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SANDA Project D4.2: Report on new nuclear reaction data evaluation

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The nuclear reaction models used to perform evaluations are different fow low and high energies and implemented in different codes. The code developments are therefore described in two different sections. The first one provides details on the CONRAD code, and focusses on the resonances analysis of major as well as minor actinides, while the second section discusses various modifications included in the TALYS code, and illustrates how these modifications have been applied to obtain a new evaluation of 239Pu.

1. Evaluation of the neutron-induced cross sections of actinides using the CONRAD code

A longstanding evaluation work was undertaken within the JEFF community to provide new resolved resonance range of actinides for JEFF-4. The evaluation work presented in this section was performed with the CONRAD code.
The CONRAD code is an object-oriented software tool developed at CEA since 2005 [dsj21]. It aims at providing nuclear reaction model calculations, data assimilation procedures based on Bayesian inference and a proper framework to treat all uncertainties involved in the nuclear data evaluation process. In the resonance range of the neutron cross sections, the models relies on the R-Matrix formalism. For example, CONRAD can handle the Multi-Level Breit-Wigner and Reich-Moore approximations with the LRF7 format and Brune parametrization. Many evaluations were produced with the CONRAD code. Results obtained for ^{235}U, ^{238}U, ^{239}Pu, ^{243}Am, ^{240}Pu and ^{242}Pu are presented.

1.1. Presentation of the CONRAD code

CONRAD can produce evaluated nuclear data over a wide neutron energy range. The interface for the cross section models allows using existing implementations, external codes or new models that can be introduced by users according to specific development rules. Another option allows testing miscellaneous models before their implementation in CONRAD. This option was used to generate covariance matrices for thermal scattering laws, to evaluate Thermal Neutron Constants and to performed non-model fit of high neutron cross sections. The covariances between the model parameters can be calculated by Monte-Carlo or via an analytical marginalization procedure [tam21]. A concise method for storing and communicating large covariance matrix was implemented in the CONRAD code. It relies on the AGS formalism developed at JRC-Geel [bec12].

Resonance parameters are extracted from data measured by the time-of-flight technique, for which experimental models are needed to accurately reproduce the experimental conditions. The three main corrections, which are routinely used in an evaluation work, are the Doppler effect, the response function of the facility and the multiple scattering correction. Evaluation works on actinides were the opportunity to improve these experimental corrections in CONRAD.

1.2. Results on major actinides

For fissile isotopes, the theoretical description of the resolved resonance range requires specific models. One of the latest model implemented in CONRAD concerns the treatment of the (n,γf) reaction as a competitive reaction to the direct fission process. For that purpose, additional partial widths can be added to compute the (n,γf) reaction. This model was necessary for ^{235}U and ^{239}Pu. Results obtained for ^{239}Pu(n,f) is shown in Fig. 1. Fluctuations of the prompt neutron multiplicities induced by the (n,γf) contribution are shown in Fig. 2 for ^{239}Pu and ^{235}U. Below 20 eV, our model predicts nearly equivalent fluctuations for ^{239}Pu and ^{235}U with a maximum decrease of the prompt neutron multiplicity of about 4% at the resonance energy.
For the resonance analysis of ^{238}U, we precisely studied the effect of the temperature. The Doppler effect is related to the vibrations of the atoms due to the temperature. The free gas model is the most popular Doppler model. However, many studies on ^{238}U demonstrated that this simple model fails to reproduce the temperature effects observed in the low neutron energy resonances. Fig. 3 shows that the asymmetry of the first resonance of ^{238}U can only be reproduced with a crystal lattice model that accounts for the phonon density of states associated to the UO_2 molecule. Unfortunately, if CONRAD can extract resonance parameters by using this improved model, processing and neutronic codes will not be able to take into account these parameters. Generalizing Doppler models to the dynamical behaviour of the molecules will be a step forward for nuclear applications.

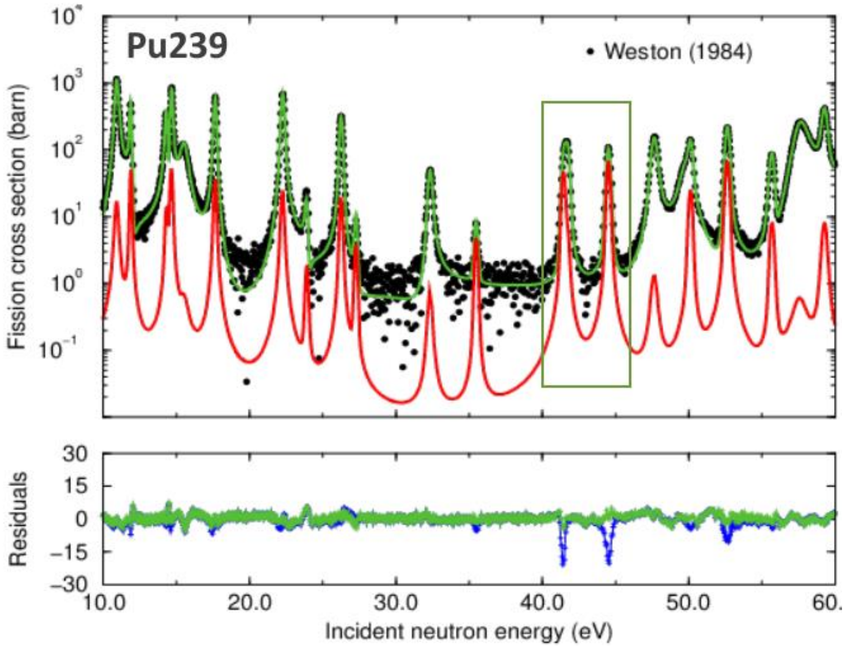

Figure 1. Contribution of the $^{239}Pu(n,\gamma f)$ reaction (red line) to the total fission reaction (green line) calculated by CONRAD with 2 additional partial widths fo spin J=0 and J=1. Residuals before (blue line) and after (green line) the adjustment of the resonance parameters are indicated in the bottom plot.

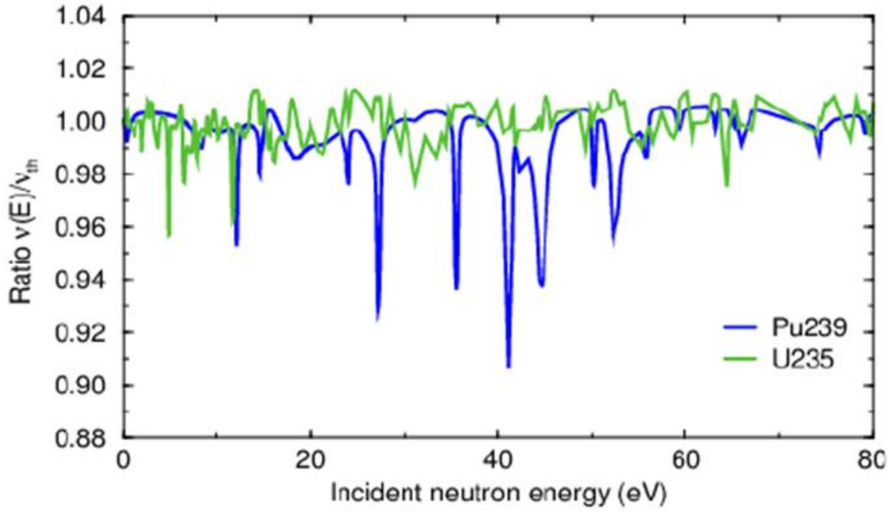

Figure 2. Comparison of the prompt neutron multiplicities for ^{239}Pu and ^{235}U calculated with CONRAD.

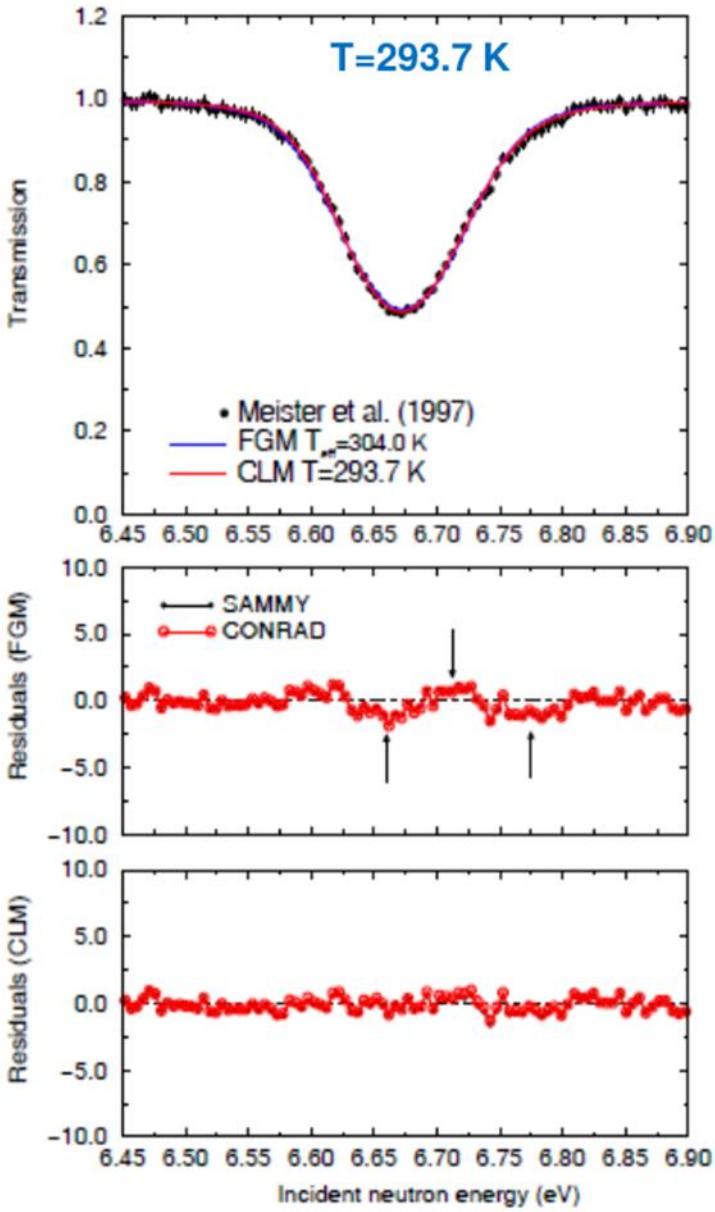

Figure 3. Comparison of the Doppler effect calculated with a free gas model (FGM) and crystal lattice model (CLM) in the case of ^{238}U transmission data measured at the JRC-Geel facility at room temperature [nog18].

For ^{239}Pu, new capture data were included in the analysis, for which new time response models were needed. The response function of the facility mainly depends on the neutron source of the facility and how neutrons are produced in the target-moderator assembly. Fig. 4. compares the ^{239}Pu resonances measured at the RPI and LANSCE facilities. The shape of the resonances is reproduced by introducing in the CONRAD calculations the time distributions shown in the right hand plot of Fig. 4. The long tail in the response function of the LANSCE facility comes from the thick spallation target. It produces the exponential behaviour observed in the right hand wing of the resonances. Such a response function makes it difficult the determination of the parameters of overlapping resonances. The analysis of ^{235}U was particularly of great interest for testing the correction due to the multiple neutron scatterings in the sample [lit13]. Figure 5 highlights the large correction that affects ^{235}U capture and fission yields measured at the RPI facility, especially in the thermal energy range.

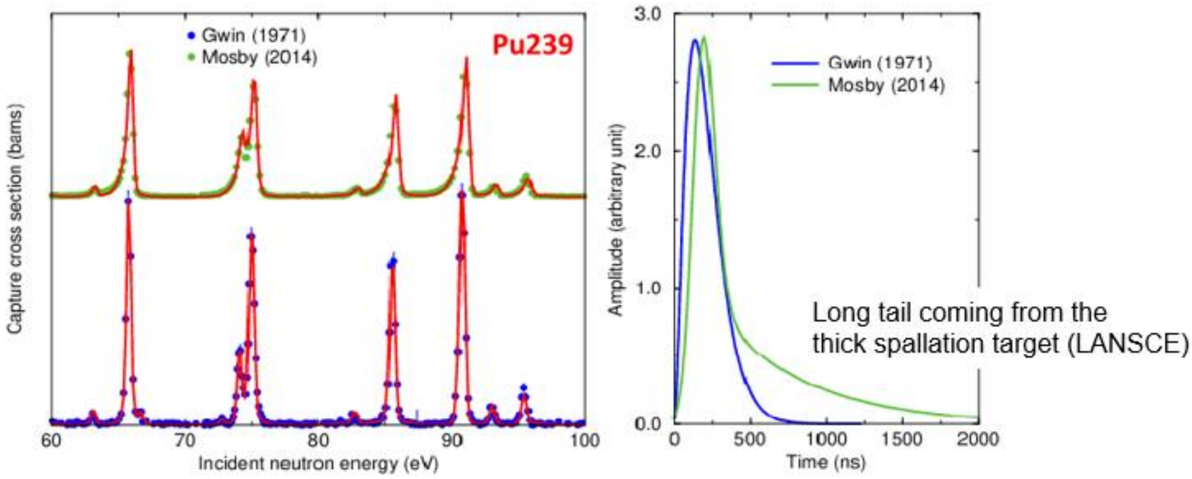

Figure 4. Resolution broadening in the case of capture data measured at RPI and LANSCE facilities. The right hand plot shows the time response function introduced in the CONRAD calculations.

The shape of the cross sections below the first resonance is of great importance in the case of well-thermalized integral benchmarks as well as for calculating neutron multiplication factors as a function of temperature. An incorrect description of the sample characteristics will have a sizeable impact on the results of these integral benchmarks.

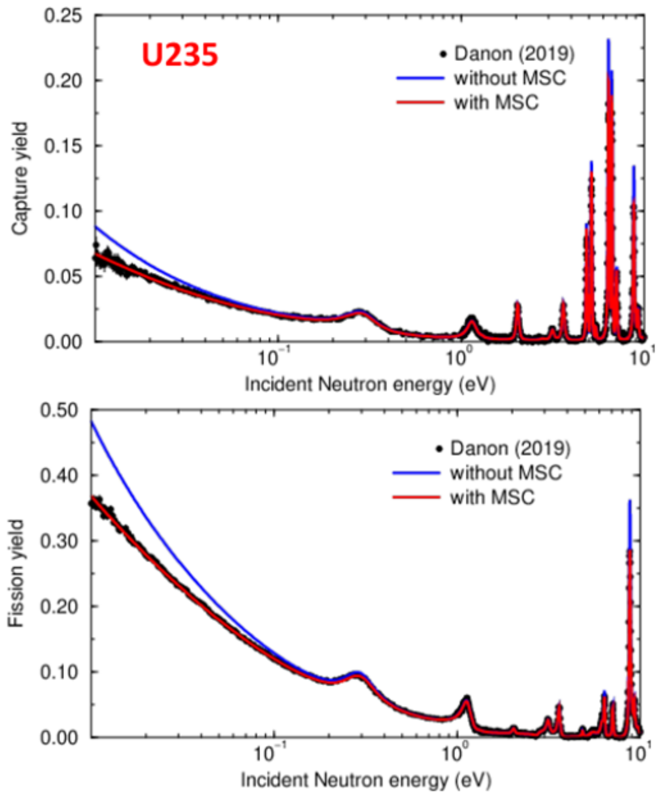

Figure 5. Effect of the multiple scattering corrections (MSC) in the case of ^{235}U capture and fission yields measured at the RPI facility.

1.3. Results on minor actinides

Evaluation work performed on minor actinides focuses on isotopes of interest for spent nuclear fuel applications. The longstanding underestimation of the 244Cm build up in UOX and MOX fuels as a function of Burn Up by the international neutron libraries motivated the revision of the capture cross sections of ^{240}Pu, ^{242}Pu and ^{243}Am. The evaluation of the resonance parameters were undertaken thanks to data retrieved from the EXFOR database [otu14]. Figures 6 and 7 compares the theoretical curves with the selected data sets.

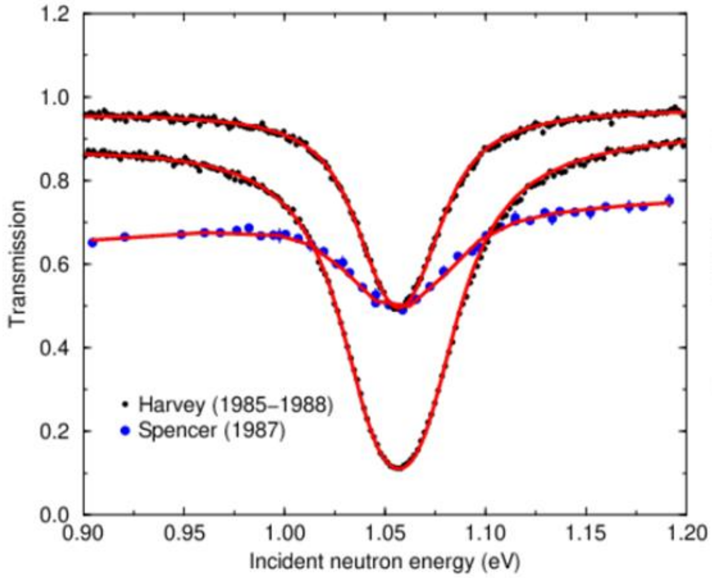

Figure 6. CONRAD analysis of three ^{240}Pu transmission experiments (1st resonance at 1.06 eV), with different response functions included in the analysis.

For the first resonance of ^{240}Pu, close to 1.06 eV, we used ^{239}Pu transmission data measured by Spencer (L=18 m, 1987) and Harvey (L=18 m, 1985-1988) in which ^{240}Pu is an impurity. Two transmission data from Kolar (L= 100~m, 1968) and a fission cross section from Weston (1984) were also included in the fitting procedure. Prior resonance parameters up to 5.7 keV were taken from the work of Bouland et al. [bou97]. The results leads to a thermal capture cross section of 285.6 barns and an increase of +4.1% of the capture resonance integral compared to JEFF-3.1.1 (I_0=8829 barns).

For ^{242}Pu, the parameters of the first resonance at 2.67 eV were derived from the total cross section of Young (1970-1971). Above 10 eV, the resonance parameters were determined with the capture yield of Lerendegui (L=185 m, 2018) and fission cross sections reported by Bergen (L=214 m, 1971) and Auchampaugh (L=245 m, 1971). The set of resonance parameters determined up to 1.5 keV leads to a thermal capture cross section 18.8 barns and an increase of +3.1% of the capture resonance integral compared to JEFF-3.1.1 (I0=1165 barns).

The evaluation of the resonance parameters of ^{243}Am were performed by using the capture yield reported by Mendoza (L=185 m, 2014), two transmission data from Simpson (1974) and the total cross section of Berreth (1970). Our analysis provides new resonance parameters up to 250 eV. The thermal capture cross section is equal to 75.5 barns. We also obtained an increase of +6.3% of the capture resonance integral compared to JEFF-3.1.1 (I_0=1902 barns).

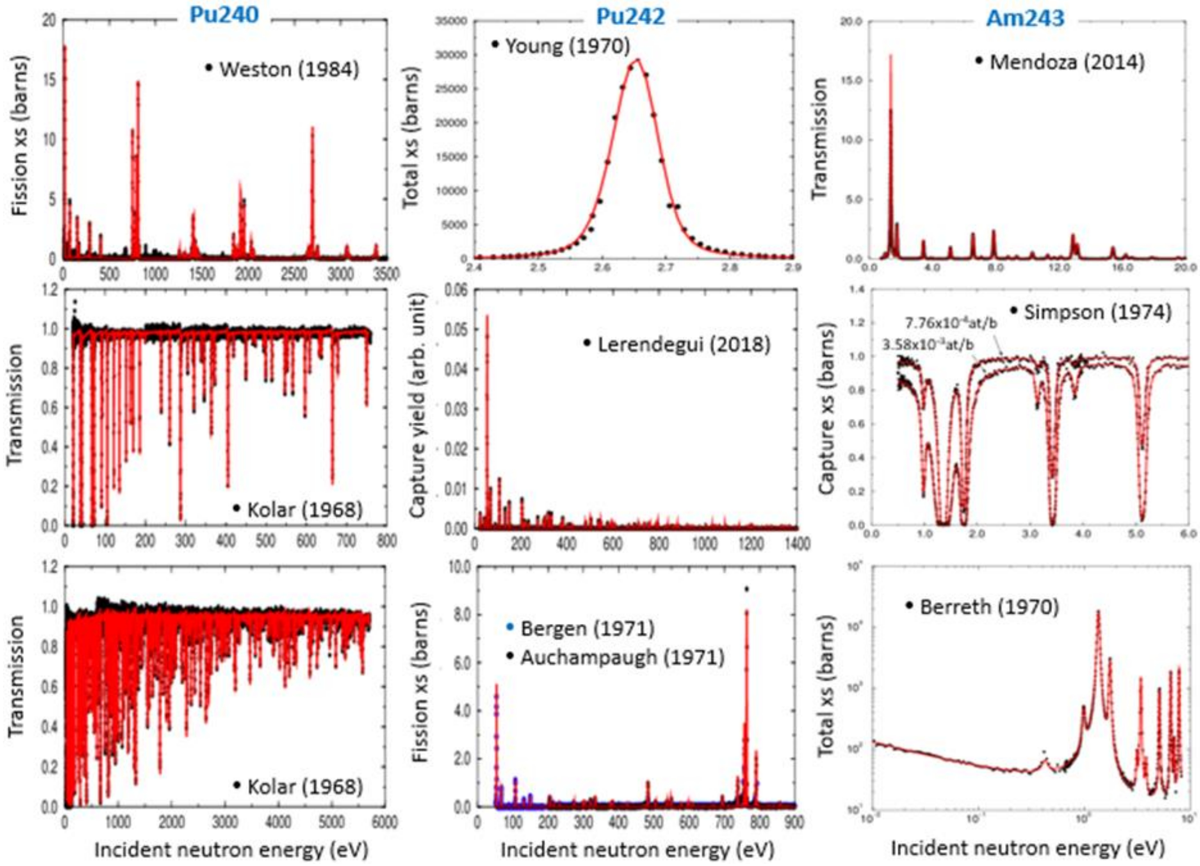

Figure 7. CONRAD results for ^{240}Pu, ^{242}Pu and ^{243}Am.

The present evaluation work confirms the systematically underestimation of the capture resonance integral of ^{240}Pu, ^{242}Pu and ^{243}Am in the JEFF-3.1.1 library. The combination of these new evaluations in Burn Up calculations increases by about +6% the production of ^{244}Cm. The underestimation of the ^{244}Cm build up at around 40 GWd/t nearly vanishes. Integral trends in term of (C/E-1) values become closer to -1.5% in average, with a dispersion of the results of about 3%.

2. High energy evaluation with TALYS

A New high-energy evaluation has been obtained for ^{239}Pu with several improvements. As usual, the starting point is the optical potential which relies, in the particular case of deformed nuclei, on coupled channels calculations. While the standard model implemented in TALYS is ECIS, the current work has been performed with a new code, called PESSA'H, which has been designed to offer the possibility to improve new coupling schemes that could not be included in ECIS, namely the so-called interband coupling "à la Soukhovitski" [Sou16]. The Ground state band has been coupled to the excited single-particle bandhead at 469.8 keV (K^{π} = ½+) giving a 9 states coupling scheme as illustrated in Fig. 8. This new optical model has then been adjusted to reproduce at best total elastic and inelastic cross sections as well as various angular distributions up to a few MeV. The adjustement of a new optical potential stems from the new width fluctuation model implemented in TALYS, namely the Engelbrecht-Weidenmuller (EW) transformation [kaw16] which modifies the way the flux is distributed between elastic and inelastic channels.

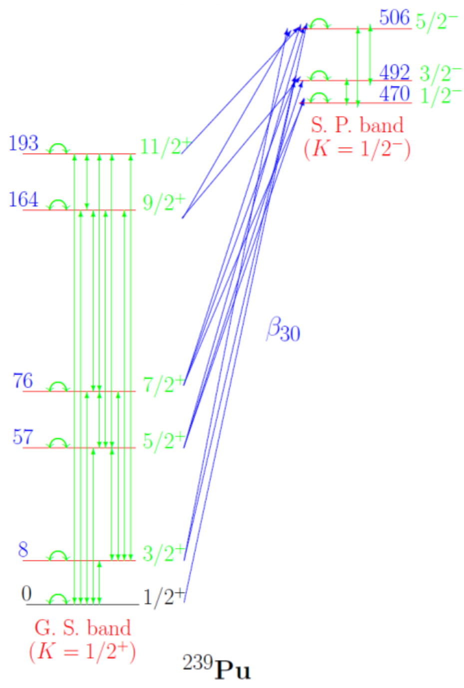

Figure 8. Coupling scheme for the ^{239}Pu optical potential.

Another modification compared to previous evaluation is the use of QRPA-based gamma strength functions [gor19] instead of the Kopecky-Uhl model traditionally employed up to now. This choice enables to reduce the need for a strength function renormalisation. As can be observed in Fig.9, the EW transformation changes the shape of the capture cross section improving the agreement with experimental data in particular between a few tens of keV up to the MeV region.

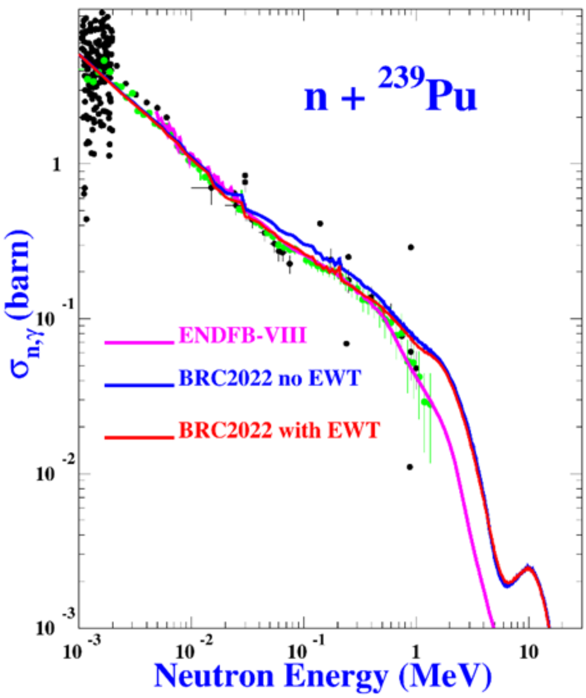

Figure 9. Capture cross section with or without the EW transformation.

The overall agreement with experimental data is illustrated in Fig.10. One can observe a rather good description of total and fission cross sections as well as (n,n'), (n,2n) and (n,3n) experimental data.

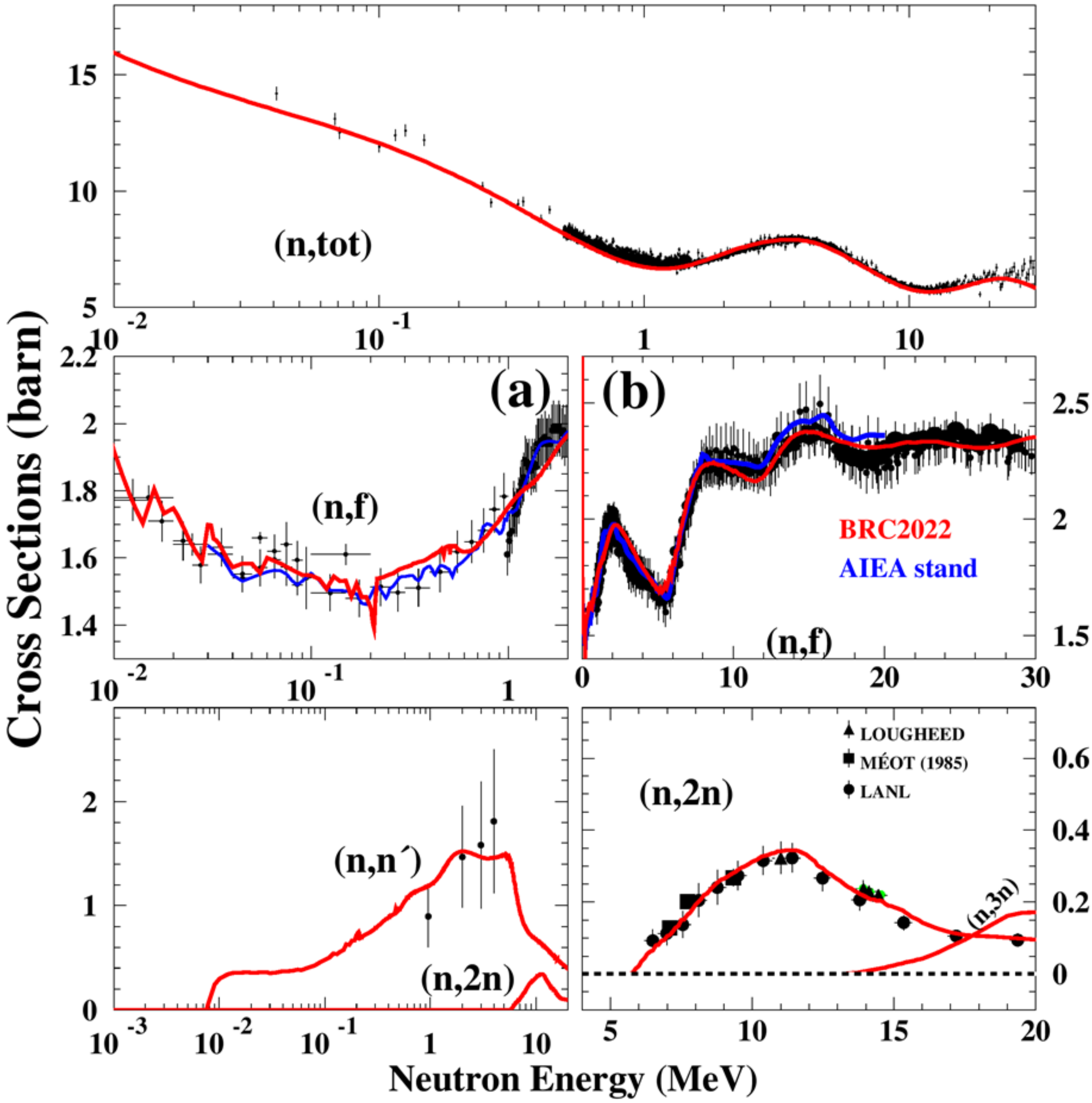

Figure 10. Cross section comparison with experiment for various induced neutron reactions on ^{239}Pu.

Differences can be observed with the fission IAEA standard, which remain globally within less than 2% as illustrated in Fig.11.

A new prompt neutron multiplicity evaluation has also been performed using Bayesian inference approach [reg22]. Compared to previous evaluations of the prompt neutron multiplicity, this work takes into accout the new experimental data obtained from the Los Alamos Chi-Nu experiment [Mar22]. In addition, the Bayesian inference was performed without assumption on the posterior distribution and based on a Markov Chain Monte Carlo method. Efforts were carried on to (I) take into account error models in the inference (ii) validate the inference using a cross reference bootstrap method. The results are illustrated in Fig.12. The outcoming evaluation turned out to be significantly different from JEFF3.3 and a normalization of 0.3% has been required on these new prompt neutron multiplicities to reproduce some of the benchmarks which have been considered to perform a first validation of the new ^{239}Pu evaluation.

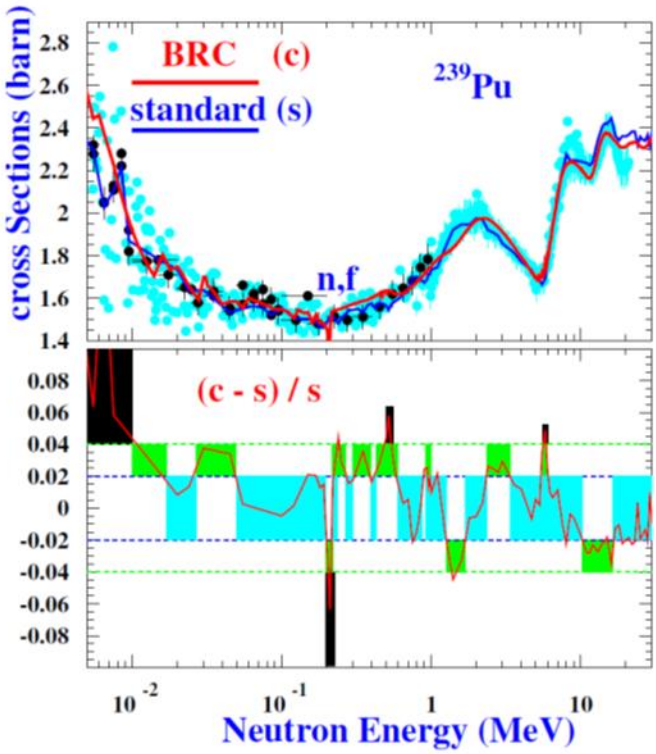

Figure 11. Comparison with the IAEA standard for the fission cross section of the present evaluation.

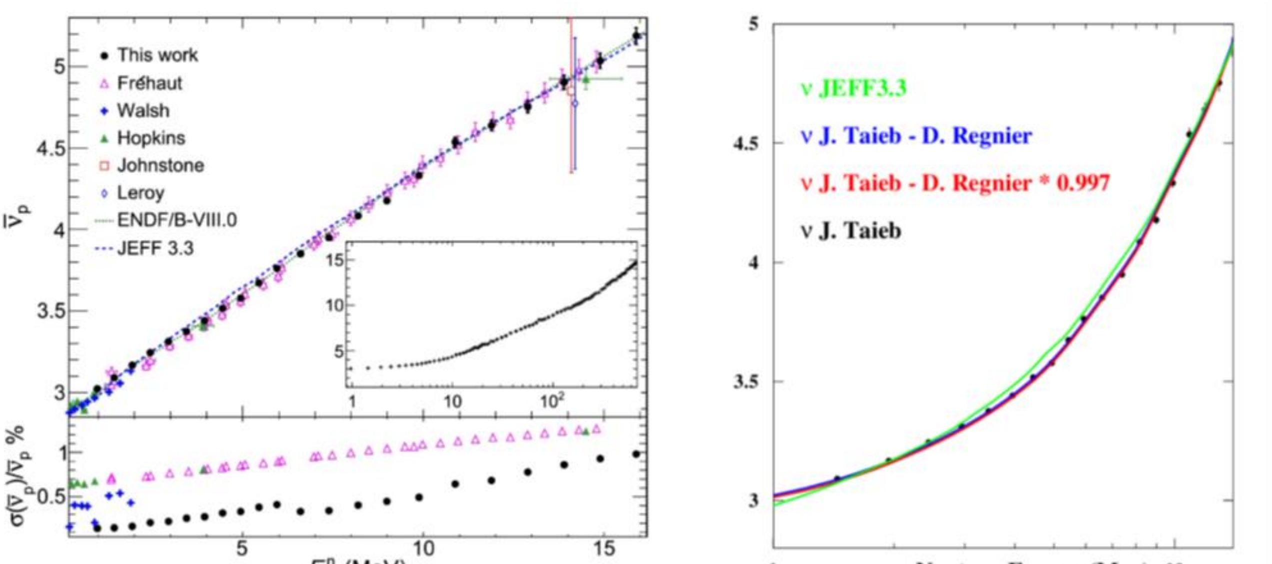

Figure 12. Left: prompt neutron multiplicities of various experiments compared to the ENDF/B-VIII.0 and JEFF-3.3 evaluations for the neutron induced fission on ^{239}Pu (figure taken from [mar22]. The lower panel shows the relative uncertainty of the measurements (without including the uncertainty coming from the ^{252}Cf standard). The P. Marini et al. experimental data show a significant reduced uncertainty. Right: prompt neutron multiplicity in the incident energy range 1 to 20 MeV. We compare here JEFF-3.3 evaluation with the raw result of the Bayesian inference (blue curve) and the renormalized results (red curve).

The latter are illustrated in Fig.13 and one can see that the new evaluation produces results of comparable quality to those obtained using either ENDFB-8.0 or JEFF3.3.

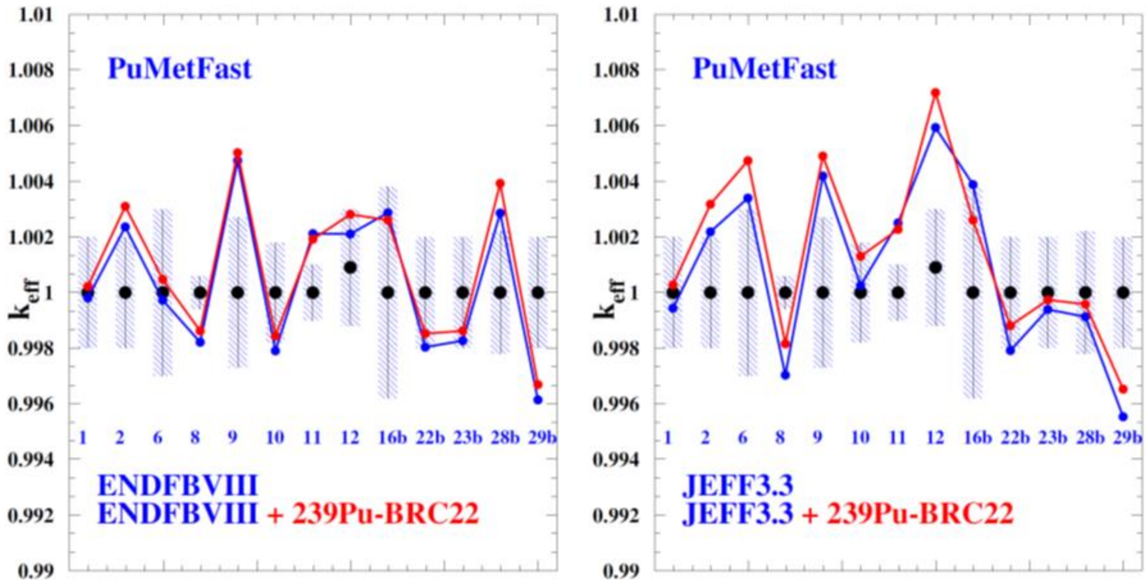

Figure 13. Comparison with various ICSBEP benchmarks replacing the ENDF/B-VIII.0 or JEFF-3.3 ^{239}Pu by our new evaluated file.

3. Reference

[dsj21] C. De Saint Jean et al., EPJ Nucl. Sci. Technol. 7, 10 (2021).
[tam21] P. Tamagno, Eur. Phys. J. A 56, 61 (2021).
[bec12] B. Becker et al., J. Instrum. 7, P11002 (2012).
[nog18] G. Noguere et al., Eur. Phys. Plus 133, 177 (2018).
[lit13] O. Litaize et al., EPJ Web of Conf. 42, 02003 (2013).
[otu14] N. Otuka et al., Nucl. Data Sheets 120, 272 (2014).
[bou97] O. Bouland et al., Nucl. Sci. Eng. 127, 105 (1997).
[sou16] E. Sh. Soukhovitski et al., Phys. Rev. C 94, 064605 (2016).
[kaw16] T. Kawano et al., Phys. Rev. C 94, 014602 (2016).
[gor19] S. Goriely et al., Eur. Phys. J. A 55, 172 (2019).
[reg22] D. Regnier, JEFF doc 2122 (2022).
[mar22] P. Marini, Phys. Lett. B 835, 137513 (2022).