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SPALLATION-FISSION COMPETITION
IN NEPTUNIUM COMPOUND SYSTEMS.

DECAY-SCHEME STUDIES

Richard Marshall Lessler
(Thesis)

October 1958

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Contents

Abstract.	4
List of Illustrations	6
List of Tables.	7
I. Introduction	8
II. Experimental Procedure.	11
A. Introduction.	11
B. Target Preparations	11
C. Target Assembly.	12
D. Chemical Procedures	14
1. Chemical Separations.	14
2. Determination of Yield	18
E. Counting Techniques and Corrections	20
1. Nucleometer.	20
2. Geiger-Müller Counter	21
3. Decay Curves	22
F. Cross Section Calculations	23
1. Standard Cross-Section Formula	23
2. Correction for Decay During Bombardment	23
3. Isotope Corrections	24
4. Errors	24
III. Results.	25
A. Spallation Cross Sections	25
1. U^{235} plus deuterons	25
2. U^{236} plus deuterons	28
3. U^{234} plus deuterons	28
4. U^{238} plus deuterons	28
B. Fission Cross Sections	34
1. Primary Fission Yields	34
2. Fission Yield Curves.	38

C.	Deuteron-Gamma Cross Sections ..	43
1.	$U^{234}(d,\gamma)Np^{236}$	43
2.	$U^{236}(d,\gamma)Np^{238}$	45
3.	$U^{238}(d,\gamma)Np^{240}$	45
IV.	Discussion.	48
A.	Spallation	48
1.	Calculation of Level-Width Ratios	48
2.	Prediction of Cross Sections	54
3.	Experimental Results.	55
B.	Fission	56
1.	Fission-Yield Curves.	56
2.	Fission Asymmetry.	56
3.	Total Fission Cross Sections	59
C.	Deuteron-Gamma Reactions.	60
D.	Summary	63
V.	Decay-Scheme Studies	64
A.	One-Hour Np^{240}	64
1.	Identification.	64
2.	Decay Properties	65
B.	The New Isotope Np^{241}	66
1.	Identification.	66
2.	Decay Properties	67
C.	Decay Properties of Np^{233}	68
D.	Decay Properties of Np^{234}	69
	Acknowledgments.....	71
	References.	72

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ABSTRACT

The results of a radiochemical study of fission and spallation products induced in U^{234} , U^{235} , U^{236} and spallation products induced in U^{238} by deuterons of less than 24 Mev are presented. The (d,n) reactions are interpreted as the result of direct interactions between the deuteron and the target nuclei, while the (d,xn) reactions are thought to be taking place primarily by compound-nucleus mechanisms. The (d,xn) reactions are seen to be affected by the large cross section for the fission reaction, showing that the excitation functions for the (d,xn) reactions are sensitive indicators of the relative fissionabilities of these nuclei. Values for Γ_f/Γ_t (based on the experimental cross sections for the (d,xn) reactions on these uranium isotopes) have been calculated. When these and other values of Γ_f/Γ_t are plotted as a function of the mass number, a systematic increase of the fissionability with decreasing mass number and increasing atomic number is seen. A method for predicting the peak values of the cross section for (d,xn) and (α ,xn) reactions on uranium and plutonium isotopes is then given for $x = 2, 3$, or 4 . The peak-to-valley ratios of the fission-yield curves for U^{234} , U^{235} , U^{236} , and U^{238} bombarded with deuterons of energies of about 23 Mev were studied, and no significant differences were found. The fission asymmetry of these reactions is seen to be consistent with the fission asymmetry of similar reactions. The experimental total reaction cross sections, fission plus spallation, are in agreement with a nuclear radius parameter of $r_0 = 1.5 \times 10^{-13}$ cm.

Excitation functions for the (d, γ) reactions of U^{234} , U^{236} , and U^{238} have been measured. These excitation functions all rise to a maximum of about a millibarn at a bombarding energy between 15 and 20 Mev. The results of these measurements are seen to be consistent with the excitation functions for (p, γ) and (α , γ) reactions.

The 60-minute neptunium activity which had previously been assigned to Np^{241} has been identified as an isomer Np^{240} . Accompanying the decay of this Np^{240} are gamma rays of energies 1160, 1000, 915, 595, 565, 435, 245, 160, and 85 kev. Neptunium-241 has now tentatively been found to have isomers with half lives of 16 minutes and 3.5 hours. A study of the decay properties of the new isotope Np^{241} and also of Np^{233} and Np^{234} are described.

List of Illustrations

1.	Microtarget assembly.	13
2.	The chemical separation.	15
3.	Spallation excitation functions for deuteron-induced reactions of U^{235}	26
4.	Spallation excitation functions for deuteron-induced reactions of U^{236}	29
5.	Spallation excitation functions for deuteron-induced reactions of U^{234}	30
6.	Spallation excitation functions for deuteron-induced reactions of U^{238}	33
7.	Fraction of chain yield as a function of (Z-Zp).	36
8.	Integrated fraction of chain yield as a function of (Z-Zp).	37
9.	Fission yield curve for 14.7 Mev deuteron-induced fission of U^{235}	39
10.	Fission yield curve for 23.4 Mev deuteron-induced fission of U^{235}	40
11.	Fission yield curve for 23.4 Mev deuteron-induced fission of U^{236}	41
12.	Fission yield curve for 23.4 Mev deuteron-induced fission of U^{234}	42
13.	(d, γ) excitation functions of U^{234} and U^{236}	44
14.	(d, γ) excitation functions of U^{238}	47
15.	Fission probability	51
16.	Ratio of fission probability to neutron evaporation probability.	53

List of Tables

I.	Nucleometer counting efficiencies used.	20
II.	U^{235} + deuterons; spallation cross sections	27
III.	U^{236} + deuterons; spallation cross sections	27
IV.	U^{234} + deuterons; spallation cross sections	31
V.	U^{238} + deuterons; spallation cross sections	31
VI.	Fission cross sections	35
VII.	U^{234} (d,r) Np^{236} cross sections	43
VIII.	U^{236} (d,r) Np^{238} cross sections	45
IX.	U^{238} (d,r) Np^{240} cross sections	46
X.	Mean level-width ratios.	50
XI.	Maximum cross sections in millibarns	54
XII.	Fission asymmetry.	58
XIII.	Asymmetry and fission cross sections for deuteron-induced fission of uranium isotopes	59

I. INTRODUCTION

A compound nuclear system has been defined by Weisskopf as the state in which some energy exchange between target and projectile has taken place, regardless of the role of the incident particle.¹ Thus it refers to states created by both direct and indirect collisions or any state in which the incident particle has been removed from its original course. This differs from the compound nucleus of Bohr, which postulates that the incident particle and its kinetic energy are absorbed by the bombarded nucleus with the result that the incident particle loses its identity.² The decay of this compound nucleus is thus essentially independent of its method of formation. The theory of compound-nucleus formation gives best agreement with experiment in the medium-energy region ($E < 50$ Mev).

One of the purposes of this study was to determine the relative amounts of fission and spallation reactions in this region, where the theory of the compound nucleus is applicable. Fission is the process whereby a heavy bombarded nucleus breaks up into two large fragments. In spallation reactions the bombarded nucleus emits only small particles such as neutrons, protons, tritons, and alpha particles. These two types of reactions compete with each other in that one reaction increases at the expense of the other. Therefore the magnitude of the spallation reactions gives a good indication of the relative fissionability of various nuclei.

A Z^2/A dependence for the fissionability of a nucleus has been predicted from the liquid-drop model.³ This Z^2/A dependence has been observed in spontaneous fission^{4,5} and slow-neutron-induced fission.⁶ The use of this parameter at higher excitation energies has not always given an accurate measurement of fissionability.^{7,8} In order to eliminate the effect of the variation of atomic number, this study was restricted to nuclear reactions of the isotopes of one element. Thus the effect of changing only A could be found. Uranium afforded the greatest number of fissionable isotopes to be studied. Accordingly, the separated uranium isotopes U^{234} , U^{235} , U^{236} , and U^{238} were bombarded

with deuterons of 8 to 24 Mev in the Crocker Laboratory 60-inch cyclotron. Cross sections for fission and spallation reactions were then determined by radiochemical separation of the products. Uranium-233 has previously been bombarded with deuterons of energies up to 24 Mev⁷ and U²³⁸ with deuterons up to 20 Mev.⁹ Therefore, five cases in which the atomic number was held constant could be studied.

Isotopes in the heavy-element region for which spallation-reaction cross section have previously been determined are U²³³, U²³⁴, U²³⁵, U²³⁶, U²³⁸, Pu²⁴², Am²⁴¹, and Am²⁴³ bombarded with helium ions¹⁰⁻¹⁶ and Th²³², Np²³⁷, Pu²³⁸, Pu²³⁹, and Pu²⁴⁰ bombarded with both deuterons^{7-9,12} and helium ions.^{7,14,15,17,18}

There are marked differences between fission caused by low-energy projectiles and fission caused by high-energy projectiles. At low energies fission is predominantly asymmetric and the primary fission fragments have a neutron excess. As the energy is increased, fission becomes more symmetric until at high energies symmetric fission predominates and the primary fission fragments have a neutron deficit. Uranium isotopes bombarded with deuterons of about 20 Mev are particularly interesting. At this energy the fission of U²³³ has been shown to be symmetric⁷ whereas that of U²³⁸ is slightly asymmetric.¹⁹ Therefore, a study of the variation of fission asymmetry with mass number might reveal something new about the fission process. Other isotopes had been bombarded and the fission asymmetry studied. These include Th²³², U²³³, U²³⁸, Pu²³⁹, and Pu²⁴⁰ bombarded with deuterons,^{7,8,19,20} and Th²³², U²³³, U²³⁵, U²³⁸, Np²³⁷, Pu²³⁸, and Pu²³⁹ bombarded with helium ions.^{7,10,12-15,17,21} All these studies, except for U²³⁸ irradiated with deuterons and Th²³² irradiated with deuterons and helium ions, had shown symmetric fission near the maximum energy obtainable on the Crocker Laboratory 60-inch cyclotron (24-Mev deuterons or 48-Mev helium ions).

The nuclear radius has been expressed by the formula $R = r_0 A^{1/3}$. Theoretical cross sections for compound-nucleus formation by deuteron-induced reactions and helium-ion-induced reactions have been calculated by Shapiro²² and Blatt and Weisskopf,²³ using various values of the

parameter r_0 . The results of previous studies^{7,13,14} agreed with a value of 1.5×10^{-13} cm for r_0 .

Measurements of the cross section for the (d,γ) reaction on U^{238} looked promising, as the 1-hour Np^{240} isomer could be formed only by this reaction and its short half life made it possible to measure a small cross section. No cross sections for (d,γ) reactions other than in very light isotopes had been reported in the literature. Kelly has determined the cross section for the (p,γ) reaction on Bi^{209} ,²⁴ and Fairhall,²⁵ Morinaga,²⁶ and Sharma²⁷ have done work on cross sections for the (α,γ) reaction in lighter elements (Ni, Xe, and Au respectively). Because measurements on U^{238} have shown the cross section for the (d,γ) reaction to be on the order of a millibarn, the cross sections for the (d,γ) reactions of U^{234} and U^{236} have also been measured.

The principal aim of the study presented herein is to determine the degree of spallation-fission competition in uranium isotopes when deuterons are used as the projectile. On the basis of these results an attempt has been made to correlate spallation-fission competition in near-by elements. Also the asymmetry of fission in U^{234} , U^{235} , and U^{236} is studied, as are the (d,γ) reactions on U^{234} , U^{236} , and U^{238} . The identification of the 1-hour Np^{240} isomer, and work on its decay scheme as well as those of Np^{233} and Np^{234} , are discussed. The tentative identification of isomers of Np^{241} with half lives of 16 minutes and 3.5 hours is described.

II. EXPERIMENTAL PROCEDURES

A. Introduction

In order to determine the cross section for a nuclear reaction, one may use the formula

$$\sigma = N/nI,$$

where σ is the cross section in cm^2 , N is the number of product atoms formed, n is the number of atoms per cm^2 of the target material, and I is the number of particles striking the target material. The use of this formula requires that all the measured particles strike the target, that the target material be of uniform thickness but not thick enough to attenuate the beam significantly, and that the half life of the products be long compared to the length of the bombardment. It is then possible to determine a cross section from the values for N , n , and I . The determination of n is discussed in the following section headed "Target Preparation," that of I under a section headed "Target Assembly," and that of N in the remainder of this chapter.

B. Target Preparation

When 1-mil-thick uranium foils were available--as for the U^{238} bombardments and a few U^{235} bombardments--it was necessary only to cut these foils into 2.20- by 4.40-cm rectangles and then weigh them in order to determine the number of atoms per cm^2 .

For U^{234} , U^{236} , and isotopically pure U^{235} (>99%), such metallic foil was not available. A thin uniform target was obtained by electro-deposition of uranium on a 10-mil 2S aluminum disc shaped in the form of a shallow "hat." The method of electrodeposition developed by Hufford and Scott²⁸ was used. A solution of a few milligrams of uranium in concentrated nitric acid was evaporated to dryness. This was dissolved in 2 ml of 0.4 M ammonium oxalate and heated. This solution was then transferred to a plating cell which contained a rotating platinum anode to insure a uniform electrodeposit of the uranium and the previously mentioned

aluminum disc as cathode. The voltage of the cell was held between 4 and 5 volts and the current adjusted by means of the electrode spacing to between 100 and 300 ma. In the 20 to 30 minutes allowed for plating, acceptable plates of from 0.1 mg/cm^2 to 0.5 mg/cm^2 of uranium were obtained over the plating area of the hats, which was around 1.30 cm^2 . To insure a sharp boundary between the plated uranium and the unplated portion of the hat, a method developed by Gibson was used.⁷ The aluminum hat was painted with a thin coat of 4A lacquer (Inter-Chemical Corporation, San Francisco, California) except for the area in the center where the uranium was to be deposited. After plating, the 4A lacquer, which had been baked on, was peeled off with the help of acetone, leaving a sharp boundary between the circle of deposited uranium and the rest of the disc. The area of the uranium circle was then determined by measuring the diameter. The amount of uranium deposited was determined by counting the uranium on the disc in an alpha-particle counter. Since the isotopic composition of the uranium had been determined previously by both a mass-spectrographic analysis and an alpha-pulse analysis of the various alpha energy groups, the specific activity of the uranium was known. From this and the geometry of the alpha counter it was possible to determine the number of atoms per cm^2 of target material. To test the uniformity of the uranium deposited, a disc with a small hole was placed over the hat and the uranium was counted for each of several positions of the disc on the hat. In most cases the deposit was found to be uniformly distributed. When the various counts did not agree to within 20%, the hat was rejected.

C. Target Assembly

The microtarget assembly used in these bombardments is shown in Fig. 1.^{8,13} The hat containing the electrodeposited target material together with an aluminum cover foil to catch recoils was placed in the microtarget. The external 24-Mev deuteron beam of the Crocker Laboratory 60-inch cyclotron was degraded to the desired energy by this cover foil and two additional degrading foils that were placed directly in the

ZN-1412

Fig. 1. Microtarget assembly. A, microtarget slot; B, microtarget;
C, collimator; D, foil holder; E, degrading foil.

microtarget assembly. The range-energy curves of Aron, Hoffman, and Williams²⁹ as interpreted by Foreman¹⁵ were used to calculate the energy of the deuterons striking the target. Since aluminum was able to withstand the heating due to the beam better than platinum, aluminum foils were used in most of the bombardments. For low-energy bombardment, for which aluminum could not provide sufficient degradation of the beam, a platinum foil was used. All foils were weighed and their dimensions measured so that the energy degradation of the beam could be calculated accurately from the thickness (in mg/cm^2).

The current I , and therefore the number of deuterons striking the target, was measured by means of the Faraday cup formed by the target-backing material and holder. This beam was recorded as a function of time on a Speedomax recorder and also integrated by the integrating capacitor of the cyclotron. To make sure all the recorded beam hit the target, a collimator of about 0.9 cm^2 cross-sectional area was placed in front of the microtarget. Also the beam pattern could be seen on the back of the aluminum hat after the bombardment and thus checked to see that all the beam had been intercepted by the target.

D. Chemical Procedures

Because of the large number of nuclear reactions, chemical purification of the desired elements was necessary. Owing to the low amount of radioactivity of many isotopes, it was not possible to separate the target into aliquots. Instead the various elements had to be separated from one another and then further purified. The chemical separation used, Fig. 2, is similar to that used by Gibson;⁷ the chemical purifications, unless otherwise noted, are taken from Meinke³⁰ or Lindner³¹. Because these purifications have been described in detail by others,^{7,13,14,17} it is unnecessary to repeat them here.

1. Chemical Separations

The electrodeposited uranium target, together with its aluminum hat and cover, was dissolved in a hydrochloric acid solution containing the appropriate carriers and tracers. In all cases zirconium and

Fig. 2. The chemical separation.

lanthanum carriers and neptunium tracer were added. A few drops of nitric acid were also added.

When only neptunium was separated, the procedure of Magnusson³² as modified by Gibson⁷ was used as follows:

Ferrous ion and hydrazine were added to the above solution in order to reduce the neptunium to the (IV) state. The solution was diluted to less than 6M acid concentration, and phosphoric acid was then added. The resulting zirconium phosphate precipitate, which carried the neptunium with it, was washed and then dissolved in a solution of hydrofluoric and nitric acids. Lanthanum carrier was then added to precipitate lanthanum fluoride, which carried the neptunium. The lanthanum fluoride was washed and dissolved in a solution of nitric and boric acids. Ammonium hydroxide was added and the precipitated lanthanum hydroxide, which carried the neptunium, was washed, dissolved in hydrochloric acid, and heated. The neptunium was reduced to the (III) state by adding hydroxylamine dihydrochloride, potassium iodide, and stannous chloride to the solution. After heating, this solution, now 1M in hydrochloric acid, was cooled and the neptunium extracted into 0.4M thenoyltrifluoroacetone (TTA) in benzene. The TTA solution was washed and then the neptunium was back-extracted into 8M hydrochloric acid. Finally the neptunium fraction was washed with benzene in order to remove any dissolved TTA and then electrodeposited³³ on a 2-mil platinum disc before counting. This neptunium separation was performed in as short a time as 15 minutes in the U^{238} bombardments when 7.3-minute neptunium was observed.

When fission products were taken out, the target solution was diluted with ice just after the target was dissolved in order to precipitate silver chloride. (See Fig. 2.) The silver chloride precipitate was then purified by the standard procedure.

The remaining solution was then treated as described previously to precipitate zirconium phosphate, which carried neptunium and protactinium. The neptunium was separated from the zirconium and protactinium when the neptunium was carried in the lanthanum fluoride precipitate. The zirconium and protactinium were eventually separated from each other

by means of an extraction with di-isopropyl ketone (DIPK); the protactinium was extracted into the DIPK while the zirconium remained behind in a hydrochloric acid solution. The protactinium and zirconium were then chemically purified.

The target solution, now void of silver, neptunium, protactinium, and zirconium, was treated with sodium hydroxide and sodium carbonate. This precipitated barium, strontium, yttrium, the rare earths, and occasionally cadmium. The remaining solution was saturated with hydrogen sulfide gas, which caused the cadmium and palladium to precipitate. This precipitate was partially dissolved in hydrochloric acid. The palladium remained as a residue and was purified by standard chemical procedures. The dissolved cadmium was then passed through a glass column packed with Dowex A-1 anion-exchange resin. The cadmium was later stripped off with 0.75M sulfuric acid and further purified by standard procedures. Meanwhile the precipitate containing barium, strontium, yttrium, rare earths, and sometimes cadmium was placed in an ice bath and saturated with hydrogen chloride gas. The residue remaining contained barium and strontium, which were then dissolved in water and reprecipitated as the chlorides. After being redissolved in water the barium was separated from the strontium by precipitation of the barium from a neutral buffered solution by addition of sodium chromate. The barium and strontium were then purified by standard procedures.

The chloride ~~supernatant~~ was then passed through the anion column used above. Yttrium and the rare earths came through in the concentrated hydrochloric acid fraction, uranium was eluted when 0.5M hydrochloric acid was passed through, and cadmium was stripped off by 0.75M sulfuric acid and purified by standard procedures.

The yttrium and rare earth fraction was further purified by precipitating the fluoride, dissolving it in a solution of nitric and boric acids and then precipitating the hydroxide. The hydroxide was dissolved in a small amount of hydrochloric acid and then placed on a column packed with Dowex-50 cation-exchange resin. The yttrium and rare earths were then separated from each other when ammonium lactate of which the pH was constantly increased was used as the eluting agent.

2. Determination of Yield

The chemical yields were determined by adding a known amount of standardized carrier for fission products and a known amount of standardized tracer for spallation products. These carriers and tracers were added to the original acid solution before the target was dissolved in it. This solution, together with the target, was heated and stirred vigorously to achieve equilibrium between the target activities and the carriers and tracers.

After the chemical separations described above, the chemical yield was determined by finding the ratio of the amount of carrier or tracer in the counted sample to the amount of carrier or tracer added to the original solution.

In the neptunium spallation products, the final electrodeposited sample was counted first in an argon-filled ionization chamber attached to a standard scaling circuit, with a 52% efficiency³⁴ to determine the relative amount of Np^{237} tracer in the sample. This worked satisfactorily except for the U^{234} bombardments and U^{235} foil bombardments, in which the alpha particle energy of the Np^{237} tracer could not be distinguished from that of the U^{234} in the target. In these cases, any U^{234} that happened to stay with the neptunium fraction would appear as Np^{237} tracer.

Therefore the following precautions were taken:

a. A large amount of Np^{237} tracer was added when practical, so that a few counts of U^{234} would not cause an appreciable error in the yield determination.

b. When the amounts of time and activity available were sufficient, a second TTA extraction was run on part of the neptunium fraction and the ratios of alpha counts to the number of counts on the nucleometer (mostly beta and K capture) were compared for both fractions. Since the TTA extraction would separate the uranium from the neptunium, the ratio of alpha counts to nucleometer counts would differ in the two fractions if the uranium were present.

c. Just before the neptunium end product was electrodeposited, a few drops of neptunium solution were placed on a 2-mil platinum disc and evaporated. The ratios of alpha counts to nucleometer counts on

the evaporated and electrodeposited discs were compared. Since it was unlikely that neptunium and uranium would electrodeposite with exactly the same efficiency, the ratios on the discs should differ if uranium were present.

d. These electrodeposited neptunium discs were cut into two or more pieces when sufficient activity was present. Since the activity was in general not deposited uniformly over the disc, it was possible for some parts of the disc to have relatively more neptunium and other parts of the disc to have relatively more uranium if both elements were present. This would again cause the ratios of alpha counts to nucleometer counts to differ in the different pieces of the disc.

In most of the bombardments, no uranium was observed in the neptunium fraction. In those few bombardments in which uranium was detected in the neptunium fraction, it was found that the neptunium cross sections calculated were so small that it was obvious that some uranium was present. In other words, when the neptunium was contaminated with uranium, the uranium was present in a relatively large amount, and therefore could be easily detected. When this happened and only one spallation cross section was determined, this determination gave a lower limit to the cross section. If more than cross section was determined, then relative cross sections were obtained.

For fission products, the final purified carrier was slurried with acetone or ethyl alcohol into a tared 2-mil aluminum shallow "hat." The slurries were then dried and weighed to determine the yield of the carriers. The rare earths, yttrium, and zirconium were weighed as oxides, cadmium as the sulfide, silver as the chloride, barium as the chromate, strontium as the carbonate, and palladium as the dimethylgloxime. After being weighed, the samples were coated with a few drops of Zapon lacquer (Atlas Powder Co., North Chicago, Ill.), diluted with ethyl acetate, to prevent any loss of material. These samples were then counted in a standard Geiger-Müller counter.

E. Counting Techniques

1. Nucleometer

The nucleometer³⁶ is a methane-flow windowless proportional counter. Because of its high counting efficiency, it was used for counting isotopes decaying by electron capture. Plateaus were taken for the isotopes counted in this investigation. It was found that an operating voltage of 4000 volts was satisfactory for all these isotopes. Counting efficiencies used for various isotopes are given in Table I.

Table I

Nucleometer counting efficiencies used		
<u>Isotope</u>	<u>Mode of decay</u>	<u>Counting efficiency</u>
Np ²³³	E.C.	0.60 ^a
Np ²³⁴	E.C.	0.65 ^b
Np ²³⁵	E.C.	0.70 ^c
Np ²³⁶	E.C., 43%; β^- , 57% ^d	0.90 ^e
Np ²³⁸	β^-	0.80 ^a

^a Assumed by Ref. 7.

^b Determined by Ref. 7.

^c Determined, this study, by using the alpha branching ratio of 3.5×10^{-5} as determined by Hoff.³⁸ Thomas¹⁰ has found an alpha branching ratio of 1.23×10^{-5} and a counting efficiency of 0.41. Recently Gindler, Huizenga, and Engelkemeir⁷⁶ have found an alpha branching ratio of 1.59×10^{-5} . Measurements by this author show that an alpha branching ratio of 1.23×10^{-5} requires a counting efficiency of about 0.25. However, a branching ratio of 3.5×10^{-5} corresponds to a counting efficiency of about 0.70. Since most of the counting efficiencies that have been determined for isotopes in this region are much nearer 0.70 than 0.25, the former value is used.

^d Ref. 37.

^e Determined, this study by using 57% for β^- branching.

2. Geiger-Müller Counter

The Geiger-Müller counter consists essentially of an end-window Amperex 100-C tube, filled with a mixture of chlorine and argon, attached to a standard scaler. Negatron emitters were counted in a thick-walled lead case surrounding the counting tube. There were five fixed positions or shelves in which the sample could be placed. When possible, Shelf 2, which was approximately 2 cm from the counting tube and for which the fixed geometry was 4.12%, was used.

The following counting corrections were necessary to convert counts per minute into disintegrations per minute. The details involved in determining the values of these corrections have been discussed by others.^{7,8,13}

a. Geometry and counting-efficiency correction (G). The geometry of Shelf 2 of the counter used was determined to be 4.12% by the counting of a weightless standard of known disintegration rate. It was assumed that all beta particles that entered the counting tube were counted. If this assumption is in error, the counting efficiency correction is automatically included in the geometry correction.

b. Coincidence correction (C). A coincidence correction is needed for high counting rates. This is because of the dead time of the counter after a beta particle has been counted. This correction is 0.45% per thousand counts per minute.³⁴

c. Air-window correction (AW). Some beta particles are absorbed by the air between the sample and the counting tube and by the mica window in the counting tube. To compensate for this, an "air-window" correction was made.¹³ This correction varies with the energy of the beta particle emitted.

d. Backscattering correction (BS). Some beta particles may be scattered by the aluminum backing material of the sample. The curves of Burtt were used to correct for this.³⁹

e. Self-scattering and self-absorption corrections (SSSA). Because of the sample thickness, some beta particles are scattered and absorbed by the sample itself. The empirical curves of Hicks and Gilbert⁴⁰ and of Stevenson, Gilbert, and Hutchin⁴¹ were used to correct for this. For

isotopes not included in these curves, the curves of Nervik and Stevenson,⁴² interpolated for summed atomic numbers, were used. This correction is also based on the decay scheme of the isotope and is discussed and shown in more detail by others.^{8,13,14} Even though a given sample may not be of uniform thickness, the average thickness was used to determine the correction.

Counts per minute (cpm) were converted to disintegrations per minute (dpm) by the formula

$$\text{dpm} = (\text{cpm} + C) (AW)/(BS) (SSSA) (G).$$

3. Decay Curves

a. Resolution. If one wishes to obtain the amount of activity of the various components of a decay curve, the decay curve must be resolved. In most cases in this work the curve was resolved by subtracting off the longer-lived components. When two half-lives were too similar for this to be accurate, a least-squares fit of the data was made on the IBM type 650 data-processing machine. The equation

$$\frac{dN}{dt} = Ae^{-\lambda_1 t} + Be^{-\lambda_2 t} + Ce^{-\lambda_3 t} + D;$$

where A, B, C, and D are constants to be determined, was used for this purpose.

b. Corrections for daughter activity. When the half-life of a daughter activity was the same as or shorter than that of the parent, the decay curve could not be resolved. Therefore, the following formula was used:

$$N_2 = [\lambda_1 / (\lambda_2 - \lambda_1)] N_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}),$$

where N_2 is the number of atoms of the daughter at time t , N_1^0 is the number of atoms of the parent present at the final chemical separation time, and λ_1 and λ_2 are the decay constants of the parent and daughter respectively. Curves for the counting corrections previously mentioned for fission products were made for the individual fission products. These curves contained all the counting corrections and also the correction for daughter activity at equilibrium.⁸

F. Cross-Section Calculations

1. Standard Cross-Section Formula

The formula used to calculate the number of atoms present at the end of the bombardment was

$$N = \text{dpm} / \lambda \cdot Y,$$

where N is the number of atoms present at the end of bombardment,

dpm is the number of disintegrations per minute of the isotope counted, extrapolated to the end of bombardment,

Y is the chemical yield of the isotope, and

λ is the decay constant of the isotope (in min^{-1}).

When the half-life of the activity considered was long compared with the length of the bombardment, the formula used to calculate the cross section was

$$\sigma = \frac{N}{5.73 \times 10^{34} \cdot n \cdot I} \text{ in cm}^2,$$

where n is the number of milligrams per cm^2 of target, and I is the integrated deuteron beam hitting the target (in microampere hours).

2. Correction for Decay During Bombardment

For activities with half-lives of the same order of magnitude as the length of bombardment, corrections had to be made for the decay of the activity during bombardment. The following formula was used for this purpose:

$$N = \frac{N_{t_0} \lambda t_b}{1 - \exp(-\lambda t_b)},$$

where N_{t_0} is the number of atoms present at the end of the bombardment,

t_b is the length of bombardment (in minutes), and

λ is the decay constant (in min^{-1}).

This formula requires that the target be hit by a steady beam during the bombardment. A record of the beam versus time was taken to see if this condition was met. If the beam was not steady, the various steady sections of the beam were considered separately and the following formula was used:

$$N = N_{t_0} \left[\frac{e^{\lambda t_b} \cdot \int_0^{t_b} I \, dt}{\int_0^{t_b} I e^{\lambda t} \, dt} \right]$$

3. Isotope Corrections

Since the U^{234} and U^{236} were not isotopically pure, cross sections due to other uranium isotopes were subtracted out. Except for the (d,γ) reactions, these corrections were quite small. The isotopic compositions of the various uraniums used were

U^{234} : 93.97% U^{234} , 4.33% U^{235} , 1.70% U^{238} ;
 U^{235} : 99.94% U^{235} , 0.038% U^{238} , 0.02% U^{234} ;
 U^{236} : 94.90% U^{236} , 4.52% U^{235} , 0.54% U^{238} , 0.04% U^{234} ;
 U^{238} : >99.9% U^{238} .

These compositions were verified by means of mass analyses.⁴³ To find the U^{234} content accurately, alpha-pulse analyses were run on the relative activities of the alpha groups of various energies.

4. Errors

The measured spallation reaction cross sections are accurate to within 25% and the measured fission cross sections to within 50% unless otherwise noted. The errors involved in the cross sections for the (d,γ) reactions vary and therefore are quoted individually. The errors varied because reactions involving the isotopic impurities in the uranium target in some cases produced a sizable amount of the measured (d,γ) reaction products. When this contribution was subtracted out, the final cross section for the (d,γ) reaction was reduced considerably. One of the principal errors in measuring the cross sections for spallation reactions was due to uncertainty of the counting efficiencies of electron-capture isotopes in the nucleometer. As previously mentioned, some of these counting efficiencies were assigned values. The 25% limits of error assigned to the spallation cross sections do not include errors due to counting efficiencies.

One of the principal errors in measuring the fission reaction cross sections was due to the errors involved in absolute beta counting.

The largest of these was in most cases due to the self-scattering and self-absorption correction. The errors involved in this correction probably varied from 5% to 30%.⁷ The remaining corrections used in converting counts per minute into disintegrations per minute were determined to within 10%.

The determination of the number of milligrams per cm^2 of the uranium deposited on the target should be good to within 10%. The accuracy of measurement of the cyclotron beam current is about 3%.⁴⁴

A 10% limit of error may be applied to the determination of chemical yield and 20% to the resolution of decay curves into the various components and their extrapolation back to the end of bombardment.

Other errors should be less than 10% except for a few specific cases among the fission products. These are discussed later under fission-product results.

III. RESULTS

A. Spallation Cross Sections

1. U^{235} Plus Deuterons

The spallation cross sections for U^{235} bombarded with deuterons are given in Table II.

The cross sections given for the (d,n) reaction include only the 22-hr Np^{236} isomer.

Cross sections in parenthesis are relative cross sections. They were normalized to the (d,n) excitation function curve.

The cross sections given for the (d,2n) reaction are upper limits. They were determined by assuming that the residual electron-capture activity remaining after the decay of the shorter-lived neptuniums was due to Np^{235} .

The excitation functions are shown in Fig. 3. The absolute limits of error excluding uncertainties in counting efficiencies are estimated at 25%.

Fig. 3. Spallation excitation functions for deuteron-induced reactions of U^{235} .

Table II

U^{235} + deuterons; spallation cross sections (in mb)				
E_d (Mev)	Reaction			
	(d,n)	(d,2n)	(d,3n)	(d,4n)
9.3	1.49	4.45	-	-
10.7	3.86	12.8	0.754	-
10.2-12.7	-	18.9	2.02	-
12.7-14.9	-	13.9	4.83	-
14.7	4.66	-	8.40	-
14.7	4.74	12.6	8.60	-
17.2	5.04	10.3	14.8	-
18.2-19.9	(5.29)	(8.83)	(24.7)	-
20.6	5.70	-	25.0	0.315
23.4	6.67	-	10.4	-
23.4	6.49	-	11.4	-
23.4	6.66	-	12.5	4.15
Q(Mev)	2.45	-3.20	-10.20	-16.22

Table III

U^{236} + deuterons; spallation cross sections (in mb)			
E_d (Mev)	Reaction		
	(d,2n)	(d,3n)	(d,4n)
8.7	2.99	-	-
11.9	43.7	-	-
14.8	31.9	28.8	-
16.7	23.7	45.6	-
18.9	24.7	56.4	1.94
20.7	23.2	45.5	3.36
22.6	17.7	21.7	7.02
23.4	16.5	-	11.8
Q(Mev)	-4.10	-9.70	-16.88

2. U²³⁶ Plus Deuterons

The spallation cross sections for U²³⁶ bombarded with deuterons are given in Table III.

The cross sections given for the (d,2n) reaction include the 22-hr Np²³⁶ isomer only. The Np²³⁶ activity was counted on the nucleometer. The activity of the Pu²³⁶ daughter was counted on the alpha-pulse analyzer. By this method the counting efficiency of the nucleometer for Np²³⁶ was determined to be 90% for a beta branching of 57%. The results of the nucleometer counts on the basis of this 90% counting efficiency are given.

The cross sections given for the (d,3n) reaction are upper limits. The decay of the Np²³⁵ product has been followed only long enough to indicate that the actual values of the cross sections for the (d,3n) reaction are within 50% of the values shown.

The excitation functions are shown in Fig. 4. The absolute limits of error excluding uncertainties in counting efficiencies are estimated at 25% except for the (d,4n) excitation function. Because a relatively large amount of the measured Np²³⁴ activity was due to the (d,3n) reaction on the isotopic impurity U²³⁵ in the target, the limits of error for the (d,4n) excitation function are estimated at 50%.

3. U²³⁴ Plus Deuterons

The spallation cross sections for U²³⁴ bombarded with deuterons are given in Table IV.

The cross sections given for the (d,n) reaction are upper limits, as they were determined by assuming the residual electron-capture activity was due to Np²³⁵.

The cross sections in parenthesis are relative cross sections. They were normalized to the (d,2n) excitation function.

The excitation functions are shown in Fig. 5. The absolute limits of error excluding uncertainties in counting efficiencies are estimated at 25%.

4. U²³⁸ Plus Deuterons

The spallation cross sections for U²³⁸ bombarded with deuterons are given in Table V.

Fig. 4. Spallation excitation functions for deuteron-induced reactions of U^{236} .

Fig. 5. Spallation excitation functions for deuteron-induced reactions of U²³⁴.

Table IV

$U^{234} + \text{deuterons; spallation cross sections (in mb)}$			
E_d (Mev)	Reaction		
	(d,n)	(d,2n)	(d,3n)
8.4	<2.8	1.41	-
12.3	-	23.6	-
13.6	-	31.2	-
14.8	12.9	-	2.33
16.0	-	(29.2)	(5.15)
16.4	12.1	29.2	8.33
17.1	-	-	12.9
17.5	13.3	26.1	16.8
18.9	-	(22.5)	(19.2)
20.0	-	(20.1)	(17.6)
20.6	-	18.9	14.9
23.4	13.4	12.3	7.44
Q(Mev)	2.22	-4.91	-10.90

Table V

$U^{238} + \text{deuterons; spallation cross sections (in mb)}$				
E_d (Mev)	Reaction			
	(d,n)+(d,p)	(d,n)	(d,2n)	(d,4n)
0-4.7	>0.38	-	-	-
1.8-6.3	>0.58	-	-	-
6.1-9.2	>9.3	-	2.93	-
9.7-13.0	>102	-	48.3	-
14.5-16.8	178	≤45	39.7	0.048
21.4-23.4	100	≤20	32.7	27.9
Q(Mev)	-	3.11	-3.30	-15.40

The cross sections given for the sum of the (d,n) and (d,p) reactions were obtained by counting the 2.33-day Np^{239} activity. When the Np^{239} was separated from the (d,p) product, U^{239} , before the 23.5-minute U^{239} had decayed completely, only lower limits on the cross sections for the (d,n) plus (d,p) reactions are given.

Upper limits on the cross sections for the (d,n) reaction were determined by two methods. One method was to separate the Np^{239} from the U^{239} twice. The first time the neptunium was separated before the U^{239} had completely decayed, and the second time after all the U^{239} had decayed. The sum of the cross sections for the (d,p) and (d,n) reactions was determined from the second separation and the individual cross sections for the (d,p) and (d,n) reactions were then determined from the first separation. The second method was to subtract out the cross section due to the (d,p) reaction on U^{238} as determined by Slater⁴⁵ from the sum of the cross sections for the (d,p) and (d,n) reactions as determined from the second separation. Since most of the Np^{239} was due to the (d,p) reaction, small errors in the determination of the cross sections for the (d,p) or the sum of the (d,p) and (d,n) reactions made large differences in the (d,n) reaction cross section. Therefore only the upper limit of the cross section for the (d,n) reaction was measurable.

The cross sections for the (d,2n) reaction were determined by alpha-pulse analysis of the Pu^{238} daughter after all the 2.10-day Np^{238} had decayed.

The cross sections given for the (d,4n) reaction include only the 22-hr isomer of Np^{236} . They were determined by alpha-pulse analysis of the Pu^{236} daughter after all the 22-hr Np^{236} had decayed. A beta-branching of 57% was used.

The absolute limits of error are estimated at 25%. The excitation functions are shown in Fig. 6 together with similar excitation functions determined by Crane.⁹

Fig. 6. Spallation excitation functions for deuteron-induced reactions of U^{238} .

B. Fission Cross Sections

1. Primary Fission Yields

A primary fission product is defined as the fission-product nucleus formed in the fission act before subsequent beta decays have taken place. For fission induced by low-energy neutrons the primary fission products have considerable neutron excess, and they beta-decay toward stability. For fission induced at high energies, the most probable fission product is often stable or even neutron-deficient.^{46,47}

For the studies undertaken in this work the primary fission products usually have considerable neutron excess, and beta-decay toward stability. Therefore the particular isotopic cross section measured experimentally usually includes essentially the total yield for a particular mass number. Thus if short-lived predecessors are allowed to decay to a beta-emitting nuclide near stability, this nuclide represents essentially the total chain yield for this particular mass number. However, some nuclides are not sufficiently close to stability to include most of the chain yield. A correction must be applied to the measured cross sections of these nuclides in order to obtain the total chain yield. One would prefer to measure the independent yields of each member of the various beta-decay chains. However, it is possible to measure by radio-chemical methods the primary yields of only a few scattered nuclides each of which is shielded from the beta decay of its isobars by a stable or long-lived isotope.

Such measurements were made on Y^{90} , La^{140} , and Pr^{142} and the results are given in the bottom of Table VI. Attempts were made to measure the primary fission yields of Y^{91} , Ag^{112} , and Pr^{143} . In each of these cases the ratio of primary fission yield to the total chain yield was so small that this primary-yield isotope was masked by the daughter of the beta-decaying isobaric parent. The fractions of the total chain yields of these isotopes are plotted in Fig. 7. Because of the small fraction, only an upper limit could be determined in some cases. The data and curves of Gibson,⁷ Thomas,¹⁰ and Vandebosch¹² are also plotted in Fig. 7. Fig. 8 is obtained by integrating the curve in Fig. 7.

Table VI

Fission cross sections (in mb)								
Isotope	Target and E_d							
	$U^{235} + d,$ 14.7 Mev		$U^{235} + d,$ 23.4 Mev		$U^{236} + d,$ 23.4 Mev		$U^{234} + d$ 23.4 Mev	
	σ_{meas}	σ_{corr}	σ_{meas}	σ_{corr}	σ_{meas}	σ_{corr}	σ_{meas}	σ_{corr}
Sr^{89}	18.4	18.6	28.1	28.4	22.8	23.0		
Sr^{91}	20.3	20.9	27.4		23.7			
Y^{91}	0		3.8		7.2			
$Sr^{91} + Y^{91}$	20.3	20.9	31.2	31.5	30.9	31.2		
Y^{93}			41.7	42.8	38.8	39.7		
Zr^{95}	40.3	41.2	53.8	54.9	83.0	83.8	73.9	76.0
Zr^{97}	49.8	52.0	55.4	57.6	85.5	88.2	69.6	74.9
Pd^{109}	7.42	7.51	12.9	13.0	28.3	28.3	16.3	16.7
Ag^{111}	9.30	9.40	21.7	22.0	27.7	28.0	22.1	22.6
Pd^{112}	9.42	9.52	17.8	18.6	26.5	27.4	18.2	19.6
Ag^{113}	5.83	6.00	14.1	14.5	17.3	17.5	15.4	16.1
Cd^{115}	12.7		27.7				24.0	
Cd^{115m}	≤ 0.941		≤ 1.52				≤ 2.58	
$Cd^{115} + Cd^{115m}$	13.6	13.9	29.2	29.8			26.6	27.4
Ba^{140}	25.8	34.4	30.1	46.4	60.5	68.2		
Ce^{141}	34.5	35.5	44.0	45.3	90.8	91.7		
Ce^{143}	25.1	26.4	30.3	34.5	64.0	66.0		
Ce^{144}	20.1	22.1	24.3	29.3	57.2	60.3		
Nd^{147}					33.1	33.8		
Eu^{156}	0.720	0.810	1.08	1.30	2.17	2.24		
Eu^{157}			1.17	1.62	1.63	1.72		
Total fission		930		1320		1690		1590
Total reaction		960		1350		1740		1620
Primary fission yields								
Y^{90}	<0.147		<0.609		<0.236			
La^{140}	1.56		3.49		5.56			
Pr^{142}	<0.704		<0.510		1.91

MU-13583

Fig. 7. Fraction of chain yield as a function of $(Z-Z_p)$.

MU-13584

Fig. 8. Integrated fraction of chain yield as a function of $(Z - Z_p)$.

The fractions of total chain yield are plotted according to the chain positions predicted on the basis of the assumption that the most probable primary fission product, Z_p , for a mass chain has the same neutron to proton ratio as the fissioning nucleus. Gibson has made a study of the charge distribution in medium-energy fission and concluded that the charge distribution in fission at these energies is not completely described either by the equal charge displacement noted at low energies^{48,49} or by the constant charge-to-mass ratio that may occur in very-high-energy fission.⁴⁶ He found, however, that at these energies the hypothesis of constant charge-to-mass ratio gave a better correlation.

The apparent fissioning nucleus is estimated from the best values for the center of symmetry of the fission-yield curves. The value of Z_p was first obtained by assuming a certain fissioning nucleus. This value of Z_p was then used with Fig. 8 to correct the fission-product cross sections. A new fission-yield curve was plotted. The cross sections were reflected about the center of symmetry which gave the best fitting curve. Twice the value of the mass number of this center of symmetry was equal to that of the apparent fissioning nucleus. The new Z_p was then used to correct the fission-product cross sections. This procedure was repeated until a constant value of Z_p was obtained.

2. Fission-Yield Curves

The fission-product cross sections for U^{235} , U^{236} , and U^{234} bombarded with deuterons of energy E_d are given in Table VI. Here σ_{meas} is the cross section (in millibarns) measured experimentally, while σ_{corr} is this cross section after the chain-yield corrections have been applied. The σ_{corr} is the cross section plotted in Fig. 9 through 12. The measured fission cross sections are accurate to within 50%. Errors due to the chain-yield corrections are small. The chain-yield corrections are less than 10% except for Ba^{140} , Ce^{144} , Eu^{156} , and Eu^{157} ; in these isotopes the corrections ranged from 20 to 50%.

The total fission cross sections in Table VI are obtained by summation of the fission mass yield curve. By summing the experimental fission and spallation cross section, one obtains the total reaction cross section. Theoretical cross sections for compound-nucleus formation

MU-13767

Fig. 9. Fission yield curve for 14.7 Mev deuteron-induced fission of U^{235} .

Fig. 10. Fission yield curve for 23.4 Mev deuteron-induced fission of U^{235} .

Fig. 11. Fission yield curve for 23.4 Mev deuteron-induced fission of U^{236} .

Fig. 12. Fission yield curve for 23.4 Mev deuteron-induced fission of U^{234} .

are given by Shapiro as a function of the nuclear radius parameter.²² In each case these experimental results correspond to a nuclear radius parameter of $r_0 = 1.5 \times 10^{-13}$ cm except for U^{235} bombarded with 23.4-Mev deuterons, which corresponds to $r_0 = 1.4 \times 10^{-13}$ cm. Within the experimental limits of error, these values of r_0 are the same.

C. Deuteron-Gamma Cross Sections

1. $U^{234}(d,\gamma)Np^{236}$

The cross sections for the (d,γ) reaction on U^{234} ($Q = 7.80$ Mev) are given in Table VII. The cross sections given include only the 22-hr Np^{236} isomer. They were determined by alpha-pulse analysis of the Pu^{236} activity after all the 22-hr Np^{236} had decayed. The excitation functions are shown in Fig. 13. Because of the low counting rate of the Pu^{236} and the large amount of Pu^{236} formed from the U^{235} and U^{238} isotopic impurities in the U^{234} , the limits of error are quite large. The limits of error given in Table VII were determined by estimating a 10% uncertainty in the number of counts per minute of Pu^{236} and 10% uncertainties in the cross sections which had to be subtracted from the total measured cross section. Since below 14 Mev most of the Pu^{236} was produced by the (d,n) reaction on U^{235} and above 20 Mev by the $(d,4n)$ reaction on U^{238} , cross sections for the (d,γ) reaction were measureable only between 14 and 20 Mev.

Table VII

$U^{234}(d,\gamma)Np^{236}$ Cross sections		
E_d (Mev)	σ_{meas} (mb)	σ_{corr} (mb)
14.8	0.0283	0.080 ± 0.06
16.4	0.570	0.355 ± 0.08
17.5	0.0613	0.372 ± 0.09
18.9	0.720	0.440 ± 0.10
20.0	0.794	0.380 ± 0.15

Fig. 13. (d, γ) excitation functions of U^{234} and U^{236} .

2. $U^{236}(d,\gamma)Np^{238}$

The cross sections for the (d,γ) reaction on U^{236} ($Q = 7.94$ Mev) are given in Table VIII. A least-squares method was used to resolve the 2.2-day neptunium from the 22-hr Np^{236} and 4.4-day Np^{234} . The cross section due to the 2.2-day neptunium formed from the (d,n) and $(d,2n)$ reactions on the isotopic impurity U^{238} was then subtracted out. The Pu^{238} activity was alpha-pulse analyzed after all the Np^{238} had decayed. Since the large Pu^{236} peak (5.75 Mev) partially overlapped the small Pu^{238} peak (5.49 Mev), only an upper limit for the Pu^{238} could be determined. The cross sections given in Table VIII are those determined by counting the neptunium activity except where cross sections measured by this means were larger than the upper limit determined by alpha-pulse analyzing the plutonium activity. In this case the latter values were used. The excitation functions are shown in Fig. 13. The limits of error given in Table VIII were determined by estimating a 10% uncertainty in the number of counts per minute of the Np^{238} activity and 10% uncertainties in the cross sections that had to be subtracted from the total measured cross section.

Table VIII

$U^{236}(d,\gamma)Np^{238}$ Cross sections		
E_d (Mev)	σ_{meas} (mb)	σ_{corr} (mb)
16.7	1.91	$0.67 \pm .31$
18.9	2.43	$1.38 \pm .34$
20.7	2.30	$1.20 \pm .31$
22.6	1.39	$0.61 \pm .21$
23.4	0.81	$0.15 \pm .15$

3. $U^{238}(d,\gamma)Np^{240}$

The cross sections for the (d,γ) reaction on U^{238} ($Q = 8.20$ Mev) are given in Table IX. These cross sections are given for the formation of the 1-hr Np^{240} isomer alone, for the formation of the 7.3-min Np^{240} isomer alone, and for the total (d,γ) reaction as determined from the sum

Table IX

$U^{238}(d,\gamma)Np^{240}$ Cross sections (in mb)			
Energy range in foil (Mev)	(d, γ), 61-min Np^{240}	(d, γ), 7-min Np^{240}	Total (d, γ)
1.8-6.3	1.57×10^{-3}	4.14×10^{-3}	5.71×10^{-3}
6.1-9.3	21.0×10^{-3}	35.1×10^{-3}	56.1×10^{-3}
9.7-13.0	0.440	0.490	0.930
14.5-16.8	0.448	1.180	1.630
21.4-23.4	0.115	0.636	0.751

of these two cross sections. The excitation functions are shown in Fig. 14.

The cross sections given for the formation of the 1-hr isomer have 50% limits of error. In all the neptunium decay curves an activity with a short half life of the order of 7 minutes was found in low abundance. This short-lived activity and its low abundance made positive identification of the 7.3-min isomer impossible. If this activity was due to the 7.3-min neptunium then the cross sections given are within a factor of 2 of the correct values. If this activity was due to an impurity then these cross sections are upper limits. The decay of all the neptunium samples was followed for several weeks and in all cases no impurities were observed. As a further check, U^{238} was bombarded with deuterons of energy less than 4.7 Mev. At this energy no detectable amount of 7.3-min or 1-hr neptunium should be formed, since the cross section for compound-nucleus formation is quite small. The experimental results confirmed this, as only the decay of 2-day neptunium could be detected.

Fig. 14. (d, γ) excitation functions of U^{238} .

IV. DISCUSSION

A. Spallation

1. Calculation of Level-Width Ratios

One may correlate fission and spallation competition in various isotopes by calculating their level-width ratios or branching ratios for neutron emission, Γ_n/Γ_t , and for fission Γ_f/Γ_t . The following treatment assumes that these ratios remain constant over the energy range considered and that the course of the reactions can be described by compound-nucleus theory.

According to the compound-nucleus theory of Bohr, the cross section for the (d,2n) reaction at the peak of the excitation curve may be expressed as

$$(1) \quad \sigma_{A-2,Z-1}(d,2n) = \sigma_c G_n(A,Z) G_n(A-1,Z) P(E,n)$$

where $\sigma_{A-2,Z-1}$ is the cross section for the (d,2n) reaction on the original nucleus consisting of A-2 nucleons and Z-1 protons.⁵⁰

$$G_n(A,Z) = \left(\frac{\Gamma_n}{\Gamma_t} \right)_{A,Z}$$

refers to the compound nucleus, where Γ_n is the level width or branching ratio for neutron emission and Γ_t is the "total" width. Thus G_n is the probability of a neutron's evaporating from a particular nucleus.

Similarly, $G_{n(A-1,Z)}$ refers to the probability of evaporation of a neutron from the nucleus remaining after one neutron has evaporated from the compound nucleus. Likewise, $P(E,n)$ is the probability of evaporating exactly n neutrons from the compound nucleus. For this equation, $n = 2$. Thus at the maximum cross section $P(E,n)$ approaches unity, because in general the energy at this maximum cross section is only around 6 Mev greater than the Q of the reaction. Here σ_c refers to the cross section for formation of the compound nucleus at the energy of the bombarding deuteron.

Similar equations may be derived for other (d,xn) and (α ,xn) reactions. As an example, for the (d,3n) reaction on U^{235} , we have

$$(2) \quad \sigma_{U^{235}}(d,3n) = \sigma_c G_{nNp^{237}} G_{nNp^{236}} G_{nNp^{235}} P(E,3).$$

If we restrict ourselves to the energy at which this cross section is a maximum, $P(E,3)$ approaches but does not necessarily equal unity. As a first approximation $P(E,3)$ is set equal to unity. Also at this energy, most of the reaction proceeds through compound-nucleus formation, as is required by this equation. The equation may be further simplified by defining geometric mean values as follows:

$$\bar{G}_{A,A-1} = \sqrt[2]{G_A G_{A-1}} = \bar{G}_{A-1/2},$$

$$\bar{G}_{A,A-1,A-2} = \sqrt[3]{G_A G_{A-1} G_{A-2}} = \bar{G}_{A-1},$$

$$\bar{G}_{A,A-1,A-2,A-3} = \sqrt[4]{G_A G_{A-1} G_{A-2} G_{A-3}} = \bar{G}_{A-1 1/2}.$$

The equation for $\sigma_{U235}(d,3n)$, when solved for $\bar{G}_n$, becomes

$$(3) \quad \bar{G}_{nNp236} = \sqrt[3]{\frac{\sigma_{U235}(d,3n)}{\sigma_c}}.$$

The calculated values of $\bar{G}_n$ or $1-\bar{G}_n$ can next be plotted against $\bar{A}$. When $(1-\bar{G}_n)$ was plotted, it was found that most existing data for (d,xn) and (α,xn) reactions, where $x = 2, 3$, or 4 , fell within experimental error, on a straight line for a given Z of the compound nucleus. The lines for the various Z 's are parallel to one another, showing that the mass effect was the same for isotopes with Z between 93 and 96. No correction is necessary between even-even or even-odd nuclei. The lines of constant Z became closer to one another as Z increased. The quantity $(\partial \bar{G}_n / \partial Z)_A$ varied from -0.20 at $Z = 91$ to -0.04 at $Z = 96$; $\bar{G}_n$ was increased by 0.04 for each neutron added regardless of Z . These effects are valid only for the range for $Z = 91$ to $Z = 97$, where the isotope considered is reasonably close to the beta-stability line. The mean probability of fission, $\bar{\Gamma}_f / \bar{\Gamma}_t$, as approximately by $1-\bar{G}_n$, is tabulated in Table X and plotted against A of the compound nucleus in Fig. 15 for constant Z of the compound nucleus.

Table X

Target	Reaction	Z	Mean level-width ratios			Reference
			$\bar{A}$	$\left(\frac{\bar{\Gamma}_f}{\bar{\Gamma}_t}\right)$	$\left(\frac{\bar{\Gamma}_f}{\bar{\Gamma}_n}\right)$	
Th ²³²	d, 4n	91	232-1/2	0.47	0.9	9
Th ²³⁰	α , 4n	92	232-1/2	0.70	2.3	12
Th ²³²	α , 4n	92	234-1/2	0.56	1.3	15
U ²³³	d, 3n	93	234	0.80	4.0	7
U ²³³	d, 2n	93	234-1/2	0.78	3.5	7
U ²³⁴	d, 3n	93	235	0.76	3.2	This work
U ²³⁴	d, 2n	93	235-1/2	0.76	3.2	"
U ²³⁵	d, 3n	93	236	0.74	2.8	"
U ²³⁵	d, 2n	93	236-1/2	0.75	3.0	"
U ²³⁶	d, 3n	93	237	0.66	1.9	"
U ²³⁶	d, 2n	93	237-1/2	0.64	1.8	"
U ²³⁸	d, 4n	93	238-1/2	0.64	1.8	"
U ²³⁸	d, 2n	93	239-1/2	0.59	1.4	"
U ²³³	α , 4n	94	235-1/2	0.87	6.7	10
U ²³³	α , 3n	94	236	0.90	9.0	10
U ²³³	α , 2n	94	236-1/2	0.88	7.3	10
U ²³⁴	α , 4n	94	236-1/2	0.84	5.2	11
U ²³⁵	α , 4n	94	237-1/2	0.80	4.0	12
U ²³⁵	α , 3n	94	238	0.80	4.0	12
Np ²³⁷	d, 3n	94	238	0.78	3.5	12
Np ²³⁷	d, 2n	94	238-1/2	0.86	6.1	12
U ²³⁵	α , 2n	94	238-1/2	0.82	4.6	12
U ²³⁶	α , 4n	94	238-1/2	0.77	3.3	12
U ²³⁸	α , 4n	94	240-1/2	0.65	1.9	51
Pu ²³⁸	d, 3n	95	239	0.82	4.6	8
Pu ²³⁸	d, 2n	95	239-1/2	0.87	6.7	8
Pu ²³⁹	d, 3n	95	240	0.77	3.3	7
Np ²³⁷	α , 3n	95	240	0.78	3.5	7
Np ²³⁷	α , 2n	95	240-1/2	0.82	4.6	7
Pu ²³⁹	d, 2n	95	240-1/2	0.80	4.0	7
Pu ²⁴⁰	d, 3n	95	241	0.77	3.3	8
Pu ²⁴⁰	d, 2n	95	241-1/2	0.73	2.7	8
Pu ²³⁸	α , 4n	96	240-1/2	0.88	7.3	50
Pu ²³⁸	α , 2n	96	241-1/2	0.84	5.2	50
Pu ²³⁹	α , 4n	96	241-1/2	0.84	5.2	50
Pu ²³⁹	α , 3n	96	242	0.80	4.0	50
Pu ²³⁹	α , 2n	96	242-1/2	0.76	3.2	50
Pu ²⁴⁰	α , 4n	96	242-1/2	0.82	4.6	18
Pu ²⁴⁰	α , 3n	96	243	0.81	4.3	18
Pu ²⁴⁰	α , 2n	96	243-1/2	0.71	2.4	18
Pu ²⁴²	α , 4n	96	244-1/2	0.70	2.3	50
Pu ²⁴²	α , 2n	96	245-1/2	0.34	0.5	50
Am ²⁴³	α , 4n	97	245-1/2	0.68	2.1	16
Am ²⁴³	α , 2n	97	246-1/2	0.67	2.0	16

MU-16153

Fig. 15. Fission probability.

This method of plotting the data seems quite satisfactory. It is sensitive enough to show the general trend of the fissionability, but insensitive enough so that experimental errors do not effect it too greatly. For example, a recent change in the measured peak value of the $(\alpha, 2n)$ reaction on Pu^{240} from 23 mb to 41 mb changed $1-\bar{G}_n$ from 0.79 to 0.71. If the measured energy of the maximum of the $(d, 2n)$ reaction on Np^{237} were changed from the present value of 15 Mev to the more probable energy of 12 Mev and the maximum cross-section value left unchanged, $1-\bar{G}_n$ would change from 0.86 to 0.78.

A more sensitive method is to plot $\bar{\Gamma}_f/\bar{\Gamma}_n$ as approximated by $(1-\bar{G}_n)/\bar{G}_n$ against $\bar{A}$. Such a plot is shown in Fig. 16. It should be noted that the linear relationships shown in Fig. 15 should not be preserved in Fig. 16. Plots of $\bar{\Gamma}_f/\bar{\Gamma}_t$ and $\bar{\Gamma}_f/\bar{\Gamma}_n$ against $Z^2/\bar{A}$ failed to show a $Z^2/\bar{A}$ dependence for the fissionability of nuclei with $Z^2/\bar{A}$ greater than 37.0.

It should also be stressed that the lines of constant Z in Fig. 13 are entirely empirical and are valid approximations only for the region in which experimental data show them to be applicable. As these lines of constant Z approach higher or lower fissionabilities, they must depart from linearity. Otherwise there would be isotopes with fission probabilities of greater than one or less than zero.

In a few cases we may calculate the individual level-width ratios instead of the mean level-width ratios. For similar excitation energies, the ratio of cross sections is calculated as follows: for the $(d, 2n)$ reaction on U^{234} and the $(d, 3n)$ reaction on U^{235} we have

$$(4) \quad \frac{\sigma_{\text{U}^{234}}(d, 2n)}{\sigma_{\text{U}^{235}}(d, 3n)} = \frac{G_{\text{nNp}^{236}} G_{\text{nNp}^{235}}}{G_{\text{nNp}^{237}} G_{\text{nNp}^{236}} G_{\text{nNp}^{235}}} \frac{\sigma_c \text{U}^{234}}{\sigma_c \text{U}^{235}} = \frac{1}{G_{\text{nNp}^{237}}} \cdot \frac{\sigma_c \text{U}^{234}}{\sigma_c \text{U}^{235}}.$$

Here $G_{\text{nNp}^{237}}$ may be determined by substituting into the expression the peak cross sections for the $(d, 2n)$ reaction on U^{234} and the $(d, 3n)$ reaction on U^{235} together with the σ_c 's at these energies. This calculation gives a value of 0.30 for $G_{\text{nNp}^{237}}$. Similar calculations give values of 0.25 for $G_{\text{nNp}^{236}}$ and 0.47 for $G_{\text{nNp}^{238}}$. These values are in good agreement with the corresponding mean level widths.

MU-16154

Fig. 16. Ratio of fission probability to neutron evaporation probability.

2. Prediction of Cross Sections

One may now estimate the $\bar{G}_n$ for various isotopes, with the result that the cross sections for given reactions may be predicted by solving, for example, equation (3) for σ rather than $\bar{G}_n$.

For this purpose an equation for the lines in Fig. 13 was determined. For $Z = 93$ and $Z = 95$ the lines may be represented by the equation $1 - \bar{G}_n = 0.12Z - 0.04A - 1.02$. Then, for example, the cross section for the (d,2n) reaction on U^{235} may be calculated as follows:

$$(5) \quad \sigma_{235}(d,2n) = (0.04A - 0.12Z + 2.02) (0.04A - 0.12Z + 1.98) \sigma_c,$$

where A and Z refer to the compound nucleus and thus are 237 and 93 respectively. Also, σ_c may be found from data by Shapiro²² for deuteron reactions and by Blatt and Weisskopf²³ for helium-ion reactions. To determine the energy at the maximum cross section the following empirical approximations were used: For (d,2n) reactions, this peak energy was taken as $Q + 8.1$ Mev; for (d,3n) and (d,4n) reactions as $Q + 9.2$ Mev. For all alpha reactions, $Q + 9.0$ Mev was used to find the peak energy. The results of these calculations together with the experimental results are given in Table XI.

Table XI

TABLE XI

Maximum cross sections (in mb)

Isotope	Reaction												
	$\sigma(d,2n)$		$\sigma(d,3n)$		$\sigma(d,4n)$		$\sigma(\alpha,2n)$		$\sigma(\alpha,3n)$		$\sigma(\alpha,4n)$		
	exp	calc	exp	calc	exp	calc	exp	calc	exp	calc	exp	calc	
U^{233}	16	17.3	14	12.8	-	-	5	9.0	1.1	1.54	.4	.9	0.1
U^{234}	33	33.2	20	21.1	-	-	-	13.8	-	3.84	1.0	0.42	
U^{235}	19	20.6	26	32.4	-	-	16	18.2	8	6.76	2.5	1.20	
U^{236}	42	41.5	56	47.4	-	-	-	21.7	-	12.1	-	2.55	
U^{238}	48	43.2	-	84.0	31	36.6	-	33.4	-	26.1	21	8.9	
Pu^{238}	11	24.0	10	6.68	-	-	15	13.1	24	3.84	.4	0.41	
Pu^{239}	29	25.4	22	13.1	-	-	$12\frac{1}{2}$	18.2	$4\frac{1}{2}$	6.54	1.0	1.20	
Pu^{240}	41	42.4	20	22.3	-	-	41	22.2	7	11.4	1.6	2.66	
Pu^{242}	-	-	-	-	-	-	103	33.4	-	25.8	8	8.35	

A comparison between the experimental and calculated cross sections for (d, xn) reactions on uranium shows that all these cross sections agree within experimental error. The calculations predict correctly that the $(d, 2n)$ cross section is larger than the $(d, 3n)$ cross section on U^{234} and that, conversely, on U^{235} and U^{236} the $(d, 3n)$ cross sections are larger than the $(d, 2n)$ cross sections. The calculations predict that the $(d, 2n)$ cross section on U^{234} is greater than that on U^{235} and that there is an orderly increase of $(d, 3n)$ cross sections with increasing A . Unfortunately, the calculations do not do so well on other elements. There is only one point in Fig. 15 that failed to fall on the constant- Z lines within experimental error. It is the $(\alpha, 2n)$ reaction on Pu^{242} . The calculated cross section is quite poor for this reaction.

3. Experimental Results

The increase in the spallation cross section with increasing mass number is clearly seen from the cross sections for the $(d, 2n)$ and $(d, 3n)$ reactions. The only unusual excitation function is the $(d, 2n)$ reaction on U^{234} ; it peaks at a higher energy (14 Mev) than the other $(d, 2n)$ cross sections (12 Mev) and is larger than the cross section for the $(d, 2n)$ reaction on U^{235} . This is because the absolute value of the Q for the $(d, 2n)$ reaction on U^{234} is almost 2 Mev greater than on U^{235} . Therefore, one would expect this cross section to peak almost 2 Mev later in U^{234} . Consequently, the cross section will be larger, since most cross sections for $(d, 2n)$ reactions on uranium are cut down by the Coulomb barrier. In U^{234} at the peak energy of 14 Mev, Shapiro's²² value of the total cross section is 660 mb and in U^{235} at the peak energy of 12 Mev this value is 330 mb. The calculated $\bar{G}_n$ for the $U^{235}(d, 2n)$ reaction is 31% less than the $\bar{G}_n$ for the $U^{234}(d, 2n)$ reaction. Thus the maximum cross section for the $(d, 2n)$ reaction on U^{234} is calculated to be greater than on U^{235} . In Table XI the cross section calculated for the $(d, 2n)$ reaction on U^{234} is 33.2 mb and for U^{235} this cross section is 20.6 mb. The respective experimental cross sections are 33 mb and 19 mb. This effect also causes the peak cross section of $(d, 2n)$ reaction to be less than that of the $(d, 3n)$ reaction.

The (d,n) excitation functions are flat. This must be due primarily to direct interactions since the compound-nucleus theory predicts that the result of increased energy should be that $(d,2n)$ and $(d,3n)$ reactions compete more strongly with the (d,n) reaction and thus lower the cross section. The shape and magnitude of the (d,n) excitation functions are similar to those found by others^{7,8} and predicted by Peaslee⁵² for stripping reactions in which fission is not competing.

The other (d,xn) excitation functions indicate compound-nucleus formation as the principal process and are similar in shape to other (d,xn) reactions in this region of fission competition.^{7,8} The tails are attributed to direct interactions.

B. Fission

1. Fission-Yield Curves

An examination of the fission-yield curves shows that the Cd^{115} points are consistently higher than the other points in the valley. Because of the uncertainties in the determination of the fission yield curve, one cannot state whether this is actually the case. One possible cause of a hump in the valley could be that an exactly symmetric fission of an even-even nucleus will produce two identical fission fragments rather than two different fission fragments.⁵³ This will tend to give twice the fission yield to this particular isotope. The emission of neutrons during and after the fission process will tend to smear out this increased yield over several mass numbers making the effect less noticeable. It is also noted that the Pd^{109} and Ag^{113} points are lower than the other points in the valley. Characteristics such as these are probably due to uncertainties in the data. The use of a poorly determined half-life or counting corrections can easily account for consistently high or low points. Clearly, better data are required before any quantitative interpretation can be made of the fission-yield curves.

2. Fission Asymmetry

For the purpose of classifying fission as relatively asymmetric or symmetric, let us group fission reactions according to whether their

valley-to-peak ratio is less than (asymmetric), about the same as (intermediate), or greater than (symmetric) that predicted by the equation of Jones, Timmick, Phaeler, and Handley.⁵⁴ This is necessary in order to make the comparison between the various reactions, because the data for some reactions do not fall in the same energy region as those for other reactions.

Gibson⁷ has plotted valley-to-peak ratio against a function of the excitation energy for the data available for given fission reactions. The equation of Jones, et.al.,

$$\frac{Y_{\min}}{Y_{\max}} = 2.8 \exp \left[\frac{-2.9}{T} \right],$$

where $Y_{\min}$ = yield of a mass in the "valley" of the mass yield curve,
 $Y_{\max}$ = yield of a mass at one of the "peaks" of the mass yield curve,

T = nuclear temperature in Mev,

was plotted on the same graph. The reaction is then classified as symmetric, intermediate, or asymmetric according to whether the points lie above, close to, or below the curve given by the above equation. If the equation is valid, the experimental points for a given reaction should lie in a line parallel to the curve. Gibson has pointed out that where it is possible to check this, the agreement is poor. However, the equation is a convenient method for classifying the relative degree of fission asymmetry. The results of such a grouping including recent data are given in Table XII.

Table XII

Fission asymmetry		
Asymmetric	Intermediate	Symmetric
$U^{235} + \gamma$ (Ref. 54)	$Th^{230} + p$ (Ref. 56)	$U^{233} + d$ (Ref. 7)
$U^{234} + d$ (This work)	$Th^{232} + n$ (Ref. 54)	$U^{233} + \alpha$ (Ref. 10)
$U^{235} + p$ (Ref. 54)	$Th^{232} + p$ (Ref. 54)	$U^{235} + \alpha$ (Ref. 12)
$U^{235} + d$ (This work)	$Th^{232} + \alpha$ (Ref. 54)	$Np^{237} + \alpha$ (Ref. 7)
$U^{236} + d$ (This work)	$U^{233} + p$ (Ref. 56)	$Pu^{239} + d$ (Ref. 7)
$U^{238} + p$ (Ref. 54)	$U^{235} + n$ (Ref. 54)	$Pu^{240} + d$ (Ref. 8)
$U^{238} + d$ (Ref. 55)	$U^{238} + n$ (Ref. 54)	$Pu^{238} + \alpha$ (Ref. 14)
	$U^{238} + \alpha$ (Ref. 13)	$Pu^{239} + \alpha$ (Ref. 17)

Observe in Table XII that reactions induced by heavy particles lie towards the right and those induced by light particles towards the left. Thus no reactions induced by helium ions are classified as asymmetric; also, no reactions induced by neutrons or protons are classified as symmetric. In the left-hand column the compound nuclei are, in general, of lower atomic number than those in the right-hand column. Only reactions involving uranium isotopes are classified as asymmetric, while all compound systems classified as symmetric have an atomic number of 93 or greater. However, there is no good correlation with the atomic number, since all reactions involving thorium, the lowest-atomic-number nucleus in the table, are classified as intermediate. Similar compound systems fall close to one another in the table: ($U^{234} + d$) and ($U^{235} + p$) are both classified as asymmetric, ($U^{235} + n$) and ($Th^{232} + \alpha$) are both classified as intermediate, and ($Pu^{239} + d$) and ($Np^{237} + \alpha$) are both classified as symmetric.

One would normally expect that in the symmetric case the fission occurs before neutron emission, and that in the asymmetric case the fission is preceded by at least one or two neutron evaporations. Thus the degree of asymmetry should be related to the fissionability of the various nuclei. This is not always so. For example, at an excitation

energy of about 32 Mev, ($\text{Th}^{232} + \alpha$) has a peak-to-valley ratio of about 1.8, while at this same excitation energy, ($\text{U}^{236} + d$) has a peak-to-valley ratio of 5.9. Table XIII shows the peak-to-valley ratio and fission cross section for deuteron-induced fission in uranium isotopes determined in this work, together with those determined by Gibson⁷ for U^{233} and by Stevenson, et al.⁵⁵ for U^{238} .

Table XIII

Asymmetry and fission cross sections for deuteron-induced fission of uranium isotopes					
	Reaction				
	$\text{U}^{233} + d$	$\text{U}^{234} + d$	$\text{U}^{235} + d$	$\text{U}^{236} + d$	$\text{U}^{238} + d$
E_d (Mev)	23	23	23	23	20
peak/valley	1.0	4.6	3.7	5.9	4.9
σ_{fiss} (mb)	1860	1590	1320	1690	1030

Within the accuracy of the data, the fission asymmetries of these reactions, except for ($\text{U}^{233} + d$), are consistent with one another. Although, for the reasons discussed above, U^{233} should be more asymmetric than the other uranium isotopes, the great difference obtained in the asymmetry is not presently explainable. Comparison of fission-yield curves of U^{233} , U^{235} , and U^{238} bombarded with helium ions indicates that there is no significant difference in the fission asymmetry of these isotopes.⁵⁷ Thus, except in deuteron-induced U^{233} fission, the mass number alone appears to have no strong effect on fission asymmetry.

3. Total Fission Cross Sections

The total fission cross sections listed in Table VI were obtained by summing the area under the mass yield curves in Fig. 9 through 12. Within the limits of error, the total fission cross sections for U^{234} , U^{235} , and U^{236} bombarded with 23-Mev deuterons are the same.

The total reaction cross sections were obtained by adding the sums of the spallation cross sections to these total fission cross sections. These total reaction cross sections given in Table VI were

compared with theoretical cross-section curves for compound-nucleus formation as determined by Shapiro.²² These curves are given as a function of the nuclear radius parameter r_0 , where the nuclear radius is given by the relation $R = r_0 A^{1/3}$. For each of the three nuclei studied, the experimental results agree with a value for r_0 of 1.5×10^{-13} cm. This is in agreement with the experimental results of others at this laboratory.^{7,57}

C. Deuteron-Gamma Reactions

In general the excitation functions for the (d, γ) reactions given in Fig. 13 and 14 rise to a maximum of about a millibarn at bombarding energy between 15 and 20 Mev. This is similar to the behavior found in (p, γ) reactions on Bi²⁰⁹ by Kelly²⁴, and in (α , γ) reactions on Xe¹³⁶ by Fairhall²⁵, and on Ni⁵⁸ by Morinaga²⁶. The cross section for the (p, γ) reaction on Bi²⁰⁹ rises to a peak value of about 0.9 mb at a proton energy of 16 Mev. The cross section for the (α , γ) reaction on Ni⁵⁸ has a peak value of 0.40 mb at an alpha energy of about 15 Mev and on Xe¹³⁶ the peak value is 0.06 mb at about 21 Mev. Thus the cross section for this type of reaction does not appear to be decreased by fission competition. Kelly compared his results with the inverse reaction by means of detailed balancing, and found that the calculated (p, γ) reaction cross section is only 1% of his observed value at a 15-Mev proton energy. On comparing the (n, γ) reaction with the observed (p, γ) cross section at 15 Mev, which is well above the proton potential barrier, Kelly found that the estimated cross section for the (n, γ) reaction on bismuth was ten times the observed (p, γ) value. Thus the observed (p, γ) cross section is in fair agreement with other experimental data since large uncertainties were involved in estimating the cross sections for the (γ ,p) and (n, γ) reactions.

If one uses a value of 1 mb for the (d, γ) cross section on U²³⁶ at a deuteron energy of 18 Mev, one calculates by detailed balancing, neglecting statistical weights, a value of 8 μ b for the (γ ,d) cross section on Np²³⁸ at a gamma-ray energy of 24 Mev. Such a small value,

for a (γ, d) cross section is consistent with the fact that (γ, d) cross sections large enough to be positively measured have not been reported in the literature. If the (d, γ) reaction occurred by compound-nucleus formation, one would expect the cross section for the (d, γ) reaction to decrease at energies at which the (d, n) and $(d, 2n)$ reactions compete with it. The cross sections for the (d, γ) reaction on U^{234} , U^{236} , and U^{238} all peak at a higher energy than the corresponding cross sections for the $(d, 2n)$ reaction. Thus it appears that simple compound-nucleus theory does not account for the occurrence of the peak of the (d, γ) reaction at this high an energy. However, this energy is consistent with the experimental results of the inverse reaction. For instance, the cross section for the (γ, p) reaction on Pb^{208} appears to have a maximum at about 22 Mev and the (γ, pn) reaction on Pb^{208} at about 26 Mev.⁵⁸ One would then expect the (γ, d) reaction to have its peak cross section at about 24 Mev. The threshold for the (γ, p) reaction on Pb^{208} is 8 Mev and that for the (γ, d) reaction 10 Mev. Thus the peak value of the cross section appears to be 14 Mev above the threshold for (γ, d) and (γ, p) reactions. The thresholds for the (γ, d) reactions on U^{234} , U^{236} , and U^{238} are all about 8 Mev. If the cross sections for the (γ, d) reactions peaked at 14 Mev above these thresholds, they would peak at 22 Mev. From the principle of detailed balancing it follows that cross sections for these (d, γ) reactions should peak at 14 Mev. This energy is only from 2 to 5 Mev less than the energy at the peak of the cross sections for the (d, γ) reactions on U^{234} , U^{236} , and U^{238} , but 1 to 2 Mev higher than the energy at the peak of the cross sections for the corresponding $(d, 2n)$ reactions.

The method of detailed balancing is based on compound-nucleus formation. Experimental evidence on photonuclear reactions indicate that a large portion of the reaction does not proceed by compound-nucleus formation. Hirzel and Waffler⁵⁹ found that in many cases the relative yield of (γ, n) and (γ, p) reactions did not agree with compound-nucleus theory. Diven and Almy⁶⁰ found that for the (γ, p) reaction on silver, low-energy protons are emitted with equal probability at all angles, as expected from compound-nucleus theory, while high-energy protons are emitted with a pronounced angular asymmetry. Curtis et al.⁶¹ have obtained

similar results for rhodium. This angular asymmetry can be explained by an absorption of the light quantum by a proton at the nuclear surface, which results in the emission of the proton before the energy of the proton can be shared with the rest of the nucleus.²³ Since a compound nucleus is not formed in this reaction, the principle of detailed balancing is not applicable. One might expect a similar situation in (γ, d) reactions. The observed cross sections for the nuclear photoeffect are much larger than predicted by compound-nucleus theory. The main difficulty appears to be in the low gamma-absorption probability predicted by theory. Theory assumes that the main contributions to the gamma-ray absorption cross section come from the magnetic dipole and electrical quadrupole absorption only. Blatt and Weisskopf point out that at higher energy the absorption is strongly increased by electric dipole absorption. This brings the theory into qualitative agreement with experimental results.

Similarly the predicted cross sections for the inverse reactions are brought into qualitative agreement with the observed cross sections when the low gamma emission probability is raised by increasing electric dipole transitions.

Several mechanisms have been proposed to explain the experimental cross sections for (p, γ) and (α, γ) reactions. Cohen⁶² has demonstrated that the (p, γ) reaction is the result of a single high-energy gamma transition to levels within the particle binding energy of the ground state. Cohen has then shown that the experimental ratios of the probabilities for photon emission and particle emission for bismuth bombarded with 10-Mev and 22-Mev protons do not agree with compound-nucleus theory. However, theoretical values for a direct single-particle transition to bound states agree with experimental results.

Morinaga²⁶ has derived a formula from known gamma-ray widths obtained from photonuclear absorption cross sections. Only the gamma rays which led to an excited state of the residual nucleus with energy lower than the sum of its photon threshold plus 1.6 Mev were considered to be responsible for (α, γ) reactions. For energies higher than this value, the successive proton emission was considered to become

predominant. This formula shows fair agreement with the experimental (α, γ) cross sections on Ni^{58} , indicating the existence of the giant resonance. A similar calculation for the $\text{Ni}^{60}(\text{p}, \gamma)\text{Cu}^{61}$ cross section also agrees with the experimental cross section.

Ball, Fairhall, and Halpern²⁵ state that "it is sometimes assumed that the high energy photon would be due to an electric dipole transition of the incoming particle to a bound state. The fact that alpha capture cross sections are about the same size as proton capture cross sections is not consistent with such a view since alphas do not have as much of a dipole moment with respect to a typical nucleus as protons do." They then postulate that in most nuclei, levels become available from which it is likely that photons of about 20 Mev are emitted. As the excitation energy of a nucleus is increased well beyond 20 Mev, capture occurs only if a sufficiently long cascade of photon emission follows the emission of the 20-Mev photon. The probability that such a cascade is long enough to bring the excitation energy down below the particle-emission threshold decreases rapidly as the excitation energy increases. Madsen and Henley⁷⁷ have had some success using the compound nucleus theory and giant resonance behavior to account for (α, γ) reactions in xenon and nickel.

The (d, γ) reaction probably does not proceed by compound nucleus mechanism and for the most part only one high-energy photon is emitted. It appears that the unexpectedly large cross sections for the (p, γ) , (d, γ) , and (α, γ) reactions are related in some degree to the giant resonance occurring in photonuclear reactions. Before one can conclude with certainty the mechanism for these reactions, additional data are necessary.

D. Summary

By assuming compound-nucleus formation one can correlate the fissionabilities of various nuclei and roughly predict spallation cross sections in the energy region of this study.

The $(\text{d}, 2\text{n})$, $(\text{d}, 3\text{n})$, and $(\text{d}, 4\text{n})$ reactions indicate compound-nucleus formation as the principal process. These reactions are

sensitive to the magnitude of the fission reaction and indicate the relative fissionabilities of different nuclides. Thus an increase of spallation cross section with mass number is seen from these reactions.

The (d,n) reaction is primarily due to direct interactions.

The mass number alone appears to have no strong effect on the fission asymmetry of uranium isotopes.

The experimental total reaction cross sections are in agreement with a value of 1.5×10^{-13} cm for the nuclear-radius parameter.

The (d, γ) reaction does not indicate compound-nucleus formation as its principal process, but appears to have some relationship with the giant resonance occurring in photonuclear reactions.

V. DECAY-SCHEME STUDIES

A. One-Hour Np²⁴⁰

1. Identification

The 60-min neptunium activity which had previously been assigned to Np²⁴¹⁶³ has in the course of this work been identified as an isomer of Np²⁴⁰. Originally Orth and Street had bombarded a U²³⁸ target with 35-Mev helium ions from the 60-inch cyclotron. Since Hyde and Studier⁶⁴ had recently identified 7.3-min Np²⁴⁰ and the 1-hr activity was not produced in proton or deuteron bombardments of U²³⁸, this new 60 $\pm$ 2-min beta-emitting activity in the neptunium fraction was assigned to Np²⁴¹ produced by the (α ,p) reaction on U²³⁸⁶⁵. Further investigation in collaboration with Maynard C. Michel, using the time-of-flight mass spectrometer, has shown that this 1-hr activity is due to mass 240.

The following experiments were performed to identify the 1-hr activity. Two separate bombardments were made on the 60-inch cyclotron, with U²³⁸ as the target material and helium ions as the projectiles. The neptunium was chemically separated from the target, primarily by use of a TTA extraction from a 1.5N hydrochloric acid solution and then a back extraction into 8N nitric acid. A time-of-flight mass separation of the neptunium fraction was made on each bombardment. In the first bombardment a mass-240 plate was collected and on the second

bombardment, in which the time for chemical separation was reduced from 45 to 25 minutes, a mass-241 plate was collected.

The first bombardment yielded decay periods of 1 hr and 8.7 hr at mass 240. The second bombardment gave periods of 16 min and 3.3 hr at mass 241. The time from end of bombardment until the plate was counted was 2.8 hr for the first bombardment and 1.8 hr for the second. Since the 1-hr activity, which is known to be neptunium, was found only at mass 240, this half-life has been definitely assigned to Np^{240} . Thus the probability previously proposed that Np^{241} also has a 1-hr half-life now seems quite remote.

2. Decay Properties

Measurements by Orth and Street on the 1-hr neptunium activity with a beta-ray spectrometer had shown a beta spectrum of upper energy limit 0.89 ± 0.03 Mev and conversion electrons corresponding to gamma rays of energies 150, 200, 260, and 580 kev. Gamma-ray measurements performed in the course of this work on a NaI scintillation spectrometer showed gamma rays of energies 1160, 1000, 915, 580, and 435 kev, but additional gamma rays of less than 350 kev were obscured by the Np^{239} background. By gamma-gamma coincidence techniques, in collaboration with Frank S. Stephens, additional gamma rays of 245, 160, and 85 kev were found. The 580-kev gamma ray was found to be complex, with components of about 595 and 565 kev in coincidence with each other. The most energetic beta-particle group had an end-point energy of about 900 kev and is in coincidence with 1160-kev gamma radiations. No other beta-ray was observed in coincidence with the gamma rays. Thus the decay energy is measured as 2.05 Mev ($890\beta + 1160\gamma$), which is consistent with the mass assignment, since from closed decay-energy cycles¹⁵ one would estimate only 1.32 Mev for the decay energy of Np^{241} , whereas 2.05 Mev is available in Np^{240} . Also the 60-min neptunium is the lower 240 isomer, as the decay energy of the 7.3-min Np^{240} is 2.156 Mev.⁶⁶ The gamma-ray spectrum is also consistent with this mass assignment, since gamma rays of 920 and 1020 kev are seen in the decay of Am^{240} to Pu^{240} , gamma rays of 557 and 600 kev are seen in the decay of the 7.3-min Np^{240} , and a gamma ray of

150 kev is seen in the alpha decay of Cm^{244} to Pu^{240} .⁶⁷ Further support for this mass assignment is found in the determination of the excitation function for 1-hr neptunium produced by the (d,γ) reaction on U^{238} given in Part III, Results. In addition, Ritzema¹³ has now determined the excitation function for the production of 60-min neptunium from U^{238} bombarded with helium ions. Lefevre, Kinderman, and Van Tuyl⁶⁸ have since produced both Np^{240} isomers by bombarding Np^{239} with pile neutrons and set an upper limit of $< 5\%$ for the genetic lineage between the two Np^{240} isomers.

A close search of decay curves previously run on chemically separated neptunium shows no evidence of any half-life between 1-hr and 2-day neptunium. Careful resolution of these curves gave 63 ± 1 min as the best value for the half-life of Np^{240} . The 8.7-hr activity could be due to an easily ionized fission product which is collected on harmonics in the separator. However, a search of the Table of Isotopes⁶⁷ failed to reveal any fission products that could account for this 8.7-hr activity. Thus the possibility that this 8.7-hr activity is another isomer of Np^{240} (an odd-odd nucleus) cannot be ruled out. Further experiments are required to establish the identity of this activity.

B. The New Isotope Np^{241}

1. Identification

The separation of the 241 mass described above was repeated by using the chemical separation for neptunium previously described in Part II, Experimental Procedures. A 5-mil-thick U^{238} foil was bombarded in the 60-inch cyclotron with 48-Mev helium ions, and this careful chemical separation of the resulting neptunium was carried out in a period of about 1-hr. The neptunium fraction was mass-separated in the time-of-flight spectrometer with a collection full time at mass 241. This mass-separated plate was first counted 3.7 hr after the end of the bombardment, and yielded a single decay period of about 3.5 hr. It was not possible to see the 16-min activity again because of the longer separation time. Since a clean chemical separation was obtained in this experiment, it

appears that the 3.3- to 3.5-hr activity is due to Np^{241} . The fact that this activity is not found in the gross decay of the neptunium fraction is not difficult to explain, as this activity is easily masked by the much more abundant 1-hr Np^{240} and 2-day Np^{238} and Np^{239} . Both TTA extraction and anion-column separations had been used in previous bombardments and in each case the 16-min activity followed the neptunium fraction.

Recently Vandebosch has repeated experiments on the chemical identification of this 16-minute activity as neptunium, and now believes the 16-min activity assignment to Np^{241} is correct.⁶⁹ In addition he found the maximum beta-particle energy to be 1.36 Mev, which is in good agreement with the 1.32-Mev value predicted by the closed decay-energy cycles. This gives rise to a log ft value of 5.9, an allowed transition, if all the decay is to the ground state.

It is possible to have isomers of Np^{241} . One would probably expect Np^{241} to have a $5/2+$ state, as both Np^{237} and Np^{239} have this state. Since the nuclear spin of the Pu^{241} daughter is $5/2$,⁷⁰ this $5/2+$ state in Np^{241} is consistent with the log ft value of 5.9 for the 16-min Np^{241} . The log ft value for the 3.3- to 3.5-hr Np^{241} is 6.9. If all the 1.32-Mev decay energy predicted by closed decay cycles goes to the ground state. Nilsson diagrams indicate that it is also possible to have an $11/2-$ state in this region.⁷¹ Such a second state could give rise to an E3 transition, which could have a half-life on the order of 3 hr. However, another mass separation using additional chemistry, such as an anion column, is necessary before both these activities can be assigned to Np^{241} with certainty. At present both these assignments should be classified as "element probable" and "mass number certain".

2. Decay Properties

The 16-min activity has been examined on the thallium-activated sodium iodide crystal of the 50-channel scintillation spectrometer. Because of relatively large amounts of Np^{240} and Np^{239} present, only tentative results could be obtained. These indicate that gamma rays of 130, 180, and 280 kev are due to the short-lived neptunium. Large peaks

at about 60 kev and 95 kev are interpreted respectively as lead x-rays from the chamber and neptunium x-rays. It appears possible that other gamma rays also are associated with this decay, but in too low abundance to assign definitely.

C. Decay Properties of Np²³³

Originally Magnusson, Thompson, and Seaborg⁷² identified Np²³³ as an isotope with a 35-min electron-capture half life. They report an alpha-particle emission abundance of about $1.5 \times 10^{-3}\%$ and an alpha-particle energy of 5.53 Mev. No other data on the decay properties of Np²³³ have been reported in the literature. In the course of this work Np²³³ was seen in the bombardment of U²³⁵ with deuterons as the (d,4n) reaction product, and in the bombardment of U²³⁴ with deuterons as the (d,3n) reaction product. The (d,3n) reaction on U²³⁴ provided the better means to produce Np²³³. The Np²³³ so produced was chemically separated and observed in a 100-channel NaI crystal scintillation spectrometer. Np²³³ gamma-ray peaks were observed at 95, 150, 170, 205, and 410 kev. Broad peaks owing to two or more unresolved gamma-rays were observed from 230 to 310 kev and from 500 to 560 kev. In addition gamma-ray peaks at 185, 450, 630, and 720 kev may be associated with the decay of Np²³³. However, owing to the very large background of gamma-ray peaks of Np²³⁴, it was impossible to definitely assign these gamma rays to Np²³³. It was also difficult to study gamma rays of low abundance without flooding the counter, as Np²³³ has about 200 counts of K x-rays for each count of the next most abundant gamma ray. In contrast this ratio was down by a factor of 10 for Np²³⁴. Coincidence counting techniques also failed to give any further information on the decay properties, owing to lack of a sufficient amount of Np²³³ activity.

The alpha particle associated with the decay of Np²³³ was not seen, therefore an upper limit of 3×10^{-5} for the branching ratio has been calculated.

D. Decay Properties of Np^{234}

Hoff and Stephens⁷³ have done gamma-ray spectroscopy on 4.4-day Np^{234} . They observed gamma rays at 1570, 1220, 980(?) and 760 kev. By means of gamma-gamma coincidence measurements, gamma rays of 440 and ~800 kev were observed to be in coincidence with the 760 kev gamma ray. Previously, Orth and Street⁶³ had reported internal conversion electrons due to gamma rays of 177, 442, 803, and 1420 kev. Huizenga et al.⁷⁴ observed gamma rays at 445, 500, 720, 905, 1010, 1190, and 1540 kev by means of a gamma-scintillation spectrometer. They also reported conversion electrons due to transitions of 43, 109, 234, 247, 449, 517(?), 752, 813, and 1567 kev. They state that there are no conversion electrons that could be interpreted as a 1420-kev transition. In order to determine the decay scheme of Np^{234} , metallic U^{235} foil was bombarded with 14- to 20-Mev deuterons on the Crocker Laboratory 60-inch cyclotron, and the Np^{234} produced was chemically separated and observed in a 100-channel NaI crystal scintillation spectrometer. Gamma-ray peaks were observed at 1600, 1250, 1000, 750, 490, 420, 260, 220, and 100 kev. The peak at 1600 kev was broad and probably consisted of two or more gamma rays. The half-width of the peak at 1250 kev was also larger than normal, even after the scattering of the 1600-kev gamma rays was carefully subtracted. This peak also is postulated to consist of two or more gamma rays. When the effect of the Compton scattering from the 1600-kev gamma rays was eliminated, a gamma-ray peak of about 1500 kev was observed. This peak was about 1/5 as abundant as the 1600-kev peak. All the other peaks were also broader than if they had been caused by single gamma rays. However, owing to the large effect of Compton scattering from the 1600-kev and 1250-kev gamma rays, it was impossible to tell whether the remaining peaks were due to scattered or unscattered gamma rays, except for the peaks at 1000, 750, and 100 kev. They were sufficiently large to be due to actual gamma rays or x-rays. The 100-kev peak is due primarily to neptunium x-rays. Therefore, this study postulates at least three additional gamma rays for Np^{234} , one or more in the region of the previously reported gamma at 1600 kev, one at about 1500 kev, and one or

more in the region of the 1250-kev gamma ray. Because of the complexity of the gamma-ray spectrum and lack of sufficient data, no decay scheme is proposed. Contemporary measurements on the annular focusing beta-ray spectrometer by Gallagher⁷⁵ may help solve the decay scheme. It is doubtful that any further elucidation of this decay scheme can be accomplished by additional gamma-ray spectroscopy.

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