# Evaluation of Nuclear Reaction Cross Section of Some Isotopes of Plutonium at Energy Range 10-20 MeV Using OPTMAN Code

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**ABSTRACT**

*Coupled-channels optical model code OPTMAN is used as an alternative to experimental approach to evaluate the total reaction cross section for four different isotopes of Plutonium as an example of heavy rotational nuclei of the transuranium elements over an energy range of 10 to 20 MeV. The selected isotopes are the* $_{94}Pu^{238}$, $_{94}Pu^{240}$, $_{94}Pu^{241}$, *and* $_{94}Pu^{242}$. *Their choice is as a result of their importance in the modern day nuclear reactor and the energy range 10 – 20 MeV is the energy range of neutron produced in neutron generators and the maximum energy possessed by neutrons which are born in fission reaction. Results show that the percentage deviation of total cross section from ENDF values obtained for this work is less than 1 % at energy 16 MeV and above for* $_{94}Pu^{238}$, $_{94}Pu^{240}$, *and* $_{94}Pu^{241}$ *while at 18 MeV and above for* $_{94}Pu^{242}$. *This work confirms that the use of the rigid rotator is no longer a preferable approach in precise evaluations of nuclear reaction cross sections of heavy isotopes. It is observed that the nucleus of plutonium isotope is symmetric and the activities of rotation and vibrations (β – quadrupole, octupole vibrations and γ – quadrupole vibrations) cannot be ignored. This work compared well with the 6 % deviation of Basunia in 2009 and 5 % deviation of Paradela in 2011 using indirect measurement based on the surrogate ratio method and ECIS code respectively. The result of this work is found to agree with about 1 % increase in accuracy.*

## INTRODUCTION

The study of nuclear reactions is very fundamental in the field of nuclear physics. A nuclear reaction is initiated when a nucleon or nucleus collides with another nucleon or nucleus. It is a process in which two nuclei or a nucleus of an atom and a subatomic particle from outside the atom collide to produce one or more nuclides that are different from the nuclides that began the process. The knowledge of reaction cross section is therefore indispensable in the accurate determination and analysis of reaction rate. Neutron energies could be as high as 14 MeV in neutron generators and fast fission reactors, therefore in order to determine the irradiation damage to the reactor first wall candidate materials owing from neutrons. The knowledge of $(n, \gamma)$ cross section is also important to establish the gamma flux in a reactor[1]

Nuclear reaction cross sections are generally obtained either by experimental measurement or by mathematical computation. Literature affirms that most available nuclear data for nuclides of the minor actinides do not fulfill the current accuracy requirements[2]. There is therefore a need to embark on the search for a more improved nuclear data for transuranium nuclides. This work obtains cross section values that are more accurate than the existing nuclear data by the use of coupled channel computer code OPTMAN to evaluate the reaction cross sections of some isotopes of plutonium nuclides at energies between 10 and 20 MeV which constitutes a major field of nuclear physics since 10 – 20 MeV is the energy range of neutron produced in neutron generators and it is the maximum energy possessed by neutrons which are born in fission reaction.

## MATERIALS AND METHODS

The use of the coupled channel optical model code OPTMAN has been employed to obtain the total reaction cross section for the isotopes of some transuranium elements under consideration. The choice of OPTMAN code is as a result of its capability to work on optical model which has the capability to work on wide energy range of 5 – 300 MeV[3] and nuclides of heavy atoms such as the plutonium elements which is the focus of this research effort. The OPTMAN code is an off-shelve programme which works on Disc Operating System (DOS). The code consists of the main program ROTAT, 44 subroutines, 2 functions and a block data. It provided five results considerations, viz;

$D_1$ - nuclides of transuranium elements are considered to be rigid to octupole deformation which implies that the particles in the nucleus are assumed to be tightly bonded together hence has rigid shape.

$D_2$ -nuclides are soft to symmetric octupole deformations scaled by $\beta_2$ on account of relativistic kinematic only[4].

$D_3$ - the nucleus is soft to symmetric octupole deformations scaled by $\beta_2$ and on account of relativistic kinematics and potential dependence.

$D_4$ - assumes that the nucleus is soft to non symmetric octupole deformations.

$D_5$ - assumes that the nucleus is soft to symmetric octupole deformations not scaled by $\beta_2$.

The input is characterized with 14 variables titled as MEJOB MEPOT MEHAM MEPRI MESOL MESHA MESHO MEHAO MEAPP MEVOL MEREL MECUL MERZZ MERRR.

The eighth entry of the input (MEHAO) enables the programme to make a choice of what to calculate between $D_1$, $D_2$, $D_3$, $D_4$, and $D_5$. The five inputs $D_1$ – $D_5$ are thus displayed

i) **$D_1$ at 10 – 20 MeV**

02 02 05 00 01 04 02 00 00 00 01 00 01 01

The inputs of $D_1$ express an assumption that the nucleus is rigid to octupole deformations.

Note: The bold **00,** the eighth entry, the MEHAO has its value determines the particular information required to be obtained.

ii) **$D_2$ at 10 – 20 MeV**

02 02 05 00 01 04 02 02 00 00 02 03 01 01

The inputs of $D_2$ is at an assumption that the nucleus is soft to symmetric octupole deformations scaled by $\beta_2$ and on account of relativistic kinematic only[4].

iii) **$D_3$ at 10 – 20 MeV**

02 02 05 00 01 04 02 02 00 00 01 00 01 01

The input entry of $D_3$ is at an assumption that the nucleus is soft to symmetric octupole deformations scaled by $\beta_2$ and on account of relativistic kinematics and potential dependence.

iv) **$D_4$ at 10 – 20 MeV**

02 02 05 00 01 04 02 01 00 00 01 00 01 01

The input entry of $D_4$ is at an assumption that the nucleus is soft to non symmetric octupole deformations.

v) **$D_5$ at 10 – 20 MeV**

02 02 05 00 01 04 02 01 00 00 01 00 01 01

The input entry of $D_5$ is at an assumption that the nucleus is soft to symmetric octupole deformations not scaled by $\beta_2$.

## RESULTS AND DISCUSSION

The results obtained for the total reaction cross sections for the four isotopes of Plutonium elements relevant in the design and operation of new generation nuclear reactors considered for study in this work is presented and carefully discussed as follows:

### *1. Total Cross Section against Energy for $_{94}Pu^{238}$*

Figure 1 displays the graph of total cross section against energy for $_{94}Pu^{238}$. The OPTMAN code is used to obtain the total reaction cross section which is now being plotted against energy from 10 to 20 MeV. Among all the readings, only $D_1$, $D_2$ and $D_5$ (Rigid rotator) actually have values plotted against energy.

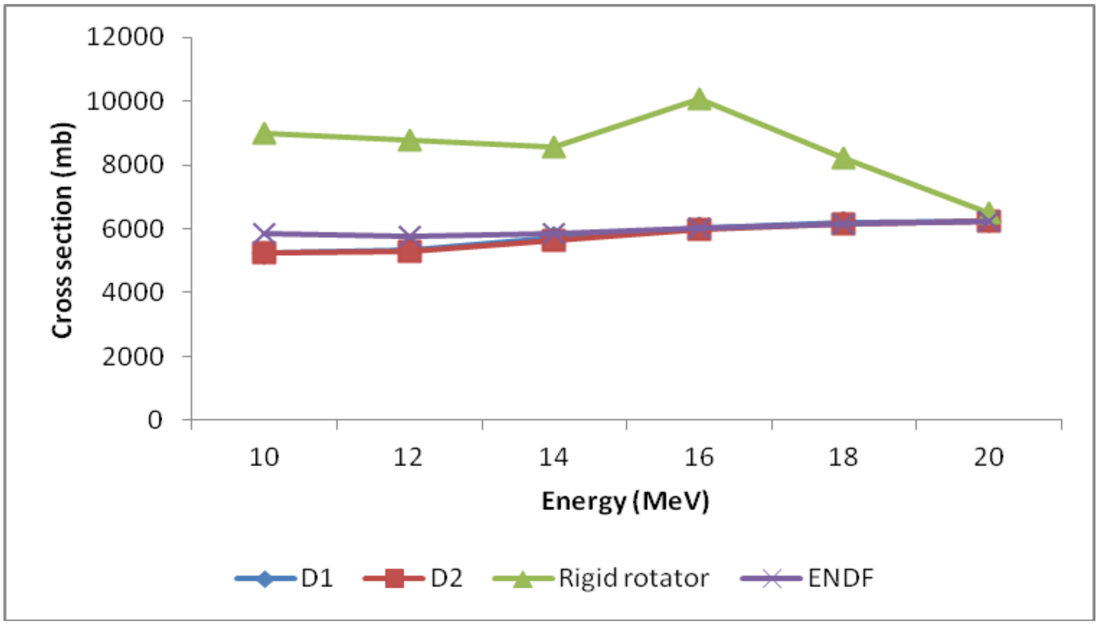


**Figure 1 Graph of Total Cross section against Energy for $_{94}Pu^{238}$**

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The plots shows $D_1$ and $D_2$ with gentle positive slope coincide at between energy 10 and 12 MeV. The deviation level between $D_1$ and $D_2$ and ENDF value is estimated to be about 10 % while it begins to gradually decrease as energy increases from 12 to 14 MeV until deviation value of less than 1 was recorded at energy 16 MeV and above which implies that the collective characteristics hardly affect cross sections at energy 16 MeV and above for $^{238}Pu$ isotope. The deviation of cross section value is 54 % for $^{238}Pu$ using the RRM-CC method which gradually reduces to 46 % at energy 14 MeV however deviation rose to 68 % at 16 MeV and thereafter began to decrease until finally to 4 % deviation at energy 20 MeV. The results obtained from RRM-CC did not follow a particular trend. Soft rotator model however records average deviation of 4 % while the Rigid rotator model records average deviation of 43 %. The use of the RRM-CC is no longer a preferable approach in precise evaluations of nuclear reaction cross sections of heavy isotopes [5]

### *2. Total Cross Section against Energy for $_{94}Pu^{240}$*

The graph of plots of $D_1$, $D_2$ against energy in comparison with ENDF values for $_{94}Pu^{240}$ using the coupled channel code OPTMAN.

The energy range is from 10 to 20 MeV. Just like the $_{94}Pu^{238}$ earlier considered, only $D_1$, $D_2$ and $D_5$ (Rigid rotator) have results which are being plotted as displayed in Figure 3

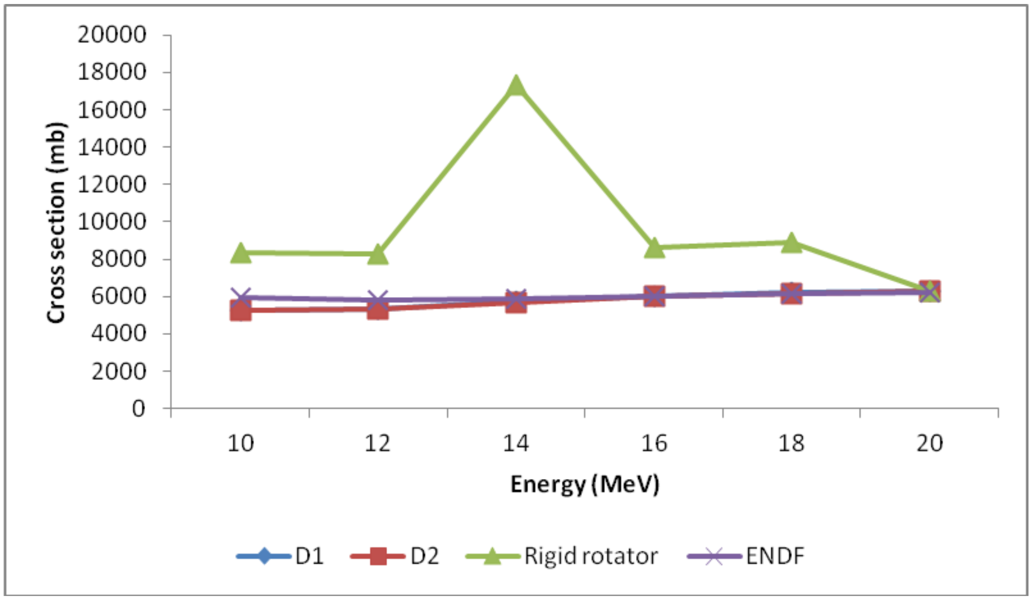


**Figure 2 Graph of Total Cross section against Energy for $_{94}Pu^{240}$**

The plots show $D_1$ and $D_2$ displaying gentle slope which coincide at between energy 10 and 12 MeV. The slope however picks a slightly higher value at energy 12 to 14 MeV than at energy 10 to 12 MeV. At energy range 10 – 12 MeV, the deviation level between $D_1$, $D_2$ and ENDF values is 11 % which gradually collapse to less than 1 % at 16 MeV energy and above which implies that the collective characteristics hardly affect cross sections at energy 16 MeV and above for $_{94}Pu^{240}$ just like $_{94}Pu^{238}$ isotope. The Rigid rotator model coupled channel records 41 % deviation at energy 10-12 MeV and sharply increased to 194.39 % at energy 14 MeV but quickly falls back to 43.16 at 16 to 18 MeV energy and sharply dropped to less than 1 % at 20 MeV showing convergence. Soft rotator model records average deviation of 4 % and the rigid rotator model records average deviation of 61 %. The Rigid rotator model is no longer a preferable approach in precise evaluations of nuclear reaction cross section of heavy isotopes [5].

### *3 Total Cross Section against Energy for $_{94}Pu^{241}$*

The plots of $D_1$, $D_2$, $D_5$ and ENDF values for $_{94}Pu^{241}$ are displayed in Figure 4 showing the comparison between values of total reaction cross section obtained for $D_1$, $D_2$ and $D_5$ using the coupled channel code OPTMAN for energy range between 10 and 20 MeV.

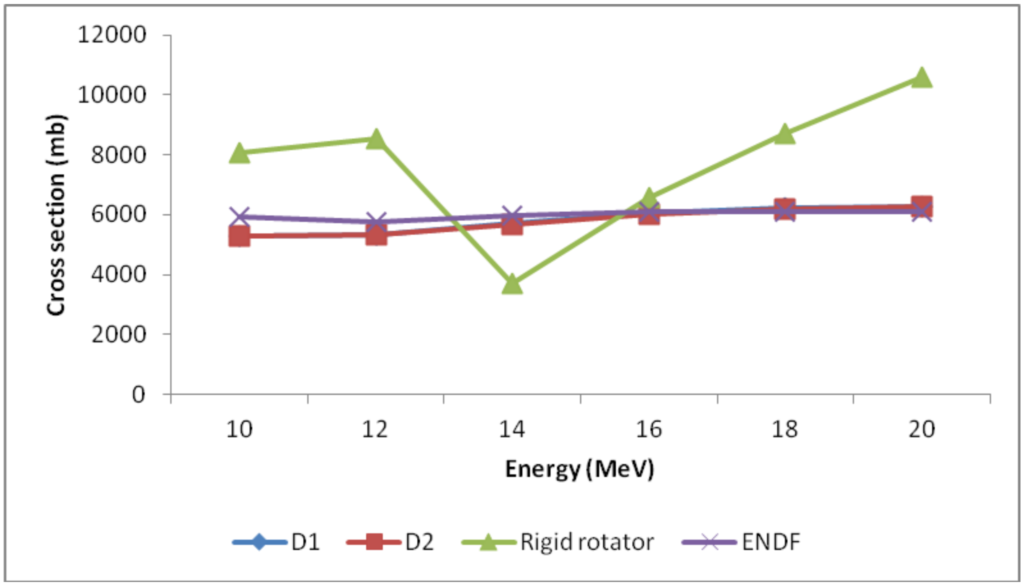


**Figure 3 Total Cross section against Energy for $_{94}Pu^{241}$**

Only $D_1$ and $D_2$ have results to show as it is the case in $_{94}Pu^{238}$ and $_{94}Pu^{240}$. The plots show a quite gentle positive slope from 10 to 12 MeV energy. The slope however becomes sharper at between 12 to 14 MeV energy such that the percentage deviation of about 11 % which is observed for $D_1$ and $D_2$ results when compared with ENDF values at energy 10 to 12 MeV, suddenly changed to about 7 % at energy 12 to 14 MeV and progressively reduced to 0.88 % at 16 MeV and above, it can therefore be inferred that the collective characteristics hardly affect cross sections at energy 16 MeV and above for $_{94}Pu^{241}$ similar to $_{94}Pu^{240}$ and $_{94}Pu^{238}$ isotopes. The rigid rotator model coupled channel records a cross section of 8076.941 mb at 10 MeV with percentage deviation of 36 % and increased to 8522.6 mb at 12 MeV with percentage deviation of 48 %. These values promptly reduced to cross section

value of 3694.6 mb with percentage deviation of 38 % and steadily continue to rise to 10603.632 mb at energy 20 MeV with maximum deviation of 73.8 %. The Soft rotator model records 5 % average deviation while the rigid rotator model records average deviation of 41 %. Result displayed confirms that the rigid rotator model coupled channel is no longer a preferable approach in precise evaluation of nuclear reaction cross sections of heavy isotopes [5].

### 4. *Total Cross Section against Energy for $_{94}Pu^{242}$*

The plots of $D_1$, $D_2$ and ENDF values for $_{94}Pu^{242}$ displaying the graphical comparison between values of total reaction cross section obtained for $D_1$ and $D_2$ using the coupled channel code OPTMAN for energy between 10 and 20 MeV is presented in Figure 5

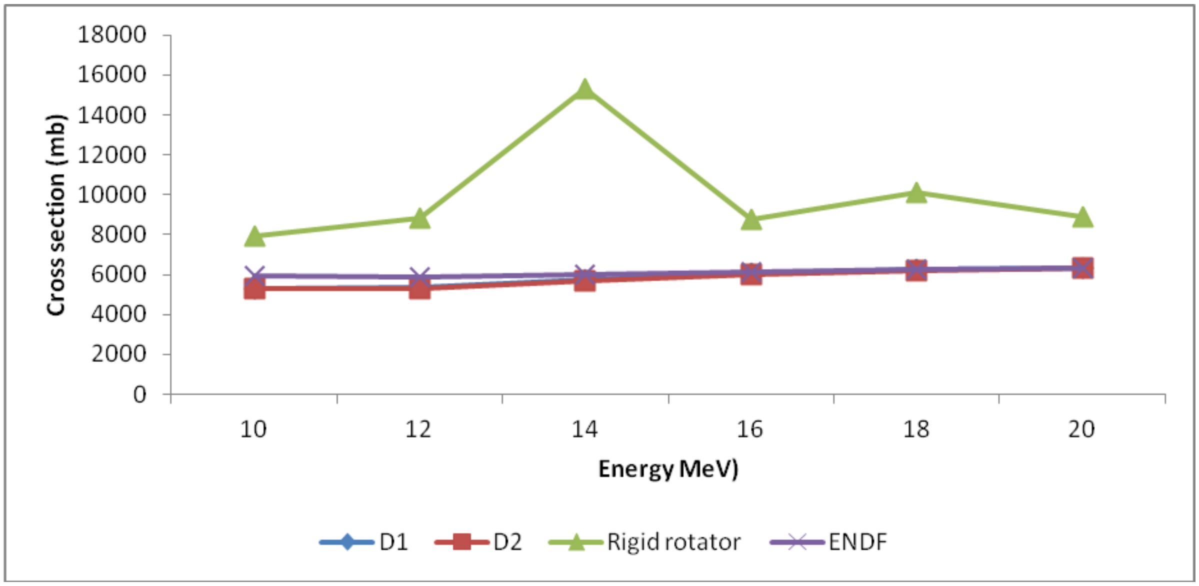


**Figure 4 Total Cross section against Energy for $_{94}Pu^{242}$**

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Only $D_1$ and $D_2$ have results for $_{94}Pu^{242}$ as it is for other isotopes in this work. The plots produce a graph with a gentle positive slope from 10 to 12 MeV energy and percentage deviation of 11 %. The slope becomes sharper at between 12 and 14 MeV thereby reducing the percentage deviation to 9 % and thereafter progressively reduced to 4 % at 14 MeV and less than 1 % at 16 MeV and above. The implication therefore is that the collective characteristics hardly affect cross sections at energy 16 MeV and above for $_{94}Pu^{242}$ unlike isotopes earlier discussed which displayed this characteristics at energy 16 MeV.

The rigid rotator model coupled channel records total reaction cross section of about 8000 mb at energy 10 MeV which increases to 8846 mb at energy 12 MeV. The rigid rotator model records its peak of 15320 mb at 14 MeV and suddenly decreases to 8744 mb at energy 16 MeV. The value rise to 10000 mb at 18 MeV and drops to 8894 mb at 20 MeV. The Soft rotator model records average deviation of about 5 % for ${}_{94}Pu^{242}$ while the rigid rotator model records an average deviation of 64 %. Results displayed confirms that the rigid rotator model is no longer a preferable approach in precise evaluations of nuclear reaction cross sections of heavy isotopes [5]. The soft rotator model coupled channel results agree with measured cross sections better than the rigid rotator coupled channel calculations.

Nuclear - induced optical model analyses have been carried out for various mediums and heavy nuclei over an energy range from 10 to 20 MeV. The rigid rotator model coupled channel allowed us to describe global tendencies of optical potential by simple functional forms and the systematical parameters. The systematic optical potential plus the soft rotator model coupled channel calculations showed better results than those of the rigid rotator coupled channel. The good results of the soft rotator model coupled channel are substantially due to the consideration of non-axial deformation which is obtained by the nuclear level structure analysis. Moreover, it was also found that there exists a clear correlation between the quadruple elasticity parameter and ground state deformation. This means that the values of the ground state deformation are likely to be predicted by the results of soft rotator model nuclear level structure analyses. It is therefore confirm that the soft rotator model coupled channel is a useful approach to give more elaborate evaluations of nuclear data.

The coupled channel optical model code OPTMAN has been used to determine the total cross section, reaction cross section, total direct cross section and scattering radius of eight isotopes of transuranium elements at energies 10 to 20 MeV. The need for this work is as a result of consistent demand for high quality data as a result of the present research directories concern strategies which is an extension of the life span of presently operating reactors, the increase of the fuel burn-up, the plutonium recycling and in particular, the incineration of actinides and long-lived fission products, the accelerator driven systems (ADS). The detailed feasibility study and safety assessment of these strategies requires the accurate knowledge of neutron nuclear reaction data. Both higher fuel burn-up and especially waste incineration options require improved and new data on actinides. Most available nuclear data for nuclides of the minor actinides (MA) do not fulfill the current accuracy requirements[6] hence the need for this work. To search for a more improved nuclear data for transuranium nuclides.

Development of nuclear energy programs around the world are presently closely related to the issue of nuclear waste treatment and storage due to the fact that a significant fraction of the high-level waste is constituted by minor actinides, in particular Plutonium (Pu), built up as a result of multiple neutron captures and radioactive decays in current nuclear reactors. Transmutation in critical, such as Gen-IV, or subcritical (ADS) systems could be a solution, in order to reduce calculation uncertainties in the design and operation of new generation reactors, high precision data on neutron induced fission cross-sections from thermal neutron energies up to several tens of MeV are required for a variety of transuranic elements.

A pressing need arise for Plutonium Pu isotopes in particular, for which the available data are scarce and show large discrepancies [7]. The new reactor concepts extend the nuclear data requirements in the fast neutron region where a more precise knowledge of most of the actinides cross sections is demanded [8]. The coupled channel optical model code OPTMAN has been used to obtain total reaction cross section for four isotopes of Plutonium as key family members of transuranium elements under study.

The following results are noted from this research effort;

i) The nucleus of Plutonium elements is symmetric
ii) The activities of rotation ($\beta$-quadrupole, octupole vibrations and $\gamma$-quadrupole vibrations) cannot be ignored. They are activities happening in the nuclides of heavy atoms such as Plutonium elements.
iii) The nucleus of Plutonium elements is with a non spherical equilibrium shape with substantial distortions from spherical shape and can therefore be referred to as deformed nuclei.
iv) The collective characteristics hardly affect cross sections in the energy range of 16 MeV and above for $^{238}$Pu, $^{240}$Pu, $^{241}$Pu and 18 MeV and above for $^{242}$Pu, which implies that the effects of collective characteristics are generally less for isotopes of Plutonium elements with lower atomic number and atomic mass. However the cross section strongly depends on the collective characteristics in the low energy region.

v) Rigid rotator model records a higher deviation of an average deviation of 46 %, soft rotator model records 4 % better than 5-6 % of Paradela [8] and Basunia [9].

vi) It is confirmed that the use of the rigid rotator coupled channel is no longer a preferable approach in precise evaluations of nuclear reaction cross sections of heavy isotopes.

vii) The soft rotator model coupled channel results agree with ENDF values better than the rigid rotator model evaluations.

viii) Findings in this work are compared with the works of other researchers and it is found to agree with about 1 % increase in accuracy.

## CONCLUSION S

With a declining number of nuclear data evaluators in the world and an increasing demand for high-quality data, there is a risk that evaluators will concentrate on producing new nuclear data to the detriment of developing new models and methods for evaluating existing data. It is essential to identify the basic physics issues that are going to be important for future nuclear data evaluation processes. At the same time, demand for new types of data, which will be needed in emerging nuclear applications, could warrant new evaluation techniques [10]. This challenge has brought about the application of the coupled-channels optical model code OPTMAN based on soft-rotator model nuclear Hamiltonian wave functions to evaluate the optical cross sections (the total, elastic and absorption cross sections), of the three optical cross sections, only the total can be compared directly with experimental data, as it is the only one that is linear in the scattering amplitude [1].

It is essential to identify the basic physics issues that are going to be important for future nuclear data evaluation processes. At the same time, demand for new types of data, which will be needed in emerging nuclear applications, could warrant new evaluation techniques that are presently only used in the context of fundamental research[10]. The quest for optical data for nucleon induced reactions (e.g. total, elastic, and inelastic scattering, nucleon transmissions and so on) on experimentally uninvestigated minor actinide nuclei, which are crucially important for design of various advanced reactor and nuclear waste transmutation systems [11]. Trans-Uranium elements (TRU) such as the Pu are built up as a result of multiple neutron captures and radioactive decays in the presently operating nuclear reactors based on the U/Pu nuclear fuel cycle. Some highly active isotopes of Plutonium elements constitute the most important hazard for nuclear waste management.

Recently, several proposals have been made to reduce the radio toxicity of nuclear waste containing Plutonium as a key member of transuranium elements. They all rely on neutron induced capture and fission of the transuranium. It is clear that any kind of waste burner system, critical or sub-critical, thermal or fast, will need to be loaded with fuel containing a large fraction of TRU. The response of these systems (e.g. criticality conditions) to the presence of TRU is directly linked to the fission and capture cross sections of the mentioned TRU isotopes. The fission cross sections of TRU are therefore fundamental elements in assessing the strategy to be followed in detailed feasibility studies of nuclear-waste transmutation. The n_TOF collaboration has proposed to perform neutron-induced fission cross-section measurements using the CERN n_TOF facility. The unsatisfactory situation of nuclear data in the presently available libraries could be substantially improved by the use of coupled-channels optical model code OPTMAN based on soft-rotator model nuclear Hamiltonian wave functions.

In some advanced fuel-cycle scenarios, the contribution of plutonium to the total fission rate can be as large as 10 % [12] and the fraction of sub-threshold fission typically of 5 % even in a fast spectrum. Therefore, both below and above threshold, plutonium isotopes represent important contributions to the global reactor neutron balance. The fission cross sections of the isotopes in consideration in this work play an important role on the precise definition of the isotopic content of the transmutation Accelerated Driven System (ADS) discharged fuel. Combining the basic motivations with the availability of target materials, it is hereby proposed to determine the neutron-induced fission cross sections of the five experimentally accessible isotopes ${}_{94}Pu^{238}$, ${}_{94}Pu^{240}$, ${}_{94}Pu^{241}$, and ${}_{94}Pu^{242}$. It is required to perform these evaluations with coupled-channels optical model code OPTMAN based on soft-rotator model nuclear Hamiltonian wave functions. The main features of the new generation reactors and the need of accurate nuclear data rest mostly on cross-sections for neutron-induced reactions [13]. For transuranic elements Plutonium (Pu), neutron-induced fission is in principle a much more efficient process, since it leads also to energy gain and a surplus of neutrons[13]. In order to reduce uncertainties in the design and operation of new generation reactors, high precision data on the cross-section for neutron-induced reactions on a variety of isotopes are required, from thermal energy to several tens of MeV. Neutron cross-sections are available by means of compilations of experimental data or through evaluated nuclear data libraries[14], such as ENDF(mostly compiled in the united states), JEFF (the European library maintained by the NEA), JENDL (the Japanese database) and BROND (the Russian one). Together with the evaluated cross-section, these libraries include information on the reliability of the data, which is mostly linked to the accuracy of the available experimental results. The present knowledge of the neutron cross-section results largely inadequate for the new applications in the field of emerging nuclear technologies, and they need to be updated with new experimental or theoretical information.

Sensitivity students performed by means of Monte Carlo simulations indicate that for most long-lived fission fragments and minor actinides, the present uncertainties are much larger than needed for the reliable design and safe operation of new generation reactors. There has been an evident need of improving the current knowledge of nuclear data [15]. Nucleon-induced optical model analyses have been carried out for various medium and heavy nuclei over an energy range from 10 MeV to 20 MeV. The RRM-CC allows the description of global tendencies of optical potential by simple functional forms and the systematical parameters. Then the SRM-CC is applied to the analyses for various transuranium nuclei. The systematic optical potential plus the SRM-CC calculations showed better results than those of the RRM-CC. The good results of the SRM-CC are substantially due to the consideration of non-axial deformation which is obtained by the nuclear level structure analysis. Moreover, it was also found that there exists a clear correlation between the quadrupole elasticity parameter and ground state deformation. This means that the values of the ground state deformation are likely to be guessed by the results of SRM nuclear level structure analyses.

It is confirmed that the SRM-CC is a useful approach to give more elaborate evaluations of nuclear data. The soft-rotator nuclear model and coupled-channels (CC) method with a coupling based on the wave functions of the soft-rotator nuclear Hamiltonian was applied to analyze simultaneously transuranium isotopes under consideration in this work for total cross section. It was found that the model could describe the basic observables i.e available experimental nuclear structure and nucleon interaction data from 10 MeV region up to 20 MeV incident energies for plutonium isotopes fairly successfully in such a uniform approach. This work showed that the approach employed here can be an extremely powerful tool for evaluation of high-energy nuclear data because the coupled-channel method based on the soft-rotator model can supplement poor number of neutron experimental data by the information obtained from the nuclear structure and proton experimental data, which are normally much more abundant and easy to be obtained. It is found that this model was flexible enough so that OPTMAN can be applied not only to a very light nucleus but can be applied very successfully to heavy (actinide) nuclei and also medium weight vibrational nuclei.

## Recommendations for Further Work

To use computer codes requires a certain craftsmanship that can only be acquired with experience. It is therefore recommended that further work be done for other isotopes of the transuranium atomic mass to study the validity of the code at that level with a view to optimizing the potential parameters, the right choice of reduced radius *r* and diffusivity *a* which give values that are in agreement with ENDF values considered as our reference values.

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