

Production and chemical separation of ^{195}Au

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Gold (Au) nanoparticles are considered useful for drug delivery systems.^{1,2)} Incorporating Au radioisotopes into Au nanoparticles enables visualizing the distribution of nanoparticles in the human body by γ -ray imaging.³⁾ Among Au radioisotopes, ^{198m}Au ($T_{1/2} = 2.272$ d) emits cascade γ rays suitable for imaging with the ability to probe local chemical environments based on perturbed angular correlation spectroscopy.⁴⁾ For preparing ^{198m}Au -incorporated nanoparticles in the future, we need to develop techniques for incorporating radioactive Au isotopes into Au nanoparticles. To this end, ^{195}Au ($T_{1/2} = 186.01$ d), which has a long half-life, is useful. Therefore, in this study, we performed the production and chemical separation of ^{195}Au .

We produced ^{195}Au in the $^{nat}\text{Pt}(d, xn)^{195}\text{Au}$ reaction at the RIKEN AVF cyclotron. A ^{nat}Pt foil (644 mg/cm², 99.95% purity) was irradiated with a deuteron beam of 24.2 MeV at an average current of 4.9 μA for 3 hours. The γ -ray spectroscopy of the ^{nat}Pt target with a Ge detector was performed 68 d after irradiation. We observed γ -ray peaks of ^{195}Au and ^{192}Ir ($T_{1/2} = 73.829$ d). The energy of the γ ray of ^{195}Au (98.9 keV, 11.2% branching ratio) is so low that self-absorption by the thick ^{nat}Pt foil was significant. Hence, we evaluated the radioactivity of ^{195}Au in the range of 8.3–11.7 MBq at the end of bombardment (EOB) based on the measurements of the γ ray from both sides of the target foil and tabulated photon absorption coefficient.⁵⁾

The ^{nat}Pt target was dissolved in aqua regia by heating. The solution was evaporated and redissolved in 2 mL of concentrated HCl. We repeated the evaporation and dissolution two more times. Then, the target dissolved in concentrated HCl was subjected to γ -ray spectroscopy. Here, the radioactivity of ^{195}Au was determined to be 9.5(7) MBq at EOB, which accounts for self-absorption. This value corresponds to a production yield of 0.65(7) MBq/($\mu\text{A}\cdot\text{h}$). It is smaller than 0.9 MBq/($\mu\text{A}\cdot\text{h}$) calculated from the recent cross-section measurements,⁶⁾ but it is consistent with 0.6 MBq/($\mu\text{A}\cdot\text{h}$) calculated from the previous measurements.^{7,8)}

We separated ^{195}Au from the ^{nat}Pt target by solvent extraction.⁹⁾ First, the target dissolved in concentrated HCl was evaporated and redissolved in 6 mL of 1 M HCl. The solution was divided into 3-mL aliquots and transferred into two centrifuge tubes. Then, we added 3 mL of ethyl acetate to each tube and shook the two tubes for 20 min. After centrifugation,

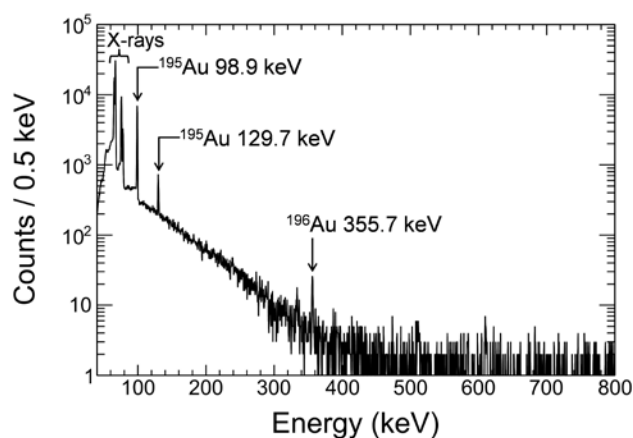


Fig. 1. γ -ray spectrum of the purified ^{195}Au sample.

the organic and aqueous phases were separated with pipettes, which were subjected to γ -ray spectroscopy. The yield of ^{195}Au extracted into the organic phase was 70(6)%. For further purification, we added

1 M HCl to the organic phase and performed further solvent extraction. The yield of ^{195}Au in the second extraction was 75(4)%. Further, we performed another extraction, and the yield of ^{195}Au was 71(4)%. Then, the organic phase was evaporated by flowing N_2 gas.

Figure 1 shows the γ -ray spectrum of the purified ^{195}Au sample. Here, the γ -ray peaks of ^{195}Au and ^{196}Au ($T_{1/2} = 6.2$ d) were observed. The radioactivity of ^{195}Au at EOB was determined to be 3.6(1) MBq, and the total chemical yield of ^{195}Au for the three-step solvent extractions was 38(3)%. The purified sample was subjected to inductively coupled plasma mass spectrometry (ICP-MS). The amount of Au was measured to be 23 ng, and thus the specific radioactivity of ^{195}Au at EOB was determined to be 157(11) MBq/ μg . The amount of Pt was 1.4 μg , indicating that the decontamination factor for ^{nat}Pt was 1.7×10^5 . The other chemical impurities (>1 μg) were Fe (1.5 μg), Cu (1.2 μg), Zn (1.4 μg), and Ba (7.0 μg). The purified ^{195}Au sample was supplied to the University of Tokyo to develop the method for incorporating it into Au nanoparticles. We are going to test the separation by extraction chromatography¹⁰⁾ to improve the chemical yield of ^{195}Au and decontamination factors for ^{nat}Pt and other impurities.

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