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ENERGY-DEPENDENT CROSS-SECTIONS FOR PARTICLES INDUCED REACTIONS ON NICKEL FOR THE PRODUCTION OF THERANOSTIC ISOTOPES ^{64}Cu AND ^{67}Cu

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ABSTRACT

The study of how a nuclear particles interaction cross-sections change with varying energies is becoming more important, because of their use in targeted radionuclide therapy (TRT) and nuclear imaging. The need for detailed study of interaction data for nuclear particles, particularly within the low energy range (0–25 MeV) is apparently overlooked. This paper employed GEANT4, a C++-based Monte Carlo simulation toolkit, to investigate the energy-dependent behavior of alpha particles, deuterons, and protons in natural Nickel and to model nuclear reactions involved in the production of theranostic radionuclides, specifically ^{64}Cu via ^{66}Ni (p, n) reaction, ^{67}Cu through ^{66}Ni (d, n) & ^{66}Ni Ni-62(a, t) reactions. The result show the shortest mean free path (MFP) of 2.697 cm at 7.5 MeV, 3.87 cm at 17.5 MeV and 4.012 cm at 17.5 MeV for alpha deuteron and proton respectively. The maximum cross section per atom (CPA) of 4.565 b at 7.5 MeV for *α,* 3.356 b at 17.5 MeV for *d* and 2.73 b at 17.5 MeV for *p*. The maximum total cross section (TCS) for *α* is 1.4175 b at 25 MeV, 1.773 b at 17.5 MeV for *d* and 1.17 b at 25 MeV for *p.* The Excitation functions for the reaction channels; ^{64}Ni (p, n) ^{64}Cu and ^{66}Ni (d, n) ^{67}Cu were compared with Experimental nuclear data (EXFOR) and Evaluated nuclear data (ENDF) libraries with good agreements. Theranostic applications utilize Cu-64 and Cu-67 as important pair radionuclides.

Keywords: energy-dependent, cross section, GEANT4, particle transport, theranostics

1.0 INTRODUCTION

Nuclear reactions occur when an incident particle interacts with a target nucleus, leading to the formation of new radionuclides or the emission of particles or radiation. The outcomes of such interactions depend on factors such as the particle energy, the nature of the projectile, and the type of reaction process involved [1] (Zubaida & Ahmad, 2024). When particles traverse matter, they interact with its constituents—electrons and nuclei—through processes such as scattering or absorption, both of which involve energy transfer or loss [2] (Loveless et al., 2020). The variety of these interactions, determined by parameters like charge, energy, and distance from the target's nucleus, highlights the complexity of particle-matter interactions. From a macroscopic viewpoint, particles may experience phenomena such as straggling, where slight deflections occur, or strong scattering events like Compton scattering. Depending on their energy and the medium, particles might interact multiple times, interact once, or pass through without interacting [3] (Alharbi, 2012). Quantum mechanics governs these interactions inherently, making precise predictions for individual particles impossible. Instead, statistical studies involving a large number of particles are necessary to observe global interaction behaviors [4] (Wilson, 1991).

The transport of nuclear particles, characterized by their movement from a source through a target material, plays a critical role in enabling nuclear reactions. The specific characteristics of each particle, such as mass, charge, and energy, dictate their interaction mechanisms. For instance, both Coulomb and nuclear forces influence protons due to their charge, while only nuclear forces affect neutrons due to their neutrality, allowing for deeper penetration and lower reaction thresholds. Similarly, alpha particles and deuterons exhibit unique transport behaviors, particularly at mid-to-high energies, where they engage in complex interactions like inelastic scattering, nuclear excitation, and particle emission. Despite the advancements in nuclear reaction theories and the expansion of global nuclear reaction databases, the need for new experimental data and consistent theoretical computations remains pressing [5] (Usman & Ahmad, 2022). The demand for diagnostic, therapeutic, and theranostic radionuclides has increased, emphasizing the necessity for accurate and comprehensive nuclear data. Such data enable the optimisation of production techniques for these radionuclides, particularly those involving alpha- and deuteron-induced reactions, which are pivotal in low-energy experiments conducted using cyclotrons and accelerators [6] (Ahmad & Koki, 2017).

A critical gap exists in understanding low-energy nuclear reactions involving nickel, particularly in terms of accurate cross-section data for interactions involving alpha particles, deuterons and protons. Addressing this gap is essential for advancing our understanding of particle transport and nuclear interactions in nickel. Furthermore, the study contributes to optimizing parameters for the enhanced production of theranostic isotopes, specifically ^{64}Cu and ^{67}Cu, which are critical for medical applications. Previous works have explored various aspects of particle interactions with nickel. For instance, **Poignant *et al*. (2016)** modelled the ^{64}Ni (p, n) ^{64}Cu reaction for the South Australian Health and Medical Research Institute (SAHMRI) Cyclotron system using GEANT4 simulations using proton reaction focusing more on product yield than particle behavior. Garrido *et al*. investigated cross-sections for proton-induced reactions on natural nickel up to 70 MeV using TALYS code, reporting good agreement with existing literature. Choinski *et al*. demonstrated ^{67}Cu production through direct ^{67}Ni (p,n) reactions, but challenges with purity suggested irradiation of natural zinc as a more efficient method [7] (Choiński & Łyczko, 2021). **Hibstie *et al*. (2019)** explored alpha-induced reactions on nickel focusing on production cross sections, while **Dauda *et al*. (2017)** reported neutron-induced excitation functions for structural materials, including nickel, in the Nigerian Research Reactor focusing on high threshold energies. Earlier, **Takacs *et al*. (1997)** measured activation cross-sections for Cu-61 production using natural nickel for nuclear medical applications but faced product purity challenges. This study aims to provide a comprehensive analysis of cross-section variations for alpha particles, deuterons, and protons interacting with nickel across the energy range of 0–25 MeV. By bridging the existing data gaps, this work enhances the understanding of particle transport phenomena and facilitates the optimisation of theranostic isotope production, contributing significantly to the fields.

2.0 THEORETICAL BACKGROUND

This study examines the interactions of alpha particles, deuterons, and protons with nickel (Ni) nuclei, focusing on their transport properties and cross sections. When high-energy particles interact with a nickel target, various physical processes occur, contingent upon the particle energy. **Elastic scattering** deflects particles without significant energy transfer, while **Inelastic scattering** involves nuclear excitation, often leading to particle emission or nuclear reactions [8] [9] (Garrido *et al*., 2021).

The mean free path is the average distance a particle travels before interacting with a nucleus. It is inversely proportional to the macroscopic cross section, indicating that shorter mean free paths correspond to higher interaction probabilities. Mathematically, it is expressed as:

$$\lambda = \frac{1}{\sum} \qquad (1)$$

where λ is the mean free path and $\sum$ is the macroscopic cross section [9] (Luoni *et al*., 2021)

In nuclear physics, the cross section quantifies the probability of a specific interaction (e.g., scattering, absorption) between an incoming particle and a target nucleus. It is given as:

$$\sigma = \frac{Number\ of\ interaction\ events\ per\ nucleus}{Number\ of\ incident\ particles\ per\ unit\ time} \qquad (2)$$

Cross sections are typically measured in barns (1 barn = 10^{-24} cm^2) [10] (Zubaida & Ahmad, 2019)

The total reaction cross section combines all possible reaction channels (elastic, inelastic, and capture processes). For a transparent sphere target, it is expressed as:

$$\sigma_R = \pi R^2 (1 - T) \qquad (3)$$

where R is the radius of the nucleus and T, the transparency parameter, is given by:

$$T = \frac{\left[1 - (1 + 2\sum R)\, e^{-2R\sum}\right]}{2R^2 \sum^2} \qquad (4)$$

while

$$R = R_0 \left(A_p^{\frac{1}{3}} + A_T^{\frac{1}{3}} \right) \qquad (5)$$

where R_0 is reduced nuclear radius parameter, A_p and A_T are the mass numbers of projectile and target, respectively. The macroscopic cross-section Σ, measured in cm^{-1}, indicates the probability of interaction per unit length. It relates to the microscopic cross section (σ) and target material density (ρ) as:

$$\Sigma = \sigma \rho \qquad (6)$$

For a hard sphere target nucleus or zero mean free paths, the total nuclear reaction cross section is given as;

$$\sigma_R = \pi {R_0}^2 \left(A_p^{\frac{1}{3}} + A_T^{\frac{1}{3}} \right)^2 \qquad (7)$$

Hadronic Models

The intranuclear cascade model simulates the initial stages of nuclear reactions, where incident particles interact with nucleons in the target nucleus. The Bertini cascade model applies to intermediate energy ranges, simulating secondary particle production due to nucleon interactions. The precompound model describes the formation of an excited nucleus prior to compound nucleus (CN) formation, essential for predicting decay and particle emissions. The evaporation model handles de-excitation of nuclei, simulating particle emission and final states, which are crucial for modeling radioisotope production.

3.0 METHOD

This study employs simulation tools to model particle (alpha, deuteron and proton) interactions with nickel nuclei and calculate cross sections for the energy range of 0 to 25

MeV. The methodology involves the selection of the GEANT4 Physics List, which incorporates the Intranuclear Cascade, Bertini Cascade, Precompound, and Evaporation models. These models simulate different reaction stages, from initial nucleon interactions to final de-excitation. The GEANT4 particle generator generates a monoenergetic particle beam of alpha particles, deuterons, and protons. The particle interactions with nickel nuclei were tracked, including scattering, nuclear excitation, and particle emission. Based on the selected models, we compute reaction cross sections and derive parameters such as the mean free path and cross-section per atom and total cross-section.

The simulation output includes data on energy deposition, secondary particle production, and cross-section variations. The simulation was repeated to ensure statistical reliability. We then compare the simulation results with EXFOR and ENDF libraries from IAEA data base to ensure accuracy and consistency.

For this paper, all uncertainties were considered uncorrelated hence, quadratically summed according to error propagation laws to get total errors. Furthermore, while some of the error sources are common to all data, others affect individual reactions. Yet, the cumulative uncertainties: statistical uncertainties of number of events (0.5 – 5%), systematic uncertainty of the beam flux (~6%) and the uncertainty of number particles generated (4 – 6%). The overall uncertainties of the cross section evaluation were in the range of 8 – 15%, and in GEANT4 code, the Analysis Manager class automatically records such errors.

4.0 RESULTS AND DISCUSSION

The observed trends highlight differences in how alpha, deuteron, and proton projectiles interact with Nickel as a target, using three different parameters which include mean free path, cross section per atom and total reaction cross-section. These differences could arise from variations in Coulomb barrier effects, resonance states, and reaction thresholds, for fig. 1 to fig. 3. For fig. 4 and fig.5, they present Excitation functions for production of theranostic pairs ^{64}Cu and ^{67}Cu.

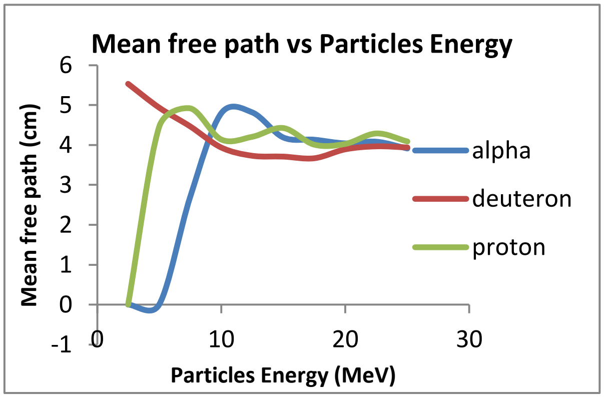

Figure 1- Mean free path variation for alpha particles, deuteron and proton on Nickel–particle reactions

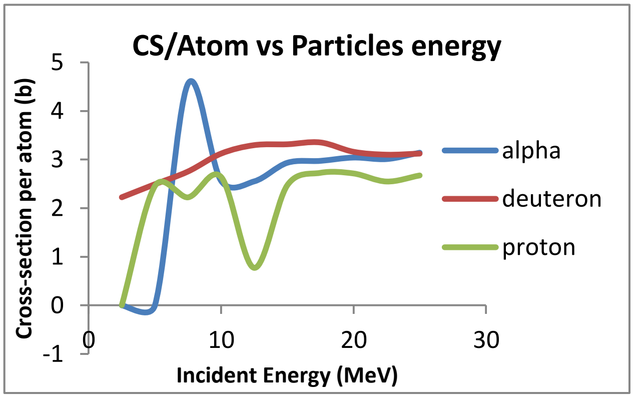

Figure 2- Cross section per atom variation for alpha particles, deuteron and proton on Nickel–particle reactions

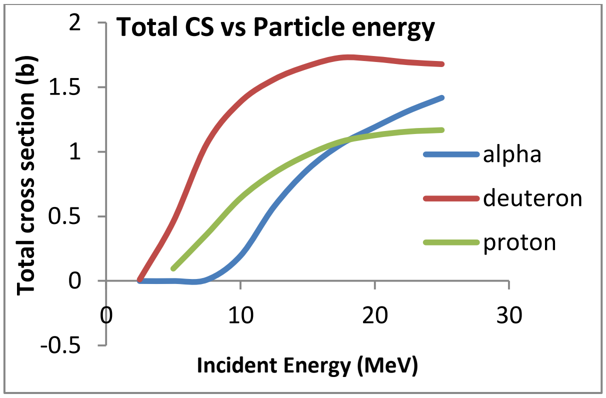

Figure 3- Total cross section variations for alpha particles, deuteron and proton on Ni – particle reactions

In figure 1, alpha particles exhibited zero mean free path up to 5 MeV indicating no interactions at this range, for proton, 2.5 MeV level exhibit zero MFP while deuteron starts with large MFP of 5.5329 cm at 2.5 MeV. Alpha particles MFP commenced with least value of 2.695 cm indicating commencement of nuclear interactions which decreases with rising MFP up to high energies and lowered slightly at 25 MeV implying level off of interactions at high energy. For deuteron the MFP steadily decreases from 5.5329 cm as incident energy increases up to 20 MeV where it rises slightly to 3.8953 cm indicating steady rise of interactions as deuteron energy increases. From 20 – 25 MeV the MFP was steady at 3.9 cm.

Proton similarly maintained steady MFP from 4.45 cm at 5 MeV to 4.01 cm at 20 MeV indicating steady rate of nuclear interactions. Then MFP rose steadily up to 4.29cm at 22.5 MeV and grounded to 4.09cm at 25 MeV symbolizing that interactions have plateaued at this point.

In figure 2 alpha particles of CPA began at 7.5 MeV with a large value of 4.5608 b which plunges in concert with incident energy to 2.9848 b at 15 MeV and then rises up to 3.142 b at 25 MeV indicating continued interactions. Comparatively, deuteron commences showcasing of CPA at 2.5 MeV with 2.2227 b which rose steadily up to 25 MeV with minor undulations, this implies sustained nuclear interactions while proton, commencing with 2.4568 b at 5 MeV, the CPA decreased to 0.7705 b at 12.5 MeV. This activity may apparently be due to Coulomb barrier effects which cause repulsion of protons at this energy. The CPA then rises again steadily up to 25 MeV where it stood at 2.6755 b.

Figure 3 shows that the total cross section for proton was zero at 2.5 MeV and up to 5 MeV for alpha particles, deuteron comparatively exhibited cross sections at all incident energy levels commencing with 0.011422 b at 2.5 MeV and rising steadily to peak of 1.7282 b at 17.5 MeV and then plummets down to 1.6779 b at 25 MeV indicating de-excitation of compound nucleus in this energy range. Alpha particles TCS began at 7.5 MeV with 0.0095 b rising steadily to its peak of 1.4175 b at 25 MeV indication continuous interactions up to peak energy. Comparatively, proton TCS started at 5 MeV with 0.094361 b and also due to continued interactions, rose steadily up to 25 MeV indicating that compound nucleus formation lies beyond the peak energy of the reaction.

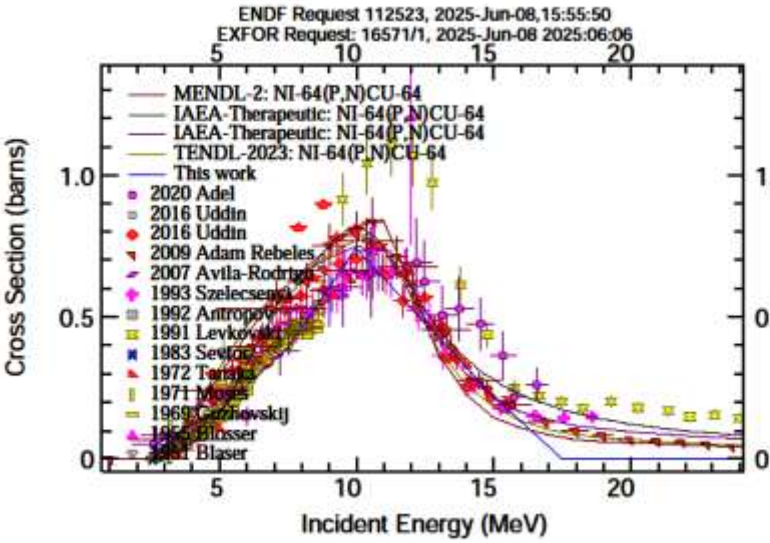

Figure 4- Excitation function for ^{64}Cu production from ^{64}Ni (p, n) reaction

Figure 4 shows that the current simulation is in good agreement with experimental data libraries including Blaser (1951), Blosser (1951), Guzhovskiskij (1969), Moses (1971), Tanaka (1972), Sevior (1983), Levkovski (1991), Antropov (1992), Szelencsenyi (1993), Avila-Rodrigh (2007), Adam (2009), Uddin (2016) and Adel (2020) and evaluated datafiles MENDL- 2 and TENDL 2 (2023). Cu-64 has a half-life of 10.701 hours, β^- energy of 191 keV (38.5%), β^+ energy of 278 keV (17.5%) and an average gamma emission of 1348.77 keV(International Atomic Energy Agency [11] (IAEA), 2021) . This makes ^{64}Cu a good candidate for theranostic application being able to perform therapeutic function because of its β emissions and can be used for SPECT on account of its γ emission.

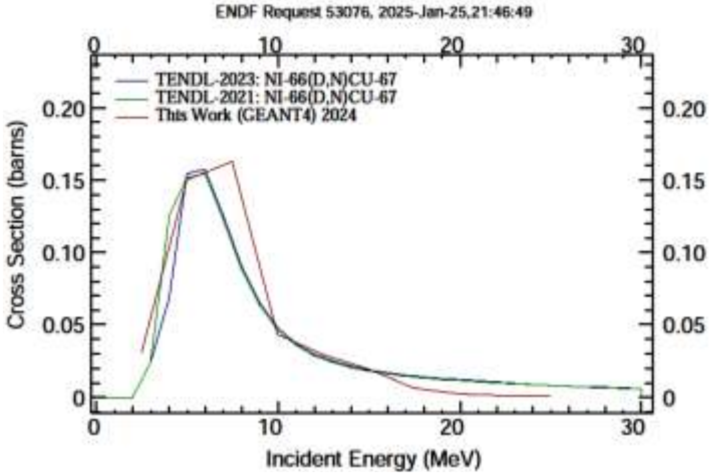

Figure 5- Excitation function for ^{67}Cu production through ^{66}Ni (d, n) reaction

Figure 5 shows deuteron reactions on^{66}Ni 66 for the production of ^{67}Cu. This work's result was compared with TENDL-2021 and TENDL-2023 works from ENDF-NDS library, all of them were in good agreement. ^{67}Cu has half-life of 61.83 hours and is a β- emitter with average electron energy of 141 keV and gamma emission of 180.6 keV [12] (Mou et al.,

2022) It can absolutely serve as a good theranostic as the gamma can be used for SPECT while β- emission can be used for radiotherapy.

The reaction channel ^{66}Ni (a, t)^{67}Cu in the current work's incident energy region of 0 – 25 MeV exhibited zero cross sections at lower energies up to 20 MeV while showing cross sections of 0.00526 barns and 0.00567 barns for energies 22.5 MeV and 25MeV respectively. Hence the result indicated in this reaction channel shows inadequacy such that full excitation function evaluation is not possible until simulation at higher energies is conducted.

5.0 CONCLUSION

The simulations of interactions of alpha particles, deuteron and protons have been conducted using GEANT4 computational code. Fundamentally, the work has unveiled the energy dependent variation of alpha, deuteron and proton reactions which provides further advance into the understanding of low energy particle transport behavior in nuclear targets. Further, in line with the objectives of this work, theranostic radionuclides ^{64}Cu and ^{67}Cu have been produced via ^{64}Ni (p, n) ^{64}Cu and ^{67}Ni (d, n) ^{67}Cu respectively, while for ^{67}Ni (a, t) ^{67}Cu reaction, higher energy simulation should be conducted to generate full excitation function. Copper radionuclides are among the most important ones in nuclear medicine for the presence of the element in many body tissues and its contribution in many metabolic processes. The scarcity of Cu radionuclides especially ^{67}Cu, and ^{64}Cu seriously hamper their application in theranostic procedures thereby limiting access of patients to the high resolution imaging of the procedure as well as the effective, efficient therapy which affords reduced cost and treatment time to patients. This work will go a long way towards tackling the problem of copper radionuclide scarcity.

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