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Statistical Analysis of Neutron Induced Capture Cross-Section in some Isotopes of Plutonium using EMPIRE 3.2 code

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Abstract
The need for a precise and accurate nuclear reaction cross-section is of growing interest because it serves as a basis for advanced reactor core simulations. In the neutron incident energy region $E_n \geq 1\ MeV$, nuclear data are scarce and grossly inadequate. With the aim of improving the nuclear data in this energy region, neutron-induced radiative capture cross-sections in each target of $^{238}Pu(n,\gamma)$, $^{239}Pu(n,\gamma)$, $^{240}Pu(n,\gamma)$, $^{241}Pu(n,\gamma)$,$^{240}Pu(n,\gamma)$ from threshold energy up to few MeV were performed using the theoretical calculation code EMPIRE 3.2. The results were in good agreement when compared with the available experimental data and the nuclear reaction cross-section was also predicted in the energy range where the measured data are scarce or grossly inadequate. The EMPIRE 3.2 predicted results were compared with the recently evaluated data of JENDL-5 and ENDF/B-VIII.0; the EMPIRE 3.2 results generally agreed closely at approximately $1-3$ % with Japanese Evaluated Nuclear Data Library JENDL- 5 compared to the Evaluated Nuclear Data File ENDF/B-VIII.0. These studies suggest the predictable performance of EMPIRE 3.2 code in predicting reaction cross section at higher energy. The results are important for a variety of applications, most especially in the study of heat effects in a nuclear reactor core.

Keywords: *Radiative capture, EMPIRE 3.2 code, Transmutation device, Reaction cross-section, Plutonium isotopes*

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I. Introduction

Nuclear reaction cross-section data is fundamental for many types of research, ranging from the testing of nuclear weapons to fusion energy research. The future development of the power generation system depends on the reliable data set of a wide energy range of incident neutron reaction cross section up to some MeV (Minato *et al.,* 2017; Olorunsola *et al.,* 2023). In an attempt to fulfill the need for nuclear data, the Nuclear Data Committee has prioritized the analyses of cross-sections (Soukhovitskii, *et al.,* 2016). But the difficulty in handling some target nuclei due to high radioactive resulted in complicated issues related to experimental research. Therefore, the model-based calculation is indispensable for the development of a nuclear library that is sufficient to predict nuclear reaction data and to completely describe the radiative captured cross-section and other competing reactions where the cross-section does not exist or grossly inadequate. (Bamikole *et al.,* 2018).

It is generally believed that radiative capture is best estimated within the Hauser- Feshbach statistical model. (Rochman *et al.,* 2016). The model assume that the capture process emerges during the formation of a compound nucleus CN in thermodynamic equilibrium, where the incident particle energy is shared uniformly among all nucleons and deexcited by releasing the emission in the form of a gamma-ray. Hauser- Feshbach formalism is a powerful tool for describing reaction cross-sections in the formation of compound nucleus CN, and is reliable for the prediction of an important cross-section of the abundance of actinides and advanced nuclear reactions (Thibault, 2016); which is primarily dependent on optical potential parameters including nuclear level density, and $\gamma - ray$ strength function as input ingredients.

In a compound nucleus, the $\gamma - ray$ strength function is important for a better description of $\gamma - ray$ emission which play a crucial role in the low incident energy range for nuclear de-excitation. The nuclear level densities are required for the estimation of the transmission coefficient in the formation of CN. For the evaluation of radiative capture data, several strength function parameterizations are also involved. The phenomenological model of Brink-Axel (Brink, 1957; Axel, 1962) with parameters dedicated to the Giant Multipole Resonance GMR based on standard Lorentzian for $f_{XL}(E_\gamma)$ energy dependency which represents the gamma-ray strength function is another important constituent of model parameters for the statistical compound nucleus CN for reliable prediction of capture cross sections.

This work studies consistent input parameters and the model that best describes neutron-induced radiative cross-section in the ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{240}Pu reaction to reproduces the experimentally measured data as much as possible which is relevant to the balancing of the actinide's reaction network in an advanced nuclear reactor. These reactions may also impact reactor performance by taking away neutrons that could otherwise cause nuclear fission, which is accountable for the transmutation of some of the diagnostic isotopes

II. Theoretical Background

The Hauser-Feshbach model is implemented in EMPIRE 3.2 (Herman et al., 2013) for the statistical description of the formation and decay of the CN (Compound Nucleus). Although, other reaction mechanisms contributed significantly; most especially at higher neutron incident energy $(E_n = 10\,MeV)$, Pre-equilibrium model is predominant.

2.1 **The Hauser- Feshbach Model**

Hauser- Feshbach theory pictures the Bohr hypothesis (Bohr, 1936) which states that compound nucleus decay is independent of its formation (Bauge et al., 2012). When applying Hauser – Feshbach model in EMPIRE 3.2 code, the compound nucleus formed through the incident channel a followed by subsequently decay via the outgoing channel b for the reaction cross section, $\sigma_{a,b}$ is given as:

$$\sigma_{a,b}(E) = \sum_{J\pi} \sigma_a^{CN}(E, J\pi) P_b(E, J\pi) \qquad [1]$$

here $\sigma_a^{CN}(E, J\pi)$ represent the formation of Compound Nucleus CN cross-section through the incident channels a in the state of spin and parity $J\pi$ and $P_b(E, J\pi)$ is compound nucleus decay probability through outgoing channel b. (Herman et al., 2013)

The Hauser- Feshbach formalism given in Equation 1 is a powerful tool to describe the reaction cross sections created by the compound nuclei. The main difficulty in the evaluation resides in the decay probability, which is defined in terms of the transmission coefficient associated with reaction channels. An optical model is used to supply the neutron transmission coefficient, which is given as

$$P_b(E, J\pi) = \frac{T_b(E_x, J\pi)}{\sum_c T_c(E_x, J\pi)} \qquad [2]$$

For estimation of the transmission coefficient for the compound nuclei, level densities are required; while describing the $\gamma - ray$ emission channel, the $\gamma - ray$ strength function also plays an important role.

2.2 **Nuclear Level Density.**

In all nuclear reaction models, the nuclear level densities are the most important statistical characteristics of excited nuclei. At low- lying energy level, a discrete spectrum is displayed by the nuclear levels. At high energies, the number of levels per unit energy becomes so high that it is difficult to experimentally resolve the differences. It is therefore, necessary to use the nuclear models for a continuous description of level density. The most used level density was derived analytically within the Fermi Gas Model FGM (Bethe, 1973)

In FGM, the intrinsic level density with spin J, and parity π and excitation energy E_x is factorise in terms of the state density ρ, spin J and parity π dependence as

$$\rho(E_x, J, \pi) = \rho(E_x)\rho(J, \pi) \qquad [3]$$

And the energy dependence is given as

$$\rho(E_x) = \frac{expS}{\sqrt{Det}} \qquad [4]$$

where S represent the entropy and Det is define in equation 4. Spin J and parity π dependence becomes

$$\rho(J, \pi) = \frac{1}{2}\frac{(2J+1)}{\sqrt{8\pi\sigma^3}} exp\left[-\frac{(J+{}^1\!/_2)^2}{2\sigma^2}\right] \qquad [5]$$

here σ^2 is the spin cut-off parameter and we assume an equal parity distribution and the dependence of the excitation energy is determined from state equation using the Fermi Gas model with the other thermodynamic functions of a nucleus on its temperature T are:

$$E_x = aT^2; \qquad S = 2aT; \qquad \sigma^2 = \Im T; \qquad Det = {}^{144a^3T^5}\!/_{\pi} \qquad [6]$$

Where a and $\Im$ are level density parameter and nuclear moment of inertia, respectively.

Excitation energy E_x is replaced with effective energy U in so as to account for odd-even effects in nuclei, which is calculated as

$$U = E_x - \Delta \qquad [7]$$

Where Δ is related to the pairing energy and the state density is

$$\rho^{FG}(E_x) = \frac{\sqrt{\pi}}{12a^{1/4}} \exp(2\sqrt{aU}) \qquad [8]$$

And the level density becomes

$$\rho^{FG}(E_x, J, \pi) = \frac{2J+1}{48\sqrt{2}\sigma^{3/2}a^{1/4}u^{5/4}} \exp\left[2\sqrt{aU} - \frac{(J+1/2)^2}{2\sigma^2}\right] \qquad [9]$$

The above equation shows that Fermi- Gas model depends on level density parameter a, spin cut - off parameter σ and pairing energy parameter Δ.

2.3 **Gamma Ray Strength Function**

The gamma ray function plays a crucial part in the compound nucleus CN for modelling capture cross section, production of gamma ray spectra and population of isometric states, which give detailed information on nuclear structure and it is used to study the nuclear reaction mechanism.

For the evaluation of radiative capture data, several strength function parameterizations are involved. Phenomenological permits the cross section for absorption by an excited state to be equated to that of the ground state. Introducing the γ −ray strength function $f_{x\ell}(E_\gamma)$, the transmission coefficient is given as (Herman et al., 2013)

$$T_{XL}^{GMR}(E_\gamma) = 2\pi . f_{XL}(E_\gamma) . E_\gamma^{2L+1} \quad [10]$$

where, $f_{XL}(E_\gamma)$ gamma ray strength function and emitted $\gamma - ray$ energy of type X ($X = E\ or\ M$ for electric and magnetic transitions) is given as

$$f_{XL}(E_\gamma) = E_\gamma^{-(2L+1)} \langle \Gamma_{XL}(E_\gamma) \rangle / D_l \quad [11]$$

where $E_\gamma^{-(2L+1)} \langle \Gamma_{XL}(E_\gamma) \rangle$ is average Reduced Partial Radiation Width and D_l denoted average Level Spacing

EMPIRE 3.2 uses the strength functions representing the spline fit of the experimental data provided by Kopecky 1993 for normalization, which are given as function of mass number for 40 < A < 260.

Moreover, six closed forms of El strength functions are included in EMPIRE 3.2 code which includes the Enhanced Generalized Lorentzian EGLO (Kopecky, 1993); Modified Lorentzians MLO 1, ML02, ML03 (Plujko, 2000) the Generalized Fermi Liquid Model (GFL) and the Standard Lorentzian SLO (Mughabghab and Dunford, 2000) Depending on the formalism chosen, the shape of $f_{XL}(E_\gamma)$ is modified, especially at low γ emission energies that are mostly responsible for the capture cross section.

For other multipolarities, the radiative strength functions for higher multipole orders F_{El}/F_{ML} are calculated using the relationships between single-particle radiative strength functions expressed in the Weisskopf (Brink, 1955) form.

For the electric transition we have.

$$F_{E(L+1)}/F_{E(L)} = CE_\gamma^{-2}[(3+L)/(5+L)]^2 \quad [12]$$

where $C = [R/(\hbar c)]^2 = 3.7\ x\ 10^{-5} A^{2/3}$ if a nuclear radius $R = 1.2A^{2/3}\ Fm$ is assumed.

For the magnetic transition, we use

$$F_{E(L+1)}/F_{E(L)} = 10\ \left[\frac{\hbar}{mcR}\right]^2 = 0.307A^{-2/3} \quad [13]$$

III. Model and Reaction Flows

Complete set of input parameters is provided in EMPIRE 3.2 code which enable default calculations for the specific user input. Optical model including coupled channel CC coupling scheme was used for our calculation and the nucleus is treated as deformed. The direct reaction and elastic scattering were evaluated with ECIS06 code (Raynal, 1971) within the frame of the generalized optical model. The optical segment and the discrete level were extracted from RIPL-2 (Capote *et al.,* 2009). Discrete population and collective levels in the inelastic scattering were, respectively, taken into account from ground state to the couple level.

Hofman, Richert, Tepel, and Weidenmueller (HRTW) model was used to account for reaction between the projectile and the target nucleus to form a compound nucleus, which subsequently emits a particle or gamma ray, with width fluctuation correction (Hofmann et al., 1975) at low energy reaction. The most vital ingredients for the compound nucleus include the transmission coefficients that arises from the optical potential model and the level densities. The transition coefficient is created from the optical model. Moreover, the gamma-ray transmission coefficients enter into the compound nucleus model that was used for capture cross-section calculation.

The modeling of the $\gamma -$ cascade in compound nucleus CN, includes the angular momentum that accounts for the competition between the γ- emission and particle emission along the de-excitation chain in the continuum and the discrete transition level with a maximum multipolarity set to 1, the most used transition is $E1, M1,$ and $E2$. The formalism of gamma-ray transmission in EMPIRE is based on the combination of the Weisskopt single-particle model (Blatt and Weisskoff, 1952) and the Giant Dipole Resonance model of the Brink -Axel hypothesis. (Axel, 1962; Brink, 1957). The GDR parameters were retrieved from the RIPL -3 library (Capote et al., 2009). The modified Lorentzian as MLO1 (Capote et al., 2009) in the closed form was computed to obtain the gamma-ray transmission coefficient from $\gamma - ray$ strength function. The $\gamma - ray$ strength function is normalized to the experimental information to deduce the value of Γ_γ and D_0 or adjusted to reproduce the radiative capture cross-section.

In the description of level densities in the continuum, the level densities that are specific to the EMPIRE 3.2 code (Herman et al., 2013) were employed. The formalism uses the generalized superfluid model (Ignatyuk et al., 1975) that distinguished two energy regions. At high energies, the level density is described by the Fermi-gas model, while at low energy, the concepts of pairing and shell effects are included in the model (Herman et al., 2013). Collective enhancements as a result of nuclear vibration and rotational are taken into account in the nonadiabatic approximation with different formulations which are parameterized to fit the available experimental data

The PCROSS module Iwamoto Hadara model includes a preequilibrium mechanism which accounts for the formation probability of the cluster exciton below and above the Fermi surface (Iwamoto and Harada, 1982) and Kalbach;s (1977) methods to calculate the nucleon and the contribution of each model at various incident energy correspond to excitation energy are thereby presented.

IV. Results and Discussions

Plutonium isotopes were investigated using the statistical modelling code EMPIRE 3.2 and the theoretical results of the reaction cross section together with available experimental data retrieved from EXFOR (Otuka et al., 2014) including the recent Evaluated Nuclear Data from ENDF/B-VIII.0 and JENDL-5 (Brown et al., 2018; Iwamoto et al., 2021) are presented in figure 1-5

Figure 1 displays the nmspectrum of capture cross section of $^{238}Pu(n,\gamma)$ against incident neutron. EMPIRE 3.2 results with available experimental data of Chyze et al., 2013; Silbert and Berreth, 1973, are given together in the Figure 1. It can be seen from Figure 1 that the predicted results from EMPIRE code are consistence with the experimental data retrieved from EXFOR (Otuka et al., 2014). Chyze et al., 2013, reported three data points above evaluated value at $E_n = 03 - 0.7\ MeV$. This may likely be linked to the error of difficulty in removing the reaction kinematic from the observed cross section. Predicted nuclear reaction cross section from EMPIRE 3.2 code compares reasonably well with Japanese Evaluated Nuclear Data Library JENDL-5 (Iwamoto et al., 2021) than Evaluated Nuclear Data File ENDF/B-VIII (Brown et al., 2018). Deviation of 3.9 % within the $E_n = 0.7 - 3.8\ MeV$ between EMPIRE 3.2 results and existing Evaluated Nuclear Data ENDF/B-VIII.0 was observed. The discrepancy may likely be traced to the too low contribution of $\gamma - strength$ function.

Figure 2 displays the response of capture cross section of ^{239}Pu against incident neutron energy. Theoretical model results of EMPIRE 3.2 code with the available experimental data of Konolov et al., 1975, and Schomberg et al., 1970 for $^{239}Pu(n,\gamma)$ reaction cross section. It can be clearly seen in figure 2 that the results of theoretical prediction from EMPIRE 3.2 reproduce the experimental data retrieved from EXFOR (Otuka et al., 2014). Predicted nuclear cross section was closely matched with Evaluated Nuclear Data reported from JENDL-5 (Iwamoto et al., 2021) throughout the energy region and compared fairly with ENDF/B-VIII (Brown, et al., 2018) at $E_n \leq 0.1\ MeV$ and toward the tail of the spectrum. Deviation of 5 % at $E_n = 0.7 - 4.8\ MeV$ exists between EMPIRE results and ENDF/B-VIII.

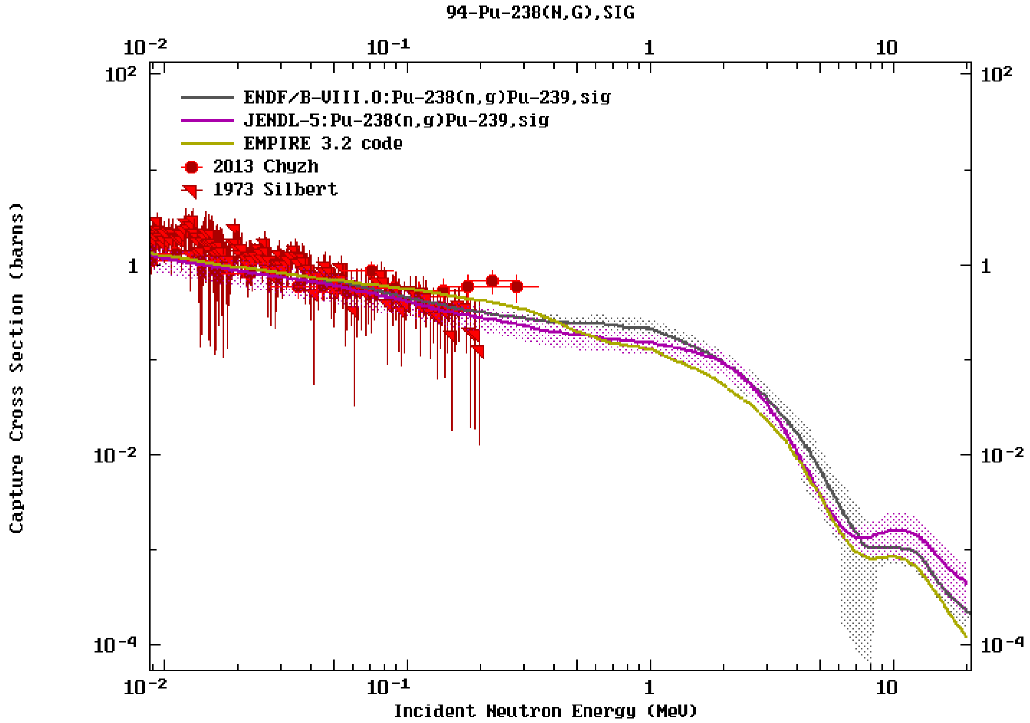

Figure 1: The spectrum of $^{238}Pu(n,\gamma)$ cross section (barns) as a function of neutron incident energy (MeV)

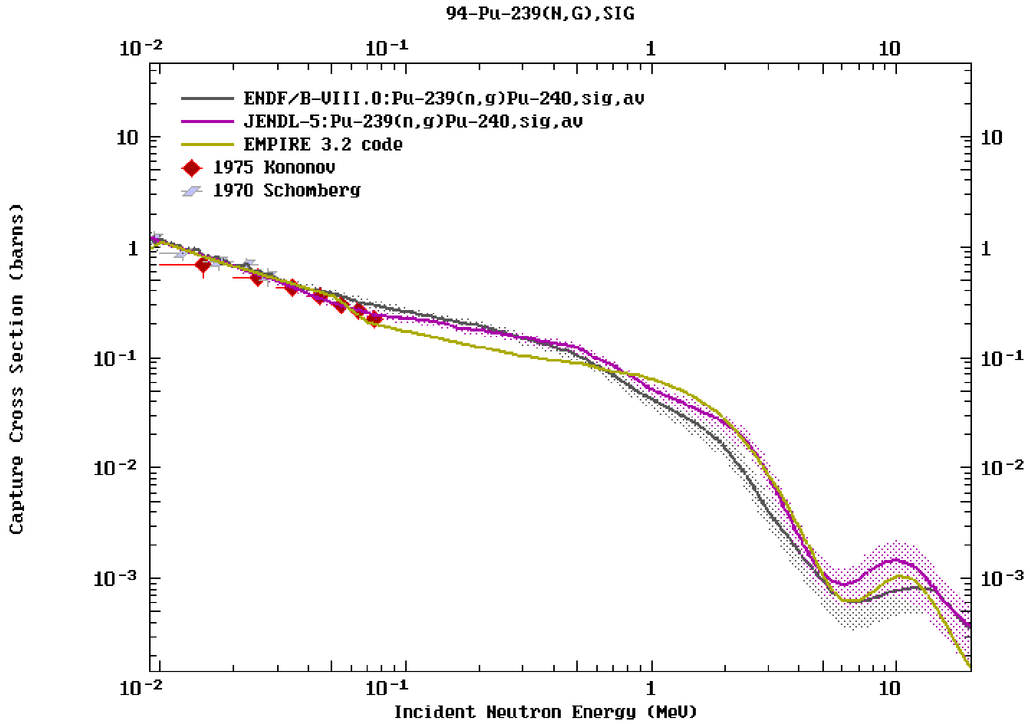

Figure 2: The spectrum of $^{239}Pu(n,\gamma)$ cross section (barns) as a function of neutron incident energy (MeV)

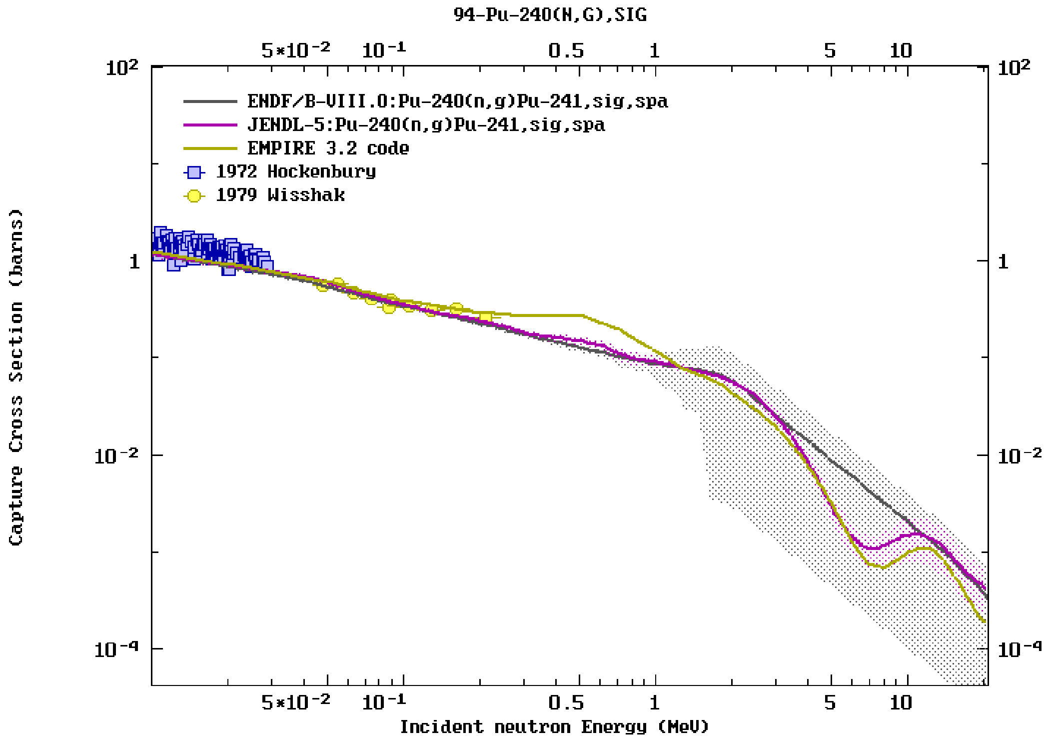

Figure 3: The spectrum of $^{240}Pu(n,\gamma)$ cross section (barns) as a function of neutron incident energy (MeV)

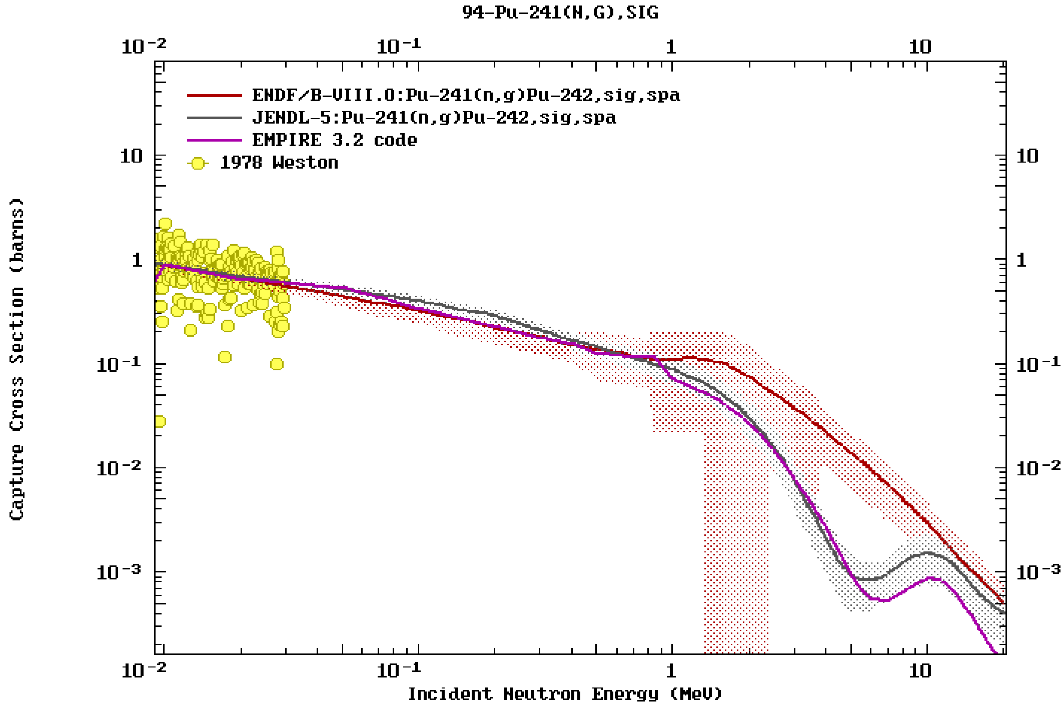

Figure 4: The spectrum of $^{241}Pu(n,\gamma)$ cross section (barns) as a function of neutron incident energy (MeV)

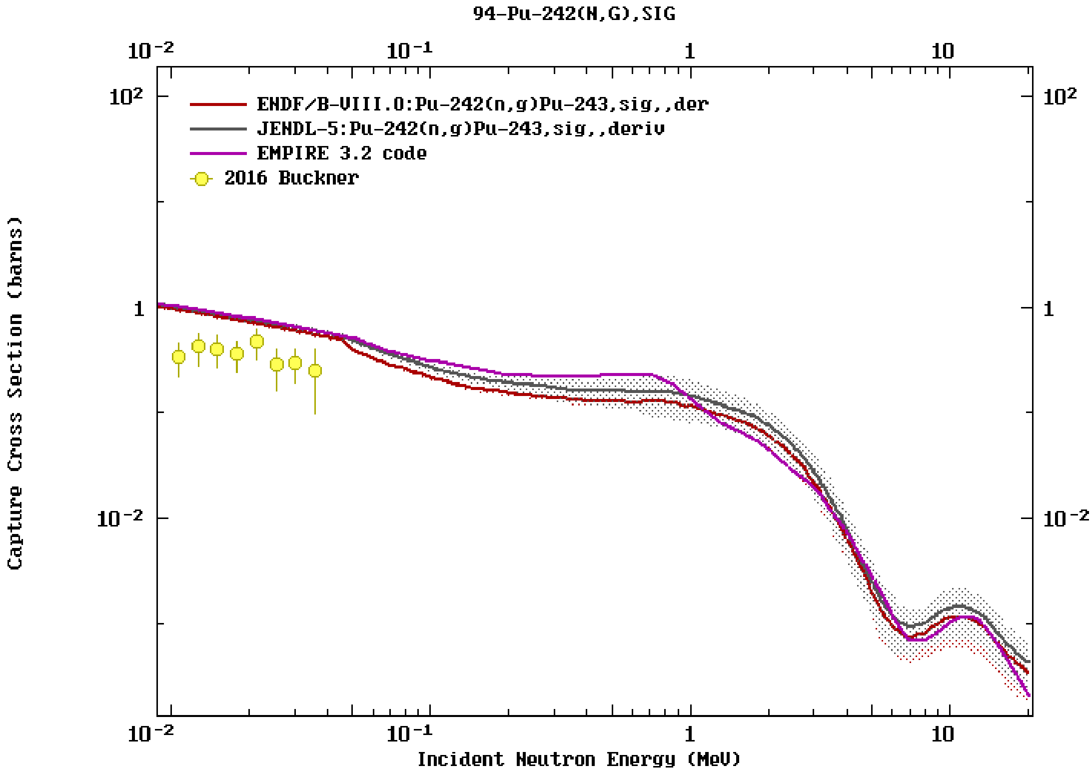

Figure 5: The spectrum of $^{242}Pu(n,\gamma)$ cross section (barns) as a function of neutron incident energy (MeV)

The spectrum of theoretical prediction using EMPIRE 3.2 code and the available experimental data from Hockenbuury et al., 1972, and Wisshak and Kaeppeler, 1979 for the reaction of $^{240}Pu(n.\gamma)^{241}Pu$ is given in figure 3. The predicted results are in accordance with the retrieved experimental values from EXFOR (Otuka et al., 2014). There is good agreement between the EMPIRE 3.2 prediction and the evaluated nuclear data file (Brown et al., 2018; Iwamoto et al., 2021). Discrepant of 2.26 % at $0.1 - 1.5\ MeV$ was exists between predicted results and JENDL – 5 and 3.6 % deviated in the incident energy $E_n \geq 3.4$ from ENDF/B-VIII. Rather, EMPIRE 3.2 result is very close to JENDL- 5 compared to ENDF/B-VIII.

In Figure 4, the spectra of theoretical prediction of nuclear reaction cross section on $^{241}Pu(n,\gamma)$ using EMPIRE 3.2 code are presented. Experimental value of Weston et al., 1978, was considered for validation of the predicted results. It can be clearly seen that EMPIRE results are in good agreement with the measured value of Weston and Todd, 1978. When compare the predicted results with the Evaluated Nuclear Data Library from JENDL and ENDF/B-VIII. The spectrum of EMPIRE 3.2 follows the same trend of JENDL and agrees with ENDF/B- VIII.0 up to an incident energy $E_n \leq 1\ MeV$. Above this energy region discrepant of 3.2 % exists between EMPIRE results and the evaluated nuclear data from ENDF/B-VIII.

Predicted cross section results for the $^{242}Pu\ (n,\gamma)$ reaction using EMPIRE 3.2 code and the experimental data of Burkner et al., 2016, with existing evaluated nuclear data from ENDF/B-VIII (Brown et al., 2018) and JENDL (Iwamoto et al., 20221) is given in figure 5. It can be seen clearly that both results from EMPIRE 3.2 and the existing Evaluated Nuclear Data Files predict a nuclear cross section higher than the measured data from Burkner et al., 2016. This may likely link to the experimental error in Burkner et al., 2016, or difficulties in removing the observed cross section from reaction kinematics or suggested to be a model deficiency.

V. Conclusion

The results presented in these studies demonstrate the performance of theoretical code EMPIRE 3.2 in the prediction of the behaviour of neutron induced reaction cross section on each of ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{240}Pu target within the (n,γ) reaction channel up to few MeV. Statistical model parameters (such as level density parameters, gamma ray strength function) were successfully use to obtained satisfactory fits to experimental data retrieved from EXFOR (Otuka, et al., 2014) within the EMPIRE 3.2 nuclear reaction code specific input (Herman et al., 2013). Moreover, the important contribution of the model arises within the reaction mechanism of the compound nucleus as the incident energy(E_n) increases. The competition of neutron captures cross section in which the gamma decay follows to the continuum and the discrete states are described within the compound nucleus including the width fluctuation corrections. At some higher excitation energies, the inelastic channel was open, which was described by the compound nucleus theory. Other's contribution reaction mechanism are Optical direct parameters used to described elastic scattering cross section shape and transition coefficient for statistical description in compound nucleus. Also, at higher incident energy, $E_n \geq 10\ MeV$ pre- equilibrium contribution to the reaction cross section where the inelastic cross sections of the discrete and continuum are possible. The predicted results were compared with the evaluated nuclear data files of ENDF/B-VIII.0 and JENDL-5 (Brown et al., 2018; Iwamoto et al., 2021). Disagreement occurs between these studies and the existing evaluated nuclear database of ENDF/B-VIII.0 (Iwamoto et al., 2018) but closed to the database of JENDL-5 in the neutron incident energy of $E_n \geq 1\ MeV$ where experimental data is scarce. The method employed for the statistical analysis of the model and parameters may likely be a potential source of uncertainties. Additional work is required to improve model parameters in order to minimized the contribution to the model uncertainties at this energy region. Nevertheless, our studies have shown that EMPIRE 3. 2 provides new cross section values that are consistence with available experimental data and in the energy level where the scissors resonance is the predominant influence is assumed. The results are therefore relevant in the studies of heating effect in nuclear reactors core which is crucial in nuclear waste management.

Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared

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