Mass distribution in the neutron induced fission of ^{232}U

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Mass distribution in the neutron induced fission of ^{232}U has been studied. The distribution though predominantly asymmetric shows the existence of a small symmetric peak. The average fission product masses for the light and heavy wing are 92.7 and 137.5 mass units, respectively. The widths of the distribution at half and one tenth maxima are 13.4 and 21.0 mass units, respectively, and the peak to valley ratio of yields is about 480. An analysis of the factors influencing the probability of the existence of a symmetric peak and the variation of peak to valley ratio as a function of the proton and neutron numbers of the fissioning nucleus has shown that these variations can be qualitatively explained on the basis of differences in the heights of symmetric and asymmetric outer barriers.

[NUCLEAR REACTION, FISSION $^{232}U(n,f)$ obtained mass yield using radiochemical and gamma spectrometric techniques, general features examined in terms of symmetric and asymmetric outer barrier heights.]

INTRODUCTION

Asymmetry in the mass distribution in low-energy neutron-induced fission of heavy elements has not been fully understood. A third peak in the symmetric region of the mass distribution is even more difficult to explain quantitatively, though it was first observed by Turkevich and Niday[1] as early as 1951, followed by Fairhall and Jenson in fission at higher energy.[2,3] Turkevich and Niday explained the occurrence of a third symmetric peak on the basis of a two mode hypothesis though the factors responsible for two modes of fission were not well understood. In an attempt to understand the influence of several parameters such as the mass, charge, and energy on the shape of mass distribution, a number of investigations with low-energy neutrons were carried out and the results were communicated from this laboratory.[4,7] The existence of small symmetric peaks in the mass distribution of reactor neutron-induced fission of ^{227}Ac and ^{232}Th showed that the contribution of the symmetric mode does not depend on the charge of fissioning nucleus as strongly as proposed by Turkevich and Niday. In a continuation of efforts to understand this feature, the mass distribution of ^{232}U has been investigated since it was considered likely to throw some light in view of its higher proton to neutron ratio among all the uranium isotopes and thus might help reveal the influence of a number of neutrons of fissioning nucleus on the symmetric contribution. The observation of a small but definite peak in the symmetric region once again indicates that the symmetric contribution depends rather indirectly on the neutron and proton number in terms of the dependence of two modes of fission and on the difference in the height of the barrier leading to symmetric or asymmetric saddle point deformations. The only reported mass yield data on neutron-induced fission of ^{232}U are those of Kemmer *et al.*[8] and are limited to the high yield assymmetric mass region only. Therefore, a detailed investigation including the low yield symmetric mass region in the neutron-induced fission of ^{232}U fission was taken up. This paper presents the mass distribution in ^{232}U fission and a qualitative analysis of these features.

EXPERIMENTAL

^{232}U obtained from the Radiochemical Centre, Amersham, U.K., contained 99.14% ^{232}U, 0.85% ^{233}U, and 0.004% ^{234}U by weight percent. Thin targets of ^{232}U (~10 μg) and ^{235}U (~2 μg) were prepared by electrodeposition from an isopropyl alcohol medium.[11] These were covered with 0.0025 cm thick super-pure aluminum foils and wrapped together in aluminum foil and irradiated in the CIRUS reactor for 24 hours with a neutron flux of $\sim 1\times 10^{13}$ n/cm^2 sec. At the irradiation position the cadmium cutoff ratio for ^{232}U fission is about 0.5 determined in separate experiments. In other words, one out of every two fissions is caused by epicadmium neutrons.

The comparison method relative to ^{235}U fission was used to determine the mass yields employing the thin target-recoil-catcher technique. For these measurements ^{99}Mo was used as an internal standard. The yields of fission products in the high yield asymmetric mass region were determined by high resolution γ spectrometry and those in the low yield symmetric region were determined by radiochemical methods. The details of γ-spectrometric methods are given elsewhere.[10] For the low yield fission products the desired fission products were radiochemically purified and their decay followed on "Sugarman" type end window gas flow β proportional counters. A low background counter with the background of one cpm was used for counting samples of ^{111}Ag, ^{115}Cd, and ^{121}Sn.

CALCULATION OF YIELDS

In the comparison method, the yield of a fission product in ^{232}U fission ($Y_{x/2}$) is related to its yield in ^{235}U fission ($Y_{x/5}$) and the activities formed in the fission of two isotopes by Eq. (1) are

$$Y_{x/2} = Y_{x/5} \frac{Y_{Mo/2}}{Y_{Mo/5}} \frac{A_{Mo/5}}{A_{Mo/2}} \frac{A_{x/2}}{A_{x/5}}, \qquad (1)$$

where

$$\frac{A_{Mo/5}}{A_{Mo/2}} \frac{A_{x/2}}{A_{x/5}} \text{ is defined as } R_x .$$

Y denotes the fission yield and A the activity formed in fission and the subscripts 2 and 5 stand for ^{232}U and ^{235}U, respectively. The values of the fission yields in the fission of ^{235}U were taken from the recent compilation by Meek and Rider.[11] The fission yields relative to ^{99}Mo for ^{232}U were obtained assuming that the fission yield of ^{99}Mo is 6.14% (same as in the fission of ^{235}U). A correction of the order of (2–5%) was applied for the fission contribution from ^{233}U. These fission yields along with the fission yields of the complementary fission products were plotted and the two wings were separately normalized to 100%. The normalized fission yields along with those reported by Kemmer *et al.*[8] are provided in Table I. The neutron emission data in the case of ^{233}U from Apalin *et al.*[12] have been used for determining the complementary fission products. Figure 1 shows the variation of the fission yields as a function of the fission product mass.

ANALYSIS OF ERRORS IN FISSION YIELD MEASUREMENTS

The errors in the determination of fission yields are composed of two factors: (a) the errors in the R_x values and (b) the errors in a subsequent computation of the fission yields. In the γ spectrometric method probable errors are due to the Compton subtraction in calculating the peak area and counting statistics. The samples were prepared and followed for their decay such that the errors due to these factors were less than 2 to 3% (the details are given elsewhere[10]). As for the radiochemical determination of the R_x values, the errors are due to chemical yields of separated

TABLE I. Values of R_x and percent chain yield obtained in the present work along with values of Kemmer *et al.* (see text).

Fission product	Rx value	Percent chain yield present work	Kemmer *et al.*, Ref. 8
^{89}Sr	1.939 ± 0.040 [a]	6.01 ± 0.12	...
^{91}Sr	1.855 ± 0.052	7.12 ± 0.20	7.43
^{95}Zr	1.490 ± 0.014	6.30 ± 0.06	6.30
^{97}Zr	1.263 ± 0.017	4.94 ± 0.06	4.99
^{99}Mo	1.000	4.15	4.22
^{103}Ru	0.522 ± 0.010	1.10 ± 0.02	1.09
^{105}Ru	0.360 ± 0.040	0.24 ± 0.03	...
^{111}Ag	0.758 ± 0.048	0.009 ± 0.0006	...
^{115}Cd	2.420 ± 0.014	0.017 ± 0.0008	...
^{121}Sn	0.940 ± 0.080	0.008 ± 0.0007	...
^{125}Sn	5.600 ± 0.020	0.092 ± 0.003	...
^{127}Sb	5.733 ± 0.022	0.358 ± 0.018	...
^{131}I	2.030 ± 0.016	3.82 ± 0.03	4.13
^{132}I	1.717 ± 0.037	4.69 ± 0.10	4.84
^{133}Xe	1.260 ± 0.010	5.64 ± 0.05	5.63
^{135}Xe	1.250 ± 0.036	5.24 ± 0.08	6.40
^{140}Ba	1.718 ± 0.028	7.08 ± 0.11	7.04
^{141}Ce	1.865 ± 0.010	7.12 ± 0.44	6.61
^{143}Ce	1.353 ± 0.083	5.28 ± 0.32	4.68
^{147}Nd	0.917 ± 0.022	1.39 ± 0.03	1.15

[a] The ± on the number shows experimental scatter.

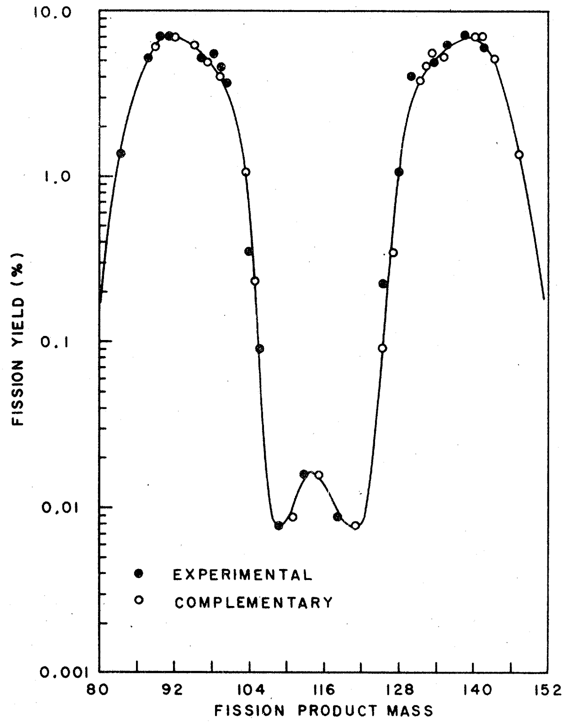

FIG. 1. The mass distribution in the neutron induced fission of ^{232}U.

fission products, counting statistics, and self-absorption. The errors due to correction for chemical yield and self-absorption were not more than 3% since the weights of the samples were of the same order and in the range of 20 to 60 mg. The errors due to counting statistics for the fission products in the high yield region were of the order of 2 to 3%. In the case of ^{111}Ag, ^{115}Cd, ^{121}Sn, and ^{125}Sn the counting rates were low, and though the decay of the samples was followed by using a low-background counter for long durations, the errors due to counting statistics were of the order of 5 to 7%. Thus, the errors in R_x values for ^{111}Ag, ^{115}Cd, ^{121}Sn, ^{125}Sn are of the order of 10 to 12% and in the rest of the cases, of the order of 5%. Errors in the subsequent computation of the fission yields are due to the assumed yields in the fission of ^{235}U and ^{233}U. These errors as given by Meek and Rider are of the order of 1 to 10%. Thus the overall errors in the high yield asymmetric region are expected to be of the order of 4 to 8% and that in the symmetric region of the order of 12 to 15%.

RESULTS AND DISCUSSION

The mass distribution in the neutron-induced fission of ^{232}U is predominantly asymmetric, but shows the existence of a small peak in the symmetric region as shown in Fig. 1. The average fission product masses of the light and heavy wings are 92.7 and 137.5 mass units, respectively. The width of the distribution at a half and a tenth of the maxima are 14.5 and 21.0 mass units, respectively. The peak to valley ratio is about 480. The difference between the sum of the average fission product masses and the mass of the compound nucleus (233) gives a value of 2.8 for the average number of neutrons emitted per fission.

The observation of a small but significant symmetric peak in the fission of ^{232}U leads to interesting observations on the general trends in the mass distribution in the low-energy fission of actinide isotopes. As mentioned earlier the existence of symmetric peaks in the fission of ^{227}Ac and ^{232}Th can be taken as an indication of a week dependence of symmetric fission on the charge of the fissioning nucleus. The presence of a symmetric peak in the ^{232}U fission further confirms this. Compared to ^{232}Th, ^{232}U has two more protons and two fewer neutrons and the excitation energies are very nearly the same (6.3 and 6.5 MeV, respectively) from calculations using the mass formula of Seeger and Howard.[15]

If the charge of the fissioning nucleus is an important factor in deciding the shape of the mass distribution as hypothesized earlier,[1] then in the case of ^{232}U a symmetric peak is unexpected; the existence of a symmetric peak indicates that the decreases in the neutron number more than compensates the effect of an increase in the proton number of the fissioning nucleus. This stronger dependence on neutron number can be examined only in a qualitative way. In low-energy fission, the relative contribution of symmetric and asymmetric components is expected to depend on the relative heights of symmetric (E_s) and asymmetric (E_a) outer barriers at the saddle point deformations.[14,15] Qualitatively, for a large value of (E_s-E_a) the symmetric contribution does not show up while below a certain value (E_s-E_a) it does. The values of E_s and E_a have been calculated for even-even nuclei by Moller and Nix[16]; in Figs. 2(a) and 2(b) the difference (E_s-E_a) as calculated by them is plotted as a function of the neutron and the proton number of the fissioning nucleus in the region of actinides and two distinct features emerge from these plots: First, (E_s-E_a) varies more sharply with the neutron number than the proton number of the fissioning nucleus. Second, the maxima in (E_s-E_a) occur around $N = 146$ and $Z = 92$. Figure 3 shows a plot of the peak to valley ratio in the low-energy fission of actinide nuclei as a function of the mass

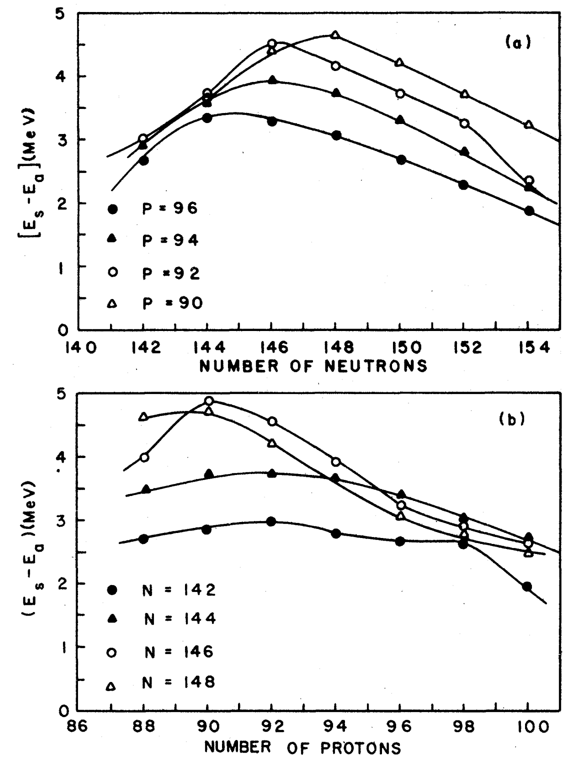

FIG. 2. Variation of $(E_s - E_a)$ as a function of (a) neutron and (b) proton number of the fissioning nucleus.

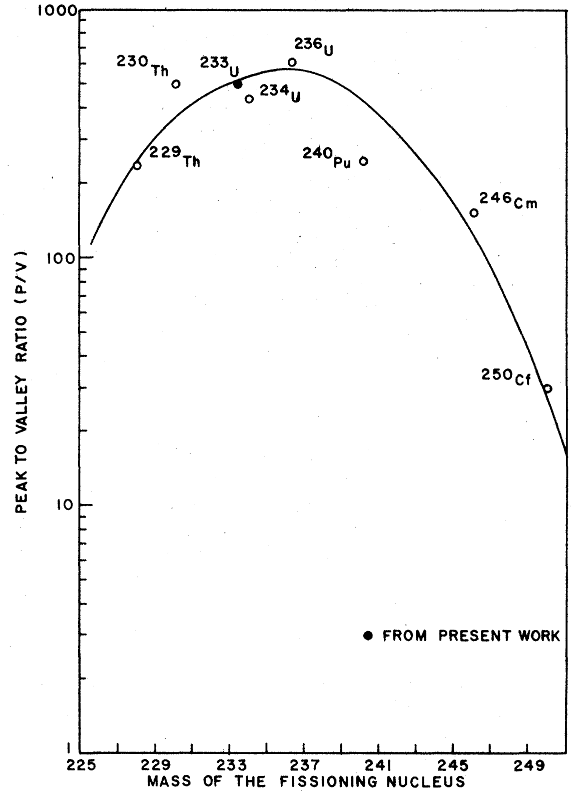

FIG. 3. Variation of P/V as a function of mass of the fissioning nucleus.

number of the fissioning nucleus. It is seen that the highest peak to valley ratio is observed around ^{235}U ($N = 144$, $Z = 92$) and on either side of this the peak to valley ratio decreases. The peak to valley ratio is expected to increase with an increasing value of $(E_s - E_a)$. Figures 2(a) and 2(b) show that $(E_s - E_a)$ decreases on either side of $A = 236$, and this qualitatively explains the maximum of the peak to valley ratio around $A = 236$ and its variation as a function of A plotted in Fig. 3. A similar dependence of the peak to valley ratio on $(E_s - E_a)$ was shown by Moller and Nix.[17]

The existence of the peak corresponding to the symmetric fission in both $^{232}_{90}Th^{142}$ and $^{232}_{92}U^{140}$ is qualitatively explained on the basis of variation of $(E_s - E_a)$ with neutron and proton number. The decrease in $(E_s - E_a)$ due to a decrease in the neutron number from 142 to 140 is more than the increase in $(E_s - E_a)$ due to an increase in the proton number from 90 to 92. Thus the overall $(E_s - E_a)$ in both the fissioning systems is nearly the same and this causes mass distributions in ^{232}U fission similar to that in ^{232}Th fission. Further work on the determination of mass yields in the region of thorium and uranium isotopes with excitation energies of the order of outer symmetric barrier heights would be interesting.

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