Neutron-induced fission of ^{232}Th near threshold

S. T. Lam, L. L. Yu, H. W. Fielding, W. K. Dawson, and G. C. Neilson
Nuclear Research Centre, The University of Alberta, Edmonton, Alberta, Canada T6G 2N5
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Fission of ^{232}Th was induced by a pulsed beam of monoenergetic fast neutrons. A heavy-ion surface barrier detector was used for the detection of fission fragments. Fragment mass was determined from measured fragment energy and fragment flight time. Fragment mass distribution and correlation of fragment kinetic energy with fragment mass were obtained for neutron incident energies of 1.6, 3.1, and 5.2 MeV. A strong dependence on shell energy is suggested from the increase of the most probable total kinetic energy of the fission fragments with increase in excitation energy of the compound nucleus.

[NUCLEAR REACTIONS Fission ^{232}Th(n,f), $E_n = 1.6$, 3.1, and 5.2 MeV natural target; measured fragment mass distribution and fragment kinetic energy versus fragment mass.]

I. INTRODUCTION

In fast neutron induced fission of ^{232}Th, resonance structures have been observed in the fission yield curve at neutron energies just above the threshold energy for fission. The fine structures at $E_n \sim 1.5$ and ~ 1.6 MeV were interpreted as due to the rotational bands built on vibrational levels in the third well of a triple hump potential.[1] Auchampaugh *et al.*[2] recently determined possible K values for the different rotational bands and their relative strengths from the angular dependence of the fission cross sections at resonance energies of $E_n \sim 1.4$, ~ 1.6, and ~ 1.7 MeV. If the fissioning of the compound nucleus at the resonance energies is from a triple hump potential and the fissioning at off-resonance energies is from a conventional double hump potential, the effects due to the difference of these two potentials may be inferred by observing the fission products at energies on and off the resonances.

In this investigation the fragment mass distributions and correlations of fragment kinetic energy with mass were measured for ^{232}Th fission at neutron energies of 5.2, 3.1, and 1.6 MeV. At neutron energies greater than 3 MeV, the excitation energies of the compound nucleus are sufficiently high above the fission barrier so that the fission yield curve in this energy region should be free of resonance structure. Our purpose was to investigate whether any anomaly could be observed in the measured fission-fragment mass distribution or fragment kinetic energies at the $E_n = 1.6$ MeV resonance as compared with those at $E_n = 5.2$ and 3.1 MeV.

II. EXPERIMENTAL METHOD

The experimental setup is described in Ref. 3. A pulsed neutron beam was produced by the ^{3}H(p,n)^{3}He reaction using a pulsed proton beam and a liquid nitrogen cooled tritium gas cell. The pulsed neutron beam was allowed to bombard a large spherical ^{232}ThF$_4$ target of 66 cm^2 in area and 490 μg/cm^2 in thickness. Fission fragments were detected by an Ortec heavy-ion silicon surface barrier detector situated at the center of curvature of the target. Fragment energy and fragment flight time were measured in coincidence for neutron energies of 5.2, 3.1, and 1.6 MeV. A total of about 5 000 fission events were recorded on magnetic tape for each neutron energy.

The procedure for data analysis is also described in Ref. 3. Fragment masses were deduced from fragment energies and fragment flight times. Fission-fragment mass distributions and correlations of fission-fragment kinetic energy versus fragment mass were obtained for the three neutron energies.

III. RESULTS AND DISCUSSION

A. Fission-fragment mass distribution

The experimental fragment mass distributions for neutron energies of 5.2, 3.1, and 1.6 MeV are shown in Fig. 1. The errors in the figure are statistical errors. Each point represents the fission yield over a mass interval of 2 u. The mass resolution was about 5 u, due mostly to the large variation of neutron flight times from the gas cell to the large thorium target and the short flight path for fission fragments. This geometry was necessary because of the low count rate of fission fragments. The fragment mass distribution at each neutron energy is normalized to 100%.

Table I gives the results of the present measurement. The peak-to-valley ratios in the mass distributions were obtained from the averages of the yield at the peak and valley regions. They are 129 ± 35, 63 ± 11, and 29 ± 3 for $E_n = 1.6$, 3.1, and 5.2 MeV, respectively. In comparison with a previous measurement,[4] which used two surface barrier detectors to detect the fission fragments in coincidence, the first two ratios are smaller than the previous values, while the third agrees within one standard deviation. Considering the very few counts in the valley region, the difference is acceptable, since they are at most three

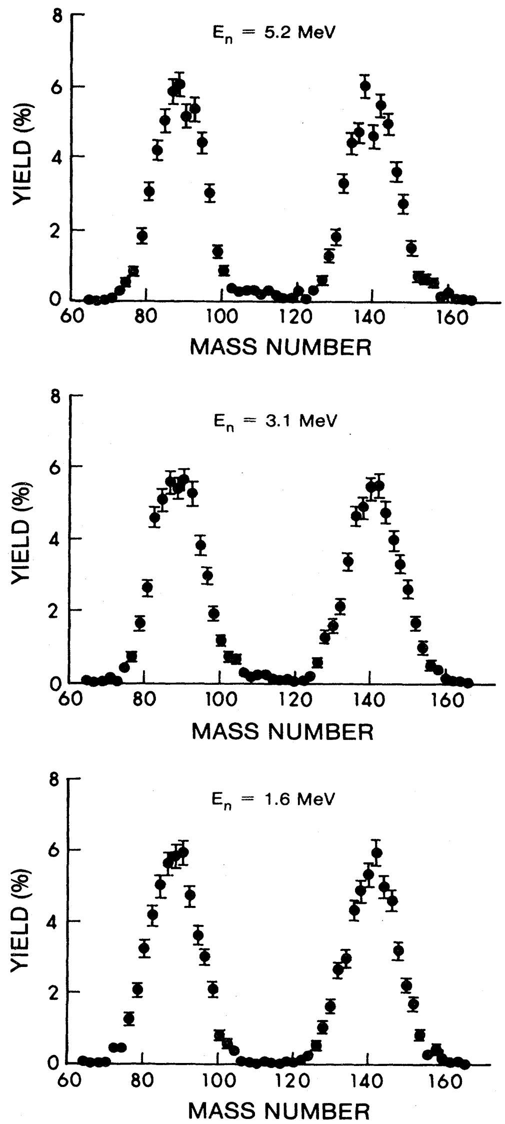

FIG. 1. The measured fragment mass distributions of fast neutron induced fission of ^{232}Th at $E_n = 5.2$, 3.1, and 1.6 MeV.

standard deviations away. The reduction of the peak-to-valley ratio with excitation energy has been explained as due to the weakening of shell effects with increasing excitation energy. Apart from the variation of the peak-to-valley ratio, there is no other significant difference in the shape of the measured mass distributions at these three neutron energies. The centroids of the light group and the centroids of the heavy group agree approximately within 1 u for the three neutron energies (see Table I). The average value for the light group is 89 u, and that for the heavy group is 141 u. These are post-neutron values. If neutron emission is taken into consideration, then the present value for the heavy group is slightly larger than that of Ref. 4; however, they agree with one another within the present systematic error of 5 u.

B. Fission fragment kinetic energy

The correlations of fragment kinetic energy versus fragment mass for the three neutron energies are shown in Fig. 2. The fragment kinetic energies shown in the figure are not corrected for the fragment energy loss of about 3 MeV in the target. Since the valley of the fragment mass distribution lies in the region of $A = 106-124$, there are large fluctuations in the valley region of Fig. 2 due to poor statistics. The mean values of the kinetic energies for the light and heavy fragment groups were determined from the data by excluding the events in the valley region. They are tabulated in Table I. These energies have been corrected for fragment energy loss in the target. From these values, the post-neutron most probable total kinetic energy, $\overline{\text{TKE}}$, of the fission fragments was determined. Using the expression as given in Ref. 5 and taking the average number of neutrons per fission to be 2.35, the pre-neutron $\overline{\text{TKE}}$ was calculated. They are all tabulated in Table I. The errors in the table are statistical errors only.

There appears in the literature a discrepancy in the reported values of pre-neutron $\overline{\text{TKE}}$ from the ^{232}Th(n,f) reaction. Figure 3 shows the $\overline{\text{TKE}}$ values as determined by the previous and present measurements. Surface barrier heavy-ion detectors were used in all cases to measure the energy of the fission fragment. A ^{252}Cf source was used in the energy calibration of the present measurement and the measurement of Holubarsch *et al.*,[6] while in the measurement of Trochon *et al.*[4] and Sergachev *et al.*,[7] thermal neutron fission of a ^{235}U target was used. The pulse-height defect of the heavy-ion detector was then corrected by a method similar to that described by Schmitt *et al.*[8]

The errors of $\overline{\text{TKE}}$ in Fig. 3 are statistical errors for the previous measurements, since only statistical errors were given with their values. The statistical errors were about 100 keV or smaller in the measurements of Holubarsch *et*

TABLE I. Fission-fragment masses and kinetic energies of the light and heavy groups of ^{232}Th(n, f) at $E_n = 1.6$, 3.1, and 5.2 MeV. Subscripts L and H denote, respectively, the light and heavy groups. The peak-to-valley ratio, P/V, of the mass distribution and the most probable total kinetic energy, $\overline{\text{TKE}}$, are also tabulated. The number of fission events at each neutron energy is about 5000. The errors in the table are due to counting statistics only.

E_n (MeV)	$\langle M_L \rangle$ (u)	$\langle M_H \rangle$ (u)	P/V	$\langle E_L \rangle$ (MeV)	$\langle E_H \rangle$ (MeV)	$\overline{\text{TKE}}$ (MeV) post	pre
$1.6^{+0.10}_{-0.13}$	88.4±0.2	141.5±0.2	129±35	95.76±0.02	63.10±0.08	158.86±0.08	160.63±0.08
3.1±0.15	89.3±0.2	141.5±0.2	63±11	96.86±0.02	64.17±0.08	161.03±0.08	162.83±0.08
5.2±0.25	89.0±0.2	140.3±0.2	29±3	98.08±0.02	65.40±0.10	163.48±0.10	165.30±0.10

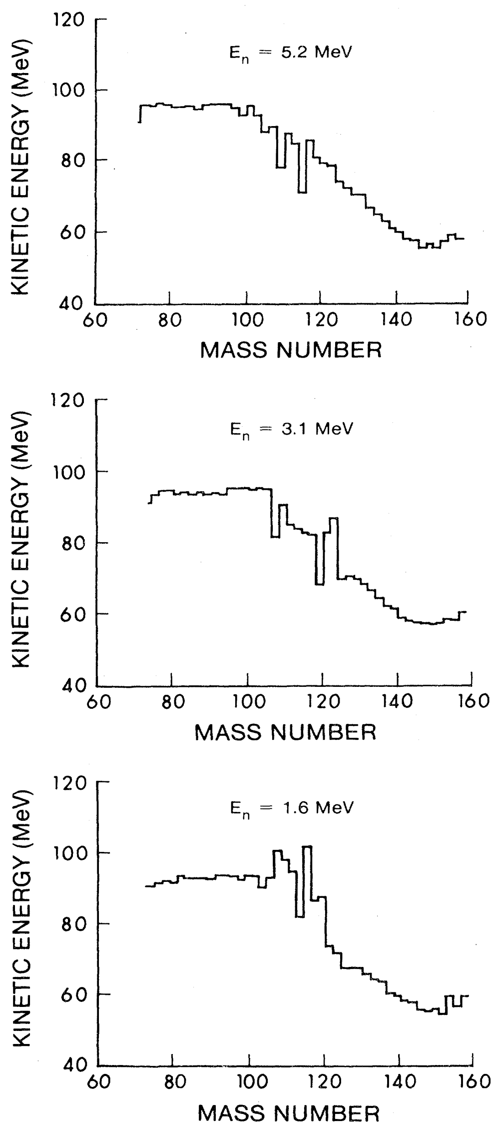

FIG. 2. Correlations of fission-fragment kinetic energy with fission-fragment mass from fast neutron induced fission of ^{232}Th at $E_n = 5.2$, 3.1, and 1.6 MeV.

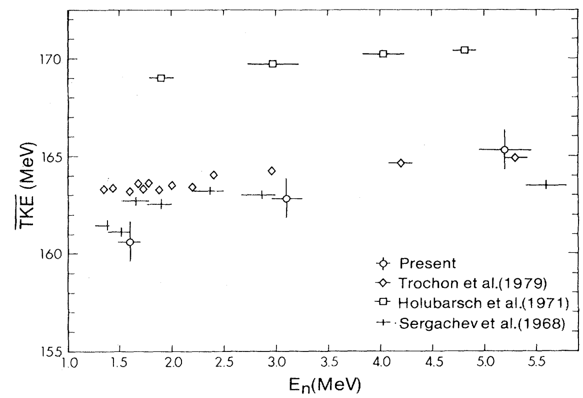

FIG. 3. Most probable total kinetic energy, $\overline{\mathrm{TKE}}$, of fission fragments from the $^{232}\mathrm{Th}(\mathrm{n},f)$ reaction as determined by different experimental groups at different neutron energies.

al. and Trochon *et al.*, while in the case of Sergachev *et al.* slightly larger errors were reported. The statistical errors in the present measurement were also about 100 keV. However, the main source of error in the energy determination was due to the correction of pulse-height defect of the heavy-ion detector. The systematic uncertainty in the present measurement was, therefore, estimated to be about 1 MeV. This is comparable to the uncertainties for ^{252}Cf and thermal neutron fission of ^{235}U, which are reported in the literature as 0.5 and 1.7 MeV, respectively. Hence the uncertainty of $\overline{\mathrm{TKE}}$ from the present measurement is shown in Fig. 3 as 1 MeV instead of the much smaller statistical error. The horizontal bars of the data points represent the neutron energy spread in the measurements.

In the measurement of Sergachev *et al.*, they found that the $\overline{\mathrm{TKE}}$ increases smoothly from $E_n = 1.65-5.6$ MeV and there was a sudden decrease in $\overline{\mathrm{TKE}}$ by about 1.5 MeV between $E_n = 1.65$ and 1.51 MeV. In the measurement of Holubarsch *et al.*, their $\overline{\mathrm{TKE}}$ also increased with neutron energy but their values were about 6 MeV higher than those of Sergachev *et al.* Holubarsch *et al.* suggested that the difference in $\overline{\mathrm{TKE}}$ from the two sets of measurements might be due to the use of different sources for energy calibration. Trochon *et al.* recently carried out another measurement. They assumed that the systematic error was small and ascribed the uncertainty of the $\overline{\mathrm{TKE}}$ as being due to counting statistics only. Because of the small uncertainty, they concluded that the $\overline{\mathrm{TKE}}$ at the resonances were smaller than those off the resonances by 0.3 to 0.4 MeV. They also pointed out a large discrepancy between their values and those of Sergachev *et al.* for E_n below 1.6 MeV. By converting the change of $\overline{\mathrm{TKE}}$ to the change of average number of prompt neutrons per fission $\overline{\gamma}_p$, Trochon *et al.* concluded that their result agreed with that of Caruana *et al.*,[9] who measured $\overline{\gamma}_p$ as a function of E_n. The sudden decrease of $\overline{\mathrm{TKE}}$ for E_n below 1.6 MeV in the measurement of Sergachev *et al.* gave too large an increase in $\overline{\gamma}_p$ when compared with the measurement of Caruana *et al.*

In the measurement of $\overline{\mathrm{TKE}}$, the corrections of the different effects are not all well known and exact corrections are difficult. It appears that both Sergachev *et al.* and Trochon *et al.* underestimated the uncertainty of their $\overline{\mathrm{TKE}}$ by considering statistical errors alone. The $\overline{\mathrm{TKE}}$ at $E_n = 1.51$ MeV is 161.20 ± 0.18 MeV in the measurement of Sergachev *et al.* and the $\overline{\mathrm{TKE}}$ at $E_n = 1.6$ MeV in the measurement of Trochon *et al.* is 163.22 ± 0.07 MeV. With a systematic error of 1 MeV as estimated in the present measurement, the present value of 160.6 ± 1.0 MeV for $E_n = 1.6$ MeV agrees within error with the measured value of Sergachev *et al.* at $E_n = 1.51$ MeV. At E_n higher than 1.6 MeV, the three sets of measurements from Sergachev *et al.*, Trochon *et al.*, and the present investigation give results agreeing almost within a systematic uncertainty of 1 MeV.

The spread of E_n was much smaller in the measurement of Trochon *et al.* Since the variation of $\overline{TKE}$ is smooth except around $E_n = 1.6$ MeV, the larger spread of E_n in the present measurement and the measurement of Sergachev *et al.* would not affect the value of $\overline{TKE}$ significantly at the higher neutron energies. However, the measurement around $E_n = 1.6$ MeV is in a critical region. The present value of $\overline{TKE}$ measured at $E_n = 1.6^{+0.10}_{-0.13}$ MeV indicates that it is possible to have a sudden decrease of about 2 ± 1 MeV in $\overline{TKE}$ between subthreshold fission and fission through open channels as observed by Sergachev *et al.*

The total kinetic energies of the different fission pairs were also determined for the present neutron energies of 1.6, 3.1, and 5.2 MeV. Since the mass resolution in the measurement was about 5 u, the fragment kinetic energies were averaged over this mass interval and the total kinetic energy, TKE, of each fission pair, with a mass resolution of 5 u, was obtained by adding the average kinetic energy of the light fragment group to that of the corresponding heavy fragment group. The total kinetic energies determined this way are tabulated in Table II. These are the post-neutron values. Since the number of neutrons associated with different mass splitting is not known, the pre-neutron TKE values are not calculated. The correction factor should be about the same as that for $\overline{TKE}$ in Table I and should be about 1%. The errors of TKE in Table II are the square roots of the sums of squares of deviations from the means, weighted by the number of fission events in the mass interval. Hence the errors become quite large on approaching the tails of the fragment mass distribution. The TKE at symmetric fission is not shown in Table II because of the very few events in this region. However, the data in the valley region do show the trend that the TKE for the symmetric pair is smaller than the TKE for the pair where the heavy fragment group is around the doubly magic number of $A = 132$.

In spite of the large errors in Table II, the TKE of individual fission pairs seems to increase with neutron energies, in agreement with the observation for $\overline{TKE}$. The trend of increase is shown in Fig. 4 for the three fission pairs starting with the pair (86—90 140—144) at the peak of the fragment mass distribution to the pair (96—100 130—134) which contains the masses in the vicinity of $A = 132$.

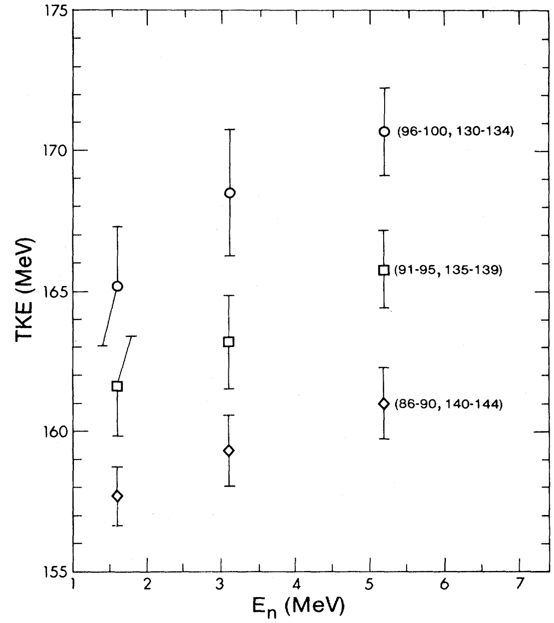

FIG. 4. A plot of the post-neutron total kinetic energy, TKE, versus the incident neutron energy, E_n, for the three fission pairs, (86—90 140—144), (91—95 135—139), and (96—100 130—134).

The increase of $\overline{TKE}$ with excitation energy in the fissioning of the compound nucleus, ^{233}Th, is quite unique, since the known $\overline{TKE}$ values for nuclei in this mass region either remain constant or decrease with excitation energy. This increase in $\overline{TKE}$ with excitation energy has perhaps been explained by the scission-point model of Wilkins *et al.*[10] In this description, the compound nucleus, ^{233}Th, must be at a high intrinsic temperature, so that the shell correction energy decreases as the excitation energy increases. This causes the potential energy surface to relax

TABLE II. A tabulation of the post-neutron total kinetic energies, TKE, of the different fission pairs for $E_n = 1.6$, 3.1, and 5.2 MeV. Since the fragment mass resolution is 5 u, the fragment masses are grouped into intervals of 5 u. The errors in the total kinetic energies are the square roots of the sums of squares of deviations from the means.

Fission pair (mass number)		Total kinetic energies (MeV) $E_n = 1.6$ MeV	$E_n = 3.1$ MeV	$E_n = 5.2$ MeV
71—75	155—159	155.4±4.7	156.4±4.5	158.1±6.7
76—80	150—154	153.8±2.0	157.8±1.0	158.4±2.3
81—85	145—149	155.5±1.2	156.8±0.6	157.9±1.4
86—90	140—144	157.7±1.1	159.3±1.3	161.0±1.3
91—95	135—139	161.6±1.8	163.2±1.7	165.8±1.4
96—100	130—134	165.2±2.1	168.5±2.3	170.7±1.7
101—105	125—129	166.3±3.7	171.3±3.2	171.9±2.6
106—110	120—124	175.2±12.7	179.1±13.7	174.0±5.7

towards that due to the liquid drop term. If there is a secondary minimum close to the lowest minimum in the potential energy surface at low excitation energy, an increase in excitation energy will increase the probability of fission via this secondary minimum. If this secondary minimum favors an overall smaller elongation of the compound nucleus as the excitation energy increases, the separation between the two nascent fragments at scission will become smaller. Hence the $\overline{\mathrm{TKE}}$ increases as the excitation energy increases. The comparatively large increase in $\overline{\mathrm{TKE}}$ observed in the fission of ^{233}Th suggests that the dependence on shell energy is stronger in ^{233}Th than any other nuclei in this mass region.

IV. CONCLUSION

In the present investigation of fast neutron induced fission of ^{232}Th, we do not observe any particular difference in the fragment mass distribution at $E_n = 1.6$ MeV as compared with those at $E_n = 3.1$ and 5.2 MeV.

Our measurement of the fragment kinetic energies at these three neutron energies supports the previous observation that the $\overline{\mathrm{TKE}}$ increases with excitation energy. Our value of $\overline{\mathrm{TKE}}$ at $E_n = 1.6$ MeV agrees with the subthreshold measurements of Sergachev *et al.*, indicating the possibility of a sudden decrease of $\overline{\mathrm{TKE}}$ by about 2 ± 1 MeV for subthreshold fission as compared with fission through open channels.

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