Lawrence Berkeley National Laboratory

Recent Work

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

Permalink
https://escholarship.org/uc/item/9tn6j4kj

Author
Eads, Donald L.

Publication Date
1959
SPALLATION REACTIONS OF PLUTONIUM-240
WITH HELIUM IONS AND
PLUTONIUM-242 WITH DEUTERONS

TWO-WEEK LOAN COPY

This is a Library Circulating Copy
which may be borrowed for two weeks.
For a personal retention copy, call
Tech. Info. Division, Ext. 5545
DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.
SPALLATION REACTIONS OF PLUTONIUM-240 WITH HELIUM IONS AND PLUTONIUM-242 WITH DEUTERONS

Donald L. Eads
(Master's Thesis)
January 1959

Printed for the U. S. Atomic Energy Commission
SPALLATION REACTIONS OF PLUTONIUM-240 WITH HELIUM IONS AND PLUTONIUM-242 WITH DEUTERONS

Contents

Abstract ................................. 3
I. Introduction .............................. 4
II. Method and Apparatus .............................. 6
   A. Pu240 with Helium Ions ....................... 6
      1. Target Assembly .............................. 6
      2. Target Preparation .............................. 6
      3. Helium-Ion Beam .............................. 6
      4. Chemical Procedures .............................. 8
      5. Counting Instruments .............................. 8
      6. Calculations of Cross Sections ...................... 9
   B. Pu242 with Deuterons ....................... 10
      1. Target Assembly .............................. 10
      2. Target Preparation .............................. 10
      3. Deuteron Beam .............................. 10
      4. Chemical Procedures .............................. 12
      5. Calculation of Cross Sections ...................... 13
III. Results ................................. 15
IV. Discussion ................................. 20
   A. A Model for Competition Between Neutron-Evaporation and Fission in the Heaviest Elements ...................... 20
   B. Evaluation of the Neutron-Emission Branching Ratios G_n for the Curium Isotopes ...................... 22
   C. Remarks on the Theoretical Calculations and Some Comparisons of the Pu(α,xn) Reactions ...................... 23
   D. Pu242(d,2n) Cross Sections ...................... 29
Acknowledgments .................. 31
References ................................. 32
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 \( ^{240}\text{Pu}(\alpha,\text{xn}) \) products produced by bombarding \( ^{240}\text{Pu} \) with 22- to 46-Mev helium ions. Excitation functions are given for the \( (\alpha,2\text{n})\text{Cm}^{242} \), \( (\alpha,3\text{n})\text{Cm}^{241} \), and \( (\alpha,4\text{n})\text{Cm}^{240} \) reactions. The \( ^{240}\text{Pu}(\alpha,\text{xn}) \) 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 \( R_0 \) of \( 1.5 \times 10^{-13} \) cm.

The \( ^{240}\text{Pu}(\alpha,4\text{n}) \) reactions were used to evaluate the ratio of \( \Gamma_n/\Gamma_t \) for the curium isotopes.

The \( ^{242}\text{Pu}(d,2\text{n})\text{Am}^{242,242m} \) 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

Donald L. Eads

Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California
January 1959

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 ≥ 90). Spallation, the emission of a various number of nucleons or light nuclei (e.g., 1d2, 1t3, or 2He4), is divided into compound-nucleus and direct-interaction reactions.

Bohr's original compound-nucleus theory7 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}±3 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.\textsuperscript{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.\textsuperscript{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.\textsuperscript{4,12,13}

This paper is composed of two parts: (a) The comparison of the Pu\textsuperscript{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, $\Gamma_n/\Gamma_t$, 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\textsuperscript{242m} formed in the Pu\textsuperscript{242}(d,2n) reaction and, for several bombarding energies, the yield of the Am\textsuperscript{242} isomer.
II. METHODS AND APPARATUS

A. Plutonium-240 with Helium Ions

1. Target Assembly

The recoil technique used in the determination of the $^{240}\text{Pu}(\alpha,\text{xn})$ cross sections is similar to that described by Harvey et al.\textsuperscript{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.\textsuperscript{10}

The unknown $^{240}\text{Pu}(\alpha,\text{xn})$ cross sections were determined by using the $\text{Cm}^{244}(\alpha,2n)\text{Cf}^{246}$ reaction,\textsuperscript{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% $^{240}\text{Pu}$, 12.20% $^{239}\text{Pu}$, and 0.583% $^{241}\text{Pu}$ by weight) and 0.2 microgram of $\text{Cm}^{244}$ per cm$^2$ from a 0.4 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\textsuperscript{16} and modified by Glass.\textsuperscript{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,\textsuperscript{18} or an energy
Fig. 1. Recoil-target assembly used in measuring the $Pu^{240}(\alpha,\alpha)$ cross sections.
of 48 ± 0.5 Mev. The range-energy curves of Aron et al.\textsuperscript{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 N HCl with just enough HNO\textsubscript{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 N HCl (+ 0.1 N HNO\textsubscript{3}) and passed through a column of Dowex 1 anion resin 3 mm in diameter and 5 cm long. 12 N HCl (+ 0.1 N HNO\textsubscript{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 N NH\textsubscript{4}Cl (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\textsuperscript{2} at 4 volts.

**Plutonium.** The plutonium was desorbed by eluting with 12 N HCl made 0.1 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\textsuperscript{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. 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 $\sigma$ for a nuclear reaction is defined by the equation

$$\sigma = \frac{N}{nI_t},$$

where $N$ is the number of product nuclei formed during the bombardment, $n$ the number of target nuclei per cm$^2$, and $I_t$ 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$^{240}(\alpha,\alpha n)$ reactions were related to the known Cm$^{244}(\alpha,2\alpha n)$Cf$^{246}$ cross section at the same energy by the expression

$$\frac{\sigma_{\text{Pu}^{240}(\alpha,\alpha n)}}{\sigma_{\text{Cm}^{244}(\alpha,2\alpha n)}} = \frac{A_{\text{Cm}^{244}}(2\alpha - \alpha n)}{A_{\text{Cf}^{246}}} \times \frac{t_{1/2}}{t_{1/2}^{2\alpha - \alpha n}} \times \frac{C_{\text{Pu}^{240}}} {C_{\text{Pu}^{244}}^{2\alpha n}}.$$

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.

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

$$A = \frac{d}{m} f_1 f_2 f_3,$$

where $(d/m)$ 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,
\[ f = \frac{t}{t_{1/2}} \times \frac{0.693}{1-e^{-\lambda t}} \]

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

\[ f_2 = e^{-\lambda(\Delta t)} \]

where \( \Delta t \) 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 illustrated 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 M NH\(_4\)Cl 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, corresponding to a maximum energy of 24 Mev.
Fig. 2. Microtarget assembly used in measuring the \( \text{Pu}^{242}(d,2n) \) 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. The organic layer was removed and washed twice with an equal volume of 6 N HCl. The aqueous phase and the two washes were combined and evaporated to dryness. The residue was dissolved in 12 N HCl (+0.1 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-amercurium fraction was eluted along with the rare earths and other fission products. The plutonium was desorbed with 12 N HCl made 0.1 N in HCl.

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 N in HF. The precipitate was washed with water containing a trace of HF and dissolved in 12 N HCl containing a trace of boric acid. NH$_4$OH 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. 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 N NH$_4$Cl 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$^{241m}$ yield, the target was stored until all the 1-hour Am$^{241m}$ isomer had decayed. The americium-curium fraction from the alcoholic HCl column was then evaporated to dryness. The residue was dissolved in 50% of 0.4 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 30% portions of the 0.4 M But. solution and transferred to the resin, washing
down the sides of the column. Trichloro-ethylene (boiling point 87°C) was condensed in a jacket surrounding the column to maintain a temperature of 87°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 μ 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 4 Cl 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 0 1 is the number of Am 242 nuclei and N 0 2 the number of Cm 242 nuclei at the end of the bombardment, the activity of the Cm 242 is

\[
-\frac{dN_2}{dt} = \lambda_2 N_2 = \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} N_0 1 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_0 2 \lambda_2 e^{-\lambda_2 t},
\]

where t is the time since the end of the bombardment and the \( \lambda \)'s are the corresponding decay constants.

The number of Am 242 nuclei N 0 1 at the end of the bombardment is

\[
N_0 1 = n_{Pu} \sigma I_t \frac{t_{1/2}}{t_b} \left[ \frac{1 - e^{-\lambda_1 t}}{0.693} \right],
\]

where \( n_{Pu} \) is the number of Pu 242 target nuclei per cm², \( I_t \) (current times bombardment time) the number of projectiles that pass through the target, \( \sigma \) 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 \((n_{Pu} \sigma t_b)\) is the number of product nuclei formed in the nuclear reaction, assuming none decayed during the bombardment. The number of Cm\(^{242}\) daughter nuclei \(N_2^0\) 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:

\[
N_2^0 = n_{Pu} \sigma t_b - N_1^0 = n_{Pu} \sigma t_b (1 - f), \tag{3}
\]

after substituting Eq. (2) for \(N_1^0\).

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

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

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

Solving for \(\sigma\) in Eq. (4), one obtains

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

Corrections that have been neglected in this experiment are:

The decay during the 4-hour bombardment of the daughter Cm\(^{242}\) \((t_{1/2} = 162\) 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$^{240}$(α,xn) Cross Sections

The cross sections for the reactions Pu$^{240}$(α,2n) Cm$^{242}$, (α,3n) Cm$^{241}$, and (α,4n) Cm$^{240}$, determined by comparing the relative yields of Cm$^{242}$, Cm$^{241}$, and Cm$^{240}$ respectively with the yield of the Cm$^{244}$(α,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$^{244}$(α,2n) monitor reaction were known to 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$^{242}$m 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

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

<table>
<thead>
<tr>
<th>E (Mev)</th>
<th>($\alpha,2n$)Cm$^{242}$</th>
<th>($\alpha,3n$)Cm$^{241}$</th>
<th>($\alpha,4n$)Cm$^{240}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>23.2</td>
<td>3.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>24.0</td>
<td>4.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>26.5</td>
<td>18.8</td>
<td>0.72</td>
<td></td>
</tr>
<tr>
<td>28.0</td>
<td>41.1 (40.9)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>29.6</td>
<td>19.1</td>
<td>2.6</td>
<td>0.0013</td>
</tr>
<tr>
<td>30.6</td>
<td>12.7</td>
<td>3.6</td>
<td>0.012</td>
</tr>
<tr>
<td>34.0</td>
<td>13.1</td>
<td>6.5</td>
<td>0.24</td>
</tr>
<tr>
<td>36.2</td>
<td>12.4</td>
<td></td>
<td>0.22</td>
</tr>
<tr>
<td>37.4</td>
<td>14.3</td>
<td>6.5</td>
<td>0.35</td>
</tr>
<tr>
<td>39.0</td>
<td>10.0</td>
<td>5.0</td>
<td>0.55</td>
</tr>
<tr>
<td>40.4</td>
<td>6.1</td>
<td>5.3</td>
<td>0.77</td>
</tr>
<tr>
<td>44.5</td>
<td></td>
<td>3.3</td>
<td>1.20</td>
</tr>
</tbody>
</table>
Fig. 3. Excitation functions for the \( ^{240}\text{Pu} (\alpha,2n)^{242}\text{Cm} \), 
\((\alpha,3n)^{241}\text{Cm} \), and \((\alpha,4n)^{240}\text{Cm} \) reactions.
Table II

Pu$^{242}$(d,2n) cross sections (in millibarns)

<table>
<thead>
<tr>
<th>E (Mev)</th>
<th>(d,2n)Am$^{242m}$</th>
<th>(d,2n)Am$^{242}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>12.0</td>
<td>11.9</td>
<td>30</td>
</tr>
<tr>
<td>13.9</td>
<td>25.1</td>
<td>45</td>
</tr>
<tr>
<td>14.4</td>
<td>21.1</td>
<td></td>
</tr>
<tr>
<td>15.0</td>
<td>12.2</td>
<td></td>
</tr>
<tr>
<td>16.4</td>
<td>12.0</td>
<td></td>
</tr>
<tr>
<td>19.0</td>
<td>11.1</td>
<td></td>
</tr>
<tr>
<td>22.2</td>
<td>13.1</td>
<td></td>
</tr>
<tr>
<td>23.3</td>
<td>16.0</td>
<td></td>
</tr>
</tbody>
</table>
Fig. 4. The excitation function for the $^{242}\text{Pu}(d,2n)^{242m}\text{Am}$ reaction and two cross sections (0) for the $^{242}\text{Pu}(d,2n)^{242}\text{Am}$ 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. The Monte Carlo method was used to evaluate 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 $\epsilon$ 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 excitation energy.

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

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

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

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

$\Delta_x$ is the energy, in units of $T$, above the threshold for the emission of $x$ neutrons,

$$ \Delta_x = (E^* - \sum_{i=0}^{x} B_i)/T. \quad (8) $$
where \( \sum_{i=1}^{x} B_i \) is the sum of the binding energies of the \( x \) emitted neutrons and all of the other terms maintain their meaning. Then \( \Delta_{x+1} \) is the energy above the threshold for the emission of \( x+1 \) neutrons,

\[
\Delta_{x+1} = \frac{E^* - \sum_{i=1}^{x+1} B_i}{T},
\]

(9)

where \( \sum_{i=1}^{x} B_i \) 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 \( \Delta_x^f \) is substituted for \( \Delta_{x+1} \) in Eq. (6) for \( P(E^*,x) \),

\[
\Delta_x^f = \frac{E^* - \sum_{i=1}^{x} B_i - E_{th}}{T},
\]

(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 \((\alpha,xn)\) reaction is then given by the equation

\[
\sigma(\alpha,xn) = \sigma_c(\alpha)\ G_{n1}\ G_{n2} \ldots\ G_{nx} P(E^*,x) T(E^*),
\]

(11)

where \( \sigma_c \) is the cross section for forming the compound nucleus at the bombarding helium-ion energy (several sources are available for the \( \sigma_c \) 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

\[
G_n = \frac{\Gamma_n}{\Sigma \Gamma_i},
\]

(12)

where \( \Gamma_n \) 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

\[ \sum \Gamma_i = \Gamma_n + \Gamma_f, \]

where \( \Gamma_f \) is the fission level width.

B. Evaluation of the Neutron-Emission Branching Ratios

\( G_n \) for the Curium Isotopes

Vandenbosch \(^1\) and co-workers \(^5,26\) have devised a method for calculating individual \( \Gamma_n/\Gamma_f \) values. The assumptions on which their method is based are

(a) \( \Gamma_n/\Gamma_f \) is independent of the excitation energy for excitation energies well above the neutron-emission threshold.\(^1,30\)

(b) \( \Gamma_n/\Gamma_f \) for even-even nuclei is twice as great as \( \Gamma_n/\Gamma_f \) for even-odd nuclei.\(^27\)

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

(i) \( \Gamma_n \) increases with decreasing neutron-binding energy.

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

Using these assumptions, Vandenbosch et al.\(^5\) derived a formula for calculating the individual \( \Gamma_n/\Gamma_f \) values for the \( x \)th neutron leaving the compound nucleus,

\[ \left( \frac{\Gamma_n}{\Gamma_f} \right)_x = \frac{1.93a}{(1.3)^x} \frac{G_n}{1-G_n}, \]

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

\[ \bar{G}_n = G_{n1} G_{n2} G_{n3} G_{n4} = \frac{\sigma (\gamma,ln) \ pK}{1200 \ mb}. \]
Vandenbosch\textsuperscript{10} did not use \( \sigma_c \) in the calculation of \( \bar{\sigma}_n \) 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.\textsuperscript{31}

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

\[
\bar{\sigma}_n = \frac{\Gamma_n}{\Gamma_f} \left( \frac{\Gamma_n}{\Gamma_f} \right) ,
\]

where \( \Gamma_n/\Gamma_f \) 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 \( \text{Pu}(\alpha,xn) \) Reactions

The two parameters that must be selected to proceed with the Jackson-type calculations are the nuclear radius \( R_0 \), as related to \( \sigma_c \), and the nuclear temperature \( T \). The \((\alpha,2n)\) reaction occurs in the region where the slope of the compound-nucleus-formation cross section \( \sigma_c \) versus \( E_\alpha \) 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 \((\alpha,xn)\) excitation functions. Experiments\textsuperscript{1,10,32} in the transuranium elements led to the selection of \( 1.5 \times 10^{-13} \) 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 \( \text{Pu}(\alpha,xn) \) excitation functions calculated by the modified Jackson method are illustrated in Fig. 5. The experimental cross sections for \( \text{Pu}^{238} \), \( \text{Pu}^{239} \), and \( \text{Pu}^{242} \) \textsuperscript{1} and \( \text{Pu}^{240} \) 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 \( \sigma = \sigma (\alpha,2n), \Delta = \sigma (\alpha,3n), \) and \( \epsilon = \sigma (\alpha,4n) \) for the \((\alpha,\text{xn})\) reactions of \(\text{Pu}^{238}, \text{Pu}^{239}, \text{Pu}^{240}, \) and \(\text{Pu}^{242}\.\)
(α,xn) curves, yielding better agreement with experiments in the heavy-element region.

The theoretical Pu(α,xn) excitation functions are only qualitative 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\textsuperscript{209}(α,xn) excitation functions\textsuperscript{33} were used as an example. A nuclear temperature of 1.35 Mev and an R\textsubscript{0} of 1.5 x 10\textsuperscript{-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\textsuperscript{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\textsuperscript{2}/A decreases.

The net result of these two factors is a rapid increase in the ratio of $\Gamma_n$ to $\Gamma_f$ with increasing A. A summary of the $\Gamma_n$ to $\Gamma_f$ 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 experimental results.

The variation with Z\textsuperscript{2}/A of the product of the $G_n$ values (G\textsuperscript{4}_n) 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\textsuperscript{238}(α,4n) peak cross section were used to evaluate the G\textsuperscript{4}_n 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 (G\textsuperscript{4}_n) line and the (α,4n) line are the same, within the accuracy of the calculations, indicating that the value of $\sigma_c F(E^*,x)$ should be the same.
Fig. 6. Theoretical excitation functions (solid lines) and experimental cross sections \( \sigma (\alpha,2n) \) and \( \phi = \sigma (\alpha,3n) \) for the Bi\textsuperscript{209} (\( \alpha,\text{xn} \)) reactions.
Fig. 7. The variation of the peak Pu(α,xn) cross sections and the variation of the product of the $G_n$ terms $\left(\overline{G}_n^4\right)$ involved in the (α,4n) reaction with $Z^2/A$. The straight line is the theoretical variation of $\left(\overline{G}_n^4\right)$ with $Z^2/A$ calculated from the Pu$^{238}(\alpha,4n)$ peak cross section. The experimental points are the $\left(\overline{G}_n^4\right)$ values calculated from their corresponding Pu(α,xn) peak cross section.
Table III

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

<table>
<thead>
<tr>
<th>Isotopes of Plutonium</th>
<th>238</th>
<th>239</th>
<th>240(^c)</th>
<th>242</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\frac{\Gamma_n}{\Gamma_f}$ (^b)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\frac{\Gamma_n}{\Gamma_f}$</td>
<td>0.071</td>
<td>0.073</td>
<td>0.062</td>
<td>0.069</td>
</tr>
<tr>
<td>$\frac{\Gamma_n}{\Gamma_f}$</td>
<td>0.186</td>
<td>0.190</td>
<td>0.160</td>
<td>0.178</td>
</tr>
<tr>
<td>$\frac{\Gamma_n}{\Gamma_f}$</td>
<td>0.121</td>
<td>0.124</td>
<td>0.104</td>
<td>0.116</td>
</tr>
<tr>
<td>$\frac{\Gamma_n}{\Gamma_f}$</td>
<td>0.314</td>
<td>0.321</td>
<td>0.270</td>
<td>0.301</td>
</tr>
<tr>
<td>$\frac{\Gamma_n}{\Gamma_f}$</td>
<td>0.204</td>
<td>0.209</td>
<td>0.176</td>
<td>0.194</td>
</tr>
<tr>
<td>$\frac{\Gamma_n}{\Gamma_f}$</td>
<td>0.530</td>
<td>0.542</td>
<td>0.457</td>
<td>0.509</td>
</tr>
<tr>
<td>$\frac{\Gamma_n}{\Gamma_f}$</td>
<td>0.344</td>
<td>0.353</td>
<td>0.297</td>
<td>0.331</td>
</tr>
<tr>
<td>$\frac{\Gamma_n}{\Gamma_f}$</td>
<td>0.895</td>
<td>0.917</td>
<td>0.772</td>
<td>0.861</td>
</tr>
<tr>
<td>$G_n$</td>
<td>0.067</td>
<td>0.068</td>
<td>0.058</td>
<td>0.064</td>
</tr>
<tr>
<td>$G_n$</td>
<td>0.157</td>
<td>0.160</td>
<td>0.138</td>
<td>0.151</td>
</tr>
<tr>
<td>$G_n$</td>
<td>0.108</td>
<td>0.110</td>
<td>0.094</td>
<td>0.104</td>
</tr>
<tr>
<td>$G_n$</td>
<td>0.239</td>
<td>0.243</td>
<td>0.213</td>
<td>0.232</td>
</tr>
<tr>
<td>$G_n$</td>
<td>0.169</td>
<td>0.173</td>
<td>0.150</td>
<td>0.164</td>
</tr>
<tr>
<td>$G_n$</td>
<td>0.346</td>
<td>0.352</td>
<td>0.314</td>
<td>0.337</td>
</tr>
<tr>
<td>$G_n$</td>
<td>0.256</td>
<td>0.261</td>
<td>0.229</td>
<td>0.249</td>
</tr>
<tr>
<td>$G_n$</td>
<td>0.472</td>
<td>0.476</td>
<td>0.436</td>
<td>0.463</td>
</tr>
<tr>
<td>$\sigma$(mb) peak</td>
<td>0.30</td>
<td>0.80</td>
<td>1.22(^c)</td>
<td>8.60</td>
</tr>
</tbody>
</table>

\(\text{\(^a\)The experimental values are underlined; all other values were extrapolated.}\)

\(\text{\(^b\)The subscript refers to the isotope of curium involved (i.e. 9 = 239, 0 = 240, 1 = 241, etc.)}\)

\(\text{\(^c\)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 \( \text{Pu}(\alpha,4n) \) 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 \( \frac{G_n}{\gamma} \) 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 \( \text{Pu}(\alpha,2n) \) 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. \( \text{Pu}^{242}(d,2n) \) Cross Sections

The \( \text{Pu}^{242}(d,2n)\text{Am}^{242m} \) excitation function is similar to the other \( \text{Pu}(d,2n) \) 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 \( \text{Am}^{242} \) yield in favor of the \( \text{Am}^{242m} \) 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 \( \text{Pu}^{242}(d,2n) \) reaction because the spins of the two isomers of \( \text{Am}^{242} \) 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(α,4n) reactions.

<table>
<thead>
<tr>
<th>Pu isotope</th>
<th>$\sigma_{c}P(E^*,x)$ (mb)</th>
<th>$P(E^*,x)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>238</td>
<td>1350</td>
<td>0.649</td>
</tr>
<tr>
<td>239</td>
<td>1356</td>
<td>0.666</td>
</tr>
<tr>
<td>240</td>
<td>1341</td>
<td>0.674</td>
</tr>
<tr>
<td>242</td>
<td>1274</td>
<td>0.660</td>
</tr>
</tbody>
</table>
Acknowledgments

I should like to acknowledge the interest and guidance of Chancellor Glenn T. Seaborg.

I am deeply indebted to Dr. Bernard G. Harvey, without whose encouragement and personal advice this work would not have been completed.

I thank Dr. Richard A. Glass for his guiding hand in the early part of this work.

I am grateful to Dr. T. Darrah Thomas for the very helpful discussions on the Jackson-type calculations.

I appreciate the many interesting discussions with Ernest Valyocsik, Robert J. Silva, and other members of the nuclear reactions group.

I thank Bob Cox, John Wood, Peter McWalters, W. Bart Jones, and the other members of the Crocker 60-inch cyclotron crew for their co-operation in setting up the targets for bombardment.

I also wish to thank Lilly Hirota and Patricia Howard for typing the Multilith for this thesis.

I appreciate the assistance of the Health Chemistry Group, under the direction of Nelson B. Garden, during the course of this work.

Last, but by no means least, I want to thank my wife, Claudia, and other members of my family for their encouragement and understanding.

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


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


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


This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission to the extent that such employee or contractor prepares, handles or distributes, or provides access to, any information pursuant to his employment or contract with the Commission.