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New Evaluation of the ²³²Th Resonance Parameters in the Energy Range 0 to 4000 keV

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Abstract

Neutron resonance parameters of ²³²Th were obtained from a Reich-Moore SAMMY analysis of high-resolution neutron transmission measurements performed at the Oak Ridge Linear Accelerator (ORELA) by Olsen et al. in 1981 and of high-resolution neutron capture measurements performed recently at the Geel Linear Accelerator (GELINA, Belgium) by Schillebeeckx et al. and at nTOF (CERN, Switzerland) by Aerts et al. The ORELA data were analyzed previously by Olsen using the Breit-Wigner multilevel code SIOB, and their results were used for the ENDF/B-VI evaluation. In our new analysis of the Olsen neutron transmissions using the modern computer code SAMMY, better accuracy is obtained for the resonance parameters by including recent experimental neutron capture data in the experimental data base. The experimental data base and the method of analysis are described in the report. Neutron transmissions and capture cross sections calculated with the resonance parameters are compared to the experimental values. A description is given of the statistical properties of the resonance parameters. The new evaluation produces a decrease in the capture resonance integral, and improves the prediction of integral thermal benchmarks.

KEYWORDS: resonance parameters, neutron cross section, statistical properties

1. Introduction

The most recent evaluation of ²³²Th resonance parameters was performed in 1982 by D. K. Olsen [1] for ENDF/B-VI. The evaluation was mainly based on parameters obtained from the analysis of transmission measurements performed by Olsen et al., [2] Ribon, [3] and Rahn et al. [4] and of capture measurements performed by Forman et al. [5] and Macklin et al. [6] with additional information taken from the evaluations made by Derrien, [7] and Keyworth and Moore. [8] The Olsen evaluation used a smooth background cross section to represent the contribution of the truncated external resonances and of the part of the capture cross section due to the missed p-wave resonances. The aim of the present work was to complete the Olsen evaluation with the computer code SAMMY [9] by adding to the experimental data base the recent capture measurements performed at Geel Linear Accelerator (GELINA, Belgium) by Schillebeeckx et al. [10] and at the neutron time-of-flight(n_TOF) facilities of the Conseil Europeen pour la Recherche Nucleaire (CERN, Geneva) by Aerts et al. [11] In the present paper, a brief description of the experimental data base is given. The method of analysis is outlined. Examples of neutron transmission and cross section calculated with the evaluated resonance parameters are compared

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to the corresponding experimental data, and statistical properties of the resonance parameters are described.

2. The experimental data base

Two independent series of transmission measurements were performed by Olsen et al. at ORELA. The first series of measurements were performed at the 22-m flight path in the neutron energy range 0.01 to 16 eV, with ²³²Th metallic samples of thicknesses ranging from 0.0387 to 0.193 at/b. The second series were performed at the 40-m flight path in the neutron energy range 10 eV to about 100 keV with ²³²Th metallic samples of thicknesses ranging from 0.000161 to 0.193 at/b. The GELINA capture measurements were performed at a 58.4-m flight path in the neutron energy range from 10 eV to about 100 keV; the thickness of the ²³²Th sample was 0.00318 at/b. The CERN-n-TOF measurements were performed at a 185-m flight path in the neutron energy range 1 eV to 1 MeV; the thickness of the sample was 0.00411 at/b. Both GELINA and n_TOF effective capture cross sections could be normalized by using the saturated resonances of the low-energy part of the data.

Several other experimental results were considered for evaluation in the thermal and lowenergy range: (1) Chrien et al. [12] capture cross sections (1979); (2) Lundgren [13] capture cross sections (1968); (3) Little et al. [14] total cross sections (1981); (4) Kobayashi et al. [15] total cross sections (1984). In the present evaluation, all experimental capture data in the thermal region were re-normalized to 7.35 b at 0.0253 eV. The value of 7.35 b was the result of the evaluation performed recently by Trkov. [16] Renormalization of the thermal total cross section was not needed because the experimental data of Olsen, Little, and Kobayashi were results of absolute measurements leading to cross sections with accuracy better than 1%. The capture cross section data of Chrien et al. and of Lundgren et al. in the thermal energy range were corrected for self-shielding and multiple scattering effects by the authors of the experiments. They agree in shape in the energy range up to about 1 eV neutron energy but disagree at higher energy.

3. Conditions of the analysis

The analysis code SAMMY uses the Reich-Moore formalism to calculate the cross sections in the resolved resonance energy range and Bayes method to fit the experimental data. The fit of the experimental data base is performed sequentially, the covariance matrix generated from the fit of a data set is used as input to fit the next data set. The calculated cross sections are modified by Doppler and experimental resolution broadening, and by other experimental effects which are selfshielding and multiple scattering in the samples, background, and normalization corrections. Doppler broadening of the resonances was calculated by the Free Gas Model (FGM) of Lamb. [17] Because all experimental data were taken at room temperature with metallic samples, the value of 298 ± 5 K was used for the effective temperature of the samples. This value was obtained from a Debye temperature of 165 K recommended by Ribon. [3] The usual components of the Time-Of-Flight (TOF) experimental resolution, that is the neutron burst width, the TOF channel width and the distribution of the flight path length, were treated by the Gaussian-distribution option in SAMMY. The corresponding parameters are generally well known. Another part of the experimental resolution relates to the neutron slowing down and to the detectors (neutron detector or γ -rays detector). This part, which should be described by an asymmetrical function, is represented in SAMMY by the exponential $\exp(-t/\Delta t)$ where Δt is the so-called exponential folding

width in microseconds. The exponential folding width is not well known and could vary with neutron energy. In the present evaluation this parameter was obtained by preliminary fits of isolated resonances or group of resonances in each experimental data set.

The contribution of the resonances external to the energy range 0 eV to 4 keV was approximated by two fictitious large resonances at -2 keV and 6 keV. The parameters of these resonances and the correlated effective scattering radius R' were obtained by preliminary SAMMY fits of the experimental transmission data. The value of 9.69 ± 0.03 fm was obtained for R', consistent with the value of 9.71 ± 0.08 obtained by Olsen from a SIOB analysis of the same experimental data, [2] the value of 9.65 ± 0.25 fm obtained by Kobayashi [18] from measured total cross section near 24 keV, and the value of 9.65 ± 0.10 fm recommended by Ribon [3] from a resonance analysis of a thick-sample transmission data.

ENDF/B-VI resonance parameters values were used as prior values in this analysis. This set of parameters contained 241 s-wave resonances and 192 p-wave resonances in the neutron energy range 0 to 4000 eV. The set of p-wave resonances was far from complete since the average level spacing was 20.8 eV compared to 5.5 eV inferred from the s-wave average spacing. Olsen has shown [1] that the ²³²Th resonance parameters that he evaluated for ENDF/B-VI produced average capture cross sections too small by a factor of 2 at 2 keV and by a factor of 4 at 4 keV, compared to the values calculated from a reasonable p-wave strength function. In the Olsen evaluation, the missing contribution was compensated by a smooth background cross section. To avoid the use of a background cross section in the present evaluation, a complete set of p-wave resonances was generated. This set, in addition to the p-wave resonances present in ENDF/B-VI and identified in the experimental neutron transmission and capture data, included fictitious resonances whose parameters were chosen at random in order to roughly comply with the Wigner distribution of level spacings and the Porter-Thomas distribution of reduced neutron widths. These resonances are too small to appear in the statistical fluctuations of the experimental data or are hidden by the large resonances. This method of analysis was used previously for the ²³⁸U resonance parameter evaluation in the energy range 0 to 20 keV [19,20].

4. Results of the analysis: transmission and cross section data

Figures 1 and 2 show the data in the thermal and low-energy range. Below 0.1 eV, important deviations are observed between the calculated total cross section and the experimental values. Attempts to reproduce the average total cross section by negative energy resonance parameters in the energy range near 0.0253 eV have failed. In this energy range, all the experimental data are in good agreement. They show important structures due to the Bragg diffraction of neutrons. Extinction effects, which are microcrystalline effects, could cause a reduction of the coherent Bragg scattering and therefore a diminution of the measured total cross section.

The calculated capture cross section at 0.0253 eV is 7.35 b, which is the value recommended by Trkov, and could be adjusted by small variations of the parameters of the resonance at -3.51 eV. In the energy range above 1 eV, the Chrien capture data are larger than those of Lundgren and n_TOF. The calculated cross section of the present evaluation follows the shape of Lundgren and n_TOF. The ENDF/B-VI evaluation follows the shape of Chrien; this agreement was obtained by Olsen by using a smooth background and not by the resonance parameters.

Figure 3 shows an example of the SAMMY fits to experimental transmission in the energy range 1500 to 1750 eV. In general, agreement between the average experimental transmission and the calculated values is better than 1.5%, corresponding to 0.06 b in the effective total cross section

of the thickest sample, which is within the experimental error bars given by Olsen. Example of SAMMY fit of the effective capture cross sections is shown on Fig. 4. The average effective capture cross sections calculated with the resonance parameters are compared to the corresponding experimental data in Table 1. The average capture cross sections were also obtained by Aerts et al. [11],[21] in the energy range above 1 keV from the same experimental data by using the computer code SESH [22] for calculation of the self-shielding and multiple scattering corrections. When averaged over the energy range 1 to 4 keV, the Aerts results agree within 1% with the SAMMY calculation. However, when compared in energy intervals of 500 eV, about \pm 5% differences are observed. These differences are due to the fact that the code SESH calculates the corrections from resonance parameters generated by the Monte-Carlo method, while SAMMY calculates the corrections from the current resonance parameters.

Energy range (eV)	GEEL effective cross section Exp.(b) Calc.(b) Diff.(b)			<u>N-TOF effective cross section</u> Exp.(b) Calc.(b) Diff.(b)		
20–50	7.65	8.86	1.21	7.316	7.486	0.170
50-100	6.09	6.37	0.28	5.357	5.478	0.121
100-500	5.19	5.38	0.19	4.627	4.487	-0.140
500-1000	2.90	2.62	-0.30	2.443	2.484	0.041
1000-1500	2.23	1.88	-0.35	1.817	1.805	-0.012
1500-2000	2.37	2.00	-0.37	1.946	1.931	-0.015
2000-2500	1.99	1.61	-0.36	1.557	1.563	0.006
2500-3000	1.90	1.56	-0.36	1.513	1.506	-0.007
3000-3500	1.63	1.27	-0.36	1.243	1.237	-0.006
3500-4000	1.45	1.10	-0.35	1.086	1.081	0.006

Table 1: Average experimental and calculated effective capture cross sections

4. Results of the analysis: the resonance parameters

The present evaluation uses 243 s-wave resonances and 667 p-wave resonances for the description of the cross sections in the energy range 0 to 4000 eV. In addition to the large fictitious resonances at -2 keV and 6 keV, which account for the far-off external resonances, a ladder of 7 resonances at negative energy and of 7 resonances in the energy range 4.0 to 4.1 keV was used to improve the fit in the thermal energy range and just below 4000 eV, respectively. The angular momentum assignments of the resonances seen in the experimental data were the same as those in ENDF/B-VI. At the beginning of this analysis, the p-wave spin assignments were chosen randomly with a two spin state population ratio of about 2. In some cases, the fit of the experimental capture data required reassignment of the spin. In the final set of p-wave resonances, 236 resonances have spin 1/2, and 431 resonances have spin 3/2.



Figure 1: Experimental and calculated total cross sections in the thermal and low-energy range.



Figure 2: Experimental and calculated capture cross sections in the thermal and low-energy range.

Statistical properties of the resonances parameters agree reasonably well with the Wigner distribution of the level spacings and with the Porter-Thomas distribution of the reduced neutron widths. Examples are given in Fig. 5 showing the distribution of the level spacings. About 95% of the resonances assigned s-wave are seen in the experimental data. Only 30% of the resonances assigned p- wave are seen in the experimental data. The distribution of the reduced neutron widths

of the resonances having 99% chance to be s-wave, and of the p-wave reduced neutron widths larger than 0.002 eV are shown in Fig. 6 and Fig. 7, with the corresponding Porter-Thomas distribution. Accurate values of the average level spacing and of the neutron strength functions can be inferred from these distributions (see Table 2).

The capture widths of the large s-wave resonances were allowed to vary in the SAMMY fits in all energy ranges of the analysis. Above 400-eV neutron energy, the results show strong fluctuations and a tendency to decrease on average. To avoid the effect of the systematic deviations, the average capture width should be evaluated in the energy range 0 to 400 eV only. The weighted average value calculated by SAMMY is 25.65 ± 0.10 meV from transmission data only and 25.24 ± 0.08 meV from transmission and capture data. These values compare to the value of 25.2 ± 0.5 meV obtained by Olsen from the transmissions in the same energy range and to the value of (24.4 ± 2.0) meV evaluated by Olsen for ENDF/B-VI.

The capture resonance integral calculated from the present evaluation is 82.34 b in the energy range 0.5 eV to 4000 eV, compared to 83.62 b calculated from ENDF/B-VI. Both values agree with the evaluation of Greneche [23], 83.42 ± 1.9 b, in the same energy range.



Figure 3: Results of SAMMY fits to the Olsen experimental transmission data in the energy range 1500 to 1750 eV. The solid lines represent neutron transmission calculated with the resonance parameters.



Figure: 4 Effective capture cross sections in the energy range 650 to 750 eV. The solid lines represent the corresponding data calculated with the resonance parameters.



Figure 5: Differential distribution of the resonance spacings compared to the Wigner distributions.



Figure 6: Integral distributions of the reduced neutron widths of the resonances having 99% of chance to be s-wave in the energy range 0 to 2 keV (left curve) and in the energy range 0 to 4 keV (right curve).



Figure 7: Integral distribution of the p-wave resonance reduced neutron widths larger than 0.002 eV.

	Average level spacing (eV)	Neutron strength function ($\times 10^4$)	Average capture width (meV)
s-wave p-wave	$\begin{array}{c} 16.5 \pm 0.3 \\ 5.52 \pm 0.20 \end{array}$	$\begin{array}{c} 0.829 \pm 0.075 \\ 1.48 \pm 0.15 \end{array}$	25.25 ± 1.00

Table 2: Recommended values of average resonance parameters

6. Conclusions

Neutron resonance parameters of ²³²Th were reevaluated in the energy range 0 to 4 keV from a Reich-Moore SAMMY analysis of the most recent experimental neutron transmission and neutron capture data. The experimental data base consisted of the transmission measurements performed at ORELA by Olsen et al. in 1980, effective capture cross section measurements performed at GELINA by Schillebeeckx et al. in 2002, and the effective capture cross section measurements performed to TOF by Aerts et al. in 2003. The previous evaluation was performed by Olsen for ENDF/B-VI in 1981. The present evaluation brings important improvements concerning the following points:

1. Two recent high-resolution effective capture cross section measurements (Geel and nTOF) were available for the present evaluation. By taking advantage of the ability of SAMMY to accurately calculate multiple scattering effects in the thorium samples, we were able to calculate the average capture cross section to better than 3% accuracy.

2. In the energy range 1.0 to 10 eV, the new resonance parameters reproduce the experimental capture cross sections inferred from the nTOF measurements, in agreement with the early measurements of Lundgren; the ENDF/B-VI evaluation added a smooth cross section to the contribution of the resonance parameters to agree with the measurements of Chrien et al. The capture cross section calculated in the present evaluation is about half as large as the ENDF/B-VI value and is closer to ENDF/B-V which was based on Lundgren data in the energy range 0.1 to 3 eV.

3. The contribution of the external resonances is fully represented by two fictitious resonances at -2 keV and 6 keV; in the ENDF/B-VI evaluation this contribution was included as a smooth background cross section.

4. The present set of resonances is complete in that the statistical properties of the parameters agree with the Wigner distribution of the level spacings and with the Porter-Thomas distribution of the reduced neutron widths. This, particularly, allows the p-wave neutron capture cross section to be fully represented by the resonance parameters. In the ENDF/B-VI evaluation, compensation for the missing p-wave resonances was calculated from the p-wave strength function and added as smooth background cross section.

The covariance matrix for the resonance parameters was also generated in the present evaluation. Results are presented in another paper at this conference[24].

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