⁶⁷Cu Radioisotope Production with using ⁷⁰Zn(p,α)⁶⁷Cu Nuclear Reaction on Cyclone-30 High Current Cyclotron

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1. Introduction

Copper radionuclides, 62 Cu, 64 Cu, 67 Cu are considered to have potential for application in nuclear medicine[1]. Among them, 67 Cu is an important therapeutic isotope due to emitting strong beta particles and suitable particle range in tissue with a mean energy of 141keV for treatment and long half-life of 61.9hr for labeling with ligands and biomolecules, γ -rays of 91.3keV(7.0%), 93.3keV(16.1%), and 184.6keV (48.7%) for distribution measurement of 67 Cu in the tissue with SPECT scanner[2], [3].

Several nuclear reactions with charged particle irradiation in the accelerator, i.e., ${}^{68}Zn(p,2p){}^{67}Cu$, ${}^{67}Zn(d,2p){}^{67}Cu$, ${}^{64}Ni(\alpha,p){}^{67}Cu$ and ${}^{70}Zn(p,\alpha){}^{67}Cu$ have been investigated for ${}^{67}Cu$ production. Based on consideration of the production yield and radioisotopes impurities level in ${}^{67}Cu$ solution, the characteristics of cyclotron beam in our institute, the latter nuclear reaction is best for ${}^{67}Cu$ production even though the target material, the enriched ${}^{70}Zn$ is very expensive.

2. Methods and Results

2.1 Materials

All reagents used for production were of analytical grade. Enriched ⁷⁰Zn (isotopic purity 99.5%) was purchased from Isoflex, Russia. The reagents for Au plating for protect Cu plating from dissolving Zn target, KAu(CN)₂ was purchased from Merck and KH₂PO₄, EDTA was bought from Aldrich. Boric acid and NaCl for ⁷⁰Zn electrolyte were obtained from Merck. All ion exchange resin, anion (AG1-x8) and cation (AG50w-x8) were supplied from Bio-Rad. Dithizone and CCl₄ for solvent extraction were bought from Merck and Aldrich.

2.2 Preparation of Electrolyte for Au Plating and Enriched ⁶⁴Ni Material on Cu Cooling Plate

The electrolyte for Au plating on Cu cooling plate was made of 300mg of KAu(CN)₂, 2g of KH₂PO₄ and 3g of EDTA in 500ml water. The electrolyte for ⁷⁰Zn plating on the Au-coated Cu cooling plate was prepared with 1.2g of ⁷⁰Zn, 0.5g of boric acid and 1g of NaCl in 80ml water.

2.3 Electroplating of Au and enriched ⁷⁰Zn on Cu Cooling Plate

Cu cooling plates were cleaned with abrasive wool, rinsed with water, acetone and air-dried. Two Cu plates were mounted in the plating vessel home-made. Under vigorous stirring (900rpm / 10:10 seconds) with PE rod and applying the constant current (60mA) with dc power supply on Cu plates (cathode) in the plating vessel for 12hrs, Au was coated on the plates with 9.5mg/cm² of thickness. Careful observation of the Au surface has been done to confirm no crack on the surface. Au coated Cu cooling plate was mounted on the Zn electroplating device, the ⁷⁰Zn electrolyte was poured on it and the constant current (200mA) was applied on Cu cooling plate for 6hrs. The cathode current efficiency of the Ni plating was about 50%.

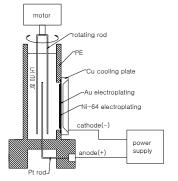


Fig.1. Schematic drawing of ⁷⁰Zn electroplating device.

2.4 Proton Irradiation on the ⁷⁰Zn Target

The proton beam irradiation on the Zn target was done with Cyclone-30 accelerator. The beam energy was controlled to 23MeV and the beam current was increased up to 200μ A step by step.

2.5 Irradiated ⁷⁰Zn target dissolving and Chemical Separation of ⁶⁷Cu from ⁷⁰Zn with solvent extraction and ion exchange resin

After the irradiation, Zn target was dissolved with 30ml of 5N HCl on the dissolving device (Fig.2.) without heating. 450ml of water was added to 64 Ni solution to dilute the normality of its hydrochloric acid to 0.5M.

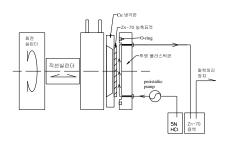


Fig. 2. Schematic drawing of the target dissolving device.

The chemical separation of ⁷⁰Cu from ⁷⁰Zn target was performed with 250ml of 0.01% dithizone in CCl₄[4]. In order to remove remained impurity RI completely in the organic phase, the organic phase was washed with 0.5N HCl and finally ⁷⁰Zn was recovered with the back extraction with 20ml of 7N HCl. For further purification of the final ⁶⁷Cu solution, the anion exchange resin (AG1- x8) was applied. The gamma-ray of RI in the dissolved ⁷⁰Zn solution and the final ⁶⁷Cu solution were measured with HPGe detector coupled with MCA. ⁶⁷Cu, ⁶⁷Ga, ⁶²Zn, ^{69m}Zn, ^{195m}Hg and ¹⁹⁵Hg were found on the dissolved solution (Fig. 3) and only ⁶⁷Cu was on the final ⁶⁷Cu solution (Fig. 4).

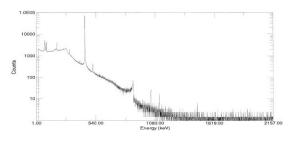


Fig. 3. Gamma-ray spectrum of the dissolved ⁷⁰Zn solution.

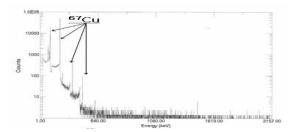


Fig. 4. The gamma-ray spectrum of the ⁶⁷Cu solution.

2.5 Chemical process of ⁷⁰Zn recovery for recycling.

⁷⁰Zn in the aqueous phase after solvent extraction and in the electrolyte after electroplating was recovered with anion (AG1-x8) and cation (AG50w-x8) resin quantitatively.

3. Conclusions

⁶⁷Cu was produced with ⁷⁰Zn(p,α)⁶⁷Cu nuclear reaction in high current cyclotron cyclone-30. For high beam current irradiation on ⁷⁰Zn target, the electroplating method was successful and physical status of the ⁷⁰Zn was good, but unfortunately melting on the surface was found during 200µA proton irradiation on the target. The efficient chemical separation method for ⁶⁷Cu separation from irradiated ⁷⁰Zn target using solvent extraction (0.01% dithizone in CCl₄-HCl) followed by ion exchange method and the recycling process for the enriched ⁷⁰Zn material were developed

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