

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

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The long-lived  $^{133}\text{Ba}$  isotope (half-life  $T_{1/2} = 10.51$  y) is the only Ba isotope commercially available from Japan Radioisotope Association. Since  $^{133}\text{Ba}$  is produced at a nuclear reactor, its specific radioactivity is low with a typical value of approximately  $0.5 \text{ MBq}\mu\text{g}^{-1}$ . Barium-135m with  $T_{1/2} = 28.7$  h can be produced from the  $^{133}\text{Cs}(\alpha, x)^{135\text{m}}\text{Ba}$  reaction by using a cyclotron. Barium-135m emits a single 268.2-keV  $\gamma$ -ray, which would be useful for radiotracer studies of Ba, especially for single-photon-emission computed tomography (SPECT).<sup>1)</sup> In this work, we investigated a procedure to produce  $^{135\text{m}}\text{Ba}$  of high specific radioactivity by using the  $^{133}\text{Cs}(\alpha, x)^{135\text{m}}\text{Ba}$  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  $240\text{-mg cm}^{-2}$  thickness at a pressure of  $2 \times 10^3 \text{ kg cm}^{-2}$  for 3 min. The CsCl pellet covered with a  $10\text{-}\mu\text{m}$  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  $3.0 \mu\text{A}$  at the RIKEN AVF cyclotron. During the beam irradiation, the target was cooled with circulating helium gas ( $30 \text{ L min}^{-1}$ ) and water ( $1.5 \text{ L min}^{-1}$ ). 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,  $^{135\text{m}}\text{Ba}$  was chemically separated from the target material and by-products such as  $^{135}\text{La}$  and  $^{132}\text{Cs}$  by using a chromatography column filled with the Eichrom Sr resin<sup>2)</sup> (Fig. 1). The radioactivity and radionuclidic purity of the purified  $^{135\text{m}}\text{Ba}$  were determined by  $\gamma$ -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  $\gamma$ -ray spectrum of the purified  $^{135\text{m}}\text{Ba}$  is shown in Fig. 2. Only Ba isotopes of  $^{131}\text{Ba}$ ,  $^{133}\text{Ba}$ ,  $^{133\text{m}}\text{Ba}$ , and  $^{135\text{m}}\text{Ba}$  were identified. The radioactivity of  $^{135\text{m}}\text{Ba}$  was determined to be  $2.25 \text{ MBq}$  at the end of bombardment (EOB). The chemical yield of  $^{135\text{m}}\text{Ba}$  was greater than 96%. Decontamination factors of  $^{135}\text{La}$  and  $^{132}\text{Cs}$  from  $^{135\text{m}}\text{Ba}$  were evaluated to be 103 and 105, respectively. The radionuclidic purity of  $^{135\text{m}}\text{Ba}$  was approximately 68% at the EOB. The major radionuclidic impurity was  $^{133\text{m}}\text{Ba}$  ( $T_{1/2} = 38.9$  h) which was produced in the  $^{133}\text{Cs}(\alpha, x)^{133\text{m}}\text{Ba}$  reaction. Referring to the excitation functions for the  $^{133}\text{Cs}(\alpha, x)^{135\text{m}}\text{Ba}$  and  $^{133}\text{Cs}(\alpha, x)^{133\text{m}}\text{Ba}$  reactions in the TENDL-2015 library,<sup>3)</sup> it is expected that the radionuclidic purity of  $^{135\text{m}}\text{Ba}$  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)

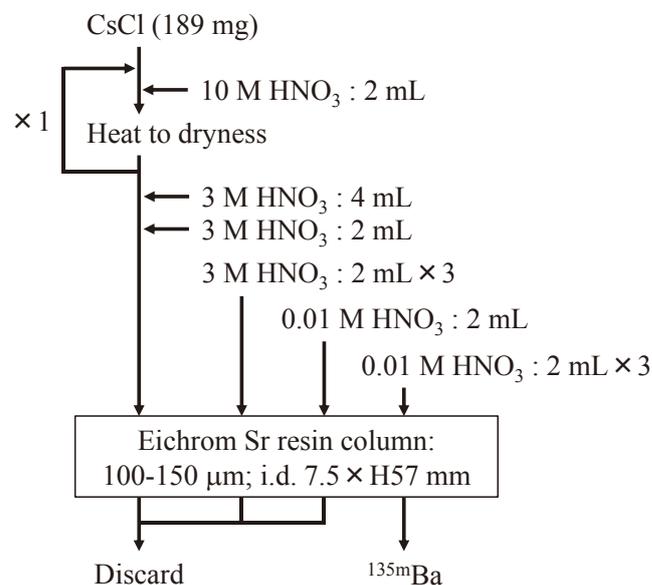


Fig. 1. Chemical separation procedure of  $^{135\text{m}}\text{Ba}$  from the CsCl target.

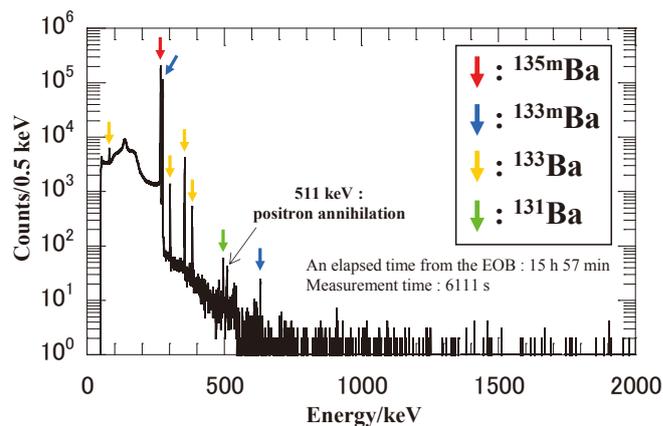


Fig. 2.  $\gamma$ -ray spectrum of purified  $^{135\text{m}}\text{Ba}$ .

were detected among the elements having atomic number  $Z \geq 20$  in the purified  $^{135\text{m}}\text{Ba}$  with a contamination level > 100 ng. The specific radioactivity of  $^{135\text{m}}\text{Ba}$  was then  $23 \text{ MBq}\mu\text{g}^{-1}$  at the EOB. This specific radioactivity is two orders of magnitude larger than that of the commercial  $^{133}\text{Ba}$ .

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

### References

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