Progress of ²¹¹At production at the RIKEN AVF cyclotron

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²¹¹At is one of the most promising radionuclides for targeted α -particle therapy. The most common method for ²¹¹At production is to irradiate natural Bi with α particles via the ²⁰⁹Bi $(\alpha, 2n)^{211}$ At reaction. We have been developing production technologies of $^{211}\mathrm{At}$ at the RIKEN AVF cyclotron. $^{1-3)}$ In our current method, $^{3)}$ a metallic $^{209}\mathrm{Bi}$ target with a typical thickness of 20 mg/cm^2 on a 1-mm Al backing is irradiated with a 28.0-MeV α beam at an angle of 15° to the beam axis. During the irradiation, the target is cooled with circulating water $(1.5 \text{ L/min}, 10^{\circ}\text{C})$ and He gas (30 L/min). The maximum beam intensity of 10 particle μA (p μA) can produce ²¹¹At with a thick target yield of 48 MBq/p μ A h. After the irradiation, the target was placed in a quartz tube, was heated up to 850° C, and kept for 10 min with O₂-gas flowing at 10 mL/min. 211 At sublimated from the target was trapped by a PFA tube cooled at -96° C. 200–400 μ L $CHCl_3$ was used to wash the PFA tube to collect ²¹¹At in a glass v-vial. Finally, the CHCl₃ solution was dried to obtain a dry 211 At in the v-vial with N₂-gas flowing at 100 mL/min.

The demand for RIKEN ²¹¹At is increasing every year. It is desirable to increase the production yield of ²¹¹At by irradiating the ²⁰⁹Bi target with a more intense beam. Due to the poor thermal conductivity (9.79 W/(m·K)) and a low melting point (272°C) of Bi, an effective cooling system is essential. In this work, we modified the channel of the cooling water to increase its flow rate from 1.5 to 4.0 L/min. We also lowered the temperature of the water from 10 to 5°C. However, we could prepare only one dry ²¹¹At in a single drying and it took approximately 15 min to dry 200 μ L of CHCl₃. To increase the productivity of the dry ²¹¹At, we developed a rapid drying apparatus that makes it possible to simultaneously produce 4 v-vials of dry ²¹¹At within less time.

The ²⁰⁹Bi targets with thicknesses of 17.7 mg/cm² and 18.6 mg/cm² were irradiated for 40 mins with a 28-MeV α beam using the previous and new irradiation systems, respectively. The beam intensity was 25 particle μ A. After the irradiations, the targets were subjected to γ -ray spectrometry to determine the activity of ²¹¹At.

In a separate experiment, 642 MBq of ²¹¹At in 300 μ L of CHCl₃ was divided into 3 v-vials of 100 μ L each and dried simultaneously in the rapid drying apparatus with N₂-gas flowing at 3 L/min at room temperature and 100 kPa. The N₂ gas was aspirated by a chemical pump and exhausted through 1 M Na₂S₂O₅ solution and a charcoal trap (Fig. 1(b)). The v-vials

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Fig. 1. (a) Photo and (b) schematic of rapid drying apparatus for 211 At.

were subjected to $\gamma\text{-ray}$ spectrometry before and after drying.

Figures 2(a) and 1(b) show photos of the irradiated ²⁰⁹Bi targets with the previous and new irradiation systems, respectively. The target cooled with 1.5-L/min water at 10°C was damaged severely. The produced ²¹¹At activity of 503 ± 16 MBq is only 61% compared with the theoretical yield.⁴⁾ The damage of the target cooled with the 4.0-L/min water at 5°C with the new system seems smaller, and the produced ²¹¹At activity of 667 ± 23 MBq is 82% of the theoretical yield.⁴⁾ As expected, a higher flow rate and lower temperature of cooling water help to produce ²¹¹At more quantitatively. Our current production yield of ²¹¹At (1.0 GBq/h at 28.0 MeV with a 25-particle μ A α beam) was increased by a factor of 2.1 compared to our previous one by a 10-particle μ A α beam irradiation.¹)



Fig. 2. Photos of the 209 Bi targets irradiated with the previous (a) and new (b) irradiation system at the 1.5 and 4.0 L/min flow rate of cooling water, respectively.

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Each 100 μ L of the ²¹¹At CHCl₃ solution in the 3 v-vials was dried within 5 min. The activities of ²¹¹At before and after drying were consistent with each other after the decay corrections. With the rapid drying apparatus, the time required to dry ²¹¹At was significantly reduced.

References

- 1) N. Sato et al., RIKEN Accel. Prog. Rep. 50, 262 (2017).
- 2) S. Yano et al., RIKEN Accel. Prog. Rep. 50, 263 (2017).
- Y. Wang *et al.*, RIKEN Accel. Prog. Rep. 53, 192 (2019).
- 4) S. M. Qaim *et al.*, IAEA Technical Report Series No.473, IAEA, Vienna, Austria (2011).