Production of no-carrier-added barium tracer of $^{135m}$Ba

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The long-lived $^{133}$Ba isotope (half-life $T_{1/2} = 10.51$ y) is the only Ba isotope commercially available from Japan Radioisotope Association. Since $^{133}$Ba is produced at a nuclear reactor, its specific radioactivity is low with a typical value of approximately 0.5 MBq µg$^{-1}$. Barium-$^{135m}$Ba with $T_{1/2} = 28.7$ h can be produced from the $^{133}$Cs(α, $x$)$^{135m}$Ba reaction by using a cyclotron. Barium-$^{135m}$Ba emits a single 268.2-keV γ-ray, which would be useful for radiotracer studies of Ba, especially for single-photon-emission computed tomography (SPECT). In this work, we investigated a procedure to produce $^{135m}$Ba of high specific radioactivity by using the $^{133}$Cs(α, $x$)$^{135m}$Ba reaction and no-carrier-added chemical separation.

CsCl powder (Sigma-Aldrich; chemical purity > 99.999%) was pressed into a disk of 10-mm diameter and 240-µg cm$^{-2}$ thickness at a pressure of $2 \times 10^4$ kg cm$^{-2}$ for 3 min. The CsCl pellet covered with a 10-µm Al foil (chemical purity > 99.99%) was placed on a target holder. The target was irradiated for 30 min with a 50-MeV alpha beam having an intensity of 3.0 µA at the RIKEN AVF cyclotron. During the beam irradiation, the 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 a circle of diameter approximately equal to 2 mm at 2 Hz to avoid local heating of the target by using electromagnets on the beam line of the RIKEN AVF cyclotron. After the irradiation, $^{135m}$Ba was chemically separated from the target material and by-products such as $^{135}$La and $^{132}$Cs by using a chromatography column filled with the Eichrom Sr resin$^2$ (Fig. 1). The radioactivity and radionuclidic purity of the purified $^{135m}$Ba were determined by γ-ray spectrometry using a Ge detector. The chemical purity and specific radioactivity were evaluated by chemical analysis using an inductively coupled plasma mass spectrometer (ICP-MS). The γ-ray spectrum of the purified $^{135m}$Ba is shown in Fig. 2. Only Ba isotopes of $^{131}$Ba, $^{133}$Ba, $^{135}$mBa, and $^{135m}$Ba were identified. The radioactivity of $^{135m}$Ba was determined to be 2.25 MBq at the end of bombardment (EOB). The chemical yield of $^{135m}$Ba was greater than 96%. Decontamination factors of $^{135}$La and $^{132}$Cs from $^{135m}$Ba were evaluated to be 103 and 105, respectively. The radionuclidic purity of $^{135m}$Ba was approximately 68% at the EOB. The major radionuclidic impurity was $^{133}$mBa ($T_{1/2} = 38.9$ h) which was produced in the $^{133}$Cs(α, $x$)$^{135m}$Ba reaction. Referring to the excitation functions for the $^{134}$Cs(α, $x$)$^{135}$Ba and $^{133}$Cs(α, $x$)$^{135m}$Ba reactions in the TENDL-2015 library,$^3$ it is expected that the radionuclidic purity of $^{135m}$Ba can be increased at lower beam energies. In the ICP-MS analysis, only Cu (1280 ng), U (160 ng), Zn (140 ng), and Ba (100 ng) were detected among the elements having atomic number $Z \geq 20$ in the purified $^{135m}$Ba with a contamination level > 100 ng. The specific radioactivity of $^{135m}$Ba was then 23 MBq µg$^{-1}$ at the EOB. This specific radioactivity is two orders of magnitude larger than that of the commercial $^{133}$Ba.

Based on the present results, we estimate that approximately 80 MBq of the no-carrier-added $^{135m}$Ba could be produced after 24-h irradiation of the 240-µg cm$^{-2}$ CsCl target with the 50-MeV and 3-µA alpha beam. The expected specific radioactivity is approximately 830 MBq µg$^{-1}$.

References