^{62}ZN RADIOISOTOPE PRODUCTION BY CYCLOTRON

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Abstract

Natural Cu target was irradiated with proton beam in the energy range of 15 to 30 MeV at a beam current of 100 μA for 15 min. In this irradiation radioisotope of 62Zn was produced as a generator and then decayed to ^{62}Cu radioisotope. The ^{62}Cu is emitting β+ and known as PET radioisotope. Excitation function of ^{62}Cu via ^{nat}Cu (p, 2n) ^{62}Zn, ^{62}Cu and ^{63}Cu (d, 3n) ^{62}Zn reactions were calculated using Alice and Talys codes and then were compared with the reported measurement by experimental data and ENDF-2013 data. Production yield versus target thickness were evaluated with attention to reaction cross section data obtained from Alice and Talys codes, and stopping power and range of protons in target materials using SRIM code. The production yield also examined experimentally and found that the optimum irradiation yield achieved to be 5.9 mci/μAh at protons of 100 μA current and 30MeV energy. A radiolabeling process also was performed using $^{62}Zncl_2$ and antitumor compound, Bleomycin (BLM) as a possible tumour imaging particle tracking.

INTRODUCTION

The short- lived generator produced β+ emitter ^{62}Cu (T= 9.7 min), has found application in blood flow studies in heart and brain using positron emission tomography (PET). This is of special interest for clinics having PET but no cyclotron. This radio isotope is obtained via the ^{62}Zn/^{62}Cu generator system, the parent ^{62}Zn (T=9.3h) being produced via the ^{63}Cu (p, 2n) process at medium-sized cyclotron [1]. Considering that cyclotron available for routine radioisotope productions accelerate particles of proton and very seldom deuteron that the maximum of proton's energy is 30 MeV and that of deuteron is 15 MeV and only the reaction resulting from proton and deuteron are accomplishable for generating zinc-62. Our irradiated target was natural copper and contains both ^{65}Cu and ^{63}Cu therefore, we'll analyse the reaction creating from ^{65}Cu and ^{63}Cu in this study. The aim of this work was the investigation of excitation function of ^{nat}Cu (p, 2n) ^{62}Zn→ ^{62}Cu and ^{63}Cu (d, 3n) ^{62}Zn reactions and their comparison with ENDF-2013 database.

The Alice/ Talys Code

The ALICE/ASH code is a modified and advanced version of the ALICE/91 code. This code has been written to study the interaction of intermediate energy nucleons and nuclei with target nuclei. The code calculates energy and angular distributions of particles emitted in nuclear reactions, residual nuclear yields, and total nonelastic cross-sections for nuclear reactions induced by particles and nuclei with energies up to 300 MeV. The parameters used in the ALICE/ASH code are as follow: (i) the Weisskopf–Ewing model for equilibrium calculations (ii) The hybrid model and geometry dependent hybrid model (GDH) for pre-equilibrium emissions [2]. TALYS aims to comprise important aspects such as physical quality, flexibility, robustness, completeness and efficiency into one software package [3].

Calculation of the Physical Yield and the Target Thickness

Theoretical physical yield can be calculated by the following equation:

$$Y = \frac{N_L \cdot H}{M} I(1-e^{-\lambda t}) \int_{E_1}^{E_2} \left(\frac{dE}{d(\rho x)}\right)^{-1} \sigma(E) dE \quad (1)$$

where Y is the product activity (in Bq) of the product, N_L is the Avogadro number, H is the isotope abundance of the target nuclide, M is the mass number of the target element, r (E) is the cross-section at energy E, I is the projectile current, dE/d (ρx) is the stopping power, λ is the decay constant of the product and t is the time of irradiation.

To achieve the optimum physical dimensions of the target such as the thickness, the SRIM codes [4] (the stopping and range of ions in matter); were accomplished. SRIM is a group of programs which calculate the stopping and range of ions (up to 2 GeV/amu) into matter using a quantum mechanical treatment of ion-atom collisions. This calculation is produced very efficient by the use of statistical algorithms which allow the ion to make jumps between calculated collisions and then averaging the collision results over the interfering gap.

The physical thickness of the target layer is chosen in such a way for a 90° geometry beam/target to ensure that the incident beam exits the target layer with a predicted energy; so the required thickness of the layer will be smaller with a coefficient 0.1.

ENDF-2013

The ENDF Evaluated Nuclear Data Formats are used all over the world to encode nuclear data evaluations for use in research and nuclear technology.

RESULT AND DISCUSSION

^{63}Cu (p,xn)^{63,62}Zn

Figure 1 show the excitation function obtained from the reaction of ^{63}Cu(p,xn)^{63,62}Zn .Considering this figure we'll find out that the cross section of ^{62}Zn generation, that is our desirable reaction is the best in the energy limit of 14 MeV to 30 MeV .This radioisotope isn't generated in the energy below than 14MeV and ^{63}Zn is one of our

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parasite reaction with a very high cross section and half-life of nearly 38.1 min .If we perform the chemistry steps after 4 or 5 half-life , we won't face the ^{63}Zn parasite reaction.

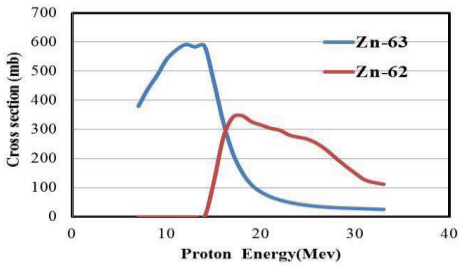

Figure 1: Excitation function of the ^{63}Cu (p, xn) ^{63,62}Zn process calculated by Talys code.

^{63}Cu (p,xnp)^{63,62,61}Cu

Figure 2 show the excitation function obtained from the reaction of ^{63}Cu (p,xnp)^{63,62,61}Cu and^{63}Cu is the natural copper. Having a successful chemistry we can also separate ^{62}Cu and ^{61}Cu during chemical processes.

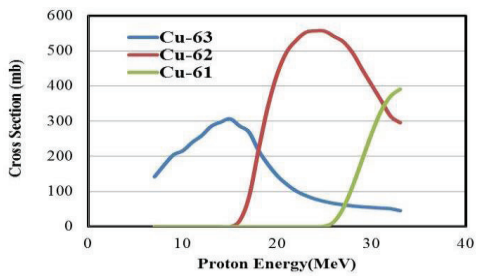

Figure 2: Excitation function the ^{63}Cu (p, xnp) $^{63, 62,61}$Cu process calculated by Talys code.

^{63}Cu (p, 2pxn) $^{59, 60, 61,62}$Ni

Figure 3 show the excitation function coming from the reaction of ^{63}Cu (p, 2pxn) $^{59, 60, 61,62}$Ni. As the cross section of Ni impurities is very low, in comparison of desirable reaction, practically, Ni impurities doesn't make any nuisance for us, and if dose, we can separate these parasite reaction during chemical processes.

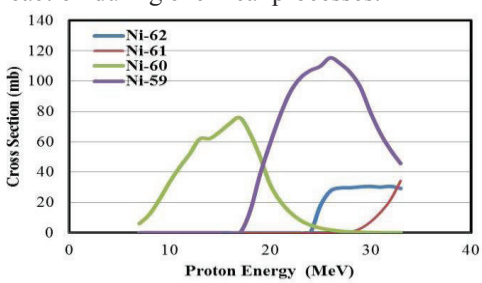

Figure 3: Excitation function of the ^{63}Cu (p, 2pxn)$^{59, 60, 61,62}$Ni process calculated by Talys code.

^{65}Cu (p, xn) $^{65, 64,63}$Zn

Figure 4 show the excitation function coming from the reaction of ^{65}Cu (p,xn)^{65,64,63}Zn. Considering Figure 4 we'll see that ^{64}Zn is one of the stable reactions and doesn't make any nuisance for us. ^{65}Zn is result of EC & β^+ decay changes into ^{65}Cu, that is the very natural copper and doesn't make any nuisance for us .^{63}Zn has already been discussed in Figure 1.

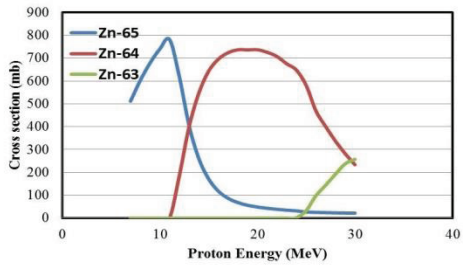

Figure 4: Excitation function the ^{65}Cu (p,xn)^{65,64,63}Zn process calculated by Talys code.

^{65}Cu (p,xnp)^{65,64,63}Cu

Figure 5 show the excitation function coming from the reaction of ^{65}Cu (p,xnp)^{65,64,63}Cu .^{65}Cu & ^{63}Cu are among stable isotopes and separate in chemical processes .

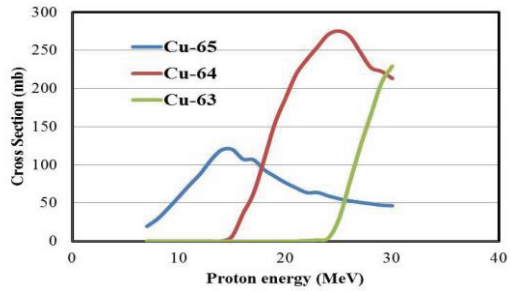

Figure 5: Excitation function of the ^{65}Cu (p,xnp)^{65,64,63}Cu process calculated by Talys code.

^{65}Cu (p, 2pxn) $^{59, 60, 61,62}$Ni

Figure 6 show the excitation function coming from the reaction of ^{65}Cu (p, 2pxn) $^{59, 60, 61,62}$Ni .As the cross section of Ni impurities is very low , then we'll see that detector can't detect the picks resulting from nickel at all , so we don't practically face the parasite reaction .

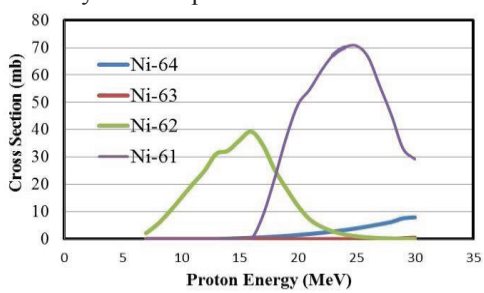

Figure 6: Excitation function of the ^{65}Cu (p, 2pxn) $^{59, 60, 61,62}$Ni process calculated by Talys code.

^{63}Cu (d, 3n) ^{62}Zn

Figure 7 shows the excitation function of ^{63} Cu (d, 3n) ^{62}Zn. According to this diagram we will see that the cross section of this reaction is zero to the energy 16 MeV. As

deuteron's energy is maximum 15 MeV in cyclotron, so we won't have any ^{62}Zn desirable reaction.

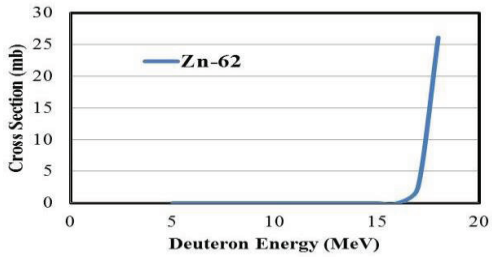

Figure 7: Excitation function of the ^{63}Cu (d, 3n) ^{62}Zn process calculated by Talys code.

Figure 8 shows the comparison between Alice code and evaluated data. The cross section resulting from Alice code is higher than evaluated data; this difference is because of existing error in low energy of used nuclear models.

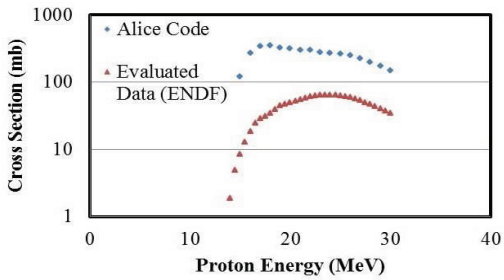

Figure 8: The comparison between Alice code and evaluated data.

After making cyclotron ready and regulating the current and energy targets are bombarded by the current of 100 μA and for 15 minutes .During bombardment the back of targets gets cool by the water current with a pressure of nearly 100 atmosphere [5].

Now a series of targets, already coated with different thicknesses, are bombarded to obtain both their amount of activity and yield. Considering Table 1 we'll find that only beneath the nickel layers contain ^{60}Cu and ^{61}Cu impurities result from nickel decay. Beneath the gold layers only ^{63}Zn impurities is found .We won't find any ^{60}Cu and ^{61}Cu is final product, this means ^{60}Cu and ^{61}Cu have disappeared in chemistry process. After bombarding the copper target by proton particle it is transferred to the related Hot Cell and 20-30cc 8 N HNO_3, depending on the thickness of the copper layer, is used for solving the target. After solving the target in HNO_3 solution , now it's necessary to substitute Cl^- ion for NO_3^- ion existing in solution resulting from washing , so that it's separable by (Dowex (1×8)-100-200 mesh-Cl^- form) resin. So the efficiency coming out from Cu is 98.08%. The physical yield of ^{62}Zn was obtained 5.9 mci/μAh and the required thickness of target is 118μm for 6° geometry beam toward the target.

Table 1: ^{62}Zn Physical Yield for Various Energies

Energy (MeV)	Substrate Materials	Available Radioisotope after Bombardment	Yield (mci/μAh)	%Yield /Yield (30)
18	Au	^{62}Zn, ^{63}Zn	0.25	4.24
20	Ni	^{62}Zn, ^{63}Zn, ^{60}Cu	0.87	14.75
22	Au	^{62}Zn, ^{63}Zn	1.25	21.12
25	Au	^{62}Zn, ^{63}Zn	2.67	45.3
27	Ni	^{62}Zn, ^{63}Zn, ^{60}Cu	4.28	72.5
30	Ni	^{62}Zn, ^{63}Zn, ^{60}Cu, ^{61}Cu	**5.9**	100

Imaging

Considering the high potential of the generator of ^{62}Zn radioisotope and clinical use of its diagnostic complexes, production of bleomycin zinc-62 was considered. Full experiment was done to determine the best temperature, acidity, time and reactor's concentration. ^{62}Zn BLM imaging performed in the tumour-bearing mice showed a distinct accumulation of the radiotracer in the tumour, while in the first and second hour a high background in liver and kidney was observed. Figure 9 shows absorption of radioactive material 2hours after injection.

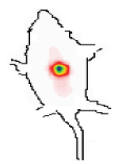

Figure. 9: Co-incidence images 2 h after injection of ^{62}Zn BLM.

CONCLUSION

In order to study practicality of the cyclotron production 62 Zn, excitation function of ^{nat}Cu (p, 2n) ^{62}Zn, ^{62}Cu and ^{63}Cu (d, 3n) ^{62}Zn reaction were calculated By Alice and Talys Codes, and compared with the date taken from the ENDF-2013 database. By virtue of excitation function and SRIM code, physical yield and target thickness were calculated for each reaction. Total labelling and formulation of [^{62}Zn] BLM took about 60 min, with a yield of 95-97%. HPLC and TLC showed that radiochemical purity of the 62Zn-labeled components was > 95%. Since the physical half-life of 62Zn is longer than most of other PET tracers, ^{62}Zn bleomycin, is a PET tracer with a suitable half-life, benefiting from PET advantage.

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