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INTEGRAL DATA ASSIMILATION ON U235 & U238 NUCLEAR DATA AND IMPACT ON FCA-IX SPECTRAL INDICES TO REASSESS MINOR ACTINIDES FISSION CROSS SECTIONS

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ABSTRACT

Critical mass calculations of various HEU-fueled reactors show there is space for improvement in current U235 & U238 nuclear data evaluations and covariances in the fast energy range. This work makes use of Bayesian Inference method as implemented in the CONRAD code. Experimental database used includes ICSBEP Uranium based critical experiments and benefits from recent re-analyses of MASURCA and FCA-IX criticality experiments (with Monte-Carlo calculations) and of PROFIL irradiation experiments. These last ones provide very specific information on U235 & U238 capture cross sections. Our integral experiment assimilation work notably suggests a 30% decrease for U235 capture around 1-2 keV, a 10% increase in the URR when using JEFF3.1.1 as “a priori” data. The work was focused on JEFF-3.1.1 U235 & U238 evaluations but also on minor actinides fission cross sections. An underestimation of 3-4% for Np237 fission and 4% for Am241 in JEFF3.1.1 is suggested by FCA-IX fission chamber results, in agreement with recent differential measurements. For Pu242 fission, FCA-IX C/E highlights a need for re-normalization for some differential measurements, as an overestimation of around 10% is observed. For Pu238 fission, the dependency on spectrum of the C/E calculated allows us to identify an overestimation of this cross section in JEFF-3.1.1 below the threshold.

KEYWORDS: assimilation, integral experiment, CONRAD, minor actinides.

1. INTRODUCTION

Critical mass calculations of various HEU-fueled reactors display large discrepancies in C/E values, depending on flux spectra, fuel enrichment, structural material present and so on. These C/E values, calculated with the Monte-Carlo code TRIPOLI-4 [1], are shown in Figure 1 and Table I gives some specifications about fuel and structural materials present in each configuration.

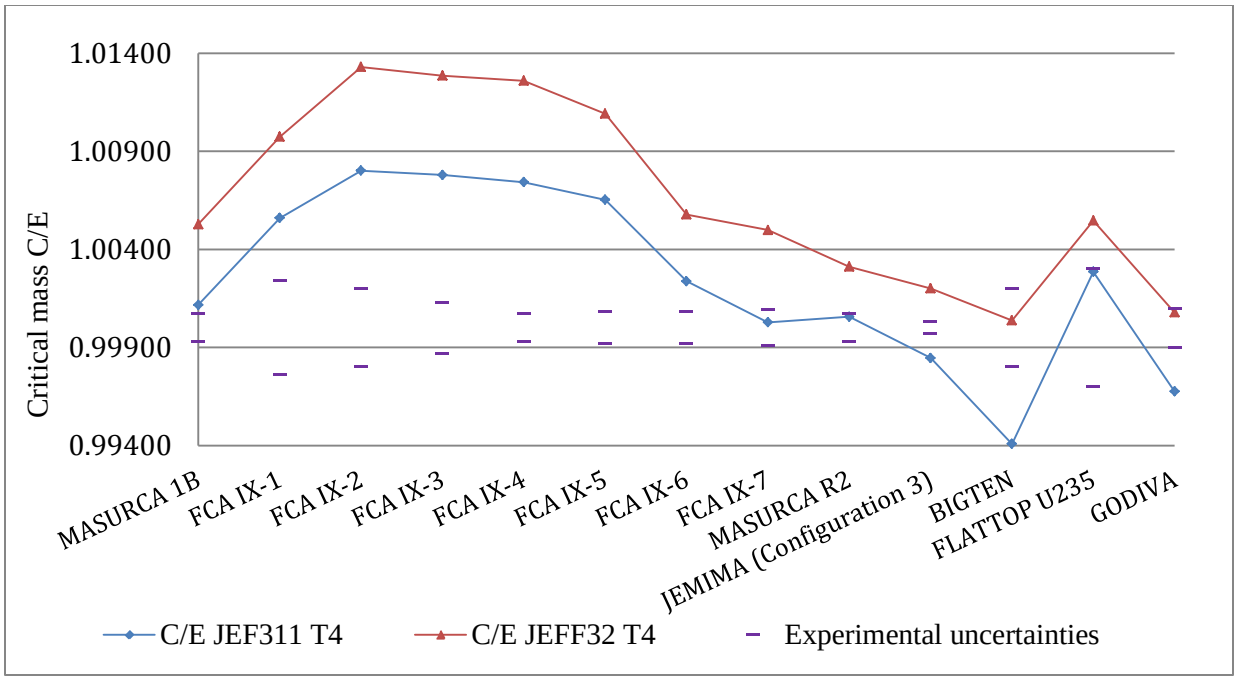


Figure 1. Critical mass C/Es compared to experimental uncertainties for Uranium-fueled configurations (using JEFF libraries).

Table I. Specifications on fuel enrichment and structural materials for each configuration.

Configuration	Fuel enrichment	Structural material and diluent	Configuration	Fuel enrichment	Structural material and diluent
FCA-IX 1	93%	Graphite	MASURCA 1B	30%	Graphite
FCA-IX 2	93%	Graphite	MASURCA R2	30%	Sodium
FCA-IX 3	93%	Graphite	JEMIMA (configuration 3)	Alternation of HEU (93.4%) fuel and Unat disks	Steel
FCA-IX 4	93%	Stainless Steel	BIGTEN	10% in average (with Udep reflector)	Steel
FCA-IX 5	93%	Stainless Steel	FLATTOP-U235	93% (with Unat reflector)	-
FCA-IX 6	93%	Stainless Steel	GODIVA	94%	-
FCA-IX 7	20%	-			

Figure 1 underlines an overestimation of critical mass for Uranium-fueled configurations with JEFF-3.2 library, except for ICSBEP benchmark BIGTEN and GODIVA [2]. When compared to JEFF-3.1.1 calculations, C/Es calculated with JEFF-3.2 are systematically higher. Discrepancy between the two sets of calculations goes from ~250pcm up to ~630pcm for BIGTEN. Moreover, large C/E are observed for FCA-IX configurations [3] (overestimation up to ~800pcm), BIGTEN and GODIVA when using the JEFF-3.1.1 library. Except for FLATTOP-U235, all critical masses for configurations with HEU fuel exceed experimental uncertainties when using JEFF-3.1.1. Given that MASURCA 1B and FCA-IX configurations have similar spectra (as they both contain graphite) but significantly different Uranium

enrichment, the large discrepancy observed in their C/E values (using either JEFF-3.1.1 or JEFF-3.2) rise concerns as to possible compensating errors between U235 and U238 evaluation in the JEFF libraries in the fast and epithermal energy range.

2. INTEGRAL EXPERIMENTS ASSIMILATION

Considering the very large C/E values presented in Section 1, it seems relevant to use integral data assimilation to attempt to identify which nuclear data are responsible for these discrepancies. This was done using the CONRAD code from CEA [4], which can solve analytically Bayes' theorem.

The JEFF-3.1.1 library was chosen as the a priori as it gives more satisfying results than JEFF-3.2 for Ura nium configurations, as seen in Figure 1. For our assimilation work, we used critical mass C/Es of MASU RCA 1B, FCA-IX cores 1 to 7, FLATTOPU235 and GODIVA, as well as variation of concentration ratio C/E from PROFIL-2A irradiation experiment [5] [6].

The nuclear data fitted through assimilation are U235 and U238 capture, elastic, inelastic cross sections as well as their fission spectrum $\boldsymbol{\chi}$ and multiplicity $\boldsymbol{\upsilon}$. U235 and U238 fission were not fitted, as JEFF-3.1.1 evaluation are in good agreement with Neutron Standard from IAEA [7] for these cross sections.

Nevertheless, as this will be mentioned in the following sections, high uncertainties associated to fission spectra can have an impact on assimilation result. Also, as differences in JEFF-3.1.1 and JENDL-4.0 carbon evaluations were found to have a non negligible impact for some critical masses used in this work, we ran CONRAD calculations considering these alternatives[8]. For these reasons, in section 3 are presented sets of trends that include all of these alternatives (for fission spectra and carbon evaluation). C/E used in the assimilation work were calculated using the Monte-Carlo code TRIPOLI-4 (except for PROFIL's variation of concentrations ratios, calculated with ECCO/ERANOS) and 33-group sensitivity coefficients to nuclear data were calculated using the ECCO/ERANOS code [9]. For nuclear data covariance matrices, we used COMACV1.0 [10], except for U235 $\boldsymbol{\upsilon}$ for which we used the COMMARA-2.0 matrices.

Figure. 2 displays post-assimilation C/Es for critical mass compared with prior JEFF-3.1.1 C/E values. As for PROFIL-2A irradiation experiment, we give average C/E value along with associated experimental uncertainties : C/E $(\frac{U_{235}+U_{236}}{U_{238}}) \approx 0{,}9993 \pm 2\%$ and C/E$(\frac{U_{236}}{U_{235}}) \approx 1{,}005 \pm 1{,}7\%$.

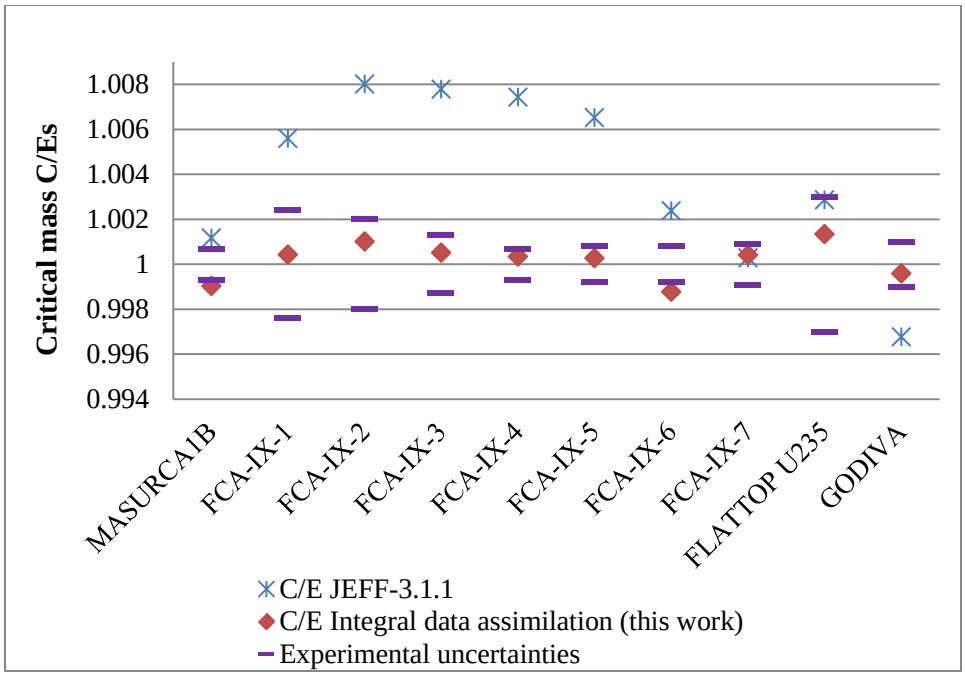


Figure. 2. Comparison between prior (JEFF-3.1.1) and posterior C/E values

Post-assimilation C/Es are well-included in 1σ experimental uncertainties, except for FCA-IX 6, which however remain in 2σ total uncertainties. This means there exists an optimal set of cross sections for the experimental database taken into account, as no inconsistency between C/E had been found.

3. COMPARISON OF ASSIMILATION TRENDS WITH DIFFERENTIAL MEASUREMENTS

In order to discuss reliability of the trends on cross sections suggested through the integral data assimilation, we compared them to differential measurements from the EXFOR database [11]. Trends are given relative to JEFF-3.1.1.

Assimilation results suggest significant change on U235 capture: a ~30% decrease around 1-2 keV and a ~10% increase in the Unresolved Resonance Range (URR) when using JEFF3.1.1 as “a priori” data. This is shown in Figure 3, along with prior and posterior uncertainties. We can observe that from 1keV to 500keV, posterior uncertainties are sufficiently low to consider assimilation trends as possible recommendations for a change in U235 capture cross section. As mentioned earlier, the set of trends displayed in Figure 3 takes into account uncertainties on graphite evaluation and whether fission spectra should be fitted or set to JEFF-3.1.1.

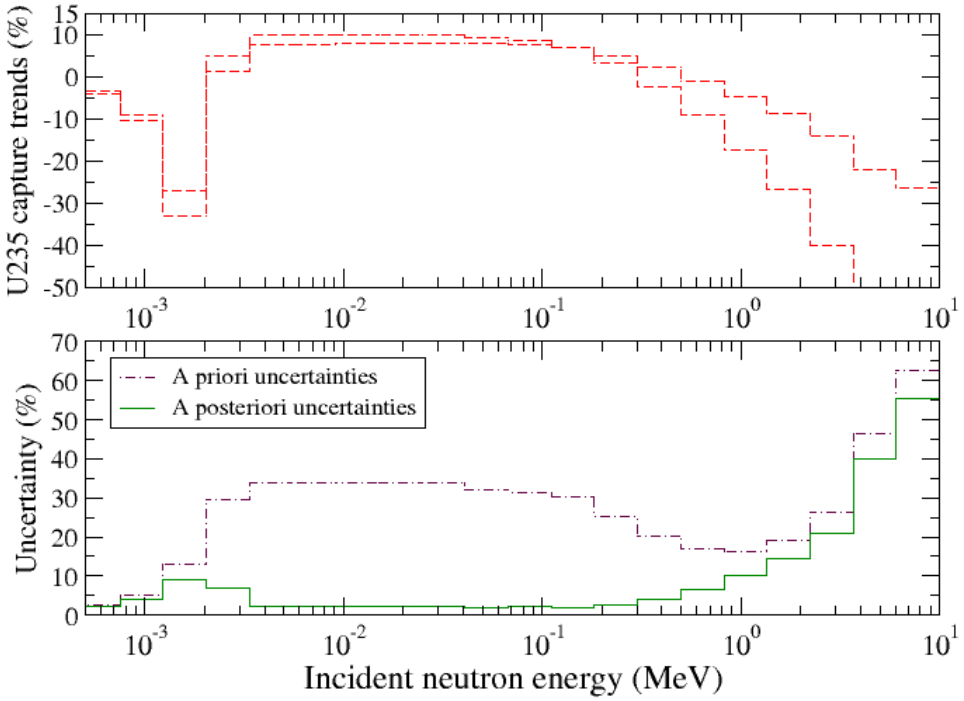


Figure 3. Trends from assimilation work for U235 capture (relative to JEFF-3.1.1) compared with a priori and a posteriori nuclear data uncertainties.

Focusing on the end of Resolved Resonances Region (RRR) from 1keV to 2.25keV, we compared our assimilation trends in this energy range with recent differential measurements made at RPI. Figure 4 display results of these measurements as published in reference [12] (they are not currently available in the EXFOR database) with a comparison to ENDF/B-VII and JENDL-4.0. It is to be noted that for U235 capture cross section, JEFF-3.1.1 and ENDF/B-VII.1 are identical. This graph shows that Danon measurements at RPI are in good agreement with our assimilation results as they suggest a -33% decrease of U235 capture cross section from JEFF-3.1.1 at 2keV.

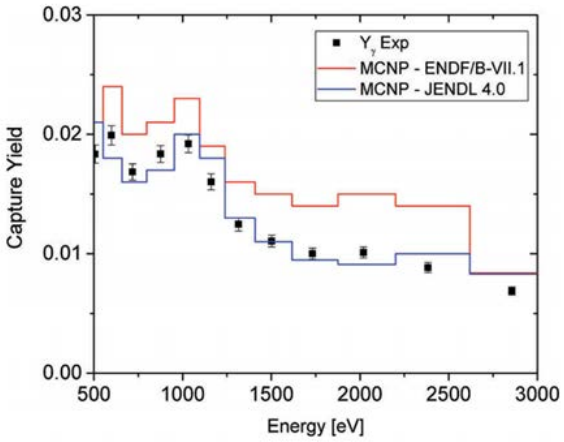


Figure 4. Results of differential measurements from Danon et al. from reference [12] for U235 capture from 0.5 to 3 keV, compared with ENDF/B-VII.1 and JENDL-4.0 evaluations.

In the Unresolved Resonance Range (URR), most recent measurement performed by Jandel et al. [13] at LANSCE with the DANCE detector are consistent with assimilation trends from 1keV to 1MeV (see Figure 5).

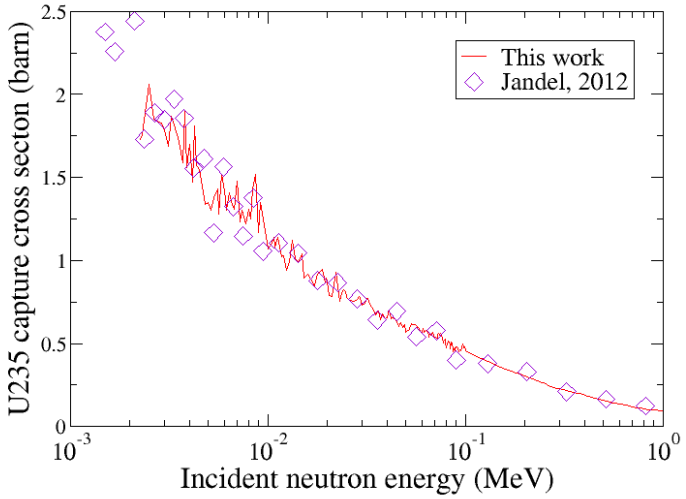


Figure 5. Results of differential measurements from Jandel et al. [13] for U235 capture from 0.5 to 3 keV. Comparison with assimilation results applied to JEFF-3.1.1 point-wise evaluations (red line).

Comparing now assimilation results to JEFF-3.3t3 (in Figure 6), one can see that they agree well in the end of RRR (considering that assimilation results uncertainties in this range is around 10%). In the URR, from 10 keV to 100 keV, JEFF-3.3t3 proposes a higher increase from JEFF-3.1.1 (around 20%) than our assimilation results.

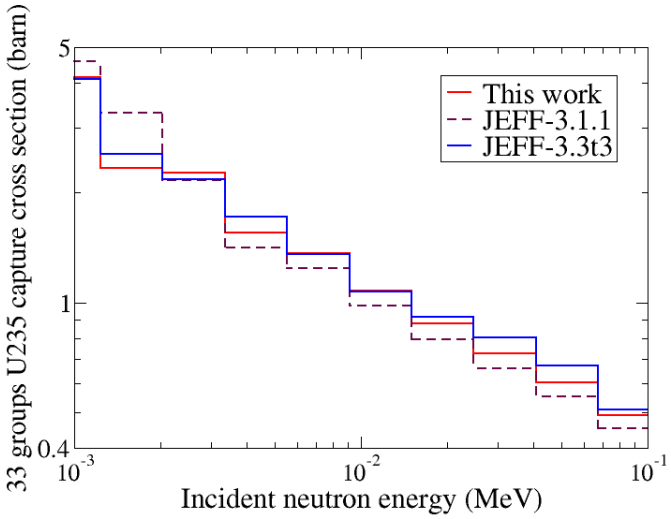


Figure 6. 33-group assimilation results for U235 capture compared with "a priori" JEFF-3.1.1 and JEFF-3.3t3 evaluation.

Apart from that, it was found that assimilation results for U238 capture and inelastic cross sections are highly dependent on the fission spectra used. Nevertheless, results for U238 inelastic suggest a 4 to 8 % decrease in the plateau region (from 1 MeV to 6 MeV), which goes in the same direction as CIELO and JEFF-3.3t3 evaluations. A modification of these cross sections as suggested by assimilation results can have a significant impact on fast reactor calculations [8].

4. IMPACT ON FCA-IX SPECTRAL INDICES

4.1. FCA-IX spectral indices

The FCA-IX experimental program [3] is composed of seven HEU fuel plate assemblies which display various flux spectra covering a large energy range in the epithermal to fast energy domain, as shown in Figure 7. Spectral indices, which are the ratio of several minor actinides fission rates (Np237, Pu238, Pu242, Am241, Am243 and Cm244) over Pu239 fission rate, were measured in the centre of the seven cores using fission chambers [14]. Absolute measurement of these fission rates ratios permits to overcome issues due to fission chambers calibration.

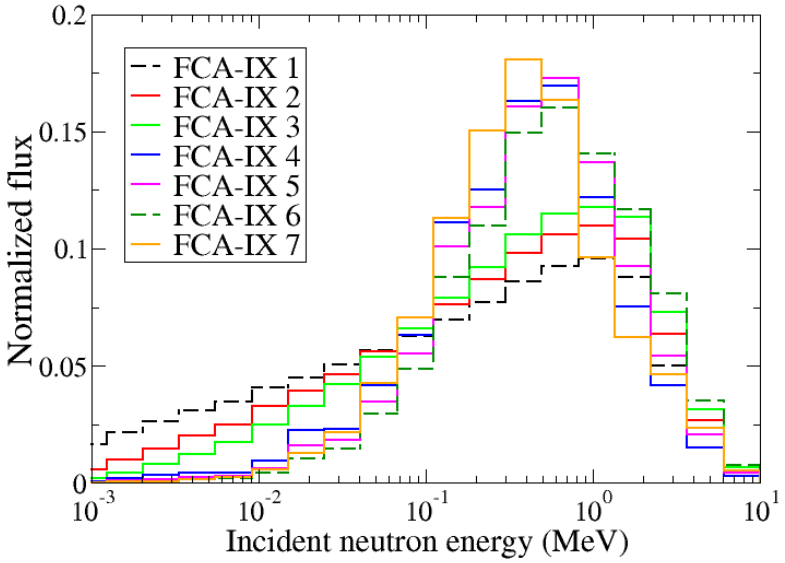


Figure 7. 33 groups flux spectra for FCA-IX cores 1 to 7

This set of measurements is thus particularly interesting for issuing recommendations on minor actinides fission cross sections around 1 MeV. We calculated spectral indices with the ECCO/ERANOS code, using JEFF-3.1.1 library. Experimental uncertainties for each spectral index are given in Table II.

Table II. Range of experimental uncertainties for each spectral indices.

	IS_F37/F49	IS_F48/F49	IS_F42/F49	IS_F51/F49	IS_F53/F49	IS_F64/F49
Exp. Uncertainties	0.4-0.9%	1.5-2.2%[1]	0.3-0.7%	0.4-1%	0.3-0.8%	0.9-2%

[1] Except in FCA-IX 6, for which total experimental uncertainties associated to Pu238 spectral index reach 6%.

4.2. Impact of U235&U238 assimilation work on FCA-IX spectral indices

As the assimilation work done on U235&U238 nuclear data resulted in significant change in some major cross sections (U235 capture, etc.), we quantified the impact of these trends on the calculation of FCA-IX spectral indices. For any of the FCA-IX configurations, the impact of trends suggested by assimilation work on U235/U238 on C/E is not large (1 to 3% change on spectral indices values) when compared to discrepancies from C/E=1. A summary of C/E results (taking into account of this impact) is given in Figure 8.

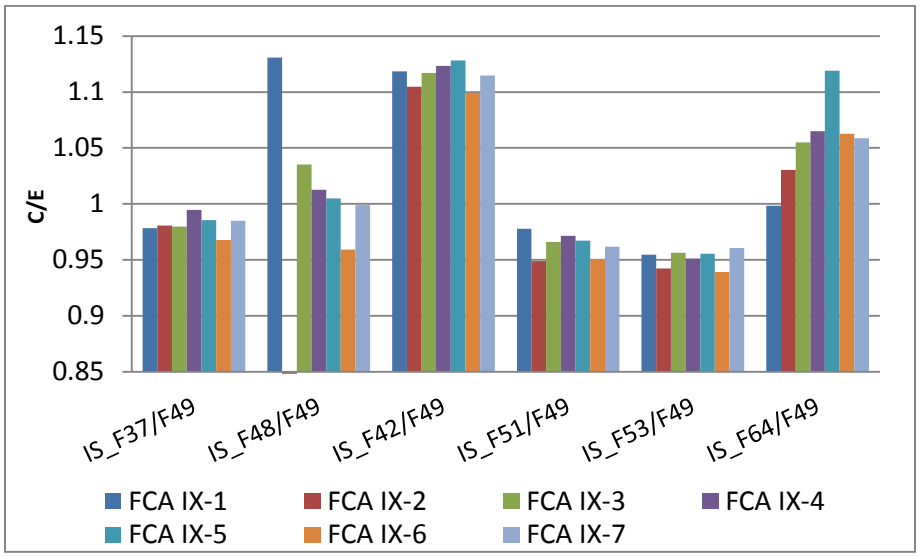


Figure 8. Spectral indices C/E (corrected for assimilation trends from our assimilation work) for FCA-IX cores 1 to 7. Experimental data for Pu238 spectral index in FCA-IX are not available.

For Np237, Pu242, Am241, and Am243 spectral indices, discrepancy from C/E~1 are more or less the same for all FCA-IX configurations. This means FCA-IX integral data can possibly detect normalization issues in differential measurements used for JEFF-3.1.1 evaluations. For Pu238 and Cm244 spectral indices, we observe a change on C/E values depending on the spectrum in which it was calculated.

5. DISCUSSION ON MINOR ACTINIDES FISSION CROSS SECTION

The work on FCA-IX spectral indices presented in previous sections allows us to make recommendations on minor actinides fission cross sections. Indeed, we showed that indirect contribution due to great change in U235 and U238 cross sections were small compared to C/E discrepancies. Trends on minor actinides fission from FCA-IX spectral indices measured in various energy spectra are discussed in this section in the light of a comparison with differential measurements from the EXFOR database [11].

5.1. Am241 fission cross section

FCA-IX results on Am241 fission suggest a 4% increase from JEFF-3.1.1. Figure 9 shows a comparison of JEFF-3.1.1 and JEFF-3.3t3 evaluations with differential measurements from Dabbs [15] and Aleksandrov [16] [17]. JEFF-3.1.1 evaluation is consistent with Dabbs measurements. However, in the threshold and plateau region, total experimental uncertainties reach 4-5%. Aleksandrov (1983) seems in agreement with integral data trends but only one measurement is available at 2.9MeV in the EXFOR database. New differential measurements with reduced uncertainties are needed. However, JEFF-3.3t3 evaluation suggests an increase of Am241 fission in the plateau region, in better agreement with Aleksandrov measurements.

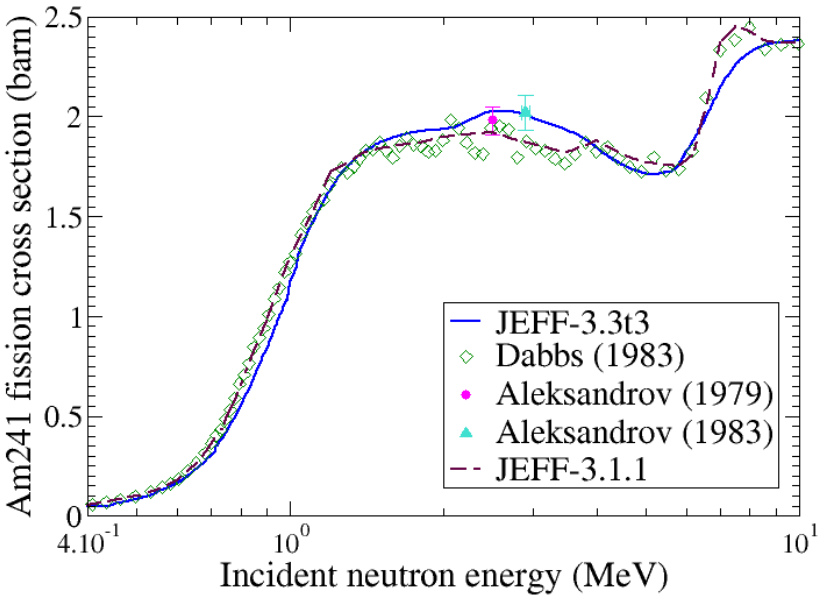


Figure 9. Comparison of JEFF-3.1.1 and JEFF-3.3t3 evaluations for Am241 fission cross section with differential measurements

5.2. Pu242 fission cross section

For Pu242 fission cross section, FCA-IX spectral indices indicate a 10% overestimation in JEFF-3.1.1. Figure 10 displays JEFF-3.1.1 evaluation, which is based on Weigmann measurements [18], and compares it to Tovesson measurements [19]. In the plateau region (from 1 to 5 MeV), Weigmann measurements differ from Tovesson's of a factor 1.05. As, FCA-IX integral data, Tovesson measurements thus push towards a decrease of JEFF-3.1.1 cross section, though at a lower level. Experimental uncertainties for Tovesson measurements in the range of energy of interest are of 2-3%.

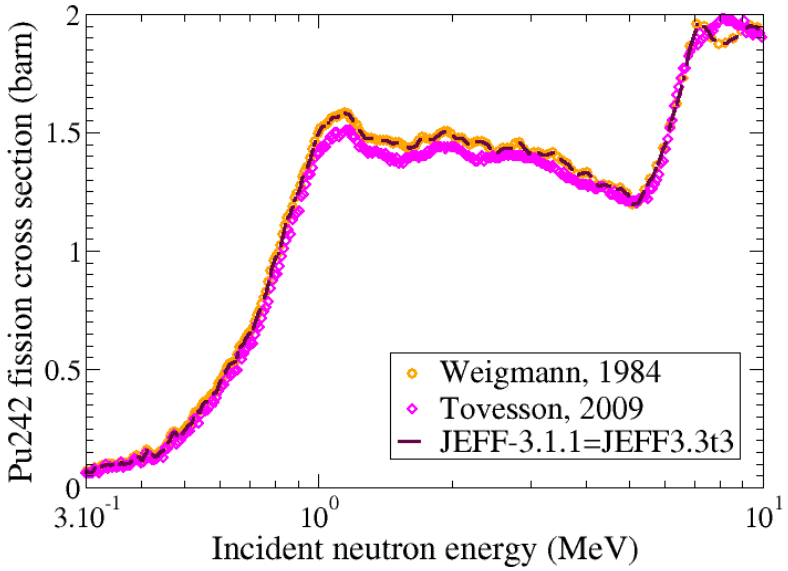


Figure 10. Comparison of JEFF-3.1.1 and JEFF-3.3t3 evaluations for Pu242 fission cross section with differential measurements.

5.3. Np237 fission cross section

Np237 spectral indices are underestimated of around 3% in FCA-IX configurations. JEFF-3.1.1 evaluation is compared to recent differential measurements from Diakaki [20] in Figure 11. From 500 keV to 1.4 MeV, Diakaki measurements are higher than JEFF-3.1.1. Notably the bump around 1MeV is underestimated of around 4% when compared to differential measurements. Note that for this energy range, total experimental uncertainties associated to Diakaki are of 2.6% (including 1σ statistical uncertainties of 2%). It is worth pointing out that ENDF/B-VII.1 is in good agreement with Diakaki measurements for this energy range, as this evaluation has been chosen for JEFF-3.3t3.

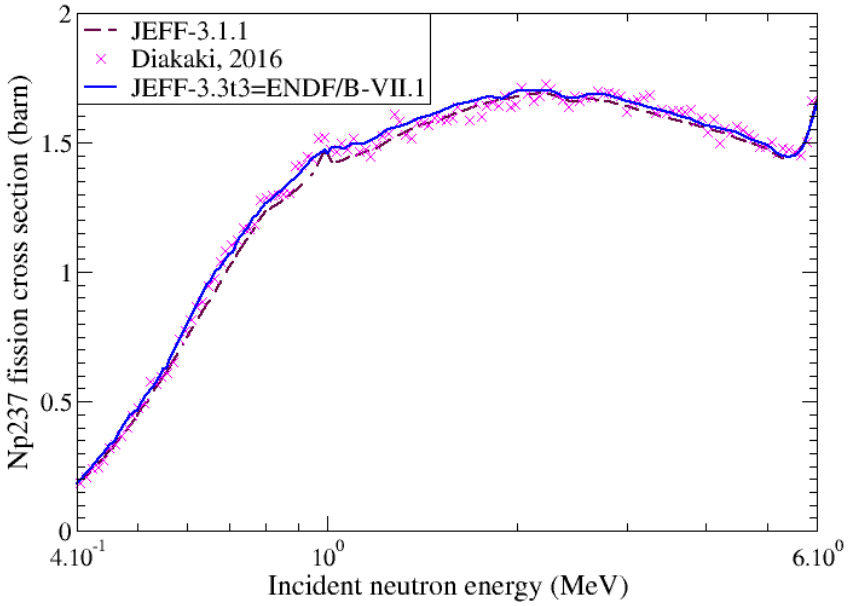


Figure 11. Comparison of JEFF-3.1.1 and JEFF-3.3t3 evaluations for Np237 fission cross section with differential measurements.

5.4. Pu238 fission cross section

For Pu238 spectral indices, we observe that C/E discrepancies depend on the spectrum. This indicates that Pu238 fission is overestimated in JEFF-3.1.1 evaluation below the threshold, since FCA-IX 1 and 3, whose spectra are the softest, are larger than 1. For FCA-IX 4, 5 and 7, C/Es are equal to 1. For FCA-IX 6, the 6% experimental uncertainties are too high to make any comment.

In the case of Pu238 fission cross section, current evaluations sensibly differ. In Figure 12 (left), JEFF-3.1.1 evaluation is compared to JENDL-4.0 from 1keV to 100keV. In RRR, JEFF-3.1.1 clearly overestimates Pu238 fission cross section. Indeed, as one can see from Figure 7, FCA-IX 1 to 3 are more sensitive to this energy range than FCA-IX 4 to 7, which explains their larger C/E. When calculating FCA-IX spectral indices with JENDL4.0, this spectrum effect disappears [21]. However, C/E values calculated with JENDL-4.0 are underestimated of 5% for all FCA-IX configurations. The impact of changing Pu239 fission evaluation on Pu238 spectral index calculation should be small compared to C/E discrepancies since main differences between JEFF-3.1.1 and JENDL-4.0 evaluations are below 25keV for this cross section, and no spectrum effect is found for JENDL-4.0 spectral indices calculations.

In Figure 12 (right), JEFF-3.1.1 is compared to JENDL-4.0 and differential measurements from Drake [22], Silbert [23] and Budtz-Joergensen [24]. These differential measurements, which were all used for JENDL-4.0 and JEFF-3.1.1 evaluation processing, have respectively experimental uncertainties of 8-9%, 5-10% and 3-4 %. Due to lower uncertainties, JENDL-4.0 follows Budtz-Joergensen measurements well from 0.1 to 5 MeV. However, FCA-IX integral data suggest a 5% increase in Pu238 fission cross section for JENDL-4.0 after the threshold. For JEFF3.3t3, JENDL-4.0 evaluation was chosen.

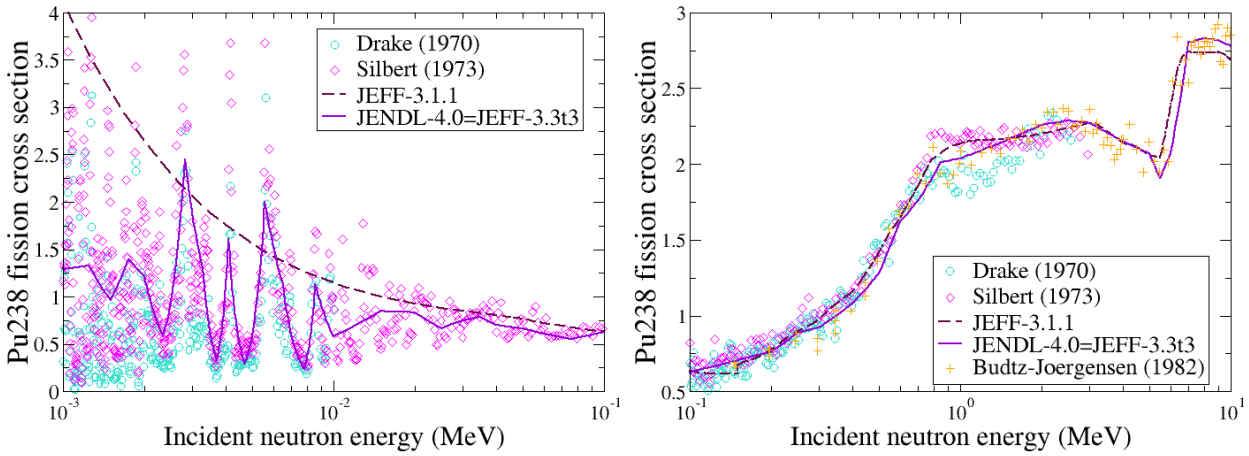


Figure 12. Current evaluations compared with differential measurements from 1keV to 100keV (left) and from 100 keV to 10 MeV (right)

6. CONCLUSIONS

C/E values from a great variety of critical masses as well as PROFIL sample irradiation experiments were used in a Bayesian inference approach as implemented in the CONRAD code to investigate cross sections that might need reassessment. Notably, trends suggested for U235 capture, which are in agreement with recent differential measurements made at RPI and LANSCE, confirm that significant modifications are needed for this cross section in JEFF-3.1.1. JEFF-3.3t3 seems to go in the right direction with a decrease of around 25% from JEFF-3.1.1 in the end of RRR and an increase of around 20% in the URR.

Such modifications can have a significant indirect impact on reaction rates measured in zero power reactor cores such as it was done in FCA-IX cores. However, FCA-IX spectral indices calculations were not so much affected by these changes on U235 capture cross sections. This enabled us to be more confident about making recommendations for some minor actinides (Am241, Np237, Pu238 and Pu242) fission cross sections using these spectral indices. These were made by comparing C/E of FCA-IX spectral indices to differential measurements. The study of FCA-IX spectral indices suggests that Am241 and Np237 fission cross section in JEFF-3.1.1 are underestimated in the plateau region, and it seems that JEFF-3.3t3 is going in the right direction for these isotopes. Pu242 fission cross section of JEFF-3.1.1 is overestimated of around 10% according to FCA-IX integral results. FCA-IX spectral indices suggest a 5% underestimation of Pu238 fission cross section above the threshold for JENDL-4.0 evaluation, whereas JEFF-3.1.1 gives satisfying results for this energy range. Below 100 keV, JEFF-3.1.1 highly overestimates this cross section. Further assimilation work on Pu239 cross sections, including fission, should allow us to consolidate these results on minor actinides fission cross sections.

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