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Original Article

Study on (n, α) reactions for the production of ^{51}Cr, ^{89}Sr, ^{99}Tc, ^{131}I, ^{133}Xe, ^{137}Cs and ^{153}Sm radioisotopes used in nuclear medicine

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ABSTRACT

Nuclear medicine seems to be a decent choice of medicine in the recent decade. The radioactive isotopes ^{51}Cr, ^{89}Sr, ^{99}Tc, ^{131}I, ^{133}Xe, ^{137}Cs and ^{153}Sm are extremely essential in nuclear medicine. The excitation functions of the ^{54}Fe (n, α) ^{51}Cr, ^{92}Zr (n, α) ^{89}Sr, ^{102}Rh (n, α) ^{99}Tc, ^{134}Cs (n, α) ^{131}I, ^{136}Ba (n, α) ^{133}Xe, ^{140}La (n, α) ^{137}Cs and ^{156}Gd (n, α) ^{153}Sm reactions were calculated in this study using the EMPIRE 3.2.3 and TALYS 1.95 nuclear codes. Additionally, the cross sections at 14–15 MeV were calculated using empirical formulae and the experimental data. The computer codes were compared to the experimental data and Empirical formulas as well as the evaluated data (TENDL 2021, JENDL 3.3, JENDL 5, JEFF 3.3, EAF 2010, CENDL 3.1, CENDL 3.2, ROSFOND 2010, FENDL 3.2 b, and BROND 3.1).

1. Introduction

Nuclear medicine is a branch of radiology that studies the structure and function of organs using small amounts of radioactive substances known as radiopharmaceuticals. This major of radiography is frequently utilized to detect and treat abnormalities that first appear relatively early in the course of a disease. Imaging in nuclear medicine is a synthesis of several disciplines, including chemistry, biology, physics, mathematics, and computer science. Demand for medical radioisotopes is growing rapidly as tens of millions of nuclear medicine procedures are performed each year [1]. Throughout the process, a little amount of a radioactive chemical is used to aid in the examination. The radionuclide is absorbed by the biological process and is known as a radioactive tracer or radiopharmaceutical [2]. Radionuclides are of different types. Examples are technetium, thallium, gallium, iodine and xenon. Depending on the objectives of the study and the part of the body being examined, the radionuclide used will vary [3].

A detailed study of the excitation function is necessary to optimize the production of radioisotopes, which allows increasing the amount of the final product while reducing radioactive contamination. In materials in a neutronic environment, (n, α)-reactions are a significant source of helium generation. Fast neutrons with energies in the MeV range react with target nuclei causing (n, α)-reactions in which the α-particles are ejected from the reacting nuclei with energies of several MeV.

Theoretical models are often used to obtain neutron cross-sections when experimental measurements at a certain energy of incident neutrons are impossible due to experimental difficulties [4–9].

In this study, the cross sections for ^{54}Fe (n, α) ^{51}Cr, ^{92}Zr (n, α) ^{89}Sr, ^{102}Rh (n, α) ^{99}Tc, ^{134}Cs (n, α) ^{131}I, ^{136}Ba (n, α) ^{133}Xe, ^{140}La (n, α) ^{137}Cs and ^{156}Gd (n, α) ^{153}Sm reactions were calculated in the neutron incident energy range between 1 and 20 MeV. The excitation functions of (n, α) nuclear reactions were obtained for each of the produced radioisotopes. The results were compared with theoretical models and experimental data found in EXFOR [10] and ENDF [11]. The excitation functions were calculated using the nuclear reaction simulation codes EMPIRE 3.2.3 [12] and TALYS 1.95 [13].

2. Methods

2.1. EMPIRE 3.2.3 code

EMPIRE 3.2.3 is a flexible system of nuclear reaction codes that consists of several nuclear models and is made to do computations with a wide variety of incident particle energies. Any particle, including heavy ions and neutrons, protons, all other ions, and photons, can be a projectile. The energy range for heavy ion

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induced reactions spans several hundred MeV and starts at the unresolved resonance region's low energy (keV) for neutron-induced reactions [14].

The formation probability of a cluster in this model eliminates free parameters and considers excitons below and above the Fermi surface. The Iwamoto-Harada model gives the probability of emission of the cluster β with spin S_β, decreased mass μ_β, and energy ε_β from a state with n excitons is giving by Ref. [15]:

$$W_\beta(E,n,\varepsilon_\beta)=\frac{2S_\beta+1}{\pi^2\hbar^3}\mu_\beta\varepsilon_\beta\sigma_\beta^{inv}(\varepsilon_\beta)\times\frac{\sum\limits_{l+m=\beta}F_{lm}^{\beta}(\varepsilon_\beta)Q_{lm}^{\beta}(p,h)w_{res}(p-l,h,U)}{w_{CN}(p,h,E)} \tag{1}$$

where $Q_{lm}^{\beta}(p,h)$ is the factor generalization of the probability of the outgoing cluster β being formed with l particles situated above and m below the Fermi surface $l+m=\beta$ [15] and the factor $F_{lm}^{\beta}(\varepsilon_\beta)$ gives the probability of formation of the cluster β as a function of its energy. This last factor is calculated in PCROSS following the unpublished parametrization based on the original calculations of the cluster formation probability by Iwamoto and Harada. $w_{CN}(p,h,E)$ the Pauli correction, $w_{res}(p-l,h,U)$ is partial state densities and $\sigma_\beta^{inv}(\varepsilon_\beta)$ is the inverse channel reaction cross section.

The pre-equilibrium spectra may be obtained as,

$$\frac{d\sigma_{a,b}}{d\varepsilon_b}(\varepsilon_b)=\sigma_{a,b}^{r}(E_{inc})D_{a,b}(E_{inc})\times\sum_n W_b(E,n,\varepsilon_b)\tau(n) \tag{2}$$

where $\sigma_{a,b}^{r}(E_{inc})$ represents the cross-section of the reaction (a, b) and the term $D_{a,b}(E_{inc})$ is depletion factor. $W_b(E,n,\varepsilon_b)$ is the probability of a particle of sort b (or γ-ray) emitting by excitation energy E of compound nucleus and energy ε_b from state with n exciton [12].

2.2. TALYS 1.95 code

The creation of the TALYS 1.95 code began in 1998 in order to capitalize on the new computers that were coming at the time and to have a tool that would be easier to update than the other available codes. It has now integrated many additional models and been made useable with other programs in order to provide a link between nuclear reaction modelling and nuclear data evaluations [13]. In this code, the pre-compound particle emission nuclear cross-section process is as shown in:

$$\frac{d\sigma_k^{PE}}{DE_k}=\sigma^{CF}\sum_{p_\pi=p_\pi^0}^{p_\pi^{max}}\times\sum_{p_\nu=p_\nu^0}^{p_\nu^{max}}w_k(p_\nu,h_\nu,p_\pi,h_\pi,E_k)\tau(p_\nu,h_\nu,p_\pi,h_\pi)P(p_\nu,h_\nu,p_\pi,h_\pi) \tag{3}$$

where σ^{CF} represents the compound nucleus formation cross section calculated by the optical model. The term τ is the average lifetime for the exciton state. The terms , $h_\pi(h_\nu)$ d , $p_\pi(p_\nu)$ represent the proton (neutron) hole number and the proton (neutron) particle number, respectively. Also, E_k and w_k are emission energy and the emission rate of a particle k, and P represents the portion of the pre-compound population that must have survived the emission's prior states in order for it to pass through the configuration of (p_π, h_π, p_ν, h_ν), averaged over time [13]. The initial neutron and proton particle numbers are $p_\nu^0=N_p$ and $p_\pi^0=Z_p$, respectively with $Z_p(N_p)$ the proton (neutron) number of incoming particles [13].

2.3. Empirical formulas

Empirical formulas have been necessary to describe nuclear structures and excited nuclear states for nuclear reaction cross-sections at a variety of energies. Additionally, nuclear reactions cause particles emission as well as the development of increasingly complicated nuclear models depend on the cross sections data. As a result, the cross sections were measured using empirical formulae. In recent decades, many number of studies and publications of empirical equations for various reaction channels have seen. In this study, we provided an empirical formula to determine the (n, α) cross-sections between 14 and 15 MeV. The cross-section equations in different mass number range are formulated as follows, as stated in previous researches:

Levkovskii's formula for the cross sections of (n,α) reactions at an energy of 14 MeV states the following:

$$\sigma_{(n,\alpha)}=a_1\left(A^{\frac{1}{3}}+1\right)^2\exp\left[-a_2\frac{N-Z}{A}\right] \tag{4}$$

Laskowskii's formula is modified by Ref. [16] for (n,α) reaction cross sections for target nuclei range $61\le$ *mass number* ≤ 105 with neutron incident energy of 14 MeV.

$$\sigma_{(n,\alpha)}=a_1\,A^2\exp\left[-a_2\frac{N-Z}{A}\right] \tag{5}$$

[17] gave the following empirical approach for calculating (n,α) reaction cross sections at a neutron incident energy of 14 MeV:

$$\sigma_{(n,\alpha)}=a_1\,(N-Z+1)\exp\left[-a_2\frac{N-Z+1}{A}\right] \tag{6}$$

The following new modified Laskowski formulas were developed by Ref. [18] after studying the systematic behaviour of 44 target nuclei with $26\le$ *mass number* ≤ 238 at neutron incident energy of 14.5 MeV.

$$\sigma_{(n,\alpha)}=a_1{*}(A^{1/3}+1)^2{*}e^{\left(-a_2{*}\frac{N-Z-3}{A}\right)} \tag{7}$$

In addition, another formulation took into account the target nuclei mass number in the range $20\le$ *mass number* ≤ 239 [19]. As a result, the phrase $C\sigma_{ne}$ has been substituted by $(A^{1/3}+1)^2$ as:

$$\sigma_{(n,\alpha)}=a_1{*}(A^{1/3}+1)^2{*}e^{\left(-a_2{*}\frac{N-Z}{A}\right)} \tag{8}$$

According to Ref. [20] calculations, the (n, α) reaction cross-sections of 125 target nuclei range of $18\le$ *mass number* ≤ 209 were as follows:

For target nuclei with atomic number ≤50

$$\sigma_{(n,\alpha)}=\pi r_0^2{*}(A^{1/3}+1)^2{*}e^{\left(\left(-a_1\left(\frac{N-Z+1}{A}\right)^2+a_2A^2+a_3f_{sh,p}-a_4\right)\right)} \tag{9a}$$

and for target nuclei with atomic number > 50

$$\sigma_{(n,\alpha)}=\pi r_0^2{*}(A^{1/3}+1)^2A^{-1/3}\left(-a_1\left(\frac{N-Z+0.5}{A}\right)+a_2f_{sh,p}-a_3\right)^3 \tag{9b}$$

Where $f_{sh,p}=(dw_n-\delta_n)-\ (dw_\alpha-\ \delta_\alpha)$, δ_n and δ_α are pairing parameter corrections for nuclei (atomic number, mass number)

Table 1
Characteristics of the produced radioisotopes.

Radioisotope	Half life	Mode of decay %	Mass Excess (MeV)	S_n (MeV)	S_p (MeV)
^{51}Cr	27.70 days	γ (100)	−51.453	9.261	9.520
^{89}Sr	50.56 days	β^- (100)	−86.211	6.364	10.891
^{99}Tc	6.10 h	β^- (100)	−87.191	8.961	6.502
^{131}I	8.01 days	β^- (100)	−87.440	8.570	7.373
^{133}Xe	5.25 days	β^- (100)	−87.640	6.642	9.222
^{137}Cs	3.71 days	β^- (92.53)	−41.933	6.170	5.824
^{153}Sm	46.28 h	β^- (100)	−7.250	5.871	8.590

Table 2
Nuclear models, calculated cross sections, EXFOR [10] and data libraries [11] of cross sections for (n, α) reactions on ^{54}Fe, ^{92}Zr, ^{102}Rh, ^{134}Cs, ^{136}Ba, ^{140}La, and ^{156}Gd at an incident neutron energy range of 14–15 MeV.

		E_n(MeV)	Cross sections (mb)						
			^{54}Fe (n, α)^{51}Cr	^{92}Zr (n, α)^{89}Sr	^{102}Rh (n, α)^{99}Tc	^{134}Cs (n, α)^{131}I	^{136}Ba (n, α)^{133}Xe	^{140}La (n, α)^{137}Cs	^{156}Gd (n, α)^{153}Sm
Nuclear Models	EMPIRE 3.2.3	14.50	145.27	8.46	34.67	6.66	2.61	13.72	1.01
	Talys 1.95	14.50	120.00	15.90	43.90	12.2	2.96	15.10	1.32
Evaluated data libraries	JENDL 5	14.50	87.52	9.49	9.06	2.06	1.32	7.65	3.29
	TENDL 2019	14.50	90.15	11.62	22.25	4.02	0.88	8.59	3.39
	JEFF 3.3	14.50	84.50	10.50	20.95	7.33	2.03	7.74	3.10
Cross section formulas	[16]	14.50	119.27	9.55	13.81	3.95	2.24	3.31	4.16
	[17]	14.50	123.58	9.79	18.69	2.27	2.59	1.83	2.72
	[18]	14.50	30.79	12.10	14.16	8.03	8.30	7.60	8.43
	[19]	14.50	108.64	6.62	8.05	2.00	1.21	1.63	1.75
	[20]	14.50	148.83	7.45	10.46	1.87	2.49	1.45	2.31
	[1]	14.50	91.12	6.93	5.37	1.21	0.75	0.85	0.95
EXFOR data	[22]	14.05	85.33 ± 4.0	–	–	–	–	–	–
	[22]	14.38	94.78 ± 4.1	–	–	–	–	–	–
	[22]	14.63	95.83 ± 4.8	–	–	–	–	–	–
	[24]	14.29	89.4 ± 5.1	–	–	–	–	–	–
	[24]	14.45	90.5 ± 4.6	–	–	–	–	–	–
	[24]	14.58	90.2 ± 4.5	–	–	–	–	–	–
	[24]	14.73	89.9 ± 4.6	–	–	–	–	–	–
	[24]	14.83	93.8 ± 4.8	–	–	–	–	–	–
	[26]	14.00	–	10 ± 0	–	–	–	–	–
	[27]	14.70	–	8.5 ± 1	–	–	–	–	–
	[28]	14.80	–	9 ± 2	–	–	–	–	–
	[31]	14.10	–	21.8 ± 1.7	–	–	–	–	–
	[34]	14.50	–	–	–	–	–	–	3.22 ± 0.48
	[33]	14.20	–	–	–	–	–	–	8.5 ± 1.3
	[35]	14.70	–	–	–	–	–	–	3.1 ± 1

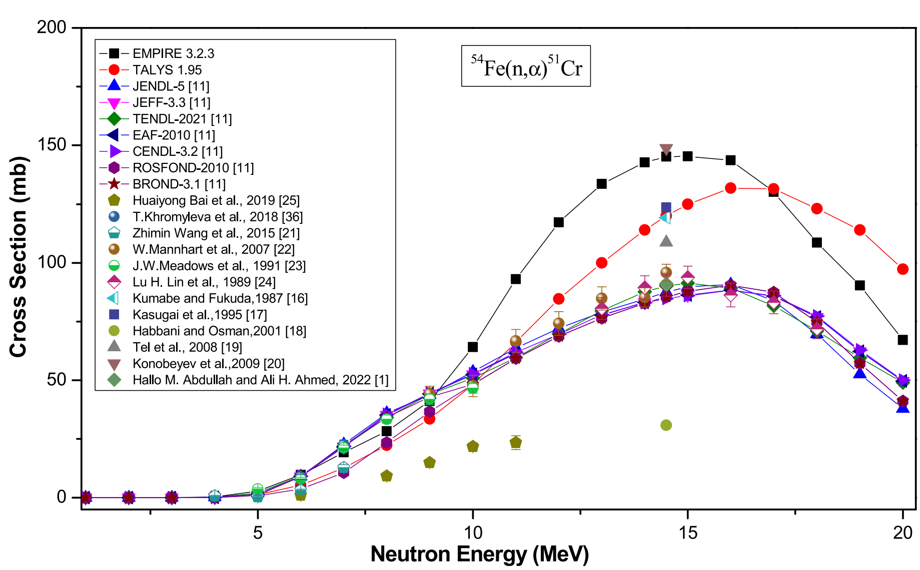

Fig. 1. Excitation function of the nuclear reaction ^{54}Fe (n, α)^{51}Cr by using empirical formulas, nuclear codes compared to experimental data.

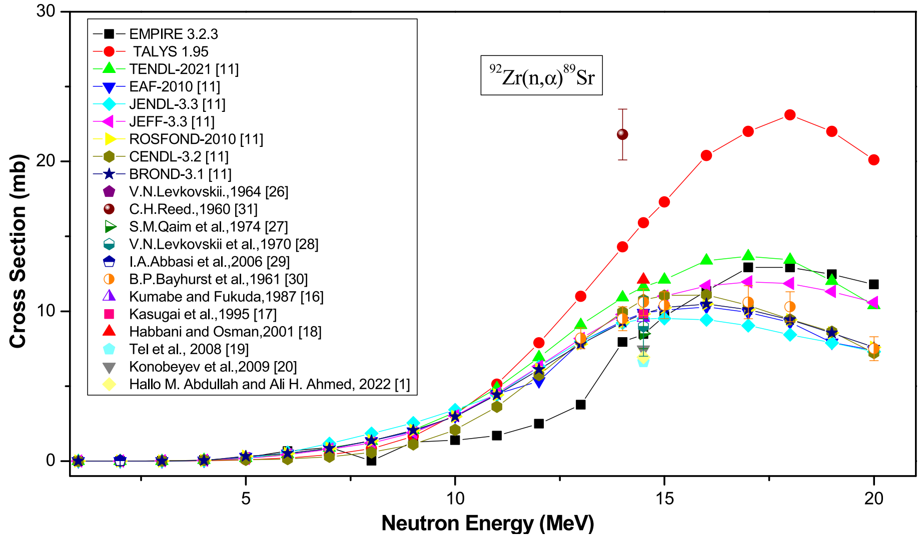

Fig. 2. Excitation function of the nuclear reaction ^{92}Zr (n, α)^{89}Sr by using empirical formulas, nuclear codes compared to experimental data.

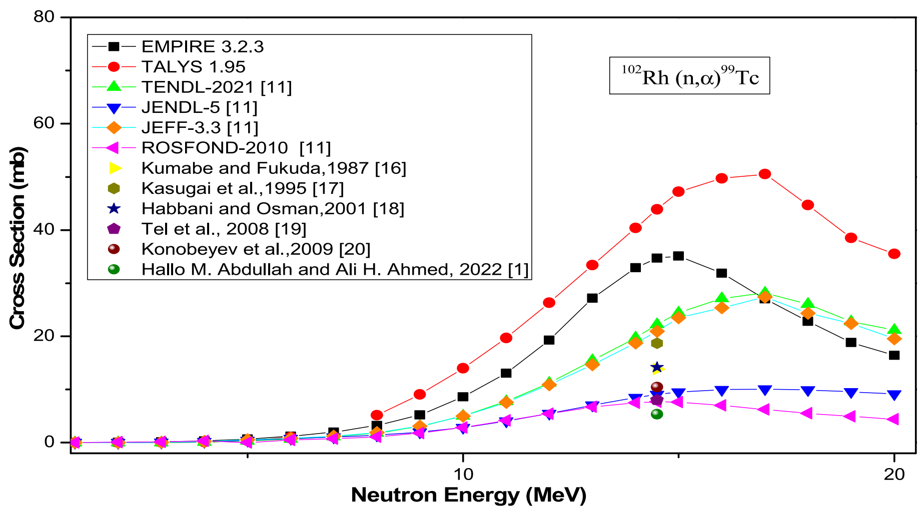

Fig. 3. Excitation function of the nuclear reaction ^{102}Rh (n, α)^{99}Tc by using empirical formulas, nuclear codes compared to experimental data.

and (atomic number −2, mass number −3) that calculated as follows: $\delta = 12A^{-1/2}$ for even-even nuclei, $\delta = 0$ for nuclei with odd A, and $\delta = -12A^{-1/2}$, for odd-odd nuclei, dw_n and dw_α are shell parameter corrections for the nuclei (atomic number, mass number) and (atomic number-2, mass number-3), respectively [20].

Connected to the topic, in our last work, we updated the Levkovskii's formula by replacing the n_e component with one modified by the free fitting parameter $(A^{1/3}+1)^{a_1}$ and introducing the average binding energy utilizing the experimental data for neutron incident energy range between 14 and 15 MeV [1]:

$$\sigma_{(n,\alpha)} = 72.9*(A^{1/3}+1)^{a_1}\left(\frac{BE}{A}\right)e^{\left(-a_2*\frac{N-Z}{A}\right)} \tag{10}$$

where a_1 and a_2 are free fitting parameters.

3. Results and discussion

In this work, radioisotopes including ^{51}Cr, ^{89}Sr, ^{99}Tc, ^{131}I, ^{133}Xe, ^{137}Cs and ^{153}Sm, which are significant in nuclear medicine, are produce through a variety of nuclear reactions. The nuclear properties of produced radioisotopes are shown in Table 1.

In addition to explaining the nuclear reaction itself, (n, α)-reaction excitation functions are required for a number of medical applications. EMPIRE 3.2.3 and TALYS 1.95 codes have been used in this work to compute the excitation functions (cross sections) for producing medical radioisotopes via ^{54}Fe (n, α)^{51}Cr, ^{92}Zr (n, α)^{89}Sr, ^{102}Rh (n, α)^{99}Tc, ^{134}Cs α)^{131}I, ^{136}Ba (n, α)^{133}Xe, ^{140}La (n, α)^{137}Cs and ^{156}Gd (n, α)^{153}Sm reactions. The Constant temperature + Fermi gas model (*ldmodel* = 1 by default) has been included in the TALYS 1.95 code. In addition, the Enhanced Generalized Superfluid Model (EGSM) and the Spherical Optical Model (DIRECT = 0) have been counted upon in the EMPIRE 3.2.3 code. Table 2 lists the numerically calculated cross sections for the (n, α) reactions at an incident neutron energy range of 14–15 MeV.

As a result, the newly computed cross sections are shown in Figs. 1–6 along with the outcomes of earlier experiments and analyzed data files (JENDL 5 [Link], JEFF 3.3 [Link], TENDL 2021 [Link], EAF 2010 [Link], CENDL 3.3 [Link], ROSFOND 2010 [Link] and BROND 3.1 [Link]).

The excitation functions of ^{54}Fe (n, α)^{51}Cr for incident neutron energy range between 1 and 20 MeV were calculated using the EMPIRE 3.2.3 and TALYS 1.95 codes, and the comparison of

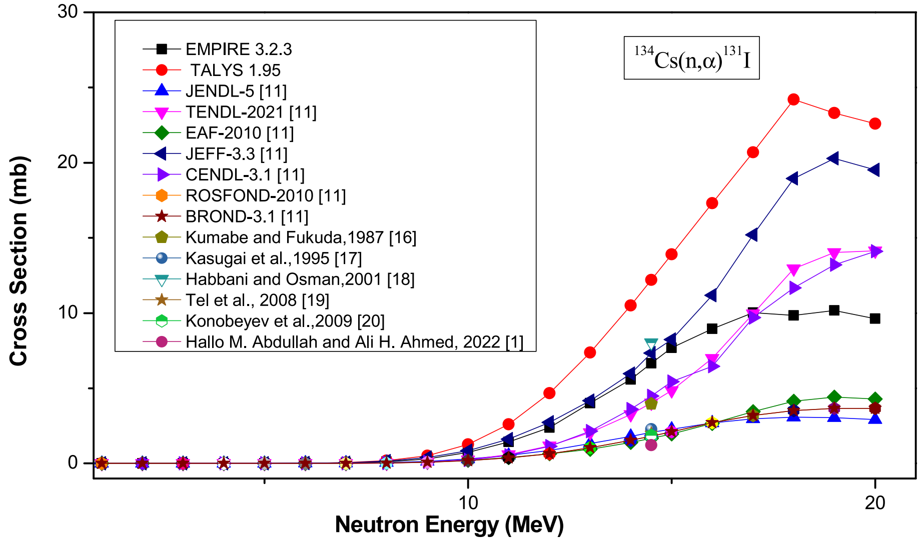

Fig. 4. Excitation function of the nuclear reaction ^{134}Cs (n, α)^{131}I by using empirical formulas, nuclear codes compared to experimental data.

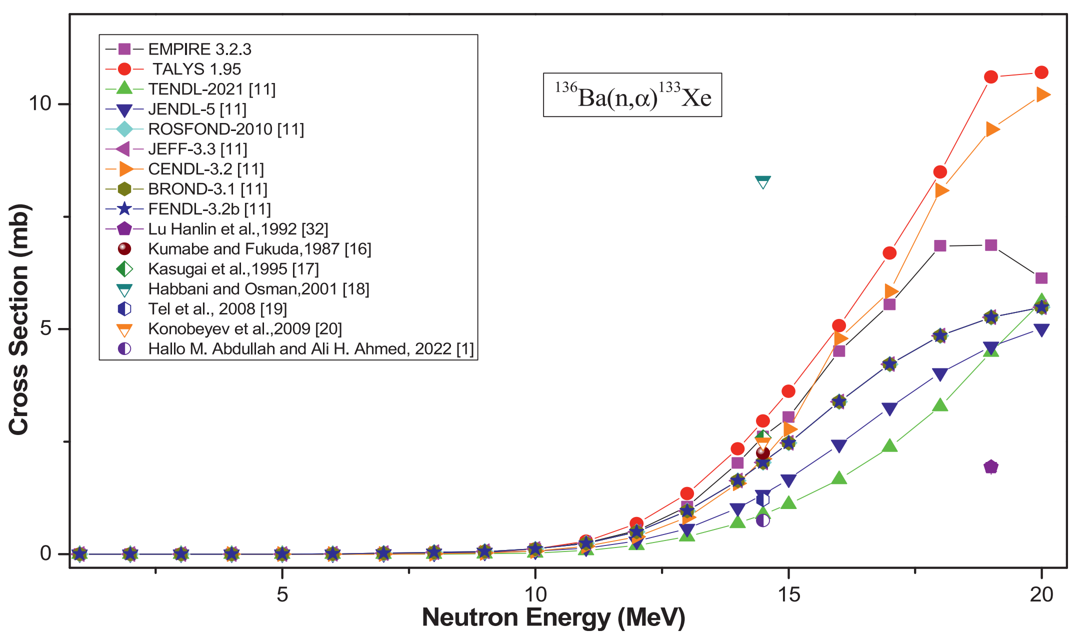

Fig. 5. Excitation function of the nuclear reaction ^{136}Ba (n, α)^{133}Xe by using empirical formulas, nuclear codes compared to experimental data.

experimental data, empirical formulas and evaluated reaction outcomes is shown in Fig. 1.

The calculated cross sections from TALYS 1.95 and EMPIRE 3.2.3 codes, the evaluated data files from (JENDL 5, JEFF 3.3, TENDL 2021, EAF 2010, CENDL 3.3, ROSFOND 2010 and BROND 3.1), the empirical formulas of [1,16,18], and [19] at neutron induced energy 14.5 MeV, are all in good agreement with the available EXFOR data [20–24]. The empirical formulae are well agreement all the experimental data except those from Refs. [18,25].

The calculated and evaluated excitation functions for the ^{92}Zr (n, α) ^{89}Sr nuclear reaction are shown in Fig. 2. The cross-section data from TALYS 1.95 and EMPIRE 3.2.3 codes are close to the evaluated data such as (JENDL 3.3, JEFF 3.3, ROSFOND 2010, CENDL 3.2, BROND 3.1 and TENDL 2021), experimental data and into evaluated and calculated curves of [26–30]. Except the data of TALYS 1.95 at energy range 13–20 MeV and one EXFOR data [31] out of the trend which might be attributed to the use of fewer and older experimental methodologies in their measurement. All empirical formulas get best result near to the calculated and evaluated reaction cross sections.

In Fig. 3, the Excitation functions of ^{102}Rh (n, α)^{99}Tc reaction are shown. The calculated data from EMPIRE 3.2.3 code were near to the evaluated data of TENDL 2021, JENDL 5, ROSFOND 2010 and JEFF 3.3, while those from TALYS 1.95 have discrepancy with the evaluated data at the energy range 10–20 MeV. All the predicted empirical data lie between the calculated and the evaluated ones.

Furthermore, Fig. 4 Depicts the excitation function data-points for the ^{134}Cs (n, α)^{131}I nuclear reaction. Excitation function calculations using EMPIRE 3.2.3 are mainly in agreement with the evaluated cross-section data between 1 and 20 MeV, with also TALYS 1.95 ones at the energy range 12–20 MeV in the out curve. All the predicted empirical data lie between the calculated and the evaluated data.

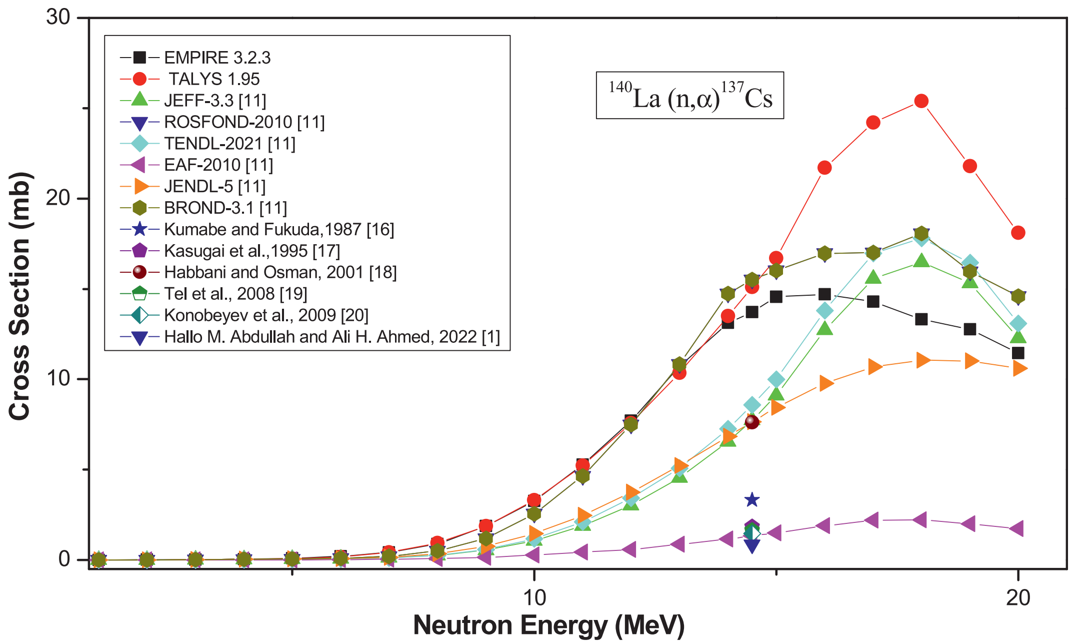

Fig. 6. Excitation function of the nuclear reaction ^{140}La (n, α)^{137}Cs by using empirical formulas, nuclear codes compared to experimental data.

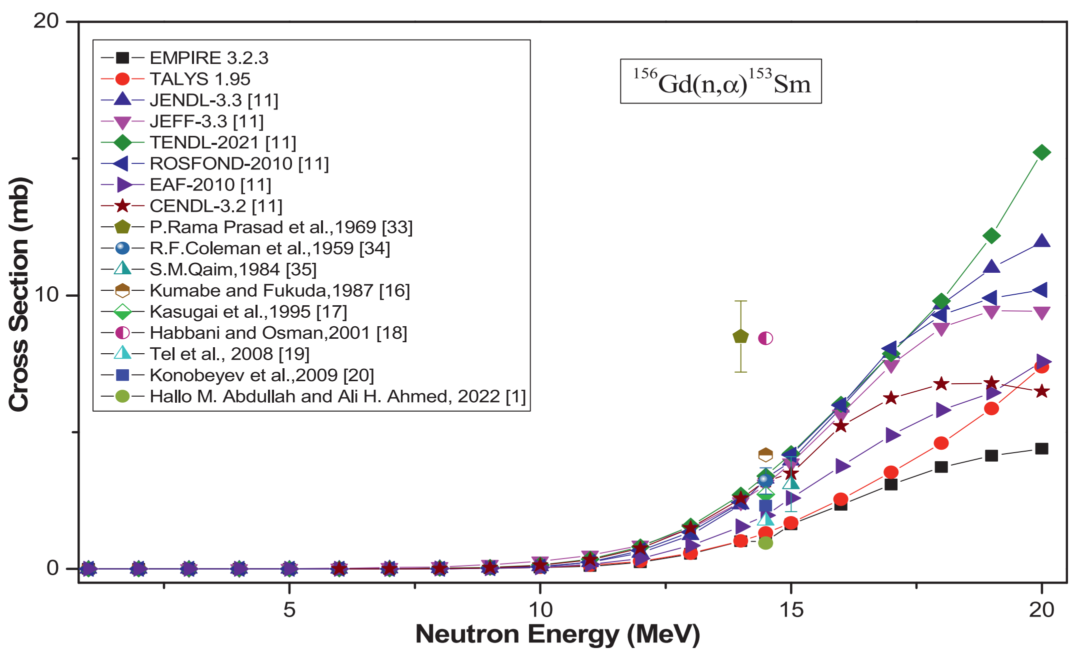

Fig. 7. Excitation function of the nuclear reaction ^{156}Gd (n, α)^{153}Sm by using empirical formulas, nuclear codes compared to experimental data.

The excitation function curve for ^{136}Ba (n, α)^{133}Xe nuclear reaction is illustrated in Fig. 5. The excitation curves obtained using the TALYS-1.95 and EMPIRE-3.2.3 codes often agree well with cross section data in the 1–20 MeV range. Experimental data [32] lie outside the curves, except for [18]. All empirical equations give significant discrepancies at 14.5 MeV.

Fig. 6 Displays the excitation function for the nuclear reaction ^{140}La (n, p)^{137}Cs. There is a scarcity of experimental nuclear reaction data for this reaction. The calculated data show results that are consistent with the evaluated nuclear reaction. but the excitation function from EMPIRE 3.2.3 code is significantly better than the TALYS 1.95 code which lies out of the curves trend at energy range 16–20 MeV. all empirical formulae have significant discrepancies.

Fig. 7 shows the calculated excitation function from EMPIRE 3.2.3 and TALYS 1.95 codes for the nuclear reaction ^{156}Gd (n, α)^{153}Sm in comparison to the evaluated data (JENDL-3.3, JEFF 3.3, TENDL 2021, ROSFOND 2010, EAF 2010 and CENDL 3.2) and EXFOR data such as [33–36]. EXFOR data are relatively few for ^{156}Gd (n, α)^{153}Sm reactions. The calculated data correspond to the calculated data and cross section data in the considered works, except for [33]. All empirical formulas give significant discrepancies, except for the formula from Ref. [18].

4. Conclusion

In this work, the calculation ^{54}Fe (n, α) ^{51}Cr, ^{92}Zr (n, α) ^{89}Sr, ^{102}Rh (n, α) ^{99}Tc, ^{134}Cs (n, α) ^{131}I, ^{136}Ba (n, α) ^{133}Xe, ^{140}La (n, α) ^{137}Cs and ^{156}Gd (n, α) ^{153}Sm nuclear reaction cross sections performed using EMPIRE 3.2.3 and TALYS 1.95 codes. These results are consistent with experimental and calculated data, as well as with data obtained using empirical formulas. The excitation function data obtained using the EMPIRE 3.2.3 code is better than the data obtained using the TALYS 1.95 code. The radioisotopes ^{51}Cr, ^{89}Sr, ^{99}Tc, ^{131}I, ^{133}Xe, ^{137}Cs and ^{153}Sm produced by the studied reactions have important and wide applications in nuclear medicine, and the present results show that they can be obtained using small cyclotrons.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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