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METHODS OF PRODUCTION AND RESEARCH ON TRANSCURIUM ELEMENTS

Stanley G. Thompson* and M. Luis Muga*

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INTRODUCTION

Isotopes of the elements berkelium and californium have now been produced in microgram amounts as a result of a program of neutron irradiation which began in 1952. At that time approximately 8 grams of Pu^{239} were subjected to a high neutron flux in the Materials Testing Reactor[1] at the Reactor Testing Station of the U. S. Atomic Energy Commission near Arco, Idaho. The Pu^{239} was rapidly transmuted to heavier isotopes and fission products. The transcurium isotopes were extracted periodically and the major parts of the plutonium, americium, and curium fractions were in every case returned to the reactor for further irradiation. Microgram amounts of berkelium and californium were obtained after about five years of irradiation at neutron fluxes of approximately 5×10^{14} neutrons cm^{-2} sec^{-1}.

The transcurium isotopes produced by neutron irradiation were used as source materials for an extensive research program in chemistry and physics, much of which has been summarized in a previous publication.[2] This research included studies of the chemical properties of the transuranium elements and investigations of nuclear properties. The isotopes have been used as targets in bombardments with charged particles to produce new isotopes and elements. The isotopes were also used in research on the mechanisms of nuclear reactions, in nuclear spectroscopy with emphasis on alpha decay, and for research on fission.

A research program on the heaviest isotopes is primarily dependent on the production of the isotopes, and on the techniques for handling them. The purpose of this paper is to describe the most important features of this work as it is done at Berkeley. Very similar procedures have been employed in laboratories at Argonne, Chalk River, Los Alamos, Harwell, Saclay, Stockholm and those of the Soviet Union. We hope to include information from these laboratories as well. In this paper we shall attempt to bring the subject up to date without including excessive detail, and to include basic information useful in research work on the transcurium elements. The developments of the last three years are emphasized.

At the time of this writing experimental work is in progress on the isolation of pure compounds of berkelium and californium and the study of their properties. More detailed and up to date information on this subject will be given in a supplement to this paper.

CHEMICAL PROPERTIES

With the exception of berkelium, which has a +4 oxidation state of about the same stability as Ce (IV), the transcurium elements through mendelevium exist predominately in the tripositive oxidation state in aqueous solutions and as such resemble each other very closely.

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*Radiation Laboratory, University of California, Berkeley, California

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Berkelium (IV) has the half-completed shell configuration $5f^7$ which has a special stability. Such an oxidation state has been found[3] by tracer experiments in which treatment with bromate ion, dichromate ion, or ceric ion caused oxidation of berkelium to a form coprecipitated with zirconium phosphate, zirconium iodate, and ceric iodate, compounds which are rather specific carriers for tetrapositive ions. The oxidation potential of the berkelium (III) - berkelium (IV) couple is about -1.6 volts.

Efforts have been made to oxidize californium at tracer concentrations with such powerful oxidants as peroxydisulfate and bismuthate ions in acid solutions and hypochlorite ions and ozone in alkaline solutions[4] without success. In the latter case the attempted oxidation was carried out in strong carbonate solutions using americium as a tracer. The results of these experiments cannot be taken as proof that oxidation will not occur with higher concentrations of californium.

The transcurium elements[5,6] exhibit about the same behavior relative to each other as do the rare earth homologues to each other. Therefore the separations of these elements from each other, from rare earths and from neighboring actinides is best accomplished by ion exchange methods.

The transcurium elements adsorb on cation exchange resin columns and can be separated from each other with buffered citrate, lactate, alpha hydroxyisobutyrate and similar eluting agents.[7] (Fig. 1) They can be eluted with 13 molar hydrochloric acid from cation exchange columns ahead of the rare earths, except for the heaviest rare earth elements.

The transcurium elements form anionic chloride complexes in 13 molar hydrochloric acid and can be adsorbed on anion resin columns. They can be partially separated from each other by subsequent selective elution.[6] The rare earth elements do not adsorb on an anion resin to an appreciable extent in 13 M HCl. Complete separations of the transcurium elements from all rare earths are made when 20% ethyl alcohol saturated with hydrochloric acid is used to elute the mixtures from columns of colloidal Dowex-50 resin (12 percent cross-linked).[6] The rare earths elute well after the actinides. Some separation of the individual actinides from each other is also achieved as shown in Fig. 2.

Negatively charged complexes are also found in concentrated solutions of ammonium thiocyanate.[8] A typical elution curve from Dowex 1 resin using ammonium thiocyanate is shown in Fig. 3. However, the desorption sequence is not simple. The order of desorption from Dowex-50 resin column is shown in Fig. 4. The rare earths are less strongly complexed by thiocyanate than are the actinides but not sufficiently so as to give a complete group separation.

Good separations of the transcurium elements from the lanthanides can be achieved by desorption of a mixture from anion exchange resins using concentrated lithium chloride solutions especially when the separation is made at elevated temperatures. The lanthanides are less strongly complexed and are eluted first.

So far, the best separations of the individual transcurium elements from each other are achieved by adsorption on Dowex-50 cation exchange resin followed by desorption with ammonium α-hydroxyisobutyrate solution at a temperature of 87°C.[7] An elution curve for these elements is shown in Fig. 1. Negatively charged complexes of the transcurium elements are formed with α-hydroxyisobutyrate and these complex ions may be adsorbed on anion resins under ideal conditions.[9,10]

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Considerable success in separating common impurities has been accomplished by desorption of mixtures from columns of Dowex 50 resin (4 percent cross-linked) using 2 M and 6 M HCl. This method is a modification of one developed by Chetham-Strode.[11] The columns are operated at 87°C and an intermediate step of elution with 2 M HCl aids in obtaining sharper actinide peaks, which are preceded by the desorption of the common elements Al, Fe, Ca and Mg.

The actinide elements can be extracted from an aqueous mineral acid solution into the immiscible solvent tributyl phosphate; the extraction coefficients achieved in this extraction are not as high as for the analogous lanthanide elements.[12] However, the coefficients, particularly for the higher members of the series, are high enough to make the use of solvent extraction by this solvent very attractive for some purposes. This is especially true of concentrated nitric acid solutions. Peppard has called attention to the remarkable analogies between the lanthanides and the actinides in this solvent extraction behavior.[13] These elements can be extracted readily into benzene as chelate complexes of α-thenoyl-trifluoroacetone[14] (T.T.A.). At pH 3.4 the extraction of berkelium and californium is roughly a factor of ten greater than that of americium and curium.

The difference in extractability of the tripositive and tetrapositive berkelium ions can be the basis for a rapid and quantitative separation from certain ions; for example, the ratio of the distribution coefficient for berkelium (IV) to the distribution coefficients for berkelium (III) and curium (III) is about 10^6 for extraction into di(2-ethyl hexyl) orthophosphoric acid - heptane from aqueous nitric acid.[15]

The coprecipitation behavior of these elements as studied at tracer concentrations is very similar to that of other actinide elements having the same oxidation states and coprecipitation is observed with such typical carrier compounds as lanthanum fluoride, lanthanum hydroxide, and ferric hydroxide.[6]

In the metallic state these elements have a sufficiently high vapor pressure to permit their vaporization from molten uranium at temperatures above 1200°C, e.g., californium produced in uranium metal by carbon ion bombardments was completely vaporized and collected on platinum counting discs, when the uranium was heated to 1200°C *in vacuo*. Similar behavior was observed for einsteinium and fermium.[16] Compounds of the elements deposited on tantalum filaments are readily evaporated onto collector plates when the filament is heated to high temperatures in vacuum.

NUCLEAR PROPERTIES OF TRANSCURIUM ISOTOPES

Since rather complete accounts of the nuclear properties of the transcurium isotopes have already been published[2,17,18] only a very brief general discussion is given here.

These isotopes are characterized by their high instability. In addition to having all the types of instability observed in lighter isotopes they are unstable toward alpha decay and spontaneous fission. In general the stability decreases as the atomic number increases resulting in shorter alpha and spontaneous fission half-lives for the heavier isotopes although the spontaneous fission half-lives show deviations from this trend.[19,20] An interesting feature of this region is the 152 neutron subshell.[21] At neutron numbers just above 152 increased instability is superimposed upon that of the normal trend. The resulting relatively higher ground state masses correlate well with an increase in the energy available for spontaneous

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fission.[22] The isotopes of the elements above fermium are so difficult to create and apparently have such short half-lives that their detection is difficult. The transcurium region is of special interest in connection with nuclear spectroscopy and particularly in the study of alpha particle decay schemes. It is a region of non-spherical nuclei for which the decay schemes are well interpreted by application of the unified model of the nucleus developed by Bohr and Mottelson.[23] Another interesting feature is the fact that some isotopes such as Cf^{254} and Fm^{256} decay primarily by spontaneous fission. The isotope Cf^{254}, because of its two month half-life for decay by spontaneous fission, has been considered as a possible explanation of the light decay curves following supernovae explosions.[24] In view of the increasing predominance of spontaneous fission at high Z and A a special problem is presented in working with large amounts of californium because of the continuous production of neutrons and fission products.

Some properties of the transcurium isotopes are listed in Table I.

PRODUCTION OF ISOTOPES BY NEUTRON IRRADIATIONS

A. General Considerations. The method by which the largest amounts of transcurium isotopes have been produced is that of neutron irradiation of the heaviest isotopes available. Many aspects of this problem have been discussed in a previous paper by Bentley et al.[25] Examples of starting materials are Pu^{239} and Am^{241}. The most important neutron source available at this time is the Materials Testing Reactor at Arco, Idaho. In this reactor neutron fluxes near 5×10^{14} neutron cm^{-2} sec^{-1} can be obtained. At these high neutron fluxes successive neutron capture reactions produce seventeenth order products (Fm^{256} from Pu^{239}). In the irradiation of fissionable materials considerable heat is produced at such fluxes. This problem has been solved by using aluminum alloys or by pressing the compounds with aluminum powder so that the heat can be removed rapidly by water cooling.

B. Choice of Starting Materials. The isotope Pu^{239} has been the most important source of the transcurium isotopes produced to date. It was chosen for this purpose because it was readily available in large amounts. It has the disadvantage that most of it is converted to fission products. Transcurium isotopes can be produced much more rapidly from Am^{241} than from the same mass of Pu^{239}. Less of the americium is converted to fission products and there is an advantage in starting with a higher mass number. However, Am^{241} is available in much smaller amounts than plutonium, and large amounts of the isotope Cm^{242} are produced very rapidly by the first order neutron capture reaction. The isotope Cm^{242} has a very high specific alpha activity, and the high level of alpha radioactivity so produced places heavier demands on the equipment and separations methods than the material produced by the irradiation of plutonium, especially if the separations are done at a time when the amount of Cm^{242} present is near its peak.

To partially circumvent the difficulty of separating transcurium elements from large amounts of Cm^{242}, it has been common practice to extract the easily separated Pu^{242} which is formed by the electron capture decay of Am^{242} and subject the Pu^{242} to further irradiation. The only adequate facilities available at this time for handling large amounts of curium are those at the University of California Radiation Laboratory at Livermore which were primarily the result of efforts by W. W. Crane and G. H. Higgins. The installation has many novel features, the description of which cannot be included here.

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High-flux space in the reactors is at a premium and the irradiation of isotopes which occupy the least reactor space is desirable. Therefore, Am^{241} and its product Pu^{242} have definite advantages. Likewise it has been advantageous to extract the transmutation products Pu^{242}, Am^{243}, and Cm^{244} from irradiated Pu^{239} after the fissionable material has been largely burned out and reinsert them in a smaller region of high neutron flux in the reactor.

CALCULATIONS OF ISOTOPE PRODUCTION

Pile cross-sections of the transcurium isotopes have now been determined independently from many experiments in which known amounts of Pu^{239}, Am^{241}, Am^{242}, Am^{243}, Cm^{244} and other isotopes have been irradiated with pile neutrons. Pile cross-sections refer to average or effective cross-sections and are measured in the neutron flux found in the body of the reactor (Pile), within or close to the active core. A pile neutron cross-section may deviate quite appreciably from a thermal neutron cross-section if the nuclide in question absorbs neutrons in the resonance region. These values, including those for plutonium, americium and curium isotopes, are compiled in Table II.

The quantities listed in Table II represent the "best fit" values chosen by this laboratory in the light of measurements made by University of California Radiation Laboratory, Berkeley, Knolls Atomic Power Laboratory, Argonne National Laboratory, University of California Radiation Laboratory, Livermore, Oak Ridge National Laboratory and Chalk River groups. These values are not to be interpreted necessarily as always being correct individually; however, they seem to give approximately correct isotopic compositions and the correct amount of Cf^{252} after a long irradiation time. It has been convenient to calculate these yield curves by means of a Growth and Decay Analog Computer (Appendix I) which makes use of an eight component system of resistors and condensers to simulate the growth, decay and destruction of nuclei along the possible path sequences. Programs have also been set up to compute the isotopic yields using Computers such as the I.B.M. 650. The reaction network showing the possible paths in forming transcurium isotopes from Pu^{242}, Am^{241}, and Cm^{244} is diagrammed in Fig. 5 (Pu^{242}, of course, is formed by successive neutron capture in Pu^{239}, and Am^{241} is formed by beta decay of Pu^{241}.)

Curves of practical interest have been made giving the estimated yields of Cf^{252} for flux levels as high as 5×10^{15} neutrons cm^{-2} sec^{-1} (Figure 6). The growth of Cf^{252} from pile neutron bombardments of Cm^{244}, Am^{241}, and Pu^{242} is shown in Figs. 6 and 7. It can be seen that the maximum yield is largest and is obtained most rapidly with the higher neutron flux levels.

In Fig. 8 is given a log log plot of the formation of Cf^{252} from Cm^{244}, Am^{243}, Am^{241} and Pu^{242} for an average flux of 1×10^{14} neutrons cm^{-2} sec^{-1}. The curve for Cm^{244} is a "best fit" curve using both calculated and experimental values of the amount of Cf^{252} formed.

For those interested in initiating a program of transcurium isotope production, Fig. 9 will be useful. The production of Cf^{252} from a Pu^{239} source is given for the probable upper and lower limit fluxes obtained in the MTR. Two experimental values (points shown in figure) for Cf^{252} yields from "napkin ring" sources are in good agreement with the calculated curves. Of equal interest is the rate of heat production from a Pu^{239} source subjected to pile neutron irradiation. (Fig. 10).

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Examination of the yield curves presented here indicates the tremendous importance of a high product of neutron flux and time. When high flux reactor space is at a premium, it can be seen that each removal of intermediate isotopes such as Pu^{242}, Am^{243}, and Cm^{244} and their re-irradiation at the highest possible flux levels is very profitable as contrasted to irradiation of larger amounts of lower mass isotopes at lower fluxes.

A typical example of the composition of isotopes resulting from an irradiation of Pu^{239} for a period of about two years at an average neutron flux of $\sim 4 \times 10^{14}$ neutrons cm^{-2} sec^{-1} is shown in Table 3. Longer irradiations do not have a large effect on the total amount of plutonium, americium and curium produced because of the "burn out" of the starting material. However, the heavier curium and transcurium isotopes increase rapidly with time because of the high order of the reactions in which they are produced. Therefore other examples of isotopic composition are not given; they may be estimated from the previously mentioned curves. Since the isotopes E^{253} and Fm^{254} are essentially in equilibrium with Cf^{252} at any given neutron flux because of their relatively short half-lives, the equilibrium values may be estimated in a simple manner from the amount of Cf^{252} present, the cross-sections and the neutron flux.

PREPARATION OF MATERIALS FOR IRRADIATION

In irradiating large amounts of americium and curium in a high neutron flux, provision may be required for heat removal just as it is in the case of Pu^{239}. The construction and dimensional design of the sample and containers were such that at the highest anticipated neutron flux, no more than one kilowatt of heat had to be dissipated per square inch of sample surface. In the case of plutonium, in which up to 350 mg samples were used for each "napkin ring" specimen, the problem was solved by preparing alloys of composition either 5% or 10% plutonium by weight in aluminum. These specimens were then clad with an aluminum jacket to give a total weight of about 20 grams and fabricated in the shape of "napkin rings", in order to allow the maximum rate of heat removal by the high velocity flow of water. A previous description of the "napkin rings" and their irradiation was given by Bentley et al.[25]

In re-irradiating Pu^{242}, Am^{243}, and Cm^{244} it is not necessary to reduce them to the metallic states and alloy them with aluminum. The oxides can be diluted with aluminum powder if necessary in a small aluminum cylinder (1/2" diameter x 5/8"). A solid aluminum plug of the same internal diameter is inserted into the cylinder and the sample is compressed (25,000 psi). A second solid aluminum plug, with a diameter slightly larger than the inside bore of the cylinder, is forced down on top of the first plug by means of a press, thus in effect "washing down" the sides of the cylinder. The cylinder edges are then folded over the second plug and tightly pressed, completing the assembly. If it is not necessary to dilute with aluminum powder to give improved heat transfer (normally the case in re-irradiating Am^{243} and curium fractions) another method is used, namely the sample solution is evaporated in a thin aluminum dish. The sample is ignited in a furnace to convert it to the oxide and the aluminum container is folded up, wrapped in a second foil and dropped into the aluminum cylinder which is then pressed into an aluminum plug as described above.

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For small samples in which self-heating offers no problem a quartz container may be used. A solution of the heavy isotope is evaporated in a 5 mm diameter quartz tube, ignited and the tube is sealed. The quartz tubes are either irradiated in sealed aluminum containers or in aluminum containers through which water may flow over the quartz surface.

CHEMICAL PROCESSING

The problem of removing the aluminum and fission products while retaining at least 95% of the actinide fraction requires more planning and equipment than any other phase in the production of heavy isotopes, both because of the high level radiation encountered and the degree of purity necessary for the final products. The importance of shielding requirements is not to be underestimated in this type of work. In addition to the radiations normally encountered (γ,β,α) the samples may emit neutrons at a high rate. For example one microgram of Cf^{252} emits about 2×10^{8} neutrons/min; this high neutron level severely limits the time during which the californium can be handled at close quarters, e.g., in micro-chemical manipulations. By interposing blocks of paraffin and cadmium, the neutron flux can be diminished; unfortunately, it is difficult to use shielding in small scale operations. The alternative is to reduce to a minimum the time necessary to complete experimentation. Before much larger amounts of transcurium elements become available, simplification of the methods and development of new types of equipment, particularly equipment of small size which may be remotely controlled, will be necessary in order to take full advantage of the opportunities for interesting experimental work with californium.

The equipment and mechanical operations used for "cave runs" in the recovery of californium in amounts ranging up to the microgram scale have received special attention and deserve a brief description. The largest scale operations involve the separation and purification of the "napkin ring" samples. The initial steps are carried out in a 6" cave box, i.e., an effective shielding of six inches of lead is provided. The box in which the irradiated materials are confined is independent of the shielding and has a separate ventilation system which draws off air from the interior, passes it through "scrubbers", condensers, and mechanical filters, and then recycles the cleaned air. A negative pressure with respect to the atmosphere is always maintained. The box has the equipment to carry out the necessary operations including centrifuge, evaporator, heating bath, storage and disposal, ion exchange columns, β-γ and neutron counters, stirrers, etc. Most of the operations are carried out using tongs and are viewed through a thick lead glass window. Electrically operated equipment is controlled from the outside as are reagents and air pressure.

The fact that spontaneous fission becomes an important mode of decay in the heaviest isotopes makes it possible to apply the very novel method of neutron counting to observe the behavior of curium and californium fractions in the chemical separations. It has been common practice to set up conventional neutron counting equipment adjacent to the boxes used in large scale operations and the chemical fractions can be placed in more or less standard positions near a neutron detector for measurement. Neutron counting is also employed throughout the course of chemical separations and purifications of the californium fractions. Thus, in this case much of the tedious work of counting samples for alpha radioactivity is unnecessary, and the hazard of taking samples out of enclosures containing high levels of radioactivity is eliminated.

After the bulk of the aluminum and fission products are separated, the sample is removed to a smaller 2" "junior cave" box. This box is equipped to handle further chemical separations in which fission products are eliminated and preliminary separations of the actinides from each other are made. Some fractions are purified for reinsertion in the MTR (reactor).

Regular gloved boxes providing only protection from alpha radioactivity are used in a supporting role for assaying, storage, and final purification. In junior cave and gloved boxes a reduced pressure is maintained inside the boxes at all times. Papers on early equipment design and health hazard problems which applied to the production of very heavy isotopes were given at the preceding conference in Geneva during 1955.[26,27,28]

Chemical steps used in the separations have been continuously modified since their inception. Fortunately, at the present time, the overall operation is more or less standardized and appears to be approximately the same at Argonne, Chalk River, Harwell, Oak Ridge, and Berkeley. The irradiated specimens are dissolved in a $NaOH$-$NaNO_3$ solution; the use of $NaNO_3$ permits dissolution of the aluminum without the evolution of hydrogen to create a dangerous atmosphere inside the cave box. The actinides are precipitated as the hydroxides and the bulk of the aluminum remains in solution, nevertheless, a small amount of aluminum and roughly half of the fission products are precipitated or carried with the actinides.

The residue is dissolved in hydrochloric acid, or in HCl containing a small amount of HNO_3 or HF depending on the circumstances and the subsequent steps of the process. The actinides and rare earths are sometimes precipitated as insoluble fluorides from a solution containing HCl and HF. The insoluble fluorides can be reprecipitated by first dissolving them in HCl containing boric acid and adding HF at this point, or they can be precipitated as insoluble hydroxides and redissolved in acid for the fluoride precipitation. Following final dissolution of the fluorides in hydrochloric acid, the plutonium and some fission products may be adsorbed on columns of anion exchange resin when the HCl concentration is in the range 6-9 M. In this case plutonium should be in the tetrapositive oxidation state and the addition of a small amount of ammonium nitrite is commonly added to accomplish this objective. Best results are obtained when the columns are operated at elevated temperatures.

The transcurium elements pass readily through the anion resin column along with the rare earths. The plutonium may be desorbed from the anion resin by passing through a solution of 6-10 M HCl containing a reducing agent such as HI or hydrazine. An alternative method is to strip the resin with dilute hydrochloric acid of concentration near 1 M, although the latter method gives less decontamination of the plutonium. It should be kept in mind that the fission products to be separated from the actinide elements are not necessarily normal fission products; they also include nuclides resulting from neutron capture on long-lived decay products of the primary fission products. In the rare earth region the heavier rare members are more difficult to separate from tripositive actinides. Thulium (Tm^{170}) is one of the elements giving trouble in the separation of rare earths.

The separation of the rare earths from the actinides is generally made at room temperature by desorption from Dowex-50 (4% cross-linked) spherical resin using saturated hydrochloric acid. In this case the mixture of actinides and rare

earths to be passed through the anion exchange column is concentrated by evaporation or by precipitation of the hydroxides and dissolution in HCl solution. In either case the concentrated solution is saturated with HCl. Then the mixture is loaded on the resin in as small a band as possible. The final, complete rare earth separations are usually accomplished using columns of Dowex-50 (12% cross-linked) colloidal resin by elution with 20% alcohol saturated with HCl gas. In the latter case even the heaviest rare earths are well separated from the actinides. Milsted[29] has been very successful in separating rare earths from the transcurium actinides by adsorbing the actinides in 13 M HCl or in 20% alcohol saturated with HCl on Amberlite IRA 400 resin. Further washing with the same reagents strips the lanthanides from the resin. Finally, the actinides are eluted in a very sharp band with 8 M HCl.

The individual actinides are normally separated from each other by adsorbing them on columns of Dowex-50 (12% cross-linked) spherical resin and desorption with buffered solutions of ammonium α-hydroxyisobutyrate. This separation is normally done at elevated temperatures (87°C) but has also been made at room temperature and at 50°C at reduced rates.

The final purification steps are often carried out by adsorbing the mixture contained in a .05 M HCl solution on columns of Dowex-50 (4% cross-linked) resin. In this way the band width of the adsorbed material is kept narrow. Initial desorption is performed with especially purified 2 M HCl which removes many common impurities. Finally the actinides are desorbed with especially purified 6 M HCl solutions. These columns are normally operated at 87°C and sharp peaks are obtained having a full width at half maximum of ~ 10-12% for a given actinide.

In the presence of organic complexing agents the normally insoluble hydroxides are not easily precipitated. Therefore, the actinide elements present in α-hydroxyisobutyrate solutions are adsorbed on Dowex-50 (4% cross-linked) resin after adding HCl to the buffered solution in an amount sufficient to give a free hydrochloric acid concentration of ~ 0.1 M. When the columns are operated at 87°C the transcurium elements can be separated, using a rather small amount of resin, from very large volumes of acidified butyrate solution because of very favorable distribution coefficients in favor of the resin.

A useful connecting step between some of the separations mentioned above (but not with solutions containing organic complexing agents) is that of co-precipitating the transcurium elements with ferric hydroxide by the addition of an excess of NH_4OH (or strong bases) to acid solutions of the elements. In this case some of the undesirable fission products and impurities may be removed. The $Fe(OH)_3$ can be dissolved in HCl and readily removed by adsorption on anion resins.

A method which has been used with some success as an alternative step to some of those described above (particularly in remote controlled operations behind heavy shielding) is that of adsorbing the mixture on the top of a Dowex-1 (8% cross-linked) resin column from a concentrated (~ 12 M) solution of LiCl. The actinides are strongly adsorbed. The columns are operated at elevated temperatures, preferably 87°C. Passage of 10 M LiCl solution through the column desorbs the rare earths first, and the individual actinides are partially separated from each other, the heavier transcurium actinides being most strongly adsorbed. Plutonium is very strongly adsorbed and can be desorbed finally with hydrochloric acid solutions. When large amounts of alpha radioactivity are present there is some increase in

neutron production due to the Li (α,n) reaction. Ammonium thiocyanate could probably be used in place of lithium chloride under similar operating conditions but has not been used in our work because of uncertainty concerning the effect of intense radiation on the decomposition of thiocyanate.

It will be seen that the two resins most commonly used are Dowex-50 (cation) and Dowex A-1 (anion), spherical (200-400 mesh size, 4-12% cross-linked) or colloidal resin, depending on requirements. An important feature necessary for reproducibility of the column separations and chemical purity of the products is the grading and cleaning of the resins which are often impure and consist of a wide range of particle sizes. The resins are graded (in the ammonium form for Dowex-50 and in the chloride form for Dowex A-1) by allowing them to settle from aqueous suspension. The fraction with a settling rate between 0.25 and 0.50 cm/min is collected and corresponds to the finer fraction of the ungraded material. This fraction is most useful for rapid separations. The resins are washed alternately with concentrated HCl and concentrated NH_4OH, rinsed with acetone or alcohol and finally with water. It is important to test characteristics of the resins under operating conditions before using them for crucial separations.

The dimensions of the resin columns are chosen to give the degree and speed of separation required and may vary in size from 0.5 mm diameter and 1.5 cm in length to more than 1.5 cm diameter and 30 cm in length. A typical column used to separate essentially "weightless" materials is shown in Fig. 11. Columns of this type are sometimes equipped with a 0.5 mm diameter platinum tip to insure a constant drop size of approximately 13 λ; sharper separations result from the fact that the retention of liquid on the tip, just after the drops fall, is minimized. Variation in drop sizes appears to be a major factor in obtaining reproducible results when individual drops are collected. With the smallest diameter columns it is most advantageous to collect liquid accumulating on the column tip at definite time intervals rather than collect the drops separately. The smallest columns have a great advantage in reducing the amounts of common impurities present with essentially "weightless" quantities of the heavy elements.

The separations described above can be combined in a number of ways to accomplish the same objectives. The order given above applies primarily to the case of irradiated Pu^{239} from which relatively large amounts of fission products must be separated. In the case of high levels of alpha radioactivity from such isotopes as Cm^{242} or Cm^{244}, it may be desirable to avoid early precipitation steps and to immediately separate the individual actinides by ion exchange; decontamination of the transcurium elements can be done later.

A factor which is often important in the efficiency of ion exchange separations is the technique of loading. It is important to begin the desorption with a narrow band of the elements to be separated. Thus it is advisable, as a rule, to avoid stirring the resin at the top when the agents for desorption are added. Sometimes a thin layer of resin is added to cover the active band and in other cases the addition of glass wool is helpful. When the loading of the column involves transfer of a solution containing a complexing agent such as concentrated hydrochloric acid, it is important to minimize the volume added and to allow the solution to pass completely into the resin before adding more complexing agent.

The most important chemical separations used at Berkeley for the transcurium element separation are those described above. Solvent extraction methods can undoubtedly be used in place of some of those described above; they have been applied very successfully to special problems.

PREPARATION OF THIN SOURCES

Thin uniform sources are useful, not only for target preparation, but for studies in nuclear spectroscopy as well. The most useful methods are those which are adaptable to handling small amounts, hence painting techniques, while used extensively with the lower actinides, have been little used in preparing transcurium sources. Extensive and useful literature exists on source and sample preparations (i.e., Reference 30). However, the methods and techniques, as applied to the transcurium elements, have not been previously summarized and are given below.

Electrodeposition is easily the most popular method not only because of the high yields obtainable but also because it is relatively simple; the deposition area is clearly defined and the method may be easily adapted to meet individual requirements. It is also not difficult to recover valuable isotopes which are not deposited initially.

The cell is made from a glass tube and the lower portion is shaped in a manner which defines the deposition area. The metal foil on which deposition occurs serves as the cathode and a soft plastic "washer" is inserted to insure a watertight seal around the base of the cell. Sometimes leakage is minimized by placing around the plating cell a container with solution at the same level as that in the cell.

The solution containing the transcurium isotope is transferred to the cell; and after addition of two drops of methyl red, the pH of the solution is adjusted to 2.0 - 2.5 and 2-3 ml of saturated NH_4Cl solution is added. Bringing the solution to the desired pH may require passing the end point several times with successively weaker solutions of NH_4OH and HCl; the final adjustment is made with ~ 0.05 M HCl solution. Alternatively, the initial solution may be evaporated to dryness and the residue redissolved in a prepared 6 M NH_4Cl (pH 2.0) solution. A platinum wire anode (wound in concentric circles at the bottom) is placed in the solution 3-5 mm above the metal plate cathode. A current density of 0.5 to 1.0 ampere/cm^2 of cathode surface is maintained for 20-30 minutes. With the current on, the pH of the solution in the immediate vicinity of the cathode becomes high enough to precipitate the actinides as hydroxides that deposit on the metal plate. The deposition is stopped by adding an excess of concentrated NH_4OH to raise the pH to approximately 9, the anode is lifted from the solution and the current is interrupted. The cathode is washed gently with water and acetone, dried and heated to a dull red in order to convert the hydroxides to the oxides.

Since most of the elements that precipitate as hydroxides or hydrous oxides are electrodeposited by this procedure, trace quantities of these elements must be excluded from the electrode surfaces, electrolyte and cell parts as well as from the target material if a pure deposit is required.

The method of vaporization from a heated filament is most often employed for the preparation of thin sources for measurements in alpha particle spectrometers. While this is a well known method for preparing thin uniform sources, it seems worthwhile to mention a few details of techniques (as described by Asaro[31]) which contribute to good results.

It should be emphasized at the outset that the isotope to be vaporized must be free of appreciable quantities of impurities. The final purification of transcurium actinides is usually accomplished by repeated adsorbtion of the mixture on Dowex-50 (4% cross-linked) resin (from .05 M HCl solutions) and desorption first with 2 M HCl and finally with 6 M HCl.

For making sources of small area a deposit of the isotope is placed on a thin tungsten filament (bent into a V shape). The area of this deposit can be made small by repeated evaporation of small portions of the isotope solution on the same area of a warmed filament. (Care must be taken to avoid spattering.) The space above the filament is then evacuated and the filament is heated to a temperature just adequate to vaporize organic residues and other volatile impurities. The surface on which the isotope is to be vaporized then is placed very close to the filament and the system is re-evacuated. The filament is "flashed" at a high temperature for one second or less. In this way it is possible to obtain transfer yields of greater than 90% with deposition in a small area. With some effort, the sources obtained are sufficiently thin so that half-widths of 1.2 Kev for alpha spectra in the 6 Mev range have been measured. A half-width of 5 Kev is the minimum obtainable for electroplated deposits, however, less effort has been applied toward obtaining the best possible resolution from these sources. There is evidence that an apparent "thickening" of the source occurs when platinum backing foils are heated to a high temperature after deposition.

Vaporized and electroplated sources have been commonly deposited on platinum, gold, and nickel foils. Electroplating on aluminum or other metals having an oxide film has been less successful. Many factors influencing the yields and character of electroplated deposits are not well understood.

A novel method for the production of thin sources has been described by Carswell and Milsted.[32] The material to be deposited is taken up in an organic solvent and is ejected in the form of a fine spray from a capillary by the application of an electric field. The droplets are evaporated in transit and the solid residue is attracted to and deposited on a suitable metal plate.

In a typical experiment a thin platinum wire was inserted as far as possible into a 0.2 mm diameter capillary previously filled with acetone containing about 10/mg/ml uranium in the form of its nitrate. The collecting plate was placed 1-2 cm below the tip of the vertical capillary and a voltage of 4-6 KV was applied. With a steady current of 0.1 to 0.2 μ amps the deposition of 5 λ of solution (containing 50 μgs uranium) was complete in about 10 minutes. The deposit, covering a well defined circular area of about one cm diameter, was flamed at 500°C, was very adherent, and showed no tendency to flake.

Modifications at the University of California Radiation Laboratory by Sikkeland et al.[33] have included shortening the distance between the capillary and collecting plate, lowering the applied voltage to 1-2 KV and placing the

entire system in a horizontal, rather than a vertical plane. In addition, the collecting foil is heated from the back side to ~ 300°C and movement of the collecting plate slowly in a two dimensional oscillation permits the selection of a small, predetermined and well-defined target area. The procedure has given excellent results in the preparation of uranium, plutonium, and curium targets.

Thin, freshly electroplated deposits of Cf^{252} have the interesting property of self-transfer from the surface. The effect has been observed with Cf^{252} deposits on gold and platinum foils that are not flamed after deposition. In these cases the rate of transfer has been observed to be as great as 1% per day. When the foils were flamed at temperatures of approximately 500°C, the rate of transfer decreased to about 0.01% per day. Advantage has been taken of this phenomenon to collect in vacuo the transferred Cf^{252} atoms on a thin plastic film such as VYNS.[34] Energy measurements can then be made on both fragments emitted in spontaneous fission processes. It was also observed, however, that the plastic films became brittle and were easily broken. The mechanism involved in transfer of the Cf^{252} has not received adequate investigation to date.

RECOIL METHODS AND FAST CHEMISTRY

In the production of transcurium isotopes by charged particle bombardments, increasing fission competition at higher Z^2/A results in smaller cross-sections for the spallation products. Therefore, in research on short-lived isotopes there was a need for increased sensitivity for separating and detecting the isotopes and, as a result, many "fast" chemistry techniques were developed. The limited amounts of target materials often available has made it advantageous to collect and separate product nuclei without damaging the target itself. Great progress in solving these problems came with the development of the "recoil method". This method makes use of the fact that product nuclei receive momentum from the incident particles and if the target film is sufficiently thin, these nuclei are ejected from it. By simply placing a "catcher" foil close to and directly behind the target material, an initial separation is achieved when the recoil nuclei are embedded in the catcher foil.

Gold, palladium and nickel foils are commonly used as collector foils. They are all good conductors, able to withstand intense beams of charged particles, and can be rapidly dissolved and separated from the actinide elements. Palladium has the additional advantage that its chloride complex is strongly colored and its behavior on anion exchange columns can be followed visually. Very thin nickel foils can be obtained (~ 0.5 mg/cm^2)(from A. D. MacKay, Inc., N.Y.), and are very useful because they have considerable mechanical strength.

The development of the deflector channel probe (Fig. 12), a target assembly designed to intercept the more intense inner beam of the cyclotron, was a significant step in the development of recoil technique. A modified version of the probe shown in Figs. 12 and 13 permits the removal of the collecting foil in less than ten seconds after the end of bombardment.

When the recoil method is used together with "fast" chemistry techniques, it is possible to study many short lived isotopes which could not be detected

otherwise. These methods were used for alpha bombardments of the isotopes Cm^{244} [11,35], Bk^{249} [36], $Cf^{249-252}$, [37,38] and E^{253} [39] in work which resulted in the discovery of many isotopes of the transcurium elements and in the discovery of the element mendelevium. A typical procedure using "fast" chemistry techniques is as follows:

The catcher foil, gold (0.1 mil) or palladium (0.05 mil), is placed in a planchet containing 25-30 λ concentrated HCl and 10 λ concentrated HNO_3 at 50°C. After dissolution is complete (15 sec.) the solution is transferred to the top of a very small Dowex A-1 anion column filled with resin and maintained at 87°C (described previously). The method of transfer, washing and elution (with 8 M HCl) has been described earlier in discussing anion column operations. Five drops of eluant is sufficient to elute the active fraction from a column 2 mm in diameter and a few centimeters in length. The drops are collected in a heated planchet and the gold (or palladium) is retained on the column. The eluate is evaporated to dryness by heating and directing an air jet gently over the liquid. This fraction can be collected on a platinum disk, flamed, and subjected to alpha pulse analysis or the actinides can be electroplated on counting discs. For further separation the residue, usually before flaming, is dissolved in a minimum of 0.05 M HCl and transferred to the top of a cation column from which α-hydroxyisobutyrate is used to elute the desired fractions, as described earlier. The eluate is caught on platinum plates (one drop per plate) and evaporated. The plates are heated to a dull red in a flame and are often counted in an ion chamber connected to a differential pulse height analyzer. The total time from end of bombardment until counting begins has been less than four minutes for some cases in which both the anion and cation column separations were made.

Exploratory experiments for short-lived alpha particle emitting isotopes in the transcurium region are sometimes made, using thin plastic films to collect the recoil atoms. Best results have been obtained using "Mylar" (obtained from DuPont) or "Terelene" (obtained in England) films of thickness 0.3 - 1.0 mg/cm^2. These films have good mechanical properties and are able to withstand intense beams (~ 50 μam/cm^2) for 20 minutes without significant deterioration. The plastic film is removed, placed on a platinum plate and stuck to the plate with a few drops of acetone. By bringing the plate into the center of a gas-air flame, the solvent and plastic are burned off very rapidly without excessive spattering and the counting disc flamed. The plate is then inserted in the counting chamber. Total time from end of bombardment has been as little as 36 seconds and some yields have been measured at more than 90%. A similar method employing Tygon films has been described by a group[40] reporting the production of element 102.

When the recoil method is applied to targets which are themselves very radioactive, the detection of short-lived transmutation products is sometimes obscured by the larger amount of radioactivity due to the target atoms which are transferred with a relatively large cross-section. The untransmuted target atoms receive much less momentum transfer from the bombarding particles, and so have a short range compared to the transmutation products. Interference from transferred target atoms can be virtually eliminated, without significant absorption of the transmutation products, by depositing a thin layer of a material such as aluminum over the target area.[40]

In the collection of recoil atoms from a bombarded target, it is essential that the target be thin relative to the ranges of the recoil atoms if a large

fraction of these atoms are to be collected. The results of an investigation of the ranges and straggling of recoil atoms have been published by Leachman and Atterling.[41] In this paper formulae are given which allow the calculation of these quantities. The results are in reasonably good agreement with the experimental values obtained by helium ion and heavy ion bombardments of heavy nuclei. Experiments have shown that for reactions involving compound nucleus formation in target isotopes, such as Cm^{244} with 40 Mev helium ions, about 90% of the recoil atoms escape if the target thickness is 10 micrograms cm^{-2}.[42] The recoil ranges in heavy ion bombardments are proportionately larger because of greater momentum transfer. The momentum transfer is less for reactions involving direct interaction mechanisms and the ranges are reduced accordingly. In this case the recoil ranges are less than those calculated using the formula of Leachman and Atterling.[41]

Another important factor to be considered in designing a recoil target assembly is the angular distribution of the recoil atoms. Harvey, Wade, and Donovan[42] have found that for the reaction Cm^{244} $(\alpha,2n)$ Cf^{246}, using targets of a few micrograms ... cm^{-2} thickness, over 90% of the recoiling atoms are collected if the solid angle intercepted by the catcher foil is at least 20 degrees. (See Fig. 14). Broader angular distributions are obtained for thicker targets and for reactions involving direct interaction mechanisms, e.g., the (α,p) reactions. In the case of reactions involving compound nucleus formation, the angular distribution of recoil products is more strongly peaked in the forward direction, e.g., in the $(\alpha,3n)$, $(\alpha,4n)$, and (C,xn) reactions in the heavy isotope region.

PRODUCTION OF ISOTOPES BY CHARGED PARTICLE BOMBARDMENTS

Charged particle bombardments of heavy nuclei provide an important means of creating and investigating transcurium isotopes. References to this work have been mentioned in the previous section. In addition, a rather complete description and interpretation of the reactions induced by helium and deuterium ions is given by Thomas, Harvey, and Seaborg[43] (to be published in the proceedings of this conference.) These authors successfully employ a modified Jackson[44] model in which the effect of fission competition is included to explain spallation cross-sections of compound nucleus type reactions.

In contrast, heavy ion reactions have been studied less extensively. Ghiorso and Sikkeland[45] have prepared an article (to be published in the proceedings of this conference) reviewing the present status of research on heavy ion reactions at Berkeley. Inasmuch as both experimental and theoretical aspects are covered in their paper, it will be unnecessary to discuss this subject here.

CONCLUDING REMARKS

The present paper is the result of an attempt to summarize the information available on the production of the transcurium isotopes. Methods and techniques used for research in this field are also included. We hope the information will be useful to those working in the field or contemplating such research. The possibility exists that some of the methods and techniques can be applied to research in other fields.

~~Two important subjects were inadequately covered in the above discussion because of interesting work now in progress which is nearing completion. In April 1958, the element berkelium was isolated for the first time and some of its macroscopic properties were measured. Interesting work with heavy ions is also in progress in the production of isotopes and in the study of reaction mechanisms. These subjects will be discussed in supplements to this paper.~~

Transcurium isotopes are being used in studies of spontaneous fission and induced fission. Their use in nuclear spectroscopy research promises to give much information concerning nuclear structure. In view of the interesting properties of transcurium elements and their isotopes it is likely that much advancement will occur in the future. With advances in basic research in this field it is reasonable to expect that practical applications will follow. The eventual availability of large sources of Cf^{252} (which decays by spontaneous fission) should present many possibilities for important applied research and even industrial applications. These possibilities for the uses of transcurium isotopes seem to justify a much larger effort to produce them than has so far been underteken.

ACKNOWLEDGMENTS

We wish to thank T. C. Parsons, Francis McCarthy, Raymond Gatti, and Lilad Phillips for the preparation of many of the isotopes mentioned in this paper. We also wish to thank Jean Rees and Patricia Howard for their valuable contributions in the preparation of this manuscript. We are grateful to others too numerous to mention for their invaluable aid.

Table I. Nuclear Properties of the Transcurium Isotopes

Nuclide	Type of decay	Half-life	Energy of most abundant α or β group (Mev)
Bk^{243}	α (0.15%) E. C. (99+%)	4.5 h	6.55
Bk^{244}	α (6×10^{-3}%) E. C. (99+%)	4.4 h	6.67
Bk^{245}	α (0.11%) E. C. (99+%)	5.0 d	6.15
Bk^{246}	E. C.	1.8 d	
Bk^{247}	α	$\sim 10^4$ y	5.51
Bk^{248}	β^- (70% E. C. (30%)	~18 h	0.65
Bk^{249}	β^- (99+%) α ($\sim 10^{-3}$%) S. F.	290 d $\sim 10^5$ y 6×10^8 y	0.08 5.40
Bk^{250}	β^-	3.13 h	1.9
Cf^{244}	α	25 m	7.17
Cf^{245}	α (30%) E. C. (70%)	44 m	7.11
Cf^{246}	α S. F.	35.7 h 2.1×10^3 y	6.753
Cf^{247}	E. C.	2.5 h	

Table I (continued)

Nuclide	Type of decay	Half-life	Energy of most abundant α or β group (Mev)
Cf^{248}	α S. F.	250 d 7×10^3 y	6.26
Cf^{249}	α S. F.	~400 y 1.5×10^9 y	5.82
Cf^{250}	α S. F.	~12 y 1.5×10^4 y	6.024
Cf^{251}		long (β stable)	
Cf^{252}	α (98%) S. F. (2%)	2.2 y 66 y	6.12
Cf^{253}	β^-	~20 d	0.27
Cf^{254}	S. F.	~65 d	
E^{246}	α	7.3 m	7.35
E^{248}	α (~0.3%) E. C. (99+%)	25 m	6.87
E^{249}	α (0.13%) E. C. (99+%)	2 h	6.76
E^{250}	E. C.	8 h	
E^{251}	α (0.53%) E. C. (99+%)	1.5 d	6.48

Table I (continued)

Nuclide	Type of decay	Half-life	Energy of most abundant α or β group (Mev)
E^{252}	α	~140 d	6.64
E^{253}	α S.F.	19.3 d 7×10^5 y	6.63
E^{254m}	β^- (99+%) E.C. (~0.1%) S.F.	37 h >10 y	1.04
E^{254}	α	~320 d	6.43
E^{255}	β^-	24 d	
E^{256}	β^-	56 d	
Fm^{248}	α	0.6 m	
Fm^{249}	α	~30 m	7.7
Fm^{250}	α	30 m	7.43
Fm^{251}	α (~1%) E.C. (99%)	7 h	6.89
Fm^{252}	α	22.7 h	7.04
Fm^{253}	α (11%) E.C. (89%)	3.0 d	6.95
Fm^{254}	α S.F.	3.24 h 220 d	7.20

Table I (continued)

Nuclide	Type of decay	Half-life	Energy of most abundant α or β group (Mev)
Fm^{255}	α S. F.	21.5 h >60 y	7.08
Fm^{256}	S. F.	~3.5 h	
Mv^{256}	E. C.	~30 m	
$102^{251,253}$	α	~10 m	8.5
102^{254}	α	3 s	

Table II. Comparative Pile Cross Section (in barns)

	Pile capture cross section	Pile fission cross section
Pu^{239}	385	765
Pu^{240}	550	4
Pu^{241}	350	1100
Pu^{242}	52	
Pu^{243}	170	
Am^{241} ($\rightarrow Am^{242}$)	225	
($\rightarrow Am^{242m}$)	675m	
Am^{242}	4500	3500
Am^{243}	137	
Am^{244}		
Cm^{242}	25	5
Cm^{243}	250	1400
Cm^{244}	25	
Cm^{245}	200	1500
Cm^{246}	15	
Cm^{247}	200	200
Cm^{248}	2	
Cm^{249}		
Bk^{249}	400	(assume negligible)
Bk^{250}	5000	5000 (assumed)

Table III. Isotopic Composition*

Element	Weight found (mg)	Mass number	Percent composition
Pu	12	238	0.15
		239	0.04
		240	0.36
		241	0.16
		242	99.2
		244	~0.06
Am	2	243	99.9
Cm	5	242	0.6
		243	---
		244	94.14
		245	1.9
		246	3.4
		247	---
		248	<0.15
Bk	8×10^{-7}	249	100
Cf	2×10^{-6}	250	~50
		252	~50
E	8×10^{-9}	253	100

* 100 mg Pu^{239}, integrated flux $\cong 2 \times 10^{22}$ neutron/cm^2.

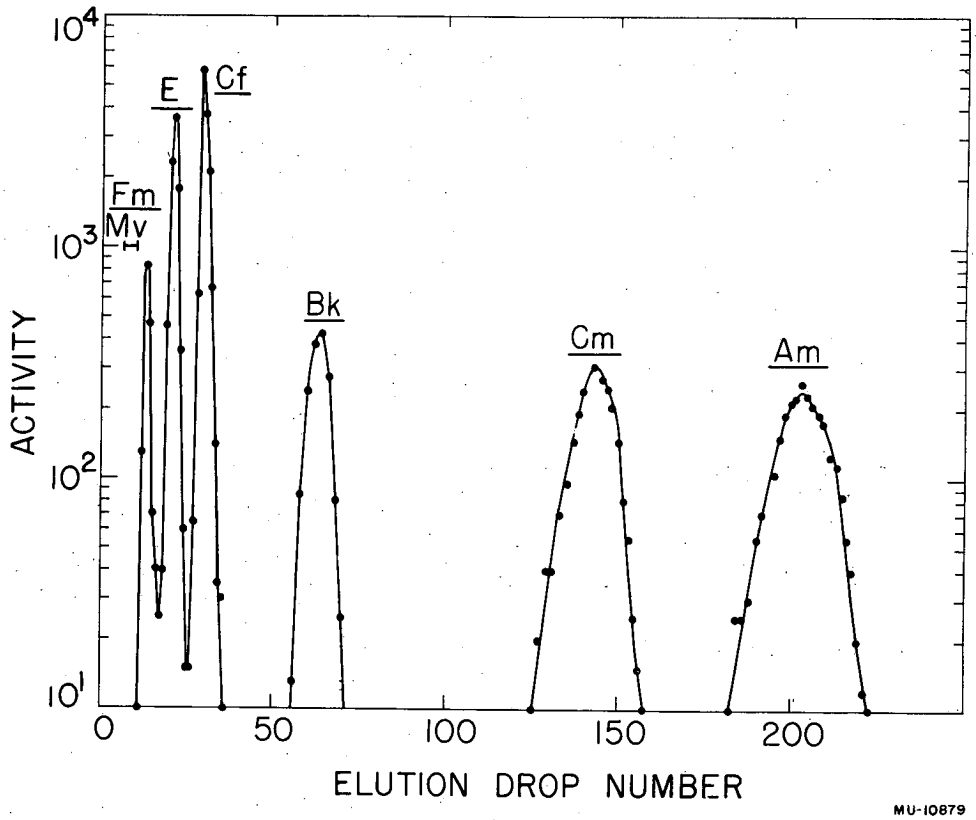


Fig. 1. Elution of tripositive actinides from Dowex-50 with ammonium α-hydroxy isobutyrate.

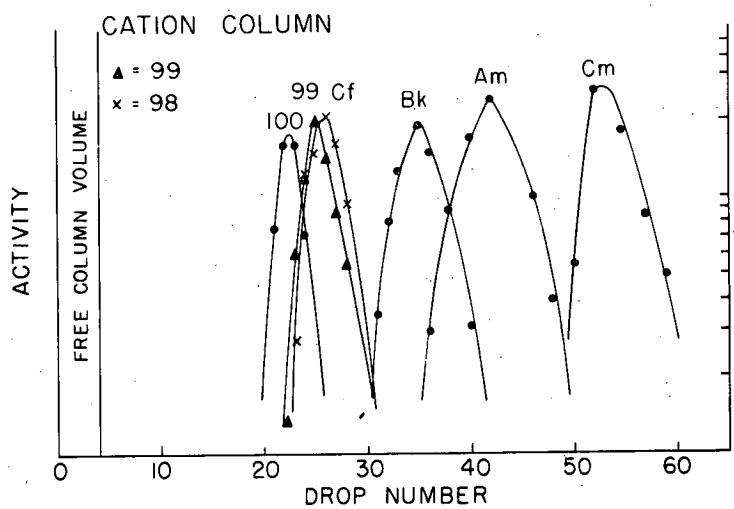


Fig. 2. Elution of trace amounts of actinide elements from a 5 cm length by 3 mm diameter column of Dowex-50 12 percent cross-linked colloidal cation exchange resin. Eluting agent – 20 percent alcohol – 12.5 molar hydrochloric acid.

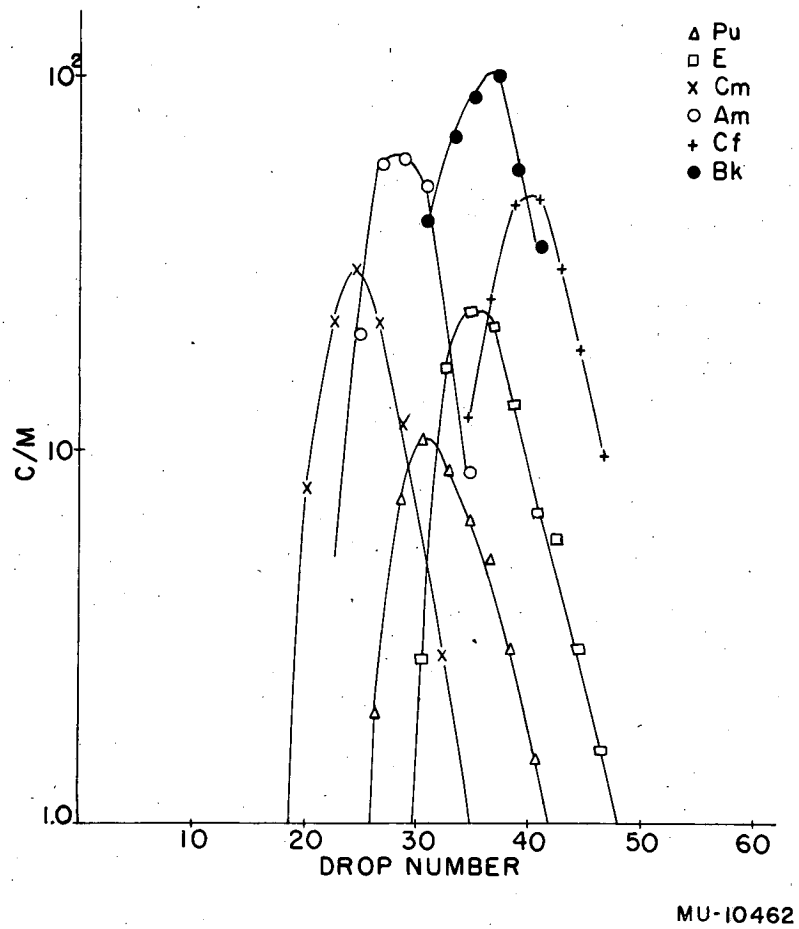


Fig. 3. Elution of actinides from Dowex-1 with 2 M ammonium thiocyanate at 87^0 C.

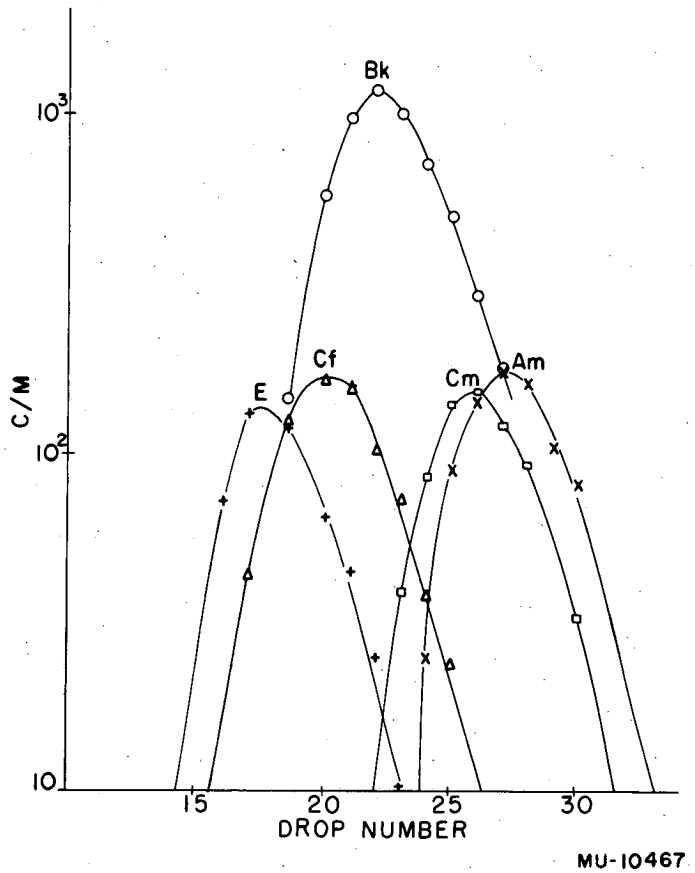


Fig. 4. Elution of actinides from Dowex-50. Resin bed 2 mm diameter by 5 cm length. 1.8 M ammonium thiocyanate eluant at 87° C.

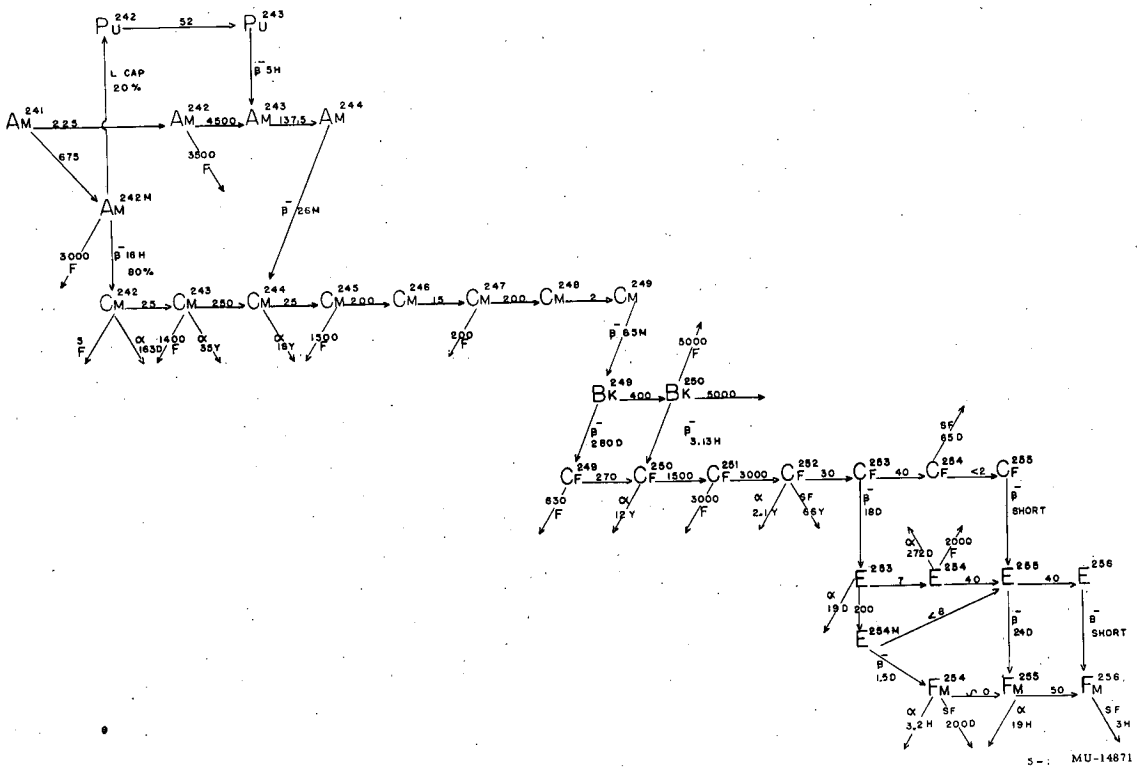


Fig. 5. Reaction sequence showing the paths by which Cf^{252} can be formed by pile neutron irradiation of Am^{241} in the MTR.

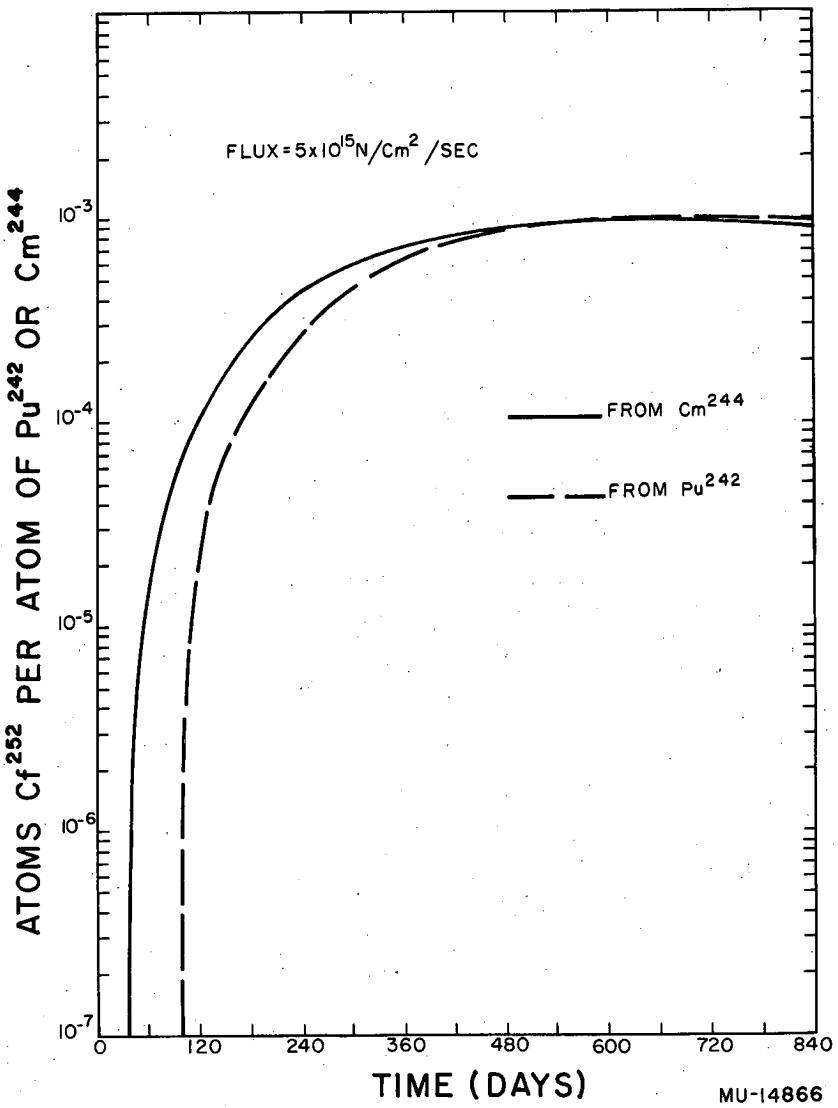


Fig. 6. Production of Cf^{252} from Pu^{242} and Cm^{244}.

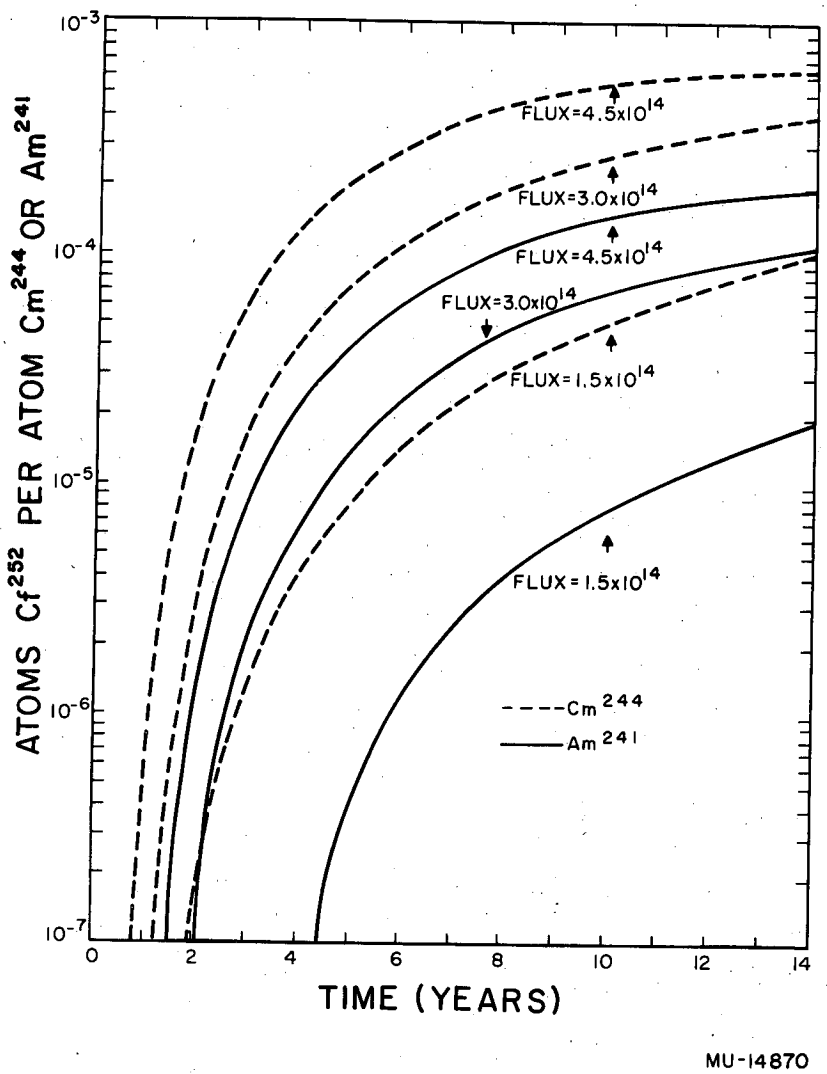


Fig. 7. Production of Cf^{252} from Am^{241} and Cm^{244} at different flux levels.

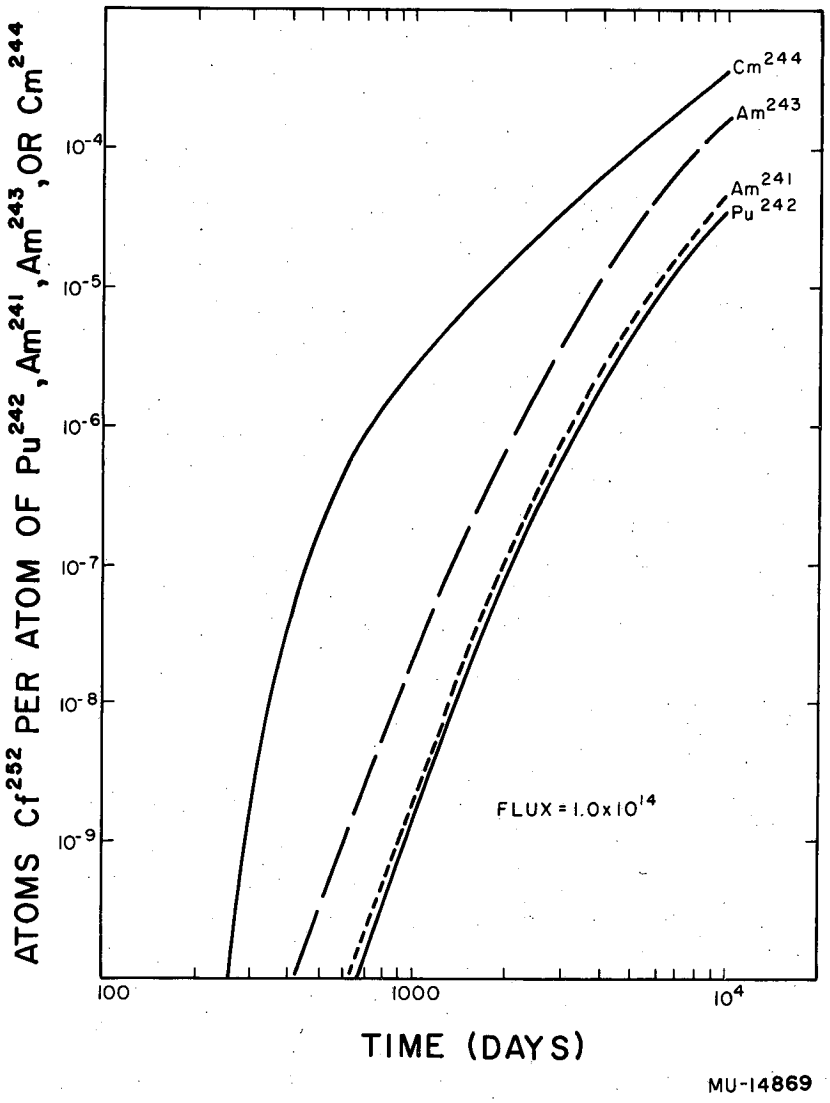


Fig. 8. Production of Cf^{252} from various starting materials as a function of time.

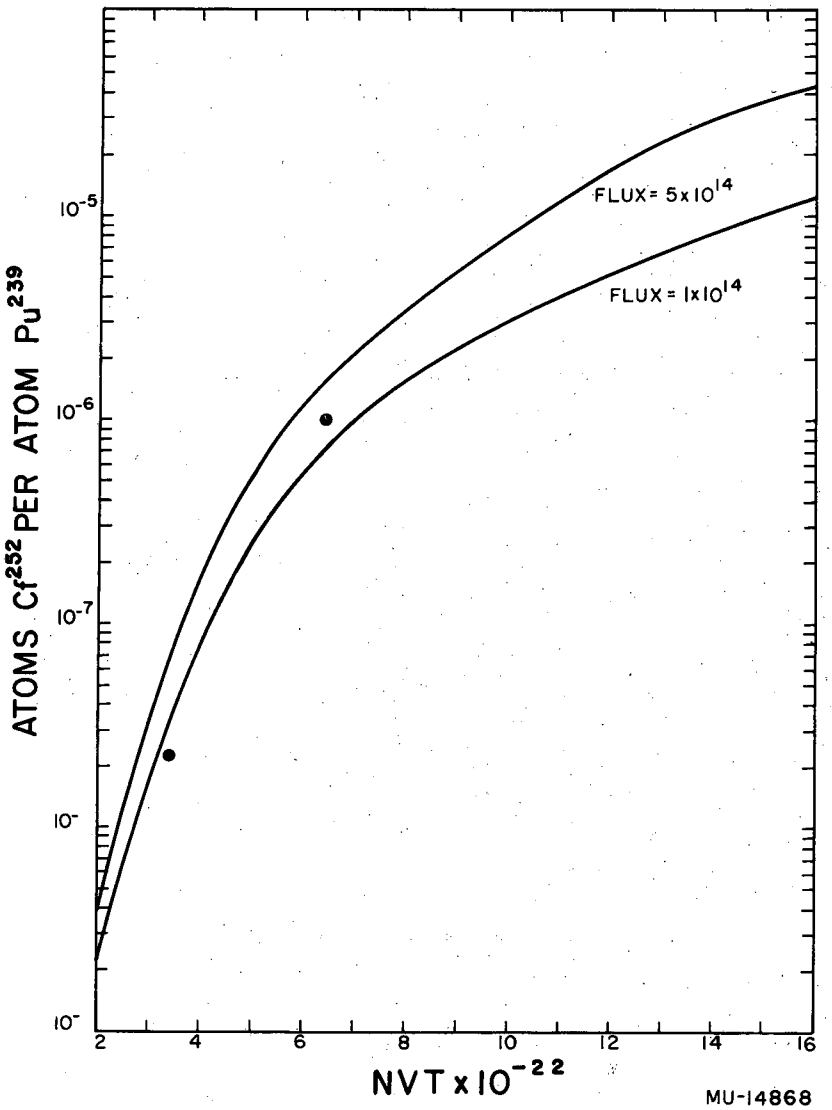


Fig. 9. Production of Cf^{252} from Pu^{239} for various integrated fluxes, (neutrons cm^{-2}).

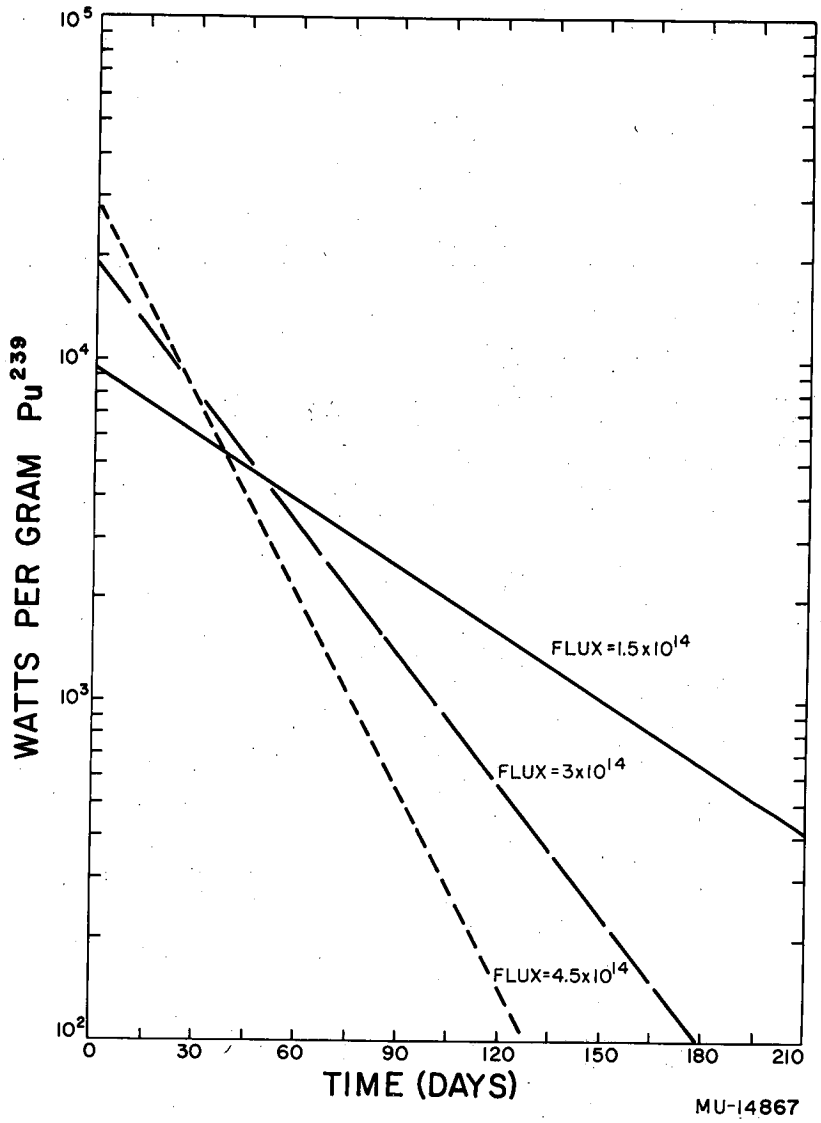


Fig. 10. Energy produced in "burn-up" of Pu^{239} as a function of time.

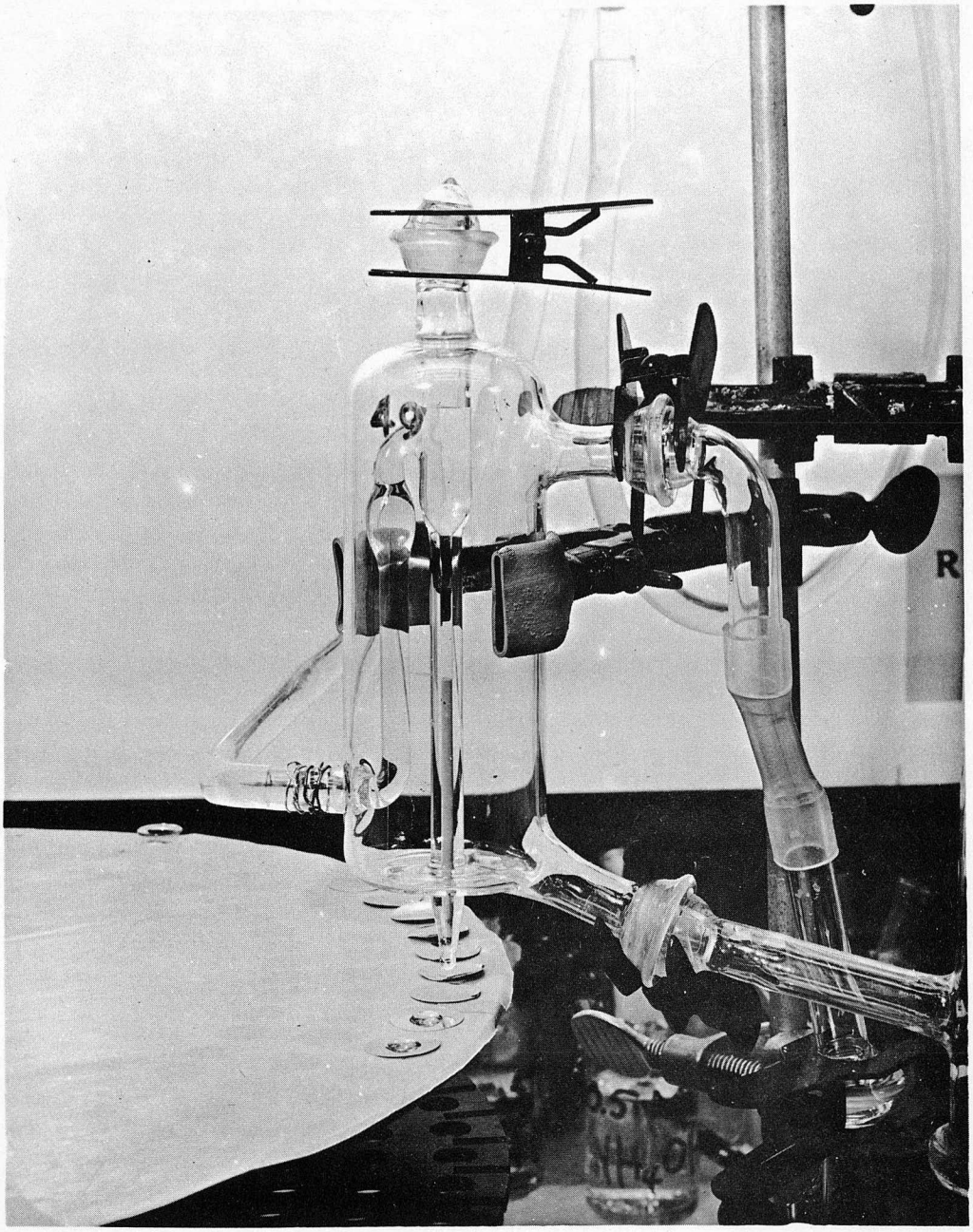


Fig. 11. Ion exchange resin is used to fractionate a mixture of the actinide elements. The resin, located at the bottom half of the inner tube, is maintained at a constant temperature by circulating trichlorethylene vapor through the outer jacket. The actinides pass through the resin at characteristic rates and emerge in sequence from the bottom where they are collected on metal plates for analysis.

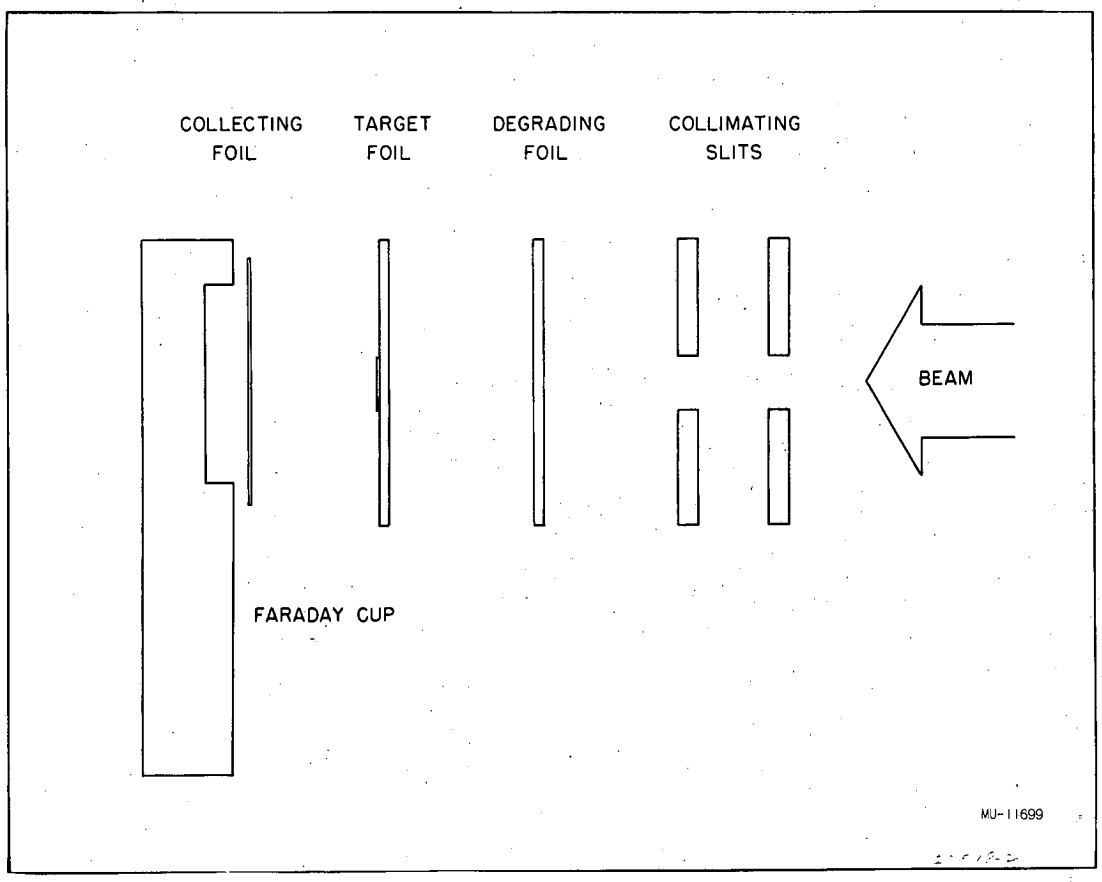
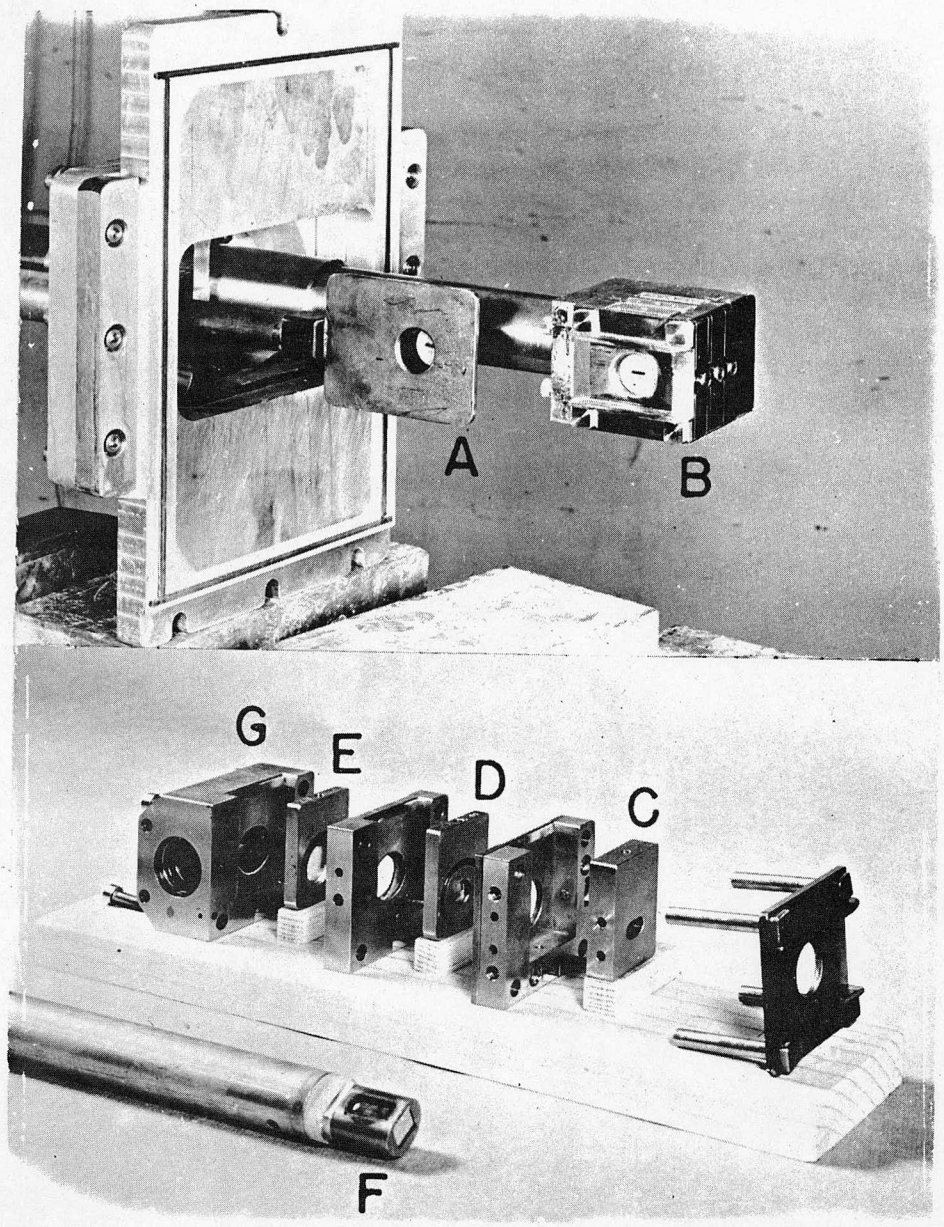


Fig. 12. Schematic diagram of the deflector-channel probe.



ZN-1504

Fig. 13. Internal Deflector Beam Probe. A. Precollimator. B. Target head. C. Collimator. D. Removable degrading foil. E. Target. F. Catcher foil and Faraday cup. G. Faraday cup holder.

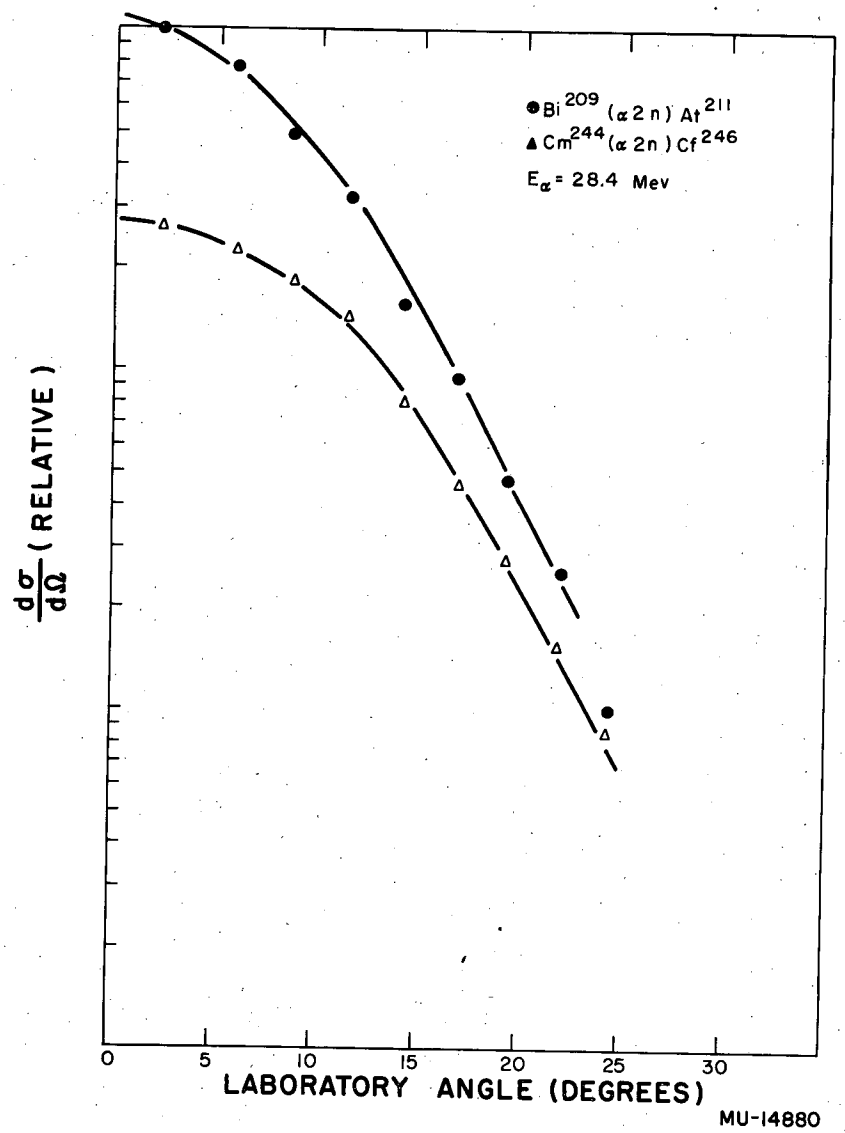


Fig. 14. Angular distribution of recoil atoms.

APPENDIX I

The Growth and Decay Analog Computer was designed and constructed by H. P. Robinson of the University of California Radiation Laboratory. Since its description is not available in previously published literature a brief description is given here. This computer is a device to reproduce by electrical analog the growth and decay of members of a radioactive decay chain. Let the chain with associated decay constants be represented by:

$$A \xrightarrow{\lambda_1} B \xrightarrow{\lambda_2} C \xrightarrow{\lambda_3} \cdots\cdots M \xrightarrow{\lambda_m} N \xrightarrow{\lambda_n}$$

Assuming unit atoms of A at time zero, and none of the other components present initially, the differential equations are:

$$\frac{dA}{dt} = -\lambda_1 A$$

$$\frac{dB}{dt} = \lambda_1 A - \lambda_2 B$$

$$\frac{dC}{dt} = \lambda_2 B - \lambda_3 C$$

$$\vdots$$

$$\frac{dN}{dt} = \lambda_m M - \lambda_n N$$

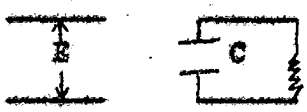
and the general solution is

$$N = \lambda_1\lambda_2\cdots\lambda_m \left[\frac{e^{-\lambda_1 t}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)\cdots(\lambda_n-\lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)\cdots(\lambda_n-\lambda_2)} + \cdots + \frac{e^{-\lambda_n t}}{(\lambda_1-\lambda_n)(\lambda_2-\lambda_n)\cdots(\lambda_m-\lambda_n)}\right]$$

For a large number of components, and especially when two or more λ's are almost equal, the numerical evaluation of this equation is laborious. An alternative method of expressing the differential equations is in terms of Laplace Transforms:

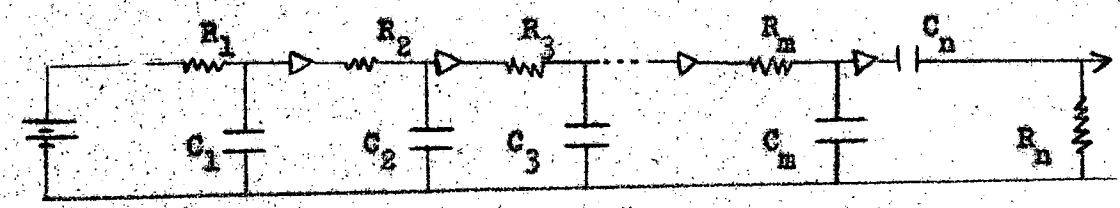
$$V_N = \frac{1}{S} \cdot \frac{\lambda_1}{S+\lambda_1} \cdot \frac{\lambda_2}{S+\lambda_2} \cdots \frac{\lambda_m}{S+\lambda_m} \cdot \frac{S}{S+\lambda_n}$$

where V_N is the transform of the nth component. An electrical analog of the first differential equation is simply a condenser discharging into a resistor:



$$\frac{dE}{dt} = -\frac{E}{RC} \qquad V = \frac{1}{S} \cdot \frac{S}{S+\frac{1}{RC}}$$

Making $\frac{1}{RC} = \lambda_1$ provides an exact electrical analog for component A. The factor $\frac{1}{S}$ represents a unit step function at zero time which starts the process. The electrical arrangement for N component is:



The symbol —▷— indicates an isolating amplifier of unit gain, and $R_1C_1 = \frac{1}{\lambda_1}$, $R_2C_2 = \frac{1}{\lambda_2}$ ---- $R_nC_n = \frac{1}{\lambda_n}$.

The transfer function of this network is:

$$\frac{R_1C_1}{R_1C_1S + 1} \cdot \frac{R_2C_2}{R_2C_2S + 1} \cdot \frac{R_3C_3}{R_3C_3S + 1} \cdot \text{----} \cdot \frac{R_mC_m}{R_mC_mS + 1} \cdot \frac{R_nC_nS}{R_nC_nS + 1}$$

and with a unit step voltage $\frac{1}{S}$ applied to the input, the output voltage is an exact representation of the growth and decay of the nth component.

In Gadac provision is made to insert the differentiating circuit (—|⊢⧖) at any component, in order to get as large a voltage output as possible. The results must be multiplied by, say λ_3/λ_n, if for example, the third component is given the differentiating arrangement. The output voltage is applied to a Speedomax recorder, and complete curves can be drawn. A timer is also provided so that the computer can be stopped at any predetermined time and the result read on the recorder. This permits taking readings of yields at times too short for the Speedomax to respond to, for example, one-tenth second. The convenience of the timer is such that many results are taken in this manner, even when no question of recorder response time is involved.

For the problems ordinarily encountered, the condensers are 10 microfarad polystyrene units and the resistors are one megohm Helipots. Provision is made to introduce additional resistance in series with the Helipots when needed.

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