

Mass- and Charge-Distribution Studies in the Fission of Au^{197} by Intermediate-Energy Helium Ions*

F. L. LISMAN,† H. W. BRANDHORST, JR.,‡ AND J. W. COBBLE
Department of Chemistry, Purdue University, Lafayette, Indiana

(Received 7 July 1965)

The absolute fission yields of twelve nuclides from the helium-ion-induced fission of Au^{197} have been determined radiochemically at 40.0, 37.0, and 33.5 MeV. The mass yield distributions were observed to be symmetric, and an upper limit of approximately 0.03% for an asymmetric fission mode was obtained. Independent and cumulative yield data were best correlated with the constant-charge-ratio postulate, although the charge distribution about Z_p was Gaussian and similar to those observed by others in heavier-element nuclides. The width of the mass distributions was invariant over the energy range studied, and fewer neutrons appear to be emitted than had been previously reported. The total fission cross sections are in good agreement with those obtained by others using purely instrumental techniques.

I. INTRODUCTION

THE fission of lower Z elements at compound-nucleus energies continues to be of interest. Any comprehensive understanding of the fission process would hopefully apply to all pertinent regions of the periodic table. The studies of Fairhall and his co-workers on the helium-ion-induced fission^{1,2} of radium clearly indicated the growth of the symmetric fission process with respect to the asymmetric mode in lower Z elements. These were followed by similar radiochemical studies on bismuth³⁻⁵ which indicated only the symmetric division, although Sugihara *et al.*,⁶ have more recently demonstrated the presence of a 0.3% asymmetric contribution in Bi^{209} excited by 36-MeV protons. Johansson⁷ has postulated that if asymmetric fission continues at all below $A=210$, it will again reach a maximum in the hafnium ($Z=72$) region, although Newson⁸ believes that such asymmetric modes will not be observed below $A=210$. A preliminary radiochemical study⁹ in these laboratories in 1960 on the intermediate energy helium-ion-induced fission of rhenium indicated appreciable asymmetric fission. However, these results have now been demonstrated^{10,11} to have been due to the

trace (ppm) presence of heavy-element impurities in aluminum catcher and mounting foils.

The problem of absolute-fission-product cross sections in lower- Z elements is complicated by a number of considerations. Because of the large energy dependence of symmetric fission, the lowest excitation energies are favored if asymmetric contributions are to be enhanced. However, the total and individual fission cross sections involved are in the microbarn-to-millibarn region and are rapidly decreasing at lower energies. Consequently, trace impurities of fissionable heavy elements and spallation-reaction impurities can be very misleading. Yet, radiochemical methods still appear to be superior at present in identifying the mode and other details of fission in the lower Z region, although purely instrumental techniques^{5,12} provide a valuable check on the over-all total fission cross section.

The very recent observations of Neuzil,¹³ which were interpreted as being consistent with substantial relative amounts of asymmetric fission products in gold were not in agreement with preliminary studies in these laboratories¹⁴ in 1961. Since that time, the advances in low-level counting techniques, the availability of extremely pure materials, and methods for purifying and preparing uncontaminated reagents have made possible more detailed and accurate fission studies on the intermediate-energy fission of elements below bismuth. The present communication reports results on the gold fission (Tl^{201} compound nucleus) and sets an upper limit on the helium-ion-induced asymmetric fission mode in Tl^{201} at 31.3 MeV excitation as $\leq 0.03\%$.

Further details on the charge distribution, independent yields and neutron release are also reported. At lower energies and masses, such data are extremely difficult to obtain by any other reliable means.

* Supported by the U. S. Atomic Energy Commission.

† From the Ph.D. thesis of F. L. Lisman, Purdue University, June, 1965; present address: Phillips Petroleum Company, Idaho Falls, Idaho.

‡ From the Ph.D. thesis of H. W. Brandhorst, Jr., Purdue University, August, 1961; present address: National Aeronautics and Space Administration, Lewis Research Center, Cleveland, Ohio.

¹ A. W. Fairhall and R. C. Jensen, *Phys. Rev.* **109**, 942 (1958).

² A. W. Fairhall and R. C. Jensen, *Phys. Rev.* **118**, 771 (1960).

³ A. W. Fairhall, *Phys. Rev.* **102**, 1335 (1956).

⁴ R. Vandenbosch and J. R. Huizenga, *Phys. Rev.* **127**, 212 (1962).

⁵ H. C. Britt, H. E. Wegner, and J. Gursky, *Phys. Rev.* **129**, 2239 (1963).

⁶ T. T. Sugihara, J. Roesmer, and J. W. Meadows, Jr., *Phys. Rev.* **121**, 1179 (1961).

⁷ S. A. E. Johansson, *Nucl. Phys.* **22**, 529 (1961).

⁸ H. W. Newson, *Phys. Rev.* **122**, 1224 (1961).

⁹ R. D. Griffioen, Ph.D. thesis, Purdue University, January, 1960 (unpublished).

¹⁰ M. E. Davis, Ph.D. thesis, Purdue University, January, 1963 (unpublished).

¹¹ C. Menninga, Ph.D. thesis, Purdue University (to be published).

¹² J. R. Huizenga, R. Chaudhry, and R. Vandenbosch, *Phys. Rev.* **126**, 210 (1962).

¹³ E. F. Neuzil, *J. Inorg. Nucl. Chem.* **27**, 3 (1965).

¹⁴ H. W. Brandhorst, Ph.D. thesis, Purdue University, August, 1961 (unpublished).

II. EXPERIMENTAL PROCEDURE

A. Bombardment Conditions

Thin target foils (1.5 mg/cm^2) were prepared by vacuum evaporation of high-purity gold¹⁵ ($\geq 99.999\%$) from a hot tungsten wire onto high-purity radiochemically analyzed aluminum foil. The thickness of such foils was determined by weighing, and had a surface density uniform to $\pm 7\%$. Radiochemical analysis of the aluminum foils indicated Ni and Fe and approximately 40×10^{-9} of fissionable heavy elements (presumably U or Th). Consequently, in those bombardments, where such impurities were objectionable, self-supporting gold targets of $5\text{--}10 \text{ mg/cm}^2$ were used. These foils were prepared by evaporation onto polycarbonate plastic films, which were then dissolved. Thick foil target assemblies were used only to obtain relative cross sections. Subsequent study indicated that the energy dependence of the individual fission cross sections were sufficiently similar so as to introduce only minor errors by such a procedure.

Target assemblies of the stacked-foil type were used on the external beam facilities of the Argonne National Laboratory 60-in. cyclotron. Range-energy relationships

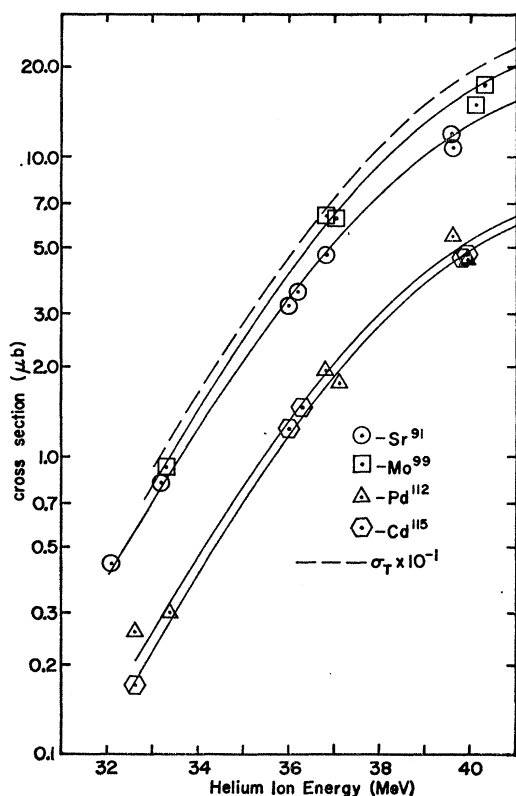


Fig. 1. Variation of isobaric chain cross sections with energy.

¹⁵ United Mineral and Chemical Corporation, New York, New York.

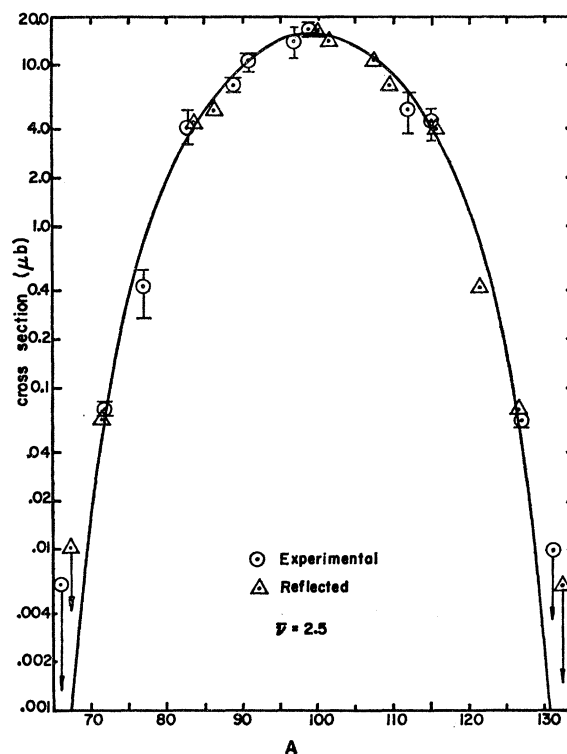


Fig. 2. Fission mass-yield curve for 40.0-MeV helium ions.

based on the work of Bichsel *et al.*,¹⁶ were used to determine the energies of the incident helium ions. When aluminum foils were used to collect recoil fission products, they were also dissolved along with the target foil. When gold foils alone were used the recoils from each face were assumed to compensate for any such loss and only the target foil was dissolved. High-current target holders¹⁰ permitted the use of up to $20 \mu\text{A}$ currents and accumulated total beams of $45 \mu\text{A h}$ were normally used. The emergent helium-ion beam from the cyclotron (about 43 MeV) had an energy spread of about 0.40 MeV, FWHM (full width at half-maximum). The maximum amount of various absorbers used increased this spread to 0.65 MeV, FWHM. Other studies¹² on the total fission cross section have indicated that such dispersions result in less than a 5% error in the cross section data at 30.0 MeV, the lowest energy used.

However, the large energy dependence of the fission cross section with energy made it extremely important to either reproduce exactly the energy of the cyclotron beam on replicate bombardments, or else to normalize cross sections to a reference energy. The former procedure was not practical; the second method requires empirical calibration of the variation of some of the individual-fission-product cross sections with energy. Such a normalization curve is shown in Fig. 1. The corrections from one run to another normally amounted to

¹⁶ H. Bichsel, R. F. Mozley, and W. A. Aron, *Phys. Rev.* **105**, 1788 (1957).

TABLE I. Cross sections for helium-ion-induced fission of Au¹⁹⁷.

Energy (MeV)	Isotope	Mass	40.0		37.0		33.5	
			σ (μb)	σ_{corr} (μb)	σ (μb)	σ_{corr} (μb)	σ (μb)	σ_{corr} (μb)
	Ni ⁶⁶		≤ 0.006	≤ 0.006				
	Zn ⁷²	66	0.071 ± 0.005		0.015 ± 0.005		0.0067 ± 0.0014	
	As ⁷⁷	72	0.43 ± 0.15	0.074	0.45 ± 0.18	0.015	0.037 ± 0.009	0.0067
	Br ⁸²	77	0.21 ± 0.04	0.43				0.037
	Br ⁸³		4.2 ± 1.1		$0.032 (\pm 0.010)$		0.182 ± 0.028	
	Sr ⁸⁹	83	7.49 ± 0.56	4.2	1.22 ± 0.04	1.22		0.182
	Sr ⁹¹	89		7.49	3.2 ± 1.5	3.2	0.44 ± 0.11	0.44
	Sr ⁹¹	91	10.6 ± 1.2	10.7	3.83 ± 0.69	3.83	0.67 ± 0.18	0.67
	Nb ⁹⁶		0.289 ± 0.059					
	Zr ⁹⁷	97	13.0 ± 2.9	14.1	5.25 ± 0.27	5.58	0.734 ± 0.091	0.765
	Mo ⁹⁹	99	16.3 ± 2.1	16.3	6.34 ± 0.09	6.34	0.92 ± 0.41	0.92
	Pd ¹¹²	112	4.6 ± 1.4	5.2	1.85 ± 0.13	2.01	0.279 ± 0.028	0.294
	Cd ¹¹⁵	115	4.5 ± 1.0	4.5	1.47 ± 0.30	1.47	0.26 ± 0.12	0.26
	Sb ¹²⁷	127	0.021 ± 0.002	0.063	0.008 ± 0.0007	0.021		
	I ¹³¹		≤ 0.01					

less than 20%, for an energy spread of ± 0.4 MeV from the reference energies, except for a few relative yields obtained with thick targets.

This method of correction assumes that the energy dependences of the isobaric chains are the same; the data in Fig. 1 seem to confirm this assumption, particularly since the curves for the individual chains (isobars) also parallel that of the total fission cross section determined elsewhere.¹²

B. Radiochemical Procedures

Standard radiochemical procedures¹⁷ common to higher Z fission were used in this study, except that much higher levels of decontamination from extraneous spallation products were required. In general only, a few isotopes were removed from each of the seventeen targets irradiated, although a number of replicate runs were made. Half-life, constant specific activity, and in some cases, parent-daughter isotope "milking" procedures were used to establish the purity of the separated fission products.

Measurements of radioactivity were carried out by beta counting in either low-background anticoincidence shielded Geiger counters¹⁸ or thin-window flow proportional counters. The former counters had reliable backgrounds of about 0.15 counts/min. Many of the isotopes involved were standardized directly by 4π beta and 4π beta-gamma coincidence techniques¹⁷ similar to

those used in our previous heavy-element studies.^{19,20} From such data, simulated corrections for back-scattering, self-absorption, foreshattering, and effective geometry were generated for Nb⁹⁶, Ni⁶⁶, Zn⁷², Ag¹¹², Zr⁹⁷, and Br⁸³.

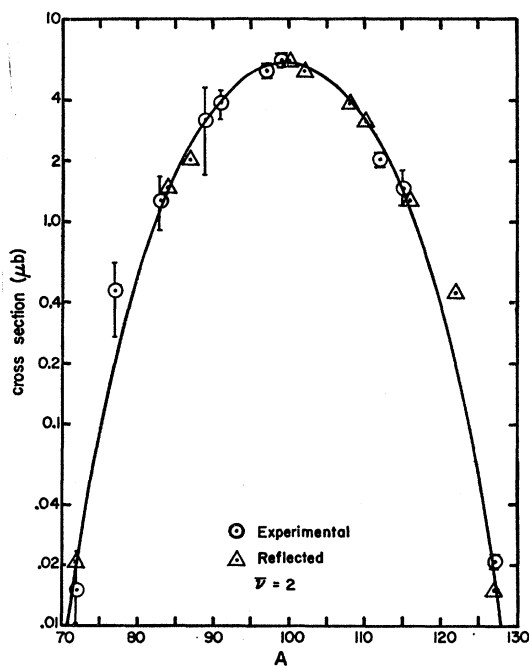


FIG. 3. Fission mass-yield curve for 37.0-MeV helium ions.

¹⁷ F. L. Lisman, Ph.D. thesis, Purdue University, June, 1965 (unpublished).

¹⁸ H. W. Andre, Ph.D. thesis, Purdue University, June, 1964 (unpublished).

¹⁹ R. Gunnink, L. J. Colby, Jr., and J. W. Cobble, *Anal. Chem.* **31**, 796 (1959).

²⁰ R. Gunnink, Ph.D. thesis, Purdue University, February, 1959 (unpublished).

TABLE II. Data on the intermediate-energy helium-ion-induced fission of Au¹⁹⁷.

Incident energy (MeV)	$\bar{\nu}$	T ²⁰¹ excitation energy	(FWHM) ^a	Total-fission cross section (μ b) (radiochemically)	Total-fission cross section (μ b) (instrumental)	
					Huizenga ^b	Raisbeck ^c
40.0	2.5 \pm 0.5	37.7	22 \pm 2	191 \pm 32	180 \pm 27	160 \pm 15
37.0	2.0 \pm 0.5	34.8	21 \pm 2	72 \pm 18	75 \pm 11	57 \pm 6
33.5	1.5 \pm 0.5	31.3	20 \pm 2	10.5 \pm 2	13 \pm 2	8 \pm 1

^a Full width at half-maximum of the mass distribution.

^b Errors are those estimated by the present authors and include the original published error plus errors introduced in interpolating the published data (see Ref. 12).

^c Errors are for counting statistics only (see Ref. 43).

Observed counting rates were converted to isotopic cross sections by applying the appropriate corrections and factors for half-life, counting efficiency, chemical yield, decay during bombardment, beam current, and target surface density. Conversion of isotopic fission cross sections to isobaric fission cross sections will be discussed in detail later in this communication.

III. EXPERIMENTAL RESULTS

The experimental data are summarized in Table I. Errors indicated include either the standard deviations for replicate bombardments, or reasonable estimates from the nature of the isotope involved (indicated parenthetically). The corrected cross sections represent the chain or isobaric yields. Mass-yield curves were constructed by the reflection method making certain assumptions (see Discussion) based on neutron emission as a function of fission product mass. These curves are illustrated in Figs. 2-5, and a summary of the characteristics of each mass distribution is given in Table II.

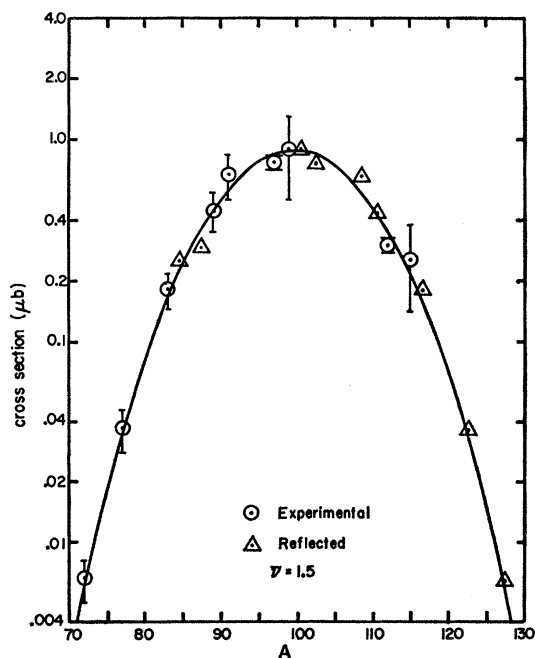


FIG. 4. Fission mass-yield curve for 33.5-MeV helium ions.

IV. DISCUSSION

A. Charge Distribution

It is beyond the scope of this communication to discuss in detail the applicable charge distribution functions in detail. In general, however, there seems to be rather widespread acceptance that such distributions are essentially Gaussian, centered about Z_p .²¹⁻²⁶ The three main charge-distribution postulates—minimum nuclear potential energy (MNPE), constant charge ratio (CCR), and equal charge displacement (ECD)—do, however, predict different values of Z_p in many mass regions of interest. Consequently, the corrections for independent yield and the conversion of isotopic cross sections into isobaric values may depend upon the particular postulate being used to estimate Z_p .

The data on independent and cumulative yields obtained in this research were used to help decide which of these three methods of estimating Z_p appeared to be applicable to this fission system.

Table III is a summary of the experimental fractional and cumulative chain yields for seven cases involving five isotopes and two energies. For the ECD calculation of Z_p , the mass equation of Green²⁷ was minimized with respect to charge for the desired mass. In the MNPE treatment, the fission fragment pair at the moment of scission was assumed to be represented by tangent spheres. Their internuclear distance was calculated by a Coulombic model from the average kinetic-energy release as experimentally determined for the He³-induced fission of Au¹⁹⁷.⁵ This method of calculation has been outlined by McHugh.²⁶

A comparison of the calculated and experimental yields and those obtained in this research is given in Table III and clearly indicates the good agreement using the CCR procedure. This does not necessarily rule out

²¹ L. E. Glendenin, C. D. Coryell, and R. A. Edwards, *The Fission Products*, National Nuclear Energy Series (McGraw-Hill Book Company, Inc., New York, 1951), p. 52.

²² R. H. Goeckermann and I. Perlman, *Phys. Rev.* **76**, 628 (1949).

²³ H. M. Blann, *Phys. Rev.* **123**, 1356 (1961).

²⁴ C. D. Coryell, M. Kaplan, and R. D. Fink, *Can. J. Chem.* **39**, 646 (1961).

²⁵ A. C. Wahl, R. L. Ferguson, D. R. Nethaway, D. W. Troutner, and K. Wolfsberg, *Phys. Rev.* **126**, 1112 (1962).

²⁶ J. A. McHugh, Ph.D. thesis, University of California, Berkeley, 1963 (unpublished).

²⁷ A. E. S. Green, *Phys. Rev.* **95**, 1006 (1954).

TABLE III. Experimental fraction and cumulative chain yields compared to theory.

Isotope	Energy (MeV)	f_{exp}	MNPE	$(Z-Z_p)$	CCR	$f_{\text{theory}}(Z-Z_p)$	ECD	$(Z-Z_p)$
^a Br ⁸²	40.0	0.065±0.013	0.122	1.18	0.050	1.52	0.088	1.32
^a Nb ⁹⁶	40.0	0.019±0.004	0.018	1.80	0.018	1.80	0.018	1.80
^a Br ⁸²	37.0	0.034±0.010	0.108	1.25	0.034	1.62	0.068	1.40
^b Zn ⁷²	40.0	0.96 ±0.07	0.81	0.10	0.96	0.60	0.88	0.30
^b Sr ⁹¹	40.0	1.04 ±0.12	0.96	0.70	1.00	0.90	0.98	0.80
^b Sb ¹²⁷	40.0	0.34 ±0.03	0.68	-0.20	0.32	-0.80	0.50	-0.50
^b Sb ¹²⁷	37.0	0.42 ±0.04	0.73	-0.10	0.38	-0.70	0.60	-0.32

^a Independent yield.^b Cumulative yield.

the possibility that some other MNPE combinations of internuclear distances and energy distributions at the moment of scission could also be made to agree with the observed data. The present yield data are not extensive enough to preclude the general applicability of the MNPE treatment to low- Z fission. However, it is clear that the very simple CCR postulate which does not introduce any arbitrary constants results in a very satisfactory procedure in the present case. A similar situation has now been observed in at least five other fission systems in our laboratories^{10,20,28-33} to data.

It is important to recognize the possibility of a cyclic argument that can be misleading in such an analysis. In many radiochemical fission studies, and certainly in the present case, the data are not so extensive to fix independently and directly the absolute yields of each isobaric chain. Consequently, the chain yield must be fixed before the fractional chain yield of any specific isobar of that chain can be determined. The chain yield is usually extrapolated from the mass yield curve, which, however, may depend upon the method used to convert isotopic yields to mass yields. There are a number of methods to resolve this difficulty, but most of them depend upon fixing the mass-yield curve in those mass regions where none of the corrections as given by any postulate for independent yields are large. As one proceeds to nuclides of lower masses and charges, the fission products are nearer to the line of stability, and, in general, correction factors become larger. However, the curves are changing into single symmetric curves, and fewer mass points are required to define the shape.

In the present research As⁷⁷, Br⁸³, Sr⁸⁹, Sr⁹¹, Mo⁹⁹, and Cd¹¹⁵ have isotopic yields which are representative of $\geq 99\%$ of the isobaric yields by any charge distribution analysis, and consequently the conclusions summarized in Table III appear to be valid. It should also be noted that the resulting total-fission cross section is in reason-

able agreement with values obtained directly by purely instrumental techniques, considering the magnitude of the numbers involved.

The independent yield data have been fitted to a Gaussian of the form as suggested by Wahl *et al.*,²⁵ (Fig. 6).

$$f = (c\pi)^{1/2} \exp[-(Z-Z_p)^2/c]. \quad (1)$$

The value of $c=0.95$ from this research is in good agreement with similar treatments for the thermal neutron fission of U^{235,25} for helium-ion-induced fission of Th^{232,26} and the fission of Au¹⁹⁷ with C¹² ions.²⁸ It is interesting to note the wide variation in mass of the fissioning nucleus to which the form of Eq. (1) seems to apply, particularly since the low-energy fission of U²³⁵ is largely an asym-

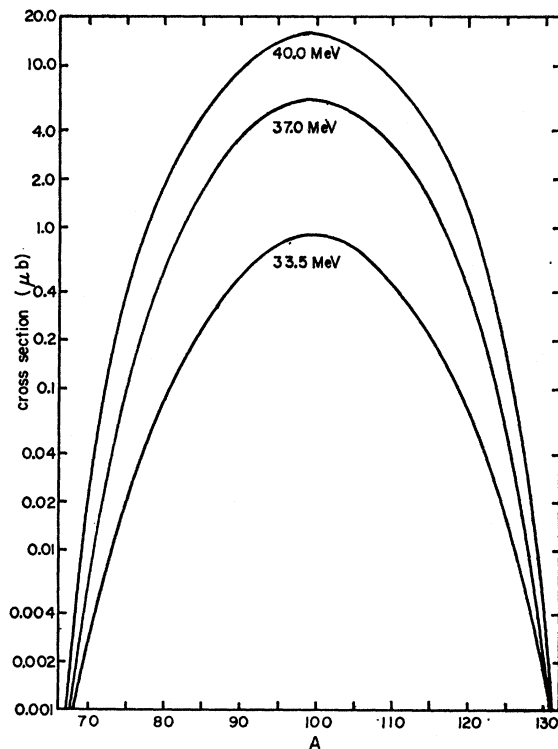


FIG. 5. Composite mass-yield curve for the fission of Au¹⁹⁷ with 40.0, 37.0, and 33.5-MeV helium ions.

²⁸ L. J. Colby and J. W. Cobble, Phys. Rev. **121**, 1410 (1961).

²⁹ R. Gunnink and J. W. Cobble, Phys. Rev. **115**, 1247 (1959).

³⁰ L. J. Colby, M. L. Shoaf, and J. W. Cobble, Phys. Rev. **121**, 1415 (1961).

³¹ J. A. Powers, Ph.D. thesis, Purdue University, January, 1962 (unpublished).

³² M. L. Shoaf, Ph.D. thesis, Purdue University, February, 1960 (unpublished).

³³ L. J. Colby, Ph.D. thesis, Purdue University, March, 1960 (unpublished).

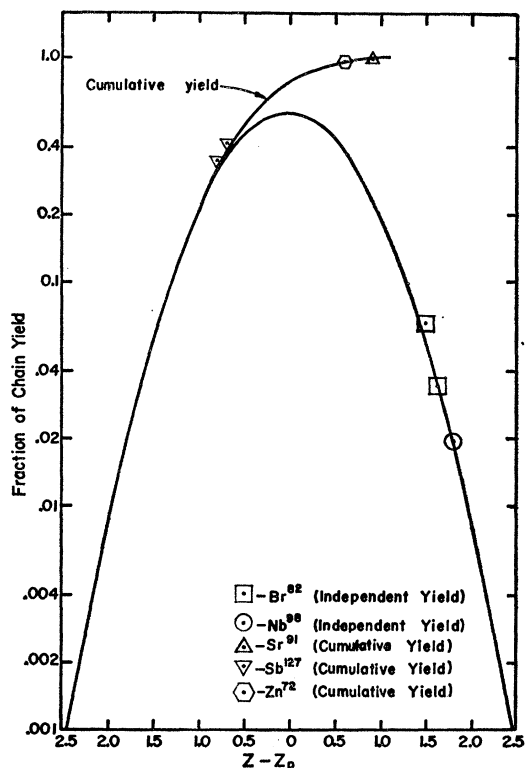


FIG. 6. Charge-distribution curve for the intermediate helium-ion-induced fission of gold using the constant-charge-ratio (CCR) postulate. Solid lines are from Eq. (1) for both independent and cumulative yields.

metric process, while the present case involves $\geq 99\%$ symmetric fission.

One other fact is worth noting concerning the independent yields of ${}_{41}\text{Nb}^{96}$. Similar Nb⁹⁶ data obtained by Wahl³⁴ and Pappas³⁵ from the thermal neutron fission of U²³⁵ were observed to be abnormally low. This fact might have been evidence for the existence of an unknown isomer of ${}_{41}\text{Nb}^{96}$, or alternatively, the complementary fission fragment may have favored a 42-50 division at the expense of a 41-51 division. In the present research, the complementary fission fragment of ${}_{41}\text{Nb}^{96}$ corresponds to a 41-38 proton division, resulting in strontium isotopes which are removed from any known closed proton shell. The present observations of normal Nb⁹⁶ yields at 40.0 MeV in this research (Fig. 2) indicate that, within experimental error, the closed proton shell effect postulated by Wahl is a more reasonable explanation of his Nb⁹⁶ yield.

B. Mass Distribution

The mass distributions obtained in this research indicate (Figs. 2-5) that the divisions of mass resulting from the fission of Tl²⁰¹ at excitation energies of 37.7,

³⁴ A. C. Wahl, *J. Inorg. Nucl. Chem.* **6**, 263 (1958).

³⁵ A. C. Pappas, *Proceedings of the First United Nations International Conference on the Peaceful Uses of Atomic Energy* (CERN, Geneva, 1955), Vol. 7, pp. 19-26.

34.8, and 31.3 MeV are essentially symmetric. Figure 7 indicates the variation of the total-chain cross sections with various mass ratios, (M_H/M_L), for 33.5-MeV helium ions. If any significant contribution from an asymmetric fission mode were present, an effect would be anticipated in this curve such as that seen in the data for the proton-induced fission of Bi²⁰⁹ previously reported by Sugihara *et al.*⁶ The Bi²⁰⁹ data demonstrated a small, but apparently real asymmetric contribution of approximately 0.3%. The change of shape in the vicinity of M_H/M_L about 1.7 is readily apparent for the bismuth data.

In addition to this analysis the absence of detectable amounts of Ni⁶⁶ and I¹³¹ fission products also precludes the presence of any significant amounts of asymmetric fission. It appears that the recently reported data of Neuzil¹⁸ which indicates the presence of a significant asymmetric mode in the Au¹⁹⁷(He⁴, f) reaction is best explained as a purity problem.

An interesting property of the mass distribution resulting from the Au¹⁹⁷(He⁴, f) reaction was the observed invariance of the full width at half-maximum of the mass distribution with change of excitation energy (Table II). Fairhall and Neuzil¹⁸ proposed a correlation for low- Z

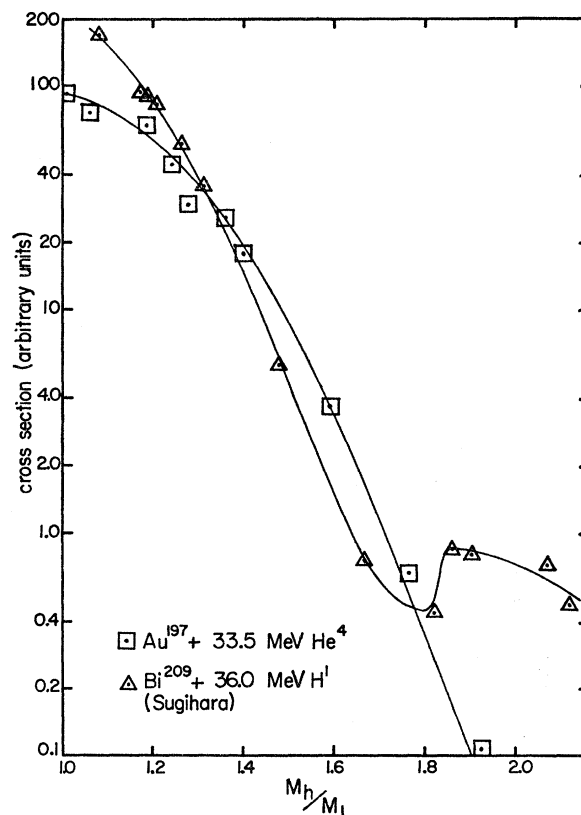


FIG. 7. Variation of isobaric cross sections with mass ratio for the 33.5-MeV helium-ion-induced fission of Au¹⁹⁷ and the 36.0-MeV proton-induced fission of Bi²⁰⁹ (see Ref. 6).

¹⁸ E. F. Neuzil and A. W. Fairhall, *Phys. Rev.* **129**, 2705 (1963).

symmetric fission which suggested that the full width at half-maximum of a symmetric mass distribution is given by

$$W_{1/2} = E_{SP}^* + 7 \quad (2)$$

where E_{SP}^* is the excitation energy of the compound nucleus at the saddle point and is taken to be:

$$E_{SP}^* = E_{CN}^* - E_f \quad (3)$$

E_{CN}^* is the excitation energy of the compound nucleus and E_f is the height of the fission barrier (in MeV). This correlation predicts a significant energy dependence for $W_{1/2}$ and does not appear to be consistent with the present research. At present, it is not at all clear why the mass distributions for this system are insensitive with respect to change of width with energy. The problem is under further investigation in these laboratories for even lower values of Z .

C. Neutron Emission

Approximate and average values for neutron emission were inferred by symmetry considerations of the mass-yield curve. The work of Terrell³⁷ on the neutron emission from U²³⁵, U²³³, and Cf²⁵² fission fragments suggests that this procedure is subject to error, since in these systems neutron emission was consistent with a step function and was not uniform for each fragment pair. However, more recent work of Britt and Whetstone³⁸ suggests that for the intermediate energy helium-ion-induced fission of U²³³, Th²³⁰, and Th²³² neutron emission occurs from the initial fission fragments approximately in proportion to the mass of the fragments. The approximately equal cross sections obtained in this research for Zn⁷² and Sb¹²⁷ indicate that the neutron emission from the fission fragments in the present system is indeed in approximate proportion to the fragment masses. While the use of the symmetry reflection method for determining $\bar{\nu}$ assumes that neutrons are emitted equally from each fragment, the error so introduced by using this method would only be of the order of 0.25 neutrons for the most nonsymmetric mass split studied (i.e., the 127-74 mass division) and is well within the experimental accuracy quoted of ± 0.5 neutrons. The values of $\bar{\nu}$ obtained in this research (Table II) are lower than those previously reported^{36,13} from more fragmentary data.

Britt *et al.*,⁵ have suggested that $\bar{\nu}$ can be calculated by considering the total energy balance involved in the fission process, and have estimated the excitation energy of the fission fragments from both the total energy release and the kinetic energy release. The excess energy is then considered dissipated in the emission of gamma radiation and neutrons. From this reasonable approach, values of $\bar{\nu}$ for the fission of Tl²⁰¹ have been calculated as 2.6, 2.2, and 1.9 for helium ion energies of 40.0, 37.0,

TABLE IV. Predicted values of $\bar{\nu}$ for the fission of various nuclides with 40.0 MeV helium ions.

Target	Compound nucleus	E_n^* (MeV) ^a	$\bar{\nu}$
Au ¹⁹⁷	Tl ²⁰¹	24.1	2.9
Pt ¹⁹⁴	Hg ¹⁹⁸	23.0	3.4
Pt ¹⁹⁶	Hg ¹⁹⁹	25.0	3.4
Pt ¹⁹⁶	Hg ²⁰⁰	25.6	3.2
Ir ¹⁹⁸	Au ¹⁹⁷	24.3	3.5
Os ¹⁸⁸	Pt ¹⁹²	17.4	2.5
Re ¹⁸⁷	Ir ¹⁹¹	21.2	3.1
Os ¹⁹⁰	Pt ¹⁹⁴	21.4	2.6
W ¹⁸⁴	Os ¹⁸⁸	22.7	2.8
Ta ¹⁸¹	Re ¹⁸⁵	20.0	2.9
Lu ¹⁷⁵	Ta ¹⁷⁹	18.2	2.6
Tm ¹⁶⁹	Lu ¹⁷³	18.1	2.8

^a E_n^* is the energy available for neutron emission.

and 33.5 MeV, respectively. These are in good agreement with those obtained experimentally (Table II).

Estimates of $\bar{\nu}$ for elements of even lower Z would be of interest to further radiochemical investigations of the fission process. Calculations similar to those of Britt *et al.*,⁵ have been carried out for elements as low as thulium excited with helium ions of 40.0 MeV. The results of these calculations are summarized in Table IV. In making the calculations, the average kinetic-energy release was obtained from a plot of experimental^{5,38,39} values of $\langle E_K \rangle$ versus Z^2/A . Mass distributions were assumed to be symmetric, and the total energy release was approximated as an equal mass division. The average neutron-binding energies are from the mass tables of Cameron.⁴⁰ Emitted neutrons were assumed to have a kinetic energy of 1.2 MeV⁵ and the average energy loss due to gamma-ray emission was set at 7.5 MeV,⁴¹⁻⁴³ and the neutron yields so calculated are probably accurate to ± 0.5 neutrons.

SUMMARY

1. The mass distribution resulting from the fission of Au¹⁹⁷ with 40.0-, 37.0-, and 33.5-MeV helium ions is symmetric; an upper limit for an asymmetric fission mode has been set as $\leq 0.03\%$.

2. The width at half-maximum of the mass distribution is essentially invariant over the energy interval studied (40.0-33.5 MeV).

3. Independent and cumulative yield data are most easily correlated with the constant-charge-ratio postulate.

4. The charge distribution about the most probable charge (Z_p) appears to be the same as that observed in heavy element fission and independent of the mode of fission.

³⁹ J. P. Unik and J. R. Huizenga, Phys. Rev. **134**, B90 (1964).

⁴⁰ A. G. W. Cameron, Chalk River Report CRP-690, AECL-433, 1957 (unpublished).

⁴¹ J. C. D. Milton and J. S. Fraser, Phys. Rev. Letters **7**, 67 (1961).

⁴² J. C. D. Milton and J. S. Fraser, Phys. Rev. **111**, 877 (1958).

⁴³ G. Raisbeck, this Laboratory (private communication).

³⁷ J. Terrell, Phys. Rev. **127**, 880 (1962).

³⁸ H. C. Britt and S. L. Whetstone, Phys. Rev. **133**, B603 (1964).

5. The number of prompt neutrons emitted is much smaller than had been previously reported.

6. The total radiochemical fission cross sections at excitation energies of 37.7, 34.8, and 31.4 MeV are found to be 191.3, 72.4, and 10.5 μb , respectively, and are in satisfactory agreement with previously published instrumental values.

ACKNOWLEDGMENTS

The authors wish to acknowledge the assistance and helpful discussions of C. Menninga, G. Raisbeck, and N. Wogman. The cooperation of M. Oselka and the staff of the Argonne National Laboratory 60-in. cyclotron is also greatly appreciated.

Polarization of Neutrons from the $D(d,n)\text{He}^3$ Reaction*

F. O. PURSER, JR., J. R. SAWERS, JR.,[†] AND R. L. WALTER

Duke University, Durham, North Carolina

(Received 24 May 1965)

Angular distributions of the polarization of neutrons from the $D(d,n)\text{He}^3$ reaction have been obtained at several deuteron energies from 1.9 to 3.7 MeV. The polarization at 45° c.m. ranged from -0.109 ± 0.013 at 1.9 MeV to -0.027 ± 0.015 at 3.7 MeV. Similar measurements presently available in the literature are reviewed and shown to fall into two conflicting sets, one of which is confirmed by the present measurement. All presently available angular distributions of the differential polarization have been fitted to the expansion $P_1(\theta)\sigma(\theta) = \sum_n a_n \sin 2n\theta$, and except for a previous measurement at 0.375 MeV, there is no evidence for terms of higher order than $n=2$ in the expansion. The coefficients of this expansion have been tabulated to permit interpolation of the differential polarization over deuteron energy and reaction angle.

INTRODUCTION

A LARGE number of experiments concerning the properties of the $D(d,n)\text{He}^3$ reaction have been performed since this reaction was recognized as a very useful source of monoenergetic neutrons. Theoretical analyses of the experimental results have followed several approaches. The cross-section data for energies above 5 MeV have been interpreted in terms of a stripping interaction with considerable success.^{1,2} Konopinski and Teller³ analyzed the cross-section data below 2 MeV using the method of partial waves and were able to conclude that a large spin-orbit interaction is involved. Wolfenstein⁴ noted that, if this is true, a polarization of the outgoing neutrons would be produced. Measurements of the predicted polarization ensued and now it is well established that indeed a polarization does exist. However, there is still some question as to the magnitude of the polarization. Below 2 MeV some of the discrepancies in the reported values can be attributed to the uncertainty in the properties of the polarization analyzers employed. Above 2 MeV the situation is not at all clear even though nine independent experiments have been conducted. Collecting all the polarization data reported for energies above 2 MeV and for reaction

angles near 45° c.m. on a single graph illustrates a curious feature. From a glance at the graph one is inclined to divide the values into two groups of points, through each of which a smooth curve varying monotonically with energy can be drawn. These two curves indicate polarizations which differ by more than 0.20 over most of the energy range from 2 to 11 MeV and which in fact differ in sign from about 4.5 to 8 MeV. Since the usefulness of the $D(d,n)\text{He}^3$ reaction as a source of partially polarized neutrons depends on an accurate knowledge of the polarization, and, since any suitable theoretical interpretation of this reaction will be expected to explain the value of the neutron polarization, it appeared desirable to perform an experiment which could aid in determining which, if either, of the sets of data are reproducible.

Although the Duke electrostatic accelerator is limited to energies below 4 MeV, it appeared that the disagreement in the 2- to 4-MeV region was large enough that an accurate experiment would test the reliability of much of the reported data. (In the interim between reporting our results⁵ prior to multiple-scattering calculations and submitting the present paper, two experiments by Bondarenko and Ot-Stanov⁶ and Babenko *et al.*⁷ also were conducted with this purpose in mind.)

* Work supported by the U. S. Atomic Energy Commission.

[†] National Science Foundation Fellow.

¹ W. W. Daehnick and J. M. Fowler, *Phys. Rev.* **111**, 1309 (1958).

² M. D. Goldberg and J. M. LeBlanc, *Phys. Rev.* **119**, 1992 (1960).

³ E. J. Konopinski and E. Teller, *Phys. Rev.* **73**, 822 (1948).

⁴ L. Wolfenstein, *Ann. Rev. Nucl. Sci.* **6**, 43 (1956).

⁵ F. O. Purser, J. R. Sawers, Jr., and R. L. Walter, *Bull. Am. Phys. Soc.* **8**, 320 (1963). J. R. Sawers, Jr., F. O. Purser, and R. L. Walter, *ibid.* **9**, 33 (1964).

⁶ I. I. Bondarenko and P. S. Ot-Stanov, *Zh. Eksperim. i Teor. Fiz.* **47**, 97 (1964) [English transl.: *Soviet Phys.—JETP* **20**, 67 (1965)].

⁷ N. P. Babenko, I. O. Konstantinov, A. P. Moskalev, and Yu. A. Nemilov, *Zh. Eksperim. i Teor. Fiz.* **47**, 767 (1964) [English transl.: *Soviet Phys.—JETP* **20**, 512 (1965)].