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INVESTIGATION OF OPTIMAL MODEL AND PARAMETERS FOR DETERMINATION OF NEUTRON INDUCED REACTION CROSS-SECTION ON ISOTOPES OF PLUTONIUM

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Abstract

Statistical nuclear reaction calculation code EMPIRE 3.2.3 was used to evaluate neutron induce reaction total and fission cross section on plutonium isotopes ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu which are important in modern nuclear reactors. Optimal model shows the contribution from direct reaction, pre-equilibrium and compound mechanism as particular reaction appear and disappear with increase in incident energy for both total and fission reaction cross section up to $20\ MeV$. Level density parameter contribute significantly for the complete description of the whole energy range. EMPIRE results are generally, in good agreement with existing evaluated nuclear data and available experimental data retrieved from EXFOR for total cross section. But for neutron induce fission reaction; EMPIRE results reproduced the experimental data better than the existing evaluated nuclear data from Evaluated nuclear data file ENDF/B-VIII.0, Joint Evaluated Fission and Fusion Nuclear Data Library JEFF- 3.3, and Japanese Evaluated Nuclear Data Library JENDL with standard deviation of 2.12 % at $E_n \leq 0.5\ MeV$. Consequently, EMPIRE 3.2.3 code has proved to be reliable in prediction of total and fission reaction cross sections in the energy grid where there is no experimental report.

Keywords: Cross section, EXFOR, EMPIRE code, Plutonium isotopes, Level density

1.0 Introduction

Information about total and fission reaction cross section are needed because they serve as basis ingredients in many advanced nuclear applications. However, literature reported that experimental data are scarce and inadequate (Trkov, et al., 2017). consequently, evaluated data have been inconsistent and not maintained with the same level of accuracy (Nobre, et al., 2019). Most available nuclear data for nuclides of the minor actinides do not fulfill the current accuracy requirements, therefore there is need to embark on the search for a more improved nuclear data for transuranium nuclides (Bamikole and Agu, 2018). Most especially, both long- lived and non -fissile of plutonium isotopes that contributed to accumulation of nuclear waste

This work determine the best model and parameters for neutron induced nuclear reaction on ^{238}Pu, ^{239}Pu, ^{240}Pu, $^{241}Pu, and$ ^{242}Pu using EMPIRE 3.2 the statistical nuclear reaction code (Herman, et al., 2007) from threshold energy up to few MeV with the aim to verify the consistency of models of nuclear data; prediction was made where measurements data are either available or not existence to improve the nuclear reaction data for future application. We reconcile the evaluation techniques and microscopic physics by adjusting the parameters, to fits the most recent and consistent experimental data retrieved from EXFOR (EXFOR, 2017). Theoretical calculation results from EMPIRE 3.2.3 were compared with available experimental results and current nuclear-evaluated data (Brown *et al.*, 2018; Iwamoto *et al.*, 2021; Plompen, *et al.*, 2020) to check the consistency based on the contributing reaction mechanism i.e (optical model, compound nucleus and pre-equilibrium models). The structure of this work is organized as follows: The theoretical consideration is presented in section 2. Nuclear level density in section 3, input parameters, and models used is presented in section 4 while sections 5 and 6 respectively present the result and discussion, and conclusion.

2.0 Theoretical Considerations

Most of the interactions in the continuum range have the probability of non-negligible occurrence. The total cross-section or direct interaction is calculated with the optical model. For neutron–nucleus interaction, the Schrodinger equation is introduced to describe Optical Potential (Raynal, 2003) which is given as:

$$\left[-\frac{\hbar^2}{2\mu}\bar{V}^2 - V(\vec{r}) - E\right]\varphi(\vec{r}) = 0 \qquad [1]$$

And $\mu = \frac{mM}{m+M}$, the reduced mass of the system and colliding masses of two particles are m and M respectively. And kinetic energy of the system is represented by E. Although, for optical potential, several phenomenology potentials can also be considered for calculations. For convenience calculation, the optical potential is written in the form

$$V(r) = V_c(r) + Uf_s(r) + iWg(r) + \left(\frac{\hbar^2}{m_\pi c}\right)^2 \frac{1}{r} U_s \frac{df_s}{dr}\, l.\sigma \qquad [2]$$

Where $V_c(r)$ represent nucleus, electrostatic potential included in the proton optical potential only. U, W and U_s are the real, imaginary spin -orbit potentials depth. the scalar product of the intrinsic and orbital angular momentum operation is represented by quantity $l.\sigma$ and is given as

$$l.\sigma = l \qquad \text{for } j = l + \tfrac{1}{2}$$

$$= -(l+1) \qquad \text{for } j = l - \tfrac{1}{2}$$

The form of $f_i\ (r)$ and $g(r)$ define the wood – Saxon potential shape. This is defined as

$$f_i(r) = \frac{1}{1 + \exp(r - R_i/a_i)}$$

$$g(r) = -4a_i d/dr f_i(r) \qquad [3]$$

Where parameters are the radius $R_i = r_i A^{1/3}$ and A is atomic mass with the diffuseness a_i for each of the potential component

When complex nuclear potential is introduced in the radial part of Schrödinger equation, the total elastic scattering is defined as

$$\sigma_T = \sigma_E + \sigma_R = \frac{\pi}{k^2}\sum_{lj}(2j+1)(1 - Res_l^j) \qquad [4]$$

Optical model helps to separate the incident projectile flux into three main components. Such as shape elastic, the direct elastic and reaction cross section and it also serve as a basis for evaluation process by providing neutron transmission coefficient used in Hauser- Feshbach theory and pre- equilibrium model.

EMPIRE 3.2.2 implemented Hauser-Feshbach for statistical model (Hauser and Feshbach, 1952), where the parity coupling and angular momentum are taking into consideration. The reaction cross section in Hauser – Feshbach model is given as

$$\sigma_{a,b}(E) = \sum_{J\pi} \sigma_a^{CN}(E, J\pi) P_b(E, J\pi) \quad [5]$$

The formation of compound nucleus CN cross section is represented by $\sigma_a^{CN}(E, J\pi)$ for spin and parity $J\pi$ state associated with incident channel a, while excitation energy E_x in channel b of CN decay probability is $P_b(E, J\pi)$. The decay probability is determined in term of transmission coefficients written as

$$P_b(E, J\pi) = \frac{T_b(E_x, J\pi)}{\sum_c T_c(E_x, J\pi)} \quad [6]$$

Where T_b is transmission coefficient for CN decay into channel b with some energy E.

The compound nucleus mechanism has been incorporated into the model of Weisskopf – Ewing and Hauser-Feshbach (Weisskopf and Ewing 1940; Hauser and Feshbach, 1952)

3.0 Nuclear Level Density

Level densities is required in modeling of nuclear reaction, especially when discrete levels information is missing for higher excitation energy. The most used level density is derived within Fermi Gas Model (FGM) developed from Bethe (1956) which depend on parameter a. EMPIRE described level density with several models with respect to parametrization, in which three of them are phenomenological which includes Generalized superfluid model GSM (Ignatyuk *et al.*, 1993), Enhanced Generalized, superfluid model EGSM (D'Arrigo *et al.*, 1994) and Gilbert Cameron model GCM (1965) which account for shell correction and fourth one is based on microscopic model (Goriely, *et al.*, 2007).

In Fermi gas (Bethe, 1956), the energy dependent level density parameter a at higher excitation energy takes into account the shell effects are written as

$$a(E_x) = \tilde{a}\left[1 + \delta W \frac{1-\exp(-1\gamma U)}{U}\right] \quad [7]$$

Where shell correction energy in MeV is given as δW. The asymptotic level density value $\tilde{a}$ is written as:

$$\tilde{a} = \alpha A + \beta A^{2/3} \quad [8]$$

With the parameters of $\alpha = 0.06926, and\ \beta = 0.2828$ obtained from a simultaneious fit level density to all average spacing parameters D_0 in the RIPL (Capote et al., 2009). For systematic shell parameter γ is given as:

$$\gamma = \frac{0.433}{A^{1/3}} \quad [9]$$

The constant temperature total level density $\rho_T^{tot}(E_x)$ below some matching energy E_x^{match}, is

$$\rho_T^{tot}(E_x) = \frac{1}{T}\exp\left(\frac{E_x - E_0}{T}\right) \quad [10]$$

When this expression is match with Fermi gas expression, the best fit of the cumulative discrete levels is possible in the nucleus. The level density over the entire energy range is smoothly obtained

4.0 Input Parameters and Model.

Couple channels CC optical model for couple levels and distorted wave Born Approximation DWBA for uncouple discrete levels were employed to determine the population of collective levels for incident and outgoing channel respectively for elastic and reaction cross section. Ground state and the couple level was taking into consideration for calculation of transmission coefficient for further used in statistic hauser- Feshba and pre-equilibrium model (Raynal, 1971). We selected optical model parameter OM proposed by Capote *et al* (2008) with RILP 2408 to described the interaction of proton and neutron with actinide nuclei up to $200\ MeV$ to evaluate total reaction cross sections up to 20 MeV

In compound nucleus model, Hauser – Feshbach (1952) formalism and statistics model of Hofmann -Richert-Tepel- Weidenmuller (HRTW) is implemented with full gamma – cascade for cross section evaluation (Hofman, *et al.*, 1975). Angular momentum and parity coupling are taken into account along with fission channel. Among other formalism to described gamma ray strength function was inputted within the Modified Lorentzian model (Plujko, 2000). In other to reproduced the experimental data accurately, we retrieved discrete level density and $\gamma - ray$ strength functions from RIPL-2 (Capote *et al.*, 2009) Individually for each isotope of interest.

Enhanced General Superfluid model (EGSM) (D' Arrigo *et al.,* 1994) were used to described both equilibrium and saddle point level density, which picture the structure of nuclear at low energy region.

Pre – equilibrium model was calculated by considering PCROSS exciton model Iwamoto – Harada (1982) that contained nucleon, gamma and emission of cluster inclusive. Other relevant parameters were tested to reproduced the available experimental data to predict the cross section where the measurement data were scarce or not exist.

5.0 Results and Discussion

Direct reaction part is calculated with ECIS code includes in EMPIRE reaction code. (Raynal. 1971). We selected coupled-channel Optical Model Potential (OMPs) proposed by Capote et al. (2008) which contain the dispersive terms with inclusion of non- local contribution. We compared the prediction of this potential with available experimental data from EXFOR (EXFOR, 2017) and existing evaluated nuclear data from Evaluated nuclear data file ENDF/B-VIII.0 (Brown, *et al.*, 2018), Joint Evaluated Fission and Fusion Nuclear Data Library JEFF- 3.3 (Plompen *et al.*, 2020) and Japanese Evaluated Nuclear Data Library JENDL (Iwamoto et al., 2020) for the total reaction cross section on $^{238}Pu,\ ^{239}Pu,\ ^{240}Pu,\ ^{241}Pu, and\ ^{242}Pu$ as displayed in figure 1 – 5. Our results agreed well with the experimental value form Harvey, et al. (1988) as shown in figure 2 also in figure 3 with the experimental report from Poenitz, and Whalen (1983). We obtained a reasonable agreement between our model and existing evaluated nuclear data in the figure 1-5. Deviation of 1% was observed within $E_n \leq 1\ MeV$ in EMPIRE results and existing ENDF/B-VIII.0 (Brown, *et al.*, 2018) as shown in figure 4 and figure 5. The accurate prediction of our results clearly shows that couple channel model in RIPL-2408 we can confidently achieve a good description of total reaction cross section where the experimental data is scarce.

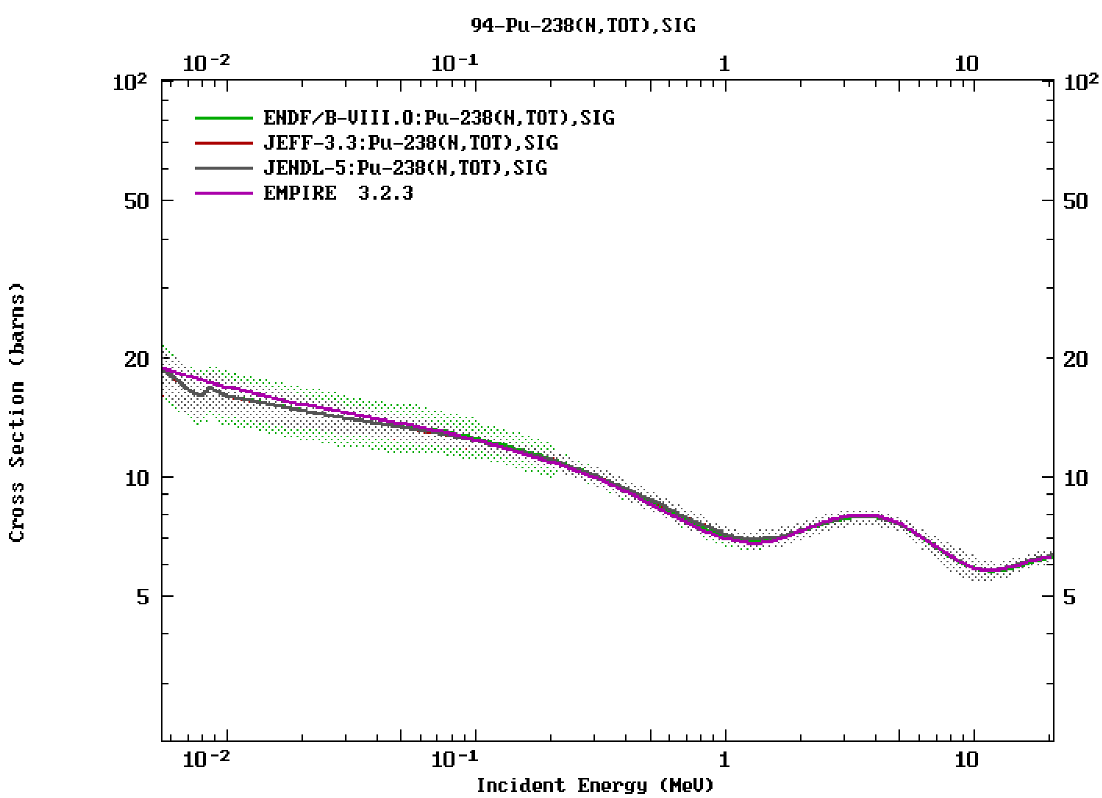

Figure 1

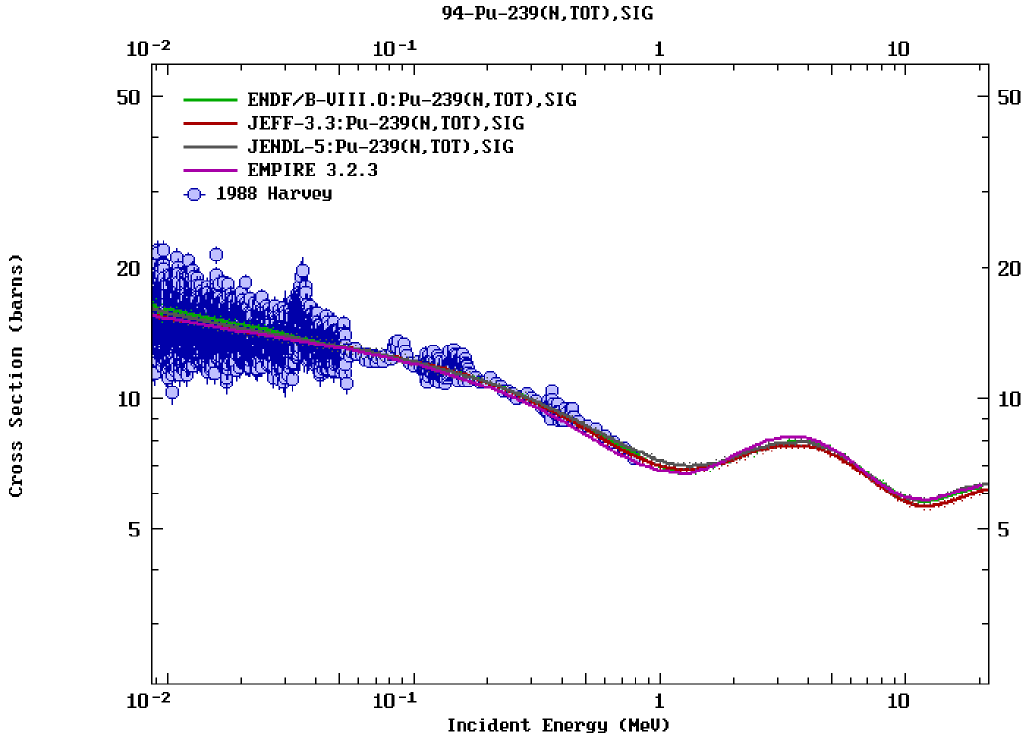

Figure 2

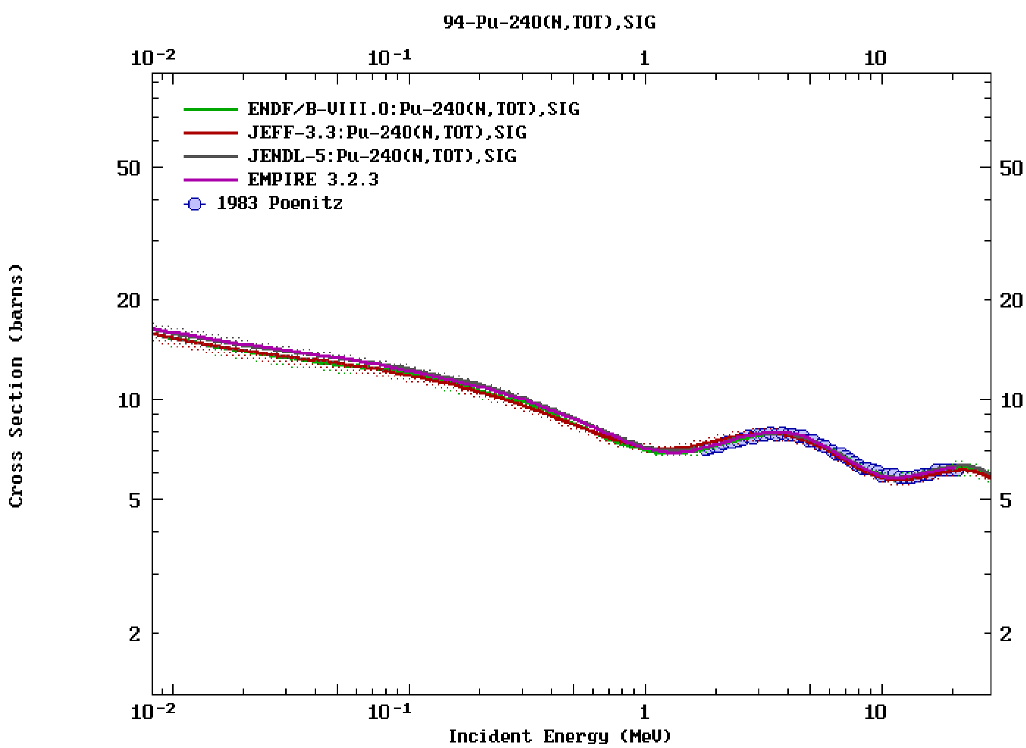

Figure 3

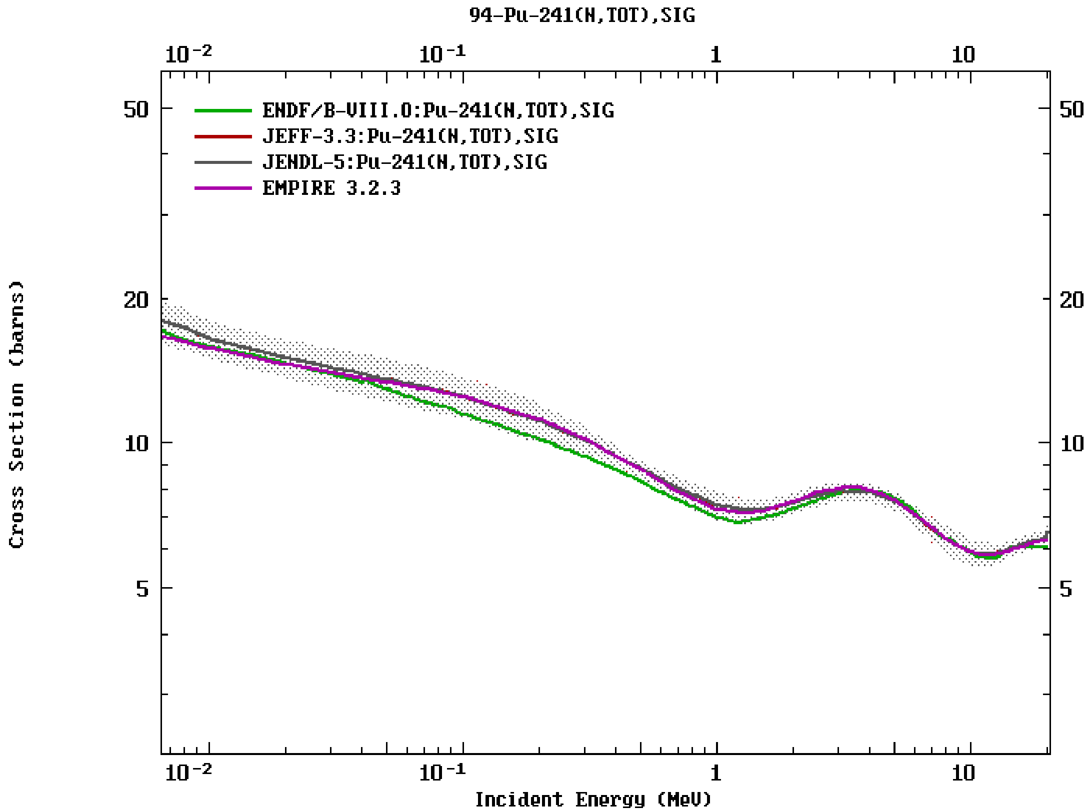

Figure 4

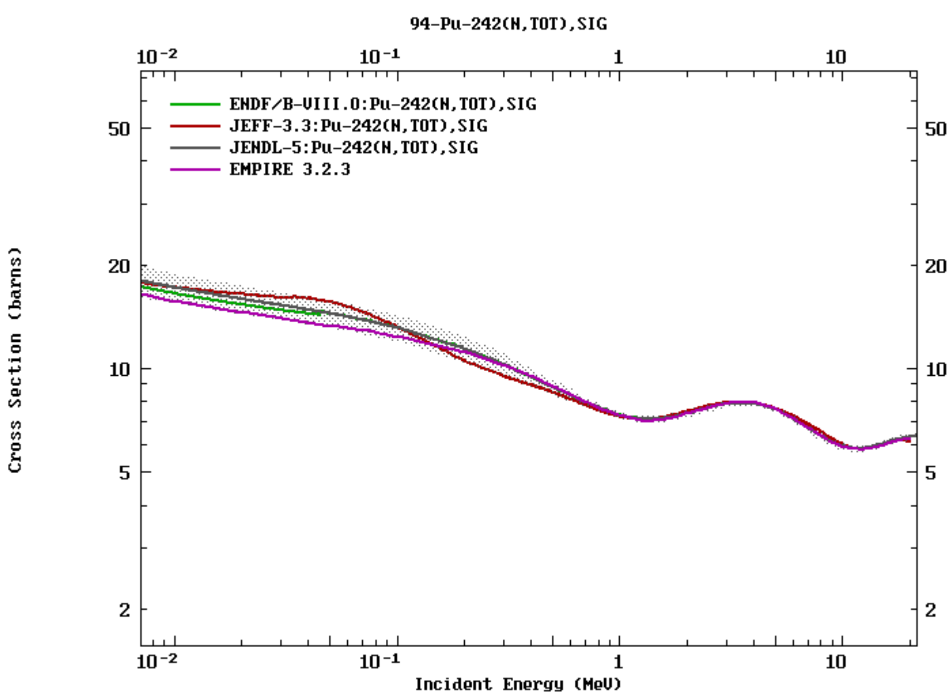

Figure 5

The optical model is not only used for calculating total and elastic cross section reaction; but, can also apply in transmission coefficient which is used in statistical Hauser-Feshbach and pre-equilibrium model.

Statistical model for description of fission cross section is the product of formation of compound nucleus cross section $\sigma_{CN}(E^*, J, p)$ and a fission probability, $P_f(E^*, J, p)$, depending on the compound nucleus of excitation energy $E^* = E_n + B_n$. Where E_n and B_n are neutron incident energy and compound nucleus binding energy respectively. The parity p and angular momentum J is defined in equation 4 which is implemented in EMPIRE reaction code. Other parameters such as discrete state, fission barrier parameters including the information parameters on microscopic and phenomenological for deduction of the fission cross section are contain in RIPL -2 (Capote *et al.,* 2009). Different model and parameters were varied to fits the experimental data retrieved from the EXFOR (Exfor, 2017) for each isotope of interest. The information adopted for fission parameters in EMPIRE reaction code (Herman *et al.,* 2007) were considered for consistency of important reaction channel. The recommended empirical value from RIPL- 2 (Capote *et al.,* 2009) were adopted for discrete transition state and multi – humped fission barrier was smoothly joined by parameterization. Different level densities and parameters for fission as presented in default EMPIRE 3.2 code (Herman, 2007) were tested to approximate the measured data for each isotope.

Figure 6 display the response of fission cross section on ^{238}Pu against energy. Experimental report from Pai *et al.* (2015), Hughes *et al.* (2014), Granier *et al.* (2009) and Ressler *et al*., 2011 were considered for validation of our results. Our evaluated value seems to compare well at higher energy with experimental data and existing evaluated nuclear data file (Brown et al., 2018; Iwamoto *et al.,* 2021; Plompen *et al.,* 2020). But below $E_n = 0.5\ MeV$, three data point from Hughes *et al.* (2014) were seen to deviate from EMPIRE result and evaluated nuclear data. This may likely due to model defect. At incident energy $E_n \leq 0.5\ MeV$, EMPIRE 3.3.2 result was observed to deviate 3.8 % below existing evaluated nuclear data. Too low contribution from the direct reactions may likely be reason.

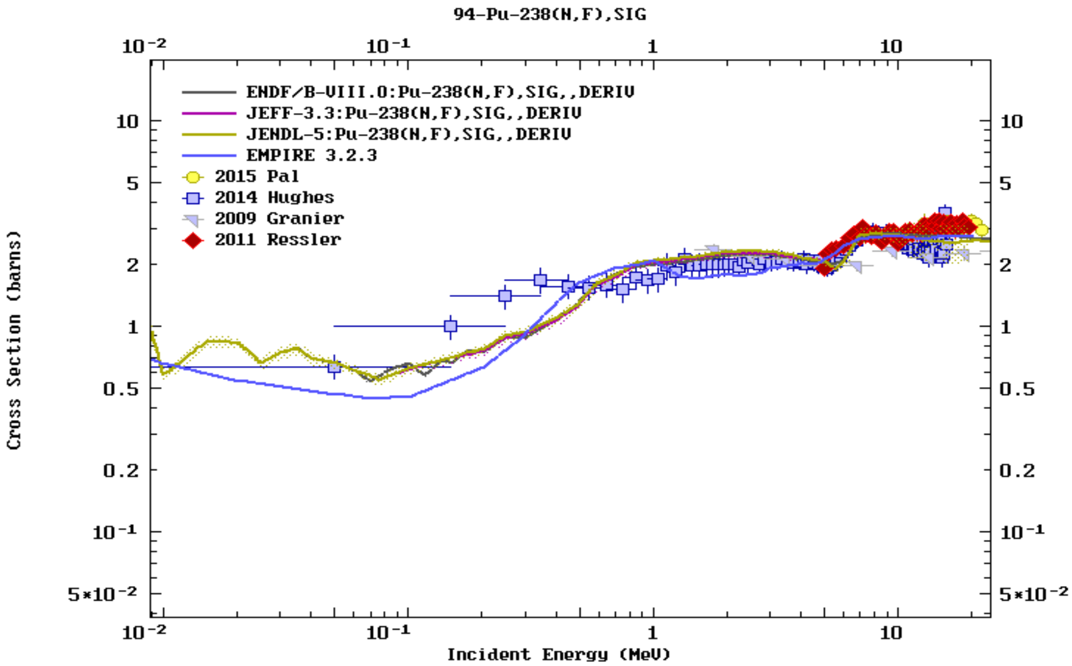

Figure 6: Result from EMPIRE 3.2.3 code in comparison with available data from EXFOR and existing Evaluated Nuclear Data Library

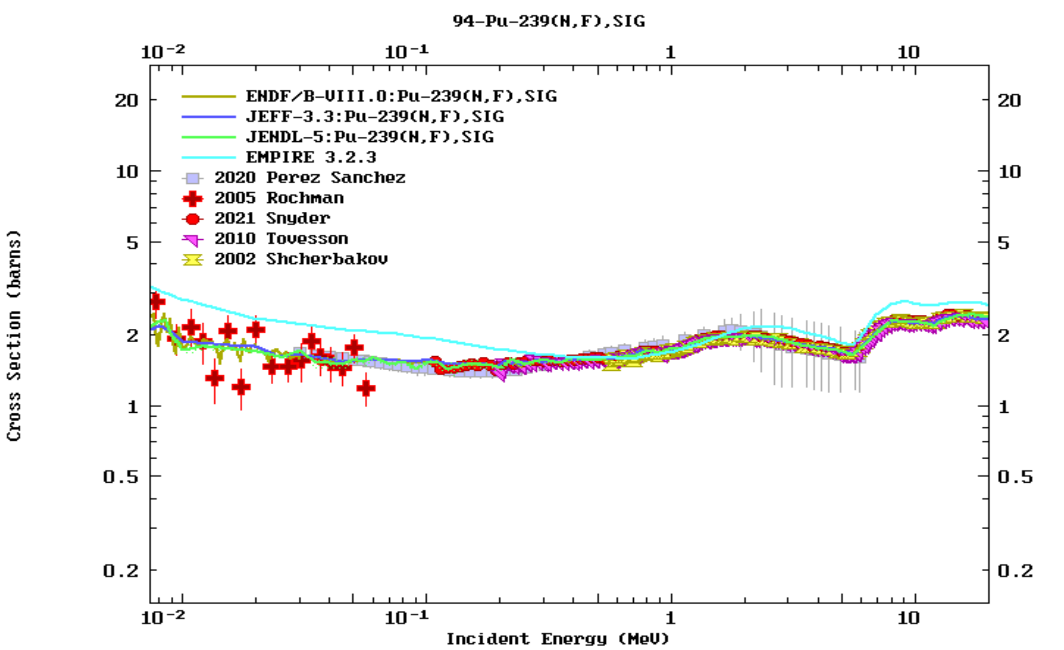

Figure 7: Result from EMPIRE 3.2.3 code in comparison with available data from EXFOR and existing Evaluated Nuclear Data Library

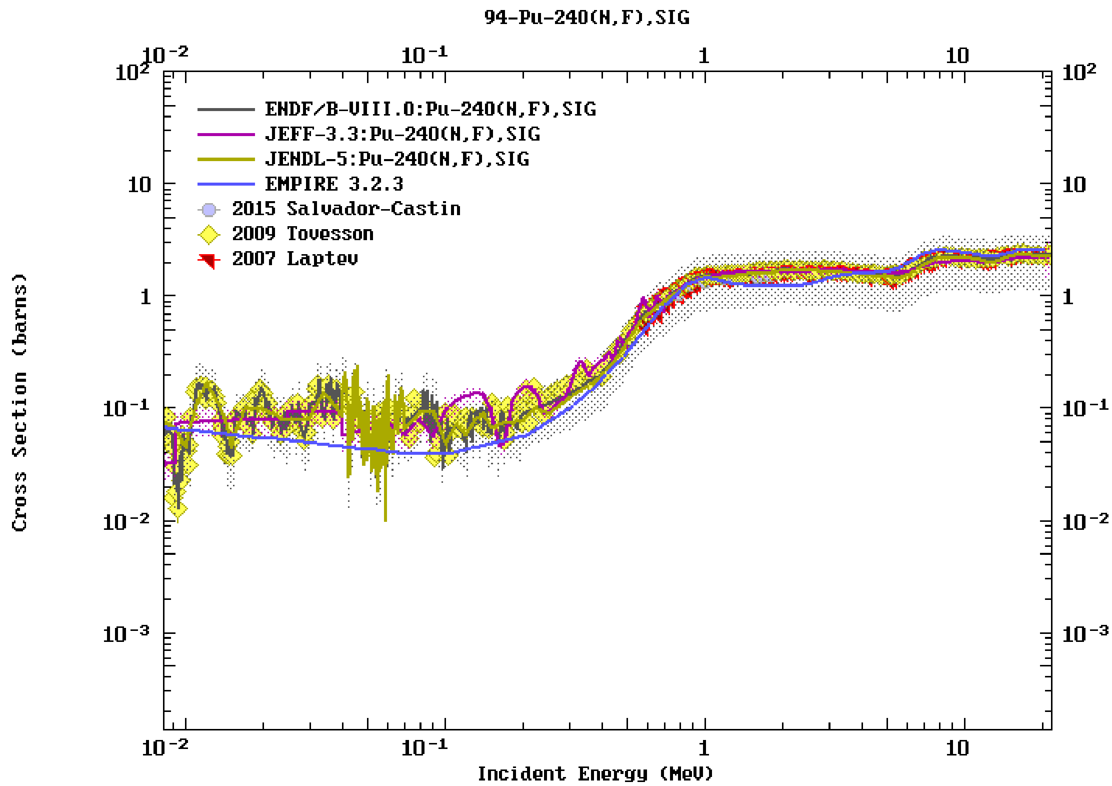

Figure 8: Result from EMPIRE 3.2.3 code in comparison with available data from EXFOR and existing Evaluated Nuclear Data Library

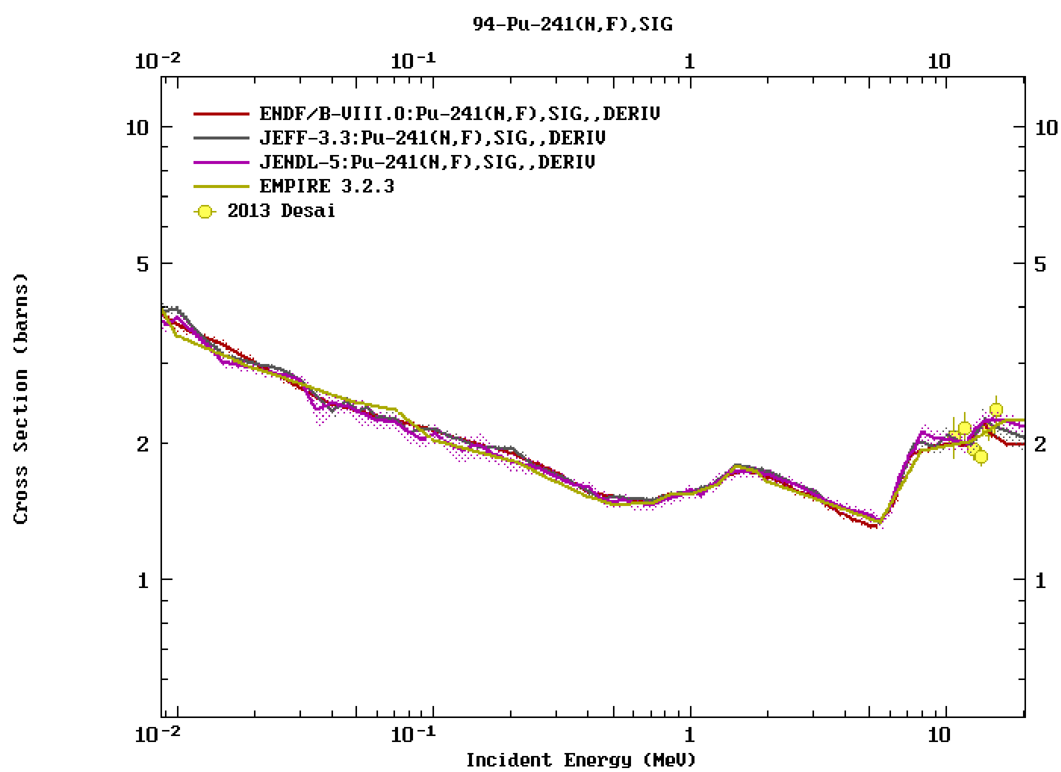

Figure 9: Result from EMPIRE 3.2.3 code in comparison with available data from EXFOR and existing Evaluated Nuclear Data Library

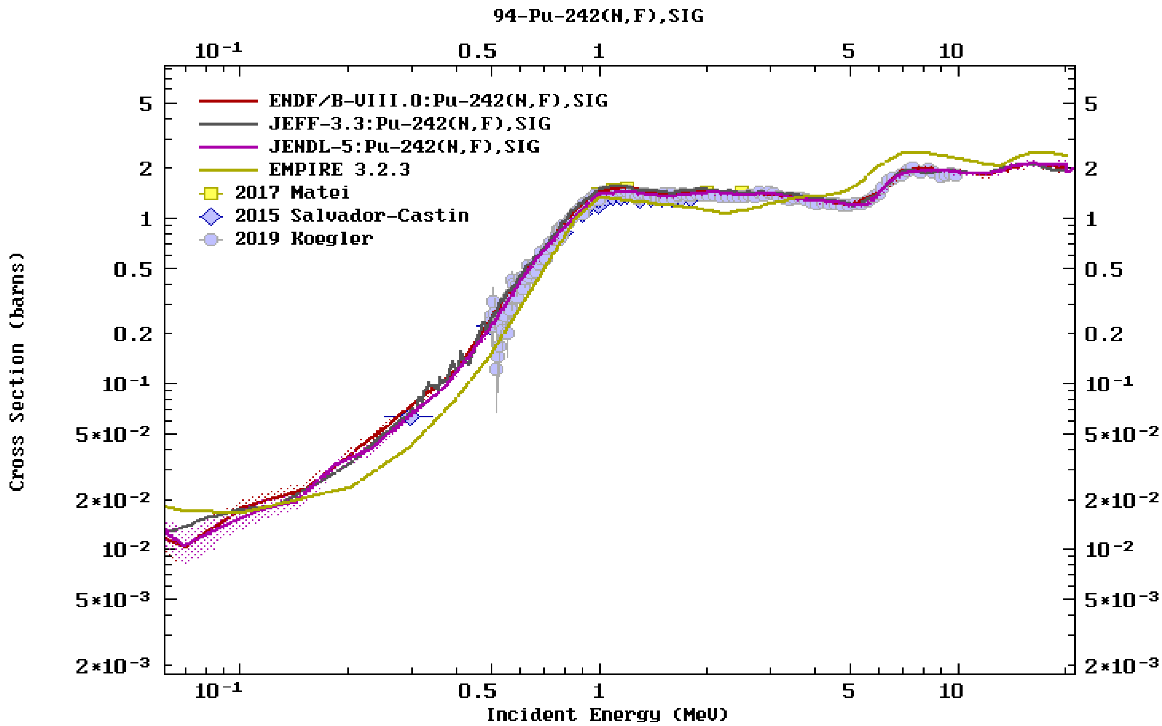

Figure 10: Figure 9: Result from EMPIRE 3.2.3 code in comparison with available data from EXFOR and existing Evaluated Nuclear Data Library

EMPIRE reaction results on $^{239}Pu\ (n,f)$ show a similar trend with existing evaluated data from and also, in good agreement with the recent experimental data from Perez, et al., (2020); Rochman et al., (2005); Synder, et al., (2021); Tovesson and Hill (2010). Shcherbakov et al., (2002). We observed deviation of 3.2 % above existing evaluated nuclear data file at $E_n \leq\ 0.5\ MeV$. Too high contribution from direct reactions may likely be reason.

Figure 7 display reaction cross section on $^{240}Pu\ (n,f)$ against incident energy. The graph shows a good agreement between EMPIRE results and experimental data from Salvador et al. (2015), Tovesson *et al*. (2010) and Laptev *et al*. (2007). It also compares well with existing evaluated data from incident neutron energy $E_n \geq\ 1\ MeV$ and deviate 1.3 % below $E_n =\ 1\ MeV$

The EMPIRE result of reaction cross section on $^{241}Pu\ (n,f)$ shows a reasonable agreement with existing evaluated nuclear data file (Brown *et al.*, 2018; Iwamoto *et al.*, 2021; Plompen et al., 2020) and experimental data from Desai (2013) throughout the whole energy range as shown in figure 9. The inelastic scatting is predominant which have both direct and compound nucleus contribution to the discrete level to the discrete states.

Figure 10 show the cross section on $^{242}Pu\ (n,f)$ against incident energy. EMPIRE result reproduced experiment data from Matoi et al. (2017), Salvador- castin et al. (2015) and Koegler et al. (2019). Deviation of 3 % occur below $E_n \leq\ 0.5\ MeV$ and 2.18% above $E_n \geq\ 4.5\ MeV$ between our results and existing evaluated nuclear data (Brown *et al.*, 2018; Iwamoto et al., 2021; Plompen *et al.*, 2020). The reason may be too high of pre-equilibrium contribution.

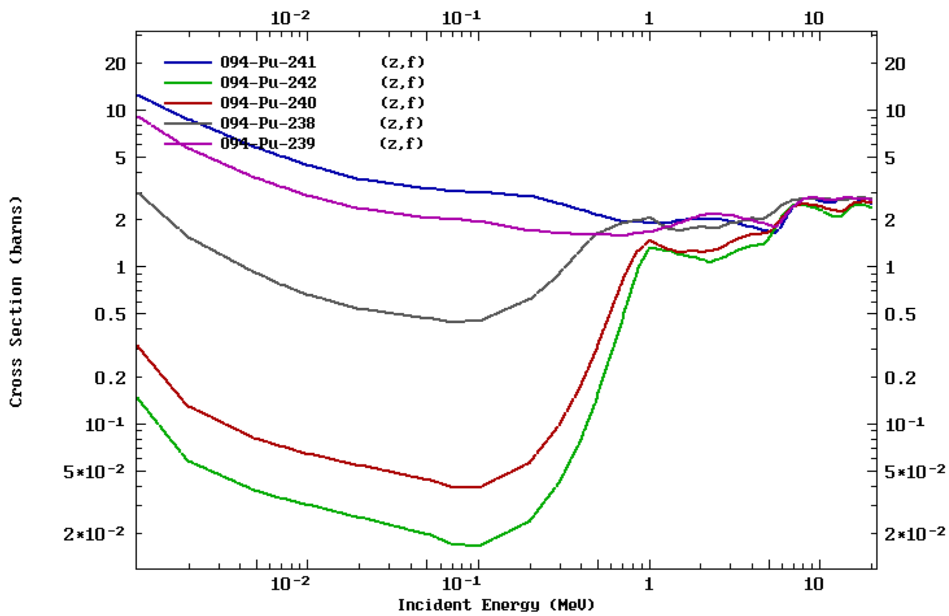

Figure 11: Comparison of results from EMPIRE 3.2.3 code on neutron induced fission cross section on $^{238}Pu\ (n,f)$, $^{239}Pu\ (n,f)$, $^{240}Pu\ (n,f)$, $^{241}Pu\ (n,f)$, $^{242}Pu\ (n,f)$

The trends of the results for reaction cross section on neutron induced fission on plutonium $^{238}Pu\ (n,f)$, $^{239}Pu\ (n,f)$, $^{240}Pu\ (n,f)$, $^{241}Pu\ (n,f)$, $^{242}Pu\ (n,f)$ is displayed in figure 11 above. The odd – A $^{239}Pu(n,f)$ and $^{241}Pu(n,f)$, cross section was seen to be flatten within $E_n\ =\ 10\ keV\ - 5\ MeV$, this may be due to the competing of inelastic channel while fissile character is displayed within the $E_n\ =\ 0.5 -\ 1\ MeV$. The similar behavior was observed in the report of (Sin, *et al.*, 2017)

Even- even target isotopes of plutonium Pu have the second maximum lower than the odd- A plutonium target owing to the odd-even effect as clearly seen in the figure 11. ZVView graphics software Zerkin (2001) were employed for all the plotting.

6.0 Conclusion

In this work, Optical Model Parameters were used to improve neutron induced total reaction cross section on $^{238}Pu\ (n,tot)$, $^{239}Pu\ (n,tot)$, $^{240}Pu\ (n,tot)$, $^{241}Pu\ (n,tot)$, $^{242}Pu\ (n,tot)$. Couple channels were generated for neutron transmission coefficient for strongly deformed nuclei. We use Enhanced Generalized Super Fluid Model (EGSM) model in its phenomenology mode including all EMPIRE input parameters for fission in defaults. Most of the contribution comes from direct, pre-equilibrium and compound nucleus reaction. For incident energy $E_n \leq 5\ MeV$ inelastic channel is first open and the predominant reaction to discrete levels have respectively compound and a direct reaction described by Distorted Wave Born Approximation (DWBA) and couple channel for deformed nuclei. At higher energies $E_n\ > 5\ MeV$ the compound nucleus is predominated and a direct like which is described with pre-equilibrium reaction to continuum. At about $E_n = 14.5\ MeV$, for the excitation to discrete state the direct component become predominant and fission reaction is possible for non -fissile nuclei. EMPIRE results were able to reproduce available experimental data and prediction was made at energy region where the experimental data is scarce. But there is need for improvement of model and parameters at $E_n \leq 0.5\ MeV$ for fission reaction cross section.

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