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Simulation of Cross Section for the Production of Copper-64

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Abstract
The radionuclide ^{64}Cu ($T_{1/2}$ = 12.7 h) is an important positron emitter, suitable for combining PET imaging and therapy. We evaluated four reactions, namely ^{64}Ni(p, n)^{64}Cu, ^{64}Ni(d, 2n)^{64}Cu, ^{68}Zn(p, αn)^{64}Cu and ^{64}Zn(d, 2 p)^{64}Cu. Data analysis was generally limited up to about 30MeV using the nuclear model code TALYS-1.6. The result compared favourably with experimental as well as other theoretical works in literature. The integral yields calculated from those data are also given. A critical comparison of the various production routes of ^{64}Cu is presented. The ^{64}Ni (p, n) ^{64}Cu reaction, utilizing a highly enriched target, is the method of choice.

Keywords: ^{6464}Cu Production, Excitation Functions, Calculate Thick Target Yield

1. Introduction

Copper-64 radioisotope ($T_{1/2}$ = 12.7h) has a large application in radiotherapy. As this isotope is a positron emission radioisotope, it is known as a particularly effective radioisotope in Positron Emission Tomography (PET) imaging study, the utility of copper-64 depends on the chemical stability in water with proper energy and half-life as gamma emitters. The radioisotopes of copper have great potential for preparing metal-chelates for medical use (Blower et al, 1996). In particular the radionuclide ^{64}Cu is very well suited: it has appropriate half-life, low β^+ end-point energy of 0.65MeV, comparable to that of ^{18}F ($T_{1/2}$ = 110 min), the most commonly used positron emitter, and practically no γ-ray. These decay properties are almost ideal for imaging. The only drawback is the relatively low β^+ abundance. On the other hand, its multiple decay modes (i.e., EC (43.8%), β^+ (17.8%) and β^- (38.4%)) renders it suitable for combining PET imaging and therapy (Sun, 1996 and Anderson et al, 1992). A pre-requisite of those studies, however, is the availability of ^{64}Cu of very high purity and high specific activity. Our goal was to investigate cross section measurements on enriched zinc and nickel for various production routes of ^{64}Cu and also to obtain the optimum production yield for the production of ^{64}Cu and ^{67}Cu on an enriched nickel and zinc target over a given range of proton-energy. In this study possible production routes of copper-64 are ^{64}Ni (p, n) ^{64}Cu, ^{64}Ni(d, 2n)^{64}Cu and ^{68}Zn(p, αn)^{64}Cu were considered. The excitation functions of these reactions reported in the literature (Tarkanyi et al 2011 and Aslam, 2010) are in good agreement with this theoretical excitation function calculation of TALY-1.6 code.

1.1 Method

1.1.0 Taly's Calculations

Taly's is a computer code system for analysis and prediction of nuclear reaction. The basic objective behind its construction is the simulation of nuclear reactions that involve neutrons, photons, deuterons, tritons, ^{3}He- and alpha-particles, in the 1KeV-1GeV. The most important parameters involving nuclear theory, which were directly used in theoretical calculations of the excitation functions of reactions under consideration over a wide range of energy extending up to 30MeV. The default optical model potential (OMPs) of TALYs for protons and neutrons are from the local and global parameterization by (Koning and Delaroche, 2003) whereas OMPs for deuterons, tritons, helion and alpha particles are based on the folding approach (Hauser and Feshbach, 1952). Depending on the structure of nuclei, calculation for direct reactions can be performed by coupled channel method, the distorted wave born approximation, weak- coupling model for giant resonance description. In all the calculation the default options for the direct reaction were used. The compound nucleus was treated within the frame–work of Hauser-Feshbach model along with the width fluctuation correction model of Moldauer, (1980) the pre- equilibrium reaction calculation were performed by exciton (Koning and Duijvestjin, 2004).

1.1.1 Generation of Nuclear Reaction Cross Section

In the estimation of optimum energy range for the production of copper-64, cross section of the ^{64}Ni (p, n) ^{64}Cu ^{64}Ni(d, 2n)^{64}Cu, and ^{68}Zn(p, αn)^{64}Cu which are function of their respective particle energies were generated using Taly's code. This was done by feeding the code with input parameter such as the incident particle, the particle energy, the appropriate target and the atomic mass of the target after which the data is submitted to code to run. After processing, the cross section of the various reaction channels are grouped and displayed into the output of the code. Using theoretical cross section values, the thick target yields of copper-64 can be calculated using SRIM-2013 for the stopping power. The thick target yield(y) is calculated by using Simpson's numerical integration.

$$y = \left(1 - e^{-\lambda t}\right)\frac{IN_a}{M} x \int_{E_1}^{E_2} \frac{\sigma(E)}{S(E)} dE \qquad (1)$$

Where N_a is the Avogadro number, M is the target atomic weight of the target element, $\sigma(E)$ is the reaction cross section as a function of energy, λ is the decay constant of the product, t is the time of irradiation, I is the projectile current and S(E) is the target stopping power (SRIM, 2013) expressed in unit $MeVcm^2g^{-1}$ [13].

1.1.2 Results and Discussion

1.1.2.1 Evaluation of Cross Sections of ^{64}Ni (p, n) ^{64}Cu Reaction

The database for this reaction is fairly strong and 13 data sets reporting experimental cross section were found in literature. But out of the 13, three were consider namely (Tarkanyi 2010), (Aslam, 2009) and (Szelecsenyi et al, 1993) for ^{64}Ni (p, n) ^{64}Cu reaction within the energy range of 5-20MeV. From figure 1 it can be seen that the cross section values obtained by Tarkanyi are slightly higher but they are in good agreement with this work. Despite the difference in cross section values, the trend of shape formation for all the excitation functions is almost the same. Therefore it can be deduced that the optimum energy range for this reaction falls within 7-12Me

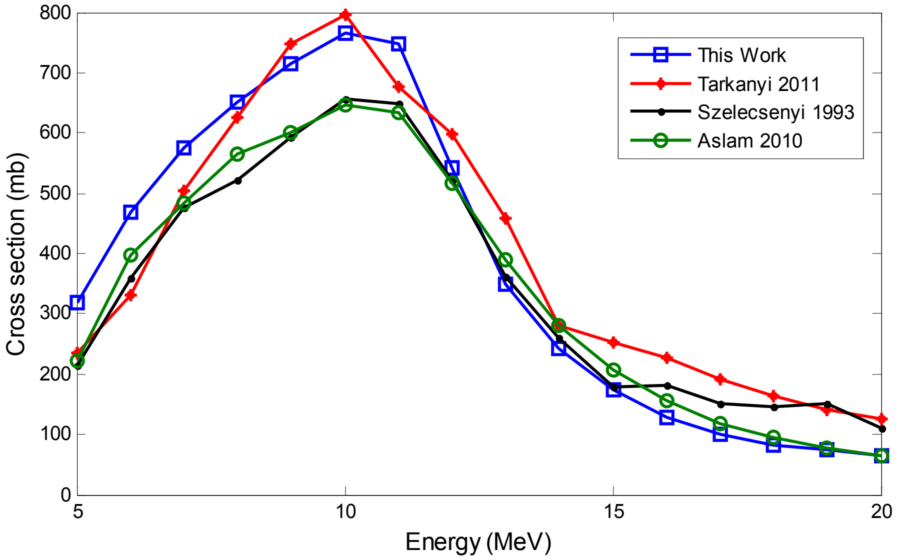

Figure 1 Comparison of excitation function for ^{64}Ni (p, n) ^{64}Cu reaction.

1.1.3 Evaluation of Cross Sections of ^{68}Zn (p, αn) ^{64}Cu Reaction

Tarkanyi performed works to experimentally measure the production cross section of ^{68}Zn (p, αn) ^{64}Cu reaction for the production of ^{64}Cu. The excitation function of his work and this current work are presented in figure 2. It can be observed that the maximum cross section is 51.4mb. Beside the trend of formation of the graph are almost the same. It can be deduced that the optimum energy range for the production of ^{64}Cu is within 20-29MeV

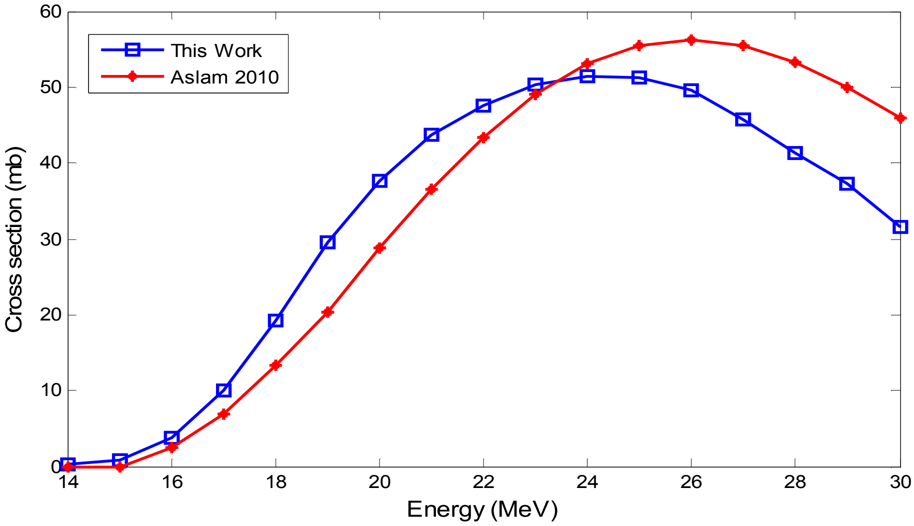

Figure 2: Comparison of excitation function for ^{68}Zn(p,αn)^{64}Cu reaction.

1.1.4 Evaluation of Cross Sections of ^{64}Ni (d, 2n) ^{64}Cu Reaction

The amounts of experimental cross section data available for ^{64}Ni (d, 2n) ^{64}Cu is not large. In a recent experiment, Tarkanyi measured the cross section using enriched ^{64}Ni as target and cross section values were also taken from Aslam. A comparison of excitation function of their work and this theoretical work are presented in figure 3. Analysis of the three graph show that there is variation of data point which may be due to unstable experimental conditions. It can also observed that result was fairly close to their work but deviation of peak were found in the energy range of 19-22MeV. The optimum energy range for the production is 10-18MeV.

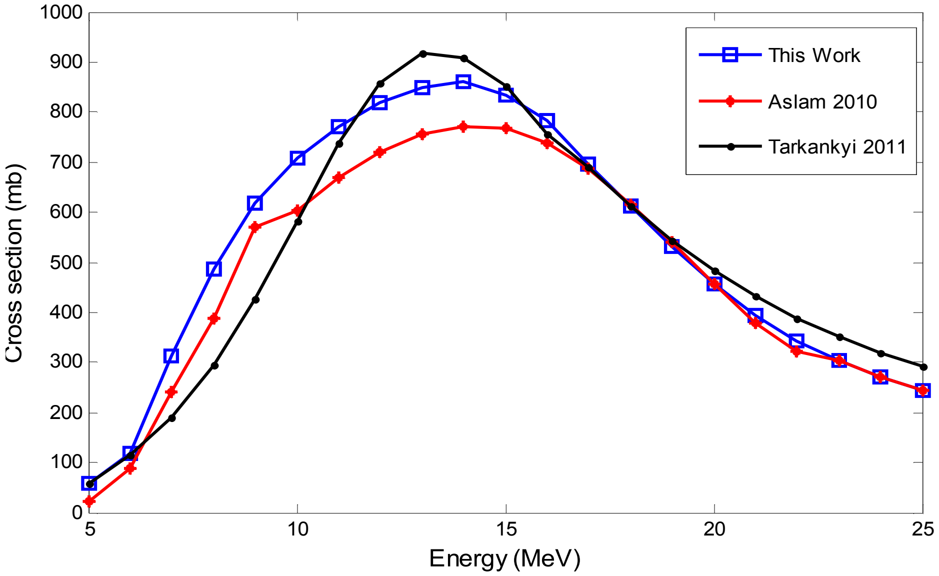

Figure 3: Comparison of excitation function for ^{64}Ni (d, 2n) ^{64}Cu reaction

1.1.5 Calculation of Thick Target Yields for Copper-64

Analyzing the available theoretical information on the excitation functions for the reactions ^{64}Ni (p, n) ^{64}Cu, ^{64}Ni (d, 2n) ^{64}Cu and ^{68}Zn(p, αn)^{64}Cu. These data could now be used for calculation of production yield using SRIM code for stopping power of ^{64}Cu via a given reaction over a certain energy range. Numerical valves of production yield in MBq/µA and particle energies in MeV were obtained from equation (1). We give the thick target yield from figure 4 to 6

From the figure below, it can be deduced that the production yield for ^{64}Ni (p, n) ^{64}Cu reaction increase with increase in proton energies. For these reaction four groups, namely (McCarthy et al, 1997), (Szelecsenyi et al, 1993), (Obata et al.2003) and Avila-Rodriguez et al, (2007) reported experimental ^{64}Cu yields. Those values are appreciably lower than the theoretical values.

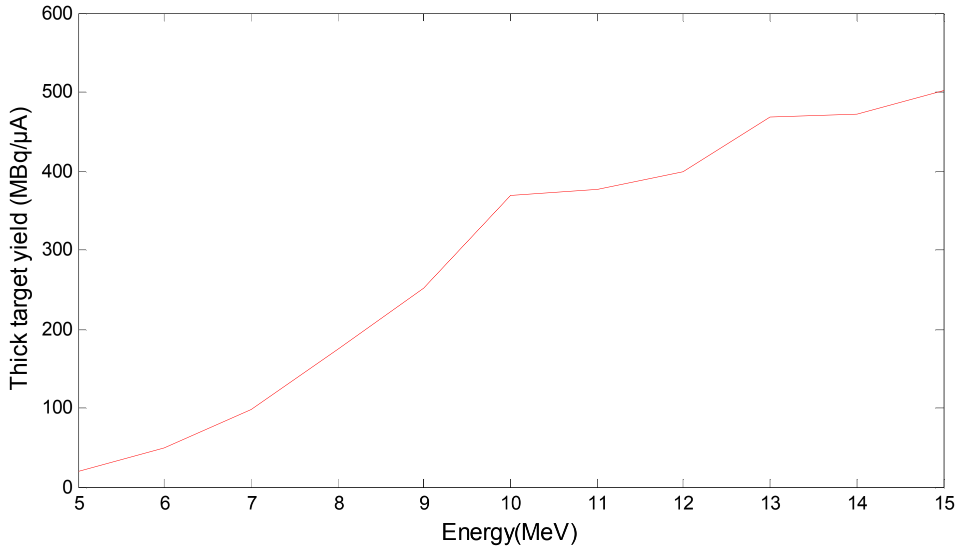

Figure 4: Calculated thick target yield for ^{64}Ni (p, n) ^{64}Cu, reaction

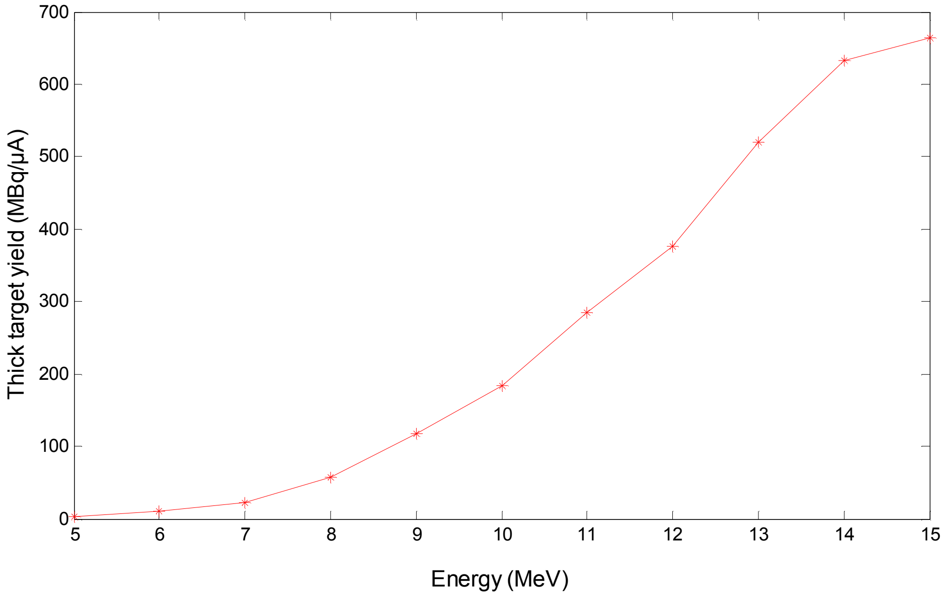

Figure 5: Calculated thick target yield for ^{64}Ni (d, 2n) ^{64}Cu reaction

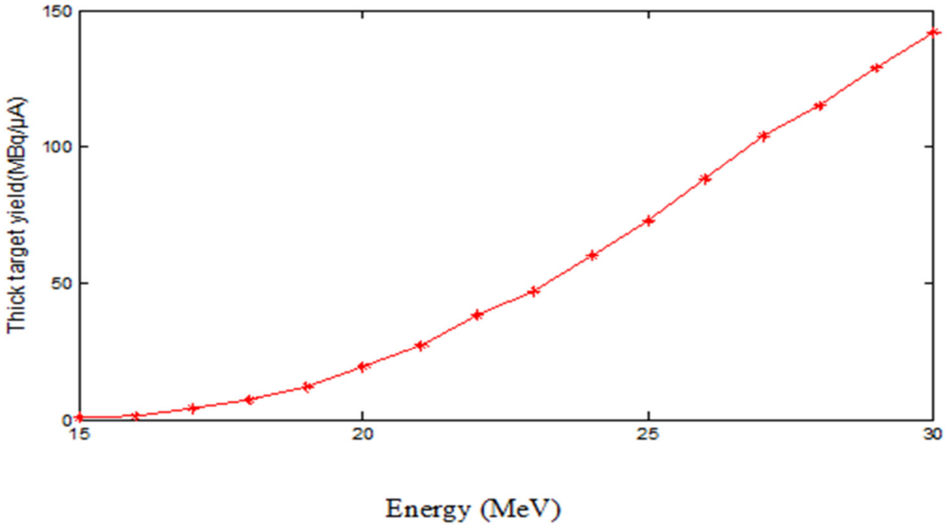

Figure 6: Calculated thick target yield for ^{68}Zn (p, αn) ^{64}Cu reaction

In the case of ^{64}Ni (d, 2n) Hermanne et al, 2007) and (Daraban et al, 2009) and also for ^{68}Zn(p, αn)^{64}Cu process (Boothe et al 1991) and (Smith et al 1996) reported experimental yields which are within the limits of the calculated yields and the values were considerably lower than the calculated yields. The reasons for low experimental yields are well known (loss of activity during irradiation and chemical processing, uncertainty in high beam current measurement, radiation damage, etc. The significance of the calculated yield is to define the ideal value which can be obtained via a given reaction.

1.1.6 Comparison of Production Yield of Copper-64

The nuclear reactions mention in this work are ^{64}Ni (p, n) ^{64}Cu, ^{64}Ni (d, 2n) ^{64}Cu and ^{68}Zn (p, αn) ^{64}Cu are the same as previously evaluated by previous researchers. For the ^{64}Ni(p, n)^{64}Cu, ^{64}Ni (d, 2n) ^{64}Cu and ^{68}Zn(p,αn) ^{64}Cu reactions, the cross sections and the thick target yield of this work are in good agreement with their work. But our values are higher than recommended values by the Tarkanyi and Aslam by 10–12%. Part of the difference arises possibly from the normalization of experimental cross section data to the new decay data of ^{64}Cu.

1.1.7 Comparison of Production Routes of Copper-64

A comparison of the various nuclear processes evaluated in this work with regard to the production of ^{64}Cu is given in Table 1. In each case the optimum energy range and the thick target yield are given below.

Table1. Comparison of production routes of Copper-64

Nuclear process	**Optimum energy range (MeV)**	**Thick Target yield (MBq/µA)**
^{64}Ni(p, n)^{64}Cu	12→ 7	99-369
^{64}Ni(d,2n)^{64}Cu	18→10	182-664
^{68}Zn(p, αn)^{64}Cu.	29→20	169-568

1.1.8 Conclusion

Copper-64 is a very important medical radioisotope. The study compared both experimental and theoretical data with the new version of Talys code 1.6 of which good agreement exit between them. Furthermore, the integral or thick target yields are estimated based on the measured excitation functions for all the investigated reactions of which the ^{64}Ni(p, n)^{64}Cu reaction is the method of choice for production of large quantities of high purity. The optimum energy range for this reaction is 12-7MeV and the thick target yield has been estimated with the same optimum energy of 99-369MBq/µAh. Finally, it is well known that for medical uses, enriched targets have to be used in the production to avoid the secondary produced unwanted impurities.

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