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Cross-Section Calculations of Medical Radioisotope ^{64}Cu via some Proton, Neutron and Deuteron Reactions

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ABSTRACT

The Copper-64 radioisotope, whose academic research continues on diagnostic and therapeutic use, was examined in this study. ^{64}Cu radioisotope is unique among other Cu isotopes for medical usage due to its low positron energy, appropriate half-life, and short tissue penetration. In cases where experimental data are missing, cross-section calculations can be used, and the existence of the cross-section data may provide various advantages in managing time, cost, and efficiency. In this context, investigated detailed cross-section calculations of the ^{64}Cu isotope. To this end, cross-sections acquired from various calculation codes were compared with the literature, and alternative production routes were investigated. Using the nuclear reaction codes TALYS and EMPIRE, cross-section data of the ^{64}Cu isotope were obtained from the ^{64}Ni(p,n)^{64}Cu, ^{65}Cu(p,n+p)^{64}Cu, ^{68}Zn(p,n+α)^{64}Cu, ^{65}Cu(n,2n)^{64}Cu, ^{64}Ni(d,2n)^{64}Cu, and ^{63}Cu(d,p)^{64}Cu reactions with the equilibrium and pre-equilibrium models. The results were compared with the available literature data from the EXFOR database.

Keywords: Copper-64, Cross-section, EMPIRE 3.2, Medical Radioisotope, TALYS 1.95.

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Introduction

Radioisotopes are a fascinating and powerful aspect of modern science. These isotopes, which are unstable and emit radiation, have revolutionized industries ranging from medicine to agriculture. Their unique properties enable them to be used in various applications, such as cancer treatment and food preservation, making radioisotopes an essential part of our daily lives. One of the most significant applications of radioisotopes is in medical imaging and cancer treatment. Radioisotopes release radiation that can kill cancer cells, while imaging techniques use radioactive tracers to pinpoint the origin and extent of the disease. Today, the research and cost-effective production of medical radioisotopes are considerable in terms of easy access and the development of cancer treatments. For this purpose, the Copper-64 radioisotope, on which academic research continues for diagnostic and therapeutic purposes, was examined in this study [1, 2]. The element Copper (Cu) exists at 1.4–2.1 mg/kg in the human body, and it is significant because it is the third most abundant metal after Iron and Zinc [3]. Cu-64 is one of the Cu isotopes, with a half-life of 12.7 hours (β+(19%), β−(40%), EC (41%)), and gamma-ray energies of 511 (35%) and 1346 (0.6%) keV, respectively [1]. It is unique among Cu isotopes for medical use due to its low positron energy (650 keV endpoint), appropriate half-life, and short tissue penetration [4]. Thanks to these features and the absence of significant additional radioactive decay, it allows image acquisition from modern Positron Emission Tomography (PET) scanners with an accuracy of a few millimeters. ^{64}Cu is a cyclotron-produced radionuclide used for diagnosis and/or therapeutic (immuno-PET and hypoxia imaging) purposes [2]. At the same time, several articles were published in which ^{64}Cu was used as a radiotracer targeting neuroendocrine, prostate, and hypoxic tumors in cancer imaging [5-9]. It was emphasized that it was a new era-opening radioisotope in PET imaging [3]. In addition to the advantages mentioned, the only disadvantage is that, due to the low branching rate, it needs to be applied in more significant amounts than other commonly used ^{18}F and ^{11}C radioisotopes to obtain the same quality image in the same tissue. There are several methods for producing ^{64}Cu in the literature, but the ^{64}Ni(p,n) reaction is the most commonly used. In this production route, incoming proton energies of 11–14 MeV are hit against an enriched ^{64}Ni target, and the production cross-sections are highest at 11 MeV (max. 600 millibarns) [1]. This production process is also compatible with the low-energy cyclotrons commonly used to produce ^{18}F and ^{11}C. As mentioned earlier, radioisotopes are generally made this way, but the low natural abundance ^{64}Ni is a drawback, making the material expensive [2].

In cases where experimental data are missing, cross-section calculations can be used, which provides more advantages in terms of time and cost. Many studies contribute to the literature with cross-section calculations [10-12]. In this context, detailed cross-section calculations of the ^{64}Cu isotope, which has many uses in the medical field due to the advantages listed above, were investigated. Therefore, different production routes were searched, and cross-sections derived from several calculation programs were compared with the experimental data of the reactions [13-16]. The production cross-section of the ^{64}Cu with the ^{64}Ni(p,n)^{64}Cu, ^{65}Cu(p,n+p)^{64}Cu, ^{68}Zn(p,n+α)^{64}Cu, ^{65}Cu(n,2n)^{64}Cu, ^{64}Ni(d,2n)^{64}Cu, and ^{63}Cu(d,p)^{64}Cu reactions were examined by using the equilibrium and pre-equilibrium models of nuclear reaction codes of TALYS 1.95 [17] and EMPIRE 3.2 [18]. In addition, calculation results and experimental data from the EXFOR [19] data library were compared. Calculations were done using the relative variance analysis method [20] to find the most compatible model with the experimental data. Finally, the results were compared with the most commonly used routes in IAEA (International Atomic Energy Agency) Radioisotopes and Radiopharmaceuticals Reports [1, 2].

Materials and Methods

The cross-section means the probability that a reaction occurs. It may not be possible to take measurements of some short half-lived nuclei. In such cases where the experimental data is insufficient or difficult to obtain, incomplete data can be completed using the results obtained by utilizing nuclear reaction codes. It is also advantageous in terms of time, effort, and cost. Many calculation programs are available to receive the cross-section data of various reactions; TALYS and EMPIRE, used in this study, are two of them. Various nuclear reaction programs are used in many studies in the literature, where more experimental data are needed to determine suitable calculation methods [21-30].

TALYS 1.95 is an open-source, free nuclear reaction analysis and prediction software. The TALYS nuclear reaction code has two related purposes. First, it is a nuclear physics tool to analyze nuclear reaction experiments. The second is to use it as a nuclear data tool by adjusting the reaction models and parameters when no measurements are available. In 2019, TALYS version 1.95 was released. It analyzes and estimates nuclear reactions with target's masses 12 and heavier neutron, proton, deuteron, triton, ^{3}He, and alpha-particle-induced within the 0.001–1000 MeV energy interval [17].

For this study, another program used to calculate the cross-section is EMPIRE 3.2. EMPIRE is a computer program comprising various nuclear models and nuclear reaction calculation codes designed to calculate the energies and particles over a wide range of energy. Deuterons, photons, nucleons, helions (^{3}He), tritons, alpha-particles, and light or heavy ions can be selected as projectiles. The possible energy range extends to several hundred MeV for the chosen particles [18].

A nuclear reaction mechanism depends on the energy of the incoming particle. Therefore, compound nuclear processes predominate in reactions in which incoming particles have energies below 10 MeV. The Hauser-Feshbach theory studies nuclear reactions resulting in the composite nucleus's decay into discrete and continuous states. In this study, equilibrium calculations have been acquired using the Hauser-Feshbach model. The following equation, which is the most basic version of the Hauser-Feshbach formula: denotes the cross-section of the compound nuclear reaction, where the input channel is represented by a, and the exit channel is β, with no spin and no angular momentum [31-32].

$$\sigma_{a\beta} = \pi \lambda_a^{2} (2\ell + 1)\, T_a T_\beta / \Sigma_i T_i \quad (1)$$

Pre-equilibrium calculations were made using the Two-Component Exciton Model, which is highly effective at explaining the high-energy section of the energy spectrum in reactions involving protons, neutrons, and alpha particles with energies between 10 and 60 MeV. However, these models accurately anticipate the emitted particles' angular distributions. In the equation below, the pre-equilibrium cross-section of a particle k with emission energy E_k, the mean lifetime τ of the exciton state, the compound nuclear cross-section σ, and the emission rate W_k are shown [33-34].

$$\frac{d\sigma^{PY}_k}{dE_k} = \sigma^{CE} \sum_{p_\pi = p_\pi^0}^{p_\pi^{max}} \sum_{p_v = p_v^0}^{p_v^{max}} W_k\,(p_\pi, h_\pi, p_v, h_v, E_k)\, \tau(p_\pi, h_\pi, p_v, h_v)\ x\ P(p_\pi, h_\pi, p_v, h_v) \quad (2)$$

The most similar model with the experimental data was determined using relative variance analysis. Equation 1 shows that the model with the slightest difference between the experimental data and the data obtained from the cross-section calculations is the best model that can be selected [20].

$$D = \frac{1}{N} \sum_{i=1}^{N} \left| S^{cal}_i - S^{exp}_i \right| / S^{exp}_i \quad (3)$$

Results and Discussion

The production cross-sections of the ^{64}Ni(p,n)^{64}Cu, ^{65}Cu(p,n+p)^{64}Cu, ^{68}Zn(p,n+α)^{64}Cu, ^{65}Cu(n,2n)^{64}Cu, ^{64}Ni(d,2n)^{64}Cu and ^{63}Cu(d,p)^{64}Cu reactions were investigated up to 60 MeV using TALYS 1.95 and EMPIRE 3.2 nuclear reaction codes with equilibrium and pre-equilibrium models. Moreover, graphical representations of the results are shown in Figs. 1-6. The Two-

Component Exciton model represented the pre-equilibrium state of the TALYS 1.95 program and the equilibrium state by the Hauser-Feshbach model. Similarly, the EMPIRE 3.2 program employed the Hauser-Feshbach model for the equilibrium state and the Exciton Model for the pre-equilibrium state. The relative variance analysis method was utilized to identify the most appropriate reaction and model for producing the ^{64}Cu after comparing the calculated results with the experimental data from the EXFOR data library. Table 1 shows the results of the relative variance analysis. Furthermore, optimal production energy intervals are shown in Table 2.

The experimental data for the ^{64}Ni(p,n)^{64}Cu reaction and the graph of the data obtained from the calculations are shown in Figure 1; experimental data, which is indicated by dots, compared with the calculation results (lines) from the studies of Rebeles et al. [35], Avila-Rodriguez et al. [36], Tanaka et al. [37] and Guzhovskij et al. [38]. The pre-equilibrium models of the EMPIRE and TALYS programs for this reaction follow similar features to the experimental data. Still, the TALYS 1.95 Two-Component Exciton model fits better with the experimental data. In addition, according to the analysis results, as indicated in Table 1, the model most compatible with the experimental data is the TALYS Two-Component Exciton Model. The optimum production energy range of the reaction is shown in Table 2 and is 8→13 MeV.

Figure 2 depicts the data of the ^{65}Cu(p,n+p)^{64}Cu reaction with a two-channel output. Experimental data of the mentioned reaction were taken from the relevant study [39] from the EXFOR database [19]. The graph shows that after 20 MeV proton energy, the TALYS Two-Component Exciton model is in almost perfect agreement with the experimental data. As can be seen from Table 1, the TALYS Two-Component Exciton model is very close to the experimental data. The optimum production energy range of the reaction is 19→24 MeV, as shown in Table 2.

Figure 3 shows another two-output channel reaction through the ^{68}Zn(p,n+α)^{64}Cu. Experimental data was obtained from the study of Hilgers et al. [40]. Again, it is seen that no model could fit the experimental data up to 15 MeV of proton energy. However, after this energy value, it can be said that the Two-Component Exciton Model is more compatible with the experimental data than the other models, and the relative variance analysis also supports this result. This reaction's optimum production energy range is 21→26 MeV (Table 2).

Figure 4 shows the production cross-section of the ^{65}Cu(n,2n)^{64}Cu reaction, which is used to generate ^{64}Cu radioisotopes with the contributions of Mannhart's [41] and Paulsen's [42] studies. EMPIRE Hauser-Feshbach and the EMPIRE Exciton models are consistent with experimental data up to a 14 MeV neutron incident energy value. According to the results of the mathematical analysis, the EMPIRE Exciton model appeared to be the closest model to the experimental data. Therefore, this graph's optimum production energy range is 12→19 MeV.

The results of reaction ^{64}Ni(d,2n)^{64}Cu studied by Daraban et al. and Hermanne et al. [43, 44] were found in a similar feature to the theoretical model results. However, the pre-equilibrium models gave results closer to the experimental data. According to the calculation results in Table 1, the EMPIRE Exciton model gave the results most proximate to the experimental data. As mentioned above, the optimum production energy range of the reaction is 12→16 MeV.

The ^{63}Cu(d,p)^{64}Cu reaction is examined in Figure 6. The EMPIRE Exciton model in the graph perfectly agrees with the experimental data [45, 46]. The optimum generation energy range is 6→9 MeV. According to the calculation results, the EMPIRE Exciton model for the reaction in question gave the closest result to the experimental data.

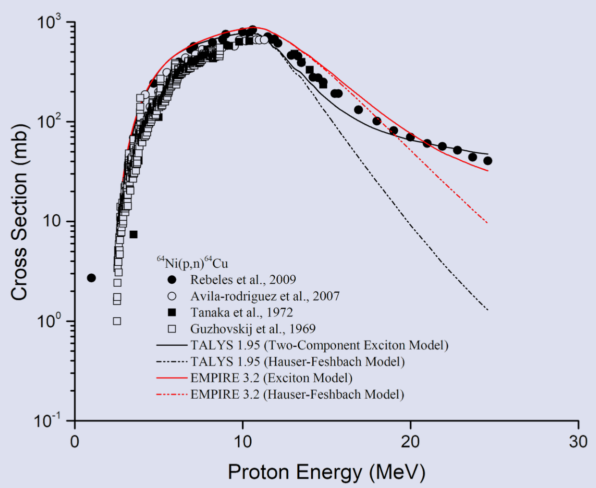

Figure 1. Cross-sections calculations of the ^{64}Ni(p,n)^{64}Cu reaction

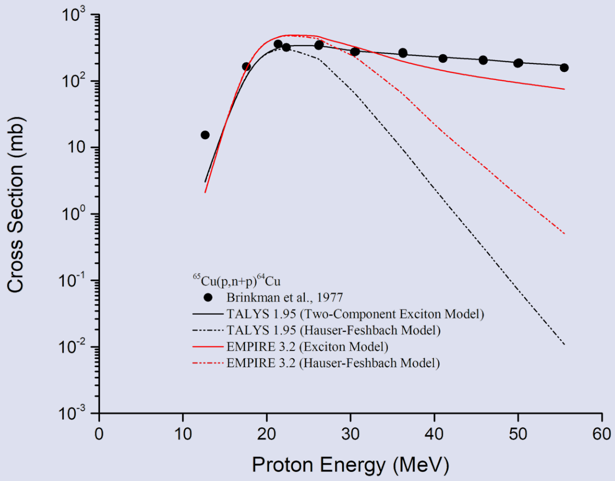

Figure 2. Cross-sections calculations of the $^{65}Cu(p,n+p)^{64}Cu$ reaction

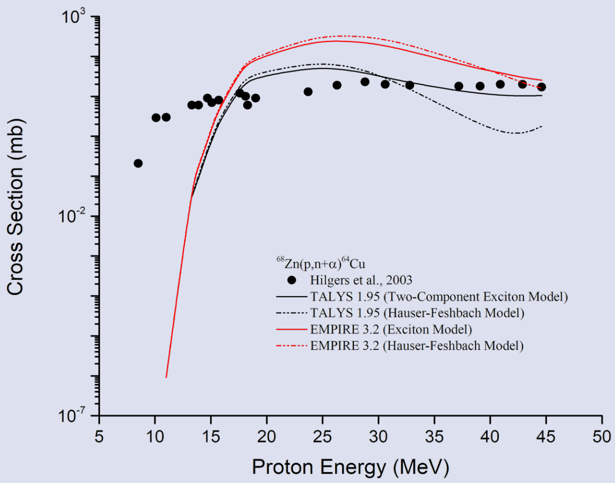

Figure 3. Cross-sections calculations of the $^{68}Zn(p,n+\alpha)^{64}Cu$ reaction

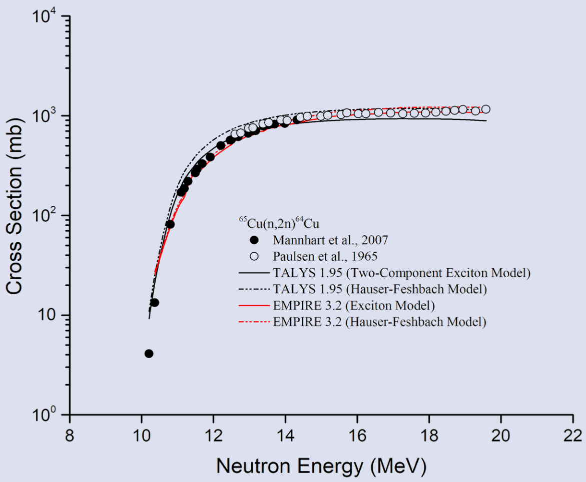

Figure 4. Cross-sections calculations of the $^{65}Cu(n,2n)^{64}Cu$ reaction

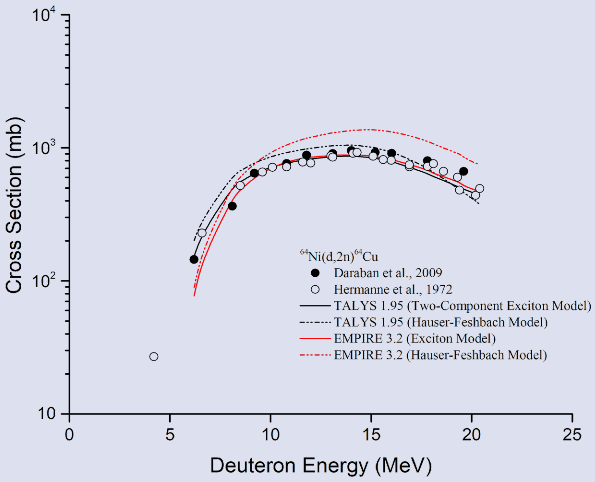

Figure 5. Cross-sections calculations of the $^{64}Ni(d,2n)^{64}Cu$ reaction

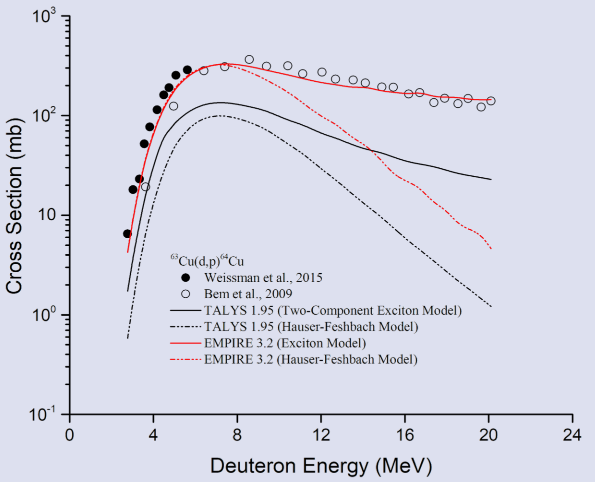

Figure 6. Cross-sections calculations of the $^{63}Cu(d,p)^{64}Cu$ reaction

Table 1. Relative variance analysis of ^{64}Cu production cross–section calculations

Reactions	TALYS Two-Component Exciton	TALYS Hauser-Feshbach	EMPIRE Exciton	EMPIRE Hauser-Feshbach
$^{64}Ni(p,n)^{64}Cu$	0.5982	0.6039	0.6173	0.6171
$^{65}Cu(p,n+p)^{64}Cu$	0.0527	0.6492	0.3759	0.5807
$^{68}Zn(p,n+\alpha)^{64}Cu$	0.8430	1.2297	4.8293	6.3229
$^{65}Cu(n,2n)^{64}Cu$	0.1116	0.1441	0.0650	0.0664
$^{64}Ni(d,2n)^{64}Cu$	0.0772	0.1716	0.0677	0.4702
$^{63}Cu(d,p)^{64}Cu$	0.7788	0.8832	0.3760	0.7472

Table 2. Optimum energy of ^{64}Cu production

Radioisotope	Production Reaction	Optimum Energy Interval (MeV)
^{64}Cu	$^{64}Ni(p,n)^{64}Cu$	8→13
^{64}Cu	$^{65}Cu(p,n+p)^{64}Cu$	19→24
^{64}Cu	$^{68}Zn(p,n+\alpha)^{64}Cu$	21→26
^{64}Cu	**$^{65}Cu(n,2n)^{64}Cu$**	12→19
^{64}Cu	$^{64}Ni(d,2n)^{64}Cu$	12→16
^{64}Cu	$^{63}Cu(d,p)^{64}Cu$	6→9

Conclusion

In this study, to contribute to the development of the ^{64}Cu radioisotope production routes, the production cross-sections of the $^{64}Ni(p,n)^{64}Cu$, $^{65}Cu(p,n+p)^{64}Cu$, $^{68}Zn(p,n+\alpha)^{64}Cu$, $^{65}Cu(n,2n)^{64}Cu$, $^{64}Ni(d,2n)^{64}Cu$ and $^{63}Cu(d,p)^{64}Cu$ reactions have been examined up to 60 MeV via equilibrium and pre-equilibrium models with TALYS 1.95 and EMPIRE 3.2 nuclear reaction codes in where the graphical representations of the outcomes are given in Figs. 1-6. Two-Component Exciton Model was used in the TALYS 1.95 program for the pre-equilibrium state, while the Hauser-Feshbach model was used for the equilibrium state. Similarly, in EMPIRE 3.2 program, Exciton Model was used for the pre-equilibrium state, while the Hauser-Feshbach model was used for the equilibrium state. Calculated results have competed with experimental data taken from the EXFOR data library.

Considering the reactions examined in this study, the cross-section calculation results obtained with the pre-equilibrium models rather than the equilibrium models are compatible with the experimental data. On the other hand, the Hauser-Feshbach models describing the equilibrium state disagreed with the experimental results, except for Figures 4–5.

Conflicts of Interest

There are no conflicts of interest in this work.

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