Measurement of the ^{232}Th(n,f) subthreshold and near-subthreshold cross section

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A measurement of the ^{232}Th(n,f) cross section for incident neutron energies between 100 eV and 1.6 MeV has been performed at the Oak Ridge National Laboratory electron linear accelerator. The weak subthreshold fission cross section found in this measurement confirms the model of a low first barrier in the triple-humped fission barrier which has been theoretically predicted for the (^{232}Th+n) system. However, the appearance of a series of plateaus in the near-threshold fission cross section region presents a challenge to current barrier calculations in the ^{233}Th compound nucleus.

[NUCLEAR REACTIONS ^{232}Th(n,f) measured cross section E=100 eV to 1.6 MeV.]

I. INTRODUCTION

In recent years considerable amounts of experimental evidence have been accumulated[1-4] showing the presence of structure in the ^{232}Th(n,f) cross section near threshold. For these structures to be interpreted as undamped vibrational levels in a double-humped fission barrier, the height, E_A, of the first barrier must be well above the neutron-binding energy, E_B, for ^{232}Th. Unexpectedly, double-humped barrier calculations, which are generally very successful in the case of heavy actinides,[5,6] yielded a value for E_A of around 4 MeV, which is lower than the neutron-binding energy of 4.786 MeV. This is the so-called thorium anomaly.[5,6]

The current explanation of the thorium anomaly is based on the fission barrier calculations of Möller[7] and of Möller and Nix.[8] These calculations, which take into account mass-asymmetry deformations, showed that for the light actinides ($N\sim142$) the fission barrier exhibits a second saddle-point split into two symmetric shallow minima at large nuclear deformations. This third well, located at a height $E_{\rm III}$ above the ground state of around 5.5 MeV,[9] can host vibrational levels which could explain the fission cross-section structures observed at neutron energies around 1.4, 1.6, and 1.7 MeV. On the basis of this picture of the fission barrier, one would expect a weak ^{232}Th(n,f) subthreshold cross section. This has been confirmed by measurements at the Rensselaer Intense Neutron Spectrometer in the neutron energy range between 1 eV and 100 keV by Block *et al.*,[10] who reported only a weak (7 μb) resonance around 2 keV.

Here we present the results of a high-resolution measurement of the ^{232}Th(n,f) cross section from 100 eV up to 1.6 MeV. The extended energy region covered in this experiment provides a comparison with previous high-energy measurements,[3,4] and also fills the gap between 0.1 and 0.6 MeV where there was no information on the ^{232}Th(n,f) cross section. The purpose of this measurement is twofold: (1) to look for broad structures in the near-threshold region at neutron energies lower than the previously observed structures[11] at 0.9 MeV, and (2) to probe the fission cross section for the possible existence of class II intermediate structures at neutron energies in the keV region.

II. EXPERIMENTAL DETAILS

A. General

The neutron-induced fission cross-section measurement for ^{232}Th was performed at the Oak Ridge National Laboratory electron linear accelerator (ORELA) (Refs. 12 and 13) using the standard water-cooled tantalum target for neutron production. Data were taken using the time-of-flight (TOF) technique, with a nominal time resolution of 0.4 ns/m. The experimental parameters for this measurement are given in Table I.

B. Fission chamber

The fission-fragment detector consisted of ten electronically independent sections. The first four and last four sections together contained a total of 5831 mg of thorium,

TABLE I. Experimental parameters.

Average electron beam power=4.5 kW
Repetition rate=150 pps
Pulse width=10 ns
Overlap filter=0.08-cm-thick cadmium plate
Flight-path length, $L=24.874$ m

Channel structure for the time-of-flight measurements

Number of channels (N_c)	Channel width, Δ_c (ns)	Energy range (keV)
1257	4	∞—127.9
5663	8	127.9—1.28
6795	16	1.28—0.128
2690	128	0.128—0.0128
4247	256	0.0128—0.00128

whereas the two central sections together contained a total of 649 mg of high purity ^{235}U.

Each thorium section consisted of eight deposits of approximately 1.2 mg/cm^2 thickness with a uranium contamination of less than 2 ppm. Each uranium section consisted of four deposits of 1 mg/cm^2 thickness. The plates, except for the end plates, were coated on both sides; the plate spacing was 0.32 cm. The chamber was filled with a mixture of 90% argon and 10% methane at a pressure of 2 atm.

C. Data acquisition

The ten outputs of the fission chamber were fed into an EG&G digital clock after appropriate signal "tagging" and pulse shaping. The TOF spectra for each section were separately stored in a SEL 810B on-line computer[13] and then combined after correction for differences in flight path among the various sections.

III. DATA REDUCTION

The count rates from the ^{232}Th and ^{235}U fission chambers were corrected for small time-independent backgrounds derived from the data below 80 eV, where the resonance structure of the ^{235}U fission cross section is well known and where the ^{232}Th fission cross section is expected to be essentially zero.

The ^{232}Th-to-^{235}U fission cross-section ratio was taken to be proportional to the net count-rate ratio and was normalized in the 1.4—1.5-MeV interval to the value 0.0519 obtained from the ENDF/B-V evaluations.[14] Finally, the cross-section ratio was converted to a ^{232}Th fission cross section using the ENDR/B-V evaluation of the ^{235}U fission cross section.[14]

Below 100 keV, the ^{235}U fission cross section has some structure which is not represented in detail in ENDF/B-V. To avoid introducing a spurious structure in the ^{232}Th data, the ratios of the ^{235}U count rate to the ^{235}U fission cross section were utilized to determine the energy dependence of the incident neutron beam. This energy dependence was found to be proportional to $E^{-0.81}$, in agreement with similar previous measurements.[15,16] The ^{232}Th fission cross section below 100 keV was then taken as proportional to the ^{232}Th count rate multiplied by $E^{0.81}$, with the proportionality constant fixed by the results above 100 keV.

A. Uncertainties

It is estimated that the systematic (rms) error amounts to about 5%. This error arises from the combination of uncertainties in normalization, background subtraction, and corrections for neutron scattering in the structural material of the fission chamber. Statistical errors range from about 60% at low energies to about 2% at high energy.

IV. RESULTS

Our results for the ^{232}Th(n,f) cross section in the energy interval between 0.6 and 1.3 MeV are shown in Figs. 1(a)—(c), together with the data of Ermagambetov *et al.*,[2] Blons *et al.*,[1] the more recent data of Behrens *et al.*,[11] and the ENDF/B-V evaluation.[14] Around 0.8 and 0.95 MeV, data of Behrens *et al.* appear to be systematically higher than our data, which is seen only as broad "should-

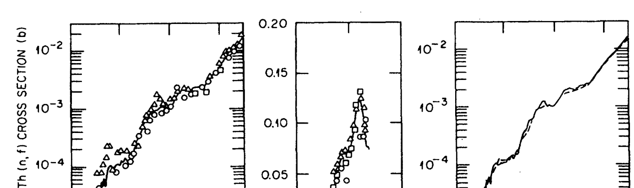

FIG. 1. The ^{232}Th(n,f) cross section obtained in this measurement (solid line), compared with the measurements of Ermagambetov *et al.* (○), Blons *et al.* (□), and Behrens *et al.* (△), over the neutron energy ranges from 0.6 to 1.3 MeV [part (a)] and from 1 to 2 MeV [part (b)]. A comparison of the present fission cross-section data (solid line) with results of the ENDF/B-V evaluation (dotted line) in the energy range between 0.6 and 1.3 MeV is shown in part (c).

TABLE II. The ^{232}Th(n,f) cross section σ_f averaged over decimal energy intervals between 0.1 and 1.6 MeV, and comparison with the results of Ref. 4.

Energy interval (MeV)	σ_f[a] (This work)	σ_f (Auchampaugh *et al.*)
0.1—0.2	3.6 ± 0.6 (μb)	
0.2—0.3	2.4 ± 0.5 (μb)	
0.3—0.4	5.4 ± 0.6 (μb)	
0.4—0.5	5.8 ± 0.6 (μb)	
0.5—0.6	5.9 ± 0.7 (μb)	
0.6—0.7	11.3 ± 1.0 (μb)	13.4 (μb)
0.7—0.8	75.6 ± 2.9 (μb)	87.2 (μb)
0.8—0.9	249 ± 6 (μb)	263.4 (μb)
0.9—1.0	919 ±16 (μb)	1085 (μb)
1.0—1.1	1.82± 0.03 (mb)	2.08 (mb)
1.1—1.2	3.16± 0.06 (mb)	3.38 (mb)
1.2—1.3	10.17± 0.18 (mb)	10.73 (mb)
1.3—1.4	37.70± 0.72 (mb)	42.10 (mb)
1.4—1.5	64.82± 1.29 (mb)	66.20 (mb)
1.5—1.6	98.96± 2.20 (mb)	99.0 (mb)

[a]The ^{232}Th fission cross section was obtained from the measured (^{232}Th/^{235}U) fission ratio, and the ENDF/B-V representation of the ^{235}U fission cross section. The errors shown are statistical errors only. It is estimated that the systematic errors amount to about 5%. These errors arise from uncertainties in normalization, in background subtraction, and corrections for neutron scattering in the structural material of the detector.

ers." This disagreement cannot be explained in terms of experimental resolution, as our resolution of 0.4 (ns/m) is slightly better than the resolution of 0.64 (ns/m) quoted by Behrens *et al.* However, the fission cross section peak at 1.6 MeV is well represented by all the data shown in Fig. 1(b).

The ^{232}Th(n,f) cross section averaged over 0.1-MeV intervals between 0.1 and 1.6 MeV is shown in Table II, together with the results of Auchampaugh *et al.*[4] These sets of data, which were taken with different neutron detectors and at different flight-path stations at the ORELA facility, are consistent within the combined uncertainties and agree to better than 10%. The low-energy results (between 100 eV and 0.1 MeV) averaged over selected energy intervals are given in Table III.

The neutron-energy interval between 1500 and 2500 eV is examined in some detail in Fig. 2 for possible evidence of class II intermediate structure in the ^{233}Th system. The locations of the *s*- and *p*-wave resonances in the (^{233}Th + n) system, given in a recent evaluation by Olsen,[17] are also shown in this figure. No statistically meaningful correlation was found between the observed fluctuations and the location of the (^{232}Th + n) levels. The average fission cross section in this interval is $\sigma_f = (0 \pm 6)$ μb.

TABLE III. The ^{232}Th(n,f) cross section averaged over selected energy intervals between 100 eV and 0.1 MeV.

Energy interval (keV)	σ_f[a] (μb)
0.1—1	−8.0± 0.0
1—5	3.0± 5.0
5—10	−2.0± 5.0
1—20	1.4± 2.8
10—20	2.3± 4.0
20—50	2.4± 2.4
50—100	2.2± 2.0
20—100	2.3± 2.0
0.1—100	2.0± 1.4

[a]Statistical errors only (on the basis of two standard deviations).

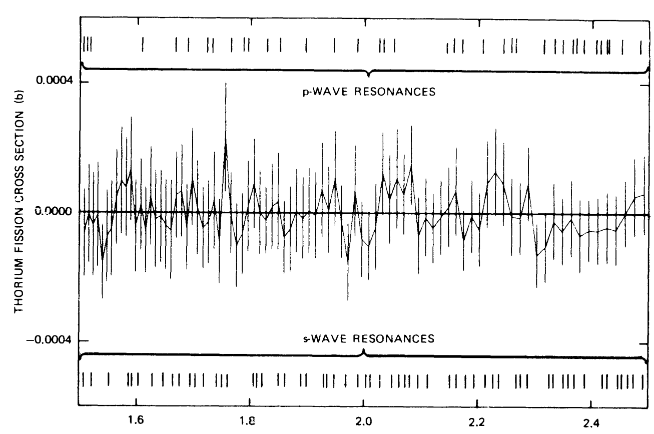

FIG. 2. The ^{232}Th(n,f) cross section in the neutron energy range between 1500 and 2500 eV. The data have been averaged over 15 channels to reduce statistical fluctuations. The error flags shown are statistical errors and correspond to two standard deviations. The vertical lines in the upper and lower regions in this figure indicate the locations of the *p*- and *s*-wave ^{233}Th levels, respectively.

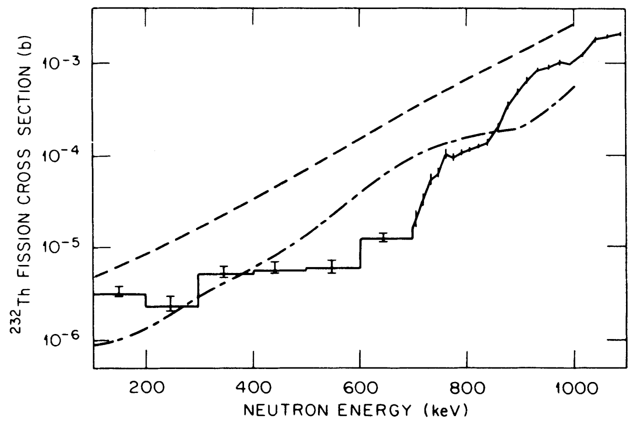

FIG. 3. The ^{232}Th(n,f) cross section in the energy range between 0.1 and 1.1 MeV. The data up to 0.7 MeV have been averaged over 0.1 MeV energy intervals. Above 0.7 MeV, the data have been averaged over two TOF channels. The error bars are statistical erros. The dashed-dot curve is a calculation based on the transition state parameters of Abou Yehia *et al.* In this calculation it was assumed that the reduced strength function for all even-l neutrons is 0.020 and for all odd-l neutrons is 0.038. The dashed curve is the result of a calculation with the COMNUC code utilizing a deformed optical model with global coefficients.

V. INTERPRETATION AND DISCUSSION OF RESULTS

The fission cross section in the neutron-energy range above 0.1 MeV is shown in Fig. 3, where the data have been averaged over 0.1-MeV energy intervals below 0.7 MeV and over two TOF channels in the neutron energy region above 0.7 MeV. Two theoretical calculations of the ^{232}Th(n,f) cross section are also shown in this figure. The dashed-dot curve is a calculation of the fission cross section which utilizes the transition state parameters of Abou Yehia *et al.*[18] to explain the structure in the fission cross section from 1 to 2 MeV. The dashed curve shows the fission cross section results derived from the code COMNUC,[19] which calculates the neutron transmission coefficients from a deformed optical model with global parameters.[20]

The measured ^{232}Th(n,f) cross section exhibits distinct plateau regions in the energy ranges 0.1—0.7, 0.76—0.82, 0.92—0.98, and 1.04—1.08 MeV. Aside from an apparent normalization difference, both theoretical calculations overestimate the fission cross section below 0.7 MeV and do not reproduce the observed plateau regions.

There are several ways to obtain a plateau in the fission cross section. The most obvious one is to assume a fully open ($T_f=1$) fission channel at the plateau region. However, calculations with $T_f=1$ produce cross sections many orders of magnitude greater than those observed in the measurement even for the neutron orbital angular momenta as large as $l=4$. In the spirit of assuming vibrational states in a shallow minimum, to obtain a "shoulder" in the cross section sitting on top of an exponentially increasing background requires that the vibrational state lie near the top of the lower of the two barriers that define the intermediate well. Otherwise, the vibrational state would appear as a narrow resonance in the cross section. Consequently, the lower barrier would have to lie just above the neutron binding energy E_B to explain, for instance, the wide plateau between 100 and 700 keV. However, the theoretical calculations by Möller and Nix[8] locate the first barrier below the neutron binding energy $E_B=4.786$ MeV and predict second and third barrer heights of about 1.5 MeV above the binding energy.

Another possibility is to assume that these plateau regions are damped vibrational states in a considerably deeper well, such as the one which might be associated with the second minimum of a triple-humped barrier, and that the first barrier is higher than current predictions. There is some evidence which corroborates this hypothesis. As pointed out in the paper by Auchampaugh *et al.*,[4] there is an apparent 15-keV-wide modulation in the cross section over the gross structure between 1 and 2 MeV that could be explained by a higher first barrier than the one predicted theoretically. In this instance, the class II states associated with the second minimum will not be completely mixed with the class I states, so that the cross section for the vibrational state in the third minimum would be modulated by the coupling of the class II and class I states. Clearly, the issue of the first barrier location with respect to the binding energy must be addressed as a compromise between the reason for the appearance of the cross-section structure mentioned above and the weak subthreshold fission cross section found in this experiment. It might be that the present data provide the first real test of the current barrier calculations related to the thorium anomaly.

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