

Subthreshold photofission of ^{235}U and ^{232}Th

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Photofission cross sections for ^{232}Th , and ^{236}U have been measured in the energy range from 3.25 to 5.75 MeV and for ^{234}U and ^{235}U at 3.5 MeV. The cross sections change by over seven orders of magnitude for this energy range. Cross section shapes are significantly different for different isotopes indicating a strong sensitivity to fission barrier parameters.

[NUCLEAR REACTIONS, FISSION Barrier shape; fission; nuclear reactions; photofission bremsstrahlung; threshold; tract counting.]

INTRODUCTION

Photofission has long been recognized as a useful method for deriving information on parameters of the fission barrier.¹ However, the method as usually applied, along with the general approach using neutrons and charged particle beams, provides information on the barriers only within about 1 MeV of the top of the barrier. Aside from isomer half-lives and excitation energies, there is very little information available on the shape of the lower portion of the barrier. However, in 1975 it was pointed out that photofission cross section measurements could be made to energies below 3 MeV and that the cross section should in general exhibit a shelf which appeared as the γ -ray energy was reduced to the point where the probability for fission through the outer fission barrier equals the γ -decay probability.² This shelf and other features of the photofission cross section shape could be used to derive valuable information on the lower portion of the fission barrier. These proposals were verified in an accompanying paper³ in which the photofission cross section of ^{238}U was measured and analyzed in the 2.75 to 5.75 MeV range. Results were also published⁴ on ^{236}U , and ^{237}Np verifying the existence of the shelf.

In this paper we report new measurements on ^{232}Th and ^{235}U in the 3.2–5.75 MeV range and of ^{234}U and ^{236}U at 3.5 MeV. We view these measurements as a step in the process of building a data base for a large number of nuclei which can then be the basis for a systematic study using methods similar to those already applied³ to ^{238}U . The purpose of this paper is to report the results and call attention to their sensitivity to the barrier parameters as reflected in the different photofission cross section shapes.

EQUIPMENT

The experimental technique is the same as that described in Ref. 3 and therefore will be described

only briefly here. Foils of the isotopes ^{232}Th , ^{234}U , ^{235}U , and ^{236}U were alternated with mica to form a sandwich. The sandwich was irradiated with bremsstrahlung from electrons accelerated with the Argonne National Laboratory electron linac in the 4 to 6 MeV range and with the NBS electrostatic accelerator at 3.5 and 4 MeV. The mica was etched and tracks counted to determine the induced fission yield. This yield, in fissions per milligram of fissile material per Coulomb of electron charge incident on the photon radiator, is given in the second and third column of Table I for ^{235}U and ^{232}Th . The uncertainty is that associated with the statistics of this track counting and the background determination of the geological spontaneous fission tracks in the mica. The two measurements at 4 MeV were averaged together for the cross section calculation. For ^{235}U the average was taken to be 46 ± 3 . For ^{232}Th the numbers are significantly different so that the error was taken to overlap the two values. At 4 MeV the yield therefore was taken as 2.1 ± 1.4 fission/C mg for ^{232}Th .

As described in Ref. 3 we used the bremsstrahlung spectrum from Dickinson and Lent⁵ to unfold the cross section from the yield. The yield in

TABLE I. A summary of experimental fission yields.

| Electron energy (MeV) | Fissions/C mg ^{235}U | Fissions/C mg ^{232}Th |
|-----------------------|--------------------------------|---------------------------------|
| 3.5 ^a | 0.42 ± 0.16 | 0.0047 ± 0.08 |
| 4.0 ^a | 41 ± 3 | 0.72 ± 0.35 |
| 4.0 ^b | 51 ± 4 | 3.5 ± 1.1 |
| 4.5 ^b | 117 ± 12 | 8.1 ± 5.0 |
| 5.0 ^b | 578 ± 92 | 106 ± 75 |
| 5.5 ^b | $4 \pm 0.3 \times 10^4$ | 2604 ± 511 |
| 6.0 ^b | $1.9 \pm 0.038 \times 10^6$ | |

^a Measured at NBS.

^b Measured at ANL.

TABLE II. Bremsstrahlung spectrum.

| E_γ (MeV) | $\varphi(E_\gamma, 6)^a$ | $\varphi(E_\gamma, 5.5)$ | $\varphi(E_\gamma, 5)$ | $\varphi(E_\gamma, 4.5)$ | $\varphi(E_\gamma, 4)$ | $\varphi(E_\gamma, 3.5)$ |
|---------------------|--------------------------|--------------------------|------------------------|--------------------------|------------------------|--------------------------|
| 5.75 | 1.2×10^{-3} | 1.5×10^{-3} | | | | |
| 5.25 | 3.9 | 1.5×10^{-3} | | | | |
| 4.75 | 7.6 | 5.7 | 1.5×10^{-3} | | | |
| 4.25 | | 8.9 | 5.0 | 1.6×10^{-3} | | |
| 3.75 | | | 9.3 | 4.6 | 1.8×10^{-3} | |
| 3.25 | | | | 8.6 | 4.8 | 1.7×10^{-3} |

^aPhotons in a half-MeV interval per 6 MeV electron.

photons per 0.5 MeV interval per electron is given in Table II for electron energy intervals of 0.5 MeV. This quantity is defined as $\varphi(E_\gamma, E_c)$ where E_γ and E_c are the γ -ray and electron energy, respectively, in MeV. The fission yield is given by the expression

$$Y_f(E_c) = n \int_0^{E_c} \varphi(E_\gamma, E_c) \sigma(E_\gamma) dE_\gamma,$$

where n is the number of atoms/cm² of the sample. By transforming the integral to a sum in half-MeV increments and using the data of Table II, a set of equations is established. These equations were solved by first calculating $\sigma(3.25 \text{ MeV})$ from $Y_f(3.5 \text{ MeV})$ and working to higher electron energy and propagating the uncertainties through the set of equations.

The resulting cross sections are given in Table III. The uncertainties listed include only the uncertainties in track counting propagated through the unfolding process. In Ref. 3 systematic uncertainties are estimated to be perhaps as large as a factor of 2.

For ^{234}U and ^{236}U the yield was too low to obtain accurate results and the error uncertainty in both cases exceeds the derived cross section value. Nevertheless the cross section at 3.5 MeV was found to be $0.3 \pm 2.5 \times 10^{-10} \text{ b}$ for ^{234}U and $1.2 \pm 1.7 \times 10^{-10} \text{ b}$ for ^{236}U . The upper limit to the cross section is perhaps useful.

TABLE III. Photofission cross sections for ^{232}Th and ^{235}U .

| Photon energy (MeV) | Cross section ^{235}U (b) | Cross section ^{232}Th (b) |
|---------------------|------------------------------------|-------------------------------------|
| 3.25 | $6.8 \pm 2.6 \times 10^{-11}$ | $7.4 \pm 129 \times 10^{-13}$ |
| 3.75 | $170 \pm 47 \times 10^{-11}$ | $3.3 \pm 2.2 \times 10^{-10}$ |
| 4.25 | $4.9 \pm 248 \times 10^{-11}$ | $4.4 \pm 10.7 \times 10^{-10}$ |
| 4.75 | $6.3 \pm 1.9 \times 10^{-8}$ | $1.6 \pm 1.4 \times 10^{-8}$ |
| 5.25 | $1.1 \pm 0.08 \times 10^{-5}$ | $4.2 \pm 1.1 \times 10^{-7}$ |
| 5.75 | $5.6 \pm 0.11 \times 10^{-4}$ | $8.6 \pm 0.25 \times 10^{-4}$ |

The cross sections of Table III are presented in Fig. 1 as a function of photon energy along with the results for ^{238}U from Ref. 4. Several features of the curves are of interest. First note that all cross sections are nearly the same at 5.75 MeV. However, the ^{232}Th decreases very rapidly so that over most of the energy range this cross section is two orders of magnitude below ^{238}U . For ^{235}U the cross section starts out nearly parallel to ^{238}U but then falls away and appears to show a peak at 3.75 MeV. All three curves bend sharply over

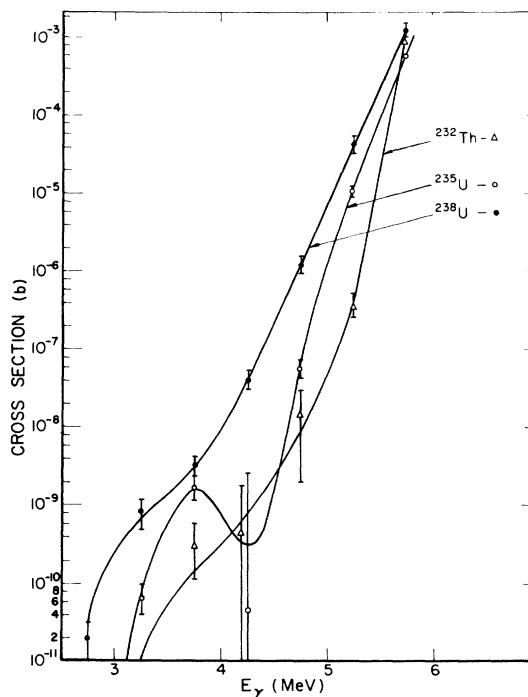


FIG. 1. The photofission cross sections of ^{232}Th , ^{235}U , and ^{238}U . The uncertainty shown includes only track counting statistics propagated through the unfolding process. A systematic uncertainty in the cross section scale might be as large as a factor of 2. The lines through the data are included to guide the eye.

again at the lower energies suggesting fission isomers near 2.7, 3.0, and 3.1 MeV for ^{238}U , ^{235}U , and ^{232}Th .

The principle limitation in extending this technique to other nuclei lies in the spontaneous fission half-life of the sample. Using the present technique we estimate that about half of the heavy isotopes now available in milligram or larger sizes can be studied. Most of the others probably can

be studied by the use of powerful pulsed beams produced by induction linac methods.⁶ Even without the induction accelerator, it appears feasible to establish a rather wide data base using this approach to study the fission barrier.

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¹See for example B. S. Bhandari and I. C. Nascimento, Nucl. Sci. Eng. 60, 19 (1976) or V. R. Huizenga, Nucl. Tech. 13, 20 (1972).

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⁶J. W. Beal, N. C. Christofilos, and R. E. Hester, IEEE Trans. Nucl. Sci. NS-16, 294 (1969).