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MEASUREMENT OF THE INTEGRAL CAPTURE AND FISSION CROSS SECTIONS FOR ^{232}TH IN THE CFRMF

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The recent evaluation of the cross-section data bases for ^{232}Th capture and fission emphasized significant normalization discrepancies between the available differential data. To help resolve the normalization discrepancies, the capture and fission integral cross sections have been measured for ^{232}Th in the fast neutron zone of the Coupled Fast Reactivity Measurements Facility (CFRMF). The cross sections are derived from the radiometric determination of the saturation reaction rates for fission and capture based on the Ge(Li) spectrometric measurement of the absolute gamma emission rates of the 537-keV and 1596-keV lines in the ^{140}Ba - ^{140}La decay and the 311.9-keV line in the ^{233}Pa decay. For capture and fission, respectively, the measured integral cross sections are 291 mb ± 3% and 19.6 mb ± 5%. The ratios of the integral cross sections computed with ENDF/B-IV thorium cross sections and the CFRMF neutron spectrum to the above values are 0.99 for capture and 0.90 for fission.

[^{232}Th(n,f), ^{232}Th(n,γ), Integral Cross Sections, CFRMF, ENDF/B]

Introduction

Recent interest in developing alternate fuel cycles for advanced reactor systems has focused on the use of ^{232}Th as a fertile material for production of ^{233}U fuel. Because a realistic prediction of the production of ^{233}U in various advanced reactor systems (LMFBR, LWBR, and others) depends on an accurate description of the ^{232}Th cross sections, much effort has been expended recently on the measurement and evaluation of the neutron cross sections for ^{232}Th. Meadows et al.[1], have prepared the ^{232}Th cross section file for ENDF/B-V using data obtained up through 1977. As stated by Meadows et al., the cross section data for the ^{232}Th (n,γ) and ^{232}Th (n,f) reactions are very poor and of dubious quality, respectively. Typically, few absolute cross-section measurements are available [none for ^{232}Th (n,γ)]. The data consist primarily of differential cross sections derived from ratio measurements. A comparison of the ^{232}Th (n,γ) cross sections obtained from ratio measurements shows discrepancies in the energy range 20 keV to 1000 keV of up to 50% between the different sets of differential data. For ^{232}Th (n,f), the cross sections based on ratio measurements have normalization problems and energy scale problems in the threshold region. In addition, when the fission ratio-determined cross sections are compared to the little absolute data available, shape discrepancies are noted for energies <1.5 MeV and >1.3 MeV. To resolve the discrepancies, Meadows et al.[1] emphasized the need for better absolute and relative measurements.

Integral measurements in well-characterized neutron fields[2], the ^{235}U fission neutron spectrum, and the fast neutron fields of the CFRMF, $\sum\sum$ facility, and BIG-10, would be helpful in resolving these normalization uncertainties and in testing the differential data. Meadows et al.[1] reference several integral fission measurements made in a reference ^{235}U fission neutron spectrum. The authors note that the measured values range from 71 mb - 83 mb and are consistent with the value of 72.8 mb calculated from their new evaluation and the ENDF/B-V ^{235}U fission neutron spectrum as compared to a value of 69.0 mb calculated using ENDF/B-IV data. No similar integral measurements are identified in the Meadows[1] evaluation for ^{232}Th (n,γ).

Beck et al.[3] have reviewed recently the fast integral data for ^{233}U and ^{232}Th. Included in the review are the results of measurements of ^{232}Th capture and fission reaction rates relative to the ^{235}U fission rate made in the Advanced Epithermal Thorium Reactor (AETR) critical experiments, in the Gas Cooled Fast Reactor Experiments for the ZPR criticals and in recent ZPPR experiments. The results reported as C/E ratios (calculated to experiment with ENDF/B-IV cross-sections used to generate the calculated ^{232}Th (n,f)/^{235}U(n,f) and ^{232}Th(n,γ)/^{235}U(n,f) ratios) indicate a spread of 0.93 to 1.12 for ^{232}Th (n,f) and 0.99 to 1.04 for ^{232}Th(n,γ).

In addition, Green[4] has reported an integral capture measurement for ^{232}Th irradiated with ^{252}Cf spontaneous fission neutrons. His measured value is approximately 13% lower than that calculated using the capture cross section from BNL-325 and his Complex Spectrum for ^{252}Cf fission neutrons.

To help in the resolution of the normalization discrepancies for the capture and fission cross sections for ^{232}Th, we have made integral capture and fission measurements in the fast neutron field of the CFRMF. The purpose of this paper is to present the necessary experimental details and the results of these measurements. More complete information is found in reference 5.

Experimental Details

Irradiation Facility

Irradiation of the thorium foils took place in the fast neutron field of the CFRMF.[6,7,8] Capable of operating at a power level of 100 KW, the reactor produces a neutron flux level of ~10^{12}n/cm^2-sec in the fast neutron experiment region. Spanning the intermediate neutron energy range (1 keV < E_n < 1 MeV) with a mean energy of 0.72 MeV, the CFRMF neutron spectrum is similar to that in a fast reactor. The neutron energy spectrum has been extensively studied by proton-recoil neutron spectrometry, ^{6}Li neutron spectrometry and neutronic calculations.

This facility has been a cornerstone irradiation facility for the Interlaboratory LMFBR Reactor Rate (ILRR) program for precise dosimetry reaction-rate measurements and for an extensive series of integral measurements for "fission-product class" materials.[6] Consequently, it has been selected by the Cross Sections Evaluation Working Group (CSEWG) as a benchmark field for integral testing of data for the Evaluated Nuclear Data File (ENDF/B).

Irradiation Package

A six hour irradiation was made in the CFRMF for each of two Reactor Experiments, 99.96% pure, 1.27 cm diameter thorium metal foils (one - 161.6 mg, 0.01 cm thick; two - 22.2 mg, 0.00127 cm thick). Each foil was wrapped in three thicknesses of 0.00127 cm aluminum foil to minimize the loss of fission gas and

stop all released fragments in the aluminum cladding. Each irradiation package included three 0.4 cm diameter, 0.0127 cm thick gold foils for monitoring the reactor power level. One gold foil was located 2.54 cm above the sample midplane and the other two were located at midplane but displaced 1 cm radially from the sample center.

Post-Irradiation Ge(Li) Spectrometry

Gamma-ray spectra were measured for each of the aluminum-clad thorium foils and the gold monitors, each positioned at a calibrated location in a Ge(Li) spectrometer (65 cm^3 closed-end coaxial detector). The Ge(Li) spectrometer and gamma-ray counting techniques followed are described in references 9 and 10. For the irradiation 1 thorium foil (161.6 mg) six spectra were measured at a source to detector distance of 25 cm for times ranging from 2 days to 21 days after the end of irradiation. For the irradiation 2 thorium foil (22.2 mg), five spectra were measured at source to detector distances of 25 cm and 10 cm for times ranging from 1 day to 110 days after the end of irradiation.

Analysis and Results

Reaction-Rate Determination

Method. The method used for the determination of the thorium capture and fission reaction rates is the radiometric method, described in some detail in references 9 and 11. Briefly, saturation reaction rates are determined by the following expression

$$RR = \frac{A}{b \cdot \varepsilon} \cdot \frac{1}{N} \cdot \frac{1}{Y} \cdot D \cdot \frac{1}{\delta} ,$$

Where A is the measured peak area, b is the gamma-ray branching ratio, ε is the detector efficiency for a point source, N is the number of atoms for isotope of interest in the sample, Y is the fission yield, D is a decay/count factor and δ includes necessary correction factors.

For this specific experiment, the capture reaction rate is based on the absolute measurement of the gamma-emission rate of the 311.9-keV gamma ray from the β^- decay of 27 day ^{233}Pa. The ^{233}Pa activity is produced from the β^- decay of 22 min ^{233}Th which is the neutron capture product for ^{232}Th. The ^{233}Pa activity was measured after a sufficient time to allow for the decay of the ^{232}Th. The fission reaction rate is based on the absolute measurement of the gamma-emission rates of the 537.4-keV gamma ray from ^{140}Ba β^- decay and of the 1596-keV gamma ray from ^{140}La β^- decay. The capture reaction rate for the gold monitor, ^{197}Au, is based on the measurement of the 411.8-keV gamma ray from the β^- decay of ^{198}Au. Decay data for these radionuclides used in the present analysis are summarized in Table I. With the exception of the half-life and branching ratio for ^{233}Pa which are from references 12 and 13 respectively, the decay data were taken from reference 11. A fission yield of 0.0777 $\pm$ 4% was used for ^{140}Ba and ^{140}La.[14]

Peak area determinations were made with the GAUSS-V computer program[15] implemented on a PDP-15 interactive graphics computer system. This analysis was consistent with that used in the measurements used to generate the efficiency curves for the Ge(Li) spectrometer as described by Gehrke et al.[10] Peak efficiencies used in the present analysis were interpolated values based on the previously measured efficiency curves.

Table I. Decay Data for ^{198}Au, ^{233}Pa, ^{140}Ba, ^{140}La

Radionuclide	$T_{1/2}$	E_γ	b_γ
^{193}Au	2.6956(10)[a] d	411.794(7)	95.52(5)
^{233}Pa	26.96(5)d	311.9	38.6(5)
^{140}Ba	12.789(6)d	537.35(5)	24.4(3)
^{140}La	40.26(2)h	1596.18(5)	95.40(8)

[a]Uncertainty in last significant digits as quoted in the references.

Correction Factors. Correction factors were estimated to account for gamma-ray self absorption in the sample foils, for gamma-ray attenuation in the aluminum cladding, for random summing/live timer losses, for coincidence-summing losses, for interferring gamma rays and for neutron resonance self-shielding. Gamma-ray self-absorption corrections of 2.6% and 1.0% were estimated for the 311.9-keV and 537.4-keV lines from the 0.01 cm thick thorium foil. All other gamma-ray self absorption and attenuation corrections for the 0.01 cm and 0.0013 cm thorium foils were less than 0.3%. The values of the mass-absorption coefficients used in the determination of these corrections were obtained using quadration log-log interpolation of the total mass absorption tables of Storm and Israel.[16]

Correction factors for random summing and the live-timer were taken from data available for the spectrometer. For the thorium related gamma-ray measurements these corrections ranged from 1.0% to 1.8% for the 161.6 mg foil and 0% to 0.4% for the 22.2 mg foil.

Two additional factors affect the accurate determination of the fission rate of Th based on a measurement of the gamma-emission rates of the ^{140}La 1596-keV line. As pointed out by Debertin et al.[17], ^{208}Tl which is at the end of the decay chain of natural thorium emits 2615 keV radiation, giving rise to a double escape peak at 1593 keV. This peak, also observed in the background spectrum due to Th present in the environment, interfers with the ^{140}La 1596-keV peak and may contribute to an error in the accurate determination of the peak area of the 1596-keV peak. Secondly, 1596-keV gammas from the ^{140}La decay are emitted in cascade with the lower energy gamma rays. For small source-to-detector distances, this results in coincidence summation losses from the 1596-keV peak. These factors were considered in the analysis as follows. The contribution of the 1593-keV interferring peak to the Gaussian area determination for the 1596-keV peak was minimized by fitting the peak with the low energy Gaussian fit limit within 1 keV of the peak channel. Fission rates computed from the ^{140}La 1596-keV peaks analyzed in this way were found to be consistent with those based on the ^{140}Ba 537-keV peaks. A correction of 1% for coincidence summing losses was required for only one of the spectrum analyses for the 0.00127 cm thick foil. This correction was determined by Greenwood[11] for a source to detector distance of 10 cm for the Ge(Li) spectrometer used in the present measurements.

Even though the CFRMF neutron spectrum in the fast zone is relatively hard, there are a significant number of neutrons in the resonance region such that

neutron resonance self-shielding corrections are required for radiative capture in thorium foils used in the present experiment. Resonance self-shielding corrections were estimated as follows. Using ENDF/B-IV data 69 group (0.25 lethargy) capture cross sections were generated with the PHROG[18] code for samples which were infinitely dilute, infinite diameter disc of thickness 0.01 cm, and infinite diameter disc of thickness 0.00127 cm. Each of these multi-group cross sections was folded with a multigroup representation of the CFRMF fast zone neutron spectrum. The spectral-averaged cross sections for the thick samples relative to that computed from the infinitely dilute sample give a measure of the integral self-shielding correction factor. For the 0.01 cm and 0.00127 cm samples, the radiative-capture self-shielding correction factors were estimated to be 0.951 and 0.987, respectively.

Reaction-Rate Results

Summarized in Table II are infinitely dilute reaction rates for ^{232}Th capture and fission in the CFRMF. The thorium capture and fission reaction rates listed under the Irradiation 1 column are the averages of values determined for each gamma ray from each of six spectra analyzed for the 161.1 mg foil. Listed under the Irradiation 2 column are average reaction rates based on the analyses of five gamma spectra for capture and three gamma spectra for fission. The thorium reaction rates listed include, where applicable, corrections for gamma-ray self-absorption, external gamma absorption, random summing/live-timer losses, coincidence summing and neutron resonance self-shielding.

In parentheses next to the thorium reaction rates are estimates of the total errors and reduced errors for the measured quantities. Details concerning the error propagation can be found in reference 5. It may be noted that the dominant errors contributing to the ^{232}Th(n,γ) reaction rate uncertainty are 1.3% in the branching ratio and 1.5% in the peak efficiency for the 311.9-keV line. Dominant errors contributing to the ^{232}Th(n,f) reaction-rate uncertainty are the 4% fission yield error and 1.5% in the peak efficiencies for the 537.4-keV and 1596.2-keV lines.

Also included in Table II are estimates of the ^{197}Au(n,γ) reaction rates at the sample location for each irradiation. These estimates were made by first correcting by 1% the reaction rates determined for each of the three monitor foils to account for axial and radial flux gradients[8] and then averaging together the three reaction rates. The gold reaction rates do not include corrections for gamma ray self absorption or neutron resonance self-shielding.

Neutron Flux Determination

The neutron flux for each irradiation was determined by correlating the sample-position estimates of the ^{197}Au (n,γ) reaction rates given in Table II to that determined by Harker[6] for a 0.0127 cm gold foil at the sample position. Harker[6] in a separate experiment had correlated the "sample position" 0.0127 cm gold reaction rate to that of a 0.005 cm gold foil at the ILRR monitor position (2.54 cm above midplane). His measurements yielded (4.785 $\pm$ 0.003) X 10^{-14} rps/a for the 0.005 cm foil and (4.859 $\pm$ 0.015) X 10^{-14} rps/a for the 0.0127 cm gold foil. From Rogers et al.[8] a reaction rate of 3.008 X 10^{-14} rps/a for the 0.005 cm ILRR monitor corresponds to a midplane flux of 7.9 X 10^{10} n/cm^2-sec $\pm$ 1.9%. Based on these correlating data and the estimated reaction rates from the present experiments, the neutron fluxes for the present irradiations were determined to be 8.78 X 10^{11}n/cm^2-sec $\pm$ (2.7%, 1.9%) for irradiation 1 and 8.73 X 10^{11}n/cm^2-sec $\pm$ (2.5%, 1.6%) for irradiation 2. In parenthesis are two error values, the first an estimate of the total error in the flux determination and the second an estimate of the error without the 1.9% flux uncertainty from the ILRR fission-chamber measurements.

Table II. Reaction rates for ^{232}Th(n,γ), ^{232}Th(n,f) and ^{197}Au(n,γ) in the CFRMF

Reaction -E$_\gamma$	Reaction Rate (reactions/sec-atom)X10^{14} Irradiation 1	Irradiation 2
^{232}Th(n,γ)		
- 311.9	25.64[a] (2.5[a],1.5[b],0.3[c])	25.31 (2.2,1.0,0.4)
^{232}Th(n,f)		
- 537.4	1.716 (4.5,2.0,0.5)	1.727 (4.7,2.5,1.4)
-1596.2	1.715 (4.3,1.6,0.4)	1.706 (4.5,2.1,1.2)
-(537+1596)	1.72 (4.2[d],1.2[e])	1.71 (4.3,1.6)
^{197}Au(n,γ)		
- 411.8	33.9 (1.9,1.1)	33.74 (1.6,0.4)

[a] Total percentage error for the above reaction rate. Definition applies for all reaction rates listed.

[b] Reduced error which for Th(n,γ) case does not include a 1.5% peak efficiency error or a 1.3% branching ratio error. For the Th(n,f) case the 4% fission yield error is excluded. For the Au(n,γ) case the 1.5% peak efficiency error is excluded.

[c] Internal error computed from the averaging indicated in the text.

[d] Total percentage error for the above reaction rate which is the weighted average of the above two rates.

[e] Reduced error which does not include the 4% fission yield error.

Integral Cross Sections

Integral cross sections for capture and fission of ^{232}Th irradiated in the CFRMF were obtained by dividing the capture and fission reaction rates given in Table II by the corresponding neutron fluxes given in the previous paragraph. The fission rates used in Table II were those based on the (537+1596) averages. For the first irradiation the integral capture and fission cross sections were determined to be 292 mb $\pm$ (3.7%, 2.4%) and 19.5 mb $\pm$ (5%, 2.3%). The first error in parenthesis for each of these cross sections is the total percentage error at the 1-sigma level. The second error in parenthesis for the capture cross section does not include the 1.5% peak efficiency, 1.3% branching ratio and 1.9% fission-chamber flux uncertainties. For the fission cross section the second error does not include the 4% fission yield error and the 1.9% fission chamber flux uncertainty. Based on averages of the results from both irradiation experiments, the measured integral capture cross section for ^{232}Th in the CFRMF is 291 mb $\pm$ 3.1%. Similarly, the measured integral fission cross section

for ^{232}Th in the CFRMF is 19.6 mb ± 4.7%.

Discussion

As stated earlier, integral measurements are helpful in sorting out normalization difficulties with differential data. Based on the measured integral data from this experiment a simple integral test of the ENDF/B-IV capture and fission cross sections was made as follows. Estimates of the ENDF/B-IV spectral-averaged cross sections were computed using the 620 group capture and fission cross sections from the ENDF/B-IV Dosimetry Library[17] and a 620 group representation of the CFRMF neutron spectrum as given by Harker et al.[6] Calculated integral cross sections are 287 mb for capture and 17.6 mb for fission. The resultant calculated-to-experimental ratios are then 0.99 for capture and 0.90 for fission. These ratios indicate that the integral values for capture deviate by less than the uncertainty in the cross section measurement. However, for fission the deviation is approximately twice the uncertainty in the measured integral value. The capture calculated-to-experimental ratio from the present work is consistent with the corresponding ratios given by Beck et al.[3] However the fission calculated-to-measured ratio is consistent with the lower ratio values given in that review of integral data. The present measurements suggest that the normalization of the ENDF/B-IV capture cross section is consistent with the present integral measurement but the normalization of the ENDF/B-IV fission cross section is too low by 10%.

With the advent of covariance files for many cross sections important to reactor technology the measurements presented here can be used for other than simple integral checks of ENDF/B data, namely, the adjustment of multigroup cross sections based on measured reaction rates. Such analyses require as a-priori information: a multigroup representation of the neutron spectrum, a flux covariance matrix which realistically treats the uncertainties and energy dependent correlations in the flux, multigroup representations of all cross sections and covariance matrices for those cross sections. The results of such analyses would be helpful in evaluating the point-wise differential data base.

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