BNL-94240-2010

Fission cross section calculations of actinides with EMPIRE code

M. SIN*, P. OBLOZINSKY[1], M. HERMAN[1], and R. CAPOTE[2]

University of Bucharest, Bucharest, Romania
[1]National Nuclear Data Center, Brookhaven National Laboratory, Upton, NY, USA
[2]International Atomic Energy Agency, Nuclear Data Section, Wagramer Strasse 5, A-1400 Vienna, Austria
[*]Corresponding author. E-mail : mihaela.sin@g.unibuc.ro

Presented at the
International Conference on Nuclear Data for Science and Technology

April 2010

National Nuclear Data Center
Brookhaven National Laboratory
P.O. Box 5000
Upton, NY 11973-5000
www.nndc.bnl.gov

DISCLAIMER

Fission cross section calculations of actinides with EMPIRE code

M. SIN*, P. OBLOŽINSKÝ[1], M. HERMAN[1], and R. CAPOTE[2]
University of Bucharest, Bucharest, Romania
[1]National Nuclear Data Center, Brookhaven National Laboratory, Upton, NY, USA
[2]International Atomic Energy Agency, Nuclear Data Section, Wagramer Strasse 5, A-1400 Vienna, Austria
[*]Corresponding author. E-mail : mihaela.sin@g.unibuc.ro

Received
Accepted for Publication

The cross sections of the neutron induced reactions on ^{233,234,236}U, ^{237}Np, $^{238-242}$Pu, ^{241,243}Am, $^{242-246}$Cm carried out in the energy range 1 keV - 20 MeV with EMPIRE code are presented, emphasizing the fission channel. Beside a consistent, accurate set of evaluations, the paper contains arguments supporting the choice of the reaction models and input parameters. A special attention is paid to the fission parameters and their uncertainties.

KEYWORDS : ND2010, Nuclear Data, ENDF

1. INTRODUCTION

At ND2007 several new features of EMPIRE code [1] have been presented. Among them, we mention only the capabilities of treating the actinide nuclei and of producing sensitivity matrices to be used in covariance calculations [2-3]. Since then, a recursive method to calculate the fission coefficient for multi-humped barrier was developed [4] and a new version of the Reference Input Parameter Library (RIPL-3) became available [5]. In the present work we made use of these accomplishments to find the right selection of reaction models and physically grounded input parameters for the evaluation of the nuclear data for actinide nuclei. The models and the parameters are presented in Section 2. The results of the neutron cross sections of 16 actinides in the energy range 1 KeV - 20 MeV calculated with EMPIRE code are presented in Section 3 with emphasize on the fission channel. In the same section are given details on the fission parameters and their uncertainties.

2. MODELS AND PARAMETERS

2.1 Optical Model

The latest extensions of the dispersive coupled-channel optical model (DCCOM) was employed to describe the nucleon interaction on deformed targets. The calculations of the direct cross sections and neutron transmission coefficients calculations have been performed with RIPL-2408 [5] optical model potential. It is isospin dependent, contains a dispersive term including non-local contribution and covers 1 KeV – 200 MeV energy range and 31 actinide nuclei [3,5].

2.2 Compound Nucleus

Hauser-Feshbach and Hofmann–Richert–Tepel-Weidenmüller versions of the statistical model considering multiple-particle emission and full gamma-cascade are employed to treat the compound nucleus mechanism [1].

2.3 Direct and Preequilibrium Emission

Different combinations of direct excitation of the collective levels in the continuum by DWBA method, pre-equilibrium emission considered within one-component exciton model (PCROSS), which includes nucleon, gamma and cluster emission [1,2], deformation dependent Tamura Udagawa Lenske multistep direct [7] and multistep compound model (MSD+MSC) were tested.

2.4 Fission

The fission mechanisms associated to the different degrees of damping of the vibrational states accommodated by the minima of the fission path are accounted for by a formalism developed within the optical model for fission which describes transmission and absorption through multi-humped barriers using a recursive method [6]. It can provide an accurate description of the experimental fission cross sections, including the resonant structure at low energies. This fission formalism implemented in EMPIRE proved to meet the requirements of an accurate fission cross section evaluation, therefore in this work we focused mainly on the fission parameters. These parameters describe: (i) the fundamental fission barrier, (ii) the barriers associated to the discrete transition states and (iii) the barriers associated to the transition states in continuum.

RIPL contains information about these parameters deduced on phenomenological and microscopic bases. For the fundamental fission barrier there are: (a) empiric heights and widths describing the humps of the fission barrier parameterized as two decoupled parabolas and (b) microscopic Hartree-Fock-Bolgolyubov (HFB) numerical description of the deformation potential along the fission path. For the discrete transition states there are recommendations regarding the excitation energies, parity and spin projection along the symmetry axes for each saddle point depending on the odd-even type of the nucleus. According to RIPL, the transition state densities can be either calculated within GSM formulation for which there are recommendations regarding the main parameters and collective enhancement factors specific to the nuclear shape asymmetry at each saddle point, or one can use the numerical global prescription obtained on HFB microscopic bases [5,8]. Details about the fission parameters used in the present calculations are given in the next section.

3. RESULTS AND DISCUSSIONS

An analysis of the experimental data for the actinide nuclei in EXFOR reveals that for most of them, only the fission cross section is available in the fast neutron range. Performing a theoretical prediction without having the constraints imposed by the experimental data for different reaction channels, especially for the total cross section, makes it difficult to obtain reasonably accurate results. What would be needed in such cases are models and parameterizations with sound physical grounds, tested and validated globally or regionally, one can rely on. For the present calculations this role was played by the optical model potential and by the Enhanced Generalized Super-fluid Model (EGSM) used to describe the continuous excitation spectra of the nuclei at equilibrium deformation. For the rest of the parameters we took as starting values RIPL-3 recommendations which in most of the cases coincide with EMPIRE's defaults [1,5].

In Fig. 1 are presented some examples of good agreement between calculated and experimental total cross sections (taken from EXFOR as all the experimental data in all the plots included in this paper). This agreement increases the confidence in the RIPL-2408 [5] optical model potential's prediction accuracy, important especially where experimental data are scarce or lack completely. It must be mentioned that in almost all cases the total and elastic cross sections predicted by EMPIRE are supported by recent evaluations (ENDF-VII.0, JENDL/AC and ROSFOND).

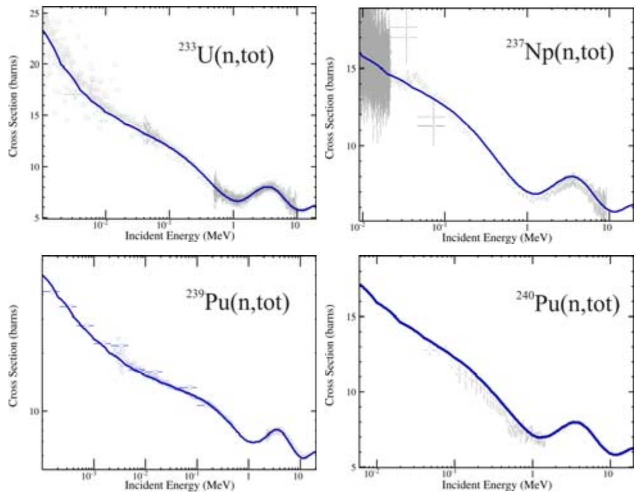

Fig. 1. Total reaction cross sections for ^{233}U, ^{237}Np, ^{239,240}Pu

Different ways of describing direct and preequilibrium emission have been tested. In the end, two approaches with equivalent impact on the fission cross section were selected: DWBA+PCROSS and MSD+MSC. The results presented in this paper are obtained with the first combination of models, the calculations with MSD+MSC being under way. The differences between them are visible when comparing the tails of inelastic cross section, the (n,2n) cross sections and obviously the energy spectra of the emitted particles. In Fig.2 are presented the cross sections of (n,2n) process for ^{239}Pu and ^{241}Am.

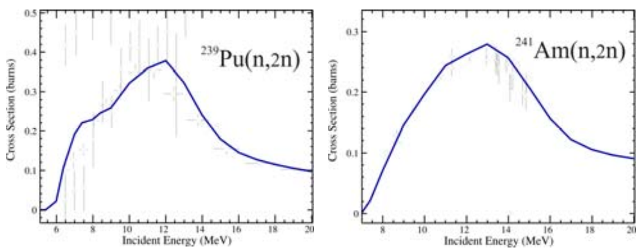

Fig. 2. Cross sections of (n,2n) process for ^{239}Pu and ^{241}Am

An important channel with significant impact on fission at low energies is the radiative capture. Among the closed formulae for the gamma ray strength function developed by Plujko and included in RIPL [5], we selected Modified Lorentzian 1 (MLO1) which was validated in previous actinides' evaluations. By using MLO1 normalized to the experimental data at low energies, a proper choice for the number of discrete excited levels for the involved nuclei and EGSM for the level density functions, a good description of the this channel can be obtained, as can be seen in Fig.3.

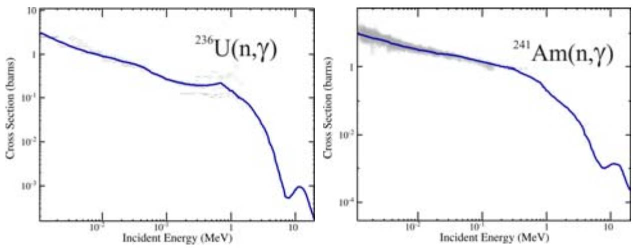

Fig. 3 Radiative capture cross sections for ^{236}U and ^{241}Am

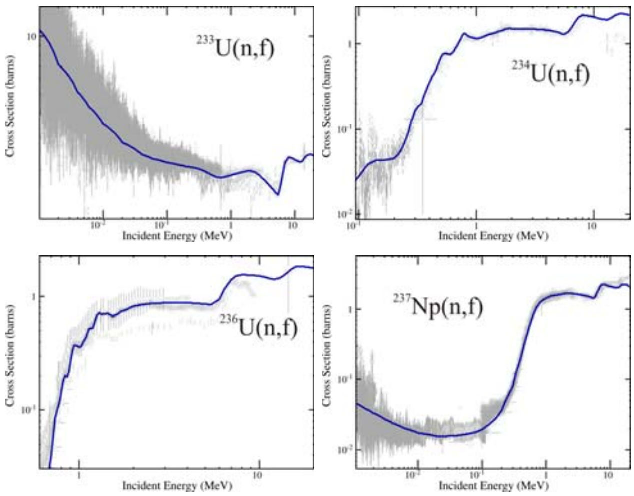

Fig. 4. Fission cross sections for ^{233,234,236}U and ^{237}Np

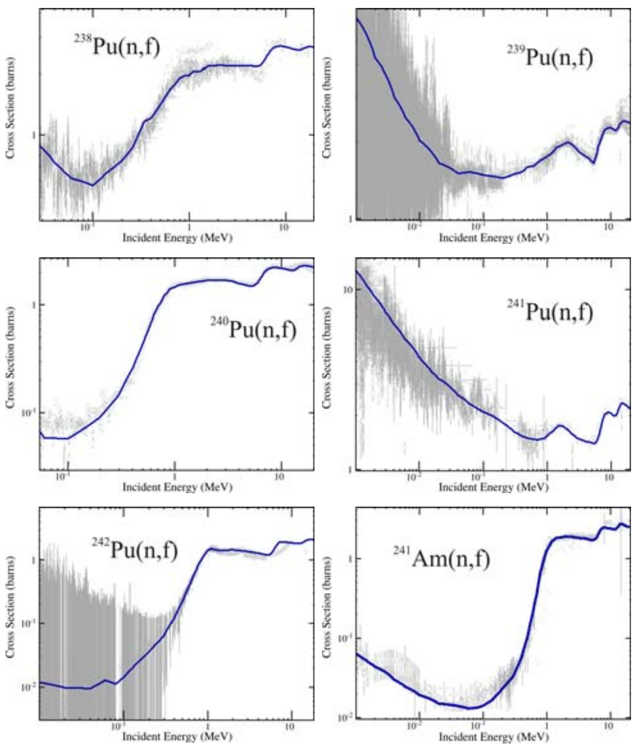

Fig. 5. Fission cross sections for $^{238\text{-}242}$Pu and ^{241}Am

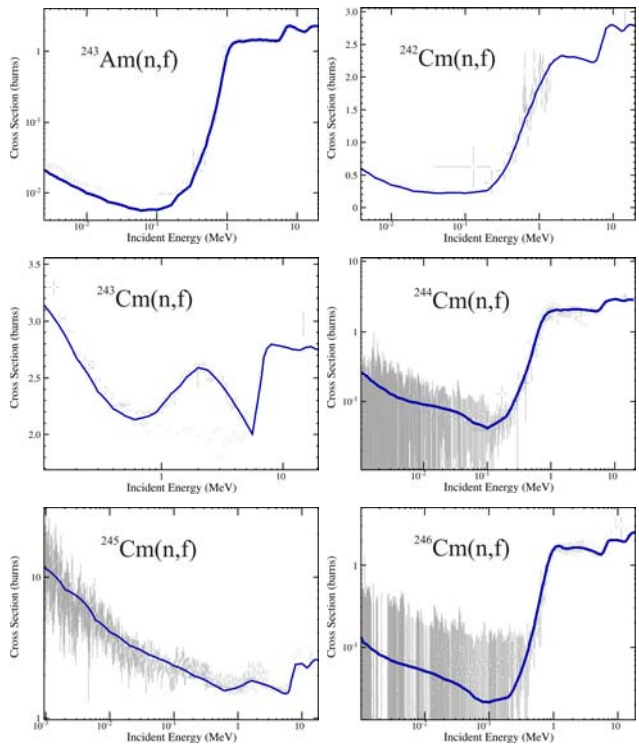

Fig. 6. Fission cross sections for ^{243}Am and $^{242\text{-}246}$Cm

Once a proper treatment for the competing reactions settled, one could focus on fission. In Section 2.4 were mentioned the information included in RIPL which concern the fission parameters. Making use of all this information, we adopted a procedure to establish the fission parameters which takes into account their interrelations and ensures consistency for all reaction channels.

The multi-humped fission barriers have been parameterized by parabolas smoothly joined [1]. For the barriers associated to the discrete transition states (including fundamental) we adopted as starting values the empirical recommendations from RIPL, completed with information regarding the isomeric well suggested by the microscopic calculations and with an imaginary potential required by the optical model for fission. There were cases, such as ^{234,236}U where we departed from RIPL recommendations by using triple-humped barriers to reproduce the structure of the experimental fission cross sections (Fig.4).

The consistent treatment of all reaction channels was ensured by using for the transition state densities the same EGSM formulation used for the normal states. It was adapted to describe the transition state densities by considering the deformation and collective enhancement specific for each saddle point. RIPL recommendations for the main parameters (shell corrections, condensation energy etc.) have been adopted. Some quantities, such as the vibrational enhancement, have been normalized with factors estimated from preliminary comparisons with the experimental data. All these fission parameters represented the default fission input of EMPIRE code.

In the next stage, these starting values were adjusted to fit the experimental data for each nucleus independently. Considering that the above mentioned fission parameters are associated mainly to the structure properties of the nucleus, they should be independent of the way the nucleus was populated. This was tested by performing, when possible, calculations for chains of isotopes (Figs. 4-6). More reliable and consistent constraints can be imposed in such cases, ensuring that for a given nucleus, the same set of parameters describes, within uncertainty limits, the first, second or third chance fission. Additional information on the parameters', and on the reaction models' consistency have been obtained by studying the photo-fission cross section of the nuclei of interest for which experimental data are available. In Fig.7 are presented the photo-fission cross sections for four plutonium isotopes. This cross-checking procedure conduct towards an improved set of fission parameters (which eventually will become the new default input), a validation of the reaction models and parameters and more accurate cross section evaluations. It provides also information about the model parameter uncertainties, important for covariance calculations.

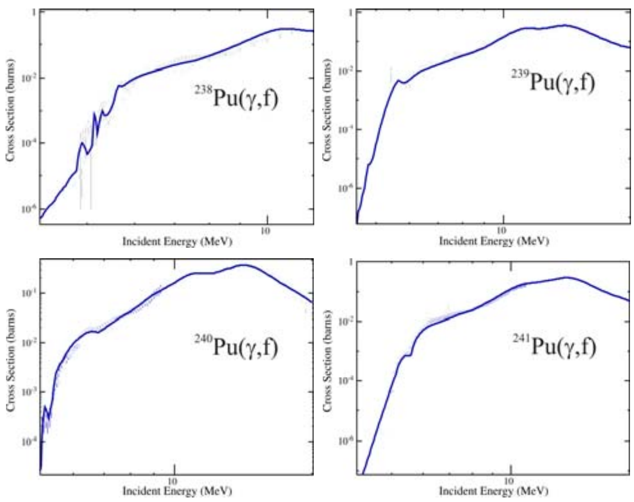

Fig. 7. Photo-fission cross sections for $^{238-241}$Pu

The methodology for generating cross section covariances in the fast neutron range employs a sensitivity matrix produced with EMPIRE and uses it in the KALMAN code for determining covariances while taking into account relevant experimental data [3]. Our sensitivity studies showed that the parameters with the biggest impact on the fission cross section are the heights and widths of the fundamental barriers and four parameters entering EGSM formulae. Assessing the uncertainty for a model parameter is a difficult task, considering that even its definition is not very clear. Therefore, we associated to each of the parameters the cross sections are more sensitive to a dispersion representing the difference of the parameters needed to describe the fission of given nuclei populated in different ways or using different reaction models, and, in the same time, the limits within parameters can vary while the resulting cross sections are still in a reasonable agreement with the experimental data. The dispersion values we considered reasonable are: 5% (approx. 200 KeV) for the barrier heights, 10% (50-100 KeV) for the widths, and from 2% to 10% for the parameters entering the transition state densities. These limits might be reduced if new, more rigorous calculations would be carried on. The final values could be considered as parameter's uncertainties.

4. CONCLUSIONS

The most important results of this work, beside a new set of consistent accurate evaluations, are: the validation of the reaction models implemented in EMPIRE and of a part of the recommendations in RIPL; an updated fission input library for EMPIRE and valuable information regarding the uncertainty of the fission parameters to be used in determining covariances. This accomplishments may be considered a step towards fulfilling the dream of replacing an entire library of cross sections with a consistent, short set of input files and a nuclear reaction code.

ACKNOWLEDGMENTS

REFERENCES

[1] M.Herman, R. Capote, B.V.Carlson, P. Oblozinsky, M. Sin, A. Trkov, H. Wienke, V. Zerkin, "EMPIRE: Nuclear Reaction Model Code System for Data Evaluation", *Nucl. Data Sheets*, vol **108**, nr.12, p.2655 (2007)

[2] R. Capote, M. Sin, A. Trkov, M. Herman, B.V. Carlson, P. Obložinský, "Extension of the nuclear reaction model code EMPIRE to actinides' nuclear data evaluation", Proc. Int. Conf. on Nuclear Data for Science and Technology, April 2007, Nice, EDP Sciences, 255 (2008)

[3] M. Herman et al, „EMPIRE's ultimate expansion: resonances and covariances", Proc. Int. Conf. on Nuclear Data for Science and Technology, April 2007, Nice, EDP Sciences, 207 (2008)

[4] R. Capote, E. Soukhovitskii, J.M. Quesada, and S. Chiba, "Lane consistency of the dispersive coupled-channel optical model potential", Proc. Int. Conf. on Nuclear Data for Science and Technology, April 2007, Nice, EDP Sciences, 239 (2008)

[5] R. Capote, M. Herman *et al*, „RIPL – Reference Input Parameter Library for Calculation of Nuclear Reactions and Nuclear Data Evaluations", *Nuclear Data Sheets,* **110,** 12, pp 3107, (2009)

[6] M. Sin, R. Capote, Transmission through multi-humped fission barriers with absorption: A recursive approach *Phys.Rev.C* **77**, 054601 (2008)

[7] H. Wienke, R. Capote, M. Herman, and M. Sin, "Deformation dependent Tamura Udagawa-Lenske multistep direct model",

Phys. Rev. C **78**, 064611(2008)
[8] S. Goriely