for much helpful guidance and encouragement. Thanks are also due to Professor E. S. Rost and Professor J. A. Wheeler for stimulating discussion and criticism.

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[2]The deformational parameter ϵ and most other symbols are defined as in Nilsson[1]; $\epsilon > 0$ corresponds to prolate spheroidal deformations, with $1+\epsilon$ roughly equal to the ratio of major and minor axes for ϵ small, and $\epsilon = 0.95\beta + 0(\beta^2)$.

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[4]The $n=7$ proton and $n=8$ neutron oscillator shells have been included in the calculations in their unshifted locations, with $k=0.05$, and $\mu=0.60$ and 0.45, respectively.

[5]This procedure was recommended by S. A. Moszkowski, in Handbuch der Physik (Springer-Verlag, Berlin, 1957), Vol. 39. Calculations in which half the total potential energy is subtracted from this sum, as originally suggested by Nilsson,[1] lead to similar conclusions.

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[10]Here $\hbar\mathring{\omega}_0 = 41A^{-1/3}$ MeV. For comparison purposes, the mass parameter $B_{\mathrm{LD}} = \frac{1}{2}$ (nuclear density) (nuclear radius[5]) calculated on the liquid-drop assumption of vortex-free flow is in these units $B_{\mathrm{LD}} = 250\hbar/\mathring{\omega}_0$ for U^{238}.

[11]Recent reviews include E. K. Hyde, The Nuclear Properties of the Heavy Elements III (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1964); L. Wilets, Theories of Nuclear Fission (Clarendon Press, Oxford, England, 1964); W. J. Swiatecki, in Proceedings of the Symposium on Physics and Chemistry of Fission, Saltzburg, 1965 (International Atomic Energy Agency, Vienna, 1965). The most detailed published LD calculation of a spontaneous fission rate is by W. D. Foland and R. D. Present, Phys. Rev. 113, 613 (1958).

EVIDENCE OF TERNARY FISSION AT LOWER ENERGIES*

R. H. Iyer† and J. W. Cobble‡
Department of Chemistry, Purdue University, Lafayette, Indiana
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Data have been obtained which we believe clearly demonstrate for the first time the existence of ternary fission in lower energy (compound) nuclear processes. In defining ternary fission, events are excluded that give rise to very small fragments, such as alpha particles, which are best explained as accompanying ordinary binary fission.[1] Lower energies are defined as ≤0.2 GeV, where compound nuclear processes are dominant. Within these limits, several incidences of ternary fission determined by fission-track methods have been reported and have been summarized by others.[1,2] In these cases the data have, however, either been disputed, or were based upon too few events to be meaningful. The most recent and extensive data on the thermal neutron-induced ternary fission of U^{235} and the spontaneous ternary fission of Cf^{252} are by Muga.[3,4] However, the radiochemical evidence is not in agreement with Muga's instrumental results, the limits set by the former being from 3 to 7 orders of magnitude smaller.[2,5,6]

Excitation functions in the present research were determined for the formation of the 21.3-h Mg^{28} and the 2.9-h S^{38} from the fission of natural uranium with 20- to 40-MeV He^3 and He^4 ions. These two isotopes were chosen since they are far removed from any possible binary products at these energies, and also because they are neutron-excess species which can only be formed either in fission or by a limited number of spallation reactions. The absolute cross sections for formation and yields were measured in 1-mil uranium foils and in silver catcher foils which collected the recoiling fragments in both the forward and backward directions parallel to the beam. The experimental arrangement is shown in Fig. 1. The uranium foils, T, were of ≥99.9% purity and were analyzed for possible impurities which could give rise to Mg^{28} and S^{38} by spallation processes.[7] The foils were

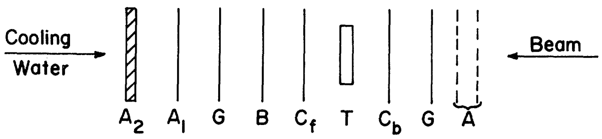

FIG. 1. Schematic diagram of target assembly. A, energy adjusting foils; G, silver guard foils; C_b, C_f, backward and forward silver catcher foils; T, 1-mil uranium target foil; B, blank silver foil; A_1, 1-mil high-purity aluminum foil; A_2, 40-mil aluminum foil.

cleaned with extremely pure nitric acid to dissolve the oxide coatings, then washed with conductivity water and absolute alcohol. They were stacked between 8- to 10-mg/cm^2 silver foils (C_b and C_f) made by evaporation of 99.9999%-pure[8] silver beads onto Lexan polycarbonate plastic film which was subsequently removed by dissolution.

Target assemblies were irradiated with He^3 and He^4 ions on an external port of the 60-inch cyclotron at the Argonne National Laboratory for one to two hours for a total of 10 to 25 μA h. Various incident energies were produced by placing additional aluminum foils, A, as shown in Fig. 1. The various foils were dissolved separately in the presence of added carriers for Mg, S, and Sr. Sr^{89} was isolated from most of the targets as an internal binary-fission monitor. The Sr^{89} cross sections indicated that the total binary-fission cross sections in He^3-induced fission are appreciably lower than those induced by He^4 at the same excitation energies. To check this point, the total He^3 fission excitation function of uranium was determined in separate experiments by a fission-track detection method using plastic films as detectors.[9,10]

The magnesium and sulfur fractions were extensively purified to obtain the necessary levels of decontamination from other fission and spallation products. Magnesium was counted as $MgNH_4PO_4 \cdot H_2O$ and sulfur as $BaSO_4$ in low background (0.12-0.18 counts/min) gas-flow Geiger counters having anticoincidence guard rings. Gross counting rates varied from about 2 to 100 counts/min depending upon the isotope and excitation energy. Identity was established by the characteristic half-lives (in most cases the decay could be followed through 4-5 half-lives) and, in the case of Mg^{28}, from the characteristic gamma-ray spectra of the Mg^{28}-Al^{28}-Si^{28} decay sequence, determined in a sodium-iodide scintillation spectrometer using multichannel analysis. The data are summarized in Table I.

The evidence seems clear that both Mg^{28} and S^{38} arise from a true ternary-fission process from the following considerations:

(1) The same fraction of Mg^{28}, 7.3%, was found in the forward catcher foil, C_f, as in the backward foil, C_b, 7.0%, in the highest ener-

Table I. Fission-product cross-section data.

Excitation energy (MeV)	Total binary-fission cross section (mb)[a, b]	Mg^{28} cross section (nb)	S^{38} cross section (nb)	Sr^{89} cross section (mb)
		He^4-induced fission		
18.56 ±1	128	≤1.9	...	...
20.0 ±1	240	5.0 ±1.5	...	...
21.4 ±1	370	16.1 ±4.8	...	...
23.6 ±1	602	35 ±10	...	7.9
27.0 ±1	910	66 ±20	≤2.7	10.9
33.6 ±0.5	1320	168 ±42	21 ± 5	...
34.0 ±0.5	1325	197 ±50	...	...
34.2 ±0.5	1330	198 ± 50	25 ± 6	11.5
34.6 ±0.5	1350	194 ± 48	17 ± 4	...
		He^3-induced fission		
32.6 ± 1	160	15.5 ± 4.6	...	1.8
38.8 ± 0.5	560	135 ± 27	...	...
39.4 ± 0.5	600	172 ± 35	24.2 ± 5	4.7

[a]The U^{238} He^4-induced total fission cross sections are from L. J. Colby, M. L. Shoaf, and J. W. Cobble, Phys. Rev. 121, 1415 (1961).
[b]The U^{238} He^3-induced total fission cross sections are from the present research.

gy He^4 bombardment. However, the silver foil used as a "blank," B, did not contain any detectable amounts of Mg^{28} (≤ 0.1 counts/min). He^3 bombardments gave similar results. This angular dependence would not be observed for spallation reactions at these energies.

(2) The only spallation reactions in the energy region of interest which can lead to Mg^{28} or S^{38} are $Mg^{26}(\alpha, 2p)Mg^{28}$, $Al^{27}(\alpha, 3p)Mg^{28}$, $Si^{29}(n, 2p)Mg^{28}$, $Si^{30}(n, He^3)Mg^{28}$, $S^{36}(\alpha, 2p)S^{38}$, and $Cl^{37}(\alpha, 3p)S^{38}$. From the low levels of magnesium, aluminum, silicon, sulfur, and chlorine (Ref. 7) and using either experimental data or reasonable estimates for the cross sections (few millibarns), it is estimated that these reactions would have a negligible effect except for the reaction $Mg^{26}(\alpha, 2p)Mg^{28}$ at the lower energies, where suitable corrections were made.[11] The fast- and slow-neutron fluxes produced in similar target assemblies have been measured in previous research from this laboratory,[12] and various types of double-capture reactions, involving secondary neutron and/or charged particles can also be ruled out.

(3) It is impossible to form either Mg^{28} or S^{38} by any of the above type of spallation reactions using He^3, with the exception of the reactions $Si^{29}(n, 2p)Mg^{28}$ and $Si^{30}(n, He^3)Mg^{28}$; the low numbers of secondary fast neutrons ($\leq 5 \times 10^{12}$ neutrons/μA h of He^3) and the small level of silicon contaminant (0.7 ppm) eliminate those possibilities. However, the observed excitation functions for both the He^3- and He^4-induced ternary-fission reactions are similar.

(4) The integral range measurements indicate that similar fractions of the binary-fission product Sr^{89}(14%) and Mg^{28}(14%) recoiled out of the thick uranium target. This indicates that the Mg^{28} fragment has a much higher kinetic energy than would be possible from spallation reactions at these energies.

(5) Some preliminary differential range measurements using He^3-induced fission also indicated the fission-productlike nature of the fragments. These experiments were performed by inserting several thin (1-2 mg/cm^2) silver foils, C_f, behind the urnaium target foil, T. At the low levels of activity involved in the present measurements it was not possible to obtain precise ranges. However, the data indicated that 30% of the Mg^{28} fragments completely penetrated one silver foil of 1.46 mg/cm^2.

Figure 2 illustrates the sharp dependence of the ratio of ternary- to total binary-fission yields of Mg^{28} and S^{38} on energy for the compound nuclei $Pu^{242*}(Z^2/A = 36.5)$ and $Pu^{241*}(Z^2/A = 36.6)$ produced by helium-ion excitation. If it is assumed that these species are typical and representative of the ternary process in heavy elements, then it is possible to make order-of-magnitude estimates on the probability of observing ternary fission in similar compound nuclei. The data given in Table I and Fig. 2 suggest that excitation energy will largely be the dominating factor in fixing the ratio of ternary/binary fission yields. Even though the present data have not tested the Z^2/A dependence of the ternary/binary ratio over any significant range, we believe it is reasonable to assume that this fissionability parameter will not be so important in fixing the ternary/binary ratio as the excitation energy. Further, it seems reasonable to assume that angular-momentum effects will be only of secondary importance in affecting the ternary/binary ratio when light particles, such as neutrons and helium ions, are used to induce fission, since such particles add only relatively small amounts

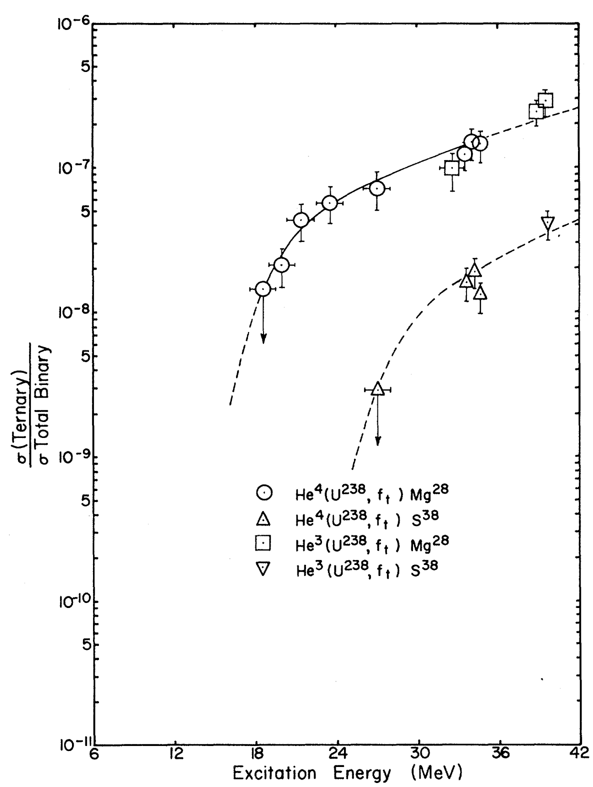

FIG. 2. The ratio of ternary- to total binary-fission cross section as a function of excitation energy for U^{238} excited with He^3 and He^4 ions.

of angular momentum to the system. For example, from the shapes of the curves of Fig. 2 and at an excitation energy of 6.5 MeV, corresponding to the thermal neutron-induced fission of U^{235}($Z^2/A = 35.9$ for U^{236*}), the ratio of the cross sections of the individual fission products to the total binary-fission cross section is expected to be of the order of $\leq 10^{-12}$, which is consistent with the previous radiochemical data.[6] This corresponds to 10^{-10}% of the total thermal binary-fission cross section, a value clearly too small to be observed by presently known means, and orders of magnitude smaller than the 10^{-5}% reported by Muga[3,4] for representative products. For S^{38}, the expected yields would be another factor of 10 lower.

In the spontaneous fission of Cf^{252}($Z^2/A = 38.1$), even allowing a generous increase (two orders of magnitude) in the ratio of ternary to binary fission with Z^2/A, the data of Fig. 2 would suggest the same order of magnitude of yields. Even if Mg^{28} and S^{38} represent only 0.1% of the total ternary-fission cross section (certainly a conservative estimate), it seems improbable that ternary fission has been observed previously in either the thermal neutron-induced or spontaneous fission in the heavy elements. It is for these reasons that we believe previously reported positive observations of lower-energy ternary fission are spurious. Similar conclusions on the thermal neutron-induced fission of U^{235} have already been published by others.[2,5,6]

From the yields in the catcher foils, a range of ~12 mg/cm^2 in uranium[13,14] and 4.5 mg/cm^2 in aluminum[15] for Mg^{28} can be estimated for the highest energy in these studies. Unfortunately, present range-energy correlations for fission products of this low mass are not very certain. A kinetic energy as high as ~100 MeV can be estimated by comparison of the ranges of binary-fission products of known kinetic energy in aluminum.[16] Alternatively, a value as small as 40-50 MeV can be obtained by interpolation of the heavy-ion-range data[17,18] for Ne^{20} and Ar^{40} in aluminum. Since the effective range of such fragments depends to some extent upon the mechanism of production, these two values supposedly represent some type of limits, although at present we consider the lower energy as more probable.

At the present time we are attempting to elucidate further the many questions suggested by these preliminary observations. It will be interesting to see if the very-high-energy ternary-fission yields proposed by others[19] at 0.4 GeV are related to the same type of ternary fission observed in this research. The data of Fig. 2 suggest that the yields of ternary-fission products should not increase too rapidly at the higher energies since the total fission cross section changes only slowly (1.5 to 3.0 b in this range).

We would like to acknowledge the assistance of Milan Oselka and his staff at the Argonne National Laboratory cyclotron for their assistance in these studies. We are particularly indebted to P. F. Thomason, J. R. Sites, and W. Laing of the Analytical Chemistry Division of the Oak Ridge National Laboratory for their detailed analysis of the uranium foil. It is also a pleasure to acknowledge helpful discussions with Mr. Grant Raisbeck of this Laboratory.

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†On leave from the Atomic Energy Establishment Trombay, Bombay, India.

‡To whom inquiries regarding this communication should be sent.

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$3p)Mg^{28}$ has been determined in separate experiments; the cross section varies from 1 to 100 μb in the 33- to 40-MeV energy range. The reaction $Cl^{37}(\alpha, 3p)S^{38}$ is expected to have even smaller values at comparable energies since it has a threshold which is ~2 MeV higher in energy. For $Mg^{26}(\alpha, 2p)Mg^{28}$, the cross-section data of R. J. Carr and R. H. Lindsay, Phys. Rev. 118, 1293 (1960), were used. We are indebted to a referee for pointing out these data to us.

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