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SPALLATION-FISSION COMPETITION IN THE NUCLEAR REACTIONS OF PLUTONIUM INDUCED BY ALPHA PARTICLES

Robert Joseph Carr
(Thesis)

April 15, 1956

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Robert Joseph Carr

Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California

April 15, 1956

ABSTRACT

Excitation functions from 25 to 47 Mev have been measured for the (α, n), (α, 2n), (α, 4n), (α, pn) and (α, p2n) nuclear reactions of Pu^{238} and the (α, 2n) and (α, 4n) nuclear reactions of Pu^{242}. Helium-ion-induced fission cross sections and yield curves have also been determined in this energy range. The results are qualitatively discussed in terms of compound-nucleus and direct-interaction models, and it is concluded that they are consistent with the assumption that fission predominates in compound nuclei of low excitation while nucleon emission predominates at higher excitation energies, with direct interaction processes--probably "knock-on" processes--occurring in the periphery of the nucleus, contributing appreciably to the total cross section.

The procedures involved in preparing macroscopic amounts of Pu^{238} by pile irradiation of Np^{237} are discussed in detail.

A new lower limit for the partial negatron decay half life of Am^{240} and new information regarding the electron-capture decay of Am^{238}, including a probable decay scheme, are appended.

I. INTRODUCTION

Glass, Cobble, and Seaborg[1,2] have recently made an extensive study of the spallation and fission reactions induced by helium-ion bombardment of Pu^{239} at energies from the reaction thresholds up to 47 Mev. This study yielded some unexpected results, notably a large (α, 2n) cross section, an (α, p2n) excitation function which at its peak greatly exceeds the highest (α, 3n) cross section, and an (α, n) cross section which changes very little throughout the range of bombardment energies.

It is desirable to obtain as much data as is possible for other comparable reactions and target materials so as to aid in the interpretation of the mechanism of these reactions. Without such information one cannot say, for example, if the relative prominence of the various reactions is determined by the statistical weights of the intermediate and product nuclei, as would be expected from any sort of compound-nucleus-nuclear evaporation model, or if they depend on the forms of the reactions *per se*.

To gain further insight into such questions, it was decided to study the fission and spallation reactions of other plutonium isotopes under the bombardment conditions used by Glass and his co-workers.

The results reported in this paper are the excitation functions up to 47 Mev for the (α, n), (α, 2n), (α, 4n), (α, pn), (α, p2n), and alpha-induced fission reactions of Pu^{238} and the (α, 2n) and (α, 4n) reactions of Pu^{242}. Comparison of the Pu^{238} and Pu^{239} excitation functions allows one to note the effect of the even-odd structure of the target nucleus, while the effect of mass separate from even-odd consideration is shown by comparison of the Pu^{238} and Pu^{242} results.

II. STARTING MATERIALS

A. Plutonium-238

Irradiation Procedures

The Pu^{238} used in these studies was prepared by slow-neutron irradiation of 150 mg Np^{237} in the Materials Testing Reactor, Phillips Petroleum Co., Arco, Idaho, which, under our bombardment conditions, has a flux originally estimated at 4×10^{14} neutrons/cm^2 sec.[3] In so high a flux, secondary reactions leading to plutonium isotopes of mass greater than 238 are important, and it was therefore necessary to choose an irradiation period short enough that corrections to the eventual Pu^{238} reaction cross sections due to these isotopes in the cyclotron target material would be small, yet long enough that several milligrams of plutonium would be produced. To this end, calculations of the amounts of various isotopes of neptunium and plutonium produced as a function of irradiation times were made on GADAC, a growth and decay analog computer described elsewhere,[4] for assumed fluxes of 2.5, 3, and 4×10^{14} neutrons/cm^2 sec. The cross sections and half lives assumed for the calculation are given on Table I--reactions not shown are inappreciable--and the results are given on Fig. 1. On the basis of these calculations, an irradiation time of 21 days was chosen.

The isotopic composition of the plutonium actually formed was: Pu^{238}, 93.79 ± 0.1%; Pu^{239}, 5.75 ± 0.1%, Pu^{240}, 0.46 ± 0.04%; Pu^{241}, less than 0.05%. This corresponds to fluxes of 1.7×10^{14} neutrons/cm^2 sec and 2.8×10^{14} neutrons/cm^2 sec on the basis of the calculated yields of Pu^{238} relative to Pu^{239} and Pu^{240} respectively (allowing, of course, for total decay of Np^{238} and Np^{239} to their plutonium daughters). The discrepancy between these two numbers is probably due to the uncertainty in the assumed cross sections.

The usual method for preparing 100-milligram samples of plutonium for bombardment in high-flux reactors is to mix powdered PuO_2 with aluminum dust and to press this mixture into an aluminum capsule. Such a target provides good heat conduction away from the fissioning plutonium. The heat produced in irradiating neptunium, however, is much less than for plutonium, and so it was possible to

Table I

Reactions included in GADAC calculation

Reaction	Cross section (barns)	Half life (days)
$Np^{237}(n, \gamma)Np^{238}$	170	
$Np^{238} \xrightarrow{\beta^-} Pu^{238}$		2.10
$Np^{238}(n, \gamma)Np^{239}$	600	
Np^{238} fission	1700	
$Np^{239} \xrightarrow{\beta^-} Pu^{239}$		2.33
$Pu^{238}(n, \gamma)Pu^{239}$	400	
$Pu^{239}(n, \gamma)Pu^{240}$	330	
Pu^{239} fission	730	
$Pu^{240}(n, \gamma)Pu^{241}$	350	
Pu^{240} fission	10	

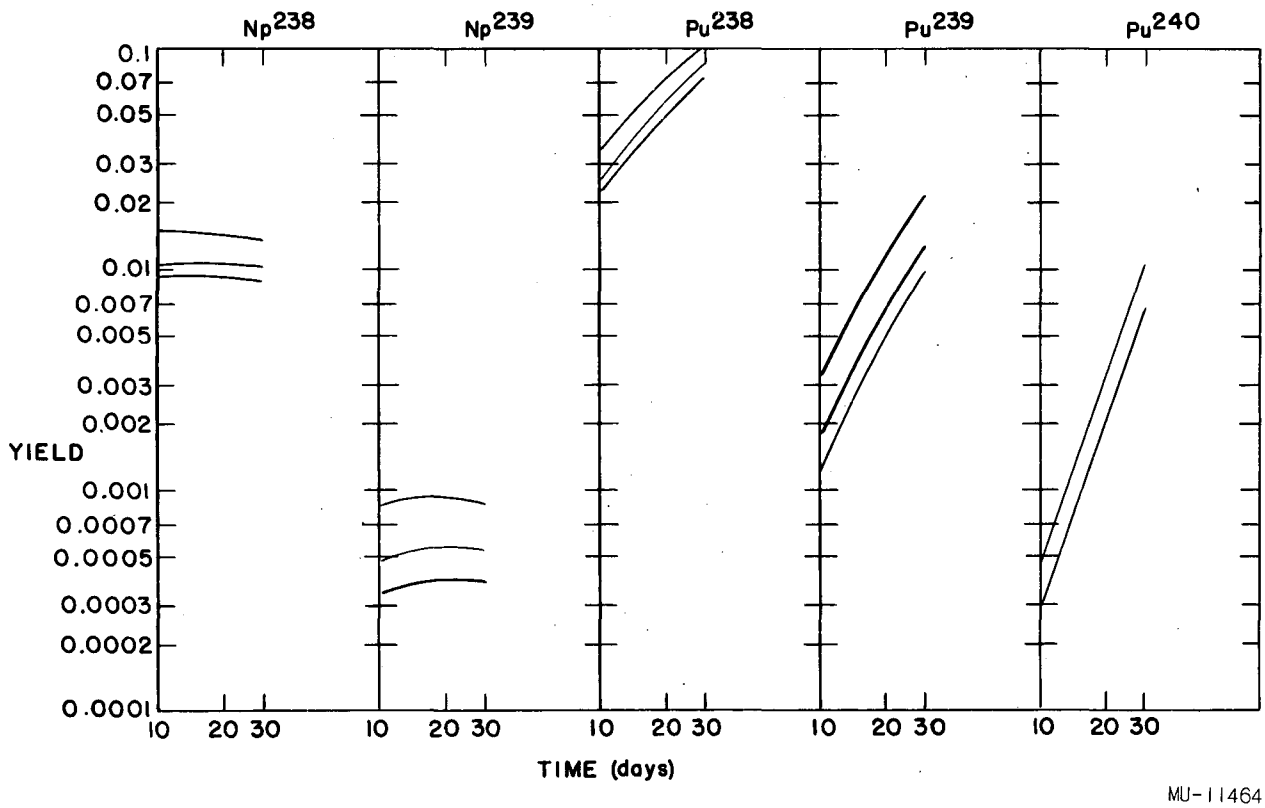

Fig. 1. Production of neptunium and plutonium, isotopes by pile irradiation of Np^{237}. The curves from bottom to top are for assumed fluxes of 2.5-, 3-, and 4×10^{14} neutrons/cm^2 sec respectively. Yields are in atoms per atom of Np^{237}.

make the target by sealing NpO_2 in two quartz glass capsules, which were then sealed into an aluminum capsule. The complications introduced into the chemical separation procedure by the presence of large amounts of aluminum were thus avoided. The lateral clearance between the quartz glass and aluminum capsules was held to 1 mil to assure good heat transfer. The outside of the aluminum capsule was water-cooled during the irradiation.

The heat calculation to justify this procedure was as follows: The greatest portion of the heat comes from fission of Np^{238} and Pu^{239}, and since the concentration of the former is approximately constant and that of the latter monotonically increasing during the irradiation period, the maximum heat production is occurring at the end of the irradiation. The rates of energy release of the various reactions at this time are tabulated in Table II; the total power is 1.85×10^9 ergs/sec.

In order to set an extreme upper limit on the temperature reached by the sample, it was assumed that the entire energy dissipation was by radiation[5] according to the Stefan-Boltzmann law, with the emissivity taken as unity and the radiating area taken as 10 times that of two 75-mg spheres of density 11 g/cm^3, the crystal density of PuO_2.[6] As the NpO_2 was granular, with the grains less than a millimeter in diameter, this assumed area is an immense under-estimation. The equilibrium temperature calculated from these assumptions is 1500° C, which is below the softening point of quartz glass.

Radiation Precautions

The radioactive hazard from the chemical operations in the preparation of the Pu^{238}, as well as in the procedure following the cyclotron bombardments, was extremely great, and therefore elaborate safety precautions were necessary.

The maximum amount of plutonium that can safely be handled directly is that decaying by alpha-particle emission at the rate of about 10^6 disintegrations/min, which, for Pu^{238}, amounts to about 10 millimicrograms. It was therefore necessary that all procedures involving plutonium be carried out in vented boxes outfitted with gloves and (or) tongs.[7]

Table II

Rate of energy release immediately before end of pile irradiation
(Flux = 4×10^{14} neutrons/cm^2 sec, 150 mg Np^{237} initially present)

Reaction	Energy release per event (Mev)	Events per second per mg reactant	Mg of reactant present	Energy release per second in total sample (Mev/sec)
$Np^{237}(n, \gamma)$	5.47	1.1×10^{11}	130	7.8×10^{13}
$Np^{238}(n, \gamma)$	6.2	6.1×10^{11}	2.1	7.9×10^{12}
Np^{238} fission	200	1.72×10^{12}	2.1	7.3×10^{14}
$Np^{238}\beta$-decay	1.3	9.67×10^{12}	2.1	2.6×10^{13}
$Np^{239}\beta$-decay	0.7	8.67×10^{12}	0.14	8.5×10^{11}
$Pu^{238}(n, \gamma)$	5.7	4.1×10^{11}	11.6	2.7×10^{13}
$Pu^{239}(n, \gamma)$	6.4	3.3×10^{11}	1.95	4.1×10^{12}
Pu^{239}fission	200	7.3×10^{11}	1.95	2.8×10^{14}
$Pu^{240}(n, \gamma)$	5.5	3.5×10^{11}	0.15	2.9×10^{11}
Pu^{240} fission	200	1.0×10^{10}	0.15	3.0×10^{11}
Total				1.15×10^{15} Mev/sec = 1.85×10^{9} ergs/sec

In addition to the large alpha radiation, the pile-irradiated sample involved an extremely high level of beta-gamma activity arising from fission-product decay. The amount of shielding necessary to reduce this to a tolerable level was calculated as follows: It was assumed that fission products were formed at a rate corresponding to a flux of 4×10^{14} neutrons/cm^2 sec irradiating Np^{238} and Pu^{239} in the amounts shown by the GADAC calculations for this flux for a 21-day period. It was further assumed that the fission products decay according to the law found to hold for the products of slow-neutron fission of U^{235},

$$A = 1.34 \times 10^{-4}\, t^{-1.03}, \quad 4 \leq t \leq 100 \text{ days},$$

where A is the fission product activity in disintegrations per minute per fission event, and t is the time elapsed since the fission occurred, expressed in days.[8] Finally, it was assumed that the average fission event is accompanied by one gamma ray of 1 Mev energy. From these assumptions, and with the help of the nomograms given by Balderston et al.,[9] the expected radiation fields for various conditions of geometry and shielding were calculated. It was found that 2 in. of lead shielding would reduce the radiation to a safe level if a 30-day "cooling period" were allowed before purification procedures were begun.

In order to evaluate this calculation method, these calculations have been remade assuming the more realistic flux of 2.3×10^{14} neutrons/cm^2 sec. By extrapolation of the GADAC calculations as a function of flux for various times, it was found that for the assumed flux the amount of Np^{238} is approximately constant at 1.27 mg (for 150 mg Np^{237} at the start of the irradiation) throughout the bombardment, except for an induction period which may be accounted for by taking the amount of Np^{238} to be zero for 2 days (one Np^{238} half life) and the constant value of 1.27 mg for the remainder of the bombardment. The amount of Pu^{239} in milligrams is given by $1.58 \times 10^{-3} t^2$, where t is the time in days since the beginning of the irradiation.

From these functions and the previously given law relating fission product decay to time elapsed since fission, one may derive the following expressions for fission product activity:

$$A_{238} = \sigma_{238} fn \cdot 1.27 \cdot 1.34 \cdot 10^{-4} \int_{2}^{t_1} (t_2 - t)^{-1.03} dt,$$

$$A_{239} = \sigma_{239} fn \cdot 1.58 \cdot 10^{-3} \cdot 1.34 \cdot 10^{-4} \int_{0}^{t_1} t^2 (t_2 - t)^{-1.03} dt.$$

Here σ_{238} and σ_{239} are the fission cross sections for Np^{238} and Pu^{239} in cm^2, n is the number of target atoms per milligram, f is the flux in neutrons/cm^2 day, and t_1 and t_2 are the times in days from the beginning of the irradiation to the end of the irradiation and the time of the fission product measurement, respectively. A_{238} and A_{239} are the fission product activities at t_2 in disintegrations per minute. Taking t_1 and t_2 as 21 and 51 days respectively, one finds that the fission product activities are 3.1 and 0.7 curies from the Np^{238} and Pu^{237} fission respectively.

Radiation fields expected from the total 3.8 curies of activity were calculated for a number of conditions of shielding and geometry for various assumed effective gamma ray energies. Comparison of these calculations with the corresponding measured values indicates that the best effective gamma-ray energies are 0.6, 0.8, and 1.0 Mev for 1, 2, and 4 in. of lead shielding respectively, if one gamma ray is assumed to accompany each decay event.

Chemical Procedure

The chemical procedure, designed for maximum recovery of the valuable untransmuted neptunium as well as separation of the plutonium, was carried out in lead-shielded vented boxes working by remote control.

After the 30-day cooling period, which allows not only for decay of much of the fission product activity but also for decay of Np^{238} and Np^{239} to plutonium, the irradiated aluminum capsule was put into a "slug opening box," where its ends were drilled out and the quartz glass capsules removed. These were scratched with a file, broken

open, and the contents washed into a 40-ml Teflon cone with a small amount of 6 M HNO_3. The cones were transferred to the "chemistry box," where the following procedure was carried out:[10]

1. The NpO_2 was dissolved by heating with successive portions 6 M HNO_3 - 0.05 M HF solution for several hours each.

2. When dissolution was complete, the solutions were combined, evaporated to about 20 ml, and saturated with NH_3 gas. Centrifuging gave a black precipitate and a green solution. The supernatant was decanted and the precipitate was washed with 0.5 M NH_4OH. The wash solutions were combined with the original supernatant. This solution, on standing overnight, gave a green precipitate of Np(V) hydroxide and a colorless supernatant solution. When the corresponding precipitate occurred in some later recovery procedures, it was found to be sufficiently free of fission products to allow it to be removed to an unshielded gloved box without further treatment. In the original separation, however, the precipitate was washed and combined with the first hydroxide precipitate.

3. The combined precipitate was dissolved in 12 M HCl. The solution was diluted to 5 M HCl, made 0.5 M in HI and 0.005 M in Fe^{++}, and warmed for about 1 hr. This reduced the neptunium to Np(IV) and the plutonium to Pu(III).

4. The solution was then made 3 M in HCl, 6 M in HF, and 1 M in KF, giving a precipitate of KNp_2F_9 and PuF_3, which carries the rare earths and, to a small extent, other fission products. The suspension was allowed to stand overnight, then cooled in an ice bath for about an hour and centrifuged. The supernatant was removed and the precipitate was washed twice with 1 M HCl-1 M HF, first warming and then cooling in an ice bath before each centrifuging. The precipitate was then dissolved in a minimum volume of Zr(IV)-HCl solution.

5. The resultant solution was again made 6 M in HF and 1 M in KF, and the above procedure of precipitating and washing was repeated. This time the final precipitate was dissolved with a saturated solution of H_3BO_3 in 6 M HNO_3. In this process, the

plutonium is oxidized by the nitrate to Pu(IV). Oxidation of the neptunium to Np(V) and Np(VI) also occurs, but this is slow and the bulk of the neptunium apparently remains unoxidized.

6. The solution was again saturated with NH_3, and the resultant precipitate was again washed as in Step 2. This time, no green solution or delayed precipitate appeared. At this point, good decontamination from all fission products except the rare earths and possibly the alkaline earths had been obtained.

7. The precipitate was dissolved in 12 M HCl and the solution was slowly forced into an anion-exchange resin column which had previously been washed with 10 M HCl. The resin used was Dowex A-2. This resin, which has a total capacity of about 2 milliequivalents per gram, absorbs neptunium and plutonium in all oxidation states higher than the trivalent in 10 M HCl and desorbs them in 2 M HCl. It does not absorb rare earths, trivalent actinides, or alkaline earths in any concentration of HCl. The column used was 0.7 cm in diameter and 13 cm long.

When the loading was completed the column was washed with 10 M HCl-0.01 M HNO_3 solution at a rate of about 20 ml/hr until all of the rare earth and alkaline earth activity had been washed through. The eluting agent was then changed to 10 M HCl-0.1 M HI-0.005 M $N_2H_4 \cdot 2HCl$ solution. This solution reduces the plutonium to Pu(III), which immediately washes through the column. The neptunium is slowly reduced to Np(IV), but is not desorbed. After about 150 ml of the solution was run through the column, the eluting agent was changed to 2 M HCl, stripping off the neptunium, as well as any oxidized plutonium that remained on the column. The neptunium and plutonium fractions were removed to an unshieded glove box for further purification.

It was found by radiochemical assay that about one-fourth of the total plutonium had not been reduced, but had remained with the neptunium. Therefore, in all subsequent purifications, reduction on the column was not attempted. Instead, the oxidized neptunium and plutonium were stripped from the anion column with 2 M HCl after the rare earths had been washed through with 10 M HCl. This

neptunium-plutonium fraction was then evaporated to a convenient volume of 6 M HCl and reduced as in Step 3 above. The reduced solution was made 10 M in HCl with HCl gas, and then rerun on the anion column, eluting the plutonium with the HCl-HI-$N_2H_4 \cdot 2HCl$ solution and the neptunium with 2 M HCl as before. This procedure gives 99.9% decontamination of the plutonium and neptunium from each other.

8. The neptunium and plutonium were further purified in the glove box by repetitions of the above procedures. All hydroxide and fluoride supernatants were assayed and alpha-pulse analyzed, and were found to contain only negligible amounts of neptunium and plutonium.

In the course of the procedure in the lead box, small amounts of solids had been left behind because of difficulty of solution or poor transfer. The above procedure was repeated on these after several milligrams of La^{+3} had been added to increase the bulk of the precipitates involved.

B. Plutonium-242-Containing Mixture

The plutonium-242 cross sections were determined by bombardment of a plutonium sample of the following isotopic composition: Pu^{238}, 58.6%; Pu^{239}, 3.4%; Pu^{240}, 0.2%; Pu^{241}, 0.02%; Pu^{242}, 37.8%.

This mixture, a milligram of which was kindly supplied by Dr. E. Kenneth Hulet of the Livermore Site of the Radiation Laboratory, was synthesized by a 7-month irradiation of Am^{241} in the Materials Testing Reactor, Phillips Petroleum Co., Arco, Idaho.

III. EXPERIMENTAL PROCEDURES

Target Preparation and Bombardment Procedures

In order to make absolute measurements of the cross sections of cyclotron-induced reactions, it is necessary either that the entire target be exposed to the cyclotron beam or that the entire cyclotron beam pass through the target. In the former case one must know the beam density in particles/cm^2 and the total mass of target material; in the latter case the area density of the target in atoms/cm^2 and the total number of incident particles are needed. Inasmuch as the external beam of the 60-in. Crocker Cyclotron is not uniform over its area, even when collimated, the second of these procedures was adopted for the Pu^{238} bombardments.

The required uniformly thick target was prepared by the electrolytic procedure given by Glass.[1] Owing to the limited amount of target material available, only 300 to 500 μg of plutonium was used in the plating cell for any one plating. The total amount of plutonium on the target varied from 60 to 230 μg as determined by counting on an ultra-low-geometry alpha-scintillation counter as well as by assay of the plutonium in the course of the chemical procedure following bombardment. The determinations by these two methods showed a systematic disagreement of about 8%; the number obtained from the low-geometry counter was high, presumably owing to errors in calibration of the low-geometry counter.

In the Pu^{238}-Pu^{242} mixture bombardments, only relative cross-section measurements were made, and therefore a uniform target was not necessary. In this case the target was made by simply slurrying a water suspension of PuF_3 or PuF_4 onto a platinum target disk and drying it under an infrared lamp. The resultant deposit adhered satisfactorily to the disk throughout the bombardment, but was very easily scraped off with a platinum rod. The total amount of plutonium on these targets was between 50 and 500 μg.

The target assembly for all bombardments was identical with that described by Glass,[1] except that the standard short-lipped target holder was found to be satisfactory. In loading the target, the target

holder invariably became contaminated with alpha activity, but satisfactory decontamination was achieved with alternate swabbings of acetone and 1 M HNO_3.

The initial energy of the external cyclotron beam changed from time to time; the particular values for the several bombardments varied a few Mev owing to cyclotron modifications made during the period in which these bombardments were carried out.[11] The energy was reduced to the desired value by placing suitably chosen weighed aluminum and (or) platinum foils over the target, with the foil nearest the target always aluminum. The resultant energies were calculated from the range-energy curve of Aron, Hoffman, and Williams[12] for aluminum and for the platinum, from a range-energy curve interpolated from the curves given for various elements by these workers.

Because the cyclotron beam was frequently not constant during bombardments, differential as well as integral beam-current measurements were made throughout each bombardment, the former being used to make corrections for decay of short-lived activities during the bombardment period. The total integrated beam ranged from 4.0 to 8.4 μah per bombardment.

Chemical Procedures[10]

After the Pu^{238} bombardments, the target was dissolved quantitatively, and radiochemically pure samples of americium, curium, and selected fission product elements were prepared. Known amounts of stable isotopes of the fission product elements being determined, and of alpha-emitting americium and curium isotopes not formed in the bombardments, were included in the target solution. The fission product samples were prepared as pure compounds from whose weights the chemical yields were calculated. The actinide yields were calculated from the count rates and alpha-pulse analyses of the final americium and curium samples.

Cross sections for the spallation products were determined for each bombardment. The fission cross section was approximated for all the bombardments by measuring cross sections for the formation of strontium, cadmium, and barium isotopes. In addition, cerium, neodymium, europium, and terbium cross sections were

measured for several of the bombardments. From these points, a fission yield curve for each bombardment energy could be estimated with little ambiguity.

The chemical procedure, where the more extensive fission product analysis was made, was as follows:

The target assembly was placed in a glove box and the bombarded target and the aluminum and platinum front foils were removed. The bulk of the plutonium on the target plate was loosened by scraping with a platinum stirring rod and washed with concentrated HNO_3 into a beaker already containing the measured carriers and tracers. The amounts of carriers used were: cadmium, 20 mg; strontium and barium, 10 mg each; cerium, neodymium, europium, and terbium, 5 mg each. The actinide tracer consisted of known amounts of about 30 disintegrations/min of Am^{243} and 150 disintegrations/min of Cm^{244}, along with some unavoidable Am^{241} and Cm^{242}.

When all the target material that loosened easily was in the beaker, the solution was made 6 M in HNO_3 and 0.05 M in HF and heated until the solution was clear. Then the aluminum target plate itself, as well as the aluminum foil nearest the target, were placed in the solution. These contained a large fraction of the fission product nuclides from recoil. Concentrated HCl was carefully added until the aluminum was completely dissolved. The solution was again heated until it was perfectly clear. Its volume was measured and small measured aliquots were removed and later prepared for counting to assay the total amount of plutonium in the solution.

In the procedure up to this point, which took about an hour or less, there had been no separation of any of the nuclides being determined or of any of their precursors having half lives longer than several seconds. The next step, which involved some fission product separation, was not begun until at least a full hour from the end of bombardment had elapsed. This allowed quantitative decay of all the precursors except some rare earth isotopes--which, however, undergo no appreciable separation from one another until much later.

After this decay period, the solution was made about 3 M in NaOH and 0.5 M in Na_2CO_3 from saturated solutions of these reagents.

This converts the aluminum to aluminate ion, which is retained in solution, and gives a precipitate of plutonium, americium, curium, and rare earth hydroxides and alkaline earth carbonates. In addition, the cadmium is partially precipitated; however, $Cd(OH)_2$ is slow to form and some cadmium remains in solution, lowering the yield of this element. The mixture was centrifuged and the supernatant liquid set aside in a hot bath. The precipitate was washed with water and dissolved in concentrated HCl. The resultant solution was saturated with HCl gas in an ice bath, forming a dense, white precipitate of $SrCl_2$ and $BaCl_2$. The mixture was centrifuged and the precipitate dissolved in water and then reprecipitated by again saturating with HCl in the ice bath. This procedure was repeated until the alkaline earth fraction contained about 10^6 disintegrations/min of alpha activity, at which point it was removed as a solution to a hood for further purification.

The first two HCl supernatants--which contain the actinides, lanthanides, and cadmium--were combined, diluted to 10 M HCl, and loaded onto a Dowex A-2 anion resin column, 5 cm long and 0.5 cm in diameter. (The behavior of the lanthanides and actinides on such a column has been discussed in the preceding section.) Cadmium, because it complexes extremely strongly with chloride ions, is strongly absorbed by Dowex A-2 from HCl solutions of any concentration above 0.1 M. It is desorbed by dilute H_2SO_4.

After the column was loaded, it was washed with 10 M HCl-0.01 M HNO_3 solution, which removes the americium, curium, and lanthanides. The plutonium was then stripped off with 2 M HCl.

By this time, an appreciable amount of $Cd(OH)_2$ had precipitated from the $NaOH$-Na_2CO_3 solution. This precipitate was centrifuged, washed, dissolved in HCl, and loaded onto the column. The column was then washed further with 2 M HCl to remove contaminants that were introduced in this step; then the cadmium was removed with 0.75 M H_2SO_4.

The cadmium and americium-curium-lanthanide fractions were radioassayed, and if they were sufficiently free of plutonium contamination they were removed from the box for further purification.

In the few cases in which they were not, a second anion-column purification was made.

In the hood, the cadmium and alkaline earth fractions were subjected to the procedure described by Glass,[1] giving samples of CdS, $BaCrO_4$, and $SrCO_3$. These were slurried onto small, flat, tared aluminum dishes in appropriate solvents (acetone for the $SrCO_3$, ethanol for the other two), dried under an infrared lamp, weighed, and covered with a thin layer of diluted Zapon lacquer (applied as a dilute ethyl acetate solution) to prevent loss of the precipitate in sample handling.

The fraction containing the lanthanides and trivalent actinides was treated as follows: The solution was heated to boiling to reduce the HCl concentration, then made basic with NH_3 gas, precipitating the lanthanide and actinide hydroxides. These were washed and dissolved in a small volume of concentrated HCl. This was transferred to a plastic cone, diluted with an equal amount of water, and treated with several drops of concentrated HF, giving the actinide and lanthanide fluorides, which were washed and dissolved in H_3BO_3-HNO_3 solution. The hydroxides were then reprecipitated, washed, and redissolved in concentrated HCl.

The solution was then saturated with HCl and loaded on a 4% cross-linked Dowex-50 cation-exchange resin column, 7 cm long and 1 cm in diameter. A few millimeters of Dowex A-2 anion-exchange resin was packed at the bottom of this column to remove the contaminating plutonium, which had been oxidized to Pu(IV) during the hydroxide precipitation.

The actinides were eluted with a 20% ethanol-80% water solution saturated with HCl.[13] The flow rate was about 4 drops per min, and fractions of about 20 drops were collected. Small aliquots of these were counted for alpha decay and electron-capture activity. After the actinides had been eluted, the eluting agent was changed to 6 M HCl, which was forced through the column as quickly as possible to strip off the lanthanides.

The actinides, which were usually in the twelfth to sixteenth fractions, were combined and evaporated to dryness to assure complete

elimination of alcohol and excess HCl. The americium and curium were then separated from each other (as well as from small amounts of heavy lanthanide fission products which may have been carried into this fraction) by elution with 0.4 M lactic acid adjusted to a pH of 4.32 with ammonium hydroxide, from a 15- by-0.2-cm 12% cross-linked Dowex-50 cation-exchange resin column, thermostated at 87° C, according to the procedure given by Thompson et al.[13] The eluant was collected in single drops on platinum plates, which were then counted for alpha- and electron-capture activity. The activity was removed from the curium plates with hot 6 M HCl containing 0.05 M HF and vaporized in vacuo from a hot tungsten filament to a platinum plate. This procedure gives extremely thin plates that yield excellent alpha pulse analyses. The americium plates were similarly vaporized if they seemed to have an appreciable amount of salts or oxides (presumably impurities in the lactic acid) on them; otherwise the spots containing the activity were simply cut from the various plates and mounted (with cellophane tape) on a single aluminum plate.

The lanthanide fission products were separated by lactate elution from cation-exchange columns by the continuously varying pH method developed by Nervik.[14] The procedure was as follows: The lanthanides were precipitated with NH_4OH, washed, dissolved in a minimum volume of concentrated HCl, diluted to around 0.5 molar acid, and equilibrated with three successive 0.5 ml portions of 4% cross-linked Dowex-50 cation-exchange resin. The resin was rinsed in degassed water, slurried in a minimum amount of degassed 1 M ammonium lactate-lactic acid solution of pH 3.2, and placed on top of a column of the same resin 45 cm long and 0.7 cm in diameter, thermostated at 80° C by a water jacket. The eluting agent, which was forced through the column at a rate of 4 drops per min and collected in 24-drop fractions, was supplied to the column from a 500-ml reservoir originally containing 1 M lactic acid adjusted to a pH of 3.2 with ammonium hydroxide. A solution of 1 M lactic acid adjusted to a pH of 5.0 dripped into this reservoir (which was continuously magnetically stirred) from another reservoir above at a rate of 4 drops per min. Thus the volume in the lower reservoir

was constant, but the pH increased steadily at the rate of roughly 0.02 pH unit per hr. Since the rate of lanthanide elution increases with increasing pH, the effect of this procedure is to give satisfactory separation of the earliest (heavy) lanthanides without requiring an excessively long time for the separation of the late ones. The total time for such a run was usually about 42 hr, with terbium (the heaviest element of interest) usually requiring some 16 hr for elution.

The first time this column was operated, aliquots of each fraction were counted in a Geiger counter, verifying quantitative separation of all the lanthanides of interest from their neighbors. In succeeding runs, the lanthanides of interest were located by the formation of precipitates upon the addition of saturated ammonium oxalate solution and identified by their order of elution and by the colors of their ignited oxides.

The oxalates were washed, ignited to the oxides in porcelain crucibles, transferred to weighed aluminum dishes, weighed, and coated with Zapon lacquer in preparation for counting.

In those bombardments in which lanthanides were not included among the fission products determined, about 0.1 mg lanthanum carrier was included in the target solution in place of the other lanthanide carriers, and an alcoholic-HCl column 0.2 cm in diameter and 10 cm long was used in place of the larger one.

For the Pu^{242} bombardments, in which neither fission cross sections nor absolute values for the spallation cross sections were determined, the target material was simply dissolved in HNO_3-HF solution without attempt at quantitative dissolution or recovery of products in the backing plate, and put through the anion-exchange column, fluoride precipitation, small alcoholic-HCl column, and lactate-column procedures described above. Only the curium fraction was examined.

Radioactivity Measurements

As soon as the fission product samples were prepared (about 8 hr after the end of bombardment for the cadmium, strontium, and barium, and about 3 days for the lanthanides), they were put on an automatic counting wheel, which is a device for counting a number of

samples in rotation for a preset time interval (15 min in this procedure). Since no more than seven samples (in addition to a blank position giving background counts) were ever counting on the wheel at one time, each sample was counted at least once every two hours. The detector was a standard calibrated end-window argon-filled halogen-quenched Geiger-Mueller tube held above the sample at an effective geometry of about 3%. A cylindrical lead shield surrounded the counting tube and extended down to within a few millimeters of the counting wheel itself. After removal from the counting wheel, the samples were manually counted at appropriate intervals (from daily to semimonthly) by use of a similar counter at similar geometry.

Electron-capture decays of both americium and curium were followed automatically at 20-min counting intervals with windowless methane-flow proportional counters coupled to Nucleometers[15] for at least 24 hr, after which the counting was continued at appropriate intervals by hand.

For Pu^{238} bombardments at energies where the (α, 4n) product, Cm^{238}, was expected, the curium fraction was divided and the decay of this nuclide was followed in a portion of it on the 48-channel alpha pulse-height analyzer.[16] For those energies at which the (α, 3n) product, Cm^{239}, should be important, both fractions were divided and a sample of each followed on the 50-channel gamma scintillation spectrometer, which uses a cylindrical thallium-activated sodium iodide crystal as detector.[17] It was hoped that Cm^{239} could be determined by decay of some characteristic gamma ray or growth of the Am^{239} daughter gamma rays at 230 and 280 kev. The americium sample spectrum was followed so as to obtain an intensity standard for these two gamma rays. However, the intensities were insufficient for any conclusions to be drawn.

At some convenient time later, careful alpha pulse analyses and alpha decay rate measurements were made, the latter by use of a conventional windowless argon-flow ionization chamber.

For the Pu^{242} bombardment, the only data needed were a careful alpha pulse analysis of the curium fraction.

IV. TREATMENT OF DATA

In interpreting the decay data, it is first necessary to resolve the decay curves into their components and extrapolate these to the end of bombardment. From the over-all counting efficiency one can calculate the disintegration rate at the end of bombardment, which is then divided by the decay constant and the chemical yield to give the number of atoms of the particular nuclide present at the end of bombardment. For nuclides with sufficiently short half lives, a correction must be applied to correct for decay during the course of the bombardment, giving the required quantity N, the number of nuclides of this mass formed during the bombardment. The cross section is then calculated from the formula

$$\sigma = \frac{NA}{In} \quad ,$$

where I is the total number of bombarding particles that have struck the target, A is the target area in cm^2, n is the total number of target atoms, and σ is the cross section in cm^2.

For the fission product activities, the counting efficiency consists of factors for geometry, the counting efficiency of the counting tube itself for radiations that manage to get into it, absorption of radiation by the counter window and the air between it and the sample, self-scattering and self-absorption by the sample, and back-scattering by the aluminum plate on which the sample is mounted. The geometry, counter-tube efficiency, and air and window absorption tend to decrease the counting rate, the others (for the sample thicknesses encountered in this work) to increase it.

The geometry and counter-tube efficiency were taken as a single factor, different for the different Geiger counters. The air-window corrections for the counters used were those of Ritsema.[18] For the activities of Cd^{115} and Ba^{140} (in equilibrium with their respective daughters), the corrections for back-scattering and self-scattering-self-absorption were in a single term determined experimentally for each of these nuclides as a function of sample thickness.[19] For the other nuclides, the two corrections were estimated from the data of Burtt[20] and Nervik and Stevenson[21]

respectively. Since the counting wheel and the sample shelves of the nonautomatic Geiger counters were of aluminum, sufficient backing to achieve saturation back-scattering was present at all times. The count rates of the Ba^{140}-La^{140} equilibrium mixtures, decaying with the Ba^{140} half life and extrapolated to the end of bombardment with this half life, were divided by 2.15 to take the daughter activity into account. The Cd^{115} N's were divided by 1.43 to take into account the formation of Cd^{115m} and the partial transient equilibrium of In^{115m}, while the Sr^{91} N's were divided by 1.07 to account for the partial transient equilibrium of Y^{91m}.

Since the alpha pulse analyzer and the alpha counters have counting efficiencies that are independent of the alpha particle energy, the absolute disintegration rates of the alpha emitters were calculated directly from the alpha peak intensity ratios and the alpha counting rate of the americium and curium fractions. The alpha-counter counting efficiency enters only in the determination of the absolute disintegration rate of the added tracers. In the Cm^{241} calculations, the electron-capture branching was of course taken into account, the branching ratio (alpha decay rate divided by electron-capture decay rate) being 0.0093.[2] For the Cm^{238} calculations, the partial alpha half life of Cm^{238} was estimated from the alpha systematics, and the corresponding branching ratio of 0.018 alphas per disintegration was used in the calculations.

No determination of the (α, 3n) cross sections was made. Cm^{239}, the product of this reaction, is known to have a half life in the neighborhood of 3 hr, but this value is not known with precision and an additional isomer of 10- to 15-hr half life is suspected.[23] Since the daughter half-life is 12 hr and the 2.5 hr Cm^{238} activity is usually also present, resolution of the curium Nucleometer curves is not possible.

The americium Nucleometer decay curves were resolved into their Am^{240} and Am^{239} components, sometimes by the usual graphical method but more often by Biller's graphic-analytic method,[24] after corrections for the alpha-decay contribution to the Nucleometer activity had been made. The Nucleometer counting efficiencies were assumed to be 80% on the basis of the work of Glass et al.[2]

It must be remembered that since the first americium-curium separation takes place some 4 hr after the end of bombardment, much of the Am^{239} in the americium samples was formed by Cm^{239} decay. Thus the apparent (α, p2n) cross section is an upper limit and approximates a lower limit for the sum of the cross sections for this and the (α, 3n) reactions.

The Pu^{242} reaction cross sections were calculated from the yields of their products relative to the yield of Cm^{240}, which is formed by the Pu^{238}(α, 2n) and Pu^{239}(α, 3n) reactions. In the $Pu^{238-242}$ mixture bombardments, the Pu^{238}(α, n) cross sections were calculated from the relative yields of Cm^{240} and Cm^{241} (using the Pu^{238}(α, 2n) cross section as a standard, and taking into account the small contribution of the Pu^{239}(α, 2n) reaction to the latter). The values found are in satisfactory agreement with the more directly measured values, providing an internal check on the consistency of the data.

V. RESULTS

The results are tabulated on Tables III to V and presented graphically on Figs. 2 to 6.

In the drawing of the fission yield curves, reflected as well as experimental points were considered. In making the reflections we assumed that three neutrons were emitted from the primary compound nucleus before or during fission in the bombardments at 33 Mev or less, four neutrons at 37 Mev and 42 Mev, and six neutrons at 47 Mev.

In the three cases in which rare earth fission product cross sections were measured, it was noted that the Ba^{140} cross section was always low, and always by the same amount--a factor of about 1.6. The reasons for this discrepancy are not clear. A partial reason is undoubtedly an independent yield of those nuclides of mass 140 having fewer neutrons than the barium isobar. Ba^{140} has a greater neutron-to-proton ratio and a smaller deviation from the most probable charge for the primary fission product of a given mass number (Z_p) than any other nuclide studied in this work. Thus, by either the postulate of unchanged charge distribution or the postulate of equal charge displacement, it is to be expected that the error due to independent yield of higher-Z isobars in a given chain would be higher for Ba^{140} than for any other nuclide studied. The magnitude of the discrepancy is much greater than one calculates following the method of Glendenin, Coryell, and Edwards, whose treatment, however, may not apply to the considerably excited fissioning nuclei encountered in this work.[22]

Regardless of the reason for the low barium values, the consistency of the discrepancy is such that it seems justifiable to assume it to be systematic, and accordingly, in drawing the fission yield curves, the true cross section for formation of mass-140 fission products was always assumed to be 1.6 times the experimental Ba^{140} cross section.

Table III

Plutonium-238 fission product cross sections (millibarns)

Element determined	Mass number	Alpha energy (Mev) 25.2	28.7	30.2	33.0	36.6	42.2	47.4
Sr	91		14.4		16.6	11.6	11.2	29.2
Cd	115	10.0	22.9	28.2	43.0	38.2	43.2	57.5
Ba	140	10.0	11.8	16.6	19.5	22.4	15.4	18.8
Ce	143			24.6		34.5	21.5	
Nd	147			17.2		23.3	20.0	
Eu	156			2.8		3.6	3.7	
Tb	161			0.48		1.9	1.0	
Total fission cross section		430	640	980	1100	1000	1000	1400

Table IV

Plutonium-238 spallation product cross sections
(millibarns)

Reaction	Product	Alpha energy (Mev) 25.2	28.7	30.2	33.0	36.6	42.2	47.4
α, n[b]	Cm^{241}	4.6	7.1		6.0	3.1	2.5	2.8
α, 2n	Cm^{240}	14.6	13.6	9.9	8.9	4.7	4.3	3.5
α, 4n	Cm^{238}					< 0.002	0.19	0.26
α, pn(α, d)	Am^{240}		2.6	1.9	3.3	15.3	12.5	8.0
α, p2n[a](α, dn; α, t)	Am^{239}		3.0	5.2	6.9	26.8	22.0	18.4

[a]Not corrected for Am^{239} formed indirectly by decay of Cm^{239}, the (α, 3n) product.

[b]See also Table V

Table V

Plutonium-242 spallation product and Pu^{238} (α, n) reaction cross sections
(Calculated relative to Pu^{238} (α, 2n)Cm^{240})
(millibarns)

Reaction	Product	Alpha energy (Mev) 23.6	25.9	27.1	28.5	32.8	38.8	43.5
Pu^{242}(α, 2n)	Cm^{244}	103.0	116.3	70.0	67.8	30.0	24.1	35.2
Pu^{242}(α, 4n)	Cm^{242}	0	0	0	0	1.8	8.6	8.3
Pu^{238}(α, n)	Cm^{241}	6.8	8.2	6.5	7.8	6.2	2.9	2.8

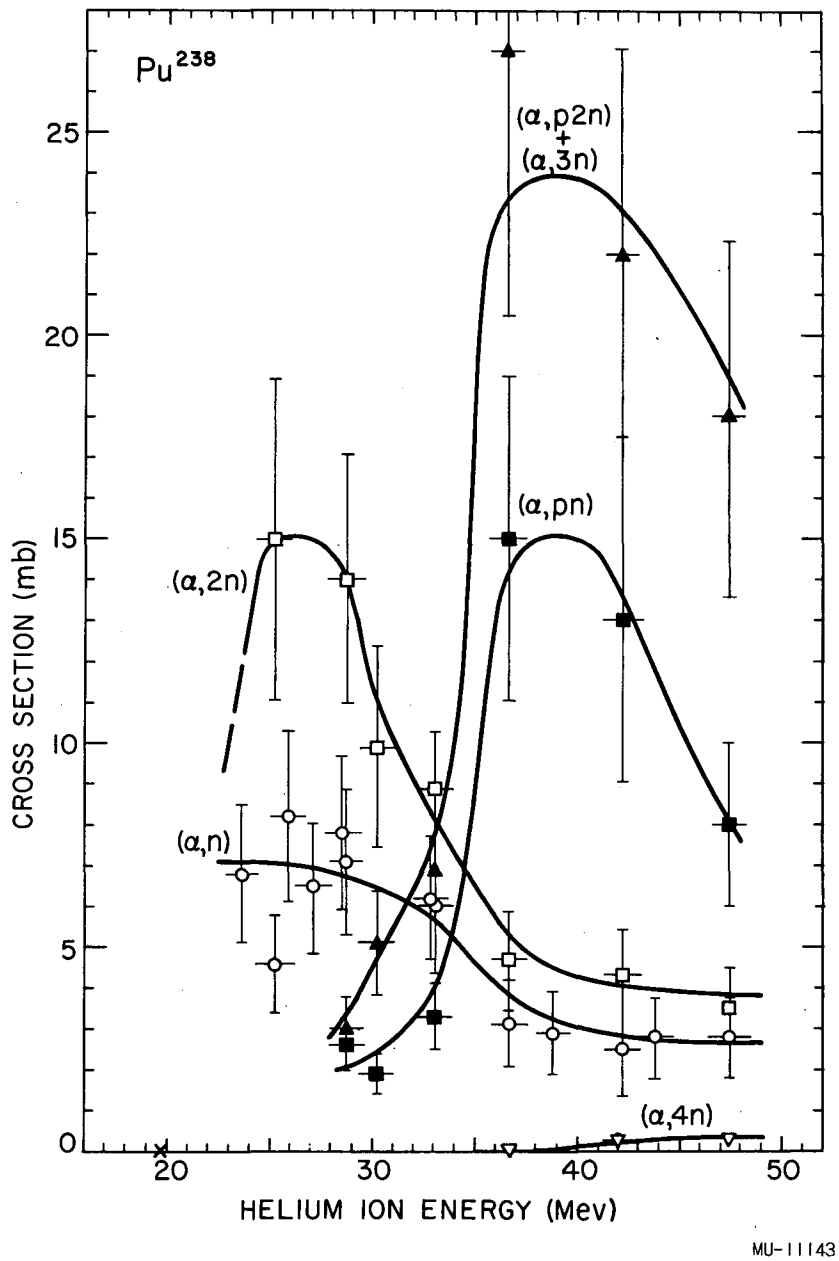

Fig. 2. Pu^{238} spallation excitation functions.

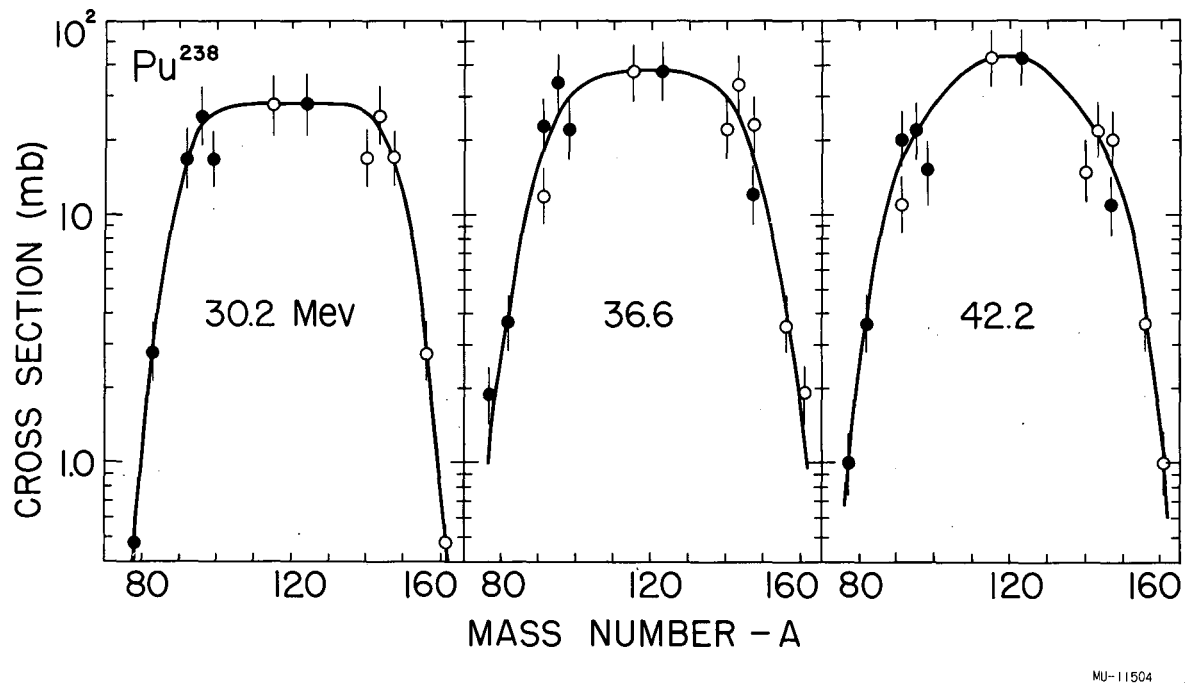

Fig. 3. Pu^{238} fission yield curves. Open circles: experimental points; solid circles: reflected points.

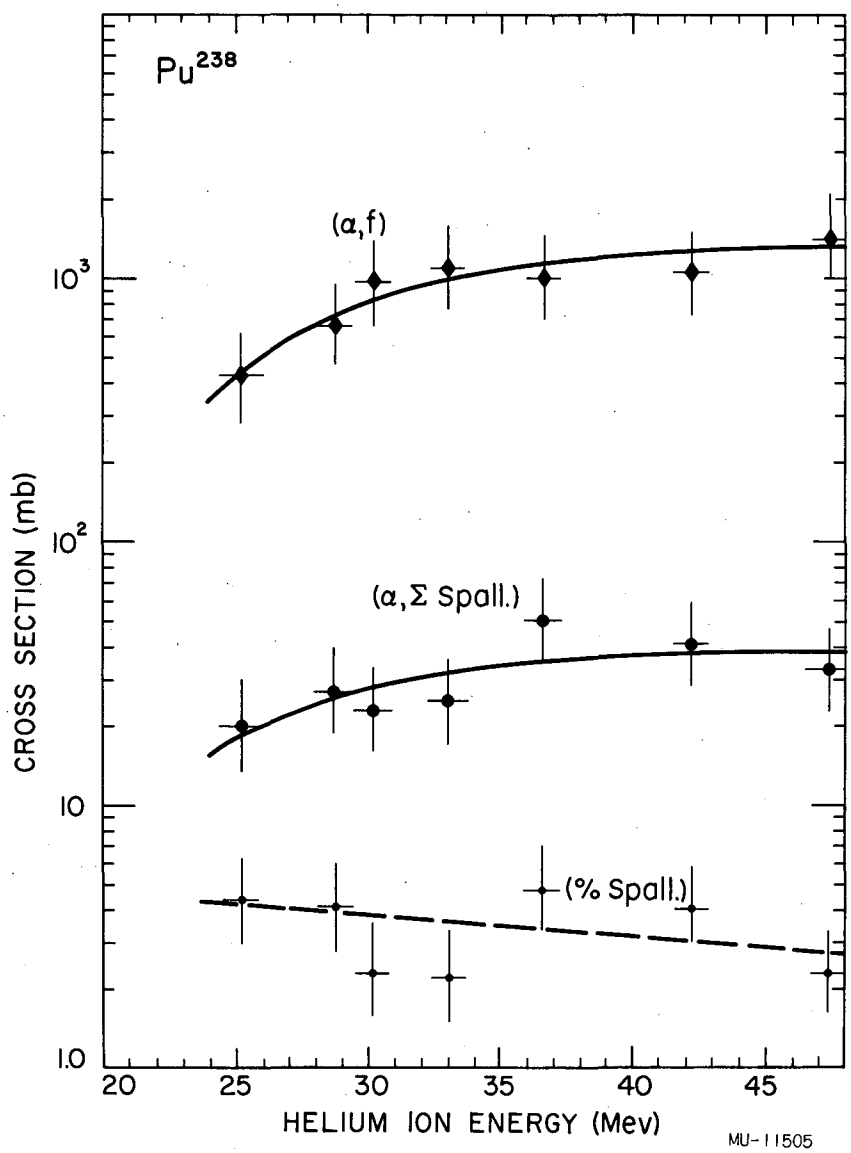

Fig. 4. Pu^{238} fission excitation function and total spallation excitation function. The latter includes only those spallation reactions which have actually been determined.

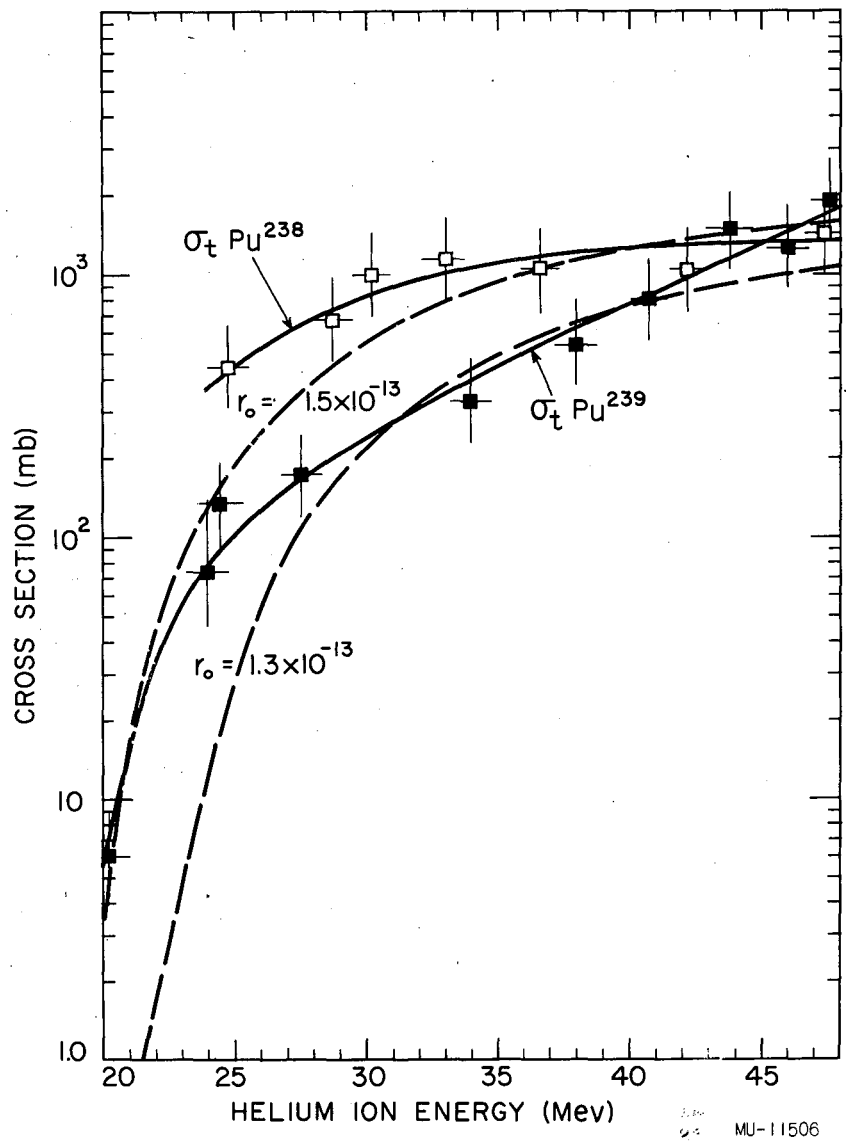

Fig. 5. Pu^{242} spallation excitation functions.

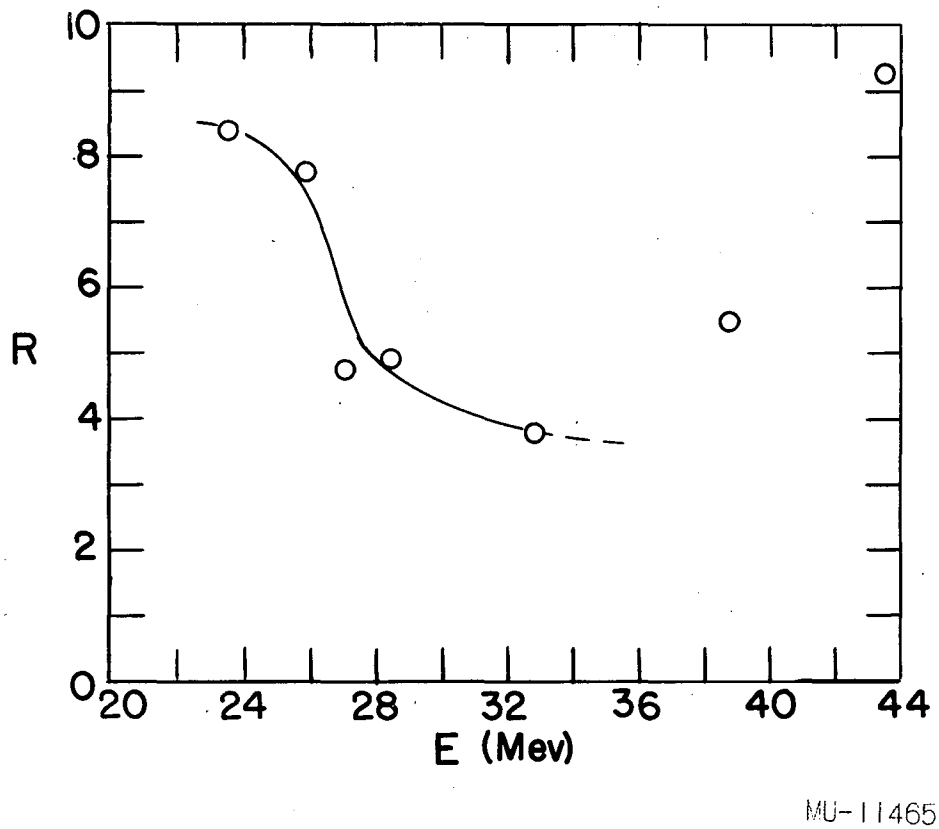

Fig. 6. Ratio of $Pu^{242}(\alpha, 2n)$ cross section to $Pu^{238}(\alpha, 2n)$ cross section versus bombarding energy.

Sources of error in the experimental results may be divided into two classes: errors in those measurements which have to do with the target and the bombardment and are thus common to all the cross sections measured in any one bombardment, and errors in determination of the numbers of atoms of various kinds formed in a bombardment.

Considering first the errors having to do with target and bombardment: measurement of the total integrated current through the target was by means of integrators which were calibrated before each bombardment and which are known to be accurate to 1%. The determination of the number of target atoms has been previously discussed. On the basis of the consistency between results of the two methods of determination, assay of the target solution, and direct counting of the target itself, error from this source may be assumed to be about 8%. That the beam had passed through the target completely was assured by visual inspection of the target. (In the bombardment at 36.6 Mev, the beam partially missed the target. The results of this bombardment have been arbitrarily normalized.)

Errors arising from measurements of the target area, and--more important--the assumption of uniform target thickness are difficult to assess a priori. The scatter in the total fission cross section values (the details of which are qualitatively reflected in the scatter of cross sections for various individual fission products) arises primarily from this source, as no other source of error is large enough to account for it. This uncertainty is about 20%. None of these errors, of course, affects the relative values of the cross sections for the various reactions at any one given bombardment energy.

Of the errors affecting the calculated values of N (the number of product atoms formed), some are common to all determinations of a given nuclide whereas others only apply to a particular bombardment.

Of the systematic errors, the most serious are probably in the counting efficiencies, both for fission products and for those actinides which were determined by electron-capture counting. For the nuclides for which self-scattering-self-absorption curves were known, i.e., Cd^{115} and Ba^{140}, errors from this source are negligible because the conditions for which the curves were determined were the same as those for which they were applied. For those fission product nuclides in which the corrections were interpolated from the data of Stevenson and Hicks, more uncertainty exists. However, there was rarely a difference of more than about 10% in the corrections for the two scatterers these workers considered, and so--allowing some validity to the interpolation--an error of more than about 5% from this source seems unlikely. Strictly speaking, these errors are, of course, not systematic as the counting-plate thicknesses varied from sample to sample. However, the thicknesses were usually in the range where the correction was varying slowly, so that nonsystematic discrepancies are probably negligible.

For the Nucleometer-counted actinides, Am^{239} and Am^{240}, the assumed counting efficiency of 80% may be open to question. The counting efficiency for neither of these nuclides was measured directly, but the value is known to be correct for Cm^{241} by comparison of Nucleometer count rate with rate of growth of the alpha emitting daughter, Am^{241}. All three of these nuclides have complicated decay schemes and probably all involve a large abundance of Auger and/or conversion electrons, so one expects a similarity in Nucleometer counting efficiency; however, the assumed value might be incorrect by as much as, say, 20%.

Of course, the calculated cross sections for formation of Cm^{241} and Cm^{238} depend on their alpha branching ratios. The value for the former, a direct measurement involving only alpha counting, is completely reliable for the present purposes, but the Cm^{238} value, based on an extrapolation of the alpha systematics, is probably no more certain than about 20%.

The other sources of error, those pertaining to a particular nuclide in a particular bombardment, arise in resolution of decay curves and pulse-analysis curves. Probable errors in resolving fission product decay curves, estimated by inspection of those curves, are as follows: Sr^{91}, 15%; Cd^{115}, 5%; Ba^{140}, 8%; Ce^{143}, 12%; Nd^{147}, 4%; Eu^{156}, 10%; Tb^{161}, 25%. For the europium and terbium, the primary source of error is poor statistics. In fact, in the latter, all samples were below background. In the strontium samples, difficulty was encountered in resolving the activity from the short-lived and complicated Sr^{92}-Y^{92} mixture, and, in some cases, from an unidentified long-lived contaminant. In fact, in two of the cases, the contaminant was present to such an extent that the 9.7-hr Sr^{91} could not be resolved at all.

Most of the curves of course contained activities from other isotopes of the same element. However, the uncertainties in those activities were so much greater than those for the nuclides listed above that they did not in general contribute to the over-all accuracy and accordingly have not been quoted.

In the resolution of the americium decay curves, correction was first made for alpha emitters by use of the alpha counting rate and the known Nucleometer counting efficiency of alpha particles. The error arising in the subsequent resolution of Am^{239} from Am^{240} was about 10%. Error in resolving the americium pulse-analysis curves is about 5%.

In the curium pulse analyses the Cm^{240} and Cm^{244} peaks were well resolved and errors in the Cm^{240}/Cm^{244} ratio are about 5%. However, the intervening Cm^{241} peak is very much smaller, owing to the small alpha branching of this nuclide, and the error in the Cm^{241}/Cm^{244} ratio, both from poor resolution and from poor statistics, is about 20%.

Sources of error other than those discussed above--such as impurities in fission product samples, weighing errors, and uncertainties in absolute alpha counting, Geiger tube calibration, or air-window absorption corrections--are quite negligible.

On the basis of the above discussion, the estimated errors are about 20% in the Cd, Ba, Nd and Cm^{240} cross sections, 25% in the Sr, Ce, and Eu cross sections, 30% in the Am^{239}, Am^{240}, and Cm^{241} cross sections, and 35% in the Tb cross sections. The uncertainty in the integrated fission cross sections is about 25%. These uncertainties of course refer to the absolute cross-section values. Relative values, either at one bombardment energy or for one product nucleus, are less uncertain.

Since the errors in pulse analysis of the curium plates from Pu^{242} bombardment are small, the uncertainties of the Pu^{242} reaction cross sections are equal to those of the Pu^{238} and Pu^{239} cross sections on which they are based.

VI. DISCUSSION

A quantitative discussion of the results presented here is, unfortunately, impossible. The primary reason is the lack of a really satisfactory quantitative theory for even the simple reactions of non-fissionable nuclei.[25,26] The statistical "compound nucleus" model requires a formula giving the level density of excited nuclei. The most usual expression for the level density is of the form expected for a degenerate Fermi gas, $C \exp 2\sqrt{aE}$, where C and a are parameters which vary slowly with mass number for a given nuclear type and E is the excitation energy; but there is disagreement as to whether nuclear type should be accounted for by a constant factor before the whole expression[27] or by an added term under the radical in the exponent.[28] Furthermore, experimental measurements leading to the level-density function indicate that the degenerate gas form in itself is inaccurate.[29] Also, considerations of cross sections of reactions involving charged-particle emission,[30,31,32,33] and of relative cross sections of (α, n) and $(\alpha, 2n)$ reactions,[34] indicate the inadequacy of the compound-nucleus model, regardless of level-density formulas. No satisfactory alternative theory exists.

In addition to the problems raised by spallation reactions in the absence of fission, a number of fundamental facts about the fission reaction itself are lacking. Chief among these is the dependence of fissionability upon nuclear type.[35]

Despite these uncertainties, however, it is quite possible to discuss the qualitative implications of the experimental data, though few unambiguous conclusions can be reached.

Considering first the fission excitation functions, we note that the magnitude of the integrated cross section is significantly higher than that found in the Pu^{239} bombardments,[2] but consistent with the general trend established by the alpha-induced fission cross sections for Th^{232} and U^{235} at about 35 Mev.[36,37]

More significant than the magnitudes of the fission cross sections, perhaps, is the fact that the fission excitation function rises more sharply and begins to level off more abruptly for Pu^{238}

than for Pu^{239}. In this respect, the Pu^{238} function resembles the U^{238} fission-excitation function,[18] suggesting that we may be dealing with an effect of nuclear type. This may be related to the fact that Pu^{239}, being an even-odd nuclide, has a nonzero spin and may therefore have a sticking probability of less than unity at even the lowest energies. However, the small value, 1/2, of the Pu^{239} spin makes this explanation very questionable. In regard to this consideration of nuclear type as the determining factor in the rise of the reaction cross section, it should be noted that the (α, 2n) excitation function, whose leading edge may be presumed to reflect to some extent the reaction excitation function as a whole, peaks several Mev higher for Pu^{239},[2] than for either of the even-even target nuclides.

The shapes obtained for the fission yield curves at the various energies are somewhat surprising in terms of previous experience. In alpha-induced fission of Th^{232},[36] and Pu^{239},[1] peak-to-valley ratios of 2 are found for bombardment energies of around 37 Mev. In proton-induced fission of natural uranium[38] and Th^{232},[39] these ratios occur for bombardment energies of 17.5 Mev and above 21 Mev respectively, corresponding in terms of the excitation energy of the primary compound nucleus to alpha-bombardment energies of 27.5 Mev and above 31 Mev. In strong contrast to these results, the fission yield curves for Pu^{238} show a peak-to-valley ratio of only about 1.5 at 25 Mev, the lowest energy studied, and no valley at all above 30 Mev.

In considering the spallation reactions, it is necessary to have some sort of picture of the way in which fission competes with spallation in the de-excitation of compound nuclei. One such picture, used by Glass[1] and by other workers, relates the fissionability of a compound nucleus to its Z^2/A value by analogy with spontaneous fission.[40, 41] While this treatment cannot be ruled out theoretically or experimentally, it seems open to question because of the fundamental difference between spontaneous fission and fission of nuclei excited to energies above the fission threshold. The Z^2/A value has been related to the penetrability of the Coulomb barrier in spontaneous fission, whereas the determining factor in fission of excited nuclei

is probably not the penetrability (which probably varies by a factor of only 10 or so over the range of excitation energies at which fission is important), but something related to the frequencies of the various oscillatory modes leading to fission relative to the probability per unit time of a single nucleon's receiving the momentum needed to boil off.

Instead of using this picture, we will discuss the results on the basis of a purely qualitative model which assumes that--except for energies near the respective thresholds--fission is very fast compared with gamma emission, but slow compared with neutron emission. One then has the following very simple situation: A compound nucleus excited to an energy above the effective neutron threshold will emit a neutron (or perhaps a proton or other small particle), a compound nucleus excited to an energy below this but still above the effective fission threshold will undergo fission, and a compound nucleus excited below the effective fission threshold will de-excite by gamma emission. Thus one may think of the energy levels of a compound nucleus as having a "fission band" in which fission will occur, with a "gamma-emission band" below and a "spallation region" above. The effective fission threshold, which marks the lower limit of the fission band, may be assumed, on the basis of plots of slow-neutron fission-to-capture ratios versus excitation energy, to be a few hundred kilovolts above the fission threshold as determined by neutron reaction data[35,41] while the effective neutron threshold, the fission band's upper limit, may, on the basis of Morrison's statement about neutron emission versus gamma emission in compound nucleus decay,[42] be several (or perhaps hundreds of) kilovolts above the neutron binding energy.

Since, for a given nuclear type, the fission threshold (from slow-neutron data or consideration of spontaneous fission rates) is constant or nearly so,[35,41] while the neutron binding energy decreases with increasing mass, the fission bands decrease in width (and more strongly in number of levels included) with increasing mass for a given element. Effects of nuclear type are of course important with regard to the upper limit of the fission band; the effect on the lower limit is not known.

What has been said here applies to compound nuclei. The question of fission competition in direct-interaction mechanisms (e.g., stripping or "knock-on" processes) is quite different. In the first approximation, it would seem that fission is less important relative to spallation in direct-interaction processes than in compound-nucleus processes because fission (except very asymmetric fission) must involve a large part of the nucleus. It must be noted, however, that a direct-interaction process will usually leave a somewhat excited residual nucleus which is effectively a compound nucleus, subject to the same modes of de-excitation as a comparable compound nucleus more directly formed.

In considering competition between various nuclear reactions, it is well to bear in mind the following rather obvious facts:

1. Competition among reactions proceeding by compound-nucleus mechanisms occurs in the usual manner.

2. There may or may not be competition among reactions proceeding by direct-interaction mechanisms (i.e., an increase in the cross section for one such reaction may or may not cause a corresponding decrease in the cross sections of others), depending on the particular mechanisms involved.

3. There may or may not be competition between reactions proceeding by direct interaction and compound-nucleus formation; however, such competition must be with compound-nucleus formation as a whole and not with any one particular compound-nucleus reaction. This is a direct consequence of the definition of a compound nucleus.

With these considerations in mind, the experimental spallation-excitation functions will be considered.

Of the Pu^{238} excitation functions, the one which corresponds most to expectation is the (α, 2n), which shows a well-defined peak followed by a long tail. The rise of the peak reflects the rise of compound-nucleus formation as a whole; its fall reflects the competition of fission and (α, 3n) reactions--but not (α, pn) and (α, p2n) reactions which, as we shall see, arise primarily from direct interaction mechanisms. To state this more mechanistically, as the bombarding energy increases, the probable excitation energy of the

intermediate excited Cm^{240} nucleus also increases until, at energies beyond 27 Mev, it becomes more and more likely that it is excited in the fission band or spallation region rather than the de-excitation band. The tail is to some extent, of course, the result of compound-nucleus mechanisms in which unusually large amounts of energy have been carried off by the emitted neutrons, but it seems likely that the bulk of it is the result of direct interaction, presumably a "knock-on" type of reaction occurring in the periphery of the nucleus. That the tail is more prominent relative to the peak in this case than in the corresponding reactions of nonfissionable nuclei is a direct consequence of the supposition that fission is more likely by compound-nucleus mechanisms than by direct-interaction mechanisms. Fission thus provides, to some extent, a "sorting out" of spallation reactions occurring by the two mechanism types.

The $Pu^{242}(\alpha, 2n)$ excitation function is, throughout the energy range, several times greater in magnitude than the corresponding Pu^{238} function, as one expects from the fact that Cm^{245} has a considerably smaller neutron binding energy and therefore narrower fission band than Cm^{241}.[42] (Alternatively, one may argue that the heavier nuclide has a smaller Z^2/A.) The shapes of the two functions seem at first glance to be similar. A more sensitive comparison may be made by plotting the ratio of the $Pu^{242}(\alpha, 2n)$ cross section to the $Pu^{238}(\alpha, 2n)$ cross section as a function of energy, as has been done in Fig. 6. From our previous discussion, one expects a decrease in this ratio as one passes from the energy range of predominantly compound-nucleus mechanism to the region of predominantly direct-interaction mechanism, and, indeed, this is shown by the experimental results. The subsequent increase at the highest energies is quite surprising. It may be partly related to the fact that Cm^{243} is not resolved from Cm^{244} by the alpha pulse analyzer (the ratio of specific activities is such that the former is counted one-third as efficiently as the latter, atom for atom) and partly to experimental error; but, at any rate, it seems unlikely that it is related to questions of compound-nucleus versus direct-interaction mechanisms.

Comparison of the (α, 2n) functions of Pu^{238} and Pu^{239} is not fruitful because the differences are not far (if at all) out of the range of experimental error. Suffice it to say that the two functions are similar in shape; the fact that the former falls more steeply than the latter may be an effect of nuclear type.

The (α, n) excitation function is quite surprising. Whereas the corresponding Pu^{239} function varies slowly and smoothly throughout the energy range, the Pu^{238} function starts out at a fairly constant, surprisingly high level, falls rather abruptly concomitantly with the fall of the (α, 2n) and the rise of the (α, pn) and (α, p2n) [and presumably (α, 3n)] functions, and levels off at a new value still considerably greater than the corresponding one for the Pu^{239} target. Why there should be this difference between the two targets is hard to say, but since these reactions proceed, as we shall see, by direct-interaction mechanisms, presumably in the periphery of the nucleus, it is perhaps not too startling to find a considerable effect of the presence or absence of an unpaired nucleon in the target nucleus; i.e., an effect of nuclear type.

The abrupt fall of the (α, n) function is most likely an effect of the rise of one or more competing reactions. These cannot be compound-nucleus reactions because, on a compound-nucleus basis, the (α, n) peak should occur 2 or 3 Mev above the (α, 2n) threshold energy, which is 19 Mev; i.e., in an energy range made inaccessible by the Coulomb barrier of the target nucleus toward the bombarding particle. It must therefore be the effect of direct-interaction competition, presumably by the (α, pn) and (α, p2n) reactions. This conclusion places certain limitations on the possible mechanisms of these reactions.

The (α, pn) and (α, p2n) reactions--or, more precisely, the reactions leading to Am^{240} and Am^{239}--are extremely complex. In the first place, the latter have been only partly separated from the (α, 3n) reaction, and the reported cross sections are more nearly the total for the (α, p2n) and (α, 3n) reactions than the values for the (α, p2n) alone. Furthermore, these reactions may involve complex-particle emission; i.e., what we have been calling the (α, pn) reaction

includes the (α, d), while the (α, p2n) includes the (α, dn) and (α, t) reactions. Where more than one particle is emitted, any order of emission is possible in principle. The -Q values, in Mev, for these reactions are: (α, pn), 18.5; (α, d), 16.3; (α, p2n), 24.2; (α, dn), 22.0; (α, t), 15.7; (α, 3n), 26.7.

For Pu^{239}, in which the (α, 3n) and (α, p2n) reactions have been studied separately, the former has an excitation function of a type that would be expected for a compound-nucleus process, whereas the latter rises fairly steeply to a surprisingly high value and tends to level off there. Inasmuch as the sum of these functions is a curve quite similar to that found for the corresponding Pu^{238} function (though less steep), we will assume that the same situation exists in that case.

The (α, pn) and (α, p2n) functions both rise steeply in the region from around 30 to 35 Mev. The former peaks around 40 Mev, while the latter is really not well enough defined to estimate a peak position, or even to state unequivocally that a peak exists at all. It seems quite clear that these reactions cannot originate to any great extent from compound-nucleus paths involving individual nucleon emission, since their cross sections are so much greater than those of the reactions involving the evaporation of only neutrons to the same number of total nucleons; i.e., the (α, 2n) and (α, 3n) reactions, which are subject to no Coulomb barrier in the emission steps. In the (α, p2n) reaction, this interpretation is also out of the question on the basis of the energy of rise of the function. If the mechanism were single-nucleon emission from a compound nucleus, the rise would be several Mev above the sum of the -Q value and the height of the Coulomb barrier (about 12 Mev); i.e., around 40 Mev.

Another possibility is a compound-nucleus mechanism involving deuteron or triton emission. This suggestion is unattractive because the time presumably required to form such a particle in the nucleus would probably make it impossible for this to compete with nucleon emission; also, the advantage over neutron emission on the basis of lower -Q value is balanced by the disadvantage of having a Coulomb barrier to surmount.

If one accepts the Z^2/A picture, rather than the band picture, for fission competition in compound-nucleus mechanisms, the objections to the compound-nucleus mechanisms just discussed are somewhat mitigated by the fact that the intermediate excited nuclei in the reactions involving charged-particle emission are less subject to fission than their counterparts in the neutron-out reactions, by virtue of their lower Z^2/A values. It is difficult to say if as large an effect as is observed may be accounted for in this way. It seems an unlikely explanation because it does not account for the fact that a corresponding prominence of charged-particle-out reactions relative to neutron-out reactions is observed for nonfissionable target nuclei.

A third type of possible mechanism is stripping; i.e., a process in which only part of the bombarding helium ion interacts with the target nucleus, the rest of it passing by without interaction. This type of mechanism, so important in deuteron-induced reactions, seems unlikely in this study because of the great stability and compactness of the helium ion.

All the possible mechanisms mentioned above have the additional objection that they offer no competition to the (α, n) reaction, and thus leave the shape of its excitation function unexplained.

The most attractive alternative is a direct interaction in the periphery of the nucleus. Presumably the incoming helium ion interacts with a few nucleons, perhaps breaking to give a deuteron or triton which may then escape from the nucleus if the energetic situation is favorable. Alternatively, nucleons may be emitted. Such a process does afford competition to the (α, n) reaction, as is observed. It should be noted that this type of direct interaction, like the compound-nucleus process, involves surmounting of the Coulomb barrier and so should have the same effective threshold. In either case, the Coulomb barrier may be lowered for deuteron and triton emission if these particles are polarized in emission.[43] Reactions by the two mechanisms should differ in the energy spectra of the emitted particles and consequently in the shapes of the excitation functions. The direct-interaction mechanism should give a function whose rise

and fall is slower than in the corresponding compound-nucleus case. However, the quantitative vagueness of this expectation and the uncertainty in the experimental data make application of this consideration impossible here. The direct-interaction mechanism, of course, implies less fission competition than the compound-nucleus mechanism. This consideration speaks in its favor in this case.

Since the (α, pn) and (α, p2n) functions seem to rise at almost the same energy and since this energy is very close to the threshold for the latter reaction, the indication is that the (α, p2n) reaction is proceeding primarily by triton emission rather than by deuteron or proton emission.

It has undoubtedly struck the reader that the interpretations given here are far from unique. What has been shown, in the experimental results, is that certain effects of mass and nuclear type do exist, but explanation that is more than speculation must await a larger body of data on nuclear reactions of all types and a better state of knowledge concerning the nature of the most fundamental nuclear processes.

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APPENDIX

I. A SEARCH FOR NEGATRON EMISSION IN THE DECAY OF AMERICIUM-240

The results of closed-cycle calculations[42] of heavy-element decay energies are ambiguous concerning the relative beta-instability of Am^{240} and Cm^{240}. The calculations indicate that electron capture of Cm^{240} is exoergic by 50 kev (neglecting the electron binding energy); however, the uncertainty in this figure is probably around 100 kev.

Experimentally, Higgins[44] has searched for both negatron emission of Am^{240} and electron capture of Cm^{240} and, on the basis of his negative results, has set upper limits of 2×10^{-5} negatron emission events per Am^{240} disintegration and 5×10^{-3} electron-capture events per Cm^{240} disintegration.

In the work described herein, a sample of Am^{240} decaying at the rate of 4.6×10^{6} disintegrations/min (as determined by Nucleometer counting with the counting efficiency assumed to be 70%) was prepared by deuteron irradiation of Pu^{239} with subsequent chemical purification similar to that described in the body of this thesis. After 26 days, this sample was counted overnight in the ultra-low-background chamber of the alpha-pulse analyzer. An upper limit of 0.02 disintegrations/min was set for alpha particle emission at 6.26 Mev, the energy of Cm^{240} alpha particles. This limit corresponds to an upper limit of 1×10^{-7} for the negatron-emission-decay branching of Am^{240} decay and a lower limit of 5×10^{4} years for the partial half life for this process.

APPENDIX

II. ELECTRON CAPTURE DECAY OF AMERICIUM-238

The electron-capture decay of Am^{238} is of unusual interest among the many heavy electron-capturing nuclides because the energy available for this process is almost uniquely high (2.22 Mev) for heavy nuclides not near closed nucleon shells,[42] and the lower-level structure of the daughter nuclide has been studied (through the alternative routes of alpha decay of Cm^{242} [45] and negatron decay of Np^{238} [46, 47]) with results which are of interest in terms of the collective model of the nucleus.[48]

This nuclide was produced by bomardment of Pu^{239} with 27- and 35-Mev protons in the 184-in. synchrocyclotron. The bombardment procedure was that of Higgins.[44] The chemical procedure for isolating the americium was essentially that given in the body of this thesis for Pu^{242} targets, except that no lactate elution from a cation column was necessary because no curium was formed in these bombardments.

The decay was studied by gamma pulse analysis, using a thallium-activated sodium iodide crystal scintillation counter and a xenon-filled proportional counter, in each case coupled to a 50-channel pulse analyzer; and by gamma-gamma coincidence measurements using two thallium-activated sodium iodide crystal scintillation counters, one coupled to a single-channel pulse analyzer, the other to a 50-channel pulse analyzer. In addition, alpha pulse analysis was done to indicate the amount of Am^{237} in the samples.

Prominent gamma rays of 580 ± 25 and 980 ± 40 kev were observed in the americium fraction in all bombardments at both 27 and 35 Mev. Plotting the logarithms of the integrated photopeaks at 580 and 980 kev from bombardments at the two energies as a function of time, and--for the lower energy peak at the lower energy bombardment--making a small (less than 10%) correction for a 12-hr (Am^{239}) component, indicated a half life of 1.86 ± 0.09 hr. This value is probably more reliable than that of Higgins (2.1 hr),[44] inasmuch as the latter was determined by resolution of a three-component decay

curve in which the longer-lived components constituted about two-thirds of the initial activity.

The assignment of these high-energy gamma rays to Am^{238} rather than Am^{237} was made because the intensity of these two gamma rays relative to that of the 6.01-Mev alpha particle of Am^{237},[44] as measured in the alpha pulse analyzer, decreased by a factor of more than 20 in increasing the bombarding energy from 27 to 35 Mev, whereas their abundances relative to one another did not change within the statistical accuracy of the determination (about 3%).

The gamma spectrum in the region from 150 to 400 kev was completely obscured by the very prominent photopeaks at 225 and 275 kev belonging to Am^{239}.

Photopeaks having 1.9-hr components were also observed at 14, 18, and 102 kev. The first two of these correspond to the L_α and L_β x-rays of plutonium; the last probably includes not only plutonium K x-rays, but also gamma rays corresponding to transitions between the 146- and 44-kev levels of Pu^{238}, known from Np^{238} and Cm^{242} studies.

In order to establish the relationships between the high-energy gamma rays in the decay scheme, gamma-gamma coincidence studies were made, and in the course of these, a new gamma ray at about 1300 kev and a possible one at 340 kev were found. The coincidences observed are given in Table VI.

Table VI

Americium-238 gamma-gamma coincidences

Gate	Coincident gammas (Signal)
950 kev	950 kev 550 (340 ?)
570	1240 920 550 340
ca. 1300	540

The energy measurements of the coincident gamma rays are of necessity very crude, as the inherent certainty in the instrument is only about 4%, the counting rates were very low (of the order of three counts per minute for some of the peaks), and the short half life of the nuclide drastically limits the amount of time available for the measurements.

In interpreting these coincidence data, it is important to remember that what is actually being measured is coincidences between scintillations of certain energies, whatever their origins may be. Thus, with the gate counter set at 580 kev, one is measuring coincidences not only with the 580-kev gamma ray, but also with Compton-scattered electrons of this energy from the 980-kev gamma ray. Thus, the apparent self-coincidence of the 580-kev gamma ray can be explained as a manifestation of the 980-kev–580-kev gamma cascade. The observed relative abundances of the 980-kev-580-kev and 580-kev–580-kev coincidences are consistent with this interpretation. The apparent 580-kev–340-kev coincidence is due primarily to coincidences of the photon and electron from Compton scattering of the 980-kev gamma ray.

The decay scheme indicated by these data is given on Fig. 7. On this same figure, for comparison purposes, is the level diagram of Pu^{238} as indicated by the decay of Np^{238} and Cm^{242}.[47] (In this diagram, probably only one of the two levels indicated by dotted lines is real.) It should be noted that many transitions are observed between the band of levels at ground and that near 1 Mev in the decay of Np^{238}; there are at least four and more likely five gamma transitions of energies between 927 and 1029 kev, and these would appear as a single photopeak on equipment such as that used in the study described here. Therefore it is possible and extremely probable the 980-kev gamma ray of Am^{238} is also complex, and, indeed, it is likely that all the observed gamma rays are complex and all the levels indicated are really bands of levels. The resolution of these gamma rays and the consequent determination of the individual energy levels requires beta spectroscopy of conversion

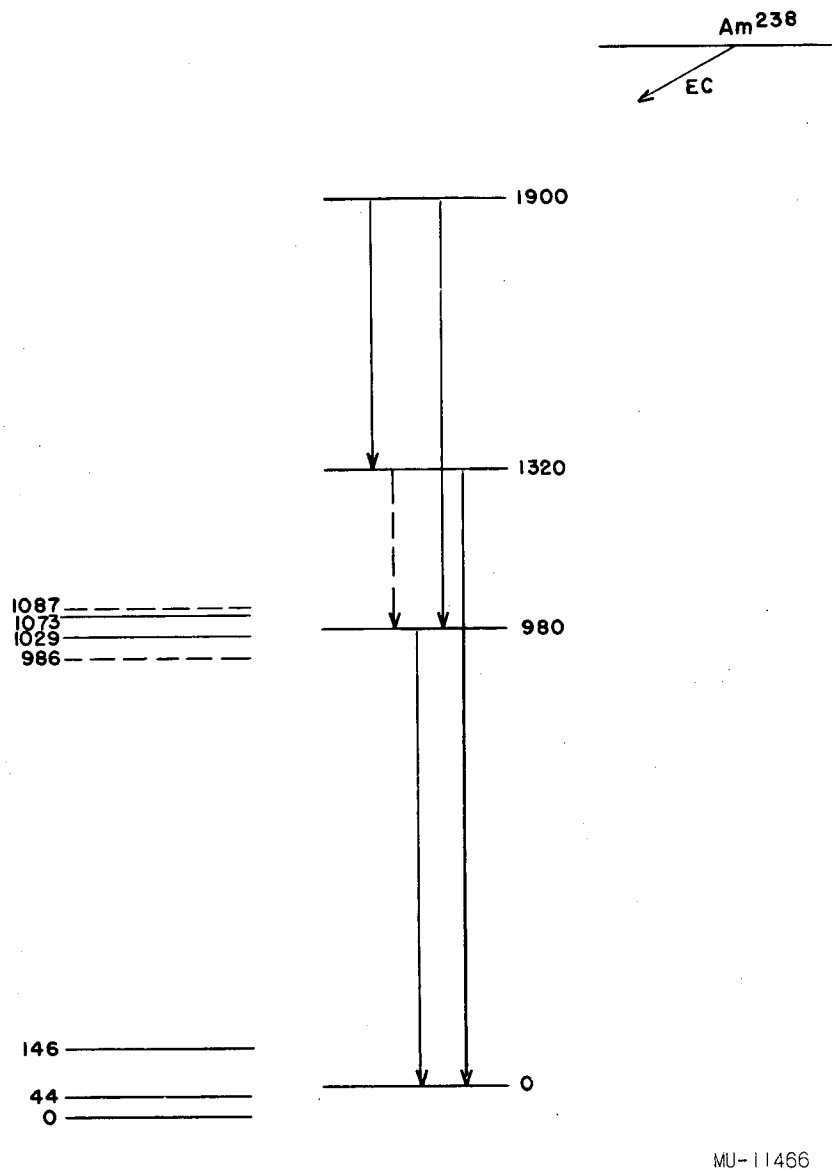

Fig. 7. Am^{238} decay scheme. Levels of Pu^{238} as deduced from alpha decay of Cm^{242} and negatron decay of Np^{238} are given for comparison (see Ref. 47).

electrons or photoelectrons, which in turn requires samples of considerably greater intensities than can be made by the methods used in this study.

The rather imprecise character of this work makes any extensive attempt at interpretation meaningless; however, it is perhaps worth pointing out that if the band of levels around 1 Mev corresponds to a first vibrational excitation according to the collective model for the nucleus, a second vibrational band is expected at a little less than twice this energy.[48] This is what has been found. However, the interpretation of the band around 1 Mev has been discussed by Rasmussen and his co-workers[47] and they have concluded on the basis of gamma lifetimes and beta $\underline{ft}$ values that this band does not constitute a vibrational band, despite the facts that the spins and parities are what is expected for a gamma vibrational band and that the energy of this band is of the order expected from theory.

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