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NUCLEAR REACTIONS OF URANIUM INDUCED BY 5.7-Bev PROTONS; RADIOCHEMICAL YIELDS OF LIGHT ELEMENTS

BERKELEY, CALIFORNIA

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ABSTRACT

Uranium was bombarded with 5.7-Bev protons. Formation cross sections are reported for products in the mass region $7 < A < 66$. The peak yields for each mass number were found to occur along the stability line or slightly to the neutron-excess side of it. The data fit in well with current ideas concerning processes involved in high-energy nuclear reactions.

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I. INTRODUCTION

Since the completion of the Bevatron at Berkeley, studies have been made of the fission-spallation products of several elements when exposed to a 5.7-Bev proton beam. These target elements include uranium,[1] copper,[2] tantalum,[3] indium,[4] and several light elements. Shudde studied products from uranium in the mass region $52 < A < 210$;[1] this work is concerned with very light products, in the mass region $7 < A < 66$. It was considered desirable to extend the data on uranium into the low-mass region at a high bombarding energy in order to obtain a more complete description of the processes involved in high-energy nuclear reactions.

In the last several years, as particle-accelerating machines of higher energies have become available, many radiochemical studies of nuclear reactions have been made. Several reasonably complete summaries of these studies have been compiled and are available in the literature.[1,5]

At low incident-particle energies ($E_p \lesssim 50$ Mev) the observed results can be explained quite adequately by the Bohr compound-nucleus theory.[6] This model describes a nuclear reaction as taking place in two steps: (a) the incident particle is absorbed by the target nucleus, forming a compound system, while the kinetic energy of the projectile is distributed among the other nucleons; (b) the compound system decays according to various possible modes of particle emission, independently of the method of formation. The first step is a relatively "slow" process; the kinetic energy of the incident particle is distributed dueing a time that is long compared with the time required for the particle to traverse the nucleus.

At higher incident-particle energies (50 Mev < E_p < 400 Mev) the data are no longer consistent with the compound-nucleus model. Excitation functions at these higher energies vary more slowly than would be predicted from a compound-nucleus mechanism. Serber has advanced the hypothesis that at higher energies the kinetic energy of the incoming particle is dissipated in collisions with individual nucleons, resulting in a nucleon cascade which occurs in a time comparable to the time required for the incident particle to traverse the nucleus.[7] Nucleons are knocked out of the nucleus with a continuous distribution of energies up to that of the incident particle, leaving the nucleus with a wide range of excitation energies. The nucleon cascade is then followed by a relatively slow de-excitation process in which nucleons and light fragments are evaporated. The increase with energy of the mean free path of a nucleon in the nucleus causes the nucleus to become increasingly transparent to the incident particle as the bombarding energy is raised, and thus the amount of excitation imparted to the nucleus increases only slowly. Studies by Folger, Stevenson, and Seaborg on the fission of uranium by 340-Mev protons[8] and the high-energy studies by Lindner and Osborne[9] indicate that at high energies fission is preceded by the evaporation of up to 25 nucleons, and that several different species may contribute to the total fission yield.

At incident-particle energies above 400 Mev, other trends become evident. Products that require large amounts of excitation energy for their formation show steeply rising excitation functions.[10] Meson production also becomes significant in this energy region. Wolfgang and others[10] have introduced the term "fragmentation" to describe a process in which π mesons are produced and reabsorbed, depositing large amounts of excitation energy in relatively small zones within the nucleus. Light fragments are then emitted in a time that is short compared with the lifetime of a compound nucleus; these fragments subsequently de-excite by evaporation. It is expected that the fragmentation process plays an increasingly important role in the production of light species (and their heavy partners) as the bombarding energy is raised into the multi-Bev region.

II. EXPERIMENTAL PROCEDURES

A. Target Arrangement

The usual target arrangement was a stack of four foils: a 1-mil aluminum guard foil, a 3-mil aluminum monitor foil, another 1-mil aluminum guard foil, and a 5-mil uranium target foil. The total thickness of this foil stack was 270 mg/cm^2. When products lighter than aluminum were to be examined, a uranium guard foil was placed between the second aluminum guard foil and the target foil.

All foils were precisely machined to dimensions of 0.746 to 0.749 in. by 2 ± 1/16 in. The edges of the stacked foils were aligned as carefully as possible with the aid of a magnifying glass, and the foils were held together with several small pieces of scotch tape. The foil stack was then inserted into a lucite holder which was attached to an aluminum probe head designed to fit the Bevatron target ram. The foil stack protruded 1.25 in. beyond the end of the lucite holder. After bombardment the foils were removed from the holder and used for radiochemical analysis.

B. Target Materials

Natural uranium foil was used as the target material. Spectrographic analysis showed the presence of 0.005% iron, 0.001% nickel, and 0.0001% manganese. The aluminum foil contained approximately 0.5% silicon, 0.05% iron, and 0.01% magnesium. Impurities present in these amounts make negligible contributions to the products formed from the uranium and aluminum foils during a bombardment.

C. Beam Monitors

In order to calculate cross sections for the products formed from uranium, it was necessary to have some measure of the number of protons striking the actual target area. This was accomplished by comparing the activities of the various nuclides formed in the uranium

foil with the Na^{24} activity formed in the aluminum monitor foil by the reaction Al^{27} (p,3pn) Na^{24}. The monitor foils were allowed to cool for about 24 hours, and then their radiations were counted directly in a proportional counter. The 15.0-hour Na^{24} activity was the only one observed in these foils after the shorter-lived activities had died out.

A number of workers have determined cross sections for the reaction Al^{27} (p,3pn) Na^{24} from threshold up into the Bev region.[11-16] In this work the cross section was assumed to be constant from 2.0 to 5.7 Bev and equal to 10.5 millibarns. The error in this value is estimated as ± 30%.

In the early stages of this work nickel was separated from the uranium foil in each bombardment and the cross section for the reaction U^{238} (p,--) Ni^{65} was determined relative to the 10.5-mb cross section for Al^{27} (p,3pn) Na^{24}. This was done with the idea of eventually using Ni^{65} as an internal monitor in bombardments in which it would be undesirable to employ a stack of aluminum foils. The radiochemical purification procedure for nickel was relatively simple, and the 2.56-hour Ni^{65} activity could very easily be resolved from the gross decay curves. One bombardment was carried out with a bare 5-mil uranium foil as the target; the results obtained using Ni^{65} as the internal monitor agreed well with results obtained previously using the external aluminum monitor. A value of 2.8 mb was found for the reaction U^{238} (p,--) Ni^{65} at 5.7 Bev; this is in good agreement with Shudde's data.[1]

D. Chemical Procedures

In all the chemical procedures used in this work the uranium target foil was dissolved in concentrated HNO_3 containing known quantities of various elements acting as inert carriers for the radioactive species to be investigated. The resultant solution was then adjusted to 4 M in HNO_3 and the uranyl nitrate was extracted with a mixture of 30% tributyl phosphate and 70% carbon tetrachloride. The elements to be investigated were then separated chemically from the aqueous phase. Most of the chemical purifications used are modifications of those used by other workers.[17-19]

The purified precipitates were dried and mounted, and after counting were analyzed by the Analytical Chemistry Group to determine the chemical yields.

Beryllium

Beryllium was first removed as the hydroxide with ammonia. The precipitate was dissolved in concentrated HCl, Fe(III) was added, and the solution was made almost neutral with NaOH. It was then poured slowly with vigorous stirring into another cone containing enough 20% NaOH to give the resultant solution a base concentration of about 5%. The solution was digested and the $Fe(OH)_3$ was centrifuged out. One milligram more of Fe(III) was added to the supernate, the mixture was stirred vigorously, and then it was centrifuged. The $Fe(OH)_3$ was saved for recovery of any beryllium that might have been carried down. The supernate containing beryllate ion was neutralized with HCl, and $Be(OH)_2$ was reprecipitated with ammonia. This precipitate was dissolved in 3 N NaOH and the $Fe(OH)_3$ scavenging step was repeated. A saturated solution of NH_4Cl was added to the supernate from the $Fe(OH)_3$ scavenge, and the mixture was digested to reprecipitate $Be(OH)_2$, which was centrifuged, washed, and redissolved in the minimum amount of HCl. About 15 ml of a pH-6 Na_2SO_3-$NaHSO_3$ buffer was added. Beryllium was extracted into 30 ml of a 0.4 M solution of thenoyltrifluoroacetone in benzene with vigorous stirring for 20 minutes. The organic phase was then washed twice with water, twice with 8 N HNO_3, once with water, twice quickly with 1 N NaOH, and once quickly with water. Beryllium was then re-extracted into a 2:1 mixture of concentrated formic acid—concentrated HCl. This mixture was neutralized and $Be(OH)_2$ was precipitated with ammonia, washed with water, and dried. Chemical yields were about 30%.

Sodium

Sodium was removed from the target solution by precipitating NaCl from a solution saturated with HCl in an ice bath. The NaCl was dissolved in water; ferric, barium, and lanthanum carriers were added; NH_3 was bubbled through the solution; and a small amount of a freshly

prepared saturated solution of $(NH_4)_2CO_3$ was added. Potassium, rubidium, and cesium holdback carriers were added to the supernate, which was then saturated with HCl in an ice bath. The precipitate of NaCl was centrifuged and redissolved in water. This solution was made 0.5 N in HCl, copper carrier was added, and CuS was precipitated with H_2S. The supernate was boiled to remove H_2S, and NaCl was precipitated with HCl, then redissolved in water. This solution was made slightly ammoniacal; calcium, zinc, and magnesium carriers were added, a freshly prepared ammonium phosphate solution was added; and the mixture was centrifuged. To the supernate were added nickel and manganous carriers. The solution was saturated with H_2S and centrifuged, and the supernate was boiled. Potassium, rubidium, and cesium carriers were added and the solution was saturated with HCl in an ice bath to precipitate NaCl. The NaCl was dissolved and reprecipitated. The final precipitate was washed twice with ether and filtered. Chemical yields were 40% to 60%.

Magnesium

Magnesium was initially removed by the addition of $(NH_4)_2HPO_4$ to the ammoniacal target solution to precipitate $MgNH_4PO_4$. The precipitate was dissolved in the minimum volume of HCl, the solution was adjusted to 0.1 M in HCl, and the mixture was passed through a 6-mm-by-10-cm column packed with Dowex-50 cation-exchange resin. The eluate was discarded. The column was washed with water and magnesium was eluted with 6 N HCl. CuS and then $Fe(OH)_3$ scavenges were performed upon this eluate. The supernate from the hydroxide scavenge was adjusted to pH 4 with acetic acid; calcium, strontium, and barium carriers were added; and the oxalates were precipitated with saturated oxalic acid. The oxalate precipitation was repeated twice. The supernate containing magnesium was evaporated to dryness, excess oxalate was destroyed with HNO_3, and the residue was dissolved in water. $MgNH_4PO_4$ was precipitated from this solution and the whole procedure was repeated. The final $MgNH_4PO_4$ precipitate was washed twice with water and twice with ethanol. Chemical yields were about 20%.

Silicon

Silicon was present in the target solution as fluosilicate ion. Boric acid was added to complex the fluoride ion and silicic acid gel was precipitated by digestion of the solution with concentrated $HClO_4$. The gel was dissolved in NaOH solution and a titanium scavenge was performed. The solution containing silicate ion was then acidified with HCl and digested with $HClO_4$ to reprecipitate silicic acid. The solution, scavenging, and reprecipitation steps were repeated twice. The final silicic acid precipitate was washed with dilute HCl, water, and ethanol. Yields were about 50%.

Phosphorus

Phosphorus, as phosphate, was removed from the target solution by precipitating zirconium phosphate. This precipitate was washed with water and dissolved in 0.2 ml of 48% HF. To this solution were added 10 ml of 3 N HCl and one milligram of As(V) carrier. The mixture was saturated with H_2S while being heated in a steam bath. Lanthanum carrier was added to the supernate to precipitate LaF_3. To this precipitate were added 4 ml of concentrated HNO_3 and 5 ml of ammonium molybdate reagent. The ammonium phosphomolybdate precipitate was dissolved in 0.5 ml of NH_4OH. Ten milliliters of water and 4 drops of 30% H_2O_2 were added, and the solution was stirred vigorously. Zirconium phosphate was precipitated from this solution by the addition of 10 ml of concentrated HCl followed by zirconium carrier. The zirconium phosphate was separated and the procedure was repeated twice, the second time only through the precipitation of ammonium phosphomolybdate. This precipitate was dissolved in 1 ml of concentrated NH_4OH; then 2 ml of citric acid solution (0.5 g/ml) plus 10 ml of magnesia mixture were added. NH_4OH was added dropwise until $MgNH_4PO_4$ started to precipitate, and then 4 ml of NH_4OH were added in excess. The mixture was allowed to stand in an ice bath for 10 minutes. The $MgNH_4PO_4$ was centrifuged off, washed with 1:20 NH_4OH, and redissolved in dilute HCl. The precipitation of $MgNH_4PO_4$ was then repeated and the final precipitate was washed with 1:20 NH_4OH, 50% ethanol, and 95% ethanol. Chemical yields were about 30%.

Sulfur

Sulfur, as sulfate ion, was separated by precipitating $BaSO_4$ from the target solution, which had been made 1 N in HCl. This precipitate was metathesized for 30 minutes in a steam bath with a 20% NaOH solution saturated with Na_2CO_3. The supernate was then neutralized with HNO_3 and scavenged with $Fe(OH)_3$ and $La(OH)_3$. The supernate from the hydroxide scavenge was made slightly acidic and a AgCl scavenge was performed. To the solution containing sulfate ion were then added Se(IV) and Te(IV) carriers followed by about 1 g of chromous acetate. The mixture was digested and filtered. $BaSO_4$ was precipitated from the filtrate with $BaCl_2$, and the metathesis was repeated. The solution containing sulfate ion was acidified, the selenium and tellurium scavenge was repeated, and $BaSO_4$ was reprecipitated. This final precipitate was washed with water and ethanol. Chemical yields were 20% to 45%.

Chlorine

The target foil was placed in a small distilling flask along with the chlorine carrier (NaCl). Concentrated HNO_3 was added and the system was immediately closed. The liberated gases were passed into a receiving vessel containing 5 ml of 6 N NaOH solution plus several drops of H_2O_2. The distilling flask was heated for several minutes after the target foil had completely dissolved. The solution in the receiving vessel was neutralized with HNO_3 and then made slightly basic with NH_4OH. Fluoride carrier was added and CaF_2 was precipitated by the addition of $Ca(NO_3)_2$ solution. The supernate was made 0.5 N in HNO_3, bromide carrier and about 0.3 g of ammonium persulfate were added, and the solution was boiled until all the Br_2 had been evolved. AgCl was then precipitated by the addition of excess $AgNO_3$. The centrifuged AgCl was dissolved in concentrated NH_4OH, and Fe(III) (as the nitrate) was added to precipitate $Fe(OH)_3$. Holdback carriers of copper, nickel, cobalt, and zinc were added to the supernate and the solution was acidified with HNO_3. Excess silver ion was added to the acidic solution to ensure complete precipitation of the AgCl. The final precipitate was washed with water and acetone. Chemical yields were about 60%.

Potassium

The target solution was adjusted to pH 5 with acetic acid and cooled in an ice bath, and $K_3Co(NO_2)_6$ was precipitated by the addition of excess $Na_3Co(NO_2)_6$. The precipitate was washed and dissolved in 1 ml of concentrated HCl. HNO_2 was boiled off, the solution was diluted to 10 ml, and a CuS scavenge was performed. H_2S was boiled out of the supernate; ferric, lanthanum, and barium carriers were added; the solution was made ammoniacal; and 1/2 ml of saturated $(NH_4)_2CO_3$ solution was added. Nickel and manganous carriers were added to the supernate and the solution was saturated with H_2S. After centrifuging, the supernate was acidified with HCl and boiled, and the chlorides and ammonium salts were destroyed by evaporating to dryness with HNO_3. Cesium and rubidium holdback carriers were added before the evaporation. The residue was taken up in 5 ml of 72% $HClO_4$ and the mixture was evaporated to dryness. Again, 5 ml of $HClO_4$ were added to the residue and the mixture was washed into a small heavy-walled cone with several portions of chilled absolute ethanol. The cone was cooled in an ice bath and the precipitated $KClO_4$ was centrifuged off. This precipitate was dissolved in a small amount of water, the solution was evaporated to dryness, and the evaporation and precipitation with $KClO_4$ were repeated. The final $KClO_4$ precipitate was filtered and washed with ethanol. During one of the runs an extra step was incorporated into the procedure.[20] Following the first $HClO_4$ precipitation, the $KClO_4$ was dissolved in water, cesium and rubidium carriers were added, and the solution was passed through a 6-mm-by-12-cm column packed with acid-washed Duolite C-3 cation-exchange resin. Potassium was eluted from the column with 0.1 N HCl, a process which took an inconveniently long time. With this concentration of acid a good separation can be achieved between potassium and rubidium; when a higher concentration was used in order to reduce the elution time, it was found that potassium and rubidium came off at best very close together, and, in a few cases, together. Also, the use of a flame photometer was required in order to determine which fractions contained potassium. The method was rather time-consuming and did not seem to give an appreciable improvement in radiochemical purity, therefore it was not used in subsequent bombardments.

Calcium

Calcium was precipitated from the ammoniacal target solution as $CaCO_3$. The precipitate was dissolved in the minimum volume of HNO_3, the solution was diluted to 10 ml with water, and a AgCl scavenge was performed. The supernate was adjusted to 0.1 M in HCl and a CuS scavenge was performed. The supernate was boiled to remove H_2S, ferric carrier was added, and $Fe(OH)_3$ was precipitated by the addition of NH_4OH. Nickel and cobalt carriers were added to the ammoniacal supernate and the solution was saturated with H_2S. The supernate from the basic sulfide scavenge was boiled and then a 4% solution of $(NH_4)_2C_2O_4$ was added. The CaC_2O_4 precipitate was dissolved in a small amount of dilute HNO_3, and barium and strontium carriers were added. Thirty milliliters of white fuming HNO_3 was added and the mixture was cooled in an ice bath. The mixed barium and strontium nitrates were centrifuged off and the supernate, containing calcium, was evaporated to 1 to 2 ml. Barium and strontium carriers were added to the supernate and the treatment with fuming HNO_3 was repeated. The supernate was evaporated to 2 to 3 ml and made up to 30 ml with water. This solution was made ammoniacal and CaC_2O_4 was precipitated by the addition of $(NH_4)_2C_2O_4$. The precipitate was dissolved in HNO_3 and 2 ml of 1 M $NaBrO_3$ solution was added. This solution was diluted to 30 ml and made strongly ammoniacal; then $CaCO_3$ was precipitated by the addition of $(NH_4)_2CO_3$. The $CaCO_3$ was dissolved in HNO_3 and the solution was diluted to 25 ml. Lanthanum carrier was added and $La(OH)_3$ was precipitated by the addition of NH_4OH. Finally, CaC_2O_4 was precipitated from the supernate by the addition of $(NH_4)_2C_2O_4$, and was washed with water and ethanol. Chemical yields were about 20%.

Manganese

Manganese carrier was added to the target solution as manganous ion. The 4 M HNO_3 solution from the uranium extraction was evaporated to dryness with HCl to destroy the HNO_3. The residue was taken up in water and the resulting slightly acidic solution was saturated with SO_2 to ensure that all the manganese was present as manganous ion. After boiling of the solution to expel excess SO_2, excess solid $KBrO_3$ was added

and the mixture was heated until MnO_2 precipitated. The MnO_2 was dissolved in concentrated HCl. This solution was boiled to expel Cl_2, and was then passed through a 6-mm-by-10-cm column packed with Dowex A-2 anion-exchange resin previously washed with concentrated HCl. The eluate was evaporated to reduce its volume and was made 1 N in HCl. Silver carrier was added and the AgCl was removed by centrifugation. The supernatant solution was then scavenged with CuS. The supernate from this operation was evaporated to dryness with HNO_3. The residue was dissolved in water and the solution was made 0.1 N in HNO_3. Excess solid $NaBiO_3$ was then added to oxidize manganous ion to permanganate. NaOH was added until the solution had a pH of 10, and a $Fe(OH)_3$ scavenge was performed. The supernate was made 1 N in HNO_3 and the permanganate was reduced to the plus-two state by the addition of oxalic acid. Excess solid $KBrO_3$ was added and the solution was heated until MnO_2 precipitated. This precipitate was washed with dilute HNO_3 and the purification procedure was repeated. After the second purification cycle the MnO_2 was dissolved in dilute HNO_3 containing some H_2O_2, the solution was diluted to 10 ml with water, and MnO_2 was reprecipitated with $KBrO_3$. The final precipitate was washed with water and acetone. Chemical yields were less than 20%.

Iron

The target solution containing ferric ion as carrier was made basic with NH_4OH. The $Fe(OH)_3$ that precipitated was washed and then dissolved in a small volume of concentrated HCl. Five milligrams of gallium carrier was added and the solution was passed through a 6-mm-by-10-cm column packed with Dowex A-2 anion-exchange resin previously washed with concentrated HCl. The column was washed once with 12 N HCl and once with 6 N HCl, and iron was eluted with 0.5 N HCl. Molybdenum, gallium, antimony, arsenic, germanium, tellurium, and tin carriers were added to the eluate and the solution was saturated with H_2S. The supernate was made basic with NH_4OH and was again saturated with H_2S. The FeS precipitate was dissolved in the minimum volume of concentrated HNO_3. Tungsten and barium carriers were added, a few drops of concentrated H_2SO_4 were added, and the precipitate was discarded. Five milligrams of

gallium carrier was added to the supernate and $Fe(OH)_3$ was precipitated by the addition of NaOH. The $Fe(OH)_3$ was dissolved in concentrated HCl. The solution was made 7.5 N in HCl and the iron was extracted into isopropyl ether. The ether phase was washed with 7.5 N HCl and was then evaporated over water. Molybdenum, gallium, antimony, tin, and germanium carriers were added to this solution and $Fe(OH)_3$ was precipitated by the addition of NaOH. The $Fe(OH)_3$ was dissolved in HCl and the extraction and precipitation steps were repeated. The purified $Fe(OH)_3$ was washed with water and was filtered. Chemical yields were 30% to 50%.

Nickel

The target solution containing nickel carrier was made basic with NH_4OH, a saturated solution of dimethylglyoxime was added, and the nickel dimethylglyoxime precipitate was filtered off, washed, and dissolved in the minimum volume of concentrated HNO_3. The solution was boiled, diluted with water, and adjusted to a pH of 1. A AgCl scavenge was performed; this was followed by a mixed copper and palladium sulfide scavenge. The supernate was boiled to expel H_2S, ferric carrier was added, and $Fe(OH)_3$ was precipitated by the addition of NH_4OH. Nickel dimethylglyoxime was precipitated from the supernate by the addition of dimethylglyoxime, and the purification procedure was repeated. The purified nickel dimethylglyoxime was washed with dilute NH_4OH, water, and ethanol. Chemical yields were 30% to 60%.

E. Sample Mounting

The final purified precipitate was washed into a filter chimney with an inside diameter of 18.5 mm. The precipitate was collected on a circle of Whatman Filter Paper No. 42 of 7/8-in. diameter, supported by a 7/8-in. diameter sintered glass frit. The total sample area was 2.7 cm^2. After washing, the precipitate was dried for 15 minutes at 110°C. The filter paper holding the sample was then placed on a 1-in.-square piece of double-sided masking tape which had previously been centered

on a standard aluminum sample holder, 2.5 by 3.5 in. by 350 mg/cm^2. A 1-in.-square piece of rubber hydrochloride film (0.6 mg/cm^2) was then placed over the sample and stuck on the edges to the masking tape. With this arrangement portions of the sample could not be lost during the counting operations.

The aluminum monitor foils were cut into three pieces of equal size and each piece was mounted on an aluminum plate with double-sided tape. These foils were not covered with rubber hydrochloride film.

III. TREATMENT OF DATA

A. Cross-Section Calculations

The rate of formation and decay of a single radioactive species during a bombardment is expressed by the differential equation

$$\frac{dN}{dt} = In_t\sigma - \lambda N,$$

where I is the number of protons hitting the target per unit time,

n_t is the number of target nuclei per square centimeter,

σ is the formation cross section of the product in square centimeters,

λ is the decay constant of the product,

N is the number of atoms of the product.

Under the conditions that the beam current I is constant and that N equals zero at the beginning of the bombardment, the above equation has the solution

$$\sigma = \frac{N^0\lambda}{In_t\,(1-e^{-\lambda t})},$$

where t is the duration of the bombardment and N^0 is the number of atoms of the product present at the end of the bombardment.

In this study, all cross sections were compared with that for Na^{24} production in aluminum. Letting the subscript "1" denote the nuclide of interest and letting "2" denote Na^{24} produced in the aluminum foil, we have

$$\sigma_1 = \frac{A_1^0}{A_2^0} \cdot \frac{W_2}{W_1} \cdot \frac{M_1}{M_2} \cdot \frac{(1-e^{-\lambda_2 t})}{(1-e^{-\lambda_1 t})} \cdot \sigma_2 ,$$

where the A^0's are the disintegration rates at the end of bombardment,

the w's are the weights of the foils used,

the M's are the atomic weights of the foil materials.

Shudde gives an expression that is useful for the interrupted bombardments sometimes experienced on the Bevatron.[1]

B. Beta Counting

An end-window methane-flow proportional counter with an 1.0-mg/cm^2 aluminized mylar window was used to count beta radiation.

Several corrections must be applied in converting observed counting rates to disintegration rates. Of these, the most important are geometry, air-and-window absorption, backscattering, self-scattering and self-absorption, radiation abundance, and chemical yield. Coincidence corrections for the type of counter used are negligible up through 10^5 counts per minute and were neglected. It was assumed that all beta particles that penetrated the counter window were counted.

Owing to the low activities encountered in this work, all samples, including monitors, were counted on the top shelf of the sample holder. It was calculated, from data provided by Dr. D. W. Barr,[21] that this shelf has a geometry of 0.33.

The air-and-window absorption factors, f_w, were calculated by use of the formula

$$f_w = e^{-\mu x} ,$$

where x is the air-plus-window thickness in mg/cm^2 and μ is the absorption

coefficient. Values for μ were calculated by use of the formula provided by Gleason, Taylor, and Tabern,[22]

$$\mu = 0.017 \quad E_m^{-1.43} ,$$

where E_m is the maximum beta-particle energy.

Backscattering factors, f_B, were taken from Burtt's data for negatron backscattering.[23] Positron counting rates were increased by 10% because the backscattering effect is smaller for positrons than for negatrons.[24]

Self-scattering and self-absorption factors, f_S, were obtained from the data of Nervik and Stevenson.[25]

Data on radiation abundance and beta-particle energies were taken from the compilations by Hollander, Perlman, and Seaborg,[26] and by Way et al.[27] The correction factor for radiation abundance, f_A, was unity for all the negatron emitters studied here.

With these correction factors, the expression for the disintigration rate of a nuclide is

$$D = \frac{A}{0.33 \times f_w \times f_B \times f_S \times f_A \times Y} ,$$

where D is the disintegration rate, A is the observed counting rate, and Y is the chemical yield.

C. Gamma Counting

For gamma-ray counting, a sodium iodide (thallium-activated) crystal 1.5-in. in diameter by 1-in. high was used with a 50-channel pulse-height analyzer. A thin beryllium window separated the crystal from the sample being counted.

Several corrections must be applied in converting gamma-ray counting rates to disintegration rates. They are counting efficiency, geometry, escape-peak loss, radiation abundance, and chemical yield.

Counting efficiencies for the size of crystal used at Berkeley have been determined by Kalkstein and Hollander;[28] their data were used exclusively in this work.

Geometry factors were also taken from the data of Kalkstein and Hollander.[28]

The integrated photopeak counting rates were corrected for escape-peak loss by means of the data of Axel.[29]

Radiation abundances were taken from the compilation by Hollander, Perlman, and Seaborg.[26]

The disintegration rate of a nuclide is then related to its gamma-ray counting rate by the equation

$$D = \frac{A}{CE \times G \times EP \times R \times Y},$$

where D is the disintegration rate,

A is the observed counting rate,

CE is the counting efficiency,

G is the geometry factor,

EP is the correction for the escape-peak loss,

R is the radiation abundance,

Y is the chemical yield.

D. Decay Characteristics

Beryllium

The 478-kev gamma rays accompanying the decay of 53-day Be^7 were counted in the gamma scintillation counter. The abundance of this radiation was taken to be 12%.[30] No beta activity was detected in the beryllium samples.

Sodium

The negatron activity from 15.0-hour Na^{24} was detected in a proportional counter. In samples from long bombardments the 15.0-hour activity tailed into several counts per minute of a long-lived activity which decayed only slightly over a long period of time. This was

attributed to 2.6-year Na^{22}, which decays 90% by positron emission. Both the 0.51-Mev annihilation radiation and the 1.28-Mev gamma ray from the decay of Na^{22} were easily detected on the scintillation spectrometer.

Magnesium

The only activity observed in the magnesium samples was 21.3-hour Mg^{28}. The negatrons from this nuclide were counted in equilibrium with those from the 2.30-minute Al^{28} daughter. The counting was done on a proportional counter.

Silicon

The silicon samples were counted in a proportional counter. The only activity observed was that of 2.62-hour Si^{31}, a negatron emitter.

Phosphorus

The phosphorus samples were counted in a proportional counter to detect the negatrons from 14.4-day P^{32} and 24.8-day P^{33}. Least-squares analysis and Biller plots were used to resolve the two activities.

Sulfur

The first two sulfur samples examined in this work were separated from foils that had been allowed to cool for several days after having been bombarded. As a result, the only activity observed in these samples was that of 87.1-day S^{35}, which decays completely by negatron emission. However, in two later bombardments the samples were purified and counted immediately, and both showed the 170-minute activity of S^{38}, which was discovered recently by Nethaway and Caretto.[31] The samples were counted in a proportional counter, and the assignment of the 170-minute activity to S^{38} was confirmed by using the scintillation spectrometer to detect the 1.88-Mev gamma ray[31] from the decay of this isotope.

Chlorine

The decay of 32.4-minute Cl^{34m} was studied by observing the 510-kev annihilation radiation from the positrons emitted in 100% of the

decays of this nuclide. The disintegration rate of Cl^{34m}, as determined in this manner, was converted to the corresponding proportional-counter counting rate. The latter was then subtracted from the gross proportional-counter decay curves to give the combined activities of 37.3-minute Cl^{38} and 55-minute Cl^{39}, which are both negatron emitters. The individual activities of Cl^{38} and Cl^{39} were then resolved by least-squares analysis.

Potassium

Least-squares analysis was used to resolve 12.5-hour K^{42} and 22.4-hour K^{43} from proportional-counter decay curves. An activity with a half-life of about 30 minutes was also observed in the potassium samples, and was attributed to 22-minute K^{44} and 34-minute K^{45}.[32] However, the data were never sufficient to resolve this short activity into two components.

Calcium

The calcium samples were counted in a proportional counter over a time sufficiently long so that the decay curves showed only the 164-day activity of the negatron emitter Ca^{45}. This activity was subtracted from the gross decay curves and a least-squares analysis was done to separate the activities of 4.8-day Ca^{47} and its daughter, 3.43-day Sc^{47}.

Manganese

The gross proportional-counter decay curves were easily resolved into the activities of 5.72-day Mn^{52} and 2.58-hour Mn^{56}. Mn^{52} decays 33% by positron emission, and Mn^{56} decays by negatron emission.

Iron

The activities of 8.3-hour Fe^{52} and 45.1-day Fe^{59} were easily resolved from the proportional-counter decay curves. Fe^{52} decays 59% by positron emission and was counted in equilibrium with 21.3-minute Mn^{52m}, which in turn decays 98.5% by positron emission. Fe^{59} is a negatron emitter.

Nickel

Only two activities were observed in the nickel samples. The negatron-emitting 2.56-hour Ni^{65} was easily resolved on proportional-counter decay curves from 56-hour Ni^{66}, which was counted in equilibrium with 5.1-minute Cu^{66}.

IV. RESULTS AND DISCUSSION

The cross sections measured are presented in Table I and are shown graphically in Fig. 1. Yields are in millibarns and are based on the value of 10.5 mb assumed for the reaction Al^{27} (p,3pn) Na^{24} at 5.7 Bev. Errors due to counting statistics, uncertainties in counting corrections, analysis of decay curves, etc., are estimated to be about ± 20%. This does not include the ± 30% error estimated for the absolute-cross-section standard.

The results obtained in this study, when combined with Shudde's work on uranium fission induced by 5.7-Bev protons,[1] are consistent with present ideas concerning high-energy reactions. The cross sections measured by Shudde are presented in Table II.

At lower energies ($E_p \lesssim 0.40$ Bev) the products from bombardment of the heavier elements (e.g., lead, bismuth, thorium, and uranium) fall into several clearly defined mass regions. The spallation region includes nuclides within 30 or 40 units of A of the target; the formation cross sections of these nuclides decrease sharply with decreasing A. There is a corresponding region composed of extremely -low-mass nuclides (neutrons, protons, and helium nuclei) which are the partners of the heavy spallation products; in this second region yields decrease sharply with increasing A. The fission region lies between these two regions and is separated from them by valleys of very low yields.

Table I

Cross sections for production of nuclides from uranium bombarded by 5.7-Bev protons

Nuclide	$T_{1/2}$	E_γ	Cross section (millibarns)	Type of Yield*
Be^{7}	53d	.48	15.	C
Na^{22}	2.6y	1.28	2.4	C
Na^{24}	15hr	2.7, 1.3	12.	C
Mg^{28}	21.3h	1.35, .9	3.8	C
Si^{31}	2.6h	1.26	2.4	C
P^{32}	14.5D	no γ	1.8	I
P^{33}	25D	no γ	0.87	C
S^{35}	87D	no γ	3.7	C
S^{38}	?		0.77	C
Cl^{34m}	34m	2.1, 1.1	1.1	C
Cl^{38}	37m	2.1, 1.6	4.5	I
Cl^{39}	55m	3.1	2.5	C
K^{42}	12.5h	1.5, .3	7.3	I
K^{43}	22hr	.6	4.5	C
$K^{44,45}$	<		2.9	C
Ca^{45}	160D	no γ	5.6	C
Ca^{47}	4.7D	1.3, .8	1.6	C
Mn^{52}	5.7D	1.4, .9	1.3 (ground state only)	~ I
Mn^{56}	2.58h	1.8, 2	7.1	C
Fe^{52}	8hr	1.4, .4	0.045	C
Fe^{59}	45D	1.1, 1.4	3.7	C
Ni^{65}	2.56h	1.5, 1.1	2.8	C
Ni^{66}	56hr	?	1.6	C

*C and I are used to designate cumulative yields and independent yields, respectively.

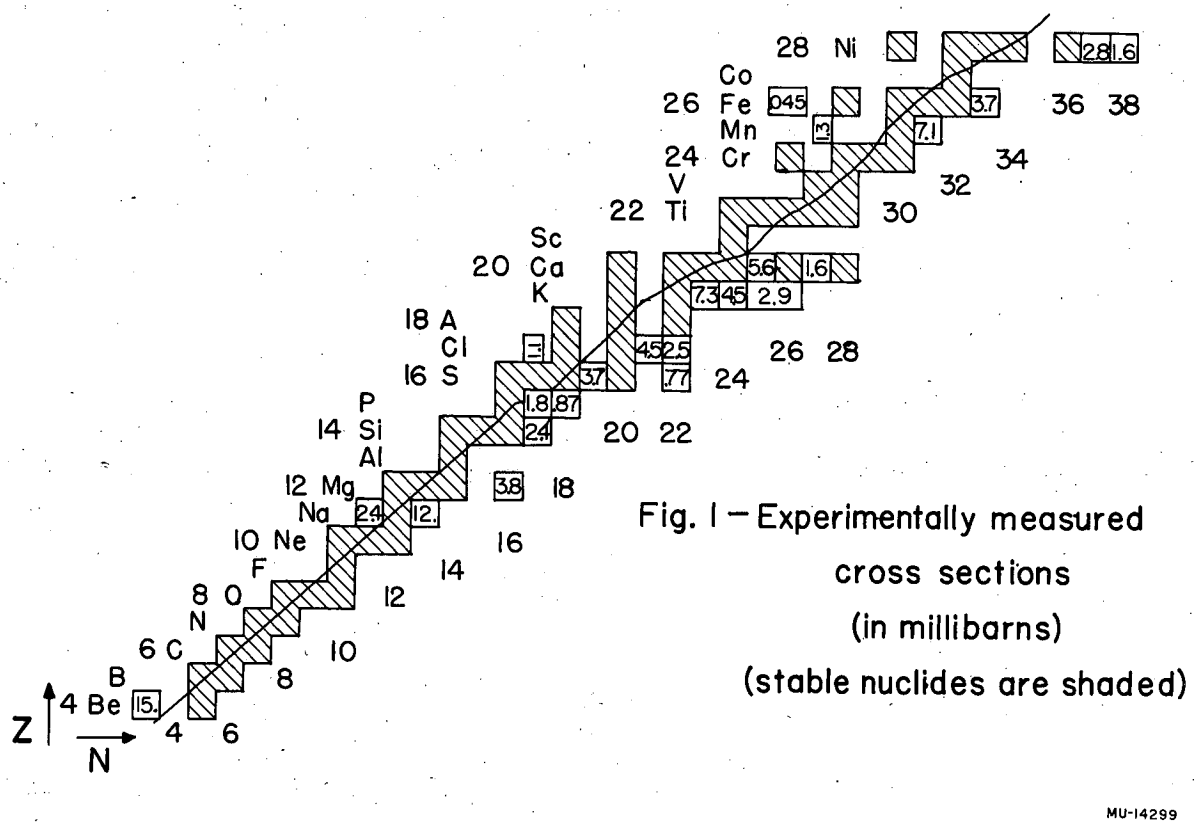

Fig. 1. Experimentally measured cross sections (in millibarns) (Stable nuclides are shaded).

Table II

Cross sections for production of nuclides from uranium bombarded by 5.7-Bev protons (measured by R. H. Shudde[1])

Nuclide	Cross section* (millibarns)	Type of Yield
Mn^{52}	0.68	~ I
Mn^{56}	6.3	C
Fe^{59}	2.3	C
Co^{61}	6.5	C
Ni^{65}	2.8	C
Ni^{66}	1.2_5	C
Cu^{61}	0.53	C
Cu^{64}	5.4	I
Cu^{67}	4.0	C
Zn^{69m}	2.0	C
Zn^{72}	0.68	C
Ga^{68}	7.1	I
Ga^{72}	5.4	~ I
Ga^{73}	6.1	C
Mo^{99}	42.	C
Ru^{103}	28.	C
Ru^{105}	28.	C
Ru^{106}	25.	C
Pd^{109}	23.	C
Pd^{111m}	2.7	C
Pd^{111}	7.2	C
Pd^{112}	1.1_5	C
Ag^{110m}	3.0	I
Ag^{111}	$21._5$	C
Ag^{112}	9.5	~ I
Ag^{113}	15.	C
Ag^{115}	7.3	C

(continued)

Table II (continued)

Nuclide	Cross section* (millibarns)	Type of Yield
Cd^{115m}	9.5	C
Cd^{115}	28.	C
Cd^{117m}	13.	C
In^{111}	5.9	C
In^{114m}	4.8	I
Ba^{128}	20.	C
Ba^{140}	32.	C
La^{140}	0.9	I
$La^{132,133,141}$	29.	-
Ta^{178}	5.9	I
Ta^{183}	8.6	C
Pb^{204m_2}	15.	~ I
Pb^{209}	3.9	C
Pb^{212}	0.47	C
Bi^{210}	3.0	C

*Cross sections presented in Table II have been adjusted to a value of 10.5 mb for the monitor reaction. Shudde's original figures were based on a value of 8.3 mb.

As the bombarding energy is raised, several trends become evident. Comparisons of yields of products resulting from the bombardment of uranium with 340-Mev,[8] 480-Mev,[33] 3.0-Bev,[34] and 5.7-Bev[1] protons; of bismuth with 340-Mev,[35] 480-Mev,[33] 660-Mev,[36] and 2.2-Bev[37] protons; and of lead with protons in the energy range 0.6 to 3.0 Bev,[10] show that as the bombarding energy is increased, significant changes occur in the yields-vs-mass curves. The over-all curves begin to "flatten out" with increasing energy, and the deep valleys between the fission and spallation regions progressively become shallower. Quantitatively, chain yields in the fission region tend to remain about the same within an order of magnitude, while the yields of products in the valleys may increase by several orders of magnitude. Wolfgang et al.[10] have found that in bombardment of lead with 3.0-Bev protons the cross sections for forming a product of any mass number below the target mass are the same within an order of magnitude, in striking contrast with results obtained at lower energies.[33,35] Thus the fission and spallation regions are no longer well-defined, and it is sometimes very difficult to assign specific mechanisms to the production of various nuclides.

These same trends are apparent when yields of products obtained in this work are compared with yields obtained from uranium at lower proton energies. The yield of Na^{24} at 5.7 Bev is 200 times as high as that at 0.34 Bev;[8] the P^{32} yield is 250 times as high as at 0.48 Bev;[33] the Fe^{59} yield is 20 times that at 0.34 Bev;[8] while the yields of nickel isotopes are 2 to 5 times as high as at 0.34 Bev.[8]

Wolfgang et al. have found that the excitation function for Na^{24} production from lead increased only from 2 mb to about 4 mb between 2.0 and 3.0 Bev, while the yields of P^{32}, Mg^{28}, and F^{18} leveled off at a value of about 0.5 mb in the same energy range.[10] The yields of Na^{24}, P^{32}, and Mg^{28} obtained in this work are 6 to 9 times as high as the yields from lead at 3.0 Bev.

Caretto and Hudis have determined excitation functions for production of F^{18} and Na^{24} from various target elements at proton energies up to 6.0 Bev.[38] The curves rise steeply from a threshold in the neighborhood of 0.5 Bev and begin to level off at values of the order of millibarns

above 3.0 Bev. The sharp rise from a threshold at about 0.5 Bev suggests that a different mechanism comes into play in the Bev region, and lends credibility to the fragmentation mechanism. Curves of Na^{24} yields vs target mass for bombarding energies of 1.0 Bev, 2.0 Bev, and 6.0 Bev also exhibit interesting features.[38] The curves all show discontinuities near mass 170. The curves for the two lower energies decrease steadily to this point and then rise steeply; the 6.0-Bev curve has a minimum near mass 100 and a slight peak at mass 170, after which the yields increase slowly. If one is willing to accept the meson-production-and-reabsorption process postulated by Wolfgang et al.,[10] the increasing yields for heavy target nuclei could be explained by saying that as the target mass (and nuclear size) increase, the mesons have a greater probability of being reabsorbed and making their energy available for fragmentation. The high yields from low-mass targets are probably due to the familiar cascade-evaporation mechanism.

With the evidence of Caretto and Hudis in mind, the yields from uranium at 5.7 Bev are not inconsistent with the much lower yields from lead at 3.0 Bev. Not only is there an energy effect, but also the higher mass of uranium would contribute to higher yields.

In connection with this, Pate and Friedlander have determined cross sections for a number of protactinium and astatine isotopes produced in the bombardment of uranium with 3.0-Bev protons.[34] The protactinium yields are lower by factors of 2 to 4 than those at 340 Mev,[39] while the yields of the neutron-deficient protactinium isotopes are very low and drop off sharply with distance from the stability line. (Some of the cross sections reported are 0.17 mb for Pa^{227}, 0.64 mb for Pa^{228}, and 2.4 mb for Pa^{229}.[34]) On the other hand, the yields of astatine isotopes (At^{209}, At^{211}) with approximately the same neutron-proton ratios as the very light protactinium isotopes are of the order of a millibarn.[34] The high astatine yields, relative to the protactinium yields, are difficult to explain in terms of spallation followed by successive alpha decays. However, if it is assumed that relatively large fragments are broken off from an excited protactinium nucleus in a fast process, producing high yields of entities in the region of astatine, then correspondingly high

yields would be expected in the low-mass region. High yields are indeed observed in the low-mass region in this work and in other high-energy studies mentioned previously.

Close examination of the data presented in Table I, combined with some admittedly crude interpolations, indicates that the maximum yields for each mass number occur along the stability line or slightly to the neutron-excess side of it. This is consistent with the fragmentation mechanism. This mechanism would be fast compared with the lifetime of a compound nucleus, and the neutron-proton ratios in the initial fragments are expected to be the same as in the undisrupted nucleus. Thus light fragments ($A \sim 40$) would be initially neutron-rich. In the de-excitation process the evaporation cascade would tend first toward the stability line and then, owing to the low Coulomb barriers toward proton emission in this region, along the stability line.

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