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# THE FISSION CROSS SECTION OF $^{239}$Th AND $^{232}$Th RELATIVE TO $^{235}$U



by

J. W. Meadows



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# THE FISSION CROSS SECTION OF $^{230}$Th AND $^{232}$Th RELATIVE TO $^{235}$U

J. W. Meadows
Argonne National Laboratory
9700 South Cass Avenue
Argonne, Illinois 60439, USA

The fission cross sections of $^{230}$Th and $^{232}$Th have been measured relative to $^{235}$U from near threshold to near 10 MeV. The weights of the thorium samples were determined by isotopic dilution. The weight of the uranium deposit was based on specific activity measurements of a $^{234}$U-$^{235}$U mixture and low geometry alpha counting. Corrections were made for thermal background, loss of fragments in the deposits, neutron scattering in the detector assembly, sample geometry, sample composition and the spectrum of the neutron source. Generally the systematic errors were ~1%. The combined systematic and statistical errors were typically 1.5%.

(NUCLEAR REACTIONS, FISSION $^{230}$Th(n,f), $^{232}$Th(n,f), cross sections relative to $^{235}$U(n,f), threshold to 10 MeV.)

## Introduction

Several years ago a measurement program was started at the ANL Fast Neutron Generator with the goal of measuring the fission cross sections of the longer-lived fissionable isotopes relative to $^{235}$U with accuracies of the order of 1 percent. This paper reports the results for $^{230}$Th and $^{232}$Th. These isotopes have received little attention in the past, particularly when compared to the more common uranium isotopes. Inspection of the CSISRS file[1] does show a number of data sets for the $^{232}$Th fission cross section including two recent ratio measurements [2,3] but there are only four references[4-7] to $^{230}$Th and the agreement between the measurements is poor.

## Experimental Method

The experimental method and techniques for making those measurements have been described previously.[8] Briefly, the relative fission rates of pairs of $^{235}$U and $^{230}$Th or $^{232}$Th samples were measured by placing them back-to-back in a double ionization chamber positioned near a monoenergetic neutron source. Neutrons with energies above 5 MeV were produced by the D(d,n)$^{3}$He reaction. Lower energy neutrons were produced by the $^{7}$Li(p,n)$^{7}$Be reaction. A pulsed and bunched charged-particle beam was used to obtain a pulsed-neutron source and fast timing techniques selected those fissions suitably correlated with the neutron pulse. The data was stored in two time-by-pulse-height arrays for later inspection when time windows and pulse-height discrimination levels were set. Windows were set over the time peak and also to one side to permit simultaneous measurement of any background due to thermal and epi-thermal room-return neutrons. The data from the individual detectors was corrected for the discriminator bias and background. The fission ratios were corrected for neutrons from secondary source reactions such as $^{7}$Li(p,pn)$^{6}$Li, $^{7}$Li(p,n)$^{7}$Be$^{*}$ and D(d,pn)D and for scattering from the ionization chamber and the neutron source assembly. The differences in the geometry and the neutron transmission for the two samples were eliminated by making a second measurement with the sample positions interchanged. Although this greatly reduced the effect of momentum transfer, the fission ratios were explicitly corrected for residual momentum effects as well as deposit thickness and fission fragment angular distribution effects. The background produced by (d,n) reactions with the gas target assembly was measured with an empty gas cell before and after each measurement with a full cell.

Table I. The Isotopic Analyses of the Samples in Weight Percent.

| Material | U-235 | | | Th-230 | Th-232 | |
|---|---|---|---|---|---|---|
| Sample No. / Isotope | 5-2 | 3 | SST-5 | (51 thru 54) | (31 thru 36) | 1 |
| 230 | ——— | ——— | ——— | 99.516 | 0.383 | 0.0 |
| 232 | ——— | ——— | ——— | 0.484 | 99.617 | 100.0 |
| 234 | 1.028 | 0.028 | 0.852 | ——— | ——— | ——— |
| 235 | 98.403 | 99.856 | 93.244 | ——— | ——— | ——— |
| 236 | 0.447 | 0.062 | 0.334 | ——— | ——— | ——— |
| 238 | 0.122 | 0.054 | 5.570 | ——— | ——— | ——— |
| Area Density mg Th/U/cm$^2$ | 0.164 | 0.077 | 0.081 | 0.075 - 0.155 | 0.131 - 0.569 | 0.127 |

## Samples

Most of the samples were prepared by molecular depositon using a method similar to that described in Ref. 9. Thin layers of uranium or thorium, 2.54 cm in diameter, were deposited onto 0.25 mm thick stainless or 0.13 mm thick platinium plates. One of the uranium deposits, SST5, was made by vacuum evaporation of $UF_4$ and one of the thorium samples, Th-232-1, was made by evaporation of thorium metal. The isotopic analyses of all materials used is given in Table I.

Absolute fission-cross-section ratios were measured using $^{235}U$ sample 5-2. The weight used for this deposit, based on specific activity measurements of the original material and on low geometry alpha counting, was 0.8316 mg. This deposit was one of those used in a recent series of measurements where reference $^{235}U$ deposits from several laboratories were intercompared.[10] The weight based on the intercomparison was 0.833 mg. The other uranium deposits were only used in shape measurements.

The weights of most of the thorium samples were based on specific activity measurements. Enough $^{230}Th$ was added to the $^{232}Th$ material to give a convenient alpha count rate and a number of $^{230}Th$ and $^{232}Th$ deposits were prepared. The alpha count rate of each deposit was measured in a low geometry alpha counter whose geometry factor was calculable from precisely measured dimensions. The weights of some of the deposits were determined by the isotopic dilution technique. The $^{230}Th$ half-life obtained from the $^{232}Th$ samples was 75600 ± 330 y. For the $^{230}Th$ samples it was 75900 ± 300 y. The best previous measurement gave 75200 ± 1600 y.[11] The weight of Th-232-1 was not determined accurately and it was only used for shape measurements.

The exact composition of these deposits is uncertain. The approximate composition of Th-232-1 is probably fairly well represented by ThO. The other thorium deposits should be $ThO_2$, however the actual weight of the deposits are ~6 percent larger than the weights of $ThO_2$ calculated from the isotopic dilution results. These deposits were heated to 700 deg. C but that was not high enough to ensure that they would not be slightly hydroscopic so this excess weight suggests that their approximate composition might be closer to $ThO_2 \cdot H_2O$. The composition of uranium samples 5-2 and 3 is unknown but may be some uranyl oxide hydrate.

## Deposit Thickness Correction

In principal the deposit thickness correction can be calculated from the deposit composition and fission fragment range and angular distribution data. In practice, as discussed above, the deposit composition may not be known very well and if the correction is significant it is best to measure it for a particular set of samples.

The effects of deposit thickness, fission fragment angular distribution and momentum transfer cannot be separated. Approximate formulas for estimating losses in thin deposits are developed in Ref. 12. In the measurements reported here the ratio of the fission rates of two deposits was obtained by averaging two measurements, one made with the thorium deposit facing the neutron direction and one made with the uranium deposit in that position. Under these conditions the first order momentum terms cancel and the average specific fission rate of the thorium deposit relative to the reference deposit can be approximated by

$$S_{obs} = S_{true}(1 + C_u - C_{Th})$$

$$C = P(\pi/2)\left(\frac{t}{2R} + \frac{R\gamma^2}{4t}\right) \qquad t > R\gamma$$

where t is the deposit thickness, R is the fragment range, $\gamma$ is approximately the ratio of the incident neutron momentum to the total momentum of the fission fragments and $P(\pi/2)$ is the normalized fragment angular distribution function evaluated at $\pi/2$. $P(\theta)$ can be expressed as a power series in $\cos^2\theta$ and for this purpose one term is usually sufficient although additional terms may be necessary in limited energy regions. Coefficients were obtained from data given in Refs. 13 thru 15. For $t > 1.5R\gamma$ the $\gamma^2$ term may be dropped and the effective value of R can be obtained from the slope. Specific fission rates were measured for a series of thorium deposits ranging from 0.13 to 0.57 mg Th/cm$^2$ with the results shown in Fig. 1. The effective value of R was determined to be 5.1 ± 0.3 mg Th/cm$^2$.

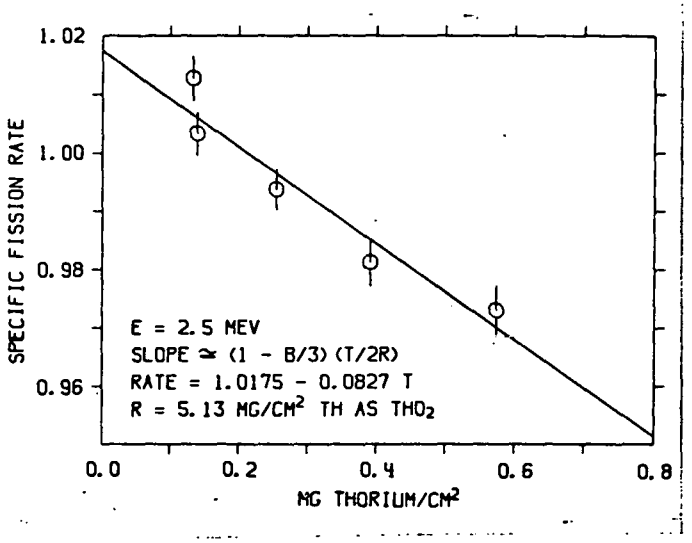


Fig. 1. The dependence of the specific fission rate on deposit thickness for thorium deposits.

Previous measurements for the uranium deposits gave 4.1 mg U/cm$^2$ for the effective range in sample 3 and 5-2 and 4.7 mg U/cm$^2$ for SST5.[10]

Calculated ranges based on the data of Alexander and Gazdik[16] and Niday[17] were generally in good agreement. The calculated value for $UF_4$ was 4.7 mg U/cm$^2$ in very good agreement with the measurement. In the previous section it was suggested that the composition of the thorium deposits might be approximately represented as $ThO_2 \cdot H_2O$. The calculated range for this material, 5.2 mg Th/cm$^2$, is in good agreement with the measured value.

## Results

The results are shown in Figs. 2 and 3. Points are plotted at average energies and all energy resolutions are full widths at half height of the calculated energy distributions. Error and resolution bars are shown where they exceed the size of the data symbol.

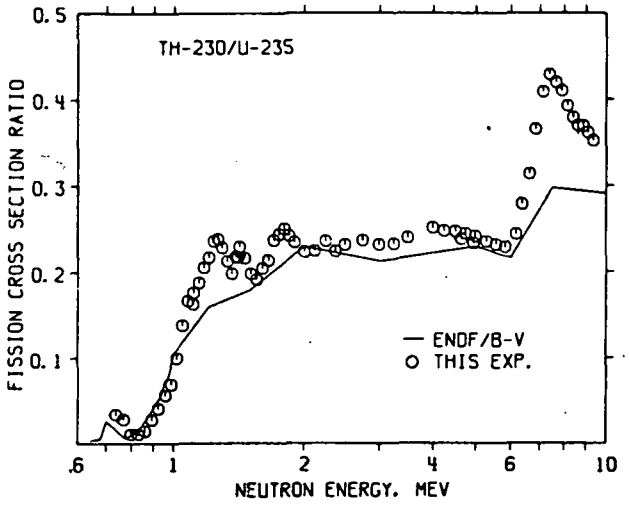


Fig. 2. The $^{230}$Th to $^{235}$U fission cross section ratios.

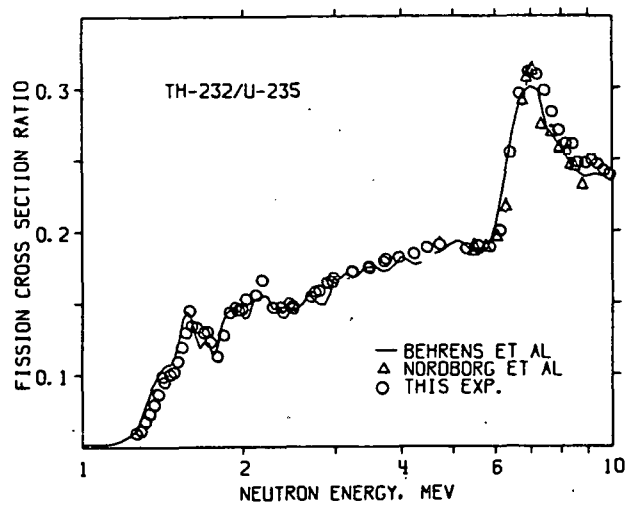


Fig. 3. The $^{232}$Th to $^{235}$U fission cross section ratios.

Figure 4 shows the combined error, excluding the fission counting errors, as a function of energy for the $^{232}$Th measurement. The error for the $^{230}$Th measurement is similar. There is a basic error of ~0.8 percent due to the uncertainty in the ratio of the sample masses and an additional 0.2 percent error in the correction for neutron scattering in the detector structure. The large errors near 5 MeV and 10 MeV are caused by the uncertainties in the correction for the lithium and deuteron break-up reactions. The correction for the $^{7}$Li(p,n)$^{7}$Be* reaction is significant in the threshold region but its yield relative to the ground state reaction is well known so the error from this source does not exceed 0.5 percent.

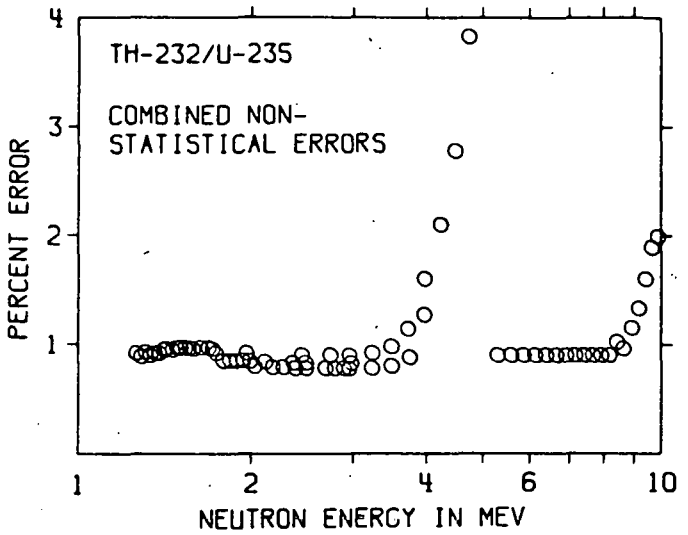


Fig. 4. The energy dependence of the principal systematic errors for the $^{232}$Th to $^{235}$U fission cross section ratios.

The counting errors are typically 1 percent except in the threshold region where the thorium fission rate was low. Points used for normalization have counting errors of ~0.5 percent. This error includes the counting statistics and also any uncertainty in the extrapolation to account for fission below the bias level. This correction ranged from ~0.5 to ~5 percent depending on the deposit thickness. In making the extrapolation the counts per channel at the bias level were assumed to remain constant down to zero pulse height. It is doubtful if this is true and such a procedure may introduce a systematic error when only one of the fission detectors is considered. However this procedure was used for both detectors and also used in determining the deposit thickness correction so any systematic effect should largely cancel in the ratio. Other corrections and errors are discussed in Ref. 8.

The results for $^{230}$Th are shown in Fig. 2. There have been few other measurements for this isotope. Muir et al.[4] made measurements relative to $^{239}$Pu using a bomb shot as a pulsed source. When the data from the present experiment is converted to cross sections using ENDF/B-V the agreement is fairly good. The other measurements[5-7] are in very poor agreement.

The results for $^{232}$Th are shown in Fig. 3 and compared with two recent ratio measurements. The agreement with the data of Behrens et al.[2] is fair. The two data sets differ by a consistent 3 to 4 percent in magnitude and by ~20 keV in energy in the threshold region. The agreement with the data of Nordborg et al.[3] is fairly good up to ~7 MeV but above the energy the difference is ~5 percent.

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