

Investigation of Nuclear Reaction Cross Section by Proton, Deuteron, and alpha Particles Induced Reaction for the Production of Important Radioisotopes for PET Imaging

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Abstract: The study aimed to investigate the nuclear reaction cross section of reaction induced by proton, deuteron, and alpha-particle for the formation of radionuclides which are used in PET imaging. Positron emission tomography (PET) is an imaging technique that plays an important role in the diagnosis and monitoring of cancer cells using appropriate positron-emitting radionuclides. Production of medical radioisotopes is one of the most important tasks in the field of nuclear technology. To produce medical radioisotopes such as: $^{44}_{Sc}$, $^{64}_{Cu}$ and $^{124}_{I}$ which are used in PET imaging, the reaction channels $^{44}_{Ca}(p,n)^{44}_{Sc}$, $^{64}_{Ni}(d,2n)^{64}_{Cu}$ and $^{121}_{Sb}(\alpha,n)^{124}_{I}$ with proton, deuteron and alpha particle energy ranges from 10MeV to 70MeV were selected. For the theoretical calculation of the nuclear reaction cross section EMPIRE nuclear reaction model code was employed and the experimental data were taken from EXFOR data source. It's found that the theoretical result fits nicely with the experimental value within the selected energy range.

Keywords: Cross section, nuclear reaction, medical Radioisotopes, PET imaging.

1 Introduction

Nuclear physics is the field of physics that studies the building blocks and interaction of atomic nuclei. Most nuclear physicist researches to investigate nuclear energy and examine the medical properties of nuclear radiation or document the breakdown of radioactive nuclear particles. The most commonly known application of nuclear physics is nuclear power generation and the other applications in many fields including nuclear medicine, magnetic resonance imaging, nuclear weapons, ion implantation, material engineering, military defense, astronomy, and radio-carbon dating in geology and archaeology [1].

The cross-section data of nuclear reactions is the quantitative characteristic of nuclear reactions which can provide information on the properties of excited nuclear states and different reaction mechanisms. Nuclear data are also necessary for the development of new concepts for medical isotope production and many other related applications. The nuclear reaction mechanism is the basic tool to study the properties of the nucleus as well as the behaviors of the nuclear force, and structure and understand

any other phenomena related to the nucleus [2].

For the production of positron-emitting radionuclides, such as $^{121}_{I}$, $^{64}_{Cu}$, $^{44}_{Sc}$ the structure of the target ($^{51}_{Sb}$, $^{64}_{Ni}$, $^{44}_{Ca}$) nucleus was modified after being bombarded by the charged particles (proton, deuteron, alpha particle). Characteristics of these charged particles, such as positron emission probability and required energy to initiate a bombardment, have an influenced on the possible outcomes of corresponding reactions.

In the present work the calculation of nuclear reaction cross-section by proton, deuteron, and alpha-particles induced reaction to produce the radioisotopes used in PET such as; I, Cu, and Sc at projectiles energy range 10Mev up to 70Mev. More experimental nuclear reaction data are needed to determine the optimum irradiation for the production yield of various radioisotopes. One of the important applicable aspects of nuclear technology is the production of artificial radioisotopes for diagnosis and therapeutic purposes in the medical field. The production of radioisotope artificially in a laboratory has a crucial role in medical diagnostic imaging or therapy. Stable targets can also be irradiated to form medically useful radioisotopes by

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using a charged particle accelerator, such as a cyclotron. In this case, the projectile (incident) particle is typically an alpha, deuteron, or proton, which gets accelerated radioisotopes by using a charged particle accelerator, such as a cyclotron. In this case, the projectile (incident) particle is typically an alpha, deuteron, or proton, which gets accelerated to high energies (10–100MeV) before bombarding a stable target. The radionuclides produced by cyclotrons are mainly neutron deficient and they decay by electron capture (EC) or positron (β^+) emission. Another mechanism is using a Nuclear reactor (n,γ), Nuclear reactors occur by exposing appropriate target material to the neutrons in the reactors, thereby causing a nuclear reaction to occur. Or it is possible to produce radioisotopes by bombarding the target nuclei with a neutron from a nuclear reactor. Radioisotopes produced by nuclear reactors are generally neutron-excess (rich) radionuclides. They mostly decay by β^- emission and are especially suitable for radiotherapy purposes. Nuclear reactors used to create medical radioisotopes operate on the concept of nuclear fission [4]. The last mechanism is by the help of a generator: A (radio-active) generator is along half-life radionuclides called 'Parent' which decay to a short half-life radionuclides called 'daughter' that can be used for imaging procedures. These long-lived radionuclides, when needed the 'daughter' radio nuclides combined with radiopharmaceuticals for design application [5]. A generator allows a short-lived daughter radionuclide to be separated from a longer-lived parent radionuclide as it decays for immediate use in the clinic.

The medical uses of radionuclides can be broken down into two general categories, imaging and radiotherapy. Imaging can be further divided into single photon emission computer tomography (SPECT) and positron emission tomography (PET). Positron emission tomography (PET) and single-photon emission computed tomography (SPECT) provide a means of examining regional cerebral blood flows, metabolism activity, and pharmacology *in vivo* under both resting and activating conditions.

In other words, PET is not only used in the diagnosis of cancer tissues but also plays a significant role in monitoring, staging, and following steps after the therapy. Epilepsy and Alzheimer's are other illnesses that can be diagnosed by PET.

The computer code EMPIRE is employed to produce theoretical cross-section data and to analyze it. Since the cross-section data is crucial to obtain an optimal specific activity by adjusting the irradiation parameters, such as target geometry and incident beam energy, we will compare the evaluated photonuclear data with the experimental measurements and then implement these data into the simulation as reliable as possible. The most important parameter in EMPIRE code is the exciton number and the level density parameter and the energy range 10MeV to 70MeV were taken by comparing the result with the experimental data from EXFOR data library sources IAEA [3].

Positron Emission Tomography (PET) Imaging Technique

Positron Emission Tomography (PET) is an imaging method in nuclear medicine based on the use of a weak radioactively marked pharmaceutical to image certain functions of a human or animal body organs. PET images display the spatial distribution of the radiopharmaceutical, also called tracer, thus allowing drawing conclusions about metabolic activities or blood flow. Therefore, PET is a functional imaging technique that has applications in oncology, cardiology, and neurology, for example to monitoring tumors or visualizing coronary artery disease. PET is a well well-known diagnostic method for investigating metabolic and physiological activity. Diagnosis of Alzheimer's disease has been investigated using an imaging agent that targets β amyloid plaque burden, but a new approach highlights altered copper homeostasis. ^{64}Cu Can be used as a new alternative method for the noninvasive diagnosis of Alzheimer's disease using PET.

2 Experimental Section

2.1 Materials

2.1. EMPIRE CODE

EMPIRE code is a highly reputable nuclear reaction model. It was developed by Herman. EMPIRE is a computer code used for the simulation of cross-section data for the light particle-induced reaction. The code provides complete and precise information on the reaction cross section based on different reaction mechanisms, i.e. direct reaction, pre-equilibrium reaction, and compound reaction. The EMPIRE code comprises various nuclear models and is designed for calculation over a broad range of energies, targets, and incident particles. The targets are calcium, nickel, and antimony. The projectile can be a neutron, proton, deuteron, alpha particle, or any heavy ion (IH) or photon. The energy range for this thesis is 10MeV to 70MeV but extends from certain Kev to several hundred MeV for heavy ions. EMPIRE CODE is a modular system of nuclear reaction codes for the comprehensive modeling of nuclear reactions using different theoretical models, so it is used to determine the covariance of the calculated data between the theoretical and experimental data [7]. In the present work, the spherical optical model through the code ECIS03 which calculates the elastic scattering and direct reactions induced by the light particles $A \leq 4$ in the frame of the generalized optical model was used. The optical model, discrete levels, and deformation parameters were retrieved from the RIPL-2 library. The direct channel calculations were performed by using the coupled channels model or the distorted wave-born approximation (DWBA) method. EMPIRE contains both the quantum mechanical (MSD/

MSC) and classical models (DEGAS, PCROSS, HMS) to describe the pre-equilibrium reaction. EMPIRE also contains the DEGAS module based on the exciton model for the description of pre-equilibrium reactions with angular momentum conservation [8]. EMPIRE CODE is used for theoretical investigation of nuclear reactions as well as for nuclear data evaluation work. EMPIRE is a nuclear reaction model code that accepts nucleons and several light ions including α -particles and deuterons in the incoming and outgoing channels [9].

EMPIRE is intended to be a general, flexible, and easy-to-use tool for basic research and evaluation of nuclear data. It also has a possibility of combining several theoretical approaches choosing among alternative input parameters and evaluating an extended set of observations. In a single run, nuclear data evaluation was facilitated by the ENDF and ENDF formatting file verification and graphical comparisons with experimental data [10]. The ENDF option in the input file of EMPIRE allows printing all necessary information to the output file.

2.1.1 Level density

Quantum mechanics says that the energy is quantized which generates the energy states. The energy states per unit energy interval are called nuclear level density. The nuclear level density influences the shape and the height of the calculation excitation function. This parameter plays a crucial role in the nuclear reaction mechanism. If the nuclear level density is higher, then the spacing between the energy states is small. Ideally, if it is infinity, then the state will be a continuum. This parameter has given great importance to the development of nuclear modular codes. The nuclear level density is given by

$$a = \frac{A}{K}$$

Where A is the mass of the radioisotope ^{124}I , ^{64}Cu , ^{44}Sc and **K** is the level density [11-15]. There are different level density models given in TYLYS and EMPIRE codes.

2.1.2 Mean Free Path

When a particle passes through a material, it undergoes collision that may change its direction of motion. The average distance between these collisions is called the mean free path (l). It describes the probability of the reaction cross-section for a particular reaction. The mean free path was the mean distance for particles to be removed from the beam. Intensity decreases due to the beam interactions with atoms or nuclei in the target. The mean free path has to be related to the number of atoms per unit volume, referred to as the number density N. The relation is the inverse proportionality. The proportionality constant multiplying N is called the cross-section σ . The relation between l, n, and σ is:

$$l = \frac{1}{N\sigma}$$

Where l is the mean free path, N is the density of the material and σ is the cross-section.

2.2 Methods

The theoretical nuclear reaction cross-section data were obtained with the help of EMPIRE Nuclear reaction cross-section calculation model by using appropriate adjustment of different parameters like exciton number, level density, and energy range, and the experimental cross-section data were gathered from nuclear reaction data using EXFOR. The comparative analysis was done. To show the correlation between the theoretical and experimental cross-section values. The numerical value of the measure of the probability of occurrence of nuclear reaction with the corresponding energies was tabulated and plotted as a nuclear reaction cross section in millibarn(mb) versus the corresponding projectile energy in Mega electron volt (MeV) for each selected reaction channel by using origin 9.1 software.

3 Results and Discussion

In the present study, the calculation of the reaction cross section for the production of important radioisotopes for PET imaging, and the excitation functions of three reactions were studied. These are $^{44}\text{Ca}(p,n)^{44}\text{Sc}$, $^{64}\text{Ni}(d,2n)^{64}\text{Cu}$, and $^{121}\text{Sb}(\alpha,n)^{124}\text{I}$ carried out in the energy range of 10MeV to 70MeV by proton, deuteron and alpha particles beam. The numerical cross-section data with the corresponding energies were presented in Tables 3.1, 3.2, and 3.3 below. The comparison of theoretical and experimental excitation functions is shown below by black and red solid lines respectively.

3.1 Production of ^{44}Sc

One of the very recent radionuclides of interest for PET imaging is ^{44}Sc (T1/2 = 3.97 hours, 100% $\text{EC}\beta^-$, 94% β^+ av 632 KeV, 100% γ 1157 KeV). ^{44}Sc can also be used independently in peptide-based imaging, as well as antibody labeling and small protein labeling.

^{44}Sc shows superiority over Gallium-68, given that the routine production method is well established. It does not require an expensive $^{68}\text{Ge}/^{68}\text{Ga}$ generator; it has a half-life suitable for logistics, a lower positron energy, and a slightly higher β^+ branching ratio. ^{44}Sc is produced when a projectile (proton beam) strikes the target calcium (^{44}Ca) and emits single neutron (n).

Table 3.1: Theoretical and experimental cross-section for the reaction channel $^{44}\text{Ca}(p,n)^{44}\text{Sc}$.

Energy(MeV)	$\sigma_{\text{Experimental}}(\text{mb})$	$\sigma_{\text{theoretical}}(\text{mb})$
10	606	562
10.8	662	552.2
11.6	672	542.23
12.4	689	535
13.2	600	509
13.9	428	458.09
15.3	355	322.37
16	274	258.4
16.5	239	219.07
17.2	195	178.24

Produced when the projectile deuterons strike a target element nickel (^{64}Ni) and emits two neutrons. However, the most abundant production method is by bombarding a solid target nickel with a proton due to the relatively high deuteron energy. The solid target nickel (Ni), its compound or alloys has large industrial applications which are perceived to be generally, due to its physical and chemical characteristics. This radioisotope is corrosion-resistant, silvery-white, and physically lustrous. The calculated excitation functions of $^{64}\text{Ni}(d,n)^{64}\text{Cu}$ reaction are compared with the experimental result and shown by the graph in Fig 3.2 below.

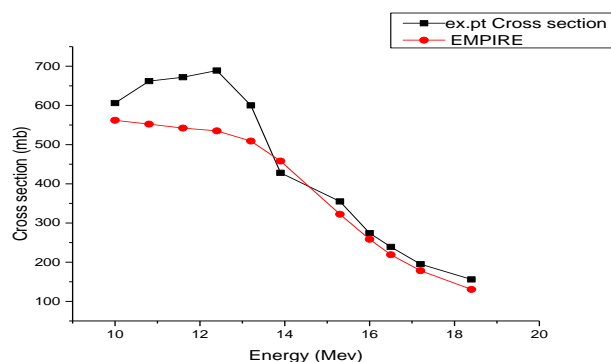


Fig. 3.1: Experimental and Theoretical excitation function for the reaction channel $^{44}\text{Ca}(p,n)^{44}\text{Sc}$.

From fig 3.1 above, the experimentally measured values, the reaction cross-section of pre-equilibrium nuclear reaction starts at minimum energy of the projectile about 10 MeV with 606mb and reaches its maximum peak point at energy about 12.5 MeV with 689mb and then falls for increasing value of projectile energy by making a long tail. But the theoretical reaction cross-section value starts at a maximum cross section 562mb at a projectile energy of 10MeV fall down by making along tail for the higher projectile energy. Comparatively, the experimental and theoretical nuclear reaction excitation functions give the best fit and have similar features at higher energies.

3.2 Production of ^{64}Cu

Copper-64 ($T_{1/2} = 12.8\text{h}$, 42.5%EC, 18% β^+ 270keV, 39% β^- 190keV, 0.5% γ 1345KeV). It is one of the most commonly used in the USA. Its low positron energy and little γ emissions make the image resolution high, and high-yield and high-purity production methods are developed. ^{64}Cu is one of the important biomedical radionuclides that is used for the labeling of radiopharmaceuticals for PET imaging. The radioisotopes of ^{64}Cu are due to the three modes of decay important for both PET and therapy. ^{64}Cu is

Table 3.2: Theoretical and Measured cross-section for the reaction $^{64}\text{Ni}(d, 2n)^{64}\text{Cu}$.

Energy(MeV)	$\sigma_{\text{Experimental}}(\text{mb})$	$\sigma_{\text{theoretical}}(\text{mb})$
10	644	883
10.8	762	980.65
11.8	883	1039.06
13.1	905	1093
14.02	955	1092
15.2	923	1066
16.02	908	1033.81
17.8	800	905.27
19.6	664	752.67

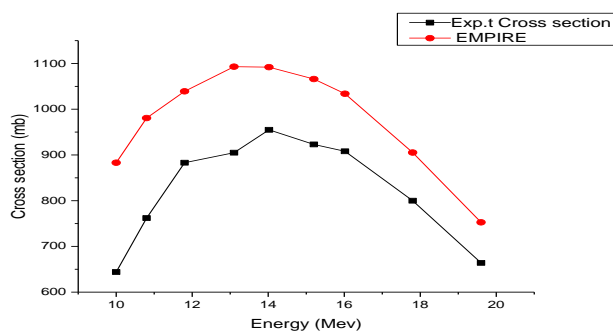


Fig. 3.2: Experimental and Theoretical excitation function for the reaction $^{64}\text{Ni}(d, 2n)^{64}\text{Cu}$.

As observed in Fig. 3.2 above the theoretically calculated values of pre-equilibrium nuclear reactions cross-section start with the energy of the projectile 10 MeV with 883mb and reaches its maximum peak point at 13 MeV with 1093mb and then fall for increasing the energy of the projectile. Similarly, the Experimentally measured value of the excitation function starts at the minimum energy of the projectile and reaches its maximum peak point at 14Mev with 955mb and falls with increasing energy of the projectile. At lower and higher energies the calculated

values of pre-equilibrium cross-section fits with the experimentally measured values. From the graph, the compound nucleus cross-section calculation gives better agreement with the experimentally measured values as compared to pre-equilibrium. As seen from the graph in the energy range between 12MeV to 14MeV the theoretical calculated value fairly fits with the experimental measured values because of the different parameters.

3.3 Production of ¹²⁴I

¹²⁴I is the only long half-life (4.18d) positron emitter radioisotope of iodine. ¹²⁴I is the most important radioisotope for thyroid image. Due to its long half-life, stability, and radiation emission ¹²⁴I is used in PET applications and oncological and non-oncological fields. ¹²⁴I is produced when a projectile (alpha particle) strikes a target element antimony (¹²¹Sb) and emits a single neutron (n). The theoretically calculated excitation functions of ¹²¹Sb(α, n) ¹²⁴I reaction are compared with the experimental result.

Table 3.3: Theoretical and measured cross section for the reaction ¹²¹Sb (α, n) ¹²⁴I.

Energy(MeV)	$\sigma_{\text{Experimental}}(\text{mb})$	$\sigma_{\text{theoretical}}(\text{mb})$
12.18	5.3	10.33
15.29	69	208.13
18.23	149	269
18	246	325
19.65	327	324.39

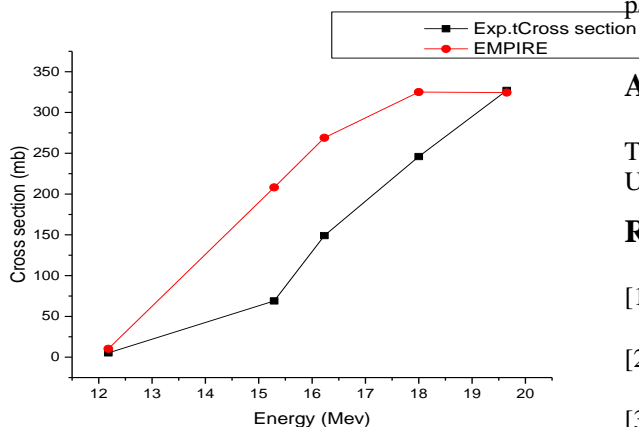


Fig. 3.3: Experimental and Theoretical excitation function for the reaction ¹²¹Sb (α, n) ¹²⁴I

From Figure 3.3 the theoretically calculated values start at a minimum energy 12MeV goes straight upward until the incident energy is 18MeV with 325mb cross section. Then

move nearly uniformly between in the energy range 18MeV to 19MeV. Similarly, the experimentally measured value starts with a minimum incident energy of 12MeV goes straight up then increases sharply until it reaches the maximum peak point with the incident energy of 19MeV with a cross-section of 327mb. It is found that the theoretically calculated value and the experimentally measured value of the pre-equilibrium in the energy range 15MeV to 19MeV are fit. The maximum cross-section value is about 327mb which is around 19.65MeV. The best production of ¹²⁴I cross-section at range between 18MeV to 19MeV. Around 19MeV and 65MeV both the experimental cross section and theoretical cross section are the best fit.

4 Conclusions

In the present work, theoretical reaction cross section of ⁴⁴Ca (p, n) ⁴⁴Sc, ⁶⁴Ni (d, 2n) ⁶⁴Cu, and ¹²¹Sb (α, n) ¹²⁴I have been carried out using nuclear reaction model EMPIRE in the incident energies ranges from 10MeV to 70MeV. The overall result shows good agreement in the selected energy range with the experimental results obtained from EXFOR library. The most appropriate production energy range for ⁴⁴Ca (p, n) ⁴⁴Sc is 11MeV to 13MeV. The maximum production of ⁴⁴Sc radioisotope occurs at the energy of 12.5MeV with 689mb nuclear cross section by a beam of proton. Similarly, the maximum production range of ⁶⁴Cu is 13MeV to 15MeV. The maximum probability of occurrence for the production of ⁶⁴Cu is at the energy of 14MeV with the corresponding cross-section value of 955mb by a beam of deuteron. The most appropriate production of ¹²⁴I is an energy range of 16MeV to 19MeV and maximum production occurs at the energy of 19MeV. The resulting radioisotopes ⁴⁴Sc, ⁶⁴Cu, and ¹²⁴I have important and wide applications in nuclear medicine particularly for PET imaging.

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