

New fit of thermal neutron constants (TNC) for $^{233,235}\text{U}$, $^{239,241}\text{Pu}$ and $^{252}\text{Cf(sf)}$: Microscopic vs. Maxwellian data

Vladimir G. Pronyaev¹, Roberto Capote^{2,a}, Andrej Trkov², Gilles Noguere³, and Anton Wallner⁴

¹ PI Atomstandart at SC Rosatom, Moscow, Russian Federation

² NACP-Nuclear Data Section, International Atomic Energy Agency, Vienna, Austria

³ CEA, DEN, DER, SPRC, Cadarache, 13108 Saint-Paul-lez-Durance, France

⁴ Australian National University, Department of Nuclear Physics, Canberra, Australia

Abstract. An IAEA project to update the Neutron Standards is near completion. Traditionally, the Thermal Neutron Constants (TNC) evaluated data by Axton for thermal-neutron scattering, capture and fission on four fissile nuclei and the total nu-bar of $^{252}\text{Cf(sf)}$ are used as input in the combined least-square fit with neutron cross section standards. The evaluation by Axton (1986) was based on a least-square fit of both thermal-spectrum averaged cross sections (Maxwellian data) and microscopic cross sections at 2200 m/s. There is a second Axton evaluation based exclusively on measured microscopic cross sections at 2200 m/s (excluding Maxwellian data). Both evaluations disagree within quoted uncertainties for fission and capture cross sections and total multiplicities of uranium isotopes. There are two factors, which may lead to such difference: Westcott g-factors with estimated 0.2% uncertainties used in the Axton's fit, and deviation of the thermal spectra from Maxwellian shape. To exclude or mitigate the impact of these factors, a new combined GMA fit of standards was undertaken with Axton's TNC evaluation based on 2200 m/s data used as a prior. New microscopic data at the thermal point, available since 1986, were added to the combined fit. Additionally, an independent evaluation of TNC was undertaken using CONRAD code. Both GMA and CONRAD results are consistent within quoted uncertainties. New evaluation shows a small increase of fission and capture thermal cross sections, and a corresponding decrease in evaluated thermal nu-bar for uranium isotopes and ^{239}Pu .

1. Introduction

An IAEA project [1] to update the 2006 Neutron Standards [2] is close to conclusion. This project constitutes an important contribution to the CIELO collaboration [3]. Traditionally, the Thermal Neutron Constants (TNC) for neutron-induced reactions on fissile nuclei and $^{252}\text{Cf(sf)}$ nu-bar have been evaluated separately using a generalized least-square fit, and released within the updated Neutron standards. The TNC comprise 17 nuclear constants: elastic, fission and capture cross sections, and total fission neutron multiplicities of thermal-neutron induced reactions on ^{233}U , ^{235}U , ^{239}Pu , ^{241}Pu as well as $^{252}\text{Cf(sf)}$ total fission neutron multiplicity (nu-bar). Additionally, if we consider reactor spectra experiments (Maxwellian data), then we can derive the values of Westcott factors for absorption and fission (8 additional constants).

TNC are fundamental neutron-induced reaction data, which have been evaluated at the IAEA since the pioneering works by Westcott [4], Hanna [5] and Lemmel [6,7]. Lemmel already pointed out in 1975 an existing discrepancy between results of the fit using microscopic data only (2200 m/s), and the overall result that employs both microscopic and Maxwellian data, especially for the ^{235}U thermal constants [6]. Lemmel evaluations have been followed by efforts by Divadeenam and Stehn [8,9] and Axton [10].

In a comprehensive paper published in 1984 Divadeenam and Stehn used both Maxwellian and 2200 m/s data to derive a new set of TNC constants [8]. They assumed Maxwellian prompt fission neutron spectra (PFNS) in their analysis of reactor spectra (see Table 26, p.386 of Ref. [8]); a similar approach was used in previous analysis by Lemmel [6,7]. ^{235}U PFNS mean fission neutron energy $\langle E \rangle = 2.03(2)$ MeV was used in Ref. [8]. This value is discrepant with the latest recommendation of $\langle E \rangle = 2.00(1)$ MeV for thermal-neutron induced fission of ^{235}U [11–13] based on a non-model least-square fit of thermal-neutron induced fission data. A similar situation arises for ^{233}U and ^{239}Pu PFNS.

There is a clear need to derive TNC based on microscopic (2200 m/s) data alone, therefore avoiding possible biases in the analysis of integral data taken in critical assemblies and reactors. This task is undertaken in the present work.

2. Results of microscopic (MIC) TNC evaluation with CONRAD code

The code used by Axton in his evaluation [10] together with original experimental datasets was unavailable. We used code CONRAD [14] and all microscopic experimental data (including variances and uncertainties cross-correlations) tabulated by Axton in his report [10]. Two options were possible: least-squares fit using the AGS

^a e-mail: Roberto.CapoteNoy@iaea.org

Table 1. Comparison of the results reported by Axton [10] (ALL means both microscopic and Maxwellian data are used, MIC means microscopic data only) and calculated with the CONRAD code by using the AGS formalism and the marginalization procedure using only microscopic data (2200 m/s data) in the fit.

Thermal constants	Axton ALL	Axton MIC	CONRAD-AGS MIC	CONRAD-AGS MIC marginal. N=1000.	
²³³ U	σ_s	12.2(0.7)	12.3(0.7)	12.3(0.7)	12.3(0.7)
	σ_f	530.7(1.3)	533.3(2.4)	530.4(1.9)	530.2(1.9)
	σ_γ	45.5(0.7)	42.0(1.8)	44.9(0.8)	45.1(0.9)
	v_t	2.495(.004)	2.486(.005)	2.491(.005)	2.493(.005)
²³⁵ U	σ_s	16.0(1.1)	16.2(1.2)	15.8(1.2)	15.8(1.3)
	σ_f	582.8(1.1)	585.1(1.6)	584.1(1.5)	584.3(1.8)
	σ_γ	99.0(0.7)	96.1(1.7)	98.1(1.1)	97.9(1.2)
	v_t	2.433(.005)	2.426(.005)	2.429(.004)	2.426(.005)
²³⁹ Pu	σ_s	7.90(0.97)	7.90(0.98)	7.99(0.99)	8.01(0.99)
	σ_f	747.6(2.0)	748.5(2.6)	748.6(2.7)	744.3(3.2)
	σ_γ	271.3(2.1)	270.4(3.2)	270.4(2.8)	271.7(2.9)
	v_t	2.882(.005)	2.879(.006)	2.881(.006)	2.887(.006)
²⁴¹ Pu	σ_s	12.2(2.6)	12.0(2.6)	12.0(2.5)	12.2(2.6)
	σ_f	1012(7)	1021(11)	1018(12)	1012(16)
	σ_γ	361.3(4.9)	361.6(6.2)	364.1(6.3)	361.5(7.3)
	v_t	2.946(.006)	2.941(.007)	2.943(.006)	2.943(.007)
²⁵² Cf	v_t	3.768(.005)	3.764(.005)	3.766(.005)	3.761(.005)

formalism for presentation of the uncertainties [15], and the Monte Carlo version of the marginalization procedure [16].

The AGS formalism is used at the JRC-GEEL (Belgium) for storing and communicating large experimental covariance matrices [15]. The microscopic data selected by Axton are reported according to the AGS formalism, in which uncorrelated and correlated sources of uncertainties are clearly distinguished. In the report of Axton [10], 64 sources of correlated uncertainties were identified and well documented. All the reported experimental information and uncertainties were introduced in the iterative fitting procedure of the CONRAD code. Final results are reported in the ‘‘CONRAD AGS MIC’’ column of Table 1.

Results obtained with the AGS formalism were verified with a Monte Carlo procedure that relies on the so-called ‘‘marginalization’’ technique [16]. The marginalization was implemented in the CONRAD code to avoid problems when highly correlated data are introduced in the fitting procedure. The principle is similar to the AGS formalism. Correlated and uncorrelated uncertainties are treated separately in a two-step CONRAD calculation. The Monte Carlo approach consists in sampling the different sources of correlated uncertainties and to repeat the adjustment of the thermal constants with CONRAD. The rightmost column of Table 1 reports the average values obtained after N=1000 samples using Cholesky decomposition of the experimental covariance matrix reconstructed with information reported by Axton [10]. The Monte Carlo calculated mean values using marginalization are in good agreement with those obtained via the AGS formalism, but sometimes with slightly increased uncertainty.

The results of the fits, in comparison with the original evaluation by Axton done simultaneously for 0.0253 eV (microscopic) and thermal spectrum averaged

(Maxwellian) data and only for microscopic data are presented. An overall good agreement with Axton’s results is obtained. The magnitude of the uncertainties reported by Axton is confirmed by the CONRAD calculations. The main differences in mean values with Axton MIC results are observed for the fission and capture cross section of ²³³U and ²³⁵U as shown in Table 1 in bold shrift.

The origin of observed discrepancies is not fully understood. One of possible problems could be the differences in the 2200 m/s data set used by Axton and introduced in the CONRAD calculations (Axton used 101 measurements, while 102 measurements were selected for the CONRAD analysis). As we see from Table 1, exclusion of the Maxwellian data has a little influence on evaluated values when the CONRAD code is used, while Axton’s fit shows visible differences between these two cases. This discrepancy was also observed and discussed by Lemmel [5].

3. Additional data used in the combined fit of TNC with other standards data

GMA evaluation can use one of the derived TNC results shown in Table 1 as a prior in the combined Bayesian fit with all standard cross sections [1,2]. TNC data are coupled with other thermal standards (e.g., ¹⁰B(n,α) and ⁶Li(n,α) cross sections) through measured ratios, and with other standards at energies above 0.0253 eV through the measurements of cross section shape, which include the 0.0253 eV point and data at higher energies. Standard input data include 27 datasets presenting energy dependent absolute and shape ratios of cross sections including the 0.0253 eV point for ²³⁵U and 12 datasets for ²³⁹Pu (with 5 common data sets). New measurements of TNC, which can be used in the standards evaluation, have been available since Axton’s evaluation. The most important one is the microscopic measurement of the ratio of capture to fission cross section (alpha) for ²³⁵U at 0.0253 eV by Adamchuk et al. [17] with 2.1% uncertainty; additionally, there are three measurements of ratio ²³³U, ²³⁹Pu and ²⁴¹Pu to ²³⁵U at 0.0253 eV done at LANL and CERN n_TOF with estimated uncertainties between 2 and 3%.

One of the problems of using Axton’s fit with microscopic data as a prior in a least-square fit is the absence of covariance (correlation) matrix for this case in Axton report [10]. Instead, we used Axton microscopic data with the same correlation matrix obtained by Axton in the fit of microscopic and macroscopic data (neglecting cross-correlations to macroscopic quantities). To check the impact on GMA evaluated values, Axton correlation matrix was replaced by the correlation matrix obtained with CONRAD code using only microscopic data. It was found, that the difference in the evaluated constants was well within the limits of their uncertainties. The largest difference in the evaluated central values was about 20% from their relative uncertainty.

Another potential problem in Axton fit is the absence of capture cross section data or measured alpha value at 0.0253 eV for fissile targets. Adamchuk et al. [17] measured an alpha value of 0.1690 ± 0.0035 at 0.0253 eV using a neutron beam with a mechanical selector (chopper), but this set was published in 1988 well after the Axton report [10]. However, Axton used data from post-irradiation experiments (PIE) by Lounsbury [18] (one year

irradiation of samples) and later re-analyzed using Monte Carlo simulations by Beer et al [19]. It is remarkable that Beer et al. ^{235}U alpha value of 0.1697 ± 0.0029 is in good agreement with Adamchuk et al. data of Ref. [17]. Such agreement is surprising considering that g-factors for fission and capture cross sections evaluated by Westcott [20] are not currently considered to be very accurate despite the very low quoted uncertainties at the time (0.1% for ^{235}U fission and 0.3% for ^{239}Pu fission).

It is expected that modern Reich-Moore analysis of the resonance region in fissile nuclei can produce more accurate g-factors. In fact, calculated g-factors of 0.97934 and 0.9913 for ^{235}U thermal-neutron induced fission and capture, respectively (IAEA CIELO ^{235}U evaluation, December 2016), differ by 0.2% for fission and by 0.5% for capture from those derived by Axton [10]. For ^{239}Pu featuring a much stronger first resonance, the difference of Axton and modern g-factors can be even larger (0.8% for capture and 0.5% for fission). The main reason of such differences is the rather strong deviation from $1/v$ energy dependence near the thermal region for many actinides. This is complicated further by the presence of a first strong resonance at hundreds meV range, by the position and widths of bound state resonances, by resonance-resonance interference, and by a possible deviation of neutron spectra from the Maxwellian shape at the experimental setup. Because of this, we undertook combined GMA fits using a non-informative prior for ^{235}U capture cross section (assigning a large uncertainty). The same prior was applied in CONRAD's fits.

4. TNC evaluation results

The results of the TNC evaluation in the combined GMA fit with the use of different priors (pre-evaluated microscopic data obtained by Axton and with CONRAD code) are presented in Table 2.

Comparing columns one and two in Table 2 we see that the use of Maxwellian data leads to much lower uncertainty of capture cross sections, but also to an undesirable effect: a reduction of thermal fission and an increase of thermal total nubar.

In other words, the 2200 m/s measured fission cross sections are systematically higher than the corresponding values derived from a combined use of Maxwellian data.

Comparing the four or fifth column (GMA fit with CONRAD prior) in Tables 1 and 2, we can observe the impact of experimental cross section data above the thermal region on the least-square fit (both Axton and CONRAD fits used only thermal data, while the GMA fit uses the whole database used in Neutron Standard fit [1,2]). The general trend is a small increase of fission cross sections, especially for ^{235}U and ^{239}Pu targets. This trend is observed also when we compare the results of TNC evaluation of previous standards (2006), based on Axton's fit of microscopic and Maxwellian data, with present results, based on Axton's fit of microscopic data alone.

5. Conclusions

Despite small differences between GMA evaluated results for different priors, all evaluated data agree within quoted uncertainties. The results of the GMA fit using CONRAD

Table 2. Comparison of TNC obtained in the combined GMA fit with different pre-evaluated constants. GMA fit with Axton's microscopic evaluation is proposed to be used as recommended and standard TNC data (highlighted in blue).

Thermal constants	GMA fit with Axton ALL	GMA fit with Axton MIC	GMA fit with CONRAD-AGS	GMA fit with CONRAD-AGS & marginal.
^{233}U	σ_s	12.1(0.1)	12.2(0.7)	12.3(0.7)
	σ_f	531.2(1.3)	534.5(2.4)	530.4(2.2)
	σ_γ	45.6(0.7)	41.9(1.7)	44.9(0.8)
	ν_t	2.4968 (0.0035)	2.4853 (0.0054)	2.4908 (0.0049)
^{235}U	σ_s	14.09(0.22)	14.09(0.22)	14.07(0.22)
	σ_f	584.3(1.0)	587.2(1.4)	585.5(1.1)
	σ_γ	99.4(0.7)	99.3(2.0)	99.0(2.0)
	ν_t	2.4355 (0.0023)	2.4250 (0.0045)	2.4289 (0.0047)
^{239}Pu	σ_s	7.80(0.96)	7.82(1.00)	7.98(0.98)
	σ_f	750.0(1.8)	752.1(2.2)	750.3(2.5)
	σ_γ	271.5(2.1)	270.4(3.1)	271.0(2.7)
	ν_t	2.8836 (0.0047)	2.8775 (0.0060)	2.8809 (0.0057)
^{241}Pu	σ_s	12.1(2.6)	11.95(2.6)	12.0(2.5)
	σ_f	1014(7)	1024(11)	1018(11)
	σ_γ	361.8(5.0)	361.8(6.2)	364.2(6.0)
	ν_t	2.9479 (0.0054)	2.9400 (0.0064)	2.9430 (0.0058)
^{252}Cf	ν_t	3.7692 (0.0047)	3.7635 (0.0049)	3.7660 (0.0049)

prior are closer to the TNC 2006 evaluation [2] that employed both microscopic and Maxwellian data. All fits have higher uncertainties than in 2006 evaluation as we excluded the Maxwellian data from the current fit. We recommend using the TNC obtained in a GMA fit with Axton microscopic data used as a prior (shown in bold blue in Table 2).

Additional work including the CONRAD fit of microscopic and macroscopic data and a comprehensive analysis of g-factors obtained in modern resonance evaluations using latest TOF experiments is strongly encouraged.

All of the work described in this paper represents the outputs from evaluation initiatives agreed at the consultants' meetings organized under the auspices of the International Atomic Energy Agency. The authors would like to thank all colleagues at their own research institutes for encouraging their involvement, and staff of the IAEA Nuclear Data Section for preparing for and organizing the coordinated research project.

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