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ORELA CONTRIBUTION TO THORIUM-CYCLE NUCLEAR DATA*

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ORELA CONTRIBUTION TO THORIUM CYCLE NUCLEAR DATA*

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Abstract

The measurements of direct importance to the $^{232}\text{Th}/^{233}\text{U}$ fuel cycle using neutrons from the Oak Ridge Linear Accelerator facility are gathered together and discussed. These measurements were done in response to specific data discrepancies, as part of generic programs, and for basic fission physics studies. In particular, completed transmission and capture work on ^{232}Th has yielded the most accurate parameters for the first four s-wave resonances; the largest average capture width, 25.2 meV; and the largest s-wave strength function of recent measurements. These results allow improved agreement between differential and integral capture rates. Moreover, the ORNL ^{252}Cf $\bar{\nu}$ measurement of unprecedented accuracy and ^{233}U $\bar{\nu}$ ratio measurement give a ^{233}U prompt $\bar{\nu}$ (thermal) of 2.490 ± 0.009 neutron/fission. This result allows a much more satisfactory understanding of the ^{233}U 2200 m/s constants. In addition, a ^{232}Th γ -ray production measurement provides needed cross sections for shielding, and important data for both application and fission physics were obtained from ^{232}Th fission and both ^{231}Pa and ^{234}U fission and total cross section measurements. Data requirements and discrepancies suggested from this work are discussed.

Introduction

In addition to providing neutron cross-section (CS) data on fission products, structural materials, coolants, control materials, standards and the $^{238}\text{U}/^{239}\text{Pu}$ fuel cycle, measurements from the Oak Ridge Electron Linear Accelerator (ORELA) facility have made a substantial contribution to our present knowledge of nuclear data directly required for the $^{232}\text{Th}/^{233}\text{U}$ fuel cycle. This paper gathers together and discusses these contributions which also include evaluation. Some data requirements and discrepancies apparent from this work are also discussed.

The incentives to perform these measurements were both varied and mixed. The proliferation concerns in the late 70's over the $^{238}\text{U}/^{239}\text{Pu}$ fuel cycle sparked a renewed interest in the $^{232}\text{Th}/^{233}\text{U}$ fuel cycle and a review of the underlying differential CS data base. Some ORELA measurements and evaluations were performed mostly as specific responses to known discrepancies in this data base. Other ORELA measurements important for the $^{232}\text{Th}/^{233}\text{U}$ fuel cycle were performed mostly as part of larger generic programs in which a fission parameter or CS type was measured for all relevant isotopes. Finally some CS important to the $^{232}\text{Th}/^{233}\text{U}$ fuel cycle were obtained from measurements motivated primarily from basic physics concerns. The material in this paper is organized somewhat in terms of the above incentives.

First, after a brief description of ORELA, the ^{232}Th total¹ and capture^{2,3} CS measurements and evaluations^{4,5} will be discussed. This work was in response to the apparent underprediction by differential data of the integrally measured thorium capture rates. The ^{233}U total CS from 0.01 to 1.0 eV was also measured⁶ to resolve known discrepancies in the data base. Secondly, the ORELA ^{233}U $\bar{\nu}(E)$ results and ^{232}Th γ -ray production results will be discussed. The ^{233}U $\bar{\nu}(E)$ measurement^{7,8} and normalizing ^{252}Cf $\bar{\nu}$ measurement⁹ were part of a comprehensive program to provide definitive $\bar{\nu}(E)$ values for the major fissile isotopes. Thorium was one of 27 materials for which secondary γ -ray production CS were measured.¹⁰ Finally, the results from the ^{232}Th fission CS,^{11,12} ^{231}Pa fission,¹³ and total¹⁴ CS, and ^{234}U fission and total CS¹⁵ measurements will be described. This work was motivated primarily by basic fission physics; specifically, the question of the shape of the fission barrier in the light actinide region. The various measurements discussed in this paper are listed in Table ..

The ORELA Facility

The ORELA facility shown in Fig. 1 was designed, constructed, and brought into operation in the late 60's and was optimized to be a high performance pulsed neutron source for time-of-flight (TOF) measurements, spanning energies from the meV region to the MeV region. ORELA¹⁶

Table 1. ORELA Thorium Cycle Measurements

Measurement	Energy Range	Ref
^{231}Pa total CS	10 meV - 10 keV	14
^{231}Pa (n,f)	0.4 eV - 12 MeV	13
^{232}Th total CS	8 meV - 100 keV	1
^{232}Th (n, γ)	100 eV - 4 keV	2
^{232}Th (n, γ)	2.6 keV - 800 keV	3
^{232}Th (n,f)	100 eV - 1.6 MeV	11
^{232}Th (n,f)	0.9 MeV - 10 MeV	12
^{232}Th (n, $\alpha\gamma$)	300 keV - 20 MeV	10
^{233}U total CS	10 meV - 1 eV	6
^{233}U $\bar{\nu}$ ratio to ^{252}Cf	10 meV - 10 MeV	7,8
^{252}Cf absolute $\bar{\nu}$		9
^{234}U total CS	3 eV - 8.9 MeV	15
^{234}U (n,f)	3 eV - 8.9 MeV	15

consist of four L-band linac sections with an unloaded beam energy of 178 MeV. Each section is 4.1 m long and is driven by a Litton LS081 klystron producing 25 MW of peak RF power. The gridded-gun injector produces peak currents in excess of 30A for pulses as short as 4 ns. Approximately half of this current is accelerated and stopped in the water-cooled and water-moderated Ta target assembly¹⁷ shown in Fig. 2. The resulting electron bremsstrahlung from the Ta plates produces photoneutrons, many of which are moderated to lower energies by the target cooling water.

These moderated and unmoderated neutrons are used for TOF measurements along 10 evacuated flight paths with 18 measurement stations at distances from 9 to 200 m from the target-moderator. Pulse widths from 4 to 40 ns and pulse repetition rates from 25 to 1000 Hz are routinely used. At the maximum pulse width and repetition rate ORELA produces 1.2×10^{14} n/s with 60 kW of beam power. A prebuncher presently under development may allow over 50% of this maximum beam power to be compressed into 4-ns-wide pulses.¹⁸ Over the last decade ORELA has been the major U.S. facility to provide the required differential CS data for both thermal and fast reactors.¹⁹

^{232}Th Total and Capture CS

Accurate knowledge of the ^{232}Th total and capture CS is required mostly to determine the shielded capture CS for ^{233}U production at all

Figure 1. ORELA layout showing the underground accelerator, target room, flight paths, and short-flight-path measurement stations.

Figure 2. ORELA water-cooled and water-moderated Be-clad Ta target assembly.

neutron energies. In the mid 70's there were many discrepancies associated with these CS. Measured capture, scattering, and total CS in the thermal region seemed inconsistent among themselves.²⁰ Moreover, in the resolved resonance region, 5 eV to 4 keV, there were only two relatively complete sets of neutron widths: those of Rahn et al.²¹ and Ribon.²² Unfortunately, at energies greater than 1 keV the neutron widths of Rahn et al.²¹ are systematically smaller than those of Ribon.²² In addition, although the older capture-width measurements²¹⁻²⁴ gave a remarkably

consistent average capture width, on a resonance by resonance basis the agreement was not that good. Finally, analysis of critical experiments seemed to indicate a ^{232}Th capture problem. Ullo et al.²⁵ tested various ^{232}Th evaluations against $^{232}\text{Th}/^{235}\text{U}$ thermal reactor benchmarks and found them to predict smaller cadmium capture ratios and shielded capture effective resonance integrals than were experimentally measured. Singh et al.²⁶ came to the same conclusion for the ENDF/B-V evaluation.²⁷ A similar problem exists for fast reactor ^{232}Th capture.²⁸ This possible systematic underprediction of ^{232}Th capture was opposite to the possible overprediction of the better studied ^{238}U capture. In order to better understand and possibly eliminate some of these discrepancies and inconsistencies, both transmission¹ and capture^{2,3} measurements using ORELA were made over the ^{232}Th resonance region.

Transmission Measurements

Transmission intensities as a function of neutron TOF were measured at 22, 40, and 155 m from the ORELA target-moderator assembly employing 1.0-mm-thick (22 and 40 m) and 12.7-mm-thick (155 m) Li-glass disks which were viewed edge on by two PMT. Table 2 lists some of the experimental parameters. Various sample thicknesses were constructed from eight 165-mm-diameter metallic ^{232}Th disks with thicknesses ranging from 0.05 mm to 25.4 mm.

After deadtime correction and subtraction of small backgrounds, the 22-m and 40-m spectra were normalized utilizing the number of beam bursts, transmission and CS calculated, and statistical errors propagated. Their energy scales were adjusted to coincide with that of the ORELA 155-m ^{238}U transmission measurement.²⁹ The 155-m data have not yet been reduced to final transmission spectra.

Below 20 eV CS were calculated from the 22-m transmissions, two sets of five samples each, and combined using both statistical and systematic uncertainties as weights. The systematic uncertainties were obtained assuming possible 0.5, 10.0, and 5.0% errors in the transmission normalizations, backgrounds, and deadtimes, respectively. The resulting CS is compared with the ENDF/B-V evaluation²⁷ in Fig. 3 and is believed accurate to $\pm 0.3\%$. From about 0.1 to 1.0 eV the two agree within uncertainty.

Table 2. Experimental Parameters for ORELA ^{232}Th Transmission Measurements

Parameter	22 m	40 m	155 m
Flight path (m)	22.52	40.01	155.94
Pulse rate (pps)	25	150	500
Pulse width (ns)	30	12	15
Beam power (kW)	1	3.5	12
Pb γ -filter (mm)	51	19	6
Time filter (kg/m ²)	Mn-22.9 Au-60.8	B-1.1	B-9.3
Bgd filters (b ⁻¹)	Cd-0.00037 U-0.0188	Al-0.237 U-0.0188	Al-0.158 Mn-0.0024
^{232}Th samples (b ⁻¹)	0.19308 0.15440 0.11588 0.07720 0.03868	0.19308 0.11588 0.03868 0.012566 0.004080 0.0013949 0.0004537 0.0001608	0.19308* 0.11588 0.03868 0.012566 0.004080*

*Transmissions only for broad energy groups.

Above 1.0 eV the difference is larger and probably not important and may be due to resolved resonance parameters. Below 0.2 eV the difference is probably due to the extinction effect from Bragg Scattering. The 22-m ORELA total cross section and the recent RPI total cross section³⁰ agree everywhere within 1%.

In order to obtain resonance parameters, the eight 40-m transmission spectra were simultaneously least-squares shape fitted using the computer code SIOB^{29,31} which employs the multi-level Breit-Wigner approximation. Considerable care was taken to account for distant levels so that the accurately measured transmission normalizations, $\pm 0.5\%$, could be exploited in the fits. The data below 2 keV were divided into 15 energy regions and the CS for each region was calculated with the s-wave contribution from resonances extending 250 eV beyond the fitted region explicitly included. These CS were Gaussian Doppler broadened giving eight transmissions which were resolution broadened and least-squares fitted to the data by varying resonance parameters, the effective radius, and the resolution width. Several iterations of this procedure produced neutron widths and resonance energies for 176 resonances from 13.1 to 2006.2 eV, capture widths for the first 19 s-wave resonances, and an effective radius of 9.72 ± 0.06 fm. Figure 4 shows the fit around the 21.8 to 23.5 eV doublet. The same CS parameters fit all eight transmission spectra.

In order to verify the widths for the low-energy s-wave resonances, the two sets of five-sample, 22-m transmissions were also least-squares fitted. The widths from these fits are compared with those from the 40-m data in Table 3 which shows good agreement between the three measurements. The uncertainties in Table 3 are the usual least-squares fitting uncertainties, propagated from the statistical scatter of the experimental data, multiplied by three.

Although the final data reduction and analysis of the 155 m transmission data has not been completed, preliminary transmissions have been calculated. Figure 5 compares the 155-m preliminary transmission spectra through the 0.03808 b⁻¹ sample with that from the 40-m measurement. The difference in resolution is obvious and the 155-m results should provide resonance parameters up to at least 6 keV. The completed transmission work gives the largest s-wave strength function from 0.5 to 2.0 keV that has been reported. More importantly, the 19 low-energy s-wave capture widths have an average value of 25.2 meV, which is substantially larger than previous measurements. Together these results imply a larger capture CS than previous measurements.

Figure 3. Comparison between the ORELA measured ^{232}Th total neutron CS and the ENDF/B-V evaluation. The upper figure is a direct comparison, whereas the lower figure shows the ratio averaged over several data points. The plotted errors are statistical only. The systematic error is believed to be less than 0.3%.

Table 3. ORELA ^{232}Th Neutron and Capture Widths (meV)*

Resonance	40 m w/B	20 m w/Mn	20 m w/Au	Average
21.80 eV Γ_n	2.08 ± 0.03	2.06 ± 0.08	2.07 ± 0.07	2.08 ± 0.02
Γ_γ	25.2 ± 0.4	25.4 ± 1.0	25.6 ± 1.0	25.3 ± 0.4
23.46 eV Γ_n	3.80 ± 0.04	3.85 ± 0.10	3.89 ± 0.10	3.82 ± 0.04
Γ_γ	27.0 ± 0.5	26.8 ± 1.0	26.7 ± 1.0	26.9 ± 0.4
59.50 eV Γ_n	3.87 ± 0.06	3.74 ± 0.20		3.86 ± 0.06
Γ_γ	24.3 ± 0.9	27.2 ± 2.8		24.6 ± 0.9
69.21 eV Γ_n	43.0 ± 0.2	43.9 ± 0.5		43.2 ± 0.2
Γ_γ	24.0 ± 0.3	24.9 ± 0.9		24.1 ± 0.3

*Listed uncertainties are uncorrelated only. The neutron and capture widths have additional 1% and 2% systematic uncertainties, respectively.

Capture Measurements

Two ^{232}Th capture measurements have been completed with ORELA using common samples and the same experimental apparatus. The low energy measurement of Perez et al.² concentrated on neutron energies from 100 eV to 4 keV, and used both 0.00082 and 0.00274 b⁻¹ samples.

Figure 4. Simultaneous least-squares fit to the eight 40-m transmission spectra, plotted around the 21.8- to 23.5-eV doublet of ^{232}Th . The dip at 18.8 eV is from a ^{186}W impurity.

Figure 5. Transmission spectra through a 0.03868 b^{-1} sample of ^{232}Th . The 40-m results, data points with error bars, have been analyzed yielding resonance parameters up to 2 keV. The 155-m results, histogram, will yield resonance parameters up to at least 6 keV.

The high energy measurement of Macklin and Halperin³ concentrated on neutron energies from 2.6 to 800 keV and used only the 0.00082 b^{-1} sample.

The samples were 5.2×2.6 -cm rectangular metallic foils, fabricated from ^{232}Th electromagnetically separated from natural thorium, to reduce the γ -ray activity associated with contaminants. Regrowth of the activity was interrupted every one or two months by chemical purification on an ion absorption column. A sample was mounted normal to the neutron beam at 40.12 m between two C_6F_6 liquid scintillators used as a γ -ray detector. The pulses from this γ -ray detector were processed by application of a pulse-height weighting scheme giving a detector response proportional on average to the total emitted photon energy. The shape of the incident neutron flux was obtained with a 0.5-mm-thick Li-glass scintillator. The capture yield was assumed to be proportional to the ratio of the pulse-height-weighted γ -ray detector count rate to the incident neutron flux as determined from the Li-glass count rate.

For the low-energy measurement of Perez et al.² the proportionality constant was obtained by normalizing the measured capture yields to calculated capture yields over the saturated peaks of the ^{232}Th s-wave levels at 113 and 121 eV. The capture yields around these two levels were computed as a function of neutron energy with the MULTSCA code³² using ENDF/B-V resonance parameters.³⁷ The computer code MULTSCA employed Monte Carlo techniques to include the effects of resonance self shielding and neutron multiple scattering in the calculated capture yields which also are both resolution and Doppler broadened. With this normalization, the measured capture yields were compared from 0.1 to 4.0 keV with MULTSCA calculated capture yields using ENDF/B-V resonance parameters summed with the auxiliary ENDF/B-V "smooth" capture CS. Figure 6 shows the comparison from 2.0 to 2.5 keV. The solid line represents the calculated yield. The measured yields have an estimated normalization uncertainty of $\pm 3\%$. Clearly from 2.0 to 2.5 keV the ENDF/B-V evaluation underpredicts the ^{232}Th capture yield, particularly for the thin sample which is more sensitive to resonance parameters because of its smaller finite sample effects. Simple integration from 1.0 to 4.0 keV of the measured and calculated capture yields from the thin sample indicates that the ENDF/B-V evaluation under predicts ^{232}Th capture by approximately $10 \pm 3\%$.

Figure 6. Comparison between measured and calculated ^{232}Th capture yields for 0.00082 and 0.00274 b^{-1} sample thickness. The calculations were done with the computer code MULTSCA using ENDF/B-V resonance parameters and include the effects of Doppler broadening, self shielding, multiple scattering, and resolution broadening. Figure from the work of Perez et al. (Ref. 2).

The high energy measurement of Macklin and Halperin³ for the 0.00082 b^{-1} sample was also normalized to give an effective capture CS by the saturated resonance technique using primarily the Au 4.9-eV resonance. These data were least-squares fitted to obtain both resolved and unresolved resonance parameters. Figure 7 shows the fit for unresolved resonance parameters. These data were simultaneously corrected and fitted,³³ on the average, for neutron multiple scattering and resonance self-shielding, in terms of theoretical distribution functions with strength functions as free parameters. The corrected capture CS from 2.6 to 112.6 keV were well fitted as shown in Fig. 7 with $10^4 S_0 = 0.86 \pm 0.10$, $10^4 S_1 = 1.48 \pm 0.07$, $10^4 S_2 = 1.12 \pm 0.06$, and $10^4 \Gamma_0/D = 14.7 \pm 0.5$. The uncertainties shown are statistical standard deviations from the least-squares adjustment. The value found for S_0 corresponds to an average capture width of $25.0 \pm 0.8 \text{ meV}$ for an s-wave level spacing of 17.0 eV .

Figure 7. Least-squares fit to the ^{232}Th capture CS measured with a 0.00082 b^{-1} sample. The measured CS is shown as a histogram and has been corrected for finite sample effects. The smooth curve is the calculated average CS from the listed strength functions. Figure from the work of Macklin and Halperin (Ref. 3).

In addition, the effective CS from 2.6 to 10.3 keV was least-squares fitted³⁴ in order to obtain resolved resonance parameters. Upper Fig. 8 shows a direct comparison of the measured effective capture CS with that calculated from ENDF/B-V resonance parameters²⁷ and includes the effects of Doppler broadening, resonance self shielding, neutron scattering, and experimental resolution. These results also indicate that the ENDF/B-V resonance parameters do not provide enough capture CS. A much improved fit obtained by least-squares adjustment of the resonance parameters is shown in lower Fig. 8. Resonance energies and capture widths were obtained for large resonances below 4 keV using the neutron widths from the Keyworth and Moore evaluation.³⁵ Resonance energies and capture areas were obtained for weak resonances and resonances above 4 keV.

The data shows 14 large s-wave resonances from 2.63 to 3.05 keV which have a weighted mean capture width of 26.9 meV . From these resonances weighted a mean capture width of $25.5 \pm 1.2 \text{ meV}$ was derived by estimating and subtracting the capture area from small unobserved p-wave resonances. This average is in excellent agreement with the low energy ORELA transmission average of $25.2 \pm 0.6 \text{ meV}$.¹

Discussion and Evaluation

In this subsection the results from the ORELA ^{232}Th transmission and capture measurements are compared to those from other workers and a new evaluation of the resolved resonance parameters which heavily weights these new results is described. This evaluation,⁴ ENDF/B-V mod-2 for

Figure 8. Measured ^{232}Th resonance neutron capture CS from a 0.00082 b^{-1} sample. The smooth curve in the upper figure is a calculated CS using ENDF/B-V resonance parameters and includes Doppler broadening, self shielding, multiple scattering, and resolution broadening. The smooth curve in the lower figure is a least-squares fitted CS, resulting in substantially increased capture widths. Taken from the work of Macklin and Halperin (Ref. 3).

^{232}Th , has been accepted to be a modification of the ENDF/B-V ^{232}Th evaluation²⁷ and was prompted by the serious underprediction of ^{232}Th capture by ENDF/B-V as suggested by integral measurements, as directly measured by the ORELA capture experiments, and as implied by the ORELA resonance parameters from transmission.

Table 4 compares the parameters from ORELA for the first four s-wave resonances with those from other measurements^{21-24,36-41} and with evaluations.^{4,27,35,42,43} These resonances provide about 68% of the infinitely dilute capture resonance integral. There are very large discrepancies between some of the measured widths and many of the older measurements have large uncertainties. The ENDF/B-V mod-2 evaluated widths are simply the inverse-variance-weighted average on a resonance by resonance basis of the Table 4 measurements. The ORELA widths have small uncertainties; however, it is important to appreciate that the evaluated widths are almost independent of this measurement. The evaluated uncertainties are considerably reduced by the ORELA results. Both the ORELA measured and evaluated widths are also in excellent agreement with the recent results of Chrien et al.³⁶ and are similar to those of the

Table 4. Widths (meV) for the First-Four ^{232}Th s-Wave Resonances

Measurement	21.8 eV		23.5 eV		59.5 eV		62.2 eV	
	Γ_n	Γ_γ	Γ_n	Γ_γ	Γ_n	Γ_γ	Γ_n	Γ_γ
Olsen (80)	2.08 $\pm$.03	25.3 $\pm$ 0.6	3.82 $\pm$.06	26.9 $\pm$ 0.7	3.86 $\pm$.07	24.6 $\pm$ 1.0	43.2 $\pm$ 0.5	24.1 $\pm$ 0.6
Chrien (79)	2.10 $\pm$.10	24.0 $\pm$ 1.5	3.70 $\pm$.20	26.0 $\pm$ 1.5	4.00 $\pm$.30	25.0 $\pm$ 2.0	41.2 $\pm$ 3.0	22.6 $\pm$ 2.0
Halperin (75)			3.72 $\pm$.11					
Rahn (72)	1.91 $\pm$.09	20.0 $\pm$ 2.0	3.24 $\pm$.24	25.0 $\pm$ 2.0	3.93 $\pm$.23	25.0 $\pm$ 2.0	44.0 $\pm$ 2.0	25.0 $\pm$ 2.0
Forman (71)					4.01 $\pm$.31	22.7 $\pm$ 6.0	42.5 $\pm$ 1.7	21.9 $\pm$ 2.8
Ribon (69)	1.96 $\pm$.08	21.3 $\pm$ 3.7	3.64 $\pm$.12	28.2 $\pm$ 2.9	3.93 $\pm$.08	23.6 $\pm$ 0.7	44.0 $\pm$ 0.5	20.5 $\pm$ 1.0
Bollinger (68)	2.09 $\pm$.03	26.5 $\pm$ 1.2	4.11 $\pm$.05	25.2 $\pm$ 1.1	3.77 $\pm$.15	31.7 $\pm$ 2.9	42.0 $\pm$ 0.8	23.5 $\pm$ 1.4
Bhat (67)	2.61 $\pm$.17	22.4 $\pm$ 2.4	4.17 $\pm$.25	22.0 $\pm$ 2.6	4.41 $\pm$.27	16.2 $\pm$ 2.3	42.9 $\pm$ 2.6	21.5 $\pm$ 4.7
Asghar (66)	1.88 $\pm$.05	24.6 $\pm$ 1.2	3.41 $\pm$.08	29.9 $\pm$ 1.6	3.34 $\pm$.09	23.2 $\pm$ 2.0	41.4 $\pm$ 1.2	21.2 $\pm$ 1.0
Haddad (64)	2.10 $\pm$.20	21.0 $\pm$ 5.0	4.00 $\pm$.30	22.0 $\pm$ 4.0	4.00 $\pm$.40	22.0 $\pm$ 7.0	47.0 $\pm$ 15.0	24.0 $\pm$ 2.0
Paievsky (64)	2.60 $\pm$.20	30.0 $\pm$ 2.0	3.70 $\pm$.20	27.6 $\pm$ 1.5	4.50 $\pm$.30	37.0 $\pm$ 5.0	38.0 $\pm$ 6.0	26.0 $\pm$ 5.0
ENDF/B-V mod-2	2.06 $\pm$.03	25.1 $\pm$ 0.6	3.85 $\pm$.06	26.6 $\pm$ 0.5	3.80 $\pm$.05	24.0 $\pm$ 0.6	43.2 $\pm$ 0.3	23.0 $\pm$ 0.6
ENDF-V (80)	2.02	23.0	3.88	25.0	4.04	23.2	44.0	21.9
ENDF-IV (65)	2.00	25.9	3.74	25.9	4.00	25.9	42.0	25.9
LASL (78)	1.97 $\pm$.05	20.9 $\pm$ 11.3	3.72 $\pm$.15	19.0 $\pm$ 7.7	3.92 $\pm$.10	20.1 $\pm$ 7.2	44.0 $\pm$ 1.0	22.9 $\pm$ 1.5
Derrien (74)	2.02 $\pm$.06	24.5 $\pm$ 2.0	3.88 $\pm$.40	26.6 $\pm$ 1.5	3.90 $\pm$.15	23.7 $\pm$ 1.0	43.2 $\pm$ 1.0	21.9 $\pm$ 0.7

1974 evaluation of Derrien.⁴³ Considering the importance of these four resonances for $^{232}\text{Th}/^{233}\text{U}$ thermal reactors, at least one additional precision measurement of these parameters should be performed. Only two such measurements have been completed during the last decade.

The s-wave neutron widths from the transmission work are summarized in Fig. 9 where local strength functions for 0.5-keV intervals are plotted as percent differences from the ENDF/B-V evaluation.²⁷ Also shown in Fig. 9 are the Ribon²² and Rahn et al.²¹ measured values and the Derrien⁴³ and Keyworth and Moore³³ evaluated values. Above 0.5 keV the ORELA measurement gives the largest strength function which has been either measured or evaluated. A direct comparison of the ORELA transmissions to those calculated from the ENDF/B-V evaluation indicate that the strength function discrepancy shown in Fig. 9 persists to 4 keV.¹ It is not known whether these systematic differences are from the measured transmission spectra or from the method of analysis. The ENDF/B-V mod-2 evaluation for ^{232}Th attempts to correct for these systematic differences and gives neutron widths below 2 keV which yield an s-wave strength function midway between the ORELA¹ and Ribon²² results. Above 2 keV the evaluated neutron widths are on average 5-15% larger than those of the ENDF/B-V evaluation.

Table 5 compares the ORELA measured average capture widths with those from other measurements and with evaluations. The results of Harwell,²³ Ribon,²² LANL,²⁴ (using Ribon's Γ_γ 's) and Columbia²¹ on average are in remarkably good agreement and are an astonishing 20% lower than the ORELA capture widths. However, if only the first four capture widths are compared, this discrepancy is reduced to 7%, and the BNL³⁶ average for the first four resonances is only 3% smaller than the ORELA result.

It is difficult to determine the origin of this large difference. The ORELA capture widths cannot be explained by reasonable uncertainties in the Doppler temperature, resolution width, background, or normalizations. In addition, as shown in Table 3 these capture widths, at least for a few resonances, are not unique to a single transmission measurement. Clearly, this is a major uncertainty which requires additional measurements. All s- and p-wave resonances in the ENDF/B-V mod-2 ^{232}Th evaluation⁴ except for the first 17 s-wave resonances are given a capture width of 24.4 ± 2.0 MeV, which allows either the old or new results to be correct.

These ORELA measured and evaluated resonance parameters, particularly the capture widths, seem to alleviate some of the known difficulties and problems with the earlier ^{232}Th CS results. These widths improve the agreement between calculated and integrally measured dilute capture resonance integrals I_γ . Greneche⁴⁴ reports an evaluated $I_\gamma = 85.8 \text{ b} \pm 0.9 \text{ b}$ (experiment) $\pm 1.6 \text{ b}$ (normalization) from a review of the available integral measurements. Both the ENDF/B-V²⁷ and Derrien⁴³ evaluation give $I_\gamma = 83.7 \text{ b}$, which is acceptable but low. The ENDF/B-V mod-2 evaluation⁴ gives $I_\gamma = 86.1 \text{ b}$. In addition, these parameters reduce the discrepancy between calculated and measured cadmium capture ratios for

Figure 9. Comparison of ^{232}Th local s-wave strength functions for 0.5 keV intervals between the ORELA, Ribon, and Rahn et al. measurements and the ENDF/B-V, LANL, and Derrien evaluations. The local strength functions are plotted as percent differences from the ENDF/B-V values. The total uncertainties in the ORELA local strength functions are 1.7% and 2.3% below and above 1 keV, respectively. Above 0.5 keV, the ORELA results give the largest strength function that has been measured.

two of the BNL ^{233}U -ThO₂-fueled water-moderated exponential assemblies.⁴⁵ Moreover, the ORELA measurements give an average capture width that is nearly equal to that of the first four resonances. Table 6 summarizes the average parameters for the resolved resonance region for the ENDF/B-V mod-2 evaluation.

Table 5. Average Capture Widths (meV)

Measurement	Γ_γ
ENDF/B-V mod-2 first-four	24.7
Olsen (0 to 350 eV)	25.2 ± 0.6
Macklin (2.6 to 4.0 keV)	25.5 ± 1.2
Rahn (0 to 2.5 keV)	21.2 ± 1.2
Ribon (0 to 300 eV)	21.8 ± 0.8
Asghar (0 to 900 eV)	21.5 ± 0.8
Forman (0 to 2.0 keV)	21.3 ± 3.0
ENDF/B-V mod-2	24.4 ± 2.0
ENDF/B-V	21.1
ENDF/B-IV	25.9

Table 6. Average Parameters from Resolved Resonance Evaluation

s-wave resonances	
$\langle \Gamma_\gamma \rangle$	24.4 ± 2.0 meV
$10^4 S^0$	0.83 ± 0.08
D_0	16.4 ± 1.0 eV
R	9.71 ± 0.04 fm
p-wave resonances	
$\langle \Gamma_\gamma \rangle$	24.4 ± 2.0 meV
$10^4 S^1$	1.6 ± 0.3 eV ^h
D_1	5.69 ± 0.35 eV

²³³U Low Energy Total CS

Over the energy region from 0.1 to 0.2 eV the total CS inferred from the ORNL-RPI ²³³U capture and fission measurements⁴⁶ is systematically less than the bulk of the total CS values from direct transmission measurements.⁴⁷ In order to clarify this discrepancy, neutron transmission spectra through 0.0031 and 0.0061 b⁻¹ samples of 99.76% enriched ²³³U were measured from 10 meV to 1 eV by Harvey et al.⁶ Neutrons were detected at 17.87 m with a 12.7-mm-thick Li-glass detector. Although the results from the measurement are not definitive, they seem to be discrepant with the ENDF/B-V evaluation,⁴⁸ which is based primarily on the ORNL-RPI capture and fission results.⁴⁶

Measurement of ²³³U $\mu(E)$

In the mid 70's ORNL began a major two-step program to measure the absolute value of the average number of prompt fission neutrons per fission $\bar{\nu}(E)$ as a function of incident neutron energy for the major fissile isotopes. This program is essentially complete. To first order, the sensitivity of the effective multiplication of a reactor or critical assembly to the spectrum-averaged and fuel-averaged $\bar{\nu}$ is unity. One step of this program was to measure the ratios $R(E)$ of the neutron induced prompt $\bar{\nu}(E)$ for ²³³U, ²³⁵U, ²³⁹Pu, and ²⁴¹Pu to that for spontaneous fission of ²⁵²Cf. These measurements by Gwin et al.^{7,8} did not require an absolute neutron detector efficiency calibration. The other step was to precisely measure the absolute prompt $\bar{\nu}$ from the spontaneous fission of ²⁵²Cf. This normalizing measurement by Spencer et al.⁹ required an absolute neutron detector efficiency calibration.

Fission neutrons for both the ²³³U $R(E)$ and ²⁵²Cf $\bar{\nu}$ measurements were detected with the large liquid scintillator tank shown schematically in Fig. 10. This tank at 85 m contained 910 liters of gadolinium-loaded NE-224 and was viewed by twelve RCA 4522 PMT's. Fast neutrons from the tank center were thermalized and absorbed within about a 15- μ s time constant. Neutron multiplicities were determined by counting summed PMT pulses with a fast scaler for a time interval following an initiating background, proton recoil detector, or fission pulse. Each event was stored according to the initiating pulse, often as a function of incident neutron TOF, and the associated number of neutrons counted by the scaler. A fission event was defined by a coincidence between the tank response to the prompt fission γ -rays and the fragment ionization pulses from the fission chamber.

Figure 10. Schematic of the ORNL 910-liter, gadolinium-loaded scintillator tank used for ²⁵²Cf $\bar{\nu}$ and ²³³U $R(E)$ measurements. The ²⁵²Cf fission chamber and NE-213 proton recoil detector as shown were used for the ²⁵²Cf $\bar{\nu}$ measurement. For the ²³³U $R(E)$ measurement the tank through tube contained a fission chamber containing ²³³U and ²⁵²Cf and no graphite plugs. Figure from the work of Spencer et al. (Ref. 9).

²⁵²Cf $\bar{\nu}$ Measurement

For the ²⁵²Cf $\bar{\nu}$ measurement⁹ the tank through tube as shown in Fig. 10 contained a small NE213 proton recoil detector illuminated by the ORELA beam and a small ²⁵²Cf fission chamber positioned outside of the ORELA beam. Neutron multiplicities from ²⁵²Cf were counted for 80- μ s intervals after a fission event. Concurrently the NE-213 detector positioned at the tank center was used to calibrate Monte Carlo calculated tank efficiencies. In particular, a scattered neutron was signaled by its associated recoil proton pulse. Since both the incident neutron TOF and recoil proton pulse height were measured, both the scattered neutron energy and angle were determined. Since only one neutron is scattered per recoil event, these data measured the tank efficiency as a function of angle and neutron energy. These normalizations gave a tank efficiency of 95% for a ²⁵²Cf. In order to study the effects of delayed fission γ -rays, data were taken with the tank bias at both 1-MeV and 3-MeV equivalent γ -ray energy.

After detailed corrections for pileup, delayed fission γ -rays, background, other effects, and a comprehensive uncertainty analysis, a value of $\bar{\nu} = 3.773 \pm 0.007$ was obtained. Table 7 compares the ORNL value for $\bar{\nu}$ (total) = $\bar{\nu}$ (prompt) + 0.009 ± 0.004 with the current status^{49,50} of other prompt measurements.⁵¹⁻⁶⁰ The ORNL result has an uncertainty of 0.2%, nearly a factor of two smaller than any other measurement. Although the ORNL result is larger than most of the older manganese bath values listed in Table 7, it is in reasonable agreement with the recent result of Smith et al.⁵⁹ The uncertainty in ²⁵²Cf $\bar{\nu}$ contributes directly to the uncertainty in ²³³U $\bar{\nu}(E)$.

Table 7. $\bar{\alpha}(\text{total})$ Values for ^{252}Cf

Measurement	Current Value
Liquid Scintillator	
Asplund-Nilsson et al.	3.792 ± 0.040
Hopkins and Diven	3.777 ± 0.031
Boldeman	3.755 ± 0.016
Spencer et al.	3.782 ± 0.008
Manganese Bath	
White and Axton	3.815 ± 0.040
Axton et al.	3.743 ± 0.019
De Volpi and Porges	3.747 ± 0.019
Aleksandrov et al.	3.747 ± 0.036
Bozorgmanesh	3.744 ± 0.023
Smith et al.	3.764 ± 0.014
Boron Pile	
Colvin et al.	3.739 ± 0.021

^{233}U $\bar{R}(E)$ Measurement

For the ^{233}U $R(E)$ measurement of Gwin et al.⁷ the tank through tube, without the graphite plugs, contained a 5.1-cm-diameter fission chamber with foils fully illuminated by the ORELA beam. This fission chamber contained one deposit of ^{252}Cf and 18 deposits of 99.97% enriched ^{233}U with a total mass of 40 mg. Both ^{252}Cf and ^{233}U fission pulses initiated neutron counting for a 50 μs time period. The ^{233}U fission pulses were measured as a function of neutron TOF, and the tank bias was set at 0.9 MeV equivalent γ -ray energy.

Figure 11 compares the resulting prompt $\bar{R}(E)$ ratios to those from other measurements⁶¹⁻⁶⁴ and the ENDF/B-V evaluation.⁴⁸ The dashed line in Fig. 11 is a linear least-squares fit to the ORNL data from 0.5 keV to 2.0 MeV and is given by $R(E) = (0.6621 \pm 0.0010) + (0.030 \pm 0.002)E$. No energy dependence of $\bar{r}$ was observed in the eV region. The difference between the ORNL results and the Boldeman et al. results⁶⁴ from 0.1 to 1.0 MeV is not clearly understood. The final $R(E)$ measurement has been completed using fission chambers with ^{233}U , ^{235}U , ^{239}Pu , ^{241}Pu , and ^{252}Cf plates.⁸ This measurement gives $R(\text{thermal})$ for ^{233}U of 0.660 ± 0.002 , a value which is in excellent agreement with the 1975 Lermal evaluation,⁶⁵ 0.662 ± 0.002 , and the Boldeman and Dulton result,^{64,66} 0.6601 ± 0.0015 . The ORNL ^{252}Cf and ^{233}U results give prompt $\bar{\alpha}(\text{thermal}) = 2.490 \pm 0.009$ neutrons per fission. The ENDF/B-V evaluation contains 2.4873 neutrons per fission. This ORNL work has greatly reduced the uncertainty in $\bar{r}$ values. The resulting thermal $\bar{r}$ values have allowed a more satisfactory understanding of the 2200 m/s constants for fissile materials.⁶⁷

Thorium γ -Ray Production CS

Over the last decade a continuing measurement program at ORELA has produced the bulk of the absolute CS data for γ -ray production from neutron interactions in various materials including thorium. CS for γ -rays with energies from 0.3 to 10.5 MeV produced by neutrons with energies from 0.3 to 20.0 MeV have been measured for Li, C, N, O, F, Na, Mg, Al, Si, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Nb, Mo, Ag, Sn, Ta, W, Au, Pb, and Th. Complete details of the experimental method are given in Ref. 68. References for the various materials are listed in Ref. 10. These results are used primarily for shielding calculations; however, they are also used to constrain and verify model calculations and evaluations, particularly for non-fissile materials.

Figure 12 shows the γ -ray production CS for Th as measured by Morgan.¹⁰ Neutrons from ORELA operated at full power were incident on a 0.0050 b^{-1} slab of thorium metal mounted at 47 m and orientated at 45° . Gamma rays from the sample were detected in a heavily shielded $12.5 \times 12.5\text{-cm}$ NaI detector at 125° with respect to the incident beam direction. Each event was recorded in a two parameter array containing γ -ray pulse height as a function of the incident neutron TOF. After

Figure 11. Measured ratios $\bar{R}(E)$ of prompt $\bar{\alpha}(E)$ for ^{233}U to that from spontaneous fission of ^{252}Cf . The dashed line is a linear least-squares fit to the ORELA results. The smooth line is the prompt $\bar{r}$ ratio from the ENDF/B-V evaluation. Figure from the work of Gwin et al. (Ref. 7).

correcting for shadow-bar measured backgrounds, the spectra were unfolded in broad neutron energy bins using the FERD code⁶⁹ with measured response functions for the NaI detector. The resulting CS shown in Fig. 12 were corrected for finite sample effects using Monte Carlo techniques. The uncertainties shown in this figure are from counting statistics only. The neutron fluence was measured with a calibrated thick organic scintillator giving an additional overall normalization uncertainty of $\pm 1\%$.

^{232}Th Fission CS

Two measurements of the $^{232}\text{Th}(n,f)$ CS have been completed with ORELA. The low-energy measurement by Perez et al.¹¹ was performed at 24.9 m with a multiplate fission fragment ionization detector containing both thorium and ^{235}U . This measurement from 0.1 keV to 1.6 MeV filled the gap between the RLI-4S lead spectrometer results of Block et al.⁷⁰ and several published measurements in the MeV region. The high-energy measurement of Auchampaugh et al.¹² was performed at 41.7 m with a six-celled, sealed gas, fission fragment, scintillation detector also containing both thorium and ^{235}U . From this measurement the fission CS was determined from 0.9 to 10 MeV; however, the primary purpose of the work was to measure fission fragment anisotropies over the 1.4-, 1.6-, and 1.7-MeV structures in the fission CS. These structures are speculated⁷¹ to be undamped vibrational levels from a tertiary well of a triple humped fission barrier as calculated by Moller and Nix.⁷²

Figure 13 compares results from the two measurements over their overlap region from 0.9 to 1.6 MeV. Both fission CS's were normalized to that of ^{235}U and there is good agreement between the two data sets. These results satisfy the ^{232}Th fission CS need for reactor application. In addition, as shown in Fig. 13, the low-energy measurement of Perez et al.¹¹ shows a series of plateaus from 0.1 to 1.1 MeV which seems to present a challenge to current fission barrier calculations. No subthreshold fission CS in the keV region was observed within the sensitivity of the measurement.

The high-energy fission CS and fission fragment anisotropy measurement of Auchampaugh et al. is described in detail in Ref. 12. Anisotropy results were obtained from TOF spectra measured with three grids placed over the thorium deposits which limited the maximum fission fragment emission angles to 23.4° , 33.7° , and 51.7° . Both the fission CS and fission fragment anisotropy results over the 1.4, 1.6, and 1.7 MeV structures are consistent with those from recent calculations assuming a triple humped fission barrier for ^{232}Th .¹² However, the same calculations do not reproduce the plateaus shown on Fig. 13.

Figure 12. Double differential CS for γ -ray production from neutron bombardment of thorium. The CS are given in b at 125° as a function of incident neutron energy and outgoing γ -ray energy per steradian per MeV of γ -ray energy. The CS are the average over the neutron energy groups as shown. Figure from the work of Morgan (Ref. 10).

Figure 13. Fission CS of ^{232}Th from 0.1 to 1.5 MeV as measured at 25 m. The smoother curve from 0.9 to 1.6 MeV is the ORELA 40-m results of Auchampaugh et al. The various plateaus present a challenge to fission CS calculations. Figure from the work of Perez et al. (Ref. 11).

^{231}Pa Fission and Total CS's

The primary motivation for these measurements was also basic fission physics and the question of a possible third minimum in the fission barriers of the thorium region. These CS are also required for the $^{232}\text{Th}/^{233}\text{U}$ fuel cycle since ^{231}Pa plays an important role in the undesirable generation of ^{232}U .⁷³

^{231}Pa Fission CS

Two measurements of the ^{231}Pa fission CS relative to the ^{235}U fission CS were made by Plattard et al.¹³ The high resolution measurement at 41.7 m spanned neutron energies from 0.1 to 12.0 MeV. The low resolution measurement at 18.3 m spanned neutron energies from 0.4 eV to 10 keV. Both measurements detected fission events with the gas scintillator chamber employed for the ORELA 40-m ^{232}Th results. This fission chamber contained 265 mg of ^{231}Pa with less than 0.1 ppm of fissile contaminants. One cell of the fission chamber contained 130 mg of ^{235}U for normalization.

Figure 14 compares the resulting ^{231}Pa fission CS from 0.12 to 0.42 MeV with the results of Sicre.⁷⁴ The most striking feature of this CS is the considerable amount of fine structure observed with the high resolution obtainable from ORELA. Together, this high resolution fission CS and the Sicre⁷⁴ low resolution fission fragment angular distributions at least partially support, but by no means prove, the speculation that some of the fractionated vibrational resonances shown in Fig. 14 arise from a third minimum in the ^{232}Pa fission barrier.¹³

From the low-resolution measurement fission widths were obtained for 80 resonances below 58 eV. The average fission width for resonances below 15 eV is about $8 \mu\text{eV}$ which is three or four orders of magnitude smaller than those generally measured for fissile nuclei. The fission resonances seem uniformly distributed over energy and not clustered into groups. These results are consistent with direct penetration through a double humped fission barrier which may or may not have a shallow minimum in the second barrier.¹³

^{231}Pa Total CS

In order to obtain more accurate resonance parameters, Hussein et al.¹⁴ measured neutron transmission spectra through 0.00034 and 0.00079 b^{-1} samples of ^{231}Pa . The samples consisted of packed Pa_2O_5 powder and were cooled to liquid nitrogen temperature. Measurements were made at both 18 m and 80 m using a 12.7-mm-thick Li-glass detector. Using the R-matrix code MULTI,⁷⁵ resonance energies and neutron widths were

Figure 14. Fission CS of ^{231}Pa between 0.12 and 0.42 MeV. The ORELA data are plotted with error bars and were measured at 40 m with a gas scintillator chamber. The data of Sicre are plotted without error bars. Figure from the work of Plattard et al. (Ref. 13).

obtained for 137 resonances below 120 eV. These resonances give an average capture width of 40 ± 2 MeV, an average s-wave level spacing of 0.47 ± 0.05 eV, and an s-wave strength function of $0.90 \pm 0.10 \times 10^{-4}$. These results will allow an improved evaluation for ^{231}Pa .

^{234}U Fission and Total CS

Both the total and fission CS of ^{234}U were measured from 3 eV to 8.9 MeV by G. D. James et al.¹⁵ This work was performed primarily to study the intermediate structure in the ^{234}U fission CS resulting from a double humped fission barrier. Fission events were detected at 20 m with a multi-plate fission fragment ionization chamber containing 181 mg of ^{234}U . This chamber also contained 20 mg of ^{235}U which was used to determine the shape of the incident neutron flux. The overall CS normalization was determined from previous ^{234}U measurements. The total CS was measured at 78 m with a 12.7-mm-thick Li-glass detector using a 0.00645 b^{-1} sample cooled to liquid nitrogen temperature and consisting of 5.69 gm of ^{234}U in the form of U_3O_8 .

Neutron widths were extracted from the transmission spectrum using the Atta-Harvey area analysis program,⁷⁶ assuming a constant Γ_γ of 40 meV. Fission widths were obtained from the fission peak areas. Altogether parameters for 118 resonances below 1.5 keV were obtained which yielded a class I level spacing of 10.6 ± 0.5 eV and s-wave strength function of $0.86 \pm 0.11 \times 10^{-4}$. In addition, two class II resonances at 580 and 1227 eV were studied in detail and a class II level spacing of 2.1 ± 0.3 keV was derived.

Summary and Conclusions

Total, capture, fission, $\bar{\nu}$, and γ -ray production measurements important to the $^{232}\text{Th}/^{233}\text{U}$ fuel cycle have been completed using the ORELA facility. These measurements, relevant to both thermal and fast reactors, were done in response to specific data discrepancies, as part of generic programs, and for basic physics studies.

The most important nuclear data for $^{232}\text{Th}/^{233}\text{U}$ fuel cycle thermal reactor application are the CS in the thermal region and corresponding 2200 m/s constants and the parameters for the low energy ^{232}Th resonances. The ORNL ^{232}Th $\bar{\nu}$ and ^{233}U $R(E)$ work gives a prompt $\bar{\nu}$ (thermal) of 2.490 ± 0.009 n/fission. This result of unprecedented accuracy allows a much more satisfactory understanding of the 2200 m/s constants for ^{233}U . ORELA transmission measurements have provided needed low-energy total CS's for ^{233}U and ^{232}Th . The ORELA ^{232}Th transmission and capture measurements give the most precise parameters for the first four s-wave resonances which provide 68% of the dilute capture resonance integral and also give both a larger average capture width and a larger s-wave strength function than previously measured. These results provide better agreement between measured differential and integral capture rates and should be confirmed with additional measurements.

In addition, the thorium γ -ray production measurements provides needed CS's for shielding. The ^{232}Th fission work and the ^{231}Pa and ^{234}U fission and total CS work provide needed data for both for application and for understanding the fission process. The ORELA facility has made a substantial contribution to thorium cycle nuclear data.

There is, however, a clear need for additional thorium fuel cycle measurements, particularly on ^{233}U . The capture CS of ^{233}U has not been measured from 2 keV to 30 keV. This gap in the basic nuclear data should be filled. Below 0.5 eV the ^{233}U capture fission and total CS are inconsistent.⁴⁷ This discrepancy should be resolved. The shape of the ^{233}U fission neutron spectrum, and also that of ^{233}U and ^{239}Pu , is a major source of uncertainty.²⁵ This uncertainty should be substantially reduced with definitive measurements.

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