EPJ Web of Conferences **281**, 00027 (2023) https://doi.org/10.1051/epjconf/202328100027
CW2022

Experimental spectrum averaged cross sections (SACS) in ^{252}Cf(sf) neutron field and its impact on the evaluation of neutron standards

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Abstract. A new evaluation of spectrum averaged cross sections (SACS) of ^{235}U, ^{238}U, and ^{239}Pu measured in the ^{252}Cf(sf) reference neutron field is presented and found to be consistent with the original Mannhart SACS evaluation in IRDF-2002. The comprehensive vetted experimental database that includes SACS ratio and absolute SACS measurements of major actinides is being used to update the SACS database employed as input of the new GMApy code to derive the Neutron Standards. An update of the current neutron standards based on Time Projection Chamber (TPC) shape data, a new comprehensive uncertainty quantification, and revised SACS experimental database is proposed which result in a 0.7% increase of the evaluated ^{239}Pu(n,f)/^{235}U(n,f) cross-section ratio in the 1–5 MeV energy region. The increase is due to a 0.3% reduction of the standard ^{235}U(n,f) cross section and a 0.4% increase of the ^{239}Pu(n,f) reference cross section in the 1–5 MeV energy region. Those changes are well within estimated USU fission cross-section uncertainties of 1.2%, but are relevant for the evaluated mean values.

1 Introduction

Evaluation of Neutron Standards have been a critical component of ENDF/B library releases. The Neutron Standards have been coordinated by the IAEA since early 2000s and have been incorporated into the ENDF/B-VII.0 [1] and ENDF/B-VIII.0 releases [2]. The latest IAEA Neutron Data Standards were released in 2017 (will be called STD 2017 in this paper) [3, 4] and were adopted for the ENDF/B-VIII.0 library. Since very early on, the Neutron Standards have been derived using the GMA code originally developed by W. Poenitz with an extensive database of vetted experimental data [5, 6] and represent our best-known cross sections derived by a non-model evaluation [7]. The GMA standard input database also included the simplest integral data, namely spectrum averaged cross sections (SACS) of ^{239}Pu(n,f) and ^{235}U(n,f) reactions measured in the ^{252}Cf(sf) neutron field.

Note that the reference ^{252}Cf(sf) neutron spectrum was originally evaluated by Mannhart [8] based on time-of-flight measurements and has been validated as a reference neutron field in IRDFF-II library [9, 10]. We can use the reference ^{252}Cf(sf) neutron spectrum [9, 10] to convolute evaluated Neutron Standard cross sections to derive SACS and corresponding uncertainties. At the same time Mannhart evaluated the experimentally measured SACS in the ^{252}Cf(sf) spectrum for the IRDF-2002 library [11]. A comparison of evaluated fission SACS measured in the ^{252}Cf(sf) spectrum for dosimetry reactions ^{235}U(n,f), ^{238}U(n,f), and ^{239}Pu(n,f) versus fission SACS derived from Neutron Standards is given in Table 1.

The fission spectral indexes (SI) ^{239}Pu(n,f)/^{235}U(n,f) and ^{238}U(n,f)/^{235}U(n,f) in the ^{252}Cf(sf) neutron spectrum are listed in Table 2 with the corresponding C/E values showing an underestimation larger than 2%. Such underestimation is close to the lower bound of the estimated uncertainty of the C/E ratios assuming the calculated and measured values to be independent. This underestimation of the SI compared to measured values deserves an investigation.

Additionally, it was pointed out by Casperson [12] that a similar underestimation is observed in C/E of fission SI measured in the neutron field of well-understood fast critical assemblies Godiva and Flaptop-U [13, 14]. These assemblies show an excellent C/E for more than 20 different dosimetry reactions [9] demonstrating our very good understanding of the neutron fluxes in these critical assemblies in a broad energy range. The comparison of measured versus calculated SI in Godiva and Flaptop-U critical assemblies are shown in Tables 3 and 4, respectively. There is an agreement in the observed underestimation of fission SI in ^{252}Cf(sf) and critical-assemblies' neutron fields.

Finally, a 2% normalization difference between the measured ^{239}Pu(n,f)/^{235}U(n,f) absolute cross-section ratio using a Time Projection Chamber (TPC) detector to the Neutron Standard was recently discussed [15][1].

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[1] Note that the TPC ratio data were treated as relative ratios (shape data) in a recent ^{239}Pu re-evaluation.

Table 1: Fission SACS calculated C in a ^{252}Cf(sf) reference neutron spectrum [10] versus measured data E (represented by the Mannhart evaluation [11]), and the corresponding C/E with uncertainties.

Reaction	IAEA STD 2017 $\equiv C$[1] [barn]	Mannhart eval. [11] $\equiv E$[2] [barn]	C/E[3]
^{235}U(n,f)	1.2267(1.2%)	1.210 (1.2%)	1.014(1.70%)
^{238}U(n,f)	0.3215(1.3%)	0.3257(1.6%)	0.987(2.06%)
^{239}Pu(n,f)	1.7978(1.3%)	1.812 (1.4%)	0.992(1.91%)

[1] Evaluated ^{239}Pu(n,f), ^{238}U(n,f), and ^{235}U(n,f) cross sections assumed fully correlated, which is an approximation.
[2] Manhhart evaluated correlations [11]:
corr(^{239}Pu,^{235}U)=0.28, corr(^{238}U,^{235}U)=0.15.
[3] Calculated C/E uncertainties derived assuming C and E are independent, which is an approximation.

Table 2: Fission SI in ^{252}Cf(sf) neutron spectrum.

Spectral Index (SI)	IAEA STD 2017 $\equiv C$[1] [barn]	Mannhart eval. [11] $\equiv E$[2] [barn]	C/E[3]
^{239}Pu(n,f)/^{235}U(n,f)	1.465(1.2%)	1.498(1.7%)	0.978(2.0%)
^{238}U(n,f)/^{235}U(n,f)	0.262(1.2%)	0.269(1.8%)	0.973(2.2%)

[1] SI C (ratio) uncertainties of IAEA STD 2017 were derived considering full correlation of numerator and denominator as stated in Table 1, which is an approximation.
[2] SI E evaluated by Mannhart include evaluated correlations as listed in Table 1.
[3] SI C/E uncertainties assumed uncorrelated C and E quantities, which is an approximation.

Table 3: Fission SI in Godiva (HEU-MET-FAST 001 [14]) critical assembly.

Reaction	IAEA STD 2017 $\equiv C$[1] [barn]	Measured SI [13] $\equiv E$ [barn]	C/E[1]
^{239}Pu(n,f)/^{235}U(n,f)	1.3841(1.7%)	1.4152(1.0%)	0.978(2.0%)
^{238}U(n,f)/^{235}U(n,f)	0.1584(1.7%)	0.1643(1.0%)	0.972(2.0%)

[1] C/E uncertainties derived assuming numerator C and denominator E are independent, which is an approximation.

Table 4: Fission SI in Flaptop-U (HEU-MET-FAST 028 [14]) critical assembly.

Reaction	IAEA STD 2017 $\equiv C$[1] [barn]	Measured SI [13] $\equiv E$ [barn]	C/E
^{239}Pu(n,f)/^{235}U(n,f)	1.3615(1.7%)	1.3847(0.9%)	0.985(1.9%)
^{238}U(n,f)/^{235}U(n,f)	0.1450(1.7%)	0.1492(1.1%)	0.972(2.0%)

[1] C/E uncertainties derived assuming numerator C and denominator E are independent, which is an approximation.

Observed differences may be due to deficiencies in the Neutron Standards input data, in the Neutron Standard evaluation methodology or in both. Biases in measured SI in critical assemblies and in the measured absolute TPC cross-section ratio are also possible. The goal of this work is to study the sources of these differences, and whether we can improve Neutron Standards fission cross sections and associated reference cross sections like the ^{239}Pu(n,f).

2 New evaluation of measured SACS in the ^{252}Cf(sf) neutron spectrum

The updated experimental database of ^{235}U(n,f), ^{238}U(n,f), and ^{239}Pu(n,f) cross sections and corresponding ratios measured in the ^{252}Cf(sf) neutron spectrum are compiled in Table 5. The corresponding experimental covariance matrix was constructed from the analysis and uncertainty quantification of the SACS experimental data similarly as done by Mannhart [11]. In fact, Mannhart provided in Ref. [11] most of the listed coefficients, we only needed to add Schröder data. Note that we also had the original Mannhart input used in his SACS evaluation provided by Mannhart himself. That makes his evaluation fully traceable. Corresponding experimental correlation coefficients are zero by default[2] except those listed in Tables 6, 7, 8, 9, and 10. The most important difference from Mannhart evaluation [11] is the addition of newly measured data by Schröder at NIST in 1985 [16].

With the updated experimental SACS database listed above we have used Mannhart BAYES code (employed in his original evaluation) to produce a new evaluation with results listed in Table 11 and corresponding evaluated correlations listed in Table 12. Both evaluations show good agreement well within their one-sigma quoted uncertainties. An small absolute increase of evaluated values is observed. Ratios of evaluated cross sections agree extremely well. Evaluated ratio uncertainty are reduced due to the very strong positive correlations derived for the ratio numerator and denominator. The obtained agreement shows the robustness of Mannhart SACS evaluation. Therefore, the newly updated SACS experimental database was validated, and can be used in the new GMA fit aiming at improving the normalization of GMA fission cross section fits, as will be shown in next section.

3 Update of the Neutron Standard evaluation with new experimental SACS data

The IAEA standard cross sections 2017 [3] are taken as the reference for the updated evaluation of the fission cross sections which will be discussed in this Section. All presented calculations were done with the new GMApy code developed by G. Schnabel at the IAEA and fully validated vs the original GMA FORTRAN code.

The GMApy evaluation was carried out in the full energy range of data; discussions in the following are restricted to the impact of the SACS experimental inputs on the normalization of the evaluated fission cross sections. Note that the new GMApy code is necessary to use measured SACS ratios that the original GMA code can not use directly. We focus on the fast energy region from 1 up to 5 MeV of incident neutron energy for comparison, being the neutron energy region that includes the maximum of the ^{252}Cf(sf) reference neutron spectrum [8, 10] and covers a significant percentage of the outgoing neutrons.

In Figs.1–3 the ratios to the IAEA reference cross sections are shown in the energy region from 1 MeV up to 5 MeV for the following cases:

1. **std2017-input**: Same input as in STD 2017, but recalculated with the GMApy code correcting identified glitches.
2. **additional-input**: New GMApy fit based on updated uncertainty quantification of ^{239}Pu data [17–19] and new TPC experimental shape data [15, 20, 21]. The fitted ^{239}Pu cross sections are used in the INDEN ^{239}Pu evaluated file pu239p55 available at the INDEN webpage (https://nds.iaea.org/INDEN/).

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[2] Reflecting assumed lack of correlations by Mannhart [11].

Table 5: Experimental ^{235}U(n,f), ^{238}U(n,f), and ^{239}Pu(n,f) SACS in ^{252}Cf(sf) neutron spectrum: updated database.

Reaction index	Reaction	SACS [barn]	SACS uncert. [%]	Reaction label	Reference
1	^{235}U(n,f)	1216	1.62	U5F	Grundl 1983 [22, 23, 25]
2	^{235}U(n,f)/^{238}U(n,f)	3.73	1.20	U5F/U8F	Grundl-Gilliam 1983 [22, 24, 28], inverse = 0.2681
3	^{235}U(n,f)/^{239}Pu(n,f)	0.666	0.90	U5F/PU9F	Grundl-Gilliam 1983 [22, 24], inverse = 1.502
4	^{239}Pu(n,f)/^{235}U(n,f)	1.500	1.60	PU9F/U5F	Heaton 1976 [25]
5	^{238}U(n,f)/^{235}U(n,f)	0.2644	1.32	U8F/U5F	Heaton 1976 [25]
6	^{238}U(n,f)/^{235}U(n,f)	0.269	1.20	U8F/U5F	Schröder 1985 [16]
7	^{239}Pu(n,f)/^{235}U(n,f)	1.500	0.80	PU9F/U5F	Schröder 1985 [16]
8	^{235}U(n,f)	1234	1.45	U5F	Schröder 1985 [16]
9	^{235}U(n,f)	1215	1.79	U5F	Davis/Knoll 1978 [26]
10	^{239}Pu(n,f)	1790	2.26	PU9F	Davis/Knoll 1978 [26]
11	^{238}U(n,f)/^{235}U(n,f)	0.2741	1.66	U8F/U5F	Adamov 1977 [27]
12	^{239}Pu(n,f)/^{235}U(n,f)	1.475	1.50	PU9F/U5F	Adamov 1977 [27]

Table 6: Experimental correlations estimated by Mannhart [11] for Grundl-Gilliam SACS data [22–25].

Reaction or Spectral Index	U5F [%]	SI U5F/U8F [%]	SI U5F/PU9F [%]
^{235}U(n,f)	100		
^{235}U(n,f)/^{238}U(n,f)	23	100	
^{235}U(n,f)/^{239}Pu(n,f)	-9	36	100

Table 7: Experimental correlations for Heaton et al. [25].

Spectral Index	SI PU9F/U5F [%]	SI U8F/U5F [%]
^{239}Pu(n,f)/^{235}U(n,f)	100	
^{238}U(n,f)/^{235}U(n,f)	60	100

Table 8: Experimental correlations for Schröder et al. [16].

Spectral Index	SI PU9F/U5F [%]	SI U8F/U5F [%]
^{239}Pu(n,f)/^{235}U(n,f)	100	
^{238}U(n,f)/^{235}U(n,f)	39	100

Table 9: Experimental correlations for Davis et al. [26].

Spectral Index	SI PU9F/U5F [%]	SI U8F/U5F [%]
^{239}Pu(n,f)/^{235}U(n,f)	100	
^{238}U(n,f)/^{235}U(n,f)	59	100

Table 10: Experimental correlations for Adamov et al.[27].

Spectral Index	SI PU9F/U5F [%]	SI U8F/U5F [%]
^{239}Pu(n,f)/^{235}U(n,f)	100	
^{238}U(n,f)/^{235}U(n,f)	39	100

Table 11: Evaluated experimental SACS in ^{252}Cf(sf) neutron spectrum vs Mannhart original evaluation [11]. These values together with correlations listed in Table 12 represent our best experimental knowledge and are independent of the evaluated ^{252}Cf(sf) neutron spectrum.

Reaction	This work [barn]	Mannhart eval. [11] [barn]	new/old eval.
^{235}U(n,f)	1.221 (0.91%)	1.210 (1.2%)	1.009
^{238}U(n,f)	0.327 (1.06%)	0.3257(1.6%)	1.004
^{239}Pu(n,f)	1.826 (1.03%)	1.812 (1.4%)	1.008

Table 12: Evaluated experimental SACS correlations in ^{252}Cf(sf) neutron spectrum of major-actinide fission cross sections corresponding to mean values and uncertainties listed in Table 11.

Reaction	U5F [%]	U8F [%]	PU9F [%]
^{235}U(n,f)	100		
^{238}U(n,f)	80	100	
^{239}Pu(n,f)	87	82	100

3. **additional-input + updated SACS**: In addition to new data the vetted SACS experimental database (see Sec.3) was used as input in the GMApy fit.

Case 1 in Figs.1–3 uses exactly the same input deck as the STD 2017 fit (labelled **std2017-input** and represented by the red line). The goal is to assess the impact of the new GMApy code and of few coding glitches which were found and corrected. The impact on ^{235}U(n,f) cross sections is up to +0.1%, on ^{238}U(n,f) cross sections – +0.2% and up to +0.15% on ^{239}Pu evaluated fission cross sections. The overall impact of the new code and corrected bugs was found to be very small.

Case 2 in Figs.1–3 represents the advanced uncertainty quantification of ^{239}Pu(n,f) GMA input database [17–19] and the addition of new TPC experimental data with very low uncertainties [15, 20, 21] used as shape data (labelled

additional-input and represented by the blue line). The impact of these changes on ^{235}U(n,f) cross sections is a reduction of up to 0.2% in the evaluated fission cross sections, on ^{238}U(n,f) cross sections – a reduction of 0.25% is observed, and an increase of up to 0.35% on ^{239}Pu evaluated fission cross sections.

Case 3 in Figs.1–3 represents the addition of the updated SACS experimental database listed in Table 5 and corresponding correlations listed in Tables 6, 7, 8, 9, and 10. It is labelled as **additional-input+SACS** and represented by the orange line. The new SACS data further decrease the ^{235}U(n,f) evaluated cross section by an additional 0.15%. That results in a total change of ^{235}U evaluated cross section by -0.35% relative to the IAEA STD 2017. No change in evaluated cross sections due to the use of updated SACS is practically observed for ^{238}U(n,f) and ^{239}Pu(n,f) reactions.

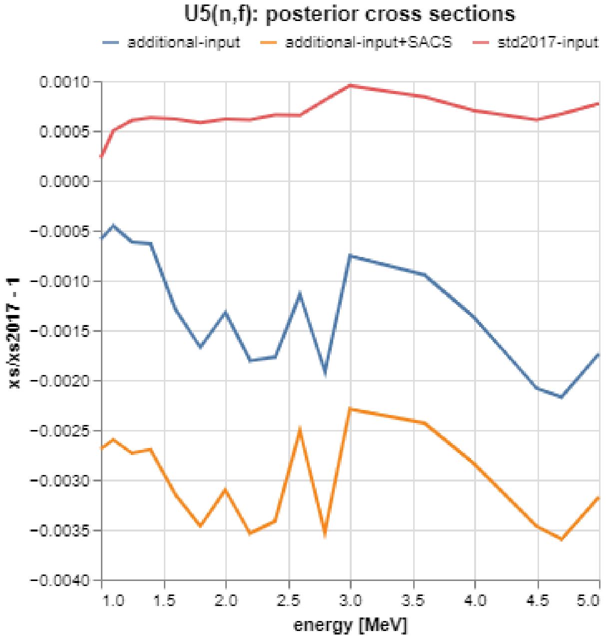

Figure 1: Evaluated ^{235}U(n,f) cross sections relative to the IAEA STD 2017 [3] from 1 MeV up to 5 MeV.

Summarizing, a reduction of 0.35% is observed of the evaluated fission cross sections of ^{235}U, whilst a 0.35% increase is obtained for the evaluated ^{239}Pu fission cross section. These changes led to a ^{239}Pu/^{235}U evaluated fission cross-section ratio increase of almost 0.7% as shown in Fig. 4. This ratio increase improves the agreement with published TPC absolute ^{239}Pu/^{235}U ratio [15]. Without the update in the experimental SACS database the absolute ^{239}Pu/^{235}U ratio increase is reduced to 0.45%. The corresponding change in the ^{238}U/^{235}U evaluated fission cross-section ratio is smaller going from 0.1% up to 0.3% in the energy region corresponding to the first chance fission plateau as shown in Fig. 5.

Newly evaluated GMApy cross sections that include the TPC ratio measurements as shape data and a revised experimental SACS database can be convoluted with the reference ^{252}Cf(sf) neutron spectrum [8, 10] to calculate the SACS which can be compared with the SACS evaluated directly from measured data listed in Table 11. This comparison is shown in Table 13. It can be seen that the agreement of SACS derived from standard and reference cross sections is already within the uncertainty of the updated SACS evaluation based on direct SACS measurements; small underestimation of the evaluated SACS average values remain to be investigated in future works. Additional review of measured absolute cross sections and

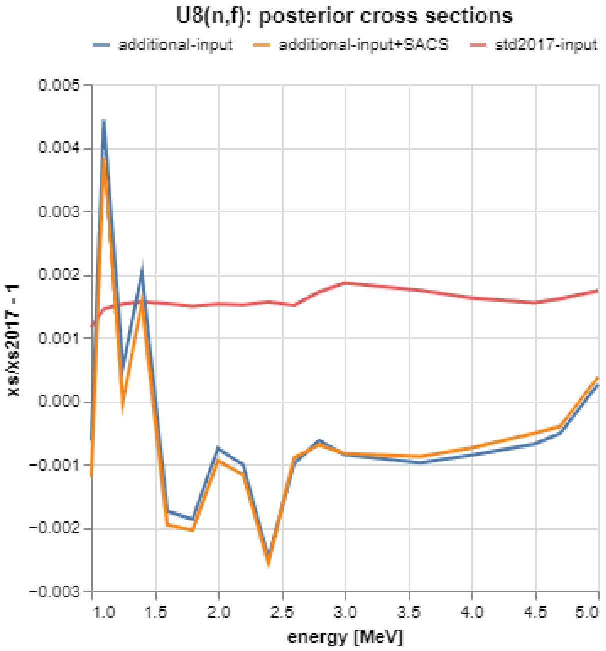

Figure 2: Evaluated ^{238}U(n,f) cross sections relative to the IAEA STD 2017 [3] from 1 MeV up to 5 MeV.

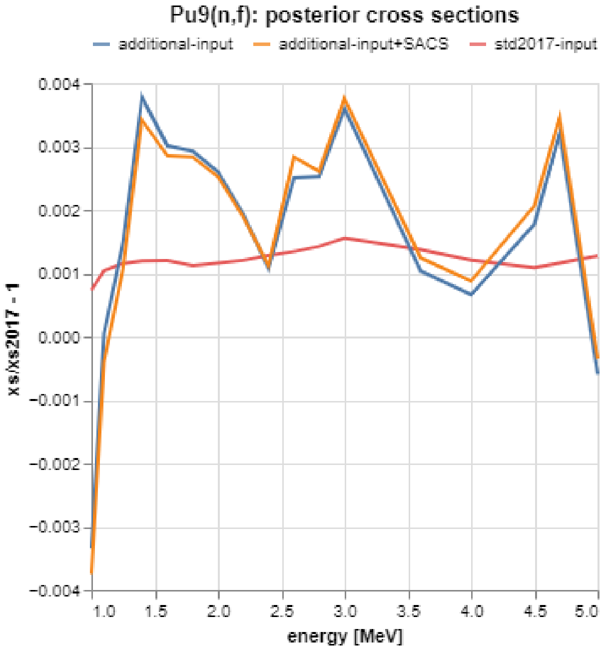

Figure 3: Evaluated ^{239}Pu(n,f) cross sections relative to the IAEA STD 2017[3] from 1 MeV up to 5 MeV.

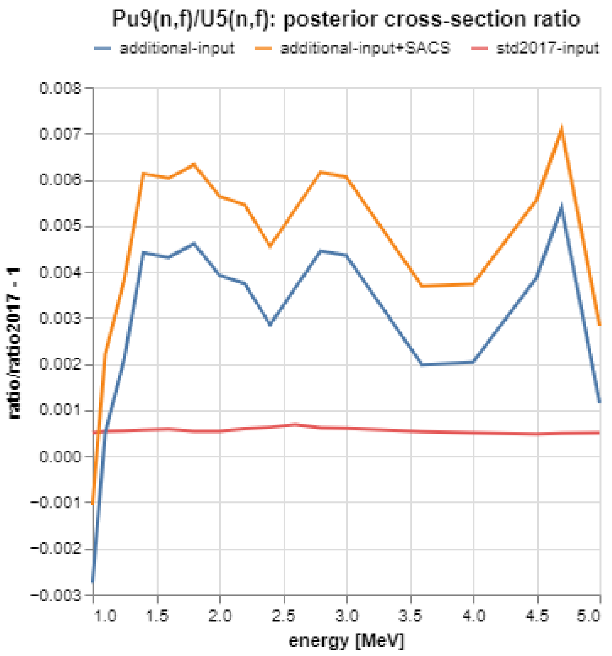

Figure 4: Evaluated ^{239}Pu(n,f)/^{235}U(n,f) cross-section ratio relative to the IAEA STD 2017 cross-section ratio [3].

absolute cross-section ratios involving ^{239}Pu targets is ongoing to further refine the experimental GMApy input database.

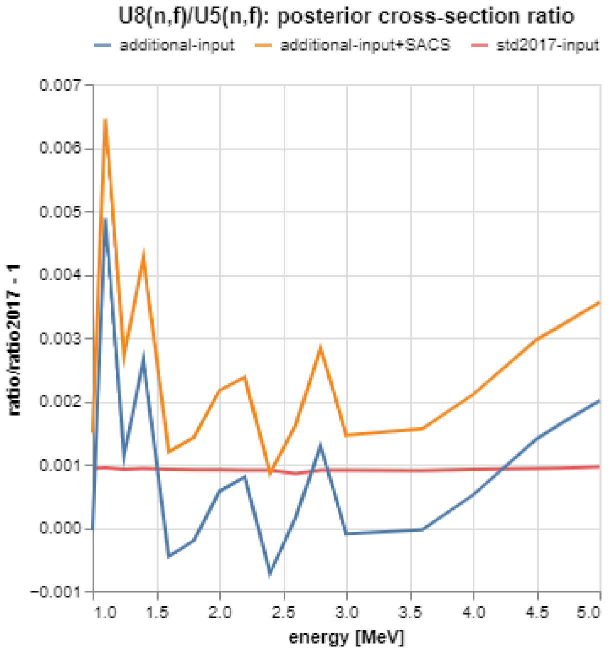

Figure 5: Evaluated ^{238}U(n,f)/^{235}U(n,f) cross-section ratio relative to the IAEA STD 2017 cross-section ratio [3].

Finally, it should be noted that the evaluated total uncertainties of the major-actinide fission cross sections remains practically unchanged from the uncertainties evaluated in the IAEA STD 2017 without considering the Unrecognized Sources of Uncertainties (USU) [3, 29]. Newly evaluated uncertainties are very weakly affected by the use of the updated experimental database and updated uncertainty quantification including the new experimental SACS input database. For the ^{235}U(n,f) cross section the total uncertainty in the first fission plateau (1–5 MeV) is about 0.5%; for the ^{239}Pu(n,f) – about 0.65–0.7%. Additional studies on the impact of energy-dependent USU on evaluated cross sections are warranted. However, energy-dependent USU are not expected to affect evaluated SACS in the ^{252}Cf(sf) spectrum.

Table 13: Calculated C fission SACS (based on GMApy fit) in a ^{252}Cf(sf) reference neutron spectrum [8, 10] vs evaluated SACS data based on experimental data E (see Table 11). The C/E values are given in the last column.

Reaction	This work GMApy $\equiv C$ [barn]	This work, SACS exp. $\equiv E$ [barn]	C/E[1]
^{235}U(n,f)	1.221 (0.3%)	1.221 (0.91%)	1.000 (0.96%)
^{238}U(n,f)	0.323 (0.4%)	0.327 (1.06%)	0.988 (1.13%)
^{239}Pu(n,f)	1.803 (0.4%)	1.826 (1.03%)	0.987 (1.10%)

[1] C/E uncertainties derived assuming C and E are independent quantities.

4 Conclusions

A vetted experimental database of ^{235}U(n,f), ^{238}U(n,f), and ^{239}Pu(n,f) spectrum averaged cross sections (SACS) measured in the ^{252}Cf(sf) reference neutron field [8, 10] is developed (see Table 5 and correlations in Tables 6–10). The experimental SACS database includes SACS ratio and absolute SACS measurements of neutron-induced fission reactions on the three major actinides using the new capabilities of the GMApy code. The new database was used to derive a non-model SACS evaluation in the ^{252}Cf(sf) neutron field that agrees within 1% with the original Mannhart SACS evaluation (see evaluated results in Tables 11 and 12). This agreement is well within the quoted experimental uncertainties. The vetted experimental SACS database is also used to update the input of the GMApy code employed to derive the Neutron Data Standards.

An update of the current neutron standards based on new TPC shape data, advanced uncertainty quantification of ^{239}Pu(n,f) reactions, and vetted SACS experimental database is carried out. The update results in a reduction of 0.3% and 0.25% of the evaluated ^{235}U(n,f) and ^{238}U(n,f) cross sections, respectively. A 0.45% increase is obtained for the evaluated ^{239}Pu fission cross section. Therefore, a significant 0.7% increase is derived for the ^{239}Pu(n,f)/^{235}U(n,f) ratio normalization. These changes are within Standard USU fission cross-section uncertainty of 1.2% [3, 29], but the obtained changes improve the C/E of fission spectral indexes in Godiva (HEU-MET-FAST 001) and Flaptop (HEU-MET-FAST 028) critical assemblies, and also improve the agreement of standard cross sections with published TPC ^{239}Pu(n,f)/^{235}U(n,f) absolute ratio data [15].

New SACS evaluations of fission cross sections in the ^{252}Cf(sf) neutron field obtained in this work are listed in Table 13. Our SACS value for the ^{235}U(n,f) of 1221 mb, newly derived from the GMApy fit, is exactly equal to the one derived by Otsuka and Iwamoto [30, 31]. For the

^{238}U(n,f) SACS value, a +2.2% difference to Otsuka and Iwamoto evaluation remains; our evaluated SACS value of 323 mb is consistent with the experimental SACS evaluated value of 327 mb. Finally, for ^{239}Pu(n,f), our new SACS value of 1803 mb is 0.7% below the evaluated value by Otsuka and Iwamoto of 1814 mb, which represents about 1.5 times our evaluated uncertainty without USU. Our newly evaluated ^{239}Pu(n,f) SACS value is 1.3% lower than the experimental SACS evaluation value of 1826 mb, which represents about 3 times our evaluated uncertainty without USU. Further investigation of the ^{239}Pu(n,f) absolute and absolute cross-section ratio experimental database used in the GMApy fit and a better uncertainty quantification are warranted to address these discrepancies.

Acknowledgements

The IAEA is grateful to all participating laboratories for their assistance in the collaborative INDEN network and for supporting the IAEA meetings and activities. Work described in this paper would not have been possible without contributions from the IAEA Member States. Authors would like to thanks D.L. Smith for his guidance and always sensible advices.

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