Chemical separation of theranostic radionuclide ^{111}Ag produced in ^{nat}Pd$(d,x)^{111}$Ag reactions

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Silver-111 (^{111}Ag, $T_{1/2}$ = 7.45 d) is a candidate radionuclide for theranostics (therapeutics + diagnosis). The nuclide emits β-particles (maximum β energy: 1036.8 keV), which can be applied to tumor therapy, and γ-rays with energies of 342 keV (branching ratio: 7%) and 245 keV (branching ratio: 1.33%), which can be used for imaging by single photon emission computed tomography (SPECT). However, ^{111}Ag has been rarely applied in the field of nuclear medicine. Toward the nuclear medical use of ^{111}Ag, we previously measured the production cross sections of ^{nat}Pd$(d,x)^{111}$Ag reactions.[1] The energy at the peak of the excitation function of the ^{nat}Pd$(d,x)^{111}$Ag reactions was approximately 9 MeV with a cross section of approximately 40 mb, which is consistent with the reported results.[2–4] In this study, ^{111}Ag was chemically separated from a metallic ^{nat}Pd target through anion-exchange chromatography for future nuclear medical application of ^{111}Ag.

^{111}Ag was produced by irradiating a stack of 6 metallic ^{nat}Pd foils (purity: 99.95%, thickness: 0.10 mm, Nilaco Corp., Japan) with a 24-MeV deuteron beam from the RIKEN K70 AVF Cyclotron. The average beam intensity was approximately 200 nA, and the irradiation time was 30 min. After irradiation, one of the irradiated ^{nat}Pd targets was dissolved in a mixed HNO_3 and HCl solution. After evaporation to dryness, the residue was dissolved in 1 M HNO_3 solution and then fed into an anion exchange column (Muromac 1 × 8, 200–400 mesh, NO_3^- form, Φ10 mm × 11 cm). Firstly, ^{111}Ag was eluted by 1 M HNO_3 solution. According to the literature,[5] Ag shows no adsorption in 1 M HNO_3 solution on an anion-exchange resin, while Pd adsorbs on the resin. After the elution of ^{111}Ag, ^{nat}Pd was stripped by concentrated HNO_3 solution. Each eluted fraction (approximately 1 mL) was subjected to γ-ray spectrometry with a Ge detector for determining ^{111}Ag radioactivity by using the 342-keV γ peak of ^{111}Ag. Because this γ peak was partly overlapped by the 345-keV γ peak of ^{105}Ag ($T_{1/2}$ = 41.29 d), which was simultaneously produced in the ^{nat}Pd(d,x) reactions, the area of the 342-keV γ peak of ^{111}Ag was calculated by Gaussian fitting. The elution behavior of ^{nat}Pd was checked using the 172-keV γ peak of ^{111m}Pd ($T_{1/2}$ = 5.5 h), which was also produced in the ^{nat}Pd(d,x) reactions.

Figure 1 shows the elution curves of ^{111}Ag and ^{111m}Pd obtained in this study. The elution of ^{111}Ag by 1 M HNO_3 solution started from fraction #11 and almost finished at fraction #16 before the start of elution of ^{111m}Pd, showing the clear separation of ^{111}Ag from the ^{nat}Pd target. By changing the eluent to concentrated HNO_3 after fraction #20, the elution of ^{111m}Pd started from fraction #28. Very little ^{111}Ag and ^{111m}Pd radioactivities remained on the anion-exchange column after completion of the separation. The recovery yields of ^{111}Ag and ^{111m}Pd were approximately 99% and 98%, respectively. This high yield for ^{111}Ag is quite suitable for nuclear medical use, which requires large radioactivity for tumor therapy. Because many radioactive isotopes of Ag are produced in the ^{nat}Pd(d,x) reactions, the use of an enriched ^{110}Pd target is essential for the selective production of ^{111}Ag in the ^{110}Pd$(d,2n)^{111}$Ag reactions. However, enriched ^{110}Pd targets are quite expensive, and it is important to recycle the ^{110}Pd target after the separation of ^{111}Ag. Therefore, the high recovery yield for ^{111m}Pd obtained in this study is also favorable for the separation of ^{111}Ag in medical use. The present results will lead to preclinical studies of therapeutic effects with ^{111}Ag.

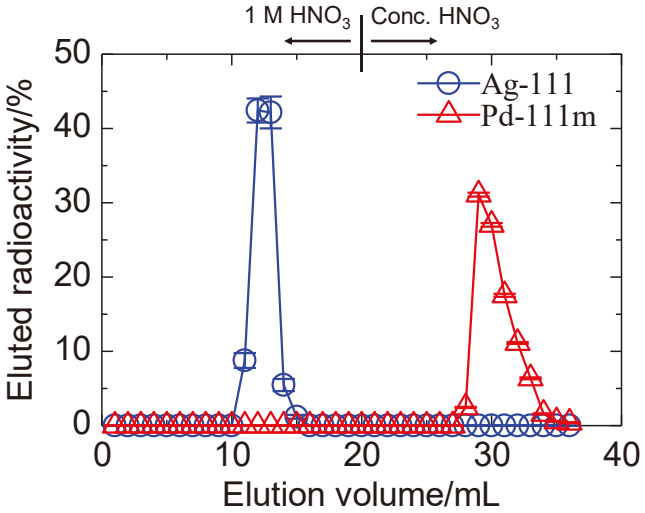

Fig. 1. Elution curves of ^{111}Ag and ^{111m}Pd in anion-exchange chromatography.

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