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Publication Date

1957-11-01

UCRL 8047

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Radiation Laboratory
Berkeley, California

Contract No. W-7405-eng-48

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Robert Vandenbosch and Glenn T. Seaborg

November, 1957

Printed for the U. S. Atomic Energy Commission

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Contents

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November 1957

ABSTRACT

A semi-empirical equation for fission threshold has been extended to include the effects of unpaired nucleons on the rate of spontaneous fission. Excitation functions for the $(\alpha,4n)$ reactions of Ra^{226}, Th^{230}, and U^{236} have been measured. These results and reported cross sections for other $(\alpha,4n)$ reactions in the heaviest elements have been analyzed in terms of fission and neutron-emission competition to obtain mean values of Γ_n/Γ_f. These mean values of Γ_n/Γ_f have been correlated with neutron binding energies and fission thresholds.

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*This work was performed under the auspices of the U. S. Atomic Energy Commission. It is based in part on work done by Robert Vandenbosch in partial fulfillment of the requirements for the Ph.D. degree at the University of California.

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SOME CONSIDERATIONS ON THE PROBABILITY OF NUCLEAR FISSION

I. INTRODUCTION

In their original considerations of the fission process employing the liquid drop model, Bohr and Wheeler[1] showed the potential importance of a fissionability parameter Z^2/A which represents the ratio of the nuclear Coulomb repulsive energy to the stabilizing nuclear surface energy. This parameter has been used to designate the relative tendency of different heavy nuclei for thermal-neutron-induced fission.[2,3]

The probabilities for fission deduced from fast neutron (3-5 Mev) fission cross sections have been correlated with Z^2/A [4,5] and also with the difference between the fission threshold and neutron binding energy.[6] Huizenga, Gindler, and Duffield correlated the relative photofission yields (~15 Mev) of different nuclei with Z^2/A.[7,8] In the present paper we shall investigate further the applicability of this parameter to the description of the relative probability for fission of various nuclei in the intermediate energy range up to about 50 Mev.

The total fission cross section for the reaction of charged particles with heavy elements is not a very sensitive measure of the relative fissionability of different nuclides, as the fission cross section usually accounts for more than 80% of the total reaction cross section and does not vary much from nuclide to nuclide.[9,10] However, the effect of fission competition on spallation reactions, particularly those occuring by compound nucleus mechanisms, is quite a sensitive measure of fissionability.[9] The $(\alpha,4n)$ reaction is particularly sensitive to fission competition as fission has had four chances to compete with neutron emission along the evaporation chain. It is also quite likely that the $(\alpha,4n)$ reaction proceeds almost exclusively through a compound nucleus mechanism. For these reasons the $(\alpha,4n)$ reaction was chosen to investigate the applicability of the parameter Z^2/A to fission competition in the heavy elements at moderate excitation energy. In addition to making a literature survey of measured $(\alpha,4n)$ excitation functions of fissionable elements, a few isotopes were chosen for additional study. These experiments are described briefly in Section II, and a discussion of the results in terms of these considerations follows in Section III.[11]

In connection with this description as well as in connection with the description of other results obtained in our general program of investigation of spallation-fission competition in the heaviest elements[10] values for the fission energetic thresholds for the various nuclei have been needed. A number of methods for calculating fission thresholds are available[3,12,13] and one of the simplest[3] is based on a comparison with the spontaneous fission decay rates for even-even nuclides. The applicability of this method of calculating fission thresholds is further extended in Section A of this paper where the effects of different nuclear types are considered and a concept of activation energy for fission is discussed.

A. Activation Energy for Fission

It has been shown that spontaneous fission lifetimes for even-even nuclides have an exponential dependence on the parameter Z^2/A.[14-16] Several years ago a semi-empirical equation[17] for the fission barrier, E_b, was derived from an empirical equation for observed spontaneous fission lifetimes and from theoretical considerations on the barrier penetration probability for spontaneous fission. It was assumed that the form of an equation given by Frankel and Metropolis,[12] $T = 10^{-21} \times 10^{7.85\ \Delta E}$ seconds, governing the dependence of spontaneous fission half life on the fission barrier is approximately correct. This led to the expression $E_b = (19.0 - 0.36\ Z^2/A)$ Mev. This is applicable only to intermediate compound nuclei of the even-even type because the relationship between observed spontaneous fission lifetimes and Z^2/A applies only to this nuclear type. Even-odd and odd-even nuclides are retarded in their rate of spontaneous fission decay by an average factor of about 10^3, and the decay rates of odd-odd nuclides are retarded by a factor of about 10^5. The equation given by Frankel and Metropolis,[12] $T = 10^{-21} \times 10^{7.85\ \Delta E}$ seconds, predicts that each factor of ten increase in half life corresponds to an increase of about 0.13 Mev in barrier height. This is consistent with the fission lifetimes of U^{238} in its ground state and at the fission threshold. Assuming a fission lifetime of about 10^{-14} seconds at the observed photofission threshold of 5.1 Mev, the lifetime of U^{238} excited to 5.1 Mev is approximately 10^{37} times shorter than the fission lifetime of U^{238} in its ground state, which has a spontaneous fission half life of about 10^{16} years. This corresponds to 0.136 Mev change in barrier height for each factor of ten change in fission lifetime, in satisfactory agreement with the value predicted by the equation of Frankel and Metropolis. Use of their predicted value indicates that fission barriers for even-odd and odd-even nuclides are higher than even-even nuclides by about 0.4 Mev, and for odd-odd nuclides are higher by about 0.7 Mev. Thus, the relationship becomes

$$\text{(I) } E_b = (19.0 + 0.36\ Z^2/A + \epsilon)\ \text{Mev}$$

where $\epsilon = 0$ for even-even nuclides, $\epsilon = 0.4$ for even-odd and odd-even nuclides and $\epsilon = 0.7$ for odd-odd nuclides.

Due to the barrier-penetration nature of the fission process, induced fission will be observed at the point below the barrier where the time for fission becomes comparable with the time for gamma emission, i.e., in a time of about 10^{-14} seconds. The required energy of activation, E_a, will be less than the barrier height, E_b, which represents a fission time of some 10^{-21} seconds. Thus, if we again use the relationship that each factor of ten in rate corresponds to some 0.13 Mev of energy, it follows that E_a, is, in general, some 0.9 Mev less than E_b.

The energy difference B_n (neutron binding energy) minus E_a (calculated) has been tabulated in Table I, and the correlation with slow-neutron fission is surprisingly good. The nuclides which show a positive energy difference have a fission cross section greater than about one barn, and the nuclides with a negative (B_n minus E_a) energy difference have fission cross sections below this arbitrary line of demarcation for slow-neutron fissionable nuclides. When the value of E_a exceeds the neutron binding energy, B_n, leading to a negative value for (B_n minus E_a) in Table I, this should be equal to the neutron threshold for fission. From the table, the following nuclides should have the indicated thresholds for neutron-induced fission: Th^{232} (0.9 Mev), Pa^{231} (0.4 Mev), U^{234} (0.3 Mev), U^{236} (0.3 Mev), U^{238} (0.9 Mev), and Np^{237} (0.3 Mev). Fission thresholds are not sharp due to the barrier penetration nature of the fission process and therefore experimentally determined thresholds depend somewhat on the sensitivity of the measuring technique. The following thresholds have been experimentally determined:[18] Th^{232} (1.1 Mev), Pa^{231}

Table I. Correlation of slow neutron fissionability with activation energy for fission and corresponding neutron binding energy.

Nuclide	E_b* (Mev)	E_a** (Mev)	B_n*** (Mev)	B_n-E_a (Mev)	Slow Neutron Fissionability****	Source of Slow Neutron Fission Cross Section[a]
Ra^{226}	7.1	6.2	4.5	-1.7	-	
Ra^{228}	7.2	6.3	4.8	-1.5	-	
Ac^{227}	7.2	6.3	5.0	-1.3	-	
Th^{227}	6.2	5.3	7.1	1.8	+	
Th^{228}	6.7	5.8	5.4	-0.4	-	
Th^{229}	6.3	5.4	6.7	1.3	+	
Th^{230}	6.8	5.9	5.0	-0.9	-	
Th^{232}	6.9	6.0	5.1	-0.9	-	
Th^{233}	6.5	5.6	6.1	+0.5	+	
Th^{234}	7.0	6.1	4.6	-1.5	-	
Pa^{230}	6.5	5.6	6.8	1.2	+	
Pa^{231}	6.8	5.9	5.5	-0.4	-	
Pa^{232}	6.6	5.7	6.7	1.0	+	
Pa^{233}	7.0	6.1	5.2	-0.9	-	
U^{230}	6.2	5.3	5.9	0.6	+	
U^{231}	5.9	5.0	7.3	2.3	+	
U^{232}	6.3	5.4	5.9	0.5	+	
U^{233}	6.0	5.1	6.8	1.7	+	
U^{234}	6.4	5.5	5.2	-0.3	-	
U^{235}	6.1	5.2	6.4	1.2	+	
U^{236}	6.5	5.6	5.3	-0.3	-	
U^{238}	6.6	5.7	4.8	-0.9	-	
U^{239}	6.3	5.4	5.9	0.5	+	
Np^{234}	6.1	5.2	6.8	1.7	+	
Np^{236}	6.2	5.3	6.8	0.5	+	
Np^{237}	6.6	5.7	5.4	-0.3	-	
Np^{238}	6.4	5.5	6.2	0.7	+	
Np^{239}	6.7	5.8	5.1	-0.7	-	

(continued)

Table I. (continued)

Nuclide	E_b^{*} (Mev)	E_a^{**} (Mev)	B_n^{***} (Mev)	$B_n - E_a$ (Mev)	Slow Neutron Fissionability[****]	Source of Slow Neutron Fission Cross Section[a]
Pu^{236}	6.0	5.1	6.1	1.0	+	b
Pu^{238}	6.1	5.2	5.6	0.4	+	
Pu^{239}	5.8	4.9	6.4	1.5	+	
Pu^{240}	6.2	5.3	5.4	0.1	+	c
Pu^{241}	5.9	5.0	6.2	1.2	+	
Pu^{242}	6.3	5.2	5.1	-0.1	-	d
Am^{241}	6.2	5.3	5.6	0.3	+	
Am^{242m}	6.0	5.1	6.3	1.2	+	
Am^{242}	6.0	5.1	6.3	1.2	+	
Am^{243}	6.4	5.5	5.2	-0.3	-	e
Cm^{242}	5.8	4.9	5.7	0.8	?	
Cm^{243}	5.4	4.5	6.7	2.2	+	
Cm^{244}	5.9	5.0	5.7	0.7	?	
Cm^{245}	5.5	4.6	6.4	1.8	+	
Cf^{249}	5.1	4.2	6.6	2.2	+	
E^{254}	5.5	4.6	5.8	1.2	+	f

[*] Potential barrier for fission.

[**] Activation energy for fission.

[***] Neutron binding energy for nuclide with mass number A + 1.

[****] The + denotes cross section for fission is greater than about 1 barn.

The - denotes cross section for fission is less than about 1 barn.

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[a] Except when noted otherwise all of the cross sections were taken from the compilations of Ref. 18 or Huizenga, Manning, and Seaborg, The Actinide Elements, edited by G. T. Seaborg and J. J. Katz, (McGraw-Hill Book Co. Inc., New York, 1954) Chap. 20, National Nuclear Energy Series, Vol. 14A, Div. IV, p. 839.

[b] J. R. Huizenga, private communication (1957).

[c] Hulet, Bowman, Michel and Hoff, Phys. Rev. 102, 1621 (1956).

[d] W. C. Bentley *et al.*, Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955 (United Nations; New York, 1956) Vol. 7, p. 261.

[e] Hulet, Hoff, Bowman and Michel, Phys. Rev. 107, 1294 (1957).

[f] S. G. Thompson *et al.*, unpublished results, 1955.

(0.4 Mev), U^{234} (0.3 Mev), U^{236} (0.6 Mev), U^{238} (0.9 Mev), and Np^{237} (0.3 Mev). It can be seen that the agreement between the predicted and the experimentally determined threshold values is good.

Recently a method for exciting nuclei to less than the neutron binding energy by the (d,p) reaction has been developed to measure fission thresholds.[19] The fission threshold is obtained by measuring the energy spectrum of protons in coincidence with fission events induced by deuterons of known energy. Preliminary results[20] indicate that U^{235} undergoes fission at an excitation energy of about 1.2 Mev below that given to it by an added slow neutron, in good agreement with the predicted value of 1.2 Mev suggested by Table I.

The E_a values calculated from an equation using a straight line dependence of spontaneous fission half lives on Z^2/A can be only approximate at best, because the rate for this process depends on more complicated factors than just a dependence on Z^2/A. Although the parameter Z^2/A accounts for the general trend of spontaneous fission lifetimes, it has been pointed out that for a given value of Z the half life goes through a maximum as A varies.[21] In addition it has been noted that there is an increase in the spontaneous fission rate for nuclides with more than 152 neutrons.[22] Swiatecki[13,23] has successfully related these deviations from a simple Z^2/A calculation by considering the energy difference between a smooth saddle point energy surface (as a function of Z and N) and the actual experimental ground state masses.

II. EXPERIMENTAL RESULTS

Excitation functions for the $(\alpha,4n)$ reactions of Ra^{226}, Th^{230}, and U^{236} were measured using the external beam of the Crocker Laboratory 60-inch cyclotron. The radium used was isotopically pure Ra^{226}. The thorium had an isotopic composition of 87.85 ± 0.1% Th^{230} and 12.22 ± 0.1% Th^{232}. The uranium had an isotopic composition of 94.9% U^{236}, 0.04% U^{234}, 4.52% U^{235}, and 0.54% U^{238}. The targets were prepared by electrodeposition of the various materials onto gold or aluminum foils. The uranium and radium targets were dissolved after each bombardment and plutonium and thorium fractions, respectively, were isolated radiochemically. A recoil technique, similar in principle to that described by Harvey, et al.,[24] was used for the thorium cross section measurements. This permitted the use of the same target for all of the bombardments. A small amount of Pu^{239} was also deposited in the thorium target and the Pu^{239} $(\alpha,3n)$ Cm^{240} reaction, for which absolute cross sections have been measured,[9] was used as a monitor reaction to determine the collection efficiency of the heavy-element-nuclei recoils. The catcher foils were dissolved and the uranium and curium fractions were isolated radiochemically. The amounts of the various alpha-emitting products were determined by use of 52%-geometry ionization counters and multi-channel alpha-pulse-height analyzers.

The cross sections determined for the Ra^{226} $(\alpha,4n)$ Th^{226} reaction are listed in Table II and illustrated in Fig. 1. The estimated limits of error of ± 20% are due principally to uncertainties in determining the amount of target material which was bombarded.

The cross sections for the Th^{230} $(\alpha,4n)$ U^{230} reaction, corrected for recoil efficiency, are listed in Table III and illustrated in Fig. 2. The results from the monitor reaction Pu^{239} $(\alpha,3n)$ Cm^{240} indicated an average recoil collection efficiency of 80 ± 5% for all of the bombardments. The estimated limits of error for the corrected $(\alpha,4n)$ reaction cross sections are ± 7%.

The cross sections for the U^{236} $(\alpha,4n)$ Pu^{236} reaction are listed in Table IV and illustrated in Fig. 3. The contribution of the Pu^{236} produced by the $(\alpha,3n)$ reaction from the U^{235} present in the target has been subtracted. Any appreciable contribution of Pu^{236} from the decay of Np^{236} was eliminated by removing neptunium chemically very soon after the bombardment. The estimated limits of error are listed in the table.

Table II. Ra^{226} $(\alpha,4n)$ Th^{226} cross sections (mb) as a function of helium-ion energy.

E (Mev)	σ (mb)
35.6	110±20
38.2	270±50
39.4	260±50
39.4	380±80
40.8	500±100
42.7	420±80
44.7	490±100
45.5	200±40

Table III. Th^{230} $(\alpha,4n)$ U^{230} cross sections (mb) as a function of helium-ion energy.

E (Mev)	σ (mb)
38.0	2.8±0.2
40.0	10.2±0.7
41.2	12.5±0.9
42.6	12.9±0.9
43.3	12.2±0.8
44.2	11.5±0.8

Table IV. U^{236} $(\alpha,4n)$ Pu^{236} cross sections (mb) as a function of helium-ion energy.

E (Mev)	σ (mb)
34.5	0.11±0.1
38.4	2.0±0.2
42.0	4.1±0.4
45.6	3.6±0.4

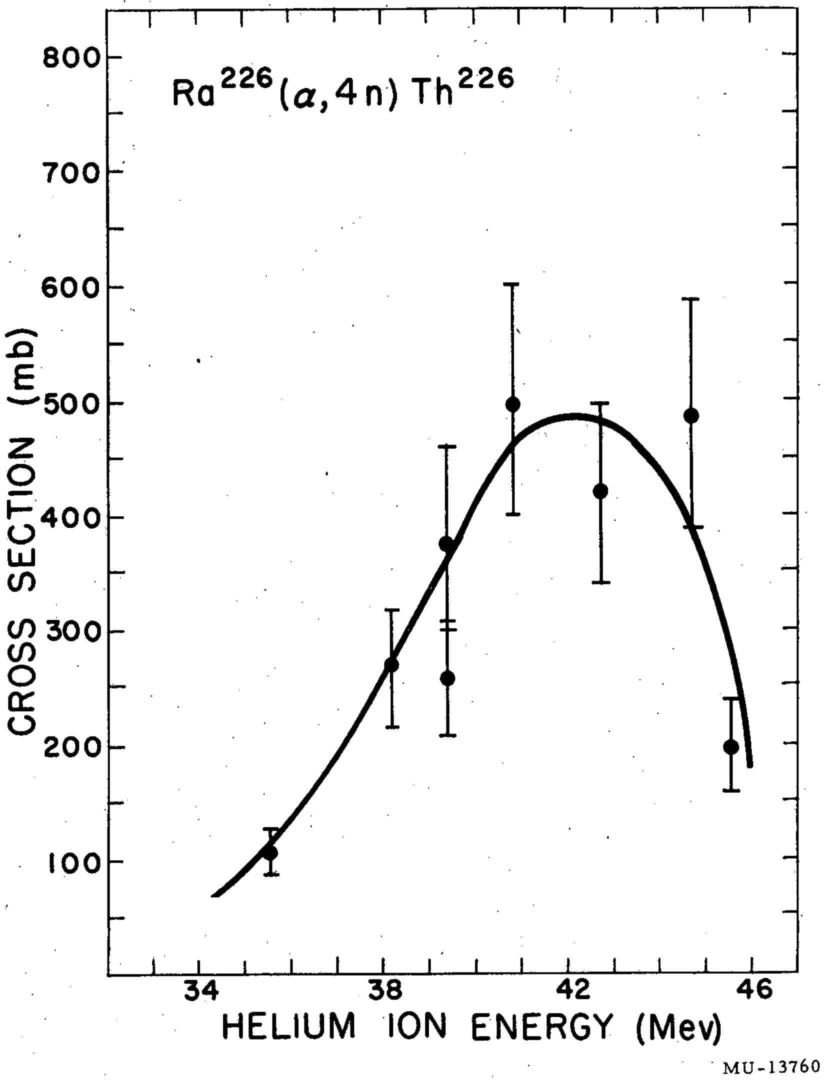

Fig. 1. Excitation function for the Ra^{226} $(\alpha,4n)$ Th^{226} reaction.

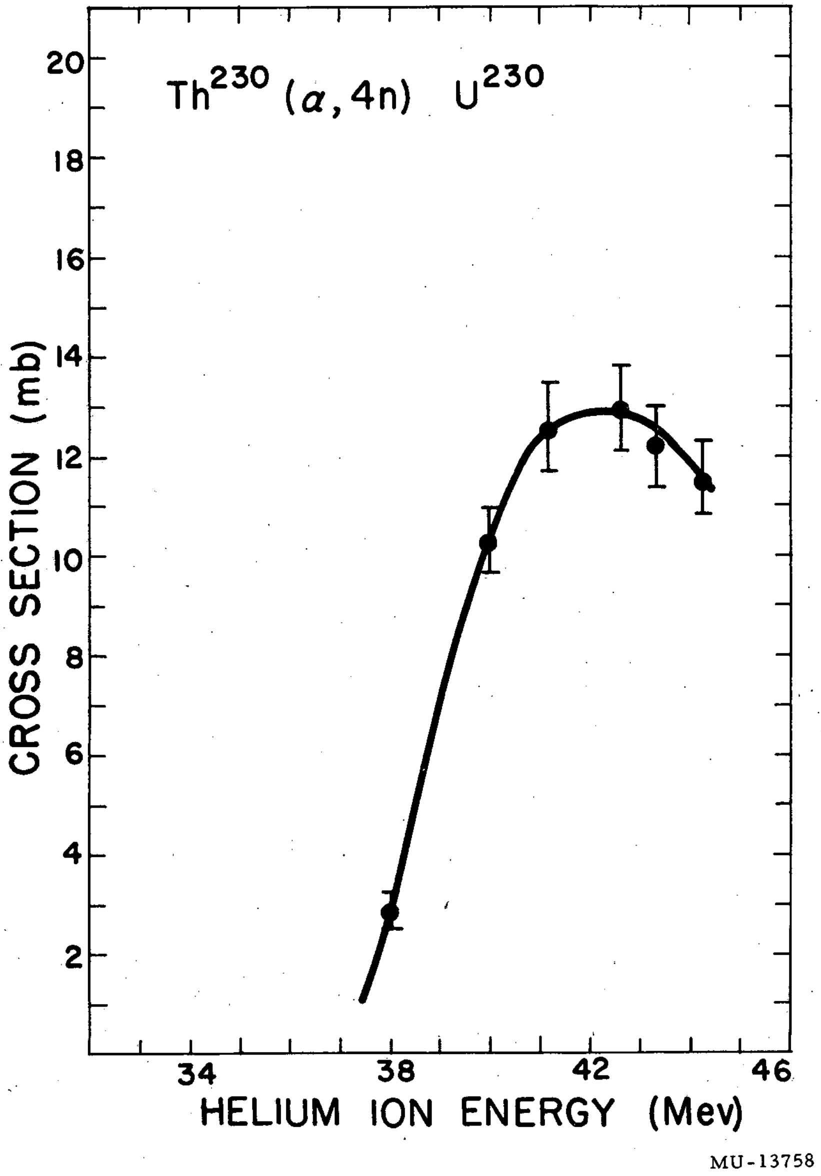

Fig. 2. Excitation function for the Th^{230} (α,4n) U^{230} reaction.

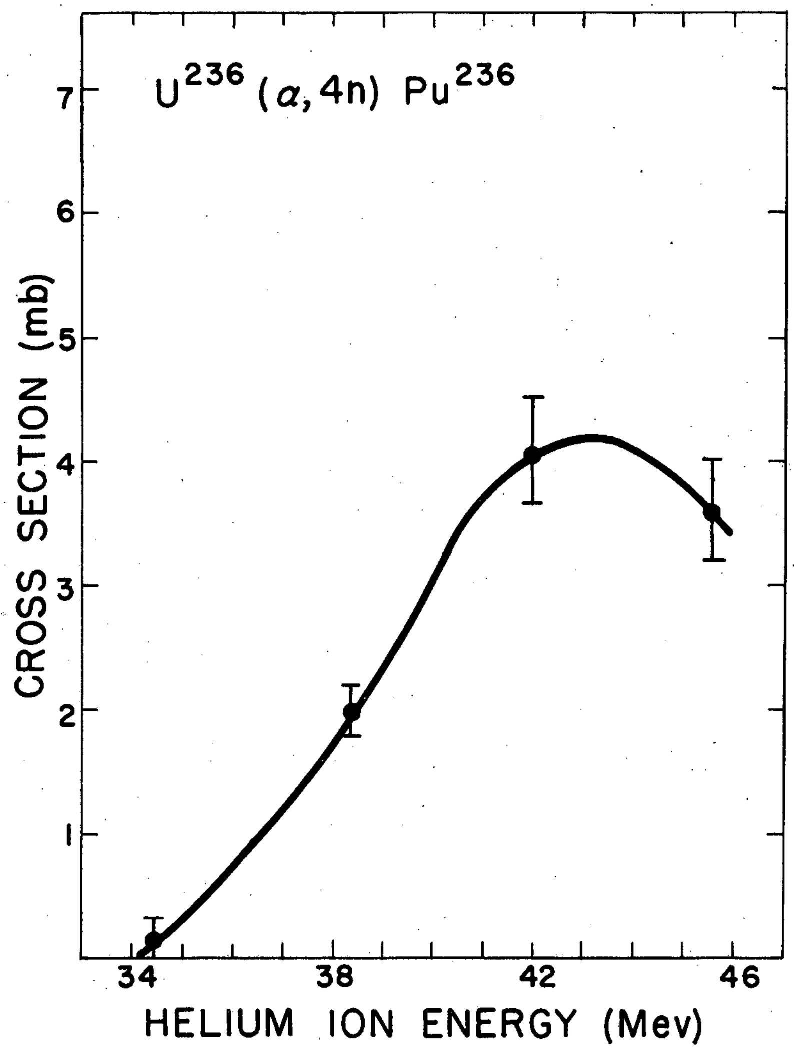

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Fig. 3. Excitation function for the U^{236} $(\alpha,4n)$ Pu^{236} reaction.

III. DISCUSSION

The excitation functions shown in Figs. 1 to 3 are illustrative of the large variations in cross sections for $(\alpha,4n)$ reactions of fissionable nuclides. In order to arrive at some semi-quantitative measure of the effective competition between neutron-emission and fission, we will attempt to relate the observed cross sections to partial level widths for the various modes of decay of the compound nucleus, which are in turn inversely related to the mean lifetimes of the compound nucleus with respect to the different modes of decay. The deduction of level width ratios (branching ratios) from α,xn excitation functions has been described by Glass et al.[25] If we assume that for excitation energies above the fission threshold and neutron binding energy the width for gamma ray de-excitation, as well as for proton and other charged particle emission, is negligible, we can write the expression for the neutron branching ratio (level width for neutron emission divided by total level width for all the possible products of the disintegration of the compound nucleus) as $\Gamma_n/(\Gamma_n + \Gamma_F.)$ This ratio will hence forth be designated as G_n. The cross section for the $(\alpha,4n)$ reaction at the peak of the excitation function can then be written as

$$\sigma\ (\alpha,4n) = G_{n_1} G_{n_2} G_{n_3} G_{n_4} \sigma_T$$

where the subscripts 1, 2, 3, and 4 refer to the branching ratio for the emission of the 1st, 2nd, 3rd, and 4th neutron. Since the neutrons are evaporated with a distribution in kinetic energy, one does not expect the cross section corresponding to the peak of the $(\alpha,4n)$ excitation function for a non-fissionable nucleus to be equal to the cross section for compound nucleus formation. Thus we must use for σ_T the cross section one would expect for the $(\alpha,4n)$ reaction at its peak if fission were not competing. This value has been estimated to be 1.2 barns from $(\alpha,4n)$ excitation functions of lead isotopes.[26] This has been used for all nuclei considered and although this choice is somewhat arbitrary it will not introduce any appreciable uncertainty in our comparisons.

All of the available cross sections for $(\alpha,4n)$ reactions have been summarized in Table V, including those reported in this work. The source of the data is listed in the last column of the table. Whenever possible the

Table V. Heavy element cross sections for the $(\alpha,4n)$ reaction and mean values of neutron to fission width ratios derived from these cross sections.

Target Nuclide	σ (mb)	$G_n = \Gamma_n/\Gamma_t$ mean	Γ_n/Γ_F mean	Reference
Ra^{226}	500	0.80	4.0	28
Th^{230}	13.0	0.32	0.48	28
Th^{232}	55	0.46	0.86	29
U^{233}	0.6^a	0.15	0.18	10
U^{234}	1.0	0.17	0.21	30
U^{235}	2.5	0.21	0.27	10
U^{236}	4.2	0.24	0.32	28
U^{238}	21	0.37	0.58	31
Pu^{238}	0.3^a	0.13	0.15	9
Pu^{239}	0.9^a	0.17	0.20	9
Pu^{240}	0.8^b	0.16	0.19	32
Pu^{242}	8.6	0.29	0.41	9
Am^{243}	14	0.34	0.52	33
Cm^{244}	0.3^b	0.13	0.15	34
Bk^{249}	6.0^b	0.27	0.36	24
Cf^{252}	2.2	0.21	0.27	35

[a] Cross section is very approximate.

[b] Lower limit, as excitation function is still rising at highest energy for which a cross section is reported.

value of the cross section corresponds to that at the peak of the excitation function. All of the data available have been listed, although some of the data are approximate or preliminary in nature. The third column lists the geometric mean values of G_n obtained from the relation

$$\bar{G}_n = \sqrt[4]{\frac{\sigma\,(\alpha,4n)}{1,200}}$$

where the cross sections are given in millibarns. Again assuming that the total width $\Gamma_T = \Gamma_n + \Gamma_F$, mean values of Γ_n/Γ_F have been calculated and are listed in the third column. From this type of experimental data we cannot isolate the fission width explicitly, but only the ratio of the neutron width to the fission width.

Examination of the mean values of the neutron-emission width to fission width ratios reveals that for a given atomic number, the ratio Γ_n/Γ_F increases with increasing mass number. This trend appears to be much stronger than that predicted by the parameter Z^2/A, and thus is probably closely related to the fact that neutron binding energies show a general trend to decreasing systematically with increasing mass number. The ratio of neutron-emission to fission widths deduced from the cross sections for $(\alpha,4n)$ reactions of uranium and plutonium isotopes are shown as a function of mass number in Fig. 4. Batzel's values[5] derived from fast-neutron fission cross sections of various uranium isotopes are shown for comparison. It is seen that the rate of change of the neutron to fission width ratio with mass number is approximately the same for uranium, plutonium and curium compound nuclei.

By making some simplifying assumptions, it is possible to derive approximate theoretical formulae for the fission width and neutron-emission width.[1,27] In particular, the treatment involves some assumptions which are not valid at low excitation energy. By assuming that the level density parameters of the parent nucleus--apart from the excitation energy dependence--are the same as those of the fissioning nucleus at the saddle point and adopting a Fermi-gas model of the nucleus, Fujimoto and Yamaguchi[27] have given the fission width as

$$\Gamma_F(E) \approx \frac{T}{2\pi} \exp \frac{-E_f}{T}$$

where E is the excitation energy, E_f is the fission threshold, and the nuclear temperature T is taken as being proportional to the square root of the excitation energy. In the same approximation the neutron width is given as

$$\Gamma_n(E) \approx \left(\frac{1}{2\pi}\right) \left(\frac{A^{2/3}}{K'}\right) \left(T^2 \exp \frac{-B_n}{T}\right)$$

where $K' = \dfrac{\hbar^2}{2\,m\,r_0^2} \simeq 10$ Mev and B_n is the neutron binding energy.

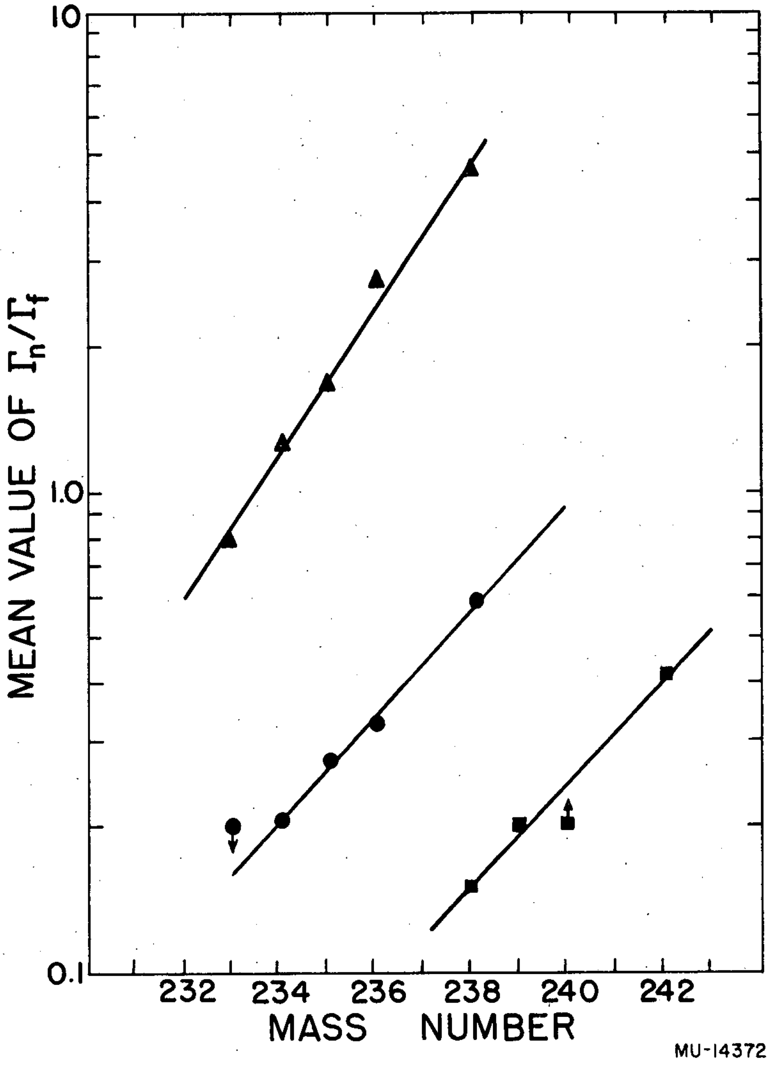

Fig. 4. Mean value of Γ_n/Γ_f derived from $(\alpha,4n)$ cross sections of uranium isotopes (circles) and plutonium isotopes (squares) <u>vs</u> mass number of target nuclide. Batzel's values of Γ_n/Γ_f derived from fast neutron cross sections for uranium isotopes (triangles) are shown for comparison.

Combining these expressions gives the following relation:

$$\frac{\Gamma_n}{\Gamma_F} = \frac{TA^{2/3}}{10} \exp \frac{E_f - B_n}{T} .$$

If one uses the fission activation energies obtained from equation (I) and a reasonable value for the nuclear temperature, (1-2 Mev) one obtains from this equation values of Γ_n/Γ_F which are several times larger than the experimental values listed in Table V.

However, the qualitative behavior predicted by this relationship may be compared with experiment. Since

$$\frac{\bar{\Gamma}_n}{\bar{\Gamma}_F} = \sqrt[4]{\left(\frac{\Gamma_n}{\Gamma_F}\right)_1 \left(\frac{\Gamma_n}{\Gamma_F}\right)_2 \left(\frac{\Gamma_n}{\Gamma_F}\right)_3 \left(\frac{\Gamma_n}{\Gamma_F}\right)_4}$$

we may write

$$\frac{\bar{\Gamma}_n}{\bar{\Gamma}_F} = \frac{T\bar{A}^{2/3}}{10} \sqrt[4]{\exp \sum_{i=1}^{4} \frac{(E_f - B_n)_i}{T}}$$

where the subscripts have the same meaning as before. Taking the logarithm,

$$\ln \frac{\bar{\Gamma}_n}{\bar{\Gamma}_f} = \ln \frac{T\bar{A}^{2/3}}{10} + 1/4 \sum_{i=1}^{4} \frac{(E_f - B_n)_i}{T}$$

it is seen that the logarithm of the neutron to fission width ratio should be a function of the difference between the fission threshold and the neutron binding energy.

In Fig. 5 we have plotted the logarithm of the neutron to fission width ratios listed in Table V <u>vs.</u> the difference between the sum of the four neutron binding energies and the sum of the four fission activation energies for the compound nuclei encountered in the evaporation chain. The fission activation energies were calculated using the formula presented in the first part of this paper, and the neutron binding energies are those calculated by B. M. Foreman, Jr., and listed by Hyde and Seaborg.[36] Considering the approximations in both the theoretical treatment and the analysis of experimental data, the correlation appears to fit quite well except for the point

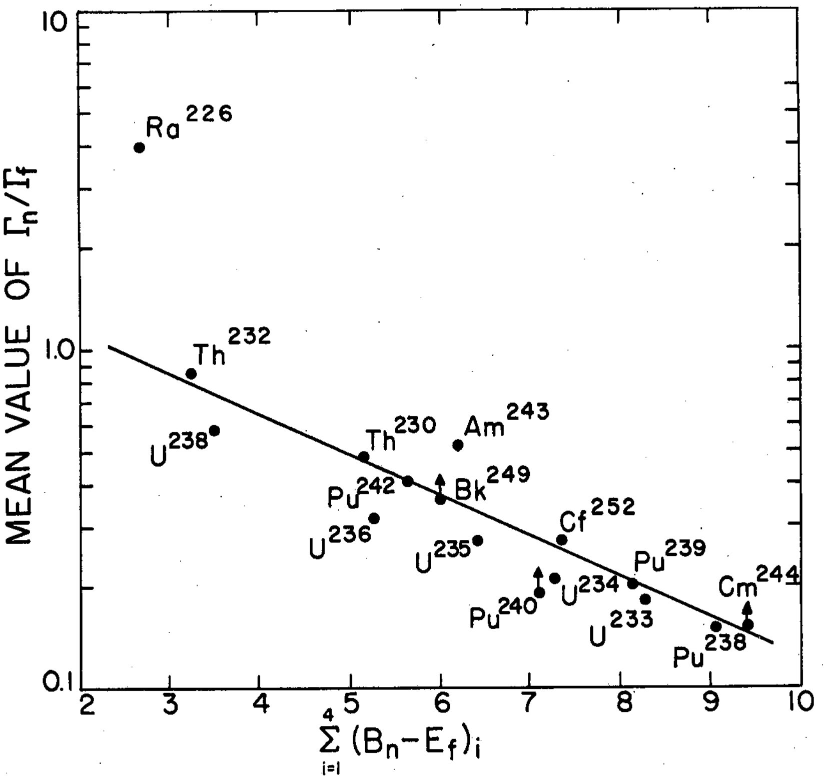

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Fig. 5. Mean values of Γ_n/Γ_f from $(\alpha,4n)$ cross sections vs difference between sum of the four neutron binding energies and the sum of the four fission activation energies for compound nuclei- encountered in the evaporation chain. The labels refer to the target nucleus and the circles with arrows indicate a lower limit.

representing helium-ion induced reactions of Ra^{226}. It would appear that for elements lighter than thorium these simple relationships are not a good approximation. All of the target nuclides represented in Fig. 5 are of even atomic number except for Am^{243} and Bk^{249}. It might be expected that these points would be high relative to even atomic number targets on the basis of the relative level densities of the products formed by neutron evaporation. For an $(\alpha,4n)$ reaction of an even-even or even-odd target nuclide, two of the residual nuclei formed by neutron evaporation are of even-even nuclear type and two are of even-odd nuclear type. However for target nuclides with odd atomic number, two of the residual nuclei formed are odd-even and two are odd-odd. Since odd-A and odd-odd nuclei are believed to have higher level densities than even-even nuclei, neutron evaporation might be expected to be more prominent for targets with odd atomic number. This effect would probably be most important at the last stage or two of the evaporation process. Meadows[37] has experimentally confirmed an effect of this nature in the yields of (p,pn) and (p,2n) reactions.

In the analysis of $(\alpha,4n)$ cross sections to obtain neutron-emission to fission width ratios, it has been assumed that there is no large variation of the neutron-to-fission width ratios with excitation energy. Experimentally it is rather difficult to obtain information on this problem. However, the rather flat plateaus observed in fast-neutron-induced fission excitation functions indicates that the relative probability for neutron-emission and fission is not strongly dependent on excitation energy for this relatively narrow range of excitation energies. Batzel[5] has analyzed the data for the 340-Mev proton-induced spallation of uranium[38] and concludes that the assumption that Γ_n/Γ_F is independent of excitation energy is a better approximation than the assumption that the probability of emission of a neutron increases much more rapidly as a function of excitation energy than does the probability of fission. If one considers the mean value of Γ_n/Γ_F obtained from an $(\alpha,4n)$ cross section to approximate that of the intermediate product half-way along the neutron evaporation chain, it is possible to compare the mean values of Γ_n/Γ_F obtained from $(\alpha,4n)$ cross sections with values from fast neutron fission in the two cases for the compound nuclei U^{234} and Pu^{240}. The mean value of Γ_n/Γ_F obtained from cross sections for the Th^{232}

$(\alpha,4n)$ U^{232} reaction is 0.87 for an approximate average excitation energy of 20 Mev, while that from U^{233} plus fast neutrons is 0.8 for an excitation energy of 10 Mev. Similarly the value of Γ_n/Γ_F from cross sections for the $U^{238}(\alpha,4n)$ Pu^{238} reaction is 0.58 for an average excitation energy of about 20 Mev, while that from Pu^{239} plus fast neutrons is 0.76 for an excitation of 10 Mev. Although comparison of the Γ_n/Γ_F values obtained from the two types of information can only be approximate, the relative probability for fission compared with neutron-emission does not seem to be strongly dependent on excitation energy.

ACKNOWLEDGEMENTS

The authors are indebted to the various people who have made available unpublished results quoted in this paper. Helpful discussions with B. M. Foreman, G. E. Gordon, T. D. Thomas and S. Vandenbosch are gratefully acknowledged. The authors wish to thank J. R. Huizenga for reading a draft of this paper and making helpful suggestions.

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