Competition between Fission and Neutron Emission in Excited Heavy Nuclei†

MANFRED LINDNER
Lawrence Radiation Laboratory, University of California, Livermore, California

AND

ANTHONY TURKEVICH
Enrico Fermi Institute for Nuclear Studies, University of Chicago, Chicago, Illinois

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The yields of the disintegration products of uranium and thorium bombarded with 340-Mev protons have been calculated from the intranuclear-cascade results of Metropolis et al. together with several alternative treatments of neutron evaporation. Several different assumptions concerning fission as a competing process were tested. The yields resulting from these different assumptions have been compared with the experimental results of Lindner and Osborne. The results point to the nonvariance of the quantity Γ_n/Γ_f with change in nuclear excitation in the energy range below 100 Mev. They also indicate that the model used for calculating these high-energy reactions in heavy nuclei is poor for estimating yields of nuclei very close to the target nucleus (previously noted) and of nuclei with charge much less than that of the target nucleus. This inadequacy appears to be associated with the cascade stage of the calculation.

INTRODUCTION

In recent years, there has been a growing interest in the competition between nucleon emission and fission in excited nuclei of heavy elements ($Z \geq 90$). Batzel[1] and Huizenga[2] have independently calculated the ratio of fission width to neutron evaporation width, Γ_f/Γ_n, for fissionable nuclei excited to about 10 Mev above their ground states. Batzel used these parameters to show that the assumption of energy independence of the Γ_f/Γ_n ratios gave a better fit to the data of Lindner and Osborne[3] on the spallation-product yields of uranium and thorium bombarded with 340-Mev protons than did the assumption that Γ_f/Γ_n was small at high excitations. Because no information was available at the time concerning the spectrum of nuclei (and their excitation distributions) resulting from the cascade process, Batzel had to make the simplifying assumption that U^{237} was the "typical" product of the nucleonic cascade giving rise to all residual uranium nuclei; similar assumptions were made for residual nuclei of protactinium and thorium.

More recently, Vandenbosch and Huizenga[4] have summarized in some detail the Γ_f/Γ_n ratios from all current experimental sources, including neutron-, charged-particle-, and photon-induced fission and neutron emission. These authors, further, have employed the Monte Carlo nucleonic cascade data of McManus and Sharp[5] in conjunction with the neutron-evaporation calculations of Jackson[6] and their own energy-independent Γ_f/Γ_n values, and they indicate that satisfactory agreement with the individual spallation yields of Lindner and Osborne results.

On the other hand, Shamov,[7] in an independent analysis of the same experimental data,[3] concluded that fission is the final act of cooling and is preceded by the emission of neutrons. His conclusions were based on the assumptions that excited U^{237} and Pa^{237} nuclei were the sole precursors of observed uranium and protactinium nuclides, respectively, and that the excitation distributions for U^{237} and Pa^{237} decreased linearly with excitation energy.

Recently, Monte Carlo calculations on the nucleonic cascade process have been reported[8] which are more extensive and more realistic than previous calculations. These calculations give both the number and types of excited nuclei, together with the distribution in excitation energies for each product resulting from the cascade process in U^{238} bombarded with 82-, 158-, 236-, and 450-Mev protons. Even though these cascade calculations have not yet been extensively tested, it was felt that the excitation distributions from them, together with a reasonable model for nucleon (neutron) evaporation, would allow a more rigorous test of the behavior of Γ_f/Γ_n in nuclei in different states of excitation.

The calculation will be presented in some detail for the case of protons incident on U^{238}. This calculation has the following order:

1. (a) Determination of the yields of the cascade products $Z^A(E^*)$ at various incident-proton energies.
   (b) Determination of the excitation-energy distribution for the various product nuclei of the cascade.
2. Calculation of nucleon evaporation from a given $Z^A(E^*)$.

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

[1] R. E. Batzel, University of California Radiation Laboratory Report UCRL-4303, February 25, 1954 (unpublished).
[2] J. R. Huizenga, Phys. Rev. **109**, 484 (1958).
[3] M. Lindner and R. N. Osborne, Phys. Rev. **103**, 378 (1956).
[4] R. Vandenbosch and J. R. Huizenga, *Proceedings of the Second United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, 1958*. (United Nations, Geneva, 1958), A/conf. 15/P/688.
[5] H. McManus and W. T. Sharp (private communication).
[6] J. D. Jackson, Can. J. Phys. **34**, 767 (1956).
[7] V. P. Shamov, Doklady Acad. Nauk S.S.S.R. **103**, 593 (1955).
[8] N. Metropolis, R. Bivins, M. Storm, A. Turkevich, J. M. Miller, and G. Friedlander, Phys. Rev. **110**, 185 (1958).

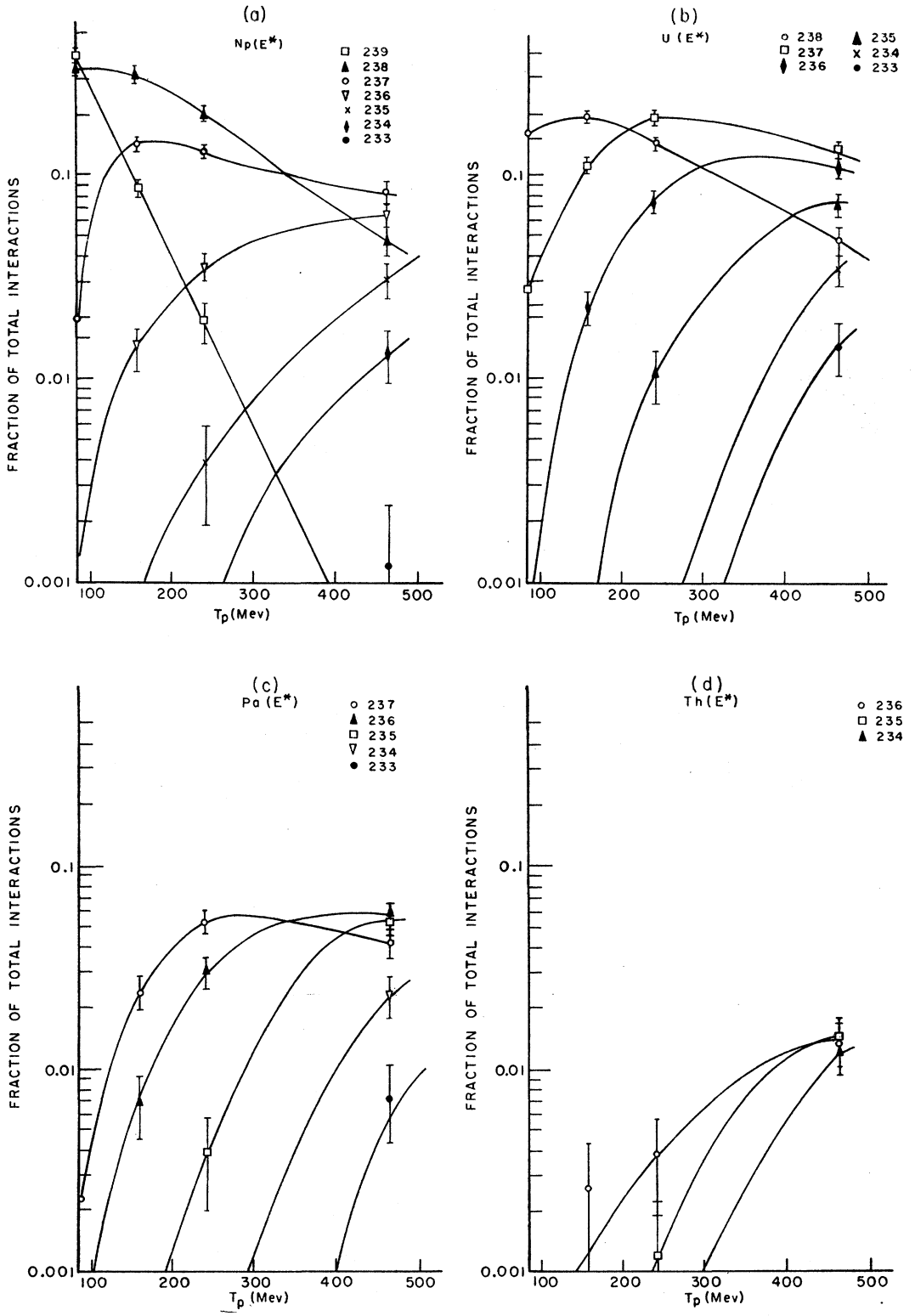

FIG. 1. The energy dependence of the probability of formation of the principal products of the intranuclear cascade produced by protons incident on U^{238}. These numbers were obtained from the calculations of Metropolis *et al.* (reference 8).

3. Testing of several possible assumptions concerning the variation of Γ_f/Γ_n with energy; comparison with experiment.

The experimental data used for comparison with the calculation were primarily those of 340-Mev protons incident on U^{238}, although some use of experimental data at lower energies, and from thorium bombardments, has also been made.

DISTRIBUTION OF NUCLEI FROM THE CASCADE

Among other information, the printouts on the intranuclear cascades[8] record the numbers of neutrons and protons ejected, and the excitation energies of the residual nuclei. The fraction of the U^{238} cascades leading to specific product nuclei as a function of proton energy has been taken from the calculations of Metropolis *et al.*,[8] and is summarized in graphical form in Fig. 1 for isotopes of Np(E^*), U(E^*), Pa(E^*), and Th(E^*).

The probabilities of formation of cascade-product nuclei of Z lower than 90 or for $A<233$ were so small as to be ignored in subsequent calculations. Cross sections for the formation of a specific nuclide as a result of the cascade were obtained from these curves by multiplying by the geometric cross section. This was taken to be 2.04×10^{-24} cm^2.[8]

EXCITATION DISTRIBUTIONS

The excitation-energy distributions for a given nuclide (and a given incident-proton energy) were obtained from the same calculations and were plotted in normalized form. In this form the abscissae are fractions of the maximum excitation energy possible for the specific nuclide, and the ordinates are the fractions of the nuclides found with this fraction of the maximum excitation energy. The latter energy is that which a nucleus would possess if all cascade nucleons were ejected with just the "cutoff" (i.e., minimum) kinetic energy.[8] This procedure proved to be of great convenience in interpolating the shapes of the excitation distributions at 340-Mev incident energy, the latter being the energy at which much of the comparison with experiment was made, but for which there were no direct Monte Carlo cascade data. These normalized distributions are given in Fig. 2(a) through (l) for the nuclides of subsequent importance in this treatment. It is to be noted that these figures are the smoothed-out curves for each distribution, and do not show the scatter of the data from the Monte Carlo results, except in the case of Fig. 2(l). The dashed curves are the values interpolated for 340-Mev incident-proton energy

Because the fraction of events leading to thorium isotopes was small, all cascades leading to this element were combined without regard to mass number. The resultant distribution, in Fig. 2(l), was assumed to be valid for the only isotopes of importance for thorium, i.e., those of masses 236, 235, and 234.

In the calculations of the yields of the products from the proton bombardment of thorium, it was assumed that the intranuclear cascade would result in the same changes in mass and charge as were produced in uranium, and that the excitation-energy distributions would be given for the appropriate residual nuclei by the curves of Fig. 2.

NUCLEON-EVAPORATION CALCULATIONS

In the phase following the nucleonic cascade, nuclear de-excitation occurs through nucleon evaporation.[9] Application of the Weisskopf model[10] to heavy nuclei at excitations extending up to 100 Mev has been made independently by Heckrotte[11] and by Jackson.[6,12] For purposes of the present work, Heckrotte's treatment has been adopted, partially because his calculations were carried out for uranium nuclei. However, a comparison of the two treatments shows that both predict similar probability distributions for evaporation of an integral number of neutrons as a function of the energy of excitation, in the region 0–100 Mev.

That the region of excitation energies below 100 Mev is the important one for the present study can be seen

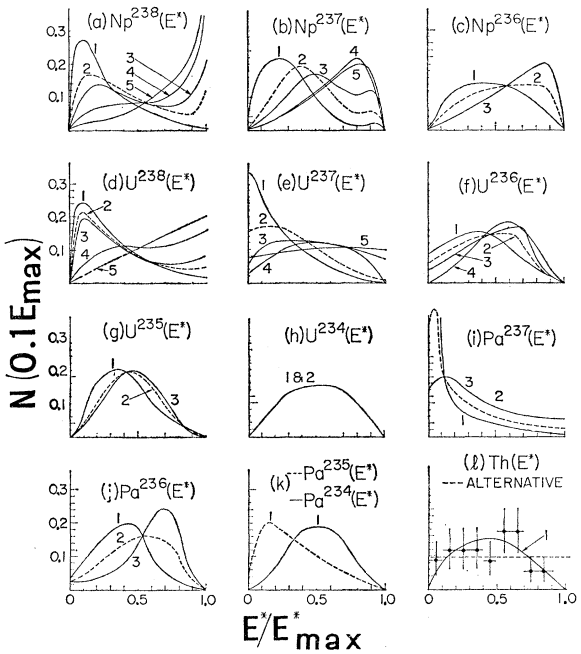

FIG. 2. The excitation-energy distribution of the principal products of the Monte Carlo cascade (from data of Metropolis et al., reference 8). U^{238}+protons. Proton energy (T_p): Curve 1, 460 Mev; Curve 2, 340 Mev; Curve 3, 236 Mev; Curve 4, 158 Mev; Curve 5, 82 Mev.

[9] R. Serber, Phys. Rev. **72**, 1114 (1947).
[10] V. F. Weisskopf, Phys. Rev. **52**, 295 (1937).
[11] W. Heckrotte, University of California Radiation Laboratory Report UCRL-2184 Rev., December 18, 1953 (unpublished).
[12] J. D. Jackson, Can. J. Phys. **35**, 21 (1957).

TABLE I. Calculated cross sections (mb) for uranium nuclei resulting from neutron evaporation following the intranuclear cascade. U^{238}+340-Mev p; no fission.

Evaporation scheme	Uranium mass number 238	237	236	235	234	233	232	231	230	229	228
Jackson	2.5	21.1	32.3	37.1	38.1	40.0	40.8	41.0	41.0	41.1	41.4
Heckrotte	3.2	24.1	24.2	25.8	32.3	35.8	38.9	41.6	42.8	44.2	45.6
10 Mev/n	3.3	28.3	38.1	40	41.2	41.6	42.4	43.2	43.6	43.4	43.0
σ_{obs} [a]		~50					≤4		0.35	0.060	0.038

[a] See reference 3.

from the fact that the mass numbers of the nuclides experimentally determined extend only about ten mass units below the heaviest of the products of the nucleonic cascade. Since, as a rough guide, the average excitation energy carried away by a neutron in the evaporation process[6] is given by

$$\langle \Delta E \rangle_{Av} = B_n + 2T \sim 10 \text{ Mev},$$

it is clear that the lightest observed nuclides must arise from nucleonic cascade products that have excitation energies of approximately 100 Mev or less. The treatments of both Jackson and Heckrotte indicate that proton evaporation from heavy nuclei can be safely neglected at this energy. Heckrotte's data on the average number of neutrons emitted as a function of the excitation energy, and his data on the distribution in the number of neutrons emitted at excitations of 50 and 100 Mev, have been used to construct curves giving $P_n(E^*)$, the probability of emission of n neutrons, as a function of the excitation energy, E^*. These curves are presented in Fig. 3. These distributions have been calculated for a constant binding energy and so do not show the secondary effects of alternation of neutron binding energies or of the increase in neutron binding energies with decrease in mass number.

For comparison, Fig. 3 also presents, by dotted lines, Jackson's predictions for the function $P_n(E^*)$, based on a value of 4.0 for the ratio of average neutron binding energy to nuclear temperature, $\bar{B}/T$, and in which $\bar{B}=6.35$ Mev. The latter is the average neutron binding energy for the mass region from U^{228} to U^{238}. Binding energies were taken from the tables of Cameron.[13] These curves are seen to be nonidentical, although similar. The effect of these differences will be discussed subsequently.

An even simpler assumption concerning the evaporation process is that, on the average, the number of evaporated neutrons is one-tenth the available excitation, since an evaporating neutron will decrease available excitation energy by about 10 Mev. That this is approximately true may be verified from Fig. 3.

Neutron-evaporation calculations have also been performed by Dostrovsky *et al.*,[14] employing the Monte Carlo method. However, these calculations were not as detailed in the region of excitation up to 100 Mev as were those of either Jackson or Heckrotte and were not used.

The distribution of resultant de-excited uranium nuclei has been calculated from the Monte Carlo cascade results under the assumption that fission does not occur, for (a) the Heckrotte probabilities, (b) the Jackson probabilities, and (c) the assumption of a 10-Mev de-excitation increment per neutron evaporated. Results are shown in Table 1. For future discussion, observed cross sections are also tabulated from the results of Lindner and Osborne.

It becomes quite obvious that, except near the region of the target nucleus, the yields obtained by all three calculations are similar. Differences of the order of twenty or thirty percent do not appreciably alter the outcome of the present work. Although the third treatment would appear quite satisfactory for these calculations, the Heckrotte probability curves were the ones usually used. However, in at least one instance in which comparison of other parameters was desired, it was convenient to use this simplified concept of 10-Mev de-excitation per neutron evaporated. In all cases, however, allusion will be made to the particular neutron-evaporation scheme that was employed.

In considering neutron evaporation from excited protactinium and thorium nuclides, the distributions $P_n(E^*)$ were modified for the slightly lower average binding energies for these elements in the region studied. Binding energies were taken from the tables of Cameron.[13]

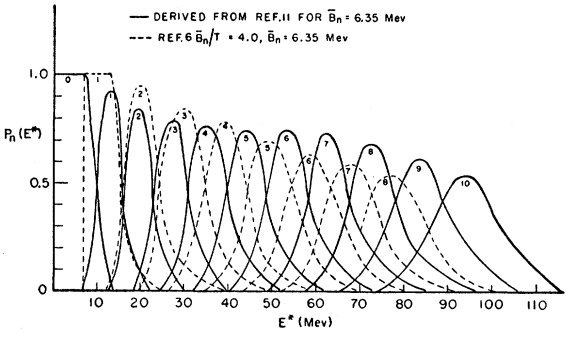

FIG. 3. Dependence of multiplicity in neutron evaporation upon excitation energy in U^{238}.

[13] A. G. W. Cameron, Atomic Energy of Canada Limited Report CRP-690, 1957 (unpublished).

[14] I. Dostrovsky, P. Rabinowitz, and R. Bivins, Phys. Rev. **111**, 1659 (1958).

TABLE II. Final cross sections (mb) of U nuclides following the cascade, neutron evaporation, and fission for U^{238}+340- Mev protons.

G from curve	Assumption	238	237	236	235	234	233	232	231	230	229	228
A	1[a]	3.3	26.7	29	21	13.7	7.9	4.1	1.9	0.77	0.27	0.074
B	1[a]	3.3	26.7	29	22	14.4	8.3	4.1	1.7	0.53	0.13	0.025
B	2[a]	3.3	27	30	22	17	14	10.5	7.9	5.4	3.4	1.8
B	3[a]	3.3	26.6	28	20	10	2.2	0.3	0.03			
B	1[b]	3.2	23.1	18.8	14	11	7.0	3.6	1.5	0.50	0.13	0.026
Experimental			~50					≤4		0.35	0.06	0.038

[a] These calculations used the simplified (10 Mev/nucleon) evaporation calculation.
[b] This calculation used the Heckrotte evaporation-calculation technique.

FISSION AS A COMPETITIVE PROCESS FOLLOWING THE CASCADE

In Table I the small values for the observed cross sections of the light U isotopes relative to the calculated values show the effect of fission as a competing reaction in the de-excitation process of the heavy elements.[1,12] Four alternative assumptions concerning the energy-dependence of the fission competition have been made, and the results of calculations, in which $G \equiv \Gamma_n/(\Gamma_n+\Gamma_f)$ is applied to each neutron-evaporation probability, have been compared with the experimental results.[3] These assumptions are:

1. That Γ_f/Γ_n is a function only of nuclear type, and does not vary with energy in the region $E^* \geq \sim 100$ Mev. Such an assumption has already been reported by two investigators[1,4] as representing the best fit to the data.

2. That fission competes only in the last stages of neutron evaporation, i.e., $\Gamma_f/\Gamma_n=0$ for, say, $E^*>20$ Mev, but $\Gamma_f/\Gamma_n \neq 0$ for $E^*<20$ Mev. This approximates the conclusion of Shamov[7] from the same data.[3]

3. That fission occurs to the exclusion of neutron emission above some specific energy, taken arbitrarily here to be 40 Mev, i.e., $\Gamma_f/\Gamma_n=\infty$ at $E>40$ but Γ_f/Γ_n is finite at $E<40$ Mev.

4. That the ratio Γ_f/Γ_n is some smoothly varying function of the energy in the region of excitation considered ($E^* \leq 100$ Mev). For this purpose, the recent treatment of Dostrovsky, Fraenkel, and Rabinowitz[15] has been adopted. This will be considered in detail later in the paper.

The values for Γ_f/Γ_n used in assumptions 2 and 3, when they are neither zero nor infinite, are those used in assumption 1.

For the first three assumptions, the procedure was tested in detail only for isotopes of uranium produced by 340-Mev protons incident on U^{238}. The reasons for this selection are several-fold: (a) At low excitation energies, the supporting parameters, Γ_f/Γ_n, are considerably better known for uranium nuclides than for the other elements tested. (b) The test should be more sensitive for uranium than for either protactinium or thorium, since the effect of fission appears greater for U than for elements of lower Z. Neptunium isotopes might have provided a still more sensitive test but the experimental yields were lacking for comparison.

The comparison of assumptions 1, 2, and 3 has also been made only for the simplified evaporation treatment (that each stage diminishes the excitation by 10 Mev). For assumption 1, however, the calculations were also performed using the Heckrotte evaporation probabilities.

It thus remains to establish a set of values for the parameters Γ_f/Γ_n for each uranium nuclide appropriate to the evaporation sequence. For this purpose there are available several tabulations.[1,2,4] Those of Huizenga[2] were taken; his results, in the form of $\log(\Gamma_n/\Gamma_f)$ as a function of Z^2/A, are presented in Fig. 4. It is apparent that the data might be equally well represented by the curve (A) or the straight line (B). Since the extrapolated values (high Z^2/A, corresponding to $U^{228,229}$, etc.) are of great importance for the purposes of this paper, calculations were made with both A and B.

Table II summarizes the results of these calculations, in which fission competes with neutron evaporation. The first column indicates the curve (A or B) of Fig. 4 that had been used in the calculation of $G=\Gamma_n/(\Gamma_f+\Gamma_n)$ for the isotopes of uranium involved. The second

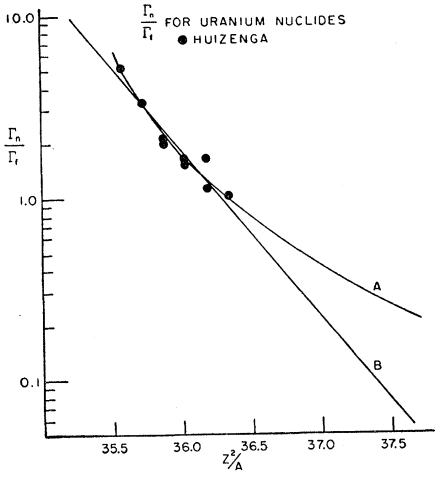

FIG. 4. Γ_n/Γ_f for uranium nuclides (from Huizenga, reference 2).

[15] I. Dostrovsky, Z. Fraenkel, and P. Rabinowitz, *Proceedings of the Second United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), A/Conf. 15/p/1615.

column specifies the energy dependence of Γ_f/Γ_n used (assumptions 1 to 3, above). The following columns give the calculated yields, in mb, of the various isotopes of uranium, down to mass 228. All of these calculations except the last used the simplified (10 Mev/nucleon) evaporation calculation. The last used the Heckrotte evaporation-calculation technique. Finally, the last line lists the experimental data available.

Inspection of Table II shows that the calculated U^{237} cross section is considerably lower than that observed. This discrepancy cannot be attributed to fission, since even the cross section without fission is too low, by almost a factor of two. This failure of the cascade calculations of Metropolis *et al.*[8] to reproduce (p,pn) cross sections has been noted before.[8,16]

Table II also shows that, although curves A and B of Fig. 2 lead to fairly similar spallation yields in this mass region (above $A=227$), curve B appears to give better agreement with experiment, and was used in further calculations. Comparison of the results of assumptions 1, 2, and 3 shows that constancy of Γ_f/Γ_n with energy (assumption 1) gives significantly better agreement than the extreme energy dependences described by assumptions 2 and 3.

Inspection of the details of the calculation, moreover, indicates that excited U^{237} and U^{236} nuclei resulting from the cascade are the most important contributors to the formation of U^{230}, U^{229}, and U^{228}. Thus, the calculation is sensitive to the choice of Γ_f/Γ_n values for these two nuclei in the excitation range 60–90 Mev. Finally, there appears to be little difference between the two treatments for the evaporation phase ($B1$ and $B1H$). Only at high mass numbers is there a significant difference between the "simplified" and the Heckrotte treatments for neutron evaporation.

1. Γ_f/Γ_n from Dostrovsky *et al.*[15]

Although a constancy of Γ_f/Γ_n with energy variation appears consistent with experiment, it cannot be ruled out that the data might be equally well represented by some energy-dependence other than the "step" type of behavior tested in assumptions 2 and 3. Recently, Dostrovsky *et al.*[15] have calculated, by Monte Carlo methods, the competition between fission and neutron emission in nuclei of mass 200–230, for excitation energies 50–600 Mev, and have obtained good agreement with observed fission cross sections. It therefore seemed worthwhile to test the expression they used for Γ_f/Γ_n. Fission was introduced into their calculations through the equation (constants herein evaluated)

$$\frac{\Gamma_f}{\Gamma_n}=\frac{3.05}{A^{\frac{2}{3}}(E-E_n)}\left(2\left[\frac{1}{10}A(E-E_f)\right]^{\frac{1}{2}}-1\right)+\exp\left\{2\sqrt{\frac{A}{10}}([(E-E_f)^{\frac{1}{2}}-(E-E_n)^{\frac{1}{2}}])\right\}, \quad (1)$$

in which expressions for Γ_f and Γ_n were derived from Bohr and Wheeler[17] and from Weisskopf,[10] respectively. A is the mass number, E the excitation energy, E_n the neutron binding energy (taken from the mass tables of Cameron,[13] and E_f the fission threshold (taken from Frankel and Metropolis[18]).

Use of this formula complicated the calculation reported here considerably, since the probability of surviving fission is a function not only of the mass and charge of the nucleus but also of the excitation energy. For this reason calculations were carried out only for the production of U^{232} and U^{228}, based on the Heckrotte evaporation probabilities. Calculated cross sections of 29 mb and 4.4 mb were obtained for U^{232} and U^{228}, respectively. These values are to be compared with experimental values of 3 and 0.03 mb, respectively.

It is evident that the equation used by Dostrovsky does not produce enough fission during the evaporation phase to give final spallation yields in agreement with experiment. Examination of this equation, moreover, shows that for the mass region of interest here, the fission competition, and hence the results of the calculations, is not unlike that of assumption 2.

It must be concluded, therefore, that if Γ_f/Γ_n is some smoothly varying function of the excitation energy, that variation does not seem to be satisfied by the Dostrovsky equation for uranium nuclei at excitations of less than 100 Mev. Conversely, it is strongly indicated that Γ_f/Γ_n is independent of energy over this range of excitation.

2. Determination of Fission Competition in other Fissionable Elements

As was pointed out earlier, one of the reasons that uranium was chosen for initial comparison with experiment was that the Γ_f/Γ_n values at low excitation energies for many of its isotopes had previously been determined.[1,2,4] Similar information for other elements, although available, is far less complete. This information has recently been summarized by Vandenbosch and Huizenga.[4] Their data are indicated by the points of Fig. 5.

In view of the relative paucity of such data for Np, Pa, and Th isotopes, it seemed worthwhile to arrive at an analytical expression for Γ_f/Γ_n for the isotopes of these elements which would best explain the differences between the observed cross sections and those calculated without fission competition. No systematic effort was made to test the energy dependence of these quantities.

The data points of Huizenga and Vandenbosch in Fig. 5 indicate that $\log(\Gamma_n/\Gamma_f)$ is an approximately linear function of A for uranium and that this linear dependence might well hold, with the same slope, for other elements.

[16] S. S. Markowitz, F. S. Rowland, and G. Friedlander, Phys. Rev. **112**, 1295 (1958).

[17] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).

[18] S. Frankel and N. Metropolis, Phys. Rev. **72**, 914 (1947).

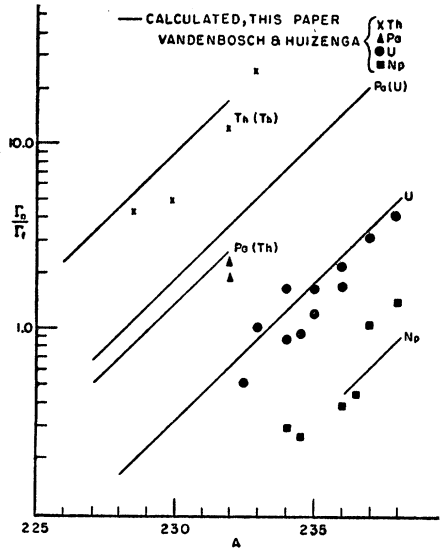

FIG. 5. Γ_n/Γ_f as a function of A for fissionable nuclides (points from reference 4).

In the present investigation, an attempt was made to determine this linear relationship for Np, Pa, and Th, produced from U and Th bombarded with 340-Mev protons. The algebraic form assumed for each element was

$$\ln(\Gamma_n/\Gamma_f)(A,Z)=a(Z)+0.335A, \qquad (2)$$

where the slope, 0.335, was obtained from the uranium data.[2,4] A given experimental spallation cross section, together with a calculated yield in which fission competition was omitted, defined a set of $\ln(\Gamma_n/\Gamma_f)$ values for the mass region between the heaviest cascade product and the evaporation product for that element.

This calculation was carried out numerically, using the evaporation procedure of Heckrotte for those cases in which fission had appreciably lowered the spallation yield. The results are summarized in Table III. This table lists for each bombarded target (column 1), the spallation product (columns 2 and 3) and the observed cross section (column 4). Column 5 shows the cross section expected without fission competition. Column 6 gives the Γ_n/Γ_f value obtained by this procedure at the reference mass, taken arbitrarily to be 230 (except for the case of Np^{236} formed from a uranium target, in which the reference mass is 237). It is to be emphasized, however, that each calculation determined a set of Γ_n/Γ_f values consistent with Eq. (2) for the mass regions between the highest mass-number cascade product and the final product of the evaporation stage.

There are several features of interest in the last column of Table III. The value of Γ_n/Γ_f for Pa^{230}, for example, was calculated from two spallation yields from the thorium bombardment, and from two yields from uranium bombardment. The thorium bombardments determine the Γ_n/Γ_f curve for the protactinium isotopes 232 down to 228; the uranium bombardments determine it from mass 237 down to 228. In view of the approximate nature of the calculations and of the experimental errors, the agreement is quite satisfactory. This is also true for the two values of Γ_n/Γ_f calculated for Th^{230} from the yields of Th^{227} and Th^{226} from thorium spallation. The Γ_n/Γ_f values for thorium are probably the least significant; since relatively little fission occurs in thorium, the values of Γ_n/Γ_f are very sensitive to the parameters in Table III.

The thorium yields from uranium bombardment could not be used to calculate an independent value of Γ_n/Γ_f for Th^{230} because of the very large uncertainties in the cascade results for this element [see Figs. 1(d) and 2(l)].

FINAL SPALLATION YIELDS

The calculated straight-line functions for $\log(\Gamma_n/\Gamma_f)$ are plotted in Fig. 5, together with the values for individual nuclei given by Vandenbosch and Huizenga. The line for thorium is obtained from the average of the two values of Table III. Similarly, the two lines for protactinium are obtained from the averages of the values obtained from U and Th targets. Each line is drawn to cover approximately the mass region contributing to the spallation yields used in the calculation. The line for uranium nuclides is taken from Fig. 4, Curve B, since those values were found in Table II to yield satisfactory agreement with experiment. The agreement of the calculated lines with the Vandenbosch-Huizenga points is satisfactory, and the method represents an independent means of calculating Γ_n/Γ_f. Since nuclei with excitation energies between 50 and 100 Mev contribute in this calculation, the agreement with low-energy Γ_n/Γ_f data implies a lack of strong energy dependence of Γ_n/Γ_f for thorium and protactinium as well as for uranium.

As a final check on these calculations, the Γ_n/Γ_f values were then taken from the straight-line functions of Fig. 5 and were used to calculate the revised cross sections for nuclide formation, which include the effect of fission competition. For Pa, the average of two lines in Fig. 5 was used. The results for uranium spallation are shown in Fig. 6; those for thorium spallation in Fig. 7. Observed values are given by the points. The

TABLE III. Γ_n/Γ_f values calculated from σ_{obs} and Eq. (2).

Target	Prod. Z	Prod. A	σ_{obs} [a]	σ_{calc} no fission	Γ_n/Γ_f at reference mass 230
U^{238}	Np	236	1.70	7.4 (237)	0.606 (237)
	Pa	228	1.70	11.7	1.98
	Pa	227	0.71	11.9	1.71
Th^{232}	Pa	228	1.7±0.2	13.1	1.32
	Pa	227	1.0	11.6	1.46
	Th	227	22±5	40.0	10.2
	Th	226	17±0.3	40.9	8.1

[a] See reference 3.

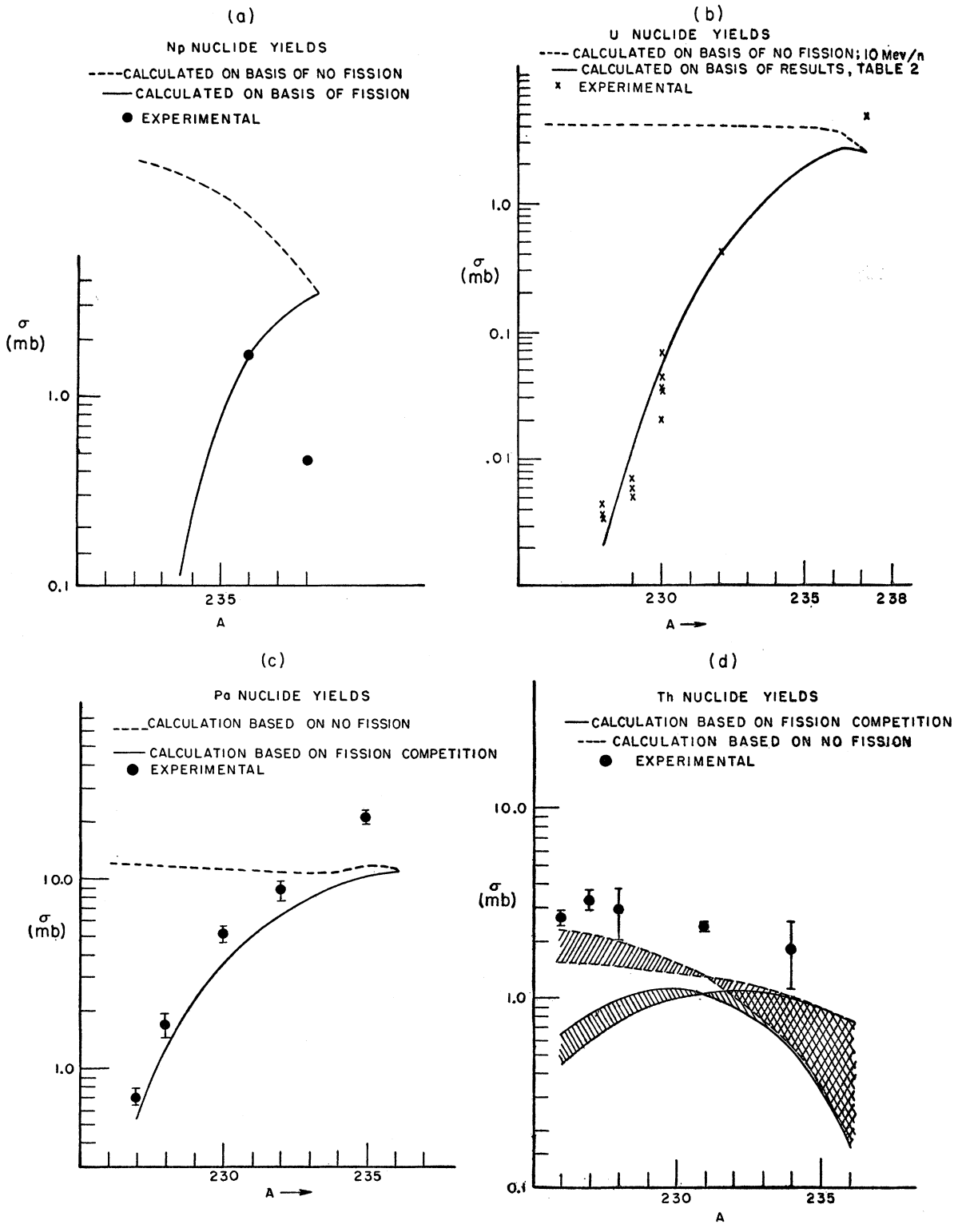

FIG. 6. Calculated yields of nuclides from U^{238} spallation with 340-Mev protons (including the effect of fission). Points are experimental data from reference 3.

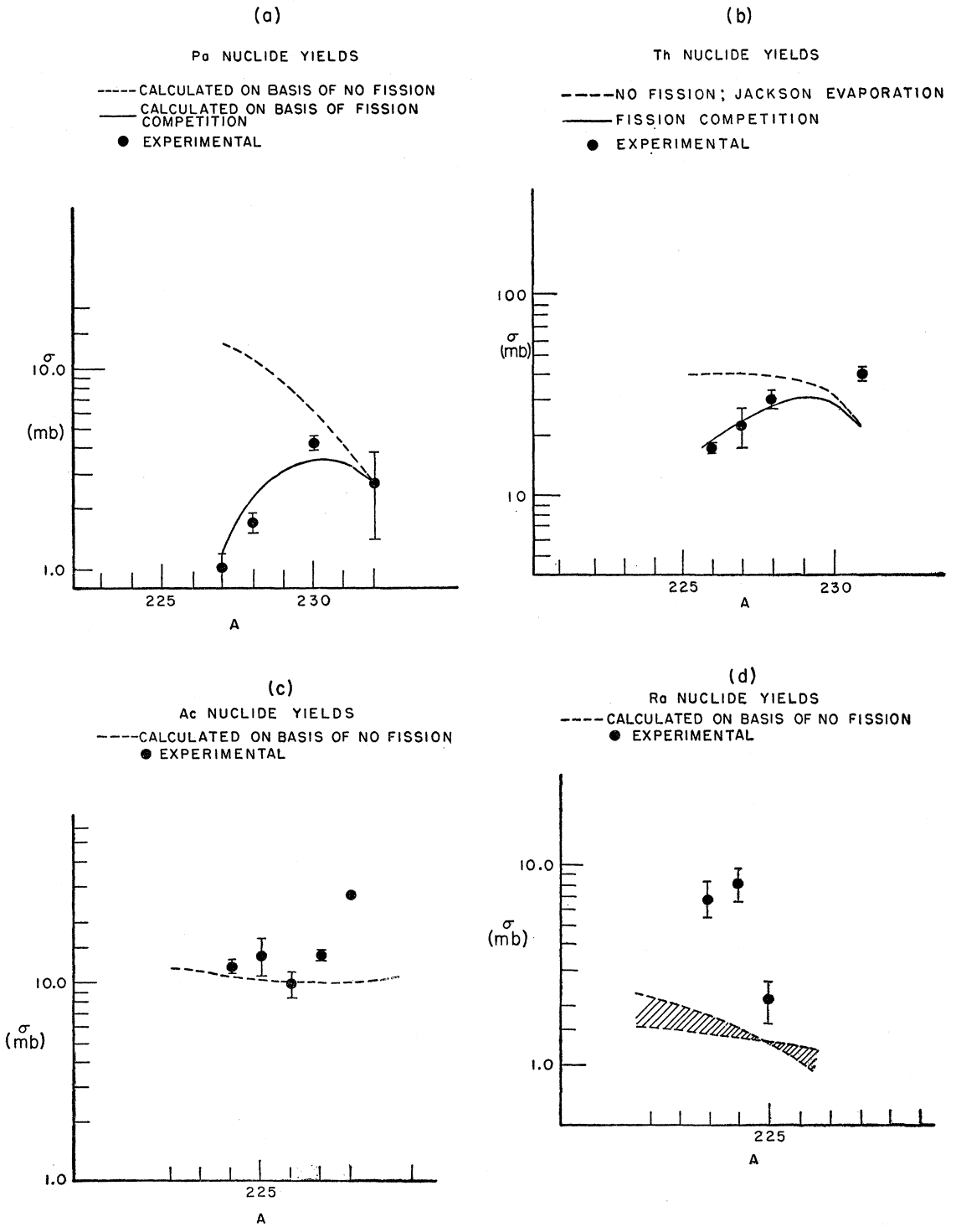

FIG. 7. Calculated yields of nuclides from Th^{232} spallation with 340-Mev protons (including the effects of fission). Points are experimental data from reference 3.

dashed lines give the yields calculated without invoking fission competition, while the solid line in each case includes this effect. Except for Fig. 7(b), the Heckrotte evaporation treatment was used.

The values calculated for uranium isotopes in Fig. 6 were taken from Table II. The shaded portions of Figs. 6(d) and 7(d) represent the areas over which the calculation is uncertain due to the possibility of two

extreme choices for the excitation distribution in Fig. 2(l).

In Fig. 7, the values calculated for actinium and radium isotopes were assumed to be those which would be found in Pa and Th isotopes if fission did not occur. From the results of Fig. 5 and Vandenbosch and Huizenga's[4] reported value of 10^4 for Γ_n/Γ_f for Ra^{227}, it may be inferred that neglect of fissionability in Ac and Ra is a fair approximation.

DISCUSSION OF RESULTS

In their gross features, the calculated cross sections from 340-Mev protons incident on thorium and uranium agree sufficiently well in magnitude and in slope with the experimental values that one may conclude that (a) the nucleonic cascade model holds reasonably well for the uranium-thorium region; (b) several methods of calculating neutron evaporation give results consistent with experiment; and (c) the ratio of the fission width to the neutron width of a nucleus appears to be independent of the excitation energy—at least up to an excitation of about 100 Mev.

On the other hand, there seem to be discrepancies between theory and observation that lie outside the limits of experimental error, and it remains to consider the possible sources of these discrepancies. Calculated results low by a factor of two in the case of the (p,pn) cross sections are noted. Markowitz, Rowland, and Friedlander[16] have recently measured the (p,pn) excitation functions for a wide variety of elements and have compared these with the predictions of the Monte Carlo nucleonic cascade,[8] followed by simple evaporation. They find discrepancies of a factor of two to three at 0.4 Bev. A similar effect was noted by Yule and Turkevich[19] for $Cu^{65}(p,pn)Cu^{64}$. No entirely satisfactory explanation has as yet been reported for the disagreement. Therefore, the present disagreement noted for the yields of U^{237} and Th^{231} will not be considered further in this work.

The mass-yield curve for uranium nuclides in Fig. 6 is not very informative for the higher mass numbers because no yields were experimentally observable in this region (except for U^{237}). On the other hand, the corresponding region for thorium spallation [Fig. 7(b)] is well covered by experimental data. The Heckrotte evaporation treatment leads to calculated cross sections that are systematically low, relative to the experimental values. The use of the Jackson evaporation treatment gives somewhat better agreement for this case. However, the Jackson treatment gives poorer agreement for Pa nuclide yields in thorium spallation, so that there seems to be no clear-cut choice between the two evaporation calculations.

In view of the reasonable agreement between calculations and experiment for the (p,pxn) reactions in thorium, it is difficult to understand the very large discrepancy between the calculated and the observed (p,n) cross section in uranium targets that results in the case of Np^{238}. Use of the Jackson calculations improved the agreement by only a fraction of the discrepancy, about 30% for this case. In view of the large reported difference in the observed cross sections for Pa^{232} from Th and Np^{238} from U (3.2 vs 0.46 mb, respectively) it seems just possible that the experimental value for Np^{238} might be in error.

The calculated mass-yield curve for protactinium in uranium spallation is systematically lower than the experimental values. It is only slightly changed if the Jackson calculations are substituted for those of Heckrotte. The corresponding $(p,2pxn)$ reactions (Ac) measured in thorium behave similarly. Possible explanations for the larger observed cross sections for these reactions will be discussed presently.

The $(p,3pxn)$ yields (Th from U or Ra from Th) are shown by the shaded areas in Figs. 6(d) and 7(d). These spreads represent the extremes in interpretation of the cascade data shown in Fig. 2(l) occasioned by the poor statistics for these events. It is obvious, however, that neither interpretation approaches agreement with the very much larger observed cross sections. The discrepancy cannot be due to an excessive evaluation of the effect of fission. since the observed yields for thorium nuclides formed in uranium spallation are even larger than the yields calculated under the assumption that fission did not occur (region bounded by dashed lines). The effect that a somewhat lower proton "cutoff" energy[8] (in the initial Monte Carlo cascade calculation) would have on the cascade-product distributions was then considered. It was concluded that this effect could not contribute sufficiently enhanced yields of thorium nuclides to give agreement with experiment.

The effect on the thorium yields of proton evaporation from excited protactinium nuclei was next considered. Values of Γ_p/Γ_n were calculated from Heckrotte.[11] Although the effect was appreciable for the lowest observed thorium mass numbers—e.g., about 40% for Th^{226}—the corrections are still far too small to account for the observed disagreement between calculated and observed values.

Another possible source of the discrepancy between observed and calculated $(p,3pxn)$ yields lies in the original Monte Carlo calculations, which considered only protons and neutrons to be involved in intranuclear collisions. However, there is good evidence that tritons and helium isotopes are also involved in the cascade process. Bailey[20] has studied the energy, angular distributions, and formation cross sections for secondary particles emitted in the bombardment of C, Al, Ni, Ag, and Au with protons and alpha particles of approximately 200 Mev. He has shown that while the low-

[19] Quoted by Metropolis *et al.* (reference 8).

[20] L. E. Bailey, Ph.D. thesis, University of California Radiation Laboratory Report UCRL-3334, March 1, 1956 (unpublished).

TABLE IV. Calculated yields (mb) for U and Pa nuclides at 236 and 100 Mev.

	236-Mev protons				100-Mev protons	
U	σ_{calc}(U)	σ_{obs}(U)	σ_{calc}(Pa)	σ_{obs}(Pa)	σ_{calc}(U)	σ_{obs}(U)
238	6.8	···	···	···	8.6	···
237	32.9	72	9.0	···	21.0	~90
236	35.8	···	10.3	···	17.5	···
235	27.3	···	9.3	13.8	17.1	···
234	18.5	···	7.7	···	14.9	···
233	10.2	···	5.7	···	9.6	···
232	4.9	···	4.0	···	5.6	···
231	2.0	···	2.8	···	2.5	···
230	0.6	0.37	1.9	4.6	0.64	0.41
229	0.14	0.073	1.3	···	0.018[a]	0.046
228	0.026	0.034	0.8	···	0.002[a]	0.012
			0.5	0.7		

[a] Not including contribution from Np(E^*) compound nucleus.

energy secondary alpha particles have properties consistent with an evaporation model, the high-energy alpha particles behave as though they had been produced in the original cascade; their cross section is strongly angle-dependent, the intensities are larger than predicted by an evaporation model, and the calculation of the center-of-mass velocities gives unreasonably high values. Since the production, in the cascade, of thorium from uranium (or radium from thorium) represents only about two percent of all the cascade interactions [Fig. 1(d)]—or about 35–40 mb—, only a small contribution from such neglected effects as $U^{238}(p,\alpha pxn)$Th* in the original cascade could conceivably be the source of the discrepancy.

It should be pointed out that the difference between the observed and calculated results discussed above are almost certainly due to neglected secondary effects. For this reason, these processes should not contribute significantly to the formation of uranium, neptunium, and protactinium nuclides; only products formed in low abundance should be affected.

CROSS SECTIONS AT OTHER ENERGIES

The results of Lindner and Osborne[3] include data from uranium bombarded by protons of energies down to 100 Mev. Calculations similar to those carried out for 340-Mev protons were also performed for protons of 100 and 236 Mev. For these calculations, the Heckrotte evaporation treatment was used; the E^*_{max} values from 100-Mev protons would have been too small to evaporate sufficient neutrons to arrive at the observed nuclides U^{229} and U^{228} had the assumption been made that each neutron carried off an average of 10 Mev of excitation energy. The values of Γ_n/Γ_f were taken from the straight lines of Fig. 5. The results are shown in Table IV for U and Pa nuclides from 236-Mev protons incident on U, and for U nuclides from 100-Mev protons on U.

Table IV indicates that, for 236-Mev protons incident on uranium, the calculated results for the yields of uranium and protactinium nuclei agree moderately well with experimental values [except for the (p,pn) case discussed above]. At 100 Mev the calculated yield for U^{230} is likewise acceptable. However, the agreement deteriorates significantly at lower masses, the calculated value of U^{228} being a factor of six lower than the observed value.

This failure of the calculation to predict the yields of the lightest uranium nuclei at 100 Mev deserves comment. Since the experiments of Lindner and Osborne were performed with natural uranium, reactions of the type $U^{235}(p,p7n)U^{228}$ might contribute. However, although at sufficiently low energies the formation of the lightest isotopes must be primarily from U^{235}—on energetic grounds—, quantitative calculation indicates that at 100 Mev this is still a negligible contribution.

Proton evaporation from the significant fraction of the cases at 100 Mev in which compound nucleus formation occurs [Fig. 1(a)] may make a sizable contribution near threshold. Calculations showed that for the formation of U^{229} this mechanism contributes 0.012 mb to the calculated cross section, but contributes only 1×10^{-3} mb to the formation of U^{228}. Examination of the calculation, however, indicates that a good estimation cannot easily be made for the formation of U^{228} from U^{238} bombarded with 100-Mev protons; close to the energetic threshold, the calculation is much too sensitive to the details of the neutron-evaporation model. For example, at 106 Mev the calculated cross section would be about 9×10^{-3} mb. This sensitivity to bombarding energy is not present at higher energies or for heavier products.

We conclude, therefore, that the model used in this paper predicts satisfactorily the yields of U^{230}, U^{229}, and U^{228} when uranium is bombarded with protons of energy varying from just above 100 Mev to 340 Mev. This general agreement supports the conclusion that fission competition with neutron evaporation is independent of energy for excited uranium nuclei (primarily U^{238*}, U^{237*}, U^{236*}) up to about 85 Mev. The reproduction of the energy dependence of the formation cross sections of these nuclei in this energy range is, however, primarily a check on the cascade calculations of Metropolis *et al.*[8] in providing the proper number of residual nuclei in appropriate excitation states, rather than an independent check on the fission competition during the evaporation stage.

From the data of Lindner and Osborne,[3] measurable yields in U^{238} spallation are indicated for such nuclides as Pa^{227}, Th^{227}, Th^{228}, Ac^{225}, Ac^{226}, and Ra^{224} at energies as low as 100 Mev. These cannot be reconciled with the present cascade-evaporation model which predict essentially zero yields for such nuclides at 100 Mev. Indeed, it is questionable whether the observed actinium yields from uranium are consistent with the cascade data, even at 340-Mev incident energy. Observed radium yields certainly are not.

It thus appears that the production of the low-yield elements having charges two or more lower than the target element, especially at low energies, cannot at present be satisfactorily explained by the model used. If we confine ourselves to the two-stage model of nuclear reactions, we must conclude that the trouble most likely is in our ideas of what happens in the cascade stage—evaporation theory seems hardly likely to provide enough charged-particle emission to lead to these elements. The first nonequilibrium stage must lead to a larger charged-particle ejection than predicted by the calculations of Metropolis *et al.*

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Cross Section for the Reaction $C^{13}(\gamma,n)$ at 6.4 Mev

R. D. EDGE*
Oak Ridge National Laboratory, Oak Ridge, Tennessee and the University of South Carolina, Columbia, South Carolina
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Using radiation from the $F^{19}(p,\alpha\gamma)$ reaction at 874-kev proton energy, the cross section for the reaction $C^{13}(\gamma,n)$ was found to be 94.1 ± 10 microbarns. The neutrons emitted from a target enriched in C^{13} were detected by an arrangement of BF_3 counters embedded in wax to give a 4π geometry.

THE reaction $C^{13}(\gamma,n)$ has previously been investigated using betatron bremsstrahlung radiation by Cook *et al.*[1,2] The cross-section curve shows a plateau at 8 to 10 Mev and a peak at around 15 Mev.

C^{13} is in many respects similar to Be^9. Both can be considered as consisting of an alpha-particle core with an extra "orbital neutron," and it has been suggested[3] that the peak in cross section found immediately above threshold in $Be^9(\gamma,n)$ might also occur in $C^{13}(\gamma,n)$. The difficulty of performing bremsstrahlung experiments at these low energies would have precluded observing this in the experiments noted above. Accordingly, an attempt was made to measure the cross section using the approximately monochromatic radiation from the $F^{19}(p,\alpha\gamma)$ reaction.

The proton target consisted of a layer of calcium fluoride deposited by evaporating the water from a slurry of this material placed on a copper backing. The target was water cooled, and the proton beam from the 3-Mv Van de Graaff generator centered by observing the hot spot. A stainless steel target tube $1\frac{1}{2}$ inches in diameter was used and the beam was stopped down to less than $\frac{1}{2}$-inch diameter.

Carbon, in elementary powder form enriched in C^{13}, was contained in two thin-walled nickel cylinders, 2 cm in diameter and 6 cm long. These cylinders had been heated for a long period, until they were of constant mass, to drive off occluded water vapor. The carbon samples were analyzed by the ORNL chemists and shown to consist of 56.5% C^{13}. Two cylinders identical in shape and size to the C^{13} cylinders had equivalent quanties by weight of ordinary graphite (effectively C^{12}) enclosed, to act as blanks, and two more were filled with heavy water. The detector has been described by Johnson, Galonsky, and Ulrich.[4] It was a 4π detector consisting of eight BF_3 counters, 1 inch in diameter by 6 inches long, embedded in paraffin on a 4.4-inch diameter circle around the beam axis, and operated in parallel by a conventional stabilized power supply and linear amplifier. The paraffin moderator was a cadmium-covered cube 17 inches on a side, with a 2-inch layer of paraffin outside. The optimum conditions for gamma-ray discrimination[4] were used.

A four-inch cylinder of sodium iodide (*Tl*) attached to the usual photomultiplier counting setup was placed approximately three meters from the target in order to monitor the gamma radiation and eliminate uncertainties in the yield per proton arising from variations in target thickness.

In performing the experiment, three runs were required in order to obtain a single set of results: In the first run, the C^{13} cylinders were placed in a standard position as close to the target as possible. The proton beam was switched on, and counts recorded by the BF_3 counter and NaI counter in a given time, generally fifteen minutes. In the second run, the C^{13} was replaced by the similar C^{12} cylinders, and in the third run by the D_2O cylinders. In all, eighteen sets of results were obtained.

* Oak Ridge Institute of Nuclear Studies Summer Research Participant 1959.
[1] B. C. Cook, A. S. Penfold, and V. L. Telegdi, Phys. Rev. **104**, 554 (1956).
[2] B. C. Cook, Phys. Rev. **106**, 300 (1957).
[3] E. Guth, (private communication).
[4] C. H. Johnson, A. Galonsky, and J. P. Ulrich, Phys. Rev. **109**, 1243 (1958).