Accelerator production and chemical separation of theranostic radionuclide ^{141}Ce

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Because all of the therapeutic radionuclides used in clinical practice in Japan are imported from other countries, domestic production using accelerators is desirable for the stable supply of therapeutic nuclides.

Cerium-141 (^{141}Ce, $T_{1/2}$ = 32.5 d) is a candidate radionuclide for theranostics (therapeutics + diagnosis) and can be produced in the ^{138}Ba$(\alpha, n)^{141}$Ce reaction using accelerators. This nuclide emits β-particles (maximum β energy: 580.7 keV), which can be applied to the therapy of tumors, as well as 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). In particular, ^{141}Ce is expected to be useful for SPECT imaging because the energy of the γ-ray of ^{141}Ce is similar to that of ^{99m}Tc (140 keV), which is the most widely used SPECT nuclide in clinical application. However, ^{141}Ce has been rarely applied to nuclear medicine. In this study, a suitable target material of ^{nat}Ba for the accelerator production of ^{141}Ce was investigated. A chemical separation method for ^{141}Ce from the irradiated ^{nat}Ba target was also investigated through column chromatography with a Ln resin (extraction chromatographic resin with di(2-ethylhexyl) phosphoric acid).

For the determination of a suitable target material of ^{nat}Ba, $BaCl_2$ and BaO pellets (both approximately 100 mg) were irradiated with a 29-MeV alpha beam (beam intensity: 1.3 particle μA, irradiation time: 10 min) using the RIKEN K70 AVF cyclotron. The irradiated ^{nat}Ba targets were subjected to γ-ray spectrometry with a Ge semiconductor detector. In the chemical separation of ^{141}Ce from the ^{nat}Ba target, 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 3 mL of 0.01 M HCl solution and then fed into a Ln resin column (5-mm diameter × 50-mm height). ^{nat}Ba was eluted by 0.01 M HCl, following which ^{141}Ce was eluted by 1 M HCl solution by referring to the literature.[1] Each eluted sample (1 mL) was subjected to γ-ray spectrometry with the 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 inductively coupled plasma mass spectrometry (ICP-MS).

In the production of ^{141}Ce from the $BaCl_2$ target, high radioactivities of short-lived 38 K ($T_{1/2}$ = 7.636 min) and ^{34m}Cl ($T_{1/2}$ = 31.99 min) were observed. These by-products are considered to be produced from ^{nat}Cl in the $BaCl_2$ target and α beam. Because of these by-products, the radiation dose from the irradiated $BaCl_2$ target was quite high, and the γ-ray of ^{141}Ce was only observed after the decay of these by-products. On the other hand, in the γ-ray spectrum of ^{141}Ce produced with BaO, no such interference nuclides were observed, and the γ-ray of ^{141}Ce was observed soon after the end of irradiation. Therefore, BaO is considered to be a suitable target material for the production of ^{141}Ce by the α beam.

The elution curves for ^{nat}Ba and ^{141}Ce from the Ln resin are shown in Fig. 1 (BaO was used as the target material). ^{nat}Ba was eluted before ^{141}Ce with 0.01 M HCl. After the elution of ^{nat}Ba, ^{141}Ce was recovered by elution with 1 M HCl. The recovery yield for ^{141}Ce was greater than 99%. The contamination of ^{nat}Ba into ^{141}Ce fraction was calculated as less than 3 μg in the ICP-MS measurement: the separation factor of ^{141}Ce for ^{nat}Ba is greater than 10^4.

In the next study, the radiopharmaceutical labeling of ^{141}Ce will be investigated.

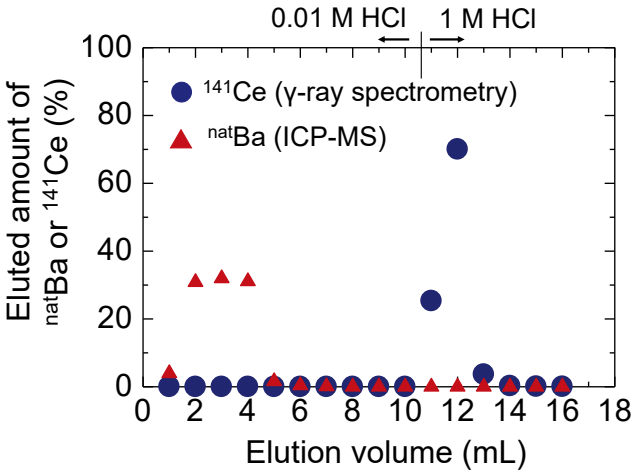

Fig. 1. Elution curves of ^{nat}Ba and ^{141}Ce in chromatographic separation with Ln resin.

Reference

1) E. P. Horwitz, C. A. A. Bloomquist, J. Inorg. Nucl. Chem. **37**, 425 (1975).

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