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Re-examining intermediate resonant structures in the ^{242}Pu fission cross section

Carole Chatel^{1,2,4*}, Olivier Bouland², Gilles Noguère², Maria Diakaki³, and Ludovic Mathieu⁴

¹Institut Pluridisciplinaire Huber Curien, 23 rue du Loess, 67200 Strasbourg, France

²CEA, DES/IRESNE/DER/SPRC/LEPh, 13008 Saint Paul Lez Durance, France

³National Technical University of Athens, Department of Physics, Zografou Campus, Athens, Greece

⁴Laboratoire de Physique des 2 infinis, 33170 Gradignan, France

Abstract. The study of the ^{242}Pu fission cross section has become more important since this isotope is present in Pressurized Water Reactor Uranium Oxide and Mixed Oxide fuel, as well as in fast neutron reactors fuel. This nucleus has been also selected for monitoring the high neutron flux RJH reactor (CEA-Cadarache). For all these reasons, the fission cross section structure of this fertile isotope, and especially its class-II states resonances were investigated in this work. Two class-II states have been identified in the Resolved Resonance Range (RRR) and their parameters have been supplied to the CONRAD code. 68 class-II states have been identified in the Unresolved Resonance Range (URR) and their parameters have been added in the TALYS code data library to reproduce as well as possible the experimental fission cross section. The methodology to identify and insert the class-II states in RRR and URR is described here.

1 Introduction

Produced in the Pressurized Water Reactor (PWR) Uranium Oxide (UOx) fuel by successive neutron captures on ^{238}U , the ^{242}Pu is present in nuclear fuel. Indeed, as no plutonium isotopic separation is made, the ^{242}Pu is present from the beginning in the Mixed Oxide fuel (MOx) type and is expected to be present in the fuel of some of the generation IV fast neutron reactors. Hence, for monitoring reactor cores as well as for more accurate calculations of the rate of the different isotopes in radioactive waste, fission cross sections are important to evaluate. Moreover, the high neutron flux Jules Horowitz Reactor (RJH) built in CEA-Cadarache, France, includes a flux monitor system for high neutron energy (above 1 MeV) which is based on a ^{242}Pu fission chamber. This isotope has been selected for its fertile character. For all these reasons, the knowledge of the ^{242}Pu fission cross section has become of higher importance. This implies a deeper investigation of the ^{242}Pu compound nucleus.

We decided to study the states lying in the second fission well (see Section 2), the so-called class-II states. Indeed, only few studies [1,2] have already been performed and no resonance parameters are available for a reconstruction using Hauser-Feshbach codes like TALYS.

Therefore, we will revisit the present methodology, add class-II states first in the Resolved Resonance Range (RRR) and finally in the Unresolved Resonance Range (URR).

2 Class-II states characteristics

A way of schematizing the compound nucleus potential well along fission is to draw it as a function of its elongation. Following Strutinsky [3], the potential well follows a “fluctuating” path as displayed in Fig. 1.

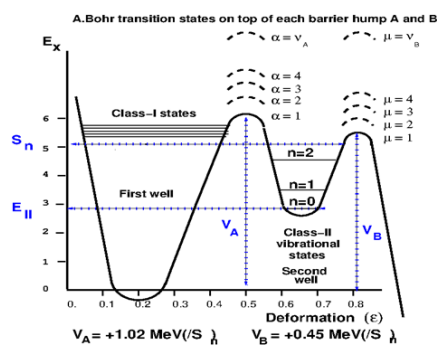


Fig. 1. Potential well of a heavy compound nucleus as a function of its elongation during the fission process. From [4].

The first well, at an elongation of about 0.2, is the main well. The states lying inside are, in fission terminology, called the class-I states and are essentially responsible for the resonances observed in the Resolved Resonance Range (RRR), region limited for the ^{242}Pu at a neutron energy of 1.15 keV. As the spacing between them decreases whereas their width increases with energy, only a gathering of these states - or the largest - can be observed in the Unresolved Resonance Range (URR, for ^{242}Pu from 1.15 keV to 40 keV).

* Corresponding author: carole.chatel@iphc.cnrs.fr

The saddles, typically lying at a deformation of 0.5 and 0.8 respectively (see Fig. 1), are commonly described as fission barriers. They assign the fertile or fissile character of an isotope by comparison to the neutron separation energy (S_n). Following A. Bohr [5] postulate, fission barriers are described in terms of transition states. Fig. 2 shows the fertile character of the ^{242}Pu compared to the fissile ^{239}Pu isotope.

The states lying inside the second well (0.6-0.7 of elongation) are the so-called class-II states. These states are well-observed in fission cross sections. The spacing between class-II states is about 50 times larger than the one between class-I states. The class-II states are more easily recognized in the URR than in the RRR.

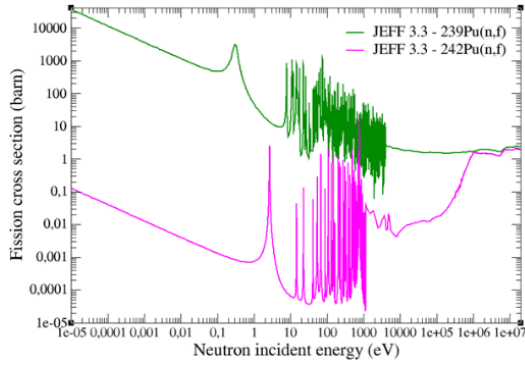


Fig. 2. ^{242}Pu fission cross section (fertile isotope, pink lower curve) compared with ^{239}Pu fission cross section (fissile isotope, green upper curve) as a function of the neutron energy. Data from the JEFF3.3 evaluation [6]

3 Class-II states in the RRR

3.1 Identification and parametrization

To identify the class-II states, one must first select the data that will be used as reference. However, in EXFOR, no data set covering the entire RRR is available. The only data available are those of Bergen *et al.* [7], starting at 50 eV, and those of Auchampaugh *et al.* [8] starting at 200 eV. For this reason, our reference will be the JEFF3.3 evaluation [6], based on the Weigmann *et al.* data [1] which are missing in EXFOR below 300 keV.

Assignment of class-II states in the RRR rely on evaluated data. Commonly, resonances are described by their widths, including the fission width, but not by their class. The fission width of a resonance Γ_f follows the R-matrix [9] definition Eq. (1):

$$\Gamma_f = 2\gamma_f^2 \quad (1)$$

with γ_f the reduced fission width amplitude, phenomenologically fitted. This definition does not include a fission penetrability and then remains phenomenological. The local amplification of the penetrability due to a nearby class-II state will thus be characterized by an artificial amplification of the fission width. When drawing the cumulative curve of fission widths as a function of the energy, the presence of class-II states manifests by steps as can be seen in Fig. 3.

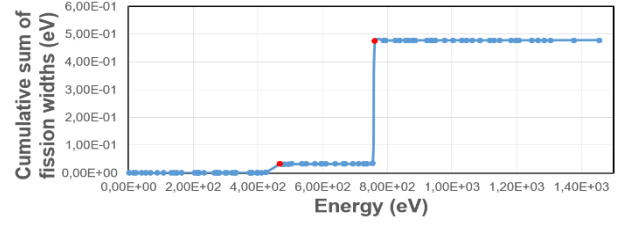


Fig. 3. Cumulative of the ^{243}Pu fission widths. The two red dots are observed class-II states.

One can observe in Fig. 3 the presence of two step structures manifesting the presence of two class-II states in the RRR, at the energies of 468 eV and 762 eV (marked with the red dots).

The present work aims to include the actual fission penetrability for those two class-II states as described in Eq. (2):

$$\Gamma_f = 2P_f \gamma_f^2 \quad (2)$$

The local amplification of the penetrability created by the presence of a class-II states, P_{CII} , can be described by a Lorentzian shape [10] Eq. (3):

$$P_{CII} = \frac{\sigma_{CII}}{4} \frac{\Gamma_{CII}}{(E - E_{CII})^2 + \frac{\Gamma_{CII}^2}{4}} \quad (3)$$

with E_{CII} being the class-II state observed energy, Γ_{CII} , the class-II state width and σ_{CII} the class-II state magnitude. To derive those parameters, the steps observed in Fig. 3 can be fitted, thanks to Eq. (4):

$$S_{CII} = \alpha + \sigma_{CII} * \text{Arctan} \left(\frac{E - E_{CII}}{\Gamma_{CII}} \right) \quad (4)$$

where α is a constant parameter used to adjust the curve's magnitude. The final parameters for both class-II states are listed in Table 1.

Table 1. Class-II states parameters identified in the RRR.

Energy (eV)	447	759
Γ_{CII} (eV)	3	0.04
σ_{CII}	0.0112	0.12854

3.2 Evaluation in the CONRAD code

To fit the ^{242}Pu fission cross section in the RRR, the code used is the CONRAD code [11], developed at CEA of Cadarache. This code is based on the R-Matrix formalism [9].

The JEFF3.3 evaluated file for ^{242}Pu relies on the Multi-Level-Breit-Wigner approximation. Since this approximation is obsolete for fertile nuclei, using a more powerful description such as the Reich-Moore formalism is interesting. The improvement lies in the inclusion of interferences between channels. Switching directly from one to the other approximation does not allow for a proper reproduction of the cross section. The readjustment of the fission parameters is described in [12]. The final result is presented in Fig. 4 (green curve). Unfortunately, a proper fit of the fission cross section in the valleys between resonances is not possible since the

residual background is larger than the cross section, thus the experimental information is not reliable.

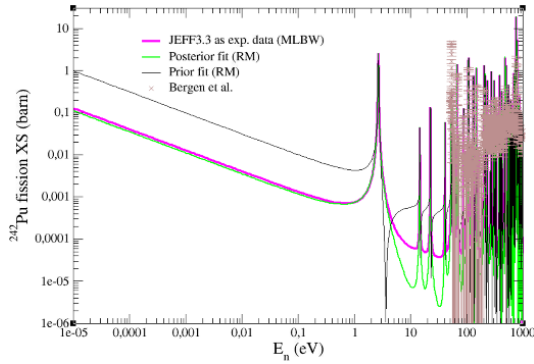


Fig. 4. ^{242}Pu fission cross section in the RRR prior (black curve) and posterior (green curve) to the resonance parameters fit to the Reich-Moore approximation. Reference: JEFF3.3 evaluation [6] (pink thick curve).

The parameters obtained with Eq. (4) for the two class-II states are supplied to CONRAD to include the meaningful description of the local enhancement of the fission cross section. Following a second parameters adjustment, the final result obtained is shown in Fig. 5 (green curve) with a zoom over the 762 eV class-II state. One can see that the class-I resonances are properly fitted. Between the resonances, the final fit (green curve) shows a slightly smoother description of the cross section than the prior one.

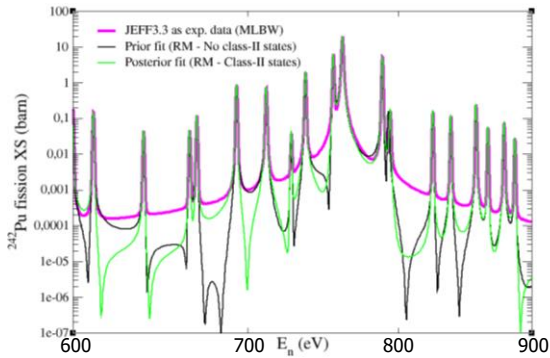


Fig. 5. ^{242}Pu fission cross section in the RRR zoomed over the 762 eV class-II state prior (black curve) and posterior (green curve) to the inclusion of the class-II states parameters with the Reich-Moore approximation. Reference: JEFF3.3 evaluation [6] (pink thick curve).

One should note that a similar result to the one shown in Fig. 5 have been obtained by trial and error parameters giving fission widths of 50 and 21 eV.

4 Class-II states in the URR

A similar work was performed in the URR. This region for the ^{242}Pu covers the range [1.15 keV-40 keV]. This study was able to extend the URR up to 100 keV.

4.1 Identification

As class-I states become unresolved in the URR, no fission width is released in the evaluated files. It is thus not possible to use them as prior to identify class-II

states. Nonetheless, a similar work than the one in the RRR can be done by direct input of the fission cross section. Indeed, by making a cumulative sum of the fission cross section, one can identify step structures, fit their features with Eq. (4) and finally supply them to an appropriate input file.

However, with this methodology, class-II states cannot be discriminated from large or gathered class-I states. Although significant improvement of the URR treatment was made in present work, additional work is needed for an exact identification of each observed resonance.

JEFF3.3 being an average description of the fission cross section, no help can be expected from this reference evaluation for an “intermediate structure” resonances description, as shown with the pink curve from Fig. 6. Differential measurements are, on the contrary, of valuable support. Two data sets are available in EXFOR covering the entire URR. As the data from Auchampaugh *et al.* [8] show the best energy resolution, this data-set will be used in this work. They are presented with their uncertainties in Fig. 6.

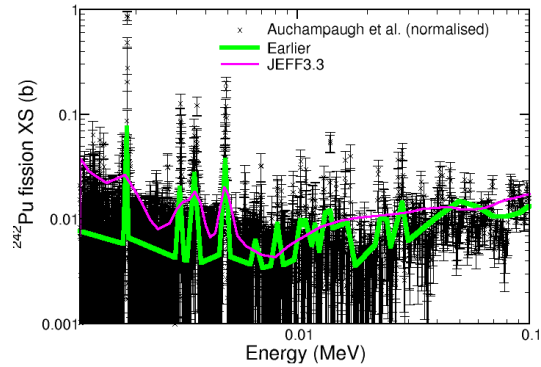


Fig.6. ^{242}Pu fission cross section in the URR. Data from Auchampaugh *et al.* [8] with uncertainties (black crosses); JEFF3.3 evaluation [6] (pink upper curve) and earlier evaluation work done by G. Noguere with TALYS [13] (thick green lower curve).

The cumulative sum of the fission cross section is therefore made. The actual steps are identified and fitted with respect to Eq. (4); each step representing a class-II state or in some cases, a broad class-I state. The results are shown in Fig. 7. Between 1.15 keV and 100 keV, 68 steps have been identified.

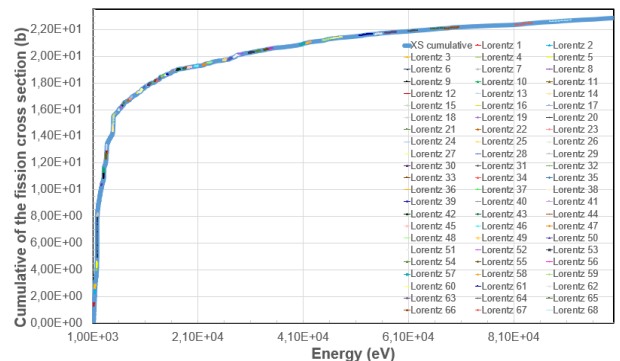


Fig. 7. Cumulative sum of the ^{242}Pu fission cross section in the URR (light blue continuous curve). Colored curves correspond to fitted steps.

4.2 Intermediate structure resonances over the URR

Once class-II states parameters corresponding to the intermediate structure of the second well have been identified, the URR fluctuations of the fission cross section were reasonably described. In the URR, our reference code is the TALYS1.4 [10] version modified by G. Noguère. This version relies on a Lorentzian penetrability formalism as described by Eq. (3) to treat class-II states. The reproduction of the ^{242}Pu fission cross section in the URR by G. Noguère is presented in Fig. 6 (green curve). The latter work was used as prior to this work. The resonances and the magnitude of the curve are obtained by fictive and very broad class-II states parameters added in a class-II states input file.

The present parameters (as shown in Fig. 7) derived from (4) are then added in the TALYS class-II states input file. Part of Noguère's fictitious resonance parameters above 40 eV were preserved to maintain the magnitude of the fission cross section since not all class-II states of high energy could be resolved. The final reproduction of the fission cross section in the URR is presented in Fig. 8 (blue thin curve).

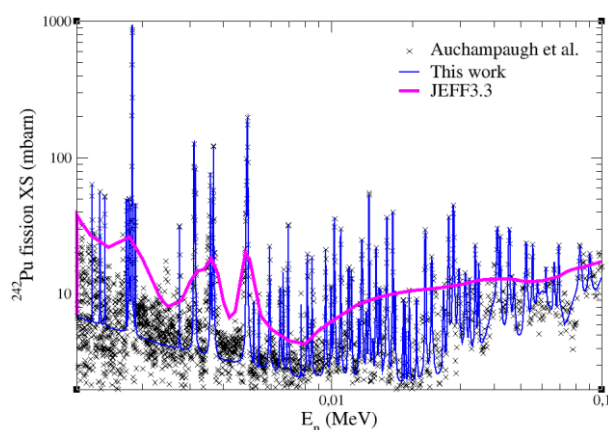


Fig. 8. ^{242}Pu fission cross section in the URR. Data from Auchampaugh *et al.* [6] (black crosses); JEFF3.3 evaluation (pink thick curve) and most complete present work (blue thin curve).

5 Conclusion

Class-II states lying in the second fission well bring the intermediate structure observed in the fission cross section of fertile actinides. To-date, they are rarely described for the theoretical reproduction of a fission cross section. During this work, class-II states of the compound ^{243}Pu nucleus have been characterized in the RRR and URR. Parameters have been extracted assuming Lorentzian penetrability for addition in evaluated data files.

Since no available data covers the whole RRR, the JEFF3.3 evaluation has been used as reference. Class-II states were taken into account by local amplification of fission widths. Hence, a cumulative sum of the fission widths allows for a better identification of class-II states manifested by step structures. Fitting these steps by a Lorentzian provides parameters needed for an explicit description of the class-II states in evaluation codes. The

JEFF evaluation of ^{242}Pu is based on a Multi-Level-Breit-Wigner (MLBW) approximation, that is obsolete. Conversion of the current MLBW parameters to the Reich-Moore formalism has been performed. However, one can note that in addition to the lack of available data in this energy range, the large uncertainties between resonances do not allow for a proper fitting of the channel interferences between resonances. The new class-II state parameters were added in the CONRAD code to reproduce the intermediate structure fission cross section over the RRR.

In the URR, as no fission widths are available, a cumulative sum of the fission cross section has been performed to identify class-II states leading to observable step structures. The present method does not allow for a discrimination between resolved class-II states and gathering of class-I states nor large resolved class-I states. To perform this study, the data set of Auchampaugh *et al.* has been used. Over the URR extended to 100 keV, 68 steps have been fitted and resonances parameters added to the TALYS input file to reproduce the fluctuating cross section in the URR. Additional studies or new experimental data are now needed to determine the actual origin of the identified resonances.

This work is the first input in an evaluated file (JEFF3.3) of class-II states information. A special evaluated data files format is now required to manage these new set of parameters.

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