

## Present status of $^{211}\text{At}$ production at the RIKEN AVF cyclotron

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$^{211}\text{At}$  is one of the most promising radionuclides for targeted alpha radiotherapy owing to its suitable half-life of  $T_{1/2} = 7.214$  h and high alpha-particle emission probability of 100%.<sup>1)</sup> We have been producing  $^{211}\text{At}$  via the  $^{209}\text{Bi}(\alpha, 2n)^{211}\text{At}$  reaction at the RIKEN AVF cyclotron.<sup>2,3)</sup> In this paper, we report improved production technologies of  $^{211}\text{At}$ , which could reduce the separation time from 2 h to 1 h and yield a neutral dry  $^{211}\text{At}$  product instead of the confused  $^{211}\text{At}$  species in the solution.

Figure 1 shows the production process for dry  $^{211}\text{At}$ . A metallic  $^{209}\text{Bi}$  target ( $> 99.999\%$ ,  $20\text{ mg/cm}^2$ ) was prepared through vacuum evaporation on an Al plate (thickness: 1 mm). As shown in Fig. 1(a), the Bi target was placed at an angle of  $15^\circ$  with respect to the beam axis. The target was cooled with water ( $10^\circ\text{C}$ ,  $1.5\text{ L/min}$ ) and He gas ( $30\text{ L/min}$ ) during the irradiation. An alpha beam with an energy of  $29.0 \pm 0.2\text{ MeV}$  was delivered from the RIKEN AVF cyclotron. The precise beam energy was measured using a time-of-flight detector.<sup>4)</sup> A beam-wobbling system was used to rotate the beam spot on the target and to prevent heat concentration. After the beam passed through a Be window ( $18.1\text{ }\mu\text{m}$ ) and He cooling gas ( $65\text{ mm}$ ,  $1.1\text{ bar}$ ), the beam energy on the target was  $28.0 \pm 0.2\text{ MeV}$ , as calculated with LISE (ver. 11.0.72). The beam intensity ( $4\text{--}32\text{ }\mu\text{A}$ ) and irradiation time (2 to 8 h) was varied to meet the users' requirements. Subsequently,  $^{211}\text{At}$  was separated from the Bi target by using a dry distillation technique (see Fig. 1(b)). The irradiated Bi target was placed on a copper tray in a quartz tube and heated up to  $850^\circ\text{C}$  in 10 min, following which it was kept for another 15 min and finally cooled down naturally.  $^{211}\text{At}$  sublimated from the target was transported from the quartz tube through a quartz capillary (*i.d.* =  $2\text{ mm}$ , length =  $13\text{ cm}$ ) to a PFA cold trap (*i.d.* =  $1\text{ mm}$ , length =  $100\text{ cm}$ ) by oxygen gas ( $10\text{ mL/min}$ ). The PFA cold trap was cooled to  $-96^\circ\text{C}$  to collect the gaseous  $^{211}\text{At}$ . As shown in Fig. 1(c), after the distillation, the capillary and the PFA trap tube were washed with chloroform (FUJIFILM Wako Pure Chemical Corporation, Cat. No.: 033-15721, Infinity Pure,  $200\text{--}400\text{ }\mu\text{L}$ ). The eluate collected in a 1-mL V-shaped glass vial was then dried up by dry  $\text{N}_2$  gas ( $100\text{ mL/min}$ ) at room temperature (see Fig. 1(d)). The chemical yields of the dry  $^{211}\text{At}$  products were determined by measuring the  $687\text{ keV } \gamma$ -ray ( $I_\gamma = 0.261\%$ ) with a Ge detector. The chemical purity of the products and the decontamination factor of  $^{209}\text{Bi}$  from  $^{211}\text{At}$  were evaluated with chemical analysis using inductively coupled plasma mass spec-

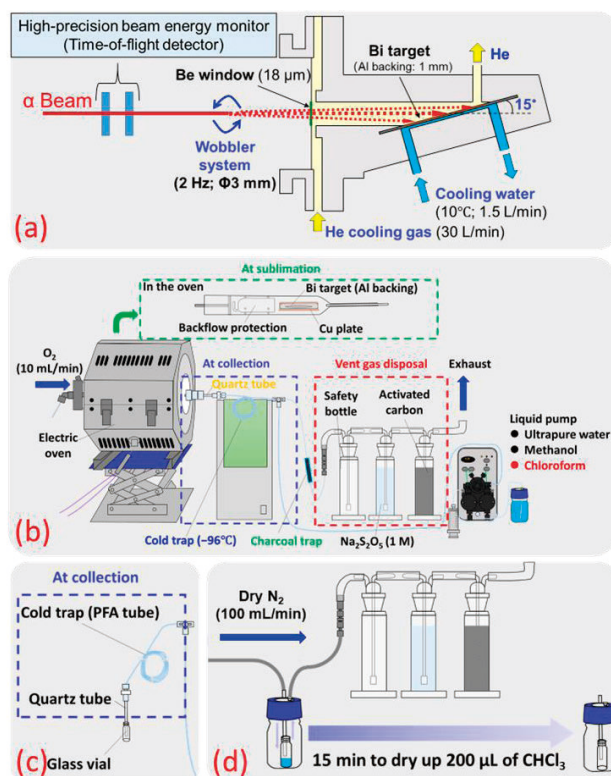


Fig. 1. Production process of dry  $^{211}\text{At}$ .

trometry (ICP-MS).

We confirmed that the radioactivities of  $^{211}\text{At}$  produced in the targets agreed well with those estimated from thick target yields in the literature.<sup>5)</sup> The atomic ratio  $^{210}\text{At}/^{211}\text{At}$  was determined to be less than  $2.1 \times 10^{-6}$  at the end of bombardment (EOB), which satisfied the medical requirement of  $< 1 \times 10^{-3}$  at EOB.<sup>4)</sup> The typical chemical yield was 80%. The chemical impurities ( $> 1\text{ ng}$ ) in  $200\text{ }\mu\text{L}$  of chloroform were Cu ( $43.84\text{ ng}$ ), Al ( $38.90\text{ ng}$ ), Zn ( $23.55\text{ ng}$ ), and Bi ( $6.70\text{ ng}$ ). The decontamination factor of  $^{209}\text{Bi}$  for dry  $^{211}\text{At}$  was determined to be  $8.3 \times 10^{-8}$ .

By using the current production technology of  $^{211}\text{At}$ ,  $1\text{ GBq}$  of  $^{211}\text{At}$  can be produced through 1.5-h irradiation with a  $32\text{-}\mu\text{A}$  alpha beam ( $27.8\text{ MeV}$  on target). Dry  $^{211}\text{At}$  can be prepared 4 h after EOB. We have been distributing  $^{211}\text{At}$  to 14 users in Japan for the research and development of nuclear medicine.

### References

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