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Binary and Ternary Fission of U^{238} Induced by Intermediate-Energy He^3 Ions*

K. W. MacMurdo† and J. W. Cobble
Department of Chemistry, Purdue University, Lafayette, Indiana 47907

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Cross-section data for 33 nuclides have been obtained radiochemically and used to construct a mass-yield curve for the fission of U^{238} induced by 31-MeV He^3 ions. The mass range studied was $24 \leq A \leq 212$ and the yields varied over seven orders of magnitude. The data for Na^{24}, Mg^{28}, Si^{31}, S^{38}, Ca^{47}, Au^{199}, Pb^{209}, and Pb^{212} confirm the existence of ternary fission in compound-nuclear processes at intermediate energies, as previously reported from these laboratories. These data also establish the transition region between ternary and highly asymmetric binary processes at $A \sim 45$. Recoil-range data indicate some small but significant differences between He^3- and He^4-induced ternary fission. On the assumption that there is a peak in the ternary mass-yield curve at $A \sim 24$, the total ternary-fission cross section for 31-MeV He^3 ions on U^{238} is $\sim 3.3 \times 10^{-30}$ cm^2, and $\sigma_{\text{ternary}}/\sigma_{\text{binary}} \sim 4.4 \times 10^{-6}$.

I. INTRODUCTION

In previous studies from these laboratories[1,2] certain low-mass isotopes in the range $24 \leq A \leq 40$ were observed in very low yields from the fission of U^{238} induced by He^4 ions. These yields, along with recoil-range data, clearly suggested that a true ternary fission process was being observed in the compound-nucleus energy region. The excitation function obtained for the ternary process was consistent with this proposal, showing a sharp cutoff at an excitation energy below $E^* \sim 18$ MeV. These energy studies also suggested that the ternary process would not take place below this threshhold, e.g., in the thermal-neutron-induced fission of uranium, unless the phenomenon was highly sensitive to other factors such as Z, A, and angular momentum.

Although preliminary experiments[3] on the He^4-induced fission of Th^{232} seemed to discount any special sensitivity of the ternary process to Z and A, it seemed worthwhile to study in detail other mass systems both to reconfirm the previous observations and to vary some of the nuclear parameters. In this paper we report a detailed study of the fission yields of U^{238} induced by He^3 ions over as wide a range of mass as is possible by radiochemical techniques. If the ternary process is highly dependent upon some extremely small direct-interaction part of the He^4 total cross section, then use of He^3 ions would not necessarily be expected to give the same ratio of ternary to binary yields at the same excitation energy. Further, at the same excitation energy, the angular momentum imparted by He^4 and He^3 ions to the compound nucleus should differ by a small but significant amount. In this latter respect, it is of course important to vary this momentum as much as possible, but it should be remembered that there are severe practical limitations to the problem. Use of protons (and deuterons) for detailed mass-yield studies in the ternary region at the necessary excitation energies is almost precluded by the relatively high cross sections for many "direct"-type (p, xn, yp) spallation reactions and by the great difficulty of removing impurities below a few parts per million. Use of fast neutrons is not practical, since there are no intense enough sources sufficiently free of a thermal-neutron background to observe the ternary process at all. The situation with regard to use of heavier ions is more complex and is currently being investigated.

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† Minnesota Mining and Manufacturing Co. Fellow, 1967–1968. This paper is taken from the Ph.D. thesis of K. W. MacMurdo, Purdue University, Lafayette, Ind., 1969. Present address: Savannah River Laboratory, E. I. DuPont de Nemours and Co., Aiken, S. C.
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TABLE I. Fission-product cross sections for U^{238} fission induced by 30.6-MeV He^3 ions.

Fission product	Isotopic measured cross sections	Isobaric cross sections[a]	$\bar{\nu}$
Na^{24}	197±39 nb	197±39 nb	
Mg^{28}	173±12 nb	173±12 nb	
Si^{31}	63±12 nb	63±12 nb	
S^{38}	16.5±3.3 nb	16.5±3.3 nb	
Ca^{47}	≤1.6 nb	≤1.6 nb	
Mn^{56}	450±68 nb	450±68 nb	3
Ni^{66}	15.9±1.6 μb	15.9±1.6 μb	4
Zn^{72}	97±10 μb	97±10 μb	4
As^{77}	1.0±0.1 mb	1.0±0.1 mb	5
Br^{83}	2.6±0.3 mb	2.6±0.3 mb	5
Sr^{89}	9.2±0.9 mb	9.2±0.9 mb	5
Sr^{91}	14.5±2.2 mb	14.5±2.2 mb	5
Zr^{95}	25.7±2.6 mb	25.7±2.6 mb	6
Zr^{97}	27.7±2.8 mb	27.7±2.8 mb	6
Mo^{99}	31.8±4.8 mb	31.8±4.8 mb	6
Ru^{105}	37.0±5.6 mb	37.0±5.6 mb	6
Ag^{111}	18.4±2.8 mb	18.4±2.8 mb	6
Cd^{115}	14.7±3.0 mb	14.7±3.0 mb	6
Ba^{140}	20.8±4.0 mb	26.7±4.0 mb	6
Ce^{143}	24.1±3.6 mb	24.1±3.6 mb	5
Sm^{153}	2.9±0.4 mb	2.9±0.4 mb	5
Eu^{157}	1.06±0.16 mb	1.08±0.16 mb	5
Gd^{159}	680±110 μb	680±110 μb	5
Er^{171}	15.2±2.3 μb	17.2±2.6 μb	4
Tm^{173}	5.3±0.7 μb	5.6±0.8 μb	4
Yb^{175}	3.04±0.46 μb	3.04±0.46 μb	4
Lu^{177}	1.32±0.22 μb	1.32±0.22 μb	4
Lu^{179}	1.00±0.15 μb	1.09±0.16 μb	3
Ta^{183}	211±32 nb	211±32 nb	3
Re^{189}	≤46 nb	≤46 nb	3
Au^{199}	≤4.0 nb	≤4.0 nb	0
Pb^{209}	≤9.0 nb	≤9.0 nb	0
Pb^{212}	≤1.8 nb	≤8.2 nb	0

[a] Calculated from the isotopic cross sections using the CCR rule of charge distribution.

II. EXPERIMENTAL

Cyclotron irradiations of 1-mil natural uranium foils were carried out at the Argonne National Laboratory using the experimental arrangement described previously.[1,2] The total He^3 current involved was 10–30 μA h and the incident He^3 beam energy on the top of the stacked foil target was 32 MeV, giving an average bombarding energy in the target of 30.6 MeV. The target foils had been analyzed spectroscopically for possible impurities which could interfere with observation of the fission fragments. Catcher foils, placed on both sides of the uranium target foil, were prepared from vacuum evaporation of very pure silver beads (United Mineral and Chemical Corp., New York, N.Y.) or from special purity aluminum foils (Cominco, Inc., Spokane, Wash.) which contained ≤0.3 ppm of silicon. It is perhaps worthwhile to note that observation of these very-low-yield nuclear processes essentially depends upon success in reducing impurity levels in the foils so that the "background" due to spallation reactions is not too large. While use of He^3 ions has a distinct advantage over He^4 ions as bombarding nuclei in this respect, it was not possible to isolate Na^{24} and Si^{31} from the target foil but only from the recoil catcher foils. Finally, it should be pointed out that there is no way to form Mg^{28} or S^{38} by any type of spallation process at these energies except the $Si^{30}(n, He^3)Mg^{28}$ and $Si^{29}(n, 2p)Mg^{28}$ reactions. The measured low numbers of fast neutrons incident on the target[3] ($\leq 5\times 10^{12}$ neutrons/μA h of He^3) and the known level of silicon contaminate in the target (10 ppm) eliminate even these possibilities.

In computing the Na^{24}, Si^{31}, and S^{38} cross sections, the assumption was made that the same fraction of the total activity recoiled out of the uranium target as was experimentally observed in this research for Mg^{28} (5.5% in both the forward and backward directions). In computing the binary cross sections, a recoil-loss correction factor was calculated for each nuclide from the approximate relationship[4]

$$R \approx 2W(F_f + F_b), \qquad (1)$$

where R is the experimentally determined[5] range of the U^{236*} binary fission fragments in uranium in mg/cm^2, W is the thickness of the uranium target foil in mg/cm^2, and F_f and F_b are the fractions of activity recoiling out of the target in the forward and backward (to the beam) directions, respectively. F_f and F_b were assumed to be equal at these energies.

Standard radiochemical techniques[6–8] were modified for use in the separations, and were repeated many

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times for the lower-yield isotopes to obtain the required level of decontamination from the gross amounts of high-yield binary fission products. The details of the radiochemical procedures are given elsewhere.[2,9,10] Briefly, these consisted of dissolving the target and/or catcher foils in the presence of known amounts of suitable carriers, followed by exhaustive separation and purification cycles. Eventually the nuclide was precipitated in a suitable stoichiometric form, dried, weighed and counted. The high-yield binary products were counted in standardized gas-flow Geiger counters. The low-yield binary and ternary products were counted in low-background (0.12–0.20 counts/min) gas-flow Geiger counters fitted with surrounding anticoincidence guard counters (manufactured by Tracerlab, Inc., Boston, Mass.). Radiochemical purity was established by the characteristic decay and by constant specific activity tests. In many cases it was possible to follow the decay through 4–6 half-lives; resolution of the decay curves from the always-present contaminates was accomplished in an objective manner by computer analysis. It was not uncommon to have to repeat the chemical purification steps many times, frequently requiring times of 12–15 h, resulting in chemical yields of ~35%, to obtain the desired degree of radio chemical purity.

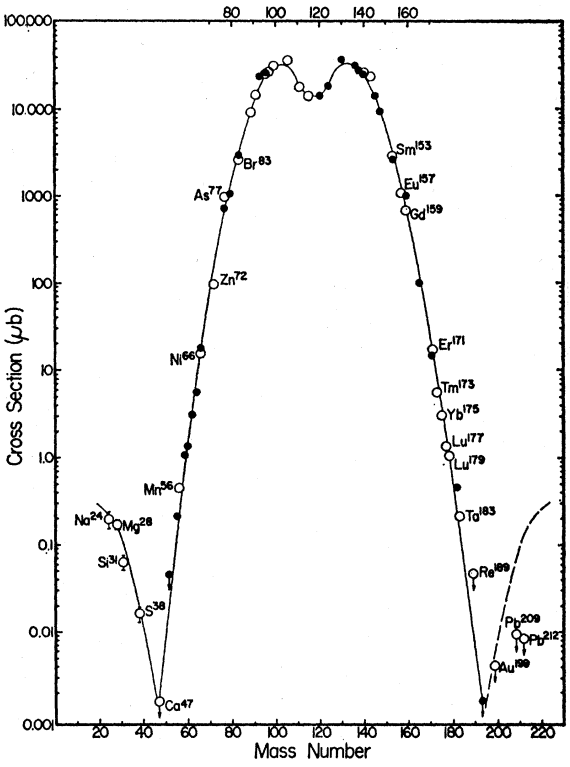

FIG. 1. Fission mass-yield distribution for U^{238} excited by 30.6-MeV He^3 ions. Data are from Table I. The dashed curve represents the heavy-mass yields expected from reflection of the light-mass-yield curve from hypothetical binary events. A value of $\bar{\nu}=0$ taken from Fig. 2 was used in this reflection. Closed symbols represent reflected data points.

III. EXPERIMENTAL RESULTS

Table I summarizes the experimental cross sections measured in this research for the fission of U^{238} induced by He^3 ions with an average bombarding energy of 30.6±0.5 MeV. The cross sections listed have been corrected for recoil losses as described previously; additional corrections had to be made to the Na^{24}, Si^{31}, and Pb^{209} data for contributions from He^3-ion- and secondary-neutron-induced spallation reactions on target and catcher foil impurities. The constant-charge-ratio[11] (CCR) rule was used to estimate the isobaric yields. This procedure has been used previously in these laboratories to treat isotopic yields in the He^4-ion-induced fission of many fission processes at moderate excitation energies. A preliminary mass-yield curve was constructed from those masses where the isotopic yields and isobaric yields were identical. This curve is independent of any charge distribution recipe. The other isobaric yields were then entered and the values of $\bar{\nu}$, the average number of neutrons emitted per fission, were obtained. These values of $\bar{\nu}$ are listed in column 4 of Table I. The complete mass-yield curve is shown in Fig. 1. The "reflected" or complementary points are indicated by closed circles in the usual manner. Even though no activity could be ascribed to the Au^{199}, Pb^{209}, and Pb^{212} fission products, the upper limits were corrected for charge distribution as if these species might have resulted from a highly asymmetric binary process.

Table II summarizes the recoil data for Mg^{28} and Sr^{89} fission fragments obtained in this research and from the data of Iyer and Cobble[9] in the 39-MeV α-induced fission of U^{238}. These data indicate that only about 11% of the total Mg^{28} activity in the He^3-induced fission of U^{238} ends up in the catcher foils, compared to the corresponding value of about 14% for the He^4-induced fission of U^{238}.[2,9] Similar comparison of Sr^{89} recoil data indicates less noticeable differences between the He^3- and He^4-induced fission of U^{238}. In the latter case, the recoil differences may not be significant, because of the errors involved in the determination of the recoil fractions. However, the recoil differences obtained in the He^3- and He^4-induced fission of U^{238} are apparently real. The average range of the Mg^{28} fission fragments in the He^3-induced fission is 9.2±0.4 mg/cm² of uranium, while in the He^4-induced fission, the average range of Mg^{28} fragments is 11.9±0.4 mg/cm² of uranium.

[9] R. H. Iyer, Ph.D. thesis, Purdue University, 1967 (unpublished).
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[11] R. H. Goekerman and I. Perlman, Phys. Rev. 76, 628 (1949).

TABLE II. Fission-product-range recoil data.

Isotope	Bombarding particle	E_{lab} (MeV)	F_F (%)	F_B (%)	F_F/F_B	R (mg/cm² of U)[b]
$M^{28}g^{8}$ [a]	He^4	39.0	7.3	7.0	1.04	12.3
Mg^{28} [a]	He^4	39.5	7.0	7.0	1.00	12.0
Mg^{28} [a]	He^4	38.9	6.5	7.0	0.93	11.6
Mg^{28} [a]	He^3	29.7	5.5	4.7	1.16	8.8
Mg^{28}	He^3	30.6	5.5	5.6	0.98	9.5
Mg^{28}	He^3	30.3	5.4	5.4	1.00	9.3
Sr^{89} [a]	He^4	39.0	7.7	7.0	1.10	12.6
Sr^{89}	He^3	29.9	6.6	...	...	11.3
Sr^{89}	He^3	30.3	6.5	6.5	1.00	11.2

[a] Data obtained from Ref. 9.

[b] Refers to the range of the fission fragment in mg/cm² of uranium as calculated from $R \approx 2W(F_F+F_B)$, where F_F and F_B refer to the fraction of the total activity found in the forward and backward catcher foils, respectively.

The total binary-fission cross section of U^{238} induced by 30.6-MeV He^3 ions is estimated by integration of half the area under the mass-yield curve to be 750±75 mb.

At present there is no way to decide how to use the admittedly limited ternary-yield data to construct a complete mass-yield curve for ternary fission. However, a linear plot of the cross sections versus mass for $A \leq 47$ suggests that Na^{24} may be near or at the peak of such a ternary distribution. The Na^{24} and Mg^{28} experiments had been designed to test just this point. If, in fact, such a peaked distribution exists, the total integrated ternary cross section $\sigma_{ternary}$ at this energy is $\sim 3.3 \times 10^{-30}$ cm².

The errors quoted for the individual cross sections were estimated from replicate determinations and uncertainties in the corrections for contributions from spallation impurities, counting efficiency estimations, decay curve resolutions, and in chemical yield determinations.

IV. DISCUSSION

The mass distribution (Fig. 1) and recoil studies, summarized in Table II, essentially confirm previous studies in these laboratories[1,2] on the light fragments and their fission-product nature. Although there are not many possible complementary binary fragments that can be measured radiochemically, limits on both lead isotopes are clearly far below those predicted from symmetrical reflection of the light-mass yields (dashed line in Fig. 1). In addition, the transition region between the light fragments and the left wing of the binary products has been defined even more clearly, and indicates a discontinuity in the total mass-yield distribution. Although only four light isotopes have been measured, these four are in agreement with a smooth mass-yield curve, and such yields do not appear to be greatly influenced by any special nuclear stability of the products being formed (e.g., $_{20}Ca^{47}$).

The average range of Mg^{28} was estimated from Eq. (1) to be 9.2±0.4 mg/cm² in uranium corresponding to an average kinetic energy of ~32 MeV, estimated from range data for Ne^{20} and Ar^{40}.[12,13] Such an energy cannot result from a spallation process under the bombarding conditions used in this research. The forward-backward recoil studies also support this conclusion.

The possibility was also considered that the observed light fragments might result from two successive binary events. Assume, for purposes of discussion, that the initial binary division of the Pu^{241}* compound nucleus excited to 40 MeV yields Re^{190}* with a formation cross section of 10 nb. It can be shown by a calculation similar to that to be presented in a later part of this discussion that this Re^{190}* fragment would not be expected to retain sufficient energy to undergo further fission. However, even if the Re^{190}* fragment retained as much as 40 MeV of excitation energy, the extremely small value of Γ_f/Γ_n of $\sim 5 \times 10^{-7}$ [14] for this nucleus at 40 MeV would preclude the detection of any further fragmentation. In all of these major respects, the He^3- and He^4-induced fission appear to be quite similar. The ternary yields are lower for the He^3 case by a factor of 2 (at the same excitation energies), following almost exactly the lower binary yields.

The range data provide one interesting difference between the He^3 and He^4 irradiations. The fraction of Mg^{28} which recoils from the 1-mil uranium foil is significantly smaller for the He^3 bombardments (11 versus 14% for He^4). We tentatively suggest that this small difference may be due to the differing amounts of angular momentum imparted by He^3 and He^4 projectiles on the U^{238} nucleus. The studies of Bochagov *et al.*[15] indicate that greater angular momentum might result in greater forward-backward anisotropy. This small sensitivity to angular momentum provides an interesting probe for further studies on the dynamics of the ternary process.

The binary-fission mass-yield curve has now been studied over a sufficiently wide range of mass to see clearly the change in the average number of neutrons $\bar{\nu}$ emitted at different A_H/A_L ratios. In reflecting the binary-yield data to obtain Fig. 1, it was possible to delineate regions of mass that could be associated with

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[14] G. M. Raisbeck and J. W. Cobble, Phys. Rev. **153**, 1270 (1967).

[15] B. A. Bochagove *et al.*, Yadern. Fiz. **3**, 461 (1965) [English transl.: Soviet J. Nucl. Phys. **1**, 323 (1965)].

various values of $\bar{\nu}$. The midpoint of these mass regions and the corresponding values of $\bar{\nu}$ are plotted in Fig. 2. The solid line in this figure was calculated on the basis of the energetics of the system. In this calculation, the distance between the fragments at scission is used as an adjustable parameter. The total excitation energy E_x available for neutron emission is assumed to be the difference in the total mass energy released in fission, ΔE, and the sum of the total fragment kinetic energy, E_k, and that energy removed by prompt γ emission, E_γ. ΔE is estimated using the mass tables of Hillman.[16] The charges of the light and heavy primary fission fragments $Z_{p(L)}$ and $Z_{p(H)}$ are computed from the CCR rule. The total kinetic energy of the two primary fission fragments is assumed to arise solely[17] from the Coulombic repulsion of the two fragments and is given by

$$E_k = Z_{p(L)} Z_{p(H)} e^2 / R, \qquad (2)$$

where $R = r_0(A_L^{1/3} + A_H^{1/3}) + \Delta$. A_H and A_L are the mass numbers of the heavy and light fission fragments, respectively, and Δ is an adjustable parameter; r_0 was set at 1.44 F. The use of Δ as an adjustable parameter has been used before in a model of the fissioning nucleus suggested by Ferguson and Read.[18] E_γ is fixed at 7.5 MeV.[17] The total excitation energy available for this emission of neutrons is assumed to be divided between the two fragments in proportion to their respective masses.[19]

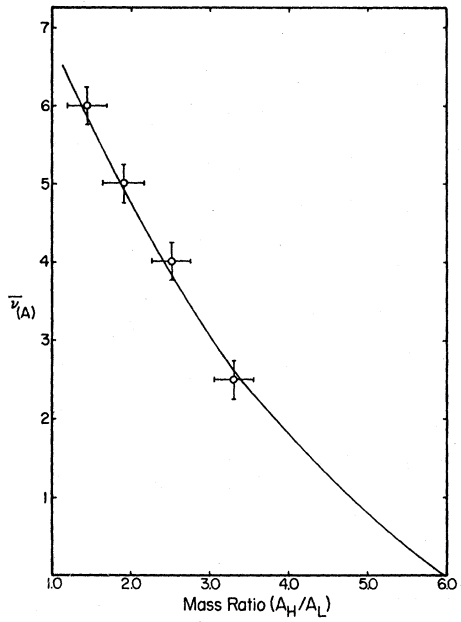

FIG. 2. Variation of the average number of neutrons $\bar{\nu}$, emitted in fission, with mass ratio. The solid line was calculated on the basis of the energetics of the system (see text).

A primary fragment of mass A excitation energy E_{x1}, and neutron binding energy B_{n1} is assumed to emit a neutron when $E_{x1} > B_{n1}$. After emission of this first neutron, the remaining excitation energy of the fragment whose mass is now $A-1$ is given by

$$E_{x2} = E_{x1} - B_{n1} - 1.2 \text{ MeV}.$$

The 1.2-MeV term represents the average neutron kinetic energy. This remaining excitation energy, E_{x2}, is now compared with the neutron binding energy of the second fragment, B_{n2}. If $E_{x2} > B_{n2}$, then another neutron is evaporated. The process continues until $E_{xi} < B_{ni}$. At this point, a fraction of a neutron is assumed emitted which is given by E_{xi}/B_{ni}.[20] This statistical simplification results in a smooth rather than a stepwise variation in $\bar{\nu}$ with mass ratio. Even-odd effects on the neutron binding energy were accounted for by using values of B_n obtained from mass tables of Hillman.[16] This simple model seems to fit the highly asymmetric binary-fission neutron data satisfactorily.

Further experiments are continuing in these laboratories in both lighter and heavier target nuclei and it may also be possible to obtain some information on the angular distribution of some of the products from this interesting form of fission.

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[18] J. M. Ferguson and P. A. Read, Phys. Rev. **150** (1966).

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[20] It should be recalled that the average energy removed by γ-ray emission, 7.5 MeV, has already been subtracted.