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

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Abstract. The CONRAD computer code is being developed by the nuclear data group of CEA Cadarache since mid-2000. It was originally designed to analyse neutron-induced reactions in the resonance energy range and then, was extended to higher energies (several MeV) treatment with inclusion of charged-particles penetration factor. In the thermal energy range, a procedure was implemented to manage the so-called Thermal Neutron Constants, especially devoted to the 239Pu, 241Pu, 233U and 235U nuclei. In the resonance range, nuclear models implemented in the CONRAD program rely on R-matrix model fits with in particular, improved treatment of the fission penetration factor and fluctuations of the prompt neutron multiplicity via a two-step (n,γf) process. Above the resonance range, the neutron continuum energy region of the observed cross sections can be analysed either with the TALYS code or with an in-house optical model code named CCCP that is followed by Hauser-Feshbach calculations according to the compound nucleus deexcitation channels. Non-model least squares fitting procedures have been also tested for neutron cross sections adjustment in the continuum energy range, and such, applied in the framework of the standard cross section group at the IAEA (Vienna). This paper will only focus on the evaluation works performed in the resonance range of minor and major actinides with special emphasis on the experimental corrections suitable to reproduce time-of-flight experiments.

1 Introduction

The CONRAD code is an object-oriented software tool developed at CEA since 2005 [1]. 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 235U, 238U, 239Pu, 241Am, 240Pu and 242Pu are used to illustrate the performances of the code.

An overview of the cross section models is presented in section 2. Section 3 shows effects of the Doppler broadening, response function of the facility and multiple scattering correction. Results obtained on minor actinides is detailed in section 4.

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

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 [3].

One of the latest models 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. 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.

3 Experimental corrections

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.

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.

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 LANSE 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 last correction discussed within the framework of this article is the correction due to the multiple neutron scatterings in the sample [5]. 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. 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.
4 Results on minor actinides

In this section, the performances of the CONRAD code are illustrated with evaluation works performed on minor actinides of interest for spent nuclear fuel applications. The longstanding underestimation of the $^{244}$Cm 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 data base [6]. Figures 6 and 7 compares the theoretical curves with the selected data sets.

For the first resonance of $^{240}$Pu, close to 1.06 eV, we have 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 (Fig. 6). 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. [7]. The results leads to a thermal capture cross section $\sigma_0=285.6$ 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 Leren-Degui (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 $\sigma_0=18.8$ barns and an increase of $+3.1\%$.
of the capture resonance integral compared to JEFF-3.1.1 ($I_0 = 1165$ barns).

The evaluation of the resonance parameters of $^{243}$Am were performed by using the capture yield reported by Mendoza ($L_1 = 185$ 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 $\sigma_0 = 75.5$ barns. We also obtained an increase of +6.3% of the capture resonance integral compared to JEFF-3.1.1 ($I_0 = 1902$ barns).

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%.

### 5 Conclusions

Evaluations works on actinides performed with the CONRAD code demonstrate that the code is mature to handle a large variety of time-of-flight data measured in various facilities. It allows handling the main experimental corrections needed to analyse these type of data, such as Doppler effect, response function of the facility and multiple scattering correction. Examples focused on $^{240}$Pu, $^{242}$Pu and $^{243}$Am highlight the needed revision of the minor actinides in the international libraries in view of predicting composition of spent nuclear fuel.

The latest theoretical developments in the CONRAD code, connected with the fission process, evidence the limits of the evaluated nuclear data format which does not allow to introduce theoretical ingredients such as the two step (n,$\gamma$) reaction. Consequently, calculations on the fly of fluctuations in the prompt neutron multiplicity as a function of temperature is still impossible. The modernization of the processing systems is a priority to take into account future theoretical development in the resonance range of the neutron cross sections.

### References