Modification of ²¹¹At production apparatus to increase its production efficiency

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²¹¹At is one of the most promising radionuclides for targeted α -particle therapy. We have been developing the production technology of ²¹¹At based on the $^{209}\text{Bi}(\alpha, 2n)^{211}\text{At}$ reaction at the RIKEN azimuthally varying field (AVF) cyclotron¹⁻⁴) and distributing ²¹¹At to approximate 20 research groups in Japan. The poor thermal conductivity (7.97 W/(m \cdot K) at 300 K) and low melting point (272°C) of metallic Bi necessitate an effective target cooling system for the production of 211 At. In the proposed method,⁴⁾ an elliptical 209 Bi metal target with an area of 4.3 cm² and typical thickness of 20 mg/cm^2 on a 1-mm Al backing is irradiated with a 28.0-MeV α (⁴He²⁺) beam at an angle of 15° to the beam axis. A beam wobbler system is used to rotate the beam spot position on the target to prevent heat concentration. During the irradiation, the target is cooled with circulating water (4 L/min; 5° C) and He gas (30 L/min). Using this method, ²¹¹At can be quantitatively produced with α beam intensities up to 10 particle $\mu A (p\mu A)$.⁴⁾ To meet the growing demand for ²¹¹At, in this study, we modified the production apparatus to realize its production with a larger intensity of 25 particle μA .

Figure 1 illustrates the schematic of the modified apparatus. The incident angle of the beam to the 209 Bi target is reduced from 15° to 10° to accept beams with a larger target area (6.5 cm²) and thinner target thickness (13 mg/cm²), whereas the effective thickness is unchanged. During the irradiation, the Bi surface is cooled with He gas (30 L/min), and the Al backing is cooled with water (4.0 L/min; 5°C) as same as the previous setup.⁴)



Fig. 1. Schematic of the modified chamber.

With the modified apparatus, two metallic ²⁰⁹Bi targets with thicknesses of 12.94 and 12.88 mg/cm² were irradiated with a 27.9-MeV α beam for 0.5 hours. The average beam intensities were 2.1 and 25.0 particle μ A, respectively. Following the irradiation, the targets were subjected to γ -ray spectrometry with a Ge detec-

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tor to determine the production yields of 211 At. The produced ²¹¹At was separated from the ²⁰⁹Bi target using a dry distillation method.^{2,3)} The ²⁰⁹Bi target irradiated with the 25.0-particle $\mu A \alpha$ beam was transferred to a quartz tube and heated up to 850°C under O_2 gas flowing at 15 mL/min. ²¹¹At sublimated from the target was transported to a cold perfluoroalkoxy (PFA) tube $(-96^{\circ}C)$ by the O₂ flow. The PFA tube was washed with 300 μ L CHCl₃ to collect ²¹¹At in a glass v-vial. Finally, the CHCl₃ solution was dried with N₂ flow (3 L/min) at $\sim 25^{\circ}$ C and 100 kPa. The chemical yield of the dry 211 At was determined through γ -ray spectrometry. The chemical purity of the dry $^{211}\mathrm{At}$ and decontamination factor of $^{209}\mathrm{Bi}$ for $^{211}\mathrm{At}$ were evaluated based on a chemical analysis using the Agilent 7900 ICP-MS.

No evidence of melting was found for both the targets (Fig. 2). The experimental thick target yields (TTYs) of ²¹¹At were 23.2 ± 0.9 and 23.1 ± 0.9 MBq/µAh with beam intensities of 2.1 and 25.0 particle µA, respectively, suggesting that ²¹¹At can be produced quantitatively with an intensity of 25 particle µA. Figure 3 depicts the experimental and theoretical TTYs of ²¹¹At in the ²⁰⁹Bi($\alpha, 2n$)²¹¹At reaction. The experimental TTY at 27.9 MeV is 94 ± 4% of the theoretical value,⁵⁾ which agrees with that obtained in our previous work (91 ± 6% at 28.4 MeV).¹⁾



Fig. 2. Photos of the irradiated ²⁰⁹Bi targets with (a) 2.1and (b) 25.0-particle $\mu A \alpha$ beams at 27.9 MeV.



Fig. 3. Experimental and theoretical thick target yields of $^{211}{\rm At}$ in the $^{209}{\rm Bi}(\alpha,2n)^{211}{\rm At}$ reaction.

The chemical yield of ²¹¹At was 77 ± 4%. The chemical impurities over 1 ng in dry ²¹¹At were Cu (13.4 ng), Al (59.0 ng), Zn (21.8 ng), Pb (8.9 ng), Fe (12.0 ng), Mg (13.1), and Bi (1.5 ng). The decontamination factor of ²⁰⁹Bi for the purified ²¹¹At was > 10⁷.

References

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