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SPALLATION REACTIONS OF PLUT0NIUM-240 WITH HELIUM IONS AND PLUTONIUM-242 WITH DEUTERONS

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Author

Eads, Donald L.

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SPALLATION REACTIONS OF PLUTONIUM-240 WITH HELIUM
IONS AND PLUTONIUM-242 WITH DEUTERONS

Donald L. Eads

(Master's Thesis)

January 1959

SPALLATION REACTIONS OF PLUTONIUM-240 WITH HELIUM

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SPALLATION REACTIONS OF PLUTONIUM-240 WITH HELIUM IONS AND PLUTONIUM-242 WITH DEUTERONS

Donald L. Eads

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

January 1959

ABSTRACT

Radiochemical studies were made of the $Pu^{240}(\alpha,xn)$ products produced by bombarding Pu^{240} with 22- to 46-Mev helium ions. Excitation functions are given for the $(\alpha,2n)$ Cm^{242}, $(\alpha,3n)$ Cm^{241}, and $(\alpha,4n)$ Cm^{240} reactions. The $Pu^{240}(\alpha,xn)$ reactions are compared to those of the other plutonium isotopes with regard to their dependence on the fission parameter Z^2/A and their agreement with theoretical excitation functions. The latter were calculated by employing a modified Jackson-type calculation with a nuclear temperature of 1.50 Mev and an R_o of 1.5 x 10^{-13} cm.

The $Pu(\alpha,4n)$ reactions were used to evaluate the ratio of Γ_n/Γ_t for the curium isotopes.

The $Pu^{242}(d,2n)Am^{242,242m}$ reactions were investigated by radiochemical methods using 11- to 24-Mev deuterons, and the excitation functions are given.

SPALLATION REACTIONS OF PLUTONIUM-240 WITH HELIUM
IONS AND PLUTONIUM-242 WITH DEUTERONS

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I. INTRODUCTION

In the field of nuclear reactions, spallation-fission competition has attracted considerable attention.[1-6] Fission, the splitting of a heavy nucleus into two or more medium-weight fragments, usually accompanied by the emission of neutrons, accounts for about 90% of the reactions in the heavy-element region ($Z \geq 90$). Spallation, the emission of a various number of nucleons or light nuclei (e.g., $_1d^2$, $_1t^3$, or $_2He^4$), is divided into compound-nucleus and direct-interaction reactions.

Bohr's original compound-nucleus theory[7] asserts that the nuclear reaction consists of two stages:

1. The formation of the compound system C,

$$A + a = C*$$
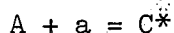
(where A and a are the target nucleus and projectile respectively, and C* is the excited compound nucleus).

2. The disintegration of the compound system into the products of the reaction

$$C* = B + b$$
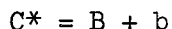
(where B and b are the product nucleus and emitted particle or particles respectively).

Bohr assumed that

1. The compound nucleus has a lifetime that is long (about $10^{-16\pm3}$ second) compared with the time for a proton to cross the nucleus (about 10^{-22} second).

2. The products of the reaction are independent of the manner in which the compound nucleus is formed; the mode of disintegration of the compound system is dependent only upon its energy.

The predictions of this theory have been verified by comparing the excitation functions of isotopes bombarded with different projectiles to form the same compound nucleus with the same excitation energy.[8-10]

At bombarding energies below approximately 50 Mev, nuclear reactions are often of the compound-nucleus type. The compound system dissipates its excitation energy by the evaporation of nucleons. At energies above 50 Mev, Serber suggested that the incident particle might interact with nucleons in the struck nucleus in the same way as it interacts with free, unbound nucleons.[11] The direct interactions, occurring within the nucleus, knock out one or more nucleons, often leaving little of the energy of the incoming particle within the nucleus as excitation energy. Pickup and stripping reactions are other types of direct interactions. The direct-interaction mechanism is responsible for the large cross sections for charged-particle emission at lower energies.[4,12,13]

This paper is composed of two parts: (a) The comparison of the $Pu^{240}(\alpha,xn)$ reactions with those of the other plutonium isotopes with respect to the dependence of the cross sections on the fission parameter Z^2/A; the neutron emission ratios, Γ_n/Γ_t, of the curium nuclei involved in the reactions; and agreement with theoretical excitation functions; (b) The determination of the excitation function for the production of Am^{242m} formed in the $Pu^{242}(d,2n)$ reaction and, for several bombarding energies, the yield of the Am^{242} isomer.

II. METHODS AND APPARATUS

A. Plutonium-240 with Helium Ions

1. Target Assembly

The recoil technique used in the determination of the Pu^{240} (α,xn) cross sections is similar to that described by Harvey et al.[14] A 2-mil gold foil, on which the plutonium was electroplated, was mounted so that the beam of helium ions passed through the foil before striking the plutonium target material. The target should be thin, so as to reduce to a minimum the scattering and absorption of the reaction products by the target material. A 0.1-mil gold foil was placed about 0.8 cm behind the target, in the evacuated chamber, to catch the recoiling products. The recoil block, including collimator, degrading foils, target, and catcher foil, is illustrated in Fig. 1. The same equipment was also used, and described by Vandenbosch.[10]

The unknown Pu^{240}(α,xn) cross sections were determined by using the Cm^{244}(α,2n)Cf^{246} reaction,[15] of known cross section, as a monitor. The recoil efficiencies of all the reaction products were assumed to be the same.

2. Target Preparation

The target was prepared by the electrodeposition of 20 micrograms of plutonium (87.17% Pu^{240}, 12.20% Pu^{239}, and 0.583% Pu^{241} by weight) and 0.2 microgram of Cm^{244} per cm^2 from a 0.4 $\underline{M}$ ammonium oxalate solution onto a 2-mil gold foil. A current of 100 ma per cm^2 at less than 4 volts resulted in better than 80% yields. After the plating the plutonium hydroxide is carefully flamed to convert it to the oxide. The electrodeposition method used was developed by Hufford and Scott[16] and modified by Glass.[17]

3. Helium-Ion Beam

The target was mounted in the recoil block and bombarded with helium ions from the Crocker Laboratory 60-inch cyclotron. The beam of ions has a maximum range of 226 to 232 mg/cm^2 of aluminum,[18] or an energy

CATCHER-FOIL POSITIONER

TARGET

ALUMINUM
WATER-COOLED

ABSORBER

GRAPHITE
COLLIMATOR

WATER-COOLED
FARADAY CUP

He ION BEAM

CATCHER-FOIL
HOLDER

He COOLING

VACUUM
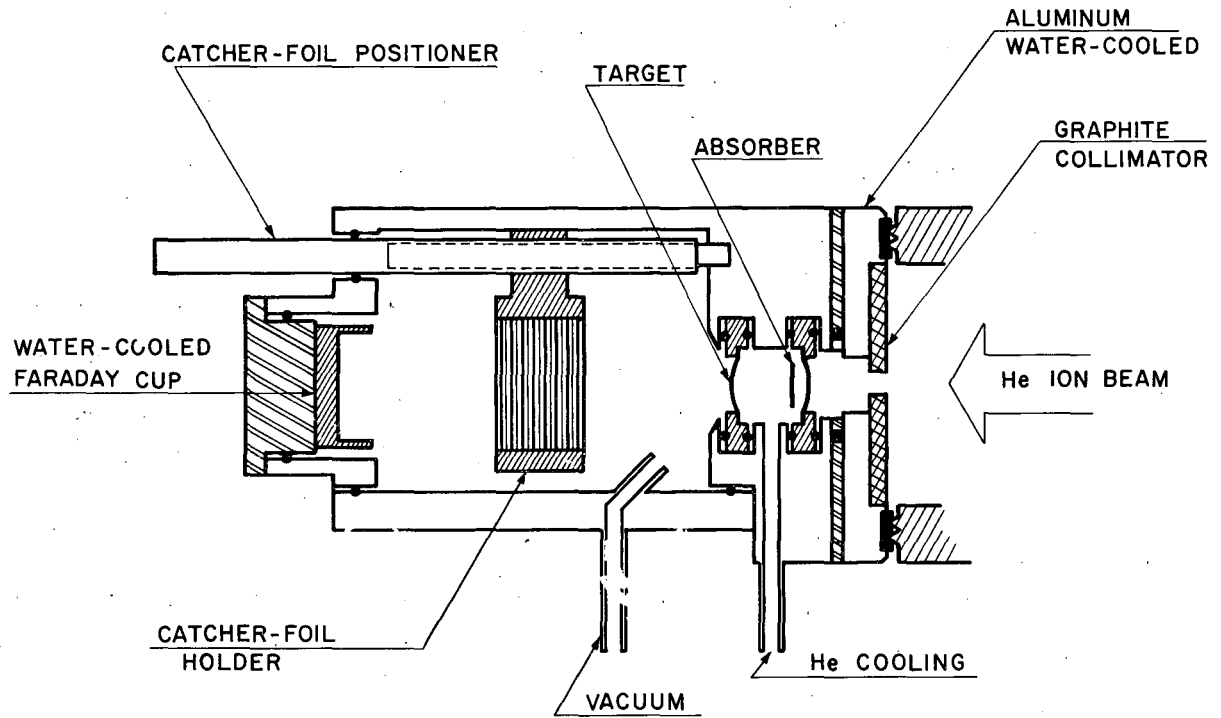
MU-13709

Fig. 1. Recoil-target assembly used in measuring the
$Pu^{240}(\alpha,xn)$ cross sections.

of 48 ± 0.5 Mev. The range-energy curves of Aron et al.[19] were used to determine the thickness of aluminum required to degrade the beam to the desired energy. Absorbers were weighed and the surface area measured to determine their thickness, with vernier calipers used as a check.

The beam intensity, known to 0.5%, was never allowed to exceed 6 microamperes, to prevent excessive heating of the target. The target foil was cooled by a jet of helium gas. A Faraday cup, located behind the recoil catcher, was used to determine the quantity of helium ions that passed through the target. A collimator insured that all the beam registered passed through the target.

4. Chemical Procedures

Dissolution of the gold recoil catchers. The 0.1-mil gold recoil catcher was dissolved in 12 $\underline{N}$ HCl with just enough HNO_3 added to complete the dissolution. The plutonium, which was knocked out of the target and caught on the catcher foil, is oxidized to the IV state.

Curium and californium. The resulting solution was evaporated to dryness. The residue was dissolved in two drops of 12 $\underline{N}$ HCl (+ 0.1 $\underline{N}$ HNO_3) and passed through a column of Dowex 1 anion resin 3 mm in diameter and 5 cm long. 12 $\underline{N}$ HCl (+ 0.1 $\underline{N}$ HNO_3) was used to elute the isotopes of curium and californium from those of plutonium and gold which remained on the column. The curium-californium fraction was evaporated to dryness, transferred to a plating cell with 5 $\underline{N}$ NH_4Cl (pH of 4.8), and electro-deposited on a 2-mil platinum foil for alpha counting. Yields of better than 90% could be obtained by using 1 amp per cm^2 at 4 volts.

Plutonium. The plutonium was desorbed by eluting with 12 $\underline{N}$ HCl made 0.1 $\underline{N}$ in HI to reduce the plutonium.

5. Counting Instruments

Gross alpha counters. Gross alpha counting of samples was done in an argon-filled ionization chamber with an accompanying scaling circuit. This instrument has a counting efficiency of 52%. A low solid-angle counter was used for very active samples of more than 10^6 dpm. The counting efficiency of this instrument was standardized regularly.

Differential pulse-height analyzer. The alpha particles from the reaction products were analyzed on a 48-channel alpha-pulse analyzer.[20] Energy calibration was made by means of samples of known alpha-emitting substances and a pulse simulator.

6. Calculation of Cross Sections

The cross section σ for a nuclear reaction is defined by the equation

$$\sigma = N/nIt,$$

where N is the number of product nuclei formed during the bombardment, n the number of target nuclei per cm^2, and It the number of incident particles passing through the target (I being the beam intensity and t the duration of the bombardment).

The cross sections for the $Pu^{240}(\alpha,xn)$ reactions were related to the known $Cm^{244}(\alpha,2n)Cf^{246}$ cross section at the same energy by the expression

$$\frac{\sigma\ Pu^{240}(\alpha,xn)}{\sigma\ Cm^{244}(\alpha,2n)} = \frac{A\ Cm^{(244-xn)}}{A\ Cf^{246}} \times \frac{t_{1/2}\ Cm^{(244-xn)}}{t_{1/2}\ Cf^{246}} \times \frac{n\ Cm^{244}}{n\ Pu^{240}},$$

where A is the activity of the product nuclei (assuming none has decayed), $t_{1/2}$ is the half life of the product nuclei, and all other terms maintain their meaning. The half lives and alpha decay energies were taken from a table of isotopes.[21]

If the product has a short half life, then A must be expresses as

$$A = (\overline{d/m})\ f_1\ f_2\ f_3,$$

where $(\overline{d/m})$ is the average rate of decay of the product measured with the pulse analyzer, f_1 the correction for product decay during the bombardment, f_2 the correction for product decay from the end of the bombardment to the beginning of the pulse analysis, and f_3 the correction for product decay during the pulse analysis. The f_1 and f_3 corrections are of the same form,

$$f = \frac{t}{t_{1/2}} \times \frac{0.693}{1-e^{-\lambda t}} \quad ,$$

where t is the length of the bombardment for f_1 or length of pulse analysis for f_3, and $t_{1/2}$ and λ are the half life and decay constant respectively for the product nuclei under consideration. The f_2 correction is given by

$$f_2 = e^{-\lambda(\Delta t)} \quad ,$$

where Δt is the time interval between the end of the bombardment and the beginning of the pulse analysis and is positive.

B. Plutonium-242 with Deuterons

1. Target Assembly

The microtarget assembly used in this investigation is illus- trated in Fig. 2. Each target was covered with a 0.1-mil gold foil to catch any product nuclei recoiling in a direction backward with respect to the beam. Two targets and aluminum energy-degrading foils were mounted in the target assembly.

2. Target Preparation

The Pu^{242} targets were prepared by the electrodeposition of 2 to 10 micrograms of plutonium (99.84% Pu^{242}, 0.15% Pu^{240}, and 0.01% Pu^{238} by weight) per cm^2 from a 5 $\underline{N}$ NH_4Cl solution, having a pH of 4.8, onto a 2-mil gold foil. A current of 1 amp per cm^2 at less than 4 volts resulted in better than 80% yields.

3. Deuteron Beam

The mounted targets were bombarded with deuterons accelerated by the Crocker Laboratory 60-inch cyclotron. The deuteron beam had a maximum range of 457 to 468 mg/cm^2 of aluminum,[18] corresponding to a maximum energy of 24 Mev.[19]
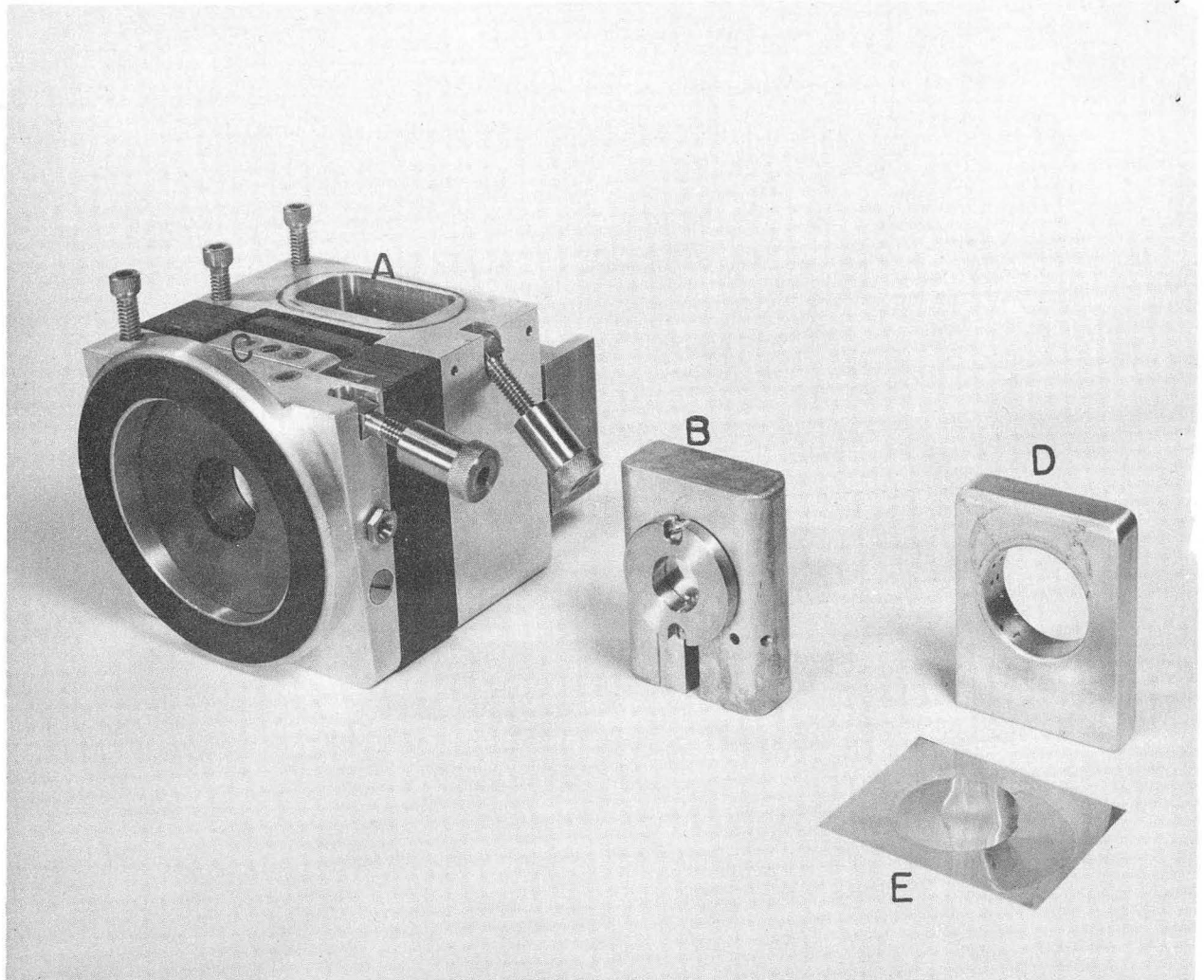
ZN-1412

Fig. 2. Microtarget assembly used in measuring the
$Pu^{242}(d,2n)$ cross sections. A, microtarget slot;
B, microtarget; C, collimator; D, foil holder;
and E, degrading foil.

4. Chemical Procedures

Dissolution of target. The Pu^{242} target and gold cover foil were dissolved in aqua regia and Am^{241} yield tracer added.

Plutonium. The gold was extracted by agitation of its solution with an equal volume of ethyl acetate.[22] The organic layer was removed and washed twice with an equal volume of 6 $\underline{N}$ HCl. The aqueous phase and the two washes were combined and evaporated to dryness. The residue was dissolved in 12 $\underline{N}$ HCl (+0.1 $\underline{N}$ HNO_3) and passed through a column of Dowex 1 anion resin 3 mm in diameter and 5 cm long. The plutonium, which was oxidized to the IV state in the aqua regia, was adsorbed, and the curium-americium fraction was eluted along with the rare earths and other fission products. The plutonium was desorbed with 12 $\underline{N}$ HCl made 0.1 $\underline{N}$ in HI.

Americium and curium. Americium and curium were separated from the fission products by coprecipitation with LaF_3; 0.4 mg of lanthanum carrier was added to the americium-curium fraction, and the resulting solution made 4 $\underline{N}$ in HF. The precipitate was washed with water containing a trace of HF and dissolved in 12 $\underline{N}$ HCl containing a trace of boric acid. NH_4OH was then added to precipitate the hydroxides, which were dissolved with alcoholic HCl (80% HCl and 20% ethanol saturated with HCl gas). This solution was passed through a Dowex-50 cation resin eluting with alcoholic HCl.[23] The rare earths were adsorbed while the americium and curium isotopes passed through. The solution containing americium and curium was evaporated to dryness. The residue was dissolved and transferred to an electroplating cell with 5 $\underline{N}$ NH_4Cl having a pH of 4.8 A current of 1 amp per cm^2 and a potential of 4 volts were applied to the cell.

Americium and curium separation. In order to determine the Am^{242} yield, the target was stored until all the 16-hour Am^{242m} isomer had decayed. The americium-curium fraction from the alcoholic HCl column was then evaporated to dryness. The residue was dissolved in 50λ of 0.4 $\underline{M}$ alpha-hydroxy-isobutyric acid (But.) with a pH of 4.38 and transferred to a column of Dowex-50 cation resin. The resin had a settling rate of 0.5 to 1.0 cm per minute. The tube, which contained the activity, was washed with two 30λ portions of the 0.4 $\underline{M}$ But. solution and transferred to the resin, washing

down the sides of the column. Trichloro-ethylene (boiling point $87^{\circ}C$)
was condensed in a jacket surrounding the column to maintain a temper-
ature of $87^{\circ}C$.

The actinide elements were eluted with the 0.4 M But. with a
flow rate of one drop per minute. A drop size of about 13 λ was obtained
with a small-bore platinum-tipped column. Any detectable amount of the
Cm^{242}, produced in the decay of the Am^{242m}, was removed by passing the
americium fraction through a column a second time. Detailed operational
procedures for this column are given in Reference 23. Two milliliters of
HNO_3 was added to the americium solution, and the mixture was evaporated
to dryness. The residue was dissolved in 5 N NH_4Cl with a pH of 4.8 and
the americium electrodeposited in the same way as described in the
preceding section.

5. Calculation of the Cross Sections

In this experiment the cross section was determined as a function
of the daughter activity Cm^{242} formed from the decay of the Am^{242} produced
in the nuclear reaction. If N_1^O is the number of Am^{242} nuclei and N_2^O the
number of Cm^{242} nuclei at the end of the bombardment, the activity of the
Cm^{242} is

$$-\frac{dN_2}{dt} = \lambda_2 N_2 = \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} N_1^O (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^O \lambda_2 e^{-\lambda_2 t} , \qquad (1)$$

where t is the time since the end of the bombardment and the λ's are the
corresponding decay constants.

The number of Am^{242} nuclei N_1^O at the end of the bombardment is

$$N_1^O = n_{Pu} \sigma It_b \left[\frac{t_{1/2}}{t_b} \times \frac{1 - e^{-\lambda_1 t}}{0.693} \right] , \qquad (2)$$

where n_{Pu} is the number of Pu^{242} target nuclei per cm^2, It_b (current times
bombardment time) the number of projectiles that pass through the
target, σ the cross section for the production of Am^{242} in units of area,
and the content of the bracket, the fraction of the product which does not

decay during the bombardment. The term $(n_{Pu} \sigma It_b)$ is the number of product nuclei formed in the nuclear reaction, assuming none decayed during the bombardment. The number of Cm^{242} daughter nuclei N_2^O at the end of the bombardment (assuming none decayed) is equal to the number of atoms of the Am^{242} product that decay during the bombardment:

$$N_2^O = n_{Pu} \sigma It_b - N_1^O = n_{Pu} \sigma It_b \left(1 - f\right) , \tag{3}$$

after substituting Eq. (2) for N_1^O.

The daughter activity at time t after the bombardment can then be expressed, after substituting Eqs. (2) and (3) into Eq. (1), as

$$-\frac{dN_2}{dt} = \lambda_2 n_{Pu} \sigma It_b \left[\frac{\lambda_1}{\lambda_2 - \lambda_1} (f) (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + (1-f) e^{-\lambda_2 t} \right], \tag{4}$$

where $f = (t_{1/2}/t_b) \times (1 - e^{-t_b})/0.693$.

Solving for σ in Eq. (4), one obtains

$$\sigma = \frac{-dN_2}{dt} \Big/ \left\{ \lambda_2 N_{Pu} It_b \left[\frac{\lambda_1}{\lambda_2 - \lambda_1} (f) (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + (1-f) e^{-\lambda_2 t} \right] \right\} . \tag{5}$$

Corrections that have been neglected in this experiment are:

The decay during the 4-hour bombardment of the daughter Cm^{242} ($t_{1/2}$ of 162 d).

The transmutation of the product Am^{242} and daughter Cm^{242} by the projectiles.

The small cross section of about 20 mb, the small number of these nuclei available, and the length of the bombardment make these corrections negligible. The decay that occurs during sample counting was neglected in the determination of the Am^{242m} because the counting rate of Cm^{242} permitted a very short counting time compared to the half life involved.

III. RESULTS

$Pu^{240}(\alpha,xn)$ Cross Sections

The cross sections for the reactions Pu^{240} $(\alpha,2n)$ Cm^{242}, $(\alpha,3n)$ Cm^{241}, and $(\alpha,4n)$ Cm^{240}, determined by comparing the relative yields of Cm^{242}, Cm^{241}, and Cm^{240} respectively with the yield of the Cm^{244} $(\alpha,2n)$ Cf^{246} reaction, are given in Table I and illustrated in Fig. 3. The Pu^{240} (α,xn) cross sections were corrected for the production of the same products from the Pu^{239} (α,xn) reactions.[1]

Sources of Error

The cross sections of the Cm^{244} $(\alpha,2n)$ monitor reaction were known to 15%.[15] The uncertainty in the target thickness was 10%. This was determined by alpha-counting equal sections of the target through a hole in an absorber. The calculated errors of the counting instruments were verified throughout the experiment and did not vary more than 1 or 2%. The error introduced by any difference in the recoil efficiency or range between the Cf^{246} and the products being measured was estimated to be small. The error involved in the determination of the target area is not encountered in the recoil technique. The total calculated error for these excitation functions was ± 18%.

$Pu^{242}(d,2n)$ Cross Sections

The cross sections for the Pu^{242} $(d,2n)$ reactions are given in Table II and illustrated in Fig. 4.
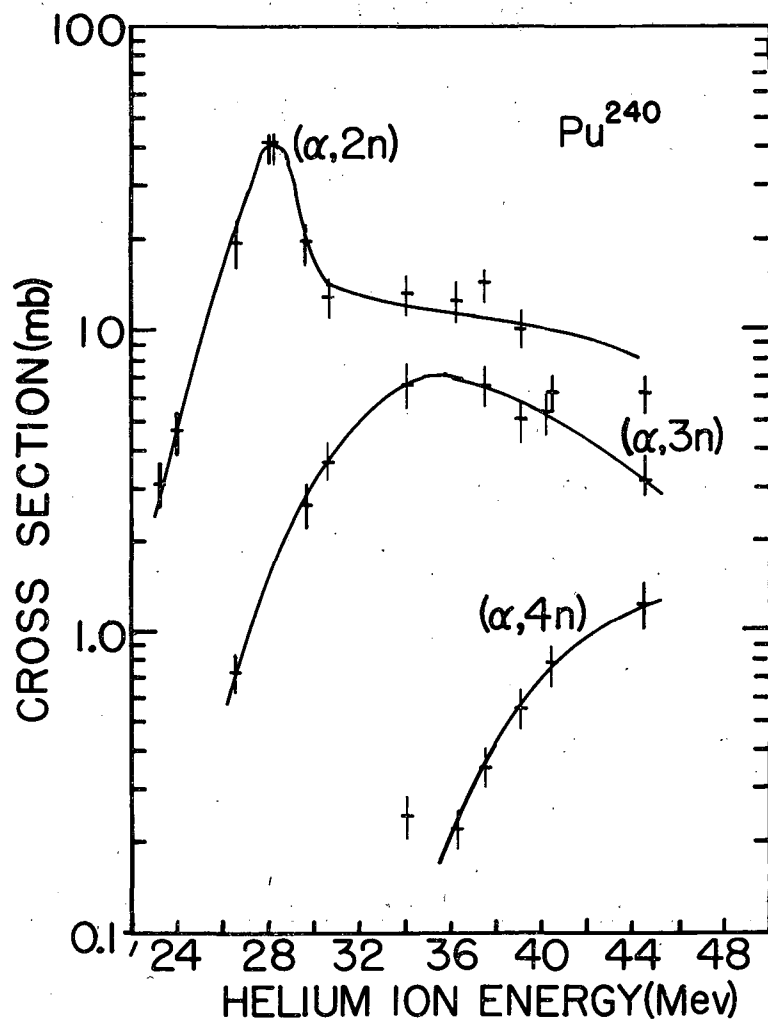
Sources of Error

Errors encountered in this experiment included: yield determination, 10%; counting statistics, 4 to 15%; and inherent errors in the detectors, 1 to 2%. The total error for the production of Am^{242m} was estimated as ± 20%.

The largest error in the determination of the cross sections for the production of Am^{242} with a half life of 100 years was due to the low counting rate of the daughter Cm^{242} and to background difficulties. The estimated total error for these cross sections was ± 40%.

Table I

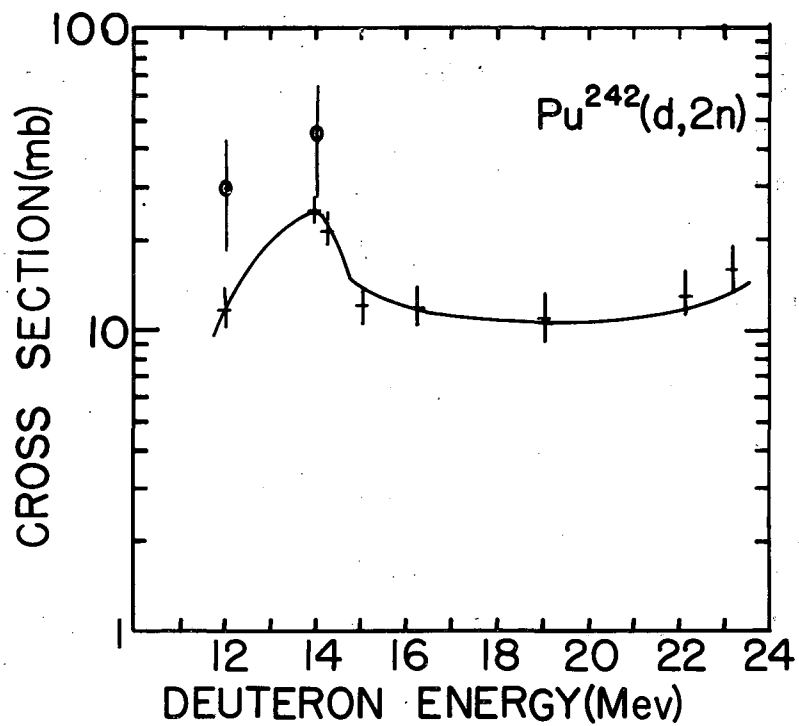| E (Mev)	$(\alpha,2n)Cm^{242}$	$(\alpha,3n)Cm^{241}$	$(\alpha,4n)Cm^{240}$
23.2	3.1		
24.0	4.5		
26.5	18.8	0.72	
28.0	41.1 (40.9)		
29.6	19.1	2.6	0.0013
30.6	12.7	3.6	0.012
34.0	13.1	6.5	0.24
36.2	12.4		0.22
37.4	14.3	6.5	0.35
39.0	10.0	5.0	0.55
40.4	6.1	5.3	0.77
44.5	6.1	3.3	1.20

$Pu^{240}(\alpha,xn)$ cross sections in millibarns

Fig. 3. Excitation functions for the $Pu^{240}(\alpha,2n)Cm^{242}$, $(\alpha,3n)Cm^{241}$, and $(\alpha,4n)Cm^{240}$ reactions.

Table II

$Pu^{242}(d,2n)$ cross sections (in millibarns)		
E (Mev)	$(d,2n)Am^{242m}$	$(d,2n)Am^{242}$
12.0	11.9	30
13.9	25.1	45
14.4	21.1	
15.0	12.2	
16.4	12.0	
19.0	11.1	
22.2	13.1	
23.3	16.0	

Fig. 4. The excitation function for the $Pu^{242}(d,2n)Am^{242m}$ reaction and two cross sections (0) for the $Pu^{242}(d,2n)Am^{242}$ reaction.

IV. DISCUSSION

A. A Model for Competition Between Neutron
Evaporation and Fission in the Heaviest Elements

Jackson has developed a schematic model for (p,xn) reactions in heavy nuclei considering both the direct interaction and compound-nucleus processes.[24] The Monte Carlo method[25] was used to evapuate the relative probabilities of the prompt processes. After the emission of the prompt nucleons, the residual nucleus loses one or more nucleons by evaporation, losing energy until the excitation energy is below the binding energy of the next nucleon. No Monte Carlo calculations have been made for incident helium ions; therefore, the contribution of direct-interaction processes has been ignored in this work.

Jackson's assumptions are

(a) The emission probabilities for charged particles are very small compared with the emission probability for neutrons, because of the high Coulomb barrier in heavy nuclei.

(b) At excitations of the order of 15 Mev or higher the probability of emission of a neutron with energy ϵ is approximately proportional to $\epsilon \exp(-\epsilon/T)$, where T is the nuclear temperature.

(c) The nuclear temperature is assumed to be independent of the exciation energy.

The probability that a nucleus with excitation energy E* will emit exactly x neutrons is given by

$$P(E^*,x) = I(\Delta_x, 2x-3) - I(\Delta_{x+1}, 2x-1), \tag{6}$$

where $I(z,n)$ is Pearson's incomplete gamma function:

$$I(z,n) = (1/n!) \int_0^z x^n e^{-x} dx, \tag{7}$$

Δ_x is the energy, in units of T, above the threshold for the emission of x neutrons,

$$\Delta_x = (E^* - \sum_1^x B_i)/T, \tag{8}$$

where $\sum_{i}^{x} B_i$ is the sum of the binding energies of the x emitted neutrons and all of the other terms maintain their meaning. Then Δ_{x+1} is the energy above the threshold for the emission of x+1 neutrons,

$$\Delta_{x+1} = (E^* - \sum_{1}^{x+1} B_i)/T, \tag{9}$$

where $\sum_{1}^{x+1} B_i$ is the sum of the binding energies of the x emitted neutrons plus that of the next neutron.

Vandenbosch et al.[5] have modified the Jackson method to account for the fission contribution when the excitation energy of a nucleus is above the fission threshold but below the neutron binding energy of the last neutron emitted in the reaction. Thus Δ_x^f is substituted for Δ_{x+1}, in Eq. (6) for P (E*,x),

$$\Delta_x^f = (E^* - \sum_{1}^{x} B_i - E_{th})/T, \tag{10}$$

where E_{th} is the fission threshold.[26]

To account for the fission competition at each stage of neutron evaporation, neutron-emission branching ratios are introduced into the equation. The cross section for an (α,xn) reaction is then given by the equation

$$\sigma\,(\alpha,xn) = \sigma_c\,(E_\alpha)\,G_{n1}\,G_{n2}\,\cdots\,G_{nx}\,P\,(E^*,x)\,T\,(E^*), \tag{11}$$

where σ_c is the cross section for forming the compound nucleus at the bombarding helium-ion energy (several sources are available for the σ_c function;[27-29] Weisskopf's values[29] were used in these calculations). The term P (E*,x) is the probability for the occurrence of the reaction, T (E*) the probability that the emitted particle will penetrate the potential barrier (T = 1 for neutrons), and the G_n terms the neutron emission branching ratios of the nuclei involved in reaching the final product.

We have $\qquad G_n = \Gamma_n/\Sigma\,\Gamma_i,$ $\qquad\qquad\qquad$ (12)

where Γ_n is the neutron level width and $\Sigma\,\Gamma_i$ is the sum of all the level

widths of the possible de-excitation modes. Because the probability of charged-particle emission is negligible in the heavy-element region, we have

$$\Sigma \, \Gamma_i \; = \; \Gamma_n + \Gamma_f \; ,$$

where Γ_f is the fission level width.

B. Evaluation of the Neutron-Emission Branching Ratios G_n for the Curium Isotopes

Vandenbosch[10] and co-workers[5,26] have devised a method for calculating individual Γ_n/Γ_f values. The assumptions on which their method is based are

(a) Γ_n/Γ_f is independent of the excitation energy for excitation energies well above the neutron-emission threshold.[1,30]

(b) Γ_n/Γ_f for even-even nuclei is twice as great as Γ_n/Γ_f for even-odd nuclei.[27]

(c) Aside from even-even and even-odd effects, there is a general trend for Γ_n/Γ_f to vary with mass number:

   (i) Γ_n increases with decreasing neutron-binding energy.

   (ii) Γ_f decreases with decreasing Z^2/A or increasing A.

Using these assumptions, Vandenbosch et al.[5] derived a formula for calculating the individual Γ_n/Γ_f values for the xth neutron leaving the compound nucleus,

$$\left(\frac{\Gamma_n}{\Gamma_f} \right)_x \; = \; \frac{1.93a}{(1.3)^x} \; \frac{\bar{G}_n}{1-\bar{G}_n} \; ,$$

where a equals $\sqrt{2}$ for even-even nuclides and $1/\sqrt{2}$ for even-odd nuclides.

The mean neutron-emission branching ratio is defined by the expression

$$\bar{G}_n^4 \; = \; G_{n1} \, G_{n2} \, G_{n3} \, G_{n4} \; = \; \frac{\sigma \, (\alpha,4n) \; pk}{1200 \; mb} \; .$$

Vandenbosch[10] did not use σ_c in the calculation of $\overline{G}_n$ for the $(\alpha, 4n)$ reaction, which is assumed to be wholly a compound-nucleus reaction. He used 1200 mb as the cross section for the $(\alpha, 4n)$ reaction at its peak if fission were not competing. This is an estimated value obtained from the $(\alpha, 4n)$ excitation functions of the lead isotopes.[31]

The neutron-emission branching ratio for a particular curium isotope is then

$$G_n = \frac{(\Gamma_n/\Gamma_f)}{1 + (\Gamma_n/\Gamma_f)} \, ,$$

where Γ_n/Γ_f is the corresponding ratio of the neutron-level width to the fission-level width for the isotope under consideration.

C. Remarks on the Theoretical Calculations and Some Comparisons of the Pu(α, xn) Reactions

The two parameters that must be selected to proceed with the Jackson-type calculations are the nuclear radius R_0, as related to σ_c, and the nuclear temperature T. The $(\alpha, 2n)$ reaction occurs in the region where the slope of the compound-nucleus-formation cross section σ_c versus E_α is greatest and is therefore more sensitive to the selection of R_0. This offers a good method, in elements in which direct interaction is not so prominent, to determine the appropriate R_0 for the calculation of the remaining (α, xn) excitation functions. Experiments[1,10,32] in the transuranium elements led to the selection of 1.5×10^{-13} cm for R_0. The effect of increasing the nuclear temperature is to increase both the value of the peak cross section and the helium-ion energy to which it corresponds. The best fit was obtained by using a nuclear temperature of 1.50 Mev.

The theoretical Pu(α, xn) excitation functions calculated by the modified Jackson method are illustrated in Fig. 5. The experimental cross sections for Pu[238], Pu[239], and Pu[242],[1] and Pu[240] are presented for comparison. The modified P(E*,x) reduces the high-energy side of the
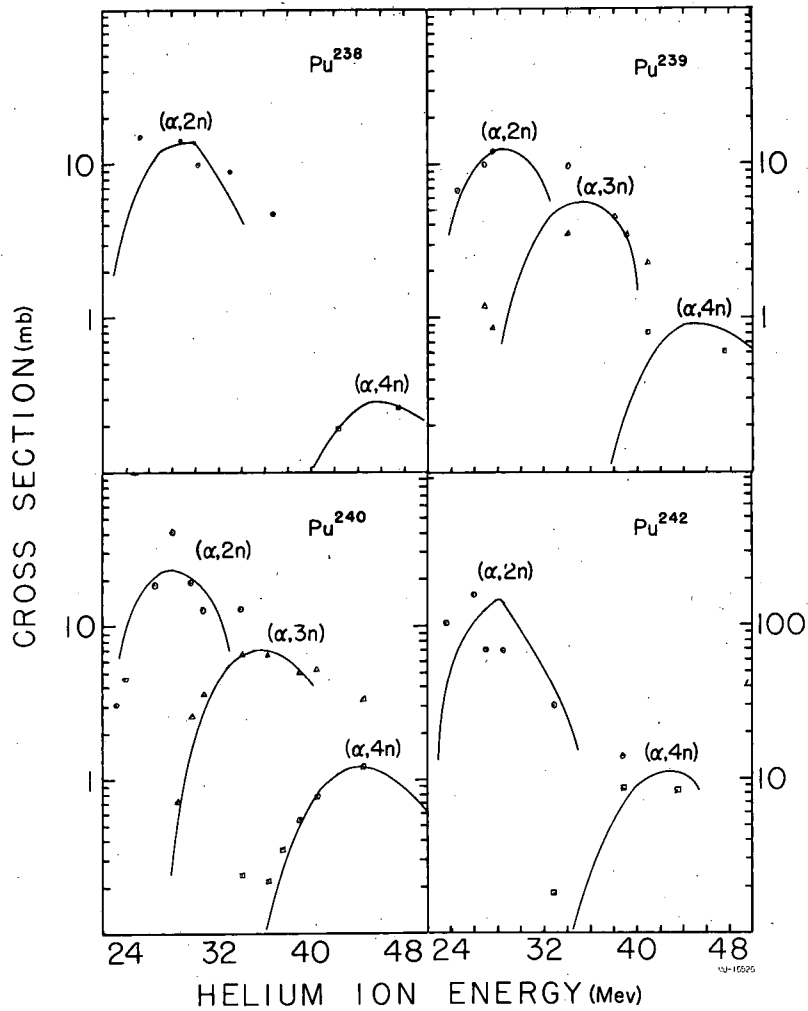
Fig. 5. Theoretical excitation functions (solid lines)
and experimental cross sections [0 = σ (α,2n),
△ = σ (α,3n), and = σ (α,4n)] for the (α,xn)
reactions of Pu^{238}, Pu^{239}, Pu^{240}, and Pu^{242}.

(α,xn) curves, yielding better agreement with experiment in the heavy-element region.

The theoretical Pu(α,xn) excitation functions are only quali-tative because of the assumptions required to make these calculations. The agreement between the theoretical and experimental cross sections in the nonfission region is illustrated in Fig. 6; the Bi^{209}(α,xn) ex-citation functions[33] were used as an example. A nuclear temperature of 1.35 Mev and an R_0 of 1.5 x 10^{-13} cm were used. Better agreement was obtained because the direct-interaction contribution is negligible and the mean neutron-emission ratio is unity in each case. The unmodified Jackson-type calculation was therefore employed. The neutron-binding energies and masses of the nuclides encountered in these calculations were taken from Reference 34.

The variation of the peak Pu(α,xn) cross sections with Z^2/A is illustrated in Fig. 7. Two factors contribute to this variation:

(a) The neutron-binding energy of the compound nuclei involved in the reaction generally decreases as A increases, and

(b) Fission competes less favorably as A increases or Z^2/A decreases. The net result of these two factors is a rapid increase in the ratio of Γ_n to Γ_f with increasing A. A summary of the Γ_n to Γ_f ratios and the corresponding G_n values for the curium isotopes 239 through 246, obtained from the Pu(α,xn) peak cross sections, is given in Table III. The terms not involved in the reaction under consideration were extrapolated from the values obtained by using the method outlined in Section B. The extrapolated values agree remarkably well with those obtained from ex-perimental results.

The variation with Z^2/A of the product of the G_n values ($\overline{G}_n^4$) of the curium nuclei involved in reaching the (α,4n) reaction product is shown in Fig. 7 for comparison. The G_n values obtained from the Pu^{238}(α,4n) peak cross section were used to evaluate the $\overline{G}_n^4$ value for the other Pu(α,4n) reactions. This eliminated the dependence of the value on its corresponding Pu(α,4n) cross section. The slopes of the ($\overline{G}_n^4$) line and the (α,4n) line are the same, within the accuracy of the calculations, indicating that the value of σ_c P(E^*,x) should be the same
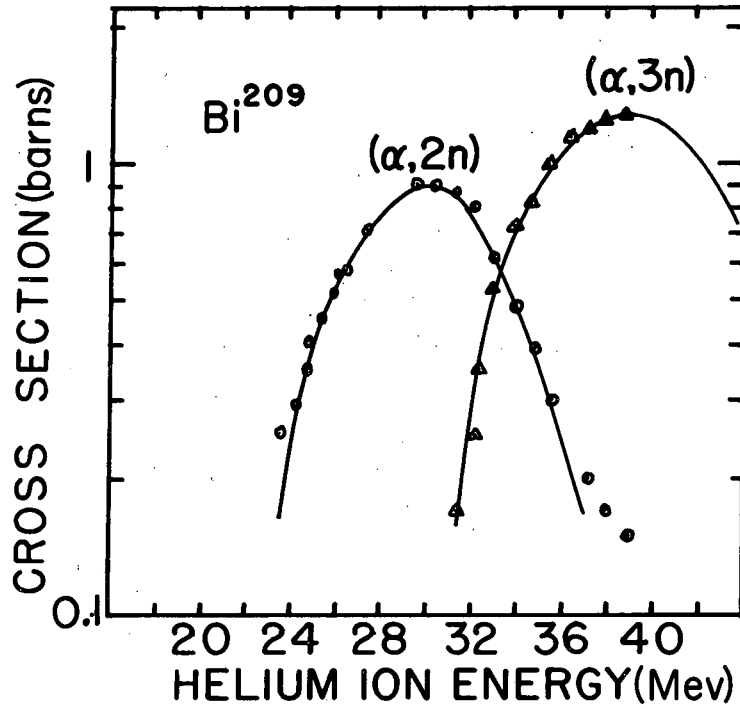
MU-16527

Fig. 6. Theoretical excitation functions (solid lines) and experimental cross sections [0 = σ (α,2n) and △ = σ (α,3n)] for the Bi209 (α,xn) reactions.
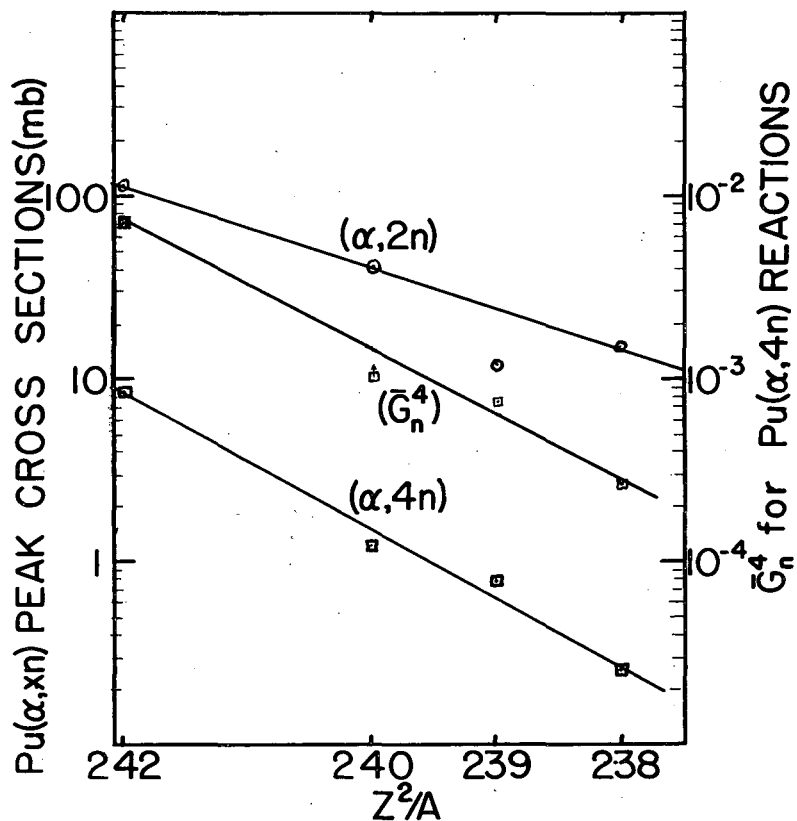
MU-16528

Fig. 7. The variation of the peak Pu(α,xn) cross sections
and the variation of the product of the G_n terms $(\overline{G}_n^4)$
involved in the (α,4n) reaction with Z^2/A. The straight
line is the theoretical variation of $(\overline{G}_n^4)$ with Z^2/A
calculated from the Pu^{238}(α,4n) peak cross section.
The experimental points are the $(\overline{G}_n^4)$ values calculated
from their corresponding Pu(α,xn) peak cross section.

Table III

Ratios of neutron level width to fission level width, Γ_n/Γ_f, and neutron-emission branching ratios G_n of the curium isotopes 239 to 246 as calculated from the $Pu(\alpha,4n)$ peak cross sections.[a]

Ratio	Isotopes of Plutonium			
	238	239	240[c]	242
$(\Gamma_n/\Gamma_f)_9$ [b]	0.071	0.073	0.062	0.069
$(\Gamma_n/\Gamma_f)_0$	0.186	0.190	0.160	0.178
$(\Gamma_n/\Gamma_f)_1$	0.121	0.124	0.104	0.116
$(\Gamma_n/\Gamma_f)_2$	0.314	0.321	0.270	0.301
$(\Gamma_n/\Gamma_f)_3$	0.204	0.209	0.176	0.194
$(\Gamma_n/\Gamma_f)_4$	0.530	0.542	0.457	0.509
$(\Gamma_n/\Gamma_f)_5$	0.344	0.353	0.297	0.331
$(\Gamma_n/\Gamma_f)_6$	0.895	0.917	0.772	0.861
G_{n9} [b]	0.067	0.068	0.058	0.064
G_{n0}	0.157	0.160	0.138	0.151
G_{n1}	0.108	0.110	0.094	0.104
G_{n2}	0.239	0.243	0.213	0.232
G_{n3}	0.169	0.173	0.150	0.164
G_{n4}	0.346	0.352	0.314	0.337
G_{n5}	0.256	0.261	0.229	0.249
G_{n6}	0.472	0.478	0.436	0.463
$\sigma(mb)$ peak	0.30	0.80	1.22[c]	8.60

[a] The experimental values are underlined; all other values were extrapolated.

[b] The subscript refers to the isotope of curium involved (i.e. 9 = 239, 0 = 240, 1 = 241, etc.)

[c] The excitation function is not at its peak, which is estimated to be 1.50 mb.

for all the plutonium isotopes. A summary of these terms is given in Table IV. The probability $P(E^*,x)$ for the $Pu(\alpha,4n)$ reaction is 0.662 ± 0.008, and $\sigma_c P(E^*,x)$ has an average value of 1330 mb. The use of this value in Eq. (14) instead of 1200 mb would decrease $\bar{G}_n$ by only 2.5%.

Fission competition increases as the atomic number Z of the nucleus under consideration increases. This results in a reduction in the cross section for the compound-nucleus $(\alpha,2n)$ reaction in the heavy-element region. At bombarding energies higher than the energy corresponding to the peak of the $(\alpha,2n)$ excitation curve, the reaction mechanism becomes predominantly one in which one neutron is knocked out with a large portion of the excitation energy, and fission competes with only a one-neutron evaporation step. Therefore the events proceeding by the compound-nucleus mechanism are affected more than those proceeding by direct interaction. This is evident in the increased prominence of the direct-interaction tail on the $(\alpha,2n)$ reaction in the heavy-element region. The tail on the $Pu(\alpha,2n)$ reactions is about 25% of the peak cross section. This would indicate that the direct-interaction and compound-nucleus processes are increasing to the same extent as A increases.

D. $Pu^{242}(d,2n)$ Cross Sections

The $Pu^{242}(d,2n)Am^{242m}$ excitation function is similar to the other $Pu(d,2n)$ excitation functions.[2,3] There is a compound-nucleus peak followed by a direct-interaction tail. At higher energies the excitation function appears to increase again. This could be the result of either a decrease in the Am^{242} yield in favor of the Am^{242m} or a change in the reaction mechanism. Theories have been developed on the relative yields of two isomers formed in a nuclear reaction as a function of their spin and excitation energy.[35-38] No correlation could be made in the $Pu^{242}(d,2n)$ reaction because the spins of the two isomers of Am^{242} are not known. The change in mechanism could be from (a) a reaction in which one neutron was knocked out followed by the evaporation of a second neutron to (b) a reaction in which both neutrons are knocked out.

Table IV

The probability of occurrence and the product of this probability and the compound-nucleus-formation cross section at the peak of the $Pu(\alpha, 4n)$ reactions.

Pu isotope	$\sigma_c P(E^*,x)$ (mb)	$P(E^*,x)$
238	1350	0.649
239	1356	0.666
240	1341	0.674
242	1274	0.660

Acknowledgments

References

1.   Glass, Carr, Cobble, and Seaborg, Phys. Rev. $\underline{104}$, 434 (1956).

2.   E. V. Luoma, Deuteron-Induced Spallation and Fission Reactions in Plutonium Isotopes (Master's thesis), UCRL-3495, Nov. 1956.

3.   Walter M. Gibson, Fission and Spallation Competition from the Intermediate Nuclei Americium-241 and Neptunium-235 (Ph.D. thesis), UCRL-3493, Nov. 1956.

4.   Wade, Gonzales-Vidal, Glass, and Seaborg, Phys. Rev. $\underline{107}$, 1311 (1957).

5.   Vandenbosch, Thomas, Vandenbosch, Glass, and Seaborg, Phys. Rev. $\underline{111}$, 1358 (1958).

6.   Joseph A. Coleman, Several Spallation Reactions of Uranium-238 Plus Helium-4 (Master's thesis), UCRL-8186, Feb. 1958.

7.   N. Bohr, Nature $\underline{143}$, 330 (1939).

8.   S. N. Ghoshal, Phys. Rev. $\underline{80}$, 939 (1950).

9.   W. John, Jr., Phys. Rev $\underline{103}$, 704 (1956).

10.   Robert Vandenbosch, Fission and Spallation Competition in Ra^{226}, Th^{230}, U^{235}, and Np^{237} (Ph.D. thesis), UCRL-3858, July 1957.

11.   R. Serber, Phys. Rev. $\underline{72}$, 1114 (1947).

12.   Jose Gonzales-Vidal, Survey of Tritium-Producing Nuclear Reactions (Ph.D. thesis), UCRL-8330, June 1958.

13.   E. B. Paul and R. L. Clarke, Can. J. Phys. $\underline{31}$, 267 (1953).

14. Harvey, Chetham-Strode, Ghiorso, Choppin, and Thompson, Phys. Rev. 104, 1315 (1956).

15. Alfred Chetham-Strode, Jr., Light Isotopes of Berkelium and Californium (Ph.D. Thesis), UCRL-3322, June 1956.

16. D. L. Hufford and B. F. Scott, The Transuranium Elements; Research Papers, Ed. by Seaborg, Katz, and Manning, National Nuclear Energy Series, Plutonium Project Record, (McGraw-Hill Book Company, Inc., New York, 1949), Vol. 14B, p. 1149.

17. Richard A. Glass, Studies in the Nuclear Chemistry of Pu, Am, and Cm, and the Masses of the Heaviest Elements (Ph.D. thesis), UCRL-2560, April 1954.

18. Peter McWalters and Robert Cox, (University of California), Crocker Laboratory, private communications, July 1957.

19. Aron, Hoffman, and Williams, Range-Energy Curves, AECU-663, May 1951.

20. A. Ghiorso, A. H. Jaffey, H. P. Robinson, and B. B. Weissbound, The Transplutonium Elements: Research Papers, Ed. by Seaborg, Katz, and Manning, National Nuclear Energy Series, Plutonium Project Record (McGraw-Hill Book Company, Inc., 1949), Vol. 14B, p. 1226.

21. Strominger, Hollander, and Seaborg, Revs. Modern Phys. 30, 2, 585 (1958).

22. W. W. Meinke, Chemical Procedures Used in Bombardment Work at Berkeley, UCRL-432 and Addendum, 1949.

23. Thompson, Harvey, Choppin, and Seaborg, J. Am. Chem. Soc. 76, 6229 (1954).

24. J. D. Jackson, Can. J. Phys. 34, 767 (1956).

25. McManus, Sharp, and Gellman, Phys. Rev. 93, 924A (1954).

26. R. Vandenbosch and G. T. Seaborg, Phys. Rev. 110, 507 (1958).

27. J. M. Blatt and V. F. Weisskopf, Theoretical Nuclear Physcis (John Wiley and Sons, New York, 1952).

28. M. M. Shapiro, Phys. Rev. 90, 171 (1953).

29. V. F. Weisskopf, Lecture Series in Nuclear Physics, MDDC-1175, Los Alamos Scientific Laboratory, 1943, p. 106.

30. R. E. Batzel, High-Energy Fission and Spallation of Uranium (Ph.D. thesis), UCRL-4303, Feb. 1954.

31. W. John, Jr., Phys. Rev. 103, 704 (1956).

32. T. Darrah Thomas, Spallation-Fission Competition from the Compound System U^{233} plus He^4 (Ph.D. thesis), UCRL-3791, July 1957.

33. Elmer L. Kelly, Excitation Functions of Bismuth (Ph.D. thesis), UCRL-1044, Dec. 1950.

34. Bruce M. Foreman, Jr. and G. T. Seaborg, Nuclear Thermodynamics of the Heaviest Elements, II, UCRL-8051, Aug. 1957; also Bruce M. Foreman, Jr., Spallation and Fission in Th^{232} and the Masses of the Heaviest Elements, (Ph.D. thesis), UCRL-8223, April 1958.

35. E. Segre and A. C. Helmholz, Revs. Modern Phys. 21, 271 (1949).

36. A. W. Fairhall and C. D. Coryell, Phys. Rev. 87, 215 (1952).

37. Harris B. Levy, I. Isomeric States of Bismuth-210.; II. Relative Yields in the Formation of Nuclear Isomers, UCRL-2305, Aug. 1953.

38. Meadows, Diamond, and Sharp, Phys. Rev. 102, 190 (1956).