Title

Spallation Fission Competition in the Heaviest Elements

Authors

Thomas, T.D.
Harvey, B.G.
Seaborg, G.T.

Publication Date

1958

UCRL 3989 Rev.

[redacted]LY

UNCLASSIFIED

UNIVERSITY OF CALIFORNIA

Radiation Laboratory

BERKELEY, CALIFORNIA

UNCLASSIFIED

[redacted]Y

~~OFFICIAL USE~~ ONLY

UNCLASSIFIED

SPALLATION FISSION COMPETITION IN THE HEAVIEST ELEMENTS

T. D. Thomas, B. G. Harvey, and G. T. Seaborg[*]

I. INTRODUCTION

Studies of reactions induced in fissionable elements by deuterons and helium ions of moderate energies have yielded information both on the mechanisms of these reactions and on the dependence of the relative probabilities of particle evaporation and fission on such parameters as Z, A, and particle-binding energies.[1-7] It has been found that although most of the reactions involve formation of a compound nucleus, the reactions that survive fission best are those going by a direct interaction without the formation of a highly excited compound nucleus.

II. EXCITATION-FUNCTION MEASUREMENTS

The most useful information has been obtained from a study of the effect of fission competition on spallation, or nonfission, reactions. It has been found that fission accounts for approximately 90% of the total reaction cross section for reactions induced in the heaviest elements by charged particles with energies of 10 to 50 Mev. Hence a large change in fissionability is reflected in a relatively small change in the total fission cross section. On the other hand, a large change in fissionability will cause a correspondingly large change in the few percent of the total cross section taken up by spallation reactions.

EXPERIMENTAL RESULTS

We now consider the cross sections for spallation reactions. Figure 1 shows the excitation functions for (α, xn) reactions of U^{235},[5] and Fig. 2 shows the excitation functions for the (d, xn) reactions of Pu^{239}.[6] Both of these sets of curves are typical of the results obtained for similar studies of the reactions of other heavy-element targets. Cross sections for the (α, n) reaction are of the order of 1 millibarn and are fairly independent of bombarding energy. The excitation functions for the other (α, xn) reactions have peaks and show a characteristic decrease in the height of the peak as x increases. The excitation functions for (d, xn) reactions are similar in shape to those for (α, xn) reactions although somewhat greater in magnitude.

---

[*] Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California.

UNCLASSIFIED

~~OFFICIAL USE~~ ONLY

A comparison of the cross sections for a given (α, xn) reaction ($x > 1$) of different isotopes of the same element shows that the relative probability of fission and particle emission depends very strongly on mass number. In Fig. 3 the excitation functions for (α, 4n) reactions of uranium isotopes are shown; note the large increase in peak height as the mass number of the target is increased. (The curves shown are calculated by the use of a simple evaporation model, which will be discussed in detail below.)

Illustrating the spallation reactions in which charged particles are emitted, Figs. 4 and 5 show cross sections for the (α, p) and (α, p2n) reactions of various targets. A striking similarity of the shapes and magnitudes of the curves for a given type of reaction is observed, as well as a similarity of the curves for the (α, p) reactions to those for the (α, n) reactions. In general, the maximum observed cross section for an (α, p2n) reaction is higher than the maximum observed cross section for the (α, 3n) reaction of the same target - a puzzling result in view of the barrier against the emission of charged particles.

DISCUSSION

A first step in an explanation of the results described is to divide the reactions into those in which the projectile and target unite to form a compound nucleus and those in which an initial direct interaction takes place and most of the available energy is carried off by an emitted particle (or particles). If a compound nucleus is formed, the excitation energy must be carried off by particle evaporation. The higher the excitation energy, the greater will be the number of particles which will have to be evaporated, and hence the longer the time required for the nuclear de-excitation. The longer the time, the greater will be the probability that the excited nucleus will be able to organize itself into a fission mode of oscillation. Hence, since an (α, 3n) reaction involves a longer evaporation chain than does an (α, 2n) reaction, the maximum cross section is lower for the former than the latter. On the other hand, if most of the available energy is carried off by a direct interaction, the residual nucleus may have an excitation energy so low that fission cannot compete with gamma emission. At worst, fission is able to compete only along a very short evaporation chain.

Dividing the reactions into those involving compound-nucleus formation and those involving direct interactions, we may put in the first category the (α, xn) reactions ($x > 1$) and in the second such reactions as (α, n), (d, n), (α, p), (α, pn), and (α, p2n). There are borderline reactions such as (d, 2n) and (α, 2n); the prominent high-energy "tails" on the excitation functions for these reactions suggest that direct interaction is contributing to the cross section for these reactions.

COMPOUND-NUCLEUS REACTIONS

A simple evaporation model, proposed by Jackson,[8] has been modified to take into account the effect of fission competition on neutron evaporation. Jackson makes the assumptions (1) that only neutrons are emitted from a compound nucleus, (2) that if neutron emission is energetically possible, a neutron is emitted, (3) that the probability of emission of a neutron with a kinetic energy ϵ is proportional to $\epsilon \exp(-\epsilon/T)$, and (4) that the nuclear temperature, T, is constant during the evaporation. To take fission into consideration, we have made the following additional assumptions:[5] (1) the branching ratio $\Gamma_n/(\Gamma_n + \Gamma_f)$ is not dependent on excitation energy for energies well above the binding energy of the last neutron, and (2) a nucleus having an excitation energy greater than the fission threshold but less than the binding energy of the last neutron will undergo fission. The cross section for a given (b, xn) reaction is given by

$$\sigma\,(\mathrm{b,xn}) = \sigma_c\, G_1\, G_2 \ldots G_x\, [I(2x-3,\ \Delta_x) - I(2x-1,\ \Delta_x^f)]\ .$$

In this expression, σ_c is the cross section for the formation of a compound nucleus from the projectile b and the target. G is the branching ratio $\Gamma_n/(\Gamma_n + \Gamma_f)$, and the subscripts 1, 2, and x refer to the compound nucleus that exists before the evaporation of the first, second, etc., neutron. I is Pearson's incomplete gamma function defined by

$$I(p,u) = \frac{1}{p!}\int_0^u y^p\, e^{-y}\, dy$$

and the Δ_x is defined as:

$$\Delta_x = \frac{E - \sum_{i=1}^{x} B_i}{T}$$

and Δ_x^f as:

$$\Delta_x^f = \frac{E - \sum_{i=1}^{x} B_i - E_{th}}{T}\ .$$

Here E is the initial excitation energy, B_i is the binding energy of the ith neutron, and E_{th} is the energy at which fission begins to compete with gamma emission. (In making calculations, we have taken neutron binding energies as calculated by Foreman and Seaborg[9] and have used the formula given by Vandenbosch and Seaborg[10] to estimate the fission activation energies.)

The curves shown in Fig. 3 have been calculated by this method, with a nuclear temperature of 1.35 Mev and values of σ_c based on the tables given by Blatt and Weisskopf ($r_0 = 1.5$).[11] The factor necessary to normalize the calculated curves to the experimental values is $(G_1\, G_2\, G_3\, G_4)$. We define an average value of $\Gamma_n/(\Gamma_n + \Gamma_f)$ as

$$\overline{G}_n = (G_1\, G_2\, G_3\, G_4)^{\frac{1}{4}}\ ,$$

and

$$\overline{\Gamma_n/\Gamma_f} = \overline{G}_n/(1-\overline{G}_n)\ .$$

On the basis of the measured cross sections for $(\alpha, 4n)$ reactions of many elements in the heavy-element region, we may correlate the quantity $(\overline{\Gamma_n/\Gamma_f})$ with the parameters Z and A. Such a correlation is shown in Fig. 6.[10] The increase of $(\overline{\Gamma_n/\Gamma_f})$ with increasing A is due partly to the decrease in the fissionability parameter Z^2/A as A increases and partly to the decrease in neutron-binding energies as A increases.

Knowledge of the dependence of $(\overline{\Gamma_n/\Gamma_f})$ on A shown in Fig. 6 enables us to estimate values of (Γ_n/Γ_f) for individual nuclides once a value of $(\overline{\Gamma_n/\Gamma_f})$ is known, and hence to calculate values for the cross sections of all the (α, xn) reactions by the use of the evaporation model described above. The results of such a calculation for reactions induced in U^{235} by helium ions are shown in Fig. 7.[5] The values of G_n for the isotopes Pu^{235}, Pu^{236}, Pu^{237}, Pu^{238}, and Pu^{239} that were used for this calculation are given in Table I, together with values for heavier plutonium isotopes derived from an analysis of the excitation function for the reaction $U^{238}(\alpha, 4n)Pu^{238}$.[7]

Table I

Neutron branching ratios for plutonium isotopes

Nuclide	$\Gamma_n/(\Gamma_n + \Gamma_f)$
Pu^{242}	0.57
Pu^{241}	0.34
Pu^{240}	0.44
Pu^{239}	0.23
Pu^{238}	0.32
Pu^{237}	0.15
Pu^{236}	0.21
Pu^{235}	0.09

In calculating the values shown in Table I, we have multiplied the calculated value of Γ_n/Γ_f by $\sqrt{2}$ if the isotope was even-even and by $1/\sqrt{2}$ if even-odd. According to Weisskopf[12] the level density of an even-odd nucleus for a given excitation energy will be approximately twice that of an even-even nucleus of the same excitation. The factors of $\sqrt{2}$ and $1/\sqrt{2}$ are used to adjust the individual calculated values of Γ_n/Γ_f for this effect, since these values represent averages over both nuclear types.

It is to be noted that at high energies of the bombarding particle the experimental cross sections for the (α, n) and $(\alpha, 2n)$ reactions are considerably higher than those predicted by the simple evaporation model. This phenomenon may be interpreted as being due to a direct interaction in which one neutron with high energy is knocked out by the incident particle, leaving insufficient excitation energy for the evaporation of more than one subsequent particle.

The success of the simple evaporation model in reproducing the major features of the excitation functions for the (α, xn) reactions suggests that the assumptions made regarding variation of Γ_n/Γ_f with energy and nuclear type are approximately correct. These assumptions are (1) that Γ_n/Γ_f is independent of excitation energy for energies well above the threshold for neutron emission; (2) that, except for effects due to nuclear type, Γ_n/Γ_f for a given value of Z increases by 30% for a unit increase of A; and (3) that the level density of an even-even nuclide is one-half that of an even-odd nuclide for a given excitation energy.

III. STUDIES OF FRAGMENTS EMITTED IN SPALLATION REACTIONS

Studies of the reactions thought to involve a direct interaction between the projectile and a few nucleons in the nucleus have followed two lines: work has been done to identify the particles emitted in the direct interaction, and measurements have been made of the angular distributions of the heavy fragments recoiling from the target.

TRITON PRODUCTION

It has been possible to identify radiochemically tritium produced when gold, thorium, and uranium are bombarded with 32-Mev protons, 24-Mev deuterons, and 48-Mev helium ions.[3] Bombardments have been made on stacked foils of the target materials, and the amount of tritium produced in each foil has been measured. Figure 8 shows the results of such measurements.

Two features of these curves are to be noted. First, the curves for a given reaction are about the same in both shape and magnitude for all three targets, suggesting that fission is not competing with this sort of reaction. Second, the cross section for the (α, t) reaction reaches its maximum in a foil not reached by the helium-ion beam. Evidently the tritons have been emitted with fairly high kinetic energies. Hence, the residual nuclei are left with a low excitation energy, and further reactions such as neutron evaporation and fission are unlikely.

In the case of Th^{232} and U^{238}, values for the cross section of the $(\alpha, p2n)$ reaction have been measured radiochemically. As is shown in Table II, the integrated values of these cross sections are very close to the integrated values for the (α, t) reactions: this agreement suggests that most of the $(\alpha, p2n)$ reaction goes by a mechanism in which a triton is emitted.

Table II

Integrated (α, t) cross sections

Target	Tritons/incident particle	
	Tritium measurements	Heavy-fragment measurements
Th^{232}	$(1.56 \pm 0.13) \times 10^{-5}$	1.27×10^{-5}
U^{238}	$(1.23 \pm 0.09) \times 10^{-5}$	1.08×10^{-5}

Further work is in progress to measure the energies and angular distributions of the light particles emitted in direct interactions.

RECOIL STUDIES

As mentioned above, the high-energy "tail" observed on excitation curves for (a, 2n) reactions may be due to direct interactions in which the first neutron is emitted with high energy. We find further support for this explanation in the angular distributions and kinetic energy spectra of the products from (a, 2n) reactions.

The kinetic energies and angular distributions of the At^{211} and Cf^{245} recoils from the reactions Bi^{209} (a, 2n) and Cm^{244} (a, 2n) are consistent with the formation of a compound nucleus followed by isotropic evaporation of neutrons only when the incident helium has an energy of not more than a few Mev above the energetic threshold of the reaction. Experiments show that for incident-helium-ion energies greater than this, however, the momentum of the recoil nuclei is as low as one half of the momentum of the helium ion: the momentum is presumably balanced by the direct ejection of a high energy neutron in the forward direction. Both the angular distributions and cross sections in this energy region are consistent with the emission of the second neutron by evaporation.

REFERENCES

1. Glass, R. A., Carr, R. J., Cobble, J. W., and Seaborg, G. T., Spallation-Fission Competition in Heaviest Elements; Helium-Ion-Induced Reactions in Plutonium Isotopes, Phys. Rev. 104: 434 (1956).

2. Harvey, B. G., Chetham-Strode, A., Jr., Ghiorso, A., Choppin, G. R., and Thompson, S. G., New Isotopes of Einsteinium, Phys. Rev. 104: 1315 (1956).

3. Wade, W. H., Gonzalez-Vidal, J., Glass, R. A., and Seaborg, G. T., Spallation-Fission Competition in Heaviest Elements; Triton Production, Phys. Rev. 107: 1311 (1957).

4. Sikkeland, T., Amiel, S., and Thompson, S. G., Spallation Reactions of Californium-252 with Helium Ions, submitted to Physical Review for publication (1958).

5. Vandenbosch, R., Thomas, T. D., Vandenbosch, S. E., Glass, R. A., and Seaborg, G. T., Spallation-Fission Competition in Heaviest Elements; Helium-Ion-Induced Reactions in Uranium Isotopes, submitted to Physical Review for publication (1958).

6. Gibson, W. M., Fission and Spallation Competition from the Intermediate Nuclei Americium-241 and Neptunium-235, Ph. D. thesis, University of California, June, 1957; also available as UCRL-3493 (November, 1956).

7. Unpublished work of Wade, W. H., Coleman, J. E., Foreman, B. M., Jr., Gonzalez-Vidal, J., Gordon, G. E., Lessler, R. M., Luoma, E. V., and Vandenbosch, R., University of California Radiation Laboratory.

8. Jackson, J. D., A Schematic Model for (p, xn) Cross Sections in Heavy Elements, Can. J. Phys. 34: 767 (1956).

9. Foreman, B. M., Jr., and Seaborg, G. T., Nuclear Thermodynamics of the Heaviest Elements. II., submitted to the J. Inorg. Nucl. Chem. (1958).

10. Vandenbosch, R., and Seaborg, G. T., Some Considerations of Nuclear Fission, to be published in Phys. Rev.

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

12. Weisskopf, V. F., Lecture Series in Nuclear Physics, MDDC 1175, U. S. Government Printing Office (1947).

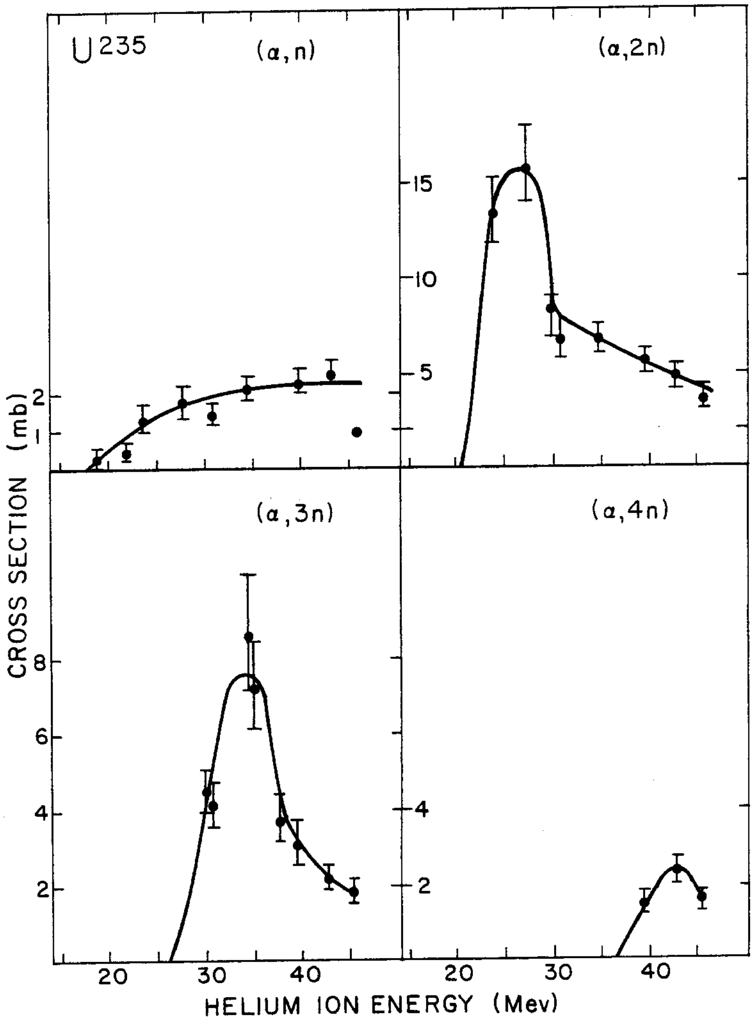

Fig. 1. Excitation functions for (α, xn) reactions of U^{235}.

UNCLASSIFIED

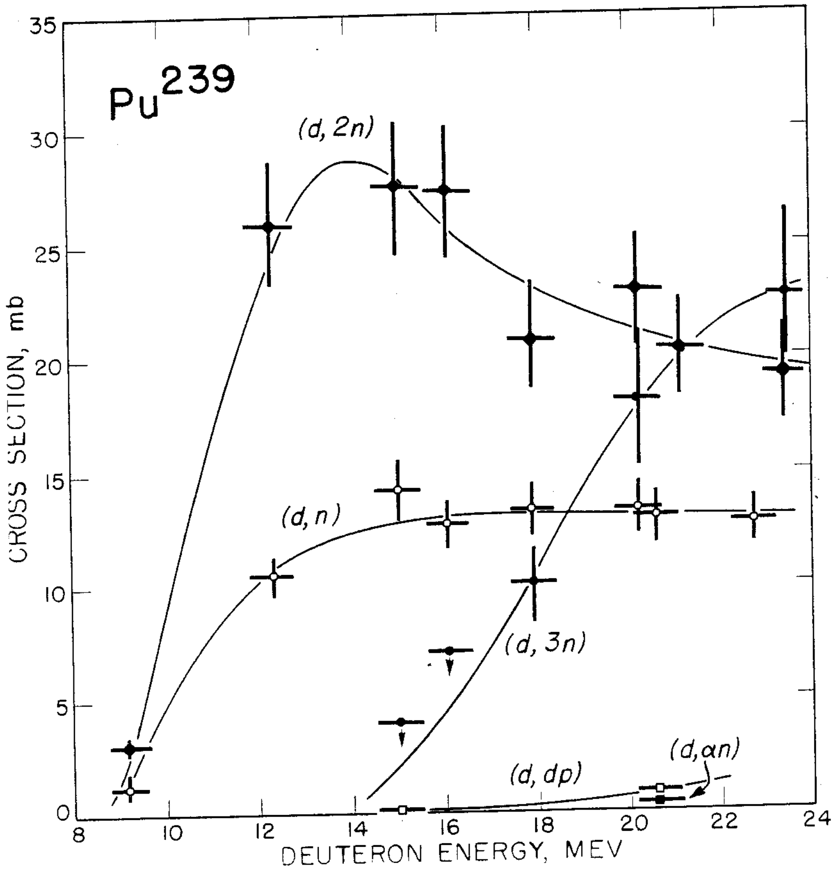

Fig. 2. Excitation functions for (d, xn) reactions of Pu^{239}.

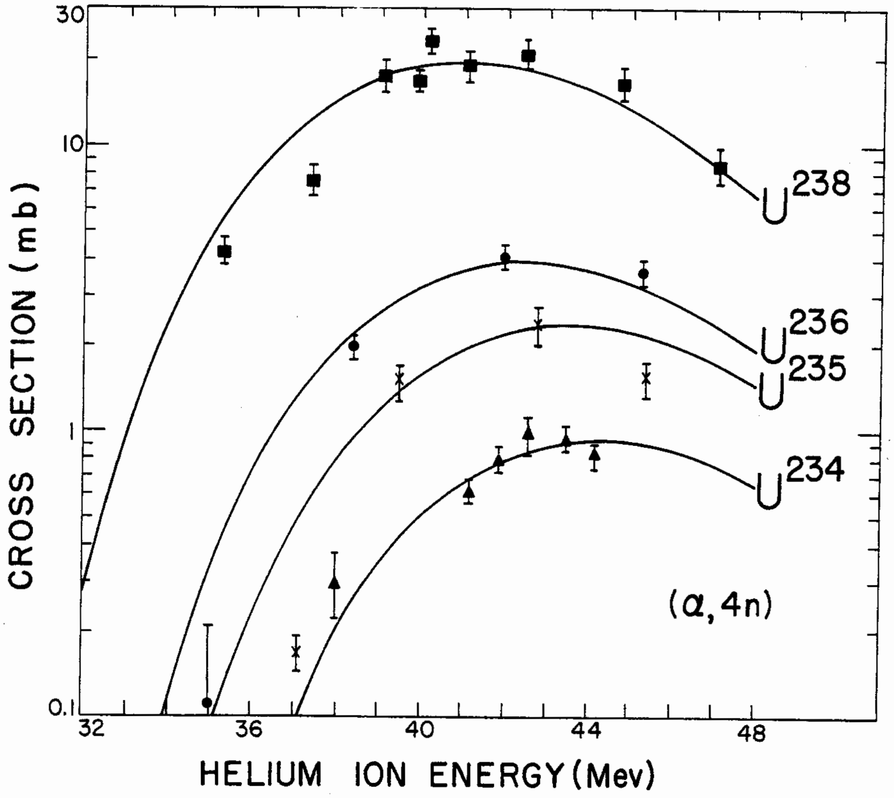

Fig. 3. Excitation functions for (α, 4n) reactions of uranium isotopes.

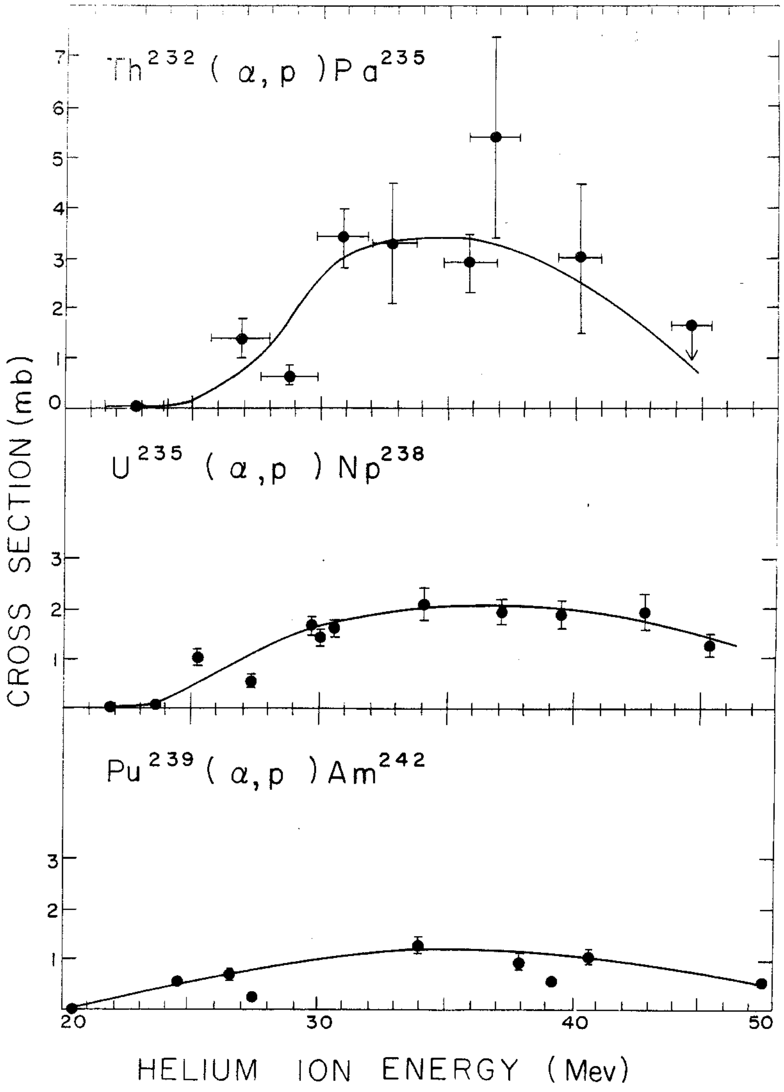

Fig. 4. Excitation functions for (α, p) reactions.

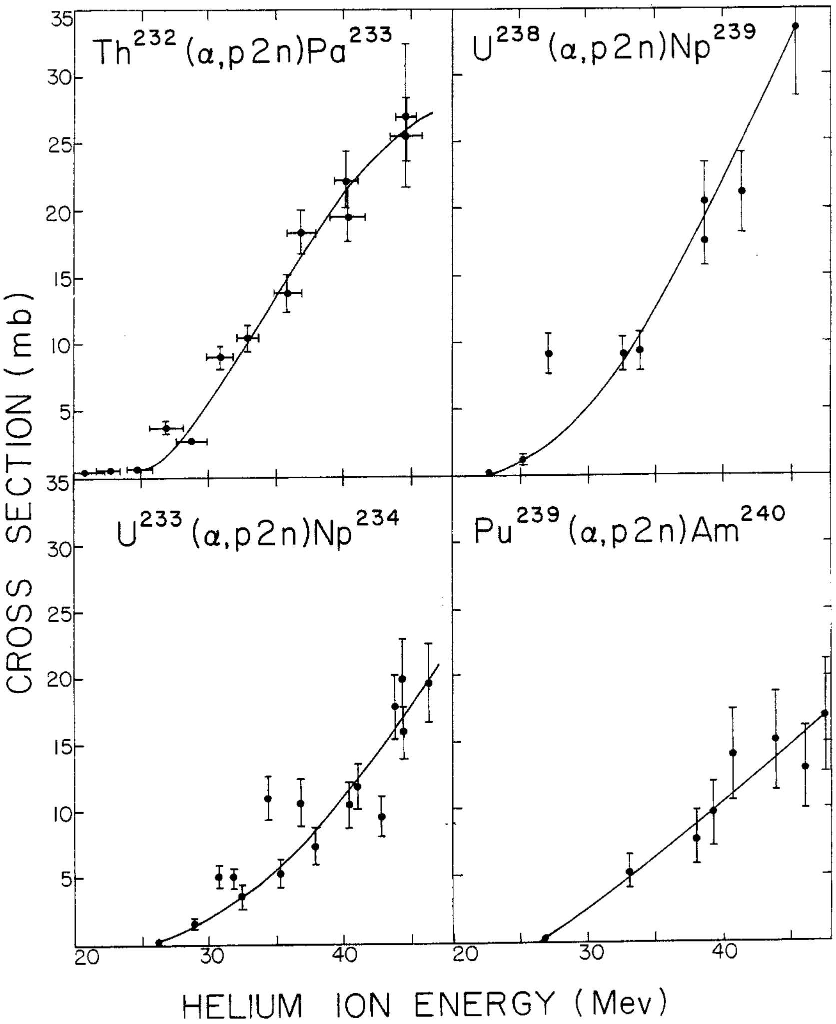

Fig. 5. Excitation functions for (a, p2n) reactions.

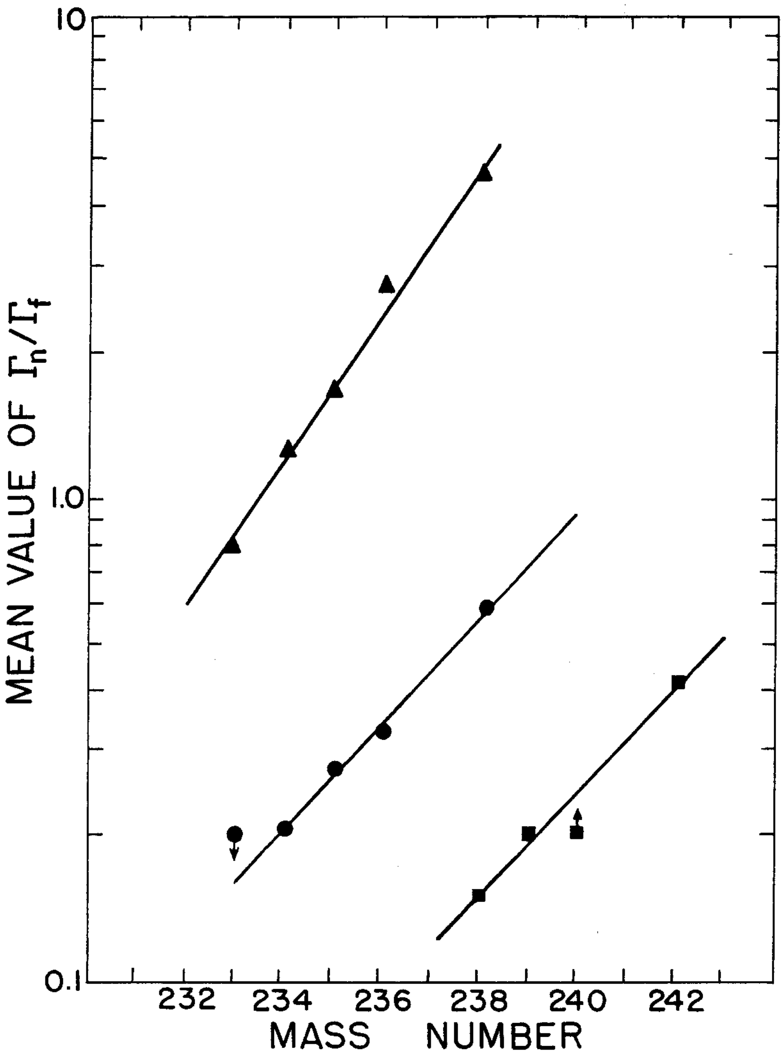

Fig. 6. Dependence of $(\overline{\Gamma_n/\Gamma_f})$ on Z and A.

▲ uranium isotopes
● plutonium isotopes
■ curium isotopes

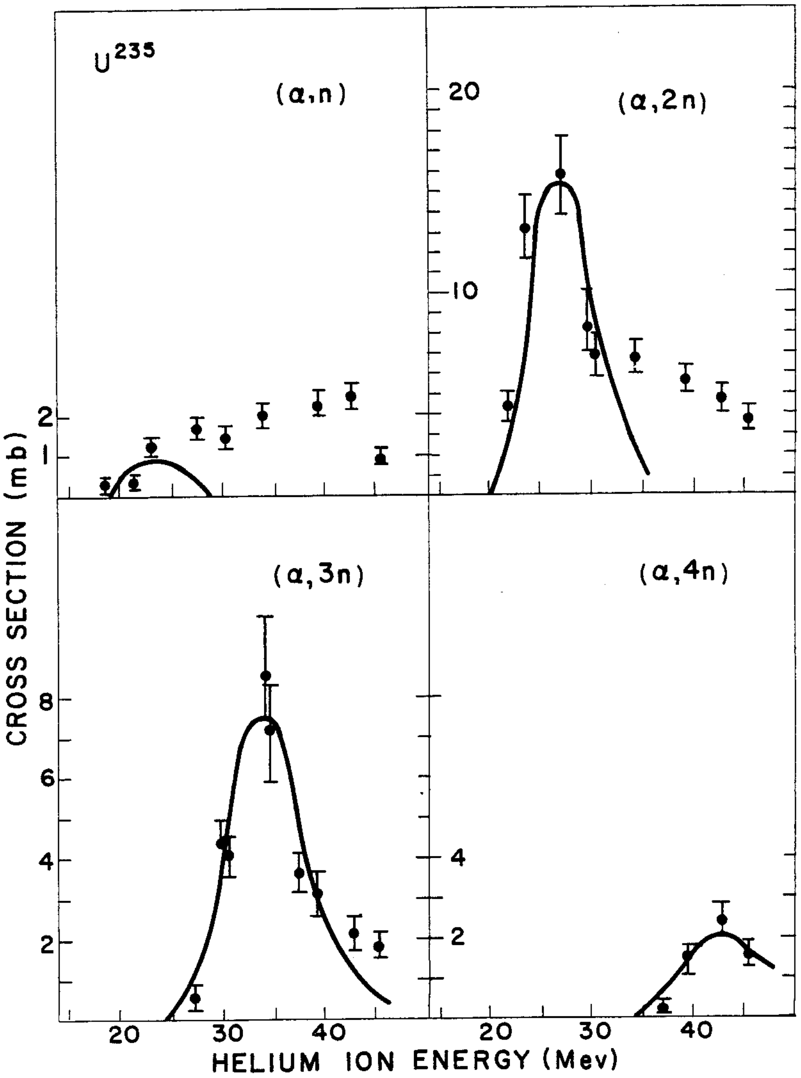

Fig. 7. Comparison of calculated and experimental cross sections for (α, xn) reactions of U^{235}.

UNCLASSIFIED

~~[illegible] ONLY~~

UNCLASSIFIED

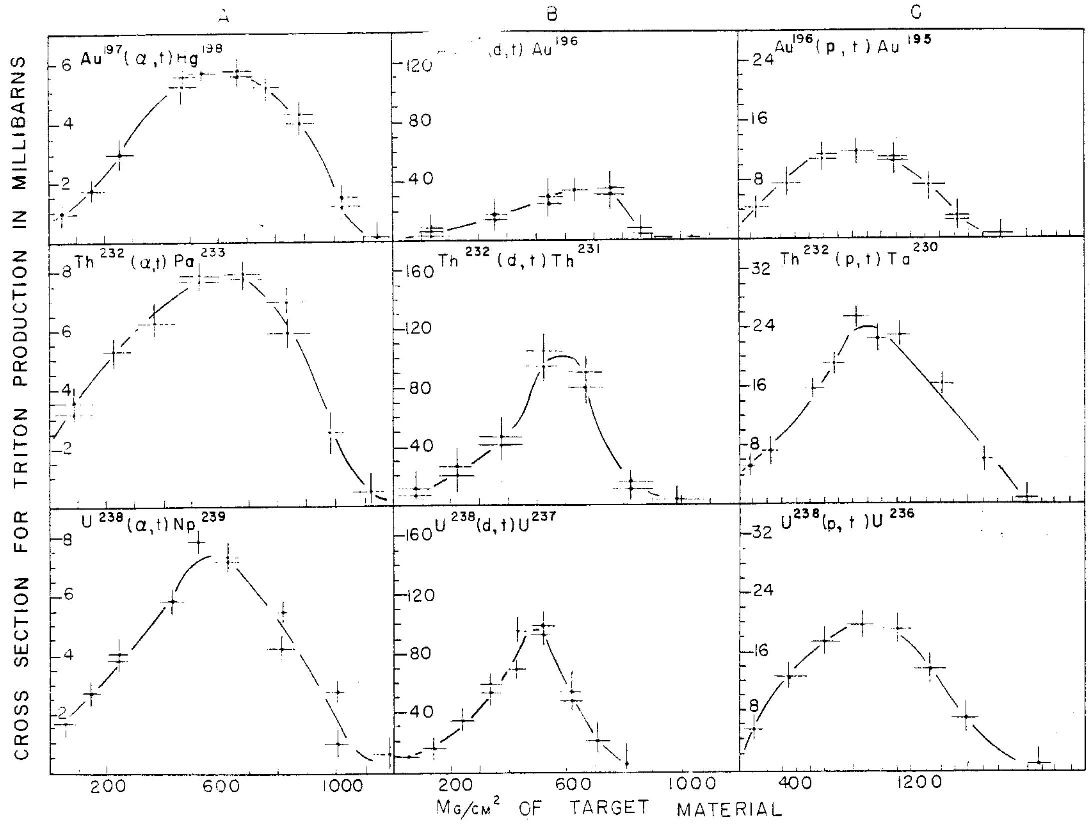

Fig. 8. Tritium production by charged-particle bombardment of Au, Th, and U.

UNCLASSIFIED

~~OFFICIAL USE ONLY~~