



## Original Article

Study on (n,p) reactions of  $^{58}\text{Ni}$ ,  $^{99}\text{Tc}$ ,  $^{99}\text{Ru}$ ,  $^{131}\text{Xe}$ ,  $^{133}\text{Cs}$  and  $^{186}\text{Os}$  radioisotopes used in medicine

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## ABSTRACT

In the last decade, nuclear medicine appears to be a good choice of medicine.  $^{58}\text{Co}$ ,  $^{99}\text{Mo}$ ,  $^{99}\text{Tc}$ ,  $^{99}\text{Re}$ ,  $^{133}\text{Xe}$  and  $^{186}\text{Re}$  are very important radionuclides for nuclear medicine. In this study, the excitation functions of  $^{58}\text{Ni}$  (n, p)  $^{58}\text{Co}$ ,  $^{99}\text{Tc}$  (n, p)  $^{99}\text{Mo}$ ,  $^{99}\text{Ru}$  (n, p)  $^{99}\text{Tc}$ ,  $^{131}\text{Xe}$  (n, p)  $^{131}\text{I}$ ,  $^{133}\text{Cs}$  (n, p)  $^{133}\text{Xe}$  and  $^{186}\text{Os}$  (n, p)  $^{186}\text{Re}$  nuclear reactions were calculated at neutron energies between 1 and 20 MeV using TALYS 1.95 and EMPIRE 3.2 nuclear codes. Furthermore, the cross sections were calculated with the empirical formula derived in our past study at 14–15 MeV. The obtained results were compared with the measured values in EXFOR library, and with the evaluated data of (JENDL-4.0/HE, JEFF-3.3, TENDL-2019, ENDF/B-VIII.0, IRDFF-II, JENDL/ImPACT-18). The results are in good agreement with those of the evaluated data libraries and experimental results and indicates that these radioisotopes can be produced by smaller cyclotrons.

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## 1. Introduction

Nuclear medicine is a branch of medicine that employs radiation to give diagnostic knowledge about personal function or treatment options. Every year, tens of millions of nuclear medicine operations are conducted, and the need for medical radioisotopes is steadily growing [1].

Nuclear medicine involves the use of radioactive materials to examine the function of organs and tissues in the body, as well as to treat and eliminate damaged or diseased tissues and organs. It is used to create images of the heart, liver, bones, thyroid and a variety of other organs, as well as to treat damaged organs and cancer tumours [2]. The effective manufacturing and use of these radioisotopes extends to cancer treatment, heart, and even psychotherapy through imaging techniques that may plan will cover on the function of every significant tissue and organ in the human body [1]. There are a lot of radionuclides that are employed in nuclear medicine are generated in accelerators, nuclear reactors, or cyclotrons, and their manufacture is a critical and an ever problem [3].

Optimizing the radioisotope created requires a thorough understanding of the excitation function, which assists in maximizing

the yield of the desired product while reducing radioactive contaminants [4]. In cyclotrons, the (n, p) reactions play a significant role in the production of isotopes. They benefit from the fact that neutrons are easily accelerated and that they may reach the target nucleus from the proton's direction, reducing Coulomb repulsion. when a proton is also one of the particles emitted. As a reason, theoretical models are most often used to calculate neutron cross-sections when experimental results at certain incidence energies are unavailable due to experimental difficulties [5,6].

In the present study, the cross sections for  $^{58}\text{Ni}$  (n, p)  $^{58}\text{Co}$ ,  $^{99}\text{Tc}$  (n, p)  $^{99}\text{Mo}$ ,  $^{99}\text{Ru}$  (n, p)  $^{99}\text{Tc}$ ,  $^{131}\text{Xe}$  (n, p)  $^{131}\text{I}$ ,  $^{133}\text{Cs}$  (n, p)  $^{133}\text{Xe}$  and  $^{186}\text{Os}$  (n, p)  $^{186}\text{Re}$  reactions were measured in neutron energies up to 20 MeV. For different radioisotopes used in the nuclear medicine listed in Table 1, the EMPIRE 3.2 and TALYS 1.95 codes were used to calculate the excitation functions of neutrons caused by nuclear reactions [7]. The results were compared with theoretical models and experimental data present in the ENDF and EXFOR. In this study, the excitation Function calculations were used to nuclear reaction simulation codes EMPIRE 3.2 and TALYS1.95 [8,9].

## 2. Methods

## 2.1. EMPIRE 3.2 code

EMPIRE 3.2 is a flexible set of nuclear reaction codes that can calculate a wide range of energies and incident particles and

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**Table 1**  
Property of the product radioisotopes.

Reaction product	Half life	Mode of decay %	Mass Excess (MeV)	$S_n$ (MeV)	$S_p$ (MeV)
Co-58	70.86 days	EC(100)	-59.84	8.58	6.96
Mo-99	2.74 days	$\beta^-(100)$	-85.97	5.92	9.73
Tc-99	6.00 h	$\beta^-(100)$	-87.19	8.96	6.50
I-131	8.00 days	$\beta^-(100)$	-87.44	8.57	7.37
Xe-133	5.24 days	$\beta^-(100)$	-87.64	6.64	9.22
Re-186	3.70 days	$\beta^-(92.53)$	-41.93	6.17	5.82

contains a number of nuclear models. The system may be utilized for theoretical nuclear reaction research as well as nuclear data analysis. The code takes into account the major nuclear reaction models, like the optical model, pre-compound, compound, fission and direct reactions developed by the nuclear studies. The pre-equilibrium process proposed in the exciton model is included in the PCROSS module as given by: [10,11],

$$-q_{r=0}(n) = \lambda_+(E, n+2)\tau(n+2) + \lambda_-(E, n-2)\tau(n-2) - [\lambda_+(E, n) + \lambda_-(E, n) + L(E, n)]\tau(n) \quad (1)$$

Where the term  $q_{r=0}(n)$  is the composite nucleus's initial occupancy probability in the state with the exciton number, for particles and  $\gamma$ -rays,  $n$ .  $L(E, n)$  is the overall emission rate integrated across emission energy for particles (neutrons, protons, and clusters) and  $\gamma$ -rays, the terms  $\lambda_-(E, n)$  and  $\lambda_+(E, n)$  are two parameters represent nuclear decay transition rates to adjacent states [8]. The pre-compound spectra at Empire code is given by,

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

where  $\sigma_{a,b}^r(E_{inc})$  is the cross-section of the reaction (a,b) and  $D_{a,b}(E_{inc})$  is the depletion factor.  $W_b(E, n, \epsilon_b)$  represents the probability of a particle emission of sort b (or  $\gamma$ -ray) with energy  $\epsilon_b$  from state with  $n$  exciton and excitation energy  $E$  of compound nucleus (CN) [8].

## 2.2. TALYS 1.95 code

The reaction model code TALYS 1.95 is commonly used in the evaluation of nuclear reaction experiments and nuclear structure. TALYS 1.95 employs a variety of reaction models, including optical, compound, direct, fission models and pre-compound [12]. In the TALYS code 1.95, the nuclear cross-section for the pre-compound particle emission process is as shown in:

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

where the parameter  $\sigma^{CF}$  is the cross section for the compound nucleus formation calculated via the optical model. The parameter  $\tau$  is the average lifetime for the exciton state. The parameters  $h_\pi(h_\nu)$  d ,  $p_\pi(p_\nu)$  represent the proton (neutron) hole number and the proton (neutron) particle number, respectively. Also, the parameters  $E_k$  and  $w_k$  are emission energy and the emission rate of a particle k. The parameter P denotes the part of the pre-compound population for the emission to survive the previous states and passes through the  $(p_\pi, h_\pi, p_\nu, h_\nu)$  configuration, averaged over time [9]. The initial neutron and proton particle numbers are  $p_\pi^0 = N_p$  and  $p_\nu^0 = Z_p$ , respectively with  $Z_p(N_p)$  the proton (neutron) number of

incoming particles [21,33].

## 2.3. Empirical formulas

The nuclear reaction cross-section for various energies have been required for describing nuclear structures, separation energy, average binding energy, and excited nuclear states [10]. Moreover, cross section data is essential for the construction of more advanced nuclear models as well as the description of particle emission process resulting from a nuclear reaction. As a consequence, empirical equations were used to calculate the cross sections. In recent decades, several researchers have investigated and published empirical equations for a variety of reaction channels. In a recent study, we presented an empirical formula for calculating the (n,p) cross sections at 14–15 MeV. As said by a previous study, the cross-section formulae counting isotopic effects, are as follows:

Ait-Tahar (1987) modified the Levkovskil formula for (n, p) reaction cross section at a neutron incident energy of 14 MeV and the target nuclei range  $40 \leq A \leq 239$  [13]:

$$\sigma(n, p) = 0.0437(A^{1/3} + 1)^2 \exp\left[-28.78 \frac{N-Z+1}{A}\right] \quad (4)$$

At the incident energy of neutron equal 14 MeV, Kasugai et al. (1996) proposed a new empirical formula for reproducing the (n, p) reaction cross sections [14]. This formula is a modification of the original Levkovskiii formula, with the nonelastic cross-section portion replaced with one that includes the neutron number minus atomic mass plus one all square.

$$\sigma(n, p) = 3.56048(N-Z+1)^2 \exp\left[-83.7984 \frac{N-Z+1}{A}\right] \quad (5)$$

Broeders and Konobeyev (2006) developed a novel equation to determine the (n, p) reaction cross-sections for 125 target nuclei in the range  $40 \leq A \leq 209$  and  $Z \leq 50$  at a neutron energy of 14.5 MeV [15].

$$\sigma_{(n,p)} = a_1 * (A^{1/3} + 1) * e^{\left(A^{0.5} \left(-a_1 \frac{N-Z+1}{A} + \frac{a_2 Z}{A^{1/3}} - a_3\right)\right)} \quad (6)$$

Yigit (2018) [16] continued to investigate the empirical formula for 91 nuclei with  $9 \leq A \leq 239$  at neutron energy ranges of 14–15 MeV and came up with the following new modified Levkovskil formulae:

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

Also, Ahmed (2018) considered the proton separation energy  $s_p$  as a dependent physical parameter by the (n, p) reaction cross-section along with the mass number of the nuclei (A) in the new formulation [17]. Furthermore, the part  $C\sigma_{ne}$  has been replaced by  $A^2 S_p$  as follow:

$$\sigma_{(n,p)} = a \cdot A^2 \cdot S_p \cdot e^{\left( b \cdot \frac{N-Z}{A} \right)} \quad (8)$$

In our previous study (2022) used the average binding energy as a reliant physical parameter with the neutron number of the target nuclei in the (n, p) reaction cross-section [18]. Besides, the modification in the first term of main empirical formula changes the cross-section of the (n, p) reactions.

So that, the part  $C\sigma_{ne}$  has been replaced by  $(N - Z + 1) \left( \frac{BE}{A} \right) N^{a_2}$  as:

$$\sigma_{(n,p)} = a_1 (N - Z + 1) \left( \frac{BE}{A} \right) N^{a_2} \exp \left[ -a_3 \frac{N - Z + 1}{A} \right] \quad (9)$$

### 3. Results and discussion

In this study, a variety of nuclear reactions to produce radioisotopes such as <sup>58</sup>Co, <sup>99</sup>Mo, <sup>99</sup>Tc, <sup>99</sup>Re, <sup>133</sup>Xe and <sup>186</sup>Re have been investigated, which are important in nuclear medicine, Table 1.

The calculated excitation functions for producing medical radioisotopes from <sup>58</sup>Ni (n, p) <sup>58</sup>Co, <sup>99</sup>Tc (n, p) <sup>99</sup>Mo, <sup>99</sup>Ru (n, p) <sup>99</sup>Tc, <sup>131</sup>Xe (n, p) <sup>131</sup>I, <sup>133</sup>Cs (n, p) <sup>133</sup>Xe and <sup>186</sup>Os (n, p) <sup>186</sup>Re reactions were obtained using the cross sections evaluated by TALYS 1.95 and EMPIRE 3.2.3 nuclear codes. The default set of input parameters utilized for the TALYS 1.95 codes included the exciton model for pre-equilibrium emission, the back-shifted Constant temperature + Fermi gas model for the level density parameter, and the global optical model potential (ldmodel = 1). While in the EMPIRE 3.2.3 code, the Enhanced Generalized Superfluid model (EGSM, level density equal zero: LEVDEN = 0), and the spherical optical model by default (DIRECT = 0) have been dependent. Number of points in the outgoing energy grid (NEX = 80), the quantum statistical Multi-Step-Direct model (MSD = 0), the quantum statistical Multi-Step-Compound model (MSC = 0), the exciton model (PCROSS = 1.5), and the specific spherical optical potential (OMPOT = -523). The evaluated cross sections for (n,p) reactions with neutron energy of 14.5 MeV are shown numerically in Table 2. In which the evaluated data files (JEFF-3.3, JENDL/ImpACT-18, TENDL-2019) are also presented.

Thus, the newly calculated cross sections are included in Figs. 1–6 with the previous experimental data results in addition to the evaluated cross sections. The calculation of excitation function has been extended to neutron energies up to 20 MeV For the neutron induced reaction <sup>58</sup>Ni (n, p) <sup>58</sup>Co by using TALYS 1.95 and EMPIRE 3.2.3 codes, and the comparison of empirical formulas, experimental data and calculated results are shown in Fig. 1, in which best production range was 2–18 MeV.

The calculated values from TALYS 1.95 and EMPIRE 3.2.3 codes are in excellent agreement with the evaluated data files (JEFF-3.3, JENDL/ImpACT-18, TENDL-2019), EXFOR data (Buczko et al., 1995) [19], (Filatenkov, 2016) [20] (Mannhart et al., 2007), [21] (Semkova et al., 2004), and (Xiaolong Huang et al., 1999) [22], and empirical formulas of (Abdullah and Ahmed, 2022) (Kasugai et al., 1996), and (Broeders and Konobeyev, 2006).

Fig. 2 shows the calculated and evaluated excitation functions for the <sup>99</sup>Tc (n, p) <sup>99</sup>Mo nuclear reaction. The cross-section data from TALYS 1.95 and EMPIRE 3.2.3 codes are close to the evaluated data of (JEFF-3.3, JENDL/ImpACT-18, TENDL-2019), and experimental data of (Filatenkov, 2016) [20] (Ikeda et al., 1994), [23], (Qaim, 1973) [24]. But the results of (Golchert et al., 1965) [25] and (Remiar et al., 2009) [26] lied out of the curves. On the other hand, empirical formulas for (mahmud and Ahmed, 2022) and (Kasugai et al., 1996) yields result data near to the calculated and evaluated data.

In Fig. 3, the resulted Excitation function of <sup>99</sup>Ru (n, p) <sup>99</sup>Tc reaction have been shown. The calculated data of EMPIRE 3.2.3 agrees with the evaluated data of JENDL/ImpACT-18 especially at the energy range of 1–12 MeV. The experimental data of (Kanno et al., 1993) [27] and (Kielan et al., 1993) [27] records large incongruities with the calculated and evaluated data at the energy range of 13–16 MeV, which may ascribed to the older experimental techniques used in their measurement. The results of involved empirical formulae shows reasonable agreement except that of (Broeders and Konobeyev, 2006) at 14.5 MeV energy which was far from the trend.

The comparison of the evaluated data (JEFF-3.3, JENDL/ImpACT-18, TENDL-2019), experimental nuclear reaction data, and excitation functions from TALYS 1.95 and EMPIRE 3.2.3 codes for the nuclear reactions <sup>131</sup>Xe (n, p) <sup>131</sup>I have been shown in Fig. 4. Despite the fact that these reactions have few experimental values, more trustworthy results can be achieved. The results of the calculated data follow the trend of the evaluated nuclear reaction model, but the estimation of EMPIRE 3.2 is much better. The experimental data of (Kondaiah et al., 1968) [28] and (Sigg et al., 1976) [29] lies on the curves of evaluated data at 14 MeV. The resulted empirical data of (Mahmud and Ahmed, 2022), (Kasugai et al., 1996), (Yigit, 2018) and (Ahmad, 2018) are better than those of (Broeders and Konobeyev, 2006).

The obtained excitation function curves for <sup>133</sup>Cs (n, p) <sup>133</sup>Xe nuclear reaction are shown in Fig. 5. The excitation curve using TALYS 1.95 and EMPIRE 3.2.3 are mostly in good agreement between 1 and 20 MeV with the evaluated cross-sectional data. All experimental data are in very good agreement except that of (Bormann et al., 1960) [30]. All empirical formulas reveal large discrepancies except that of (Yigit, 2018) at 14.5 MeV.

The resulted excitation function for <sup>186</sup>Os (n, p) <sup>186</sup>Re reactions

**Table 2**

The calculated, Evaluated and Empirical cross sections of the (n,p) reactions on <sup>58</sup>Ni, <sup>99</sup>Tc, <sup>99</sup>Re, <sup>131</sup>Xe, <sup>133</sup>Cs and <sup>186</sup>Os at neutron induced energy of 14.5 MeV.

		Energy (MeV)	Cross sections (in mb)					
			<sup>58</sup> Ni(n, p) <sup>58</sup> Co	<sup>99</sup> Tc(n, p) <sup>99</sup> Mo	<sup>99</sup> Ru(n, p) <sup>99</sup> Tc	<sup>131</sup> Xe(n, p) <sup>131</sup> I	<sup>133</sup> Cs(n, p) <sup>133</sup> Xe	<sup>186</sup> Os(n, p) <sup>186</sup> Re
Nuclear Models	EMPIRE 3.2	14.5	358.48	18.33	57.50	5.763	13.5	3.05
	Talys 1.95	14.5	273.63	18.61	37.4	13.8	14.51	6.79
Evaluated data libraries	JENDL/ImpACT-18	14.5	275.68	12.33	49.39	23.46	10.01	6.87
	TENDL-2019	14.5	312.42	15.15	78.32	7.71	19.47	5.99
	JEFF-3.3	14.5	299.3	13.99	78.35	7.71	10.08	5.99
Cross section formulas	(Ait-Tahar, 1987)	14.5	234.05	23.64	42.27	8.29	9.05	8.75
	Kasugai et al., 1996)	14.5	311.01	16.86	39.42	3.62	4.16	4.13
	Broeders and Konobeyev,2006)	14.5	270	1.65	13.69	0.14	0.23	12.54
	(Yigit,2018)	14.5	176.44	26.44	41.73	11.364	12.16	11.76
	(Ahmad,2018)	14.5	262.10	21.18	55.67	10.79	8.45	12.49
	(Abdullah &Ahmad,2022)	14.5	352	17.39	50.99	7.81	6.25	9.83

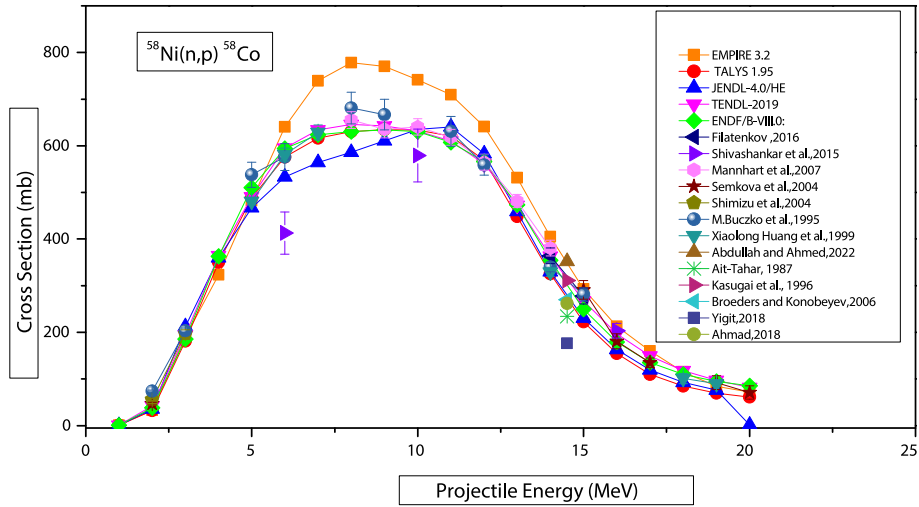


Fig. 1. Excitation function of the  $^{58}\text{Ni}(n, p)^{58}\text{Co}$  reaction by using nuclear codes, empirical formulas and EXFOR data.

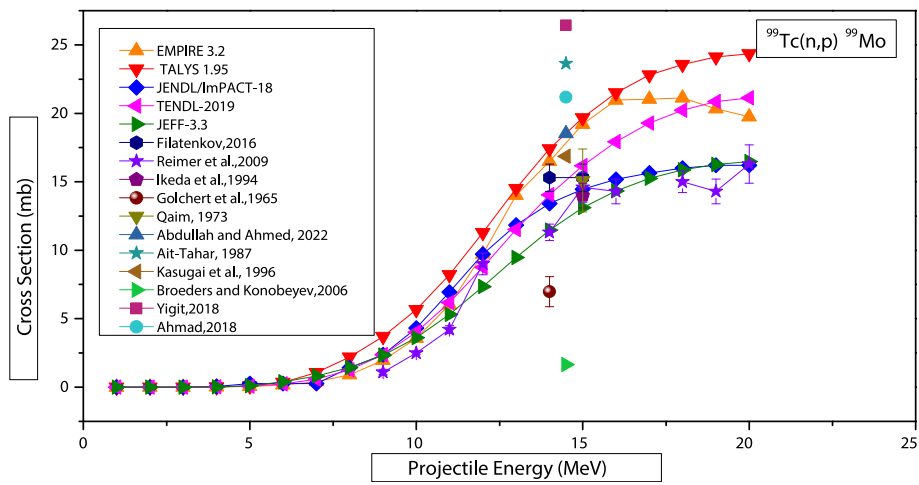


Fig. 2. Excitation function of the  $^{99}\text{Tc}(n, p)^{99}\text{Mo}$  reaction by using nuclear codes, empirical formulas and EXFOR data.

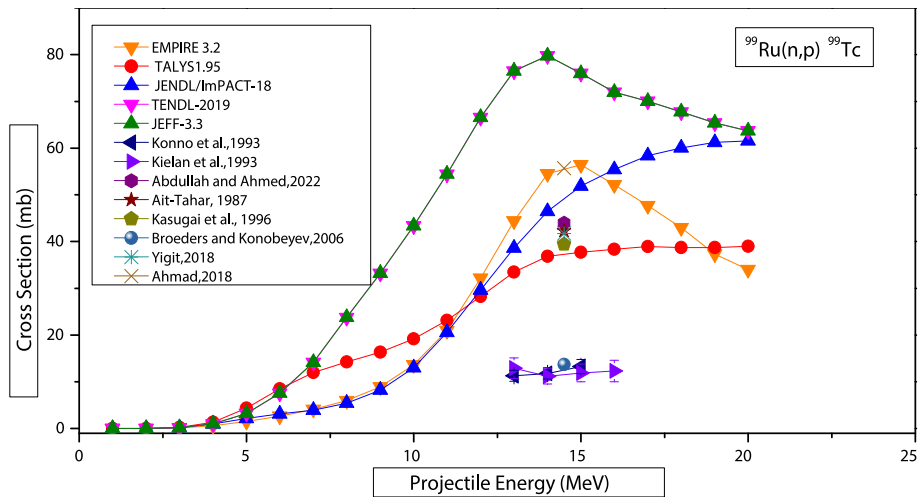


Fig. 3. Excitation function of the  $^{99}\text{Ru}(n, p)^{99}\text{Tc}$  reaction by using nuclear codes, empirical formulas and EXFOR data.

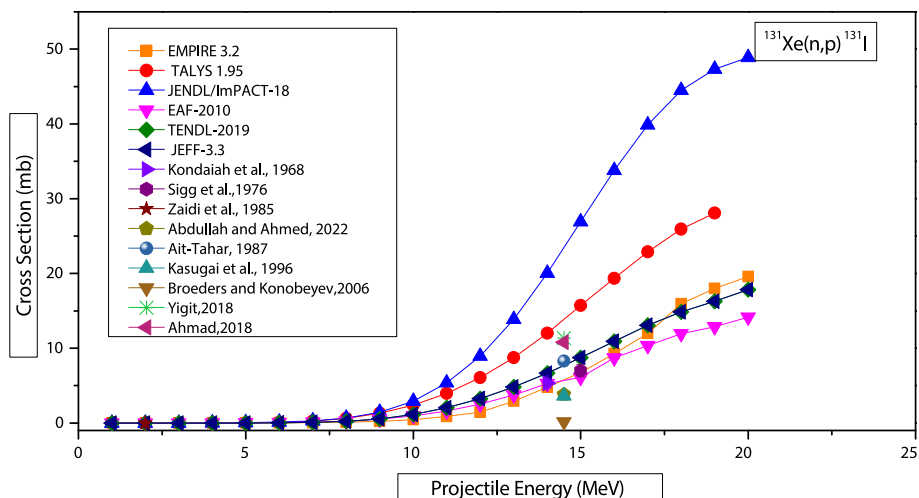


Fig. 4. Excitation function of the  $^{131}\text{Xe} (n, p)^{131}\text{I}$  reaction by using nuclear codes, empirical formulas and EXFOR data.

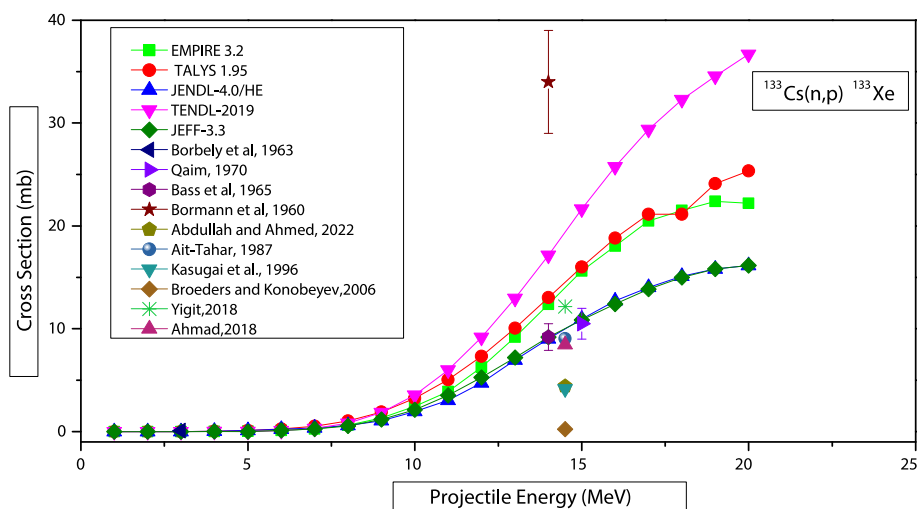


Fig. 5. Excitation function of the  $^{133}\text{Cs} (n, p)^{133}\text{Xe}$  nuclear reaction by using nuclear codes, empirical formulas and EXFOR data.

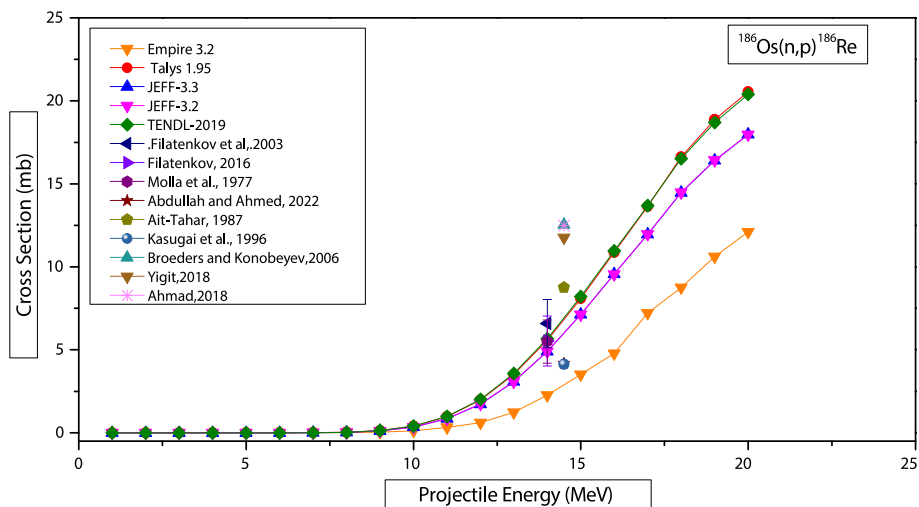


Fig. 6. Excitation function of the  $^{186}\text{Os} (n, p)^{186}\text{Re}$  nuclear reaction by using nuclear codes, empirical formulas and EXFOR data.



are shown in Fig. 6 (experimental nuclear reaction data is scarce). The calculated nuclear reaction model reflects the trend of the evaluated cross-sectional data, although the TALYS 1.95 calculation is significantly better. Except for (Kasugai et al., 1996) at 14.5 MeV, all empirical formulae have considerable discrepancies. All experimental data are in very good agreement except that of (Filatenkov et al., 2003) [31]. The cross sections calculated with TALYS 1.95, EMPIRE 3.2.3 and the experimental nuclear reaction data are often in good agreement.

#### 4. Conclusions

In this study, the calculated excitation functions of  $^{58}\text{Ni}$  (n, p)  $^{58}\text{Co}$ ,  $^{99}\text{Tc}$  (n, p)  $^{99}\text{Mo}$ ,  $^{99}\text{Ru}$  (n, p)  $^{99}\text{Tc}$ ,  $^{131}\text{Xe}$  (n, p)  $^{131}\text{I}$ ,  $^{133}\text{Cs}$  (n, p)  $^{133}\text{Xe}$  and  $^{186}\text{Os}$  (n, p)  $^{186}\text{Re}$  reactions have been carried out using nuclear reaction models of TALYS 1.95 and EMPIRE 3.2 nuclear codes. The results are mostly in a good agreement with the Evaluated data libraries, cross section formulas and experimental results from the EXFOR data. The obtained radioisotopes  $^{58}\text{Co}$ ,  $^{99}\text{Mo}$ ,  $^{99}\text{Tc}$ ,  $^{99}\text{Re}$ ,  $^{133}\text{Xe}$ , and  $^{186}\text{Re}$  obtained have essential and widespread application in nuclear medicine, and the results illustrated in Figs. 1–6 indicate that they can be produced by smaller 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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