

DETERMINATION OF THE CROSS SECTION DATA FOR THE TRANSMUTATION OF LONG-LIVED FISSION PRODUCTS THROUGH NEUTRON CAPTURE USING EMPIRE 3.2 CODE

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Abstract

Transmutation is one of the preferred path way of dealing with LLFP in fission reactors therefore, in this work, the calculation of neutron capture reaction on some LLFPs which are problematic in radioactive waste management in the neutron energy range of 0 – 20 MeV where performed using the EMPIRE 3.2 Code. The EMPIRE Code is a hybrid of several codes for addressing different reaction mechanisms of particle-induced reactions. Results of neutron capture reaction cross sections on LLFPs determined using the EMPIRE Code are compared with experimental data were available to select the most appropriate model for predicting the neutron capture reaction cross sections on the LLFPs that are scanty and discrepant. The results of the calculated cross sections are in good agreement with the measurements from EXFOR data of IAEA. The comparison of the calculated and the measured cross sections indicates that the models used in the calculation are suitable for practical applications.

Keywords: Cross section, Transmutation, Long-Lived Fission Products, Neutron Capture.

1. INTRODUCTION

The need for the transmutation of Long-Lived Fission Products (LLFPs) cannot be over emphasized considering the health implication of the uncontrolled exposure to the radiations. As of today, there is no set of consistent measured or evaluated cross section data that covers all the LLFPs which are problematic radioactive waste in spent nuclear fuel. Therefore evaluation of a set of consistent, accurate and precise cross section data, based on sound physics, is required for the transmutation of these isotopes for safe management and disposal. In nuclear transmutation technology, various kinds of particle beams from accelerators and neutron flux in nuclear reactors are used. The set of evaluated data thus have the important advantage of providing insight into the important physical considerations for these isotopes. For all transmutation studies a large number of additional nuclear data, which have not yet been available, is needed [1].

Of the possible nuclear reactions in the reactor, fission is the best in terms of energy production (about 200 MeV) and sustainable number of neutrons. The second best alternative is capture. It also releases energy, but much less (about 8 MeV) and photons which do not induce chain reactions. Thus capture is the choice only for elements which cannot fission, like the rest products after fissions, e.g technetium or iodine. Neutron Capture reaction on LLFPs results in nuclei that are often stable or short lived decaying into stable nuclei. The models used are presented in section two. The result of the neutron capture calculated with neutron energy up to 20 MeV using EMPIRE 3.2 code is presented in section 3.

2. MATERIALS AND METHOD

The backbones of the EMPIRE system are bash-shell UNIX scripts that provide for seamless console operation of EMPIRE on Linux, Mac OS X, and Microsoft Windows with GNU gfortran compiler installed. The graphical interface provides for an easy operation of the system on Linux, Mac OS X and virtual Linux machines running on Microsoft Windows.

For this work, the EMPIRE 3.2 (Malta) was installed on a Fedora Operating System on an Acer Laptop with Intel CORE i3-

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380M processor - 2.53 GHz, 3MB L3 cache, 4 GB DDR3 Memory and 320 GB Hard Disk Drive. The PC was partitioned and WINDOWS and LINUX Operating Systems were installed to dual boot. The EMPIRE 3.2 modular code was installed on the LINUX Fedora 16 Operating System. As a part of the system software requirements, C++-compiler, gnuplot, Tcl/Tk, itcl were installed before the installation of the EMPIRE 3.2 Code. EMPIRE code files are available for download on <http://www.nndc.bnl.gov/empire/> and consists of .tgz (compressed) files and the installation script (install.sh). The .tgz files are: EMPIRE-3.2-MALTA.tgz, C4-latest.tgz, Active Tcl 8.4.19.6.295590-linux-ix86.tgz, and Install.sh – the installation script. Out of these files only empire-3.2-MALTA.tgz is required to run calculations. The installation can be done using the graphical or non- graphical method. The installation method used for this work was the Non-graphical Installation, done as follows:

First, the files were downloaded and placed in a temporary directory (e.g. ~/empire-tmp/). Then the setup.sh script was run with the following command: \$ sh install.sh and followed the on screen instructions. The installation script detected the optional files in temporary directory and installed them.

To run empire, a working subdirectory was created from empire root as ~/empire-3.2-malta/ads and changed to it. Then the EMPIRE GUI was launched with the command: empire3

The models used to describe the reactions include a non-dispersive phenomenological global optical model potential employed to describe the neutron interaction with the spherical targets. ECIS06 optical model was employed to calculate the direct reaction and the neutron transmission coefficients with a neutron optical model potential with RIPL number 2405 by [2]. The optical model potential is relativistic covering 1KeV to 200MeV energy range and 70 nuclei with mass range from 27 to 209.

The exciton model PCROSS as used as the pre-equilibrium model to calculate the gamma emission of the reactions at the pre-equilibrium level.

Hauser-Feshbach [3] and Hofmann-Richard-Tepel-Weidenmüller - HRTW [4] versions of the statistical model with full gamma-cascade were employed to treat the compound nucleus. The EMPIRE default nuclear level density, Enhanced Generalized Superfluid Model - EGSM, was used for the description of the nuclear structure. The discrete levels were taken from the RIPL-3 level file, based on the 2007 version of Evaluated Nuclear Structure Data File - ENDSF [5]. The gamma ray strength function Plujko Modified Lorentzian (MLO) RIPL-2 was used. The electronic dipole (E1) strength function was set to RIPL modified Lorentzian version 1 while Giant Dipole Resonance parameters (GDR) were taken from RIPL/Exp.data+Plujko systematic. All the deformation parameters are to take care of any deformation that could arise as a result of excitation.

These models and parameters fully describe the reactions and give good results as compared with the measured data from EXFOR in section 3.

3. RESULTS AND DISCUSSION

The results of the neutron capture reactions are presented as discussed below:

$^{79}\text{Se}(n,\gamma)^{80}\text{Se}$ has only one point measurement of the cross section by [6]. The one point is well predicted by this work as shown in Fig. 1. The measured data area is shown clearly in Fig. 1 (b).

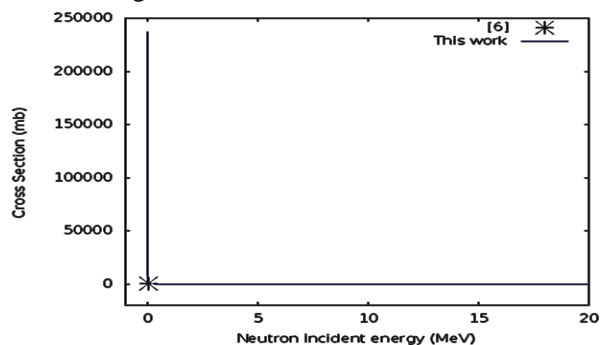


Fig 1: (a). The excitation function of $^{79}\text{Se}(n,\gamma)^{80}\text{Se}$ up to 20 MeV compared with measured data.

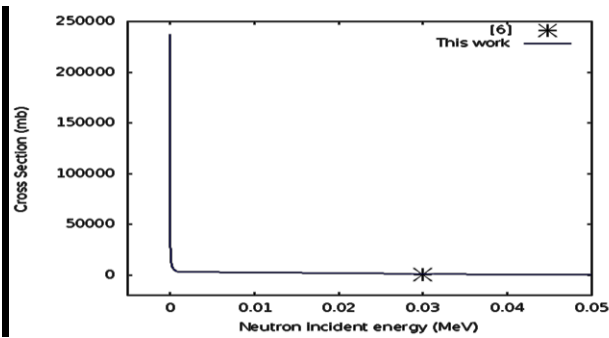


Fig. 1: (b). The excitation function of $^{79}\text{Se}(n,\gamma)^{80}\text{Se}$ in the region of available measured data.

Most of the cross section measurements of $^{90}\text{Sr}(n,\gamma)^{91}\text{Sr}$ coincide and are well predicted by this work as shown in Fig. 2.

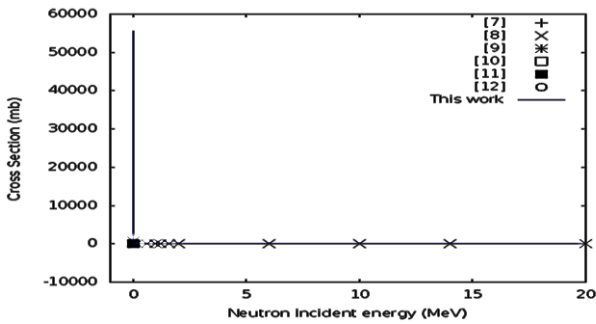


Fig. 2: (a). The excitation function of $^{90}\text{Sr}(n,\gamma)^{91}\text{Sr}$ with energy up to 20 MeV.

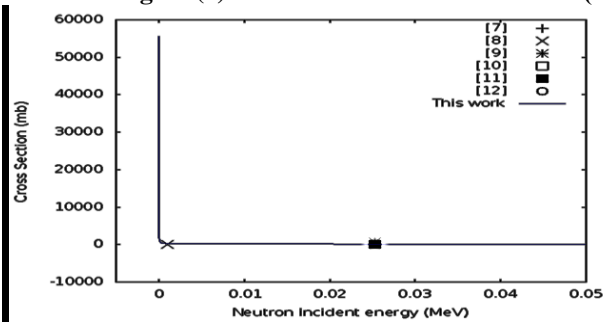


Fig. 2: (b). The excitation function of $^{90}\text{Sr}(n,\gamma)^{91}\text{Sr}$ showing the measured data area.

The measured cross sections of $^{93}\text{Zr}(n,\gamma)^{94}\text{Zr}$ are well predicted by this work as shown in Fig. 3.

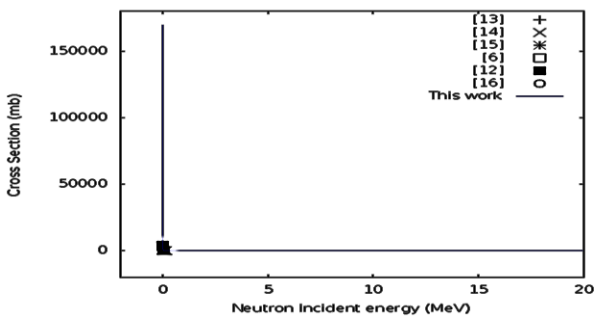


Fig. 3: (a). The excitation function of $^{93}\text{Zr}(n,\gamma)^{94}\text{Zr}$ with energy up to 20 MeV.

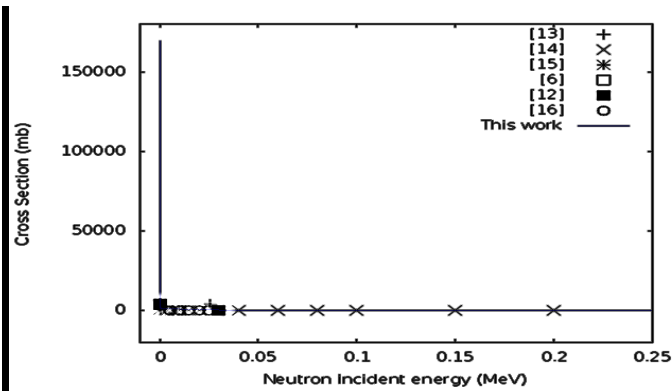


Fig. 3: (b). The excitation function of $^{93}\text{Zr}(n,\gamma)^{94}\text{Zr}$ showing the measured data area.

$^{107}\text{Pd}(n,\gamma)^{108}\text{Pd}$ reaction has measured cross sections beyond 20 Mev. Generally , the measured reaction cross sections are well predicted by this work as shown in Fig. 4.

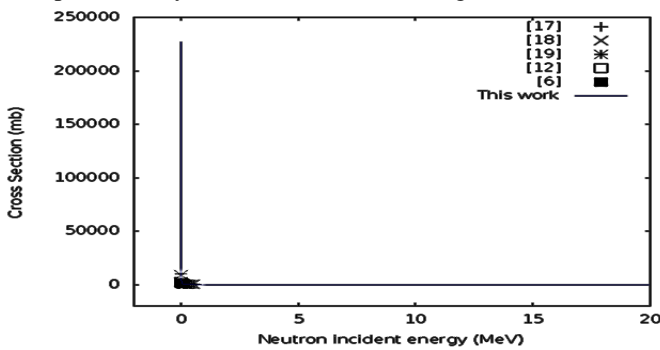


Fig. 4: (a). The cross section of $^{107}\text{Pd}(n,\gamma)^{108}\text{Pd}$ with energy up to 20 MeV.

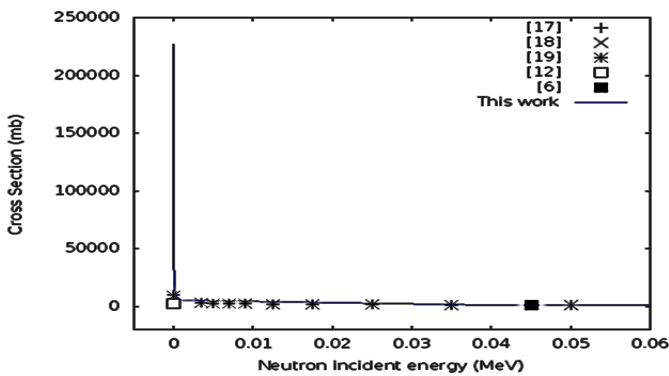


Fig. 4: (b). The excitation function of $^{107}\text{Pd}(n,\gamma)^{108}\text{Pd}$ showing the measured data area.

$^{99}\text{Tc}(n,\gamma)^{100}\text{Tc}$ has measurements up to 20 MeV. The measurements are well predicted by this work as shown in Fig. 5.

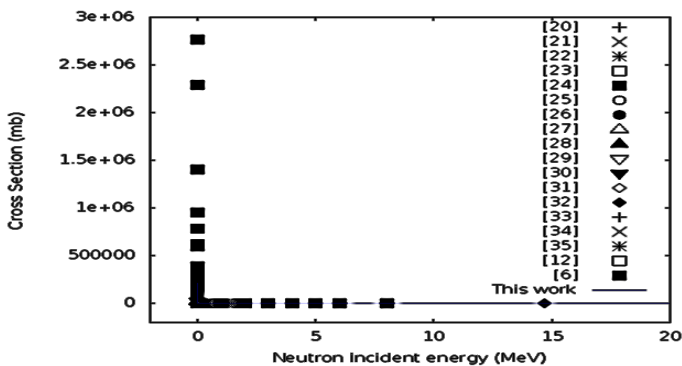


Fig. 5: The excitation function of $^{99}\text{Tc}(n,\gamma)^{100}\text{Tc}$ with energy up to 20 MeV.

The cross sections of $^{126}\text{Sn}(n,\gamma)^{127}\text{Sn}$ are as shown in Fig. 6. The one point each measured by [36] and [6] are well predicted by this work.

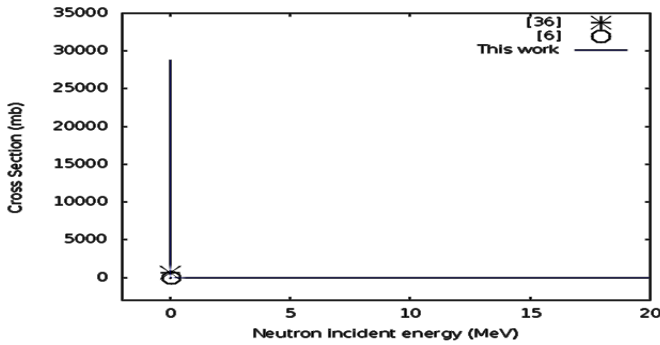


Fig. 6: (a). The excitation function of $^{126}\text{Sn}(n,\gamma)^{127}\text{Sn}$ with energy up to 20 MeV.

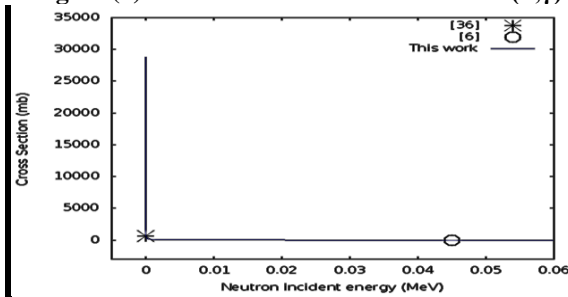


Fig. 6: (b). The cross section of $^{126}\text{Sn}(n,\gamma)^{127}\text{Sn}$ showing the measured data areas.

The measured cross sections of $^{129}\text{I}(n,\gamma)^{130}\text{I}$ are well predicted by this work as shown in Fig. 7.

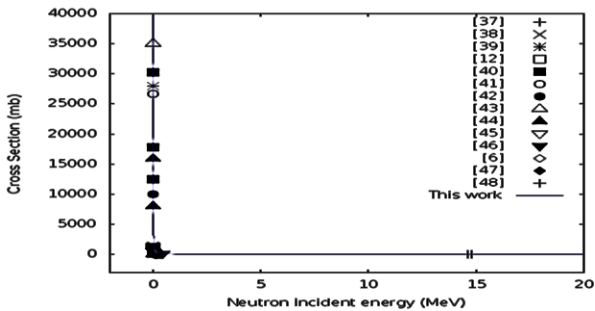


Fig.7: (a). The excitation function of $^{129}\text{I}(n,\gamma)^{130}\text{I}$ with energy up to 20 MeV.

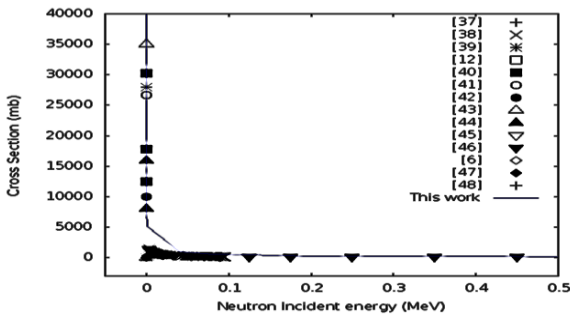


Fig. 7: (b). The excitation function of $^{129}\text{I}(n,\gamma)^{130}\text{I}$ showing the measured data area.

The $^{135}\text{Cs}(n,\gamma)^{136}\text{Cs}$ reactions has measurements up to 1MeV and they are all well predicted by this work as shown in Fig. 8.

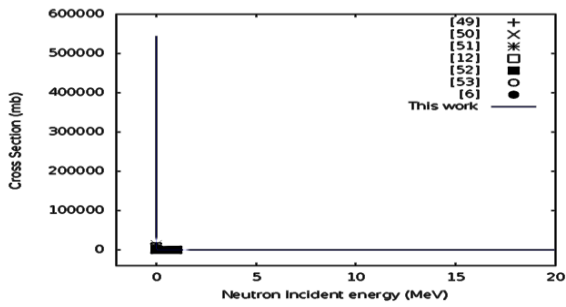


Fig. 8: (a). The excitation function of $^{135}\text{Cs}(n,\gamma)^{136}\text{Cs}$ with energy up to 20 MeV.

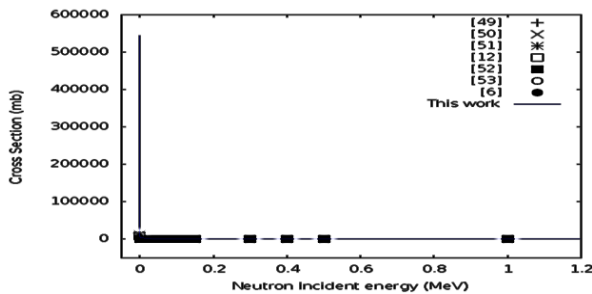


Fig. 8: (b). The excitation function of $^{135}\text{Cs}(n,\gamma)^{136}\text{Cs}$ showing the measured data area.

The measurements of $^{137}\text{Cs}(n,\gamma)^{138}\text{Cs}$ cross sections are very close and are over predicted by this work as shown in Fig. 9.

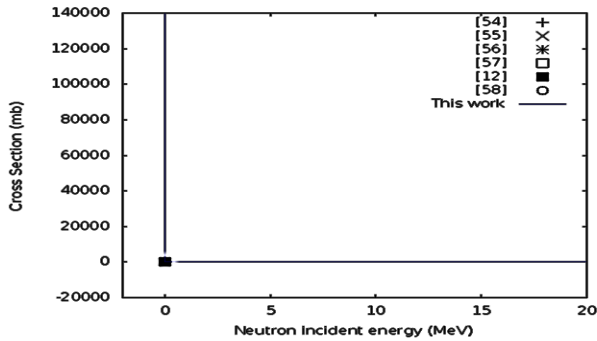


Fig. 9: (a). The excitation function of $^{137}\text{Cs}(n,\gamma)^{138}\text{Cs}$ with energy up to 20 MeV .

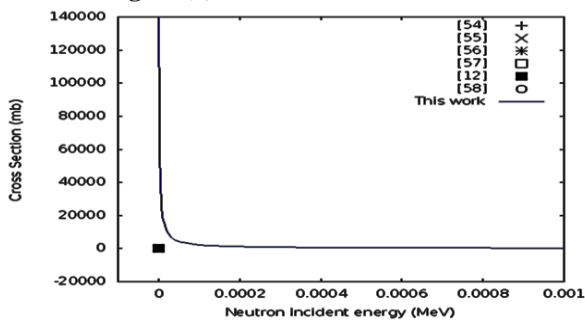


Fig. 9: (b). The cross section of $^{137}\text{Cs}(n,\gamma)^{138}\text{Cs}$ showing the measured data area.

$^{151}\text{Sm}(n,\gamma)^{152}\text{Sm}$ reaction has measurements with high energy of about 1MeV as shown in Fig. 10. All the measurements are well predicted by this work.

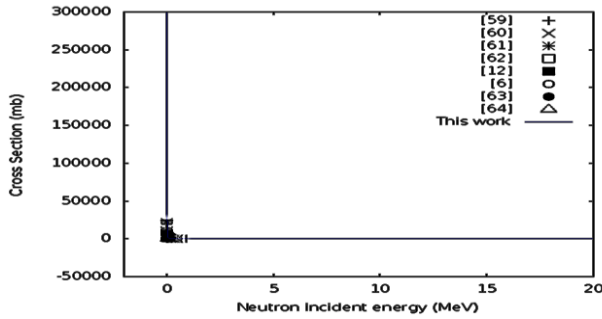


Fig. 10: (a). The excitation function of $^{151}\text{Sm}(n,\gamma)^{152}\text{Sm}$ with energy up to 20 MeV .

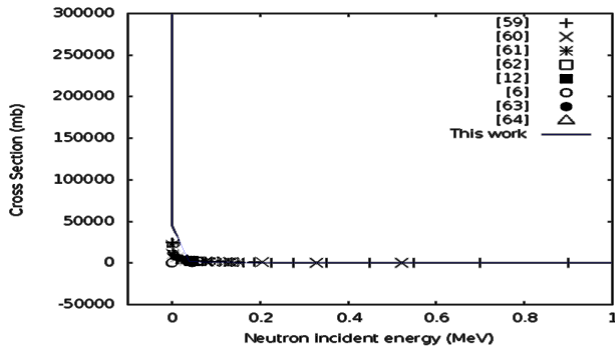


Fig. 10: (b). The excitation function of $^{151}\text{Sm}(n,\gamma)^{152}\text{Sm}$ showing the measured data area.

It could be seen that generally the (n,γ) reactions show continuous increase of neutron absorption as the energy decreases and there is no threshold energy as can be seen in Fig.11

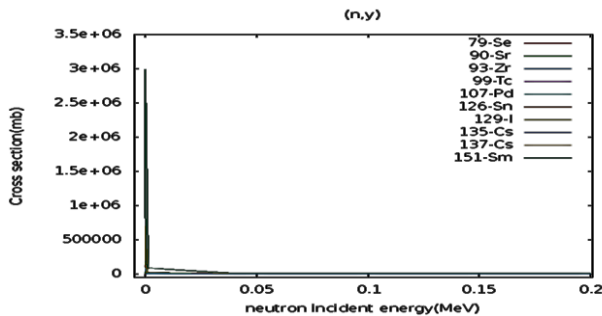


Fig. 11. Comparison of all the (n,γ) reaction cross sections of LLFPs.

Fig. 1 to 11 present the excitation functions for the neutron induced reactions on ^{79}Se , ^{90}Sr , ^{93}Zr , ^{99}Tc , ^{107}Pd , ^{126}Sn , ^{129}I , ^{135}Cs , ^{137}Cs and ^{151}Sm from threshold to 20 MeV. Most of the available measurements are within the thermal energy range and the evaluated results drop steeply within the thermal region confirming that the (n,γ) reaction is an open channel within thermal energy range and not favoured by higher energy.

The results of the neutron capture cross section show that the models used in the calculations accurately describe the measurements except for $^{137}\text{Cs}(n,\gamma)^{138}\text{Cs}$ (Fig. 9b) where the calculation over predicts the measured data. The spherical optical model in which the direct population of collective levels in the incident channel is suppressed, suitably describes the optical model contribution. The spherical, relativistic, non dispersive potential which cover isotopes within $13 \leq A \leq 83$, $29 \leq A \leq 209$ and 0 – 200 MeV incident energy was suitably used for this spherical optical model contribution as the neutron potential.

Furthermore, the compound nucleus has been accounted for by an advanced implementation of the Hauser Feshbach theory with exact angular momentum and parity coupling which also suitably accounted for the full γ -cascade in the residual nuclei which is necessary for a good description of radiative capture cross section. This compound nucleus mechanism also suitably account for the correlation between incident and exit channels in elastic scattering through the HRTW width fluctuation correction which was tested and optimized at 3 MeV. The Enhanced Generalized Superfluid Model improved the cross section result by suitably using the super-fluid model below critical excitation energy and the Fermi Gas model above. In this case the improvement is due to a proper accounting for the spin distribution in the Fermi Gas model by including a more accurate treatment of high angular momenta which are important in some reaction. The Gamma Ray Strength Function was also properly accounted for with a suitable form of the Modified Lorentzian.

For the pre-equilibrium contribution, the phenomenological exciton model PCROSS appropriately accounted for gamma emission at the pre-equilibrium level.

For the observed poor agreement with measurement in $^{137}\text{Cs}(n,\gamma)^{138}\text{Cs}$ (see Fig. 9(b)), the measurement may be better accounted by the use of a couple channel option for the optical model or the selection of an appropriate potential or a more descriptive gamma ray strength model. It should be noted that ^{137}Cs , as one of the principal long-lived fission products responsible for most of the radioactivity of spent nuclear fuel is a strong emitter of gamma radiation and has a very low rate of neutron capture, as shown by the measurement and this evaluation, such that it cannot be feasibly disposed through the (n,γ) transmutation and must be allowed to decay.

Generally, the results show that EMPIRE-3.2 has very strong capability of describing the available experimental results and provides suitable models and parameter description for the evaluation of cross section of LLFP useful in the design and operation of a reactor system for transmutation purposes.

CONCLUSION

The results of the determination of the cross section data of (n,γ) reaction on LLFPs is thus significant in the context of the nuclear data base available for the isotopes. The comparison of the calculated results with the measured data shows that EMPIRE-3.2 code is quite suitable in predicting the cross-section of the (n,γ) for neutron energy up to 20 MeV region. Optimal models and parameters which give results in good agreement with standard EXFOR cross section data for proton induced and neutron capture reactions were determined to include spherical optical model with RIPL number 2405 for neutron incident channel. This is enhanced with HRTW set to 3 MeV. Enhanced Generalized Super-fluid Model approach of the level density accounted for the nuclei structures. Modified Lorentzian (MLO 1) for the gamma ray strength function accounted for the gamma radiation is an appropriate model. Further enhancement is obtained with PCROSS to account for the pre-equilibrium contribution. The results show that EMPIRE 3.2 has a strong predictive abilities of the (n,γ) reaction on LLFPs.

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