

Evaluation of the ^{232}Th Neutron Cross Sections between 4 keV and 140 keV

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Abstract. An evaluation of the ^{232}Th total and capture cross sections has been performed in the energy region between 4 keV and 140 keV. The evaluation results from a simultaneous analysis of capture, transmission and self-indication measurement data, including the most recent capture cross section data obtained at the GELINA facility of the Institute for Reference Materials and Measurements at Geel (B) and at the n-TOF facility at CERN (CH). The experimental data has been analysed in terms of average resonance parameters exploiting two independent theoretical approaches – the Characteristic Function model and the Hauser-Feshbach-Moldauer theory. The resulting parameters are consistent with the resolved resonance parameters deduced from the transmission measurements of Olsen et al. at the ORELA facility.

INTRODUCTION

The thorium-uranium fuel cycle is very appealing because the natural resources of thorium considerably exceed those of uranium and because this cycle limits the build-up of highly radioactive transuranium nuclides. Furthermore Accelerator Driven Systems, based on the Th – U fuel cycle, are studied to incinerate the waste of the first generation of nuclear power plants¹.

The nuclear data relevant to the thorium fuel cycle do not have the same level of accuracy as those of the U – Pu fuel cycle. In particular, an analysis of the available nuclear data reveals that the status of the $^{232}\text{Th}(n,\gamma)$ cross-section is far from the requested 2 % uncertainty level². Recently, the need of improved thorium nuclear data initiated a Co-ordinated Research Project (CRP) “Evaluated Nuclear Data for the Th–U Fuel Cycle” organized by the International Nuclear Data Committee (INDC) of the IAEA in Vienna.

In this paper we present an evaluation of the ^{232}Th average total and capture cross-section in the energy region from 4 keV up to 140 keV. The evaluation results from a simultaneous analysis of capture,

transmission and self-indication measurements, including the most recent capture data of Borella et al.³ and Aerts et al.⁴, which were obtained at the time of flight facilities GELINA at Geel (B) and n-TOF at CERN (CH), respectively.

$^{232}\text{Th}(n,\gamma)$ CROSS SECTION MEASUREMENTS AT GELINA

Recently, $^{232}\text{Th}(n,\gamma)$ cross section measurements for the unresolved resonance region (URR) were carried out at a 14.36 m measurement station of GELINA³. The capture events were detected by two C_6D_6 liquid scintillators and the shape of the neutron flux was monitored by a ^{10}B ionisation chamber. A pulse height weighting function was used to provide proportionality between the detection efficiency for a neutron capture event and the total energy released in the capture cascade. The weighting function was determined by Monte Carlo simulations⁵. The data were internally normalized at the peak of the well-isolated and nearly saturated resonance at 23.5 eV, with about 1 % peak transmission. As compared to a normalization to a reference sample such as gold, an internal

normalization has the advantage of eliminating systematic uncertainties due to variations of detector and accelerator operating conditions and due to the possible errors introduced by the weighting function when dealing with different gamma spectra shapes and sample thicknesses.

In Ref. 3 the influence of the weighting function and the normalisation was investigated by analysing the data with different weighting functions and threshold levels. Moreover, the influence of the resonance parameters of the 23 eV resonance on the normalisation was verified. It was shown that using an internal normalisation the systematic uncertainty related to the normalization and weighting function is limited to less than 1%.

To deduce the average capture cross section from the experimentally determined capture yield one has to correct for the self-shielding and multiple scattering effects. These corrections were studied by both analytical expressions⁶ and Monte Carlo simulations. For the analytical expressions we used the HARFOR code^{7,8} to calculate the self-shielding corrections. This code was primarily developed for the parameterization of cross sections in the URR (see the next section). We also used the Monte Carlo code SESH, developed by Fröhner⁹. This code takes into account the sample geometry and accepts average resonance parameters as input parameters (i.e. the average resonance spacings, neutron strength functions and average radiation widths) to create resonance structured cross sections. In addition, the correction factors were calculated with two other Monte Carlo simulation codes, i.e. SAMSMC¹⁰ and MCNP 4C2¹¹. The resonance parameters to create resonance structured cross sections for SAMSMC were generated by the LADDER module, implemented in the SAMMY package. The results with MCNP were obtained using the probability tables which are incorporated in the code.

In Figure 1 we compare the results for a 0.5 mm thick metallic ²³²Th disc, with a 80 mm diameter. The self-shielding factor is expressed as:

$$\frac{(1 - e^{-n\sigma_t})}{n\sigma_t} \quad (1),$$

and the multiple scattering correction is represented by μ . The data in Figure 1 reveal that the corrections for the sample used at GELINAc can be determined within about 0.5 %.

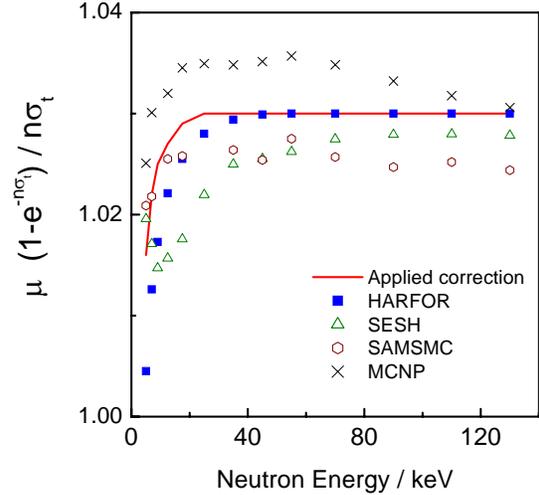


FIGURE 1. The self-shielding and multiple scattering correction as a function of neutron energy for the ²³²Th sample used in Ref. 3.

EVALUATION IN THE UNRESOLVED RESONANCE REGION

In the URR average cross sections can be parameterized by statistical models, implemented in e.g. the FITACS¹² and HARFOR^{7,8} code. The former is based on the Hauser-Feshbach statistical reaction theory including width - fluctuations following the Moldauer prescription. The latter, which was developed at the INRNE Sofia (BG), relies on the characteristic function of the R-matrix element distributions and uses the Reich-Moore approximation of the R-matrix theory. This model provides an exact relationship between the average partial cross sections and the average R-matrix element, and supplies analytical expressions for the related cross section functionals, such as transmission and self-shielding factors. Parameters for determination in the URR are the mean level spacing for s-wave resonances D_0 , the neutron strength functions S_ℓ , the average radiation widths $\langle \Gamma_\gamma \rangle_\ell$ (with ℓ the orbital angular momentum of the incoming neutron) and the effective scattering radius R' . To determine reliable average resonance parameters one needs besides the capture cross-section an additional complementary data set¹³ such as the results of transmission and self-indication function measurements in the URR.

TABLE 1. The Evaluated Average Resonance Parameters of ^{232}Th for the URR

R' / fm	D_0 / eV	S_l ($\times 10^{-4}$)			$\langle \Gamma_\gamma \rangle_t$ / meV			Ref.
		0	1	2	0	1	2	
9.43	17.385	0.94	2.15*	1.15	21.3	21.3	21.3	Maslov et al. ²⁶
9.43 (0.2)	17.60 (0.50)	0.94	1.96 (0.20)	1.24 (0.12)	24.88 (0.30)	25.50 (0.20)	24.88	HARFOR ^{7,8}
9.52 (0.2)	17.60	0.94	1.83 (0.02)	1.26 (0.05)	24.08 (0.24)	24.52 (0.20)	24.08	FITACS ¹²

Since the present version of HARFOR does not include the in-elastic scattering reaction, the application of the code is limited to about 50 keV. We derived the mean level spacing D_0 , the average radiation widths $\langle \Gamma_\gamma \rangle_{0,1}$, the neutron strength functions S_1 and S_2 and the effective scattering radius R' from a simultaneous fit of our capture data together with the transmission data of Uttley et al.¹⁵ and the self-shielding factors measured by Oigawa et al.¹⁴. The radiation width for d-wave resonances was fixed at the value for s-wave resonances and the neutron strength function for s-wave resonances $S_0 = 0.94 \cdot 10^{-4}$, as deduced from RRR. The results are listed in Table 1 and compared with the evaluation of Maslov et al.²⁶.

We also used the SAMMY code¹², which incorporates the FITACS algorithms, to parameterize the average capture cross section between 4 and 140 keV. We fitted our capture data together with the capture data obtained at the n-TOF facility of CERN and cross sections of Ref. 15-23. In the analysis we also adjusted a normalisation factor, which we varied within the uncertainty limits quoted in the corresponding reference. A summary of the different data sets together with the adjusted normalisation factors are given in TABLE 2. In Fig. 2 we compare the results of the evaluation with the experimental data.

Table 2 and Figure 2 demonstrate that there is a very good agreement between the GELINA and n-TOF data, both in shape and absolute value. This good agreement confirms again that the use of an internal normalisation resonance together with a pulse height weighting function accounting for the sample characteristics reduces systematic errors.

For a good description of the capture cross section we had to decrease the radiation widths by about 4 %, compared to the values deduced by HARFOR. From a

given set of average resonance parameters both codes produce an identical total cross section. However, differences related to the treatment of neutron width fluctuation corrections result in a different capture cross section and therefore different values for the deduced radiation widths.

The average resonance parameters resulting from our analysis are consistent with the values deduced from a statistical analysis of resolved resonance parameters. The average radiation width for p-wave neutrons does almost not differ from the one for s-wave neutrons, as already suggested by Olsen et al.²⁴. For the effective scattering radius we obtain values $R' = 9.4$ (0.2) and 9.5 (0.2) fm. This value relates to a distant level parameter $R_\infty = -0.128$, which is consistent with the results of optical model calculations. The neutron strength function for p-wave resonances is also close to the value reported by Corvi et al.¹⁶.

TABLE 2. The Data Sets Used in the Evaluation and the Adjusted Normalisation Factors.

	Cross Section Data	Ref.	Normalisation
Total	Gregoriev et al.	16	1.108 (0.006)
	Phoenitz et al.	17	1.042 (0.005)
	Iwasaki et al.	18	1.023 (0.004)
	Uttley et al.	15	1.018 (0.005)
Capture	Borella et al.	2	1.000 (0.001)
	Aerts et al.	3	1.005 (0.004)
	Kobaiashy et al.	19	0.915 (0.004)
	Phoenitz et al.	20	1.023 (0.004)
	Macklin et al.	21	0.944 (0.003)
Inel.	Vertebnyj et al.	22	1.258 (0.022)
	Fujita et al.	23	1.070 (0.046)

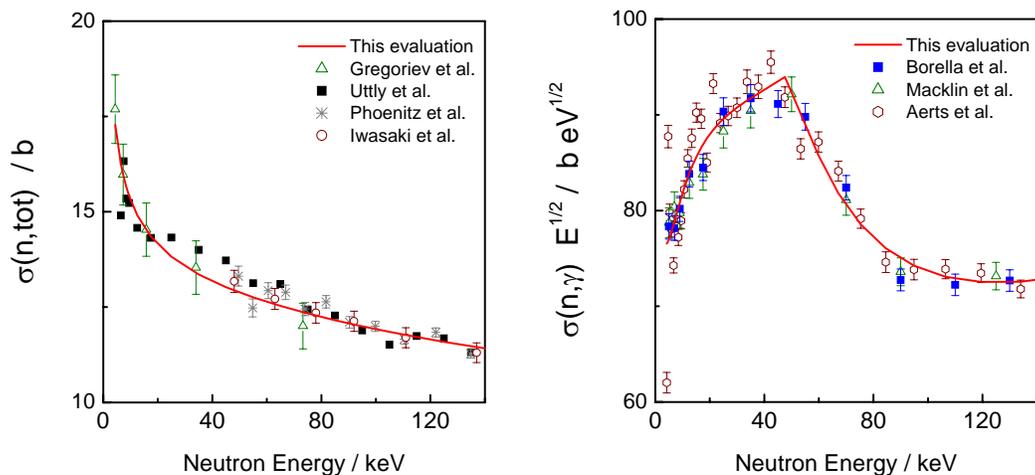


FIGURE 2. A comparison of the experimental total (left) and capture (right) cross-sections with the results of the evaluation using FITACS. The capture cross section is multiplied by $E^{1/2}$.

CONCLUSIONS

A new evaluation of the ^{232}Th average total and capture cross section has been performed from 4 keV up to 140 keV, using the statistical models implemented in the FITACS and HARFOR codes. The evaluation includes the most recent data obtained at GELINA and n-TOF facility of CERN. The resulting average resonance parameters for the unresolved resonance region are consistent with the resolved resonance parameters reported by Olsen et al.²⁴ and Corvi et al.²⁵.

REFERENCES

- Bowman, C., *Annu. Rev. Nucl. Part. Sci.*, **48**, 505 (1998).
- Kuzminov, B.D., and Manokhin, V.N., Proc. Int. Conf. Nuclear Data for Science and Technology, Trieste, Italy, 19-24 May 1997, Part. II, (1997), p. 1167
- Borella, A., Volev, K., Schillebeeckx, P., Corvi, F., Koyumdjieva, N., Janeva, N., and Lukyanov, A., *Nucl. Sci. Eng.*, (accepted).
- Aerts, G., Günsing, F., and the n-TOF collaboration, *these proceedings*.
- Abbondanno, U., et al., *Nucl. Instr. Meth.*, **A521**, 454 (2004).
- Lukyanov, A., Koyumdjieva, N., Janeva, N., Volev, K., Brusegan, A., Schillebeeckx, P., Lobo, G., and Corvi, F., in *Mathematical and Computational Sciences: A Century in Review*, Gatlinburg, Tennessee, April 5-11, (2003)
- Koyumdjieva, N., Janeva, N., and Lukyanov, A., *Z. Phys.*, **A353**, 31 (1995).
- Koyumdjieva, N., Janeva, N., and Volev, K., *Nucl. Sci. Eng.*, **137(2)**, 194 (2001).
- Fröhner, F., Report GA-8380, Gulf General Atomic (1968).
- Larson, N., And Volev, K., “Validation of Multiple-Scattering Corrections in the Analysis Code SAMMY” in *PHYSOR 2002*, Seoul, Korea, October 7 – 10, (2002).
- Briesmeister, J., LA-13709-M, (2000).
- Larson, N., ORNL/TM-9179/R6 ENDF-364, July 2003
- Fröhner, F., Haddad, E., Lopez, W., and Friesenhahn, S., in *Neutron Cross Sections and Technology*, Washington D.C., **vol. I**, pp. 55-66, (1966).
- Oigawa, H., Fujita, Y., Kobayashi, K., Yamamoto, S., and Kimura, I., *J. Nucl. Sci. Technol.*, **28(10)**, 879 (1991).
- Uttley, C., Newstead, C., and Diment, K., “Neutron Strength Function Measurements in the Medium and Heavy Nuclei”, Proc. Conf. Nuclear Data for Reactors, 17 – 21 October, Paris, **vol. 1**, p. 165, (1966).
- Grigoriev, Yu., et al., EXFOR : entry num. 41372.
- Poenitz, W., Whalen, J., and Smith, A., *Nucl. Sci. Eng.*, **78**, 333 (1981).
- Iwasaki, T., Baba, M., Hattori, K., Kanda, K., Kamata, S., and Hirakawa, N., in *Contribution to the Specialists’ Meeting on Fast Neutron Scattering on Actinide Nuclei*, R-NEANDC, EXFOR: entry num. 21766, (1981).
- Kobayashi, K., Fujita, Y., and Yamamuro, N., *J. Nucl. Sci. Technol.*, **18 (11)**, 823 (1981).
- Poenitz, W., and Smith, D., ANL/NDM-42, (1978).
- Macklin, R., and Winters, R.R., *Nucl. Sci. Eng.*, **78**, 110 (1981).
- Vertebnyj et al., IAEA-410, p.257, EXFOR: entry num. 40929, (1986).
- Fujita, Y., et al., EXFOR entry num. 12858.
- Olsen, D., Ingle, R., and Portney, J., *Nucl. Sci. Eng.*, **82**, 289 (1982).
- Corvi, F., Pasquariello, G., and Van Der Veen, T., “p-wave assignment of ^{232}Th resonances” in *Proc. Int. Conf. Neutron Physics and Nuclear Data*, Harwell, UK, p. 712, (1978).
- Maslov, V., et al., INDC(BLR)-016, IAEA, (2003).