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DEUTERON-INDUCED SPALLATION AND FISSION REACTIONS IN PLUTONIUM ISOTOPES

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UCRL-3495

UNIVERSITY OF CALIFORNIA

Radiation Laboratory
Berkeley, California

Contract No. W-7405-eng-48

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(Master's Thesis)

November 1956

Printed for the U. S. Atomic Energy Commission

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ABSTRACT

Spallation products produced in bombardments of Pu^{238} and Pu^{240} with 9 to 23 Mev deuterons were isolated by chemical methods. In addition, fission products of Pu^{240} were isolated. Various isotopes of the elements separated were identified by their radioactive properties. Fission-yield curves at various energies are presented as well as absolute fission and spallation excitation functions. A marked suppression of the (d,2n) and (d,3n) reactions of Pu^{238} was found in comparison to those of Pu^{240}, apparently due to the increased fissionability of Pu^{238}. The cross section for the (d,α) reaction of Pu^{238} was determined for one deuteron energy. The usual increase of symmetry in the fission-yield curves of Pu^{240} was observed with the minimum vanishing with approximately 19 Mev deuterons.

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I. INTRODUCTION

This work comprises a radiochemical study of the fission and spallation reactions of Pu^{238} and Pu^{240} induced by deuterons ranging from 9 to 23 Mev in energy. The deuterons were accelerated in the Crocker Laboratory 60-inch cyclotron.

The reactions of bombarding particles with nuclei can be classified as elastic scattering, inelastic scattering, capture, spallation, and fission. With charged particles, only spallation and fission can be studied by radiochemical methods. In spallation reactions small particles such as protons, neutrons, tritons, and alpha particles are emitted from the bombarded nuclide. In fission reactions the bombarded nuclide breaks into two large fragments in addition to emitting neutrons. These different reactions can be considered as a competition in which one reaction can become prominent only at the expense of the other types, since the sum of the cross sections for the individual reactions approaches the geometric cross section of the bombarded nuclide. This does not hold, of course, for thermal energies where the de Broglie wave lengths of the particles are very important or for energies above 50 Mev where the nucleus becomes somewhat "transparent" to the bombarding particles.[1] Factors such as atomic number, mass number, odd-even character of the target nuclei, and the energy of the bombarding particle determine which reaction predominates.

Mechanisms for spallation fit into two categories, low energy (< 50 Mev) and high energy (> 50 Mev). In spallation induced by low-energy particles the mechanism is usually described by the compound-nucleus theory. According to this theory the incident particle loses its kinetic energy in the first few collisions with nucleons in the bombarded nucleus and is then

held by nuclear forces. The energy contributed by the incident particle is rapidly distributed among all the nucleons. The breaking up takes place only after a relatively long time because a large number of collisions is necessary before sufficient energy can be concentrated on any one nucleon to allow it to escape. It is readily seen that the number of nucleons that can be emitted increases with the energy of the incident particle. According to the compound-nucleus theory the method of de-excitation should not depend on the way the compound nucleus is formed but only on its excitation energy, angular momentum, and parity.[2] Comparison of the excitation functions found by bombarding Cu^{63} with protons and Ni^{60} with helium ions shows agreement with the compound-nucleus theory.[3] Agreement with the theory is also obtained from the comparison of the excitation functions of Pu^{239} bombarded with deuterons and those of Np^{237} bombarded with helium ions.[4] However, there are experimental results of other reactions which indicate that compound-nucleus formation may not be taking place.[5-12] For example, the angular distribution of neutrons produced in (p,n) reactions is strongly peaked forward.[5]

At higher energies (> 50 Mev) the compound nucleus breaks down and other mechanisms such as the direct-interaction mechanism must be employed.[1] The compound-nucleus theory postulates that proton emission is much less probable than neutron emission from high-Z targets because Coulombic repulsion tends to keep protons inside the nucleus. If there is direct interaction, even at low energy, of the bombarding particles with the target nucleons, then the probability for proton emission is greater.[13]

Fission reactions with various bombarding particles have been observed in all parts of the periodic table.[14] Among heavy elements and for low bombarding energies there seems to be a relationship between Z^2/A of the target nucleus and the ease with which the nuclide fissions.[15] As the atomic number of the target increases, the energy requirements for fission to occur decrease. High-energy protons, deuterons, and helium ions were required to produce fission in copper, tantalum, and other medium-weight elements,[16-19] whereas thermal neutron fission occurs in Pu^{239}, U^{235}, and U^{233}. As in spallation, there are marked differences between fission reactions induced by low-energy projectiles and those induced by high-energy projectiles. Low-energy fission is asymmetric and

the primary fission fragments have a neutron excess and are all beta-particle emitters. Some features of the mechanism of low-energy fission can be described by the liquid-drop model,[15] although this model cannot explain the asymmetry. At higher energies fission becomes symmetric and the primary fission fragments have a lower neutron-to-proton ratio than in asymmetric fission. Goeckermann and Perlman[16] proposed that 10 to 12 neutrons are evaporated before fission takes place for 190-Mev deuteron-induced fission of Bi^{209}.

Since fission in the medium-Z and low-Z areas of the periodic table requires high-energy projectiles and even then low cross sections are observed, the area in which to study fission and spallation reactions is the heavy-element region. In this region projectiles of low energies may be used, and the shift from asymmetric fission to symmetric fission can be observed. At the same time there is a lower number of possible spallation products, which makes the study much easier. The work reported here is part of a series of studies of medium-energy fission and spallation reactions of heavy nuclei with helium ions and deuterons. This series was undertaken with the hope of finding systematic trends in the various excitation functions with change of mass, atomic number, and nuclear type of the target material. Part of this series is completed--, namely Pu^{239} bombardments with helium ions,[20] Pu^{238} and Pu^{242} bombardments with helium ions,[21] U^{238} bombardments with helium ions,[22] Pu^{239} and U^{233} bombardments with deuterons,[23] and Np^{237} bombardments with helium ions.[23] In the present study excitation functions for deuteron-induced reactions of Pu^{238} and Pu^{240} were studied to reveal the change of the excitation functions with change of mass when Z is not constant. According to one concept,[30,31] in going from Pu^{240} to Pu^{239} to Pu^{238}, Z^2/A increases, and thereby the fission cross section should increase, decreasing the cross section remaining for the other possible reactions, mostly spallation. The (d,2n), (d,3n), and (d,f) reactions were observed in Pu^{240}, and the (d,n), (d,2n), and (d,3n) reactions in Pu^{238}. One value for the (d,α) cross section of Pu^{238} was obtained at 20 Mev.

II. EXPERIMENTAL PROCEDURES

A. Target Preparation

A uniform target is required in order to calculate absolute cross sections in the case where all the beam hits the target. There are several methods of obtaining a uniform deposit, including sublimation and vaporization, but it was believed that electrodeposition resulted in less loss of valuable target material and required less time and equipment. The electrodeposition method used was developed by Hufford and Scott[24] and modified by Glass.[20]

Both Pu^{238} and Pu^{240} were plated by the same method. About 200 μg of plutonium in an acid solution was oxidized to plutonium (VI) by adding 1 M sodium bromate and evaporating to dryness. The excess bromate was destroyed by adding two to three drops of concentrated nitric acid and again evaporating to dryness. The residue was then dissolved in about 1 ml of 0.4 M ammonium oxalate and the resulting solution transferred to a plating cell. The anode of this cell was a platinum stirring rod and the cathode a 10-mil aluminum hat-shaped foil. If the potential drop across the cell was kept at about 4 v and the current at 100 to 200 ma, from 30 to 100 μg of platinum (as plutonium (IV) hydroxide) could be plated in 30 minutes.

The area of the target was determined by measuring several diameters.

Two methods of determining the amount of plutonium plated were used. The targets were counted in a low-geometry alpha counter for which the geometry and counting efficiency had been determined to better than 1 percent. Also the dissolved target solutions were assayed after bombardment. In almost all cases these two methods checked within 5 percent. In general the radiometric assays were lower than those obtained by direct counting, as has been noted by others.[20,23]

In the Pu^{238} bombardments the plutonium used contained 93.8 percent Pu^{238} and 5.8 percent Pu^{239}, and small amounts of Pu^{240} and Pu^{241}.[21] The plutonium used in the Pu^{240} bombardments contained 12.20 percent Pu^{239}, 87.172 percent Pu^{240}, 0.583 percent Pu^{241}, and 0.044 percent Pu^{242}.

B. Target Assembly

The target assembly used in the Pu^{240} bombardments was identical to that used by Glass.[20] Aluminum and platinum foils were used to degrade the external 24-Mev deuteron beam of the Crocker Laboratory 60-inch cyclotron. The foils were placed in an air-cooled block in front of a water-cooled pistol-grip target holder. It was found that the heating effect of the beam caused the platinum foils to soften and become bulged and thinned or to burn through entirely. Because of this, aluminum foils were used wherever possible. Beam patterns were taken before each bombardment to insure that all the beam was striking the target. The pistol-grip target holder acted as a Faraday cup, and beam integrators were used to measure the total beam current that struck the target. In this way the number of deuterons bombarding the target was determined.

The energy of the bombarding deuterons was determined by the thickness and kind of degrading foils used. All foils were carefully weighed and measured and the energy of the beam then calculated from range-energy data.[25]

The target assembly used in the Pu^{238} bombardments is illustrated in Fig. 1. This microtarget assembly has better cooling and is easier to assemble than the pistol-grip assembly. The degradation of the beam and the integration of the beam were the same as those used in the Pu^{240} bombardments.

A 1-mil aluminum cover foil was used on each target to catch fission recoils, and was dissolved along with the target.

C. Chemical Procedures

Extensive chemical procedures are necessary because of the many possible reactions induced in the target material itself and in the aluminum backing plate. Because of the small amount of valuable target material available and the low counting rates that were present in products in many cases, it was not possible to take aliquots. It was necessary to separate each of the desired products from the others and then to purify each one radiochemically. The large amount of aluminum from the target hat and cover foil introduced additional problems. The initial chemical procedures were carried out in a "glove box" because of the intense alpha radioactivity of the target material.

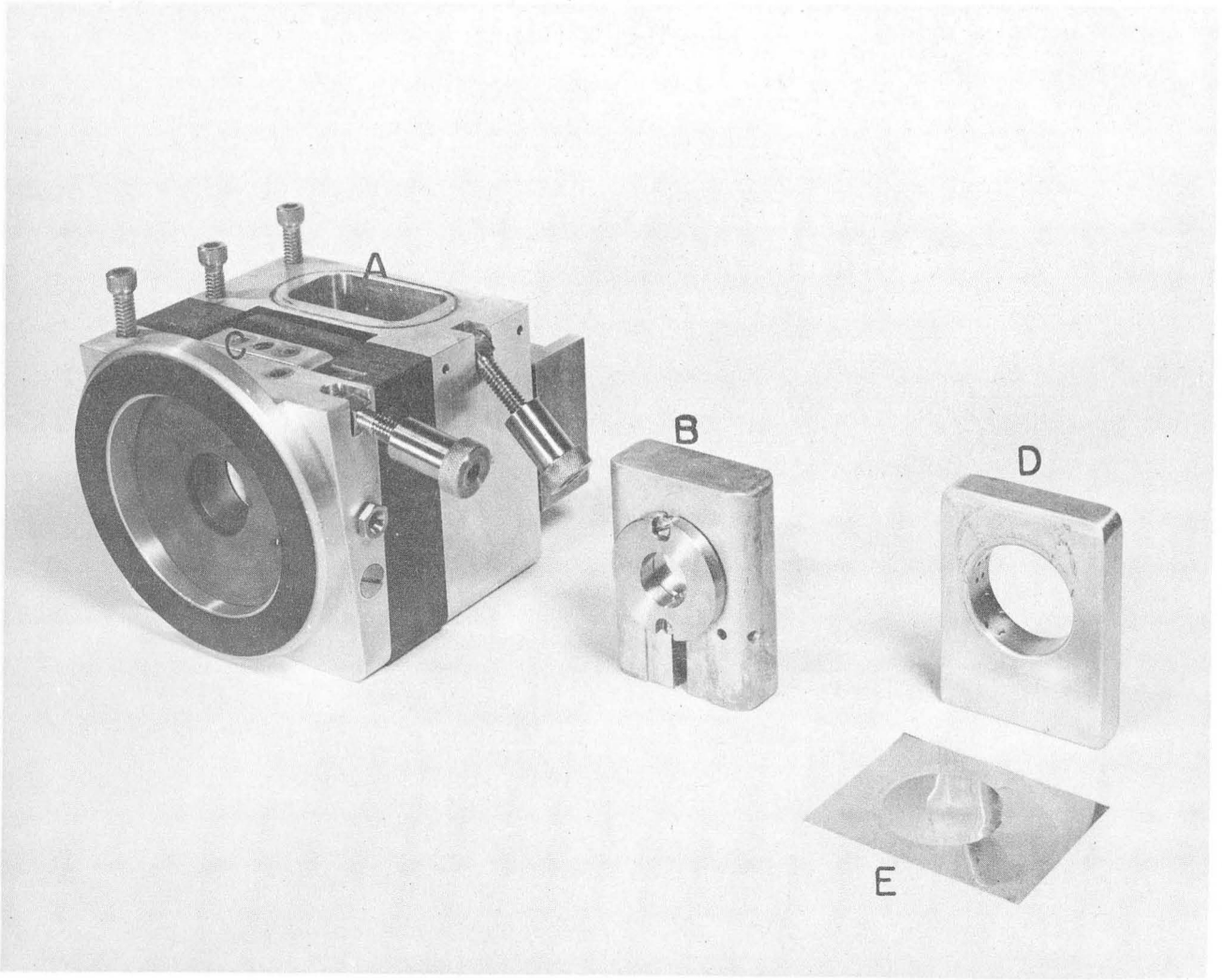

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Fig. 1. Microtarget assembly. A Microtarget slot, B Microtarget, C Collimator, D Foil holder, E Degrading foil.

1. Target Dissolution.

The target was placed in a beaker containing the carriers and tracers for the products removed. The plutonium target material, which was plated as plutonium (IV) hydroxide, was usually converted by the heating effect of the beam to the refractory oxide, which is not readily dissolved. It was found that when the solution was made 6 M to 10 M with concentrated nitric acid and about 0.01 M in flouride with hydrofluoric acid, the target material was readily dissolved when the solution was heated to about 80°C. After the plutonium was all dissolved the aluminum backing plate and cover foil were dissolved by addition of hydrochloric acid. At this point the solution was diluted to a known volume and radiometric assays were taken to check on the amount of target material present.

2. Spallation-Product Chemical Procedures.

The procedures used were modifications of ones already reported.[20,22,23]

In the Pu^{240} bombardments sodium hydroxide and sodium carbonate were added to the target solution to precipitate barium, strontium, part of the cadmium, palladium, and the rare earth elements as the hydroxides or carbonates. Americium and plutonium were carried on the precipitate. The aluminum and part of the cadmium remained in the supernate. The precipitate was dissolved in hydrochloric acid and saturated with hydrogen chloride gas to precipitate barium and strontium chloride. This precipitate was dissolved twice and re-precipitated with hydrogen chloride gas to free the barium and strontium of alpha radioactivity. The combined chloride supernates were passed through a (3 mm x 2.5 cm) glass column packed with Dowex A-1 anion-exchange resin. The americium and rare earth elements pass through the column in the concentrated hydrochloric acid fraction and the plutonium, cadmium, and palladium are adsorbed on the resin. The plutonium was desorbed by use of 1 M hydrochloric acid, and after the column had been washed with a small amount of water the palladium and cadmium were desorbed with 0.75 M sulfuric acid.

No fission products were taken out in the Pu^{238} bombardments, but the steps taken were similar to those of the Pu^{240} bombardments. Barium and lanthanum were added to the dissolving beaker as carriers.

The hydroxide-carbonate was precipitated and dissolved in hydrochloric acid. The solution was saturated with hydrogen chloride gas and the supernate passed through a column packed with Dowex A-1 anion-exchange resin. Since speed was more essential in the Pu^{238} bombardments, only one precipitation of the chloride was made. As before, the americium and rare earth elements passed directly through the column. The plutonium was desorbed with concentrated hydrochloric acid plus 0.1 M hydroiodic acid. The neptunium was then desorbed with 1 M hydrochloric acid.

Americium. Hydrofluoric acid was added to the americium and rare earth elements fraction to precipitate the fluoride. When no rare earth elements were being taken out as fission products about 0.5 mg of lanthanum was added to carry the americium. The fluoride was dissolved in nitric and boric acids and the hydroxide precipitated with an excess of ammonium hydroxide. The hydroxide precipitate was dissolved with a minimum amount of concentrated hydrochloric acid containing 20 percent absolute ethanol. This solution was saturated with hydrogen chloride gas. The americium was separated from the rare earth elements by passing them through a glass column packed with 4 percent cross-linked Dowex-50 cation-exchange resin. A solution of 80 percent concentrated hydrochloric acid and 20 percent absolute ethanol saturated with hydrogen chloride gas was used as an eluant.[20] The americium was eluted in about 5 column volumes.

Neptunium. Attempts were made in each of the Pu^{238} bombardments to separate a neptunium fraction in order to observe the (d,α) reaction. Only one attempt was successful, owing to the gross activity of plutonium present.

The neptunium from the Dowex A-1 anion-exchange column was made about 5 M acid with nitric and hydrochloric acids. Zirconium carrier, ferrous ion, and hydrazine dihydrochloride were added so that the concentration of ferrous ion was 0.005 M and hydrazine dihydrochloride was 0.1 M. The solution was heated at 50°C for about 5 minutes in order to reduce neptunium to the (IV) state and plutonium to the (III) state. Orthophosphoric acid was added to precipitate zirconium phosphate, which carried the neptunium. Plutonium remained in the supernate. The phosphate precipitate was dissolved in nitric and hydrofluoric acids. The solution was then taken from the "glove box". About 2 mg of lanthanum was added

to precipitate lanthanum fluoride, which carried the neptunium. The fluoride was dissolved in nitric and boric acids and ammonium hydroxide was used to precipitate the hydroxide. The precipitate was dissolved with hydrochloric acid and the solution adjusted to 1 M hydrochloric acid, 0.2 M hydroxylamine and heated for one minute. Stannous chloride and potassium iodide were added to ensure reduction of neptunium to the (IV) state and plutonium to the (III) state. The neptunium was extracted with 0.4 M TTA (thenoyltrifluoracetone) in benzene and then back-extracted with 8 M hydrochloric acid.

3. Fission-Product Chemical Procedures.

The fission-product isolation procedures are modifications of procedures listed by Meinke.[26]

Cadmium. The cadmium that remained in the hydroxide supernate was precipitated as the sulfide with hydrogen sulfide gas. The precipitate was dissolved in concentrated hydrochloric acid and saturated with hydrogen chloride gas. The solution was passed through a glass column packed with Dowex A-1 anion-exchange resin, which adsorbed the cadmium. The column was washed with 1 M hydrochloric acid and with water. The cadmium was desorbed with 0.75 M sulfuric acid. This cadmium solution was added to the first cadmium solution, which also contained palladium. At this point the solution was taken from the "glove box". The solution was diluted to 0.5 M acid and the sulfides precipitated with hydrogen sulfide. Cadmium sulfide was dissolved with concentrated hydrochloric acid, which does not dissolve the palladium sulfide. An antimony scavenge precipitation was made from a 2 M hydrochloric acid solution. The final cadmium sulfide precipitation was made from a 0.5 M hydrochloric acid solution.

Palladium. The palladium sulfide from the cadmium chemistry was dissolved in concentrated nitric and hydrochloric acids. An iron scavenge and a silver scavenge were performed and the final palladium precipitation was made with dimethylglyoxime.

Barium. The barium and strontium chloride precipitate from the spallation-products procedure was dissolved in water and taken from the "glove-box". The solution was buffered to a pH of 5 and barium chromate precipitated with strontium remaining in solution. The barium

chromate precipitate was metathesized to barium carbonate with sodium carbonate. The barium carbonate was dissolved and a strontium holdback agent was added. Barium chromate was again precipitated as the final product.

Strontium. The supernate from the first barium chromate precipitation was neutralized with ammonium hydroxide and strontium carbonate precipitated by adding sodium carbonate. The precipitate was dissolved and a barium chromate scavenge precipitatation was made. Strontium was precipitated in its final form as the carbonate.

Rare earth elements. Nervik's method[27] of separating the rare earth elements was used in the bombardments from which rare earth elements were to be taken out as fission products. The rare earth elements were desorbed from the Dowex-50 cation-exchange resin (americium procedure) with 6 M hydrochloric acid. The hydroxide was precipitated with ammonium hydroxide and then was dissolved with a minimum of hydrochloric acid. The solution was diluted to about 15 cc with water and was then equilibrated with about 1 cc of Dowex-50 cation-exchange resin. This resin was placed on top of a glass column, 9 mm by 70 cm, packed with Dowex-50 cation-exchange resin. The eluant used was ammonium lactate of continuously varying pH (of from pH 3.2 to pH 5.0). After the rare earth elements were eluted they were each precipitated as the oxalate which was then ignited to the oxide.

D. Mounting of Samples

1. Actinide Elements.

Americium samples for the Pu^{240} bombardments were prepared by a volitilization of the samples from a tantalum filament onto 2-mil platinum plates 1 inch in diameter. The volitilization took place in an evacuated system and was done at a temperature of about 1800° C. This method produced thin samples but required considerable expenditure of time. Occasionally a thick coating of tantalum oxide was deposited on the plate, introducing thick-sample errors. A new method of plating tracer quantities of actinide elements has been developed and was used to plate the americium and neptunium samples for the Pu^{238} bombardments. In this method the activity is electroplated from a 4 M ammonium chloride solution which has been adjusted to the acid end-point of methyl red with

ammonium hydroxide and hydrochloric acid. A platinum plate acts as the cathode and a platinum wire is used as the anode. A current of about 2 amperes is used with the voltage adjusted to 6 to 8 volts. After plating for 5 to 10 minutes the reaction is quenched with ammonium hydroxide before the current is shut off. Samples obtained from this method were thinner than those obtained from volitilization. This was determined by visual observation and by the energy resolution of alpha particles. The nucleometer was used to follow neptunium and americium decay.

2. Fission Products.

Fission products were transferred in an ethanol or acetone slurry to weighed 2-mil aluminum hats and dried under a heat lamp. After being weighed, the samples were coated with a thin layer of zapon lacquer to prevent loss. A Geiger-Müller counter was used to follow the decay of fission products.

E. Counting Instruments

1. Alpha Counter.

An argon-filled ionization chamber attached to a scaling circuit was used for gross alpha counting of tracers and plutonium assays. This instrument has a counting efficiency of 52 percent for thin samples.

2. Alpha Pulse Analyzer.

The tracers on actinide sample plates were checked for purity on a 48-channel alpha pulse analyzer.[28] This instrument consists of electronic circuits which pick up amplified pulses produced by alpha particles in a methane-filled ionization chamber. These circuits sort the pulses according to size and record them as a function of energy on 48 separate registers.

3. Gamma Analyzer.

Analysis of gamma rays was performed on a 50-channel gamma-ray pulse analyzer. A thallium-activated sodium iodide scintillation crystal was used as a detector.

4. Geiger-Müller Counter.

All the fission products were beta-particle emitters and were

counted on a Geiger-Müller counter. The counting unit used was an end-window Amperex 100 C tube filled with a mixture of chlorine and argon. This was attached to a standard scaling unit. Background was reduced by housing the tube and sample holder in a thick-walled lead case. Samples could be placed on any one of five fixed positions below the counter tube. Whenever the activity of the sample permitted, Shelf 2 was used. Shelf 2 was about 2 cm from the window of the counter tube. The geometries of the two counters used were 3.09 percent and 4.23 percent. The dead-time value is 0.45 percent per 1000 counts per minute.

5. Nucleometer.

Isotopes decaying by electron capture were counted in a Nucleometer. This instrument contains a methane flow-type windowless proportional counter. The high counting efficiency of this instrument makes it especially favorable for counting electron-capture isotopes. Each isotope has its own plateau on this counter and determinations have been made[23] which show that 3900 volts is the operating plateau for the isotopes investigated in this study. A counting efficiency must be determined for each isotope.

III. TREATMENT OF DATA

A. Yield Determination

A known amount of alpha-emitting isotope was added to the target solution for each actinide that was to be taken out. Yield was determined from the amount of that particular isotope remaining in the isolated actinide. In the Pu^{240} bombardments, Am^{241} was produced by the beta-particle emission of the Pu^{241} in the target material. Because of the large alpha-background build-up with time, it was necessary to separate the americium and plutonium, just before electroplating the target, shortly before each bombardment. Since the proportion of Pu^{241} in the target material was well known, it was possible to determine the yield by noting the time of separation before electroplating and the time of separation of the americium and plutonium in the chemical procedure of the bombardment. Am^{243} was added to several bombardments to check this method of yield determination and in all cases the yield was the same.

For each of the fission products isolated a known amount of inactive carrier was added and the yield was determined by weighing the separated fission product.

B. Isotope Identification

Energy and half-life data were used to identify isotopes. Energy data was used solely in determining the amount, on each counting plate, of alpha-emitting isotope that was added for yield determination. Usually the beta-particle emitters and electron-capture isotopes were identified by their half-lives which were resolved from decay curves. This could not be done for Am^{237} ($t_{1/2}$ = 1.3 hours) and Am^{238} ($t_{1/2}$ = 1.86 hours). These isotopes were separated by the method of least squares, an analytical method of separating decay curve components.

C. Decay Curve Resolution

Approximately five counts were taken during each half-life period of the shortest-lived isotope in each sample. After a correction for counter dead time and background was made, the counts per minute were plotted versus time. In cases where several activities were contained in one sample, the curve was resolved by subtracting out the activities.

D. Calculation of Disintegrations per Minute

Counts per minute in an alpha ionization chamber were converted to disintegrations per minute by dividing the counts per minute by the geometry factor for platinum mounted samples, 0.52.

The disintegrations per minute of isotopes counted in the Nucleometer were determined by dividing the counts per minute of each isotope by its counting efficiency. One of the biggest problems with this instrument was this counting efficiency, particularly with the electron-capture isotopes. All the actinide element isotopes studied in this work were electron-capture in whole or in part. Each isotope has its own counting efficiency which appears to change with the method of preparing the counting plate. The counting efficiency values used in this work are listed in Table I. Work by Gibson[23] indicated that the counting efficiencies of electroplated samples approaches 100% so that a value of 90% was assumed for the electroplated americium samples.

Table I

Nucleometer Counting Efficiency Values

Isotope	Mode of Decay	Method of Sample Preparation	Counting Efficiency
Np^{236}	E.C. 43%, β^- 57%	Electroplated	0.65[a]
Am^{237}	E.C.	Electroplated	0.90[b]
Am^{238}	E.C.	Electroplated	0.90[b]
Am^{239}	E.C.	Electroplated	0.90[b]
Am^{239}	E.C.	Vaporized	0.60[c]
Am^{240}	E.C.	Vaporized	0.91[c]

a. Ref. 32

b. Assumed

c. Ref. 23

The conversion of counts per minute on the Geiger-Müller counter to disintegrations per minute was accomplished by use of the formula:

$$(d/m)_A = \frac{(c/m)_t}{g} \quad \frac{1}{S},$$

where $(d/m)_A$ is the disintegrations per minute of the isotope studied, $(c/m)_t$ is the total counts per minute in equilibrium, g is the geometry of the counter used, and S is determined by using the following formula:

$$S = F_1 S_1 + F_2 S_2 + F_3 S_3 + \ldots\ldots ,$$

where each F is the ratio of the abundance of the specific beta particle or conversion electron to the total abundance of beta particles of the parent nuclide and S_1 is determined from the formula:

$$S_1 = \frac{(BS)_1 \ (SSSA)_1}{(AW)_1 \ (C_{eff})} ,$$

where the correction factors are as set forth below.

1. AW - Air-Window Correction

Between Shelf 2 and the inside of the counting tube some of the beta particles were absorbed or scattered by the air or by the mica window of the tube. The air thickness was 2.4 mg/cm^2 and the window thickness was 3.5 mg/cm^2. The mass adsorption coefficient is proportional to Z/A

of the absorber[33] so that for the light elements, where Z/A is approximately constant, this quantity is nearly independent of the absorber. By extrapolating aluminum absorprion curves back 5.9 mg/cm^2 from the counting rate obtained with zero absorber, the air-window correction can be obtained. Ritsema[22] has done this for beta particles of various energies and the values for the air-window corrections were taken from that tabulation. In general the correction is less than 10%.

2. C_{eff} - Counting Efficiency Correction

It was assumed that all beta particles that entered the counting tube would count. (Therefore $C_{eff} = 1.0$).

3. BS - Backscattering Correction

Correction factors determined by Burtt[34] for beta particles scattered from a saturation aluminum backing were used. For beta particles of greater than 0.8 Mev of energy this factor is 1.28.

4. SSSA - Self-Scattering and Self-Absorption Correction

One of the biggest problems involved in converting counts per minute on the Geiger-Müller counter to disintegrations per minute was the evaluation of the SSSA correction factors. Since a separation determination must be made for each isotope of each sample thickness, much tedious work was necessary to interpolate the data of Stevenson and Nervik[35] in the cases where the SSSA effects were not directly determined by Hicks and Gilbert.[36] Stevenson and Nervik determined the variation of the SSSA factor with beta-particle energy and thickness of sample, using homogeneous mixtures of carrier-free beta-particle emitters and inactive salts. As described by Ritsema,[22] the interpolation was made on the basis of summed atomic numbers of the anion and cation of the sample.

It has been a project of the chemistry group at this laboratory to determine the correction factors which vary for each isotope, combine them into one factor (S), and plot this factor (S) as a function of sample thickness. Curves of this type have been made from the data of Stevenson and Nervik[35] for nearly all fission-product isotopes studied and Fig. 2 shows a comparison of this interpolated data with that of Hicks and Gilbert[36] for Ba^{140}. In the figure the upper curve is the actual function for the S factor.

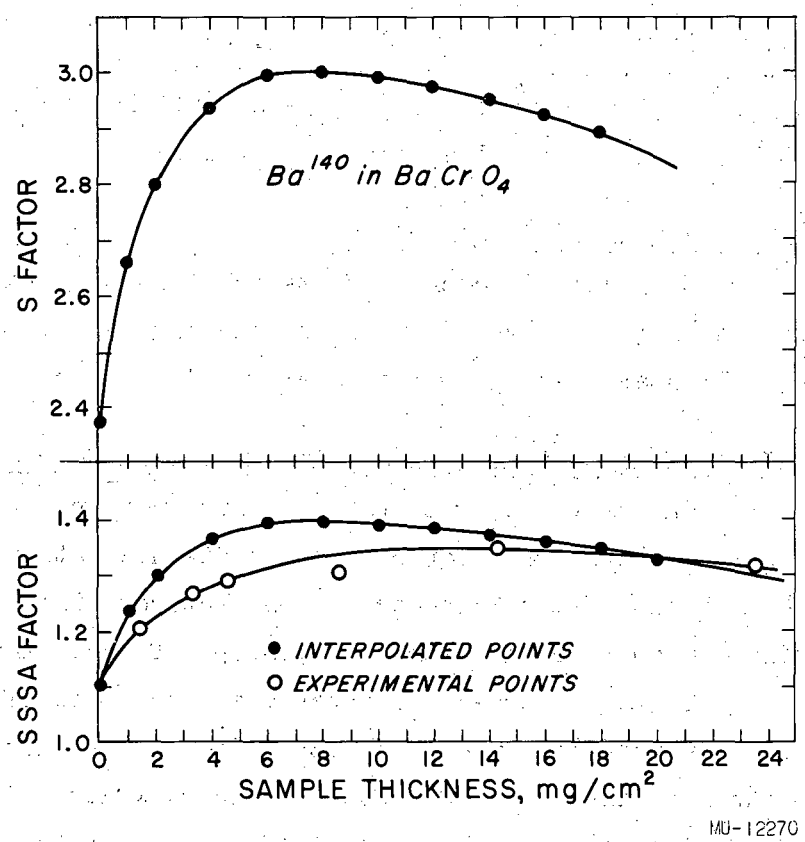

Fig. 2. SSSA and S factor corrections for Ba^{140}. The experimental data are those of Hicks and Gilbert.[36]

The work of Hicks and Gilbert contains only the SSSA corrections for Ba^{140}, whereas the S factor contains the SSSA, AW, and BS correction factors for Ba^{140} and its daughter La^{140}. With this curve it is only necessary to take the total counts per minute of Ba^{140} and La^{140} in equilibrium and divide by the S factor and the geometry of the counter to obtain disintegrations per minute. The two lower curves show a comparison of the experimental and interpolated values for the SSSA corrections of pure Ba^{140}.

5. g - Geometry Correction

The geometry factor is the ratio of the solid angle subtended by the window of the Geiger tube and the total solid angle around the sample. This factor was determined by using known standards and was found to be 3.09% and 4.23% for Shelf 2 of the two counters used.

E. Parent-Daughter Relationships

Corrections for daughter beta activities were made by using the formula:

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^{o} \left(e^{-\lambda_1 t} - e^{-\lambda_2 t} \right),$$

where N_2 is the number of daughter atoms present at time t, N_1^{o} is the number of atoms of parent nuclide produced in the bombardment, and λ_1 and λ_2 are the decay constants of the parent and daughter respectively.

F. Cross-Section Calculations

When the half-life of the isotope was long compared to the length of bombardment, the number of atoms produced was calculated by means of the formula:

$$N = d/m \; \frac{t_{1/2}}{0.693 \; (\text{chemical yield})}$$

where d/m is the number of disintegrations per minute at the end of bombardment and $t_{1/2}$ is the half-life of the isotope. In cases where the half-life of the isotope was short and appreciable decay took place during the bombardment, the formula used was:

$$N = \frac{(d/m) \times t_b}{(\text{chemical yield}) \times (1-e^{-\lambda t_b})}$$

where d/m is as before, t_b is the length of bombardment, and λ is the decay constant of the isotope.

Cross sections were then calculated from the formula:

$$\sigma = \frac{N}{n/cm^2 \ (It)}$$

where N is the number of atoms produced, n/cm^2 is the density of the target atoms, and It is the total number of deuterons striking the target.

IV. RESULTS

A. Spallation Cross Sections

The spallation cross sections of Pu^{240} are listed in Table II and those of Pu^{238} in Table III. Energy thresholds are listed below the reactions. These thresholds were calculated from the formula:

$$\text{Threshold energy} = -Q \frac{M_c}{M_r} ,$$

where M_c is the mass of the compound nucleus formed and M_r is the mass of the target nucleus. Q is the nuclear reaction energy and was determined from the formula:

$$Q = c^2 (\Sigma M_r - \Sigma M_p) ,$$

where ΣM_r is the sum of the reactant masses and ΣM_p is the sum of the product masses. The masses used in the threshold calculations were taken from the compilation of Hyde and Seaborg.[37] Fig. 3 shows the spallation excitation functions for Pu^{240} and Fig. 4 shows those for Pu^{238}. Both Table II and III and Figs. 3 and 4 have been corrected for the Pu^{239} content of the target material using Gibson's[23] data.

Table II

Pu^{240} + d Spallation Cross Sections (millibarns)[a]

Product Reaction Threshold (Mev)	Am^{240} d,2n 4.50	Am^{239} d,3n 10.34
Energy (Mev)[b]		
12.4	17.2	2.9
13.7	46.8	7.8
14.0	35.2	9.4
15.5	24.3	10.9
17.3	19.8	26.5[c]
19.4	17.4	19.9
21.2	18.2	
22.1	16.8	18.9
23.3	16.0	12.9

a. ± 10 -- 20%

b. ± 0.5 Mev

c. Upper limit

Pu^{238} + d Spallation Cross Sections (millibarns)[a]

Table III

Product Reaction Threshold (Mev)	Am^{239} d,n -1.77	Am^{238} d,2n 5.23	Am^{237} d,3n 11.31	Np^{236} d,α -4.03
Energy (Mev)[b]				
9.1	1.5	1.1		
12.4	7.5	9.5	3.6	
14.2	12.7	11.1		
15.9	13.9	8.9	7.6	
18.0	13.9	10.2	6.6	
20.1	13.8	8.8	9.0	5.5
22.2	13.3	4.6	11.3	
23.3	15.1	6.1	8.7	

a. ± 10 - 30%

b. ± 0.5 Mev

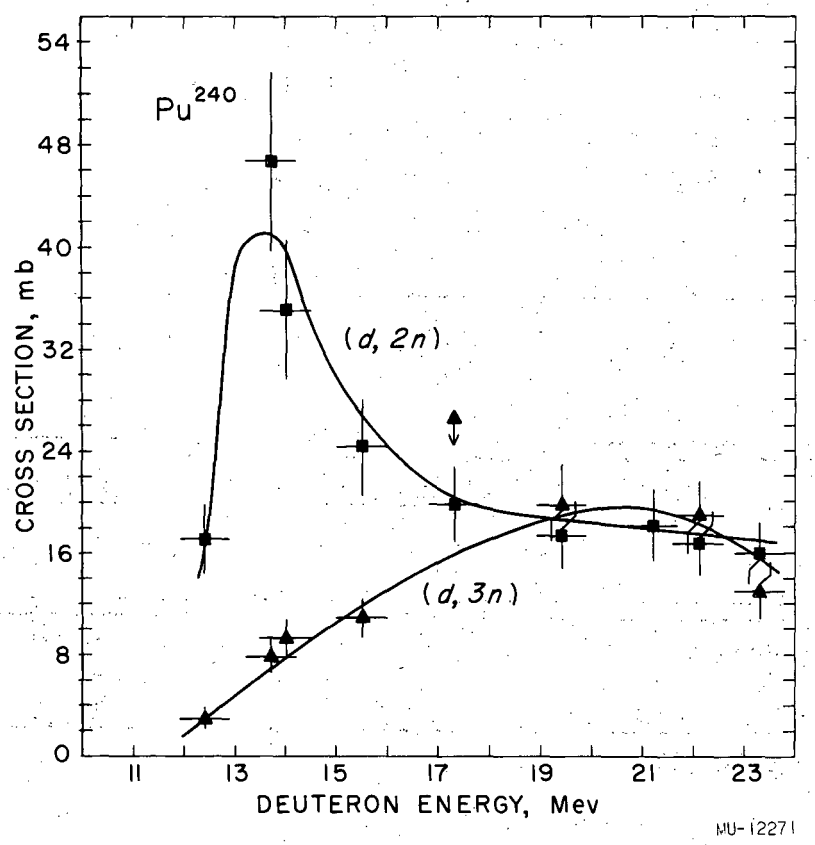

Fig. 3. Excitation curves for the (d, 2n) and (d, 3n) spallation products formed in bombardments of Pu^{240} with deuterons.

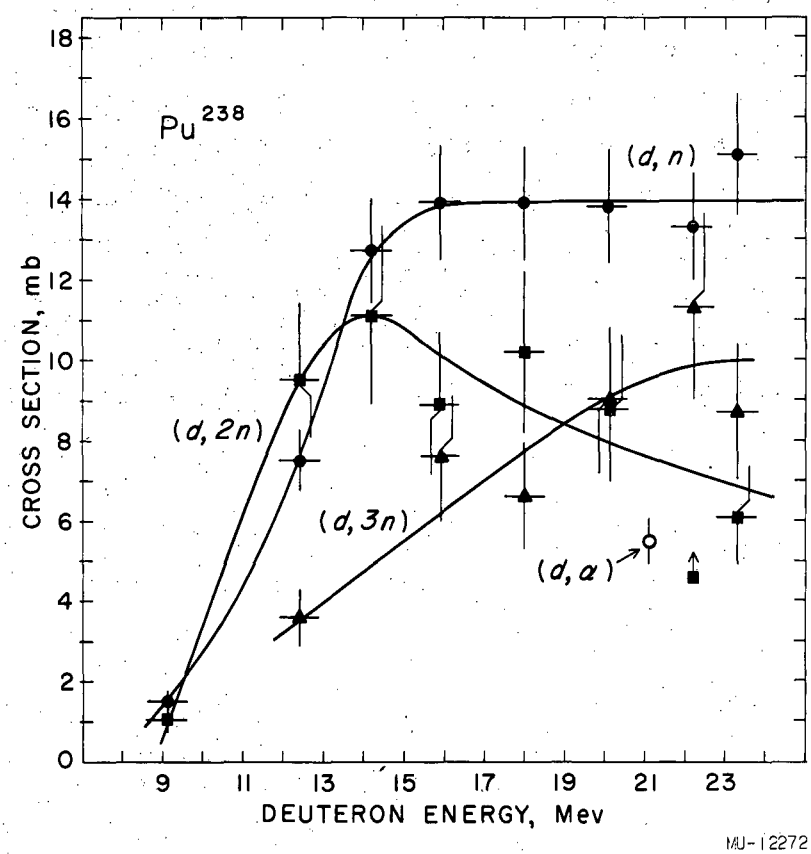

Fig. 4. Excitation curves for some spallation products formed in bombardments of Pu^{238} with deuterons.

B. Fission Product Cross Sections

It has been previously assumed that the fission product cross sections determined represented the total yield of the mass chain.[20-22] Gibson[23] has found that this is not valid at higher energies and has determined the factors necessary to correct for any losses through primary fission to products in the mass chain beyond the product measured. Fission product cross sections for Pu^{240} are listed in Table IV. Gibson's data for mass chain yield corrections were used and both the uncorrected and corrected cross sections are tabulated. Fig. 5 presents the curves of corrected fission product yield versus mass and a comparison of the curves at various energies is given in Fig. 6. The total fission, spallation, and theoretical total reaction cross section excitation functions of Pu^{240} are compared in Fig. 7. The fission cross sections are not corrected for the Pu^{239} content of the target material.

Table IV

Pu^{240} + d; Fission Product Cross Sections (millibarns)[a]

Deuteron Energy (Mev)[b]	10-11[e]		12.4		13.7		15.4		19.4		21.2		23.3	
Isotope	C	D	C	D	C	D	C	D	C	D	C	D	C	D
Sr^{89}	1.95	1.95	4.70	4.70	7.18	7.18	10.7	10.7	10.5	10.5	14.2	14.2		
Sr^{91}	3.01	3.01	8.20	8.20	12.5	12.5	15.3	15.3	19.3	19.3	20.6	21.0		
Pd^{109}			9.00	9.00										
Pd^{112}			6.21	6.41										
$Cd^{115m}+Cd^{115}$	2.78	2.78	7.20	7.20	11.0	11.0	29.4	29.4	38.9	38.9	38.2	38.2	53.9	53.9
Cd^{117}	2.45	2.51	5.61	5.80	10.7	11.0	28.9	30.1	35.6	36.3	43.0	45.2	51.8	55.0
Ba^{139}	7.9	8.5	19.6	20.9	22.1	23.6	44.1	49.4	37.6	42.2	47.3	55.3	53.9	65.7
Ba^{140}	5.1	6.0	12.7	14.9	17.3	20.3	26.6	32.5	30.6	37.4	30.1	37.7	34.1	47.1
Ce^{141}									44.7	44.7				
Ce^{143}									38.3	40.6				
Ce^{144}									29.6	33.2				
Nd^{147}					11.3	11.5			23.0	23.9				
Eu^{156}					1.71	1.81			4.58	5.04				
Eu^{157}					1.44	1.60			4.85	5.68				
Tb^{161}									0.79	0.85				
Total fission cross section	157		367		526		995		1172		1300			

a. ± 20 - 30%
b. ± 0.5 Mev
c. Uncorrected cross section for the isotope
d. Cross section corrected for mass chain yield
e. Degradation foil bulged making energy uncertain

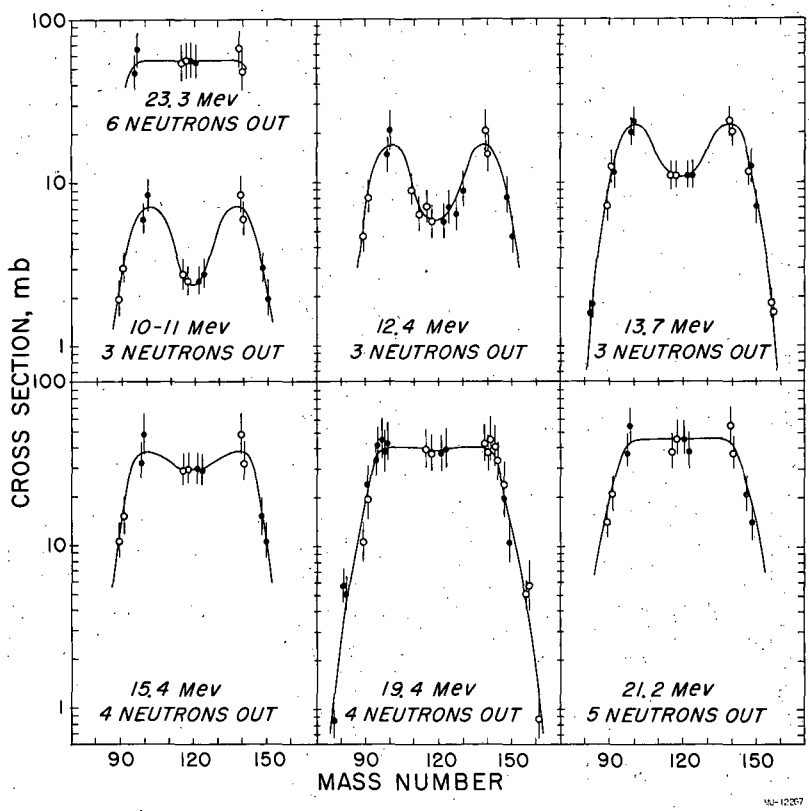

Fig. 5. Fission yield versus mass curves for Pu^{240} bombarded with deuterons. Open circles are experimental points and solid circles are reflected points.

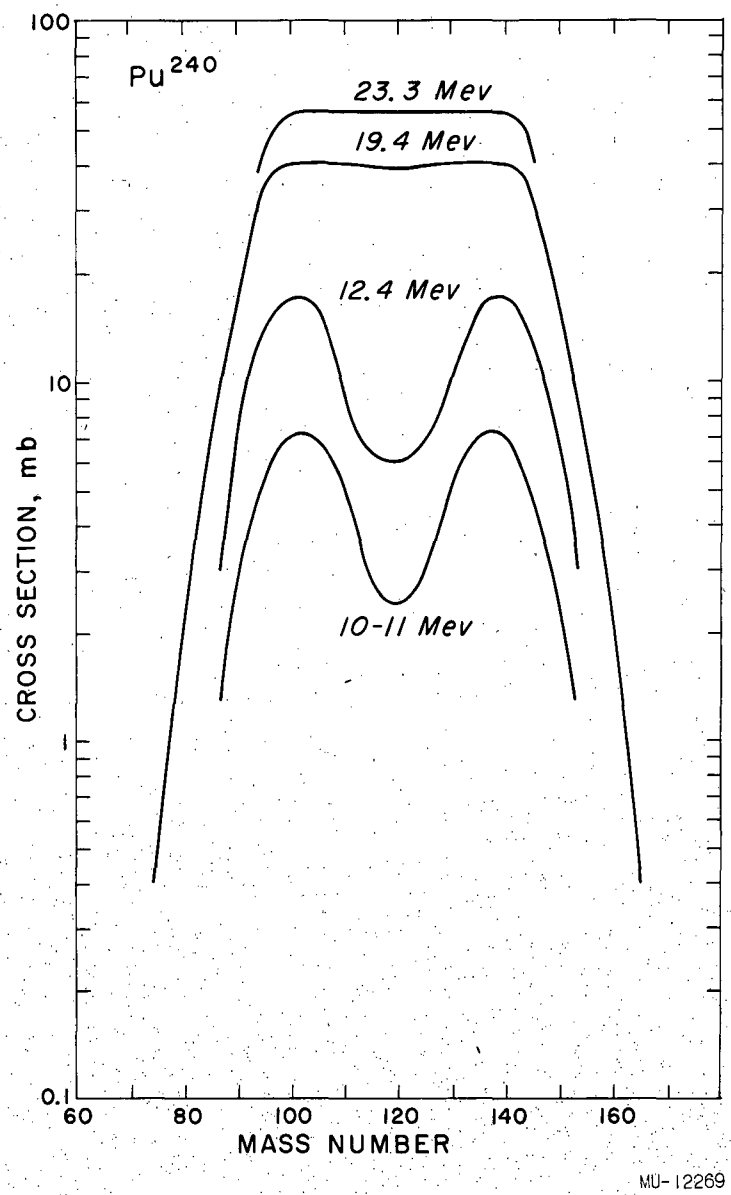

Fig. 6. Variation of fission yield versus mass curves with energy.

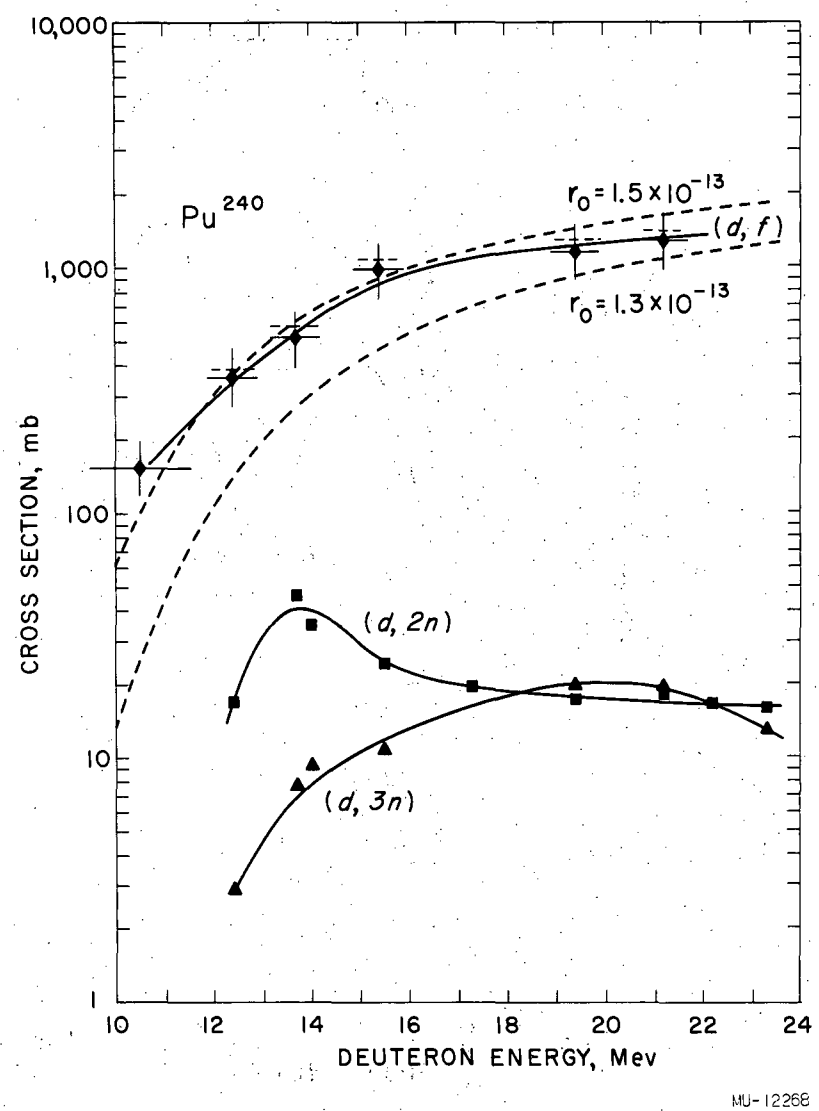

Fig. 7. Comparison of the total fission, spallation, and theoretical total reaction cross section excitation functions for Pu^{240}.

V. DISCUSSION

A. Spallation Reactions

The (d,n) reaction excitation function for Pu^{238} is similar to that found by others[23, 38-40] in that it rises to a maximum value slightly above the barrier and is flat from there out to the highest bombardment energies. The main difference in the (d,n) reaction excitation functions for isotopes of elements in various parts of the periodic table is the maximum cross sections. As the Z of the bombarded nuclide increases, the maximum decreases. For example, the (d,n) reaction excitation function for I^{127} reaches 180 mb,[40] for Bi^{209}, 30 mb,[39] and for uranium and plutonium, about 13 mb.[23,38]

Peaslee[41] has calculated theoretically the (d,n) reaction excitation function for Bi^{209} and obtained fairly good agreement with the experimental data of Kelly and Segre.[39] The calculated (d,n) curve did not become entirely flat but continued to rise at a relatively slow rate. In this calculation, Peaslee considered that the (d,n) reaction was entirely stripping. Glass[42] has extended Peaslee's calculations to the heavy element region and found that the curve rises to about 20 mb for plutonium and uranium, which is in rough agreement with the experimental data.[23,38] A comparison of the (d,2n) and (d,3n) reaction excitation functions for Pu^{240}, Pu^{239} (Gibson[23]), and Pu^{238} shows a marked decrease in going from Pu^{240} to Pu^{238} whereas a comparison of the (d,n) excitation function of Pu^{239} and Pu^{238} shows no differences within the limits of experimental error. This is further evidence that the (d,n) reaction is mainly stripping and is not affected by the proposed[43] increasing fissionability of the compound and intermediate nuclei with decreasing A. It was not possible to determine the (d,n) reaction cross sections for Pu^{240} because of the growth of (d,n) product, Am^{241}, from the Pu^{241} in the target material.

A surprising result is the marked decrease of the (d,2n) reaction of Pu^{238}. The peak value for the (d,2n) reaction is only 11 mb compared to 28 mb for Pu^{239} and 41 mb for Pu^{240}. Even if a value of 60% is taken for the counting efficiency of electroplated Am^{238} instead of the 90% assumed, the cross section reaches only 16.5 mb for the (d,2n)

reaction of Pu^{238}. More will be said about this in relation to the fission cross sections.

In Pu^{240} bombardments a new value of 51.0 ± 1.0 hours for the half-life of Am^{240} was determined. The Am^{240} was in high abundance in all bombardments and was the longest lived isotope in the samples. The determinations took place over 3 - 5 half-lives in each bombardment and in every case the half-life found was 50 to 52 hours.

Difficulty was encountered in separating the Am^{237} and Am^{238} activities in the Pu^{238} bombardments. Higgins[44] has reported a half-life of ~1.3 hours for Am^{237} obtained by alpha pulse analysis and Carr[21] has reported a half-life of 1.86 ± 0.09 hours for Am^{238} obtained by following the decay of prominent gamma rays. Attempts were made in this study to find a prominent gamma ray in Am^{237} but were without success. As was observed by Higgins, the decay curve for the americium activities had a half-life of ~1.5 hours after subtracting out the Am^{240} (from the (d,n) reaction of the Pu^{239} in the target material) and the Am^{239} components. In the bombardment at 9.1 Mev, which is below the threshold for the (d,3n) reaction, this residual activity had a half-life of 1.81 hours which is in good agreement with Carr's value of 1.86 ± 0.09 hours. An analytical method of determining the half-life of Am^{237} was attempted but proved to be too insensitive. The method of least squares was used in separating the Am^{237} and Am^{238} activities assuming that the half-life of Am^{237} was 1.3 hours. The activities were also separated by this same method assuming a half-life of 50 minutes for the Am^{237}. In this latter case, the cross sections for the (d,3n) reaction were highly scattered so it was assumed that the 1.3 hour value was more nearly correct. The values obtained for the (d,2n) cross sections did not vary appreciably from each other using either 50 minutes or 1.3 hours for the half-life of Am^{237}. The cross sections for the (d,3n) reaction of Pu^{238} are thus dependent not only upon an assumed counting efficiency but also upon the half-life of Am^{237}, which needs to be better determined.

An attempt was made to define an excitation function for the (d,α) reaction of Pu^{238}. Owing to the gross alpha activity of Pu^{238} present and the relatively ineffective chemical procedures available for the separation of plutonium and neptunium, only one experiment was

successful. A cross section of 5.5 mb was determined for the (d,α) reaction at 20.1 Mev.

All (d,xn) cross sections for both Pu^{238} and Pu^{240} were corrected for Pu^{239} content using data of Gibson[23] for the Pu^{239} cross sections.

B. Fission Yields

The fission yield curves are shown in Fig. 5 and Fig. 6. The usual minimum in the curve is observed at low energies and disappears at about 19 Mev. Within the limits of experimental error, at higher energies the curves could be drawn with a slight valley or a slight peak. Work by Gibson[23] on Pu^{239} in which more points on the top of the curve were determined indicates that the curve is flat.

The fission yield curves and total fission for Pu^{240} are the same as those for Pu^{239} within the limits of experimental error. A more sensitive test for the comparison of the fissionability of Pu^{238}, Pu^{239}, and Pu^{240} is in the spallation excitation functions. The (d,2n) reactions are probably compound nucleus type reactions to a large extent which are in direct competition with fission. It is then reasonable that a decrease in the (d,2n) reaction (for example) indicated a relative increase in the fission level widths of compound and intermediate nuclei. By this reasoning fission increases from Pu^{240} to Pu^{238}. This appears at first to be a Z^2/A effect[15] but is in reality more of an A effect since the values for the (d,2n) reactions of uranium isotopes[23,38] are about the same as for the plutonium isotopes. If fission decreased by a Z^2/A effect in going from plutonium isotopes to uranium isotopes then a large increase in the (d,2n) reaction would be observed.

The theoretical total reaction cross section excitation function[45] for Pu^{240} is compared with the fission and spallation curves in Fig. 7. The fission cross section itself is as great as the theoretical curve for an r_o value of 1.5×10^{-13} cm. The addition of all possible spallation reactions would probably raise the experimental total cross section slightly above the theoretical curve indicating that the nuclear radius is larger than that calculated with an r_o value of 1.5×10^{-13} cm, assuming theoretical treatment is inadequate.

Fission cross sections were corrected for mass chain yield by a method reported by Gibson.[23] No attempt was made to correct the fission cross sections for Pu^{239} content since the fission yield curves for Pu^{240} and Pu^{239} are essentially the same.

ACKNOWLEDGMENTS

I wish to thank Professor Glenn T. Seaborg for his interest and guidance. I wish to thank Bruce M. Foreman, Jr., Walter M. Gibson, T. Darrah Thomas, Mrs. Susanne R. Vandenbosch, Robert Vandenbosch, and Dr. William Wade for technical assistance and helpful discussions. The assistance of Mrs. Therese K. Pionteki in counting the samples is appreciated, as is the aid of the Health Chemistry Group and particularly that of Roberta Delk and Marshall Lombardo. The cooperation of Professor Joseph G. Hamilton, the late G. Bernard Rossi, W. Bart Jones, and the crew of the 60-inch cyclotron is gratefully acknowledged. Finally, I wish to thank Dr. Richard A. Glass for his aid, advice, and encouragement.

This work was performed under the auspices of the U. S. Atomic Energy Commission.

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