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Nuclear model calculation of excitation functions of neutron induced reactions on the structural materials of the miniature neutron source reactor (Nigeria Research Reactor 1)

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ABSTRACT

Nuclear models calculation of excitation functions was performed using the nuclear theoretical model code EXIFON on the impurity nuclides of the Be-reflector of the MNSR (NIRR-1). Elements with concentration greater than 100 ppm were considered in the energy range of 0-20 MeV. Result of the calculated excitation functions obtained with the code EXIFON were compared with the experimental data retrieved from IAEA-Nuclear Data section EXFOR data library. Some of the results of the calculated excitation functions agree well with the experimental data, while some disagree. The good agreement was obtained for nuclides with magic number of neutrons and protons. Shell structure effects were not taken into consideration during calculation with theoretical model code. This might be the reason for disagreement. It is also noted that most of the data retrieved from EXFOR data library were incomplete, scanty, and are measured around 14 MeV. This exposed the short coming of using measured data for calculation of excitation functions. On the other hand the result of calculated excitation function with the Code EXIFON seems to give good result. Empirical research method was largely used in determining the success of this research.

***Keywords*:** EXIFON, EXFOR, MNSR, NIRR-1

1. INTRODUCTION AND BACKGROUND OF THE STUDY

Research reactors have played a significant role in the development of scientific and technical infrastructure in many countries. In developing countries, low power research reactors are acquired for training and research aimed at the introduction of Nuclear Power technology. (Jonah *et al*, 2005). In Nigeria, the first nuclear research reactor, a Miniature Neutron Source Reactor (MNSR) code named Nigeria Research Reactor – 1 (NIRR-1) is sited at CERT, ABU. Presently, there are eight commercial Miniature Neutron Source Reactors (MNSR) in the world and Nigeria Research Reactor – 1 (NIRR-1), which went critical for the first time on February 03, 2004 is the eighth The core of NIRR-1 with a fuel enrichment of over 90% was designed to last for approximately 10 years (Balogun, *etal*, 1999). Frequent refueling is avoided by the addition of Beryllium shims (Be) to compensate for the fuel burn up and long time poisoning effect. To achieve efficient shimming, the commercial (MNSR) has been designed with an atomic H/U^{235} ratio of 197 in the fuel lattice leading to further core under moderation (Jonah *et al* 2005). As expected, there is enhanced contribution from fission neutron at the irradiation channel, via threshold reaction on the Be shim serving as a reflector. Fe, Al, N, Mg, Si, Ni, Zn, Cu, Cr, with concentration greater than 100 ppm (part per million), impurities are known to be present in the B_e – reflector of the MNSR (SAR, 2005). The life time of the structural materials of the MNSR is put at 20 years by the manufacturer, during which this impurity elements are exposed to neutrons of energy between 0 to 20 MeV. Nuclear transmutation induced by these energetic neutrons causes damage in the material. The knowledge of cross section data due to (n , p) and (n, α) reaction on these elements could give information on the helium and hydrogen gas formation in the reflector. Finally, presence of these significant amount of impurities in the Be – reflectors can cause possible long – lived induced radioactivity of the structural material irradiated for 20 years or more (use time of the radiation) from radio nuclides of Ca, Fe , Co, Cs and Eu which are responsible for long-lived activity.

2. PURPOSES OF THE RESEARCH

The purpose of this research work is to achieve the following;

1. To use Nuclear model of statistical multi-step direct and multistep compound (SMD and SMC) for calculation of excitation function (n,α) and (n, p) reaction for impurities in the Be-reflector.
2. To compare the calculated results of the excitation functions (n, p) and (n,α) reactions induced on the impurities of Be-reflector with the experimental value in the EXFOR DATA library, in order to test the suitability of the model used.
3. To use the experimental data to investigate the suitability of the theoretical models upon which the EXIFON is based for elements of interest.
4. Experimental cross section data of some of these elements in the energy range of 0 – 20*MeV* are incomplete and discrepant, so theoretical calculation of excitation function can assist in the provision of cross section data.

a. Previous Works

The energy and angular distribution of pre-equilibrium nucleon from 14 *MeV* neutron energy with target Nuclei $^{51}V_i$ to $^{58}N_i$ are analyzed by multi-step compound (MSC) theory formalism of Feshback, Kerman and Koonin (FKK) and Geometry Dependent Hybrid (GDH) model by M. Avrigeamu and P.E. Hudgson in 1992. The same parameter set were used for both of these calculation with no free parameter, the calculations are compared with experimental data and the applicability of theory assessed.

Similar work was carried out by Jonah 2004, for the use of shell structure effect in neutron cross section calculations by theoretical model code. Others are inelastic scattering of neutron using statistical model by Walter Hauser and Herman Feshback (1952) and statistical multi-step reaction model for nuclear data by H. Kalka in (1991)

b. Present Work

In this work, the nuclear model calculation of excitation function will be based on the 'EXIFON' code and will be performed to determine the activation cross section data for (n, p) and (n , α) reaction, with neutron incident energy from (0 – 20*MeV*), in step of 2*MeV* on the impurities in the Be reflector. This will be done for impurity elements ^{54}Fe, ^{55}Fe, ^{56}Fe, ^{57}Fe, ^{24}Mg, ^{25}Mg, ^{26}Mg, ^{27}Al, ^{28}Si, ^{29}Si, ^{30}Si, ^{58}Ni, ^{60}Ni, ^{61}Ni, ^{62}Ni, ^{63}Ni, ^{14}N, ^{15}N, ^{63}Cu, ^{65}Cu, ^{55}Mn, ^{64}Zn, ^{66}Zn, ^{67}Zn, ^{68}Zn, ^{70}Zn, ^{50}Cr, ^{52}Cr, ^{53}Cr, ^{54}Cr, ^{59}Co, ^{16}O, ^{17}O, ^{18}O, ^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb, base on the isotopic abundance.

The analytical model is statistical multi step code which is based on many body theories and the random matrix physics. It utilized the statistical multi step direct and multi step compound reaction for the prediction of emission spectra, angular distribution and activation cross section taking into account equilibrium, pre-equilibrium as well as direct processes. It is a fast code which predicts the cross section data from global parameter set, in order to access the excitation functions of the impurity element. Detailed description of the model used in the 'EXIFON' code has been given in a report by H. Kalka (1991) and Kalka *et al* (1990).

3. MATERIALS AND METHOD

For this research, the structural material of the MNSR to be taken into consideration is the Beryllium Reflector (Be), which consists of annular beryllium reflector, top Beryllium shim and bottom Beryllium Reflector. The aim of using Beryllium shim is to compensate for fuel burn up, avoid frequent refueling and to reflect fast and slow neutrons. The inner diameter of the annular Be reflector of NIRR-1 is 23.10 cm while the outer the diameter of the annular Be-reflector is 43.5 cm the height of the annular Be-reflector is 23.85cm and the thickness of the bottom Be-reflector is 5cm (Akaho, 2000). For calculation of the excitation function of neutron induced reaction on the structural material of MNSR (NRR1). We consider the different Isotopes of the impurities in the 'Be' reflector based on the percentage abundance. This impurity composition by percentage was obtained from the SAR, 2005:

3. 1. The (n.p) Reaction Channel

The cross section of each impurity nuclides with neutron was calculated in (n, p) reaction channel. The outgoing particle in this reaction is charged, occurrence of the reaction depends on the height of coulomb barrier which increases in energy from 1 Mev, for Z = 3 to 12Mev for Z = 90. For impurities in the Be-reflector, the (n.p) reaction cross section was calculated for the following reactions:

1)	a. ^{56}Fe (n p)^{56} Mn	b. ^{57}Fe (n p)^{57} Mn	c.^{58}Fe (n p)^{58} Mn	
2)	a. ^{24}Mg (n p)^{24} Na	b. ^{25}Mg (n p)^{25} Na	c. ^{26}Mg (n p)^{26} Na	
3)	^{27}AL (n p)^{27} Mg			
4)	a. ^{28}Si (n p)^{28} AL	b. ^{29}Si (n p)^{29} AL	c. ^{30}Si (n p)^{30} AL	
5)	a. ^{63}Cu (n p)^{63} Ni	b. ^{65}Cu (n p)^{65} Ni		
6)	a. ^{14}N (n p)^{14} C	b. ^{15}N (n p)^{15} C		
7)	a. ^{50}Cr (n p)^{50} V	b. ^{52}Cr (n p)^{52} V	c. ^{53}Cr (n p)^{53} V	d. ^{54}Cr (n p)^{54} V
8)	a. ^{64}Zn (n p)^{64} Cu	b. ^{66}Zn (n p)^{66} Cu	c. ^{67}Zn (n p)^{67} Cu	d. ^{68}Zn (n p)^{68} Cu
	e. ^{70}Zn (n p)^{70} Cu			
9)	a. ^{58}Ni (n p)^{58} Co	b. ^{60}Ni (n p)^{60} Co	c. ^{61}Ni (n p)^{61} Co	d. ^{62}Ni (n p)^{62} Co
	e. ^{63}Ni (n p)^{63} Co			
10)	a. ^{204}Pb (n p)^{204} Ti	b. ^{206}Pb (n p)^{206} Ti	c.^{207}Pb (n p)^{207} Ti	d. ^{208}Pb (n p)^{208} Ti
11)	^{59}Co (n p)^{59} Fe			
12)	^{55}Mn (n p)^{55} Cr			
13)	a. $^{16}_{8}O$ (n p)^{16} N	b. ^{17}O (n p)^{17} N	c. ^{18}O (n p)^{18} N	

3. 2. The (n. α) Reaction channel

Similarly for the different Isotopes of the impurities in the Be-reflector, the (n, α) reaction cross section were calculated. In this reaction the coulomb barrier is higher than in (n.p) reaction therefore it is not prominent at low energy of neutrons. The coulomb barrier increases with increasing atomic number from 1Mev at Z = 2 to 20Mev at Z = 90. This data were calculated for the following reactions:

1. $^{54}Fe\,(n,\alpha)^{51}Cr$, $^{56}Fe(n,\alpha)^{53}Cr$ $^{57}Fe(n,\alpha)^{53}Cr$ $^{58}Fe(n,\alpha)^{55}Cr$
2. $^{24}Mg\,(n,\alpha)^{21}Ne$, $^{25}Mg(n,\alpha)^{23}Ne$ $^{26}Mg(n,\alpha)^{23}Ne$
3. $^{27}AL(n,\alpha)^{24}Na$
4. $^{28}Si\,(n,\alpha)^{25}Mg$, $^{29}Si(n,\alpha)^{26}Mg$ $^{30}Si(n,\alpha)^{27}Mg$

5. $^{63}Cu\ (n,\alpha)^{60}Co,\ ^{65}Cu(n,\alpha)^{62}Co$

6. $^{17}N\ (n,\alpha)^{11}B,\ ^{15}N(n,\alpha)^{12}B$

7. $^{50}Cr\ (n,\alpha)^{47}Ti,\ ^{52}Cr(n,\alpha)^{49}Ti\ ^{53}Cr\ (n,\alpha)^{50}Ti\ ^{54}Cr\ (n,\alpha)^{51}Ti$

8. $^{64}Zn(n,\alpha)^{61}Ni,\ ^{66}Zn(n,\alpha)^{63}Ni\ ^{67}Zn(n,\alpha)^{64}Ni\ ^{68}Zn(n,\alpha)^{65}Ni\ ^{70}Zn(n\alpha)^{64}Ni$

9. $^{58}Ni\ (n,\alpha)^{55}Fe,\ ^{60}Ni(n,\alpha)^{57}Fe,\ ^{61}Ni\ (n,\alpha)\ ^{58}Fe\ ^{62}Ni\ (n,\alpha)\ ^{59}Fe,\ ^{63}Ni\ (n\ \alpha)\ ^{60}Fe$

10. $^{204}Pb\ (n,\alpha)^{201}Hg\ ,\ ^{206}Pb\ (n,\alpha)^{203}Hg,\ ^{207}Pb\ (n,\alpha)\ ^{204}Hg\ ^{208}Pb\ (n,\alpha)\ ^{205}Hg$

11. $^{59}Co(n,\alpha)^{56}Mn$

12. $^{55}Mn(n,\alpha)^{52}\nu$

13. $^{16}O\ (n,\alpha)^{13}C,\ ^{17}O(n,\alpha)^{14}C\ ^{18}O(n,\alpha)^{15}C$

3. 3. Installation and Running of Exifon Code

In this work, the theoretical model code EXIFON which is based on the formalism of optical potential (OM) of the statistical multistep direct (SMD), statistical multistep compound (SMC) and multi particle emission processes (MPE) was used for calculation of excitation function of the neutron induced reaction to the impurities Nuclide in the Be-reflector. The code is a command prompt programme. The excitation functions were calculated from a plot of cross section Vs Energy. The values of the cross section were given in 10^{-3} barns, while that of energies is in MeV. The (n, p) reaction for the various impurity nuclides was considered as (n, pγ,) data for comparison with measured cross section data in EXFOR data library. Similarly, the result of (n, α) reaction was considered in (n, αγ) data. Both results were used in plotting a graph of excitation function (a graph of cross section vs Energy).

In order to calculate the excitation function, threshold reaction via (n, pγ,) and (n, αγ) reaction in the energy range of 0 – 20Mev is considered in step of 2Mev at each projectile (neutron) energy.

A simple two body interaction is assumed and a single state particle density of (n, p, α) with reduced mass given by:

$$g = 4\ \rho\ (E_F)$$

where the factor 4 takes into cognizance of spin and Isospin degeneracy $\rho(E_F) = (4.8 \times 10^{-3}Fm^{-3}MeV^{3/4})r_o{}^3AE^{1/2}$

3. 4. Standard Parameter Set

Other global parameters set for the excitation function are:

- Strength of delta interaction Fo = 27.5 Mev.
- Radius Parameter r_o = $1.21 + 4.0A^{-2/3} – 15A^{4/3}Fm$

- Potential depth V_o = 52.03 Mev.
- Fermi Energy E_F = 33 Mev.
- Paring shift A = 12.8 $A^{-1/2}$Mev.
- Phonon (Breit – Wigner) width Δw = 1.4 Mev.
- Optical model (Wilmore-Hodgson for Neutron Huizenga – Igo, for alphas, Perey et al, for proton)

3. 5. Modifications

In calculation the following parameter can be changed, strength of the residual interaction F_o, radius parameter r_o, Fermi energy E_f, phonon width Δ_w, and the global OM parameter set for proton in addition the pairing shifth Δ and Δ^{eff} can be modified (also for SMD processes). The pairing energy has the most influence on description of emission spectra especially on activation cross section.

3. 6. Access to EXFOR Library

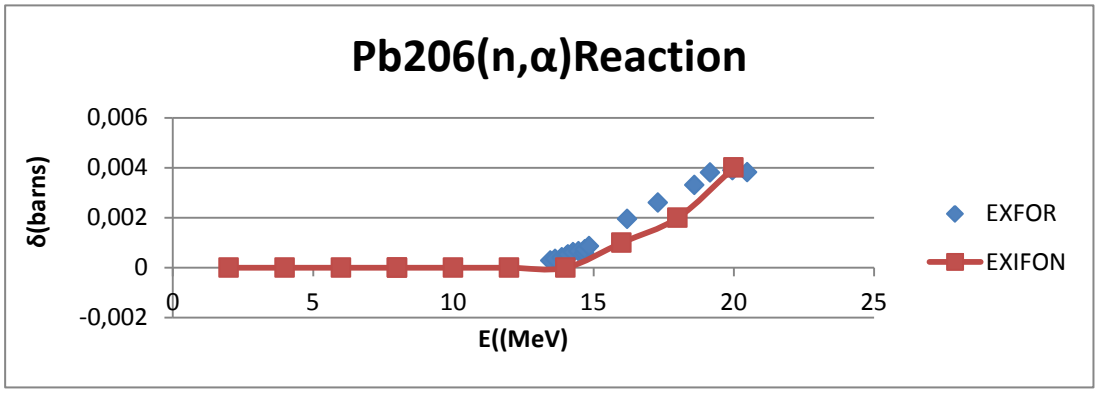
The EXFOR is available for interactive retrievals through World Wide Web (www) and telnet (one reaction at a time). Also, on two CD-ROM versions (same data base, different retrieval software) both developed by nuclear data library (NDS) of the IAEA complicated retrievals are available individually on request from IAEA –NDS (diskette, tape, file transfers, e-mail, and print out) EXFOR output are in formats and these include standard format (Exchange formats), computational format, for plotting and further processing from the IAEA and online plots for inter comparison with evaluated data "BNL325" "ZV View".

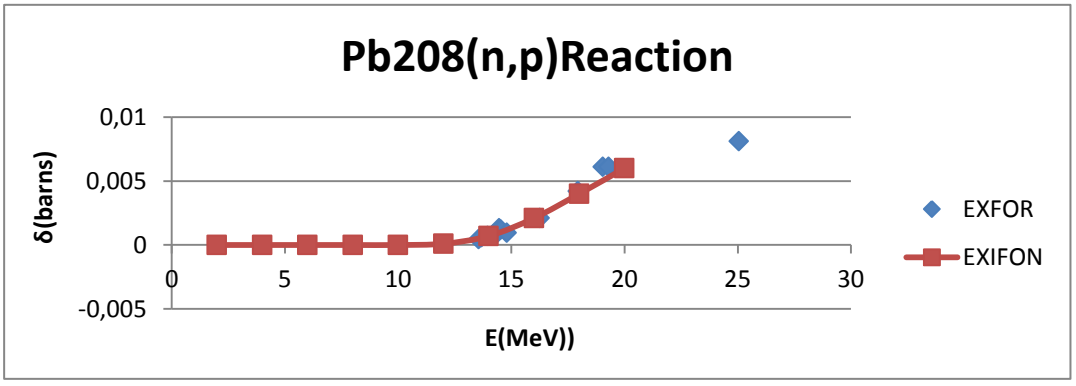
Finally a plot of both EXFOR and EXIFON results of excitation function will be used for this research work. From which further analysis can be give insight to the suitability of each model.

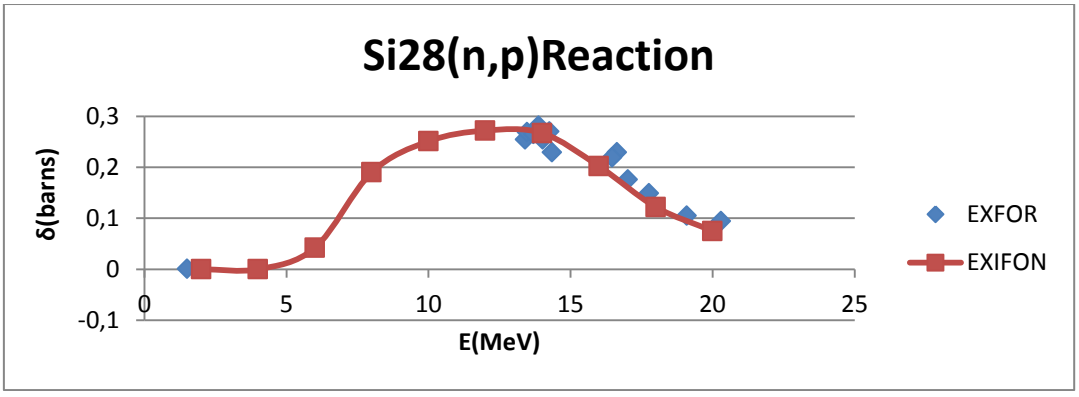
4. RESULTS AND DISCUSSION

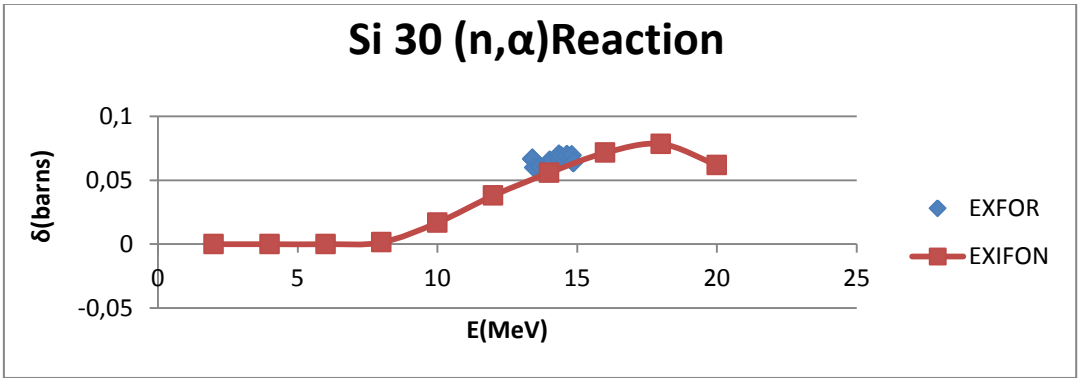
The theoretical model code EXIFON was used to determine the excitation function of the nuclides of impurities in the Be-reflector of NIRR-1.

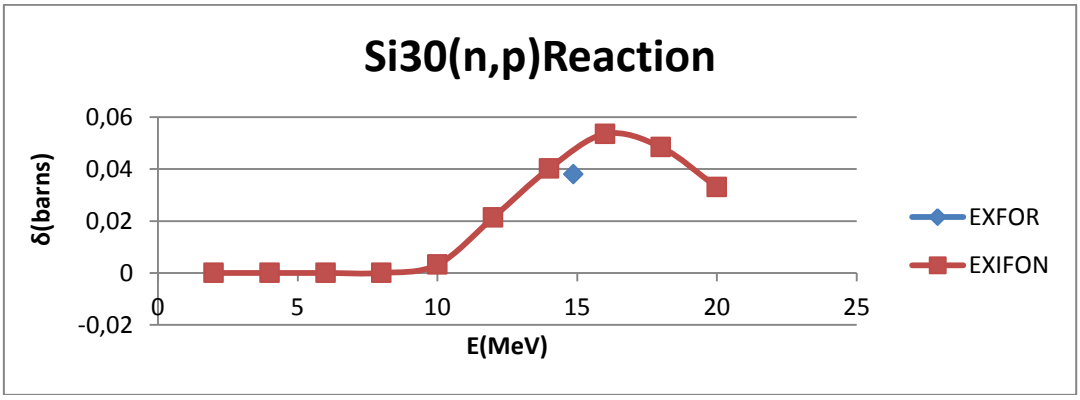
The graphs below shows some of the calculated excitation functions versus experimental result from EXFOR data library via (n, α) (n p) reaction channels.

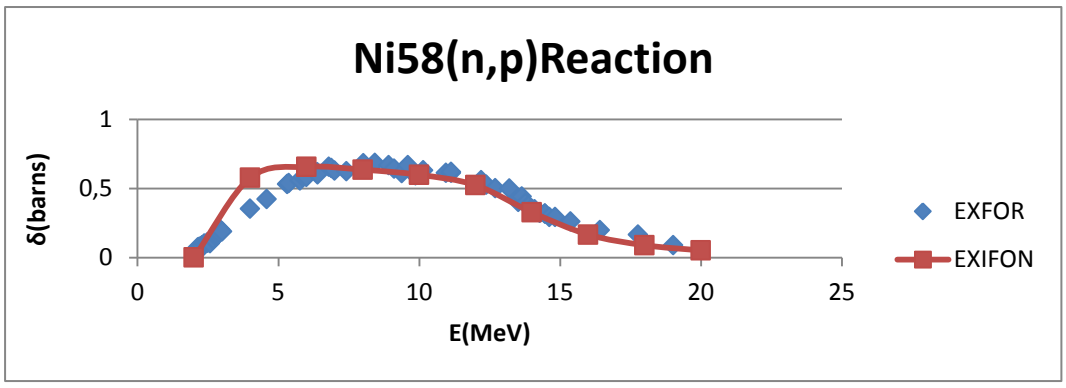


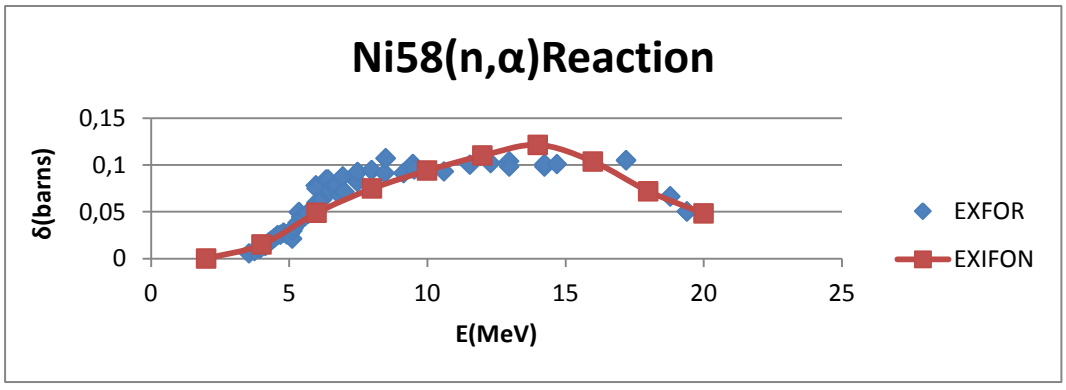

Pb208(n,p)Reaction
δ(barns)
0,01
0,005
0
-0,005
0
5
10
15
20
25
30
E(MeV))
EXFOR
EXIFON

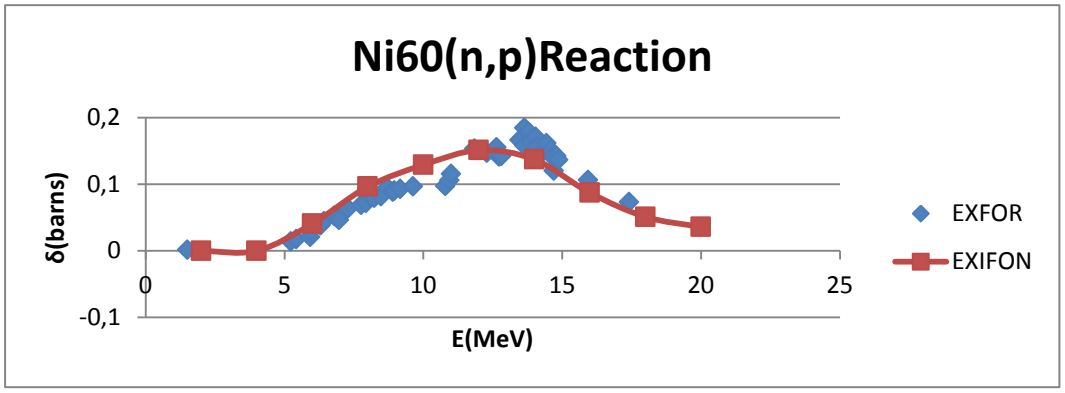

Si28(n,p)Reaction
δ(barns)
0,3
0,2
0,1
0
-0,1
0
5
10
15
20
25
E(MeV)
EXFOR
EXIFON

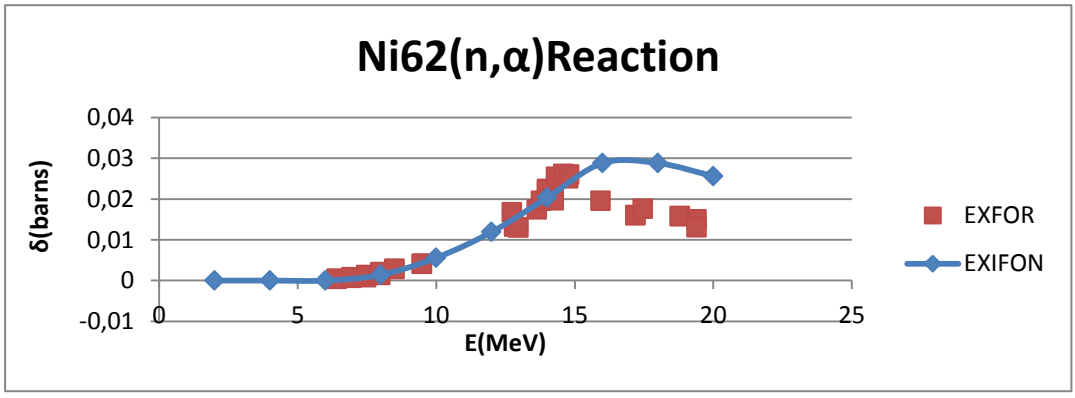

Si 30 (n,α)Reaction
δ(barns)
0,1
0,05
0
-0,05
0
5
10
15
20
25
E(MeV)
EXFOR
EXIFON


Si30(n,p)Reaction
δ(barns)
0,06
0,04
0,02
0
-0,02
0
5
10
15
20
25
E(MeV)
EXFOR
EXIFON


Ni58(n,p)Reaction
δ(barns)
1
0,5
0
0
5
10
15
20
25
E(MeV)
EXFOR
EXIFON


Ni58(n,α)Reaction
δ(barns)
0,15
0,1
0,05
0
0
5
10
15
20
25
E(MeV)
EXFOR
EXIFON


Ni60(n,p)Reaction
δ(barns)
0,2
0,1
0
-0,1
0
5
10
15
20
25
E(MeV)
EXFOR
EXIFON


Ni62(n,α)Reaction
δ(barns)
0,04
0,03
0,02
0,01
0
-0,01
0
5
10
15
20
25
E(MeV)
EXFOR
EXIFON

5. A SUMMARY OF THE STATUS OF CALCULATED EXCITATION FUNCTIONS USING EXFOR AND EXIFON

Target Nuclei	Percentage Isotopic Abundance (%)	Concentration in Part per million (ppm)	Status in EXIFON	Status in EXFOR (n.p)	(n α)	EXIFON Vs EXFOR (n.p)	EXIFON Vs EXFOR (n.α)
^{54}Fe	5.845	4000	Exist	Exist	Exist	Mostly agree	Agrees
^{56}Fe	91.754	4000	Exist	Exist	Exist	Mostly agree	Agrees
^{57}Fe	2.119	4000	Exist	Exist	No data	Disagree	No data
^{58}Fe	0.282	4000	Does not Exist	No data	No data	No data	No data
^{27}Al	100	3000	Exist	Exist	Exist	Agrees	Agrees
^{24}Mg	78.99	1000	Exist	Exist	Exist	Agrees	Agrees
^{25}Mg	10.00	1000	Exist	Exist	Exist	Disagree	Almost agree
^{26}Mg	11.01	1000	Exist	No data	Exist	No data	Almost agree
^{28}Si	90.23	800	Exist	Exist	No data	Agrees	No data
^{29}Si	4.683	800	Does not Exist	No data	No data	No data	No data
^{30}Si	3.087	800	Exist	Exist	Exist	Agrees	Almost agree
^{58}Ni	68.077	100	Exist	Exist	Exist	Agrees	Agrees
^{60}Ni	26.223	100	Exist	Exist	No data	Agrees	No data
^{61}Ni	1.140	100	Exist	Exist	No data	Disagree	Almost agree
^{62}Ni	3.634	100	Exist	No data	Exist	No data	No data
^{63}Ni	0.926	100	Exist	No data	No data	No data	No data
^{14}N	99.634	200	Exist	Exist	No data	Incomplete	No data
^{15}N	0.366	200	Does not Exist	No data	No data	No data	No data
^{63}Cu	69.17	200	Exist	Exist	Exist	No data	Almost agree
^{65}Cu	30.83	200	Exist	Exist	Exist	Almost agree	Mostly disagree
^{55}Mn	100	200	Exist	Exist	Exist	Disagree	Partly disagree
^{64}Zn	48.63	150	Exist	Exist	Exist	Almost agree	Disagree
^{66}Zn	27.90	150	Exist	Exist	No data	Disagree	Almost agree
^{67}Zn	14.10	150	Does not Exist	No data	No data	Disagree	No data
^{68}Zn	18.75	150	Exist	Exist	Exist	Disagree	Mostly disagree
^{70}Zn	0.62	150	Does not Exist	No data	No data	No data	No data
^{50}Cr	4.345	200	Exist	Exist	No data	Agrees	No data
^{52}Cr	83.789	200	Exist	Exist	No data	Agrees	No data
^{53}Cr	9.501	200	Exist	Exist	No data	Disagree	No data
^{54}Cr	2.365	200	Exist	Exist	Exist	Almost agree	Disagree
^{59}Co	100	100	Exist	Exist	Exist	Partly agree	Partly agree
^{16}O	99.70	2500	Exist	No data	Exist	No data	Incomplete
^{17}O	0.038	2500	Exist	No data	Exist	No data	Incomplete
^{18}O	0.200	2500	Does not Exist	No data	No data	No data	No data
^{204}Pb	1.4	30	Exist	No data	No data	No data	No data
^{206}Pb	24.1	30	Exist	No data	Exist	No data	Agrees
^{207}Pb	22.1	30	Exist	No data	No data	No data	No data
^{208}Pb	52.8	30	Exist	Exist	No data	Agrees	No data

6. CONCLUSIONS AND RECOMMENDATIONS

The nuclear model calculations of excitation function were performed with theoretical model code EXIFON. Data of the excitation function were compared with measured data from IAEA-NDS EXFOR Data library. The theoretical code EXIFON adequately account for the interaction of nucleus with neutron in the energy range of interest. Results obtained differ from experimental data in the input parameter used in EXIFON code, which may not be able to account for structures of all the nuclides considered. For nuclides with magic number of neutrons or protons for example ^{54}Fe, ^{16}O, ^{58}Ni, ^{208}Pb results obtained were found to be consistent with the experimental data. The theoretical model code was able to predict the cross section data where no experimental data exist. In cases where the calculated data deviate significantly from experiments it is recommended to use other code that are robust and more flexible in terms of input parameter such as the EMPIR-II, ALICE and GNASH. Another advantage of the theoretical model code, is that it was able to provide information of most of the cross section data for the nuclides of the impurities on the Be-reflector, where there are no measured data. For the set of target nuclides in Be-reflector of NIRR1 experimental data EXFOR data were scanty and discrepant. Hence further measurements need to be carried out at different energies so as to improve data base. Most of measured data were around 14 MeV.

The (n p) and (n α) considered in this work are to give information on the presence of H_2 and He gas formation on the Be reflector. With the exception of ^{52}Cr (n, p), ^{53}Cr(n p), and ^{64}Zn (n, p) reactions. Finally, the EXIFON code adequately reproduce experimental data for the (n,p) reaction channel. For both EXFOR and EXIFON, the following recommendations were made based on the outcomes of the research.

a. Additional work are to be conducted for the experimental data to serve as a basis for comparison between measured data and theoretical code, in order to access the suitability of the theoretical model used.
b. For other structural materials of NIRR-1, further research needs to be conducted on the activities of the impurities contained on these materials, so that it could give account of the integrity of the structural materials of NIRR-1.
c. Hence a good knowledge of cross section data of all the structural materials of NIRR-1, need to be taken into consideration, even before the commissioning of NIRR-1.
d. Similarly, the EXIFON code is suitable for the calculation of (n, α) reaction cross section except for ^{68}Zn, ^{25}Mg, ^{55}Mn, ^{16}O and ^{17}O

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