate to use as material for the studies. Because of the critical importance of solving the plutonium fluoride problem, the work will be reviewed every two days.

Thompson reported that by tomorrow eight new men will begin work in his sub-section and will require training. He explored the possibility that James, Dixon, La Chapelle, and Kohman might assist in the training, but since these individuals are very busy, I suggested that Schuler take on the training job with Katzin to provide backup. Ghiorso said the counting manual is nearly ready, and I requested that it be rushed through immediately so that it can be duplicated tomorrow morning and be available for training. A lecture series was planned for the new men. I will give the first lecture on Thursday and will be followed by Thompson and Watt on Saturday, and Dreher next Tuesday.

Cunningham said he would like about 10^{11} counts of 93^{239}, and it was suggested that he try to make arrangements at Argonne through Hughes. I reported that Turkevich has found at least 0.1% of U^{237} (compared with Pu^{239}) in pile material. We generally agreed to adopt 24,000 years as the half-life for Pu^{239} using 52% as the geometry for the "50%" chamber. It was suggested that Perlman may wish to follow suit. The specific activity on the basis of the above is 71,000 counts per minute per μg for weightless samples mounted on platinum.

Helen had coffee at Wilma's.

As the Soviets have moved into Poland, their government has announced its intention to retain the bulk of the territory that Russia obtained in the 1939 partition of Poland.

Wednesday, January 12, 1944

Frederic W. Albaugh arrived as scheduled and I am making him group leader for the Extraction and Decontamination group in Thompson's subsection. Jack Flox from Texas Company, Lockport, Illinois, also began working in Section C-I. Formerly he was a chemist for the Department of Medicine here at the University.

Manning and Orlemann have prepared an outline of the Purification and Metal Production Program presently engaged in by our section and have submitted this to me in the form of a memorandum (MUC-GTS-391) for use in the meeting of the Thomas group tomorrow. The program is divided among three major areas--analytical (a joint action of Sections C-I and C-IV), preparation of a pure plutonium compound, and studies with plutonium metal. With the larger amounts of plutonium coming, we calculate we need about 10 grams to carry out this program. A detailed outline of the projects under each program area is given as follows:

Purification and Metal Production Program

I. Analytical--Joint Action of Sections C-IV and C-I

A. Spectrochemical Analysis

1. Copper spark method for lithium, sodium, potassium, beryllium, magnesium, calcium, aluminum, silicon, and possibly boron and phosphorus--based on a preliminary separation of plutonium from these light elements.

2. Pyroelectric concentration method for lithium, sodium, potassium, beryllium, magnesium, silicon, phosphorus, boron, iron (to be used if (1) fails. It is less satisfactory.)

3. Electrodeless discharge

B. Chemical Analysis

1. Fluoride--Limit 0.6 ppm

Method: Separated as HF, determined by decrease of fluorescence of aluminum morin complex.

Status: Detection limit 0.01 microgram. Recovery of 0.1 microgram amounts erratic.

2. Boron--Limit 0.2 ppm

Method: Separated as methyl borate, determined colorimetrically.

Status: Determines 500 ppm on 1 mg uranium samples. Increased sensitivity being studied.

3. Phosphorus--Limit 350 ppm

Method: Extraction as phosphomolybdic acid, determination colorimetrically.

Status: Determines 50 ppm on 1 mg thorium samples.

4. Sulfur--Limit 10,000 ppm

Method: Removed as H_2S, determined colorimetrically by conversion to methylene blue.

Status: Determines 500 ppm in the presence of 1 mg of uranium.

5. Carbon--Limit 50 ppm

Method: Conversion to CO_2, absorption in dilute NaOH, measurement of decrease in conductivity of the NaOH.

Status: Determines 0.05 microgram of carbon. Some mechanical difficulties and occasional erratic results.

6. Chloride--Limit 200 ppm

Method: Remove as Cl_2 by diffusion, determine colorometrically.

Status: Determines 0.1 microgram of chloride. Bromide interference not completely eliminated.

7. Oxygen in Metal--Limit 80 ppm

a. Bromine volatility method

Method: Metal plus Br_2 yields volatilization of the metal as the bromide and a residue of PuO_2 primarily. The residue can be counted to estimate the oxide.

Status: Data insufficient to decide the limitations.

b. CCl_4, phosgene method

Method: The metal will be opened up by reaction with a suitable reagent such as Cl_2 or Br_2 and then reaction of the oxide with CCl_4 should yield $COCl_2$. A colorimetric determination of the phosgene will be a measure of the oxygen.

Status: Data insufficient to support conclusions.

C. Determination of the Properties of Plutonium of Analytical Importance

1. Solution of the metal.

2. Solution of the oxide.

3. Degree of separation of plutonium from the light elements by the cupferron extraction method.

4. Test pyroelectric concentration method on plutonium.

5. Develop alternates for the cupferron method of removing plutonium from the light elements.

6. Test developed chemical methods in the presence of plutonium.

D. Shotgun Methods

1. Construct High Efficiency Neutron Counters

a. BF_3 pressure ion chamber
b. Proportional BF_3 counter
c. Fission counter

E. Cooperate with the General Physics Division in the Development of Mass Spectrographic Methods

F. Cooperate in the Problem of Setting up Analytical Standards

G. Carry Out Routine Analysis for the Light Elements

1. On plutonium

2. On plutonium stand-ins

3. On a variety of reagents needed in the purification program

II. Preparation of a Pure Plutonium Compound

A. Precipitation Methods

1. Survey using plutonium stand-ins to select the most promising possibilities

2. Quantitative studies of plutonium purification on the 1 to 10 mg scale

B. Solvent Extraction Methods

1. Study distribution of plutonium tracer between various empirically chosen two liquid phase systems

2. Determination of light element distribution in two phase systems where plutonium can be satisfactorily extracted

3. Study of purification of plutonium on the 1 to 10 mg scale by systems tested in (2)

4. Study combination of solvent extraction with precipitation methods

a. Using plutonium stand-ins
b. Using plutonium on the 10 mg scale

C. Volatility Methods

1. Prepare and determine the properties of volatile plutonium compounds

a. Preparation of plutonium halides from oxide and metal
b. Determination of the composition and the vapor pressures of the plutonium halides
c. Preparation and determination of the vapor pressure of plutonium acetylacetonate

2. Study the purification attained by volatility methods

a. Using plutonium stand-ins, where possible
b. Using plutonium

3. Study techniques and materials of construction needed in extending work on 1 to 10 mg scale to the 10 to 100 gm scale

III. Studies with Plutonium Metal

A. Methods of preparing metallic plutonium--including the preparation of plutonium compounds most suitable as starting materials

1. Reduction with electropositive elements--including the fabrication of satisfactory refractories

2. Electrolysis

3. Atomic hydrogen reduction

4. Hot wire technique

B. Determination of physical properties of plutonium metal

1. Density

2. Melting point

3. Hardness

4. Vapor pressure

C. Preparation and determination of the properties of plutonium hydride

1. Density

2. Formula and structure

3. Equilibrium pressure of H_2 as function of temperature

D. Purification of plutonium metal

1. Study the purity of plutonium prepared by reduction of a pure compound with pure reducing agents in various crucible materials

2. Purification achieved by distilling out light element in vacuo

3. Possibility of purification by sublimation of plutonium metal

4. Purification by the hot wire technique

5. Test suitability of various refractories for reduction and remelting of plutonium by studies using uranium

This evening I attended the regular triweekly C-I Extraction, Decontamination, and Concentration Sub-section meeting at 7:45 p.m. in Room 209 of Eckhart Hall. The following were among those who also attended: Albaugh, Creek, Dean, Dreher, Gaarder, James, G. Johnson, Malm, Pye, Schuler, Thompson, Watt, Willard, and Yett. I discussed work at Clinton in light of my recent visit there and pointed out that the plant has been operating successfully with active material for approximately 3 weeks. Product yields of 50% to 74% have been obtained with decontamination factors of several hundred at the end of the decontamination cycle and of several thousand through the crossover steps. We feel that the product losses are not caused by the chemical methods used but rather by the lack of equipment; some of the equipment must be used repeatedly for several different steps in the process. This will not be the case at Hanford where each piece of equipment will be used for only one step in the process. The decontamination factors obtained at Clinton check very well with our results. However, the activity is still too high for the product to be handled safely in Room D. Two ways to increase decontamination were discussed: (1) follow the bismuth phosphate by-product step with a lanthanum fluoride by-product decontamination step which should increase the decontamination factor by a factor of 20;

(2) the use of scavengers, such as $Ce_3(PO_4)_4$ and Cb_2O_5, in the bismuth phosphate by-product step which should increase decontamination by a factor of 10. The scavengers MnO_2 and LaF_3 do not handle as well as Cb_2O_5 and ceric phosphate.

The Clinton pile is presently operating at approximately 600 kw. Operation at a higher level is prohibited because of inadequate cooling. The aluminum cans cannot stand temperatures greater than 200°C without developing pinhole leaks allowing the uranium inside them to corrode. If this should occur, the uranium slugs would swell and not be removable from the pile.

It was pointed out that Hogness and I plan to coordinate the work between Clinton and here. The Clinton semiworks group has identified many problems that require study, and our Chicago semiworks group has agreed to help. Because of the speedup at Site W, we do not want to duplicate work and thus waste time. Four of the problem areas that will be worked on here are: (1) a study of the effects of the Hanford aluminum-silicon slug coatings on the process; (2) a search for an alternate oxidizing agent that will not give inconsistent results in stainless steel; (3) studies on general problems involving Hanford concentrations of plutonium and fission activity; and (4) the investigation on a semiwork scale of alternate methods of concentration involving hydrochloric acid.

After my announcements I turned the meeting over to Thompson to preside. Dreher reported on decontamination results on a semiworks run of Argonne metal using bismuth phosphate and $U(C_2O_4)_2$. A decontamination factor for gamma radiation of about 400 was obtained in the final $U(C_2O_4)_2$ precipitate.

Watt described solubility measurements on plutonium peroxide. The solubility of plutonium peroxide in water is about 20 milligrams of plutonium per liter. Five measurements using 0.8 N nitric acid and 5% H_2O_2 over periods of time ranging from 15 minutes to 2 hours gave solubilities ranging from 300 to 500 milligrams per liter. Questions that still need to be answered are whether or not Fe^{+3} interferes in the peroxide precipitation; whether or not neptunium is carried by plutonium peroxide; and whether or not plutonium phosphate, which might be used as an alternate to plutonium peroxide, is soluble in hydrochloric acid.

Gordon Johnson reported that he has just completed an experiment on the 0.5 cc scale to determine whether or not the high concentrations expected at Site W will cause poor carrying with bismuth phosphate. The plutonium concentration was 25 mg/l; a total of 12.5 micrograms was used. Site W concentrations of fission products, in the form of their non-radioactive isotopes, were added. The bismuth phosphate precipitate showed 97% recovery of plutonium. In discussing extraction and decontamination, Thompson remarked that Gordon Leader has found that even at high concentrations of inactive rare earth holdback carriers there is no improvement in decontamination. Dean stated that a bismuth phosphate by-product precipitate dissolved in 10 N nitric acid was complexed with an excess of citric acid, and diluted to less than 1 N nitric acid without a precipitate forming. This procedure should help in disposing of the

waste without either the concentrated nitric acid attacking the pipes or the bismuth phosphate clogging them. Thompson remarked that the recovery of uranium is a problem to be faced at some time in the future.

A good deal of work has been done with $H_2C_2O_4$ as an alternate reducing agent; it appears successful in all the steps except possibly the prereduction step. However, the lanthanum fluoride precipitates for plutonium analysis are heavier than usual. This reducing agent has been tried in the semiworks at Clinton and seems successful.

After the meeting, Fred Albaugh and the Thompsons dropped in to our apartment for a visit, and Helen served coffee and tidbits. Stan and I were glad to see Fred, whom we knew as a fellow chemistry major and fraternity brother in our professional chemistry fraternity at UCLA.

This is a presidential election year, and today the Republicans announced that they will hold their convention in Chicago.

Thursday, January 13, 1944

I gave an orientation lecture to the new members of our section the first thing this morning.

Herbert H. Hyman, who started work with the Met Lab on January 6, transferred to Section C-I today. After receiving his M.S. degree from Brooklyn Polytechnic Institute in 1941, Hyman worked for the Huntsville Arsenal at Huntsville, Alabama, until he joined the Met Lab.

Our special monthly meeting with Charles A. Thomas on the "Coordination of Research on Final Purification and Metallurgy of Product at the Metallurgical Laboratory with the Program at Site Y" was held today. The opening session of the meeting began at 10 a.m. in Room 209 of Eckhart Hall. In addition to myself, those present were Allison, Chipman, Franck, Hogness, Kennedy, A. V. Peterson, Potratz, Cyril Smith, Thomas, and Warner. Allison, in his introductory remarks, said that since plutonium is now becoming more abundantly available, coordination of the Met Lab work with the program at Site Y is urgently needed. The objective of these meetings is to accomplish this coordination and to review personnel requirements as they relate to possible transfers of men from Chicago to Site Y. Thomas pointed out that Site Y is committed to getting plutonium into its final usable form as soon as possible after quantities become available. Because of this, Site Y needs to make use of the best available techniques and personnel as no time is available for pathfinding research or training. Basic exploratory work will be left to Chicago, Ames, and Berkeley. I, together with Hogness, Franck, and Potratz, pointed out that we appreciate the importance of the program and needs existing at Site Y and that we expect that as techniques and processes of value are developed at Chicago, these techniques and some of the persons acquainted with them will be transferred to Site Y. We emphasized that Chicago, however, still has definite and important responsibilities to the projects at Site X and Site W. It was pointed

out that we still do not have a sure-fire separation process for Site W and that this responsibility cannot be neglected if we wish to insure a successful process that will deliver the plutonium to Site Y. Also, analytical methods for plutonium still require perfection. We expressed the opinion that a definite plan upon which all can agree is essential, so that the need for transferring men to other locations can be determined sufficiently in advance to permit adequate planning. Chipman agreed that such a plan must be developed if Chicago and Site Y are to meet their obligations in the most effective manner. An allocation of these problems to Chicago and Site Y should be made. Chipman pointed out that desirable techniques and processes do not just happen--they result from planning. If we have a definite coordinated plan, transfers will seem logical to both groups and the need for transfers of personnel will be seen ahead of time. Smith and Kennedy, representing Site Y, expressed concern for the time limits imposed upon their program. There are many things which must be done before anything like gram quantities of plutonium are available, and they need all the help Chicago can give them in the way of techniques and personnel. Work at Site Y must be confined to the most promising methods and processes. For instance, information on the melting point and density of plutonium and plutonium compounds is urgently needed.

Following the opening session, the group split into two working groups--one on chemistry and micro-metallurgy and the other on metallurgy. I attended the session covering "Research Program of the Chemistry and Micro-metallurgy of 49" which began at 11:10 a.m. Also, in attendance were Allison (part time), Franck, Hogness, Johns (part time), Kennedy, Potratz, and Thomas. Following a suggestion by Thomas, the various problems related to the Site Y program were listed and discussed. The purification and metal production program outline (MUC-GTS-391) prepared yesterday by Manning and Orlemann (which I distributed during the opening session) was used to present the problem areas in the Chicago program that are related to the Site Y program. The items on the outline were discussed in the reverse order beginning with studies on the metal and ending with analytical techniques and other problems. Priorities were assigned to the projects and suggested plutonium allocations were made. In general, our Chicago Laboratory will conduct experiments on the micro and sub-gram scale. Gram and larger quantity experiments will be conducted at Site Y with plutonium or stand-ins. Density and melting point determinations of the metal were considered of high priority, with lower priorities assigned to hardness and vapor pressure determinations. It is expected that the samples of plutonium used for making the density determinations here in Chicago will be furnished to Site Y for oxygen analyses. Site Y is presently doing no work on the preparation of pure compounds of plutonium; so this work will be continued at Chicago. Of the spectrochemical analysis methods discussed, the copper spark method for light elements was considered of highest priority. With regard to chemical analytical procedures, methods for fluorine analysis, taking plutonium metal into solution, and vacuum-fusion methods for oxygen analysis were considered of highest priority. It was decided Chicago should duplicate the Site Y vacuum fusion apparatus and that duplication of effort in this area is highly desirable. Analytical methods for boron, phosphorus, sulfur, carbon, chlorine, and oxygen were given lower priority but will

be continued at about the present rate. Kennedy agreed to send Franck a sample of uranium wire already analyzed for oxygen. This will serve as a standard sample for other methods of oxygen analysis.

The session covering "Research and Development Program on the Metallurgy of 49" ran concurrently and was attended by Cyril Smith, Chipman, and Warner, and later by Allison and Johns. An outline for a comprehensive program of research and development work on the metallurgy of plutonium was prepared. Time schedules and assignments of responsibility for each problem area were discussed and agreed upon. Those in attendance wished to call the attention of the Chemistry Division to the fact that the success of the metallurgy program is very dependent upon the development of satisfactory methods for the analysis of materials involved in the metallurgical processes. In addition, the Chemistry Division must provide competent analytical services and develop methods for the recovery of plutonium from scrap, refractories, slags, and other waste materials. Chipman expressed the opinion that the obligations assumed by Site Y in the metallurgical program represent a job of considerable magnitude, and he questioned whether the number of metallurgical personnel at Site Y is large enough for this undertaking.

The joint closing session began at 4 p.m. in Room 209 of Eckhart Hall. In addition to myself, those in attendance were Allison, Chipman, Hogness, Johns, Kennedy, A. V. Peterson, Potratz, Cyril Smith, Thomas, and Warner. Thomas summarized the meeting, and it was generally agreed that the meeting had been highly useful in arriving at a unified program through the allocation of specific problem areas. All agreed to the recommendation that Chicago should duplicate the vacuum fusion method for oxygen analysis developed at Site Y. Kennedy will provide Hogness with all of the details. Also, studies on alternative methods for oxygen analysis will continue. Thomas suggested that the future monthly meetings on Final Purification and Metallurgy of Plutonium be confined to those in attendance at this meeting, plus Spedding from Ames, and a Berkeley representative to be designated by Compton.

Thompson summarized for me in a memo the recent conversation he and I had regarding possible experiments to be tried in connection with the extraction, decontamination, and concentration work. It covers the following: Further development work is suggested on the discovery made by Dean that citric acid will prevent precipitation of bismuth phosphate when nitric acid solutions containing bismuth phosphate are neutralized. The use of tartaric acid as a substitute for citric acid should also be tested. Attention should be given also to the recovery of uranium from waste solutions by means of ammonium uranyl phosphate precipitation. Thompson conducted preliminary experiments early last year that involve the addition of diammonium phosphate to the waste solution. A third experiment, which Watt mentioned at last night's meeting of the Extraction Sub-section, would be to test whether or not neptunium is carried by plutonium peroxide. Arrangements will be made with Cunningham to get some neptunium tracer for these experiments. After the reinvestigation of the peroxide method with respect to Site W operation, two alternative methods will be considered that could possibly be substituted at Hanford. The first of these is the precipitation of plutonium fluoride from

hydrochloric acid solutions of bismuth phosphate, as well as the precipitation of plutonium oxalate from similar solutions. The possibility of substituting the direct precipitation of plutonium hypophosphate from lanthanum nitrate solutions will be considered as the second alternate. Some work will be done to determine whether plutonium exists as a soluble phosphate or as a separate phase in the hydrochloric acid-bismuth phosphate solution at Hanford concentrations. The problem of whether the peroxide method will work during the early stages of operation at Hanford when the plutonium concentrations are low will need investigating. Possibly the hypophosphate method can be used in place of the peroxide method if the latter is found unsatisfactory. A yet unidentified insoluble precipitate has given some trouble in the first semiworks runs involving the precipitation of lanthanum fluoride to carry plutonium from the hydrochloric acid-bismuth phosphate solution. Additional studies are required on this problem.

The memo goes on to say that when more plutonium is available for G. Johnson's work, some experiments will be undertaken in the attempt to introduce more plutonium into a bismuth phosphate sample than that present in the sample recently prepared for x-ray study by Mrs. Mooney. It was Mooney's opinion that no conclusive results could be obtained from this first sample since the percentage of plutonium in it is too low (3.6%). Johnson plans to make a 10 cc scale run with "W" concentrations of plutonium and twice-"W" concentrations of fission products. Dean may be asked to assist in these experiments.

Thompson also states that he and Watt have had discussions with Cunningham concerning possible experiments to be performed on the 15 milligrams of plutonium that we expect to receive soon. The plutonium nitrate solution to be supplied should be quite low in nitric acid concentrations, and therefore it is hoped that during sample preparation, the solubility of plutonium nitrate can be determined.

I received a memorandum from Beaton giving the current status of contacts his group has made with three adsorbent manufacturers. (1) The Research Products Corporation (Madison, Wisconsin) contacts are Messrs. K. Slidell, M. J. Shoemaker, and M. Klieforth. A two-month subcontract with their company, just terminated, resulted in the production of a titanium zeolite that shows some promise when used as a decontamination adsorbent after Amberlite IR-1 extraction of plutonium. (2) The Resinous Products and Chemical Company (Philadelphia, Pennsylvania) contacts are F. J. Myers, R. J. Myers, D. S. Herr, and L. C. Hurd. Amberlite IR-1 is made by this company and approximately 1,000 other such adsorbent materials have been prepared by their company to date. F. J. Myers is anxious to arrange a conference to discuss separation efficiencies in the hopes that one or more of these materials will be of specific value to our program. (3) The Chemical Process Company of San Francisco, for which G. F. Mills is the contact. Mills was here on Tuesday and conferred with Beaton and others. He volunteered to attempt a laboratory synthesis of certain types of highly specific ion-exchange resins. A set of samples of these materials are to be forwarded by him to Chicago in the near future.

Willard, as Associate Section Chief, wrote to Colonel John E. O'Gara, who is chief of the Labor Branch in the Industrial Personnel Division at Headquarters, Army Services Forces in Washington, D.C. It concerns the need for the continuing services of Joe Halperin who has been with us since June 21 of last year and was ordered last Thursday to report for induction into the Army. Willard urgently requests that every possible step be taken to have Halperin reassigned or released for duty on the Project before January 27, the date when he is being ordered to go to camp.

Recommendations were submitted by Allison to General Groves concerning the future relations between the Met Lab and the Canadian group working in Montreal under the direction of H. von Halban. These include procedures for exchanging letters and reports and suggest arrangements for future visits by members of the Canadian group to Chicago and by Chicago laboratory personnel to Montreal.

The meeting of the Research Assistants of Section C-I was held at 6:45 p.m. in Room 209 of Eckhart Hall.

U.S. marines repelled a Japanese attempt to land reinforcements at night at Cape Gloucester on New Britain.

Friday, January 14, 1944

I called a meeting in my office the first thing this morning to go over with my people the urgent assignments resulting from yesterday's meetings. The key people present to help plan our program were Kirk, Willard, Baumbach, Manning, Cunningham, Orlemann, and Davidson. I informed them that we will be getting all of the plutonium shipment leaving from Clinton Laboratories on January 17, which may be as much as 20 milligrams. I indicated that the assignment of this will be essentially the same as we decided Tuesday night. We should have a decision soon on the allocation of the gram amounts expected to be available in the not too distant future. Kennedy at Site Y would like, as the first thing, to reduce PuF_4 with calcium on a gram scale and then see what we can do to remove the impurities present. He will count on us to do scouting work on all the items in the Manning-Orlemann report (MUC-GTS-391). Of great importance to Site Y will be Meyer's and S. Katz's studies concerning the preparation of the correct form of PuF_4. We must make density, melting point, vapor pressure, and hardness determinations on plutonium metal as first priority. We plan to lend the chemists at Site Y about 100 μg of plutonium metal by January 23 so they can analyze the sample for oxygen. Jasaitis will continue with the melting point work, and Rosenfels will proceed with hardness determinations. In general, we will continue to do micrometallurgy work here; we will continue the four methods for production of plutonium metal--(1) reduction by electropositive metals (Fried, Dirksen, Jasaitis), (2) hot wire technique (Jasaitis), (3) electrolysis (S. Katz, Zvolner), (4) reduction by atomic hydrogen (Karle). Chipman's people at Site B will work on these methods on the gram scale, and Eastman's group at Berkeley will work on the electrolysis method on the gram scale. Hellman and Jasaitis will work in refractories using plutonium on the

micro-scale, and Chipman's and Eastman's people will work with refractories on a larger scale; the main effort will be in Chipman's group at Site B. Work on the hydride is almost of equal priority; Fried, Dirksen, and Jasaitis will attempt to determine its formula, structure, density, methods of production, and equilibrium pressure. It is expected that any work involving quantities of plutonium in the approximately 500 gram range will be handled exclusively at Site Y.

It has been decided that all the work on the small scale recovery of plutonium for re-use, for the whole Project, will be done by our Recovery Group (Dawson's group) here in Section C-I. The question of future large scale recovery of plutonium will be decided later; perhaps it will be done here. It is possible that the electroplating of the final device will be done in Chicago.

In summary the assignment of the main tasks for the next month will be as follows: Kirk and Baumbach--general supervision; Rosenfels--hardness; Jasaitis--melting point; Zvolner--electrolysis; Karle--atomic hydrogen; S. Katz and Meyer--preparation and identification of proper PuF_4; Fried and Dirksen--reduction with electropositive metals; Hellman--refractories. S. Katz and Jasaitis will also have other assignments as outlined in our discussion.

It has been determined that the regular meetings of the inter-laboratory Purification and Metal Production coordinating group (the Thomas meetings) will be held each month on the Monday preceding the regular monthly meeting of the Project Council Information Meeting on Chemistry which is held on the third Tuesday of the month.

When I visited Perlman at Oak Ridge early this month I was greatly concerned with the health hazards involved in the handling of plutonium. Today I wrote him a letter emphasizing the need for new procedures. I suggested that he should regard the handling of plutonium as a very real problem and take necessary steps to guarantee the health of the men in his section, for whose safety he is responsible. I also recommended that he confer with Dr. Cantril as soon as possible to be sure that adequate safety steps are being taken immediately. I said it might be necessary to institute protective measures of the type used in the radium dial painting industry within the plutonium isolation rooms at Clinton. In regard to these protective measures and the health hazards of alpha emitters, I cited articles by Evans and others in the September 1943 issue of the Journal of Industrial Hygiene and Toxicology and suggested that he read them.

R. J. Myers, Director of the Resinous Products and Chemical Company, met with Beaton here today to discuss the possibility of his company supplying an ion-exchange resin capable of purifying plutonium (described to him as thorium). Myers discussed several types of resins that might have better selective adsorbent properties than Amberlite IR-1. A phenolformaldehyde resin containing carboxylic acid exchange groups should be more selective than a resin containing sulfonic acid groups, as does Amberlite IR-1. The company has also prepared a phenolic-substituted sulfonic acid resin that has adsorption characteristics quite

different from Amberlite IR-1. A melamine sulfite type of ion-exchange resin is apparently capable of forming intercomplexes with certain cations. Samples of all the above materials are available for testing and will be sent to us in the near future. Polyvinyl butyral resins have been made which act as specific adsorbents for boron, and these quite possibly might be applicable to plutonium. Myers suggested a meeting in Philadelphia within the next two weeks, at which time other security-cleared members of his staff could be available for more detailed discussions.

Allison wrote to General Groves in Washington, D.C., presenting a statement of the Met Lab's understanding about the transmission of reports and information to the Montreal group. The categories of information involved were outlined as follows:

"I. It is our understanding that we are to make available to the Canadian group information and reports covering the following topics.

1. General Nuclear Physics such as cross sections, neutrons per fission, fission energy spectrum, nuclear reactions, pile theory, mass spectroscopy, etc.

2. General Physics and Chemistry such as heat transfer, physical properties of materials, instrumentation, properties of slurries, chemical properties, fission products.

3. General Engineering such as corrosion of metals, heat exchangers, pumps, shielding (in general), coating and canning, etc.

4. Specific information on P-9 (heavy water) properties, analysis, tentative designs for P-9 piles, slurry pumps, etc., P-9 production schedules.

"II. It is our understanding that we are to withhold information covering the following topics.

1. Exact nature of X or W piles, including lattice spacing, method of cooling, method of shielding, power rating, controls, specific instrumentation.

2. Anything in the nature of time schedules for X or W, or performance data from the pile or chemical plant at X, or production rate of tube-alloy metal.

3. Chemical and physical properties of 49.

"III. We are not clear as to the following topics, but shall assume them on the forbidden list until further instructions.

1. Extraction and purification of product (plutonium), that is, actual processes as distinguished from chemical properties."

The California Project Report (CN-1233) "Chemical Research--Chemistry of Plutonium, Report for Month Ending January 10, 1944" by Latimer, Eastman, Connick, and Gofman, is being issued. H. Crandall, C. Crompton, G. Pimentel, W. Reas, and F. Stevenson report on their work

relating to the coupling of the thorium oxalate procedure to the sodium uranyl acetate process. The two proposed procedures being investigated are described as follows:

"One method consists of dissolving the thorium oxalate precipitate in an excess of $(NH_4)_2Ce(NO_3)_6$, destroying the excess Ce^{+4} ion with Cr^{+3}, and then precipitating sodium uranyl acetate by adding UO_2^{++}, $NaNO_3$, and NaAc. $(NH_4)_2Ce(NO_3)_6$ is known to oxidize Pu^{+4} very rapidly in the cold. The plutonium becomes oxidized by Ce^{+4} and is therefore carried by the $NaUO_2Ac_3$ precipitate. The thorium oxalate precipitate is readily dissolved through the oxidation of the oxalate by Ce^{+4}. It was found, in fact, that thorium oxalate could be titrated quantitatively with a solution of $(NH_4)_2Ce(NO_3)_6$ in 1 M HNO_3 if the solution was heated to 40-50°C. The indicator used was nitro-ortho-phenanthelein ferrous sulphate. Further studies showed that this reaction was complete within 30-60 minutes at room temperature..."

"Another proposed method for coupling the $Th(C_2O_4)_2$ extraction to the $NaUO_2Ac_3$ method is as follows: The $Th(C_2O_4)_2$ precipitate containing the Pu^{+4} from the extraction step is dissolved with $Na_2Cr_2O_7$ or $(NH_4)_2Ce(NO_3)_6$ plus $Na_2Cr_2O_7$. The Pu^{+6}, $Cr_2O_7^{=}$, and/or Ce^{+4} are reduced with Fe^{++} or SO_2. The acid concentration is raised to 4 M destroying any Fe^{++} present. Oxalic acid is then added, reprecipitating $Th(C_2O_4)_2$ which carries the Pu^{+4}. This $Th(C_2O_4)_2$ precipitate is dissolved with more $(NH_4)_2Ce(NO_3)_6$ which oxidizes the plutonium. The Ce^{+4} is destroyed with Cr^{+3} as described above, and $NaUO_2Ac_3$ is preciptated carrying Pu^{+6} by adding UO_2^{++}, $NaNO_3$, and NaAc. This method has the advantage of possible increased decontamination caused by the reprecipitation of $Th(C_2O_4)_2$ out of high acid and a higher oxalic acid concentration. This is possible because of the absence of UO_2^{++} at this point. The rare earth oxalates are more soluble in the higher acid, and columbium and zirconium form soluble oxalate complexes which are more soluble in the higher oxalic acid concentration."

Sheline reports on the carrying of plutonium by thorium oxalate at high plutonium concentrations. He presents data on carrying or coprecipitation of Pu^{+4} with $Th(C_2O_2)_2$ at Pu:Th ratios up to 1:2.31. The results obtained show that at each ratio of thorium to plutonium studied, the carrying of Pu^{+4} by the $Th(C_2O_2)_2$ precipitate is 97% or better.

L. J. Beaufait, Jr. and A. M. Stein report on "Coupling of the Amberlite IR-1 Adsorption Process to the $NaUO_2Ac_3$ Process." Work done by Boyd's group in Oak Ridge has shown that the Amberlite IR-1 adsorption process can be coupled to the sodium uranyl acetate process for decontamination. However, high sulfate concentration in the column elutriants preclude the use of $Cr_2O_7^{=}$ as an oxidizing agent in the first sodium uranyl acetate cycle and also makes necessary a high acetate concentration for complete precipitation of the sodium uranyl acetate. A $La(OH)_3$ crossover step has been investigated that should eliminate both of these difficulties. The plutonium carried by the $La(OH)_3$ precipitate can be removed from the elutriant solution and dissolved in nitric acid to give a sulfate-free solution for the sodium uranyl acetate process. A flowsheet is presented for the crossover process that summarizes the results obtained in their investigations on the distribution of plutonium in the entire cycle.

A 99% plutonium recovery is observed in the crossover step which is encouraging. However, no beta or gamma active fission products were used in the experiment, and it is recommended that decontamination in the Cb_2O_5-$NaUO_2Ac_3$ cycle that follows the crossover should be checked using the actual Amberlite elutriant solutions.

The Drehers, the Baumbachs, Helen, and I took the Beatons to Gus' Good Food Restaurant for dinner. Roy is transferring to Clinton Laboratories soon. We then went to see the Olson and Johnson stage show "Sons O' Fun" at the Civic Opera House. After the performance Dagmar Dreher took us backstage to meet her friend in the cast and Olson and Johnson.

A report from Berlin in today's paper says that the outskirts of Rome have been bombed by ninety Allied planes.

Saturday, January 15, 1944

This was the last day of service at the Met Lab for Sydney Gaarder and James Kroner. Gaarder and Kroner are transferring to the Clinton Laboratories and will report for duty in Knoxville on Monday. They are both single and will reside in a men's dormitory.

A letter was sent to General Groves, over the signatures of Compton and Whitaker, to inform him that the work of Private Halperin is considered of sufficient urgency that Halperin's release from the Army to the Enlisted Reserve Corps should be expedited. Groves was notified that they are sending a request and endorsement to Colonel O'Gara, of the Army Service Forces, for Halperin's release so that he may be permitted to continue his work at Clinton Laboratories as a member of the Enlisted Reserve Corps.

I received a copy of a memorandum from Hogness to Vernon on the control of (accounting for) plutonium and plutonium metal. Within the Chemistry Division the responsibility for control of both plutonium and neutron-irradiated uranium metal will be assigned to me and my section. The plutonium will be controlled through Cunningham and uranium metal through Thompson. Because nearly all of the plutonium passes through Dawson's group (supervised by Cunningham), it is logical that his group also check on the material. It is anticipated that most of the uranium metal will be received in the form of slugs, processed and checked by Dreher's group under Thompson's supervision (which will cooperate with the semiworks group in Cooper's Division). Hogness points out that the plutonium as received in the form of the active uranium metal slugs will not in all cases be isolated but will be retained in the form of concentrates. He thinks it advisable to have the hot uranium slugs from Site X consigned to me so that Section C-I may be able to keep check on them from the very beginning.

I also received a copy of a memorandum from Charles Cooper to Bellas confirming the cooperative arrangement between the Chemistry

Division and the Technical Division with regard to studies of coating removal and of the effect of coating residues upon separations and decontamination processes. Beaker-scale methods for removing coatings bonded with aluminum silicate (also zinc and zinc-tin) will be developed by the semiworks group. The methods will be checked by dissolving coated inactive slugs in the semiworks dissolver with the resulting solutions activated by the addition of active solution. Then separation and decontamination steps will be carried out both in the Technical Division's semiworks and in the Chemistry Division. Finally, coated slugs will be exposed in the Clinton pile to provide material for ultimate tests, which may be made either at Chicago or Clinton Labs, or both.

I outlined for Hogness the problems under investigation in the Physical Measurements Group of Section C-I. This was in answer to an earlier request from Doan for such information. Seventeen tasks or problem areas are described which include: Jaffey's supervision of ether extractions for plutonium isolation; the development by Bert Schoenberg of a plutonium-finding alpha counter for checking for alpha contamination; the maintenance of alpha counters by Schoenberg, Crawford, and Ghiorso; the maintenance of Geiger-Müller counters by B. Scott, P. Walsh, and L. Heller; Geiger-Müller counter tube research by Scott, Walsh, and Heller; the development of an alpha magnetic counter to satisfy Hanford requirements (most of this work will be done at Clinton because of the shortage of manpower within the group since Kirk is preparing for his move to Hanford); the preparation of a manual on the measurement of radioactivity by Jaffey, Kohman, and Crawford (work practically completed); the compilation of experimental data for a complete table and chart of the isotopes and the writing of a section on Radiochemical Techniques for the Plutonium Handbook by Kohman; assistance to Monsanto Chemical Company for setting up of a counting room (Scott has recently been spending 90% of his time in Dayton on this work); the development and application of alpha-range measuring instruments needed for the determination of ranges of possible new alpha-emitting isotopes formed in the pile so that their existence can be identified (this work is being done by Jaffey and Crawford and includes work on spherical pressure-type chambers for use with the present FP-54 electrometer and a differential ionization-type detector for use in connection with special counting equipment)--these instruments can also be used for determining composition of any mixture of alpha emitters such as the ratio of 93^{237} and 94^{239} in mixtures formed during the study of methods for the separation of neptunium from plutonium; the development by Kohman of high-efficiency neutron counting methods using pressurized BF_3 ion chambers for determining total neutron-emitting impurity in final product; the development of BF_3 proportional counters for detecting neutrons as an alternate to the above method by Schoenberg; the development of high-efficiency fission counters for detecting neutrons using UF_6, or possibly surfaces coated with uranium enriched in U^{235} (this work has not yet been started because of insufficient manpower and time); determination of the resolving power and coincidence losses of alpha counters (initial work has been completed); the determination of the half-life of 94^{238} by measurement of its decay by Jaffey and others; the construction of a nitrogen high resolving power alpha counter designed

at Site Y and the evaluation of this counter for possible use at Hanford (most of the details will be worked out at Clinton Labs since manpower is limited here); and the investigation by Crawford, Ghiorso, and Cunningham of backscattering of alpha particles as it affects the determination of counting yield and geometry of parallel plate alpha chambers and other alpha detectors.

I prepared and sent another memorandum to Stone about health hazards in working with plutonium. The text of this letter is as follows:

"Since receiving your memorandum on the above subject (dated January 8), I have given further thought and have held discussions with a number of people on this subject. Also, I have read the articles by Evans and others in the September 1943 issue of the Journal of Industrial Hygiene and Toxicology. As a result I am seriously worried about the health of the people in my section, for which I am responsible, since they will soon handle such relatively large amounts of plutonium. I wonder whether some plutonium should be made available to Dr. Hamilton for his distribution studies sooner than the couple of months or more indicated in your memorandum. I would like to suggest that five or ten milligrams out of the first one-half gram or so of material from 'X' be allocated to this purpose. In this way the experiments should get under way in about a month or so, which will be not too soon in view of our time schedule and the length of time estimated as required for these experiments.

"This problem of health hazards assumes even greater importance for Site Y where so much plutonium will be handled in so large a variety of operations. It is, of course, also important in connection with the operations which will go on at Site W, particularly those involved in its final isolation there."

I sent Vernon the Chicago Chemistry Division program for receipt of neutron-irradiated Site X uranium metal. The memorandum confirms the arrangements made with Whitaker by Hogness and me while at Clinton on January 7. Kircher has completed arrangements for the production, storage, and shipment of uranium slugs through discussions with H. M. Parker, who is now stationed at Oak Ridge, and Edward Sinclair. Sinclair has arranged for 24 slugs to be pushed from the pile each week from a channel located in a position to yield metal having the desired plutonium concentration. The first special batch of material should be ready for shipment around February 15. Sinclair will provide us with the essential details regarding the history of this and each future batch. Weekly shipments of six boxes containing four slugs each are indicated in the program. Shipments, however, will be made only by specific request and after obtaining proper authorization. I said that radiation exposure conditions, as they pertain to the packing and shipping of the material, have been discussed with Parker and criteria are now established.

The second lecture in the series for new members of Section C-I was presented by Thompson. The purpose of the lecture was to further acquaint the new individuals with the background of the Project and to familiarize them with the work that has been accomplished. The current

knowledge and experimental results concerning the chemistry of plutonium and general information on the production and separation process were presented. Watt also talked to the group about the research program on the development of methods for the isolation of plutonium, the last step in the chemical process for separating plutonium from uranium and fission products.

Stan and I caught the 11:55 p.m. Pennsylvania Railroad train from Englewood Station to Cincinnati, where we will continue on L&N R.R. to Knoxville. We plan to meet with Iz Perlman and the people in his section, and I will attend the Project Council Information Meeting on Chemistry and Technology to be held at Clinton Laboratories on Tuesday.

Sunday, January 16, 1944

Thompson and I arrived in Knoxville this afternoon. I spent the night at the Vance Coopers'.

Monday, January 17, 1944

Thompson and I met with Iz Perlman and the people in his section. We also had the opportunity to review the performance of the Bismuth Phosphate Process in the chemical separation plant including a tour of the plant. I was particularly interested to inspect the concentration procedure in Room D and the ingenious procedures for the isolation of plutonium being carried out by Vance Cooper, Louis Werner, and their groups. After the Cooper group performs the initial isolation steps, in which all kinds of unexpected impurities and debris must be handled, the final isolation, usually as the peroxide, is completed by Werner. Werner has the distinction of being the first man in history to handle in the pure form a relatively large output of plutonium from a production plant.

There was no Chemistry Division seminar this evening in Chicago because of the large number of people attending the meetings here at Clinton Laboratories today and tomorrow.

Tuesday, January 18, 1944

I attended the first Project Council Information Meeting on chemistry ever to be held at Clinton Laboratories. Others in attendance included M. F. Acken, Allison, Boyd, Brown, Burton, C. V. Cannon, Cantril, Chipman, Compton, C. M. Cooper, Coryell, Doan, Gofman, Greager, Greenewalt, A. B. Greninger, Hogness, Jeffries, Johns, W. C. Johnson, Kay, Latimer, Leverett, Mulliken, Murphy, Perlman, Potratz, Pratt, E. Shapiro, D. Smith, Spedding, Sugarman, Sutton, W. W. Watson, Whitaker, Wigner, Wilhelm, and later A. V. Peterson, and Wensel. The meeting was opened by Compton at 8:00 a.m. He, Whitaker, Allison, and Doan gave opening remarks and

welcomed the Council and the Chicago, California, and Ames delegations to Clinton Laboratories. Hogness introduced the speakers from Chicago who, in addition to myself, included Burton, Potratz, and Sugarman.

I was the first to report. I highlighted results of work we have completed on the 0.5 cc scale of the Bismuth Phosphate Process at Hanford concentrations, half-life measurements on plutonium-239, density determinations of plutonium metal, and HCl concentration methods in the semiworks. I reported that the process work done at Hanford concentrations of plutonium and fission products results in an extraction precipitate recovery of about 95 ± 5% and a final product precipitate recovery of about 97 ± 5%. Measurements made in October 1942, on the basis of a supposed counting yield of 45%, gave a value of around 20,000 years for the half-life of plutonium-239; in January 1943, a half-life of 21,300 years was determined assuming a 47% counting yield. Recently, using absolute geometry to calibrate our high geometry counters, we have determined that the counting yield is actually 52%, which can be explained by alpha backscattering of two kinds; thus, on the basis of the latter counting yield, the half-life of plutonium-239 is now established as 24,000 years. The specific activity of uranium has also been measured, giving 1,515 disintegrations per minute per milligram (which can be favorably compared with 1,500 dis/min/mg obtained by Kovarik and Adams, and 1,517 dis/min/mg reported in recent German determinations).

I reported that several runs, involving the reduction of plutonium tetrafluoride with lithium and barium, have resulted in black powder samples all giving a density of 13.01 gm, assuming the product is metal. The determinations were made by Zachariasen using x-ray diffraction techniques. Reduction of plutonium tetrafluoride with lithium at 1,000°C gave a black powder, reduction with barium at 1,100°C also gave a powder, and reduction with barium at 1,600°C resulted in small solid particles. This latter product was previously used by Kirk and gave a density greater than 15.5 gm/cm^3, using the capillary technique. I expressed my suspicion that the plutonium tetrafluoride used was actually $PuOF_2$, and that the black powder product of reduction is PuO, and not metallic plutonium. I based my suspicions on the black color and the fact that when the product was returned by Zachariasen to Kirk, it could not be transformed with H_2 to PuH_3. All this plutonium density work was done with a total supply of one milligram.

Continuing my report, I said that our Chicago semiworks ran an extraction plus one decontamination cycle; and then in place of crossover, we dissolved the bismuth phosphate precipitate of plutonium in HCl. The plutonium was then precipitated directly with lanthanum fluoride in one run, and with uranous oxalate in a second run, obtaining a yield exceeding 90% in both runs.

Burton discussed radiation effects on graphite within the proposed Hanford pile. Sugarman reported on work he is starting on minor fission products using two slugs from the Site X pile having about 1-1/2 curies of activity. Among these products there is evidence for the existence of 7-1/2-day silver. Seven day-U^{237} also is present, showing a yield of 0.2% in pile operation. Fission product yields for 86-min. Ba^{139} and 12.5-day Ba^{140} have already been determined, which are believed to be

7.2% and 5.8% respectively. Ternary fission has been looked for and could have yields as high as 5%, but no evidence for this has been obtained so far. Sugarman also reviewed the results on the two forms of bismuth phosphate and described the effect of 0.13 M Fe^{+++} which completely inhibits the conversion of the more soluble to the less soluble form.

Potratz presented the analytical results obtained on two out of 250 samples of plutonium analyzed during the month. These were a 10-microgram nitrate sample and a 49-microgram hydroxide sample purified in my section. As the samples were too small for cupferron extraction, they were analyzed directly by sparking the samples on copper electrodes and photographing the visible and ultraviolet emissions simultaneously. Except for 3,000 ppm of aluminum, the trace concentrations of 16 other elements were below the limits of sensitivity within the nitrate sample. The only positively identified trace elements in the hydroxide sample were 2,000 ppm of lanthanum and indications of calcium and sodium. The total impurities were less than 7% in the nitrate sample and about 1% in the other. Since sensitivity is limited because of the strong plutonium spectrum, there is a necessity to use cupferron extraction to remove the impurities from the plutonium. Two milligrams of plutonium are being set aside to establish this latter technique for the determination of lithium, beryllium, magnesium, and other light elements.

Latimer introduced Gofman as the Berkeley speaker. The thorium oxalate extraction process, which is described in detail in the recent Berkeley Project Report CN-1233 (Report for month ending January 10, 1944) was discussed.

Spedding introduced the Ames speakers and said that he expects the Ames group to be divided shortly. Johns reported on the work that W. H Sullivan, Amos Newton, and Frederick J. Wolter are doing on the radiochemistry of ruthenium and rhodium, the volatilization of plutonium chloride from Boyd's elutriants, the preparation of uranium metal enriched in plutonium by metal reduction, and their attempts to volatilize plutonium from uranium slugs. The reduction of uranium hexafluoride to uranium tetrafluoride is being studied because of the difficulty of storage of the former. Ruff in early studies reported that hydrogen reduces uranium hexafluoride at room temperature, but present studies at Ames give only 30% reduction even at 600°C, very little at 300°C, and none at room temperature. The tetrafluoride produced at 600°C was jet black. Johns said that SO_2 does not reduce UF_6 in the gas phase; however, NH_3 reduces UF_6 violently upon introduction, even at dry ice temperature, but can be controlled and gives quantitative reduction to a green UF_4.

Wilhelm reported on the phase diagram work for several alloys of uranium. The reduction of uranium on a 100, 50, and 25-gram scale, and corresponding casting, has been thoroughly studied. The overall yield of the magnesium reduction of green salt (UF_4) to the finished metal has gone up from about 80% a year ago to 91.2% at the present time.

Johnson introduced the Clinton Laboratories' speakers. Boyd described the process development work on his adsorption technique for the plutonium extraction process. He has observed a slightly more than 100% extraction efficiency from 20% UNH with decontamination factors of 2 for betas and 3.5 for gammas. Higher concentrations of UNH (to at least

31% UNH) may be used if the diameter of the adsorption bed is increased. Two possible ways of obtaining decontamination during the adsorption extraction process were discussed: (1) complex the fission products so that they will not be adsorbed and (2) use selective washing techniques after complete adsorption of plutonium (factors of 400 to 1,000 have been obtained in this way on several runs). Boyd advocates use of adsorption extraction followed by sodium uranyl acetate precipitation of oxidized product, and a modified California flowsheet was presented.

Perlman reported that experience to date shows that plutonium can be recovered from the Clinton separations plant in good yield without excessive effort. Clinton Laboratories has delivered batches of 1.5, 1.0, and 22.5 milligrams. Concentrations in the latter stages of the separation process are comparable to Hanford concentrations, and no unexpected difficulties have occurred. On the one milligram sample, the gamma decontamination was about 2×10^7 after about two lanthanum fluoride oxidation-reduction cycles. On the basis of this it looks as if the Hanford decontamination requirements have been met. Perlman informed the group that the colors for plutonium compounds reported in these larger quantities are olive-green for the solid peroxide and greenish for the nitrate in dilute solution which becomes yellowish when more concentrated.

Coryell talked about his fission product work at Clinton Laboratories. By application of the Bohr-Wheeler picture, he feels reassured about the minor fission products in the range of palladium to antimony. He believes that uranium-236 may have a half-life as short as 10 days and is likely to be detected in the fission product work or in alpha decay measurements on uranium irradiated within the pile. (I did not mention James's recent experiments which seem to contradict this.)

Cannon reported the successful construction of an ether extraction system with which a 100-fold concentration of fission products was obtained. He expects to handle up to 100 curies using remote control. Studies on the formation of peroxide within the water circulating through the pile were described. A steady state concentration of 1.7×10^{-4} molar was observed with ordinary water in the Site X pile.

Greager introduced the speakers for the Clinton Separations Process Development Division. Sutton reported that a gamma decontamination factor of 10^3 is now being obtained prior to concentration steps in Room D of the chemical extraction plant, but the need is for 10^4 or 10^5. Three possible remedies were discussed: the use of direct instead of reverse strike, the use of a lanthanum fluoride by-product precipitation step in the crossover cycle, and the introduction of a scavenger such as ceric phosphate. Improvements in the crossover cycle may be made by introducing a reduction procedure using oxalic acid, or by carrying out metathesis in the tank instead of the bowl. Both procedures have proved satisfactory on the basis of 10 semiworks runs. Sutton discussed the concentration procedure and said that yields of 90% to 96% for one complete lanthanum fluoride concentration cycle were obtained in two semiworks runs, using dichromate plus bismuthate oxidation and using Perlman's Mn^{II}-catalyzed oxalic acid reduction at 35°C.

Acken discussed the yields obtained in semiworks runs made with standard flowsheet conditions. Measured losses totalled a little over 26%; product recovery was 47%; an additional 22% was accounted for when both precipitators were washed with strong acid, which shows that bismuth phosphate adheres firmly to the walls of the vessels. During the discussion period that followed Acken's report, I pointed out that the total of 95% of product accounted for is in agreement with the semiworks results obtained in Chicago, where our vessel walls are also carefully cleaned.

Doan mentioned that an urgent problem recently arose which led to the formation of the new Analytical Control Division that will have responsibility for all routine analyses and for developing new controls. Doan introduced D. Smith who said the work of the Analytical Control Division falls into two areas, analytical work and improved procedures. The routine analysis of alphas and betas plus other assays, including the use of physical methods, are included in the analytical work. The development of analytical and sampling equipment and the development and analysis of ion improvement methods are to be included in the improved procedures work. The Division will consist of the Control Section, Special Analyses Section, and the Analytical Improvement Section.

Kay, in discussing Plant Operations, said, "It is a privilege to report on the successful operation of the first industrial plant for the manufacture of a synthetic element." He described the deliveries and production rates to date. The overall yield of 59% through the decontamination and crossover step compares with 57% obtained in the last ten runs in the semiworks. The decontamination factors likewise are of the same magnitude as were obtained on the semiworks scale. The unaccounted-for losses also are similar to semiworks results. From these data it can be concluded that the semiworks is a good guide to plant operation. Kay pointed out that important as the quantities produced at Clinton Labs are, their significance is over-shadowed by the demonstration that plutonium can be isolated from pile uranium metal on a large scale by essentially the same process that has been selected for Site W. No new fundamental problems have grown out of the plant operation, although some old problems have received additional attention. The processing cycle at Clinton Labs now requires approximately 36 hours; however, steps are being taken to improve the yield of product and to reduce bottlenecks, such as presently occur in dissolving the metal, and in another month Clinton Labs should be able to process a charge per day.

At the conclusion of this meeting Hogness presented information on the distribution of personnel within the Chicago Chemistry Division, which totals about 180 exclusive of administration and nonacademic personnel. The distribution for my section was listed as follows:

Section C-I (Seaborg)	Dec. 1943	Jan. 1944
Extraction and Decontamination		
(A) Mainline	16½	10
(B) Alternative and Basic	9	6
Purification	10	12
Metal Production	9	10
Services	14½	14
Total	59	52*

The "Monthly Report on the Activities of the Chemical Division" (MUC-TRH-34) was issued today in Chicago. The report gives a list of terms that will be used in the Metallurgical Project until further notice. Extraction, decontamination, concentration, and isolation will be used to represent the various process steps as applied to the Hanford operation. Purification and metal production apply only to the treatment of plutonium after it will have been delivered by Hanford. The Mainline Process refers to the bismuth phosphate-extraction-decontamination process together with the crossover step and the concentration and isolation steps not yet definitely fixed. References to other processes will be made in as specific terms as possible, for example (a) Adsorption extraction - bismuth phosphate decontamination or (b) $U(C_2O_4)_2$ extraction - sodium uranyl acetate decontamination, rather than the more vague terms wet fluoride process and adsorption process.

The report also notes work conducted in my section in cooperation with Mooney shows that high percentages of plutonium (4.5%) in bismuth phosphate give 0.1% expansion of the bismuth phosphate lattice. These results are close to the experimental error but suggest that carrying involves incorporation of the plutonium into the crystal lattice. With regard to the Mainline Process concentration and isolation, recent experiments indicate that plutonous peroxide is very rapidly dissolved by 8 N HNO_3 and that data used to draw up the present flowsheets are approximately correct. With regard to purification and metal production, experiments with uranium have shown that it is not possible to draw inferences as to the volatility of plutonium fluoride in macro quantities from experiments with tracer amounts. Approximately milligram quantities will be needed for reliable information. Microgram quantities of PuO_2 can be sublimed in HBr at 700°C. Larger quantities are still questionable since microgram and milligram quantities of uranium treated in the same way do not behave in a like manner. The report also covers a large number of other items in the research program of Section C-I as well as the research programs of Section C-II (Radiation Effects--Burton), Section III (Fission Product Chemistry--Sugarman), Section C-IV (Analytical--Potratz), and Section C-V (P-9 Slurry Studies--Hiskey).

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*Figure and distribution unreliable because of uncertainty of transfers and replacements.

Wednesday, January 19, 1944

Thompson and I continued our visits and discussion in the Chemistry Building and chemical separations plant. In the late evening we left Knoxville by train (Southern RR) to Cincinnati where we will continue on the New York Central back to Chicago.

The Project Council held a Policy Meeting beginning at 9:15 a.m. in the Conference Room of the Clinton Laboratories Administrative Building. The members of the Project Council and guests participating were: Allison, Cantril, Chipman, Compton, C. M. Cooper, Doan, Greager, Greenewalt, Hogness, Jeffries, W. C. Johnson, Kay, Latimer, Mulliken, A. V. Peterson, Pratt, Spedding, Stone, W. W. Watson, Whitaker, and Wigner. In his State of the Nation opening summary Compton said the present status of the Plutonium Project is most encouraging, but there is still a big job to do. With regard to other competing approaches for producing fissionable material, the Plutonium Project is still in the lead. He said although a transition to longer term at Project laboratories will be desirable as Hanford is completed and begins operations, our present work must be directed toward four primary objectives:

1. Securing information for successful construction and operation of the Hanford plant.
2. Supplying experimental amounts of plutonium and other materials needed for work at Los Alamos.
3. Assisting in working on problems and providing whatever aid we can during the time Los Alamos facilities (including housing) are lacking.
4. Supplying and training men for Los Alamos.

Compton said that as we look to the future and after the war, keen competition will develop toward applications of our research and technology. We may then be concerned with such problems as more efficient production of plutonium, nuclear power (the Navy is vitally interested), recovery of wastes, and the development of adequate sources for critical materials. These long-range programs must, of course, be preceded by adequate planning. Comments and suggestions for post emergency planning were requested of the Council members, to be submitted through Allison. When Wigner said that he would like to add another program to the list, the bringing together of the isotopes separation groups with the plutonium group, Stone made some remarks about how Y-12 (Lawrence's electromagnetic separation project) has been "stumbling along" on problems already long solved by the Met Lab.

Whitaker gave the status of Clinton Laboratories operation and said that the du Pont construction will be through in about six weeks. Housing in Oak Ridge is still a major problem. About 950 persons are employed on-plant at Clinton Laboratories and about

100 more are expected. The Clinton pile has been stepped up to about 840 kw and is now operating at roughly twice the power output per ton originally specified. It is expected that 0.5 gram of plutonium will be delivered during January, about 3 grams in February, and 3-4 grams in March. With the production of plutonium now rapidly increasing, Whitaker suggested that care by taken to avoid uneconomical use of manpower in recovering small amounts of it. Hogness suggested a central recovery plant. Greagor estimated that Clinton pile plutonium will eventually cost perhaps 100 man days per gram of plutonium. He suggested that such a value in terms of time be set in order to decide whether or not it is worth while recovering plutonium from some batches.

Allison said that discussions during last week have begun to clarify relations between Sites X, Y, W, and M (Montreal). Spedding, speaking for Ames, said that 800,000 pounds of uranium metal have already been made and he expects 1 million pounds by April. Stone spoke concerning the poisonous nature of plutonium. He said that plutonium is an alpha emitter, and he suspects that it will be stored after ingestion in bones. With radium, 1 to 2 micrograms is sometimes fatal. He said that plutonium may perhaps be less dangerous by a factor of 50 and that exposure to it has not as yet been proved to be accumulative. Radium in the body can be identified by detecting the radon exhaled in the breath or by measuring radiation from within the body with a Geiger counter. These methods cannot be used for plutonium detection. Compton added that for the moment we should consider plutonium as potentially extremely poisonous and that, obviously, more investigation is necessary. He believes the factor of 50 mentioned by Stone probably represents the worst case, which would correspond to a tolerance level of about 5 micrograms stored within the body.

Hogness presented the Chicago Chemistry Division report. Referring to the distribution of personnel, Hogness stated that he and I had met with Clinton Labs personnel as a Steering Committee two weeks ago (January 8) and had developed a joint program which was mutually agreeable. With regard to Sugarman's section, work will proceed on problems close to Mainline needs, but no increase is expected. Potratz's analytical work will be greatly expanded. Greagor has asked for 200 pounds each of aluminum silicon-bonded and zinc-bonded uranium slugs to be put in the Clinton pile and run through the semiworks to check coating behavior.

Johnson reviewed the problems and manpower distribution for Clinton's three chemical sections I, II, and III supervised by Perlman, Coryell, and Boyd, respectively. Greager then gave a report on the activities of the Separations Development Division (Section S-I under Acken and Section S-II under Sutton). Charles Cooper submitted a report on research within his Technical Division at Chicago. The meeting closed with Compton stating that the next two meetings of the Project Council will be held in Chicago on the regular dates (first and third Tuesdays and Wednesdays).

Thursday, January 20, 1944

Thompson and I arrived back in Chicago at 3:00 p.m.

Now that the required quantities of plutonium are becoming available, James and I plan to go to St. Louis at the end of next week and bombard Pu^{239} with deuterons in the Washington University cyclotron. Today I read a memorandum written by James last Tuesday that presents a preliminary analytical scheme for working up the plutonium target. In addition to the plutonium-239, fission products, and the possible new isotopes of plutonium and element 95, the target material will probably contain large amounts of copper, some iron, aluminum, and other elements. James has prepared a detailed scheme for processing the target material together with suggestions for alternate approaches to analysis. In an attached note, James referred to an article by Turner entitled, "The Missing Heavy Isotopes." (Phys. Rev. 57, 950, (1939)). Turner predictions suggest our deuteron-bombarded Pu^{239} should not contain 95^{239} because of decay by spontaneous fission nor should it contain 95^{240} due to its expected short alpha half-life. The isotope 94^{240} is predicted by Turner to be an alpha emitter, and it could have a half-life of about 10^5 years. James believes, however, it is desirable to look for beta activity in the 94^{240} fraction as planned. He lists a chart of the isotopes for the missing heavy elements, giving the characteristics predicted by Turner and the characteristics thus far determined by experimentation that indicate there is some agreement between theory and the currently available experimental data.

I suggested to James that an attempt be made to find long range alphas, by counting on a vacuum-type low-geometry counter with the counter set just outside the Pu^{239} range, and which James will calibrate and test in the meantime. I also mentioned other possible experiments such as looking for isotopes with short half-lives using neutrons from the Chicago cyclotron to bombard Pu^{239} and counting immediately after bombardment, the irradiation of 1 mg Pu^{239} in the Clinton pile and subsequent separation of fission products from the plutonium to a constant alpha/beta ratio, bombarding Pu^{239} with high energy neutrons, and performing a preliminary deuteron bombardment of 1 mg Pu^{239} at St. Louis to test our recovery of Pu^{239} and this proposed technique of looking for longer-range alphas. Finally, I mentioned that, eventually, bombardable amounts of 93^{237} will be available so we can look for such reaction products as 93^{236}, 93^{238}, U^{237}, 94^{238}, and 94^{237}.

On Tuesday, while I was at Oak Ridge, the Met Lab received its third shipment of plutonium from Clinton Laboratories; all of the 16.5 milligrams is assigned to our section. An additional 6.0 milligrams was sent to Los Alamos. Orlemann and Brody are purifying our plutonium by oxidizing it to the +6 oxidation state and extracting it into ether. Also on Tuesday Dreher gave his orientation lecture on our semiworks program to the new members of our section.

I received a letter from Kennedy written last Monday stating that David Lipkin and Morris Perlman are preparing to make an oxygen analysis on the plutonium metal sample that we agreed to submit to them. A sample

of plutonium known to be in form of oxide is also needed to aid in the interpretation of this experiment. Site Y does not have plutonium in sufficient quantity or purity to make this test, and Kennedy requests that we send a 20 to 50 microgram plutonium oxide sample along with the metal sample.

Another letter written by Kennedy the same day concerns a long-standing patent application made in the names of Kennedy, Segrè, and me. Negotiations are going on in Berkeley between the University Board of Regents and Lavender's office representing the Government. Both sides agree that neither the University nor the Government has any legal claim to the patents. Segrè is very much against giving the matter outright to the Government since he feels that "he is in no position to make such large donations to the United States Treasury." On the other hand, we cannot insist that outright assignment be made to the University. A compromise has been suggested that immediate assignment be made to the Government for all military rights and assign to the University all rights involving peace-time applications. Kennedy requests my comments and a list of the types of agreement that would be satisfactory and also the types of agreement I would find acceptable.

Last Tuesday Vernon sent two memoranda to Hilberry. The first summarizes our plutonium requirements and requests that all plutonium for the Chemistry Division coming from Site X be addressed to me. Reference was made to the memorandum from Hogness of January 15 that assigns the responsibility for control of both plutonium and Clinton uranium slugs to me. Fifteen milligrams of crude plutonium are earmarked for Sugarman for his studies on fission products and yields. Ten milligrams of plutonium oxide are destined for Potratz to be used in the development of analytical techniques by chemical and spectrographic methods. My section will receive 30 milligrams of plutonium as nitrate solution. This material is being manufactured this week at Site X and is the first request made using the Special Materials Request form.

The second memorandum from Vernon advises Hilberry that a Special Materials Request for 24 Site X slugs is being processed. The slugs will have a total plutonium content of about 2.5 milligrams. It was pointed out that this is a weekly recurring request for my section's regular chemical research. The memorandum also suggests that the plutonium will probably not be recoverable for future use as high processing losses are anticipated.

The regularly scheduled triweekly meeting of the Purification and Metal Production Groups of Section C-I was held in my absence last night at 7:45 p.m. in Room 209 of Eckhart Hall, presided over by Orlemann. Attending the meeting were: Baumbach, Bradshaw, Cunningham, Davidson, Dirksen, Dixon, Florin, Fred, Fried, Gilbreath, Heath, Hellman, Hindman, Hoekstra, Howland, Hyde, James, Jensen, Jasaitis, I. Karle, J. Karle, S. Katz, Katzin, Kirk, Kohman, Magnussen, Manning, Margolis, Marshall, Nickson, Patton, Rosenfels, Frederick Seitz, Sheft, Cliff Smith, Stein, Warner, Watters, Zachariasen, Zvolner, and others.

Nickson discussed the hazards of handling plutonium. He said that at present no definite pharmacological data are available and we must use experience gained with radium. Between one to several hundred micrograms of radium fixed in the body can prove fatal in a period of 5 to 10 years. The present data on radium are: (a) it is lethal, (b) it takes time to kill, (c) it specifically affects bone marrow, and (d) osteogenic sarcoma and necrosis may be caused by quantities as small as one microgram of radium deposited in the bones. To achieve this concentration perhaps 20 to 50 micrograms of radium must be ingested. It may be assumed that the plutonium will be approximately 1/20 as toxic as radium. Therefore, the lethal dose could be obtained from 200 micrograms of plutonium. At this stage, however, it must be assumed that 100% absorption will take place. Plutonium may be compared with radium since it also forms an insoluble phosphate that may be fixed in the bone. Nickson thinks the big danger lies in the inhalation of dust. More space, better hoods, more adequate ventilation, and perhaps a change of clothing will have to be provided by the Project. He noted that there is at present no good way to detect plutonium in the body and this represents a serious problem that needs to be solved.

Zachariasen presented the results of x-ray investigations to date on the crystalline structure of plutonium compounds. He gave evidence for PuO_2 (the only compound and structure that is reasonably certain), PuF_3 (from sodium reduction of so-called plutonium fluoride), the so-called black fluoride (that may actually be $PuOF_2$), Pu_2O_3 (a phase found in black fluoride reduced with lithium or barium), PuF_2 (a phase obtained by reduction of plutonium fluoride with atomic hydrogen). Kirk pointed out that the chemical and x-ray data agree substantially and that none of the results obtained by Zachariasen is inherently improbable. He emphasized that one of the troubles in metal production may be due to the presence of $PuOF_2$ and the presence of oxygen in crucibles. Davidson said that the chemical structure for the lower valent plutonium fluoride is still unsettled.

Yesterday Heath wrote a summary of his recent trip to Harshaw Chemical Company to discuss the availability and procurement of anhydrous HF. While at Harshaw he talked with W. J. Harshaw, President; Kenneth Long, Assistant to Mr. Harshaw; Carl Swinehart, in charge of fluorine research; A. R. Bozarth, Development Engineer; and James Hall, Sales Division. Two six-pound cylinders of anhydrous HF were shipped to us yesterday by express and the analysis is being supplied by Harshaw. A purity of only 99% can be guaranteed. A design for an apparatus, however, was worked out during Heath's visit for the upgrading of anhydrous HF through purification by distillation and treatment with fluorine in the gas phase. Analytical procedures and handling problems associated with both anhydrous HF and aqueous HF were discussed. Harshaw also agreed to supply typical analyses of tanked anhydrous HCl for the past two months' operation.

My office today received a copy of a memo from Arnold to Hogness giving the status of requests by the Met Lab for pure and rare chemicals. Included on the list are a number of chemicals not considered to be

critical in the quantities required by the Met Lab but that will probably be critical at Site Y where larger quantities will be required. Arrangements for purifications in quantity may be necessary either at Site Y or some other place.

Alan Florin and Roy Heath today treated a sample of plutonium dioxide with HF for 3 hours at 550°C and produced a light brown product which they expect might be PuF_4 or possibly PuF_3. Examining a similar preparation of a purple (or black) plutonium fluoride last month Zachariasen identified the product as PuF_3 or $PuOF_2$; our chemists had thought this was PuF_4.

The regular meeting of the Research Assistants of Section C-I took place at 6:45 p.m. in Room 209, Eckhart Hall.

A meeting of the Council of Section C-I was held in my office this evening. Those present, besides myself, were Albaugh, Baumbach, Beaton, Cunningham, Davidson, Dawson, Dreher, Ghiorso, Kirk, Manning, Orlemann, Thompson, and Watt. I presented a brief account of my trip to Oak Ridge and reported on the present status of operations at Clinton. I said that it is now possible to go from the metal solution step through crossover in 36 hours. Samples are then accumulated over a week or so before concentrating them. There is general satisfaction with the plant operation and through-the-plant plutonium yields have run about 50% and are now climbing higher. Approximately half of the 50% loss of plutonium in the plant is accounted for in the waste solutions. The remainder is presumably caused by materials sticking to the walls of vessels, pipes, etc. With a lanthanum fluoride by-product precipitation included in the crossover cycle, the decontamination factor through the crossover in the plant should increase to about 5×10^4. I also reported that all our work on uranium-233 is being discontinued except for that on the material now actually in the pile.

I stated that, in my opinion, the basic chemistry work here should be kept at a high priority. Up to now we have by necessity had to use an empirical approach. We should now proceed with more fundamental approaches to the problem of plutonium chemistry. We need to determine what should be done to obtain the most information in the shortest time. After some discussion it was generally agreed that several topics should receive early attention by the Basic Chemistry Group:

1. Number of valence states of plutonium in aqueous solution--direct chemical approach (use of oxidizing or reducing agents), polarographic approach, potentiometric titration.

2. Nature of ions--hydrolysis, etc., e.g., $Pu^{+4} \rightarrow PuO^{+2}$ (Kraus is working on this problem, including a study of solubility as a function of pH).

3. Spectrophotometer studies--differences in spectra of different valence states (a Beckman spectrophotometer is on order).

4. Preparation of inorganic derivatives of plutonium (in a sense this is an expansion of item No. 1).

5. Preparation of organic derivatives of plutonium (Melvin Calvin will be consulted on this subject).

6. Process chemistry--explanation of carrier behavior, possible complex formation, hydration in solution, effect of aluminum on lanthanum fluoride carrying, etc. (Thompson and Cunningham will consult on this problem).

7. Rough estimates of oxidation potentials.

Other possible experiments suggested by various members of the group were (1) pH at which hydrated PuO_2 precipitates (Kraus is working on this); (2) determination of water of hydration of various compounds; (3) search for lower halides by hydrogen reduction of the tetrahalides; (4) electrolytic reduction experiments; (5) possible applicability of a liquid ammonia chemistry.

It was suggested that Zachariasen should be given samples of PuO_3, Pu_3O_8, PuF_4, chlorides and other halides for x-ray analysis. In other words, it would be desirable to make these among the first inorganic derivatives of plutonium.

The present assignments of manpower in the Basic Chemistry Group are approximately as follows: Kraus, hydrolysis; Hindman and Howland, oxidation states; Patton and Smith, organic derivatives; Magnusson, neptunium separation.

Cunningham announced that approximately 35 micrograms of Np^{237} is now in process although not yet isolated. Arrangements will be made to procure Np^{239} tracer from Argonne at regular intervals to follow the remaining isolation steps. The purified neptunium will be used for measurements of specific activity and for determination of the principal valence states. Watt will test the carrying of neptunium by plutonium peroxide. This will require approximately 1 microgram of neptunium.

(Cunningham mentioned to me privately after the meeting that he had promised Studier a drink if he isolated 10 micrograms of Np^{237}, but now it appears that he will have to pay off with a couple of drinks.)

Last Sunday, the day after I left for Oak Ridge, Helen went to a picnic at the 55th Street Promenade with the Beatons and Maloneys. Afterwards, she and the Baumbachs visited the Drehers. On Monday, Wilma came by to see Helen, and later they went to Wilma's place for coffee. Al joined them for dinner at Pederson's Restaurant. After dinner, Helen spent the evening playing cards with the Ghiorsos at their apartment. Tuesday she stayed home, but on Wednesday she went back to the Ghiorsos' to spend the afternoon and evening. Today Helen had lunch at Adelaide Willard's home. She and I had dinner at the 51st Street YMCA with the Baumbachs. Afterwards, Nathalie spent the evening at our apartment while Harlan and I attended the meeting of the Council of Section C-I.

Allied bombers have sunk three Japanese ships in a raid on Rabaul. Bombers also have cut rail lines north of Rome.

Friday, January 21, 1944

We have just learned that one hundred micrograms of plutonium went down the drain! It happened this way. In Room 4 of the New Chemistry Building there is a small area where glassware and apparatus to be cleaned are usually stored. One of the cleaning girls who has the duty of washing dishes for the recovery group became over-zealous. Contrary to instructions, she picked up a clearly-labeled beaker outside this area and dumped the contents into the sink. It was this beaker that contained the 100 micrograms of plutonium in impure solution. As soon as the loss became known, the sewer trap was opened and a futile attempt was made to recover the lost plutonium. This is only slightly less than the amount of plutonium we so laboriously extracted from 300 pounds of neutron-bombarded UNH in the summer of 1942.

Report M-CA-1256, "Personnel of Metallurgical Laboratory as of January 21, 1944," is being issued. The following table includes the names of the technical administrative staff and the personnel of Section C-I.

ADMINSTRATION (TECHNICAL)

Laboratory Director	Samuel K. Allison	Eck 326
Associate Laboratory Director	Henry D. Smyth	Eck 327
Assistant Laboratory Director	Harcourt C. Vernon (DuPont)	Eck 407
Assistant to Laboratory Director	Thomas J. O'Donnell (Shop Foreman)	Ry Shop
Director, Division of Chemistry	Thorfin R. Hogness	NC 42
Director, Division of General Physics	William W. Watson	Eck 316
Director, Division of Health	Robert S. Stone	Site X
Director, Division of Patent Disclosures	Arthur J. Dempster (Special Assistant to Project Director)	Eck 117
Director, Division of Nuclear Physics	Enrico Fermi	Eck 304
Director, Division of Technology	Charles M. Cooper (DuPont)	Ry 257
Co-Ordinator of Information	Robert S. Mulliken (Special Assistant to Project Director)	Eck 210
Secretary to S. K. Allison	Dorothy C. Johnson	Eck 325
Secretary to T. R. Hogness	Fern L. Zimmerman	NC 42
Secretary to W. W. Watson	Irene G. Boland	Eck 331
Secretary to R. S. Stone	Kathryn J. Pattee	Eck 321
Secretary to E. Fermi	Marcia Ellis	Eck 304
Secretary to C. M. Cooper	Dorothy D. C. Dunaway	Eck 210
Secretary to R. S. Mulliken	Margaret A. Butler	Eck 210

ADMINISTRATION (EXECUTIVE)

Chief Administrative Officer	Lawrence A. Kimpton	Eck 311
Director, Argonne Department	Cyril A. Tregillus	Argonne
Director, Finance Department	William J. Vatter	Eck 330
Director, Personnel Department	Joyce C. Stearns	Ry 164
Director, Security Department	Theo E. Hall	Eck 311
Director, Service Department	Donald K. Keith	Eck 328
Assistant to Chief Administrative Officer	Galen Hunt	Eck 309
Organizational and Procedural Officer	Paul H. Howard	Site B
Secretary to Lawrence A. Kimpton	Mary A. Quintilian	Eck 311
Secretary to Cyril A. Tregillus	LaVerne K. Fessett	Argonne
Secretary to William J. Vatter	Margaret R. Fitzgerald	Eck 330
Secretary to Joyce C. Stearns	Mildred J. Thompson	Ry 163
Secretary to Donald K. Keith	Jeannette J. DeWind	Eck 328
Secretary to Paul H. Howard	Mary W. Draley	Site B

DIVISION OF CHEMISTRY

Hogness, Thorfin R.	Chief Chemist	Division Director	NC
Zimmerman, Fern L.	Secretary to T. R. Hogness		NC
Franck, James	Chief Chemist	Associate Director	NC
Mann, Norene K.	Secretary to J. Franck		NC
Johnson, Warren C.*	Principal Chemist	Associate Director	NC
Sibbald, Phoebe F.	Secretary to W. C. Johnson		NC
Lange, Howard W.	Curator		NC
Bottomley, Mary V.	Secretary to H. W. Lange		NC
Hendrickson, Claire	Secretary to J. C. Warner, Observer		NC

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*Assigned to Clinton Laboratories

SECTION C-I

Seaborg, Glenn T.	Senior Chemist	Section Chief	NC
Smith, Edrey E.	Secretary to G. T. Seaborg		NC
Willard, John E.	Senior Chemist	Associate Section Chief	NC
Manning, Winston M.	Chemist	Associate Section Chief	NC
Anderson, Elsy	Secretary to Seaborg's Section		NC
Gottlieb, Dorothy	Secretary to Seaborg's Section		NC
Hood, Alice	Librarian, Information Center		WS
Saxton, Irma N.	Secretary to Seaborg's Section		NC
Cunningham, Burris B.	Chemist	Assistant Section Chief	NC
Orlemann, Edwin F.	Chemist	Assistant Section Chief	NC
Thompson, Stanley G.	Chemist	Assistant Section Chief	NC

DIVISION OF CHEMISTRY

SECTION C-I

Group 1:

Albaugh, Frederic W.	Chemist	Group Leader	NC
Ader, Milton	Junior Chemist		NC
Hoekstra, Henry R.	Junior Chemist		NC
Gilbreath, James R.	Junior Chemist		NC
Greenlee, Roy W.	Research Assistant		NC
Johnson, Gordon L.	Research Assistant		NC
Schuler, Frederic W.	Research Assistant		NC

Group 1a:

Beaton, Roy H.	DuPont	Group Leader	WS
Fields, Paul R.	Junior Chemist		WS
La Chapelle, Theodore J.	Junior Chemist		WS
Lincoln, Dwight C.	Junior Chemist		WS
Sheft, Irving	Junior Chemist		WS
Casler, Ruth A.	Laboratory Technician		WS
Daniels, Minnie M.	Laboratory Assistant		WS

1/21/44 (cont.)

Group 2:			
Watt, George W.	Senior Chemist	Group Leader	NC
Pye, Donald G.	Associate Chemist		NC
Hopkins, Horace H., Jr.	Research Assistant		NC
Yett, Fowler R.	Research Assistant		NC
Group 3:			
Dreher, J. Leonard	Associate Chemist	Group Leader	WS
Dean, Floyd M.	Associate Chemist		WS
Katzin, Leonard I.	Associate Chemist		NC
Margolis, Asher J.	Associate Engineer		WS
Creek, George E.	Junior Chemist		WS
Flox, Jack	Junior Chemist		WS
Hyman, Herbert H.	Junior Chemist		WS
Malm, John G.	Research Assistant		WS
Winner, Bernard M.	Research Assistant		WS
Freeman, Julia	Laboratory Assistant		WS
Group 4:			
Orlemann, Edwin F.	Chemist	Group Leader	NC
Watters, James I.	Associate Chemist		NC
Dixon, Jonathan S.	Junior Chemist		NC
Hagemann, French T.	Junior Chemist		NC
Jensen, Lyle H.	Junior Chemist		NC
Brody, Bernard B.	Research Assistant		NC
Stein, Amanda P.	Research Assistant		NC
Carter, Theocian H.	Laboratory Technician		NC
Group 5:			
Davidson, Norman R.	Associate Chemist	Group Leader	NC
Bradshaw, Benjamin	Associate Chemist		NC
Karle, Isabella L.	Associate Chemist		NC
Katz, Joseph J.	Associate Chemist		NC
Heath, Roy E.	Chemist		NC
Florin, Alan R.	Junior Chemist		NC
Hyde, Earl K.	Junior Chemist		NC

1/21/44 (cont.)

Group 6:

Baumbach, Harlan L.	Chemist	Group Leader	NC
Dirksen, Alvin J.	Chemist		NC
Rosenfels, Richard S.	Chemist		NC
Fried, Sherman M.	Associate Chemist		NC
Hellman, Nison N.	Associate Chemist		NC
Jasaitis, Zene V.	Associate Chemist		NC
Karle, Jerome	Associate Chemist		NC
Katz, Sidney	Associate Chemist		NC
Meyer, Fred	Associate Chemist		NC
Zvolner, Hymen P.	Associate Chemist		NC
Frank, Ray D.	Research Assistant		NC
Foreman, Beatrice	Laboratory Technician		NC

Group 7:

Cunningham, Burris B.	Chemist	Group Leader	NC
Patton, Robert L.	Chemist		NC
Hindman, James C.	Associate Chemist		NC
Kraus, Kurt A.	Associate Chemist		NC
Smith, Clifford	Associate Chemist		NC
Howland, Jerome J.	Junior Chemist		NC
James, Ralph A.	Junior Chemist		NC
Magnusson, Lawrence B.	Junior Chemist		NC

Group 8:

Dawson, Lyle R.	Chemist	Group Leader	NC
Studier, Martin H.	Junior Chemist		NC
Baumbach, Nathalie S.	Research Assistant		NC
Beard, Walter G., Jr.	Research Assistant		NC
Belletire, Frank L.	Laboratory Technician		NC

Group 9:

Ghiorso, Albert	Associate Chemist	Group Leader	NC
Jaffey, Arthur H.	Associate Chemist		NC
Kohman, Truman P.	Associate Chemist		NC
Crawford, John A.	Junior Physicist		NC
Schoenberg, Bert	Junior Chemist		NC
Weissbourd, Bernard B.	Junior Chemist		NC

1/21/44 (cont.)

Heller, Lorraine G.	Research Assistant		NC
Scott, Benjamin F., Jr.	Research Assistant		NC
Walsh, Patricia D.	Research Assistant		NC
Le Feuvre, Arthur G.	Laboratory Technician		NC
Lenhard, Madeline S.	Clerk		NC
Service Group:			
Donlan, F. Bernard	Clerk	Supervisor	NC
Gavin, Kathleen B.	Clerk		NC
Bowman, Eleese E.	Laboratory Assistant		NC
Dreyfuss, Richard S.	Laboratory Assistant		NC
Fulan, Emil R.	Laboratory Assistant		NC
Harding, Reid B.	Bench Hand		NC
Jennings, Nellie G.	Laboratory Assistant		NC
Summers, Mildred M.	Laboratory Assistant		NC
Young, Robert B.	Laboratory Assistant		NC

Paul Kirk and R. S. Rosenfels transferred from my section to Section C-IV under Potratz as a prelude to their transfer to Site W where Kirk will be in charge of the measurement of yields of plutonium in the various steps of the Bismuth Phosphate Process in the chemical extraction plant; microchemical methods will be used, many instruments need to be designed and built, and many techniques need to be developed. With Kirk's departure, Baumbach becomes group leader of the Metal Production Group.

I learned that Oppenheimer has sent a letter to Allison, dated January 18, confirming an agreement reached between Allison and Thomas that T. T. Magel will transfer to Site Y on February 1. Oppenheimer is particularly happy to get Magel because of his former association with my section.

Orlemann gave a small sample of his purified Pu^{+6} to Wright Langham and Sol Wexler in the Analytical Section (C-IV) so they could check its purity. They came to me this afternoon, perplexed, and said that the solution turned blue (instead of green) when they added hydroxylamine. I wonder if this suggests the observation of a +3 oxidation state.

Kennedy wired me from Site Y that the amplifier design that Ghiorso wants has been sent to Allison by messenger.

1/21/44 (cont.)

Hogness asked Whitaker for a list of surplus equipment from Brown's group at Site X to be transferred to Willard and Kirk.

This evening Helen and I went to the movies and saw "Thank Your Lucky Stars" with Eddie Cantor, Dinah Shore, Humphrey Bogart, and Bette Davis.

Today's paper says that the British Royal Air Force hammered Berlin last night in the eleventh massive attack since the aerial campaign started about two months ago.

Saturday, January 22, 1944

When I came to the office this morning, I was met by Cunningham who told me that we almost lost a sizable portion of our plutonium. Last night in Room 5, Orlemann, Brody, and Dixon stayed on to make a third ether extraction of the 16.5-milligram sample of plutonium that arrived from Clinton Labs last Tuesday. Around three o'clock this morning a tube containing approximately five milligrams of the plutonium in the aqueous layer, which had been frozen in liquid nitrogen, broke in Brody's hand. Brody and the others recovered about two milligrams of this aqueous layer. Another milligram was in the washings from Brody's hands. Now Brody's hands are contaminated with plutonium, and until the alpha count goes down sufficiently, he must wear cotton gloves to avoid ingestion whenever he eats or drinks. As much as possible of the rest of the plutonium must be salvaged from Kleenex, cardboard, floor tile, rubber apron, etc. I am asking Cunningham to have members of his Basic Chemistry and Recovery Group (including Kraus and Studier) work on the recovery and purification of the plutonium involved in this accident and the further purification of the remainder of the Clinton plutonium.

However, the big news today is that Clark Hindman, using the remainder of the ether-purified plutonium from Clinton, apparently reduced Pu^{+6} with SO_2 to Pu^{+3}; or possibly, it was reduced to Pu^{+5}. The solution he obtained is purple in color.

Baumbach sent me a summary of a talk given by Zachariasen on the x-ray diffraction results on eight uranium compounds and five possible plutonium compounds. If it is assumed that the formula for each compound is correct, PuO_2 has a face-centered cubic structure and a density of 11.54 gm/cm^3; PuF_3 is hexagonal with a density of 8.01 gm/cm^3; $PuOF_2$ is hexagonal with a density of 9.24 gm/cm^3; Pu_2O_3 has a face-centered cubic structure and a density of 14.32 gm/cm^3; and PuF_2 is face-centered cubic with a density of 10.95 gm/cm^3. The PuF_3, Pu_2O_3, and PuF_2 were produced during attempts to produce plutonium metal by various reduction techniques and their composition is deduced from the x-ray diffraction data and must be considered doubtful at this stage.

Lyle Dawson left Chicago today on a trip to Oak Ridge in order to observe the processes being carried out in the concentration room at Clinton Laboratories.

I learned that a meeting was held Thursday in the office of Charles A. Thomas in Dayton, Ohio, with Thomas, Spedding, and J. C. Warner present. It was agreed, as suggested by Compton and Major Peterson, that technical supervision of the Ames Project will be transferred from Compton to Thomas, effective February 1, 1944. It has become apparent that the equipment, personnel, and past experience of the Ames Project can be used more effectively to supplement the work at Site Y than any other part of the overall project.

This afternoon I worked through a stack of personnel papers put at my disposal in order to select additional men for Section C-I. These men are presently under the jurisdiction of the U.S. Army Special Engineering Department (SED). I have been told that they will be available to work with us and that I may pick out a half dozen or so. I was happy to run across the name of Donald C. Stewart, a very capable chemist who was a fellow undergraduate student at UCLA. The other men I have selected are all highly qualified chemists, and I hope that most of them can come with us. They include Larned B. Asprey, J. W. Britain, Phillip Fineman, and Leon Leventhal.

Compton sent Allison a memo about the interchange of information between the Met Lab and the Montreal Lab and between the Met Lab and Los Alamos. Compton said that it became evident in the Project Council meeting on Wednesday that a revision is called for in connection with our interchange of information with Site Y. Mulliken will pull together all suggestions about the latter so that they may be presented when Thomas is in Chicago on February 14.

A meeting was held in Franck's office with Hiskey, Hogness, Manning, Potratz, Sugarman, and me present. Hogness reviewed the statements made by Compton at last Wednesday's Project Council Policy Meeting in Oak Ridge. Health hazards relating to the handling of plutonium were stressed. It has been requested that better ventilation be provided and linoleum floors be installed for the original New Chemistry Building (now occupied by my Section C-I). It is becoming clear that extensive renovation will be required. Hogness and I raised questions concerning insurance coverage.

Wilma visited Helen in the afternoon. The Ghiorsos dropped in to play cards with us during the evening.

German planes bombed London in a spectacular raid last night. The Soviets have driven 30 miles southwest of Leningrad and widened the Leningrad line.

Sunday, January 23, 1944

The headline in today's Sun reads "Only 16 Miles from Rome!" British and U.S. troops have landed southwest of Rome, supported by planes and ships. They met surprisingly little resistance.

Monday, January 24, 1944

This morning Hindman reported to me that yesterday when he checked the purple plutonium solution he found that it remained stable overnight. We intend to identify the oxidation state as soon as possible. Kurt Kraus has taken the lead in setting up a program for determining the absorption spectra of Pu^{+4} and Pu^{+6} in aqueous solution using our new Coleman spectrometer. This gives us a powerful method for identifying this new oxidation state; therefore Hindman and Kraus will measure its absorption spectrum and compare it with those of Pu^{+4} and Pu^{+6}. They, with the assistance of Jerry Howland, intend to try to produce it with other reducing agents and will attempt to measure its oxidation potential. The new oxidation state seems to be Pu^{+3} but since Pu^{+5} is a possibility, they will attempt to produce it by the reduction of Pu^{+4}; this would establish that it is Pu^{+3}.

Melvin Gerstein started work today in Baumbach's group as a Junior Chemist. He has been assisting Professor Schlesinger at the University of Chicago. Gerstein has completed all necessary course work for a Ph.D. but is lacking the research for a thesis. Another new arrival to Section C-I is Jacob Sedlet, who has been a chemistry student at the University of Wisconsin. Stan Thompson briefed him on the purpose of the Project. Sedlet, although surprised about the discovery and properties of Pu^{239}, said that he had heard lectures by Professor Norris Hall about radioactivity, the fissionability of U^{235}, and its potential as an energy source. He also had heard rumors about a secret nuclear energy war project. We plan to have Sedlet work for Albaugh as a Research Assistant.

Sherm Fried placed the plutonium fluoride sample, prepared by Florin and Heath on Thursday, in a capillary tube and took it to Zachariasen for analysis by x-ray diffraction. Zachariasen reports that it is isomorphous with the green form of UF_4 he analyzed last month and hence must be PuF_4.

I wrote to Kennedy explaining that we were unable to meet yesterday's deadline for the sample of plutonium metal that was to have been sent to Site Y for oxygen analysis because we have not yet been able to remove a large enough piece of the sample from the crucible. I told him that we will send it as soon as we can, together with about 100 micrograms of PuO_2 as requested in his letter of January 17.

Thompson wrote up a resume of several discussions he and I had with each other at Clinton last week about the Bismuth Phosphate Process. A summary is as follows. There is a definite trend at Clinton Labs towards improved yields in the plant that now may be approaching, through

the crossover cycle, overall yields of about 70-80%. The change back to the direct strike method of bismuth phosphate precipitation in the extraction step (i.e., addition of phosphoric acid after the addition of bismuth carrier) has resulted in precipitates that are more easily dissolved. Consequently, losses due to holdup of undissolved bismuth phosphate are reduced. The use of a fluoride by-product precipitate in the crossover cycle is now regular practice. Very promising results have been obtained in Perlman's group using scavengers in oxidized solution in the bismuth phosphate decontamination cycle. Zirconium phosphate, ceric phosphate, barium sulfate, and lanthanum fluoride were the scavengers used in one run. Studies are now underway designed to find out which scavenger is responsible for the most decontamination. Experiments just being completed by Schuler here in Chicago indicate that amounts of holdback carriers in the extraction step, of the order of about 5 mg per cc each of lanthanum, yttrium, and cerium improve the gamma decontamination by a factor of 4. The product yield, however, seems to be reduced to about 75%. The first Chicago semiworks run with a uranous oxalate concentration step using Site X uranium metal has just been completed; an overall yield of about 90% was obtained. The training program is nearly complete, and most of the new people are running the Bismuth Phosphate Process in order to become acquainted with it and the associated techniques.

Thompson goes on to recall that our discussions covered all the project assignments and the individuals who are working on them. He notes that all the problems that were assigned to us from Clinton Laboratories are now being carried on. Our major problem assignments include the HCl concentration work, the coating problem, studies of the process at Hanford concentrations, and the peroxide concentration work. Several new assignment sheets are being prepared and will include such problems as the tracing of neptunium through the Hanford concentration process using 93^{237} and also the problem of studying yield and decontamination using lower temperatures throughout the entire phosphate process. This means that about two micrograms of 93^{237} will be required in about two weeks in order to make the process studies. It is hoped that tracer amounts of 93^{239} will also be available soon. Continuous ether extraction equipment is being set up in West Stands, and it should be possible to extract and decontaminate 93^{237} by this method. Because of the increased work now being handled, Thompson will meet with Watt, Dreher, and Albaugh (group leaders for concentration, semiworks, and extraction-decontamination groups) each morning at 9 a.m. to discuss Section C-I problems and work plans. Each of these groups will meet weekly to give the members the opportunity to present and discuss their work.

Wigner sent a letter to Smyth at Princeton University (and made several copies for internal distribution at the Met Lab) enclosing a clipping from the latest Time magazine that states that no respectable scientist believes that a secret weapon will have a substantial influence on the outcome of the war. Wigner made the following comments:

"Of course, nobody knows whether a secret weapon will actually have a great influence on the present war. It seems to me, however, very dangerous to let such statements go by unchallenged. Perhaps the greatest danger that we fear in connection with the peace negotiations is that the public will not be aware of the importance of setting up an international organization to supervise armaments and to enforce its decisions. It seems to me that it would be very desirable to begin the education of the public in this direction as soon as possible, so that by the time the peace negotiations come, it should be ready to make substantial sacrifices in order to insure the creation of such an international agency. This will be a difficult task anyway, since it is only human to forget past dangers for the sake of present comfort, even if one was fully aware of the dangers. It would be much worse if sacrifices were demanded from the general public based on dangers of which they were entirely unaware up to that time.

"Naturally, it would be undesirable if a statement that dangerous secret weapons might be developed came out from Chicago. On the other hand, if it came from the staff of a well-recognized institution, which at present is not engaged in developing a secret weapon of this decisive nature, there could be no objection to it from the security point of view. Of course, I was thinking of Princeton in this connection. I would very much like to know what you think about sending an article to Time, in the name of the Department or its members who are at present in Princeton, saying that they disagree with their statement. I believe that such a statement would have a great and very healthy influence on public opinion. I would appreciate your letting me know at an early date whether or not you are intending to send a note to Time in the above sense, since if a note is to be sent it should be sent at an early date."

Beaton is again in Philadelphia today meeting with representatives of the Resinous Products and Chemical Company.

This afternoon Wilma visited Helen.

The top headline today indicates that the Allies are making great gains in the drive on Rome.

Tuesday, January 25, 1944

Ralph James prepared a standard sample consisting of 0.1 mg plutonium mounted on a beryllium counting disk (Sample 49 D-Be-1). Using the sample, he tested his vacuum low-geometry counting chamber method to be used in looking for longer range alpha particles after deuteron bombardment of similar samples. He has worked this method out following my suggestion last Thursday that we try such a technique. He concludes that his method can detect the production of alpha particles that have a range greater than 94^{239} by a fraction of a centimeter.

A three-page, detailed memorandum was sent by Willard to Squires listing some of the jobs which should be the responsibility of the

chemistry group at Site W. In general these jobs consist of emergency trouble-shooting in the case of process difficulties, improvement of process yield, improvement of decontamination procedures, improvement of methods for concentration, final isolation, and shipment of plutonium. There is a need for two or more men who are capable of designing and constructing specialized instruments but who are also experts in the techniques and problems of radiochemistry. Considerable attention must be placed on waste disposal. Willard also suggested that work be directed toward the separation of possible new transuranium isotopes from the Hanford piles. Under the conditions of pile operation, there is a possibility that significant amounts of new transuranium isotopes, such as 95^{240}, may be produced. These isotopes could have characteristics that might give rise to serious problems at Site Y. It would therefore appear necessary to identify these new isotopes and incorporate steps in the process to remove them.

Beaton returned from Philadelphia today after meeting with F. J. Myers, R. J. Myers, D. S. Herr, and L. C. Hurd of the Resinous Products and Chemical Company. He wrote up a summary of their discussion, saying that all the men are very optimistic about the chances of success in providing a resinous ion-exchanger other than Amberlite IR-1 to carry out the decontamination of "thorium" (plutonium) from by-product "impurities" such as zirconium, columbium, cerium, yttrium, and strontium. In order to expedite the work, arrangements will be made to obtain the materials on a purchase order basis rather than on a subcontract.

Cunningham sent a letter to Connick, Gofman, and Sheline in Berkeley transmitting a correction to a recent report. Cunningham says he supposes that by this time the group in California has definitely established the +3 state of plutonium. He says we have been having a lot of fun with it here and are somewhat abashed at not having pinned it down before this. On another matter, Cunningham says we have not had a chance to check the specific activity of the uranium samples they sent to us and that we probably will not be able to do so for some time. He mentions that the discrepancy between the Kovarik and Adams value and ours has been traced to backscattering of alpha particles.

Allison sent a memo to J. H. McKinley in the Area Engineer's office concerning the present unpainted concrete floors in the New Chemistry Building and the possibility that dust from these floors might carry plutonium or other radioactivity into the air and become a health problem. Allison requested that Stone's suggestion that the floors be painted and the traffic areas be covered with linoleum be carried out.

Zachariasen has issued a report, "X-Ray Diffraction Studies of Plutonium Preparations" (CK-1259), summarizing in part the work of his section for the period ending January 22, 1944. The abstract of this report is written as follows:

"X-ray diffraction patterns have revealed the existence of at least six distinct phases (A-F) in samples of plutonium compounds and of plutonium metal preparations. The crystal structure data for the six phases are given. The identification of these phases with specific plutonium compounds is considered. One phase (A, yellow oxide) is definitely PuO_2 and the so-called white fluoride (phase C) is definitely PuF_4. The black fluoride (phase B), which the chemists believed to be the tetrafluoride, is shown to be either PuF_3 or $PuOF_2$. The identification of the three remaining phases is less certain. Phase D (hexagonal) may be $PuOF_2$, PuF_3, or plutonium silicide; phase E (cubic) may be Pu_2O_3; and phase F (cubic) may be PuF_2."

Zachariasen has this to say about the black fluoride (Phase B): "Several different samples of the black fluoride have been studied. The various samples gave the same diffraction pattern, all lines of which are due to a single phase. This phase is hexagonal with lattice constants $a_1 = 4.085$ Å, $a_3 = 7.236$ Å. The observed intensities require two metal atoms per unit cell and in positions (1/3 2/3 1/4) (2/3 1/3 3/4). We have also examined a sample of black fluoride which had been heated to 1,120°C. The diffraction pattern was not changed, except for the fact that two weak additional lines due to another phase were observed.

"For reasons which will be explained in Section 3 of this report I have for the last six weeks maintained that the black fluoride probably was $PuOF_2$ or PuF_3 rather than PuF_4 as the microchemists assumed. The chemists were immediately informed of my suspicion of the black fluoride, but they were not until recently able to find any result of their own which suggested that their assumed formula was in need of revision. After repeated failures to produce metal when starting with the black fluoride also, the metal production group has recently become suspicious of the assumed composition of the black fluoride. During the last week attempts have therefore been made to produce plutonium fluoride under various experimental conditions."

He concludes this part of the report by saying, "The six phases listed above are the only ones which are definitely established. Some metal preparations give diffraction lines which cannot be ascribed to any of the phases A-F nor to slag or crucible materials. These diffraction lines have, however, been too few and too weak for interpretation purposes."

Today Helen went downtown and later had coffee at Wilma's.

The Germans have hurled a heavy blow at the Cassino front. One hundred thousand Nazis are reported to be fighting U.S. troops.

Wednesday, January 26, 1944

Last night another near catastrophe occurred with our purified Clinton plutonium. After almost everyone else had gone home from the New Chemistry Building, Kurt Kraus remained quite late to work on the further purification of our precious sample. When he turned on a micro

burner, it started whipping around because it was hooked up to the air line instead of the gas line, sending a beaker crashing to the floor. The beaker contained two 2-ml glass-stoppered tubes, one of which contained about 12 milligrams of our purified plutonium and the other a dilution of an aliquot for assay. Fortunately only the latter tube was completely shattered. The stopper on the tube holding the 12 mg of purified plutonium was sheared off, but the sample was intact. Kraus found Studier in an adjoining room and asked for assistance. They pipetted up the spilled assay solution and then cleaned up, locating the plutonium with the help of Ghiorso's "Treasure Finder," our portable alpha counter.

However, the good news is that Hindman, Kraus, Howland, and Cunningham have succeeded in establishing that the purple solution of plutonium is due to Pu^{+3}. Cliff Smith has precipitated Pu^{+3} as the fluoride, oxalate, hydroxide, and phosphate.

Today Fried reduced about 350 micrograms of "white" plutonium fluoride with calcium in a "degassed" crucible. The results were very good--he pried loose one large shiny piece (about 300 μ in diameter) which had a piece of thoria adhering to it and placed it in an evacuated tube for Zachariasen. A smaller piece, estimated at a little more than 12 micrograms, appeared bright and silvery and had no pieces of thoria attached to it. This was given to Jasaitis who, using his talents for small scale manipulations, flattened the sample (it proved quite ductile and about as hard as copper), sealed it in an evaculated capillary tube and sent it to Zachariasen's laboratory for x-ray measurements.

Ralph James is leaving at 11:50 this evening by train for St. Louis, carrying plutonium samples and equipment needed for our deuteron bombardment experiments at the Washington University Cyclotron. I will join him on Friday.

Today Warren C. Johnson is moving his family to Oak Ridge where he is Director of the Chemistry Division at Clinton. He plans to return to Chicago around June 1.

I wired Eastman requesting that he airmail to me about 3 grams of uranium disulfide for use as a refractory material here.

In a memorandum to Colonel H. C. Metcalf in the Patent Division I informed him that I have asked Katzin to help me in the handling of all patent matters for Section C-I. Manning has been assisting me in this capacity up to the present time, but he will no longer be able to take care of this work because of the increased pressures from his other duties.

Over the signatures of Allison and Compton, a letter was sent to H. D. Smyth releasing him from the responsibilities of the position

of Associate Director for the Metallurgical Laboratory at his request. Smyth was asked to retain his connection with the Metallurgical Project as a consultant. This change in appointment status was requested by Smyth since he can participate only part-time in the work of the Laboratory. Appreciation was expressed to Smyth for his past work as Associate Director, especially in connection with his guidance of the P-9 program.

At Whitaker's suggestion, letters were sent by Compton to S. T. Cantril, who is serving as Medical Director, and D. M. Smith, the newly-appointed director of the Control Analysis Chemistry Division, both at Clinton Laboratories, appointing them to serve as members of the Project Council.

Compton then informed Charles Cooper, who is secretary of the Project Council meetings, of the changes in the membership of the Project Council. It was requested that Professor Smyth's name be removed from the list of the regular members of the Council and be placed on the list of observers. Because the work of the Ames Laboratory is being transferred from the responsibility of the Plutonium Project to that of the Site Y Project, Spedding will no longer be a Project Council member but will be welcome to attend all future meetings, including the policy meetings, as an observer. It was also requested that Cooper add Cantril and D. M. Smith to the list of Project Council members.

I attended the triweekly meeting of the Basic Chemistry, Recovery, and Instruments Groups of Section C-I which met at 7:45 p.m. in Room 209 of Eckhart Hall. We were pleased that Hogness accepted our invitation to attend. Present tonight, in addition to Hogness and me, were H. Baumbach, N. Baumbach, Brody, Crawford, Cunningham, Davidson, Dempster, Dixon, Fried, Ghiorso, Hagemann, Hindman, Howland, Jaffey, I. Karle, J. Karle, J. J. Katz, Katzin, Kirk, Kraus, Magnusson, Manning, Meyer, Orlemann, Patton, Rosenfels, Cliff Smith, Studier, Thompson, Watters, Willard, and others.

In my opening remarks I stated that this exciting meeting would be mainly about Pu^{+3}, the existence of which recently has been fairly well established as a result of the availability of relatively large quantities of plutonium from Clinton Laboratories. I then turned the meeting over to Cunningham who pointed out that the existence of Pu^{+3} has been repeatedly suggested in the past. The work performed by La Chapelle in June 1943, with tracer experiments using bismuth phosphate as carrier and hypophosphite as a reducing agent, was inconclusive. Later, James used other carriers, but the evidence was incomplete. An attempt made by Cunningham to obtain Pu^{+3} through reduction with zinc, on the ultramicro scale, gave negative results. Tonight Hindman reported on his exciting, successful identification of Pu^{+3}. The first reduction to Pu^{+3} was achieved using a 1.5 ml Pu^{+6} solution containing 5 g of plutonium per liter and 0.5 M HCl, obtained from ether purification of the plutonium obtained from Clinton Laboratories. This sample probably contained some nitrate. The absorption spectrum for this sample was then determined with a Coleman spectrophotometer. The sample was reduced with SO_2 and a

purple solution was obtained, distinctly different than the well-known green color of Pu^{+4}. This meant something new had been obtained--probably Pu^{+3}. The spectrum of this solution was also determined. After a number of unsuccessful attempts to change another sample of the original Pu^{+6} solution to a green color using various reducing agents, the green color of Pu^{+4} was finally obtained through the use of hydrogen peroxide. The absorption spectrum of the green solution was determined. The purple solution was thus identified as a solution of Pu^{+3}. The three spectrophotometer absorption curves were sketched. The Pu^{+3} gives a new absorption band at 6,000 Å. A band around 3,900 Å and one around 4,100 Å are typical for Pu^{+4} and Pu^{+6}, respectively. The difference in the oxidation numbers of Pu^{+3} and Pu^{+4} was determined spectrophotometrically by titrating Pu^{+4} with the reducing agent Sn^{+2} with the use of a Beckman spectrophotometer. Advantage was taken of the fact that Pu^{+4} has a strong and sharp absorption band around 4,700 Å and Pu^{+3} does not. Attempts to bracket the potential of the Pu^{+3}/Pu^{+4} couple were made; results suggest that the normal potential of the couple is more negative than -0.53 volts. I called attention to the similarity between plutonium and uranium. The colors of the +6, +4, and +3 states are yellow, green, and purple, respectively, for both uranium and plutonium. The stability of Pu^{+4} seems to be similar to that of UO_2^{++}, and that of Pu^{+3} similar to that of U^{+4}.

Smith discussed the solubility and appearance of four Pu^{+3} compounds:

a. Fluoride. It was prepared in 1 N HF from a Pu^{+3} solution (0.25 M in HCl) by the addition of HF (final concentration of 1 N) in the absence of alkali ions and has a solubility of 0.043 gm Pu/l. This insolubility is in contrast to the behavior of Pu^{+4}, which does not yield an insoluble fluoride in the absence of alkali ions.

b. Phosphate. It is white, gelatinous, and has a solubility of 0.33 gm Pu/l. It was prepared in 1.2 M H_3PO_4.

c. Oxalate. It is very soluble in water and in oxalic acid. A solubility of 9 to 10 gms Pu/l in 0.05 M $H_2C_2O_4$ was found.

d. A blue hydroxide was precipitated with saturated ammonia and was found to have a solubility of 0.09 gm Pu/l.

Smith described another experiment in which oxygen was bubbled for one hour through a reduced plutonium solution (in 0.5 HCl) containing some SO_2 without causing oxidation of Pu^{+3} to Pu^{+4}. When the solution was warmed, oxidation took place immediately with the formation of a green solution.

Kraus mentioned inconclusive results from his attempts to reduce Pu^{+3} to a lower valence state. He also described some experiments which suggest the existence of a carbonate complex of Pu^{+4}.

Orlemann reported on the ether extraction made on the Clinton sample which contained (as received) approximately 25% thorium and 25% lanthanum. The sample solution was oxidized with dichromate, and the

Pu^{+6} was extracted into ether with the help of saturated ammonium nitrate solution. The ether was removed by evaporation over a 1 N HCl solution. The ether layer was pink in color and the water layer yellow. Spectrographic analysis of the final HCl solution showed that it contained less than 0.4% impurities. I pointed out that this method holds great promise for use in the purification of plutonium. Hindman used this solution for his first observation of Pu^{+3}.

Smith also precipitated a yellow-brown compound from the Pu^{+6} solution by the use of ammonia. It is probably $(NH_4)_2Pu_2O_7$ and has a solubility of 1.25 gm Pu/l.

During discussion that followed on Pu^{+3}, I stated that a possibility exists that $PuCl_3$ is volatile at relatively low temperatures. It is possible that separation from light elements could be achieved by using the sublimation or distillation of this compound.

Ghiorso reported on the recent reinvestigation of the half-life of plutonium and the effect of alpha backscattering. Experiments to prove that backscattering from platinum occurs were described. I pointed out that the error in the half-life of plutonium that results from the incorrect counting geometry is reflected in an error in the fission cross section for plutonium. The present specific activity for plutonium is close to 71,000 c/m/μg according to Cunningham. Crawford stated that backscattering was caused by multiple scattering rather than to single scattering. He briefly explained his method of calculating the percentage of backscattering.

Helen worked all day in the Information Division on the "Table of Isotopes."

Cassino appears to be deserted by the German troops, but they still hold the surrounding heights.

Thursday, January 27, 1944

I received a memorandum from Thompson requesting at least 10 mg of plutonium out of the approximately 100 mg allocation that we are expecting to receive from Clinton within the next few days. He needs the plutonium to carry out the necessary work on two very high priority problems: (1) testing and developing the peroxide isolation method that is to be used at Hanford and (2) testing the entire Hanford separations process at Hanford concentrations on a 100 cc scale. On this scale it will be possible to state fairly definitely whether or not the present flowsheet isolation procedure is operable. In addition, he will use one milligram of the 10 milligrams for further solubility studies of the peroxide in nitric acid solutions, and possibly to determine the effect of temperature on solubility.

Cunningham also gave me his request for plutonium. He wants a total of 15 milligrams for work on the basic chemistry of plutonium. Included in this work would be the determination of the oxidation potential for the Pu^{+3}-Pu^{+4} couple to an accuracy of at least ±0.05 v. Four

milligrams of the requested plutonium will be required in this study using three approaches: bracketing by means of known oxidation-reduction couples, direct potentiometric measurement, and a polarographic survey. Another 5 milligrams of plutonium will be used in the search for and characterization, in aqueous solution, of valence states of plutonium other than those already idenitified, such as a +5 and a lower than +3 state. One milligram of this amount will be reserved for the preparation of compounds of such other valence states as may be found. If no other states are found, the one milligram of plutonium will be used for a more extensive investigation of Pu^{+6} compounds. Two milligrams will be used for measurements of the solubilities of plutonium compounds in various valence states--particularly of those compounds that might be formed with reagents used in extraction, decontamination, concentration, isolation, or purification. The remaining four milligrams will be used to study the formation of plutonium complexes in the various valence states. This information together with the determined potentials of the plutonium couples should have practical application to the plant processes. Specifically sulfate, chloride, oxalate, and fluoride complexes will be studied.

I checked with Dreher on the progress on the second 26 pound batch of Site X uranium metal. It was received by the semiworks, has been dissolved, the bismuth phosphate extraction step performed, and the decontamination cycle is now underway. Dreher said that disposal of the increasing volume of radioactive waste solutions here in Chicago has become a major problem. As the methods used at Clinton Labs for handling the extraction step effluent and jacket solution appear promising, they will be tried here next week. Extraction and decontamination equipment has been altered to permit operation largely by remote control. It may be another ten days before the equipment to test the alternate ether extraction step will be ready for operation.

Zachariasen issued a second part of his report on the activities of his X-ray Diffraction Section for the period ending January 22, 1944 (CK-1250). He gives a short review of the lattice structures of three plutonium samples, which he classifies as phases B, E and F in Report CK-1259. Included is a description of Mrs. Mooney's work on bismuth phosphate. It reads as follows: "Diffraction patterns of two samples of bismuth phosphate precipitated with 49 were made. One was estimated to be about 100 micrograms, carrying 4.4% of 49; the other 300 micrograms, carrying 8% of 49. These samples were made and sealed in a fine glass capillary by Gordon Johnson of Thompson's group. The results of careful measurement and comparison of the cell constants with those obtained from pure bismuth phosphate fail to give an unequivocal indication of change of the cell size due to the addition of the 49. If the plutonium actually forms a mixed crystal with the bismuth phosphate, the expansion of the lattice is not more than 0.1%."

I sent a rather long memorandum to Compton outlining the possible utilization of uranium-233 by the enemy, in response to Compton's request. I give reasons why I think the Germans may be able to produce at least one uranium-233 device in the near future. I suggest this possibility

be considered and be brought to the attention of our High Command in order that steps may be taken to distribute governmental facilities of the Allied Powers in such a way that a single device alone could not deal a fatal blow. It seems highly desirable that we sustain enough effort on the uranium-233 problem in order that we will be able to evaluate any developments the enemy may claim or employ. I gave the following reasons why I think it probable that the Germans may be preparing to use a U-233 device:

(a) "We know that the Germans are making strenuous efforts to continue heavy water production in Norway, and there seems to be no other possible use for this material other than a nuclear power program. Likewise, we can infer that such efforts would not continue unless they had in mind an overall heavy water production program in other places as well as Norway, calculated to make or have made an amount sufficient for their final objective within a reasonable length of time.

(b) "U^{233} can be separated from its parent thorium by relatively well-known chemical processes. The isolation of U^{233} may be very simple compared to that of Pu^{239} or U^{235}. This is in part due to the fact that essentially only one radioactive species (Pa^{233}) is involved in the separation. I believe that the Germans could handle this separation problem in one of their large industrial chemical companies, who in turn would isolate the U^{233} with only slight modifications in their existing equipment.

(c) "The tolerance for light element impurities (which must be removed to prevent neutron production by the α,n reaction) is some ten-fold greater than that of Pu^{239}. Furthermore such purification as must be obtained can be tested with uranium, which, in contrast with Pu^{239}, would have been available for experiments during production of the actual U^{233}. Our own present knowledge of the purification and metal production of uranium leads us to the practically certain conclusion that this problem is rather easily solvable for U^{233}. If we should run into serious difficulties in the final processing of Pu^{239}, it is conceivable that the Germans might produce a final U^{233} device before we produce one from Pu^{239} even if their program were not very far along today.

(d) "Raw materials for U^{233} production are probably available to the Germans. Using a uranium-heavy water pile in conjunction with thorium, it is quite probable that they would not need to meet the high purity standards for these raw materials that we have been required to meet for use in the uranium-graphite pile. It may even be that they have not needed to make either uranium or thorium metal, but can employ compounds of these elements in the system. The construction of a heavy water pile and its operation and attendant chemical extraction work for the preparation of the U^{233} device might be carried out without requiring a serious drain on the manpower or natural resources of the enemy."

I received a copy of a letter from Allison to Thomas referring to a request from G. D. Graves of du Pont that a decision be made as soon as possible on the actual chemical compound of plutonium to be

delivered by the Hanford plant to Site Y. Specifically, an agreement must be made regarding the chemical composition, purity, and physical form of the plutonium compound to be stored at Hanford and shipped from there. The 231 Building (building where concentration step takes place) design at Hanford is now well underway, but the procurement of specific equipment is being held up because of this lack of agreement. In addition, further progress is dependent upon: (1) the development and demonstration of the process to be used to produce the compound decided upon; (2) the development of analytical methods for process control and product characterization; and (3) the specification of the hazards involved, both as to possible toxicity of the compound and to possible critical mass effects in the operation. Allison wrote that we are actively working on all the problems suggested by Graves but are not yet in a position to give answers. Presumably the best compound is the one most readily reducible to a high purity metal. Allison also said we are acutely aware of the possible toxicity of plutonium and are sending some to Dr. Hamilton at Berkeley on February 1 to initiate research on this problem.

In a memorandum to Lawrence Kimpton, the Chief Administrative Officer of the Met Lab, I announced that F. B. Donlan, who is supervisor of our Service Group, will act as the representative of Section C-I in matters pertaining to procurement, property, buildings, and grounds, etc. Until now, Manning has been acting in this capacity but because of the increased pressures on his other duties, he no longer will have time to carry out this responsibility.

Vernon distributed a memorandum to Met Lab people announcing that the Laboratory has leased 23,000 square feet of space in the Museum of Science and Industry, which may be occupied after February 5, 1944. The space is suitable for laboratory work, but radiation and poison hazards must be completely avoided because the Museum is a public building. These new facilities are about one mile from Eckhart Hall, and routine bus service between facilities is planned.

Allison wrote to General Groves about Major Furman's request that we consider the possibilities that the enemy is engaged in any project like those the U. S. is engaged in; that is, the X-10 (pile), Y-12 (calutron), and K-25 (gaseous diffusion) processes. Allison considers it improbable that the enemy may be engaged in the Y-12 process on a scale of military importance. However, since the enemy was considerably in advance of us before the war on work similar to the K-25 process, it is highly probable that if he is doing anything, this is the approach he will take. It was requested that Karl Cohen of the Columbia group be authorized to visit the Met Lab to discuss various methods of carrying out the K-25 type processes.

I held the evening meeting of the Council of Section C-I in my office at 7:45 p.m. Those in attendance were Albaugh, Baumbach, Beaton, Cunningham, Davidson, Dreher, Ghiorso, Kirk, Manning, Orlemann, Thompson, and Willard. I announced that Thompson's group will begin an exchange

of their laboratory and office space (now in the Chemistry Annex) with Sugarman's section (now in the original New Chemistry Building). After the move is accomplished, all Section C-I personnel will be together in the original New Chemistry Building. To minimize health hazards, linoleum and air filters will be installed in our wing of the building (the original New Chemistry Building). A hot lab is being designed and will be installed as soon as possible in the West Stands. Continuous type centrifuges and remote control devices are being considered for installation. Much of Dreher's process development work, as well as the 93^{239} extraction work, will make use of the new hot lab. Other active materials will be stored there also.

I announced that Katzin will take over the patent work from Manning and that Donlan will be responsible for handling office and laboratory equipment and other property problems. I also mentioned that the new counting manual, "A Manual on the Measurement of Radioactivity" (MUC-GTS-407), by Jaffey, Kohman, and Crawford, is now available for distribution and that a report on analytical methods used at Clinton, written by Knox, is also available.

The results thus far obtained on the metal production problem were discussed. The information obtained by Zachariasen on the 12 microgram sample Jasaitis gave him yesterday indicates that plutonium metal has a face-centered lattice with a lattice constant of 4.6 Å. The diffraction lines were broad. The determined density is 16.3 gm/cm^3. Zachariasen found that two phases were present: thorium dioxide and a new phase not hitherto found in earlier tests. Zachariasen believes the new phase is plutonium metal. An unsatisfactory pattern was obtained with a hydride sample. We plan to send Zachariasen a larger metal specimen and another hydride sample tomorrow. In discussions that followed it was agreed that the 16.3 gm/cm^3 value should be considered as a minimum density of plutonium as calcium or other metal impurities might be responsible for a low density value. Kirk considered the 16.3 gm/cm^3 value low by perhaps 5% and stated that metal of a higher purity is necessary before the density value can be adequately established. I suggested that the broadening of the lines could have been caused by impurities in the sample and said that a spectrographic analysis should be run upon completion of the x-ray diffraction studies. Baumbach mentioned that in a recent attempt to reduce black fluoride with calcium, with complete degassing, it was believed that plutonium metal was produced; however, it was so thinly spread in the crucible that it could not be isolated. Orlemann proposed that in order to get plutonium metal in a fused recoverable state, a high heat of reaction of the reducing agent with the fluoride is necessary. Reduction of PuF_4 is to be tried with this idea in mind. Orlemann also proposes to determine the solubilities of PuF_3 and PuF_4 in water, as a function of temperature, in order that the heats of solution may be determined.

Baumbach thinks the last metal production run gave a yield of about 60% and that the yield on a plant scale should be much larger. He stated that plutonium metal oxidizes about 60 times faster than uranium for comparable quantities. He said that Jasaitis' work with the 12 microgram sample yesterday indicates that plutonium metal has a hardness about the same as that of copper.

The schedule for distribution of plutonium was reviewed in detail. I announced that 90 milligrams of plutonium will soon be received from Clinton Laboratories. The needs for plutonium by Thompson's and Cunningham's groups were discussed. In addition Orlemann presented the requirements for the purification program and said that about 42 milligrams will be needed. Of this, 20 milligrams will be assigned to work with metallic plutonium, about 11 milligrams for studies on volatility and general dry chemistry of plutonium compounds, and the remaining 10 to 11 milligrams will be used for work with purification and analysis of plutonium compounds. Orlemann's detailed assignment of plutonium to projects within these three program areas is as follows:

I.	Work with Metallic Plutonium	20 mg
1.	Determination of density by x-ray and capillary methods	4 mg
2.	Determination of the melting point by observing the change in surface tension of fattened pieces	3 mg
3.	Determination of the vapor pressure by an effusion method	2 mg
4.	Determination of the hardness using the micro hardness tester	1 mg
5.	Preparation of the hydride, x-ray determination of its density and structure, and study of the conditions for its formation and decomposition	5 mg
6.	Studies of the purity of the metal as produced by the current techniques	3 mg
7.	Study of the fused salt electrolysis of plutonium using 100 microgram units	1 mg
8.	Study of atomic hydrogen reduction of plutonium halides using 100 microgram units	1 mg
II.	Volatility and General Dry Chemistry of Plutonium Compounds	10 to 11 mg
1.	Preparation and study of plutonium higher fluoride in an apparatus that has been shown to permit satisfactory trapping, revolatilization, and volatility estimation using 1 mg units of UF_4	4 mg
2.	Synthesis and isolation of plutonium halides from PuO_2 using HBr, HCl, CCl_4, and the mixture HCl, CCl_4, O_2 in quartz capillaries	5 mg
3.	Preparation of the chloride and the bromide by the action of Cl_2 and Br_2 on metal to permit the determination of density, composition, vapor pressure, and serve as starting materials for further halides syntheses	1 mg

	4. Systematic investigation of the preparation and properties of the IV and lower fluoride	1 mg
III.	Purification and Analysis of Plutonium Compounds	10 to 11 mg
	1. Study of the purification of plutonium by extraction with ether and nitromethane	4 mg
	2. Study of the purification of plutonium by precipitation using H_2O_2 and $(NH_4)_2MoO_4$	3 mg
	3. Preparation and determination of the stability, melting point, vapor pressure (approximate), and usefulness in extraction of the acetylacetonate	2 mg
	4. Use in analytical work	2 mg

Other miscellaneous technical items were discussed including Kirk's assertion that a micro assay method, correct to 0.1%, must be developed for use at Hanford before the time when quantities of pure Hanford plutonium become available. I agreed with Kirk that this work should begin very soon and it would be the responsibility of Kirk's analysis group at Clinton Laboratories. Ghiorso commented that it might be possible to make direct analysis using instrumental methods by the time Hanford plutonium is on hand.

The current status of 93^{239} supply was also discussed briefly. I directed that one of Dawson's men take over the task of maintaining a supply of 93^{239} for the entire section. Uranium bombardments are to be arranged at Argonne.

Dagmar and Wilma dropped in for coffee with Helen.

I left Chicago around midnight for St. Louis to join Ralph James at Washington University where we plan to bombard plutonium samples with deuterons at the cyclotron in order to look for the possible production of isotopes of element 95.

The war news today comes first from the Pacific New Guinea headquarters. The Allies have sunk six ships at Rabaul.

Friday, January 28, 1944

I arrived in St. Louis this morning and went directly to Washington University to meet Ralph James at the cyclotron. After initial preparations, the deuteron bombardment of our first plutonium sample (target No. 1) was started at 1:45 p.m. This sample consisted of 1 mg of 94^{239} salt which had been evaporated to dryness on a grooved copper plate. The bombardment ended at 4:20 p.m. with a total of 230 microampere hours.

At about 5:20 p.m. the material on target No. 1 was removed from the copper plate with nitric acid. After a few chemical manipulations a portion of the resulting plutonium fraction was counted at various positions in the low geometry counter (the mica window had broken, so

the planned procedure was modified, and the measurements made at atmospheric pressure). In measurements lasting until about 11:20 p.m., we found no evidence for an alpha particle of longer range than the Pu^{239} alpha.

The deuteron bombardment of target No. 2, 0.1 mg of plutonium on a beryllium counting disk, started at 5:30 p.m. and ended at 7:20 p.m., for a total of 242 microampere-hours. This was counted directly, the counts being taken between 7:56 p.m. and 10:33 p.m. Our conclusion is that the counts indicate that an alpha activity with a range longer than that of 94^{239} and a half-life of a few hours has been produced as a result of this bombardment. This radioactivity is very likely due to an isotope of element 95.

A new 0.1 mg Pu^{239} sample was prepared and placed in the cyclotron beam to remain overnight.

I boarded an overnight train for Chicago. James will remain in St. Louis through tomorrow to complete our experiment.

Saturday, January 29, 1944

I arrived back in Chicago at 8:00 a.m. from St. Louis. Helen told me that she had dinner at the Ghiorsos' last night.

I learned that yesterday Frank remeasured the hardness of the 12-microgram piece of plutonium and obtained a value of 88.5 Vickers. This is considered more reliable than Wednesday's determination in which Jasaitis, with Frank's help, determined that the hardness of plutonium is comparable to that of copper.

Yesterday, Willard sent a memorandum to Stearns confirming the transfer of Frederic Schuler from Section C-I to Clinton Labs. Schuler will leave for Oak Ridge next Tuesday at the same time that Gordon Johnson, Dean, and Creek make their transfers. Originally, Schuler was scheduled to leave here on the 15th of this month, but because of the possibility of losing his deferment, it was arranged for him to stay with the Met Lab until he receives his degree from the University of Wisconsin for work done here at Chicago during the semester that closed last Saturday.

A report entitled, "A Brief Summary of Methods for Extraction, Decontamination, Concentration, and Isolation of Plutonium," (CN-1315) was prepared by Thompson, Watt, Beaton, and Manning yesterday. It contains detailed summaries of the methods used in the following processes:

A. Precipitation and Solvent Extraction Methods for Extraction and Decontamination of Plutonium

   1. Bismuth phosphate process

   2. Bismuth arsenate - bismuth phosphate combination process

   3. Lanthanum fluoride process

4. Bismuth phosphate - lanthanum fluoride combination process
5. Sodium uranyl acetate process
6. Bismuth phosphate - sodium uranyl acetate combination process
7. Uranous oxalate - sodium uranyl acetate combination process
8. Ether extraction process
9. Ether extraction - bismuth phosphate combination process
10. Ether extraction - lanthanum fluoride combination process
11. Ether extraction - sodium uranyl acetate combination process
12. Columbic oxide - sodium uranyl acetate combination process

B. Adsorption Methods for Extraction and Decontamination of Plutonium

1. Adsorption extraction cycle
2. Adsorption - bismuth phosphate coupling
3. Adsorption - sodium uranyl acetate coupling
4. Adsorption - lanthanum fluoride coupling
5. All-adsorption process

C. Concentration and Isolation Processes for Use in Conjunction with the Bismuth Phosphate Decontamination Process

D. Fluoride Volatility Method for the Separation and Isolation of Plutonium

Their introduction to the report reads as follows:

"Plutonium, after its formation in the pile, must be separated from uranium (extraction) and from the radioactive fission products (decontamination). Because of the low concentration of plutonium, ~1 gm per ton of uranium at Clinton, ~250 gm per ton at Hanford, most methods of extraction and decontamination require the use of carrier compounds. It is necessary to separate the plutonium from carrier (concentration and isolation) before final purification is undertaken. The purpose of this report is to give a brief outline of several of the procedures which appear promising for large-scale separation of plutonium from pile metal.

"To reduce beta and gamma radiation to a level where personnel can safely handle the product without shielding, decontamination must reach a factor of 10^4 to 10^5 at Clinton and ~10^7 at Hanford. All of the decontamination procedures described below take advantage of the difference in chemical or physical behavior between plutonium in the lower (+4 or +3?) valence state and in the less stable, higher (+6) valence state. To attain a decontamination factor of 10^7, at least two oxidation-reduction cycles will probably be required in addition to the extraction and concentration steps which also contribute to decontamination.

"All of the procedures are probably inherently capable of giving plutonium yields of 90% or greater for each cycle and decontamination factors of 100 or more for a single decontamination cycle. A great deal of additional work must be done on any single method to establish the maximum yields and decontamination factors which can be attained in plant operation. This is true even for the bismuth phosphate and lanthanum fluoride methods which have been more thoroughly investigated in plant and semiworks operations than any of the other methods."

This is a very valuable compendium.

The fourth shipment of plutonium arrived from Oak Ridge today. Of the 100 milligrams, 65 milligrams are allocated to us for our chemical and metallurgical studies for Los Alamos, and 35 milligrams are allocated for our chemical and other work for Hanford and for the determination of fundamental physical and chemical data. Another 100 milligrams of plutonium went directly from Oak Ridge to Los Alamos.

Beaton sent a letter to F. J. Myers about the preliminary testing results on two samples of new resin materials, Amberlite XE-12 and XE-26, recently received from Myers. The XE-26 sample was entirely unsatisfactory since it immediately disintegrated into a gelatinous paste, even when soaked in water alone. With the XE-12 sample, the adsorption of "thorium" was not high enough to make use of the resin practical in either the extraction or decontamination step.

I attended a meeting of Chemistry Section Chiefs in Hogness' office. Mrs. Eunice Eyre, the deferment clerk in Personnel, was invited in to say a few words about the draft situation. She said that it is important that those men we wish to get back after induction should be assigned to the Army and that we should notify the deferment office about such individuals well in advance of their pre-induction examinations. After she left Hogness reviewed the steering committee meeting that we attended last week in Oak Ridge and the Purification meeting here with Thomas two weeks ago in light of our commitments to Site W and Site Y. Concerning reports, Hogness said that bi-monthly reports may be required in the future instead of the currently issued monthly reports. This matter will be taken up at the next meeting. It was announced that Pierce W. Selwood will be joining the Chemistry Division next Tuesday.

Helen had lunch at Alice Thompson's this noon.

Russia continues to loosen Germany's grip on the Leningrad line. The Soviets have cleared new lines around Leningrad and have taken a town 35 miles from the Estonian boarder.

Sunday, January 30, 1944

The main headlines in the Sun today deal with bombings of Germany: "1,500 Bombers Blast Nazis," "Berlin Aflame; Frankfurt Hit," and "Greatest Day Raid by U.S. Pounds Industrial Center."

Monday, January 31, 1944

James returned to Chicago at 7:00 a.m. from St. Louis. He brought me up to date on his results after I left St. Louis last Friday night. On Saturday he repaired the mica window on the low geometry counter so it could be operated in the vacuum mode. The 0.1 mg Pu^{239} sample left in the cyclotron beam overnight had not received sufficient bombardment by morning so the irradiation was continued strongly until about 4 p.m. James began counting the sample at 4:25 p.m. and continued counting periodically until around 8:30 p.m. James concluded that the results of the counts definitely show the absence of any appreciable amount of alpha-active nuclides with energy greater than that of 94^{239}. However, the data obtained do leave the possibility, and even show some evidence, of the formation of an alpha activity with a range shorter than 94^{239}. He believes the new activity is probably due to an isotope of element 95; the only other possible explanation is that there may have been greater self-absorption than in the non-bombarded sample used for comparative purposes. Continuing his measurements on Sunday, he found no evidence for decay or growth of long range alpha particles.

Robert H. Goeckermann started work in Section C-I. He has just received his B.S. in chemistry from the University of Wisconsin and will work as a Research Assistant under George Watt. A new technician, Donald B. Wetlaufer, began working in Dawson's group.

Compton wrote to Roger Williams at the du Pont Co., Wilmington, Delaware, about recent discussions with Stone relating to the fact that no provision has been made on the staff at Hanford for a medical man who is thoroughly acquainted with the physiological effects of radiation. Although Dr. William D. Norwood and Dr. Fuqua are being trained at Oak Ridge in the type of tasks that are needed to detect early physiological changes, there is no opportunity to demonstrate clinical examples of damage due to radiation. Compton pointed out that this is a special field of medicine with which most physicians are unacquainted. Several doctors on the project have had this experience, however, such as Drs. Stone, Cantril, Warren, Friedell, and Nickson. All these men are available as consultants, but Hanford should have a physician with such training continually at hand in order that dangerous conditions may be recognized promptly and, if excessive exposures occur, that immediate diagnosis will be possible.

Compton said that Stone also informed him that he is concerned about the arrangements at Hanford for taking measurements of the radiation exposure throughout the entire Site W area. He said Stone feels that the primary responsibility for these measurements should be in the hands of a Health Officer and that the instruments should be provided in accordance with the requirements that this Officer specifies. Compton mentioned Herbert M. Parker, who is currently in charge of the radiation health measurements at Clinton Laboratories, is the best man we now have available for such work. He said that Stone is agreeable to releasing Parker for service at Hanford.

1/31/44 (cont.)

Compton received a memorandum from Major A. V. Peterson confirming a change in the allocations and schedule of plutonium from Clinton Laboratories as agreed upon among Thomas, Oppenheimer, and Compton. The changes consist in the allocation of 2 grams of plutonium to Los Alamos and none to Chicago on March 1, and an allocation of 1.5 grams to Chicago and none to Los Alamos on March 15. Peterson includes a listing of all past and planned product allocations of plutonium to Los Alamos, Chicago, Berkeley, and Ames. Up to the present date 0.225 gram has been delivered by Clinton Labs (consisting of shipments to Chicago of 0.0015 gram on December 30, 0.001 gram on January 6, 0.0165 gram on January 17, and 0.100 gram on January 25, and shipments to Los Alamos of 0.006 gram on January 17 and 0.100 gram on January 25). It is expected that by May 1 of this year 14.5 grams of plutonium will have been shipped from Oak Ridge.

Harry Brown and Wendell Latimer are in town. They had dinner with Helen and me in our apartment this evening, after which the three of us attended the regular Monday evening Chemistry Division seminar held in Room 251, Ryerson Laboratory.

The U.S. fleet has launched a big attack on the Marshall Islands, bombing and shelling Roi Islet and Maloelap, Kwajalein, and Wotje Atolls.

Tuesday, February 1, 1944

This was the last day at the Met Lab for George Creek, Floyd Dean, Gordon L. Johnson, and Frederic Schuler, who are transferring to Clinton Labs. Dean had been scheduled for transfer earlier, but he asked to remain in Chicago while his wife had some intricate dental work completed.

Thomas E. Phipps started work in Section C-I. He is on leave from the University of Illinois, where he is a professor of chemistry. Phipps received his Ph.D. from the University of California in 1921, which makes him the oldest chemist in my section. Hogness and I made a special effort to obtain his services because he is an outstanding expert in vacuum techniques and, we believe, can be of great help in our program of production and measurement of the properties of plutonium metal.

Lyle Dawson, accompanied by Dr. James Nickson, left Chicago at ten o'clock this morning for Boston to visit the Luminous Engineering Company. They will study the radium dial painting industry in connection with the health hazard problem associated with plutonium.

I am using my notebook as a repository for some of my conceptions. Today I wrote the following:

"(A) The extraction, decontamination and concentration of 94^{239} is accomplished by a number of 'oxidation-reduction' procedures in which an 'upper oxidation state,' defined as fluoride soluble state or +6 state, and a 'lower state or states,' defined as fluoride insoluble states or +4 or +3 states, are the two states or two sets of states involved. I suggest that analogous methods can be worked out in which the +4 and +3 oxidation states may be involved as the 'upper' and 'lower' states respectively. Either of these may be the one in which the product is carried in the extraction step (i.e., the step in which the product 94^{239} is separated from the bulk of the uranium).

"In the decontamination and concentration steps the product 94^{239} may be carried while it is in one of these two oxidation states (+4 or +3) and the fission products may be carried (and the product not carried) while the product is in the other of these two oxidation states. This principle can be so general as to include, in addition to precipitation reactions, electrolysis procedures (in which the product is deposited when it is in one state initially and the fission products are deposited when the product is present in solution in the other state), solvent partition methods (in which the product is extracted into some solvent when it is initially present in solution in one state and the fission products are extracted into the same or a different solvent when the product is present in the other oxidation state). In the solvent partition methods the product when present in the +3 state (and the same applies, of course, when it is in the +4 or +6 state) may be complexed with some inorganic or organic complexing agent or agents so as to give rise to a 94^{239} complex which is

specifically extracted into the second solvent. The +3 oxidation state may be useful for extraction, decontamination, or concentration by dry methods (as is the case for the +4 and +6 state) by forming volatile compounds, especially halides, of this oxidation state.

"(B) The +3 oxidation state should be useful as the state of 94^{239} which is involved in purification methods, i.e., the removal of the light element impurities (e.g., Li, Be, B, etc.) which give rise to the α,n reaction in the final device and hence must be removed in order that the final device shall work properly. An insoluble compound (inorganic or organic) of the +3 oxidation state of 94^{239} may be precipitated, this precipitated compound being so chosen as to be specific or partially specific in its separation from these light element impurities. A solvent partition method, involving the +3 state, either as a simple salt or an inorganic or organic complex, may be used to purify the product or aid in the purification of the product. A volatile compound of the +3 state, especially one of the halides, might be very useful in this purification step. Some compound of the +3 state might be very useful in the metal reduction step, either by (1) the thermal reduction of some such salt (e.g., a Pu(III) halide reduced by an alkali or alkaline earth metal), (2) the electrolysis of some such fused salt probably dissolved in another fused salt medium, (3) reduction by atomic hydrogen, and/or (4) decomposition or "cracking" of a halide by contact with a hot surface."

A manpower distribution table issued by Hogness shows the following for my section.

		Persons
Thompson (Site W work, 20 men)	Albaugh, extraction and decontamination	7
	Watt, concentration	5
	Dreher, process development	8
Orlemann (Site Y work, 24 men)	Orlemann, precipitation and extraction	6
	Davidson, volatility	7
	Baumbach, metal production	11
Cunningham (24 men)	Cunningham, basic chemistry	7
	Dawson, recovery	9
	Ghiorso, instruments	8

A report of the Chemistry Division, prepared for use at the next Project Council meeting, breaks down the personnel distribution for my section in a different way:

	January	February
Basic Chemistry	7	8
Instruments	8	8
Recovery	4	7
Fundamental Process Research	4	7
Process Development	5	7

(cont.)

2/1/44 (cont.)

	January	February
Concentration and Isolation for Site W	3	5
Adsorption	5	0
Training	4	7
Purification and Analysis	7	6
Volatility and Dry Chemistry	4	7
Metal	9	11
	60	73

Helen and Peg Beaton had coffee at Wilma Ghiorso's. After work, Helen and I had dinner at Isbell's Restaurant with the Baumbachs.

More bombing reports arrive from the Pacific. Wake Island and seven other Marshall Islands have been bombed by the U.S.

Wednesday, February 2, 1944

I received a letter from Jane Dorn of the Department of Chemistry at Berkeley asking on behalf of Gofman for twelve additional reprints of my article, "Artificial Radioactivity," which appeared in Chemical Reviews in August 1940.

Based on the work done so far on the search for new heavy elements and isotopes, Ralph James and I conclude from the one-day Clinton neutron irradiations (a) the half-life of U^{236}, produced from uranium, must be greater than 10,000 years and (b) the half-life of 94^{240}, produced from 94^{239}, must be greater than one year, or less than 15 minutes. The possible products (95^{240}, 94^{240}, or 95^{239}) of the deuteron bombardment of 0.1 mg of plutonium at St. Louis must have partial alpha half-lives of (a) greater than 0.1 year to 1 year or less than 5 minutes, if the alpha particle has a range greater than 3.85 cm, or (b) greater than 0.01 year or less than 5 minutes, if the alpha particle has a range less than 3.85 cm.

I am carrying two plutonium samples to Oak Ridge with me tonight for neutron irradiation. One consists of 0.1 mg on a beryllium disk prepared by James, and the other is about 7 mg as the hydroxide prepared by Marty Studier. The purpose is to look for the possible production of Pu^{240}.

In the evening I presided with the help of Thompson at the triweekly meeting of the Extraction, Decontamination, Concentration, and Process Development Groups of my section at 7:45 in Room 209, Eckhart Hall. Present were Christiansen, Dreher, Flox, Gilbreath, Hoekstra, Hyman, Kircher, Manning, Margolis, Thompson, Watt, Willard, and others. In a discussion of the major problems at Site X, I mentioned that yields have fallen to 60% at certain stages and then gone back up to 80% in a subsequent stage, indicating holdup of product in centrifuges and tanks. Thompson pointed

out that the same equipment is used for both oxidation and reduction and for both product and by-product precipitations. There are also problems of oil contamination that present special difficulties during the concentration steps, and the crud problem that causes trouble in the metathesis step. Thompson indicated that Hanford problems now being studied by his group are carrying and decontamination with Site W concentrations of plutonium and inactive fission product elements, testing of concentration and isolation processes, and the effect of plutonium fission products on the process.

Dreher reviewed the semiworks operation, indicating that a bismuth phosphate extraction and one decontamination cycle have just been completed on Site X pile uranium with a product yield of 78.3%; he cannot account for the loss. Runs 12 and 13 are now underway. Hyman discussed work on dissolving the three different protective coatings being considered for use on Hanford slugs (zinc-tin solder, Al-Si, Zn-Al). In a discussion of the tests of the isolation steps, Watt and I agreed that 100 milligrams of plutonium are really needed for best results.

At 11:50 p.m. Dreher and I left Chicago's Englewood Station on the Penn R.R. to Cincinnati where we will again continue on the L&N R.R. to Oak Ridge. I have the two plutonium samples and will attend a meeting of the Project Steering Committee at Clinton Laboratories to review their program and ours about process work for the coming February 15-March 15 period. We shall return to Chicago Sunday.

The Marshall Islands have been invaded by U.S. troops! What is reported to be history's biggest fleet has landed on two islands — Roi and Kwajalein.

The Project Council Policy Meeting was held at 9:30 a.m. in Room 209, Eckhart Hall. Present were Allison, Chipman, Compton, C. M. Cooper, Dempster, Fermi, Greager, Greenewalt, Hilberry, Hogness, Jeffries, Miles, Mulliken, A. V. Peterson, Smyth, Stearns, Stone, Warner, C. J. Watson, W. W. Watson, Whitaker, and Wigner. Compton summarized the main tasks to which we are committed at present: (1) Get information needed to make Hanford go; (2) Train personnel for Hanford; (3) Supply materials for Los Alamos; (4) Make fundamental experiments needed by Los Alamos and which can properly be carried out at other locations. He further observed that looking down the line we must maintain a standby organization to care for the as yet undefined problems and emergencies and also look toward improvement of process and recovery and re-use of valuable materials. Further along the line will come power development, etc. Compton considers we are now at the peak of Hanford problems, and we are beginning to feel demands from Los Alamos; Hanford work will probably decrease and Los Alamos increase. Compton announced that program responsibility for Ames is now in the hands of the Site Y people. Spedding, however, will continue as a member of the Project Council.

The distribution of work effort for some of the Divisions was given by Stone for Health, at both Chicago and Clinton; Fermi for

Nuclear Physics; W. W. Watson for General Physics at Chicago; and Whitaker for Physics at Clinton Laboratories. In presenting the Clinton Laboratories report, Whitaker stated that 3/4 gram of plutonium is already out of the 205 building, with nearly a gram due for release early next week. Recovery through bulk reduction is now around 56%.

Thursday, February 3, 1944

Dreher and I arrived in Oak Ridge in the afternoon. I spent the remainder of the afternoon conferring with Perlman and visiting the laboratories in the chemistry building to talk with the chemists in his section. I am staying with the Coopers again.

Friday, February 4, 1944

The two samples I brought with me from Chicago, i.e., the 0.1 mg plutonium sample mounted on a beryllium disk and the 7 mg sample of $Pu(OH)_4$ sealed in a tube, were placed in the Clinton pile for neutron irradiation.

Hogness and I attended the Steering Committee meeting to discuss Chicago and Oak Ridge programs and distribution of manpower for process work, including isolation and concentration of plutonium. The manpower available was listed as follows. Clinton Laboratories, Perlman's section: Process Research Group--Miller, Kroner, Dam, Scott, Germany, McBride, G. L. Johnson, Kay, Halperin; Process Service Group--Hill, Bohlmann; Product Isolation and Concentration Group--V. R. Cooper, Covey, Firminhac, Gevantman, Lindner, Farabee; Product Chemistry Group--Werner, Fries. Chicago, Seaborg's section: Process Research Group--Albaugh and 6 men; Process Development Group--Dreher and 6 men; Concentration Group--Watt and 4 men.

We spent most of the meeting reviewing the tasks ahead of us. In so doing we made the following problem assignments. Clinton Laboratories--(1) Dissolution of Bismuth Phosphate Precipitates; (2) Tetravalent Uranium as a Reducing Agent; (3) The Behavior of High Concentrations of Plutonium in the Bismuth Phosphate Process (Completed. Report will be issued February 15.); (4) Use of Ceric Ion as an Oxidant in the Complete Bismuth Phosphate Process; (5) Search for New Oxidizing Agents--A Survey; (6) Oxalate as a Reducing Agent (Completed. A report will be issued February 15.); (7) Decontamination in the Crossover Cycle of the Bismuth Phosphate Process at Clinton; (8) Use of Ferrous Iron as a Reductant for Plutonium in Fluoride Solutions (Completed. Report will be issued February 20.); (9) Ceric Method of Concentration; (10) Isolation of Plutonium from Room D Concentrates; (11) Alternate Methods of Hydroxide Metathesis of Lanthanum Fluoride in the Concentration Cycles (Completed. Report will be available February 25.); (12) Uranous Oxalate Method for Concentration of Plutonium; (13) Concentration Processes Involving the Use of Hydrochloric Acid; (14) Development of Radiochemical Analysis Procedures; (15) Use of Bismuth Phosphate for Concentrating Plutonium from

Lanthanum Solutions (Completed. Report will be available February 25.); (16) Solubility of Plutonium Peroxide; (17) Investigation of Plutonium Compounds Suitable for Storage and Shipment; (18) Purification of Plutonium from Lanthanum by Peroxide Precipitation. Chicago Process Research Group--(1) Search for New Oxidizing Agents; (2) Uranous Ion as a Reducing Agent; (3) Bismuth Phosphate Process Research at W Concentrations; (4) Bismuth Phosphate Process Research--Decontamination Studies at W Concentrations of Plutonium and Fission Products; (5) Bismuth Phosphate Process Research--General Problems; (6) 93-94 Separation; (7) Crystal Structure of Bismuth Phosphate Containing Plutonium; (8) Decontamination--Mass Spectrographic Studies (with Dempster); Concentration Group--(9) Isolation at Hanford--Peroxide Method. (10) Concentration at Hanford--Fluoride Method. Chicago Process Development Group--(11) Metal Coatings--Hanford; (12) HCl Concentration for Semiworks; (13) Ether Extraction-Bismuth Phosphate Decontamination; (14) Semiworks Waste Disposal; (15) Construction of Hot Laboratory; (16) Process Service; (17) Clinton Flowsheet.

As is apparent, Clinton Laboratories does not propose to study any Hanford problems in this period. Most likely, however, Hanford problems will be started at Clinton Labs the following month. The next meeting of this committee is scheduled for March 10 and 11 here at Clinton.

Saturday, February 5, 1944

I was informed that Clinton Laboratories today made its first shipment of pile-produced plutonium to Berkeley. Latimer's group will receive 20 milligrams, and Hamilton will receive 11 milligrams for metabolism tests to determine toxicity. A much larger quantity of 475 milligrams was consigned to Los Alamos.

Dreher and I ended our visit and boarded a Southern R.R. train to Cincinnati; we will then continue on the New York Central R.R. to Chicago. I have with me the plutonium and uranium samples (small quantities mounted on beryllium or platinum counting disks) that James placed in the Clinton pile on January 9 and which were removed today.

Sunday, February 6, 1944

Dreher and I arrived back in Chicago at 1:30 p.m. After I checked in at home and dropped my bags, Helen and I got on the streetcar and rode to Brownsville. During our ride Helen told me of her activities while I was in Oak Ridge. On Thursday and Friday she had coffee at Wilma's. Thursday she had dinner at Don the Beachcomber's on the near North Side with Dagmar Dreher and Nathalie Baumbach, and yesterday Nathalie and Wilma came to our apartment to help Helen proofread the "Table of Isotopes." Following this, Helen had dinner at Thomas' Drugstore with the Ghiorsos and then played cards with them at their house.

As soon as we arrived in Brownsville, we went to the Regal Theater to see the stage show featuring the Ink Spots, Ella Fitzgerald, and Cootie Williams. We then returned to our neighborhood and had dinner at Isbell's Restaurant. Afterwards we played cards at the Ghiorsos'.

2/6/44 (cont.)

When I found time to pick up the paper, I saw the Sun headline "Yanks Take Kwajalein Isle." This is the opening offensive against Japan's pre-Pearl Harbor empire.

Monday, February 7, 1944

When I arrived at the lab I was brought up-to-date on what happened during my absence. Larned B. Asprey, a member of the U.S. Army Special Engineer Detachment (SED), who received his B.S. from Iowa State in 1940, started work in Section C-I last Thursday. Another member of SED, J. W. Britain, who earned his B.S. at Rice Institute, Houston, in 1942, joined us Saturday. Both he and Asprey will work under Dawson as Research Assistants in the plutonium recovery operation. Thursday Frank Belletire resigned to enter the Army, and one of our secretaries, Elsy Anderson, also terminated because her husband has been transferred.

Halban and Placzek of the British-Canadian Atomic Energy Project in Montreal arrived here Thursday to engage in discussions with Laboratory personnel. On Thursday afternoon, Halban and Placzek discussed with Fermi the following topics: (a) The diffusion theory developed by Placzek for the study of the density of neutrons near a boundary. (b) Pontecorvo's experiments on the neutron transport mean free path. (c) The experiments performed both at Montreal and in our project for the determination of resonance capture of neutrons. On Friday morning, Halban and Allison discussed the following topics: (a) Liaison between the two Projects concerning health protection. (b) Need of the Montreal group for an electron microscope. (c) The next visit of a delegation from Montreal, which is arranged for later this week (February 9-12). (d) The desirability of imparting information to the Montreal group on the chemistry and the physical properties of plutonium. Halban asked for 20-100 micrograms of plutonium to be used in an experiment to study the valence states of plutonium. A letter is being sent by Allison to General Groves requesting a ruling on this matter. (e) The needs of the Montreal group for uranyl fluoride. On Saturday morning, Halban and Allison discussed the mechanism of transfer of reports to the Montreal group. Halban left on Saturday afternoon, February 5, after having a brief discussion with A. C. G. Mitchell about the resonance capture experiment, during which Placzek was present. Placzek is staying until tomorrow. His conferences are almost exclusively with Wigner about the theory of the slowing down of neutrons and other theoretical matters.

On Thursday Beaton sent a letter to F. J. Myers of The Resinous Products and Chemical Company informing him that studies of an all-adsorption process are being discontinued temporarily, at least, and therefore Myers' planned activities should be postponed or suspended. Beaton indicated that we will be continuing the Amberlite IR-1 work for a short time, however, in order to determine its prospective use as a single step in the process.

Three evening meetings were held on Thursday. (1) There was a meeting at 7:45 in Room 133, Eckhart Hall, open to all scientific and technical personnel at which T. E. Hall spoke about security problems,

and Stone gave an illustrated lecture on health problems. (2) The evening meeting of the Research Assistants of my section was held at 6:45 in Room 209, Eckhart Hall. (3) At 7:45 the Council of my section met in my office and was attended by Albaugh, Baumbach, Cunningham, Davidson, Ghiorso, Orlemann, Thompson, and Willard. Willard, who presided, told me the high points of this latter meeting. The Council members were informed that Boyd's adsorption group at Clinton Laboratories has been curtailed and Beaton's group at Chicago has been dissolved. Beaton will transfer to Clinton Labs to do engineering design work on adsorption equipment. Other personnel changes: Phipps will join Baumbach's group with Seifert and Turnbull assisting him. Beaton's men have been reassigned, La Chapelle to work with Cunningham, Lincoln to work with Dreher, and Sheft to work with Albaugh. Fields in Dawson's group will maintain the supply of 93^{239}. Selwood from Northwestern University will function as a special assistant to Hogness. Schedules for regular, frequent washing of benches, desk tops, and floors are being arranged as a health measure. There was discussion of the use of glass plates and trays on desk tops for health protection. Baumbach reported on three plutonium metal specimens, 25 micrograms each, which have been given to Zachariasen after being flattened out to give a sufficient surface area for testing. The two samples that were calcium-reduced gave diffraction lines--the one reduced with lithium gave no lines; spectroscopic analysis has shown the samples are only about 80% plutonium. A low-temperature lithium reduction was made on Thursday yielding plutonium in the powdered form, but x-ray results are not yet available. Metal has also been produced by reduction of plutonium fluoride with barium; this should be better than that produced with the other reducing metals from the standpoint of impurities introduced by the reductant. Baumbach stated that Zachariasen now believes the reduction product from the black fluoride to be PuO instead of Pu_2O_3. Baumbach further reported that in a reduction of PuF_4 with atomic hydrogen, PuO_2 is not formed; hence, worry over oxygen contamination can be eliminated. Jasaitis is building a platinum foil heater for microscopic observation of the melting point of plutonium metal. Willard stated that the canning problem has priority at present. Our work shows that the aluminum-silicon bond, provisionally accepted for use at Site W, does not dissolve in HNO_3; caustic dissolves the alloy readily but the hydrogen evolved may interfere with the subsequent extraction process.

Dawson is back at the Laboratory. He returned Friday at 1:30 a.m. from his visit to Boston to study health protection procedures used in the radium industry.

Gofman and Sheline sent me a joint letter which arrived Saturday. They were following up on a recent conversation with me at Oak Ridge in which I mentioned the possibilities of positions at Site W. They asked for details of the proposition such as type of work, rank or position, and salary.

Also in the mail Saturday was a letter from Eastman saying that, in response to my wire of January 26, Bromley has prepared and sent me a sample of uranium disulfide.

2/7/44 (cont.)

Edrey Smith answered Jane Dorn's request saying that Gofman will be sent twelve copies of the reprint "Artificial Radioactivity" when additional copies are made up in a month or so.

James took alpha counts on the plutonium and uranium samples I brought back from Oak Ridge. The plutonium sample has an alpha activity the same as before irradiation, within limits of counting error. The uranium sample shows a very definite increase in alpha activity, possibly representing production of U^{236}.

Today Ray Frank measured the hardness of a 36-microgram piece of plutonium metal made by barium reduction of PuF_4 and obtained values on the Vickers hardness scale of 112, 181, 155, and 155.

I replied to Kennedy's letter of January 17, (which arrived late on February 1, the day before I left for Site X), concerning the importance of our taking the initiative in submitting patent applications covering the early work at Berkeley on the discovery of plutonium and the determination of the slow neutron fissionability of Pu^{239}. I indicated that I find it difficult to make up the lists of the types of agreements acceptable and unacceptable to me as he requested. I suggested we discuss the matter when he is here next week.

Pierce W. Selwood of Northwestern University has been appointed Assistant Director of the Chemistry Division effective today.

Manning and I attended a meeting of Chemistry Section Chiefs in Hogness's office. Selwood was also present. I reviewed my recent trip to Clinton Laboratories saying, among other things, that the chemical extraction plant there is now dissolving one-ton batches and that the current rate of plutonium production is three grams per month. Other topics of discussion included reports. It was concluded that the present system will continue, but that more summary reports will be prepared and bound separately.

Nathalie and Wilma came to our apartment this afternoon and again helped Helen proofread the "Table of Isotopes." Helen and I had dinner at the 51st Street YMCA with the Baumbachs and Ghiorsos.

U.S. troops have taken three more islands in the Marshalls.

Tuesday, February 8, 1944

Ralph James, in order to check on the apparent production of U^{236} in the 0.1 mg uranium sample No. 23-Pt-U, neutron-irradiated in the Clinton pile between January 9 and February 5, obtained some waste solution of Clinton pile-irradiated uranium from the semiworks and isolated pure uranium in the form of U_3O_8. His measurements of counting rates from this uranium give no evidence of production of U^{236}.

2/8/44 (cont.)

I sent a memo to Allison indicating that, as part of the general problem of concentration and isolation at Hanford, we are including the investigation of suitable procedures for operation at Hanford during the start-up period when there will be low plutonium concentrations. This problem was raised in a January 6 letter from G. D. Graves to Whitaker and discussed during the planning conference on the Bismuth Phosphate Process held at Oak Ridge on January 7 and 8.

The Metallurgical Laboratory monthly report for January (MUC-SKA-500) has been prepared for distribution through Allison's office. The report contains progress reports from the Divisions of Nuclear Physics, General Physics, Chemistry, Technical, and Health.

Studies by the Nuclear Physics Division on the resonance capture of neutrons by uranium indicate a less favorable reproduction factor in the homogeneous P-9 pile than previously supposed. Experiments have shown that 2.2 neutrons are emitted per fission of U^{236} (formed by neutron capture in U^{235}). At present, Nuclear Physics has an academic staff of 65 persons.

The General Physics Division studies on the radioactivities produced in water pumped through the Clinton pile are reported as they relate to Hanford. The report states that irradiated bismuth slugs received from the Clinton pile were measured for their radioactivities in preparation for polonium production at Hanford. The density of metallic plutonium as determined by x-ray diffraction is reported as being not less than 16.4 ± 0.5 gm/cm^3. General Physics currently has an academic staff of 81 persons.

In the section covering the Chemistry Division's activities, it is reported that radiation for 500 hours at Hanford levels shows no effect on aluminum corrosion in flowing water. The following additional information is included:

"Dissolution of aluminum jackets bonded with zinc can be readily accomplished without excessive loss of uranium. The zinc and aluminum carried into the decontamination process seem to have no bad effects. The Al-Si bond does not yield to the acid procedure and although NaOH will dissolve it, the H_2 evolution is a disagreeable factor with which to contend.

"Improvement in decontamination is found when hold-back carriers of lanthanum, yttrium, Ce^{+3} are used, and U^{+4} is showing good results as reducing agent in the phosphate process. Plutonium peroxide solubility measurements are confirming the basis on which the previous flowsheet was drawn up.

"A very important result of the work on plutonium is the discovery of the +3 state of oxidation. The oxidation potential of the Pu^{+3}-Pu^{+4} couple has been roughly determined as -0.54 to -0.77 volts. A rough determination has been made of the solubilities in excess precipitating agents of the fluoride in H_2SO_4, yielding the +3 state. All proposed reduction steps for the separation processes must be reviewed in the light of this new variable.

"The alternate processes of ether extraction and resin adsorption have received some attention. It has been found that an ethyl ether

extraction is an excellent procedure for final purification of plutonium from thorium, lanthanum, chromium, bismuth, platinum, and sodium. Using various precipitants on uranium solution contaminated with known elements, dissolving and oxidizing the precipitate and extracting with ether did not remove below tolerable level the elements noted (the precipitants are shown in parenthesis) boron, beryllium, sodium, phosphorus, silicon (IO_4^-); boron, beryllium, phosphorus (SO_3^{--} or $C_2O_4^{--}$); boron (MoO_4^{--}).

"The vapor pressure of $PuBr_4$ (?) has been roughly estimated (450°C) as 1/4 that of UBr_4 at 7×10^{-2} mm Hg.

"Efforts to produce plutonium metal by reduction of the white fluoride with calcium were apparently successful, yielding a malleable material, giving a new x-ray pattern resulting in a density of 16.4 ± 0.5. Reduction by barium and lithium produces what appears to be metal (malleable) but gives a still different pattern, density 16.2. The micro hardness tester indicates a hardness of plutonium similar to copper.

"Work has been begun on the preparation of standards for alpha counters for precision work. About 55 micrograms of Np^{237} have been isolated in a state of 25 to 50% purity."

The Chemistry Division reports an academic staff of 188 persons.

The Technical Division portion of the report states that a continuing main problem is the development of a satisfactory canning process for Hanford pile slugs. The single can, zinc-tin bonded and argon arc-welded, looks promising. Work is also progressing on refractory crucibles for high purity metal production, with BeO and TiN being considered. A semiworks is being constructed to evaluate the ether extraction processes that show promise. Theoretical and design work is being done on the resin adsorption processes. The semiworks unit has continued dissolution of Site X slugs for the Chemistry Division. The Technical Division personnel presently include 137 technical persons in Chicago and an estimated 51 outside Chicago.

The portion of the report covering the Health Division's activities states that out of a total of five individuals thought to have abnormally high blood counts, four are in Cunningham's group. Gross contamination of laboratory table tops has been found in the New Chemistry Building. During the month the possible toxicity of plutonium received considerable attention. Active steps are now being taken to protect individuals from plutonium hazards.

The Health Division reports an academic staff of 39½ persons.

Willard, Kircher, Christiansen, Bird, Thompson, Dreher, and I held a meeting to discuss the results of Chicago semiworks runs 11, 12, 13, and 14. Progress is being made in testing a number of variables in the Bismuth Phosphate Process. I told the group that I doubt we will be able to utilize the Clinton pile-irradiated uranium metal at the rate we have set up to receive it (250 pounds per month).

Iz Perlman is visiting the Met Lab today and tomorrow. English will follow next Monday for a four-day visit.

Helen had coffee at Wilma's. She and I attended a farewell dinner for the Beatons, who are leaving for Oak Ridge, at the Red Star Inn (1528 N. Clark).

2/8/44

Detailed reports on the fighting on Kwajalein are coming in. At the same time the Germans are pouring heavy fire on the Allied beachhead at Anzio.

Wednesday, February 9, 1944

Saturday Hyman successfully decanned an Al-Si bonded slug in 10% HNO_3, 2% HF, 0.1% $Hg(NO_3)_2$; however, the corrosion was excessive, even with only 1% HF. Today studies of the coating removal problem began in earnest with Winner assigned to study the effect of zinc on the process, Flox to study the effect of Al-Si in NaOH on the process, and Hyman to continue the study of solution in NaOH plus an oxidizing agent (to reduce the hydrogen evolution which gives rise to an explosion hazard).

"Chemical Research--Separation Processes for Plutonium. Report for Month Ending February 1, 1944" (CN-1258) is being issued. The following research is described: Creek, Schuler, and Dreher tested the effect on the Bismuth Phosphate Process of slug coating-materials containing tin. They find that insoluble metastannic acid is formed when the slugs are dissolved and that the precipitate follows through the by-product precipitate in the first decontamination cycle, where it carries 12 to 35% of the product. G. Johnson tested the Bismuth Phosphate Process with Site W concentrations of plutonium and inactive fission product elements on a 0.5 cc scale. Through an extraction and one decontamination cycle the overall yield is 97%. Hopkins and Watt include tentative flowsheets for the concentration and isolation steps at Site W and identify some of the specific problems which must be considered. Concerning nomenclature, they suggested that the steps variously referred to as "Concentration I," the "crossover cycle," and "the bulk-reduction step" be designated as "concentration" and that those operations that first result in the isolation of a compound of plutonium in pure form be designated by the term "Isolation." They also have carried out experiments on the solubility of plutonium peroxide in nitric acid and obtained results in fairly good agreement with values reported previously. They emphasize the danger of shipping plutonium in aqueous solution in closed containers because of pressure built up by hydrogen produced by the decomposition of water from the action of the alpha particles. I point out that the recent experiments confirming the existence of a stable +3 oxidation state of plutonium leads to the probability that separation processes utilizing the difference in properties of the +3 and +4 oxidation states may prove useful. The use of these two states may be significant with regard to any one of the types of process, including precipitation methods, adsorption methods, solvent extraction methods, and dry volatility methods.

Our triweekly meeting of the Purification and Metal Production Groups of Section C-I was held as usual at 7:45 p.m. in Room 209, Eckhart Hall. Present, besides Perlman and me, were Brody, Dirksen, Florin, Frank, Fried, Ghiorso, Hagemann, Jasaitis, Katz, Manning, Meyer, Orlemann, Willard, and others. Orlemann helped conduct the meeting. I summarized the present status of the determinations of plutonium metal density, stating that current values are 16.2 gm/cm^3 by the capillary method and 16.4 gm/cm^3 by the x-ray method. Fried discussed the degassing technique now used in plutonium metal production, which involves cooling in an argon atmosphere before loading the

crucible. Florin described the production of plutonium fluoride now being prepared on a larger scale; he disclosed that the so-called white fluoride is actually brownish in color and x-ray analysis shows it to have a structure similar to UF_4. Dirksen discussed the rapid thermal reduction of PuF_4 with the object of preparing spongy plutonium metal for x-ray analysis. Success has not been attained, but results indicate plutonium may have a lower melting point than uranium. I mentioned the future plans to try reduction of halides other than the fluoride and that production of the chloride by wet methods has some promise.

Frank discussed hardness tests which give values on the Vickers scale of 90 for calcium-reduced plutonium (85% assay) and 160 for barium-reduced plutonium (96% assay). For comparison, the value is 300 for uranium and 94-130 for copper. Brody spoke on solvent extraction purification methods; solvents showing promise are methyl-n-amyl ketone, methylisobutyl ketone, and methylethyl ketone-xylene mixture.

This morning Helen had coffee at Wilma's. Iz Perlman ate dinner with us in our apartment. Wilma visited Helen during our evening meeting, and afterwards Al Ghiorso and Iz dropped in for coffee.

Resistance by the Japanese has ended on Kwajalein, but Germans are launching new attacks on the Allies at Anzio in Italy.

Thursday, February 10, 1944

Working on the assignment to isolate microgram amounts of Np^{237} from cyclotron-bombarded material, La Chapelle and Magnusson have outlined a proposed separation method that involves first separating both the neptunium and plutonium from the fission products by means of a lanthanum fluoride cycle. The solution is next oxidized in H_2SO_4 with BrO_3^- for 1/2 hour which preferentially oxidizes the neptunium; afterwards, LaF_3 is precipitated, carrying away the bulk of the plutonium. The supernatant, containing the neptunium and some plutonium, is then fumed to destroy the BrO_3^-, reduced with SO_2 water, fumed again, put in solution with H_2SO_4, and lanthanum fluoride product precipitation is made. This precipitate is dissolved in H_2SO_4, the solution oxidized again with BrO_3^- for 1/2 hour, and the cycle repeated until, calculating the beta activity back to standard time, a constant alpha/beta ratio is attained.

Hagemann treated 50 micrograms of plutonium metal for 15 minutes with Cl_2 gas at 450°C today and observed the production of a fluffy residue which is robin's egg blue in color. He will submit a sample of this, sealed in a capillary, to Zachariasen for x-ray analysis.

R. S. Dreyfuss, the technician who was hired last month for Donlan, is being inducted into the Army and terminated today.

Watters wrote to Leo Brewer in Berkeley and offered to analyze carbon samples for him in response to Brewer's recent request to Orlemann.

Kohman reviewed for Philip Morrison in the Nuclear Physics Division several papers on element 93 published in German periodicals last year and the year before. He concludes that nothing in the publications is inconsistent with the view that the Germans are aware of the importance of the various heavy isotopes and that they are engaged in intensive work on them. On the other hand, he points out there is no direct evidence for this view either. Kohman further guesses, on the basis of the intensity of the neutron sources implied in the articles, that it is unlikely that the Germans have isolated and identified plutonium.

The "University of California Progress Report for January 1944" (MUC-NH-617) by Latimer and Eastman has been issued. The summary of this report is as follows:

"Investigations to assist the semiworks at X continue. A satisfactory $Th(C_2O_4)_2$ separation step has been devised and coupled with the acetate cycle. The decontamination efficiency of the acetate cycle has been improved by use of a Cb_2O_5 scavenger. Contributions have been made to the fundamental chemistry of plutonium. Work continues on analytical methods and purification by vacuum fusion in sulfide refractories. Preliminary work on product metal production progresses using plutonium tracers and uranium and thorium as stand-in metals.

"The estimated expense for January 1944 is $15,325.00 as compared to $12,900.00 for December 1943. The personnel remained at 34."

I received a suggestion from George Watt that we should adopt the system of inorganic nomenclature of the International Union of Pure and Applied Chemistry for use in our designation of plutonium compounds. He offers the following for purposes of illustration.

Assumed Formula	Name
PuF_3	Plutonium(III) fluoride
$PuBr_4$	Plutonium(IV) bromide
$PuCl_6$	Plutonium(VI) chloride
PuO_2	Plutonium(IV) oxide
K_2PuF_6	Potassium plutonium(IV) fluoride
La_2PuF_{10}	Lanthanum plutonium(IV) fluoride
$Pu(NO_3)_4 \cdot 6H_2O$	Plutonium(IV) nitrate 6-hydrate
$PuO_2(NO_3)_2 \cdot 4H_2O$	Plutonium(VI) dioxynitrate 4-hydrate
$PuO(NO_3)_2$	Plutonium(IV) oxynitrate
$NaPuO_2(C_2H_3O_2)_3$	Sodium plutonium(VI) dioxyacetate

He also points out that the recommended procedure for designation of mass numbers of isotopes involves writing the mass number as superscript to the left of the symbol, thus $^{235}_{92}U$ rather than $_{92}U^{235}$.

There was the evening meeting of the Research Assistants of Section C-I at 6:45 in Room 209, Eckhart Hall.

The 7:45 p.m. meeting of the Council of my section was attended by Albaugh, Baumbach, Davidson, Dawson, Dreher, Ghiorso, Manning, Orlemann, Thompson, Watt, Willard, and me. I emphasized the necessity for testing the Bismuth Phosphate Process at Hanford concentrations with larger quantities than have heretofore been available. In this connection, I indicated that we shall be getting 350 milligrams of plutonium about February 15 and more about March 1 if the Site X plant yield is above 50%. A one-gram sample is expected to arrive here about March 15. A space survey in the New Chemistry Building shows that we have only from 50 to 80% as much space per man as other sections in the Division and that the most crowded conditions exist for those groups where health hazards are greatest. We can expect a reallocation of space in the near future. I announced that two types of Project monthly meetings are now being held, one concerned with the development of the Hanford Process, and the other dealing with problems at Site Y.

Thompson described recent work on extraction and decontamination which involves an experiment on a 10-ml sample employing fission products concentrations at twice that expected at Hanford. The extraction cycle gives 98% yield, but the decontamination cycle gives only 50% carrying. I stressed the need for further confirmatory experiments before conclusions are drawn.

Watt reported on concentration and isolation studies by Yett and Pye using Site W concentrations involving 7.5-ml volumes; this scale of work is now possible because of our supply of plutonium from Site X. It was found necessary to use 2 N HNO_3 to dissolve the metathesized LaF_3 precipitate. There is evidence that the peroxide precipitation from 1 N acid is not complete in a one to three hour period. Davidson reported experiments by Hagemann in preparing uranium chloride, which volatilized at 450°C. Similar treatment of plutonium metal with chlorine resulted in a blue substance that is being submitted to Zachariasen for x-ray analysis. Baumbach stated that the problem of plutonium metal production is now fairly well solved. It is possible to produce usable metal in yields of 50% or more when one starts with the white fluoride. He stated that the density value of 16.4 ± 0.5 gm/cm^3, as reported by Zachariasen, is for a calcium-reduced sample assuming it has a face-centered cubic structure. Analysis shows the sample is 80% pure. Since this analysis, samples reduced by lithium or barium have been produced that give a different set of x-ray diffraction lines than the calcium-reduced samples. A density value of 16.2 gm/cm^3 has been obtained, using the capillary method, on a barium-reduced sample shown to be 96% pure. Baumbach stated it is possible that pure plutonium metal does not crystallize in the face-centered cubic form as does the oxide. Zachariasen is still anxious to get the plutonium metal in the form of a powder, but this is difficult to accomplish. Orlemann stated that purification experiments are continuing using ether or nitromethane as extraction agents.

Helen and Wilma had lunch together in our apartment.

The Soviets are closing in on Krivoi Rog in a new advance. They have also taken 30 hamlets in a drive on Luga.

Friday, February 11, 1944

Today Henry Hoekstra is beginning a very important experiment. He will test the Bismuth Phosphate Process on the 100 ml scale starting with a UNH solution containing W concentrations of plutonium and twice W concentrations of fission products. He will go through the extraction step, two complete decontamination (oxidation-reduction) cycles, and a lanthanum fluoride concentration cycle. John Malm will help him by counting the various fractions to determine the yield.

I recorded an idea in my Notebook No. 139 on the use of solvent extraction methods as concentration methods for operation in connection with either the Clinton or Hanford plants, suggesting that after the lanthanum fluoride concentration cycle, the plutonium-containing solution (also containing La^{+3}) could be contacted with a solvent immiscible with water, in which Pu^{239} is soluble and hence preferentially extractible. After thus being freed of the carrier material, the essentially pure Pu^{239} could be obtained from the solvent by evaporation or re-extraction into a water phase after, e.g., utilizing a change in oxidation state or degree of complexing. I also suggested it might be even more effective to start with an HCl or HNO_3 solution of the bismuth phosphate after the decontamination cycle.

I had a telephone conversation with Hamilton in Berkeley to discuss two planned bombardments at the 60-inch cyclotron. One is the possible bombardment of large quantities of uranium metal (order of 50 pounds) with the fast neutrons from the bombardment of beryllium or lithium with deuterons; the problem is whether this is a better source of Np^{237} from the standpoint of concentration of Np^{237} and associated fission products than neutron-irradiated uranium from the Clinton pile. The other planned bombardment is plutonium plus deuterons or helium ions to look for isotopes of elements 95 or 96. We discussed the possible use of a plutonium-gold alloy for this purpose. Joe will send me four grooved copper plates to use as backing for the plutonium targets.

Today I wrote a memo to Hogness regarding the salary and rank of Manning. To quote: "I am writing in answer to Mr. Stearns' request of February 8, 1944, for specific information on the nature of the work which Mr. Manning has been doing in the past and the nature of the increased responsibilities which are his now and will continue to be in the future. Up until a few months ago Mr. Manning was a research chemist in the Metal Production Group with no additional responsibility other than that of his own work. At that time I asked him to act in the capacity of an Assistant Section Chief for my section, and he then began to help in the supervision and planning of work for the whole section. Recently he was again asked to assume further responsibility, and he is now acting in the capacity of

Associate Section Chief for our section. In this capacity his responsibility is large; in addition to his duties to help in the administration of the section he is giving particular attention to the running of the purification part of our section's program. Thus, since our section is rather large, he is intimately concerned with the direction of the research efforts of some thirty people who are working on this phase of our program. I believe that on this basis he should certainly qualify for the title of Senior Scientist.

"As you know, Mr. Manning came to us with about a dozen years' experience since his Ph.D. degree, while his salary was calculated on the basis of four years' additional experience on the assumption that his salary be reconsidered as soon as his worth had been established. The unique rapidity with which he has risen to broad responsibility makes it apparent that he has turned out to be a man of ability even beyond our expectation, high as that was."

In a memo Dreher outlined for me a procedure for handling waste disposal from the Chicago semiworks. This is a problem of growing concern.

Jaffey and Turkevich prepared a memo comparing the production of U^{237} at the St. Louis Washington University cyclotron with U^{237} produced in the chain-reacting pile at Site X. Their calculations relate to 100 pounds of uranium metal bombarded with neutrons for 30 days. For the cyclotron operating at an average current of 200 microamperes, the yield of U^{237} would be 66 or 109 micrograms, depending on whether a lithium or a beryllium target is used; for the pile operating at an average power of 1,000 kw, the yield of U^{237} would be 733 micrograms.

Our curator for the Chemical Division, Howard W. Lange, who is the general building supervisor and who procures supplies and equipment, received orders from his draft board to report for a pre-induction physical examination next Tuesday. It is indicative of the cloud that hangs over many of the workers here. Lange is 37 years old, is married, and has been with us for two years. His technical training includes one year at Northwestern University and two years at Lewis Institute, and he holds assistant pharmacist's papers. But none of this seems to bear much weight with the draft boards. Today, Selwood wrote John McKinley in the Area Engineer's office saying that the loss of Lange will cripple the activities of the Chemical Division even more seriously than would the loss of one of the chemists.

I received a memo from J. B. Tepe of the Technical Division describing a proposed process and experimental program to separate plutonium from fission products and uranium by continuous ether extraction in a counter-current extraction column. Semiworks equipment is nearly ready for operation at this time to study the first two steps, i.e., extraction into the solvent phase, followed by re-extraction into an aqueous phase. Related chemical experiments are to be carried out jointly by Margolis of Dreher's group and R. T. Makins of Kircher's group in the Technical Division.

"Chemical Research--Final Processing of Plutonium. Report for Month Ending February 1, 1944" (CK-1169) has been issued. Here I point out that purification procedures involving Pu^{+3} may be of great value in removing

light element impurities from plutonium. The ready interconvertibility of Pu^{+3} and Pu^{+4} may make it feasible to employ both oxidation states in a complete purification scheme. It is possible that Pu^{+3} compounds will be desirable starting materials for metal production. Activities of the group involved in the final processing are described as follows: Brody, Dixon, Smith, and Orlemann performed an ether extraction of oxidized plutonium to purify 15.8 mg of rather impure material in as short a time as possible. The product of two extractions was re-extracted from the ether into 1 M HCl. Spectrochemical analysis showed total measured impurities of less than 0.667%, with particularly good separation from thorium, lanthanum, platinum, bismuth, calcium, chromium, and sodium. Fried reduced 100 micrograms of white plutonium fluoride (obtained by treatment of PuO_2 with very pure HF) in ThO_2 at 1,600°C using calcium as the reducing agent and obtained a high yield of plutonium metal in one bright piece. A similar reduction of 350 micrograms produced a single piece of 200 micrograms and a smaller one of 25 micrograms. The smaller piece was flattened by Jasaitis and both pieces given to Zachariasen for x-ray analysis. The 200 microgram piece gives lines only of ThO_2. The 25 microgram sample gives x-ray patterns probably characteristic of plutonium metal, but the lines are quite diffuse. From the x-ray pattern Zachariasen calculates a density of 16.4 ± 0.5 gm/cm^3. When the 200 microgram sample was flattened, it yielded an x-ray pattern identical with the smaller sample. Fried reacted these metal pieces with hydrogen to form the hydride. X-ray diffraction photographs of these samples show lines only for ThO_2 and PuO_2. Fried and Baumbach used about 800 micrograms of the black fluoride (obtained by treatment with HF of doubtful purity) in seven attempts to produce plutonium metal by high temperature reduction (1,600°C) with barium without success. A lower valent form of plutonium is evidently produced that is quite metallic in appearance and does not form a hydride--it may be Pu_2O_3. Fried and Jasaitis attempted to fuse about 5 micrograms of black plutonium fluoride by heating to 1,120°C in a vacuum. Fusion did not occur, but some of the material thought to be Pu_2O_3 was produced and a white sublimate formed. J. Karle treated 100 micrograms of the black plutonium fluoride with atomic hydrogen which, as shown by x-ray analysis, resulted in the production of PuO_2 and lower valent forms of plutonium, possibly Pu_2O_3, PuF_2, and another undeterminable substance. Zvolner electrolyzed quantities of plutonium up to 65 micrograms in aqueous solutions, which resulted in the deposition of plutonium as $Pu(OH)_4$ at the cathode.

Helen was home all day. A big snowstorm has been raging since yesterday, and 8.4 inches of snow has fallen. The temperature did not go above 15°F today.

Saturday, February 12, 1944

Watt has been conducting a series of tests to ascertain the effect of Fe^{+3} in inhibiting the precipitation of plutonium peroxide under conditions suitable for use in the isolation procedure at Hanford. His experiments today indicate that the influence of 0.05 M Fe^{+3} does not appear to be so great as suggested by earlier measurements.

Zachariasen reports that the sample Hagemann submitted to him the day before yesterday for x-ray analysis has the same structure as UCl_3 and therefore must be $PuCl_3$.

Allison sent a memo to Mulliken, saying that he is notifying General Groves that Mulliken is now in charge of information transmission between our Laboratory and the Montreal Laboratory. He enumerated five questions that came up when the representatives from Montreal were here last week. Two of them concern instrumentation and technique. The other three are as follows: "(3) They were much interested in the work of Dr. (Howard R.) Kratz on heat transfer through uranium, and would like data on this matter. (4) They told me that Seaborg has prepared as part of his monthly report a special chemical report of material which Seaborg considers can go to Montreal. Could you examine what Seaborg has written in the light of the instructions to which the General has agreed, and decide whether it can be transmitted? (5) The delicate question as to whether representatives of the Montreal group can look over our entire list of titles and select ones which they would like to request keeps coming up. As it is now, they have to depend on us to select things which would be of interest to them. Would you consider this problem and see what can be done?"

Katzin sent Moulton patent case S-781 by Seaborg, Thompson, and Davidson on the use of scavengers. He relayed, in the accompanying memo, my feeling that it may possibly be desirable to limit this case to the Bismuth Phosphate Process, inasmuch as similar work may be traced to the early researches of Wahl, Kennedy, and me, e.g., Report A-135, "Chemical Properties of Elements 94 and 93," March 19, 1942.

Helen went downtown with Dagmar Dreher and her son, Jerry. Later she and I had dinner out and then went to a movie.

Sunday, February 13, 1944

The Ghiorso baby, a girl, was born at 2:00 a.m. in the University of Chicago Lying-In Hospital. She was promptly named Kristine.

The big headline in today's Sun concerns the war in Italy and reads, "Allies Push Nazis Back; Fall of Rome Promised."

Helen and I visited the Museum of Science and Industry in the afternoon and then went out for dinner.

Monday, February 14, 1944

Ray Frank made hardness measurements on two samples of plutonium metal--one prepared by barium reduction which weighed 78 micrograms and the other, prepared by calcium reduction, which weighed 43 micrograms. The Vickers hardness values were 124 for the barium-reduced sample and 128 for the calcium-reduced sample.

2/14/44 (cont.)

Spof English from Clinton Laboratories and Ermon Eastman from Berkeley are visiting the Met Lab today. They will both be here through Thursday to attend meetings.

Professor Eastman showed Cunningham and me a draft copy of a Berkeley report describing the observation by Bob Connick, Bill McVey, and Glenn Sheline of the +3 oxidation state of plutonium, which apparently occurred before we observed this oxidation state here. Berkeley chemists used plutonium isolated in December from their recent large cyclotron bombardment for their work.

Gerald W. Sears, who received his Ph.D. from the University of Illinois two years ago and has been an instructor there, started working with Phipps in Baumbach's group. With a rating of Chemist his starting salary is $4200 per year.

Latimer sent me the names of two of his recent B.S. graduates, Arthur E. Stickland and Jack B. Chrisney, who would be willing to join me here at the Met Lab. He also mentioned he has suggested that Sigfred Peterson, who was graduated with highest honors from California in 1940 and is now a graduate student at the University of Minnesota, apply to me. He said of Peterson, "He is very bright, but of course his Swedish personality might be hard on you." He added a note that his group received 20 milligrams of plutonium from Clinton Labs on Friday, the day he wrote the letter, and that the glass seal is broken, "which introduces some difficulties in recovery from the Kleenex packing."

I wrote Gofman answering in detail the many questions he and Sheline raised in their February 1 letter about positions at Site W. Concerning their query as to when they might start work at Hanford, I suggested to Gofman that if he were one of Willard's three to five supervisors directing research efforts, the starting time would be in July. I also said that most of my answers apply to Sheline as well as to him.

Only six others in addition to myself attended the monthly meeting on the final purification and metallurgy of plutonium this morning: Thomas, Hogness, Selwood, Eastman, Warner, and Potratz. Because so few of us were present we decided to postpone the main meeting until next Thursday, when the other participants will be here. We had some discussion, however, on the Berkeley work, and it was decided that the Berkeley group is responsible to Chicago for the kind of investigations they are doing. Eastman pointed out that the Berkeley program (electrolysis, reduction of plutonium from plutonium halide, uranium distribution, etc.) was not covered in the minutes of the last meeting, January 13. We agreed that Berkeley, nevertheless, is to go ahead as planned. Allocation of plutonium to Hanford investigations was discussed.

Today I insisted that a meeting be held, with Nickson, Hogness, Kidd, Katz, Captain Ware, and others to discuss special dangers involved in handling plutonium. I took the position that something concrete about the health hazards must be done immediately. I have become impatient with the lack of progress. Nickson summarized the recommendation submitted to the Administration that includes more space, adequate ventilation (hoods, etc.),

and installation of linoleum on floors. He discussed the health hazards indicating that the safe tolerance for plutonium in the bone, based on radium toxicity, is about two micrograms; he did not think one would be any better off to have plutonium in the lungs rather than in the bone. In my opinion, I said, Pu(III) and Pu(IV) should not much resemble uranium in physiological behavior but should more resemble zirconium. The dangers are principally caused by plutonium-containing dusts in the air. I asked about the feasibility of a precipitron installation, but Captain Ware thought it would not be a good idea to install a precipitron in a particular room because of the need to clean it. Nickson declared that each bench and vacuum line should be enclosed in a hood. Hogness would like to see this coupled with a general air conditioning system to insure the absence of dust in the building. Unfortunately, the more extensive the building alteration program, the more it will interfere with the work and the longer it will take. Captain Ware would make no definite commitments how to resolve the matter except that he agrees to get together with Kidd and make some decisions.

Hamilton wrote from Berkeley about our telephone conversation last Friday. Concerning the fast neutron production of Np^{237}, he asked if there were figures on efficiencies of the various processes (lithium, beryllium, and pile neutrons) when all the thermal neutrons are kept out by a cadium absorber. He plans to go ahead and set up 50 pounds of uranium metal for neutron bombardment at the 60-inch cyclotron. He said he has spoken to Latimer about his, Hamilton's, idea of alloying the plutonium with gold for the target for a deuteron or helium ion bombardment in the 60-inch cyclotron. Latimer considers the idea reasonable and suggests a BeO crucible be used for isolation of the plutonium metal and production of the subsequent gold alloy. Hamilton also mentioned the possibility of plating or depositing the plutonium on the gold by sputtering and said he is sending me the four copper target plates for my possible use.

"Chemical Research--Chemistry of Plutonium. Report for Month Ending February 1, 1944" (CN-1324) is being issued today. It is devoted largely to descriptions of the recent experiments on the +3 oxidation state of plutonium. Hindman, Kraus, Howland, and Cunningham state that a solution of 8.5 mg of Pu^{+6} in HCl (prepared by Orlemann and Brody) was treated with gaseous SO_2 with the intention of reducing it to the +4 state in preparation for further work. The solution changed from light pinkish-yellow to a purple violet; the typical green color of the +4 state was not observed. A series of tests were conducted to determine whether the color is due to reduction to the +5 state, to formation of a complex chloride of the +4 state, or to reduction of a state lower than +4. One of these tests was to add NH_4OH and resulted in a pale blue-colored precipitate. It was noted that the upper surface of this blue hydroxide precipitate slowly changes to green on exposure to air. This was considered strong evidence that the plutonium in the blue precipitate is present in a valence state lower than +4. After dissolving the hydroxide precipitate in HNO_3, the purple-violet color returned. Reprecipitation of the plutonium as the peroxide led to the typical green color of the +4 state and then dissolving in HNO_3 led to the typical green color of the +4 state. They performed a series of micro-scale experiments that gave further evidence that the purple-violet color

obtained by the addition of reducing agents to the +6 and +4 states is caused by reduction to a state lower than four, presumably the +3 state. Smith and Patton prepared and measured the solubility of Pu^{+3} fluoride, phosphate, oxalate, and hydroxide. Hindman, Howland, Kraus, and Cunningham determined the absorption spectra of +6, +4, and +3 plutonium in aqueous solution using a Coleman Universal Spectrophotometer for the +6 and +3 spectra and a Beckman Quartz Spectrophotometer with a much higher dispersion order for the +4 spectra. Kraus, Howland, and Hindman carried out preliminary experiments with oxidizing and reducing agents of known potential in order to find the potential for the Pu^{+4}/Pu^{+3} couple and concluded the upper limit in acid solution is -0.45 volts. Watters and Orlemann conducted an initial experiment to determine the potential of the Pu^{+4}/Pu^{+3} couple by the polarographic method and find definite indications that it is -0.65 volts or more negative. Hindman and Howland find evidence that the oxidation number for the lower oxidation state is +3 by carrying out a spectrophotometric titration of Pu^{+4} using Sn^{+2} as the reductant in nitrate solution; the experiments are being repeated in HCl solutions. Patton obtains further evidence for the +3 state by determining the mole ratio of oxalate to plutonium in an oxalate precipitate of the lower valence compound. Brody, Dixon, Orlemann, and Smith determined the distribution of Pu^{+6} between 10 M NH_4NO_3 and ethyl ether and find it to be 0.54. Brody, Dixon, Smith, Hindman, and Orlemann find that Pu^{+3}, Pu^{+4}, and Pu^{+6} in solution do not fluoresce in a mercury vapor source of ultraviolet light. Kraus finds that if plutonium hydroxide is aged seven days, it still dissolves readily. This experiment was conducted to test the suggestion that the difficulties sometimes encountered in dissolving precipitates containing plutonium hydroxide may be due to aging. Smith carried out an experiment to estimate the vapor pressure of plutonium bromide through sublimation of a mixed uranium-plutonium bromide and finds that at 450°C the vapor pressure of plutonium (IV?) bromide is roughly 7×10^{-2} mm Hg.

"Theoretical Calculations Concerning Backscattering of Alpha Particles" (CC-1342) by Crawford was also issued today as Part II of Report CN-1324. These calculations are based on experiments performed last July by Cunningham, Ghiorso, and Jaffey (Reported in "Chemical Research. Special Chemistry of Products," CK-888), which up to now have been interpreted by assuming approximately a 52% counting yield for samples mounted on platinum or an error in the accepted value for the specific alpha activity of normal uranium as measured by Kovarik and Adams. The question arises, is it at all possible that the extra 2% (a 4% effect) in counting yield obtained last July is caused by backscattering? Crawford made calculations, based on Geiger's multiple scattering data of 1910-11, that indicate that alpha particles with a range of 3.6 cm in air are backscattered from a platinum sample mounting to the extent of 3% to 3.9%, the variation depending upon the residual range of the scattered particles. The relative backscattering coefficients for counting plates of different substances are also calculated. Taking gold as unity, the backscattering ranges from 0.99% for platinum to 0.23% for aluminum and 0.19% for quartz. It would therefore seem that when we make alpha counts of plutonium these backscattering coefficients must be considered.

The evening seminar of the Chemistry Division was held at 7:45 p.m. in Room 251, Ryerson Laboratory.

Al Ghiorso had dinner, then spent the late evening with us in our apartment after visiting Wilma at Lying-In Hospital.

The Anzio battle has been a seesaw struggle. The Soviets have won the east shore of Lake Peipus, which forms the frontier of Estonia.

Tuesday, February 15, 1944

Arthur Baer began working as a laboratory assistant with Donlan today.

As a further check on the possible production of U^{236} from neutron irradiation of uranium, James made alpha activity measurements on samples he has derived from 3 cc of nitrate solution of uranium from the Clinton pile that Spof English brought from Oak Ridge. He concludes there is no production of U^{236} observable.

Moulton has asked me for information on the conception of the idea for the use of O^{16} (i.e., O^{18}- and O^{17}-free oxygen) and C^{12} (i.e., C^{13}-free carbon) in connection with purification work. I replied that immediately after my letter of November 3, 1942, to Professor Oppenheimer (MUC-GTS-243) and Professor Oppenheimer's answer to me of November 6, 1942, (MUC-GTS-242, Series B) copies of which correspondence Moulton has, there was a great deal of discussion by many people on the possibilities of the use of separated isotopes. This idea followed obviously from a consideration and calculation of the actual light element impurity limits (such as was calculated by Teller in Memo No. 27, November 30, 1942, and by Perlman and me in the "Feasibility Report" of November 26, 1942, and in CN & CF-380, report for month ending December 15, 1942), since these calculations were based on the knowledge that O^{16} and C^{12} do not undergo the α,n reaction with 94^{239} alpha particles. I stated that I have looked for some specific written matter on the suggested use of separated O^{16} and C^{12} which might have come out at about this time, but have not found any; however, my search has been rather cursory, and probably does not indicate the absence of such written information on the Project.

Following a discussion with members of Davidson's group, Orlemann, and me, Manning recommended to Captain Ware that for health protection against plutonium the present hoods in Room 2 should be provided with much stronger blowers with separate outlets for each of the two pairs of hoods. He said the largest and most hazardous of the vacuum setups in Room 2 will be moved to another smaller room. Our work on the dry chemistry of plutonium, the preparation of fluorides, chlorides, etc., takes place in Room 2. This is the most dangerous of our chemical operations.

At 9:30 a.m. I attended the Project Council Information Meeting on Chemistry in Room 209, Eckhart Hall. Others present were Allen, Allison, Boyd, Burton, Chipman, Compton, C. M. Cooper, Doan, Eastman, English, Fermi, Garrison, Greager, Hilberry, Hiskey, Hogness, Jeffries, W. C. Johnson, Kennedy, Kircher, Levy, Mulliken, Potratz, Shapiro, Cyril Smith, Smyth, Spedding, Stone, Sugarman, Sutton, Vernon, Warner, W. W. Watson, Whitaker, Wigner, Zachariasen, and later Stearns. Allison opened the meeting and

asked Hogness to introduce each of the Section Chiefs for the Chicago Chemistry Division. I was allotted 20 minutes and was the first to speak. I summed up the work of Section C-I, the highlights being:

(1) Separation process - Hanford Concentrations. Extraction and decontamination - demonstrated on 1/2 cc scale; but consequences of existence of Pu^{+3} need to be studied further. Concentration step - now also demonstrated at Hanford concentration; isolation step - not yet demonstrated.

(2) Concentration - HCl methods (Semiworks). Worked out in laboratory previously, now in semiworks. Regular bismuth phosphate from Site X-type cycle, plus inactive bismuth phosphate to make 5 lb. per run, was dissolved in HCl and run through uranous oxalate precipitation in rubber-lined equipment, with average plutonium yield 92% and gamma activity decontamination factor of 4.5 for the step.

(3) Metal Production. We have now learned how to make PuF_4 and to reduce it both by calcium and barium. Zachariasen finds the plutonium metal reduced by calcium is cubic face-centered with density 16.4 ± 0.5 gm/cm^3 giving diffuse lines in the x-ray pattern. Plutonium metal produced by barium reduction gives a complicated pattern and a density of 16.2 gm/cm^3. Respective hardnesses resemble those of copper and iron. The melting point of plutonium metal is somewhat lower than uranium metal. Calcium, as well as barium-reduced metal, forms a plutonium hydride.

(4) Purification. Ether extraction works well with plutonyl compounds; solubility only slightly below that of uranyl compounds; $PuCl_3$ and $PuBr_3$ have been made.

(5) Basic chemistry. While processing plutonium produced at Site X, our chemists observed the +3 oxidation state. Connick and co-workers, working with cyclotron-produced plutonium, observed the +3 state somewhat earlier. They find, in 1 M HCl, an oxidation-reduction potential against +4 of -0.97 volts; in H_2SO_4, -0.77; in $HClO_4$, -0.96. (Compare $Pu^{+4} \rightarrow PuO_2^{++}$, -1.05 volts.) Complete absorption spectra of Pu^{+3}, Pu^{+4}, and PuO_2^{++} are now determined, which are valuable for determination of concentrations of ions. The change from +3 to +4 has also been observed with the polarograph.

(6) Heavy isotopes. $94^{239} + n \rightarrow 94^{240} + \gamma$ was looked for at Clinton. The sample was irradiated with neutrons a month at the center of the Clinton pile, but no change in alpha activity was found. One concludes from this result that the upper limit of possible increase of alpha particle activity due to 94^{240} formation at Hanford would be by a factor of 3. Extent of possible occurrence of $93^{239} + n \rightarrow 93^{240} \rightarrow 94^{240}$ will be determined in the pile at Site X.

In answer to Allison's question about the melting point work, I replied that in comparing UF_4 and PuF_4 reduction with calcium, the plutonium was found to form melted pieces 200° lower than did the uranium. Zachariasen reported finding a density of 15.9 gm/cm^3 in a plutonium metal sample that contains only 0.1% calcium, according to M. Fred. Burton (Section C-II), Sugarman (Section C-III), and Potratz (Section C-IV) followed me with their reports. Next W. C. Johnson introduced the Clinton Laboratories Chemistry Division speakers. English reported that, in connection with the isolation of plutonium at Clinton Labs, a number of studies on the solubility of plutonium compounds have been undertaken, viz, $Pu(IO_3)_4$ has a solubility of 5 mg/liter in 3 N HNO_3, the oxalate has a solubility of about 15 mg/liter in 5 N HCl and the peroxide is soluble to the extent of 25 mg/liter in a solution of pH 6.5.

Henri A. Levy gave a report on the new "hot" laboratory at Clinton. The hot laboratory, he said, is designed to permit carrying out most of the ordinary chemical operations at high levels of radioactivity. Construction is scheduled to be completed by March 1st, and work is ahead of schedule. Among other uses, the hot laboratory will be used to prepare solutions which have the activity of "W" solutions to make further studies on fission chains, and to prepare strong gamma sources. Strong gamma emitters are desired for studying (γ,n) reactions. All of these intended uses require the construction of an unusual building. Levy sketched the construction of the shielded cells in the laboratory at the blackboard and discussed the methods that have been developed for obtaining visibility within. A periscope equipped with a scanning mirror was displayed. This periscope can be placed at any of 42 observation points in the cells. With its use it is possible to obtain a good view of any small portion of the interior of a cell. For quick observation of a large part of a cell, a large scanning device has been developed which will be placed at the top of a cell. This device was also on display. In addition to the heavily shielded cells, other cells shielded by 6 inches to 1 foot of concrete are available for work at lower levels of activity.

At the conclusion of a report by Edward Shapiro on the production of hydrogen peroxide in water exposed in the Clinton pile, Allison called on Greager to introduce Clinton Separations Development speakers. These were Sutton, who talked about improvements in decontamination and concentration, and Greagor, who presented a report on yields obtained in the Bismuth Phosphate Process in the Clinton semiworks. Whitaker commented that yields from the last seven runs in the plant through Room D averaged 72%. Yields should be further improved by the installation of slinger rings to wash down the sides of the centrifuge with acid.

The meeting ended with reports by Eastman of Berkeley, Kennedy of Los Alamos, and Spedding of Ames. Eastman reported on Gofman's thorium oxalate extraction process, Kennedy gave a brief account of analytical methods for oxygen and fluorine, and Spedding talked about uranium trifluoride, uranium tetrabromide, and a new method of firing refractory oxides.

Today a melting point determination of plutonium metal was attempted for the first time by Jasaitis. A platinum foil was coiled into a tube, and a small piece of plutonium metal (made by barium reduction) was placed on the coil and heated. At 860°C an alloy with platinum was formed. On repetition the same observation was made.

I received a memo from Willard about the amounts of plutonium needed for necessary investigation of the Bismuth Phosphate Process at Hanford concentrations during the next month. He estimates that over a gram of plutonium would be needed for (1) development work on problems in which we have no information as yet, and (2) systematic tests to determine safe limits of operation.

Dreher and Thompson attended a meeting to evaluate the present status of coatings for Hanford slugs. Others present were members of the Technical Division--F. G. Foote, L. M. Foster, J. P. Howe, and K. G. Jones of the Coating Section and G. W. Bird and C. E. Kircher of the Engineering

Development Section. The discussion centered around the problem of removing the coating involving the aluminum-silicon bond, which is receiving the most attention in the Technical and Chemical Divisions. Preliminary experiments indicate the only feasible methods for removal of this coating will involve either nitric acid or sodium hydroxide or both. It was decided to carry out a laboratory program to determine the optimum conditions for sufficient removal of the coating to permit complete dissolution of the slug in 60% nitric acid. Experiments also will be conducted to determine the effects on the process of the presence of the insoluble material in process solutions.

Section C-I issued "Progress Report, February 15, 1944. Work of Sub-section I - Extraction, Decontamination, Concentration, and Isolation Work" (MUC-GTS-457). The subjects covered are as follows: Group I - Albaugh - Extraction and Decontamination - Report on experimental work using the Bismuth Phosphate Process on a macroscale (1-10 cc) with Hanford concentrations of plutonium and inactive fission products. Results are generally satisfactory but indicate the carrying of plutonium with bismuth phosphate at the end of a decontamination cycle in the presence of Fe^{+3} is erratic and needs further study. Group II - Watt - Concentration and Isolation - Report on two tests of the Site W concentration procedure made on a 7.5-ml scale. Overall yield is 98.3%, but much more work must be done to establish optimum operating conditions. Experiments on a much larger scale (100 ml requiring 230 mg of plutonium per experiment) should be made as soon as possible. Two tests of the Site W isolation procedure were made on a 0.1-ml scale using 8.25 mg of plutonium. It was found that the high concentrations of lanthanum present has a pronounced inhibiting effect upon completeness of precipitation of plutonium peroxide, but the peroxide isolation procedure appears operable. Group III - Dreher - Process Development and Semiworks - Report on three semiworks runs (12, 13, 14). The use of U^{+4} as the reducing agent in the 40% uranium metal solution and in the decontamination cycle appears to give complete reduction of plutonium, but product yields are only 85, 75, and 59%; hold-up in equipment is suspected. The plutonium-containing bismuth phosphate precipitates from these runs were used in three semiworks tests of concentration by $U(C_2O_4)_2$ precipitation from HCl solutions. Analysis of the $U(C_2O_4)_2$ waste shows less than 3% loss of plutonium. Studies were made of two possible Hanford slug coatings - aluminum can with aluminum-silicon bonding and aluminum can with zinc bonding. Because of the formation of an insoluble scale, no satisfactory method for removal of the former has been found. The latter coating is dissolvable, but experiments indicate that the presence of zinc and aluminum will interfere with the bismuth phosphate separations process.

Helen visited Alice Thompson in the afternoon and then went to see Wilma in the hospital. The Baumbachs dropped in for the evening.

The Allies have occupied another island of Guinea--Rooke Island in Vitiaz Strait between New Guinea and New Britain.

Wednesday, February 16, 1944

General Groves is in Chicago.

I attended a meeting of Section Chiefs today at which Ann Hill, Manager of Office Services, Information Department, talked about signature authorizations.

Cunningham presented me with a list of problems to be investigated by Groups VII, VIII, and IX. They are: Group VII (Cunningham)--Organic derivatives of plutonium (Patton); Effect of complexing on oxidation-reduction potentials of plutonium, and Hydrolysis of Pu^{+3} and Pu^{+4} in aqueous solution (Kraus); Absorption spectra of plutonium and complexes (Hindman); Properties of plutonium peroxide (Howland); Solubilities of inorganic derivatives of plutonium (Smith); Chemical properties of neptunium (Magnusson); Isolation of neptunium (La Chapelle). Group VIII (Dawson): Recovery, isolation, and purification of plutonium (Asprey); Recovery, isolation, and purification of plutonium (Beard); Recovery of plutonium (Britain); Recovery of plutonium (Wetlaufer); Recovery, isolation, and purification of plutonium (Studier); Recovery, isolation, and purification of plutonium and supervision of recovery group (Dawson). Group IX (Ghiorso): Neutron counting with BF_3 pressure ionization chamber (Kohman and Weissbourd); Neutron counting with BF_3 proportional counter (Kohman and Schoenberg); Differential alpha range chamber (Jaffey and Crawford); Spherical pressure alpha range chamber (Jaffey); Nitrogen high resolving power alpha counter (Ghiorso and Weissbourd); Geiger counter maintenance (Walsh and Golden); Alpha counter maintenance (Schoenberg and Ghiorso); Plutonium finding alpha counter--ion chamber type (Schoenberg and Ghiorso); Plutonium-finding alpha counter--proportional counter type (Golden and Walsh); Extraction work (Jaffey); Project handbook (Kohman); Foreign literature survey in connection with Philip Morrison (Kohman); Final backscattering experiments (Ghiorso and Crawford); Testing of Simpson's high resolving power proportional counter under Hanford process conditions.

Thompson, Willard, Tepe, and Maloney held a conference about the concentration and purification of plutonium by ether extraction. It was agreed that experimental work could not be started for several weeks and would consist of batch experiments to determine the optimum concentration of NH_4NO_3 to have in the aqueous phase. Probably someone in Watt's group or Dawson's group will be assigned the task.

After having dinner with Helen at the 51st Street YMCA, I attended the triweekly meeting of the Basic Chemistry, Recovery, and Instruments Groups of my section at 7:45 p.m. in Room 209, Eckhart Hall. Those present were Baumbach, Cunningham, Davidson, Dawson, Dixon, Dreher, English, Ghiorso, Heller, Hindman, Howland, Jaffey, Katzin, Kirk, Kohman, Kraus, La Chapelle, Malm, Manning, Meyer, Patton, Pye, Rosenfels, Sheft, Clifford Smith, Watt, Watters, Willard, and others. I asked Cunningham to run the meeting. Howland discussed the recent measurements on the Pu^{+3}/Pu^{+4} couple, carried out by essentially a potentiometric titration. In 1 M H_2SO_4 the potential was found to be -0.739 volts; in the 1 M HCl the potential was found to be

-0.966 volts. In 1 M $HClO_4$ and 1 M HNO_3 the readings were found to drift, but the potentials were estimated to be near that in 1 M HCl. The value in H_2SO_4 may be due to the formation by Pu^{+4} of a complex ion with sulfate. Watters reported that polarographic investigations indicate that the potential of the Pu^{+3}/Pu^{+4} couple is more negative than -0.65 volts. In all his determinations he has failed to observe a second wave which indicates that if a valence state of plutonium less than three occurs, it has to be at a potential more negative than 1.2 volts which represents the limit of his experimental set up.

Hindman reported on new absorption spectra taken with a Beckman instrument that gives much better resolution of the various peaks for the three plutonium oxidation states than observed with the Coleman spectrophotometer. The spectra show some sharp peaks; since sharp absorption lines occur only in the rare earth series, this indicates that plutonium may be a member of a new "rare earth" series. A new determination of the oxidation number of Pu^{+3} was carried out by the use of the spectrophotometer in a titration of Pu^{+4} with Fe^{+2}. The results show a potential between -0.72 and -0.76 for the Pu^{+3}/Pu^{+4} couple in sulfate solution. Patton reported on a number of new organic derivatives of Pu^{+4} and Pu^{+3}. Dixon reported that solid plutonium acetylacetonate has been prepared recently and found to be light brown and crystalline. The crystals are found to sublime easily. The melting point was determined to be about 170°C after sublimation. Ghiorso reported that the range of the alpha particles from Np^{237} has been determined to be 3.25 ± 0.1 cm in air.

Helen had Peg Beaton and Alice Thompson over for lunch. Peg mentioned that the Baumbachs will move into the Beatons' apartment (on E. 78th St.) when she and Roy transfer to Oak Ridge.

The Allies have taken possession of the Green Islands, five islands off the northern tip of Bougainville in the Solomons.

The Project Council Policy Meeting was held at 9:30 a.m. in Room 209, Eckhart Hall, attended by Allison, Chipman, Compton, C. M. Cooper, Dempster, Doan, Eastman, Fermi, Greager, Hilberry, Hogness, Jeffries, Johnson, Miles, Mulliken, Smyth, Spedding, Stearns, Stone, Vernon, Warner, W. W. Watson, Whitaker, and Wigner. In his usual "State of the Nation" message Compton stated that planning must begin for the transition of our efforts to after-Hanford objectives, although such transitions cannot begin until after June 1. Smyth has been asked to work up longer range plans; all ideas concerning such planning should be sent to him. Smyth listed the following objectives for the next five to ten years:

1. Basic research
2. Production methods
3. Atomic power
4. Military weapons
5. Miscellaneous
6. Training of men

In answer to a question by Fermi, Smyth indicated that "Miscellaneous" includes development of the use of radioactive materials. Smyth expressed some ideas regarding the possible organizational approaches to conduct these programs. He visualized:

Basic Research

University and cooperative labs - no secrecy

Production Methods

Cooperative labs, Industrial Labs
Government Labs - some secrecy

Atomic Power

Government Labs, cooperative labs - some secrecy

Military Weapons

Government Labs - complete secrecy

Miscellaneous

University, Industrial Labs - no secrecy

A cooperative lab is thought of as an activity on a scale too large to be financed by universities, with building and equipment furnished by the Government. Research would be administered by cooperation of the educational institutions in the region of the laboratory. Smyth believes it essential for vitality that research people also be in contact with students. Smyth further pointed out that a supervisory control body for all study will be needed and suggested something similar to the National Advisory Committee for Aeronautics - entirely non-political. Hogness commented that he believes the Navy will have great interest in submarine power. Hilberry felt that the laboratory for weapon development would probably be an Army or Navy responsibility with complete secrecy. Allison spoke of the need to arrange to make radioactive tracers widely available. Smyth wondered when the P-9 pile will be ready, and Vernon replied that heavy water for the exponential pile should be here soon and heavy water for the self-sustained pile will arrive perhaps two months from now.

The next business on the Council agenda was to consider various Project reports on programs as related to the distribution of manpower. These included the following: Chicago. Chemistry Division (Presented by T. R. Hogness, Director)--Section C-I (G. T. Seaborg), Section C-II (M. Burton), Section C-III (N. Sugarman), Section C-IV (H. A. Potratz), and Section C-V (C. F. Hiskey). Technical Division (Presented by C. M. Cooper, Director)--General Engineering Section (G. E. Kidd), Engineering Development Section (H. W. Bellas), Metallurgy Section (A. B. Greninger), and Optics Section (G. S. Monk). Clinton Laboratories. Chemistry Division (Presented by W. Johnson, Director)--Section I (I. Perlman), Section II (C. D. Coryell), and Section III (G. E. Boyd). Separations Development Division (Presented by O. H. Greager (Director)--Section S-I (M. F. Acken) and Section S-II (J. B. Sutton).

California. Chemical Research (Presented by E. D. Eastman, Co-Director with W. M. Latimer)--Group leaders, R. E. Connick and J. W. Gofman.

Hogness mentioned that because of probable needs of Hanford, Thompson's group will be called on to supply at least 15 men and should be expanded in order to have a small group to carry on after the middle of the year; Burton's section will be reduced by about one-half while Sugarman's, Potratz's, and Hiskey's sections will continue at about the present size. Greager stated the Clinton semiworks is preparing to carry out a straight-through operation, as will be done at Hanford without re-use of equipment. Whitaker stated that the average yield of the Clinton separations plant for the last 8 runs is 72%. Slinger rings should still further increase the yield. He also mentioned that about 10 grams of plutonium-equivalent has already been pushed from the pile. The meeting adjourned at 12:45 p.m.

Thursday, February 17, 1944

Continuing his work on the melting point of plutonium metal, Jasaitis placed a sample of flattened plutonium metal (0.50 micrograms) and a sliver of gold (for reference) on a plate of fused thoria and heated them in a vacuum. The gold melted first, at 170°C above its melting point. The plutonium finally melted at about 1,300-1,350°C. There was evidence that the gold and the plutonium alloyed with each other.

James and I discussed the use of the Szilard-Chalmers mechanism for concentrating the Pu^{240} product of a neutron bombardment of Pu^{239}. Two possible substances for irradiation are (1) a metal organic salt of plutonium such as the acetylacetonate in an organic solvent such as chloroform and (2) a water solution of ammonium plutonyl acetate.

Clinton Laboratories shipped 0.965 grams of pile-produced plutonium last Tuesday, which rounds out the total of their deliveries to date (starting December 30, 1943) to 1.5 grams. Tuesday's shipments were divided into the following portions: 600 mg to Los Alamos, 350 mg to Chicago (which we received today), of which 100 mg is to be allocated to Los Alamos studies and 250 mg to Hanford studies, and 15 mg to Spedding at Ames for metal alloying experiments.

I received a letter from Sigfred Peterson, the graduate student in physical chemistry at the University of Minnesota who was recommended to me by Latimer. Peterson is concerned about his draft status and is interested in employment with us. Willard transmitted a copy of Peterson's letter to Wayne Johnson in Personnel, saying that we know him as an exceptionally able man and urging him to take every possible step to obtain Peterson's services for us as soon as possible; he suggested telephoning Peterson.

At 9:45 I attended the morning session of the February meeting under the chairmanship of C. A. Thomas on final purification and metallurgy of plutonium, held in Room 251, Ryerson Laboratory. Others present were Chipman, Eastman, Foster, Hogness, Kennedy, Selwood, Cyril Smith, Spedding, Thomas, Warner, and later A. V. Peterson and G. Derge. In his opening remarks Thomas emphasized that, as begun with the meeting last month, the attendance will be limited to those directing work in purification and metallurgy at the Met Lab, Site Y, Ames, and Berkeley; he said that at the suggestion of A. V. Peterson and A. H. Compton the work at Ames is now under his direction. After the opening remarks by Thomas, I spoke until 11:45 on the highest priority problems in the Chemical Division, covering the following:

A. Studies with metal. Preparation of metal from plutonium fluoride. I presented data on the differences between the "white" or "light brown" and the "black" or "purple" fluoride and said that the most reasonable interpretation for the difference in color and phase is that PuF_4 tends to undergo the reaction $PuF_4 = PuF_3 + 1/2\ F_2$ at 550°C and a slight variation in the nature of the HF (slight oxidizing or reducing properties) determines whether the end product will be PuF_4 (light brown) or PuF_3 (purple). I discussed the reduction of PuF_4 with barium, lithium, and calcium and mentioned that, in general, the calcium-reduced samples seem less pure than barium-reduced samples. I discussed the x-ray evidence that the structure of plutonium metal produced by barium or lithium reduction is more complex than that produced by calcium reduction which has a face-centered cubic structure. I reviewed the density determinations made so far and cited recent experiments that indicate the melting point to be between 850°C and 1,300°C and mentioned that an apparatus for direct measurement of the melting point has been constructed and will be used as soon as sufficient product is available. Calcium-reduced metal is about equal in hardness to annealed copper, while barium-reduced metal is about equal to soft iron. I reviewed the preparation and determination of the properties of plutonium hydride and mentioned that none of the preparations has yielded interpretable x-ray photographs. I said that to date no information has been obtained on the formula, density, or equilibrium dissociation pressure of the hydride.

B. Preparation of a pure compound of plutonium. I mentioned that precipitation methods of purification have been studied using 2 mg of plutonium to carry out peroxide and molybdate precipitations. I reviewed solvent extraction and volatility methods of purification and, in connection with the latter, observed that a plutonium chloride was made in a capillary tube by the reaction of Cl_2 with 0.05 mg of metal at 450°C. There was no evidence of volatilization. The product was blue, and x-ray examination by Zachariasen indicates structure similar to that of UCl_3. By chemical analysis the formula is $PuCl_{3.0 \pm 0.3}$. I outlined a complete purification procedure as a basis for discussion. The procedure involves precipitation of Pu(IV) molybdate, dissolution in HNO_3, precipitation of Pu(IV) oxalate, dissolution in HNO_3, oxidation with dichromate, extraction of Pu(VI) with ether, transfer to aqueous solution, reduction to the +3 and +4 oxidation state, followed by either wet or dry conversion to the chloride, reduction to metal with barium, and further purification possibly by the iodide-hot wire method. In conclusion I

reviewed the work by Potratz of Section C-IV on analytical methods to determine the purity of plutonium. Chipman took the floor after I sat down and described plans for the central refractories laboratory. He introduced Gerhard Derge as Magel's successor in the Metal Production Group; Magel has been transferred to Site Y.

I also went to the afternoon session of the meeting, which commenced at 1:15 and was attended by Chipman, G. Derge, Eastman, Foster, Hogness, Kennedy, Potratz, Selwood, Cyril Smith, Spedding, Thomas, and Warner. In addition Greenewalt was present during a discussion concerned with the form in which plutonium should be stored and shipped from Site W to Site Y. The tentative decision was to ship a concentrated solution, probably nitrate, in stainless steel containers. In outlining in detail the metallurgical program at Site Y, Smith described the plans for the first two grams of plutonium to be received for metallurgical studies. Two one-gram lots of metal will be prepared by calcium reduction; experiments will be conducted to give information on melting point, density, crystal structure, expansion coefficient, specific heat, hardness, elastic modulus, yield strength, work hardening coefficient, microstructure, softening temperature, recrystallization temperature, hydride composition and properties, oxidation rate, corrosion rate, electrical conductivity, and perhaps magnetic susceptibility. Smith was followed by Kennedy who spoke on a method of analysis for fluorine, Spedding who gave a progress report on the program of the new Ames Project, and Eastman who reported on the work at Berkeley on sulfide refractories and the UBr_3-U system; refractories of lower sulfides such as $CeS_{1.4}$ and $ThS_{1.9}$ have shown excellent promise as demonstrated by their use for the melting of uranium.

A summary of the decisions reached at this meeting includes the following:

The work on density of plutonium will continue to have the highest priority in our division.

The Chemistry Division expects to have a fair value for the melting point of plutonium before March 15.

Both Chicago and Los Alamos should continue to get as much information as possible on the crystal structure of plutonium and its compounds, with possibly some exchange of samples between these two sites.

The Chemistry Division will continue to obtain information on the hardness of plutonium metal and on the structure, formula, density, and dissociation pressure of the hydride.

It is very important to have a method of assay for plutonium upon which all parties can agree. For this reason the radioactive method of assay should be improved as much as possible and an independent chemical method of assay, good to within 0.1 percent, should be developed.

Cyril Smith will be furnished CaO crucibles by Spedding, nitride crucibles by Spedding, and sulfide crucibles by Eastman.

It was concluded that the exact form in which plutonium is shipped from Site W to Site Y is not important. A concentrated solution or a wet, centrifuged precipitate would be satisfactory. Site Y wants the

plutonium in such form that it can be stored as long as one month; if a solid, it must be in a form that can be readily put into solution.

Kennedy came to our apartment for dinner before taking the train back to Los Alamos. We reviewed a great number of topics related to the chemistry programs here, at Oak Ridge and Los Alamos. We spent considerable time discussing ways to distribute any money realized from our pre-Met Lab U.S. patents (Cases 52 and 61) that would win the approval of the other inventors, Segrè and Wahl. Kennedy promised to talk over the matter with Segrè and Wahl and give each of us a resume of the most favored disposition. We talked so long that I missed the regular Council meeting of Section C-I, which met at 7:45 p.m. in my office.

The Council meeting was attended by Albaugh, Baumbach, Cunningham, Davidson, Dawson, Dreher, Ghiorso, Manning, Orlemann, Thompson, Watt, and Willard. Willard, who presided, stated that 350 mg of plutonium in the form of the dry peroxide is arriving from Site X for distribution to Thompson (200 mg), Baumbach (75 mg), Cunningham (25 mg), Orlemann (25 mg), and Davidson (25 mg). Cunningham mentioned that recent attempts by Smith to prepare standard enriched and natural uranium plates for Fermi has resulted in samples showing high alpha count presumably due to plutonium contamination. Ghiorso also reported the recent appearance of high alpha background in our counters, frequently sixty to eighty counts per minute. It is clear the recent influx of milligram quantities of plutonium into our laboratories has resulted in widespread contamination. Willard emphasized the necessity for noting the health hazard in working with plutonium. Work must be expedited on the problem of providing adequate hoods. Hogness has told us that Captain Ware will visit us tomorrow to discuss this. We must insist that a sufficient number of hoods of large air transfer capacity (1200 cu. ft. per minute) be installed, including installation above each laboratory bench where work with plutonium takes place. We are trying to hire three or four persons as soon as possible to clean the laboratories on an essentially continuous basis--in the meantime everyone must help to keep the laboratories clean of plutonium contamination. Dr. Nickson has obtained samples of cloth that can stand washing with acid--such cloth might be suitable for laboratory coats from which plutonium contamination might be removed by acid washing.

Davidson reported difficulty in preparing satisfactory samples of white (light brown) fluoride. Varying colors from purple to brown with varying weight changes have been produced. By x-ray analysis, the purple material has the same structure as the black fluoride, which is believed to be PuF_3. Baumbach stated that ever since a degassing technique has been used, no reduction of PuF_4 has failed to produce plutonium metal. Metal is now needed for a melting point determination--Jasaitis' preliminary work shows the melting point is below 1,300°C. Tests of process solutions with a colorimeter will be started by Cunningham's group in a day or two. Watt has shown that he can recover 96% of the plutonium by precipitating it as the peroxide from the lanthanum solution at the tail end of the process under process conditions. He and Howland find they can readily dissolve plutonium peroxide in acid after it has been dried at 90°C.

2/17/44 (cont.)

In the late evening Spof English left by train to return to Oak Ridge taking with him two samples of plutonium of 0.1 and 1.0 microgram, mounted on thin foils by the acetyl-acetonate technique. The foils are to be used by John E. Brolley at the pile at Clinton Laboratories to make measurements on the Pu^{239} fission cross section.

U.S. liberators have smashed Penape, a Carolina Island base, called a Japanese "Gibraltar." Nazis report that the Soviets are crossing Lake Peipus on the ice.

Friday, February 18, 1944

Manning made up a list of titles for me of all the problem assignment sheets now on file in Section C-I. There are 99 of them, classified according to the groups concerned. The group number, the leader, and representative type of investigation areas follows: Group 0 (Seaborg)--heavy isotope research; Group IA (Willard)--adsorption process; Group I (Albaugh)--Bismuth Phosphate Process research; Group II (Watt)--isolation and concentration of plutonium by various methods (proposed) at Hanford; Group III (Dreher)--collaboration on extraction problems with the Technology Division; Group IV (Orlemann)--purification of plutonium and its compounds; Group V (Davidson)--preparation and properties of plutonium compounds; Group VI (Baumbach)--production and properties of plutonium metal; Group VII (Cunningham)--chemical and physical properties of plutonium and neptunium; Group VIII (Cunningham and Dawson)--methods for the recovery of plutonium and neptunium; Group IX (Ghiorso)--instrumentation.

Zachariasen issued a memorandum (MUC-WHZ-9) today reporting his determination of the crystal structure of $PuCl_3$ by the x-ray diffraction technique. The structure is hexagonal, isomorphous with UCl_3. The $PuCl_3$ was prepared by Hagemann.

In an attempt to perform a definitive experiment on the relationship between PuF_4 and PuF_3, Florin and Heath today treated a sample of PuF_4 (as identified by Zachariasen) with a mixture of 50% HF and 50% H_2 at 550°C for 2 hours. The light brown PuF_4 turned to a purple color. It was put into a capillary tube and sent to Zachariasen for identification by x-ray diffraction.

Ralph James left the Englewood Station at 11:30 p.m. on a train trip to Washington University in St. Louis, taking with him a 10 mg plutonium target to be bombarded with deuterons in our search for transplutonium isotopes. He has prepared the target by evaporating a plutonium nitrate solution on a grooved gold target plate made by our shops.

Helen visited Wilma in the hospital this afternoon.

The two-inch headline today says, "Yanks Attack Truk!" Truk is a powerful Japanese air and naval base in the Carolina Islands.

Saturday, February 19, 1944

Jasaitis made another melting point determination using two more pieces of plutonium. The first piece seemed to show signs of melting at 1,125°C, although it did not coalesce into a sphere. The second piece showed no sign of melting. Jasaitis considers that these experiments indicate the vacuum system will have to be improved for successful runs.

Today Hoekstra and Malm completed their experiments, begun on February 10, to test the Bismuth Phosphate Process on the 100 ml scale at W concentration of plutonium and twice W concentrations of fission products. They obtained a plutonium yield of 95% which is good news indeed.

Thompson wrote a memo (MUC-GTS-465) for me concerning future work to be performed in the Chicago semiworks. He suggests the long-range program should include, as its most important part, the investigation of problems connected with the mainline process, such as coatings and alternate concentration methods. He said that if the program meets with my approval, he and the others concerned will work out a detailed schedule and will outline specifically the experiments to be performed.

Today we hired Michael J. Wolf to work as an Associate Chemist in Davidson's group. He has been working for the War Department, Sanitary Corps, Fort Benning, Georgia, and has a Ph.D. in plant physiology, received from the University of Wisconsin in 1942. Lawrence S. Bartell is another new employee. He comes from the University of Michigan where he has been a teaching fellow and he will work as a Research Assistant with Albaugh. Minnie Daniels, one of Kay Gavin's assistants, terminated today.

Helen went to a luncheon shower at Dot Maloney's for Peg Beaton.

U.S. forces have established beachheads on Eniwetok Atoll, westernmost of the Marshall Islands.

Sunday, February 20, 1944

The headline in the Sun today reads, "Warships Shell Rabaul," the first direct naval action against the two major Japanese strongholds in the Bismarck Archipelago.

Helen and I went to a show and dinner with the Baumbachs. Stan dropped by our apartment in the late evening.

Monday, February 21, 1944

A request was made today for the transfer of Frank W. Haeckl and Raymond G. Larson from Section C-II to Section C-I. Another SED man, Leon Leventhal, arrived. He has a B.S. in chemical engineering from the University of California and is being assigned to Dawson's group. Eda Kelley,

who will be a secretary for Orlemann's purification group, also began working today. Her salary will be $30.00 per week. Finally, Madeleine Lenhard, a clerk in Ghiorso's group, resigned because her husband has been transferred.

Today I received the four grooved copper plates in the mail which Hamilton, in our phone conversation February 11, promised to send me. These are to be used as backing plates for plutonium targets. We intend to bombard these targets with deuterons or helium ions at the Berkeley 60-inch cyclotron and then search for isotopes of elements 95 and 96, which to our knowledge have not yet been produced and identified.

Word came back from Zachariasen that the purple plutonium fluoride prepared by Florin and Heath on Friday is PuF_3. Thus it now appears possible to control the reduction of PuF_4 to PuF_3 and, presumably, the oxidation of PuF_3 to PuF_4.

I wrote to Coryell and asked if he is finished using the negatives, which he borrowed, for the chart of isotopes I have in my office in Berkeley. We have requests for the chart and will have to have more printed before a new chart can be made from my updated "Table of Isotopes."

"Chemical Research - Separation Processes for Plutonium. Process Development and Chicago Semiworks Operations" (CN-1353) has been issued. Authors listed are C. Kircher, E. B. Christiansen, and G. Bird of the Technical Division and S. G. Thompson and J. L. Dreher of the Chemistry Division. This is the first report on a cooperative program of process development and semiworks operation being carried out by the Engineering Development Section of the Technical Division and Section C-I of the Chemistry Division. In this cooperative program Section C-I is responsible for defining specific problems to be studied in the semiworks, for evaluating the results obtained, and for backing up the semiworks with process development studies; whereas the Technical Division is responsible for the setting up of the semiworks equipment, preparation of operating instructions, and carrying out of actual operations. The general aim of the cooperative program at Chicago is to test alternate procedures and modifications in present process procedure that may aid in the mainline development and which, because of the limitations of equipment and time, cannot be adequately tested at Site X. It is the further aim of the program to supply to the research groups at Chicago such active solutions of metal from the Site X pile as may be needed.

The problems on which the semiworks is now concentrating are: (1) study of alternate (HCl) methods of concentrating product after extraction and decontamination; (2) investigation and evaluation of methods of dissolving bonded slug jackets; (3) dissolving of uranium metal from the Site X pile and preparation of uranyl nitrate solutions for the Chemistry Division; (4) investigation of ether extraction methods as part of a separation process. A program has been set up whereby Site X will ship about 250 lb. of active uranium metal per month to the Chemistry Division at Chicago. The requirements of the semiworks will be covered by these shipments.

The report includes a summary of the first extraction, decontamination, and concentration runs made in the semiworks; a statement of the present status of development of equipment for ether extraction; and a statement of the program to test the dissolving of slug coatings. It also says that during the past month three semiworks runs (two Argonne metal samples containing 2 micrograms of plutonium per pound and one with Site X metal containing 70 micrograms of plutonium per pound) on HCl methods of plutonium concentration were carried through a dissolving step, a bismuth phosphate extraction step, a bismuth phosphate decontamination cycle, and a concentration step. U^{+4} was used as the reducing agent in both the prereduction and the decontamination steps. Each run used about 25 pounds of uranium metal. Plutonium yields of 89% and 93%, using uranous oxalate and lanthanum fluoride precipitation respectively in the HCl solution concentration step, were obtained starting with solutions of Argonne pile uranium metal, and an 80% yield was obtained in a run with the Clinton pile uranium metal using uranous oxalate in the concentration step. Glass equipment was used in these preliminary runs; a rubber-coated centrifuge and a rubber-coated tank are now available for use in future runs. Equipment for both continuous and batch ether extraction work has been built and installed by the Technical Division and will soon be given preliminary tests.

My section was responsible for the program of the evening seminar of the Chemistry Division, held at 7:45 in Room 241, Ryerson Laboratory. I reviewed our program in Section C-I. Baumbach and Cunningham spoke on the metallurgy and the basic chemistry of plutonium, respectively.

Helen worked at home all day on the "Table of Isotopes." The end of the last draft is in sight, and I was therefore able to write the following message to Professor J. W. Buchta at the University of Minnesota in Minneapolis: "Nearly two years ago, as you know, we were in correspondence about a table of radioactive isotopes, and I promised you at that time to furnish such a table for publication in the Reviews of Modern Physics. Immediately thereafter I was called to the Metallurgical Laboratory at Chicago and became so tied up that I was unable to meet this commitment, although I did continue to keep up on complete coverage of the literature. Just recently I have finally had the time to prepare this table, and it is now in such shape that I can send it to you if you still want it. It is entitled 'Complete Table of Isotopes' and includes in the one table the natural radioactive isotopes and stable isotopes as well as the artificial radioactive isotopes.

"Please let me know if you want this table now, and if so when you might be able to publish it. After some more checking of the items in the table, I could probably send it to you in about a month from now."

War reports in today's paper come from seven fronts. One of the biggest pieces of action was the U.S. bombing of German aircraft factories.

Tuesday, February 22, 1944

Although this is Washington's birthday, work continues as usual.

2/22/44 (cont.)

Hoekstra is beginning experiments today to test whether bismuth phosphate carries Pu^{+3} at W concentrations.

James returned to Chicago at 8:30 this morning from St. Louis. After checking in at his hotel on Saturday, he took our ten-milligram plutonium sample mounted on a gold backing plate to the Washington University cyclotron. The target was bombarded with deuterons continuously from 1:00 p.m. Saturday until 11:00 p.m. Sunday for a total of 5,000 microampere-hours. James brought the target back with him so that we can search for any isotopes of element 95 that might have been produced in the bombardment.

Another man is transferring from Section C-II to our section--Albert C. Krueger. Krueger, who has a Ph.D. from the University of Wisconsin, will work in Ghiorso's instrument group.

We have received news from Los Alamos that Don Mastick and Art Wahl have observed the +3 oxidation state using plutonium they received from Clinton Labs. This represents another independent discovery of Pu^{+3}.

I received a February 19 memo from Kennedy at Los Alamos addressed to Segrè, Wahl, and me that gives the essentials of the two most favored plans proposed for distribution of any benefits accruing from Patent Cases 52 and 61. One plan (referred to by Kennedy as "Plan D-1") provides that all money go into research grants or funds to be administered by the University of California for nuclear physics and chemistry studies, half to be administered by the Department of Physics or Radiation Laboratory and half to be administered by the Department of Chemistry. The other plan (Plan E-2) is similar except that one-half of the money would be divided so that financial benefit will accrue directly to specified persons.

I immediately wrote to C. D. Shane, Radiation Laboratory, Berkeley, that I and the other inventors have decided to formulate a plan of action to be carried through by us and that this plan may or may not be similar to the agreement which is being entered into between President Sproul and Dr. Bush about which I have no knowledge. I said that Dr. Kennedy will give further details of our plan to Mr. Baird through whom we hope to carry on our negotiations.

Cunningham wrote to Connick and Gofman pointing out the need for better coordination between the Berkeley and Chicago groups on work on the basic chemistry of plutonium. He proposes keeping the Berkeley group informed by means of letters which will give information pretty much as we get it, regardless of whether or not it seems reliable enough to put in a report. As an example, he says that their proof of the existence of the +3 oxidation state of plutonium antedates ours and that we regret not having their data to refer to at the time our report was issued. He is referring to our Report CN-1324. Cunningham goes on to report some measurements on the Pu^{+3}/Pu^{+4} couple made by potentiometric titration using about one milligram of plutonium. He also mentions the preliminary examinations of the absorption spectra of +3, +4, and +6 oxidation states of plutonium in various aqueous media, using a Beckmann quartz spectrophotometer. He cites Watters' work with the polarograph on the effect of

complex ion formation on the half-wave potential for Pu^{+3}-Pu^{+4}. He mentions some questionable results indicating that solutions of the +3 chloride may be dried in vacuo to yield a volatile plutonium compound having a vapor pressure at 170°C of around 10^{-6} mm Hg.

Helen dropped by the hospital to see Wilma. We had dinner this evening at the 51st Street Y.M.C.A. with the Baumbachs.

The news reports the capture of Eniwetok after stubborn Japanese opposition.

Wednesday, February 23, 1944

Laura O. Boyd was hired as a clerk for Manning's group.

I mailed Kennedy my version of a Plan D regarding distribution of financial benefits from Patent Cases 52 and 61. I proposed that all the money be used for research purposes at the University of California. Funds from Case 61 would be used for salaries and equipment for scientists working at the Radiation Laboratory at Berkeley and funds from Case 52 would be similarly used for scientists in the Chemistry Department there. I further indicated that I am willing to act with the majority of the inventors along the general lines of either his Plan D or E.

I received a memo from Thompson (MUC-GTS-474) covering possible methods of improving decontamination in the Bismuth Phosphate Process; the problem of new oxidizing agents was also covered. Thompson points out the high solubility of rare earth elements in 1 N HNO_3 and raises again the possibility of making by-product precipitates at very low acidities and product precipitates at high acidities (2 N HNO_3). With regard to work on oxidizing agents he refers to the use of sodium bismuthate with certain catalysts (cerium, cobalt, manganese) to increase the rates of oxidation; the study of such oxidizing agents as PbO_2, $PbAc_4$, Bi_2O_4, $Ag^+ - S_2O_8^{--}$; and the use of catalysts other than silver along with the persulfate.

I replied to Hamilton's February 12 letter enclosing a copy of Jaffey's and Turkevich's summary of comparative U^{237} yield data from cyclotron and pile neutrons which I discussed with him over the phone on February 11. I wrote that I see no harm in surrounding the 50 pounds of uranium metal with cadmium sheet to screen out the slow neutrons. I informed him that preliminary work on the alloying of plutonium with gold looks not too bad and that I have received the four copper target plates that he shipped. I concluded my letter by saying that I am looking forward to seeing him when he attends the Information Meeting of the Health Division on Tuesday, March 7. I mentioned that I plan to leave for Oak Ridge the day after that to stay until Saturday, March 11.

California Project Report, "Chemical Research - Chemistry of Plutonium. Report for Month Ending February 10, 1944," (CN-1360) was issued today. This report includes descriptions of the work of Bob Connick, Bill McVey, and Glenn Sheline on the discovery of the +3 oxidation state of plutonium.

Starting with 244 micrograms of plutonium isolated in December from uranium bombarded with neutrons at the cyclotron, they first added hydroxylamine to the green colored Pu^{+4} nitrate solution. After about six minutes the green solution began to take on a blue cast, and after several hours at room temperature the solution changed to a beautiful blue color. Next, three compounds of this reduced state were precipitated: hydroxide, oxalate, and a double sulfate. Finally, the oxidation number of the reduced state was established as +3 by determining the quantity of dichromate required to oxidize a known quantity of plutonium from the reduced state to the +4 state.

I attended the triweekly meeting of the Extraction, Decontamination, Concentration, and Process Development Groups of Section C-I at 7:45 p.m. in Room 209, Eckhart Hall. Those present were Ader, Albaugh, Bartell, Bird, Dreher, Fields, Flox, Gardner, Gilbreath, Goeckermann, Greenlee, Hoekstra, Hopkins, Hyman, James, Katzin, Kircher, La Chapelle, Lincoln, Malm, Margolis, Pye, Sedlet, Sheft, Thompson, Willard, Winner, Yett, and others. I turned the meeting over to Thompson who began by making a summary report. He quoted recent data from Clinton to the effect that the yield in the last eight runs amounts to 72%. At Clinton semiworks, he said, the overall yield has averaged 88%; also here at Chicago he expects to investigate the trivalent state of plutonium as applied to bismuth phosphate carrying. Albaugh reported on an extraction and decontamination run at Hanford concentrations of plutonium and twice Hanford concentrations of fission products on a 10-ml scale that gave only 51% yield of plutonium overall; this low yield was traced to the use of a reverse strike in the final plutonium precipitation. He feels that this poor result can be avoided in the future by using a direct strike, and he reported experiments that confirm this. Hoekstra reported on a 100-ml run also made at Hanford concentrations of plutonium and twice Hanford concentrations of fission products that gave a 95% yield of plutonium through two bismuth phosphate cycles using the hot direct strike, followed by a lanthanum fluoride crossover cycle. Gilbreath told about the successful use of U(IV) in the pre-reduction step and stated that he has performed several tracer experiments confirming that H_2SO_4 rather than HNO_3 can be used to achieve the acidity of 0.3 M required for reduction. Pye reported on several 7.5 ml runs on the lanthanum fluoride concentration step under Hanford conditions with plutonium recoveries between 97% and 98%. This was followed by precipitations of pure plutonium peroxide in the isolation step with the observation of yields between 97% and 98%. He plans to make three 100-ml runs of a similar nature. Hopkins has carried out four 1-ml scale runs at Hanford concentrations on the lanthanum fluoride concentration step and through the isolation step. He concludes that precipitation from 1 N HNO_3 is satisfactory and that the H_2O_2 concentration should be set at about 5% for satisfactory results. Dreher reported on semiworks runs 14, 15, and 16, testing the use of U(IV) in 1 N H_2SO_4 in the pre-reduction step, the decontamination step, and the uranous oxalate concentration method; plutonium yields of 60% to 75% have been obtained through the extraction and decontamination cycle. He has also tested the hydrochloric acid-uranous oxalate concentration procedure; results look promising. Hyman discussed the slug coating problem. Despite his efforts he has yet to develop a satisfactory method for dissolving the aluminum-silicon bonding on the slugs. Margolis reported that the semiworks

equipment for work on an ether extraction process for recovery and decontamination of plutonium has been set up.

Helen worked on the YWCA budget campaign and then visited Wilma in the hospital.

Los Angeles is facing many problems this week--an electricians' strike and a seven-inch downpour of rain in four days. Schools are closed, and 150 factories are shut down. About 150,000 homes and businesses are without power.

Thursday, February 24, 1944

Studier and Brody prepared a sample of PuO_2Cl_2, sealed it, and gave it to Willard, who left today for a visit to Clinton Laboratories, to take along for pile irradiation in the search for Pu^{240} through use of the Szilard-Chalmers mechanism.

I received a third memo from Thompson (MUC-GTS-480), entitled "Suggestions for Further Work on Concentration and Isolation." In it he reports on some experiments done at my request to test the use of uranous or thorium hypophosphate as well as the corresponding oxalates with respect to their precipitation from 5 or 10 N HNO_3 solutions of bismuth phosphate. He has been successful in precipitating thorium hypophosphate from 10 N HNO_3 and finds that the use of the oxidizing agent ceric ion in 5 N HNO_3 dissolves the thorium hypophosphate upon heating. He will continue the studies. Thompson also writes about the possible advantages of using solvent extraction methods (ether) for concentrating plutonium; he proposes that one man work on this problem very soon. He refers to an experiment by Gilbreath that shows that at Hanford concentrations it may be possible to precipitate plutonium oxalate as a pure compound from HCl solutions of bismuth phosphate. Thompson also cites work by Watt and Goeckermann indicating that bismuth can be precipitated as a hydroxide by neutralizing 10 N HNO_3 solutions of bismuth phosphate, which provides in itself a means of concentrating plutonium.

I received a copy of a memo written by Dempster to Watson on mass spectrographic techniques of analysis for light element impurities and their limits of detectability.

Compton wrote to Allison about the future program of the radiation chemistry section (Section C-II under Milton Burton). He points out that the radiation chemistry work is essentially completed, but that a strong nucleus of the group should be maintained as insurance in case future radiation chemistry problems might arise. He proposes, therefore, that the group take up problems involving our long range program and suggests studies related to power production (radiation corrosion, radiation stability of working substances) and the possible use of our process of producing intense sources of radiation in the general field of photochemistry and photosynthesis.

John Willard's efforts to extricate Joe Halperin from the Army have been partially successful. Joe was scheduled to transfer to Clinton Labs on January 14, but instead was drafted into the Army on January 6. Yesterday he arrived at Clinton Labs, but as a member of the U.S. Army Special Engineer Detachment (SED).

The evening meeting of the Research Assistants of Section C-I was held at 6:45 in Room 209, Eckhart Hall.

I held the usual evening meeting of the Council of our section at 7:45 in my office, attended by Albaugh, Baumbach, Cunningham, Davidson, Dawson, Dreher, Ghiorso, Katzin, Manning, Orlemann, Thompson, and Watt. (Willard was away on a visit to Clinton Laboratories.) I announced that our section has just acquired several new rooms, but no further expansion is possible in our wing in the New Chemistry Building unless the shops are to be moved. I mentioned that Katzin will write a survey report, about 50 pages in length, covering the history and chemistry of plutonium. Ghiorso stated the general level of plutonium contamination in the building is rising rapidly and that segregation of counters for precise work has become necessary. Manning reported on plans for the installation of additional hoods and a new ventilating system as a means of combating the growing health hazard because of plutonium contamination in our building.

An air intake addition to the New Chemistry Building will be constructed to house a large blower and heating facilities and will be located in the backyard at the alley. Ventilating engineers say that the practical upper limit for the central blower is 100,000 cu. ft. per minute; the power required for winter heating is a limiting factor. After passing through precipitators, the air will be piped into the various rooms through vents in the walls. The new hoods will have small individual ventilating fans and will have individual outlets from the building. Manning stated that considering the large number of rooms requiring new hoods and the limit on the total air intake, some system of air rationing is needed. In rooms having 24 feet of new hood space, the hood doors must not be more than half open at any time. Rooms with 12 feet or less will not be so restricted. Alternative rationing methods are (1) mechanical features to prevent a hood or pair of hoods from opening more than one-half at any time, or (2) reliance upon the honor system which would allow greater flexibility in putting large apparatus into a hood. I favor the honor system. Watt thinks the total hood space should be reduced and any attempts at air rationing abandoned.

Manning and I pointed out that it will be two or three months before the new hoods are installed. In the interim the health problem must be controlled by good housekeeping, more frequent use of the present hoods, laying of linoleum, and extensive varnishing. Orlemann stated that at Site Y the air in each room is tested every day, and the rooms are cleaned twice a day. Dawson may train a man to test the air in the New Chemistry Building. Dr. Nickson has fixed 50 micrograms as the maximum amount of plutonium to be worked with outside of a hood and has also limited to four the number of persons who may simultaneously work in any one laboratory.

2/24/44 (cont.)

I asked for an itemization of the experiments to be carried out with the supply of plutonium now on hand and got the following replies. Baumbach's group is working with 10 mg of plutonium for the determination of melting points, hydride synthesis, etc.; he can now produce metal aggregates as large as 300 micrograms by letting metal accumulate after successive reductions. Davidson is using 25 mg for volatility work, including production of chlorides and bromides for metal production work. Orlemann is using 28 mg for current purification work. Cunningham is using about 25 mg; 10 mg will be used to determine the Pu^{+3}/Pu^{+4} oxidation potential in the presence of oxalate, formate, chloride, and acetate; another 10 mg will be required to establish the existing valence state at various stages of the separation process; the remaining 5 mg will be employed in studying the oxidation potentials in the presence of organic complexing agents, in isolating a volatile chloride, and in determining the vapor pressure of plutonium acetyl acetonate. Thompson reported the Separation Sub-section is working with 190 mg of plutonium, of which 176 mg are being used by Watt for large-scale concentration and isolation runs. Albaugh's group is using 14 mg for several 100-cc scale Hanford concentration process runs and other tests. On completion of the current isolation run, Watt will release 64 mg of plutonium to Albaugh for use in one-liter scale, Hanford plutonium concentration runs. Dawson reported that the recent plutonium shipment (on February 15) assayed only 337 mg as against 350 mg attributed by Perlman. The possibility of plutonium precipitating out on the walls of the container due to low nitric acid concentration will be investigated.

I suggested that the hydrolysis of plutonium from nitric acid solutions of pH~1 might be utilized as a separation method. Thompson pointed out that hydrolysis products of basic plutonium salts could be used in a separation process only if a carrier hydrolyzing out at a low pH could be found. Cerium was suggested as a possibility.

Allison has asked us to prepare a 200 microgram uniformly thin sample of enriched U^{235}; Cunningham will do this. I suggested that selected plutonyl compounds be studied, especially PuO_3.

Helen had lunch in the downtown YWCA with Dorothy Paul and then had a coke with Mrs. Barnes.

U.S. carriers have moved the war 700 miles nearer Japan with an attach on two Japanese bases in the Marianas.

Friday, February 25, 1944

John P. Hunt is our latest "short-term" employee. Hunt has a B.S. from the University of Michigan and will transfer to Site X next month after becoming somewhat familiar with our work. A request was made today for the transfer of James Watters from Section C-I to Section C-IV.

Mulliken sent a letter to Gregory Breit at Johns Hopkins University asking for the date of receipt of Project-related papers submitted to the Physical Review in the last two years for which publication was withheld. Of the five listed, three of the papers are co-authored by me viz:

"Radioactive Element 94" by G. T. Seaborg, E. M. McMillan, J. W. Kennedy, and A. C. Wahl

"Radioactive Element 94 from Deuterons on Uranium" by G. T. Seaborg, E. M. McMillan, J. W. Kennedy, and A. C. Wahl

"Radioactive Element 94 from Deuterons on Uranium" by G. T. Seaborg, A. C. Wahl, and J. W. Kennedy. Mulliken intends to publish these papers as secret Met Lab reports. He said that in fairness to the authors he and Breit should note the date of receipt by the Physical Review editor, just as is the custom for publication in a journal.

Because I was tied up at work, Helen went alone to meet Melvin Calvin at the train station today. Later she helped Al Ghiorso take Wilma and their baby girl to their home from the University of Chicago Lying-In Hospital and then cooked dinner for them. When Helen returned home, she, Calvin, and I went down to the Loop to have dinner at Gus' Good Food Restaurant with the Drehers and the Baumbachs; then we all went to see the show "Oklahoma." Since Helen and I had only two tickets, I gave mine to Calvin; I then managed to buy a good seat from a scalper which I occupied while the others sat high in the balcony.

Helsingfors, the capital of Finland, today has been subjected to a twelve-hour raid by 600 Russian planes. Germany has announced that it has no intention of withdrawing troops from Finland, even if Finland makes peace with the Soviet Union.

Saturday, February 26, 1944

James completed the alpha activity measurements on the "94 fraction" (which might contain any element 95 on the assumption 95 is chemically like 94) and the "93 fraction" (which might contain 95 on the assumption it is like 93) that he has chemically isolated from the 10 mg of plutonium bombarded with deuterons at St. Louis on February 19 and 20. He used cold bromate solution to selectively oxidize 93 to separate it from 94. He concludes that all the alpha particle absorption curves associated with the samples definitely show the absence of long-range alpha particles, with the possible exception of one sample which shows a slight hump in its curve; however, this increased counting rate may be due to the sample being especially thick, or to spurious counts.

Buchta, who is acting editor for the Reviews of Modern Physics, replied to my letter to him of last Monday with the words, "I should be very glad indeed if you could complete the table of isotopes which was planned some years ago. I should tell you that just this morning I received a request for a table giving the atomic weights of all isotopes. The inquirer wished to have the most accurate and recent values, and asked if we had published recently a table of such values. Our publication schedule for the Reviews of Modern Physics is much disturbed, as you know, but I think we could publish the table rather promptly."

I sent a detailed memo to Hogness on the subject of providing safe working conditions for personnel in our building and laboratories with

respect to the inhalation and ingestion of plutonium. I point out that the greatest general danger to our personnel is the gradual accumulation of plutonium in the laboratories because of the unavoidable and undetected small amounts that are lost in every operation. I state that this problem can be met only by absolute cleanliness in the laboratories and propose that the following steps be taken within the next two to four weeks:

1. Put linoleum on all floors.
2. Paint or varnish walls and ceilings.
3. Remove steam coils from windows and seal all windows.
4. Provide a cleaning crew to mop every hall, office and laboratory, every laboratory bench, shelf, and hood, twice a day.
5. No laboratory unit should have more than four workers.
6. Every effort should be made to develop adequate methods for monitoring the air and the equipment in the labs.
7. Such dangerous practices as eating in the labs must be stopped.

I also include a table of the linear feet of minimum, additional hood space required for each room.

Some of the comments I make are the following:

"It is impossible to remodel the existing building or to design and build a new one in which research work on plutonium, because of its very nature, can be carried out with complete safety to the personnel. If the work were limited to routine operations, plutonium could be handled with a high degree of safety (as radium is now handled) because the type and scale of operations could be accurately predicted. By and large the personnel recognize this fact and are willing to take reasonable risks in order to do the job in time to be of value, but under existing conditions the risks are entirely unreasonable, and unless the situation is immediately improved, the work will have to be curtailed. We cannot ask for the impossible but we can only take all the steps possible within the limitations imposed by time and by the existing physical plant. The maximum amount of clean filtered air possible should be decided upon and the corresponding installation completed within sixty days. If the heating is the limiting factor, it should be remembered that the bulk of our demands is to be met within a year, and heating is a minor problem for the next eight months. Accidents such as dropping containers, vacuum-line breakage, etc. will occur. There is no fool-proof way of protecting the worker from such hazards. Every man must guard against their occurrence to the extent demanded by the type of operation. This will require adequate filter masks, rubber gloves, and general outer protection. Each group leader will have to be held very strictly accountable for maintaining the maximum degree of protection on this aspect of the problem and careless workers in any unit will have to be removed."

Thompson and I signed a Record of Invention form for "Lanthanum Phosphate Method of Separation of 94," S-1002.

Cunningham wrote to John E. Brolley at Clinton Labs, saying that we recently sent via S. G. English thin foil samples of plutonium containing approximately 0.1 and 1 micrograms. The plutonium was deposited by sublimation of the acetylacetonate onto the foils. He asked if Brolley wants some enriched U^{235} foils; if so a technique for preparing them will have to be developed.

Helen prepared and took a lunch to Wilma at her home.

Sunday, February 27, 1944

Helen, Melvin, and I had dinner at Morton's Tavern (5487 S. Lake Park), and then Melvin and I went to the New Chemistry Building so that I could show him around.

The headline in today's Sun reads, "Losses Stagger Nazi Defenses; Allies Win Air, Land Victories." This refers to the war theater in Italy, near Anzio.

Monday, February 28, 1944

I requested of Hilberry approval for a proposed experiment using the Clinton pile as a source of neutrons. I explained that we wish to search for Pu^{240}, utilizing the Szilard-Chalmers mechanism, to determine whether this isotope will be a complicating factor in the purification and use of plutonium from the Hanford pile. The experiment will consist of irradiating about 10 mg of Pu^{239} in the Clinton pile. The plutonium would be irradiated in the form of PuO_2Cl_2 in HCl solution to look for Pu^{240} as Pu^{+4}, which is chemically separable from the PuO_2^{++} and which is formed by the Szilard-Chalmers mechanism during the irradiation of Pu^{239}. Willard carried this sample with him on his visit to Clinton.

James G. Barrick from the National Carbon Company in Cleveland, Ohio, started work as a Research Assistant in Albaugh's group at $2400 per year. Barrick has an A.B. from Oberlin College.

I received copies of two letters dated February 26, written by Kennedy about Patent Cases 52 and 61. One was to Shane in Berkeley, informing him that we (Kennedy, Segrè, Wahl, and I) are proceeding with our own plan regarding the patent cases in order to eliminate any further delay. The other letter was to D. Baird in Berkeley asking him to determine, in our behalf, whether or not the Government is agreeable to our proposal, what sum they would consider a reasonable settlement, and with what office we should communicate. He emphasizes that our research leading to these cases, especially Case 52, was not sponsored by any government agency. If the Government is agreeable to this proposal in principle, Kennedy suggests that the patent applications be prepared and submitted for signing and assignment as soon as practicable. In the

meantime, he continued, we will enter into further discussions between ourselves and with members of the University about a proper disposition of the funds involved.

I had a long talk with Joe Hamilton, who is visiting the Met Lab, in particular about what we are planning to do to avoid plutonium exposure to Met Lab personnel. In a discussion about our irradiations at Berkeley, he said we can expect a neutron flux from the 60-inch cyclotron amounting to 10^{12} n/sec/cm^2 over an area of 0.5 cm^2.

"X-Ray Diffraction Results for Uranium and Plutonium Compounds" (CK-1367) by W. H. Zachariasen, has been issued and gives the results of x-ray diffraction studies of the following uranium and plutonium compounds:

UF_5	Pu, Phase I
Black UF_4	PuO_2
Green UF_4	PuO?
UF_3	Pu_2O_3, predicted structure
UO_2F_2	PuF_3
	PuF_4
$UO_4 \cdot 3?H_2O$	$PuCl_3$
$UO_3 \cdot H_2O$	
U_3O_8	$PuOF_2$??
UCl_3	PuF_2?
	PuF_{2-3}??
	PuH_3

The study of plutonium metal by Mooney and Zachariasen is reported as follows:

"Pu, Phase I. This phase was first observed in a sample of solid metal which Dr. Fried had prepared by reduction of PuF_4 with calcium. The solid piece had been flattened into a thin circular disk by Mr. Jasaitis so as to give more surface. The same phase has subsequently been observed in two other samples prepared and treated in the same manner and in a fourth sample which Dr. Fried prepared by low-temperature reduction of PuO_2 with calcium. All four samples proved to contain appreciable amounts of ThO_2 from the crucible, and the last mentioned sample also contained CaO. The observed phase which we believe is metallic plutonium (possibly with small amounts of Ca and Al, in solid solution) has a face-centered cubic structure. The lattice constant as obtained from the four samples is

$$a = 4.58 - 4.64 \text{ A}$$

The variation in the lattice constant from one sample to another may in part be ascribed to inaccuracy due to the poor quality of the diffraction pattern, but is probably mainly due to variable amounts of impurities. If it is assumed that the metal is pure plutonium, the calculated density is 15.9 - 16.4. The diffraction lines are somewhat diffuse which may indicate a transition point at a fairly low temperature. Samples of metal prepared by reduction of PuF_4 with Ba or Li do not give the diffraction pattern of Phase I, so that the existence of a second form of metallic plutonium is indicated."

Helen had lunch with Wilma, worked at the Crerar Library on the "Table of Isotopes," and then prepared dinner. Calvin and Hamilton ate with us this evening.

The evening seminar of the Chemistry Division was held at 7:45 in Room 251, Ryerson Laboratory.

Tuesday, February 29, 1944

George E. Moore, a B.S. man from Northwestern University who has completed eight months of graduate studies at the University of Minnesota, began working as a Research Assistant in Dawson's group.

Willard conferred with me about those Met Lab and Clinton Lab people who might join the group he will head when he transfers to Hanford. Asprey, Britain, Brody, Dawson, Dixon, Dreher, Fields, Gilbreath, Goeckermann, Hoekstra, Hopkins, Howland, Kohman, La Chapelle, Lincoln, Magnusson, Malm, Post, Pye, Studier, Walling, Watt, and Yett are being considered. Possibilities from Perlman's section at Clinton Labs are V. Cooper, Fries, Gaarder, Halperin, Hill, G. Johnson, Knox, Koshland, Lindner, Miller, Schuler, Stoughton, Turk, and Werner.

I prepared a summary of metal production tests showing the physical nature of the plutonium metal resulting from reduction of PuF_3, PuF_4, and PuO_2 with four different reducing agents (calcium, barium, lithium, and magnesium).

Compton wrote that he is transmitting to General Groves and others in the Army who are concerned, copies of my memorandum on the possibility of the utilization of U^{233} by the enemy. Compton added that it is his own judgment that such a U^{233} program probably would require more time and industrial effort to develop a conclusion than the Pu^{239} program. Also, he does not see any strong reason to believe that the Germans are actually engaged upon a program of U^{233} production. He does agree, however, that work on its investigation should be kept alive and considers that my suggestion of encouraging the Canadians to follow this line is well worth following up.

In a memo to Willard I quote some calculations of J. B. Work (who works at Clinton Laboratories) about the distribution of fission activity according to isotope, as functions of the time of pile operation and the

time of decay. Because decontamination studies often furnish discordant results when the age of the starting material is not known, I suggest to Willard that a code designation for the starting material used in all future tests would be helpful. Thus, "X-15-30-40" could indicate Clinton pile metal irradiated at 15 kw/ton for 30 days that is allowed to decay for 40 days before using.

Hogness asked me to advise the men in my section who receive registration forms sent out by the American Chemical Society not to answer the question regarding the nature of the work in which the recipient is engaged.

At 2:00 p.m. Ed Orlemann, Helen, and I caught a train to New York. Helen will stay in New York City while Ed and I spend part of Wednesday and Thursday at Brown University in Providence, Rhode Island, discussing the uranium halides with Professor Charles Kraus.

Wednesday, March 1, 1944

Orlemann, Helen, and I arrived in New York City at noon. Orlemann and I took a train to Providence, Rhode Island, to visit Professor Charles A. Kraus of Brown University, while Helen remained in New York City. Kraus, with E. Charles Evers as Associate Director, has a group of about 15 men engaged primarily in work with uranium chlorides. They occupy eight or nine laboratory rooms at the University. Orlemann and I reviewed their methods of preparing UCl_6, UCl_5, UCl_4, UCl_3, UF_5, UF_4, UF_3, and their work on the preparation of refractories. They reviewed with us in detail their methods of preparation and the properties they have observed for each of these compounds. Kraus believes BeO is the best crucible material for molten uranium, although ThO_2 may be satisfactory and CaF_2 is worth trying. He believes the so-called black form of UF_4 is actually UO_2 produced from traces of oxygen or water. They showed us their dry box used for this work which they think is a very good design. After our discussions we had an excellent lobster dinner with Kraus and his people.

Thursday, March 2, 1944

Orlemann and I concluded our discussion on uranium halides and refractories with Professor Kraus of Brown University and returned to New York City, meeting Helen who had been to see Helen Hayes in the play "Harriet" and had visited various stores and Radio City Music Hall. Yesterday she had lunch at Rockefeller Plaza, saw Katherine Cornell, Raymond Massey, and Henry Daniel in the play "Lovers and Friends," had dinner at Jack Dempsey's Restaurant on Broadway, and then saw Paul Robeson, Uta Hagen, Jose Ferrer, Margaret Webster in the play "Othello" in the evening. Today is Helen's birthday, and she and I went dinner dancing at the Roosevelt Grill, to the music of Guy Lombardo. Afterwards, we went to the Piccadilly Circus Bar to see and hear the "Three Suns."

Friday, March 3, 1944

Orlemann and I visited Columbia University to confer with Willard F. Libby, Homer F. Priest, P. H. Emmett, J. F. G. Hicks, and Simon Freed primarily on the preparation, properties, and handling of UF_6. This, of course, is the process gas used in their gaseous diffusion process for the enrichment of U^{235}. They have investigated its methods of preparation and properties very thoroughly and were very generous in sharing their information with us. They also described their work on UF_5 and on UF_4, which interests them primarily as an intermediate in the preparation of UF_6. They have not found any evidence for the formation of a black UF_4 nor have they succeeded in preparing UF_3, UOF_2, or UOF_4. Priest told us their best source of pure HF is Kinetic Chemical Company

(a subsidiary of du Pont). Libby claimed to have the "driest dry box in the world." We also discussed procurement of B^{10} with Martin Kilpatrick.

Helen went to Columbia University alone by subway (and returned by bus). She met Mrs. Libby and had lunch with her at Stouffers. They visited the Museum of Modern Art, the NBC Studios, the Rainbow Room, and the top of the RCA Building and then met Ed Orlemann, Bill Libby, and me downtown. After dinner in the Tudor Room of the Commodore Hotel, we all went to see Milton Berle in the Ziegfield Follies at a theater just off Broadway. This was a very entertaining production with Milton Berle at his best.

Helen, Orlemann, and I caught the train in New York City to return to Chicago. I was feeling rather queasy during dinner and at the theater, apparently an aftermath of my lobster dinner in Providence Wednesday night. Because of the severe shortage of pullman space we had to share a bedroom, and Helen and I occupied the upper berth.

Saturday, March 4, 1944

Helen, Orlemann, and I arrived in Chicago about 5:00 p.m.--our train was very late.

Sunday, March 5, 1944

The headline in this morning's Sun reads, "Forts Raid Berlin First Time." American bombers, "Flying Fortresses," blasted the outlying sections of Berlin for the first time in the war today.

I went to my office to read the papers that accumulated during my absence. The rate of appearance of reports is now so great that it is quite a problem to remain current in reading them. Among the memoranda I read were the following:

Last Wednesday Dreher described in a memo the tentative plans, as suggested by Hogness, for converting Room 217 on the second floor of the West Stands into a "hot" laboratory for the handling of several hundred millicuries of activity. The hot lab will be shared with Sugarman's Section C-III if approved. These tentative plans, which require the removal of the present laboratory bench in the center of the room, will incorporate the following features: a laboratory bench with a working area of 6 ft. by 3 ft. that will be shielded with about four inches of lead, permitting the handling of materials with activities up to 100 millicuries, perhaps up to 500. The bench top will be covered with a sheet of stainless steel which will be sloped so that spilled liquids can be flushed into a sink. At one end of the working area and within the shielding will be a No. 2 International centrifuge. To facilitate observation, adjustable mirrors will be installed. Suction set-ups for the transfer of liquids, tongs, etc., and other measures will be utilized to minimize the necessity of hands being behind the shielding. Plans

are being considered for the installation of a hood, but in order to expedite construction of the laboratory, one will not be included initially.

My office received a copy of a memo sent out Thursday by Kimpton, in the Chief Administrative Office, to Section Chiefs and other principal staff personnel, informing us that the U.S. Army is making available to this and other laboratories 500 men who have completed their ASTP training. We are asked to specify the number and kind of men we want (training and experience).

Jaffey has asked David Rudolph in the Chief Administrative Office to arrange to dispose of over 200 pounds of bombarded uranium metal, oxide, and nitrate now in the custody of Dreher in the West Stands.

Allison sent me a memorandum dated March 1, 1944, enclosing a letter from Ruhoff which gives flowsheets for the treatment of pitchblende ores. He says that the Army is interested in recovering products such as protactinium and ionium and suggests that we review the flowsheets for this purpose. He also mentioned that Dr. Meyer S. Agruss, who extracted protactinium from Joachimsthal pitchblende, might be available to help.

Last Wednesday in a letter to Oppenheimer at Los Alamos, Allison discussed the problem of plutonium hazards. In it he describes a commercial electrostatic dust precipitator being used here in conjunction with a Dersham electrometer. He points out that the lower limit of the amount of heavy metal that can be detected by this apparatus is somewhere between 10 and 50 micrograms which, although adequate for surveying uranium dust in the air, is not suitable for detecting the smaller amounts of plutonium which are hazardous. His letter continues:

"Our thoughts about the plutonium problem have not by any means crystallized as yet, but I give them to you now in their present stage.

"(1) The plutonium problem will exist here at Chicago, at Site Y, at Site X, and also at Site W. Therefore a rather large construction program is indicated.

"(2) We are proceeding on the following assumptions. (a) Five micrograms of plutonium in the lungs is a dangerous amount. (b) A man in the laboratory not doing active physical work will breathe about 10 liters per minute. (c) It would be dangerous to work 48 hours per week for two years in an atmosphere which contains 2×10^{-15} grams per cc of plutonium, or which has 5×10^{-6} alpha emissions per cc per second.

"(3) Some kind of a meter and sampling system is to be devised, such that periodic readings taken through the day will indicate whether this limit of 5×10^{-6} alphas per cc per second has been surpassed in the preceding collecting interval. At present, this will be attempted on a crude scheme of collection on filter paper and measurement in an ordinary alpha counter.

"(4) The 'whiff' meter should serve as an interpolating protection during the collection periods of the long range sampling meter. Thus if it

should turn out that a sampling meter should be read every three hours, the 'whiff' meter should give an instantaneous warning when the alpha count rises to a value such that it is dangerous to remain in the room for the succeeding three hours."

Last Wednesday the regular triweekly meeting of the Purification and Metal Production Groups of my section was held at 7:45 p.m. in Room 209, Eckhart Hall, attended by Albaugh, Arnold, Baumbach, Davidson, Frank, Fred, Hagemann, Heath, Hellman, Hoekstra, Hyde, Jensen, Karle, J. Katz, S. Katz, Katzin, Kirk, Kohman, La Chapelle, Manning, Meyer, Potratz, Rosenfels, Scott, Sheft, Willard, Winner, and Zvolner. Manning conducted the meeting. Jensen discussed studies on purification of plutonium by precipitation reactions using Th^{+4} and U^{+4} as stand-ins. Peroxide and oxalate are the best precipitates, but precipitation alone will not do the job--perhaps a combination of precipitation and solvent extraction will. A number of experiments have been performed by Jensen to determine whether trivalent plutonium can be extracted into solvents. None of the following extracted such plutonium from hydrochloric acid solutions: nitromethane, nitroethane, ethyl ether, methyl-n-amyl ketone, and methyl iso-butyl ketone. Heath described attempts to track down the reason why both PuF_3 (purple) and PuF_4 (white) are obtained when PuO_2 is treated with anhydrous HF. He reported that the HF residual gases are mostly hydrogen and that by adding oxygen to the HF, it is possible to obtain PuF_4 consistently. Hagemann described the preparation of plutonium chloride by reaction of chlorine on the metal. X-ray examination shows it to be isomorphous with UCl_3.

Katz discussed the preparation of plutonium bromide from reaction of HBr with the oxide. There is no significant volatilization at 600°, but complete volatilization at 750° of this bluish green substance. The results encourage the belief that the preparation of bromide "in situ" may be a feasible procedure. X-ray studies of this material are still inconclusive. Davidson reviewed the known facts about plutonium halides. His conclusion is that stable forms of the bromides, chlorides, and iodides may be expected to be trivalent compounds and that they may be expected to resemble the rare earths in their volatility characteristics. Manning reviewed the safety program--the present plans for sealing windows, laying linoleum, and installing additional hoods and some precipitrons.

Two Section C-I evening meetings were held last Thursday while Orlemann, Helen, and I were in New York City. One was that of the Research Assistants, who met at 6:45 in Room 209, Eckhart Hall, and the other was that of the Council members who met in my office at 7:45. In attendance at the Council meeting were Albaugh, Baumbach, Cunningham, Davidson, Dawson, Dreher, Ghiorso, Hindman, Katzin, Manning, and Willard.

At the latter meeting, Manning discussed plans to store chemicals and the like in cabinets in the halls, rather than on open shelves. Dreher reported on preparations to have workers in the West Stands change clothes on entering and to shower before leaving. Katzin announced a series of general lectures for the new men on the Project, to be given next week. It was suggested that Ghiorso speak at a number of the night meetings about contamination of counters.

Willard reported to the Council that he has been told that at Site W the first chemical separation canyon is completed to about deck level and the building for chemistry and physics research is partly completed. Between 20,000 and 30,000 workers are now employed on the site. He said that present specifications require that du Pont produce a solution in which 99% of the cation weight is plutonium; the plutonium will be shipped as nitrate in aqueous solution. At Site X, slinger rings used for the precipitator tank in cell 3 are now able to remove 60 to 80 pounds of bismuth phosphate which is three times the amount used per run. Plutonium yields have been consistently increasing, 77% having been realized through the whole process on one set of runs.

Baumbach reported that Jasaitis now has a vacuum line in operation in which melting point determination on plutonium metal will be made in tantalum cylinders (tantalum does not alloy with plutonium). In order to obtain finely divided plutonium metal for Zachariasen, plutonium hydride will be prepared and then decomposed. So far the best x-ray pictures have been obtained on plutonium metal pieces flattened by Jasaitis. Titanium nitride, the product supported by Site B as best, appears to be a promising refractory, but cerium sulfide produced by Eastman at Berkeley will also be tried.

Several new men have joined the staff of Section C-I. Last Wednesday Paul R. O'Connor, Roy G. Post, Oliver C. Simpson, Mathew T. Walling, Jr., John O. Blomeke, and Robert W. Rasmussen were hired. O'Connor comes from the University of Wisconsin and has an M.S. degree, while Post and Walling have just received B.S. degrees in chemical engineering from the University of Texas. Rasmussen and Blomeke also come from the University of Texas. Rasmussen has been an instructor there in chemical engineering, and Blomeke has completed 9 credits of work toward his M.S. in chemical engineering. Simpson has a Ph.D. in physical chemistry from the University of Illinois (1934); he is here on a leave of absence from the Carnegie Institute of Technology where he is an associate professor of physics. O'Connor is now a Junior Chemist working in Hindman's group, Rasmussen is a Junior Chemist with Dreher; Post, Blomeke, and Walling are Research Assistants in Watt's group, and Simpson will lead a high vacuum technique sub-group in connection with Baumbach's group in Orlemann's sub-section. I learned that Donald C. Stewart, a member of the U.S. Army Special Engineer Detachment and whom I knew as a student at UCLA, arrived at New Chem Friday. He, along with our other SED men, has been billeted at the Miramar Hotel on Woodlawn Avenue while awaiting arrival of some civilian clothing. Our SED men have orders not to visit the University of Chicago campus in uniform. Leon O. Morgan was hired Saturday and will work as a Junior Chemist in Albaugh's group--he comes from the University of Texas where he was an instructor and graduate student. A request was made Friday for the transfer of Louis O. Gilpatrick from Technology to Section C-I. He will work in Baumbach's group.

A two-page isolation flow chart for the peroxide isolation process has been issued by my section. It reveals that this process has an overall plutonium peroxide yield of 96%.

3/5/44 (cont.)

Allison has proposed to Captain McKinley that sealed samples of solutions of plutonium salts be taken to Northwestern University for measurements of magnetic susceptibility by Selwood.

My office has received a memo from R. S. Mulliken, Director of the Project Information Department, about our "A Manual on the Measurement of Radioactivity," written by Jaffey, Kohman, and Crawford in January. He wrote, "May I have a copy of the very interesting manual MUC-GTS-407? This manual is so valuable that I believe it should have received systematic distribution through the Information Office." He then asks for permission to reproduce it as a CL report, or possibly a CC report.

After this rather long session with reports, correspondence, etc. in my office, Helen and I had dinner at Morton's Tavern and then, for relaxation, went to see the movie, "Thousands Cheer," with Mickey Rooney, Judy Garland, and Mary Astor, at the Picadilly Theater.

Monday, March 6, 1944

This morning I gave Don Stewart the usual run-down about our program and some reading material. He told me that he really appreciated being transferred to our group since he had been undergoing final processing in Alabama preparatory to transfer to the European front as an infantry man. Stewart is being assigned to Dawson's recovery group; after a brief tour of our laboratories, I left him with Studier and Asprey.

Frederick D. Johnson, who has been working in Section C-II, has been transferred to Baumbach's metal production group.

Because my section, Section C-I, has so many groups working on diversified problems, we have organized them into three sub-sections of Section C-I. The sub-sections are: Sub-section I, Separation Processes under S. G. Thompson; Sub-section II, Purification and Metal Production under E. F. Orlemann; and Sub-section III, Basic Chemistry and Service under B. B. Cunningham. Thompson, Orlemann, and Cunningham each have been given the title of Assistant Section Chief. I have asked each one to prepare an outline of the work distribution in his sub-section. Using this information Hogness has issued a manpower distribution table for the Chemistry Division's six sections. Of a total of 212 men, 82 are in my Section C-I. The distribution for my section is as follows:

		No. of men
Thompson Site W work 23 men:	Albaugh, extraction and decontamination	10
	Watt, concentration	6
	Dreher, process development	6
Orlemann Site Y work 29 men:	Orlemann, precipitation and extraction	6
	Davidson, volatility	8
	Baumbach, metal production	15

Cunningham	Cunningham, basic chemistry	9
28 men:	Dawson, recovery	9
	Ghiorso, instruments	10

Heath has informed me he wants to conduct his research on the fluorides of plutonium more independently, and not as a member of Davidson's group. In order to accommodate his request I am creating a sub-group, under Orlemann's general direction, in which Heath will have responsibility for his, Florin's, and Zvolner's research on plutonium fluorides.

I had a conference with Ghiorso, whose group is continuing our investigation of the complete (alpha, beta, and gamma) radiation spectrum of plutonium, using plutonium from the Clinton pile.

I learned that Hoekstra completed on last Wednesday his experiment, begun on February 22, to test whether Pu^{+3} is carried by bismuth phosphate at W concentration (Bi/Pu ratio = 12). Hindman helped him determine the yield of Pu^{+3} in the bismuth phosphate precipitate, which was dissolved in HCl to form a pale violet-greenish colored solution. Their measurements with the Beckman spectrophotometer showed that the plutonium carried quite well. His alpha counting assays indicate, more quantitatively, that about 90% of the plutonium (in the form of Pu^{+3}) is carried.

Today I sent a long and detailed memorandum to Hogness summarizing our visits to Brown and Columbia Universities last Wednesday, Thursday, and Friday. In this memorandum I also covered my ideas on possible analogies between uranium and plutonium halides. I have this to say:

"Our detailed interest in the halides of uranium, of course, stems from their possible similarity to the halides of plutonium, and it seems worthwhile to try to make some generalizations along these lines, even though these be little more than guesses. It may be that some of the above described methods for the preparation of the various halides of uranium may be applicable to the preparation of the corresponding halides of plutonium. However, there seem to be certain differences in stability between corresponding halides of uranium and plutonium, and an attempt should be made to take these differences into account.

"In a very crude way, the halide of plutonium of oxidation number n corresponds in stability to the uranium compound with the same halogen where the oxidation number of the uranium is $n + 1$. Again, in a very crude way, the halide (e.g., fluoride) of plutonium of a given oxidation state corresponds in stability to the compound of uranium, of the same oxidation number, with the halogen (e.g., chlorine) of next higher atomic number.

"On this basis, for example, the PuF_6 would have a stability less than that of UF_6 and quite similar to that of UCl_6. PuF_5 would be about as stable as UF_6 or UCl_5. $PuBr_3$ would have a stability corresponding to UBr_4, and we might even expect that it be possible to form a $PuBr_2$ of stability similar to UBr_3.

3/6/44 (cont.)

"These crude considerations are summarized in the following table where each column lists compounds of comparable stability. Of course the comparison is best for adjacent rows.

	UF_6	UF_5	UF_4
PuF_6	PuF_5	PuF_4	PuF_3
UCl_6	UCl_5	UCl_4	UCl_3
$PuCl_6$	$PuCl_4$	$PuCl_3$	$PuCl_2$
--	UBr_4	UBr_3	
$PuBr_4$	$PuBr_3$	$PuBr_2$	
	UI_4	UI_3	
	PuI_3	PuI_2	

I received Coryell's reply to my letter of February 21 about the return of the California negatives. He is returning the negatives but notes that he has been unsuccessful in obtaining satisfactory prints. He asks for two dozen prints when I have more made. Coryell also mentions that he is sending me copies of notes on his atomic physics lectures for chemists there at Clinton.

Kraus from Brown University is at the Met Lab for discussions today and tomorrow, an interesting coincidence in view of my visit with him in Providence last week.

Before Helen and I left for New York, I sent Martin Kamen in Berkeley, among others, a copy of my new "Table of Isotopes" for review. He wrote asking if a more accurate value than 2.5×10^4 years for the half-life of carbon-14 (his determination) is available. He wrote, "I understand that very large yields of carbon-14 can now be obtained with your set-up, and I am wondering whether the work (of measuring the half-life) could not be repeated so that a more accurate value was obtainable." He expressed the hope that Helen is in good health and that I likewise am over the "Chicago Megrims."

Nickson urged Manning that we investigate the possibility of using metal, enamel, or glass trays for work on the chemistry of plutonium instead of depending on a bench covering of paper. In this way, he thinks, an impervious easily cleaned surface could be obtained and thus minimize contamination.

I asked Heath, J. J. Katz, Davidson, Baumbach, Manning, Jensen, and Orlemann to meet with me in my office to discuss purification problems with my visit to Brown and Columbia Universities in mind. We agreed that with the approximately 70 mg of PuF_4 now available, metal production for the determination of the melting point, density and hardness, as well as hydroxide formation and halide preparation, must be carried out as rapidly as the resumption of work in Room 37 will permit (following completion of our re-modeling program). As soon as $PuCl_3$ can be made available, studies of its reduction to metal will be made. Other programs agreed on are:

1. Heath will push experiments to determine the existence of a higher fluoride or fluorides of plutonium.

2. Hagemann will temporarily move to Room 36 so that he can carry out the preparations of plutonium bromides and iodides immediately. Providing plutonium metal for these preparations is therefore a high priority job for Baumbach.

3. The cubicles in Room 2 will be completed as soon as possible so that studies on halide production from oxides using CCl_4, HCl, and HBr can be resumed.

"Chemical Research - Production and Extraction of Plutonium. Adsorption Methods for the Extraction and Decontamination of Plutonium" (CN-1373) has been issued and covers the work of Beaton, Fields, La Chapelle, Lincoln, and Sheft. Investigations of a two-cycle, all-adsorption, extraction, and decontamination process are continuing using an Amberlite IR-1 extraction cycle. Two runs were carried out with Clinton pile uranium metal solutions and, although an overall beta and gamma decontamination in excess of 4.0×10^3 was obtained, plutonium recovery was extremely poor; impurities and a faulty pH adjustment are believed to be the cause. Preliminary work on the coupling of the Amberlite IR-1 extraction cycle to the bismuth phosphate decontamination cycle shows that, although plutonium recovery is satisfactory, gamma-decontamination factors of only 40-50 are obtained; more work is needed. Because of a recent rearrangement of the adsorption program, the search for more highly specific adsorbents is now suspended.

The "Metallurgical Laboratory Progress Report for February 1944" (MUC-NH-807) has been issued. Some of the highlights of the report are as follows:

Considerable work has been carried out by the Nuclear Physics Division during this month in an attempt to learn something about the radioactive properties of U^{236} and ascertain its rate of formation in our piles. The answers to the problem are of the greatest importance to Sites Y and W. First attempts to measure the lifetime of U^{236} have failed, indicating a value of more than 10^5 years with respect to alpha emission. Experiments to determine the rate of formation and hence the true value of the number of neutrons emitted per neutron captured in U^{235} have been initiated. The value of the absorption cross section of U^{235} has been measured in a beam geometry and found to be 640×10^{-24} cm^2 for neutrons with 2,200 meters per second velocity. Also the value of ν has been measured again, and the new value is almost identical to the one found previously namely, 2.15 compared with the previous value of 2.14. An amount of heavy water adequate for carrying out the exponential experiment with metal rods embedded in heavy water has been recently received. Experimental work started immediately, and it is expected that the first exponential experiment will be completed in about a week. The construction of the Argonne heavy water pile is progressing satisfactorily, and the machine will be ready by the time a sufficient amount of heavy water will be available for filling it. A revision of estimates of the amount

of heavy water needed for a homogeneous pile has been made. The earlier figure of about 30 tons for a pile at ordinary temperatures has now been raised to about 50 tons. Another main line of work concerns the theory of neutron diffusion, and a considerable part of the activity of the Division has been dedicated to the problem of improving our knowledge of neutron cross sections. Assistance was given in the jacketing program of the Technical Division by carrying out various calculations on the temperatures to be expected at different parts of the slugs. This involved calculations on the rib effect and on the effect of defective spots in the coating bond.

The problem of devising a suitable canning technique for heavy metal slugs for Hanford continues to occupy a large share of the time of the Technical Division. A satisfactory technique would be one in which the danger of canning failure in the pile would not be the limiting factor in the permissible operating power. Major emphasis is now being placed on aluminum silicon bonding with a welded closure. Considerable difficulties are still encountered in both the bonding and closure operations. The difficulty is that time is running out, and the Division cannot say definitely when the solution will be found. A large number of alternates to all phases of the canning process are being considered. Favorable progress continues on the studies of: (1) heat transfer between graphite blocks in mechanical contact, (2) heat transfer rates from slugs at Site W flux levels, (3) detection of leaks in Hanford water tubes, (4) aluminum corrosion laws, (5) effects of coatings on removal and subsequent canyon operations, and (6) optical instrument developments. The apparatus for the ether extraction system has been tested and, after minor modifications, will be ready to yield separation data. Contact and cooperation has been maintained with the development at Site X of the adsorption process.

Contamination with plutonium and methods of preventing same are receiving a great deal of attention. The most important procedure now is for personnel to protect themselves by extreme precautions including masks when work is not done under a hood. Hand, glove, and coat contamination studies are continuing. Final reports by the University of California Radiation Laboratory on the metabolic studies of columbium, cesium, barium, praseodymium, and emanation have been completed and will appear as CH reports during the next month. Attempts at removing yttrium and cerium from the body have so far been unsuccessful. As for health physics and radiation measurements, pocket meter service is expanding both at Clinton Laboratories and the Met Lab in Chicago. Atmospheric surveys at Clinton have shown no appreciable activity due to pile operation to date.

The research activities of Section C-I had a wide range as usual. A 10-cc scale of operations, with Site W concentrations of plutonium and twice Site W concentrations of fission products, does not confirm the microscale work on bismuth phosphate plutonium precipitation steps: incomplete carrying has resulted. Reduction with SO_2 rather than Fe^{+2} does result in complete carrying. The reasons for this departure cannot be stated at this time. A 10-cc scale of operations with Site W concentrations of plutonium and twice Site W fission products did not give expected holdup of plutonium due to oxidation, a 97% yield being obtained

for the extraction step. A repeat of this work with twice Site W concentration of plutonium gave 98% yield. Two 100-cc scale extraction-decontamination-crossover runs gave slightly conflicting results; one gave 95% overall product yield with low decontamination of both beta and gamma, the other gave better decontamination (10^5) but subnormal yield. These are inconclusive results. Important extensions of information were obtained in the following: (1) holdback carriers, (2) lanthanum phosphate precipitation, (3) acidity factor in product precipitation, (4) bismuth phosphate cycle oxidizing agents and reducing agents (U^{+4}), (5) crystal structure of bismuth phosphate carrying of plutonium, (6) effect of temperature on decontamination, (7) concentration process Site W flowsheet, (8) removal of slug coatings and effect on following processes, (9) ether extraction, (10) concentration, (11) waste disposal. Four samples of calcium-reduced plutonium metal show a face-centered cubic structure giving 16.4 ± 0.5 gm/cm^3 as the density of the metal. One sample of barium-reduced plutonium metal and one of lithium-reduced plutonium metal show a complex structure not yet analyzed. A direct method gives 16.2 gm/cm^3 as the density of metal assaying 96% plutonium.

Plutonium hydride has formed readily on every sample tested, whether barium or calcium has been reductant. One sample (of two 300 micrograms pieces) was analyzed by x-ray methods, and no lines were found attributable to the hydride; this sample was formed by reduction with barium and the hydride formed at room temperature.

In other activities of the Chemistry Division, the energy liberated in fission has been re-evaluated at 185 ± 10 Mev, as determined in the first few minutes in a metal lump. The study of the short-lived fission products continues to proceed satisfactorily. More than one-half the long-lived activities have been isolated and procedures on the others are being checked.

Total expenditures of the month of February for the Met Lab at Chicago are $631,752. Personnel employed on February 29 total 1,427 (430 academic), a net increase of 147 during the month. The mechanism of handling the Hanford designs remains unchanged, and during the month 62 drawings were approved, bringing the total to 613. The record file of Metallurgical Laboratory activities in connection with Hanford Design approval has been brought up to date. There were 8 visitors from Canada during the month. The total man-days of visitation was 25. Canadian representatives witnessed the heavy water exponential pile experiments.

The evening seminar of the Chemistry Division was held at 7:45 in Room 251, Ryerson Laboratory.

Helen visited Wilma this afternoon.

Marshal Stalin announced a new offensive in the western Ukraine under the leadership of Marshal Zhukov, and the Soviet army is only 60 miles from Rumania.

Tuesday, March 7, 1944

Our laboratories in Section C-I are in the process of being remodeled to make them safer for our experiments with plutonium and, therefore, are quite torn up. This makes it somewhat difficult to carry on our work in some of the rooms and results in some modification of our program in order to conform with the conditions. We are looking forward to the completion of this important task.

Alec E. Kelley has joined Section C-I. He has received a B.S. degree from the University of Texas and will work in Watt's group as a Research Assistant. This was the last day at the Met Lab for John P. Hunt who is transferring to the Clinton Laboratories as a research assistant. Benjamin Bradshaw plans to terminate his work at the Met Lab Thursday.

I received a copy of a March 6 memo from Allison to Hogness stating Compton has notified him that 1.4 grams of plutonium will be sent to us on March 10. Allison suggested that one gram of this material be converted into PuO_2 for Fermi to use at Argonne Laboratory to make an experimental investigation of the number of neutrons emitted per fission and the energy spectrum of the emitted neutrons.

At 9:30 a.m. I attended the Project Council Information Meeting for the Health Division in Room 209, Eckhart Hall. There were many out-of-town guests. Attending the meeting on health were Allison, Chipman, Carl, Cole, Compton, C. M. Cooper, Curtis, Curtiss, Darrow, Franck, Hamilton, Hilberry, Hogness, Jeffries, Mitchell, Mulliken, Nickson, Pardue, A. V. Peterson, Selwood, Spedding, Stone, Vernon, Warren, C. J. Watson, W. W. Watson, and Whitaker. Joining us later in order to attend the meeting on nuclear physics were Alvarez, Anderson, Bacher, Dancoff, Fermi, Friedman, Goldsmith, Hughes, Jesse, H. Jones, Kirst, Marshall, Miles, Morrison, Newson, Nordheim, Snell, Way, Weinberg, Wheeler, Wigner, and Zinn. The biological effects of external radiation and ingested radioactivity as determined by animal experiments were discussed. There ensued a long discussion on the tolerance for plutonium in the lungs where as much as 20% retention of dust was predicted. Pardue said that a preliminary investigation indicates oral ingestion leads to low absorption for all three valence states--less than 0.05%. Hamilton gave a report on his Berkeley program of investigation of the ingestion and retention of fission products.

At the conclusion of the meeting of the Health Division, the Project Council Information Meeting on Nuclear Physics started at 11:40 a.m. Present were Allison, Alvarez, Anderson, Babcock, Bacher, Borst, C. M. Cooper, Curtiss, Dancoff, Darrow, Fermi, Franck, Friedman, Goldsmith, Greninger, Hilberry, Hogness, Hughes, Jesse, Jeffries, Haydn Jones, Kirst, Marshall, Miles, Mitchell, Morrison, Mulliken, Newson, Nordheim, Pardue, Sawyer, Selwood, Smyth, Snell, Spedding, Stone, Vernon, W. W. Watson, Way, Weinberg, Weller, Wheeler, Whitaker, Wigner, Wollan, Young, and Zinn. Bacher reported that at Site Y he and his group have compared the fission neutron spectrum from U^{235} and

94^{239} using 2 grams of enriched U^{235} and 1/2 gram of 94^{239}. Within errors of measurement, the U^{235} and 94^{239} fission spectra are identical in the region between 1 and 4 Mev. Fermi asked if the half-life of plutonium has been remeasured here or at Site Y, and I replied that we have a new value of 24,000 years. Bacher added that this value is now in use at Site Y, which means a disintegration rate for 94^{239} of ~138,000 d/min/μg. Alvarez reported that he has performed a preliminary experiment to look for long-range alpha particles emitted in the neutron-induced fission of U^{235} with negative results so far.

The meeting on nuclear physics continued after lunch, and when it concluded I attended a third Project Council Information Meeting in Room 209, Eckhart Hall, this one on General Physics. Others present were Allison, Alvarez, Anderson, Babcock, Bacher, Borst, Chapman, Compton, C. M. Cooper, Curtiss, Dancoff, Darrow, Dempster, Fermi, Franck, Friedman, Goldsmith, Greninger, Hilberry, Hogness, Hughes, Jeffries, Jesse, Haydn Jones, Kirst, Marshall, Miles, Mitchell, Moon, Mrs. Mooney, Morrison, Mulliken, Newson, Nordheim, Pardue, Sampson, Sawyer, Selwood, Smyth, Snell, Spedding, Stone, Vernon, Warren, W. W. Watson, C. J. Watson, Kay Way, Weinberg, Weller, Wheeler, Whitaker, Wigner, Wilson, Wollan, Young, Zachariasen, and Zinn. In a discussion of the development of alpha particle counters to operate in the presence of strong beta particle backgrounds, I mentioned the satisfactory results we have had with alpha counters containing nitrogen gas, received from Site Y. Snell talked about the search for some of the fission products at the end of decay chains; in answer to my question he indicated no search has been made for a gallium activity in uranium fission products.

Zachariasen reported on his recent work on estimating the size of plutonium ions in trivalent and tetravalent states as compared with other such ions of the related elements. He concludes that, in the tetravalent state, thorium, uranium, cerium, and praseodymium have the same molecular structure in their compounds as plutonium. Also, in the trivalent state, uranium or any of the first six members of the rare earths will be isostructural with plutonium compounds. His values for the radii of plutonium are: $Pu^{+4}-O = 2.27$ Å; $Pu^{+3}-O = 2.45$ Å. Mrs. Mooney reported obtaining more consistent results on the change of bismuth phosphate forms. Her results confirm previous conclusions that diffraction patterns show, for digestion times less than one hour, all bismuth phosphate is of the hexagonal form; as digestion time increases, the second, insoluble form, appears and becomes increasingly predominant. The hexagonal structure of bismuth phosphate has been completely characterized; the structure is an open network with rather wide channels running along the hexagonal axis. I asked if it were possible for tetravalent plutonium to enter these open sites, to which Mrs. Mooney said, "Yes, but in absence of experimental data, it is only a speculation."

Helen worked at Crerar Library on the "Table of Isotopes" and later had an appointment with a personnel man at the laboratory to arrange for her to type the manuscript in final form here.

Helen and I attended a dinner at the University of Chicago Quadrangle Club in honor of Professor Kraus of Brown University. Among those present

were the Comptons, Hognesses, John Warners, Zay Jeffries, and James Franck.

Today's headlines report a terrible sky battle on the aerial highway between London and Berlin and over Berlin. Losses were heavy for both the Allies and the Axis.

Wednesday, March 8, 1944

In response to my request Thompson prepared the following account for me of the work distribution in Sub-section I, Section C-I, as of last Monday:

Group I Extraction and Decontamination

Albaugh	Supervision and reports.
Hoekstra Ader	Variables affecting carrying of plutonium at Hanford concentrations with bismuth phosphate.
Malm Bartell Sheft	100-cc scale runs in glass and stainless steel (with controls on Clinton metal solution) using Hanford concentrations of plutonium and fission products. Follow flow sheet conditions and measure yields and decontamination.
Gilbreath Post	Use of rare earth phosphates as scavengers in Bismuth Phosphate Process.
Sedlet	Effect of lower temperatures in Bismuth Phosphate Process on carrying and decontamination.
Greenlee	New oxidizing agents for Bismuth Phosphate Process. Studying $NaBiO_3+KMnO_4$; will do further work on PbO_2, $PbAc_4$, and Bi_2O_4.
Morgan	Breaking in--will probably work on neptunium-plutonium separations.
Gardner	Breaking in and preparing samples for mass spectrographic studies.

Group II Concentration and Isolation

Watt	Supervision and reports.
Pye Yett	Hanford concentration process--carrying by LaF_3 at various lanthanum:plutonium ratios and various HF concentrations. Also, studying dissolving of metathesis products following concentration.
Hopkins	Isolation process--studying precipitation of the peroxide at various temperatures and influence of H_2O_2 in wash solution on yield.

3/8/44 (cont.)

Goeckermann	Influence of Fe^{+3} on precipitation of PuO_4.
Haeckl	Alternate concentration process for Hanford involving direct precipitation of plutonium peroxide from solutions containing high concentrations of Bi^{+3}.
Walling Kelley	Breaking in.

Group III Process Development and Semiworks

Dreher	Supervision and semiworks assistance.
Hyman Winner Flox	Problem of coating removal and effects of scale and insoluble residues on yield and decontamination in Bismuth Phosphate Process.
Lincoln Larson	Evaluation of process modifications. At present making runs according to standard flow sheet conditions to establish base line for certain modifications to follow. Will also check behavior of 93 in process.
Rasmussen	Breaking in with Lincoln.
Margolis Malekow	Development of ether extraction process--studying at present the extent of decontamination obtained by second and third ether cycles.

I also received a memo from Thompson giving the plans for semiworks operation at Chicago until June. The problems to be studied include the uranous oxalate and lanthanum fluoride concentration methods from HCl solutions of bismuth phosphate after both U^{+4} and Fe^{+2} reduction, effect of coating residues on yields and decontamination, alternate methods of concentration, and new methods of decontamination.

In another memo Thompson gives suggestions for items to bring up before the Clinton Steering Committee meeting I will attend in Oak Ridge day after tomorrow. His reaction so far to the status of the process as it applies to Hanford are (a) there probably won't be any difficulty with carrying of plutonium by bismuth phosphate provided scaling up is satisfactory and provided some of the present conditions are not near a critical region, (b) unless some good methods of improving decontamination are found, it will be necessary to have more than two bismuth phosphate cycles.

Ralph Gardner, a Research Assistant, is a recent transferee from Section C-II to Section C-I.

I read a report to Stone, written on University of Chicago stationery during his recent visit here, from Joe Hamilton on present and future health activities at Berkeley. He listed 15 fission products that will have been studied by June 30 of this year and then outlined the anticipated program for the fiscal year 1944-45. An enumeration of the various

phases of the program to be assigned in order of priority is as follows: (1) Completion of plutonium tracer studies. (2) Completion of radioactive smoke experiments. (3) Continuation of studies concerning the removal of absorbed long-lived fission products from experimental animals. (4) Initiation of tracer studies of the long-lived fission products of low abundance (and possibly Np^{237}). (5) An investigation of the deposition and exchange reactions between the long-lived fission products and various types of soils.

I sent a summary to Hogness of the work of the past month on separations processes on a problem number basis.

At 7:45 p.m. I attended the triweekly evening meeting of the Basic Chemistry, Recovery, and Instrument Groups (Sub-section III) of my section in Room 209, Eckhart Hall. Others present were Arnold, Barrick, Cunningham, Davidson, Dawson, Dixon, Eiseman, Gilbreath, Hagemann, Heath, Hindman, Hoekstra, Howland, Hyde, Jaffey, Katzin, Kirk, Kohman, Kraus, Krueger, Larson, La Chapelle, Magnusson, Manning, Meyer, O'Connor, Patton, Phipps, Rosenfels, Sheft, Clifford Smith, Studier, Warner, Weissbourd, Wetlaufer, and others. As usual I turned the meeting over to Cunningham.

Howland reported on his work on the structure of plutonium peroxide; a titration experiment indicates a formula of PuO_4. An attempt to prepare a peroxide of Pu^{+6} failed. During the discussion I mentioned that Perlman has found it necessary to have $SO_4^{=}$ present in the precipitation of the peroxide. Kraus reported on the hydrolytic behavior of Pu(IV) in 0.01 M $SO_4^{=}$ in the pH region 2 to 6. He finds that the log of the solubility of the plutonium(IV) hydroxide is a linear function of the pH in the pH range 3 to 5.

Patton reported that an analysis of the plutonium(IV)-8-hydroxyquinolinate shows that the ratio of the quinoline to Pu(IV) is four to one. He summarized further work on the following organic derivatives of Pu^{+3}: salicylic acid, salicylic aldehyde, salicylaldoxime, α-nitroso-β-napthol, quinaldinic acid, pyrogallol, phenylarsonic acid, dithizone. I called attention to the fact that the organic derivatives are advantageous for (a) separation of plutonium from light element impurities; (b) as an aid in the analytical program substituting for cupferron, and (c) as a possible concentration method as was suggested by Hogness. Dawson mentioned that up to February 20, 20 mg of plutonium have been recovered, the usual method being LaF_3 cycles, dissolving LaF_3 precipitates by fuming with H_2SO_4 or metathesis with KOH or with K_2CO_3 and KOH, and hydroxide and peroxide precipitations. Recent use of ether extractions has reduced considerably the time required for recovery and purification.

Kohman brought up the problem of neutron counting. A counter of very high efficiency will be required for use in the "shot-gun" test to determine purity of the final plutonium product by measuring the number of neutrons emitted. I mentioned a statement by Bacher at yesterday's Project Council Information Meeting that his Site Y group has found that BF_3 of high purity doubles the sensitivity of neutron ionization chambers over that obtained with tank BF_3. Jaffey spoke about range measurement

techniques to measure the alpha particle range of a single isotope or the relative amounts of more than one isotope. The simplest method involves the use of the demultiplier vacuum low geometry chamber in which the absorption is varied by varying the air pressure. For measuring small amounts of short-range alphas in the presence of long-range alphas, it is preferable to use the differential range chamber. The disadvantage of both these chambers is the quite low geometry. For low activities it is preferable to use a chamber of 52% geometry, such as the spherical range chamber. Ghiorso again warned that contamination of the counters is developing into a very serious problem.

Helen visited Wilma in the afternoon.

At 11:30 p.m. Hogness, Selwood, and I left Chicago (Englewood Station) on a trip to Knoxville via Penn R.R. with a change to the L&N R.R. in Cincinnati.

This morning's paper reports that U.S. troops have made another landing on New Britain on the north coast of the Willaumez Peninsula.

The Project Council Policy Meeting took place at 9:30 a.m. in Room 209, Eckhart Hall. Present were Allison, Chapman, Chipman, Compton, C. M. Cooper, Dempster, Fermi, Franck, Hilberry, Hogness, Jeffries, Kimpton, Mulliken, Norwood, Smyth, Spedding, Stone, Vernon, Warner, C. J. Watson, W. W. Watson, Whitaker, and Wigner. In his "State of the Nation" summary Compton discussed Project timing and indicated that it now appears that the first bombs will use U^{235} separated by the calutron process at Y-12 in Oak Ridge, that the first quantity production of fissionable material will be 94^{239} produced by our Project, and that a combination of effort at K-25 (using the diffusion process) and Y-12 will also give a considerable quantity of U^{235} at about the same time. Fermi reminded those present that the U^{235} route is limited by the smaller amount of this naturally occurring isotope while the potential amount of 94^{239} made from the abundant U^{238} is much greater; thus if the war should last a long time--say 5 to 10 years--we would run out of uranium resources unless the more effective use of it is made. The first pile units at HEW (Hanford Engineer Works as it is now called) should be ready for operation in 4 to 5 months. After the first unit is ready to operate, some 8 to 12 months will elapse before the first 12 kg of plutonium will be available for the first plutonium bomb, which is the amount required. Hence, for purification and metal reduction developments, we have a maximum of perhaps 14 months. Compton stressed that such information as this is needed for intelligent planning, but it must not go beyond the group assembled at this policy meeting.

In a discussion of possible limits of uranium resources, Jeffries pointed out that the price of uranium in the past has been that of a by-product which has been a "drug on the market." If the price of the material were to rise to, say, $5.00 per pound at the mine, the supply would be greatly enhanced because uranium is a relatively plentiful element.

3/8/44 (cont.)

Compton summarized a discussion of staffing HEW and Site Y with Met Lab people (John and Leona Marshall, Alvarez, Zinn, and Morrison have already been requested to transfer) by saying there is evidently a unanimous feeling of those present at the policy meeting to maintain a going organization at Chicago.

Whitaker reviewed progress at Clinton Labs, stating that the current efficiency in the 205 (chemical) plant is now up to 80%. As of now, 72% of the plutonium charged into Building 205 has been accounted for out of Room D (the room where isolation of plutonium occurs). Present production through extraction is about 0.2 gram of plutonium per day and pile production is about 20 grams per month. During the course of the meeting, program reports were given by Stone for the Health Division, Fermi for the Nuclear Physics Division, and W. W. Watson for the General Physics Division.

Thursday, March 9, 1944

Hogness, Selwood, and I arrived at Clinton Laboratories this afternoon. I again am staying with the Vance Coopers.

Friday, March 10, 1944

At 10:00 a.m. I attended the first session of the two-day Chicago-Clinton Steering Committee (Chemical Conference). Others present were Brown, Doan, Greager, Hogness, W. C. Johnson, Kay, Kirst, and Squires. The Hanford Engineer Works preliminary separations process flowsheet of February 1 was surveyed and problems at various points were discussed. The main problems discussed were:

1. Feasibility of multiple metal charges in dissolver--determination of a reasonable upper limit.

2. Coating removal problem--decision between sodium hydroxide and acid coating-removal method is desirable.

3. Sludge problem and coating removal.

4. Elimination of the use of formic acid.

5. Replacement of formic acid with U^{+4} if prereduction is necessary.

6. Storage of solutions containing product and by-products after formic acid reduction.

7. Use of 2.3 grams of bismuth per liter versus 2.5 grams of bismuth per liter in the extraction step.

8. Variables in the extraction step such as the digesting temperature, the effect of iron, UNH concentration, type of strike, decontamination obtainable, and acid concentration.

9. Acid wash vs water wash for product precipitates.

10. Optimum centrifuging rate for bismuth phosphate precipitates.

11. Flexibility of washing precipitates with anything desired.

12. The feasibility of adding carbonate to the metal solution rather than the metal solution to the carbonate in waste disposal.

13. Determination as to whether or not fission activities are volatilized during the boiling of waste in the waste storage tank.

14. The desirability for the presence of fluoride ion in the fission decontamination cycle.

15. Determination of the most desirable scavengers in the decontamination cycle.

16. Striking into hot water vs striking into cold water in decontamination cycle.

17. Possibility of using lanthanum phosphate as a scavenger.

18. Desirability of iron as a reducing agent from the points of view of decontamination, product carrying, and inhibiting the formation of the insoluble form of bismuth phosphate.

19. Desirability of alternate reducing agents.

20. Waste disposal of coating solutions--the use of 50% caustic solution instead of 30%.

21. Question as to whether one should oxidize final solution in the 221 Building (Chemical Extraction Plant) or jet the dissolved cake solution to the 224 Building (Concentration area) and oxidize there. Stability of oxidized solution in storage tanks. Possibility of precipitation of bismuth phosphate in the connecting pipe lines between the two buildings.

22. Desirability of lanthanum fluoride by-product precipitation.

23. Use of manganous as a catalyst for oxalate reduction in the crossover.

24. The necessity for HF-nitric acid washes of lanthanum fluoride product precipitates.

25. The amount of excess acid necessary in order to satisfactorily dissolve lanthanum hydroxide precipitates in the plant.

26. Desirability for the presence of zirconium in the fluoride concentration cycle.

27. Difficulties in the precipitation of plutonium peroxide.

28. Desirability of devising alternate methods for final product precipitation.

Saturday, March 11, 1944

I attended the second session of the Chicago-Clinton Steering Committee (Chemical Conference) along with Brown, Greager, Hogness, Johnson, and Kay. As a result of yesterday's discussions, it was agreed that my section should take on the following assignments: 1. Search for new oxidizing agents (1 man jointly with Clinton). 2. Uranous oxalate method for concentration of plutonium (one man part-time jointly with Clinton). 3. Solubility of plutonium peroxide (two men jointly with Clinton). 4. Investigation of the chemistry of plutonium (8 men).

We caught the train to Chicago via Cincinnati (Southern R.R. to Cincinnati, then NYC R.R. to Chicago). I have with me the PuO_2Cl_2 solution which the Clinton Lab people refused to irradiate because of concern that the pressure developed by decomposition of the water would be too high for the container. Willard had taken it to Oak Ridge about two weeks ago with the intent of having it irradiated in our search for Pu^{240}.

Sunday, March 12, 1944

When we arrived in Cincinnati, we made a run to catch the streamliner train "James Whitcomb Riley" to Chicago. This has the advantage of getting us into Chicago several hours earlier than the continuing train. It is also better stocked with food. However, the connection is very tight, about a 10 minute leeway, and despite many previous attempts this is the first time we have been successful. We arrived in Chicago at noon.

Helen told me that she had spent the whole morning Friday at the Met Lab working on her employment status--the personnel office insists that she be paid, against her wishes, as a typist to work on my "Table of Isotopes." Helen had both lunch and dinner that day at the Ghiorsos. Yesterday she spent the afternoon at the Crerar Library working on the "Table of Isotopes."

This evening Helen and I had dinner at Morton's Tavern.

News from the war fronts is not in evidence on the front page of the Sun today. The top news from overseas deals with U.S. government pressure on Ireland to give up its neutrality in the war and join the Allies.

Monday, March 13, 1944

Yesterday when I returned from Clinton Labs I gave Ralph James the PuO_2Cl_2 solution I brought back with me. Today he told me that he prepared two new samples from it by first precipitating $NaPuO_2Ac_3$, then making the supernatant alkaline and precipitating $Pu(OH)_4$, thereby separating Pu(IV) and Pu(VI). Each of the precipitates was dried and sealed in a soft glass ampule which will now be irradiated in the Clinton pile. The two samples were sent by truck today.

3/13/44 (cont.)

I found a memo from Cunningham, written in response to my request last week, giving the current activities of the Basic Chemistry, Recovery, and Instrument Groups, Sub-section III of Section C-I. They are:

Basic Chemistry Group. Hindman is continuing the investigation of the absorption spectra of plutonium. Kraus is extending his investigation of hydrolysis of Pu^{+4} to 0.1 - 1 molal hydrogen ion concentration in $HClO_4$ solutions. Samples of uranyl and plutonyl sodium acetate salts have been prepared and submitted to Zachariasen for examination; the salts were found to be isomorphous. Smith has prepared samples of $Pu(III)PO_4$ and Pu_2O_3 for examination by Zachariasen. The material was amorphous and no diffraction patterns were obtained. La Chapelle has separated approximately 20 micrograms of 93^{237} from Clinton supernatant solutions. Magnusson has recovered 15 micrograms of 93^{237} from the St. Louis samples. These two samples will be combined, converted to the oxide, and submitted to Zachariasen for structure determination. Patton is working on the composition of 8-hydroxy-quinolate of Pu^{+4}.

Recovery Group. Dawson has found it saves time to make an ether extraction after oxidizing the plutonium obtained by dissolving the first LaF_3 product precipitate.

Instrument Group. Kohman and Weissbourd are working on the BF_3 pressure ionization neutron counters, and Schoenberg is assembling a multiple unit device. Jaffey and Crawford are testing the differential range chamber and ironing out the kinks in the spherical range chamber. Ghiorso is working on the nitrogen high resolving power alpha chamber. Krueger is testing a new magnetic alpha counter received from Clinton and a new design of an alpha contamination detector chamber, the "Treasure Finder." To study the backscattering of alpha particles, Scott is making a series of uranium plates with constant weight and variable spread to determine the variation in backscattering with sample thickness for U_3O_8.

The evening meeting of the Council of my section held last Thursday at 7:45 p.m. in my office was attended by Albaugh, Davidson, Dawson, Ghiorso, Hindman, Jensen, Katzin, Orlemann, Thompson, and Watt.

Willard reported that the present official attitude at the Met Lab is that the Chicago section of the Project will operate for at least 1½ years more. This is based on the presumption that Site W will not be in full operation for another year, and Site Y will require even a longer time for full operation. Remodeling the rooms of Section C-I (new hoods, center benches, etc.) will be roughly in the following order: 2,4,25,26,33,34,35, 36,27,30,32,37, plus some offices, 1,12,11,19A,3,5,6,7,8,9,10,13,16,28,29, 31,41.

In a discussion of reports, Willard stated Section C-I must uphold its reputation for turning out well-written, easily readable reports because of the necessity for selling our work to the men determining Project policy who must form their opinions on the basis of very cursory examination of a large number of written reports. There was considerable feeling among those present that group leaders and assistant section chiefs spend more time in report writing than in productive work. Thompson advocated consolidation of reports, pointing out that in his subsection alone, about thirteen reports per month are required.

3/13/44 (cont.)

It was announced that on about March 15, the lab will receive 1.3 to 1.4 g of plutonium, of which one gram will go to Fermi (for neutron yield measurements) for about one month, after which we will get it back. The remaining 300 to 400 mg will be immediately available to us and will be distributed to Thompson (200 mg), Orlemann (125 mg), and Cunningham (25 mg).

The evening meeting of the Research Assistants of Section C-I was also held as usual last Thursday in Room 209, Eckhart Hall.

Last Saturday at Hogness' request, Willard held a meeting with Cunningham, Ghiorso, Katzin, Kirk, and Watt to discuss problems involved in determining the purity of pile-produced plutonium at Hanford, to coordinate the efforts of those in the Division who will work on the problems, and to plan a program for the immediate future. After considering alternate methods of analysis, the group concluded that the most promising approach to the purity assay problem would be to weigh 10 mg samples of the dried, ignited residue from the plutonium nitrate solution on a light-weight counting dish that could be weighed on an Ainsworth balance and then placed in a counter for counting at very low geometry. It was agreed that Kirk will check the reproducibility of pipetting small volumes and also investigate potentiometric titration as an alternate to counting. Ghiorso will design and prepare a suitable low-geometry counter, and Cunningham will work on the technique of spreading 10 mg samples uniformly using uranium as a substitute for plutonium in the preliminary work.

Also on Saturday as the result of difficulties encountered in trying to load a quartz boat with a couple of milligrams of plutonium dioxide for Isabella Karle, Earl Hyde finished developing a protective box that can be used in a hood for safe loading and handling of such boats.

My office received a copy of a March 8 memo from Kircher to Hogness and Stone, transmitting for comment, the design of the batch ether extraction system being assembled in the West Stands semiworks; it can be used to concentrate product and fission products from an aqueous solution of "X" metal prepared in the semiworks dissolver.

I read a copy of a memo, dated March 10, from Zachariasen to Allison in which Zachariasen reports that the sample of sodium plutonyl acetate submitted to him by Cunningham is cubic and isomorphous with the corresponding uranyl compound, having the lattice constant: $a = 10.643 \pm 0.002$ Å. With four molecules per unit cell this gives a calculated density of 2.578 gm/cm^3.

Willard informed Allison by memo of the steps being taken in Section C-I for the assay, storage, and shipment of 250 gram lots of Hanford-produced plutonium (some questions have been raised by Greenewalt). Willard explained that tests of the corrosion of stainless steel by 30% solutions of plutonium in nitric acid will be made when sufficient quantities are available. He stated that the problem of developing a routine method for determining the impurities in the Hanford plutonium is being taken on by Kirk with the cooperation of members of Section C-I. Willard also mentioned that it is possible that alpha-emitting isotopes other than Pu^{239} may be produced

in the Hanford piles and, if this proves to be the case, these will have to be considered in writing up the final specifications for plutonium purity. He added that we are trying to determine whether or not such isotopes will be produced.

Allison wrote to Roger Williams on the subject of the reluctance of physicists to enter the employ of the du Pont organization to work at Hanford. He believes that there are three attitudes of the physicists which contribute to this reluctance: (1) They consider "their discovery of our process to be the greatest achievement of the science of physics, the culmination of three hundred years of research," and that physicists should have a prominent place in directing its development. To enter the du Pont Company in subordinate positions would diminish the prestige of physicists in our program; (2) The younger physicists are now working for physicists of world-wide reputation; there are no such men in the Hanford organization; (3) They feel that being in the employ of the du Pont Company would make it harder to obtain an academic position after the War. Allison suggests that as a compromise, scientific men leaving the Met Lab to work for du Pont should be considered "on loan" and subject to recall by the Met Lab.

My office also received a directive from Moulton on how to write laboratory notebooks in order to satisfy legal requirements.

Edgar F. Westrum, Jr. started work in Section C-I as an Associate Chemist. He has just received his Ph.D. degree from the University of California at Berkeley and is being assigned to Baumbach's group.

Neal Thurman of the du Pont Company and I had a conference to discuss which of my people might go to Hanford. We decided that offers will be made to the following on about April 1: Dreher, Goeckermann, Hoekstra, Howland, Lincoln, Pye, Studier, Thompson, and perhaps Yett. The salary considerations, etc., for Ader, Kohman, and Watt will be discussed further.

The evening seminar of the Chemistry Division was held at 7:45 in Room 251, Ryerson Laboratory. Afterwards Helen and I had pie and coffee at the Thompsons'.

Travel between Britain and Ireland has been suspended. This is an effort to isolate neutral Ireland.

Tuesday, March 14, 1944

Hyde and Davidson mixed 2.5 mg of plutonium, in the form of the dioxide prepared by heating the hydroxide to 70-100° C, with an excess of NH_4Br and treated this with pure gaseous HBr at 730° C. This resulted in the formation of a light green product which dissolved in H_2SO_4 to give a purple solution. Apparently $PuBr_3$ was formed.

My section issued a five-page report entitled "Laboratory Procedure for Bismuth Phosphate Process" (MUC-GTS-533) prepared by Dwight Lincoln.

Thirty-one steps in the process are described, covering the extraction step, two complete oxidation-reduction decontamination cycles, and the crossover lanthanum fluoride step.

The Soviets have captured Kherson, a port on the Black Sea.

Wednesday, March 15, 1944

Alvin Dirksen terminated work at the Met Lab to accept a position with the Phillips Petroleum Company.

I told Kircher, by memo, that we want to use his batch ether extraction system in the West Stands semiworks in about six weeks to work up 60 pounds of uranium metal now undergoing neutron bombardment at the Berkeley cyclotron. The plan is to recover the 93^{237} formed in this irradiation, and it is essential to achieve a concentration first.

In response to Allison's memo of March 1, I summarized Katzin's, Kohman's, and my opinions as to where in Mallinckrodt uranium ore residues it would pay to look for protactinium and ionium. I suggested that the former be sought in the residues from the radium extraction procedures. For ionium, the picture is considered much less clear. I suggested looking in the same residues as mentioned for the protactinium, another possibility being the barium fraction of the eventual barium-radium fractionation. For the ionium which follows the uranium, however, the most likely stage to look for it is in the Mallinckrodt residues. I offered to have these residues assayed for ionium.

I attended the triweekly evening meeting of the Separation Processes Sub-section I of my section at 7:45 in Room 209, Eckhart Hall. Others present were Ader, Albaugh, Arnold, Brody, Christiansen, Dixon, Dreher, Fields, Flox, Gardner, Gerstein, Gilbreath, Goeckermann, Greenlee, Haeckl, Hindman, Hoekstra, Howland, E. K. Hyde, Hyman, Katzin, Kelley, Kirk, La Chapelle, Larson, Manning, Margolis, Morgan, Post, Pye, Rasmussen, Rosenfels, Scott, Sedlet, Sheft, Thompson, Walling, Watt, Willard, Winner, Yett, and others. I asked Thompson to conduct the meeting.

Christiansen of the Technical Division reported on semiworks operations and indicated that the group is now equipped to do metal dissolving, extraction, decontamination, and concentration, starting with a 26-pound batch of metal from Site X. He showed data indicating a considerable portion of the plutonium may be oxidized when the Clinton slugs are dissolved in excess HNO_3. The use of U(IV) is found to be quite satisfactory in the preextraction reduction step. I mentioned that a prereduction step may be unnecessary if excess metal is present in the dissolver, as is true at Site X. I also reiterated the volume reduction advantages of using HCl rather than HNO_3 in the concentration procedure.

Gilbreath discussed lanthanum phosphate as a scavenger in the bismuth phosphate by-product precipitate at reduced acidity (0.1 N). Experiments show a definite improvement in gamma decontamination. Greenlee discussed alternate oxidizing agents in place of sodium bismuthate--the only one

which has proved successful so far is lead tetra-acetate. I suggested it might be wise to recheck the use of Ce(IV) as an oxidant. Although Ce(IV) has been unsatisfactory in steel equipment in the laboratory, the scale-up in the plant will reduce the steel surface-to-volume ratio by a factor of 30 which might cut down the concentration of inhibitors sufficiently to allow the oxidation to proceed.

Ader talked about hold-back carriers, covering work with lanthanum, yttrium, and zirconium. Because of poor results, further work will not be continued. Yett reported the results of three concentration runs on a 100-ml scale. There was about a 25% plutonium loss in the lanthanum fluoride-plutonium precipitation that is attributed to the use of 0.2 N HF (the flow sheet specification). Previous runs at 1 N HF give good plutonium carrying. Goeckermann reported on a peroxide isolation process run on a 10-ml scale with Site W concentrations (60 mg) of plutonium which gives an over-all yield of 96% (after the second peroxide precipitation).

Hyman discussed his work on slug coatings. Margolis, reporting on ether extraction studies, indicated poor yields using U(IV) reduction. I suggested he confer with Brody who has been using ether extraction in plutonium purification studies--perhaps a salt of some sort should be added to aid the extraction.

Thursday, March 16, 1944

Today we received a shipment of 1.4 grams of plutonium in the form of solid peroxide from Clinton Laboratories.

Ghiorso's group has now obtained conclusive evidence for the existence of gamma radiation and x-rays from highly purified Clinton-produced plutonium. Crude estimates indicate that the element plutonium emits about one gamma-ray photon per 10^4 alpha particles and one x-ray photon per 100 alpha particles.

In response to my request Orlemann presented me with the manpower assignments for his Sub-section II. The memo reads as follows:

Group IV--Purification and Analysis of Plutonium Compounds
Jensen, Group Leader

Jensen	Supervision, reports. Combination of precipitation and solvent extraction methods for plutonium purification.
Stein	Solvent extraction methods of purification using plutonium stand-ins. Study of light element distribution in systems extracting plutonium.
Brody	Solvent extraction methods of purification of plutonium. Cooperation with Section C-IV on the cupferron extraction method of analysis.
Dixon	Preparation and study of the properties of Pu(IV) acetylacetonate. Preparation of $ThCl_4$ by the reaction of HCl on thorium acetylacetonate in non-aqueous solvents.

3/16/44 (cont.)

Group V--Volatility and General Dry Chemistry of Plutonium Compounds

Davidson, Group Leader

Davidson	Supervision, reports. Liquid phase chlorination of PuO_2 in pressure bombs.
Hagemann	Halogenation of metal and preparation of halide samples for x-ray analysis. Dry box construction. Effusion apparatus for vapor pressure estimation.
J. Katz	Assisting in metal production. Study of wet methods leading to anhydrous halides.
I. Karle	Vapor phase reaction of CCl_4 with PuO_2.
E. K. Hyde	Vapor phase reaction of HBr with PuO_2 and the NH_4Br bromination of PuO_2.
Wolf	Development of accurate methods for the determination of the composition of plutonium bromides and chlorides on a 1-mg scale.

Group Va--Chemistry of the Plutonium Fluorides

Orlemann, Group Leader

Heath	Supervision, reports. Preparation and study of the higher fluoride(s) of plutonium.
Florin	Chemistry of the lower fluorides of plutonium.
Zvolner	Development of accurate methods for the analysis of fluoride in mg samples of plutonium fluorides.

Group VI--Metal Production

Baumbach, Group Leader

Baumbach	Supervision, reports.
Meyer S. Katz	Preparation of plutonium fluorides for metal production.
Dirksen (to 3/15) Fried	Vapor phase reductions of plutonium fluorides to metal with electropositive elements.
Westrum	Breaking in. To be temporarily assigned to the above problem.
Hellman	Preparation of refractories and study of their reaction with plutonium.
Fried	Determination of the density of plutonium.
Jasaitas	Determination of the melting point of plutonium.
Frank	Determination of the hardness of plutonium.
Fried	Determination of the density and structure of PuH_3 in cooperation with Zachariasen.
S. Katz	Preparation of plutonium samples for spectrochemical analysis.

3/16/44 (cont.)

High Vacuum Technique Sub-group

Simpson	Reports. Direction.
Simpson	Vacuum fusion method for oxygen determination.
Phipps Sears	Determination of the vapor pressure of plutonium compounds and metal.
Seifert	Vacuum centrifugal casting and preparation of plutonium metal.

I wrote to Darrell W. Osborne in Arlington, Virginia, asking if his situation has changed in such a way as to make the prospect of his coming with us in the near future any brighter. Osborne was a fellow classmate of mine at UCLA who received his Ph.D. in chemistry at Cal Tech in 1938. At present he is working on a project at George Washington University.

Hogness pointed out to Kimpton the difficulty in keeping qualified glass blowers and proposed that as a first step in rectifying the problem, we increase the wage scale which now ranges between $1.00 and $1.54 an hour. The Chemistry Division now has four glass blowers, but the work load is so great Hogness said we must employ two more.

Asher Margolis attended a conference on the ether extraction process called by the Technical Division. Members of the Technical Division present were C. H. Holt, A. C. Hyde, J. O. Maloney, and J. B. Tepe. This is the first of a series of planned weekly meetings for the purpose of discussing progress on the semiworks and chemical studies of the continuous ether extraction separation process. A review of the previous week's effort was reported. In the description of the progress made in chemical experiments it was stated that the plutonium yield and decontamination obtained during two extraction cycles have been investigated. Low yields of plutonium were observed with a decontamination factor greater than 200. Third cycles could not be carried out because of build-up of UX_1 concentration that interfered with measurement of fission products. Each cycle consisted of an ether extraction in the oxidized state, followed by an aqueous extraction in the reduced state.

Our report "Chemical Research-Separation Processes for Plutonium" was issued for the month ending March 1, 1944 (CN-1370). The information in this report is essentially the same as that in the memo I sent to Hogness Wednesday of last week and is as follows: G. Johnson, Malm, and Hoekstra have tested the Bismuth Phosphate Process on a 10-ml scale at Hanford concentrations of plutonium and twice the calculated quantities of fission product elements. With the use of a hot, direct strike, a 97% yield is obtained in the extraction step. Use of a hot reverse strike in the product precipitation step of the decontamination cycle results in 50% loss of the product. Hoekstra, Gilbreath, Malm, and Albaugh have studied the conditions for maximum yield in the plutonium precipitation step of the bismuth phosphate decontamination process and verified the deleterious effect of a hot reverse strike at Hanford plutonium concentrations in the absence of fission products; however, if sulfur dioxide is substituted for ferrous ion as the reducing agent, complete carrying is obtained even with the reverse strike. Albaugh

and Gilbreath have studied pre-reduction with uranous ion in 20% UNH solution containing sulfuric acid (the use of sulfuric acid being considered as a means of increasing the acidity of the dissolver solution) and find that plutonium may be completely reduced in a sulfate-complexed UNH solution by uranous ion concentrations as low as 0.003 M.

Dean, Dreher, and Margolis have made preliminary tests of the performance of the bismuth phosphate decontamination process when coupled with an ether extraction step and find that such a coupling step is operable. (This is of interest because the ether extraction process gives considerable concentration of plutonium.) Malm has studied the possibility of precipitation of lanthanum fluoride to carry plutonium from nitric acid solutions of bismuth phosphate as a means of simplifying the present separations process; he finds it to be a promising procedure. Kroner has investigated the effect of cerium(III) holdback carrier on the carrying of radioactive cerium(III) by bismuth phosphate in the absence of uranyl nitrate; indications are that it is not effective in improving decontamination.

Pye, Watt, and Yett have carried out experiments on a 7.5-ml scale to test the Site W concentration flow sheet procedure. They find that a 96% plutonium yield can be obtained using as little as 50 mg of La^{+3}/liter (flowsheet specifies 150) in the lanthanum fluoride product precipitation step. Dissolution of the metathesis product requires a final nitric acid concentration of 2 N rather than 0.16 N as originally planned. Hopkins and Watt have tested the Site W isolation flowsheet process on a 1.0-ml scale and tentatively conclude that (a) plutonium peroxide may be precipitated satisfactorily from 1.0 N nitric acid solutions in the presence of high concentrations of lanthanum, (b) the first peroxide precipitation does not reach substantial completion until 24 hours have elapsed, (c) the first peroxide precipitate is sufficiently pure that adequate washing may provide plutonium of the desired degree of purity without the necessity for a reprecipitation of the peroxide. Preliminary micro scale experiments indicate that the presence of 0.05 M Fe^{+3} inhibits precipitation of plutonium peroxide. Goeckermann and Watt have carried out experiments that indicate that dry plutonium peroxide may be a suitable form for storage and shipment.

Creek, Malm, Margolis, and Dreher have studied the concentration of plutonium from hydrochloric acid. Inasmuch as a considerable concentration of plutonium results from the dissolution of the bismuth phosphate precipitate in hydrochloric acid, it was decided to determine the plutonium yields and decontamination factors that might be expected from the use of lanthanum fluoride and of uranous oxalate when precipitated from a HCl solution of bismuth phosphate obtained after one decontamination cycle. The lanthanum fluoride precipitate gives a plutonium yield of 100% and a gamma decontamination factor of three. The precipitation of uranous oxalate gives a product yield of 97% and a gamma decontamination factor of four.

The evening meeting of the Research Assistants of my section was held at 6:45 in Room 209, Eckhart Hall.

The evening meeting of the Council of my section at 7:45 in my office was attended by Albaugh, Baumbach, Cunningham, Davidson, Dawson, Dreher, Ghiorso, Jensen, Katzin, Manning, Orlemann, Thompson, Watt, Willard, and me. I recommended the following nomenclature be used in referring to the groups of Section C-I:

3/16/44 (cont.)

Section C-I

Chemistry of the Final Products

Sub-section I

Separation Processes

Group I. Extraction-decontamination
Group II. Concentration-isolation
Group III. Process Development

Sub-section II

Purification and Metal Production

Group IV. Purification and Analysis
Group V. Volatility and General Dry Chemistry
Group Va. Fluoride Chemistry
Group VI. Metal Production
(High Vacuum Sub-group)

Sub-section III

Basic Chemistry and Service

Group VII. Basic Chemistry
Group VIII. Recovery
Group IX. Instruments and Physical Measurements

I suggested that the Sub-section Chiefs each appoint a rotating three-member Sub-section Committee, not including group leaders, to help in the administration of the sub-sections.

I then brought up the subject of the control of plutonium and called attention to the necessity to clear all group transfers of plutonium through Dawson. Dawson has been made this section's representative on a safety committee of the Chemistry Division. The question of keeping the lab clean was next discussed--two lab assistants, Martin T. Knowles and John J. Slattery under F. B. Donlan, have now been obtained for special cleaning work. It was pointed out that men wearing rubber gloves should remove them before going to drinking fountains. Careful washing of gloves with soap and water before removing is also recommended as good practice. It was also recommended that smoking and eating in the laboratories stop immediately. Some effort is being made to obtain space for eating. Possibilities are the botany laboratory across the street and the old cooperative eating place on the corner of Ellis and 56th. Baumbach suggested that thin inner gloves be obtained to wear with the rubber gloves, and Manning will look into this.

There followed a discussion of the problem of coordination, during which I pointed out that in the future all records for the x-ray work of Zachariasen should go through Jensen, as well as all samples for spectrographic analysis. The transfer of men to Hanford was mentioned. The present plan calls for Willard to head a group of 30 men, ten of whom are to go from here. I agreed to Cunningham's request that the allotment of plutonium (from the 1.4 gram batch recently received) to the basic chemistry group be increased from 25 to 40 milligrams.

Helen had coffee at Wilma's in the afternoon.

3/16/44 (cont.)

Reports are that U.S. tanks were moving into Cassino in Italy last night. The town is a smoking ruin. Cassino has been the keystone of Germany's defenses in central Italy. A new all-out drive on Rome is now under way.

Friday, March 17, 1944

Ghiorso talked to the General Physics Division about counting methods for milligram quantities of plutonium (a one milligram sample gives off about 10^8 alpha particles per minute) using low geometry counters. Techniques and instruments are being developed in Ghiorso's group as a possible means of determining the purity of Hanford-produced plutonium when weighed out in 10-mg samples routinely.

Philip Fineman, a member of the U.S. Army Special Engineer Detachment (SED), started work in Section C-I. He received a B.S. in chemistry from Berkeley in 1942 and a B.S. in chemical engineering from Virginia Polytechnic Institute this year. Fineman is assigned as a Research Assistant in Dawson's group.

I have decided to visit Berkeley at the end of this month for discussions with Latimer's group in Gilman Hall and with members of the chemistry laboratory, under the direction of Pan Jenkins, in the Radiation Laboratory.

Helen and I had dinner at Morton's Tavern with the Baumbachs.

Reports in the paper today indicate that U.S. troops made another landing on Manus Island on Wednesday morning. Manus is the largest of the Admiralty Islands.

Saturday, March 18, 1944

Janet E. Owen was hired for our secretarial group today.

I replied to Martin Kamen's inquiry of March 3, telling him there has been no further work at Chicago on the half-life of carbon-14 and suggesting we talk, during my forthcoming trip to Berkeley from March 28 to April 2, about the possibility of his making another such determination. With regard to the trip, I mentioned my interest in going through the chemistry laboratories of the Radiation Laboratory.

I next wrote to Jenkins in Berkeley about my plan to visit Berkeley and asked about any additional arrangements, in addition to filling out a V-2 form, that might be necessary to allow me to visit his laboratory and discuss the halides of uranium with Kamen and him.

Helen had coffee at Wilma's.

U.S. troops have captured Lorengau airdrome on Manus Island. The Soviets have reach a position only eleven miles from the Rumanian border.

Sunday, March 19, 1944

The top headline in this morning's Sun reads "Cassino Won by Allies; Blitz Launched at Anzio." The Soviets have cut two big gaps in German lines in the Ukraine. Two U.S. submarines were lost, making a total of 22 lost since Pearl Harbor.

Helen saw the stage show "This is the Army" with her cousin Frances Chilson. It features the original U.S. Army cast, including Captain Ronald Reagan.

Monday, March 20, 1944

Westrum is beginning a series of experiments on the reduction of PuF_4 with barium metal, performed at relatively lower temperatures than have been used recently. The usual type of tungsten wire furnace with beryllium oxide crucible was used. One run was carried out today at 850° C with 0.6-0.9 mg of PuF_4. The product had no visual appearance of metal. A second run at 950° C produced a number of shiny globules of metallic appearing substances. Of the pieces of plutonium metal produced by Westrum, one was given to Zachariasen and another was assayed by Sid Katz and showed 99% purity. The third piece was used by Jasaitis for a melting point determination, who found that it melted when heated rapidly on tantalum to 1,100° C. We are now assured that the melting point of plutonium is not higher than about 1,100° C.

Richard A. Reinhardt started work in Section C-I as a Junior Chemist in Jensen's group at $2850 per year. Reinhardt was an outstanding student at the University of California at Berkeley. Latimer, in his recommendation, noted: "Graduated with highest honors. One of our best B.S. men."

Warren Johnson from Oak Ridge, Spedding from Ames, and Eastman and Latimer from Berkeley are in town to attend the Thomas purification meetings, the Chemistry Information Meeting, etc.

Allison sent me a memo giving information on the form and chemical history of the 1.4 gram sample of plutonium received from Clinton Laboratories last Thursday. He described the chemical treatment prior to shipment as follows: "The bulk of the LaF_3 carrier was removed from the product by metathesis using K_2CO_3, and then dissolved in nitric acid. After an oxidation by silver in the presence of persulfate, the by-product LaF_3 was removed and potassium plutonous fluoride was precipitated. After a second metathesis with KOH, the product was dissolved in nitric acid, and after adjustment of the acidity, made 5% in hydrogen peroxide with subsequent precipitation of the plutonium peroxide. The peroxide was redissolved in nitric acid, the pH adjusted to about 1, and the peroxide again precipitated. It was then centrifuged and dried at 80° C."

Kohman informed Hamilton in Berkeley that within a week he should receive his requested sample of mesothorium in solution.

Willard sent Allison the comments of several of my staff members about the du Pont drawings of the Isolation Building (No. 231) at Hanford.

Allison received a memo from Compton about the transfer and accountings of plutonium shipments. Identical memos went to Whitaker and Hilberry. Compton wrote the following:

"I shall appreciate it if you will call to the attention of the men in your laboratories who have occasion to use product 49, the following principles with regard to its transfer and use:

(1) Product 49 is produced by us on order from the U.S. Government and becomes their property when turned over to the U.S. Engineers acting as their agent at Clinton. From that time forward, the Engineers are accountable for the disposition of the material.

(2) The assignments of this material are made by the U.S. Engineers to the various laboratories that have use for it on the recommendation of Messrs. Oppenheimer, Thomas, and myself.

(3) The director of the laboratory which receives this material is responsible for accounting to the Engineers for its disposition. The manner in which this accounting is to be done at the Metallurgical Laboratory and at the Clinton Laboratories should be worked out in collaboration with Mr. Hilberry.

(4) All transfers of this material from one guarded area to another shall be made only by authorization of the Area Engineer or his representative and is to be accompanied by appropriate receipt forms. Within each guarded area the man in charge of the technical work being done within the area is accountable for its disposition.

"It is hoped that all of our men will cooperate to the fullest extent in avoiding loss of this material, in ensuring its efficient use and in keeping track of its whereabouts. Especially at this time even small amounts of the product are of extreme value in furthering the nation's military operation. It is for the same reason that the greatest care must be taken to avoid any possibility of allowing any of this material to fall in the hands of an enemy agent."

Today, starting at 9:30 a.m., I attended the morning, afternoon, and evening sessions of the March meeting with C. A. Thomas on Final Purification and Metallurgy of Plutonium. Others present were Allison, Ashcraft, Chipman, Eastman, Foster, Franck, Hogness, Jeffries, Potratz, C. S. Smith, Spedding, and Warner.

I was first on the program and gave the Chemistry Division's progress report. It may be summed up as follows:

1. Methods for preparing metallic plutonium. I indicated we are having success in reducing both PuF_3 and PuF_4 when extreme care is taken to eliminate oxygen.

2. Electrolysis. I reported that we have done no work on this method during the month. A discussion ensued, bringing out the fact that this problem has a low priority at Chicago, but high priorities at Site Y and Berkeley.

3. Atomic hydrogen reduction. We discussed our disappointing results, and it was agreed by those present that work on this method should be discontinued.

4. Refractories. Small pieces of uranium metal (ca.1 mg) have been heated to 1,500°C in vacuo in crucibles of thoria, beryllia, and titanium nitride--degassing is important to prevent oxidation. In reply to a question from Smith, I stated there is no evidence of a reaction between plutonium and beryllium oxide to give beryllium when plutonium is melted in contact with beryllium oxide.

5. Physical properties of plutonium metal. We have not yet succeeded in preparing metal finely enough divided to permit ideal x-ray diffraction studies. Our melting point work was reported which indicates a melting point between 850 and 1,150°C. Our hardness values of 62 and 104 (Vickers scale) were reported. I mentioned no work has been done on vapor pressure during the month.

6. Preparation and determination of the properties of plutonium hydrides. Four pieces of barium-reduced plutonium metal (300 micrograms each) formed hydrides at room temperature with purified hydrogen. No additional information has been obtained on the formula of the hydride nor on the equilibrium dissociation pressure as a function of the temperature.

7. Preparation of a pure compound of plutonium. The survey of precipitation methods with plutonium stand-ins was discussed. Solvent extraction methods for Pu(IV) using the following solvents were covered: Methyl-n-amyl ketone, triglycoldichloride, methyl isobutyl carbinol, methyl ethyl ketone + 15% xylene. The unsuccessful effort to extract plutonium(III) into organic solvents was discussed.

8. Volatility methods were then discussed. It was noted that PuF_3 can be prepared by using HF-containing hydrogen. I mentioned that the reaction of PuF_4 and F_2 on a 5-mg scale has yielded a product of much lower volatility than UF_6. I also mentioned the successful preparation and sublimation of stable plutonium(IV) acetylactonate starting with 1 mg of plutonium.

9. Construction of high efficiency neutron counters--BF_3 pressure ion chamber; proportional BF_3 counter.

Chipman discussed the Technical Division's program; Spedding, the Ames Project; Eastman, the Berkeley Project; and Smith, the Site Y Project. Eastman, in his report, said that in the work completed at Berkeley for the period ending March 17, lower sulfides approximating $Th_2U_2S_3$ and Ce_4S_3 have been produced by reduction of ThS_2 with uranium and Ce_2S_3 with cerium, and crucibles have been prepared from them. These are bronzy in appearance, good electrical conductors, and do not vaporize rapidly nor melt below 2,200°C. He said that Berkeley proposes to manufacture crucibles of vacuum-reduced ($ThS_{1.8}$ or $CeS_{1.3}$) and metal-reduced (M_4S_3) types for test purposes.

Smith reported that two attempts at reduction of metal from fluoride have been made at Site Y. In one of them 50 mg of metal as PuF_3 was reduced with lithium in the centrifuge at 1,350°C. 20 mg of product was produced in the form of a good button. This product had a density of 16.9 ± 0.3 gm/cm^3 by the capillary displacement method. Microscopic examination showed that the sample contained approximately 40% of a second phase. He spoke of two experiments that are planned for the immediate future at Site Y: (1) Carrying out a reduction on a 1-gram scale with lithium in the centrifuge and going to better vacuum in remelting and (2) Carrying out an electrolytic reduction on 50 mg scale.

Spedding advised Site Y not to give up attempts at calcium reduction on the 1-gram scale.

At 1:15 p.m. I attended the afternoon session. The same group was present, and was joined later by Compton. A revised flowsheet for final purification and plutonium metal production, prepared by the Chemistry Section C-I, was discussed. The initial steps of the flowsheet are the same as those discussed at last month's meeting (successive precipitations of Pu(IV) molybdate, Pu(IV) oxalate, and an ether extraction cycle). The remaining steps have been changed to provide for precipitation of plutonium peroxide, followed by halogenation to either $PuCl_3$, PuF_3, or $PuBr_3$, with reduction of the halide to metal with calcium in a ThO_2 crucible. Finally, the volatile impurities are removed by their sublimation in vacuo.

Compton gave the results of a preliminary investigation he and Smyth have made on the feasibility of separating light element impurities from Pu^{239} by the electromagnetic separation method. They find it would be a big undertaking--to put 0.6 kg per day through the process, apparatus equivalent to one of the four units of the beta process of the Y-12 program would be required. Although the process seems feasible, it would require considerable development work and might not be ready in 12 months. Smith said that after discussion of the subject with others at Site Y, it was concluded that the method should be considered only as a last resort.

The formation of a group under Willard to study the whole problem of specifications and assay for plutonium was mentioned.

At 8:30 p.m. I attended the evening session of the Thomas meeting, along with the others previously named. The discussions and assignments were summarized. The general priority assignments for my section remain unchanged except as follows:

1. Work on atomic hydrogen reductions as a possible means of metal production will be discontinued.

2. A careful study will be made of the conditions for producing PuF_4 and PuF_3, including study of the equilibrium

$$PuF_3 + HF \rightleftharpoons PuF_4 + 1/2\ H_2$$

3. Vapor pressure measurements of uranium, plutonium and their compounds. Hogness and Chipman are to discuss the most urgently needed measurements and make assignments.

The "Chemistry Division Summary Report for February 1944" (CN-1471) was issued and covers the work of the six sections. The introductory note explains that up to the end of February the monthly summary reports of the Chemistry Division have been prepared principally for the Project Council Information Meeting with limited distribution. The Information Office now believes these reports should be given wider circulation and issued as CS reports. This report, then, initiates this new policy. The introduction mentions that the laboratory space made available by the reduction of academic personnel in Burton's Section C-II is being allotted to my Section C-I, and that the whole area of the New Chemistry Building is now undergoing extensive reconstruction to minimize both the health hazard and the dust contamination of plutonium. It also states, "The liaison on Hanford problems between the Clinton Laboratories and the Chemistry Division is effected by informal monthly meetings at Clinton which are usually attended by R. L. Doan, W. C. Johnson, O. H. Greager, W. C. Kay, L. Squires, G. T. Seaborg, and T. R. Hogness. The work at the various laboratories on Y problems is coordinated by C. A. Thomas and J. C. Warner largely through monthly meetings held in Chicago. The liaison on Basic and Fission Product Chemistry is effected by personal contact between the representatives of the various research units."

With respect to Section C-I, the report summarizes the status of the following problems:

1. Hanford metal coatings. Removal of aluminum-silicon bonded jacket. Effects of coating materials on the separations process.

2. Bismuth Phosphate Process Research. Conditions for maximum yield in the plutonium precipitation steps of the Bismuth Phosphate Process--Site W concentrations of plutonium.

3. Bismuth Phosphate Extraction and Decontamination. 10-cc scale runs--Site W concentration of plutonium. 100-cc scale runs--Site W concentrations of plutonium.

4. Bismuth Phosphate Process Research - Decontamination Studies. Holdback carriers. Lanthanum phosphate by-product precipitation.

5. Bismuth Phosphate Process Research - New Oxidizing Agents.

6. U^{+4} as a Reducing Agent. Pre-reduction by U^{+4}. The use of U^{+4} as a reducing agent in the decontamination cycle.

7. Site W Concentration Process. Experiments on 7.5-ml scale. Experiments on 100-ml scale. Plutonium yield as a function of the carrier-product ratio. Dissolution of metathesis products.

8. Isolation at Site W - Peroxide Method. Precipitation of plutonium peroxide from 0.5 N HNO_3. Influence of variations in the mode of addition and the quantity of hydrogen peroxide. Precipitation of plutonium peroxide at different temperatures. Precipitation of plutonium peroxide in the presence of iron. Dissolution of dry plutonium peroxide in concentrated HNO_3.

9. Survey of Solvent Extraction Methods for Plutonium Purification.

10. Determination of the Properties of Pu(IV) Acetylacetonate.

11. Purification of Th(IV) and U(IV) by Precipitation Methods.

12. Preparation and Properties of PuF_3 and PuF_4.

13. Properties of Volatile Plutonium Higher Fluoride.

14. Properties of Plutonium Halides. Halogenation of Plutonium metal. Vapor phase chlorination of PuO_2. Hydrobromination of PuO_2.

15. Reduction of Plutonium Compounds to Metal.

16. Electrolytic Metal Production.

17. Atomic Hydrogen Reduction of Plutonium.

18. Refractory Studies.

19. Plutonium Density Determinations. X-ray diffraction method. Direct capillary method.

20. Oxidation Potentials of Plutonium Couples.

21. Polarographic Study of Plutonium Oxidation - Reduction.

22. Hydrolysis of Pu^{+4} and Pu^{+3}.

23. Organic Derivatives of Plutonium.

The evening seminar of the Chemistry Division met as usual at 7:45 in Room 251, Ryerson Laboratory at which Potratz summarized the present status of analytical methods and tolerance methods.

Helen worked today at home on the "Table of Isotopes."

The Soviets have entered Rumania along a 31-mile front. From the Pacific comes word that all of the important parts of the Admiralty Islands have fallen to U.S. troops.

Tuesday, March 21, 1944

Another laboratory assistant was hired today for the Donlan group--Aquilla Parsons.

Our sodium plutonyl acetate sample is scheduled today to be placed in the Clinton pile for neutron irradiation. The purpose is to look for Pu^{240} concentrated by the Szilard-Chalmers mechanism.

Cliff Smith began today the preparation of a one-gram sample of PuO_2 mixed with 2 grams of carbon for Fermi. His starting material is 1 1/2 grams of plutonium as the nitrate in 20 ml of 1 N HNO_3. Dawson previously carried out two peroxide precipitations in order to remove as much of the Np^{237} as possible. Smith carried out two additional peroxide precipitations and saved the supernatants for 93^{237} preparation and isolation. After dissolving the second peroxide precipitate in nitric acid, he returned 0.462 grams of the plutonium to Dawson.

At 9:30 a.m. I attended a Project Council Information Meeting on Chemistry. Others present were Allison, Ashcraft, Burton, V. Bush, Cannon, Chapman, Chipman, Compton, C. M. Cooper, Coryell, Dempster, Doan, Eastman, Fermi, Franck, Greager, Hiskey, Hogness, Jeffries, W. C. Johnson, Kay, Latimer, Mulliken, Perlman, Potratz, Cyril Smith, D. M. Smith, Spedding, Stone, Sugarman, Sutton, Szilard, Thomas, Vernon, Warner, W. W. Watson, Wigner, V. C. Wilson, and Zachariasen.

I was first on the program and described the Section C-I work on Hanford problems directed by Thompson, Los Alamos problems by Orlemann, and basic chemistry by Cunningham. In connection with the discussion of the Los Alamos related work, I mentioned that yesterday Westrum produced some plutonium metal by reduction of PuF_4 by barium in a beryllium oxide crucible. One of the three metal pieces melted when heated rapidly on tantalum to 1,100° C. It now seems safe to say that the melting point of plutonium metal is lower than 1,100° C. I also said that solvent extraction methods for purification of plutonium continue to show promise as a means of purification.

In connection with the discussion of basic chemistry, I mentioned the work on determining the potential of the Pu^{+3} - Pu^{+4} couple and the study of absorption spectra for the +3, +4, and +6 valence states of plutonium. I also mentioned that during the past week conclusive evidence has been obtained for the existence of gamma radiation and x-rays from plutonium--one gamma ray photon per 10^4 alpha particles, one x-ray photon per 100 alpha particles. I said the inference could be drawn that there are two groups of alpha particles differing by a certain amount in energy, and the x-rays may come following internal conversion of a low-energy gamma ray, or the few electrons observed could possibly imply decay to some isotope of element 95. Also, other interpretations are possible. In answer to a question by Hogness, I replied that these gamma rays could have a decided effect on the decontamination factor when dealing with Site X solutions because one can begin to reach this gamma radiation level with a decontamination factor of 10^5, whereas we are looking for a decontamination factor of 10^7. It should not be troublesome with actual Hanford solutions because this gamma radiation level would correspond to a gamma decontamination factor of 10^8 after 60 days shut-down. It may be troublesome in studying the Hanford decontamination process with spiked solutions to simulate Site W concentrations.

Perlman discussed work at Clinton Laboratories on an alternate isolation method involving precipitation of the iodate. Cannon reported on the successful concentration of the fission products from one 35-megawatt-day slug by remotely controlled continuous ether extraction, ending up with 80 ml of solution which possesses 1/2 to 1 times Site W concentration of activity. The solution has strong fluorescence, and its radiation renders glass almost opaque in 4 to 5 days. I requested that some of the oxide extraction residue be sent to Chicago for use in the development of shot-gun methods.

Kay summarized the Clinton Separations Plant results, indicating the accumulative plutonium yield has gradually risen from an initial 40% to 75%, and the average decontamination factors being attained through Room D are 10^6 to 10^7. In answer to Cooper's question as to what could

be expected as a final steady state yield, Kay responded, "You can't safely extrapolate from the present data, as doing so would predict a yield of 105% by June!"

Cyril Smith reviewed the results of two attempts at plutonium metal production at Los Alamos. In one case a bomb reduction of a one-gram sample of plutonium as PuF_4 failed. In the other case, 50 mg of plutonium in the form of PuF_3 was reduced with lithium at 1,350° in a centrifuge to produce a good metallic button with a density of 16.9 ± 0.3 gm/cm^3 as determined by the capillary displacement method. The product was not pure metal and appeared to contain about 40% of a second phase. I asked whether the two phases might be two forms of metal, but Smith thought not, because there is evidence that one phase was formed in a peritectic reaction.

Herman H. Fussler in Mulliken's office, sent me a memo dated March 21 saying he has learned of my practice of sending carbon copies of my monthly reports directly to Perlman in Oak Ridge. K. H. Fagerhaugh there feels this is an undesirable practice since it involves an unidentified copy of a regularly issued report. He suggests regularizing the procedure by arranging for the Information Office to transmit directly to Perlman a copy of any report issued by my section; in this way the report will have the proper report number and the distribution list will be correct.

"Hanford Separation Process Flowsheet. Bismuth Phosphate Process" (M-CN-1478) was prepared by Katzin last Friday and issued today. This condensed, two-page flowsheet is intended to be a ready reference and represents the status of the process up to the present time. Differences between Hanford and Clinton processes are indicated.

"Chemical Research--Basic Chemistry of Plutonium Report for Month Ending March 1, 1944," was issued as CK-1371. An account of the research covered is as follows: Hindman, Kraus, and Howland have followed spectrophotometrically the stoichiometry of reduction of Pu^{+4} by Sn^{+2} and Fe^{+2} and of oxidation of Pu^{+3} by Ce^{+4}. The results confirm the conclusion given in Reports CN-1325 and CN-1360 that the oxidation number of plutonium obtained with these reducing agents is +3. A study also has been made of the absorption spectra of Pu^{+3}, Pu^{+4}, Pu^{+6}, and of U^{+4} and U^{+6} in various acid aqueous media with a view to their possible usefulness for analytical purposes. It is noted that the spectrophotometric evidence obtained to date is in agreement with the hypothesis that plutonium and uranium belong in a series, the members of which resemble each other as do the rare earths. Howland, Kraus, and Hindman also have extended the qualitative earlier experiments (CN-1324) on the potential of the Pu^{+3}-Pu^{+4} couple. At equal concentrations of the two ions the values for the couple found are: E = -0.966 v in 1.0 M HCl; E = -0.75 v in 1.0 M H_2SO_4; E = -0.95 v in 1.0 M $HClO_4$.

Also reported are Watters' polarographic experiments that indicate that the half-wave potential of the Pu^{+3}-Pu^{+4} couple may be shifted by the formation of complex ions of plutonium to the following values: -0.082 v in 0.8 M oxalic acid 0.1 N in H_2SO_4; -0.235 v in 0.18 M oxalic

acid 0.4 N in H_2SO_4; -0.872 v in 9 M NH_4F 0.1 N in H_2SO_4. Kraus has investigated the pH dependence of solubility of Pu^{+4} in 0.01 M Na_2SO_4 over the region pH 2-6. The data indicate that in the region pH 2.5-6, the principal form of the plutonium(IV) ion may be represented symbolically as $Pu(OH)_3^+$. In the region pH 2.0-2.5, $Pu(OH)_2^{++}$ begins to represent an appreciable fraction of the total plutonium. Patton has prepared the following organic derivatives of plutonium in search of reagents which could be of value in purification: plutonium(IV) benzoate, fumarate, m-nitro benzoate, salicylate, pyrogallate, 8-hydroxyquinoline, picrolonate, phenylarsonate, cinnamate; plutonium(III) benzoate, oxalate, picrolonate, and phenylarsonate. Dixon, Cliff Smith, and Hagemann have continued the work reported in Reports CK-932 and CK-1221 on the preparation and properties of plutonium(IV) acetyl-actonate, using the larger (1 mg) amounts of plutonium now available. The melting point is found to be 170°-173°C, and the vapor pressure is found to be $9.7 \pm 0.4 \times 10^{-4}$ mm Hg at 140°C. Stein, Brody, and Orlemann have determined the distribution of Pu^{+4} between 10 M NH_4NO_3 and the following immiscible organic solvents: methyl-n-amyl ketone, methyl-isobutyl ketone, phenyl cellosolve, xylene and isophorone.

Helen worked on the "Table of Isotopes" at home.

Reports of sharp fighting between Hungarian and German troops have reached London. German forces are occupying Hungary in preparation for a last ditch stand against the onrushing Soviet army.

Wednesday, March 22, 1944

Today started as an ordinary day, but by afternoon one of the most exciting discoveries of the Met Lab was revealed. It happened this way. I asked Sidney Katz to come with me to Sherman Fried's lab. I suggested that Fried demonstrate to Katz the method used to determine the density of microgram quantities of plutonium metal so that Katz can make such measurements for us. I then left the room, and Fried told me later in the day what transpired shortly thereafter.

As Fried showed Katz how to weigh a plutonium metal sample of 114 micrograms, just produced by Westrum by the reduction of PuF_4 with barium in a beryllia crucible, he explained that he would inevitably find the density to be 16 gm/cm³. Fried showed him the method for introducing the tiny sample into the micropycnometer. The rise in the meniscus of the liquid caused by displacement of the metal sample was then measured under a microscope. As they went through the arithmetic, it became clear that something was drastically wrong. The density of plutonium was calculated to be about 25 gm/cm³!

Fried, confident that he had somehow made a stupid mistake in the weight or meniscus position, prepared to check the data. Being somewhat embarrassed by the failure of the demonstration, he got Katz to verify his observations. They tried several new recently produced pieces of metal singly and in combination, but the density invariably had a value in the neighborhood of 20 gm/cm³!

Fried told me the situation was a nerve-wracking thing because he was expected to report at the regular triweekly evening meeting of the Purification and Metal Sub-section II in a few hours and would be expected to make reasonable answers to inquiries about the progress of the work in his group. He was afraid that he had made some obvious error which would be pointed out publicly during the seminar.

He and Katz worked until just before the meeting began, and their results were essentially the same as before.

Ralph James began working up a supernatant from the plutonium peroxide isolation step at Clinton in a search for element 95 or 96. La Chapelle has found some peculiar results in performing bromate cycles on these supernatants, and Ralph and I have decided to investigate.

In order to pass on decisions made at the Thomas purification meetings on Monday I held a discussion of purification and metal production problems with Baumbach, Fried, Hagemann, J. Katz, Jensen, Manning, Orlemann, and Simpson. I reviewed the information on the plutonium production work at Site Y, as reported at the Monday meeting with Thomas. The Berkeley people, I said, report production of uranium metal in 90% yield by electrolysis on the 1 gm scale in fused salts consisting of mixtures of sodium, barium, and potassium chlorides containing UCl_3. I set forth the commitments of Section C-I as outlined at that meeting with the following having high priority:

1. Continuation of the study of preparing plutonium metal from fluoride and by other methods.

2. Determination of the density and the melting point of plutonium metal.

3. Plutonium hydride studies.

4. Work on the preparation of a pure plutonium compound by precipitation methods and particularly by solvent extraction methods is to continue and will remain the sole responsibility of our Chicago group. Compton suggested obtaining an experienced man in the field of solvent extraction processing, and it was requested that we draw up specifications for this type of man we feel would be most useful in this connection.

5. The following studies in connection with plutonium hydride are to have very high priority: redetermination of the formula, the determination of the decomposition pressure, and temperature curves, and continued attempts to obtain its density.

6. The synthesis of the halides of plutonium and the determination of their composition and properties is to continue with high priority.

7. A neutron counter status report is to be assembled by Kohman.

8. One gram of pure UO_2Cl_2 is to be prepared for Zachariasen's use by the section. This problem was turned over to J. Katz.

Commitments as to the work in the immediate future for Orlemann's Sub-section II, in particular, were made as follows:

1. The metal production group will undertake the following: the determination of another pycnometric density, reduction to metal in tantalum, the determination of the melting point on a sample which is radioactively and spectrochemically analyzed before and after the determination. The studies on the hydride will be carried out as soon as possible.

2. In order to avoid the bottleneck in the preparation of fluoride for metal reduction and to implement studies of alternate methods of preparing the fluoride, all the fluoride work will be concentrated in Heath's unit. The 12 mg of oxide available will be converted to PuF_4 immediately. The 8 mg of oxide now available will be used to test other methods of preparing the fluoride. No decision was reached as to which of the alternatives here should be tried first.

In answer to a question by Baumbach as to what disposition of the available metal should be made at this time, I suggested the following division: 1/2 to be used for capillary determination of densities, the remaining 1/2 to be divided in the following way; 1/2 to melting point, 1/4 to hydride, and 1/4 to the preparation of halides by Hagemann.

Kircher of the Technical Division discussed with me Potratz's need for small quantities of semiworks-dissolved Clinton active solution, all such materials currently being charged to Section C-I. I agreed to release 3-4 liters of 40% UNH solution to Potratz every ten days for use by E. R. Russell.

I attended the meeting of our Purification and Metal Production Sub-section II at 7:45 p.m. in Room 209, Eckhart Hall, with Arnold, Fried, Fred, Hellman, Jasaitis, J. J. Katz, Langham, Orlemann, Potratz, Simpson, Westrum, and others. After my opening remarks the meeting was run by Orlemann. I summarized the recent work at Site Y relating to the preparation of plutonium metal, mentioning the one large reduction of one gram, which yielded what would appear to be mostly PuO. Fred and Langham described their work on analytical procedures. Simpson reported that the apparatus for determination of oxygen is in the process of construction and that a vacuum of 4×10^{-8} mm has been reached and a temperature of 2,278° C attained. It was agreed that after the apparatus has been tested on known oxides, the first analysis will be of the so-called lower plutonium oxide. Hellman reviewed work on testing refractories (ThO_2, Al_2O_3, MgO, BeO, TiN, ThS_2, CeS_2) for reductions and remeltings using uranium as a stand-in for plutonium. Westrum reported on three plutonium reduction runs in beryllium oxide crucibles using barium, at 800° C, 950° C and 1,150° C, respectively. The two higher-temperature runs yielded globules of what appear to be metal.

Jasaitis reviewed his experiments to determine the melting point of plutonium using a tantalum surface for carrying the plutonium; behavior of different batches of metal has been variable, but he is reasonably certain that a maximum value for the melting point can be set at 1,100° C.

When it came time for Fried to report, he cited his new density measurements by the Kirk capillary technique on the four particles of metal produced by Westrum by barium reduction in beryllium oxide crucibles. After Fried stated that the particles ranged from 116 to 190 micrograms

in weight and density measurements on three of the four samples gave values from 19.3 to 22.0, there was a great deal of excitement and, I think, some skepticism in certain quarters. These high densities that he measured are considered outside experimental error and can very well be associated with either a difference in structure or with the presence of other phases that arise from different temperature conditions during reduction. The fact that plutonium metal is of a higher density than known heretofore means that production time at Hanford need not be so long (as estimated on the basis of the lower density) to give enough material for our first plutonium bomb.

I mentioned the finding of Ghiorso and co-workers of the existence of a plutonium gamma and an x-ray. There is still no unambiguous interpretation of these phenomena. They may indicate a branching of the chain with the formation of 95 or may be associated with the decay to U^{235}.

Helen had coffee at Wilma's today.

American marines have seized Emirau Island in the St. Matthias group. Emirau is only 540 miles south of Truk.

The Project Council Policy Meeting held today at 9:30 a.m. in Room 209, Eckhart Hall, was attended by Allison, Chapman, Chipman, Compton, C. M. Cooper, Dempster, Doan, Eastman, Fermi, Franck, Greager, Hilberry, Hogness, Jeffries, W. C. Johnson, Kay, Mulliken, Don Smith, Spedding, Stone, Vernon, Warner, C. J. Watson, W. W. Watson, and Wigner.

Compton gave his usual "State of the Nation" introduction. He talked about circulating a statement summing up the proposed future program for our Project and asked that all concerned comment on it. Written comments should be in his hands by next Wednesday. He then read the statement, which covered the following fields: (1) Support of Hanford production, (2) Cooperation with Los Alamos, and (3) Investigations of new possibilities. Conant and Bush, he said, hope there will be published a statement of the scientific and technical basis of the Project. The comments especially needed are those giving manpower requirements; where possible, these should be manpower in specific fields as of 1944 and forecasts for 1945. Compton further said that he will not transmit the forecasts beyond the Project, but he needs them for preparing the final document.

Hilberry reported on his recent Hanford Engineering Works trip. Among other things, he noted, the building which will house the first pile is essentially completed; also the Chemical Separations building is nearly ready for roofing and the cells are up to the level where the piping goes in.

Hogness presented the Chemistry Division report for the Met Lab, including our manpower distribution of effort. He noted the new ventilation system for the New Chemistry Building will be ready about May 1. Just then Burton and Seitz came in, and Hogness asked Burton to present his program for further graphite work. In substance, it calls for a study of graphite disease, comparing various sources

of graphite and indicating phases of programs still requiring study, and ways to get the information. Compton was concerned about whether or not the proposed work on graphite, especially that at Site X, can be carried out with present personnel. Burton replied in the affirmative, if he can count on help from two men in the Radiation Section at Clinton Laboratories to help with the work there. Johnson reported on the Clinton Chemistry Division programs indicating that emphasis is now shifting to (1) Hanford needs; (2) fundamental studies of the Bismuth Phosphate Process; and (3) training program.

Compton raised the question of relative emphasis on analyses for low atomic number elements versus fission product analyses. After a discussion with Greager, Hogness, Doan, and Franck, Compton expressed the feeling of the Council that the most urgent phase is at present the analyses for low atomic number elements.

Eastman was asked to report on the program at Berkeley. He said that it is essentially unchanged. Of the 32 or 33 men employed, their time is divided among (1) Extraction and decontamination, (2) Fundamental chemistry of 94 and (3) Production and purification. Kay reported on Clinton pile operations and indicated that the average power level was raised recently to 825 kw, and around 72 grams of plutonium now have been made. Compton closed the meeting by emphasizing the need to avoid all indication of the connection between Chicago, Clinton, and Sites Y, and W. The meeting adjourned at 1:30 p.m.

Thursday, March 23, 1944

With the knowledge of our exciting new result on the high density form of plutonium, Hogness telephoned Los Alamos to give them the good news. I am sure they are as jubilant about it as we are.

Westrum has completed the series of experiments on the reduction of PuF_4 with barium in beryllium oxide crucibles at relatively lower temperatures than used recently. He found on Monday that a run at 950° C with 0.6 - 0.9 mg PuF_4 produced minute shiny globules of metallic-appearing substances. Two subsequent runs at 1,150° C with the same size samples gave larger metallic-appearing spheres ranging from 100 to 320 micrograms. Some of these samples were used for the recent exciting density determinations; in all cases the density continues to be much higher than previously measured and is in the vicinity of 20 gm/cm^3.

Sigfred Peterson whom Latimer highly recommended started work in Section C-I. He has been a graduate student at the University of Minnesota, after graduating with highest honors from the University of California in 1940. He will be working as a Junior Chemist in Albaugh's group. Another laboratory assistant, Elsie Mae Freeman, was hired to work in the Donlan group.

I wired Hamilton in Berkeley that the solution he requested yesterday will arrive Saturday morning on the streamliner with Eastmen and that,

if possible, he should have a station wagon meet the train. I also sent a wire to Kennedy in Los Alamos saying that the plutonium gamma rays are of much lower energy than I told Cyril Smith when he was here--the energy is down in the x-ray region.

A "Status Report on U^{233} Program" has been written by Leonard Katzin and was submitted for distribution as a Laboratory Report today. Part I covers the discovery of U^{233}, its properties and possible advantages over Pu^{239}, and the present status of its isolation. Part II is concerned with the preparation of U^{233} and Part III summarizes the chemical extraction of U^{233} indirectly, in which Pa^{233} is the intermediate, and directly.

Dreher attended the second weekly conference on the ether extraction process with George Bird, E. B. Christiansen, A. C. Hyde, Kircher, Maloney, and Tepe. It was reported that major efforts are being expended on finding a suitable reducing agent for use in the aqueous extraction step of the process. During the past week the first attempt was made to operate the continuous ether extraction system with tracer solutions, and a number of mechanical problems were identified.

"Chemistry Division Problem Assignments" was issued by Hogness as a compilation of one-page descriptions of each project now underway. My section's assignments are grouped according to the subjects "W Process Studies," "Y Process Studies," "Basic Chemistry, Recovery, and Instruments," and "Miscellaneous Studies."

The evening meeting of the Research Assistants of my section met at 6:45 in Room 209, Eckhart Hall.

The regular Thursday evening meeting held in Room 133, Eckhart Hall, was a meeting for broad attendance. Moulton discussed patents, and Wigner gave a talk "How We Came to the Present Use of Our Materials." I did not go because at the same time I was holding the regular evening meeting of the Council of my section in my office with Baumbach, Cunningham, Davidson, Ghiorso, Hindman, Jensen, Manning, Orlemann, Watt, and Willard.

I opened the meeting by pointing out that any requests for alterations in our laboratory rooms should be made now as changes after completion of the building alteration program will be difficult to obtain. Arnold has suggested that Donlan might be given a position similar to that held by Lange as Lange is to be stationed at the Armory. In this event, Kathleen Gavin would take charge of the order records for our section. Manning noted that Donlan's present draft classification would necessitate advancement or his probable loss. It was suggested that Arthur LeFeuvre, the lab technician in Ghiorso's group, be put in charge of the woodshop after Reid Harding leaves. Manning is to check the possibility that a place for the shop may be found in the new building (New Chem Annex).

The allocation of the recently received 1.4 gram sample of plutonium was discussed. The approximately 400 mg we are to receive immediately will be distributed among Albaugh (200 mg); Baumbach (75 mg); Davidson, Jensen, Heath (25 mg each); Hindman (50 mg); and Ghiorso (10 mg). The one-gram

sample (first purified by us for Fermi) will be distributed, after return by Fermi, to Albaugh (200 mg), Baumbach (400 mg), Davidson (75 mg), Jensen (75 mg), Heath (50 mg), and Hindman (100 mg). Manning reported that the Ceramics Group at Site B is to move to M.I.T. Baumbach reported that the density value of the newly-prepared plutonium metal has been checked and is definitely above 20 gm/cm^3. In this connection it was noted that the metal obtained from beryllia crucibles is not the same as that obtained from thoria crucibles; this is the key to our recent success in obtaining samples of high density plutonium. Jensen suggests that beryllium may be causing the hardening recently observed in our plutonium samples.

Helen had coffee at Wilma's.

The Germans have set up a puppet rule in Hungary under Doeme Sztojay, a quisling premier.

Friday, March 24, 1944

Ralph James prepared a sample of about 15 mg plutonium on a gold target backing plate for alpha particle bombardment at Berkeley.

Willard, Thompson, and Cunningham met to plan an investigation of the basic chemistry of plutonium under experimental conditions that approximate as closely as possible the operating conditions of the Site W flowsheet. The following program was decided upon for the next two weeks:

1. Smith will prepare, for structure determination by Zachariasen, LaF_3 containing Pu(III), LaF_3 containing Pu(IV), the double salt of LaF_3 - PuF_4, crystalline samples of Pu(III) phosphate and plutonium(IV) phosphate, bismuth phosphate containing co-deposited Pu(III), bismuth phosphate containing co-deposited plutonium(IV), Pu(VI) fluoride, and Pu(VI) phosphate.

2. O'Connor and Kraus will make solubility measurements of the phosphates, fluorides, and hydroxides of Pu(III) and Pu(IV) under process conditions through the fluoride crossover step.

3. Howland will make potential measurements of plutonium couples in process solutions.

4. Hindman will identify the valence states of plutonium obtained in the various process steps by means of spectrophotometric examinations. Howland will identify valence states in precipitates by potentiometric titration.

5. Lincoln will investigate the separation of neptunium from plutonium by the bismuth phosphate method.

6. Hindman, if time permits, will determine rates of oxidation and reduction spectrophotometrically.

"Chemical Research--Special Chemistry of Plutonium. Report for Month Ending March 1, 1944," (CK-1372) was issued. Stein, Jensen, and

Orlemann have continued the study of purification methods using thorium and uranium as stand-ins for plutonium. Florin and Heath have studied the conditions that determine whether PuF_3 (black or purple) or PuF_4 (pale brown) is formed on treatment of PuO_2 with anhydrous HF. They believe that the most reasonable interpretation of their work is that H_2 in the HF has been responsible for the conversion of PuO_2 to PuF_3 rather than to PuF_4. Meyer, Florin, and S. Katz studied the weight changes during hydrofluorination of PuO_2 samples, but find them difficult to interpret.

Hagemann has prepared plutonium chloride (blue in color) by treating plutonium metal with chlorine at 450° C. X-ray and chemical analyses indicate the composition to be $PuCl_3$. I. Karle, J. J. Katz, and E. K. Hyde have studied various chlorinating agents for the synthesis of plutonium chloride from PuO_2 and find that CCl_4 vapor at 750° C is an effective reagent. J. Katz and Hyde have further investigated the conversion of plutonium oxide to bromide and find that treatment with HBr at 600° C seems to produce $PuBr_3$. The substance sublimes in a stream of HBr at 750°-800° C. Davidson reviews the possibilities of converting a plutonium oxide, perhaps the peroxide, to a chloride using a liquid phase chlorinating agent such as PCl_5 or CCl_4 in a high pressure, bomb tube reaction.

Baumbach presents a chart summarizing the compounds that have been produced during attempts to make plutonium metal and indicating how they were made. Meyer and S. Katz review the procedures followed in preparing pure plutonium oxide for use in preparing PuF_4 and thence plutonium metal. Fried and Dirksen describe the improved techniques that resulted in a greatly increased number of satisfactory reductions. Major changes are the degassing of the unloaded crucible system, filling with argon before opening the system to the atmosphere, and placing the reducing agent above rather than below the inner crucible. The use of degassed crucibles has permitted repeated reloading of the inner crucible without removing the metal already present; subsequent firing gives excellent agglomeration of the metal so that larger single pieces can be obtained. Since adopting these improvements all reductions of PuF_4 with calcium and barium at 1,600° C have been successful in yielding plutonium metal. Also, the purple plutonium fluoride, now believed to be PuF_3, has been successfully reduced to metal with both calcium and barium at 1,600° C.

Dirksen and Fried have attempted unsuccessfully to prepare finely divided plutonium metal for x-ray examination by lower temperature reductions of PuF_4 and PuO_2. Fried and Jasaitis have measured the density of a 35.85 microgram sample of plutonium metal using a capillary pycnometer method devised by Kirk and obtained a value of 16.2 gm/cm^3. Six metal samples from 20 to 200 micrograms prepared by calcium, barium, and lithium reduction were analyzed by x-ray diffraction by Zachariasen. The four samples produced by calcium reduction show the cubic structure with a density value of 16.4 ± 0.5 gm/cm^3. The lithium-reduced plutonium and the barium-reduced plutonium have a complex structure which cannot be worked out from the present photographs. Since the calcium-reduced plutonium is of low purity (80-86%) and the barium-reduced plutonium is quite pure (96-99%), it is probable that the cubic structure is characteristic of an alloy, whereas the complex structure is characteristic of the pure metal.

Jasaitis reviews the malleability of plutonium metal and observes that it flattens quite easily much like annealed copper. Plutonium metal does not tarnish readily beyond the surface probably because of a protective oxide or nitride film. Jasaitis has studied the melting point of plutonium metal by heating 50 microgram pieces in platinum cylinders in vacuo. They have melted and formed alloys at about 850° C. Observations made during reductions of PuF_4 with lithium suggest that the melting point of plutonium is no higher than 950-1,000° C. Frank has measured the hardness of plutonium metal using an Eberbach Micro Hardness Tester and, using four metal samples of from 12 to 78 micrograms, obtains Vickers hardness values of from 88.5 to 153. Zvolner and S. Katz have continued the study of the electrolysis of uranium from fused salts on a micro scale without success. As yet, no experiments have been tried with plutonium. Hellman, Gerstein, and Frank have constructed and tested (with uranium) an apparatus for evaluating possible refractories to be used in the production of plutonium metal.

Helen had coffee with Wilma this afternoon.

A new Japanese army dramatically appearing out of the jungle has driven 60 miles into India. This is the second invasion within 48 hours. The new force is within 30 miles of the key city of Imphal.

Saturday, March 25, 1944

Pearline Boykin began working today as a technician in Dreher's group.

Cliff Smith completed the preparation of the PuO_2-graphite sample for Fermi. The sample consists of a mixture of 1.2 grams of PuO_2 (1.0 gm of plutonium) and 2.0 grams of carbon contained in an aluminum capsule.

Using the sample prepared by Smith, Kohman made a direct determination of the yield of neutrons from the bombardment of light elements (oxygen and carbon) by plutonium alpha particles. Measurements were made of the neutron emission rate with a BF_3-filled ionization chamber in a large mass of paraffin and also with a counter consisting of four BF_3-filled proportional counter tubes in a paraffin mass. He arrives at a counting rate value of 5,000 ± 1,000 neutrons per minute. Because of the large size of the PuO_2 particles (up to 200 microns), quantitative information about the carbon (α, n) yield cannot be obtained.

Willard received a copy of a letter from Johnson (Personnel) to H. S. Wade in San Francisco asking that Cliff Smith's leave of absence from Wade's company (Owens-Illinois Pacific Coast Company) be extended until September of this year.

Allison wrote to Greenewalt quoting the text of Willard's memo to him of March 13 concerning investigations planned for the assay, storage, and shipment of Hanford product.

3/25/44 (cont.)

At 5:00 p.m. I left Chicago on the Santa Fe Super Chief for Los Angeles. My ultimate destination is Berkeley.

This morning's paper brings news that the Soviet Union's first Ukranian army has broken through to the Dniester River. Allies have pried the Japanese loose from three positions in India; all are positions near Imphal, an important Allies communications base.

Sunday, March 26, 1944

En route to Los Angeles.

Monday, March 27, 1944

I arrived in Los Angeles at 10:15 a.m. and was met by Mother and Jeanette. I spent the day with my parents at 9237 San Antonio Avenue, South Gate and had potato sausage for lunch and dinner. Helen's mother came for dinner; Clayton and Rita Sheldon (friends from grammar and high school days) came over after dinner; then all of them accompanied me to the train. At 9:00 p.m. I left Los Angeles for San Francisco on the Southern Pacific Lark in Roomette 5, Car 3.

Tuesday, March 28, 1944

I arrived in San Francisco in the morning and went to Berkeley where I visited the Department of Chemistry. I discussed with Calvin his method of preparing what is possibly anhydrous UCl_4 by passing HCl gas into an anhydrous solution of U^{+4} acetyl-acetonate in benzene. We also talked about his preparation of U(IV) complexes.

During the day I talked by phone with Willard, Baumbach, Ghiorso, James, and others in Chicago to go over the progress of their work.

Melvin and Genevieve Calvin and I had dinner at Eastman's home; I am staying with the Calvins.

Wednesday, March 29, 1944

I visited the Radiation Laboratory and talked with Kamen and saw Paul Aebersold. I was shown through the various chemical laboratories by Kamen and discussed the chemical work there with Prescott, Altman, Holmes, and others. They are doing a great deal of work on the halides of uranium. Perhaps of greatest interest to us is the work on the thermal decomposition of UI_4 to metallic uranium.

I had dinner with the Calvins at Tiny's in Oakland--fried chicken now costs $1.10. After dinner I was the sole speaker at a meeting of

Latimer's and Eastman's groups. I described our Chicago Section C-I work. Then I again spent the night at Calvins'.

Thursday, March 30, 1944

I spent much time with the members of Latimer's group in Gilman Hall discussing their research program.

I talked with Gofman, Howard Crandall, and W. H. Reas about their study of a new beta particle and gamma-ray-emitting activity in pile uranium which does not seem to decontaminate by their "oxalate" method. They believe it to be an isotope of element 95. However, I wonder whether the activity might be element 61 or some mixture of isotopes.

I played golf with Melvin Calvin, had dinner at Joe and Leah Hamilton's home in San Francisco, then returned to Berkeley again to spend the night with the Calvins.

Friday, March 31, 1944

I continued my conversations with the Gilman Hall group.

Kamen and I discussed the Berkeley work on research and production of the chlorides of uranium, using UCl_4 as the process gas. They have found some evidence for the production of UCl_2 during some of their experiments. Kamen also described their procedure for purification of uranium from impurities by electrolyzing the impurities away. I also learned of the special alpha ionization chamber built by Frederickson that enables one to count alpha-particles over a beta-particle background of about one millicurie.

I had a meeting with C. D. Shane regarding possible patent rights covering the discovery and use of element 94. I explained that my personal position is that any money derived from the assignment of the patent rights to the Government would be given to the University with restrictions to ensure its being devoted to research in nuclear work, half in the Radiation Laboratory and half in the Department of Chemistry.

I again spent the night at the Calvins'.

Saturday, April 1, 1944

I had discussions with Leo Brewer, Norman Lofgren, and LeRoy Bromley about the preparation of refractories using the sulfides of cerium and thorium, a rather large program being underway. They gave me several items to take back to Chicago to test with plutonium including a crucible of "cerium brass," a crucible of "vacuum heated" cerium sulfide, a crucible of "vacuum heated" thorium sulfide, and a piece of silver-colored $ThS_{0.8}$. Brewer mentioned the titles of a number of good metallurgy books and named the best commercial sources for a number of metals and refractories.

I called Art Wahl at Los Alamos in the morning to tell him what I have suggested to Shane about the patent matter. I again talked with Shane, who believes that Bush now wants to be arbitrator rather than trustee. Kennedy called me back during the afternoon and told me that he and Art favor my proposal and that he feels that Segrè (who is away at Site X) would also. I told him of Shane's news regarding the position of Bush, but I agreed to see if Bush would be trustee in the new arrangement, while Kennedy will bring with him positive expressions of opinion of Wahl and Segrè, plus power of attorney from them, when he comes to Chicago this month. We may have to see Captain Lavender in Washington.

I had dinner at the Latimers' and spent the night at the Calvins'.

Sunday, April 2, 1944

Melvin Calvin and I played golf on the Tilden Park golf course. At 4:30 p.m. I caught the "City of San Francisco" to Chicago. During my train ride I recorded the following things to do.

1. Look for 93^{235} + 93^{236} (beta and alpha particles) from Berkeley uranium plus deuteron bombardment.

2. Look for 94^{240} (alpha) from U + He (Berkeley bombardment). Also 94^{241} (betas and alphas) — 95^{241} alphas.

3. Look for 95^{242} (alphas and betas) and 96 (alpha) from 94^{239} + He (Berkeley bombardment).

4. Look for 95 (alpha + beta) from 94^{239} + deuterons (Berkeley bombardment).

5. Look for 91^{235} from uranium plus neutrons (large Berkeley bombardment).

6. Trace the 4n + 1 series. It should include 89^{221} (alphas) + 85^{217} (alphas). Try Th^{232} (n,alpha)

$$Ra^{229} \xrightarrow{beta} Ac^{229} \xrightarrow{beta} Th^{229} \xrightarrow{alpha} Ra^{225} \xrightarrow{beta} Ac^{225} \xrightarrow{alpha} 87^{221} \xrightarrow{alpha} 85^{217} \xrightarrow{alpha}$$

7. Does 93 redissolve in $C_2O_4^{=}$?

Monday, April 3, 1944

Enroute to Chicago on the "City of San Francisco."

I drafted a memorandum to Hogness covering in detail the various observations from the visits I made to the Department of Chemistry and the Radiation Laboratory during my stay in Berkeley. I had this to say about my observations of the Chemistry Department.

"Calvin has succeeded in preparing what seems to be anhydrous UCl_4 by passing HCl gas into anhydrous solution of U^{+4} acetylacetonate in benzene. However, this supposed UCl_4 has not been analyzed yet. The U^{+4} acetylacetonate was prepared by adding acetylacetonate to an aqueous HCl solution of UCl_4 and then carefully neutralizing with NaAc until the precipitate of U^{+4} acetylacetonate forms. Slow neutralization with a mild neutralizing agent (such as NaAc) is necessary in order to prevent the precipitation of oxides or hydroxides of uranium. The U^{+4} acetylacetonate appears to be anhydrous while the Th acetylacetonate seems to contain $2H_2O$ of crystallization when prepared in this way. The U^{+4} may be an all around better stand-in than Th^{+4} for our work since it might be expected to resemble Pu^{+4} more closely, and Calvin seems to experience no difficulty with respect to oxidation to UO_2^{++} in his work with it. It might be worth trying to prepare $PuCl_3$, as well as $PuCl_4$, via this acetylacetone method, since Pu^{+3} hydrolyzes less easily and might not tend to bring along OH groups as readily as Pu^{+4}.

"Calvin has also prepared in a similar manner U^{+4} furoylacetone and finds that he can extract this compound from water into benzene, and then back into water at a higher acidity, and so on back and forth depending on the acidity. He believes that the light elements would not follow the U^{+4} in these partitions.

"In addition to the ethylenediamine-acetylacetone and -salicylaldehyde and phenylenediamine-acetylacetone and -salicylaldehyde complexes with U^{+4}, which Calvin discussed in some detail when he was in Chicago last month, he believes that the U^{+4} phthalocyanine and U^{+4} porphyrin type of compounds might be useful in solvent partition methods of purification. These latter are more stable, and if not too stable, may be useful if the ethylenediamine, etc., type are not sufficiently stable. He gave me some phthalocyanine and tetraphenyl porphyrin to bring back to Chicago to use for this purpose. Compounds of U^{+4} with these might be prepared by adding them to a glacial acetic solution of UCl_4, and then extracting them into benzene after dilution of the glacial acetic acid with the proper quantity of water. Calvin didn't mention whether he thought that it would be necessary to heat the glacial acetic acid solution.

"Calvin believes that nicotinic acid, quinaldinic acid, or orthohydroxyazobenzene (especially the latter) might be good substitutes for cupferron in the cupferron extraction procedure.

"Calvin has prepared anhydrous U^{+4} tetra-acetate by adding acetic anhydride to a solution of UCl_4 in glacial acetic acid. The solid U^{+4} tetra-acetate separates out upon heating the solution. It may be possible to prepare UCl_4 at a relatively low temperature by passing HCl gas over

this compound; however, this procedure may yield $U(OH)_4$ and acetyl chloride rather than the desired UCl_4 and acetic acid.

"It seems worthwhile to repeat with Pu^{+4}, and perhaps Pu^{+3}, a number of experiments of the type which Calvin has been doing with U^{+4} as a stand-in, perhaps after we have tested the behavior of the light elements in some of the systems.

"Calvin and I discussed the magnetic criteria for the type of electron structures of the Pu ions and the question of what might be learned from magnetic susceptibility measurements. The possible electron structures, beyond the radon-type core, for Pu^{+4} seem to be $6d^4$, $5f^16d^3$, $5f^26d^2$, $5f^36d^1$ and $5f^4$, and for Pu^{+6} seem to be $6d^2$, $5f^16d^1$ and $5f^2$. For example, the $5f^4$ structure for Pu^{+4} corresponds to the spectroscopic state $^5I_4$ and a magnetic moment equivalent to about 2.6 Bohr magnetons, if one assumes undisturbed orbital contribution as in the rare earths, and the $5f^2$ structure for Pu^{+6} corresponds to the spectroscopic state $^3H_4$ with a magnetic moment of about 3.8 Bohr magnetons on the same assumption. If some of the electrons in the ground state of the Pu ions occupy 6d orbitals the type of spin-orbit coupling and amount of orbital contribution to the magnetic moments and the effect of environment in general may be less certain. In this connection it is of interest to note that Calvin has made magnetic measurements on aqueous U^{+4} and has found a striking variation in its magnetic moment with its concentration, going from about 2.5 at moderate to more than 4 Bohr magnetons at very low concentrations. R. W. Lawrence (J.A.C.S. 56, 776 (1934)), who had measured the magnetic susceptibilities of U^{+3}, U^{+4}, U^{+6} in aqueous solution, had failed to notice this concentration effect with U^{+4}. Calvin recommended for reading, in addition to Selwood's book on Magnetochemistry, the book "Magnetochemie" written by Klemm in 1936.

"The case for the existence of 5f electrons in Pu ions seems to be strengthened by the latest spectrophotometric absorption measurements on aqueous solutions by Connick, Sheline and McVey. By using a slit width of about 10 Å they find new, very sharp, absorption peaks for Pu^{+6} (about 20 Å half-width) at 4590 Å and at 5060 Å. They are studying the light absorption of aqueous solutions of Pu^{+6}, Pu^{+4}, and Pu^{+3} in considerable detail and with excellent resolution with a Beckmann spectrophotometer. I believe that, in addition to U^{+4} and U^{+6}, they intend to study the light absorption of the difficultly maintainable aqueous solution of U^{+3}; one would think that U^{+3} might be the most interesting U ion to investigate in this manner due to its greater number of electrons and hence presumably greater possibility for the existence of 5f electrons in its structure. Stickland and Webster have made rough absorption measurements on U^{+3}, reported in a project report of some time ago. Connick has found that U^{+6} has some fairly sharp absorption bands at around 4000 Å (of course not due to 5f electrons since U^{+6} cannot have such).

"Gofman has Beaufait and Crompton working on his "oxalate" extraction-decontamination procedure. In this method the Pu is carried by precipitation with $Th(C_2O_4)_2$ out of 20% UNH (the yield of Pu is fairly good even up to 40% UNH as starting material for the extraction). In this step the rare earth and alkaline earth fission products are carried while Zr and Cb remain largely in solution. The $Th(C_2O_4)_2$ is dissolved in a 0.3 M solution of $(NH_4)_2C_2O_4$ and Ce^{+3} is added to precipitate a by-product of $Ce_2(C_2O_4)_3$,

which carries the rare earths and alkaline earths. After separating the $Ce_2(C_2O_4)_3$ the solution is acidified to again precipitate $Th(C_2O_4)_2$ which carries the Pu and leaves the Zr and Cb in solution. From this point an identical second cycle can be gone through, starting by again dissolving the $Th(C_2O_4)_2$ in a 0.3 M solution of $(NH_4)_2C_2O_4$. It should be noted that the Pu remains in the +4 oxidation state throughout this entire extraction-decontamination procedure.

"They have found one beta- and gamma-emitting activity in pile uranium which does not seem to decontaminate at all by this "oxalate" method. Gofman, Crandall and Reas have been studying in great detail this activity, which they believe to be due to an isotope of element 95. This activity has a half-life of a month or two and accounts for about 1% of the total beta-activity at about 90 days after shutdown; it amounts in weight to about 0.2% of the Pu^{239}. Its beta-particles are relatively hard, having a half-thickness of some 80-100 mg/cm^2, and in addition to a gamma-ray of some 0.5 Mev it seems to have an x-ray with an energy in the region of the uranium $K\alpha$ radiation, i.e. some 100 Kev. There seems to be some slight growth of daughter alpha-activity (element 96?) as it decays, and if this alpha-activity is actually due to the daughter of this 1-2 month beta-activity the daughter must have a half-life of some 10^5 years.

"The chemical properties of this 1-2 month beta-activity, which have led them to believe that the activity is due to element 95, seem to be different than those of any known element and seem to be about as follows. There seem to exist three oxidation states, a +2, a +3, and a +4 state, but no states higher than +4. The +3 state is carried with potassium rare earth fluoride, rare earth oxalate out of acid or alkaline solution, and potassium rare earth sulfate. The +4 state, which is obtained by oxidation of the +3 with Ce^{+4} or silver persulfate but not by HNO_3, is carried with potassium rare earth fluoride and rare earth oxalate out of acid solution but not out of alkaline solution, and is not carried by potassium rare earth sulfate. The +2 state, which is obtained by reduction of the +4 or +3 state by SO_2 or hyposulfite, is carried and not carried in just the same manner as the +4 state as described above.

"I am not convinced that the activity isn't due to element 61, or to some other rare earth such as cerium mixed with element 61, or to UX_1 mixed with some rare earth or rare earths. It certainly would be strange to have four successive beta-decays, starting with uranium, which is necessary in order to obtain a beta-emitting element 95 following neutron capture in uranium. On the other hand, the chemical evidence does seem to point to element 95. The growth of an alpha-emitting daughter, if one could be certain that it was not due to some natural radioactivity contamination such as UX_1, Ac, or RdTh, would constitute a virtual proof that the beta activity belongs to element 95.

"Frederickson has built a special alpha-ionization chamber through which nitrogen flows continuously and whose pulses are amplified by a special high resolving power circuit, with which it is possible to count alpha-particles over a beta-particle background of about one millicurie. He will send us a diagram of the circuit.

"Hamaker is studying the effect of the Hanford level of radiation on the "oxalate" and "acetate" processes by having the proper amount of 93^{239}, as the source of radiation, present right in the solutions.

"Brewer, Lofgren, and Bromley have a rather large program of study of refractories underway. They are specializing in the sulfides of cerium and thorium.

"The cerium sulfides are prepared as follows. Powdered CeO_2 is heated in a graphite crucible to about 1,400°C at which temperature the H_2S effects conversion to red Ce_2S_3. The Ce_2S_3 is then heated, as a liquid, in a vacuum at 1,900°C to produce so-called "vacuum reduced" cerium sulfide of approximate composition Ce_3S_4. This is pressed into crucibles in a Mo mold with a hydraulic press at a pressure of 10,000-15,000 pounds per sq. in., after which they are sintered at 1,700°C, and then the crucibles are ready for use. Another type of sulfide of cerium, known as "cerium brass" is prepared by heating the Ce_2S_3 with clean Ce metal (or CeH_3) in vacuum in a Mo container at 1,400°C for 2 hours, then at 2,000°C for 1 hour (the latter heating to distill out Ce and some Fe impurity). This product is "brassy" yellow in appearance, has a formula such as Ce_4S_3 or $CeS_{0.9}$, and after pressing into a crucible is sintered at 1,900°C in a Mo mold.

"Thorium sulfides of analogous compositions are prepared in much the same way. The "vacuum reduced" thorium sulfides have formulas in the range $ThS_{1.8}$, $ThS_{1.5}$, etc., while the yellow "thorium brass," produced by reduction of ThS_2 by Th metal (when ThH_3 is used the product has a silver color), has formulas such as $ThS_{0.8}$, $ThS_{0.4}$, etc. The complete study of the thorium sulfides systems, including complete phase diagrams, is given in an article by Strotzer and Zumbusch, Z.A.A.C. 247, 415 (1941). Analogous information on the sulfides of uranium is given in an article by Strotzer, Schneider and Biltz, Z.A.A.C. 245, 307 (1940). These "brasses" have a NaCl-type (distorted) lattice.

"These materials seem to offer exceedingly good hope as refractories for our job, especially the "brasses." I brought back to Chicago with me, for testing with Pu, a crucible of "cerium brass" ($CeS_{0.9}$) a crucible of "vacuum heated" cerium sulfide ($CeS_{1.38}$), a crucible of "vacuum heated" thorium sulfide ($ThS_{1.75}$) and a piece of silver-colored $ThS_{0.8}$ (from ThS_2 plus ThH_3 at 2000°C).

"All-Mo, all-W and all-Ta crucibles are fabricated for Brewer in the Radiation Laboratory machine shop; they are able to weld sheets of these materials into crucibles in spite of their high melting points. Brewer says that Ta oxidizes more readily than Mo or W and also is more reactive toward refractories. He prefers Mo for temperatures up to 2,200°C and W for temperatures above this. Good instructions for working Ta, Mo and W are given by P. Rapin, Bull. Soc. Franc. Elec., (6), 1, 347 (1941), of which I have an abstract.

"Cubicciotti and Sienko are measuring the e.m.f.'s of cells involving the couple $U \rightarrow UCl_3$ (fused) with the view toward establishing the potential for this change, and with the aim of perhaps eventually applying the technique to analogous Pu couples. They seem to be getting in the neighborhood of 2 volts for this couple.

"Webster is working on the electrolysis of metallic U out of fused salt baths such as 20% UBr-80% $SrBr_2$. He has used both porcelain and quartz containers with Pt electrodes, and at sufficiently high current densities he has obtained some very fine looking "trees" of silvery U metal.

"Thurmond is working on the U-UBr_3 melting point-composition system and has obtained evidence which could be interpreted as due to the existence of UBr_2.

"Fontana is working on the determination of the distribution of tracer Pu between metal-metal halide systems (such as Sr-$SrBr_2$, Ca-$CaBr_2$, etc.) in order to obtain information as to the relative electropositive character of Pu."

Tuesday, April 4, 1944

I arrived in Chicago at 12:15 p.m.

I learned that Ralph James has completed a series of bromate cycles on the main plutonium fraction of the sample of Clinton peroxide supernatant. The isolated neptunium fraction (about 40 micrograms of 93^{237}) was given to La Chapelle. James has found no irregularity of chemical behavior, alpha particle range, growth, or decay during these operations. This evening, Ralph began making rough alpha particle absorption measurements on a portion of sample 49 DC (our target of 3 micrograms 94^{239} bombarded with 1930 microampere-hours of deuterons in the Berkeley 60-inch cyclotron) which I brought back with me from Berkeley.

As usual when I return from a trip, Helen filled me in on all of her activities during my absence and the people she saw. Last Monday when I left for California she had dinner at Peg Anderson's with Marilyn Howe. The next afternoon she visited Dagmar Dreher and on Thursday had dinner at the Quadrangle Club with the Howes, Footes, Harry Brown, and Leo Szilard; afterwards she went to the Footes' home. The Ghiorsos invited her for dinner Friday night, and on Saturday she had lunch with Wilma. On Sunday she had dinner and supper at the Ghiorsos'. Last night she again had dinner with the Ghiorsos, and later Stan, whose apartment is across the hall from ours, dropped in to chat.

Much has transpired here at the Lab while I was away the last eight days. Here is a day-to-day account of the activities that concern my section.

Monday, March 27

The Chemistry Division seminar met at 7:45 p.m. in Room 251, Ryerson Laboratory, the eve of my departure.

Tuesday, March 28

Herman P. Robinson, a former partner of Robinson and Stephens Company in La Verne, California, started working in Section C-I. After receiving his B.S. in chemical engineering at the University of Southern California in 1934, he first went to Shell Oil Company as assistant chief chemist. Robinson is joining Baumbach's group as an Associate Chemist.

Continuing his measurements on the melting point of plutonium, Jasaitis found that a sample of the high density (20.6) form could be melted and then remelted on a tantalum strip, with the final melting point going down to 950°C.

The following three articles, co-authored by me were published as Met Lab Report CN-1488 this Tuesday. They were submitted to the Physical Review in 1942, but publication is being withheld because of the nature of the contents: "Radioactive Element 94 from Deuterons on Uranium" (Seaborg, McMillan, Kennedy, and Wahl), "Radioactive Element 94 from Deuterons on Uranium" (Seaborg, Wahl, and Kennedy), and "Radioactive Element 94" (Seaborg, McMillan, Wahl, and Kennedy).

Wednesday, March 29

Byron Malekow transferred from Section C-II to C-I and will work with Dreher in West Stands. Ellen Watts is now working as a technician with Baumbach's group.

The regular triweekly evening meeting of the Basic Chemistry, Recovery, and Instruments groups (Sub-section III) of my section was held at 7:45 in Room 209, Eckhart Hall, attended by Albaugh, Arnold, Brody, Cunningham, Davidson, Dreher, Ghiorso, Hagemann, Heath, Hindman, Howland, Jaffey, Katzin, Kirk, Kohman, Kraus, La Chapelle, Manning, Meyer, O'Connor, Phipps, Rosenfels, Scott, Thompson, and Willard, and others.

Willard opened the meeting and mentioned that the Basic Chemistry Group will be working on an investigation of the chemistry of process solutions during the next few weeks, then turned the meeting over to Cunningham. O'Connor reported his studies on the rate of precipitation of Pu(IV) phosphates and their solubilities under various conditions. Kraus discussed the solubilities of Pu(IV) in the presence of 1 M Na_2SO_4 in the pH region 2-6.5. Howland talked about his recent determinations of the potentials of the Pu(III)/Pu(IV) couples in acetic acid buffer and in 1 M HF.

Ghiorso described his evidence for the emission of very soft electrons, x-rays, and gamma rays from pure plutonium. Kohman reported on his recent measurement of the neutron emission of the one-gram plutonium oxide sample mixed with powdered graphite prepared for Fermi. Scott discussed his determination of the change of counting yield with thickness of U_3O_8. Cunningham pointed out that error introduced due to back scattering of plutonium alpha particles from platinum is not as large as that of uranium alpha particles, because only a 4% increase in the counting yield results from decreasing the sample thickness from about 0.1 mg/cm^2 to essentially zero.

Thursday, March 30

General Groves was in Chicago.

Albaugh and Thompson met with Stoughton who is visiting from Acken's section at Clinton Laboratories. They discussed extraction step variables being studied at Clinton by Apple's group (agitation factor, effect of temperature on product yields and decontamination, and effect of varying rates of addition and amounts of phosphoric acid on product yields).

Dreher, Kircher, Christiansen, Bird, A. C. Hyde, and Maloney held their weekly conference on the semiworks and ether extraction process.

Compton transmitted a draft of the program of the Metallurgical Project for the year 1944-45 to Groves. "We are now using this draft," the covering letter stated, "for shaping the specific programs of the Metallurgical Laboratory and the Clinton Laboratories which you have requested for April 15." He asked for suggestions of whatever modifications may make the program suitable for submission to Groves in final form.

The evening meeting of the Research Assistants of my section met at 6:45 p.m. in Room 209, Eckhart Hall.

Friday, March 31

General Groves was still in Chicago.

A piece of high density plutonium metal (865 micrograms) was successfully filed into finely divided form by Herman Robinson and submitted to Zachariasen who found a considerable number of x-ray diffraction lines that have not yet been interpreted.

My office received a copy of a memo from Zachariasen to Allison, in which Zachariasen states that guided by the close crystal chemical similarity he has found between plutonium and the rare earth elements, he believes that he has definitely identified Pu_2O_3 as the compound which he earlier suggested might be PuF_2.

Saturday, April 1

Effective today, Lyle Jensen was appointed leader of Group IV (Purification-Precipitation-Extraction) in Orlemann's sub-section II, and J. C. Hindman was appointed leader of Group VII (Basic Chemistry) in Cunningham's sub-section III.

A break-through was achieved in the program to find a means of dissolving the Al-Si coating of uranium slugs. Flox has found that a $NaOH-NaNO_3$ mixture is very effective, and today Hyman tested this on two Al-Si coated uranium slugs. He found that 5 M NaOH plus 3 M $NaNO_3$ results in a vigorous reaction with the evolution of ammonia (and reduced hydrogen evolution to lessen the explosion hazard) and a complete removal of Al-Si coating.

4/4/44 (cont.)

Between 11 a.m. and 12 noon a meeting of the Chemistry Division Section Chiefs and Group Leaders took place in Room 209, Eckhart Hall.

Hogness issued a summary of manpower distribution in the Chemistry Division which shows the following for my section:

Section C-I, Seaborg (97 men)

			No. of Men	
			Feb.	Mar.
Thompson "W" Work 31 men	I.	Albaugh, extraction and decontamination	10	13
	II.	Watt, concentration and isolation	6	8
	III.	Dreher, process development	6	9
Orlemann "Y" Work 33 men	IV.	Jensen, precipitation and extraction	6	6
	V.	Davidson, volatility	8	6
	Va.	Orlemann, fluoride chemistry	-	4
	VI.	Baumbach, metal production	15	16
Cunningham 30 men	VII.	Cunningham, basic chemistry	9	9
	VIII.	Dawson, recovery	9	11
	IX.	Ghiorso, instruments	10	10

As a follow-up to the discussion at the Council meeting of Section C-I on March 16, L. B. Arnold issued a notice to all chemistry personnel saying that eating is now forbidden in New Chemistry Building because of health hazards. Arnold is attempting to locate a suitable area for use as a lunch room.

Monday, April 3

Duane L. Hufford, who received a B.S. in chemistry at the University of Illinois last year, began work with Ghiorso. He has been a graduate assistant at the University of Rochester.

In a letter to Tilley, attention: Chambers, in Wilmington, Delaware, Willard enclosed a print of the details of the "desk hoods" which are currently being installed in the New Chemistry Building. He mentioned that all who have used this type of hood consider it superior to the conventional single vertical-window hood.

Manning recommended to L. H. Richmond in Personnel that Kay Gavin because of her new responsibilities (she is to assume some of Donlan's duties) be reclassified to Clerk B and that Irma Saxton receive a merit increase. He also pointed out that Slattery has been asked to take the responsibility of supervising our "special cleaning people" and referred to an agreement that Slattery should receive a $2.00 per week increase in base pay, making his pay $30 per week for 40 hours, effective April 3.

There was an evening seminar of the Chemistry Division at 7:45 in Room 251, Ryerson Laboratory.

Yett has performed an experiment to determine the minimum amount of La^{+3} necessary to use in the crossover step for the precipitation of PuF_4 at Hanford concentrations of plutonium and to determine the relative merit of using 0.5 N HF or 1 N HF. He concludes that 100 mg/liter of La^{+3} and 1 N HF are desirable concentrations for the product precipitation. A higher La^{+3} concentration would possibly improve the yield slightly, but since it is desirable to have as low a lanthanum concentration as possible, it is believed 100 mg/liter of La^{+3} will be a good compromise.

I pointed out to Stearns that Dreher started working for the Met Lab on April 2, 1943, and has been unable to take a vacation since that date because of the pressure of work. He is being transferred to Hanford within a short period of time, and he requests that he be allowed to take a cumulative vacation at the time of transfer. In another memo to Stearns I explained that Stan Thompson has been able to take only one week of vacation since starting work on October 1, 1942. He, too, is to be transferred to Hanford and is due there October 15; he wishes to take a cumulative vacation of three weeks starting September 21. Since two days traveling time is allowed, he would leave September 21 from Chicago.

The group leaders in my section and I received an April 3 list from Hogness of a proposed series of lectures by the Technical Division. He asks whether our people are interested in participating. The weekly series will begin about the second week of April and will be held in Room 133, Eckhart Hall, probably on Tuesday evenings.

Helen and I had dinner at Leonard Katzin's (829 E. 52nd Street).

Moscow radio, according to today's newspapers, has called on the Rumanian people to abandon the Germans and capitulate. The Soviets are thirteen miles inside the Rumanian border.

Wednesday, April 5, 1944

James G. Barrick has left Section C-I to go to Clinton Laboratories on a loan basis.

Ralph James is continuing to take range measurements on a portion of sample 49DC.

Heath is on a two-day trip to Harshaw Chemical Company to obtain information on the preparation of F_2, UF_4, UF_6, and BF_3, including the composition of apparatus being used there--copper, nickel, etc.

Halban and Pontecorvo of the Montreal Group are here today through Friday to discuss their heavy water experiments with Allison, Fermi, and Wigner.

I received a letter from C. D. Shane written April 3, about our continuing negotiations regarding patent rights concerning element 94. He suggests that the other inventors and I offer to deal with Dr. Bush and that the University be invited to participate in the negotiations.

He predicts, however, that the University will not participate until its representative has been able to examine the nature of the inventions and the other circumstances pertaining to them. He also thinks that Bush considers that the University may have certain rights and so may not be willing to enter into an agreement with the inventors alone.

I sent Hogness the summary of my visit to the Berkeley Radiation Laboratory and the Department of Chemistry March 25 - April 2 which I wrote on the train on my way home.

During the past month the Nuclear Physics Division completed the first series of experiments using about one ton of heavy water. Three measurements were performed, namely, (a) an attempt to determine the absorption cross section of heavy water by the sigma pile method with the result that the cross section was found to be too small to be detected with a sigma pile of the dimensions used and that the cross section is, if anything, smaller than had been estimated previously; (b) the range of fission neutrons in heavy water was measured and found to be somewhat smaller than had been expected theoretically, and the age for the reduction of fission neutrons to indium resonance energy was found to be 96 cm^2; (c) an exponential experiment was performed in order to determine the Laplacian of the lattice to be used in the P-9 pile now in construction at Argonne. The Laplacian was found to be 880×10^{-6} cm^2, in good agreement with the theoretical expectation. There are certain indications that the agreement is to some extent accidental since a satisfactory explanation of the observed Cd ratio is still missing. Work is going on in order to clarify this point. The practical conclusion is that the design of the Argonne pile need not be changed.

I received a memo from Thompson giving the work distribution in Sub-section I as of April 5. In addition to listing the problems under study, he shows the following organizational arrangement:

<u>Group I - Extraction and Decontamination</u>
Albaugh (Leader), Hoekstra, Ader, Sedlet, Bartell, Sheft, Gilbreath, Post, Morgan, Gardner, Greenlee, Peterson, Malm (on loan to Cunningham to prepare crystalline product phosphates)

<u>Group II - Concentration and Isolation</u>
Watt (Leader), Pye, Walling, Yett, Kelley, Haeckl, Hopkins, Goeckermann

<u>Group III - Process Development and Semiworks</u>
Dreher (Leader), Hyman, Winner, Flox, Margolis, Malekow, Lincoln, Rasmussen, Larson

In a memo to Hogness I summarize the work of the past month on "Separations Processes." It will be issued later this month in essentially the same form as a CN Report.

Greenewalt, Hogness, and I met to discuss procedures in the plutonium isolation step at Hanford, and the shipment of plutonium to Site Y. We reached tentative agreement that (1) Site Y will accept shipments of a solution 30% plutonium or 30% $Pu(NO_3)_4$ by weight, (2) routine analyses

other than process control will not be carried out, (3) in processing, the 250-gram batches of material will not be divided into three portions, (4) the determination of the final amounts of plutonium will be made by alpha particle counting. This latter procedure will be combined with a weighing and a determination of the density of the plutonium solutions.

Helen and I had dinner at the 51st Street YMCA with the Baumbachs.

I attended the regular triweekly evening meeting of the Separation Processes Sub-section I of Section C-I, at 7:45 in Room 209, Eckhart Hall. Others present were Ader, Albaugh, Bartell, Beard, Dreher, English, Fields, Flox, Gardner, Gilbreath, Greenlee, Hoekstra, Hopkins, Howland, Hyman, Jasaitis, Katzin, Kelley, Kraus, Krueger, La Chapelle, Malm, Manning, Margolis, Morgan, Peterson, Phipps, Post, Pye, Rasmussen, Rosenfels, Sedlet, Seifert, Thompson, Walling, Wolf, and others. As usual I turned the meeting over to Thompson to act as chairman.

English, who is visiting from Clinton Laboratories, reviewed the Clinton separations process and indicated no striking changes have been made. Some difficulties have been encountered which may be related to the insoluble (beta) form of bismuth phosphate and for this reason some work has been started with this material. He also mentioned that some work has been done on different isolation procedures and referred to the fact that the 45% K_2CO_3 used in the metathesis step leaves a solid behind that contains only about 1% plutonium. I suggested that this solid might contain element 93 and should be checked for such. Thompson mentioned plans to work on the insoluble form of bismuth phosphate in Chicago and the work being done on new oxidizing agents such as Pb_3O_4.

Albaugh reviewed the latest Bismuth Phosphate Process run at Hanford concentrations which gave 93% recovery through the crossover step. Decontamination was not as good as expected, and it appears scavengers will be necessary at Hanford.

Hoekstra enumerated nine process variables that have been tested, or will be, at Hanford concentrations in the extraction step.

During a resume by Hopkins of experiments on the concentration and isolation of plutonium, I mentioned that it has now been decided to process the entire 250 grams of plutonium from each chemistry extraction plant run at Hanford instead of dividing it into three batches. Although the critical mass may well be in the neighborhood of 250 grams, this is only true for a perfect neutron reflector; it has been deemed safe to process the entire 250 grams at once.

Dreher, reviewing process development, mentioned that Lincoln has done some work on the fate of 93^{237} in the Bismuth Phosphate Process and that four runs have given yields of 10.3, 14.7, 11.3, and 18.9% at the end of the first cycle. The loss of 93^{237} in the by-product has been only about 1%. However, only about 20% is carried in each product precipitate. The coating problem is still quite serious, and it is now believed that complete dissolution will be impossible--it is hoped merely to dislodge the scale. Margolis concluded the meeting by talking about his work on the effect of time on extraction of oxidized plutonium by ether.

4/5/44 (cont.)

After the meeting English came home with me for coffee.

War news is not in the headlines being superseded by a headline declaring that a jury in federal court has acquitted Charlie Chaplin on two counts of violating the Mann Act. The second but smaller headline deals with politics; Thomas Dewey is leading in the Republican Presidential primary in Wisconsin.

The Project Council Meeting was held at 9:30 a.m. in Room 209, Eckhart Hall, attended by Allison, Chapman, Chipman, Compton, C. M. Cooper, Dempster, Fermi, Franck, Greenewalt, Hilberry, Hogness, Jeffries, W. C. Johnson, Kimpton, McKinley, Mulliken, Smyth, Spedding, Stone, Vernon, C. J. Watson, W. W. Watson, Whitaker, and Wigner. In his "State of the Nation" message, Compton summarized his meetings of last week with General Groves and Directors of various phases of the Project and stated that it looks as though both major approaches (uranium-235 and plutonium) will be successful, will have the same order of magnitude of cost and should be producing on a plant scale at about the same time.

Great Britain is expected to build in Canada a heavy water lattice pile for the production of 49; Met Lab is expected to supply a team of perhaps 15 men to work in Canada on a loan basis. Production is not expected before 1946. Whitaker gave the general Clinton Laboratories report and indicated that the 200 area (Chemical Separations Plant) at X-10 will be shut down for about 10 days to make equipment changes. This will give an excellent check on the cleanability of equipment. The current yield of the 49 product is believed to be above 80%. Fermi and Watson each presented a "distribution of effort" for his Division. In reply to Compton's question whether or not collaboration with Los Alamos on the properties of 49 and 25 is working out, Fermi said "yes--especially since we do not always agree exactly and so the need for parallel effort is desirable." Watson commented that consideration is being given to transferring Snell, Wilson, and Wollan to other Divisions where they can be effectively employed more directly in connection with nuclear physics. In a discussion of Hanford, Greenewalt mentioned the plans to ship 49 in closed stainless steel containers of about 2 liter capacity but carrying only about 500 cc of solution. The containers will be evacuated after charging and then filled with an inert gas. It is estimated that samples can be stored in this manner for a month before any hazard from an explosive mixture develops.

Thursday, April 6, 1944

Today Earl Hyde treated a 3.93 mg sample of $PuBr_3$ with Br_2 for an hour at 400°C. He found essentially no change in weight indicating that $PuBr_4$ was not formed.

Samuel A. Jackson, III, who was discharged from the Army February 26, started work in Section C-I, Group VI (Baumbach). He received his A.B. in anthropology from the University of Michigan last year.

I wrote Eastman in Berkeley explaining that the crucibles Brewer gave me when I was visiting Berkeley were too large for some of our tests. I suggested that he send me some solid pieces of the metal-reduced powder (cerium "brass") from which we can machine our own crucibles.

I asked Hilberry to procure additional radium-beryllium and radium-boron neutron sources for Kohman's use in his work to develop sensitive neutron detectors.

Manning suggested to Hogness that he ask Compton to obtain for our section from Murphee of the Standard Oil Development Company, a man with broad research experience with organic solvents and solvent extraction processes. The program we have in mind for this man would involve particular emphasis on purification aspects of our work, but there might also be developments in relation to alternate concentration or isolation processes.

Dreher attended the regular weekly conference on ether extraction with Bird, Christiansen, A. C. Hyde, Kircher, Maloney, A. C. Miller, and Tepe. He learned that the second run of the continuous ether extraction system disclosed no mechanical difficulties. Mechanical improvements to be made to the system were agreed upon.

This evening the Baumbachs had dinner with Helen and me in our apartment.

The evening meeting of the Research Assistants of my section was held as usual at 6:45 p.m. in Room 209, Eckhart Hall.

At the laboratory-wide evening meeting of scientific personnel at 7:45 p.m. in Room 133, Eckhart Hall, Hogness spoke on the topic, "Bombs on England," describing his observations based on a recent trip to England.

The evening meeting of the Council of my section at 7:45 in my office was attended by Albaugh, Baumbach, Cunningham, Davidson, Dreher, Hindman, Jensen, Katzin, Manning, Orlemann, Thompson, Watt, and me. I pointed out that, for security reasons, no mention should be made of quantities of plutonium received or production schedules when speaking at such meetings as the Monday night Chemistry Division seminar. I reviewed the tentative agreements on shipment of product from Site W to Site Y, reached in my meeting yesterday with Hogness and Greenewalt.

Watt reported that he has prepared a small volume of solution containing 550 g/l of plutonium (element) as the nitrate; it had a density of 1.65 gm/cm^3. The series of lectures proposed by the Technological Division was discussed. Manning will see how many in the section are interested in attending. It was agreed that during this lecture

series period (beginning the second week in April), the Research Assistants' meetings should be discontinued. An additional 100 mg of plutonium was assigned to the Basic Chemistry Group for the process chemistry work.

I reviewed some of the work in progress at Berkeley, mentioning the hot-wire technique developed by Prescott for the thermal decomposition of UI_4 to produce metal and the plans to work out the application of this technique to plutonium. I also mentioned that sulfide refractories appear promising.

Baumbach reported that a number of large pieces of good plutonium metal have been produced, including a piece of 2.88 mg in weight. Pills of PuF_4 were used for reduction of them.

At 11:30 p.m. George Watt and I left Chicago Englewood Station on the Penn R.R. to Cincinnati where we will continue on the L&N R.R. to Site X. We plan to return Sunday.

Wendell Willkie has withdrawn from the primary race for the Republican nomination for the Presidency. He failed to win even one delegate to the convention in the state of Wisconsin.

Friday, April 7, 1944

Watt and I arrived in Oak Ridge in the afternoon. I attended the Steering Committee meeting at Clinton Laboratories (Chicago-Clinton Chemical Conference); others present were Harrison Brown, Greager, Hogness, W. C. Johnson, Kay, Kirst, and Squires. The status of Clinton problem assignments was discussed. A list of these problems and Clinton manpower assignments is as follows:

		Manpower
1.	Dissolution of bismuth phosphate precipitates	0
2.	Search for new oxidizing agents--a survey	1
3.	Uranous oxalate method for concentration of plutonium	1
4.	Investigation of the chemistry of plutonium	3
5.	Investigation of plutonium compounds suitable for storage and shipment	?
6.	Chemical problems of the extraction plant	1
7.	Process service work for Section C-I	3
8.	Development of radiochemical analysis procedures	1
9.	Application of decontamination studies to the design of improved steps in the complete Bismuth Phosphate Process	2
10.	The behavior of plutonium fission products in the Bismuth Phosphate Process	0
11.	Concentration processes involving the use of hydrochloric acid	0

4/7/44 (cont.)

I explained that we are training a man, at Clinton's request, to work on bismuth phosphate solubility; the dissolution of bismuth phosphate precipitates is an important problem, according to Kay. In the matter of plutonium peroxide precipitation problems, I said that Watt may bring Goeckermann and Hopkins to Site X the latter part of April for the purpose of running through the Hanford flowsheet for the concentration process, using Clinton plant solutions. Regarding the Hanford effort, Clinton will place the balance of its available manpower on Hanford orientation runs and on Hanford decontamination problems. Chicago will put three men on the variables affecting carrying at Hanford concentrations.

Saturday, April 8, 1944

In Oak Ridge. I attended another session of the Chicago-Clinton Chemical Conference, with the same participants as yesterday's session. The problem assignments at Chicago were summarized as follows:

	Manpower
Variables affecting carrying at Hanford concentrations	3
Process orientation runs	2
Low acidity scavenging	2
New oxidizing agents	1
Solubility work on $BiPO_4$	1
Concentration (dissolving and metathesizing of product)	2
Hanford peroxide concentration method	2
Concentration method for early operation at Hanford	2
Alkaline concentration methods	1
Coating dissolution problem	3
Process service work	3
Ether extraction	2
Basic process chemistry	3

Perlman, English, and I reviewed the organization of Section C-I at Clinton. Perlman and English are Section Chief and Associate Section Chief, respectively, for Section C-I here. The groups are Process Research under Koshland, Process Services under English and Bohlmann, Physical Measurements under Borkowski, Isolation under V. R. Cooper, Basic Chemistry under Werner, and Isolation (research) under Fries.

Watt and I caught the train to Chicago via Cincinnati: Southern RR to Cincinnati, NYCRR to Chicago.

Sunday, April 9, 1944

Watt and I arrived in Chicago at 3:00 p.m. During the train trip I talked to him about assuming the position of an Associate Section Chief in Section C-I.

Helen told me that she had dinner at Marilyn Howe's on Friday, followed by badminton at the U.C. Ida Noyes women's gym, and then went to the Zinns'. Yesterday she had a luncheon for Adelaide Willard, Jeanette Sawyer, Alice Katzin, Dot Maloney, Dorothy Manning, Dagmer Dreher, and Pauline Watt at our apartment. Later Marilyn Howe had dinner with her.

Monday, April 10, 1944

James told me that on Friday he completed absorption curves on an as-received portion of sample No. 49DC (plutonium plus deuterons, Berkeley bombardment) and concluded there is some indication of a long-range alpha activity.

He began working up the 7-mg sample of plutonium, in the form of $Pu(OH)_4$, that has been bombarded in the Clinton pile for about one month (Sample 49NA). I took this sample to Oak Ridge with me on February 2. The tube containing the sample broke during the bombardment, and it was first necessary for James to heat it with H_2SO_4 and HNO_3 to get rid of the paper, etc.

In other happenings Friday, Dawson summarized for Cunningham the batch ether extraction methods used with considerable success by the Recovery Group since last March 2, for the separation, isolation, and purification of plutonium. He recommends such a method be used at Site X and Hanford.

Hubert Billington began work as a technician in Hindman's group at $28 per week.

Compton wrote to General Groves in Washington suggesting he re-emphasize to the du Pont people the importance of producing, at the earliest possible moment, a few kilograms of plutonium from the first Hanford pile, even if it means sacrificing the certainty of its long, continued operation.

On Saturday, Dreher held a meeting concerning trouble with carrying of plutonium on bismuth phosphate on a liter scale, attended by Larson, Lincoln, Rasmussen, Thompson, Cunningham, and Hindman. A series of tests were agreed upon.

Donald P. Ames started work today as a Research Assistant. He comes from the Atlantic Refinery Company in Philadelphia, Pennsylvania and received his B.S. degree in chemistry this January from the University of Wisconsin.

Hagemann today carried out a bromination of three plutonium metal pieces weighing 0.1 mg. Treatment was with bromine gas in a closed capillary system at 450°C. The resulting product is in the form of green powdery lumps. The capillary was sealed off and the sample submitted to Zachariasen for x-ray analysis.

Cunningham sent a letter to Whitaker, attention Brolley, in Oak Ridge, giving the specifications of a U^{235} sample deposited on a thin aluminum foil that he has delivered to Allison. He also sent Fermi a correction in the calculated composition of the mixed plutonium oxide and graphite sample prepared for Fermi about a week ago by Cliff Smith.

Willard listed for D. M. Smith in Oak Ridge, the laboratory items and office equipment needed for his process chemistry group in the Research Building (No. 3706) at Site W.

In a joint letter to General Groves, Compton, Allison, and Whitaker submitted a proposed program of work for the Metallurgical Project until about July 1945. The letter specifies the follow major tasks:

I. Support of the Hanford 49 Production Program

II. Cooperation with Los Alamos toward use of products 25 and 49

III. Investigation of new possibilities: (improved piles; process materials; basic scientific work)

In the presentation of the proposed research program it was indicated that the following chemistry studies would be sharply increasing:

Purification of final product

Separation processes for 23, and other products

Recovery of impoverished material and by-products

Utilization of by-products

Basic chemical research

It was also stated:

"The chemical studies should be continued at about the present level. While there will be sharply reduced emphasis on separation methods, there is need for greatly increased attention to product purity and extensive new work on recovery and efficient use of the process materials. This change in emphasis will mean the increasing importance of the fundamental chemistry of our process."

I attended the evening seminar of the Chemistry Division at 7:45 in Room 251, Ryerson Laboratory.

Helen had Wilma to lunch. In the evening she put the finishing touches to the "Table of Isotopes" while I was at the seminar. It should be ready for submittal to Reviews of Modern Physics this week.

The paper today carries news of an American attack at the end of March on the string of Palau Islands, stretching for more than 70 miles. Japanese resistance was surprisingly light.

Tuesday, April 11, 1944

I received a copy of an April 10 memo from Hogness to his Section Chiefs and Group Leaders, reiterating that we have been asked by the Army not to mention in group gatherings the amount of product we have on hand, the amount of any one shipment or expectations. He said that the factors which must be guarded most in this Project are: (1) time schedules, (2) production rates, (3) relations between various sites, (4) magnitude of the operation, and (5) important personnel associated with the Project.

A memo (MUC-LAK-108) from Kimpton's office gives the following addresses for our correspondence: Site X, P.O. Box 1991, Knoxville, Tenn.; Site W, P.O. Box 100, Richland, Wash.; Site Y, P.O. Box 1663, Santa Fe, N. Mexico; du Pont, Wilmington, P.O. Box 1791, Wilmington, Del.; and our return address as P.O. Box 5207, Chicago 80, Ill.

I received and read with great interest today the University of California Project preliminary report (CN-1540) issued last Friday, "Discovery of Element 95 (and/or 96)." It reports that a new activity has been discovered in pile material that emits beta particles, x-rays, and gamma rays. The authors conclude that the observed activity represents element 95 or 96, although the possibility of a rare earth like 61, the chemistry of which is not known, cannot be ruled out. A search for alpha particle growth is in progress, and a magnetic beta-ray spectrogram will be made.

On the basis of this more detailed information I believe I now understand the origin of this activity--it is not due to element 95 or 96. I immediately wrote Gofman that I have just received his report on element 95 or 96 and have had a chance to study it. I asked whether he has satisfied himself as to the possibility of the activity being due to the fission product 57-day yttrium, inasmuch as the absorption curves of the beta and gamma radiation from this yttrium activity are not far different from his absorption curves and the chemical properties are not too inconsistent with this possibility.

I transmitted to Kennedy in Los Alamos a copy of the April 3 letter I received from Shane about our patent application plans. I wrote that I have tried to make it clear to Shane that my stand is not irreversible and that I will go along with the majority opinion of the inventors.

I also wrote to Saul Winstein at UCLA that I regret missing him when he was in Chicago last Friday as I was very anxious to talk with him. I went on to explain where he might fit in our organization, suggesting he might want to be leader of one of the groups in my section or, if he

prefers administrative work, he could consider being an assistant or associate section chief inasmuch as these positions will be vacant very soon. I said the situation with some of the positions is such that it would be much better if he could come here very soon although, even if he could come only for the summer beginning in June, I would want him.

In a memo to Orlemann, Brody urges the use of an ether extraction step for the concentration and isolation of plutonium in the Clinton and Hanford operations. He points out that ether extraction of plutonium(VI) has replaced in part the previous recovery method used by the Recovery Group and accomplishes separation and isolation of plutonium efficiently with considerable saving in time and material. He warns about the hazards of handling ether but said that they can be reduced to safe operating risks.

Manning asked Stearns to give A. Pauline Stein, a research assistant in Jensen's group, sick leave in order to care for her mother who just had a heart attack.

Helen worked all day and evening on the "Table of Isotopes" to get it ready for mailing tomorrow.

In the South Pacific the Japanese have abandoned Gasmata, air supply base on the south coast of New Britain and are in full retreat to Rabaul.

Wednesday, April 12, 1944

Robert Patton is transferring from Section C-I to work with Kirk in Section C-IV.

Today I mailed my "Table of Isotopes" to Reviews of Modern Physics. Thanks to Helen who did much of the library work and preparation, it will become available to the scientific community much earlier than if I had done all the work myself.

Hindman sent to Fries at Clinton Labs the results of a spectrophotometric examination of a plutonium iodate solution that Fries submitted to the Met Lab some time ago.

In a memo to Compton, Hogness describes his idea for a "West Point of Science" to support a continuing program of higher scientific study after the war as a means of self-defense. "History," he wrote, "is full of great men who were students of great men and in turn produced great scholars. That principle is the basic foundation for the 'West Point of Science.'"

I attended the triweekly evening meeting of the Purification and Metal Production Sub-section II of my section, at 7:45 in Room 209, Eckhart Hall. Others present included Arnold, Baumbach, Brody, Davidson, Florin, Fred, Gilbreath, Hagemann, Heath, E. K. Hyde, Jaffey, I. Karle, J. J. Katz, Kirk, Meyer, Orlemann, Scott, Stein, and Westrum. After some introductory remarks I asked, as usual, that Orlemann conduct the meeting.

Brody reviewed solvent extraction studies for removal of light element impurities. Of the solvents tried, triglycoldichloride, ethyl ether, and ethylbutylcellosolve do a fairly good job. I mentioned that the results look good, except for calcium, iron, boron, and phosphorus, and that continuous extraction ought to function well. Mrs. Stein told of her unsuccessful attempts to use complexing agents in the purification process.

Meyer mentioned difficulty in studying the weight change in the reaction: $PuO_2 + 4HF \rightarrow PuF_4 \rightarrow H_2O$ because HF attacks platinum boats; therefore gold boats are to be constructed. Meyer has treated a number of different plutonium compounds with HF at 500°C; all of them yield a green substance, presumably PuF_4. Florin described experiments to determine the effect of small amounts (0.5%) of hydrogen with HF on the conversion of the oxide to the fluoride. This mixture reduces PuF_4 to PuF_3. Work on the higher plutonium fluoride(s) was described. Using a long nickel tube with a known temperature gradient, Florin conducted fluorination at 700°C; 94% of the material was recovered from that portion of the apparatus that was at the temperature gradient 160°C-50°C.

Hyde talked about recent work on the chemistry of plutonium bromide. Reaction of PuO_2 with HBr at 700°C yields a green compound that gives a different x-ray picture than the material prepared by Hagemann using the reaction of bromine on the metal. It has been noted that hydrobromic acid reduces Pu(IV) to the trivalent state on warming to 70°C. I suggested liquid phase reactions with plutonium metal as being worthy of investigation. I. Karle discussed treatment of PuO_2 or $Pu(OH)_4$ with CCl_4 at 750°C to produce $PuCl_3$. The same difficulties in the interpretation of the x-ray data as encountered with the bromide are found here.

J. Katz described experiments relating to the preparation of $PuCl_3$ by a wet method. Aqueous $PuCl_4$ is reduced to the trivalent state with HI and the solution is evaporated to dryness in an HCl atmosphere at a pressure of 50-60 mm of mercury. The dried substance is obtained as crystals by heating to a temperature of 300°C, giving a bluish-gray substance that is water-soluble and yields a purple solution. X-ray examination indicates that the material is anhydrous $PuCl_3$.

Baumbach discussed recent work of the metal production group. Density of the metal seems to be around 20 gm/cm^3. Spectrographic analysis indicates that metal obtained by barium reduction in beryllia is quite pure, whereas metal prepared in thoria seems to contain 4-6% thorium. The melting point seems in the range 850-950°C. As a result of using the PuF_4 in the form of a pill, plutonium metal yields have been much improved, ranging as high as 86%. A piece of plutonium metal weighing 2.8 mg has been prepared. Plutonium has been volatilized from tantalum at 1,700°C which suggests that the metal may be quite volatile.

In the drive in the Crimea the Soviets have captured two of the most important points in the peninsula--the heavily fortified town of Kerch and the strategic rail junction of Dzhankol.

Thursday, April 13, 1944

Helen Pellock began working as a laboratory technician in Davidson's group.

Today, using a specially constructed glass capillary apparatus, Fried measured the amount of hydrogen absorbed in a sample of plutonium metal (containing 262 micrograms of plutonium). In this manner he determined the formula of the black plutonium hydride to be $PuH_{4.0}$.

On Monday Hagemann brominated at 450°C three small pieces of plutonium metal (approximately 100 micrograms) and submitted the sample of blue-green powdery material to Zachariasen for x-ray diffraction examination. Today Zachariasen informed Allison that the sample is anhydrous $PuBr_3$ and has the UCl_3-type structure.

"Personnel of Metallurgical Laboratory as of April 10, 1944" (M-CA-1563) has been issued. This is an alphabetical listing and includes the following as members of Section C-I:

Ader, M.	Junior Chemist	(Albaugh)
Albaugh, F. W.	Chemist	(Seaborg)
Ames, D. P.	Research Assistant	(Hindman)
Asprey, L. B.	Research Assistant-SED	(Dawson)
Baer, A. J.	Laboratory Assistant	(Donlan)
Barrick, J. G.	Research Assistant	(on loan to Clinton)
Bartell, L. S.	Research Assistant	(Albaugh)
Baumbach, H. L.	Chemist	(Seaborg)
Baumbach, N. S.	Research Assistant	(Dawson)
Beard, W. C.	Research Assistant	(Dawson)
Billington, H.	Laboratory Technician	(Hindman)
Blomeke, J. O.	Research Assistant	(Watt)
Bowman, E. E.	Laboratory Assistant	(Donlan)
Boyd, L.	Clerk	(Manning)
Boykin, P.	Laboratory Technician	(Dreher)
Britain, J. W.	Research Assistant	(Dawson)
Brody, B. B.	Research Assistant	(Jensen)
Carter, T. C.	Laboratory Assistant	(Orlemann)
Casler, R. A.	Laboratory Technician	(Jensen)
Crawford, J. A.	Junior Physicist	(Ghiorso)
Cunningham, B. B.	Chemist	(Seaborg)
Davidson, N. R.	Associate Chemist	(Seaborg)
Dawson, L. R.	Chemist	(Seaborg)
Dixon, J. R.	Junior Chemist	(Jensen)
Donlan, F. B.	Clerk	(Seaborg)
Dreher, J. L.	Associate Chemist	(Seaborg)
Fields, P. R.	Junior Chemist	(Dawson)
Fineman, P.	Research Assistant-SED	(Dawson)
Florin, A. E.	Junior Chemist	(Davidson)
Flox, J.	Junior Chemist	(Dreher)
Foreman, B.	Laboratory Technician	(Baumbach)

4/13/44 (cont.)

Frank, R. D.	Research Assistant	(Baumbach)
Freeman, E. M.	Laboratory Assistant	(Donlan)
Freeman, J.	Laboratory Assistant	(Dreher)
Fried, S. M.	Associate Chemist	(Baumbach)
Fulan, E. R.	Laboratory Assistant	(Donlan)
Gardner, R. A.	Research Assistant	(Albaugh)
Gavin, K. B.	Clerk	(Donlan)
Gerstein, M.	Junior Chemist	(Baumbach)
Ghiorso, A.	Associate Chemist	(Seaborg)
Gilbreath, J. R.	Junior Chemist	(Albaugh)
Gilpatrick, L. O.	Research Assistant	(Baumbach)
Goeckermann, R. H.	Research Assistant	(Watt)
Golden, L.	Research Assistant	(Ghiorso)
Gordon, E. N.	Laboratory Assistant	(Donlan)
Gottlieb, Dorothy	Secretary	(Thompson)
Greenlee, R. W.	Research Assistant	(Albaugh)
Haeckl, F. W.	Associate Chemist	(Watt)
Hagemann, F. T.	Junior Chemist	(Davidson)
Harding, R. B.	Bench Hand	(Donlan)
Heath, R. E.	Chemist	(Davidson)
Hellman, N. N.	Associate Chemist	(Baumbach)
Hindman, J. C.	Associate Chemist	(Cunningham)
Hoekstra, H. R.	Junior Chemist	(Albaugh)
Hopkins, H. H.	Research Assistant	(Watt)
Howland, J. J.	Junior Chemist	(Hindman)
Hufford, D. L.	Research Assistant	(Ghiorso)
Hyde, E. K.	Junior Chemist	(Davidson)
Hyman, H. H.	Junior Chemist	(Dreher)
Jackson, S. A.	Laboratory Technician	(Baumbach)
Jaffey, A. H.	Associate Chemist	(Ghiorso)
James, R. A.	Junior Chemist	(Hindman)
Jasaitis, Z. V.	Associate Chemist	(Baumbach)
Jennings, N. G.	Laboratory Assistant	(Donlan)
Jensen, L. H.	Junior Chemist	(Orlemann)
Johnson, F. D.	Research Assistant	(Baumbach)
Karle, I. L.	Associate Chemist	(Davidson)
Karle, J.	Associate Chemist	(Baumbach)
Katz, J. J.	Associate Chemist	(Davidson)
Katz, S.	Associate Chemist	(Baumbach)
Katzin, L. I.	Associate Chemist	(Seaborg)
Kelley, A. E.	Research Assistant	(Watt)
Kelley, E. J.	Secretary	(Orlemann)
Knowles, M. T.	Laboratory Assistant	(Donlan)
Kohman, T. P.	Associate Chemist	(Ghiorso)
Kraus, K. A.	Associate Chemist	(Hindman)
Krueger, A. C.	Associate Chemist	(Ghiorso)
La Chapelle, T. J.	Junior Chemist	(Hindman)
Larson, R. G.	Associate Chemist	(Dreher)
LeFeuvre, A.	Laboratory Technician	(Ghiorso)
Leventhal, L.	Research Assistant-SED	(Dawson)
Lincoln, D. C.	Junior Chemist	(Dreher)
Magnusson, L. B.	Junior Chemist	(Hindman)

4/13/44 (cont.)

Malekow, B.	Research Assistant	(Dreher)
Malm, J. C.	Research Assistant	(Albaugh)
Manning, W. M.	Chemist	(Seaborg)
Margolis, A. J.	Associate Engineer	(Dreher)
Meyer, F.	Associate Chemist	(Baumbach)
Miller, E.	Laboratory Assistant	(Donlan)
Moore, G. E.	Research Assistant	Site X
Morgan, L. O.	Junior Chemist	(Albaugh)
O'Connor, P. R.	Junior Chemist	(Hindman)
Orlemann, E. F.	Chemist	(Seaborg)
Owen, J. E.	Secretary	(Cunningham)
Parsons, A. M.	Laboratory Assistant	(Dreher)
Patton, R. L.	Chemist	(Hindman)
Peterson, S.	Junior Chemist	(Albaugh)
Phipps, T. E.	Senior Chemist	(Baumbach)
Post, R. C.	Research Assistant	(Watt)
Pye, D. G.	Associate Chemist	(Watt)
Rasmussen, R. W.	Junior Chemist	(Dreher)
Reinhardt, R. A.	Junior Chemist	(Jensen)
Robinson, H. P.	Associate Chemist	(Baumbach)
Rosenfels, R. S.	Chemist	(Baumbach/Curtis)
Saxton, I. N.	Secretary	(Manning/Willard)
Schoenberg, B.	Junior Chemist	(Ghiorso)
Scott, B. F.	Research Assistant	(Ghiorso)
Seaborg, G. T.	Senior Chemist	(Hogness)
Sears, G. W.	Chemist	(Baumbach)
Sedlet, J.	Research Assistant	(Albaugh)
Sheft, I.	Junior Chemist	(Albaugh)
Simpson, O. C.	Senior Chemist	(Baumbach)
Slattery, J. J.	Laboratory Assistant	(Donlan)
Smith, C.	Associate Chemist	(Hindman)
Smith, E. E.	Secretary	(Seaborg)
Stein, A. P.	Research Assistant	(Jensen)
Stewart, D. C.	Junior Chemist-SED	(Dawson)
Studier, M. H.	Junior Chemist	(Dawson)
Summers, M. M.	Laboratory Assistant	(Donlan)
Thompson, S. G.	Chemist	(Seaborg)
Walling, M. T.	Research Assistant	(Watt)
Walsh, P. D.	Research Assistant	(Ghiorso)
Watt, G. W.	Senior Chemist	(Seaborg)
Watts, E.	Laboratory Technician	(Baumbach)
Weissbourd, B. B.	Junior Chemist	(Ghiorso)
Westrum, E. F.	Associate Chemist	(Baumbach)
Wetlaufer, D. B.	Laboratory Technician	(Dawson)
Willard, J. E.	Senior Chemist	(Seaborg)
Winner, B. M.	Research Assistant	(Dreher)
Wolf, M. J.	Associate Chemist	(Davidson)
Yett, F. R.	Research Chemist	(Watt)
Young, R. B.	Laboratory Assistant	(Donlan)
Zvolner, H. P.	Associate Chemist	(Baumbach)

4/13/44 (cont.)

Figure 34 shows a floor plan with room assignments of our men in New Chem. (See also Figures 35 and 36.) Dreher's group, consisting of Boykin, Flox, Freeman, Hyman, Larson, Lincoln, Malekow, Margolis, Parsons, Rasmussen, and Winner, is assigned to the second floor of the West Stands (Figure 12).

The meeting of the Research Assistants of my section took place at 6:45 p.m. in Room 209, Eckhart Hall.

The evening meeting of the Council of my section at 7:45 in my office was attended by Albaugh, Baumbach, Cunningham, Davidson, Dawson, Dreher, Hindman, Jensen, Katzin, Manning, Orlemann, Simpson, Thompson, Watt, Willard, and me. Proposals for safety in the section were discussed, and the following items were considered among the most important:

1. Rubber gloves should be worn when handling plutonium and should be washed and removed before leaving the room. No one should be allowed to wear such gloves in the counter room.

2. Rooms should be kept clean.

3. Daily surveys with "Pluto" should be carried out. A number of instruments are being constructed and will be available for this work.

4. The question of whether air measurements and more nose swab counts should be made is to be determined by the Assistant Section Chiefs.

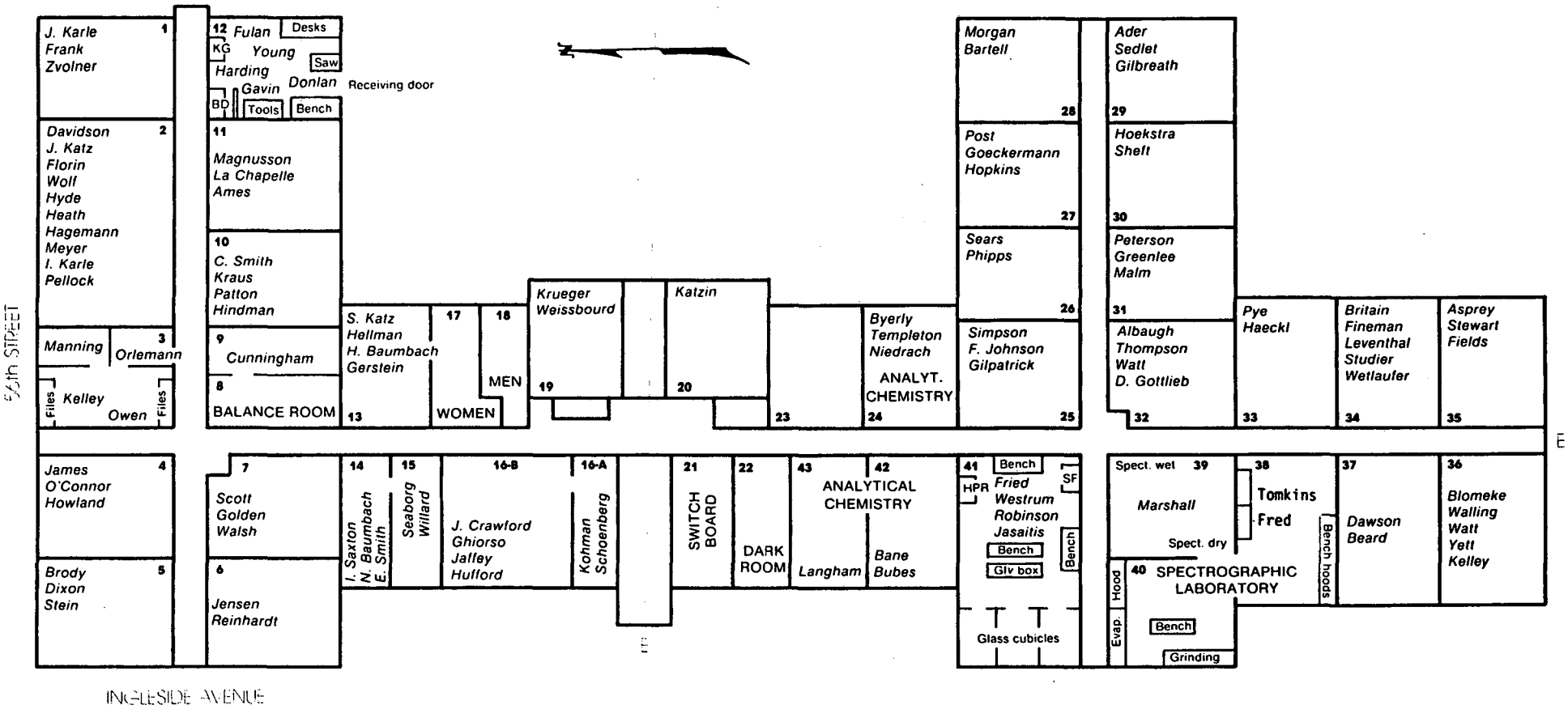
I brought up the subject of plutonium metal production, saying that thorium is produced from ThO_2 by calcium reduction. Other refractories already suggested to replace this are CaO, lanthana, and BaO. I placed thermal reduction ahead of electrolytic reduction as a method of plutonium metal preparation. Hindman reported briefly on progress in working out the basic chemistry of the Bismuth Phosphate Process. Considerable knowledge on valence states, solubilities, and potentials for various process steps is now available. Hindman suggested the section acquire a calculating machine, and we decided to do so. Simpson and I discussed possible vapor pressure measurements of plutonium metal--about two mg of metal would be needed. Fluoride vapor pressure measurements were then briefly discussed.

Helen went to Wilma's for coffee while I was conducting the Council meeting.

The Russian army in its swift offensive in the Crimea raced 46 miles in one day. They are only 46 miles from Sevastopol.

Friday, April 14, 1944

James completed a series of peroxide precipitations and lanthanum fluoride oxidation-reduction cycles to purify plutonium sample 49NA. Measurement of the ratio of alpha particle counts to beta particle counts indicates the possibility that a beta-active 94 is produced by neutron bombardment of Pu^{239}. Other possibilities are that the beta particle activity is caused by (1) some fission product which is carried by plutonium peroxide, (2) element 93, or (3) element 95 or 96 carried by plutonium peroxide.

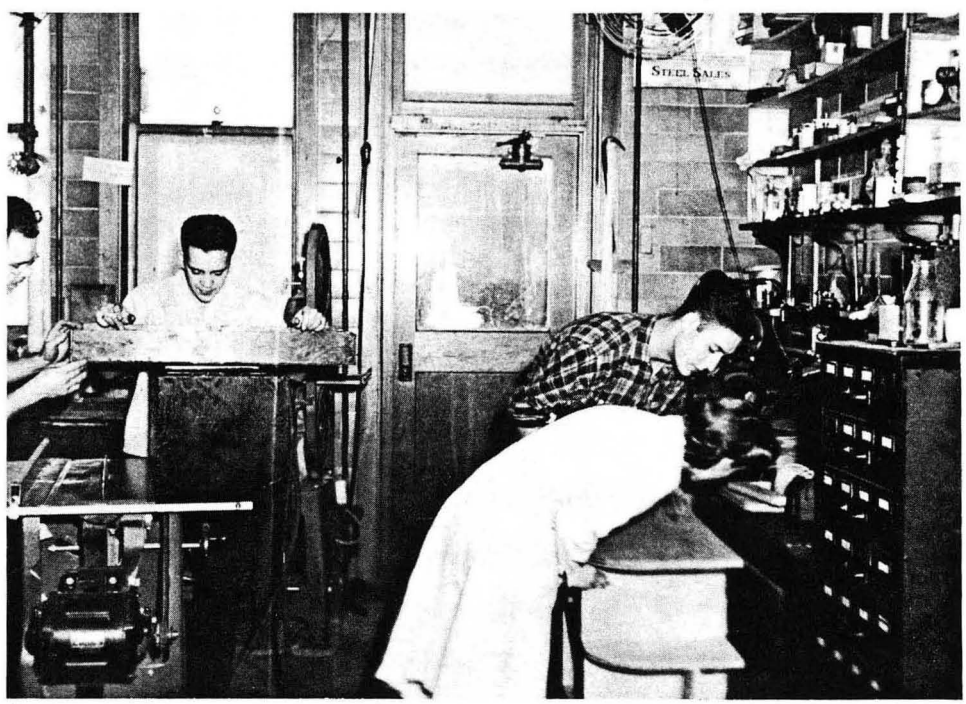


XBL 785-786

Figure 34. Section C-I room assignments in New Chemistry Building, Spring 1944.

XBB 783-3141

Figure 35. Bernie Donlan in Room 12, New Chemistry Building, Spring 1944.



XBB 783-3142

Figure 36. Reid Harding, Bob Young, Emil Fulan, and Kay Gavin in Room 12, New Chemistry Building, Spring 1944.

Westrum succeeded in producing plutonium metal by reduction of $PuCl_3$. The starting material was a sample of 150-250 micrograms, prepared by I. Karle by treating dried $Pu(OH)_4$ with CCl_4 at 750°C and resubliming from a furnace at 700°C. Westrum used a degassed inner and outer crucible system of beryllia with barium as the reducing agent. Loading of the crucible was carried out in Hagemann's dry box (a sealed metal box with heavy rubber gloves and an air lock) which was filled with nitrogen gas. Firing was at 1,240°C, and the result was an excellent yield of metal in the form of numerous very bright, shiny spheres in great abundance on the walls and sides of the inner crucible.

Cunningham sent a letter to Whitaker, attention English, in Oak Ridge, giving the alpha particle assay on a solution of plutonium tracer being sent to Clinton Labs for use in standardizing assay techniques.

Using a 93^{237} sample (lanthanum carrier) prepared by La Chapelle and Magnusson, Jaffey has been measuring the range of its alpha particles during the last couple of days. The thin sample is mounted on a platinum plate, and the range is measured by interposing thin aluminum absorbers and using a vacuum low geometry alpha particle chamber for detection. He finds a range of about 3.25 cm (equivalent air at standard conditions, 25°C and 76 cm). The measurements are made by comparison with a standard 94^{239} sample. The 93^{237} has a little 94^{239} in it that has been corrected for in arriving at the range of the 93^{237} alpha particles.

"Chemical Research--Separations Processes for Plutonium. Process Development and Chicago Semiworks Operation" (CN-1491) was issued covering work to about April 1, 1944. The following account is given: Hyman, Makins, Flox, and Winner have worked on the removal of aluminum bonded slug jackets. They find that part of the bond forms an alloy with uranium and remains as a scale on the slug after removal of the jacket either by 10% nitric acid containing mercuric nitrate (standard Clinton procedure) or by sodium hydroxide. The slug can then be dissolved in concentrated HNO_3, but the resulting solution contains most of this scale. Dislodging of the scale by acid or alkali treatment is found possible; and if the scale thus detached can be slurried from the dissolver, the final UNH solution will contain only small amounts of scale. The effects of the coating materials on the Bismuth Phosphate Process have been investigated; it is found that it does not materially alter the plutonium yields or the decontamination.

The semiworks has carried out eleven tests of the $HCl-U(C_2O_4)_2$ concentration method, with an average plutonium yield of 92% and average beta and gamma activity decontamination factors for the concentration step of two and five respectively. It is also reported that Margolis has worked on ether extraction, carrying out laboratory experiments to obtain essential fundamental data.

"Chemical Research--Special Chemistry of Plutonium. Report for Month Ending April 1, 1944" (CK-1512) was issued. The research activities reported are as follows: Jensen, Brody, and Stein summarize data obtained to date on the solvent extraction of inorganic plutonium compounds on a tracer scale. Sixteen of the solvents tested are found to extract less than 1% Pu(III) as the chloride from 5 M NH_4Cl, 1 N HCl. Of twenty-three

solvents tried, twelve extract more than 20% Pu(IV) as the nitrate from an equal volume of 10 M NH_4NO_3, 1 N HNO_3. Three solvents are found which extract more than 50% of Pu(VI) as the nitrate from an equal volume of 10 M NH_4NO_3, 1 N HNO_3.

E. Hyde, Davidson, J. Katz, and Wolf have treated oven-dried (70°C) plutonium hydroxide ($PuO_2 \cdot H_2O$) with HBr at 700°C and obtain a grass green $PuBr_3$ (2.5 mg sample). An attempt also has been made to prepare plutonium bromide by treatment of the hydroxide with NH_4Br at 350°C, but there is evidence that PuOBr is formed.

Fried and S. Katz find that reduction of PuF_4 with barium in beryllia crucibles produces plutonium metal that gives pycnometric density values consistently higher (17 to 21 gm/cm^3) than those of metal made in thoria. Zachariasen has obtained the same incomplete diffraction pattern (representing a complex, but unknown structure) from plutonium metal reduced with barium, whether the metal is made in thoria or beryllia. Fried finds that 100-300 microgram samples of plutonium metal made in beryllia or thoria react with purified hydrogen at room temperature to form a finely-divided, black powdery hydride; however, Zachariasen's x-ray analysis shows only the lines of ThO_2. Frank has made plutonium metal hardness tests and finds that metal made in beryllia crucibles (sample size from 0.116 to 0.324 mg) has an average Vickers hardness of about 260 (similar to that of uranium) in contrast to metal made in a thoria crucible with a hardness of about 100.

Jasaitis has carried out melting tests of plutonium metal using an electrically-heated tantalum surface in an evacuated chamber and finds that plutonium metal made in beryllia with a density over 20 gm/cm^3 melts at a temperature as low as 950°C. Fried and Westrum have tested the effect of heating time and temperature on barium reductions of PuF_4 in BeO and find that firing temperatures should be kept below 1,260°C. J. Karle has carried out further unsuccessful experiments on reduction of plutonium tetrafluoride by means of atomic hydrogen.

Hellman, Gerstein, and Frank have studied the reaction of various refractories with molten uranium and find that only BeO, La_2O_3, and Ce_2S_3 give uranium metal unchanged in hardness.

Helen went downtown. Afterwards she stopped in at Wilma's for coffee.

Britain and the U.S. have demanded that Sweden halt the export of ball bearings and raw materials and machines used in making bearings. Nazi ball-bearing production has been one of the prime targets of American daylight bombing in Germany.

Saturday, April 15, 1944

Today I received a very supportive letter from Robert G. Sproul, President of the University of California, dated April 11. He says, "In connection with the negotiations between the University and the United States Government with reference to Patent Case 61, I should like you to know that

in all the discussions which have taken place in the Board of Regents and between myself and Dr. Bush, the rights of the inventors, as well as the possible rights of the University, have been kept continuously in mind and vigorously supported. I thought that you might like to know this, and that it might prove to be a guide to you in the difficult decisions that you yourself will have to make in connection with this matter."

Thompson sent a memo to Vernon requesting shipment from Clinton Laboratories of three 750-ml samples of oxidized solution after by-product precipitation. These will be used for investigation of the Bismuth Phosphate Process under separation plant conditions.

Cunningham transmitted to H. H. Goldsmith, the Information Coordinator in Physics in Mulliken's office, a description written by James and English of the technique used in preparing thin uranium plates by electrolysis. According to this technique, a solution of $UO_2(NO_3)_2 \cdot 6H_2O$ in absolute ethyl alcohol is prepared. The electrolysis is performed at 20-25 volts, the current being allowed to vary as the deposition proceeds, until about 1 mg of uranium is plated out on platinum at 1 mg/cm^2 density from a volume of about 5 cc. The resulting plate of uranium is a thin yellow film of oxide which is very adherent.

Orlemann summarized for me his manpower assignments in his subsection II as of April 15, 1944. Group membership is shown as follows:

Group IV, Purification and Analysis of Plutonium Compounds: Jensen (Group Leader), Stein, Reinhardt, Brody, Dixon.

Group V, Volatility and General Dry Chemistry of Plutonium Compounds: Davidson (Group Leader), Hagemann, J. Katz, I. Karle, E. Hyde, Wolf.

Group V-A, Chemistry of Plutonium Fluorides: Orlemann (Group Leader), Heath, Florin, Meyer, Zvolner.

Group VI, Metal Production: Baumbach (Group Leader), S. Katz, Fried, Westrum, Frank, Jasaitis, J. Karle, Hellman, Gerstein. High Vacuum Technique Sub-Group: Simpson, Seifert, Phipps, Sears.

Dreher and Thompson attended the regular weekly conference on the ether extraction process, along with Kircher, Maloney, A. C. Hyde, G. Bird, Christiansen, H. Hoyt, and Tepe. According to the progress report on the semiworks operation, Run No. 4 of the continuous ether extraction system has been completed, but evaluation of the run is not yet possible. Future work will test the ether and aqueous extraction columns independently until good results are obtained. In addition, a rather extensive program of chemical experiments is being planned in an effort to discover the reasons for continued failure of the semiworks system to perform as was expected from the results of the fundamental chemical study.

Helen visited Wilma this afternoon.

Australian troops have captured Bogadjim on the north coast of New Guinea. They marched in without resistance. The bad news today comes from India, where the Japanese have cut off Imphal.

Sunday, April 16, 1944

I played golf during the morning. When I returned home, I received a telephone call from Kennedy who arrived in Chicago by plane at 2:00 a.m. He outlined to me Plan G for our patent application. Under this plan, we will request $1 million for our patents rights to plutonium. The University of California will receive one-half of the funds, to be allocated on a 50-50 basis for nuclear research in the Chemistry Department and the Radiation Lab. We will settle for less, perhaps go as low as $200,000 if necessary. If the settlement is more than $300,000, the University half of the funds will be used to establish a research corporation under the control of the four inventors. If the settlement is less, the funds will be divided four ways among the inventors. I invited him to come to the apartment at 8:30 p.m. to discuss these matters.

Helen and I had dinner at the Windemere West Hotel with the Drehers and Baumbachs.

Kennedy visited Helen and me in the evening, as planned. I said that I am favorable to Plan G and suggested that the money allotted to the University of California be held by a trustee. I also mentioned that Latimer is rather inclined to the idea of the inventors simply dividing the money. We discussed the Gofman paper on the discovery of 95 or 96. I mentioned that I had gone over the data when in Berkeley and had urged them to be more cautious in their claims. I said that I believe their unidentified activity is actually due to fission product yttrium.

Monday, April 17, 1944

C. Keith McLane started work as a Research Assistant in Section C-I. McLane has been a graduate student at the University of Wisconsin. Farrington Daniels describes him as an extraordinary student with a fine personality.

Ralph James completed another fluoride cycle on Sample 49NA. Ghiorso took beta particle absorption curves on the sample and also on an identical sample which James has prepared from some pure, non-bombarded plutonium obtained from Studier. The alpha particle activity of the two samples also has been measured. The absorption curves and the alpha/beta particle ratio of the two samples are identical. We therefore conclude that there is no long-lived beta-active plutonium produced in the Clinton pile by a one-month neutron reaction on 94^{239}. The electrons that have been observed in pure 94^{239} must be either conversion or Auger electrons and not beta particles.

Today L. B. Arnold submitted to the Section Chiefs and Group leaders a list of essential office equipment whose purchase is now restricted. Items such as pencils and typewriter ribbons may be purchased on the commercial market only in cases of emergencies and then in minimum quantities. The purchase of another 30 items for office use is forbidden unless they are clearly essential to the war effort. These include such things as carpets, pencil sharpeners, cuspidors, and smoking stands.

I received a response from Gofman to my April 11 letter in which I suggested that his new "activity" might be 57-day yttrium rather than element 95 or 96. Gofman said that, as a matter of fact, he has been considering the same possibility and that, chemically, it seems quite likely; however, it is too soon to be sure.

Cunningham outlined for me the work of his Basic Chemistry Group (Group VII) on certain phases of the co-precipitation problems of the bismuth phosphate extraction process. He says that, basically, the planned approach is to (a) determine the valence state of plutonium present at each step in the process and (b) determine the co-precipitation behavior of plutonium in this valence state under the conditions of the process at the given step. He states that, so far, the valence state of plutonium at each step in the process has been defined up to the point of the crossover. Crystalline precipitates of plutonium(III) phosphate and fluoride have been prepared and have been determined by Zachariasen and Mooney to be isomorphous with bismuth(III) phosphate and lanthanum(III) fluoride, respectively. Work is in progress on the preparation of crystalline plutonium(IV) phosphate and fluoride.

Cunningham also writes "Preliminary results suggest that the 'co-precipitation' of Pu(IV) with $BiPO_4$ is of an unusual type and does not appear to obey the laws of simple isomorphous substitution. However, extraordinarily large amounts of Pu(IV) (5% or more of the weight of the $BiPO_4$) may be precipitated with the $BiPO_4$ even from relatively dilute (0.03 g Pu/liter) solutions. Additional work will be necessary to elucidate this process more completely."

I received a copy of a memo from Szilard to Hogness proposing the testing of the purity of plutonium by observing the neutron emission of one gram of plutonium which is alloyed with ten milligrams of polonium. He goes on to say, "The neutron emission of this alloy would correspond to the neutron emission of about 500 grams of plutonium of the same purity. The polonium which is added would have to be pure but the percentage of the impurities permissible would be about twenty times as high as the percentage which is specified for plutonium. Ten milligrams of polonium correspond to an activity of about 30 curies. Such a quantity could be manufactured at X and probably obtained from one stringer of bismuth slugs after a few months of irradiation in the pile. If this proposal meets with the approval of those who are responsible for preparing pure plutonium it would be desirable to place an order for 30-60 curies of polonium with the Clinton Laboratories at an early date."

I received a copy of a letter from William Watson to Fulbright at Washington University in St. Louis in which Watson indicates that in the near future I may have samples from bombarded material which might conceivably involve another transuranium element and that he knows Fulbright will be interested in cooperating with me in obtaining the beta-ray spectrogram of any such sample.

Orlemann sent a memo to E. B. Ashcraft and H. L. Davis in Hogness' office about the selection of suitable materials for the storage and use of the reagents involved in the purification program for plutonium. He

explained that at the present time it appears that the preparation of pure plutonium metal will go through three steps. First, a purification by wet methods, i.e. precipitation and solvent extraction; second, conversion to an anhydrous halide; and third, reduction to, and purification of, the metal. In the latter two steps it seems most efficient to have the problem of testing and selecting suitable materials of construction remain part of the work being done on the development of the method for carrying out these steps because the techniques involved are more or less specialized. The problem of selecting suitable materials for carrying out wet methods of purification is another aspect of the problem of finding suitable materials for the storage of necessary reagents and for the analyses required in the preparation of pure plutonium. Quartz, pyrex, platinum, and tantalum are mentioned as materials to be studied. In a second memo to Ashcraft he gives the analytical needs of Section C-I for the past month and for the immediate future.

Watt informed Stearns that the Board of Regents, University of Texas, is extending his leave of absence from June 30 to October 31 of this year.

"Chemical Research--Separation Processes for Plutonium. Report for Month Ending April 1, 1944" was issued as document CN-1510. According to this report, Hoekstra has studied the effects of variations in flowsheet conditions in the extraction step of the Bismuth Phosphate Process in order to check the permissible limits of variations of process conditions. Site W concentrations of plutonium and twice Site W concentrations of fission elements were used on a 10-ml scale. It is found that broad variations can be tolerated in concentrations of HNO_3, H_3PO_4, and Bi(III). Hoekstra and Malm have carried out two 100-ml scale Bismuth Phosphate Process test runs (in glass equipment) through an extraction step, two decontamination cycles, and a crossover cycle which included a lanthanum fluoride by-product precipitation. Site W concentrations of plutonium and twice Site W concentrations of fission products were used. Over-all plutonium yields were 95% in the first run and 76% in the second run. Part of the product loss in the second run is accounted for by an operational error. A 10-ml scale run was carried through one decontamination cycle using two times Site W concentrations of plutonium and fission elements; product yield was 93%.

Ader has studied holdback carriers La(III), Y(III), and Ce(III) in the extraction step and finds that in every case where improvement in decontamination is obtained there is a decrease in product yield. Greenlee has tested oxidizing agents using tracer solutions of plutonium in 5 N HNO_3, testing Bi_2O_4, $Pb(C_2H_3O_2)_4$, $NaBiO_3$, and PbO_2-permanganate ion. All show promise.

Albaugh summarizes information available on the use of U(IV) in the prereduction step and on the decontamination cycles of the Bismuth Phosphate Process. Sedlet has carried out a number of experiments to see whether improved decontamination in the Bismuth Phosphate Process may be obtained through the use of lower process temperatures (50°C rather than 75°C). Yields were about the same at either temperature--no conclusions are drawn regarding the effect of temperature on decontamination.

Pye and Yett have tested the Site W concentration process and find that 0.2 N HF employed in the LaF_3 precipitation step gives only 75% carrying. Following metathesis, large insoluble residues are obtained which contain relatively large quantities of product. Goeckermann and Hopkins have tested a revised Site W isolation flowsheet on a 10-ml scale. Average yield in the first peroxide precipitation is 97.3% and in the second, 96.5%. Optimum conditions for precipitation of the peroxide appear to involve the use of 10% excess H_2O_2 added in four portions at 15-minute intervals.

"Chemical Research--Basic Chemistry of Plutonium. Report for Month Ending April 1, 1944" (CK-1511) was also issued. The following investigations were reported:

Inorganic Derivatives of Plutonium. O'Connor investigated the rate of precipitation and equilibrium solubilities of plutonium(IV) phosphates in H_2SO_4 and HNO_3. Among other things it is found that, as expected, plutonium(IV) phosphate is about 10 times as soluble in 1 M H_2SO_4 as in 1 M HNO_3. Cliff Smith studied the co-separation of plutonium(III) and lanthanum(III) as fluorides and finds it results in isomorphous incorporation of plutonium(III) in the LaF_3 lattice, according to results of x-ray examination by Zachariasen of mixed precipitates containing lanthanum-plutonium ratios of 2:1.1, 6:1, and 100:1. Howland prepared samples of sodium plutonium(VI) acetate and green plutonium(IV) peroxide for examination by Zachariasen who found the former to be isomorphous with sodium uranium(VI) acetate. The plutonium peroxide was found not to be isomorphous with uranium(VI) peroxide. Pye, Watt, and Yett studied the factors affecting the precipitation of plutonium(IV) peroxide and find its solubility continues to decrease with increasing H_2O_2 concentration in the range 0.6% to 7.5%. Both iron(III) and zirconium(IV) have an inhibiting effect on the precipitation of the peroxide. Howland undertook a determination of the composition of green plutonium(IV) peroxide and finds that there are two $O_2^{=}$ groups per atom of plutonium as shown by oxidimetric titration. Attempts to precipitate the peroxide of trivalent plutonium have failed.

Organic Derivatives of Plutonium. Dixon investigated the composition of plutonium(IV) acetylacetonate using one mg of plutonium and finds there are four acetylacetonate molecules per atom of plutonium as shown by a weighing assay. The coordination number of plutonium(IV) in this compound is eight.

Instruments. Ghiorso, using approximately 3 mg of plutonium repeatedly purified by fluoride cycles, peroxide and sodium acetate precipitation, and ether extraction, finds there is associated with Clinton pile plutonium a gamma-ray emission amounting to about 1 gamma per 8000 alpha particles (assuming 0.06% counting yield for the type of G-M tube used) and about 1 soft x-ray per 25 alpha particles, assuming 1% counting efficiency. Scott and Cunningham investigated the counting rate of U_3O_8 on platinum versus sample thickness. The counting rate varies linearly with the thickness in the range 0.17 to 0.10 mg per cm^2 but increases more rapidly at lesser thicknesses. For a thickness of 0.012 g U_3O_8/cm^2, the counting yield is about 52.7% in "50%" geometry counters.

Recovery and Service. The recovery group, composed of Asprey, Beard, Britain, Dawson, Leventhal, Stewart, Studier, and Wetlaufer, purified 1,430 mg of Clinton pile plutonium to greater than 99% purity as shown by spectrographic analysis. Of this, 38 mg of plutonium has been recovered, isolated, and purified from materials which had been used by other groups. An improved recovery method has been worked out which utilizes ether extraction after precipitation of the product with lanthanum fluoride.

At 9:30 in Room 209, Eckhart Hall, I attended the morning session of the April meeting with Dr. C. A. Thomas on the final purification and metallurgy of plutonium. Others present were Ashcraft, Chipman, Derge, Eastman, Foster, Hogness, Jeffries, W. C. Johnson, Kennedy, Selwood, Spedding, Warner, and later Compton.

Kennedy reviewed seven attempts to produce metallic plutonium at Site Y starting with PuF_3, PuF_4, and $PuCl_3$, with samples ranging from 50 mg to 1,000 mg. Calcium and lithium were tried as reducing agents while BeO, MgO, and BaO were used as refractories. Three of the attempts with $PuCl_3$ were by electrolysis with tungsten anode and cathode, using as solvent a fused mixture of $BaCl_2$-NaCl-KCl at 600°C. Success was mixed, but the electrolytic method is now considered very promising and will be given more attention than in the past.

After Eastman of Berkeley and Spedding of Ames finished with their presentations, I gave a progress report for my section; a summary of my review is as follows:

1. Methods of preparing plutonium metal. Yields of metal are greatly enhanced by forming the PuF_4 into a pill, using a hand-operated press to achieve a density of 6.2 gm/cm^3 (theoretical density is 7 gm/cm^3). A barium reduction of $PuCl_3$ in beryllia resulted in a highly satisfactory yield of plutonium metal. I suggested to Kennedy that the trouble experienced at Site Y from penetration of the product into crucibles might be eliminated by carrying out reductions at lower temperature.

2. Density of plutonium metal. Reduction with barium in beryllia gives consistently higher values than reductions in thoria. Metal with a density of 20.2 gm/cm^3 (barium reduction in BeO) has been shown by spectroscopic analysis to be strikingly pure with respect to barium and beryllium.

3. Melting point. Metal of high purity made by barium reduction of the tetrafluoride in beryllia is found to melt in vacuo at 950°C.

4. Hardness. Vickers hardness values are much higher (250-320) for metal prepared by barium reduction in beryllia than for metal made by barium reduction in thoria (100).

5. Vapor pressure of plutonium metal. The apparatus for preliminary measurements is practically completed. It is proposed to use a tantalum oven and employ the effusion method.

6. Properties of plutonium hydrides. X-ray diffraction studies have not yielded lines for samples of plutonium hydride. Plutonium hydride has been formed at room temperature by the reaction of hydrogen on a 250 microgram sample of metal made by barium reduction in beryllia. The formula calculated from this determination was $PuH_{4.0}$.

7. Preparation of a pure compound of plutonium. The results of solvent extraction methods were presented.

8. Preparation and properties of plutonium halides. On the basis of recent work, I gave answers to questions raised at the March meeting by indicating (1) H_2 will reduce PuF_4 to PuF_3; (2) pure HF will not oxidize PuF_3 to PuF_4; (3) if O_2 is present, the Pu(III) is oxidized to Pu(IV). Recent work on preparation and properties of the lower and higher fluorides, the chlorides, bromides, and iodides, was summarized.

A general discussion followed my presentation.

By noon I was home with a migraine headache. After resting, I felt somewhat better and returned for the afternoon session of the Thomas meeting, which convened in Room 209, Eckhart Hall. Ashcraft, Chipman, Derge, Eastman, Foster, Hogness, Johnson, Kennedy, Spedding, Thomas, and Warner returned also.

I presented a new possible flowsheet for production of pure plutonium metal. Changes since last month's flowsheet include the use of a solvent extraction method (ether and triglycoldichloride) as an alternate to the molybdate procedure. Also, BeO or La_2O_3 are suggested as the refractories in the reduction step, rather than ThO_2.

Ashcraft gave a report on the activities of Section C-IV, the topics being (1) "Progress on the method for determination of fluoride," (2) "Radioactive tracer studies on cupferron extraction procedure," and (3) "Accuracy of spectrographic determinations as used with cupferron extraction procedure." Chipman, Foster, and Derge reported for the Technical Division. Their subjects were, respectively, (1) "Progress of the Ceramics Group at M.I.T.," (2) "Production of pure uranium halides at Brown University," and (3) "Experiments on (plutonium) metal production."

Among the decisions reached and assignments made at the meeting are: (a) Research on plutonium metal production is more urgent at present than work on the production of a pure plutonium compound for use in metal production. The latter problem shows every promise of being solved on schedule. (b) It was suggested that the Chicago Chemistry Division should investigate the possibility of using metallic rhenium as a container material for melting product. (c) The Chicago Chemistry Division was asked specifically: (1) To attempt the reduction of a plutonium bromide, probably $PuBr_3$, during the coming month. (2) To determine the carbon content of $PuCl_3$ made by the action of CCl_4 vapor on $PuO_2 \cdot xH_2O$. (3) To obtain as much information as possible on the analyses of the various plutonium halides used for metal production. (4) To study the physical properties of product produced by the reduction of the trichloride. (5) To continue small scale scouting work on reaction, or lack or reaction, between various refractories and metallic plutonium. It was suggested that high melting point metals such as tantalum, tungsten, columbium, and rhenium be included in the search for suitable container materials, especially for use in remelting plutonium. The investigation of tantalum coated with TaN was suggested.

There was an evening seminar of the Chemistry Division at 7:45 in Room 251, Ryerson Laboratory.

The Russians are bombing and shelling Axis forces as they are fleeing Sevastopol. The Soviet forces are capturing Axis troops at the rate of 3000 per day.

Tuesday, April 18, 1944

General Groves is in Chicago.

I didn't have a chance to see Jasaitis before my report at the Thomas meeting yesterday. This morning he told me that he observed, on Saturday, the melting of a 500 microgram piece of plutonium metal on a tantalum strip in Robinson's vacuum line at about 835°C. The melting took place on top of plutonium previously melted and was aided by tapping the equipment to dislodge scum on the surface. Yesterday he used a 200-300 microgram piece and thought he observed melting, using the same technique, at as low a temperature as 810°C. In any case it seems safe to say the melting point is as low as 835°C.

Ralph James began working up the remainder of sample 49DC (1,931 microampere-hour bombardment of plutonium with deuterons at Berkeley, March 13-16) in a search for element 95 or 96 by the $K_2Cr_2O_7$ approach, on the assumption that it should be more difficult to oxidize to the VI, fluoride-soluble state, than is plutonium.

I received a copy of an April 17 memo from Mrs. Mooney to Allison, giving the results of x-ray examination of trivalent plutonium phosphate from a sample made by Cunningham's group. The compound was shown to be isomorphous with hexagonal bismuth phosphate, and to be closely similar to it in all dimensions. She went on to say: "It is highly probable that the mechanism of carrying product in the bismuth phosphate precipitation process is the formation of mixed crystals, the plutonium being carried in the trivalent state, despite its being originally in the solution in tetravalent state." She also observes that the close similarity in the dimensions of the two phosphates (Bi^{+3} and Pu^{+3}) explains why no measurable change in cell dimensions were found for bismuth phosphate precipitates carrying up to eight percent of the product. I believe that her suggestion that plutonium is carried in this manner is highly unlikely.

Manning sent Richmond an additional memo justifying our request for Kay Gavin's reclassification. He says that she has been given responsibility for handling non-technical problems which arise among Section C-I members and also for supervision of the girls who are laboratory assistants. Manning therefore recommends she be reclassified from Clerk C to Clerk B at a salary of $32.00 per week on a forty-hour basis.

In a memo to Compton, Hogness explains that my section will not be able to release Watt to go to Hanford unless we succeed in finding a suitable replacement to serve as an Associate Section Chief, a position we have been reserving for Watt when Willard goes to Hanford. Hogness says that Henry Taube of Cornell University, whom we have tried to get earlier, is the man we now want for this position and asks Compton if he would write a letter to Professor Debye requesting Taube's release.

4/18/44 (cont.)

I attended the Project Council Chemistry Information Meeting held in Room 209, Eckhart Hall. Others present were Apple, Ashcraft, Burton, Chipman, Compton, Connick, Cooper, Coryell, Davies, Doan, Eastman, Fermi, Greager, Hilberry, Hiskey, Hogness, Jacobson, Jeffries, W. C. Johnson, Kay, Kennedy, Leverett, McClinton, Mulliken, Newton, Perlman, Selwood, Shapiro, Sheline, D. H. Smith, Stearns, Sugarman, Sutton, Szilard, Thomas, Vernon, Warner, W. W. Watson, Wigner, and Zachariasen. Compton opened the meeting and called on Hogness to introduce the Chicago Chemistry Division speakers. I divided my presentation into three parts in the following manner:

1. Hanford Problems. Another 100-ml run has been made at Hanford concentration of plutonium and by-products. Stainless steel was present in the system. The run was carried from the extraction step through two decontamination steps and one crossover step with a yield of about 93%. A gamma-radiation decontamination factor of 1.5×10^5 was obtained. Under flowsheet conditions, carrying of the plutonium is good. The decontamination is somewhat short of that expected. On a large scale with synthetic solutions some deviation from flowsheet conditions can be allowed without much change in yield. Pb_3O_4 gives complete oxidation in a short time and may be considered an alternate to $NaBiO_3$ as an oxidant. It has been found that 5 to 10% K_2CO_3 added to KOH improves the metathesis.

In the isolation step we have had good results in precipitating the peroxide. The solution is kept hot with 1 N HNO_3 and 0.2 N H_2SO_4 and 10% excess H_2O_2 is added. Good precipitates that are filterable through sintered glass of medium porosity have been obtained. Arrangements have been made to ship "X" solutions to Chicago for testing the isolation and concentration steps.

More work has been done on the coating removal problem. Good progress has been made in finding reagents that will remove coating without the evolution of large amounts of hydrogen gas.

2. Los Alamos Problems. Our section has been making plutonium metal by the reduction of PuF_4 by barium. We previously used thoria crucibles but are now using beryllia crucibles. Plutonium metal produced in beryllia crucibles has shown 40 ppm of beryllium and <500 ppm of barium on analysis; whereas plutonium metal produced by reduction with calcium in thoria crucibles had a density of 16.3 gm/cm^3, plutonium metal produced with barium reduction in beryllia crucibles has shown densities as high as 20.5 gm/cm^3 by the capillary displacement method. Metal produced by barium reduction in beryllia has a hardness slightly greater than that of uranium and melts at 950°C, possibly lower; the melting point may be as low as 835°C.

I was interrupted by Fermi, who asked how much plutonium metal has been produced; he also wanted to know when a piece weighing, say 100 mg would be available. I replied that we are trying to produce pieces of this size now and probably will succeed within the next month. Thomas added that a 156-mg piece was the largest single piece yet produced at Site Y but that this piece was probably relatively impure.

I continued with my report. Plutonium trichloride ($PuCl_3$), I said, has been prepared by a "wet" method. It also has been prepared in a dry method by the action of CCl_4 vapor on PuO_2. Zachariasen reports that both samples are relatively pure, anhydrous $PuCl_3$. The "wet" method permits

the preparation of the halide at a low temperature which should be less troublesome from the standpoint of the introduction of impurities. Plutonium tribromide has been made by the action of bromine on metal. Zachariasen reports that this bromide contains approximately 10% of chloride. The chlorine is probably introduced as an impurity in the bromine. Plutonium bromide has also been made by a "wet" method. The purity and exact composition of these bromides is being investigated. Work has continued on methods for purification. The solvent extraction still looks very promising.

3. Basic Chemistry. Work in this sub-section has been directed toward study of the valence states in each step of the mainline process. If, in the pre-reduction step, solution is heated up to the proper temperature without formic acid being added, most of the plutonium is oxidized to +6 state by the HNO_3. If formic acid is added and the heating is prolonged, some of the plutonium is still oxidized to the +6 state. When Fe^{++} is added in reduction step, plutonium is reduced to +3 state. When reheated to 75° plutonium is oxidized back to the +4 state. The phosphate of +3 valent plutonium is isomorphous with bismuth phosphate; one would like to believe that product is carried in the +3 state, but the evidence from Cunningham's work makes this seem unreasonable. It appears that +4 valent plutonium is being carried, but the reason for this is not understood.

In the discussion that followed, Zachariasen said he is inclined to believe carrying is in +3 state. Hogness suggested an adsorption mechanism. Zachariasen pointed out that PuF_3 is isomorphous with LaF_3, and he believes that plutonium carried by LaF_3 must be in the +3 state.

I then stated that we are trying to produce a sample of the phosphate of +4 plutonium to submit for x-ray diffraction analysis. Doan inquired as to what difference there was in carrying of +3 and +4 valent plutonium, and I stated that there is an expected difference; that +3 valent plutonium, according to conventional wisdom, should be carried in almost any ratio, but the carrying of +4 valent plutonium should fall off as the ratio of plutonium to carrier increases. Sutton inquired concerning the valence state of plutonium after formic acid treatment, and I replied that under the conditions used formic acid yields +4 valent plutonium. I mentioned that PuF_4 is carried by LaF_3 and said that it might be due to the formation of $PuF_4 \cdot 2LaF_3$. I suggested that x-ray diffraction studies should be made.

I concluded my report by saying that further studies have been made on gamma rays and x-rays from plutonium. The gamma ray is 0.4 Mev rather than 0.6 Mev as indicated by the preliminary investigation. The previous result of 1 gamma ray for every 10^4 alpha particles has been checked. Also, there are x-rays in the 50 to 100 Kev region. We suspect there is a 65 Kev gamma ray with conversion. There seem to be some very soft electrons also. It is postulated that plutonium decays to an excited state followed by a 65 Kev gamma ray which is largely converted and a 0.4 Mev gamma ray which is not converted.

Burton and Sugarman followed me on the program to review the work of their sections, Section C-II and Section C-III, respectively. Compton next called on Doan, W. C. Johnson, and Greager to introduce the Clinton Laboratory speakers.

Perlman was the first to be introduced. He spoke about his group's study of the various phases of the Bismuth Phosphate Process as applied to Hanford problems. In general, results agree with those reported by Thompson and me. All runs were made on plant solutions, and results obtained on the extraction step have been more erratic than those obtained on synthetic solutions. Losses of product have been ~0.1% for each by-product precipitation and ~1-2% for each product precipitation. In general, the process looks good except for occasional erratic results. In the crossover step, using 5 liters of plant solution, the overall loss has been ~2%.

Perlman has followed up Cunningham's work by studying precipitation of peroxide from plant solution. Precipitation of peroxide from plant solutions in presence of SO_4^{--} helps produce a good yield. If SO_4^{--} is present the solubility of peroxide appears to be 1/2 the solubility in absence of SO_4^{--}. The precipitation of peroxide has been carried out in the presence of 1 M, 0.1 M, and 0.01 M SO_4^{--}. He believes that the +4 valent plutonium peroxide is the insoluble one and that the peroxide precipitated from +4 valent solutions is not the same as the peroxide precipitated from +3 valent solutions. Thus far his group has not been able to find any SO_4^{--} in the peroxides precipitated in presence of SO_4^{--}. It is believed that the influence of SO_4^{--} is due to the fact that it complexes with +4 valent plutonium and favors this valence state in solution. Studies on absorption spectra show that the intensity of absorption due to the +4 state is increased by addition of SO_4^{--} to the process solutions.

Of the speakers who came after Perlman, reports were presented by Davies on hot laboratory activities; Shapiro on studies concerned with extraction and by-product precipitation of Hanford solutions and with steady-state peroxide concentrations; Apple on the influence of variables on percent of plutonium going to waste in the extraction step; and Sutton on Hanford separations development studies, improvement in decontamination (Hanford), improvement in yield (Hanford), flocculation studies (Hanford and Clinton), and crossover and isolation (Hanford).

Fermi next took Compton's place as chairman and called on Eastman and Connick as the Berkeley speakers. Eastman spoke on refractories and plutonium metal production. Connick's topic was the thorium oxalate process and basic chemistry. Fermi next called on Newton of Ames, who spoke on the applications of complex formation to decontamination and purification of plutonium. Kennedy, representing Site Y, was the last speaker of the meeting to be introduced by Fermi. He chose for his review, experiments on plutonium metal production and the production of halides of plutonium, remelting studies, and shot-gun methods of analysis. Fermi inquired about the sensitivity of the neutron counters in my section, and I replied that they have a 1% efficiency now and we expect to get a 5% efficiency, that is, we expect to count 1 neutron per minute. When Fermi asked Kennedy what purity has been obtained with the plutonium metal so far, he answered that no metal made thus far is believed to be very pure.

The "Chemistry Division Summary Report for March, 1944" (CS-1557) was issued. The work of my section is summarized in the first 15 pages under the headings "Separation Process Studies," "Purification and Metal Production," and "Basic Chemistry." The remaining 11 pages are devoted

to the work of the other five sections: Section C-II, Radiation Effects (Milton Burton, Section Chief); Section C-III, Fission Product Chemistry (N. Sugarman, Acting Section Chief); Section C-IV, Analytical (E. B. Ashcraft, Section Chief); Section C-V, P-9 (C. F. Hiskey, Section Chief); and Section C-VI, Pure and Rare Chemicals (L. B. Arnold, Jr. in charge).

I attended an evening meeting at 7:45 in Room 209, Eckhart Hall, called by Compton to discuss a program for alternative chemical separation process development. Others present were Arnold, Boyd, Chipman, Connick, C. M. Cooper, Coryell, Doan, Eastman, Greager, Hilberry, Hiskey, Hogness, W. C. Johnson, Kay, Kircher, Leverett, Maloney, Manning, Perlman, D. M. Smith, Sugarman, Sutton, Thompson, W. W. Watson, Whitaker, and Willard.

Compton explained that the purpose of the meeting was to consider plans for alternative processes, with special reference to adsorption processes, that could be used at Hanford in case the straight phosphate method is not successful.

Boyd described the adsorption method as now developed at Clinton Laboratories and stated that he believes an all adsorption process could be realized.

Compton asked about the degree of confidence in the present Hanford process, to which I responded that from the laboratory point of view the phosphate process looks "entirely all right." I pointed out that the present adsorption process is aimed at the extraction step and, should a phosphate method fail here, there are many other carriers that could be used. Greagor pointed out that a good basis for confidence in the phosphate process at Hanford is that the last six semiworks runs gave 85% yield through the crossover step. After Burton stated that the levels of radiation anticipated at Hanford will not interfere with the first steps of the present Hanford process, Compton summed up the situation by saying that we now have assurance in the laboratory work of Seaborg and Burton that both concentration and radiation factors will not interfere.

At Compton's request, Hogness reviewed his memo on alternate process developments. He expressed his feeling that the present process seems like a hard way to do the job although we have no real choice for current development. In his opinion, it seems probable that much simpler, continuous processes can be devised. Solvent extraction may be the most likely, but adsorption has strong possibilities. Dry processes are attractive but because of the special conditions may be most difficult. I expressed the view that we should concentrate our process effort on phosphate and put the balance of our time on studies of fundamental chemistry.

Compton summarized the discussion by saying that we are agreed to bank on the phosphate method for Hanford; that alternative processes when worked on should be fundamentally sound; that perhaps our most immediate alternate process work should be aimed at recovering uranium for re-use; finally, that alternatives are entirely subordinate to the mainline process.

Helen went to Dagmar Dreher's for lunch and to Wilma's for coffee.

4/18/44 (cont.)

Today's paper says that the Russians are only one mile from the Bay of Sevastopol. British and Indian troops are dealing heavy losses to the Japanese near Imphal.

Wednesday, April 19, 1944

Just two years ago on April 19, Perlman and I arrived in Chicago via the "City of San Francisco."

Walter J. Blaedel began working in Section C-I today. Blaedel graduated from UCLA in 1938 and received a Ph.D. from Stanford in 1941. Since that time he has been an instructor at Northwestern University. He is being assigned to Dreher's group and is rated an Associate Chemist at $3,900 per year.

Hogness left on a trip to the East today. His itinerary includes the following: Thursday and Friday, C. A. Kraus at Brown University; Friday and Saturday, F. H. Norton at M.I.T.'s Department of Ceramics; Saturday through Tuesday noon, H. C. Urey at Columbia University; and Tuesday and Wednesday, J. B. Conant in Washington, D. C. Hogness is visiting Conant to get his comments and criticisms of the future long-range program here at the Met Lab as proposed recently by Compton in a letter to Groves. He also wishes to discuss his own suggestion for a national institute of science.

I wired Darrell Osborne in Arlington, Virginia, that I shall be in Washington April 20 and might phone him. Kennedy and I are going to Washington to try to straighten out matters concerning our joint patents.

Willard, Cunningham, Thompson, and Howland met to discuss continuation of the research begun by Hindman's Basic Chemistry Group on elucidating the chemistry of the mainline process and to extend the program for the purpose of obtaining fundamental information which might suggest lines of attack for improvement of the flowsheet. It was decided to divide the work between Cunningham's and Thompson's sub-sections. The following assignments were agreed upon: mechanisms of carrying, Smith and Malm; solubilities of plutonium in process solutions, O'Connor; solubility of bismuth phosphate under process conditions, Peterson; stability of Pu(III) in the extraction and decontamination steps, Howland; and prereduction and extraction steps (possible oxidation of Pu(IV) to Pu(VI) by hot UNH), Malm and Howland.

At 3:00 p.m. Joe Kennedy and I left Chicago by train (Penn) for our trip to Washington, D.C. As I left Helen was planning to celebrate my birthday by having dinner with the Ghiorsos.

There was a Project Council Policy Meeting at 9:30 a.m. in Room 209, Eckhart Hall. Present were Chipman, Compton, C. M. Cooper, Dempster, Doan, Eastman, Fermi, Greager, Hilberry, Hogness, Jacobson, Jeffries, W. C. Johnson, Kay, Kimpton, McKinley, Mulliken, D. M. Smith, Spedding, Stearns, Vernon, Warner, W. W. Watson, Whitaker, and Wigner. Compton opened the meeting with his customary "State of the Nation"

message. He remarked that the main program will continue at about its present level--except for health studies which must increase as the scale of operation increases. The present level is considered to be about what is needed to provide for the war effort. Our primary effort is on Hanford Engineer Works, then on Los Alamos. By the end of the year the emphasis is expected to shift toward Los Alamos, but release of pressure should make it possible to develop critical information needed to intelligently plan our future program. Proper items of a program must include improvements that are directly useful in the war program; it is not appropriate to expand facilities for work looking far down the line. Compton looks on the present staffs as shock troops who must be used as effectively as possible during the period while successful full-scale production is getting underway, but may be swung into action on emergency problems if they materialize. Compton announced that the cooperative P-9 (heavy water) pilot plant project with the British has been approved. The direction of the program will be a British responsibility and the laboratory will be manned by Britons, Canadians, and Americans. Compton also announced that Smyth has been taken on by General Groves to prepare a report on the activities of all DSM projects for general use when and if it becomes desirable. He reviewed yesterday's meeting concerning alternate separations processes and stated these conclusions: (1) Bismuth phosphate is being counted on for the Hanford Engineering Works. (2) Alternatives should be improvements rather than substitutes for phosphate. (3) Alternatives should aim at conservation and recovery of uranium. (4) Alternatives in general do not appear at the moment vital to the winning of the war.

Hogness gave the Chemistry Division report. He indicated that at least 35 men are expected to go to Hanford and that we are losing many key men who must be replaced. He also mentioned that Hiskey has been called into the Army and the remainder of Section C-V will probably be transferred to Section C-VI. Others reporting at the Policy Meeting were C. M. Cooper (Technical Division), Greager,(Clinton Separations Development Division), W. C. Johnson (Clinton Chemical Division), D. M. Smith (Clinton Analytical Group), Doan (Clinton Engineering Group), Whitaker (Clinton Laboratories in general), and Eastman (Berkeley).

Kay stated that the Clinton pile is operating at 1,100 kw. The measured losses from Building 205 (the chemical plant) are now running about 13%. The cleanup of chemical equipment removed most of the plutonium but failed to recover about 2 grams of material which, by analysis, should have been there. It is believed there were errors in the analyses.

Thursday, April 20, 1944

Kennedy and I arrived in Washington in the morning and at 10 a.m. went to Captain Lavender's office on the second floor of the Carnegie Foundation Building at 1530 P Street. He took us to see Vannevar Bush in the same building, who spoke to us about President Roosevelt having directed

him to obtain all controlling patents in these matters to facilitate international dealings. Bush emphasized they have no intention of coercing us; however, he said, since the University of California is becoming so demanding, he is forced to be legal and tough with U.C.

Afterwards, Lavender took us over to his other office in the Navy Department, located in Tempo 3 on Constitution Avenue and 17th Street (part of the Navy Department complex which houses the S-I Section). There, we talked over the scope of the inventions with Lavender, Lt. Cmdr. Roland Anderson, and Richard Radue.

After lunch at the Army-Navy Club with Lavender, Anderson, and Radue, we continued the discussions. We convinced Lavender that we, not the University of California, should be dealt with and indicated that we feel that our plan (we have in mind Plan G) would satisfy the University. Kennedy and I explained that we want to ensure support for our type of research after the war and for this reason we are not interested in small sums. We asked $1 million for Case 52 and an equal amount for Case 61, if it were just as valuable. Lavender felt that without further study he could offer a nominal sum of, say, $50,000 total. Kennedy agreed to brief Segrè and Wahl on our entire conversation. We asked Lavender to make a further study in order to arrive at a more reasonable offer, pointing out that we do not wish to appear difficult and would prefer to accept his studied estimate of the actual value of the application to any other course. After we left, Kennedy remarked that Lavender seemed not to care who receives the money (the University or the inventors). As Kennedy and I parted at the train station I agreed to telephone Shane at Berkeley, explain the whole matter, and ask for the University's cooperation. I then caught the late afternoon train back to Chicago.

Friday, April 21, 1944

I arrived in Chicago from Washington, D.C., at 9:00 this morning.

While I was away the triweekly Wednesday evening meeting of the Basic Chemistry, Recovery, and Instrument Groups (Sub-section III) of my section met at 7:45 in Room 209, Eckhart Hall, attended by Albaugh, Cunningham, Davidson, Dawson, Hindman, Howland, Hyde, Kirk, Kraus, Krueger, Larson, Malekow, Malm, Manning, Mooney, O'Connor, Orlemann, Perlman, Peterson, Rosenfels, Scott, Sheel, Simpson, Clifford Smith, Studier, Thompson, Wetlaufer, Willard, Wolf, and others. Cunningham opened the meeting and said that the meeting would be devoted to the problems concerning the valence numbers of plutonium during the various stages of the Bismuth Phosphate Process and the mechanism of carrying.

Howland stated that, on the basis of measurements of potential, the chances of having Pu(III) in the prereduction and extraction steps is very small at elevated temperatures and that, therefore, it would be necessary for Pu(IV) to be carried. During reduction with ferrous ion in process solutions, it was found that reduction of Pu(VI) to (Pu(III) is practically instantaneous at 25°C and that heating to 75°C causes reoxidation to Pu(IV). The results of a series of carrying experiments at bismuth-plutonium ratios by weight of 54:1 to 9:1 suggest a solid solution of plutonium(IV) in the

bismuth phosphate. A number of experiments regarding the lanthanum fluoride cycle were reported, including some that suggest that plutonium in reduced process solutions is present largely in the IV state.

Mrs. Mooney reported on the recent work on the crystal structure of various phosphates and the mechanism of carrying. Because of the close similarity in the dimensions of the unit cells of bismuth phosphate and plutonium phosphate, she is of the opinion that Pu(III) has to be carried. She does not see a reasonable mechanism for the carrying of Pu(IV). Although it is true that the bismuth phosphate network contains channels around the hexagonal axes and that in these channels are sites which could accommodate at the same time an equivalent number of negative ions, such a picture becomes very involved. During the discussion Cunningham inquired if the distribution ratio of plutonium between the bismuth phosphate precipitation and the solution could be estimated, but no such estimate was reported. Answering a question by Manning, Mrs. Mooney estimated that the upper limit of Pu(IV) carrying would be in the atomic ratio of 1:5 but pointed out again that the mechanism proposed is very unsatisfactory. She repeatedly raised the question as to whether the reduction of Pu(IV) to Pu(III) could at all occur in the solutions. The heated discussion that followed brought no conclusions.

Yesterday Dreher attended the weekly conference on ether extraction with Maloney, A. C. Hyde, Bird, Christiansen, Buffum, Holt, and Tepe. No experimental runs were made during the week, pending the results of chemical experiments to determine suitable oxidizing agents. These experiments show that hydroxylamine sulfate would be suitable for use as the reducing agent in the aqueous extraction column.

The evening meeting of the Research Assistants of my section was held last night at 6:45 p.m. in Room 209, Eckhart Hall. There was also an evening meeting of project-wide scientific personnel at 7:45 p.m. in Room 133, Eckhart Hall. Stearns talked on incipient effects of radiation.

Yesterday Helen had Dagmar and Wilma over to lunch and then had dinner at the Ghiorsos'.

Today, Ralph James, working on Sample 49DC (1,931 microampere-hour Berkeley bombardment of plutonium with deuterons), has found that after repeated dichromate cycles (lanthanum fluoride precipitation from $K_2Cr_2O_7$-oxidized solution, followed by metathesis with NaOH, dissolving in HNO_3, and again oxidizing with $K_2Cr_2O_7$) the alpha particle activity removed by this oxidation procedure has apparently fallen to zero, leaving a small level of alpha activity. This would indicate the presence of an alpha particle activity that is not oxidized to a fluoride soluble state by prolonged treatment with $K_2Cr_2O_7$. This could not be plutonium or neptunium; and, since thorium should be absent, the effect, if it persists through more cycles, is very probably due to an alpha-active element 95.

Helen and I played cards in the evening at Ghiorsos'.

Chinese troops fighting under Lt. General Joseph W. Stilwell have encircled a Japanese division near the jade mines of North Burma according to a war bulletin mentioned in today's newspaper.

Saturday, April 22, 1944

This morning I went to Jasaitis' lab to watch as he flattened two small pieces of plutonium metal (a few hundred micrograms), then fused them together on a tantalum strip covered with some $BaCl_2$. I was so impressed that I brought Jake Warner to the lab so he could watch as Jasaitis went ahead and remelted the fused piece; Jake was also intrigued.

James carried out additional dichromate oxidation cycles on the solution from Sample No. 49DC. A black crud formed during these operations. Pressure (range) curves on this material and on a pure plutonium sample shows that the alpha activity in the crud has the same range as Pu^{239} and is probably due to that isotope.

Willard sent a memo to Stearns regarding the extension of Stan Thompson's leave of absence from the Standard Oil Company of California. It appears that Standard Oil is willing to consider extensions until July of next year but will require a formal request every six months. Because Thompson will be going to Site W later in the year and will be on the du Pont payroll, Willard suggested that subsequent requests to the Standard Oil Company come from Stearns' office. Such requests will be needed in August of this year and again next February.

Helen and I went shopping this evening. Later the Baumbachs and Zene Jasaitis dropped in to the apartment for coffee.

War news in today's paper gives priority to the suggestion to set up a post-war world money plan. The purposes of the plan are to promote stability of nations' currencies and to facilitate world trade.

Sunday, April 23, 1944

The big headline in today's Sun reads "Finns Reject Second Peace Bid." The Finns say that they cannot meet some of the demands of the U.S.S.R.

I played golf in the morning and early afternoon while Helen had lunch with Wilma. In the evening Helen and I saw the movies, "Gang's All Here" and "The Lady" at the Frolic Theater at 55th and Ellis. Alice Faye and Carmen Miranda, along with Benny Goodman's orchestra, were featured.

Monday, April 24, 1944

In an experiment begun last Wednesday Fried has repeated, under more careful conditions, his experiment to determine the formula of plutonium hydride. The hydride was formed by treating 264 micrograms of plutonium metal with hydrogen at room temperature (28°C), and the amount of hydrogen absorbed corresponds to the formula $PuH_{2.8}$. During the course of the experiment he heated the hydride to determine its degree of decomposition at various temperatures. At the highest temperature, 360°C, the compound was about 16.6% decomposed.

4/24/44 (cont.)

Eleese Bowman, one of our laboratory assistants, transferred to the mail room today.

The evening seminar of the Chemistry Division was held at 7:45 in Room 209, Eckhart Hall.

Helen had coffee with Wilma.

The newspaper today reports new invasions in the Pacific on New Guinea. This is a 500-mile advance.

Tuesday, April 25, 1944

James carried out two more dichromate oxidations and lanthanum fluoride precipitations on the "crud" he obtained on Saturday after repeated dichromate cycles on Sample 49DC (1,931 microamphere-hour bombardment at Berkeley of plutonium with deuterons). Only 954 alpha particle counts per minute remain, and the activity is still behaving like plutonium. A range-curve will be run on the sample by Jaffey.

James recounted some of the samples prepared during the processing of sampel 49DC, in order to check for growth or decay of alpha particle activity. Results indicate no large growth or decay. He plans to count the samples again in a few days to see if the small apparent changes are real.

In order to check the possibility that element 95 has only a fluoride-soluble oxidation state, James decided to go back and investigate the plutonium-containing supernatant remaining from the first lanthanum fluoride precipitation after dissolving and oxidizing Sample No. 49DC. The plan is to carry out repeated precipitation of fluorides from reduced solution in order to remove all plutonium. If element 95 is present and if it is fluoride-soluble, this procedure will gradually separate it from the plutonium.

The "Metallurgical Laboratory Progress Report for March 1944" (MUC-SKA-603) was issued. Cited as an important development in chemistry is that "the density of our product in metallic form is higher than previously estimated. Metallic globules of density as high as 21 have been made." The work of my section is summarized as follows:

Studies on the removal of the aluminum can and bond (both aluminum-silicon and zinc processes) are continuing. Although a completely satisfactory process has not been evolved, present methods can be used.

Hanford flowsheet extraction steps have been studied for sensitivity to variations in conditions that may be encountered in practice and found to be sufficiently insensitive to: (a) nitric acid concentration, (b) phosphoric acid concentration, (c) Bi(III) concentration, (d) time of digestion, and (e) rate of addition of H_3PO_4. Similar studies of the oxidation step showed tolerable deviations in: (a) $NaBiO_3$ concentration, (b) time of oxidation, and (c) temperature of oxidation. Studies on Clinton flowsheet (with W concentrations) showed an increase from 1.1 to 12.8% loss in product with 0 to 0.6 M Fe(III) concentration in the extraction step. A 100 cc run, in which stainless steel was used in contact with the solutions, was made

and included an extraction step, two bismuth phosphate cycles, and a cross-over including lanthanum fluoride by-product precipitation. A 93% overall produce yield was obtained and is considered satisfactory.

Other studies active during the month include bismuth phosphate research--new oxidizing agents, U(IV) as a reducing agent, crystal structure of bismuth phosphate containing 94, ether extraction and decontamination, concentration by fluoride and alternates, and isolation by peroxide method. Studies on purification and metal production by solvent extraction continue to show promise. Work on the halides continues. Plutonium metal reduced by barium from PuF_4, in a beryllia crucible, gives pycnometric density values consistently higher than those from thoria crucibles. Values range between 17 and 21 gm/cm^3 leading to the belief that the density of the pure metal is above 20 gm/cm^3. Although metal made in beryllia or thoria reacts with hydrogen, it has not been possible to obtain x-ray diffraction photographs of the hydride. Hardness of the plutonium metal made in beryllia is about that of uranium rather than copper, as previously reported of metal made in thoria. The melting point of plutonium metal with a density over 20 gm/cm^3 has been observed at 950°C.

Basic chemistry during the month covered determinations of oxidation potentials of Pu(IV) - Pu(VI); acetate complexing factor ($\sim 10^9$), solubility of plutonium phosphates, isomorphism between Pu(III) and La(III) fluorides and NaPu(VI) and NaU(VI) acetates, absence of isomorphism between Pu(IV) and U(VI) peroxides, hydrolysis and solubility of Pu(IV) and Pu(VI) sulfate and perchlorates, and composition of peroxides. Further work on the valence states of plutonium at high concentration in the prereduction step of the Site W process was carried out to determine if the plutonium phosphate would precipitate by itself, and it is found that excess HNO_3 (0.8 millimoles) results in Pu(VI) whereas no excess and excess formic acid results in Pu(IV). Although the latter conditions cause precipitation of plutonium phosphate, the concentration remaining in solution is above that of Site W levels. A redetermination of α-n count from PuO_2 (1 g) + C (2 g) confirms previous neutron yield figures. An examination of the radiation spectrum from a 3 mg plutonium sample shows one gamma photon per 8,000 alpha particles and one soft x-ray photon per 25 alpha particles. Absorption curves indicate 0.3 Mev and 50 Kev energies for the γ and 51, 18, and 12 Kev for the x-rays, and some low energy electrons.

Total expenditures for the month of March for the Laboratory amount to $761,492. Personnel employed on March 31, 1944, total 1,585 (505 academic), a net increase of 158 during the month.

Helen and I had dinner at the Howes' (1321 E. 56th).

This morning's headline reads, "Hollandia Taken by Yanks." The troops found this city on New Guinea abandoned.

Wednesday, April 26, 1944

This was the last day in Section C-I for Jack Flox, Ralph Gardner, and Bert Schoenberg. They are transferring to the new Radiation Protection Group (Section C-V).

Lincoln sent Willard a detailed proposed plan of action to investigate the poor carrying of plutonium in the extraction step, tentatively attributed to oxidation of the plutonium on heating the UNH solution from the semiworks dissolver. The proposals are as follows: A. An investigation of analytical techniques planned to parallel as nearly as possible the work which Howland has in his program. B. Application of a satisfactory analytical technique to the further investigation of the difficulty in the bismuth phosphate extraction step. C. Methods of eliminating the difficulty encountered in the bismuth phosphate extraction step.

Jaffey has repeated his range measurements on the 93^{237} alpha particles using another thin sample of 93^{237} (lanthanum carrier) from which 94^{239} has been further removed by chemical separations. Completing his measurements today using the vacuum low geometry alpha particle chamber with thin aluminum absorbers and comparison with 94^{239} alpha particles, he finds a range for 93^{237} alpha particles of 3.26 cm (equivalent air at 15°C and 76 cm), corresponding to an energy of 4.8 Mev.

I met with Manning, Watt, Thompson, and Willard to plan the concentration-isolation program for the immediate future. Among the items agreed upon were:

1. Determine whether or not the use of a better holding oxidant, such as MnO_4^-, will reduce plutonium losses when the lanthanum fluoride by-product precipitation is carried out in the crossover step in stainless steel.

2. Study the use of especially pure KOH in the metathesis step as a means of avoiding impurities which may contribute to the "crud" problem.

3. Secure more data on the isolation process operated in 25-12 stainless steel.

4. Determine the behavior of Pu(VI) and Pu(III) under the conditions employed in the plutonium peroxide precipitations in the Hanford Isolation Process.

The second orientation lecture for new employees at the Met Lab was held at 7:45 p.m. in Room 133, Eckhart Hall.

I attended the triweekly evening meeting of the Separation Processes Sub-section I of Section C-I, at 7:45 p.m. in Room 209, Eckhart Hall. Others present were Albaugh, Arnold, Beard, Dawson, Dreher, W. R. Diver (a du Pont man from the Technology Division), Flox, Fred, Gilbreath, Greenlee, Haeckl, Howland, Hyman, Katzin, Kirk, Larson, Malm, Manning, Margolis, Morgan, Post, Rosenfels, Sheft, Simpson, Thompson, Watt, Watters, Willard, Winner, and Yett. Thompson introduced Diver who gave a detailed description of the plant layout at Hanford. The total area of the site is 631 square miles. There will be two chemical extraction plants or canyons in the 200 area, each 810 feet long and 70 feet wide and containing 40 cells. Bulk reduction will take place in the 224 building and final concentration and isolation in the 231 building. The control labs are housed in the 222 building.

Helen spent much of today at the YWCA.

4/26/44 (cont.)

War news these days mainly emphasizes victories; so it is a shock to read in today's paper that 2,300 airmen--British and American--have been lost in eleven days' raids.

Thursday, April 27, 1944

Richard Rosenfels has transferred to Section C-IV.

Hogness returned to Chicago at 9 a.m. from his trip to Brown University, MIT, Columbia University, and Washington, D.C.

Ralph James completed the series of LaF_3 precipitations from the SO_2-reduced solution of sample 49DC in order to see if any significant fluoride-soluble alpha particle activity would remain in the solution. The final solution contained large amounts of salt and could not be counted, but James estimates that the fluoride-soluble alpha particle activity, which is still behaving like plutonium, is no more than 350 counts per minute and may even be less. James decided to next attempt to find element 95 in sample 49DC by carrying out regular bromate cycles. This procedure assumes that 95, like 93, might be more rapidly oxidized by $KBrO_3$ than is plutonium. Thus, from a bromate-oxidized solution, the plutonium would be carried with lanthanum fluoride, whereas element 95 would not.

Margolis told me that during the past week he and Blaedel have carried out chemical experiments on ether extraction of plutonium to compare absolute ether and fat-extraction grade ether. They also studied plutonium distribution at equilibrium between UN-HNO_3-H_2O and UN-HNO_3-ether.

Hamilton wired that a sample of 5×10^5 alpha particle counts per minute of U^{232} is being sent to me from Rochester today.

There was a meeting of Albaugh, Cunningham, Hindman, Howland, Morgan, Thompson, Watt, and Willard to discuss bottlenecks developing in the cooperative effort between Sub-section I and Sub-section III to improve the bismuth phosphate extraction-decontamination process. It was agreed to temporarily assign four of Thompson's and Albaugh's men to the process chemistry research; the manpower now on this problem thus consists of Ader, Bartell, Howland, Malm, Morgan, O'Connor, Peterson, Sheft, and Cliff Smith. The problems of most urgent importance and the men assigned to them are as follows:

(1) Extraction step difficulties (occasional poor carrying). Ader and Sheft. (2) Carrying of Pu(III) and Pu(IV) by bismuth phosphate (relative effectiveness of carrying). Malm and Bartell. (3) Effect of iron on carrying by bismuth phosphate. Morgan. (4) Analysis of valence state of plutonium in bismuth phosphate precipitated from a solution of Pu(IV). Malm. (5) Determination of prevailing valence states of plutonium in process solutions and the means of stabilizing the desired valence state. Morgan or Howland. (6) Crystal structure studies (preparation of crystalline samples of Pu(IV) phosphate, fluoride, etc., for x-ray examination). C. Smith. (7) Solubility of Pu(III) and Pu(IV) compounds in process solutions. O'Connor. (8) Solubility of bismuth phosphate. Peterson.

4/27/44 (cont.)

At 1:30 p.m. the regular weekly conference on ether extraction was held and attended by Bird, R. S. Buffum, Dreher, A. C. Hyde, Holt, Kircher, Maloney, Tepe, and Thompson. It was reported that during the past week the aqueous extraction column of the continuous ether extraction system was operated successfully using 0.05 M hydroxylamine sulfate as the reducing agent. Quantitative plutonium recovery was obtained. During the coming week the ether and aqueous extraction columns will be operated independently at full dissolver activity under conditions recommended by Dreher's group.

The evening meeting of the Research Assistants of my section was held at 6:45 in Room 209, Eckhart Hall.

I held an evening meeting of the Council of my section at 7:45 in my office, attended by Albaugh, Cunningham, Davidson, Dawson, Hindman, Jensen, Katzin, Manning, Orlemann, Thompson, Watt, and Willard. I reported that it has now been decided to ship the Site W plutonium to Site Y in the form of dry plutonium (IV) nitrate hydrate rather than in aqueous solution; Watt is to investigate whether the nitrate can be melted and poured as is possible with UNH.

I also said it has been decided to stop the present Berkeley neutron bombardment (target 60 pounds of uranium metal) and ship the material to Chicago. Magnusson and La Chapelle will work out the procedure for extracting the neptunium. After some discussion, it was decided to make a practice run on a Clinton slug--this will also serve to check the amount of 93^{237} produced in the Clinton pile.

The next item discussed was the problem of cleaning the building after the workmen finish the remodeling. It will probably be necessary for the laboratory personnel to do the initial cleaning. Other items considered in the general program of cleanliness were: (1) Limitation of glass-blowing tables. There are to be two general tables in Cunningham's sub-section, two in Orlemann's and one in Thompson's. (2) All desk tops are to be clean before leaving the laboratory at night. Beakers and similar equipment are to be kept in the cabinets provided. (3) The dieners (cleaning people) will scrub daily. (4) Blackboards are to be removed. (5) No smoking will be allowed. (6) Shoes to be worn in the Section C-I area (changed in the entrance area outside the air lock) are to be provided by the personnel. Requisite ration coupons will be furnished. Surgical gowns will be worn. Each individual will have a locker (outside the air lock) where his clothes, shoes, etc., will be kept. Entrance to the Section C-I will be from the alley. (7) A "Radiation Protection Group," Section C-V, is being formed. Three members are to be from our section. The duties of this group will be similar to the equivalent group at Y, where it is responsible for such items as taking daily nose swabs, hand counts for alpha, beta, and gamma contamination, air samples, checking masks to be used for highly contaminated rooms, etc.

The question of dry-boxes for work with powdered materials was discussed. It was noted that the sample grinding carried out in Room 13 has not always been done in a box. The sub-section chiefs were advised that they should encourage planning so that a sufficient number of boxes should be available.

4/27/44 (cont.)

On the subject of reports, I noted that Katzin is preparing a review in narrative style of the basic data on the processes, etc., developed by the section. H. Baumbach is to prepare a photographic summary of the progress made in the section.

I remarked on the general lack of progress that has been made on the decontamination problem. It was suggested that a liter-scale run be made on a hot slug using a process with the best possible combination of scavengers known.

I mentioned that Warner's notes on the last Thomas meeting outlined the following commitments: (1) Try metal production from $PuBr_3$, (2) Determine the carbon content of $PuCl_3$ produced by reduction with CCl_4, (3) Determine the physical properties of plutonium produced from $PuCl_3$, (4) Test reactions of various refractories with plutonium.

It was decided that the Section C-I Council meeting is to be shifted to Tuesday nights and the sub-section meetings are to be held on Thursday nights.

Helen took a walk with Wilma.

War news is pushed out of the top headlines today by the battle between Sewell Avery of Montgomery Ward and Company and the federal government over labor problems. The government has seized Ward's Chicago plant, and the army has been called in.

Friday, April 28, 1944

Allison left at 11 a.m. on a trip to Montreal to inspect the redistilling operation of the heavy water being returned to Chicago.

"California Project, Chemical Research--Chemistry of Plutonium" (CN-1588) was issued and covers (1) the effect of beta particle radiation upon the sodium uranyl acetate extraction process for plutonium (J. W. Hamaker), (2) lanthanum fluoride analysis for $Pu^{(r)}$ in the presence of PuO_2^{++} (W. H. McVey), and (3) the adsorption of $Pu^{(r)}$ by glass at various concentrations of HNO_3, and (4) the radiocolloidal behavior of $Pu^{(r)}$ (E. L. King).

Helen shopped downtown. In the evening she and I played cards with the Ghiorsos.

Saturday, April 29, 1944

This is the last day of work at the Met Lab for Byron Malekow who is resigning.

I received a memo from Stan Thompson discussing long-range plans for further work on separations processes, possibilities which I may wish to consider at the Steering Committee meeting next month. He proposes further work on decontamination by investigating new scavengers such as lead sulfate

and by conducting some one-liter scale runs using Hanford concentrations of plutonium and by-products. He also wants to push development of improved analytical procedures for Pu(VI) to aid in studying the problem of reoxidation. He mentions that specific plans have been made for further work on the concentration (crossover) procedure (problem of product losses in the fluoride by-product precipitate) and the isolation steps (effect of the presence of Pu(III) and (VI) in the lanthanum nitrate solution, use of nitric-sulfuric acid mixtures).

I wrote C. A. Kraus at Brown University to tell him that Norman Davidson plans to visit on Wednesday, May 3, from late morning to late afternoon.

Willard sent a memo to Thompson quoting for the record a memo from Foote of the Technical Division about the specifications of ten special extrusion billets of uranium metal containing added amounts of inactive fission product elements. The nominal amounts of these elements in grams per ton are as follows: columbium, 12.5; lanthanum, 11.0; molybdenum, 22.0; ruthenium, 14.0; and zirconium, 26.5. Cerium, neodymium, and yttrium additives are still on order. Collins of Clinton Laboratories will make up aluminum-silicon coated slugs from the billets for us.

I've been told that Donald Pye is scheduled to marry Sarah Wilkins today; they will take a three-day honeymoon.

Helen and I saw "Jack London" at the Frolic theater, starring Susan Hayward and Michael O'Shea, followed by a musical.

Secretary of the Navy, Frank Knox, died today at the age of seventy.

Today's paper reports that a "mystery load" of power from the Bonneville hydroelectric project in Washington and Oregon will go into operation soon in the production of a "new weapon of warfare," according to Representative Angell, Republican from Oregon.

Sunday, April 30, 1944

Politics dominate the front page of the Sun today; MacArthur has declared that he will not be a candidate for President and requested supporters to stop activity in his behalf.

I played golf while Helen had lunch with Wilma.

Don Cooksey called with the news that he and Milicent expect a baby in September.

Norm Davidson left on a trip to New York. He will visit Professor Kraus at Brown University on May 3.

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