Speciation analysis of oxidation states of astatine extracted into ethanol-water solutions

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The short-path length and high linear energy transfer of α particles are expected to enable targeted alpha therapy for the treatment of tumor. A promising nuclide among various α emitters is ²¹¹At with a halflife of 7.21 h, which has gained popularity owing to its appropriate life and possible compounds as a halogen element. This has been the motivation behind several preclinical studies on At-chemistry¹⁾ However, the successive chemical processes for the general use of Atchemistry has not been well studied. We aim to study the wet chemistry processes of At that may form a prevailing technology.

In previous studies,^{2,3)} the At-211 radioactivity was recovered efficiently from an irradiated Bi target in the chemical systems of dodecane and ethanol solution with oxidizing agents such as N-bromosuccinimide (NBS) and bromine water, which supply Br₂. However, the mechanism of solvent extraction was not clarified in the study. Here, we performed a thin layer chromatography (TLC) experiment to investigate the oxidation states of At species extracted into the system with NBS as well as bromine water, in reference to a TLC study on At species.⁴

The At-211 radioactivity was produced via $^{209}\text{Bi}(\alpha,$ 2n) reaction at the RIKEN AVF cyclotron and delivered to Kanazawa University. The irradiated Bi target was dissolved in 2 mL of 6 M HNO₃ and diluted with 10 mL of H_2O to prepare a 1 M HNO₃ solution, from which the ²¹¹At nuclide was extracted into the dodecane solvent. Aliquots of the solution were subjected to back extractions into 90% ethanol solution with NBS or Br_2 water after 15 min of its addition or without an oxidizing agent. 10 μ L from each sample of ²¹¹At (ca. 15 to 50 Bq) obtained in the back extractions was spotted on a TLC plate (Merck Silica gel 60 F254 aluminum sheet, 10×2 cm) and developed with 50% ethanol solution for 90 min (7 cm in flow distance). After drying, the plate was exposed to an imaging plate for 10 to 20 min to perform an image analysis of its radioactivity.

The images of TLC demonstrated in Fig. 1 suggest that three different species were separated. The results of the analysis exhibit that the oxidizing agents of Br_2 water and NBS significantly increase the oxidized species of AtO_4^- instead of decreasing the others' as

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compared to the case without an oxidizing agent. This indicates that such oxidation due to the agents causes distribution among the species and enhance the distribution ratio of At during extraction. Therefore, the back-extraction favors the oxidized species instead of At⁻.

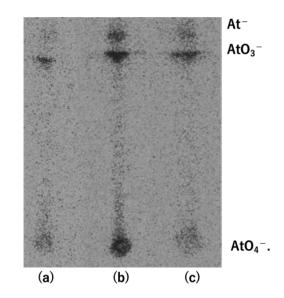


Fig. 1. Imaging of ²¹¹At radioactivity on thin-layer chromatography for (a) Br₂ water, (b) NBS, and (c) no oxidizing agent (see text for detail).

Table 1. Distributions of At chemical species in extracted solutions.

Chemical species	Br ₂ water	NBS	No addition
At ⁻	12 %	13 %	23 %
AtO ₃ ⁻	47 %	45 %	57 %
AtO ₄ ⁻	41 %	42 %	20 %

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