Specification of $^{67}$Cu produced in the $^{70}$Zn($d,\alpha n$)$^{67}$Cu reaction

S. Yano,$^{*1}$ H. Haba,$^{*1}$ S. Shibata,$^{*1}$ Y. Komori,$^{*1}$ K. Takahashi,$^{*1}$ Y. Wakisaki,$^{*1}$ T. Yamada,$^{*2}$ and M. Matsumoto$^{*2}$

Copper-67 (half life $T_{1/2} = 61.83$ h and $\beta$-decay branch $I_\beta = 100\%$) is one of the promising radioisotopes for radiotherapy and radiodiagnosis.$^{1}$ In our preliminary study,$^{2}$ about 70 kBq of $^{67}$Cu was produced in the $^{70}$Zn($d,\alpha n$)$^{67}$Cu reaction at the AVF cyclotron. The production yield of $^{67}$Cu was 4.0 MBq/µA h at 24-MeV deuteron beam energy. We also investigated a chemical purification procedure for $^{67}$Cu. The chemical yield of $^{67}$Cu was 97\%, and the decontamination factors for Ga and Zn were evaluated to be $\sim 10^3$ and $>10^3$, respectively. In this work, we developed a new irradiation chamber to produce a larger amount of $^{67}$Cu ($> 100$ MBq) with a more intense deuteron beam. About 100 MBq of the purified $^{67}$Cu was obtained and its radionuclidic purity, specific radioactivity, and chemical purity were evaluated.

A schematic of the $^{67}$Cu production chamber is shown in Fig. 1. The 24-MeV deuteron beam with an intensity of 4 µA was extracted from the AVF cyclotron. The $^{70}$Zn-enriched oxide ($^{70}$ZnO) powder was prepared as a disk with 10-mm diameter and 340-mg cm$^{-2}$ thickness at a pressure of 2×10$^3$ kg cm$^{-2}$ for 3 min. The isotopic composition of the $^{70}$Zn target was 96.87\% $^{70}$Zn, 1.55\% $^{68}$Zn, 0.09\% $^{67}$Zn, 0.55\% $^{66}$Zn, and 0.94\% $^{64}$Zn. As shown in Fig. 1, the $^{70}$ZnO disk placed on a Ta beam stopper was covered by a high-purity Al foil 10 µm in thickness. During the irradiation, the $^{70}$ZnO target was cooled with circulating helium gas (30 L min$^{-1}$) and water (1.5 L min$^{-1}$). The beam axis was continuously rotated in 3-mm diameter at 2 Hz to avoid local heating of the target using electromagnets on the beam line of the AVF cyclotron. After the 10-h irradiation, $^{67}$Cu was separated from the target material and by-products such as $^{67}$Ga, $^{68}$Zn, and $^{71}$Zn through the chemical procedure reported in Ref.$^{2}$ The purified $^{67}$Cu was obtained as 300 µL of 0.1 M CH$_3$COOH for synthesis of the $^{67}$Cu-labeled antibody.$^{3}$ The radioactivity and radionuclidic purity was determined by $\gamma$-ray spectrometry using a Ge detector. The specific radioactivity and chemical purity were also evaluated by chemical analysis using an inductively coupled plasma mass spectrometer (Agilent Technologies 7700x).

A $\gamma$-ray spectrum of the purified $^{67}$Cu is shown in Fig. 2. 135 MBq of $^{67}$Cu was produced at the end of bombardment (EOB). The major radionuclidic impurity in the purified $^{67}$Cu was $^{64}$Cu ($T_{1/2} = 12.70$ h). The radioactivity ratio $A(^{64}$Cu)/$A(^{67}$Cu) was $1.2 \times 10^{-2}$ at EOB, which decreased to $8.9 \times 10^{-4}$ at 60 h after EOB (a typical time for its application studies). The present $A(^{64}$Cu)/$A(^{67}$Cu) ratio is smaller than the typical value of 6.7 in the $^{68}$Zn($p,2p$)$^{67}$Cu reaction.$^{4}$ The radionuclidic purity of the $^{67}$Cu solution was then evaluated to be $>99.9\%$ 60 h after EOB. In the ICP-MS analysis, only Cu (2.1 ppm) and Br (1.0 ppm) were detected with concentrations $>1$ ppm among the elements having atomic number $Z \geq 20$. The specific radioactivity of $^{67}$Cu was then determined to be 220 MBq µg$^{-1}$ at EOB. Hundreds of MBq of the purified $^{67}$Cu are ready for application studies. The results of synthesis of the $^{67}$Cu-labeled antibody will be reported elsewhere.$^{3}$

References
3) K. Fujiki et al., private communication.

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$^{*1}$ RIKEN Nishina Center
$^{*2}$ Japan Radioisotope Association