UCRL 8032 Rev.

UNIVERSITY OF CALIFORNIA

Radiation Laboratory

BERKELEY, CALIFORNIA

UCRL-8032-Rev.

UNIVERSITY OF CALIFORNIA

Radiation Laboratory
Berkeley, California

Contract No. W-7405-eng-48

SPALLATION-FISSION COMPETITION IN HEAVIEST ELEMENTS; HELIUM ION-INDUCED REACTIONS IN URANIUM ISOTOPES

by

R. Vandenbosch, T. D. Thomas, S. E. Vandenbosch, R. A. Glass
and G. T. Seaborg

January 1958

Printed for the U. S. Atomic Energy Commission

SPALLATION-FISSION COMPETITION IN HEAVIEST ELEMENTS; HELIUM ION-INDUCED REACTIONS IN URANIUM ISOTOPES*

by

R. Vandenbosch,† T. D. Thomas, S. E. Vandenbosch,† R. A. Glass,‡ and G. T. Seaborg

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

January 1958

*This work was performed under the auspices of the U. S. Atomic Energy Commission. It is based in part on the Ph.D. Theses of R. Vandenbosch, University of California, Sept., 1957, and T. D. Thomas, University of California, Sept., 1957, and on the M.S. Thesis of S. E. Vandenbosch (nee Ritsema), University of California, January, 1956.

†Present address: Argonne National Laboratory, Lemont, Illinois.

‡Present address: Stanford Research Institute, Menlo Park, California.

ABSTRACT

A radiochemical study of fission and spallation products produced by bombardment of U^{233}, U^{235}, and U^{238} with 18-46 Mev helium ions has been made. As in the case of similar studies using isotopes of plutonium as targets, most of the reaction cross section is taken up by fission. Also, the pronounced increase of the total cross section for (α,xn) reactions with increasing mass number of the target that was observed for plutonium targets is observed for uranium targets.

Excitation functions for $(\alpha,2n)$, $(\alpha,3n)$, and $(\alpha,4n)$ reactions are interpreted in terms of compound nucleus formation and fission competition at the various stages of the neutron evaporation chain. The importance of neutron binding energies on the competition between fission and neutron emission is stressed. An existing model for neutron evaporation following compound nucleus formation has been extended to include the effect of fission competition.

Results of calculations based on this model show good agreement with those features of the (α,xn) excitation functions believed to result from compound nucleus formation. These calculations also show that fission usually precedes neutron evaporation for helium-ion-induced reactions of U^{233} and U^{235}. The excitation functions for the (α,n), (α,p), $(\alpha,pn + \alpha,d)$, $(\alpha,p2n + \alpha,t)$, and $(\alpha,p3n + \alpha,tn)$ reactions are discussed in terms of direct interaction mechanisms involving little competition from fission.

Fission shows an increase in symmetry with energy and becomes symmetric at about 40 Mev energy of the helium ions. There is no significant difference in the symmetry of fission for the three uranium isotopes. Total reaction cross sections, including those for both fission and spallation reactions, indicate a nuclear radius parameter r_o slightly larger than 1.5×10^{-13} cm.

SPALLATION-FISSION COMPETITION IN HEAVIEST ELEMENTS; HELIUM ION-INDUCED REACTIONS IN URANIUM ISOTOPES*

by

R. Vandenbosch,† T. D. Thomas, S. E. Vandenbosch,† R. A. Glass,‡ and G. T. Seaborg

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

January 1958

*This work was performed under the auspices of the U. S. Atomic Energy Commission. It is based in part on the Ph.D. theses of R. Vandenbosch, University of California, Sept., 1957, and T. D. Thomas, University of California, Sept., 1957, and on the M.S. thesis of S. E. Vandenbosch (nee Ritsema), University of California, January, 1956.

†Present address: Argonne National Laboratory, Lemont, Illinois.

‡Present address: Stanford Research Institute, Menlo Park, California.

I. INTRODUCTION

This paper extends the investigations of the present series[1-4] on fission and spallation reactions in the heaviest element region. Spallation reactions in the heaviest elements are particularly interesting because the fission process provides a prominent competing reaction (not found in lighter elements except at high excitation energies) which can have effects on the cross-sections of the other reactions. In addition, the fission process is interesting in its own right.

The investigations which are being pursued in the present program are primarily of target nuclides of atomic number greater than or equal to 88, where fission threshold energies are roughly comparable to nucleon binding energies. We have been concerned principally with nuclear reactions induced by particles of less than about 50 Mev energy, with the hope that at these relatively low energies the compound nucleus theory can be used as a starting point in describing the characteristics of the nuclear reactions.

Previously reported work[1-4] has indicated, first, that fission competes successfully with spallation reactions that proceed by the formation of a compound nucleus, and, second, that reactions involving the emission of charged particles proceed by direct interaction mechanisms. In particular, fission competes with neutron emission at every stage of the neutron evaporation chain. There has been noted,[1] however, a striking effect of the mass number of the target on the relative probabilities of fission and neutron emission: neutron emission competes more successfully as the mass number of the target is increased. The surprisingly large cross sections for the production of the nuclide corresponding to the $(\alpha,p2n)$ reaction have been shown to be due to the reaction (α,H^3), in which a triton, rather than three separate particles, is emitted.[3] Furthermore, it has been suggested that an appreciable fraction of the (α,xn) reactions are produced by direct interaction mechanisms.

In the first paper of this series,[1] the variation in the fission mass yield distribution with bombarding energy of helium ions was reported for plutonium isotopes. It was found that the transition from predominantly asymmetric to symmetric fission occurred at helium-ion bombarding energies between 30 and 40 Mev.

This paper will report cross-sections for helium-ion-induced reactions of U^{233}, U^{235}, and U^{238}. The study of these isotopes was undertaken to determine the effect of changing the atomic number and mass of the target nucleus, to compare with the work on the plutonium isotopes, and also to see if the striking mass effect on the spallation reactions in the plutonium isotopes is apparent for uranium isotopes. It was also hoped that a comparative study of the fission mass yield distribution in U^{233}, U^{235}, and U^{238} would shed some light on fission asymmetry.

II. EXPERIMENTAL PROCEDURES

Preparation of targets

The U^{233} used in these bombardments had an isotopic purity of approximately 96%; there was about 3% U^{238} and less than 1% U^{234} present in the material. The U^{235} generally had an isotopic purity of greater than 99.9%. The U^{238} also had an isotopic purity of greater than 99.9%. The techniques used in these experiments were generally those described by Glass et al.[1] Most of the targets were prepared by electrodeposition of 0.1 to 2 mg of hydrated uranium oxide over an area of about 1 cm^2 on a dish-shaped aluminum disk. The amount of material deposited, which was of uniform thickness, was determined by direct alpha counting, weighing, or both. These targets were then mounted in a water-cooled microtarget holder[5] which also served as a Faraday cup for beam intensity measurements.

Bombardments

Aluminum or platinum foils of measured thickness were used to degrade the helium ion beam to the desired energy.[6] The irradiations were for a period of two to three hours for each target, with beam currents of 5 to 10 micro-amperes. Because of the fact that only moderate amounts of activity were produced, the chemical separations of the various fission and spallation products were generally performed on the whole target. However, three experiments were performed in which 1-mil metallic U^{235} foils (~ 93% isotopic purity) were bombarded and one experiment was performed in which a 1-mil metallic U^{238} foil (> 99%) was bombarded. This procedure resulted in the production of sufficient activity to permit aliquots to be taken for the various fission product elements, making possible a study of a wider selection of fission-product elements and a more complete determination of the mass yield curve. The principal disadvantage of the use of uranium foils was that the uranium foil reduced the helium-ion beam energy by 3 to 5 Mev, resulting in a range in energy of the helium ions which caused the reactions.

Chemical procedures

The usual chemical procedure[5] involved dissolving the target, backing plate, and aluminum cover foil in acidic solution containing known amounts of

fission product carriers and radioactive tracers (Np^{237} and Pu^{239}) for the spallation products. First the neptunium, and then the plutonium, was removed from the target solution by coprecipitation in the IV oxidation state with zirconium phosphate under the proper oxidizing or reducing conditions. The neptunium fraction was further purified by coprecipitation with lanthanum fluoride and conversion of the fluorides to hydroxides, followed by dissolution in acid and the extraction into benzene of a neptunium (IV) thenoyltrifluoroacetone chelate complex.

The plutonium was purified by similar fluoride and hydroxide precipitations followed by an ion-exchange column step, in which the plutonium IV was first adsorbed on Dowex A-1 anion exchange resin from concentrated hydrochloric acid and then reduced to the III oxidation state and eluted from the resin. The neptunium and plutonium were either electrodeposited[7] or vaporized onto platinum counting plates. The fission products were purified by techniques adapted from those described in the compilations by Meinke[8] and Lindner.[9]

Detection of radiations

The fission products were mounted on previously weighed aluminum plates for weighing and counting. The disintegration rates were determined using end-window "Amprex" geiger counter tubes. Appropriate correction factors[10] were applied to obtain disintegration rates from the measured counting rates. The intensities and energies of alpha-emitting spallation products were measured by use of multichannel alpha-pulse analyzers. The counting rates of spallation products which decay by negatron emission or electron capture were determined with a methane-flow windowless proportional counter. Counting efficiencies for this counter have been measured or estimated for each particular isotope involved. Table I lists the nuclides produced by spallation reactions, together with their nuclear properties and counting efficiencies used in this work.

Table I

NUCLEAR PROPERTIES AND COUNTING EFFICIENCIES USED IN THIS WORK

Isotopes	$t_{1/2}$	Principal mode of decay	Percent alpha emission	Source	Proportional counter counting efficiency (percent)	Source
Pu^{232}	36 m	E.C.	11	a		
Pu^{233}	20 m	E.C.	0.12	b		
Pu^{234}	9 h	E.C.	6.16	c		
Pu^{235}	26 m	E.C.	3.0×10^{-3}	b	70 ± 14	b
Pu^{236}	2.7 yr	α	100	-		
Pu^{237}	44 d	E.C.	3.3×10^{-3}	b	79 ± 8	b
Pu^{238}	89.6 yr	α	100	-		
Np^{233}	35 m	E.C.			80 ± 20	d
Np^{234}	4.4 d	E.C.			63 ± 2	e
Np^{235}	410 d	E.C.			41 ± 4	f
Np^{236}	22 h	E.C., β^-			92 ± 20	g
Np^{238}	2.1 d	β^-			70 ± 5	h
Np^{239}	2.3 d	β^-			92 ± 5	i
Np^{240}	60 m	β^-			94 ± 6	i
U^{237}	6.75 d	β^-			80 ± 5	j

a. Estimated from the alpha systematics. I. Perlman and J. O. Rasmussen, Handbuch der Physik (Springer-Verlag, Berlin) Vol. 42, 1957.

b. Thomas, Vandenbosch, Glass, and Seaborg, Phys. Rev. 106, 1228 (1957).

c. Private communication, R. W. Hoff and F. Asaro (1957).

d. Estimated by authors.

e. By "milking" daughter U^{234} and determining its alpha disintegration rate, see Reference 11.

f. This work, mass spectrometry.

g. This work, by "milking" daughter Pu^{236} and determining its alpha disintegration rate. Percent negative beta decay (57%): T. O. Passell, Ph.D. thesis, University of California, June 1954 (unpublished); also University of California Radiation Laboratory Report UCRL-2528, March 1954 (unpublished).

h. This work, by "milking" daughter Pu^{238} and determining its alpha disintegration rate.

i. This work, by 4π-counting to determine absolute disintegration rate.

j. This work, by 4π-counting and by counting K x-rays. The number of K x-rays per disintegration was taken as 0.55, from Rasmussen, Canavan, and Hollander, Phys. Rev. 107, 141 (1957).

III. RESULTS

Spallation reactions

The cross-sections obtained at each energy for the spallation reactions of the various uranium isotopes are shown in Tables II to IV. The spallation cross-sections have been plotted as a function of helium-ion energy in Figs. 1 to 5. (Because of the scatter in the points, no curve has been given for the reactions $U^{233}(\alpha,p)Np^{236}$ and $U^{233}(\alpha,pn)Np^{235}$.) The product which was observed is indicated in the tables. In the cases where Np^{236} was the product, only the 22-hour isomer was observed. Similarly, when Np^{240} was the product only the yield for the 60-minute isomer was measured. The deviation due to random errors is believed to be about ± 10% for most of the spallation cross sections. Estimated systematic errors raise the total estimated deviation to between ± 15% and ± 25%. In the case of the U^{233} (α,pn) and $(\alpha,4n)$ reactions, the yields of the products Np^{235} and Pu^{233} were difficult to measure, and the limits of error may be as much as ± 50%.

Fission yields

The measured cross-sections for the formation of various fission product isotopes are shown in the left-hand columns of Tables V to VII. Since absolute cross-sections were not measured in the bombardments of U^{235} and U^{238} metallic foils, it was necessary to normalize these results in some way to the absolute cross-sections obtained from other bombardments. This was done by taking the average of normalization factors obtained by interpolation of smooth excitation function curves for the absolute fission yields of several isotopes.[12] The median energy of the helium ions inducing the fission in the foil bombardments was also calculated from these curves.

Gibson, Glass, and Seaborg[4] have made a preliminary study of the charge distribution in medium energy fission. Their conclusion is that the charge distribution in fission at these energies is not completely described either by the equal charge displacement noted at low energies[13,14] or by the constant charge to mass ratio which has been suggested to be occurring in very high energy fission.[15] However, the latter postulate appears to give a better correlation. A few primary yields measured in this work plus the primary yields

measured by Gibson have been used to construct a charge distribution curve which is slightly different from that of Gibson et al., but like theirs, is based on the postulate of equal charge to mass ratio.[4,11] This curve was used to correct the observed fission product cross-sections for the loss of yields of members of the same mass chain with higher atomic number, and the corrected cross-sections are shown in the right-hand columns of Tables V to VII. The mass number of the apparent fissioning nucleus used in application of the curve was estimated from the best values for the center of symmetry of the fission yield curves. Additional discussion of the problem of nuclear charge distribution in medium energy fission will be given by Gibson, Glass, and Seaborg,[4] and the problem will not be discussed further here.

Mass-yield curves for representative energies are shown in Figs. 6 to 8. The limits of error are estimated to be about ± 15% for most of the mass chains reported. However, at higher energies, particularly for U^{233}, the chain yield corrections become quite sizeable, and the errors may be somewhat greater.

The number of neutrons emitted as estimated from the center of symmetry of the fission mass yield curve is indicated in Figs. 6 to 8 and in the next to last row of Tables V to VII. It should be emphasized that the reflection of mass yield curves does not give any information as to whether the neutrons are emitted before or after the fission process takes place but includes contributions from both sources. However, some information on this subject implied by other types of data will be discussed later.

The total fission cross-sections obtained by integration of the fission mass yield curves are shown in the last row of Tables V to VII. The total fission cross-sections are compared with the summed spallation cross-sections in Figs. 9 and 10. No figure is shown for U^{238}, as it was impossible to measure yields for most of the (α,xn) reactions because of the long half lives of the products. The importance of the fission process is readily apparent from these figures.

Total cross sections

The total reaction cross-sections as obtained from the sum of the experimental fission and spallation cross-sections are shown in Figs. 11 to 13. Theoretical cross-sections for compound nucleus formation as given by Blatt and Weisskopf[16] are shown for two values of the nuclear radius parameter,

$r_o = 1.3 \times 10^{-13}$ cm and $r_o = 1.5 \times 10^{-13}$ cm. These experimental results indicate a value of the nuclear radius parameter slightly greater than $r_o = 1.5 \times 10^{-13}$ cm. There appears to be a discrepancy between the value of $r_o = 1.5 \times 10^{-13}$ cm determined in these experiments and that of 1.2×10^{-13} cm determined by electron scattering experiments.[17] The value of 1.5×10^{-13} cm is, however, consistent with values of the same parameter determined by other experiments on interactions of helium ions with nuclei and from study of the alpha decay process.[18]

IV. DISCUSSION

The general features of the excitation functions for spallation reactions in the uranium isotopes are in many ways quite similar to those that have been determined for other very heavy elements.[1,2] The cross-sections for the (α,n) and (α,p) reactions do not vary much with energy and are seldom more than a few millibarns in magnitude. The excitation functions for the (α,xn) reactions (for x greater than 1) have peaks which decrease in magnitude as x increases. The cross-sections for the $(\alpha,2n)$, $(\alpha,3n)$, and $(\alpha,4n)$ reactions of U^{233} are considerably smaller than those for U^{235}. A similar mass effect occurs in the plutonium isotopes. The cross-sections for reactions in which charged particles are emitted are quite large compared to the (α,xn) reaction cross sections.

In order to explain the relatively low cross-sections for the spallation reactions of the plutonium isotopes, Glass and co-workers have proposed that both fission and the major part of the (α,xn) reactions involve compound nucleus formation and that in the break-up of the compound nucleus fission competes more successfully than does spallation to claim the larger share of the total cross-section.[1] The decrease in the peak heights for the successive (α,xn) reactions has been interpreted to mean that fission is competing successfully at each stage of the evaporation chain in a compound nucleus reaction. Thus the peak cross-section of the $(\alpha,3n)$ reaction is lower than the peak cross-section of the $(\alpha,2n)$ reaction because in the former case fission has had three

chances to compete with neutron emission compared with two chances in the latter case. The long "tail" on the (α,xn) excitation functions and the relatively high cross-sections for the reactions involving the emission of charged particles suggest direct interactions of the projectile with a few nucleons on the nuclear surface. When a direct interaction occurs, the highly excited compound nucleus is by-passed, with the result that fission has fewer chances to compete with particle emission than when the highly excited compound nucleus is formed. Thus the products of the direct interaction type reactions often survive fission, whereas the products which are formed by evaporation of neutrons from a compound nucleus tend to be eliminated by fission. This means that excitation functions for reactions in the very heavy elements often strikingly demonstrate the importance of direct interaction mechanisms even at relatively low bombarding energies. Most of the results reported here can be explained in the framework of the ideas mentioned above.

Compound nucleus spallation reactions

The cross-sections reported for the (α,xn) reactions indicate that fission is competing more effectively in the bombardments of U^{233} than in those of Pu^{239}. Two factors affect the competition: the relative fissionability of corresponding compound nuclei and the ease with which neutrons are evaporated from corresponding compound nuclei. Fissionability increases as Z^2/A increases; the curium isotopes produced by the bombardment of Pu^{239} have higher values of Z^2/A than do the corresponding plutonium isotopes produced by the bombardment of U^{233}. The ease of neutron evaporation increases with decreasing neutron binding energy; the neutron binding energies of the curium isotopes produced by bombardment of Pu^{239} are lower than the neutron binding energies of the corresponding plutonium isotopes produced by bombardment of U^{233}.[19] Hence, the higher fissionability of the curium isotopes is apparently more than offset by the greater ease of neutron evaporation from these isotopes.

The strong effect of the mass number on the relative probability of neutron emission and fission observed in the reactions of both the uranium isotopes and the plutonium isotopes can be explained along similar lines. Since Z^2/A decreases as A increases, the ease of neutron evaporation increases. Furthermore, fission thresholds are lower than neutron binding energies in the

nuclides considered, with the result that a nucleus that has survived fission long enough to evaporate all of the neutrons that the original excitation energy would allow may still have sufficient residual excitation to undergo fission. Thus fission has an additional chance to occur when neutron emission can no longer compete. The higher the neutron binding energy and the lower the fission threshold, the larger will be the excitation energy range in which such fission can occur. Since neutron binding energies decrease and fission thresholds increase as A increases, such fission will compete less effectively as A increases. Thus, the three factors mentioned all contribute to decreasing competition from fission as A increases.

Jackson[20] has devised a schematic model for (p,xn) reactions in heavy elements. In his treatment he combines the results of Monte Carlo calculations for the probability of the various prompt processes with the results of a simplified evaporation model. His calculated cross sections show reasonable agreement with the experimental results of Bell and Skarsgard[21] and Kelly[22] for (p,xn) reactions of lead and bismuth in the energy range up to 100 Mev.

The evaporation model devised by Jackson has incorporated into it the following assumptions: (1) the neutron energy spectrum is given by $\epsilon \exp(-\epsilon/T)$ where ϵ is the kinetic energy of the neutron and T is the nuclear temperature, (2) neutron emission occurs whenever it is energetically possible, (3) proton evaporation is neglected, and (4) the nuclear temperature T is independent of excitation energy. This last assumption is an approximation; however, results calculated using this assumption agree reasonably well with results calculated using a nuclear temperature varying as the square root of the excitation energy.[20] According to Jackson, the probability that a nucleus with initial excitation energy E will evaporate exactly x neutrons is then given by

$$P(E,x) = I(\Delta_x, 2x - 3) - I(\Delta_{x+1}, 2x - 1) \qquad (1)$$

where $I(z,n)$ is Pearson's incomplete gamma function, $I(z,n) = \frac{1}{n!}\int_0^z x^n e^{-x} dx$ and $\Delta_x = (E - \sum_1^x B_i) / T$. B_i is the binding energy for the ith neutron and T is the nuclear temperature.

If we wish to extend the model given by Jackson to helium-ion induced reactions of fissionable elements, two difficulties arise. The first is that no Monte Carlo calculations have been made for the case where the projectile is a helium ion. Thus the contribution of direct interactions or similar prompt processes will for the present have to be ignored in the calculation. On the other hand, comparison of the calculated probabilities for evaporation with the experimental results can be used to estimate the contribution of direct interactions. Secondly, we must make a modification to include the effect of fission competition.

The fission competition will be considered in the framework of compound nucleus formation followed by competition between neutron emission and fission at each stage of the evaporation chain. There are two effects to consider: first, fission occurs while neutron emission is energetically possible, thus destroying nuclei during the early stages of the evaporation chain, and, second, some fission occurs after all of the possible neutrons have been evaporated, thus destroying nuclei whose excitation energy is less than the binding energy of the last neutron, but greater than the activation energy for fission, and which would otherwise have de-excited by gamma emission.

The probability that an excited nucleus will emit a neutron is given by its branching ratio[23] (level width ratio) for neutron emission $\Gamma_{n/\sum_i} \Gamma_i$ (henceforth designated as Gn). Similarly the branching ratio for fission is given by $\Gamma_{f/\sum_i} \Gamma_i$, or G_f, and the branching ratio for gamma ray de-excitation by $\Gamma_{\gamma/\sum_i} \Gamma_i$ or G_γ. The denominator, $\sum_i \Gamma_i$, contains terms for all the possible modes of decay of the compound nucleus. However, the assumptions will be made that the widths for proton evaporation and for gamma-ray de-excitation are negligible wherever neutron emission or fission is energetically possible. However, the gamma-ray branching ratio is taken as unity wherever neither fission nor neutron evaporation is energetically possible. When the excitation energy is greater than the activation energy for fission and less than the binding energy of the last neutron, G_f is taken to be unity. Hence to take into account the fission competition along the evaporation chain, we multiply the probability, P (E,x), defined above, by terms, G_{ni}, to give a new probability that the original compound nucleus will not only evaporate x neutrons but will also survive fission during the evaporation process.

After all of the neutrons have been evaporated, the residual nucleus may either undergo fission or may de-excite by gamma emission. We make the somewhat arbitrary assumption that if the residual nucleus has an excitation energy greater than the activation energy for fission it will undergo fission and that if the nucleus has an excitation energy less than the activation energy for fission it will de-excite by gamma emission. In Jackson's model, the first incomplete gamma function gives the probability that the original compound nucleus will emit at least x neutrons; the second the probability that the residual nucleus will have an excitation greater than the binding energy of the last neutron. Therefore, to account for fission competition at the final stage, we replace the last incomplete gamma function of Jackson by one giving the probability that the residual nucleus will have an excitation greater than the activation energy for fission. The result is a narrowing of the peak of the theoretical excitation functions, in better agreement with experiment.

Using the considerations, one can express the cross section for a reaction following compound nucleus formation as

$$\sigma(\alpha,xn) = \sigma_c G_{n_1} G_{n_2} \cdots G_{n_x} \left[I(\Delta_x, 2x-3) - I(\Delta_x^f, 2x-1) \right] \qquad (2)$$

where $\Delta_x^f = (E - \sum_i^x B_i - E_{th})/T$.

E_{th} is the activation energy for fission for the residual nucleus. The subscripts 1, 2--x on the G_n factor refer to the branching ratio for emission of the 1st, 2nd, --, x th neutron from the compound nucleus. σ_c is the cross section for the formation of the compound nucleus at the particular energy considered. The neutron binding energies were taken from Hyde and Seaborg,[19] and the fission activation energies were calculated from a semi-empirical equation relating fission thresholds to spontaneous fission rates.[24]

It is necessary to evaluate the G_n quantities and to choose a value of the nuclear temperature. Not a great deal is known about the variation of Γ_n/Γ_f with excitation energy and nuclear type (Z, A, even-odd character, etc.). The following assumptions about Γ_n/Γ_f will be made:

(1) Γ_n/Γ_f is independent of excitation energy for excitation energies well above the neutron emission threshold.

(2) Γ_n/Γ_f for even-even nuclei is twice as great as Γ_n/Γ_f for even-odd nuclei. (It will not be necessary to consider odd-odd products in the present calculations.)

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

The first assumption as a first approximation obtains support from the shape of excitation functions for fast neutron-induced fission and also from an analysis by Batzel[25] of high energy spallation excitation functions. The same conclusion was reached by Glass and co-workers from analysis of spallation excitation functions.[1] There is, however, some evidence that Γ_n/Γ_f increases with increasing excitation.[26] The second assumption arises from the expectation that the even-odd product of the evaporation of a neutron from an even-even nucleus has a higher level density than the even-even product from an even-odd nucleus; the factor of two used was taken from an estimate by Weisskopf.[27] Evidence for such a variation with nuclear type is presented by Vandenbosch and Seaborg.[24]

Using the foregoing assumptions together with information given by Vandenbosch and Seaborg[24] on the variations of Γ_n/Γ_f with mass number, we can derive a formula for the value of Γ_n/Γ_f for a particular plutonium isotope:

$$\left(\frac{\Gamma_n}{\Gamma_f}\right)_x = \frac{1.93\,a}{(1.3)^x}\,\frac{\bar{G}_n}{1-\bar{G}_n} \qquad (3)$$

where $a = \sqrt{2}$ for even-even nuclides

$a = 1/\sqrt{2}$ for even-odd nuclides.

The subscript x has the same significance as in Equation (2). $\bar{G}_n$ is a mean value of Γ_n/Γ_t and is defined as

$$\bar{G}_n = (G_{n_1}\, G_{n_2}\, G_{n_3}\, G_{n_4})^{1/4} . \qquad (4)$$

This quantity can be evaluated from Equation (2) if a value of the cross section for the $(\alpha,4n)$ cross section near its peak is known. (A similar set of formulae may be derived in which $\bar{G}_n$ is based on the cross section for the

$(\alpha,2n)$ reaction. Because of the poorly defined excitation function for the reaction $U^{233}(\alpha,4n)Pu^{233}$, it was necessary to base the value of $\overline{G}_n$ for the reactions of U^{233} on the excitation function for the $U^{233}(\alpha,2n)Pu^{235}$ reaction.)

Using the above considerations, one needs to choose only two parameters to calculate excitation functions for all of the possible (α,xn) reactions. These parameters are a value of $\overline{G}_n$ and a nuclear temperature T. Excitation functions have been calculated for the (α,xn) reaction cross sections of U^{233} and U^{235}. Values of $\overline{G}_n$ were determined in the manner described above to be 0.11 for U^{233} and 0.21 for U^{235}. Nuclear temperatures were chosen so that the position of the maximum of the curve calculated for the $(\alpha,2n)$ reaction for U^{233} coincided with the position of the maximum of the experimental curve, and so that the position of the maximum of the curve calculated for the $(\alpha,4n)$ reaction for U^{235} coincided with the position of the maximum of the experimental curve. The values chosen were 1.41 Mev for U^{233} and 1.35 Mev for U^{235}. The neutron branching ratios derived from the mean values of Γ_n/Γ_f are given in Table VIII. In Figs. 14 and 15 the calculated curves are compared with the experimental points. Considering the simplicity of the model, the agreement with those features of the excitation functions believed to result from compound nucleus formation is good. The agreement with the peak cross section values for the $(\alpha,2n)$, $(\alpha,3n)$, and $(\alpha,4n)$ reactions supports the assumed variation of Γ_n/Γ_f with mass number and nuclear type.

In view of the success in reproducing certain features of the spallation excitation functions using the branching ratios shown in Table VIII, it seems justifiable to use these branching ratios to calculate the fraction of the fission that occurs before the emission of various numbers of neutrons. Given an initial excitation energy of the compound nucleus, we can also calculate the average excitation energy at which fission occurs. It is assumed that the average excitation energy of the residual nucleus after the emission of a neutron is given by the initial excitation energy minus the binding energy of the neutron and minus 2 T, where the nuclear temperature T has been taken as 1.41 Mev for the spallation products of U^{233} and 1.35 Mev for the spallation products of U^{235}.

In Table IX the percentage of total fissions occurring after the evaporation of various numbers of neutrons are listed for three helium-ion

bombardment energies. The second row gives the initial excitation energy corresponding to the helium ion energy. The last row gives the average excitation energy at which fission is occurring for each of the three initial excitation energies in the case of each isotope. Calculations by Coffin and Halpern give results which are in substantial agreement with those reported here.[26]

It can be seen from Table IX that most of the fission precedes neutron evaporation for helium-ion induced fission of U^{233} and U^{235}. This conclusion is in apparent disagreement with the observations of Harding and Farley,[28] who measured the angular distribution of neutrons from the bombardment of natural uranium with 147 Mev protons. They concluded that the greater part of the neutron emission occurs before fission, with only 2.5 ± 1 neutrons being emitted from the moving fragments. However Marquez has pointed out that had Harding and Farley assumed what appears to be a more reasonable value for the average energy of the emitted neutrons, they would have found their results consistent with the neutrons' being emitted after fission.[29]

The results reported here, and by Glass and co-workers,[1] indicate that increasing the excitation energy of a compound nucleus increases the probability of the destruction of that nucleus by fission (either before or after neutron emission.) If we accept the assumption that Γ_n/Γ_f does not vary rapidly with energy, then the increased probability is due not so much to an increasing relative probability of fission with increasing excitation energy, but rather to the increased number of chances for fission to occur as the length of the evaporation chain increases with increasing excitation energy.

Direct interactions

Examination of Figs. 14 and 15 shows that almost all of the (α,n) excitation functions and the high energy part of the $(\alpha,2n)$ excitation function cannot be accounted for by a compound nucleus model. It has been mentioned earlier that direct interaction mechanisms must be important in these reactions. In general, however, it has been expected that the effect of direct interaction would be seen only at projectile energies above 50 Mev. In the reactions of non-fissionable nuclei, the prominent compound-nucleus-spallation reactions usually mask out any small effects due to direct interaction. The region of

fissionable nuclides is, therefore, a particularly good place to study the direct-interaction-spallation reactions with fairly low energy particles because the reactions which involve compound nucleus formation are largely eliminated by fission competition.

Glass and co-workers[1] concluded that products of the direct interactions survive because these reactions do not involve a highly excited intermediate nucleus. We must extend this conclusion to say that the products of the direct interactions survive because fission has a chance to compete only after a high energy particle (nucleon or complex particle) has carried off most of the energy of the incident particle. The residual nucleus is often left with too little energy to undergo fission or to evaporate another neutron. In those cases where subsequent neutron emission is possible, fission competes, in general, only once, rather than several times as in the case where a highly excited compound nucleus is formed.

One reasonable mechanism for the (α,n) and (α,p) reactions is a "knock-on" reaction in which the helium ion strikes a nucleon, which is then emitted. The product of the $(\alpha,2n)$ reaction can be formed in the following three ways: (1) by evaporation of two neutrons from the compound nucleus and (2) by ejection of the first neutron by a direct interaction mechanism followed by evaporation of the second neutron, and (3) by ejection of both neutrons by a direct interaction mechanism. The "tail" of the excitation function for the $(\alpha,2n)$ reaction is very likely due to an initial knock-on followed by the evaporation of the second neutron. Many of the direct interactions in which one neutron is knocked out will leave the nucleus with enough energy to evaporate a second neutron. Fission tends to cut down the products, but not so severely as it cuts down the products from the reaction involving the evaporation of two neutrons, since in the latter case fission has two chances to compete with neutron emission whereas in the former it has only one. The fact that the "tail" on the $(\alpha,2n)$ excitation function for U^{233} is lower than those for U^{235} and Pu^{239} is consistent with increased fission competition at the evaporation stages of the reactions of U^{233}. A comparison of the $(\alpha,2n)$ excitation functions of U^{233}, U^{235}, and Pu^{239} with those of lead shows that the peaks have been cut down by fission more than have the "tails", an observation that lends further support to the idea that the peaks, being due to initial compound nucleus formation, suffer from fission competition twice, whereas the tails, being due partly to

direct interaction, suffer from fission competition at most only once. The contribution of direct interactions to the excitation functions for the (α,3n) reaction appears to be fairly small. Reactions proceeding by direct interaction mechanisms probably contribute to the peak in the curve representing the (α,2n) cross sections and possibly to that in the curve representing the (α,3n) cross sections. It is likely, however, that the observed products of the (α,4n) reaction are due almost entirely to reactions going by a compound nucleus mechanism.

There is little doubt that the products of the (α,p2n) reaction of the heavy elements are produced almost entirely by the direct emission of high energy tritons, without the formation of a compound nucleus.[3] The yield of tritium from helium-ion bombardment of U^{238} has been measured[3] and found to be slightly larger than the amount that would be expected if the entire cross section for the (α,p2n) reaction - as measured radiochemically through the yield of the product nuclide in this work - was due to the (α,t) reaction. The cross section for the production of the nuclide corresponding to the "(α,p3n) reaction" is probably due to the reaction (α,tn). Thus the yield of tritium would be expected to be higher than the radiochemical yield of the product due to the (α,t) reaction because of the contribution of (α,tn) and (α,t fission) reactions. The observation that the yield for the product of the U^{233} (α,p3n) reaction (which includes the contribution of the U^{233} (α,4n) reaction) is much less than the yield for the product of the U^{238} (α,p3n) reaction indicates the increased fission competition in the neutron deficient isotopes.

Very little can be said about the mechanism of the (α,pn) reaction. On the basis of the data for the reaction $U^{233}(\alpha,pn)Np^{235}$ we can conclude only that the reaction occurs to an appreciable extent. In the U^{238} case, only one isomer of Np^{240} was observed; hence, we have only a lower limit for the cross section for this reaction. (An excitation function for the reaction Pu^{238} $(\alpha,pn)Cm^{240}$ was reported by Glass et al.[1]) It is tempting to suggest (by analogy to the (α,t) reaction) that this reaction occurs by the emission of a deuteron by a direct interaction; there is, however, at present no direct evidence that such is the case.

The (α,αn) reaction was the most prominent spallation reaction observed in the bombardment of U^{238} with helium ions. It is doubtful that compound

nucleus formation accounts for much of this cross section since the coulomb barrier would make it very difficult to evaporate an alpha particle. This view is supported by the low yields of (d,αn) reactions observed in the bombardment of U^{233} and Pu^{239}.[11] There are several possible alternate mechanisms. One mechanism for this reaction is a direct interaction of the bombarding particle with a neutron in the diffuse rim of the nucleus, resulting in the neutrons being knocked out without the capture of the bombarding projectile. With this type of mechanism the cross section for the (α,αp) reaction should also be fairly prominent. Another possibility is inelastic scattering of the incident alpha particle, with the excited target nucleus evaporating a neutron. With this type of mechanism, the cross section for the (α,αp) reaction should be much less than that for the (α,αn) reaction because of coulomb barrier discrimination against charged particle evaporation. Unfortunately, no cross sections for (α,αp) reactions have been studied in the heavy elements so that it is not possible to choose between the two mechanisms on this basis. Still a third possibility is a coulomb excitation process, but the probability for this does not seem to be large enough to account for the observed cross section.

Merkle[30] has measured a cross section of 70 mb for the (α,αn) reaction of Au^{197} at 46 Mev, which is quite comparable in magnitude to that found for the (α,αn) reaction of U^{238}. This would indicate that the last two mechanisms are not very likely, for in those cases one would expect that fission would compete with the neutron emission and the (α,αn) reaction would be less probable for U^{238} than for Au^{197}.

One interesting consequence of the large contribution of a direct interaction mechanism in spallation reactions for highly fissionable nuclei is illustrated in Figs. 9 and 10. The curves showing the percent of total reaction cross section due to spallation reactions is seen to decrease with increasing energy for U^{235} and Pu^{239}, while for U^{233} the curve rises at the highest energies. This is attributed to the prominence of compound nucleus type spallation reactions at the lower energies with increased chances for fission competition at the higher energies in the U^{235} and Pu^{239} reactions. However, the major part of the spallation reactions in U^{233} proceed through direct interaction mechanisms and these become more probable at higher energies.

This does not imply that there is a larger amount of direct interaction taking place for U^{233} than for U^{235} and Pu^{239}, but that the fraction of the spallation reactions that go by direct interaction is larger for U^{233} than for Pu^{239} and U^{235}.

Fission

The mass yield distributions of the fission products are shown for different helium ion energies in Figs. 6 to 8. It is seen that fission is predominantly asymmetric at low energies and appears to become more symmetric as the excitation energy is increased, in agreement with previous work.[1,4,31] However, it should be noted that the increased symmetry is not due to the asymmetric peaks moving together, but rather to an apparent increase in a symmetric mode causing the valley to rise up faster than the wings. Comparison of the fission yield curves, and particularly the valley to peak ratios (ratio of the cross section at the minimum in the yield distribution to the cross section at the asymmetric maxima) indicates that there is no significant difference in the fission asymmetry in the three uranium isotopes studied.

As seen in Figs. 9, 10, and 13, the total fission cross sections for the three isotopes are all approximately the same and account for most of the total cross section. Comparison of the fission cross sections determined in this work for helium ion induced fission of U^{235} and U^{238} with the results determined by Jungerman[32] using an ionization chamber show good agreement between the two methods.

ACKNOWLEDGMENTS

We wish to acknowledge helpful discussions with Drs. W. M. Gibson, B. G. Harvey, and W. H. Wade. The cooperation of the late G. Bernard Rossi, and of W. Bart Jones and the crew of the Crocker Laboratory 60-inch cyclotron is appreciated. The Health Chemistry Group of the Radiation Laboratory assisted in minimizing the hazards in working with radioactive materials.

Two of the authors (R.V. and T.D.T.) wish to acknowledge the support of the National Science Foundation in the form of Predoctoral Fellowships (1955-57 and 1954-57, respectively).

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

Table II

Spallation cross sections (mb) for helium-ion induced reactions of U^{233}

Reaction	α,n	α,2n	α,3n	α,4n	α,5n	α,p	α,pn	α,p2n	α,p3n
Product	Pu^{236}	Pu^{235}	Pu^{234}	Pu^{233}	Pu^{232}	Np^{236}	Np^{235}	Np^{234}	Np^{233}
20.3 (Mev)	0.18								
23.5	0.42	1.30							
26.2	0.59	3.68	0.003			0.20	1.0	0.16	
28.9	0.96	6.54	0.083			0.53	-1.8	1.63	
29.4	0.64		0.058						
30.7						0.63	3.5	5.04	
31.8	1.01	3.40	0.91			1.72	0.3	4.91	
32.4			0.39			0.64		3.52	
34.3						1.07	13.5	10.9	
35.3	0.49	1.19	0.97			0.58	2.5	5.20	0.21
36.8						1.46	6.5	10.5	
36.8			0.67						
37.8	0.52	0.94	0.48			0.74	3.5	7.25	0.11
39.0			0.54						
40.0			0.44						
40.4						0.40	4.6	10.4	1.16
41.0	0.42	1.19	0.33			0.62	14.9	11.8	0.60
42.7			0.19	0.27	0.002	0.70	2.6	9.4	
43.8						2.53	8.8	17.8	1.41
44.3	0.73		0.26			0.74	18.7	19.9	1.72
44.4			0.51	1.03		0.72		15.9	0.64
46.2	0.79		0.45	1.13		0.30	21.3	19.6	1.10
46.2		1.31	0.20	0.33					
46.2			0.15	0.34					

Table III

Spallation cross sections (mb) for helium-ion induced reactions of U^{235}

Reaction	(α,n)	(α,2n)	(α,3n)	(α,4n)	(α,5n)	(α,p)	(α,p2n)
Product	Pu^{238}	Pu^{237}	Pu^{236}	Pu^{235}	Pu^{234}	Np^{238}	Np^{236}
18.7 (Mev)	0.27						
21.9	0.36	4.43				0.02	
23.6	1.32	13.3				0.035	0.042
25.2						1.01	0.087
27.3	1.74	15.8	0.61			0.55	0.52
29.7						1.7	1.86
30.0		8.3	4.43			1.43	2.22
30.6	1.42	6.84	4.15			1.57	2.38
34.1	2.15		8.63			2.08	4.38
34.7		6.8	7.23				4.20
37.1			3.67	0.17		1.92	5.9
39.5	2.26	5.65	3.12	1.5		1.87	8.5
42.8	2.52	4.8	2.23	2.4	0.002	1.94	10.7
45.4	.91	3.5	1.86	1.55	0.034	1.21	10.5

Table IV

Spallation cross sections (mb) for helium-ion induced reactions of U^{238}

Reaction	α,pn	α,p2n	α,p3n	α,αn
Product	Np^{240}	Np^{239}	Np^{238}	U^{237}
22.6 (Mev)	0.024	0.22		
25.2	1.1	1.06		0.6
27.1	1.2	9.1		1.5
32.5	1.7	9.0		8.2
33.8	3.6	9.3		7.9
37.9	6.0			
38.6	6.1	17.5		49.2
38.6		20.5		56.2
40.0			3.8	
41.4	6.3	21.2		
43.9				56.0
45.4	5.3	33.4	8.8	74

Table V

Fission cross-sections (mb) for helium-ion induced reactions of U^{233}.

The left-hand columns list the observed yield for each isotope.

The right-hand columns list the corrected cross-section for the mass chain.

Energy (Mev)	23.5		26.2		27.8		30.7		35.3		40.4		41.0		44.3		46.2	
Isotope	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.
Sr^{89}									16	16			9.9	10	32	33	22	22
Sr^{91}									19	21			17	19	52	58	35	40
Zr^{95}	2.4	2.5	12	12	17	17	21	22	39	41	46	48	42	44	55	58	57	61
Zr^{97}	6.5	7.1	15	17	15	17	12	14	38	44	45	55	44	52	43	53	48	61
Mo^{99}	1.4	1.4													32	33		
Ru^{103}	4.8	4.8							24	24					28	28		
Ru^{105}	3.2	3.3							27	28					41	44		
Ru^{106}									27	30					45	51		
Ag^{111}	>0.29	>0.29	11	11											>44	>45		
Cd^{115}	3.3	3.4	9.9	10	15	16	32	35	41	44			43	47	68	74	74	82
Ba^{135m}			0.42	4.0					4.9	31			4.0	22	8.5	45	4.9	22
Ba^{139}	4.6	6.7	9.2	16					18	37			12	31	22	55	25	83
Ba^{140}	3.4	5.9	7.4	17					12	34			8.4	32	14	54	15	83
Ce^{141}	10	11			26	29	13	15			39	46						
Ce^{143}	8.4	10			13	17	12	18										
Ce^{144}	2.1	3.1			8.0	14	8.8	18			15	40						
Nd^{147}	2.0	2.1			8.0	9.2					15	22	3.0	4.4				
Eu^{156}					0.68	2.4					0.44	1.6						
Eu^{157}	0.04	0.07			0.94	1.9					0.48	1.8						
Tb^{161}					0.50	0.77					0.71	1.8						
Number of Neutrons	4		5		5		6		6		7		7		7		8	
Total Fission Cross-Section	184		400				1060		1270				1430		1990			

Table VI

Fission cross-sections (mb) for helium-ion induced reactions of U^{235}.

Each left-hand column lists the observed yield for each isotope.

Each right-hand column lists the corrected cross section for the mass chain.

Energy (Mev)	18.7		21.9		26.8		30.6		32.8		34.1		37.1		42.8		45	
Isotope	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.
Zn^{72}															0.48	0.52		
Sr^{89}	0.098	0.098			10	10	17	17	22	22	23	23	27	27			60	60
Sr^{91}	0.095	0.097			11	11	18	18	24	25	26	27	28	29			56	59
Zr^{95}			4.2	4.2	16	16	29	30	32	32	39	40	47	48	49	51	45	47
Zr^{97}	0.071	0.075	4.3	4.5	17	18	31	33	31	33	42	45	49	53	50	57	56	63
Mo^{99}					19	19			35	35							63	64
Ru^{103}	0.017	0.017			12	12											52	52
Ru^{105}					12	12											52	53
Ru^{106}					11	12											60	64
Pd^{112}					10	10			30	33							48	54
Ag^{111}					11	11			33	33							72	72
Ag^{112}																	48	54
Ag^{113}					11	11											76	80
Cd^{115}			0.87	0.87	12	12	18	18	36	37	38	40	48	49	51	53	60	62
Cd^{117}			0.90	0.94			15	16			35	38	45	49	57	56		
Ba^{139}	0.10	0.11			16	19	20	26			30	42	30	40				
Ba^{140}	0.10	0.13			11	15	15	24	23	36	22	41	21	40			29	63
Ce^{141}			2.1	2.1	14	14			28	28	49	52					38	42
Ce^{143}			1.5	1.6	12	13			34	39	27	34					31	41
Ce^{144}			0.98	1.1	7.4	8.9			16	23	15	22					20	34
Nd^{147}					7.1	7.5			15	16	20	22					16	19
Eu^{156}					0.55	0.63			2.1	2.6	36	4.9					2.4	3.7
Eu^{157}					0.56	0.73			1.9	2.8	2.7	4.3					1.8	3.7
Gd^{159}					0.29	0.35			0.55	0.82							1.3	2.3
Tb^{161}											0.36	0.49					0.57	1.0
Number of Neutrons	3		4		4				5		6		6		7		7	
Total Fission Cross-Section	1.8		58		420				780		1290		1490		1760		1840	

Table VII

Fission cross-sections (mb) for helium-ion induced reactions of U^{238}. Each left-hand column lists the observed yield for each isotope. Each right-hand column lists the corrected cross-section for the mass chain.

Energy (Mev)	22.6		25.2		27.1		32.5		33.8		38.6		40		43.9		45.4	
Isotope	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.	σ	σ corr.
Sr^{89}													24	24			27	27
Sr^{91}													27	28			35	36
Zr^{95}	4.7	4.7			29	29	21	21	28	28	38	38	35	35	41	41	36	36
Zr^{97}	8.0	8.2			36	37	34	35	41	42	54	56	54	56	53	56	52	54
Mo^{99}													59	59				
Ru^{103}	6.5	6.5									47	47	44	44	51	51	47	47
Ru^{105}	7.0	7.0									36	36	53	53	55	55	48	48
Pd^{112}													54	56				
Ag^{111}													43	43				
Ag^{113}													49	49				
Cd^{115}	2.6	2.6			15.4	15.4					48	48	60	60	58	58	49	49
Cd^{117}	1.9	2.0													61[b]	64[b]		
Te^{129m}													31[a]	35[a]				
Te^{132}													39[b]	74[b]				
Ba^{139}	6.5	7.0									36	41			37	45	42	51
Ba^{140}	5.8	6.7							29	36	35	43	35	43	36	50	36	50
Ce^{141}					40	40												
Ce^{143}			11.5	12.1	23	24					44	48	49	52	30	33		
Nd^{147}					15	15.4					27	28	19	20				
Eu^{156}					1.8	1.9					3.4	3.7	4.1	4.5				
Eu^{157}					1.5	1.7					2.5	2.8	2.2	2.5				
Gd^{159}													0.71	0.71				
Tb^{161}					0.29	0.31					0.46	0.48						
Number of neutrons	4		4		5		5		5		6		6		7		7	
Total Fission Cross-Section	129				890						1480		1570		1600		1500	

a. Cross-section is for one isomer only.

b. Cross-section is approximate owing to complexities in the decay scheme.

Table VIII

Neutron branching ratios used in calculating U^{233} and U^{235} (α,xn) cross sections. The numerical subscripts refer to the emission of the 1st, 2nd, ...ith neutron.

Ratio	U^{233}	U^{235}
$\left(\frac{\Gamma_n}{\Gamma_t}\right)_1$	0.12	0.23
$\left(\frac{\Gamma_n}{\Gamma_t}\right)_2$	0.17	0.32
$\left(\frac{\Gamma_n}{\Gamma_t}\right)_3$	0.07	0.15
$\left(\frac{\Gamma_n}{\Gamma_t}\right)_4$	0.10	0.21
$\left(\frac{\Gamma_n}{\Gamma_t}\right)_5$	0.04	0.09

Table IX

The percentage of total fissions occurring after the evaporation of various numbers of neutrons in the helium-ion induced fission of U^{233} and U^{235}. Calculations for three different initial excitation energies are listed in each case.

	U^{233}			U^{235}		
Helium-ion energy (Mev)	46	36	29	42	32	23
Excitation energy (Mev)	40	30	23	37	27	18
Neutrons emitted before fission						
0	88%	88%	90%	77%	78%	83%
1	9.6%	10%	10%	16%	16%	17%
2	1.8%	2%		6%	6%	
3	0.1%			1%		
Average excitation energy of fission (Mev)	38.3	28.4	22.2	34.2	24.6	16.6

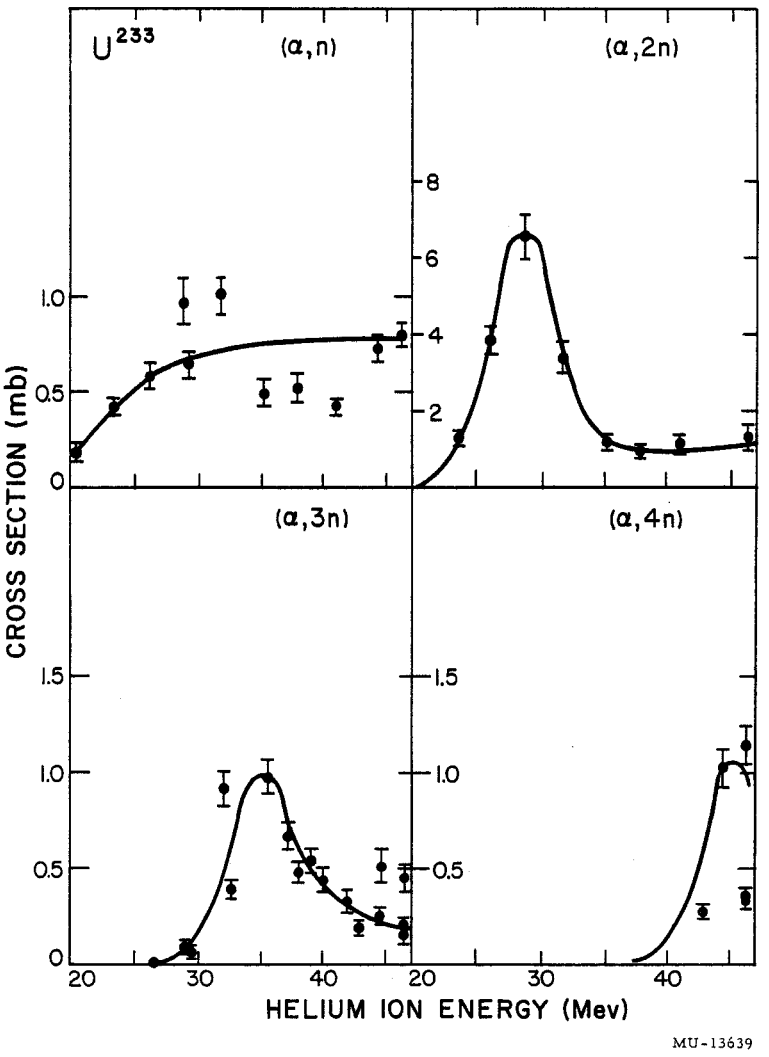

Fig. 1. Spallation excitation functions for (α,xn) reactions of U^{233}. Indicated limits of error on the (α,4n) cross sections are relative errors only.

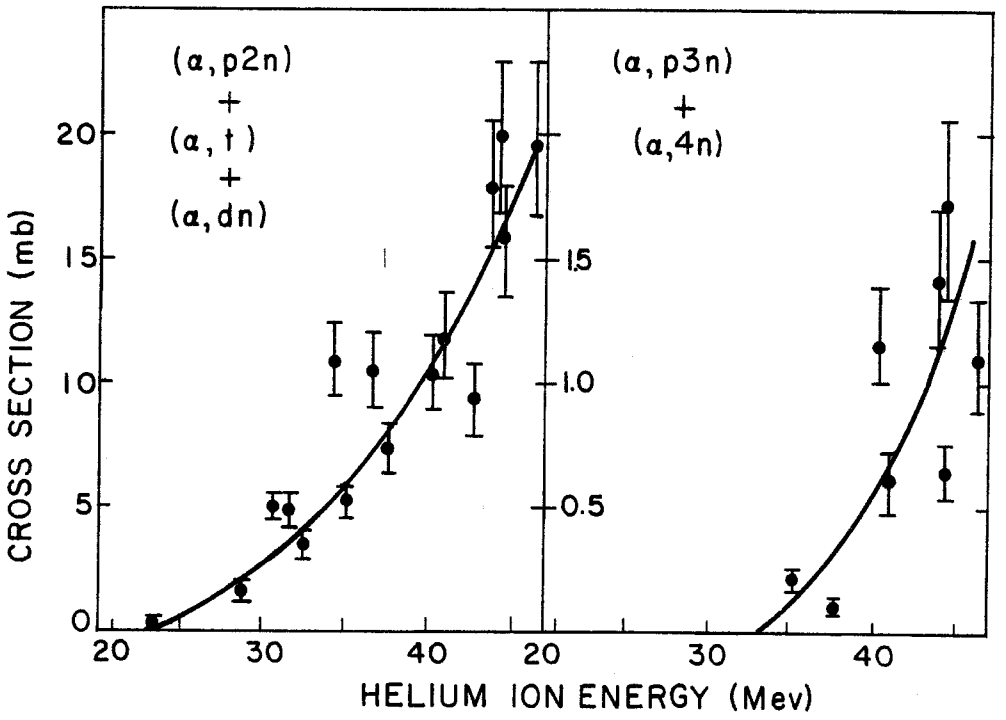

Fig. 2. Spallation excitation functions for (α,pxn) reactions of U^{233}.

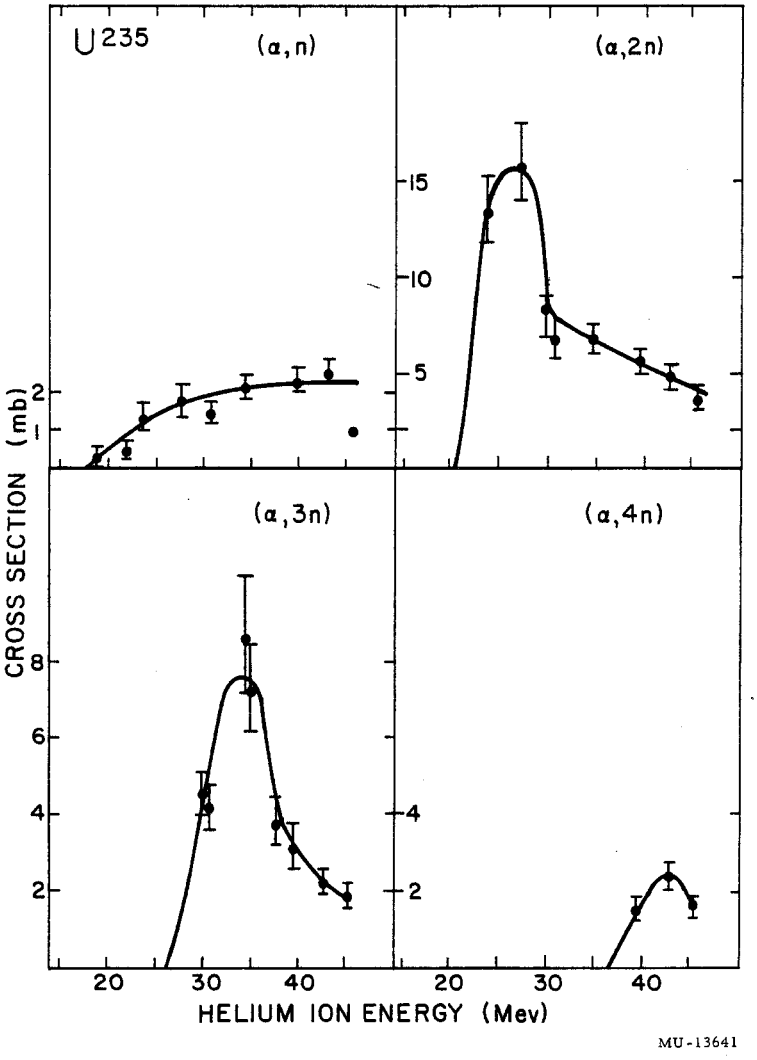

Fig. 3. Spallation excitation functions for (α,xn) reactions of U^{235}.

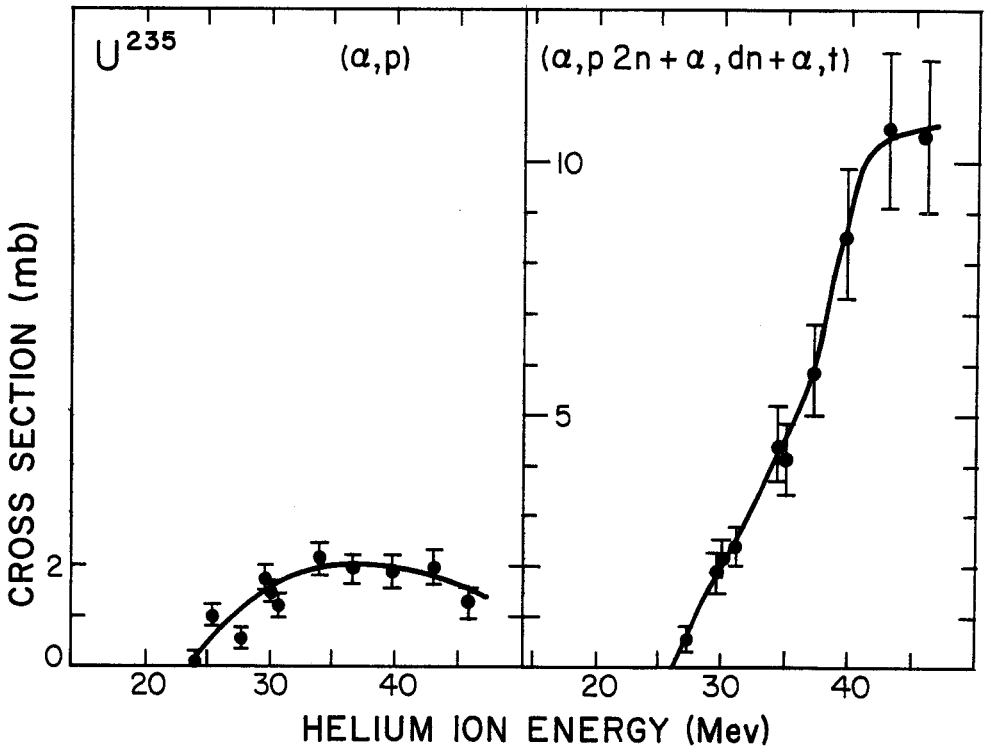

Fig. 4. U^{235} Spallation excitation functions for (α,pxn) reactions of U^{235}.

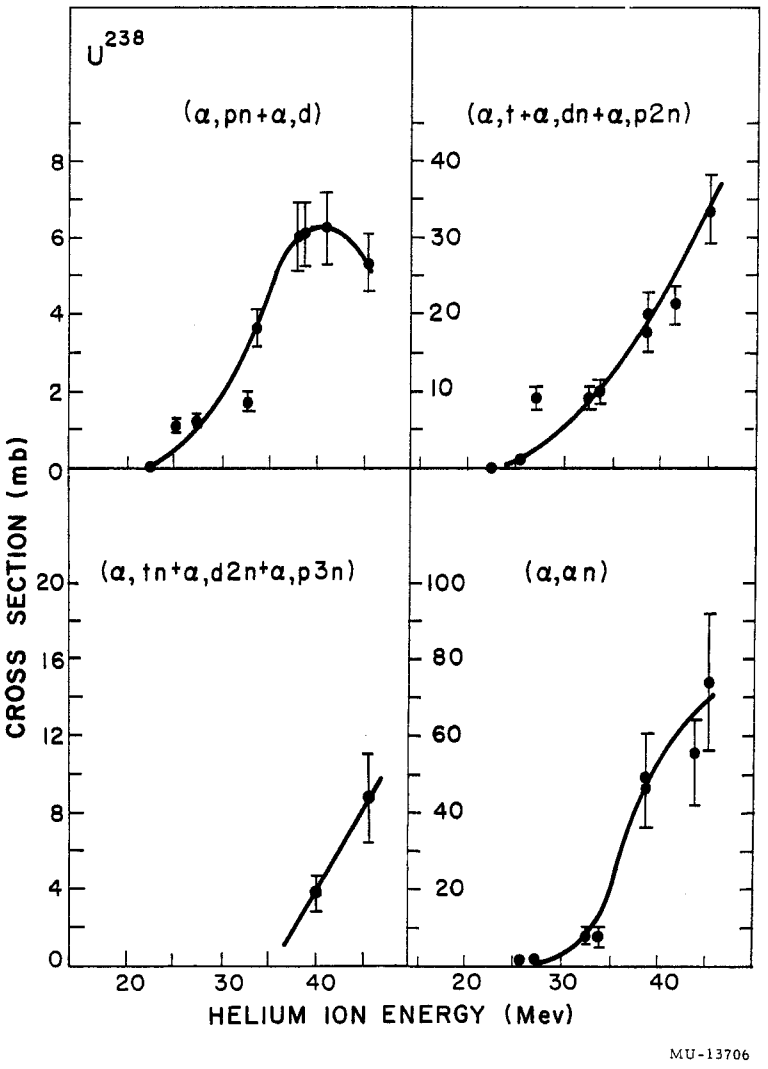

Fig. 5. Excitation functions for spallation reactions of U^{238}.

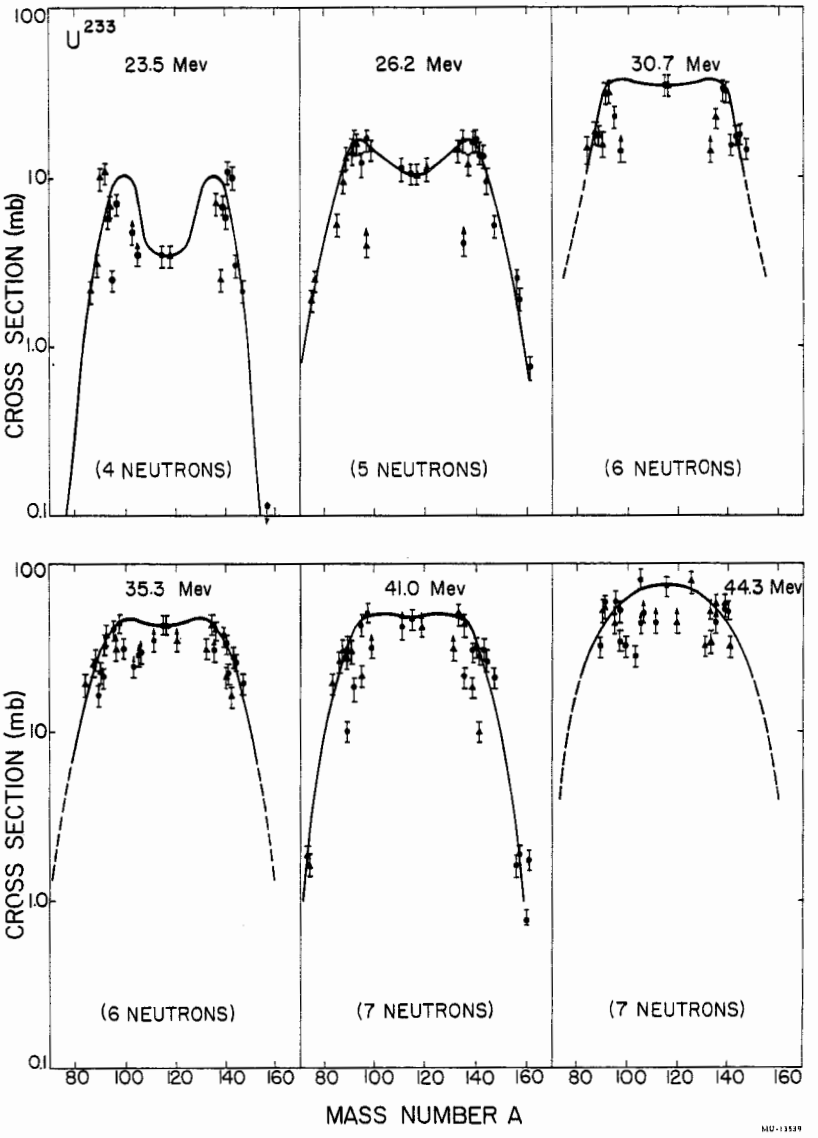

Fig. 6. Fission yield curves for helium-ion induced fission of U^{233}. The circles represent experimental points (corrected for the mass chain yield) and the triangles represent reflected points. The number of neutrons assumed emitted in reflecting the curves are indicated for each energy.

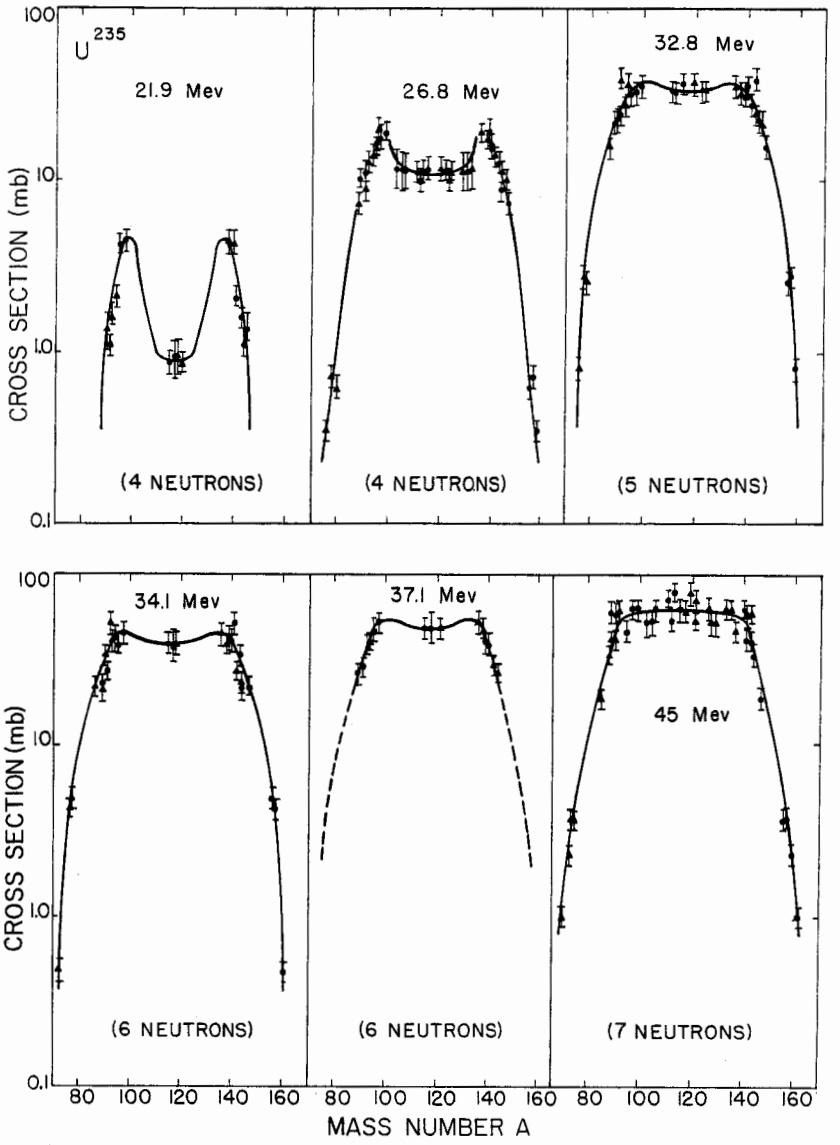

Fig. 7. Fission yield curves for helium-ion induced fission of U^{235}. The circles represent experimental points (corrected for mass chain yield) and the triangles represent reflected points. The number of neutrons assumed emitted in reflecting the curves are indicated for each energy.

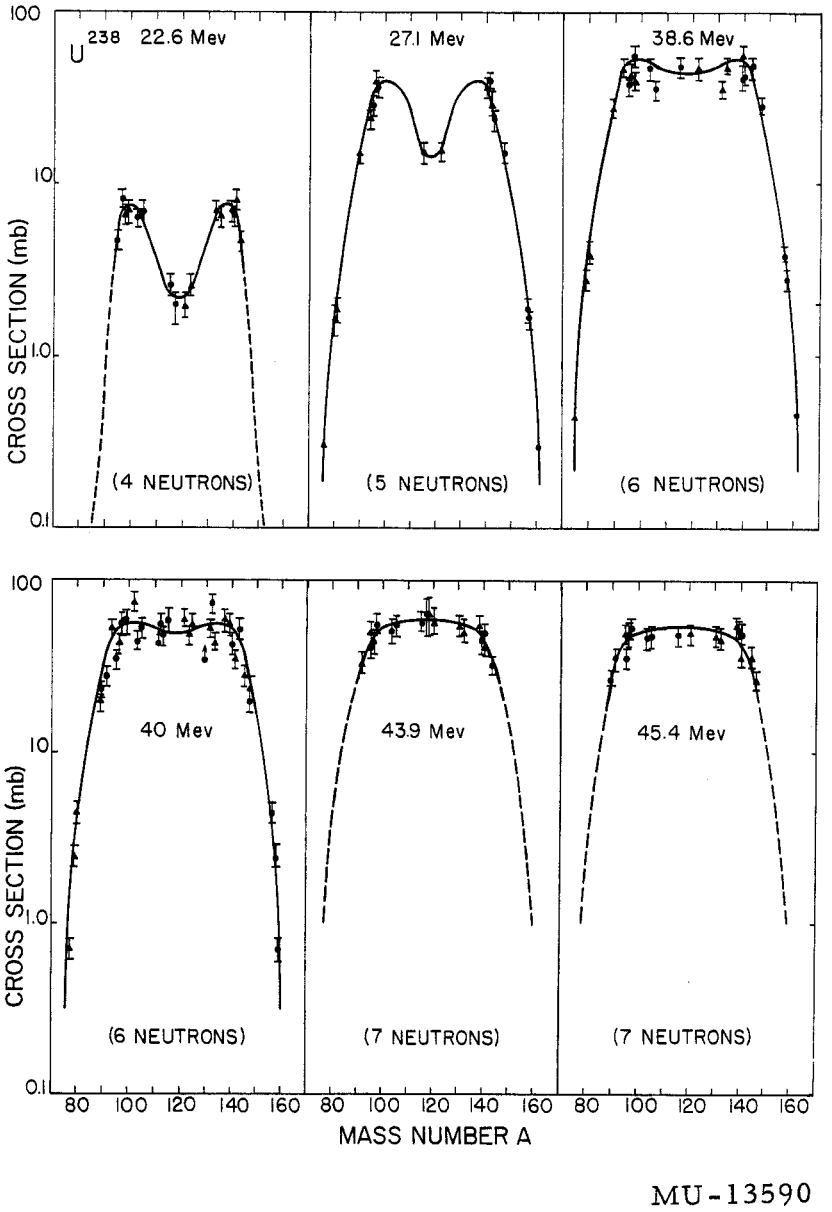

Fig. 8. Fission yield curves for helium-ion induced fission of U^{238}. The circles represent experimental points (corrected for mass chain yield) and the triangles represent reflected points. The number of neutrons assumed emitted in reflecting the curves are indicated for each energy.

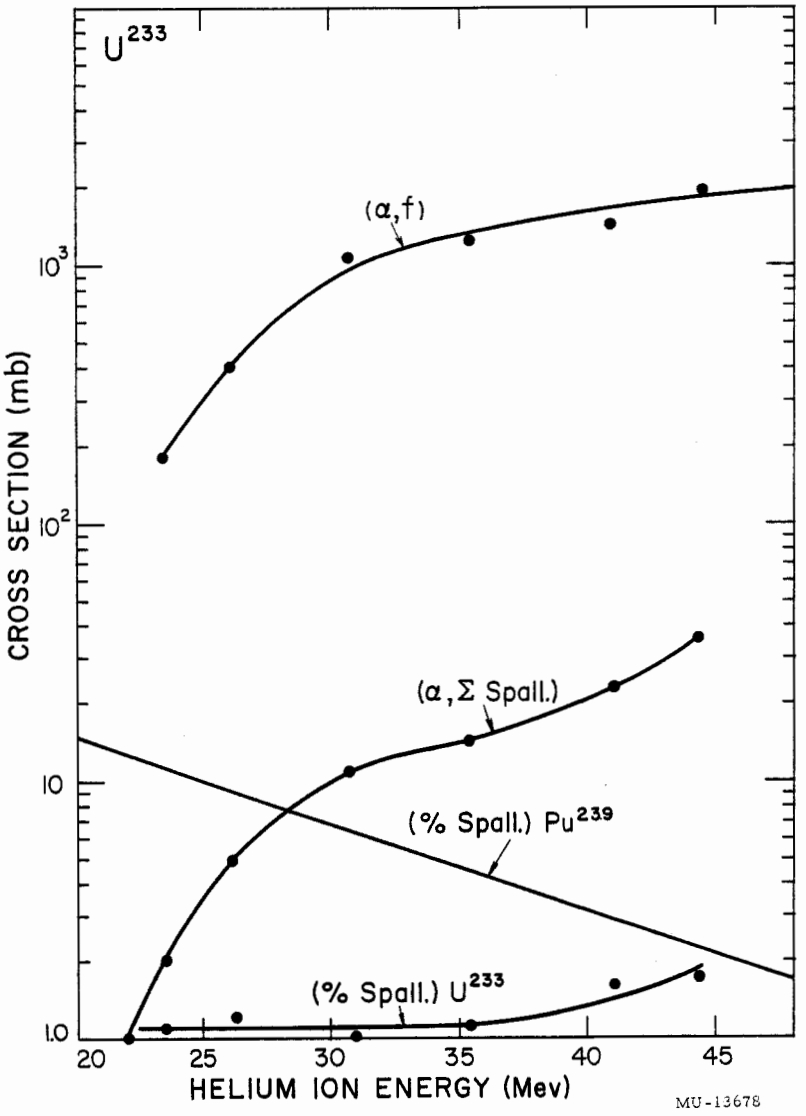

Fig. 9. Excitation functions for fission and summed spallation reactions in U^{233}. Also shown is the percent of the total reaction cross section going into spallation for U^{233} and also for Pu^{239} for comparison.

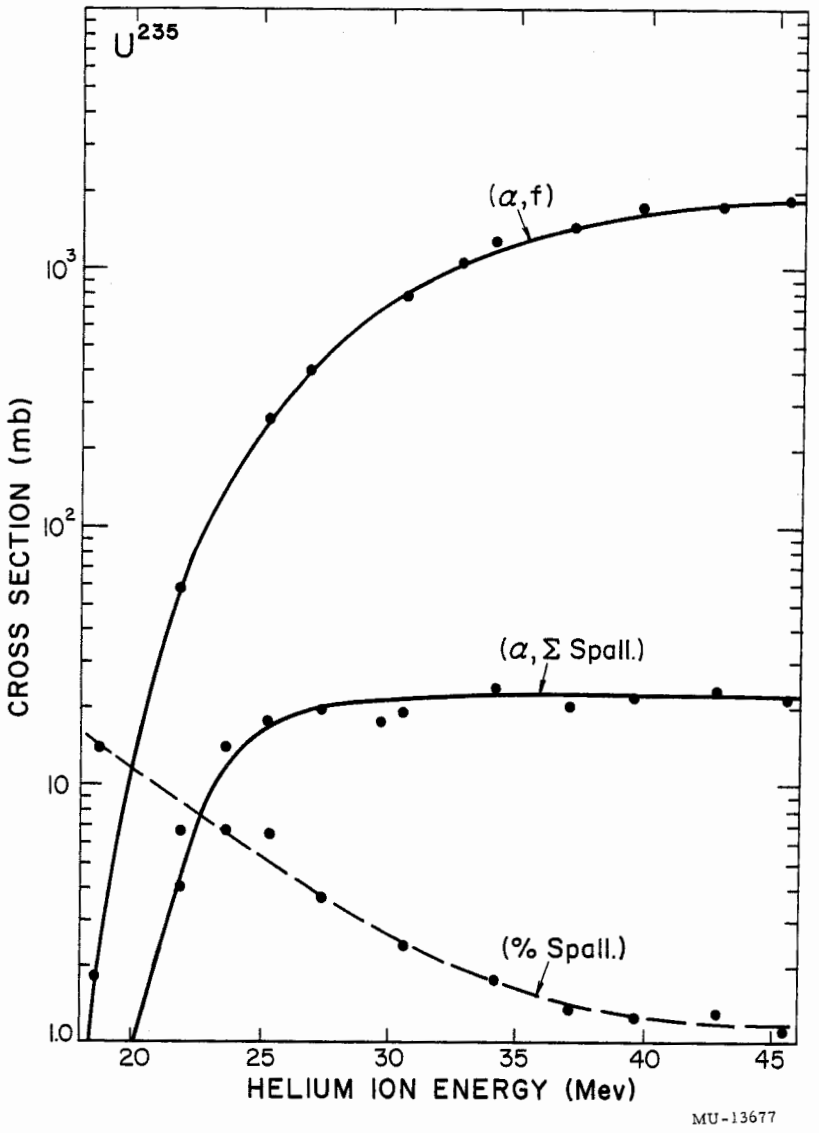

Fig. 10. Excitation functions for fission and summed spallation reaction in U^{235}. The dashed lines show the percent of the total reaction cross section going into spallation.

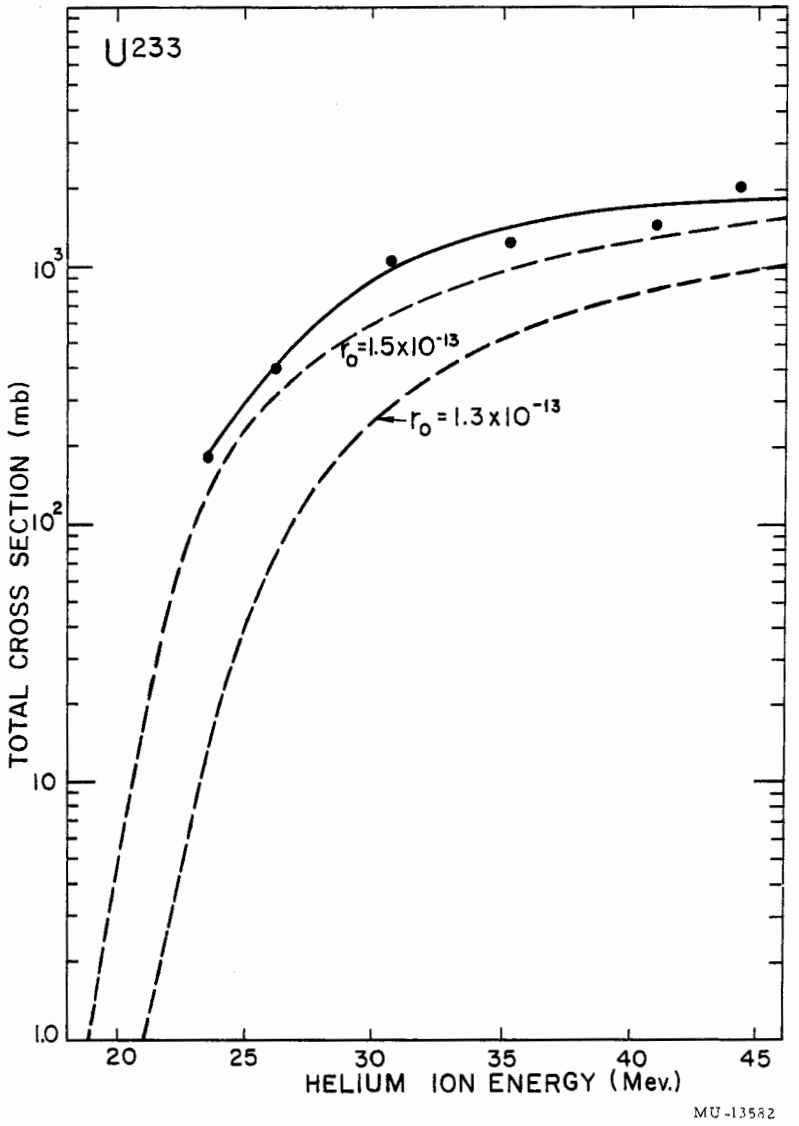

Fig. 11. Total fission yields plus the observed spallation yields for helium-ion bombardments of U^{233}. The circles represent experimental data. The dashed lines represent theoretical compound nucleus formation cross sections and were taken from reference 16.

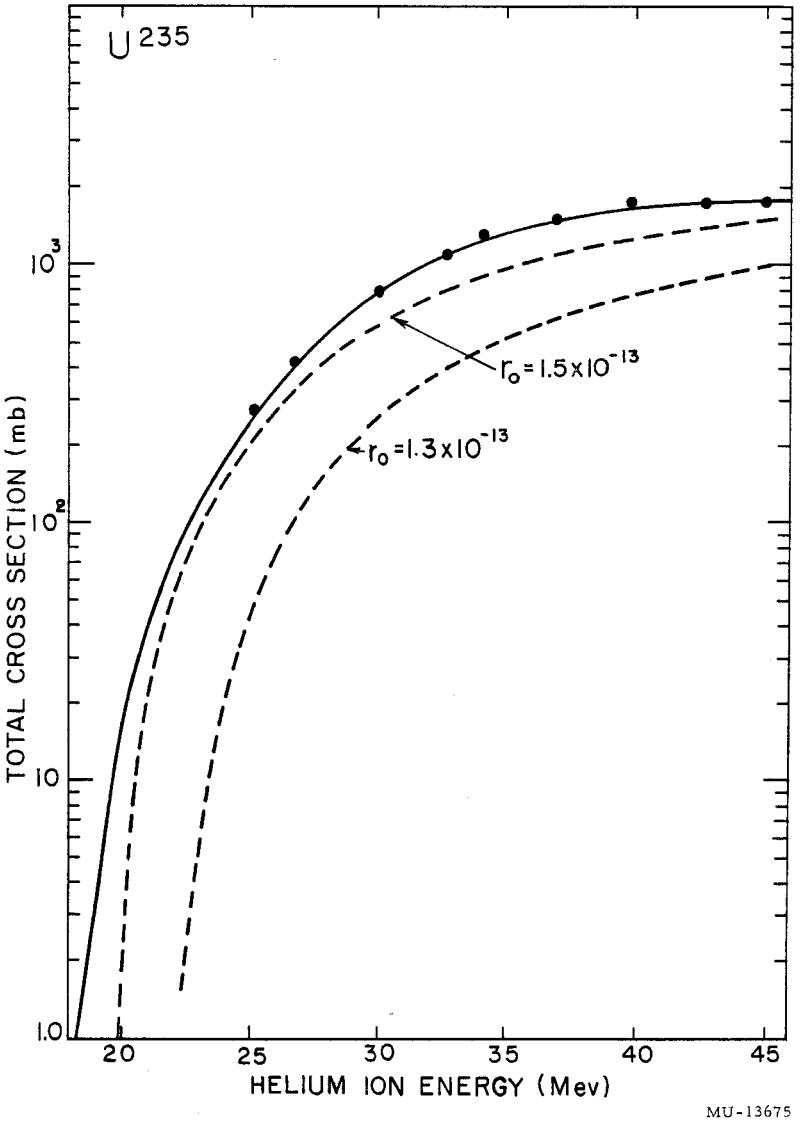

Fig. 12. Total fission yields plus the observed spallation yields for helium-ion bombardments of U^{235}. The circles represent experimental data. The dashed lines represent theoretical compound nucleus formation cross sections and were taken from reference 16.

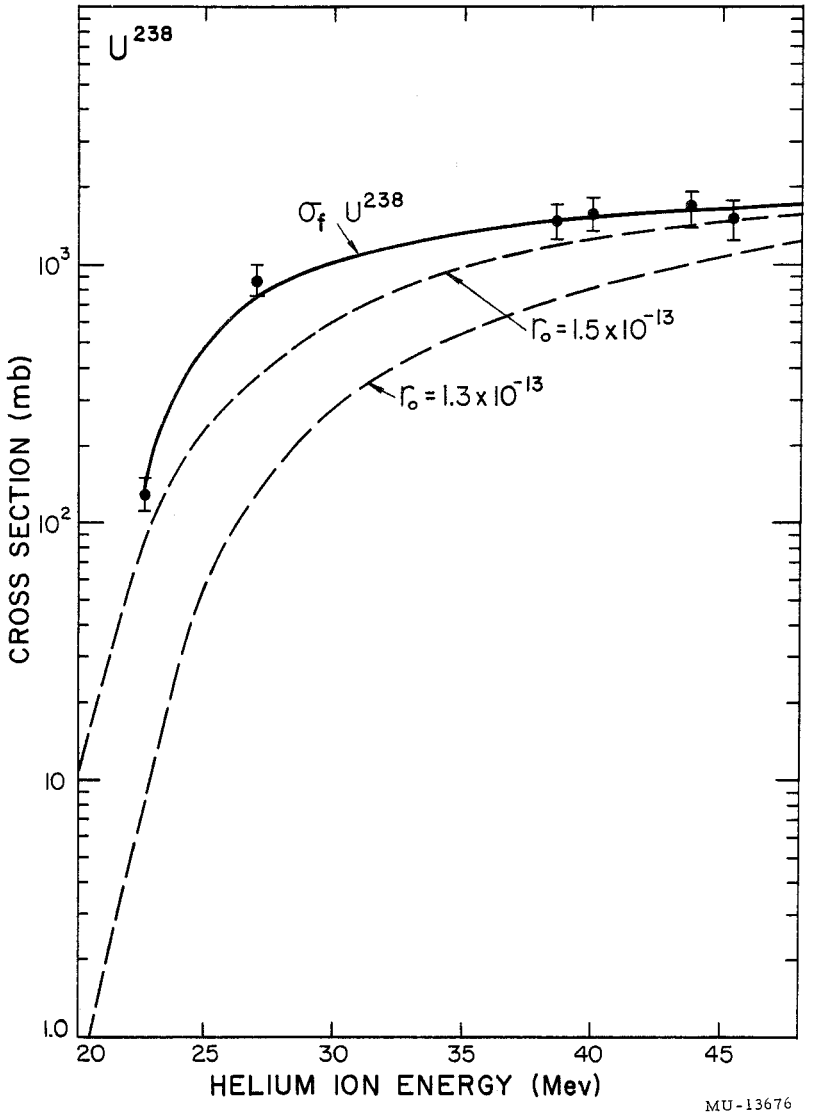

Fig. 13. Total fission yields for helium-ion bombardments of U^{238}. The circles represent experimental data. The dashed lines represent theoretical compound nucleus formation cross sections and were taken from reference 16.

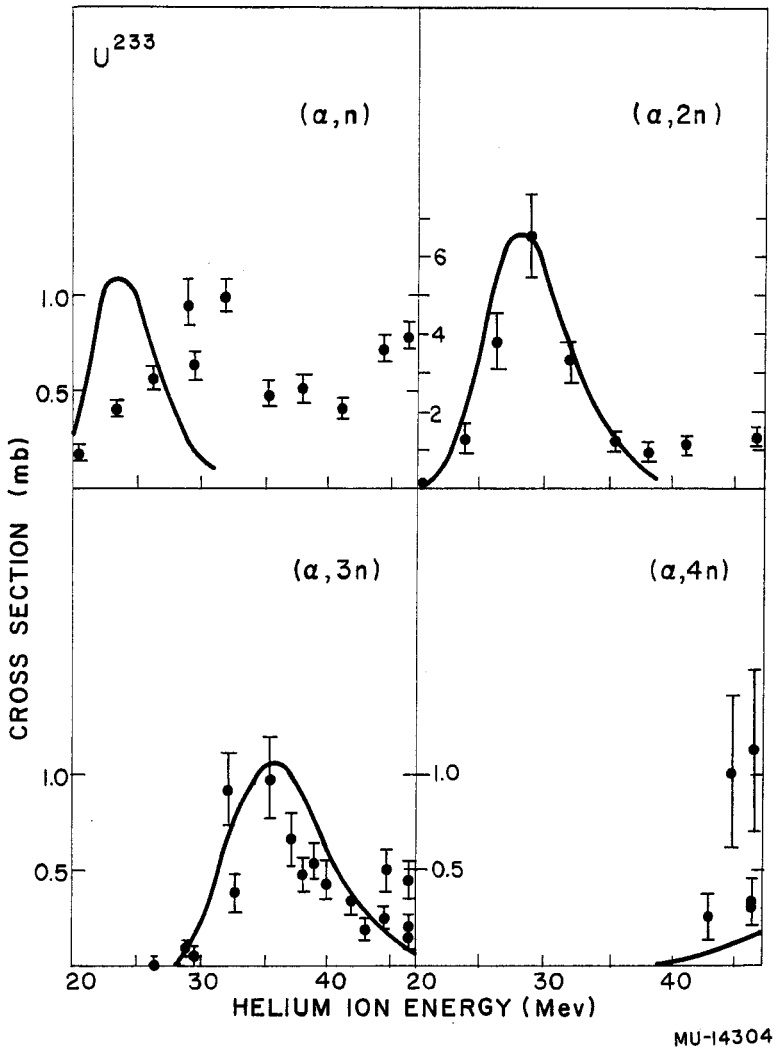

Fig. 14. Comparison of calculated and experimental excitation functions for (α,xn) reactions of U^{233}. The smooth curve represents the calculated cross sections and the actual experimental points are shown as circles.

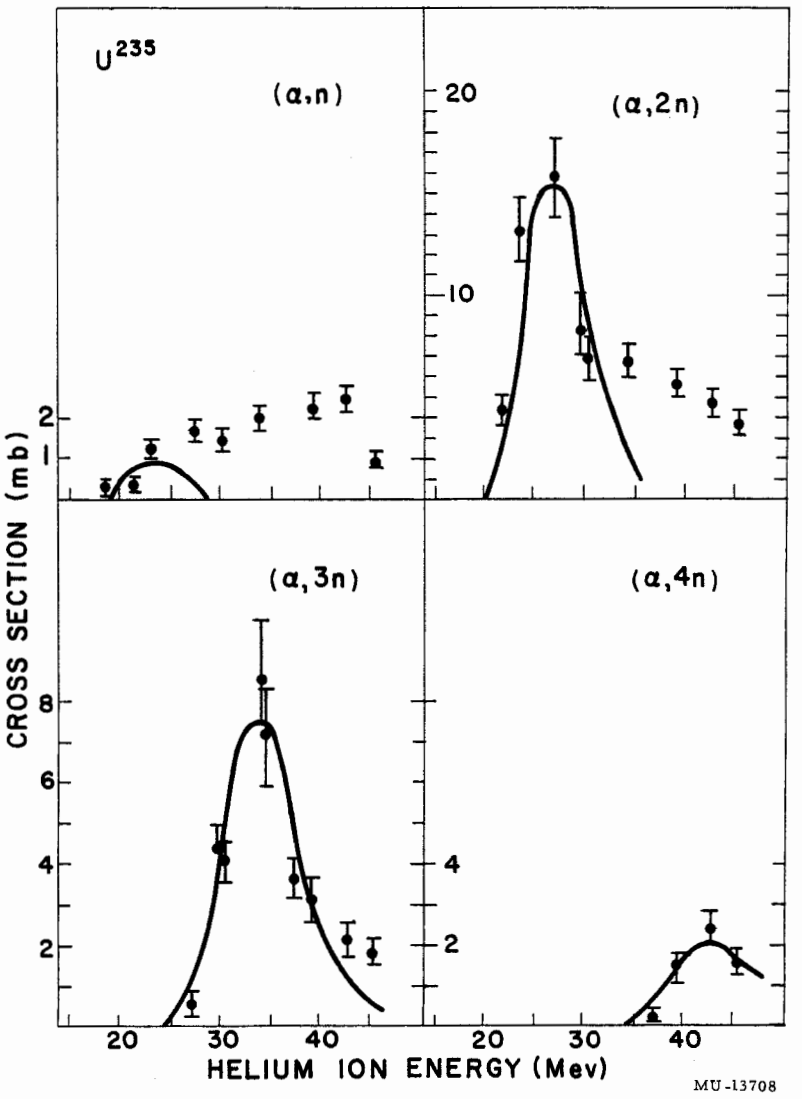

Fig. 15. Comparison of calculated and experimental excitation functions for (α,xn) reactions of U^{235}. The smooth curve represents the calculated cross sections and the actual experimental points are shown as circles.

REFERENCES

1. Glass, Carr, Cobble, and Seaborg, Phys. Rev. 104, 434 (1956).
2. Harvey, Chetham-Strode, Ghiorso, Choppin, and Thompson, Phys. Rev. 104, 1315 (1956).
3. Wade, Gonzalez-Vidal, Glass, and Seaborg, Phys. Rev. 107, 1311 (1957).
4. Gibson, Glass, and Seaborg, to be published.
5. For further details see: S. E. Ritsema, M.S. thesis, University of California, 1956 (unpublished); also available as Radiation Laboratory Report UCRL-3266, January, 1956 (unpublished).
6. The range energy curves of W. A. Aron, B. G. Hoffman, and F. C. Williams were used. U. S. Atomic Energy Commission Document AECU-6631, May, 1951 (unpublished).
7. A. Chetham-Strode, Jr., Ph.D. thesis, University of California, Feb., 1957 (unpublished); also available as University of California Radiation Laboratory Report UCRL-3322, June, 1956 (unpublished).
8. W. W. Meinke, University of California Radiation Laboratory Report, UCRL-432, August, 1949 (unpublished).
9. M. Lindner, University of California Radiation Laboratory Report, UCRL-4377, August, 1954 (unpublished). For further details, see Reference 1.
10. For further details, see Reference 1.
11. W. M. Gibson, Ph.D. thesis, University of California, June, 1957, also available as University of California Radiation Laboratory Report UCRL-3493 (Nov., 1956).
12. Additional points to those reported here have been determined. See Reference 5, and R. Vandenbosch, Ph.D. thesis, University of California, Sept. 1957; also available as University of California Radiation Laboratory Report UCRL-3858, July, 1957 (unpublished).
13. Glendenin, Coryell, and Edwards, Vol. 9, The Fission Products, (National Nuclear Energy Series, McGraw-Hill Book Co., Inc., New York, 1951), paper 52.

14. A. C. Pappas, Proceedings of the International Conference on the Peaceful Uses of Atomic Energy (United Nations Publications, New York, 1956), Vol. 7.

15. R. H. Goeckermann and I. Perlman, Phys. Rev. 76, 628 (1949).

16. J. M. Blatt and V. F. Weisskopf, Theoretical Nuclear Physics, John Wiley and Sons, Inc., New York (1952).

17. R. Hofstadter, Revs. Modern Physics 28, 214 (1956).

18. J. O. Rasmussen, Conference of Nuclear Sizes and Density Distributions, 1957. To be published, Revs. Modern Phys.

19. E. K. Hyde and G. T. Seaborg, Handbuch per Physik, (Springer-Verlag, Berlin), Vol. 39, 1957.

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

21. R. E. Bell and H. M. Skarsgard, Can. J. Phys. 34, 745 (1956).

22. E. L. Kelly, Ph.D. thesis, University of California, 1950; also available as University of California Radiation Laboratory Report UCRL-1044, (1950).

23. This treatment of branching ratios is similar to that described in Appendix II of Reference 1.

24. R. Vandenbosch and G. T. Seaborg, to be published.

25. R. F. Batzel, University of California Radiation Laboratory Report UCRL-4303, Feb., 1954 (unpublished).

26. C. T. Coffin and I. Halpern, University of Washington Cyclotron Research Report, 1957.

27. V. F. Weisskopf, Lecture Series in Nuclear Physics, MDDC-1175, U. S. Government Printing Office, 1947, pp. 106 et. seq.

28. G. N. Harding and F. J. M. Farley, Proc. Phys. Soc. (London), A69, 853 (1956).

29. L. Marquez, Proc. Phys. Soc. (London) A70, 546 (1957).

30. G. Merkle, private communication, 1957.

31. H. A. Tewes and R. A. James, Phys. Rev. 88, 860 (1952); H. A. Tewes, Phys. Rev. 98, 25 (1955).

32. J. Jungerman, Phys. Rev. 79, 632 (1950).

UNIVERSITY OF CALIFORNIA

Radiation Laboratory
Berkeley, California

June 30, 1958

MEMORANDUM

To: Distribution

From: Technical Information Division

Subject: UCRL-8032 Rev., "Spallation-Fission Competition in Heaviest Elements; Helium Ion-Induced Reactions in Uranium Isotopes" by R. Vandenbosch, T. D. Thomas, S. E. Vandenbosch, R. A. Glass and G. T. Seaborg

Please add the attached to your copies of UCRL-8032 Rev.

APPENDIX - - - - - to be added to UCRL-8032 Rev.

Vandenbosch, Thomas, Vandenbosch, Glass, and Seaborg

It will perhaps be informative to present a justification for and to outline the derivation of the model proposed by Jackson and the modification suggested here.

Jackson's model

The assumptions of Jackson's model are (1) that, if it is energetically possible for a neutron to be evaporated, a neutron will be evaporated; (2) that competition from other modes of de-excitation can be neglected; (3) that the neutron energy spectrum is given by $C\epsilon \exp(-\frac{\epsilon}{T})$, where C is a normalization constant, ϵ the kinetic energy of the neutron, and T the nuclear temperature; and (4) that the nuclear temperature is independent of the excitation energy.

From the first three assumptions we conclude that

$$\int_0^{\epsilon_{max}} C\epsilon \exp\left(-\frac{\epsilon}{T}\right) d\epsilon = 1$$

or

$$C = \frac{1}{\int_0^{\epsilon_{max}} \epsilon \exp\left(-\frac{\epsilon}{T}\right) d\epsilon},$$

where ϵ_{max} is the maximum possible kinetic energy of the neutron. For $\epsilon_{max} >> T$

$$C \approx \frac{1}{T^2}$$

and the kinetic energy spectrum of neutrons is given by

$$dR = \frac{\epsilon}{T^2} \exp\left(-\frac{\epsilon}{T}\right) d\epsilon .$$

Let us consider the probability for a nucleus with an initial excitation energy, E, to evaporate three neutrons. The probability that the first two neutrons will have kinetic energies ϵ_1 and ϵ_2 is given by the expression

$$dR = \frac{\epsilon_1}{T^2} \exp\left(-\frac{\epsilon_1}{T}\right) \frac{\epsilon_2}{T^2} \exp\left(-\frac{\epsilon_2}{T^2}\right) d\epsilon_2 \, d\epsilon_1 .$$

If the excitation energy after the evaporation of two neutrons is greater than the neutron binding energy, a third neutron will be emitted. Hence, the probability that at least three neutrons will be evaporated is

$$R_3 = \int_0^{E - B_1 - B_2 - B_3} \int_0^{E - B_1 B_2 - B_3 - \epsilon_1} \frac{\epsilon_1}{T^2} \exp\left(-\frac{\epsilon_1}{T}\right) \frac{\epsilon_2}{T^2} \exp\left(-\frac{\epsilon_2}{T}\right) d\epsilon_2 \, d\epsilon_1 ,$$

where B_1, B_2, and B_3 are the binding energies of the first, second, and third neutrons, respectively. The integration is made over all possible kinetic energies such that the excitation energy remaining after the evaporation of two neutrons is greater than the neutron binding energy.

Making the substitutions

$$\delta_1 = \frac{\epsilon_1}{T} ,$$

$$\delta_2 = \frac{\epsilon_2}{T} ,$$

and

$$\Delta_3 = \frac{E - B_1 - B_2 - B_3}{T} ,$$

we find

$$R_3 = \int_o^{\Delta_3} \int^{\Delta_3 - \delta_1} \delta_1 \exp(-\delta_1) \, \delta_2 \exp(-\delta_2) \, d\delta_2 \, d\delta_1 .$$

Performing the integration, we get

$$R_3 = 1 - e^{-\Delta_3} \sum_{n=0}^{3} \frac{\Delta_3^{\,n}}{n!}$$

$$= I(\Delta_3, 3) ,$$

where I is the incomplete gamma function defined in the body of the paper.

Similarly, it is possible to show that the probability, R_4, of evaporating at least four neutrons is given by

$$R_4 = 1 - e^{-\Delta_4} \sum_{n=0}^{5} \frac{\Delta_4^n}{n!}$$

$$= I(\Delta_4, 5) .$$

The probability for evaporating exactly three neutrons is the difference between the probability for evaporating at least three and the probability for evaporating at least four. Hence,

$$P(E, 3) = R_3 - R_4$$

$$= I(\Delta_3, 3) - I(\Delta_4, 5) .$$

The above is, of course, only a demonstration for a particular case. H. McManus[34] has shown us a rigorous proof of the last equation for the general case of evaporation of x neutrons.

Fission model

To modify Jackson's model for the case where fission is possible, we make two additional assumptions: (1) that $\frac{\Gamma_n}{\Gamma_f}$ is independent of energy, and (2) that a nucleus with an excitation energy greater than the fission activation energy but less than the neutron binding energy always undergoes fission.

The probability that a nucleus evaporates three neutrons and survives fission at each of the evaporation stages is given by

$$R_3 = \int_0^{\Delta_3} \int_0^{\Delta_3 - \delta_1} G_{n_1} \delta_1 \exp(-\delta_1) G_{n_2} \delta_2 \exp(-\delta_2) G_{n_3} d\delta_2 \, d\delta_1 ,$$

where $G_{n_1} = \frac{\Gamma_n}{\Gamma_n + \Gamma_f}$ for the compound nucleus existing before the evaporation of the ith neutron. However, since G_n is independent of energy

$$R_3 = G_{n_1} G_{n_2} G_{n_3} \int_0^{\Delta_3} \int_0^{\Delta_3 - \delta_1} \delta_1 \exp(-\delta_1)\, \delta_2 \exp(-\delta_2)\, d\delta_2\, d\delta_1$$

$$= G_{n_1} G_{n_2} G_{n_3} I(\Delta_3, 3) .$$

In the original model, P_4 is the probability that the nucleus evaporates three neutrons but still has an excitation energy greater than the neutron binding energy. To take fission into account, we must use the probability that the nucleus evaporates three neutrons but still has an excitation energy greater than the fission activation energy. Hence,

$$R_4 = G_{n_1} G_{n_2} G_{n_3} I(\Delta_3^f, 5) ,$$

where

$$\Delta_3^f = \frac{E - B_1 - B_2 - B_3 - E_{th}}{T} ,$$

and E_{th} is the activation energy for fission.[24] The probability for evaporation of exactly three neutrons is

$$P(E, 3) = R_3 - R_4$$

$$= G_{n_1} G_{n_2} G_{n_3} [\, I(\Delta_3, 3) - I(\Delta_3^f, 5) \,].$$