Improvement of chemical separation method for theranostic radionuclide ^{141}Ce

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Radionuclides are widely used in the treatment of tumors. In Japan, all therapeutic radionuclides used in clinical practice are imported from other countries. Domestic production using accelerators is desirable for a stable supply of therapeutic radionuclides.

One of the candidate radionuclides for theranostics (therapeutics + diagnosis) that can be produced by using accelerators is cerium-141 (^{141}Ce, $T_{1/2}$ = 32.5 d). This nuclide emits β-particles (maximum β energy: 580.7 keV), which can be used for tumor therapy. It also emits a γ-ray with an energy of 145.4 keV (branching ratio: 48.2%), which can be used for imaging by single photon emission computed tomography (SPECT). One of the production reactions for ^{141}Ce using accelerators is the ^{138}Ba$(\alpha, n)^{141}$Ce reaction. However, ^{141}Ce has rarely been used in the field of nuclear medicine, and investigation of accelerator production and chemical separation conditions will be necessary.

We have previously reported the accelerator production and chemical separation of ^{141}Ce.[1)] BaO was selected as a suitable target material for the production of ^{141}Ce by the α beam using an accelerator. Chemical separation of ^{141}Ce from the irradiated BaO target was also achieved through column chromatography using a Ln resin (extraction chromatographic resin with di (2-ethylhexyl) phosphoric acid). However, it took approximately 4 h for the chemical separation of ^{141}Ce because there was no eluent pumping.

In this study, the rapid chemical separation of ^{141}Ce from the Ba target using the Ln resin cartridge was investigated with increasing eluent flow rate.

^{141}Ce was produced in the ^{nat}Ba$(\alpha, xn)^{141}$Ce reaction with a 29-MeV alpha beam using the RIKEN K70 AVF cyclotron. ^{nat}BaO pellet was used as the target material. The irradiated ^{nat}Ba target (approximately 100 mg) was dissolved in 3 mL of 1 M HCl. After evaporation to dryness, the residue was dissolved in 10 mL of 0.03 M HCl solution The solution was filled into a 10 mL syringe and then injected into the Ln resin cartridge column (resin volume: 2 mL) at a flow rate of 1 drop per 1–2 seconds. Each 1 mL of the eluents was corrected with sample tubes. The ^{nat}Ba was washed out from the cartridge with 0.03 M HCl, and then ^{141}Ce was eluted with 1 M HCl solution. Each eluted sample was subjected to γ-ray spectrometry with a Ge detector for the determination of ^{141}Ce radioactivity. After measurement with the Ge detector, the concentration of ^{nat}Ba in each eluted sample was measured by ICP-MS.

The elution curves for ^{nat}Ba and ^{141}Ce, from the Ln resin cartridge, were shown in Fig. 1. The time required for the separation of ^{141}Ce with the Ln resin cartridge was less than 1 h, significantly reduced compared to the previous study (approximately 4 h) by increasing the eluent flow rate. The most of ^{nat}Ba was eluted at elution volume of around 15 mL with 0.03 M HCl. After elution of ^{nat}Ba, ^{141}Ce was recovered by elution with 1 M HCl. Although a small amount of ^{141}Ce leakage was observed around 10 mL of 0.03 M HCl, probably due to the increased eluent flow rate, the recovery yield for ^{141}Ce with 1 M HCl was as high as 96%. The contamination of ^{nat}Ba in the ^{141}Ce fractions was calculated to be approximately 0.6 μg in the ICP-MS measurement, which is also improved compared to the previous separation (contamination of ^{nat}Ba was approximately 3 μg).[1)] The separation factor of ^{141}Ce for Ba is estimated to be approximately 10^5.

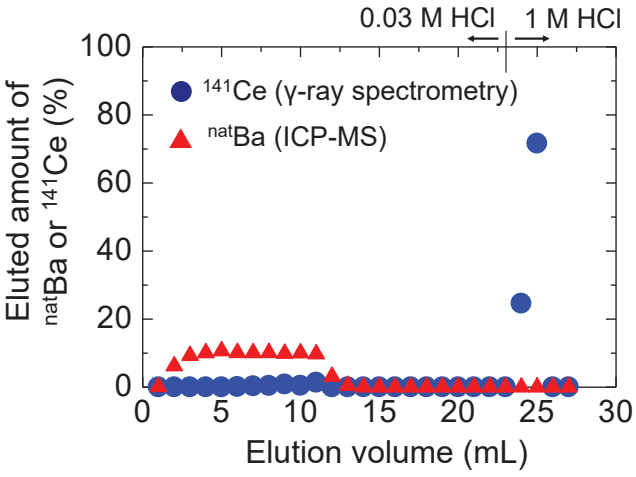

Fig. 1. Elution curves of ^{nat}Ba and ^{141}Ce in chromatographic separation with Ln resin cartridge.

In the next study, the radiopharmaceutical labeling of ^{141}Ce using DOTA (1, 4, 7, 10-Tetraazacyclododecane-1, 4, 7, 10-tetraacetic Acid), which is widely used for chelate labeling of therapeutic radionuclides, will be investigated.

Reference

1) K. Ooe *et al.*, RIKEN Accel. Prog. Rep. **55**, 130 (2022).

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