EPJ Web of Conferences **106**, 04011 (2016)
DOI: 10.1051/epjconf/201610604011

Validation of IRDFF in ^{252}Cf Standard and IRDF-2002 Reference Neutron Fields

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Abstract. The results of validation of the latest release of International Reactor Dosimetry and Fusion File, IRDFF-1.03, in the standard ^{252}Cf(s.f.) and reference ^{235}U(n$_{th}$,f) neutron benchmark fields are presented. The spectrum-averaged cross sections were shown to confirm IRDFF-1.03 in the ^{252}Cf standard spontaneous fission spectrum; that was not the case for the current recommended spectra for ^{235}U(n$_{th}$,f). IRDFF was also validated in the spectra of the research reactor facilities ISNF, Sigma-Sigma and YAYOI, which are available in the IRDF-2002 collection. The ISNF facility was re-simulated to remove unphysical oscillations in the spectrum. IRDFF-1.03 was shown to reproduce reasonably well the spectrum-averaged data measured in these fields except for the case of YAYOI.

1. Introduction

The recently released International Reactor Dosimetry and Fusion File (IRDFF) [1, 2] supersedes the International Reactor Dosimetry File IRDF-2002 [3]. With its energy extension up to 60 MeV, and in some cases even up to higher energies, and the inclusion of new reactions, this new library serves fission, fusion and accelerator driven applications. To ensure the quality of the newly released library the Nuclear Data Section (NDS) of IAEA has initiated a new Coordinated Research Project (CRP) with the objective to test, validate and optionally extend the scope of IRDFF (see summary report of the 1st CRP Meeting and the CRP web-page [4]). In March 2014 a new Version 1.03 of IRDFF was released, which included one new reaction ^{238}U(n,2n) and updates of several existing ones [5, 6].

This paper presents the validation results for the IRDFF-1.03 in the standard ^{252}Cf(s.f.) [7] and reference ^{235}U(n$_{th}$,f) [8] neutron benchmark fields. It also investigates the impact on validation of extending the integration of the spectrum-averaged cross sections above 20 MeV, since the preliminary validation of the initial release IRDFF-1.00 was done for an energy cut-off at 20 MeV [2, 9]. This paper

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also revisits older reference data obtained at the reactor facilities ISNF, Sigma-Sigma, CFRMF and YAYOI [10, 11]. Their spectra were included in the IRDF-2002 database [3], however measurements in these neutron fields were not previously incorporated into the evidence package supporting the validation of IRDFF.

2. Validation in the ^{252}Cf(s.f.) and ^{235}U(n$_{th}$,f) Fields

Validation of the IRDFF cross section in the standard and reference spectra was performed in terms of the ratio of Calculated-to-Experimental (C/E) Spectrum-Averaged cross sections (SPA). To support this purpose available experimental SPA data were systematically collected on the CRP web page [4] (see hyper-links there, e.g. "^{252}Cf(s.f.): Measured"). These data comprise mainly data points recommended by experienced evaluators (W. Mannhart and K. Zolotorev) after their renormalisation to the current standards, uncertainty analysis and eventually weighting of known experimental results. Other data included in this collection are from individual measurements. NDS systematically checks the EXFOR content for SPA and various neutron source spectra and compiles relevant data missing from this repository [4, 12].

The standard ^{252}Cf(s.f.) [7] and two reference Prompt Fission Neutron Spectra (PFNS) for ^{235}U(n$_{th}$,f), Los Alamos Madland-Nix model (implemented in ENDF/B-VII.1) [8] and Scale method [13], are shown in Fig. 1 as ratio to the Maxwellian distribution with $kT = 1.42$ and 1.32 MeV, respectively. It is worthwhile to note the general tendency of essential increasing of spectra uncertainties above 10 MeV and extremely large uncertainties in the ENDF/B-VII.1 evaluation in comparison with the Scale method for the ^{235}U(n$_{th}$,f) PFNS.

Computation of SPA was performed by the RR_UNC code [6] with the cut-off energy at 20 MeV. They were also calculated by the MCNP5 code employing dosimetry data from the IRDFF-1.03 library processed in ACE format by NJOY 2012 with latest update 32 [14]. This update solved the following processing problems: insufficiently large scratch arrays for Cd and Gd evaluations, cross sections leading to the isomeric states of ^{93m}Nb and ^{115m}In, and ^{10}B(n,α). In the Monte Carlo simulations the ^{252}Cf(s.f.) and ^{235}U(n$_{th}$,f) neutron spectra were represented in the standard SAND-II 640 groups [15] up to 20 MeV with the extrapolation up to 60 MeV with the addition of 0.2 MeV equal energy bins. The spectra extensions were accomplished by the product of a Maxwellian distribution with corresponding temperatures and linear functions in energy to get the same energy slope as below 20 MeV, Fig. 1.

The C/E ratios of SPA in the standard ^{252}Cf(s.f.) and reference ^{235}U(n$_{th}$,f) ENDF/B-VII.1 spectra as a function of representative neutron energy $E_{50\%}$ (point where energy integration of the product of spectra and cross sections reaches 50%) are shown in Fig. 2. It is seen that computation of SPA by RR_UNC and MCNP produces identical results for practically all reactions. The observed 1–2% over-prediction of MCNP with respect to RR_UNC for reactions with $E_{50\%}$ >12 MeV is explained by the extension of the neutron spectra above 20 MeV only in the MCNP calculations. The impact of the spectrum extension is seen to be stronger for the higher threshold reactions (whose SPA are not measured yet), e.g. it reaches a factor of 29X for the ^{59}Co(n,3n) reaction. This effect is less pronounced for the case of ^{235}U(n$_{th}$,f) due to its softer spectrum (mean energy 2.03 MeV) in comparison with ^{252}Cf(s.f.) (mean energy 2.12 MeV).

For the ^{252}Cf(s.f.) source, the C/E ratio fluctuates around unity as a rule within the associated total uncertainty and thus confirms the quality of the IRDFF-1.03 cross sections. However there are several obvious outliers ^{59}Co(n,γ), ^{92}Mo(n,p), ^{60}Ni(n,p) and ^{46}Ti(n,2n) (their C/E are located out of scale on Fig. 2). The likely reason are measurement errors, thus we recommend new measurements for these reactions. The spectrum component of the uncertainty becomes the dominant contributor in the total C/E uncertainties above 12 MeV, i.e. this validation could serve for the improvement of the ^{252}Cf(n,f) spectrum uncertainties at such high energies.

For the ^{235}U(n$_{th}$,f) field, the C/E ratio is close to unity for most of the reactions with $E_{50\%}$ <10 MeV as seen in Fig. 3. However there are several exceptions. Thus for the ^{10}B(n,α) reaction the IRDFF gives

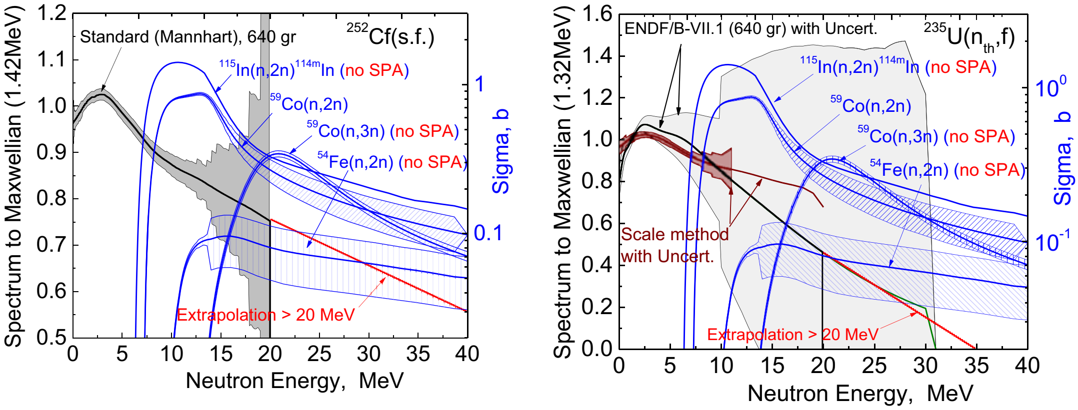

Figure 1. Neutron spectra ratios to Maxwellian distributions with indicated Temperatures (both spectra were normalised to unity) and their uncertainties: (left) ^{252}Cf(s.f.) from Standards [7]; (right) ^{235}U(n_{th},f) from ENDF/B-VII.1 [8] and Scale method [13]. Spectra extrapolations beyond 20 MeV are shown by red lines. Several high threshold IRDFF-1.03 cross sections with uncertainties are displayed in blue. Reactions, which SPA are not measured yet, have comment "(no SPA)".

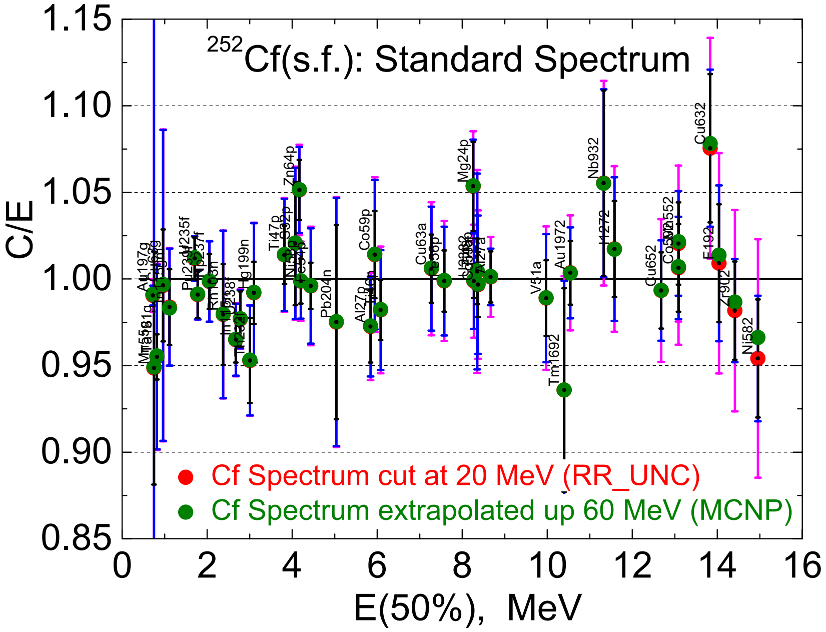

Figure 2. C/E with IRDFF-1.03 cross sections averaged in the ^{252}Cf(s.f.) field. Uncertainties bars: experimental SPA (black), IRDFF-1.03 cross sections (blue) and spectra (pink). On this and other Figs. the reactions are identified by the SAND-II abbreviations, e.g. Co592 stands for ^{59}Co(n,2n)^{58}Co.

C/E = 0.81 ±0.04. Since known experimental SPA were measured by helium-gas counting technique [8], the other alpha producing reaction ^{10}B(n,t2α) must be added. Calculating the corresponding SPA from ENDF/B-VII.1 results in C/E = 1.00. Similarly for ^{6}Li(n,t)^{4}He: addition of the (n,ndα) and (n,2npα) channels increases the C/E from 0.71 ±0.03 up to 1.03. Other reactions ^{55}Mn(n,γ), ^{238}U(n,γ), ^{139}La(n,γ), ^{31}P(n,p) and ^{238}U(n,2n) are outliers and need further investigations.

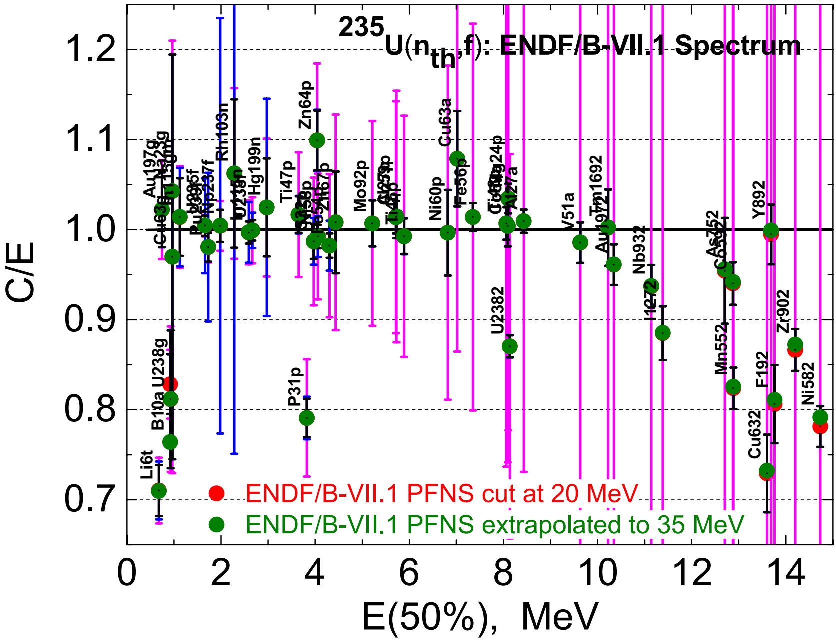

Figure 3. C/E with IRDFF-1.03 cross sections averaged in the ^{235}U(n$_{\mathrm{th}}$,f) field. Uncertainties bars: experimental SPA (black), IRDFF-1.03 cross sections (blue) and spectra (pink).

Above 10 MeV, the C/E shows a gradually increasing underestimation. The uncertainty associated with ENDF/B-VII.1 PFNS is rather large and begins to dominate in the C/E uncertainty above 4 MeV and even exceeds 50% above 8 MeV.

In the frame of an international effort to provide improved Neutron Standards [16] an attempt is ongoing to produce better evaluation of the ^{235}U(n$_{\mathrm{th}}$,f) PFNS and its uncertainty. One of the alternatives to the Los Alamos Madland-Nix model [8] is an evaluation based on known spectral measurements of PFNS, performed by N. Kornilov, the so-called "Scale method" [13]. The C/E ratios for SPA calculated with his and ENDF/B-VII.1 spectra (however without uncertainties) are shown in Fig. 4.

It is seen that the energy-integrated results reflect the shape difference between the Scale method and ENDF/B-VII.1 fission energy-differential spectra shown in Fig. 1: SPA with the Scale method PFNS gives systematically higher C/E below 2 MeV, lower between 2 and 8 MeV, and essentially higher above 10 MeV, where C/E shows a different overall trend, than with ENDF/B-VII.1. However, the spread of experimental SPA for the high-threshold (n,2n) reactions on ^{127}I, ^{55}Mn, ^{59}Co, ^{63}Cu, ^{19}F, ^{89}Y, ^{90}Zr and ^{58}Ni (all have $E_{50\%}$ between 11 and 15 MeV) is also large.

It is also worthwhile to note that even recommended experimental SPA for ^{127}I(n,2n), ^{55}Mn(n,2n) and ^{58}Ni(n,2n) reactions are different. The curved arrows in Fig. 4 show that the replacement of SPA recommended by W. Mannhart with the ones of K. Zolotarev increase the C/E by 8–15% for both PFNS. The values recommended by these evaluators could be found in their (not always published) documents, for convenience the data and proper references were collected on the CRP web-page [4].

Because of these reasons, it is difficult to make a preference for the ^{235}U(n$_{\mathrm{th}}$,f) PFNS and, in particular, to establish its shape above 10 MeV – it essentially depends on reliable and new measurements of high threshold reactions. The estimated SPA for such reactions are in the range 0.1–100 μb [4] and become unacceptable for the traditional activation technique. An accelerator mass

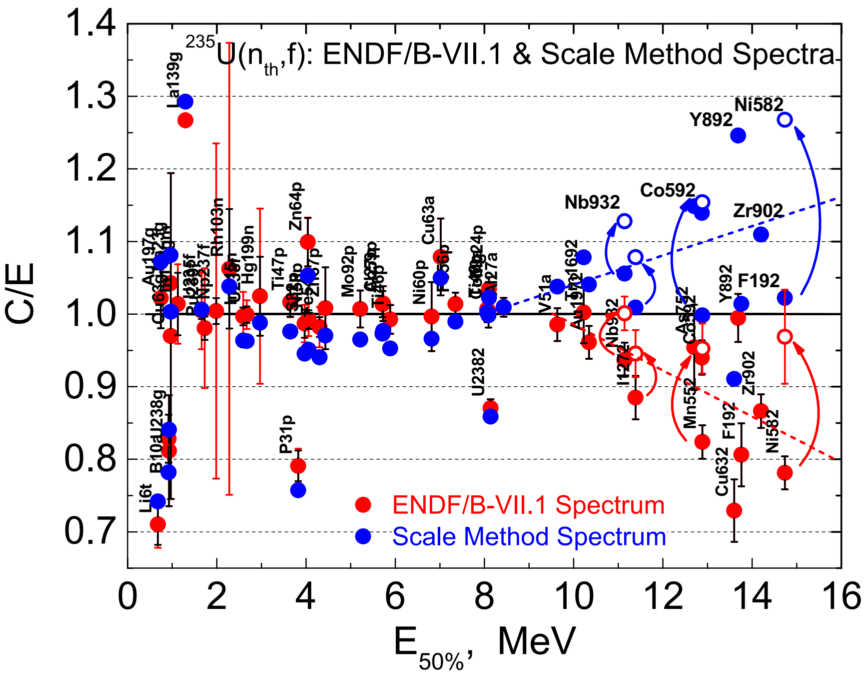

Figure 4. C/E with IRDFF-1.03 cross sections averaged in the ^{235}U(n$_{th}$,f) field from ENDF/B-VII.1 [8] and Scale method [13]. Uncertainties bars: experimental SPA (black), plus IRDFF-1.03 cross sections (red), PFNS – are excluded. Three curved arrows show the change of C/E for ^{127}I(n,2n), ^{55}Mn(n,2n) and ^{58}Ni(n,2n) when SPA recommended by W. Mannhart are replaced with K. Zolotarev values.

spectrometry [17] could be an alternative method due to its high sensitivity and the possibility to accumulate stable or long-lived isotopes.

3. Validation in the Reference Reactor Facilities

Another task of the CRP is to revisit the older reference data obtained at reactor facilities such as coupled thermal/fast uranium and boron carbide spherical assembly (Sigma-Sigma), Coupled Fast Reactivity Measurement Facility (CFRMF), Intermediate-energy Standard Neutron Fields (ISNF), and Glory hole of the Tokyo University research reactor (YAYOI) [10, 11]. Their spectra were included in the IRDF-2002 database [3] however measurements in these neutron fields were not previously incorporated into the evidence package supporting validation of the IRDFF cross section library.

In the present analysis we selected the following facilities and spectra from the IRDF-2002 collection [3, 10]: ISNF – material number MAT=4, 620 groups representation; CFRMF – MAT= 5, 460 groups; Sigma-Sigma – MAT= 7, 431 groups; YAYOI – MAT= 9, 100 groups. The IRDF-2002 spectra were copied from the IRDF-90 database [18], which was assembled in 1993. We failed to find the primary sources of numerical information for these spectra. The spectra of these facilities are displayed in Fig. 5.

The ISNF spectrum obviously has unphysical irregularities in the vicinity of 0.4 MeV. Failing to find original data, we backed to the available description of the facility [10]. According to this document the samples were irradiated within a ^{10}B shell, around which eight ^{235}U source disks were mounted symmetrically and near the cavity within the graphite column of the NBS research reactor. The graphite column was a source of thermal neutrons. The simulation model of this facility, depicted in Fig. 6, is also available [10] and gave us a chance for reanalysis. For this we used the MCNP5 code while the

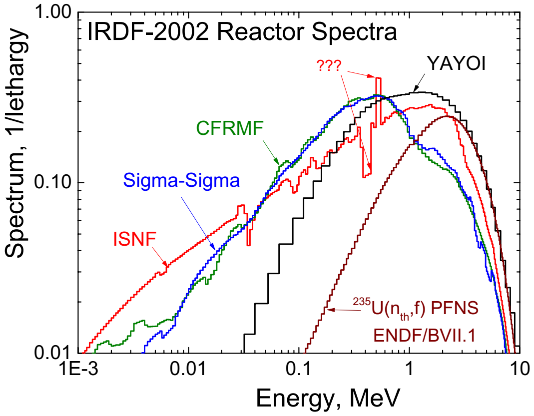

Figure 5. Reference reactor facilities spectra available in IRDF-2002 [3] and ^{235}U(n$_{\rm th}$,f) PFNS from ENDF/B-VII.1. The Question marks point to unphysical oscillations.

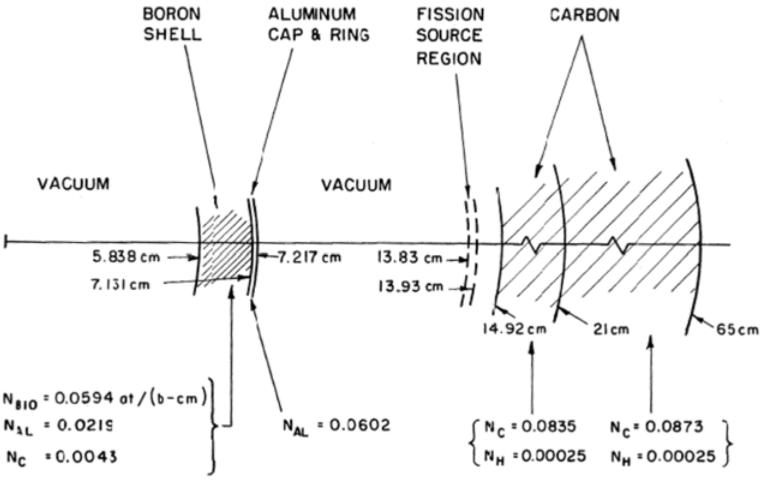

Figure 6. Geometry and material specifications of the ISNF facility used for transport calculations by authors of [10] and in the present work.

neutron transport cross sections and ^{235}U(n$_{\rm th}$,f) PFNS were taken from ENDF/B-VII.1. The calculated neutron energy distribution within the irradiation volume is shown in Fig. 7. It is seen that the shape is very close to the ISNF spectrum stored in IRDF-2002 and also exhibits resonances caused by the Al cap of the boron shell, but without oscillations near 0.4 MeV. This new spectrum calculated in the standard 640 groups presentation was used to compute SPA.

The C/E ratios for SPA are presented in Fig. 8. The experimental reaction cross sections were taken from EXFOR and checked against original publications [19–27] (they were not corrected for modern monitors or decay data). Some experimental data are spectral indices, i.e. reaction rate ratios measured relative to ^{235}U(n,f) [21, 22, 24], ^{238}U(n,f) [24] and ^{197}Au(n,γ) [25]. The energy-averaged cross sections were computed using IRDFF-1.03 and facility spectra available in IRDF-2002 or one recalculated for ISNF.

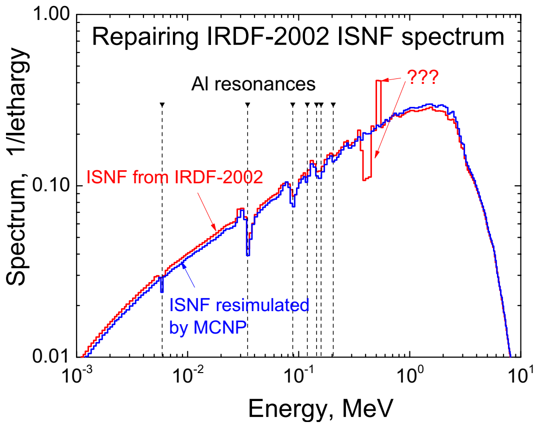

Figure 7. The ISNF facility spectrum: original from IRDF-2002 (red) and recalculated in this work (blue). Arrows indicate Al resonances.

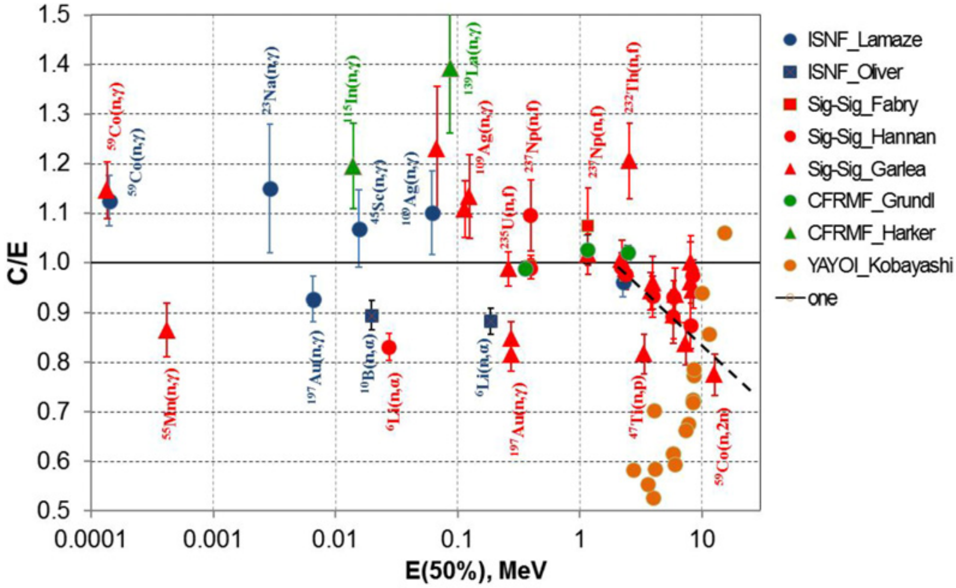

Figure 8. C/E ratios for SPA in the ISNF, CFRMF, Sigma-Sigma and YAYOI research reactor facilities.

We also tried to use the YAYOI spectrum from IRDF-2002 and SPA measured by K. Kobayashi et al. [27]. However practically all C/E ratios turned out to be below unity by 20–50%, Fig. 8. One possible reason may be the rather course 100 groups representation of energy spectrum, or a misinterpretation of the published measured values. Alternative YAYOI spectra received through private communications did not improve the agreement.

Fig. 8 displays ratio of calculated to experimental SPA or ratio of reaction rates, whereas $E_{50\%}$ is the mean response energy calculated for the SPA The error bars displayed in Fig. 8 include the uncertainty only from experimental and IRDFF-1.03 cross sections (the spectra uncertainties are not taken into account since they are not available in IRDF-2002). It is seen that deviation of C/E from unity, for most reactions and facilities (except YAYOI, whose spectrum is thought to be incorrectly presented in IRDF-2002), lies within two to three uncertainty bars.

It is interesting to observe in Fig. 5, that these research reactor benchmarks have much softer spectra than the fission source ^{235}U(n$_{\text{th}}$,f) and thus they are able to deliver validation for reactions not validated so far with the ^{252}Cf or ^{235}U(n$_{\text{th}}$,f) such as the (n,γ) reaction on ^{45}Sc, ^{55}Mn, ^{58}Fe, ^{109}Ag – the observed agreement, as seen in Fig. 8, is within 2-3 uncertainty bars. These results also confirm an underestimation of (n,α) reaction for ^{6}Li, ^{10}B observed in ^{235}U(n$_{\text{th}}$,f) PFNS and an agreement for (n,f) reaction on ^{232}Th, ^{238}U, ^{237}Np in both the ^{252}Cf and ^{235}U(n$_{\text{th}}$,f) fields.

As for the reactions sensitive to neutron energies above 10 MeV, we may compare benchmark results only for the ^{59}Co(n,2n) reaction with $E_{50\%} = 12.7$ MeV: $C/E = 0.78 \pm 0.04$ in Sigma-Sigma and $C/E = 0.94 \pm 0.02$ (ENDF/B-VII.1) or 1.14 ± 0.03 (Scale method) in the ^{235}U(n$_{\text{th}}$,f) PFNS, the spectra uncertainties are excluded from analysis. It should be noted that the Sigma-Sigma facility spectrum was calculated using the ANISN discrete ordinate neutron transport code and the older ENDF/B-III library [22].

4. Conclusions

Validation of the latest IRDFF library, version 1.03, has been performed against spectrum-averaged cross sections measured in ^{252}Cf(s.f.) and ^{235}U(n$_{\text{th}}$,f) fission neutron spectra and in several research reactor fields.

The best agreement between experimental and evaluated cross sections was observed for the standard ^{252}Cf(s.f.) spectrum. Performance in both of the representations of the ^{235}U(n$_{\text{th}}$,f) PFNS, one based on Madland-Nix model and the other derived from the time-of–flight measurements (Scale Method), shows similarly acceptable results below 10 MeV and different tendency for high threshold reactions. The final decision is difficult to make basing on the experimental spectrum-averaged data available now, thus new measurements are highly recommended. Extension of the spectra above 20 MeV increases the calculated SPA in some cases by 1–3% or even more for reactions with $E_{50\%} > 13$–14 MeV.

A collection of research reactor neutron spectra in the IRDF-2002 library was used in support of the validation of the IRDFF library. The ISNF spectrum was recalculated to correct the unphysical oscillations seen in the IRDF-2002 spectrum near 0.4 MeV. The YAYOI spectrum available in IRDF-2002 was found to be unsuitable for validation purpose. Experimental data from the above mentioned reactor facilities were used to confirm or provide new preliminary validation results of IRDFF-1.03 in addition to the validation evidence observed for the ^{252}Cf(s.f.) and ^{235}U(n$_{\text{th}}$,f) benchmark fields. However for the final validation, a complete characterization of the neutron spectra including uncertainties will be needed.

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