Variation in U^{235} Mass Yields at Neutron Energies Below 0.5 eV*

K. T. FALER AND R. L. TROMP
Atomic Energy Division, Phillips Petroleum Company, Idaho Falls, Idaho
(Received 8 April 1963)

Variations in the shape of the fission-product mass-yield distribution curve when U^{235} is irradiated with neutrons of various energies up to 0.5 eV were studied by radiochemical measurements of peak-to-valley ratios. A >30% increase in the relative amount of symmetric fission was found at the peak of the 0.29-eV resonance compared to the amount of symmetric fission produced with thermal neutrons. A decrease in the amount of symmetric fission was observed at increasing neutron energies where symmetric fission was 18% less common with epicadmium pile neutrons than with those at thermal energy. At neutron energies below 0.01 eV a sudden increase in the relative amount of symmetric fission was also found, even though there is no evidence from cross-section data for a resonance in this energy region. The results of this study are compared with those of other investigations at higher neutron energies. A discussion is presented regarding implications of mass-yield variations in the theory of fission through saddle-point states.

I. INTRODUCTION

CURRENT interest in the variation of fission-product mass-yield distributions with neutron energy in low-energy fission dates from papers by Bohr[1] and Wheeler.[2] In these papers it was suggested that at the moment when a fissioning nucleus crosses the saddle point in the potential energy barrier it has attained a shape which is axially symmetric and highly deformed. Associated with this shape one might expect to find nuclear energy levels of the rotational and vibrational types observed as the lower energy excited states of ordinary even-even nuclei[3] but with closer rotational level spacing due to large deformation of the fissioning nucleus. If this picture is correct, then the fissioning nucleus passes through a state of more or less well-understood character as it crosses the fission barrier, and the nature of the fission process might be expected to be dependent upon the character of the nuclear level at the saddle point and, thus, upon the spin state of the compound nucleus which is compatible with a given saddle-point configuration. In particular, it is believed that fission level width and relative amounts of symmetric and asymmetric fission should vary between level types. In the fission of U^{234}, the existence of such saddle-point states has been demonstrated by Lamphere.[4]

Variations in relative amounts of symmetric and asymmetric fission are sensitively observed by radiochemical determination of changes in peak-to-valley fission-product mass-yield ratios. Measurements of this variation as a function of incident neutron energy have been carried out by several investigators.[5–10] The existence of variations among different levels (fission resonances) has been well established, the peak-to-valley ratio change being as large as a factor of 5 in one case.[5]

Variations are most easily discussed and tabulated in terms of a normalized ratio R, where

$$R=\frac{A_{\rm asy}'/A_{\rm sym}'}{A_{\rm asy}/A_{\rm sym}}.$$

$A_{\rm asy}$ represents the saturation counting rate of a fission-product nuclide with a very high mass yield (usually Mo^{99}) and $A_{\rm sym}$ for a low-yield fission product produced in symmetric fission (such as Ag^{111} or Cd^{115m}). Primed counting rates indicate activity produced by neutrons at a given experimental energy and unprimed A's refer to values obtained with thermal neutrons. All data are thus expressed as values normalized to the peak-to-valley ratio obtained for the pair of nuclides at thermal energy, giving R at thermal energy a value of unity.

Although the results of measuring R at various energies were relatively straightforward for U^{233}, Pu^{239}, and Pu^{241}, the data reported for U^{235} fission have been quite confusing. It has been found that the difficulty in obtaining meaningful results has been due chiefly to the fact that R values for some resonances are greater than at thermal and some are less than at thermal.[8,10] Thus, normalizing all data to the value obtained with thermal neutrons has tended to de-emphasize values for which R was not very different from the thermal value.

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

[1] A. Bohr, in *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 2, p. 151.

[2] J. A. Wheeler, Physica **22**, 1103 (1956).

[3] B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat. Fys. Skrifter **1**, No. 8 (1959).

[4] R. W. Lamphere, Nucl. Phys. **38**, 561 (1962).

[5] R. B. Regier, W. H. Burgus, and R. L. Tromp, Phys. Rev. **113**, 1589 (1959); and R. B. Regier, W. H. Burgus, R. L. Tromp, and B. H. Sorensen, *ibid.* **119**, 2017 (1960).

[6] Los Alamos Radiochemistry Group, Phys. Rev. **107**, 325 (1957).

[7] R. Nasuhoglu, S. Raboy, G. R. Ringo, L. E. Glendenin, and E. P. Steinberg, Phys. Rev. **108**, 1522 (1957).

[8] G. A. Cowan, A. Turkevich, C. I. Brown, and the Los Alamos Radiochemistry Group, Phys. Rev. **122**, 1286 (1961); and G. A. Cowan, B. D. Bayhurst, and R. J. Prestwood, *ibid.* **130**, 2380 (1963).

[9] H. B. Levy, H. G. Hicks, W. E. Nervik, P. C. Stevenson, J. P. Niday, and J. C. Armstrong, Jr., Phys. Rev. **124**, 544 (1961).

[10] J. G. Cuninghame, G. P. Kitt, and E. R. Rae, Nucl. Phys. **27**, 154 (1961).

II. LOW-ENERGY FISSION CROSS SECTION OF U^{235}

The cross section for fission of U^{235} with neutrons of energies[11] <0.6 eV is shown as the solid curve in Fig. 1. The only resonance observed in this region peaks at an energy of 0.29 eV. The dashed lines in the figure show the cross section which is due to this resonance and the remaining "background" after it has been subtracted from the gross curves. This "background" is assumed to result from one or more negative energy resonances, that is, resonances due to levels in U^{236} which occur just below the energy of the compound U^{236} nucleus formed on capture of a zero-energy neutron by U^{235}. Both Shore and Sailor[12] and Vogt[13] have been able to describe the measured fission and total neutron cross section data in this energy region, even though they used different assumptions concerning the presence of negative energy resonances. In both approaches to fitting the data, the spins of the 0.29-eV resonance and of the 1.1-eV resonance (the next resonance observed in the cross-section measurements) are assumed to be the same. Since these spins are not known we will simply designate them as "J." In order to account for the low-energy "background" cross section, Vogt assumed the presence of a single resonance at -0.95 eV of spin J while Shore and Sailor assume two resonances, one at -1.45 eV and the other (weak) at -0.02 eV, both of which are levels of spin $J\pm1$.

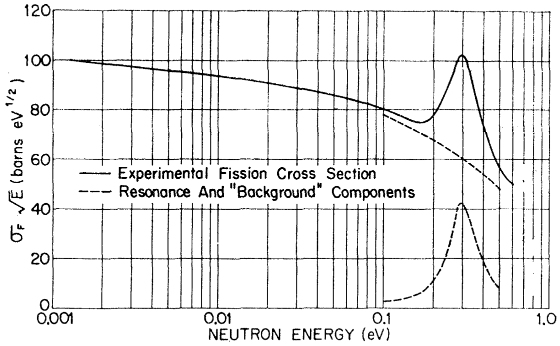

FIG. 1. Low-energy fission cross section of U^{235}.

III. EXPERIMENTAL PROCEDURE

A. Irradiations

Irradiations were carried out with samples of solid 95% U^{235} metal in three different neutron facilities. The thermal-energy neutrons were obtained at the face of the MTR thermal column with about 11 ft of graphite moderator between the reactor fuel and the samples. At this position the flux was 4.6×10^6 neutron sec^{-1} cm^{-2}. Similar irradiations were made using cadmium-covered uranium samples and results of these studies showed that $<1\%$ of the fissions were produced by epicadmium neutrons or by photofission.

Between 0.04 and 0.5 eV, irradiations were done with monoenergetic neutrons provided by the MTR beryllium-crystal neutron spectrometer. Samples were shielded in a borated-polyethylene box lined with cadmium along the entrance and exit slits and around the sample. The samples themselves were wrapped in thin aluminum foil. The number of fissions due to neutrons of energies other than that desired was determined by rocking the crystal out of the position in which the desired diffraction was produced. Backgrounds measured in this way varied from 1.0% at 0.04 eV to 3.5% at 0.5 eV.

Very low-energy neutrons were obtained by using the MTR "Cold Neutron" facility. Studies were carried out with beryllium-filtered neutrons of mean energy $\sim$0.004 eV and carbon-filtered neutrons of $\sim$0.002 eV. Fissions produced by epicadmium neutrons in this facility were below our limit of detection ($<0.08\%$). The borated-polyethylene sample box was used in these experiments also to reduce contributions from stray neutrons of undesirable energies. These neutron energies are all given in the laboratory system.

Most irradiations were carried out for periods of 2 to 3 days. The shortest one was 24 h and the longest was 104 h. During each irradiation the neutron intensities were essentially constant so that calculation of saturation values for the fission products was straightforward. Small variations in neutron fluxes was followed by noting variations in reactor power level, and most runs were also directly monitored by measuring transmission fluctuations with a BF_3 neutron counter placed behind the uranium sample. Corrections were made for small fluctuations which occurred during an irradiation, but if fluctuations amounted to more than a few percent the measurement was discontinued.

B. Chemical Separations

Following irradiations of the uranium samples, the metal was dissolved in dilute nitric acid containing silver and cadmium carriers and diluted to 100 ml. Duplicate 1-ml samples were removed and molybdenum carrier was added prior to Mo^{99} separation. The remaining solution was used for the determination of Ag^{111} and Cd^{115m} following their separations. Chemical procedures used were essentially those described by Kleinberg.[14] Samples were finally filtered onto 1-in. filter papers, covered with thin rubber hydrochloride film on standard aluminum mounts, and counting was begun about 24 h after completion of the irradiations.

[11] *Neutron Cross Sections*, compiled by D. J. Hughes and J. A. Harvey, Brookhaven National Laboratory Report BNL-325 (U. S. Government Printing Office, Washington, D. C., 1957 and 1958), Suppl. No. 1 and 2nd ed.

[12] F. J. Shore and V. L. Sailor, Phys. Rev. **112**, 191 (1958).

[13] E. Vogt, Phys. Rev. **112**, 203 (1958).

[14] J. Kleinberg *et al.*, Los Alamos Scientific Laboratory Report, LASL-1721, 1958 (unpublished), 2nd ed.

C. Counting Data

Since R values are determined by ratios of mass yields, it is not necessary to determine absolute disintegration rates. Variations in yield ratios can be established by computing saturation counting rates of samples all counted on a low-background beta counter in a standard, reproducible manner extrapolating the counting data back to the time at which the irradiation ended. Sample decay was followed for two weeks in all cases. If any significant deviation from expected decay rates was observed (attributed to radiochemical impurities) the measurement was discarded. Decay curves were obtained by least-squares analysis of the counting data. Data points were weighted by a factor proportional to the counting rate in order to give more weight to the early points of best statistical accuracy. The counting rate at time zero was computed as well as standard deviations. Of the values reported, standard deviations of all time zero counting rates were $<2\%$ except for three values which fell between 2 and 4%.

Over the long periods of irradiation there was no significant uncertainty in time deviation except that due to reactor-power fluctuation, and if this was significant the sample was discarded before chemical separations were performed. Counting rates were, of course, also corrected for chemical yields and for thickness of counting samples.

Following calculation of saturation counting rates for each set of samples, the difference between the duplicate Mo^{99} samples was checked. If these values were different by more than a few percent the data from that run were discarded. Two R values were then obtained, one for Ag^{111} and one for Cd^{115m}, by dividing their counting rates into the Mo^{99} counting rate and dividing this ratio by that measured at thermal energy. Each of these R values was treated statistically as an independent data point in computing the standard deviation among R values at each energy. Data points differing significantly from the mean R value were subjected to a "Studentized" test for extreme variation from the mean in order to eliminate points of accidental high variation. This resulted in the elimination of two points at 0.06 eV. Background neutron corrections were made at 0.04 eV, where second-order neutron diffraction had become a significant 5.2% of the beam.

IV. EXPERIMENTAL RESULTS

The experimental values obtained in this study are shown in Table I. The energies given for filtered neutrons from the cold neutron facility represent approximate mean energy based on expected filter transmission distributions. The thermal neutron energy is actually a distribution of energies from thermalization in graphite at room temperature. The energies in spectrometer irradiations are nearly monoenergetic due to the high resolution of the spectrometer. R values are the mean of four data points at each energy with their standard deviations calculated as discussed above. Qualitatively, an increase in R represents a fractional decrease in the amount of symmetric fission observed. The value given for epicadmium neutrons was obtained by Regier[15] and is the mean value based on 20 data points. All other values are from our study.

Table I. Peak-to-valley ratios for U^{235} normalized to thermal energy.

Incident neutron energy (eV)	% of σ_f due to 0.284-eV resonance[a]	Average R	Standard deviation
0.002	···	0.765	0.044
0.004	···	0.875	0.042
Thermal	···	1.000	0.02
0.04	···	0.880	0.047
0.06	···	0.783	0.021
0.10	4	0.796	0.059
0.25	28	0.730	0.022
0.28	42	0.746	0.026
0.33	37	0.708	0.022
0.50	14	0.841	0.033
0.70	···	0.752	0.074
Epi-Cd, Sm[b]		1.181	0.081

[a] Shore and Sailor (Ref. 12).
[b] R. R. Regier (private communication).

Figure 2 is a plot of R values versus neutron energy. Also shown is the fission cross section in this energy region. Vertical bars show standard deviations based on the four R values at each energy. The smooth curve is arbitrarily drawn through the data points without regard to theoretical implications. The value at thermal energy is shown with an arrow pointing upward since fission by neutrons in the distribution spectrum both above and below this value presumably give rise to lower R values, thus, depressing the value which would be measured with a monoenergetic 0.025-eV neutron beam.

V. DISCUSSION

It would be of interest to establish the maximum and minimum R values for U^{235} fission as has been done with the other nuclides studied. This study did not produce these numbers. About 45% of the fission cross section at 0.29 eV is due to the resonance itself, while the remainder of the "background" may be due to resonances of the same, different, or a mixture of both symmetry types. The minimum R values reported here of about 0.7 is lower than any previously reported, but if the "background" is due to resonances of the low-symmetric-fission type the R value for fission at this resonance could be considerably smaller. The R value of 1.2 for epicadmium neutrons is in good agreement with values obtained by other investigators,[6] and since the energy range covered is quite large and includes many resonances above 0.4 eV, it may be concluded that the R value for individual resonances of the less-symmetric-fission type is considerably larger than this

[15] R. B. Regier (private communication).

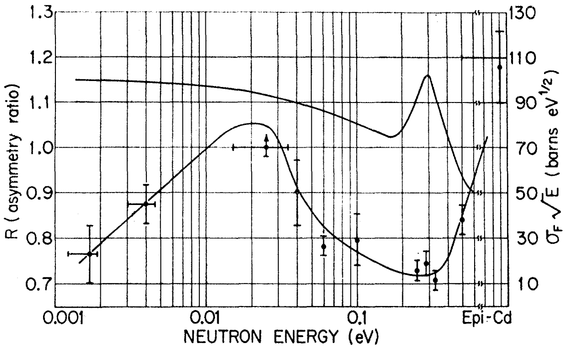

FIG. 2. Low-energy fission cross section and plot of "R" values for U^{235}.

value. The highest R value reported so far by other investigators is about 1.8.[8]

At neutron energies above 0.03 eV the general trend of the symmetry-variation curve in Fig. 2 might be expected, under the assumption that most of the "background" cross section is due to a level or levels with relatively less symmetric-type fission and that the resonance at 0.29 is of the type characterized by relatively more symmetric fission. It is quite surprising, however, to note the decrease in R as one goes to neutron energies below thermal. There is no indication in the fission cross-section data of the presence of a positive energy level at or below thermal. Even though an undiscovered level were hypothesized, its shape would have to be very unusual to account for the shape of the R value curve. It might be possible to explain this sudden dip in the curve on the basis of an interfererence effect between close-lying levels near zero energy.

A second observation of interest as seen from the data in Fig. 2 and Table I is that levels exist with R values both larger and smaller than the R value at thermal energy. On the assumption that symmetry type is dependent only on the spin of the level, neither of the published attempts to fit the cross-section data[12,13] are adequate to explain this observation since both assume that the cross section at thermal is due entirely to levels of one J value. A more plausible explanation of these observations is that the asymmetry value R is not dependent on spin alone. This is in agreement with the suggestion by Moore[16] that the character of the fission process may not be dependent on spin but rather on the fission channel; i.e., the K value (projection of the total angular momentum on the nuclear symmetry axis) of the nuclear level. Thus, levels of the same spin but belonging to different K bands might be expected to lead to fission of considerably different R values and fission-product angular distributions. It might then be possible to explain these low-energy data if the "background" fission cross section is due to resonances associated with different channels. The U^{234} study by Lamphere[4] indicates the presence of several K bands in the compound nucleus, each with different fission-product angular distributions. If fission symmetry as well as angular distribution is dependent on K, then not two but rather several R values would be expected in the fission of U^{234}. There is reason to believe that the s-wave neutron fission of U^{235} also takes place through several fission channels, i.e., through several bands with different K values. Indeed, in the recent report of Cowan *et al.*,[8] it was found that the R values of the observed resonances do not fall into only two mutually exclusive groups. Considerable evidence, then, points to channel rather than spin as the property on which relative amounts of symmetric fission depends.

[16] M. S. Moore, Trans. Am. Nucl. Soc. **5**, 21 (1962).

VI. HIGHER ENERGY STUDIES

From the nuclear explosion time-of-flight studies, Cowan *et al.*[8] have been able to determine R values for individual resonances between 7 and 61 eV. Their measured R values with low uncertainties range between (0.850 ± 4) and (1.84 ± 7)%. A majority of the observed resonances have a high R (16 are >1 and only 5 are <1). There also appears to be a bunching of levels into high and low R value types. Combining these results with those reported here, the observed range in R values for fission of U^{235} from zero to 60 eV is about 0.7 to 1.8.

Cuninghame *et al.*[10] have studied U^{235} fission with neutrons from 40 keV to 14 MeV. Up to 1 MeV their R values range from 0.72 to 1.74. At 14 MeV they report an R vaue of 0.008. Their study, in which neutrons of angular momenta>0 were able to form U^{236} compound nuclei in a large number of different spin states, again points up the inadequacy of the idea that the relative amount of symmetric fission is determined by spin alone.

ACKNOWLEDGMENTS

The authors wish to thank Dr. M. S. Moore, Dr. J. R. Smith, and Dr. R. R. Spencer for the use of the MTR crystal spectrometer and their assistance in using the instrument. Likewise, thanks are expressed to Dr. R. M. Brugger, Dr. R. E. Schmunk, and K. A. Strong for similar use of and assistance with the MTR cold neutron facility. Particular thanks are due to Dr. W. H. Burgus and Dr. M. S. Moore for their guidance and many helpful discussions during the course of this investigation.