Mass Production of ^{64}Cu with ^{64}Ni(p,n)^{64}Cu Nuclear Reaction and Target Material Recycling

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1. Introduction

^{64}Cu ($T_{1/2}$=12.7h, β^- decay: 40%, β^+ decay: 19%, E.C. decay: 41%) is one of the most useful radioisotope in the nuclear medicine due to its multiple decay mode and the intermediate half-life. Several nuclear reaction, i.e., ^{64}Ni(p,n)^{64}Cu, ^{68}Zn(p,αn)^{64}Cu and ^{64}Ni(d,2n)^{64}Cu have been investigated for production[1]. The highest could be obtained with proton irradiation on the enriched ^{64}Ni target. For mass and routine production, the ^{64}Ni target fabrication using electroplating[2], the reliable chemical separation of ^{64}Cu from the irradiated ^{64}Ni target and the effective recovery process for the recycling of very expensive enriched material ($20,000/g) and so forth are absolutely necessary to be established. In this work, we report our mass production method of ^{64}Cu with ^{64}Ni and Cyclone-30 accelerator.

2. Methods and Results

2.1 Materials

All reagents used for production were of analytical grade. Enriched ^{64}Ni (isotopic purity 96.1%) was purchased from Isoflex Russia. In order to dilute the enrichment of Ni to 25%, high purity $NiCl_2{\cdot}6H_2O$ (Puratronic^R) obtained from Alfa Aesar was added to enriched ^{64}Ni. The reagents for Au plating, $KAu(CN)_2$ was purchased form Merck and KH_2PO_4 and EDTA was bought from Aldrich. Boric acid and NaCl for Ni electrolyte were obtained from Merck. All ion exchange resin, anion (AG1-x8) and cation (AG50w-x8) were supplied from Bio-Rad. Dithizone and CCl_4 for solvent extraction were bought from Merck and Aldrich.

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

The electrolyte for Au plating on Cu cooling plate consisted of 300mg of $KAu(CN)_2$, 2g of KH_2PO_4 and 3g of EDTA in 500ml water. The electrolyte for Ni plating on the Au-coated Cu cooling plate was prepared with 1.5g of ^{64}Ni (enrichment: 25%), 1.0g of boric acid and 2g of NaCl in 90ml water.

2.3 Electroplating of Au and enriched ^{64}Ni 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/cm^2 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 Ni electroplating device, the ^{64}Ni electrolyte was poured on it and the constant current (150mA) was applied on Cu cooling plate for 6hrs. During electroplating, the thickness of ^{64}Ni deposited on the target was expected with counting ^{57}Ni activity remained in the Ni electrolyte. ^{57}Ni was produced with ^{58}Ni(p,2p)^{57}Ni reaction. The cathode current efficiency of the Ni plating was about 50%.

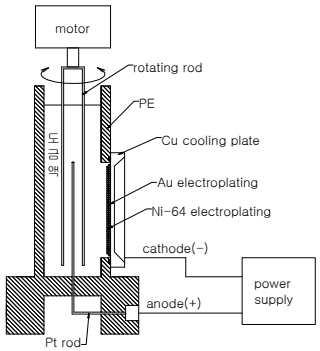

Fig.1. Schematic drawing of ^{64}Ni electroplating device.

2.4 Proton Irradiation on the ^{64}Ni Target

The proton beam irradiation on the Ni target was done with Cyclone-30 accelerator. The beam energy was controlled to 18MeV and the beam current was increased up to 150μA step by step.

2.5 Irradiated ^{64}Ni target dissolving and Chemical Separation of ^{64}Cu from ^{64}Ni with solvent extraction and ion exchange resin

After the irradiation, Ni target stayed in the hot-cell for 1day to decay out the short-lived radioactive impurities. Ni target was dissolved with circulation of 50ml of 5N HCl on the dissolving device (Fig.2.) equipped with the heater and the temperature controller. During Ni target dissolving, the temperature was maintained to 90°C to increase the dissolving speed. 450ml of water was added to ^{64}Ni solution to dilute the normality of its hydrochloric acid to 0.5M.

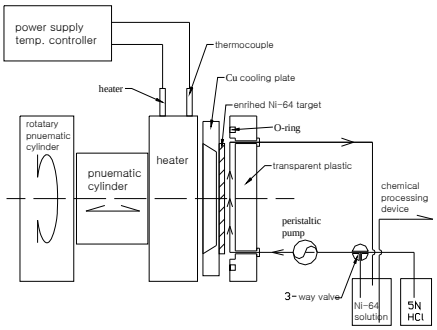

Fig. 2. Schematic drawing of the target dissolving device.

The chemical separation of ^{64}Cu from ^{64}Ni target was performed with 250ml of 0.01% dithizone in CCl_4[3]. In order to remove the impurity RI completely, the organic phase was washed with 0.5N HCl and finally ^{64}Cu was recovered with the back extraction with 20ml of 7N HCl. The gamma-ray of RI in the aqueous phase and organic phase were measured with HPGe detector coupled with MCA and ^{55}Co, ^{57}Co, ^{57}Ni and so on, were found and the separation yield in each separation process were calculated with counting ratios of them(Fig.3.).

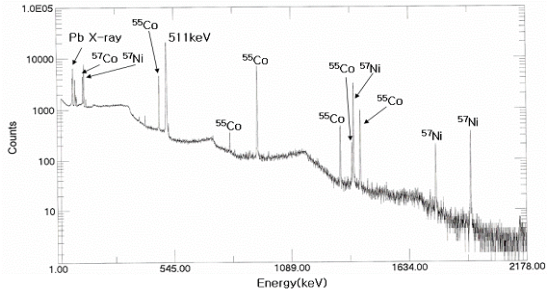

Fig. 3. Gamma-ray spectrum of the aqueous phase obtained with HPGe detector coupled with MCA

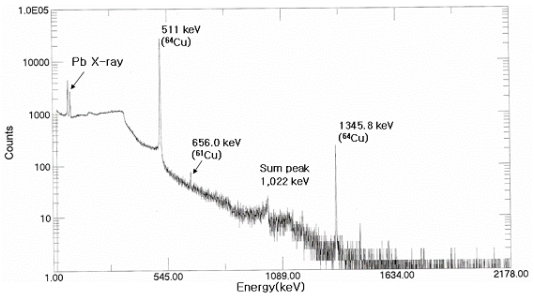

Fig. 5. Gamma-ray spectrum of the final ^{64}Cu solution.

2.5 Chemical process of ^{64}Ni recovery for recycling.

^{64}Ni remained in the aqueous phase after solvent extraction and in the electrolyte of ^{64}Ni electroplating was recovered with anion and cation exchange resin such as shown in the Fig. 4. Cation resin was used to concentrate ^{64}Ni and remove Fe ion and in order to remove ^{57}Co produced with ^{58}Ni(p,2p)^{57}Ni $\rightarrow$ ^{57}Co reaction anion resin was used[4].

4. Conclusions

The developed ^{64}Cu separation procedure from ^{64}Ni target and chemical processing for ^{64}Ni recycling is summarized in a flow chart (Fig. 4.). ^{64}Cu production yield was about 8.9mCi/μAh corrected on 96% enrichment of ^{64}Ni at EOB and are in good agreement with those predicted by Szelecscenyi[5]. The radionuclidic purity was higher than 99%(Fig. 5.). The ^{64}Ni recovery yield with anion/cation ion exchange resin was more than 98%.

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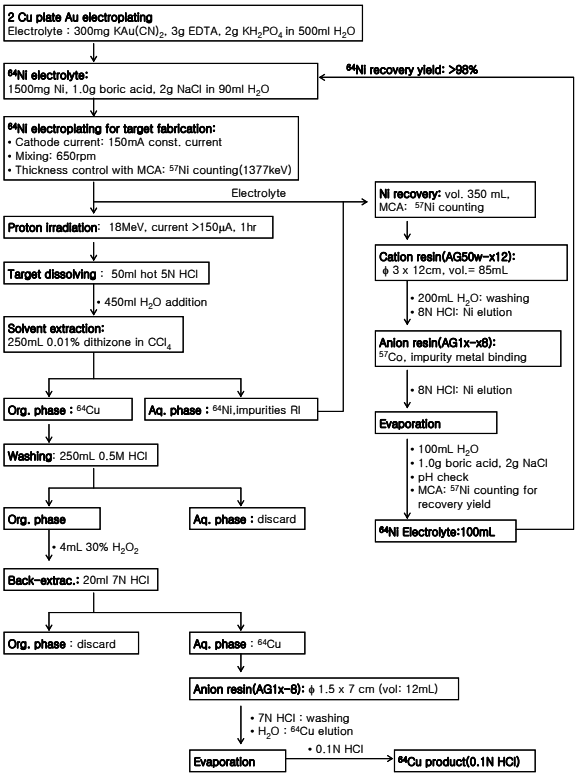

Fig. 4. Flow chart of ^{64}Cu mass production and ^{64}Ni enriched target recycling process.