Solvent extraction and speciation of astatine species via thin layer chromatography

S. Maruyama,[*1] K. Aoi,[*1] Y. Nagai,[*2] K. Washiyama,[*3] A. Yokoyama,[*2,*4] I. Nishinaka,[*5] Y. Wang,[*4] and H. Haba[*4]

The short path length and high linear energy transfer of α particles are expected to enable targeted alpha therapy for the treatment of cancer. A promising nuclide among various α emitters is ^{211}At with a half-life of 7.21 h, which has gained prominence owing to its appropriate half-life for labelling and drug pharmacokinetics and potential to synthesize labelled compounds as a halogen element. This has motivated several preclinical studies on At chemistry.[1] However, the successive chemical processes for the general use of At chemistry have not been understood well. One of the difficulties in At chemistry is the coexistence of several At species in a sample. For instance, the species of At^-, AtO_3^-, and AtO_4^- are identified in some solutions via thin layer chromatography (TLC).[2] This makes the processes complex, and the species need to be adjusted by varying the redox potential, hydrogen ion concentration, and other factors for appropriate preparation of At solutions.

We aimed to study At species in diisopropyl ether (DIPE) solvent, which was shown to extract cationic species of At in a previous study.[3] Speciation of At was performed via TLC similar to the method in Ref. 2) but in an argon atmosphere, to allow preservation of the species during the drying and development time for the TLC.

In this study, ^{211}At was produced via the ^{209}Bi$(\alpha, 2n)$ reaction at the RIKEN AVF cyclotron and delivered to Kanazawa University. The irradiated Bi target was dissolved in 3 mL of 6 M HNO_3 and mixed with an appropriate amount of H_2O to prepare 1 M HNO_3 solution,

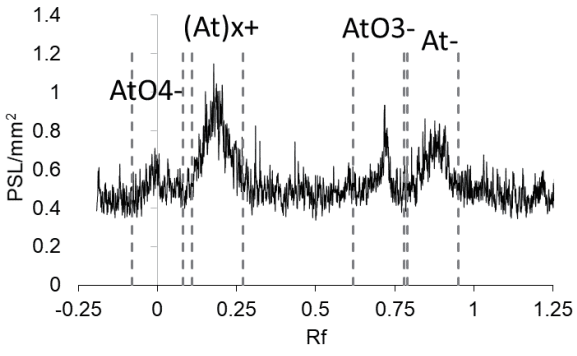

Fig. 1. TLC chromatogram of At species extracted from 9 M HCl.

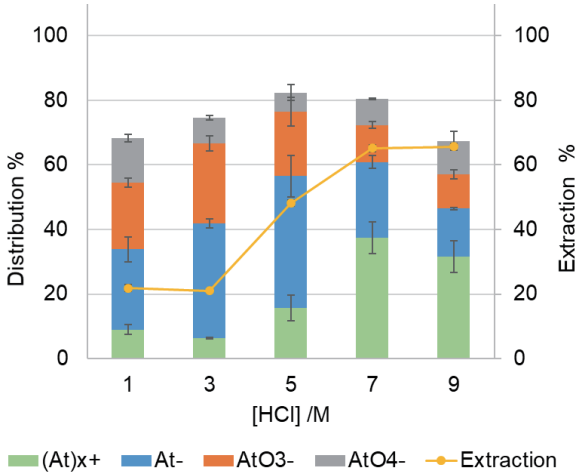

Fig. 2. Distributions of At species extracted to DIPE by TLC and their extraction ratios.

7 mL of which was used to extract the ^{211}At nuclide into 7 mL of dodecane solvent.

Aliquots of the dodecane solution were subjected to back extractions into several solutions of various HCl concentrations ranging from 1 M to 9 M. Approximately 0.5 mL of each HCl solution was subject to solvent extraction with 0.5 mL of DIPE. TLC was performed on 10 μL of each DIPE solution to determine the ^{211}At species.

Figure 1 displays the TLC chromatogram showing a new species at approximately $R_f = 0.2$, which was not found in an open air experiment, in addition to the At^-, AtO_3^-, and AtO_4^-. Figure 2 demonstrates the compositions of the At species, except for the continuum composition deduced from the TLC chromatograms, in Ref. 2), and the At extraction rates dependent on the HCl concentration. The new species increases with the increase in the HCl concentration and the extraction rate. Therefore, the species is considered to be a cation of At based on a previous study.[3]

We are in planning on applying the TLC technique to other extraction systems to identify extracted At species and clarify the mechanism of At extraction. Such information will be useful to construct chemical processes for the general use of At chemistry.

*1 Graduate School of Natural Science and Technology, Kanazawa University
*2 Institute and College of Science and Engineering, Kanazawa University
*3 Fukushima Global Medical Science Center, Fukushima Medical University
*4 RIKEN Nishina Center
*5 Quantum Beam Science Research Directorate, National Institutes for Quantum and Radiological Science and Technology

References

1) F. Guérard, J. -F. Gestin, M. W. Brechbiel, Cancer Biother. Radiopharm. **28**, 1 (2013).
2) I. Nishinaka *et al.*, J. Radioanal. Nucl. Chem. **318**, 897 (2018).
3) T. Taniguchi, Master thesis of Graduate School of Natural Science and Technology, Kanazawa University (2015).