Development of Separation for Carrier-free Astatine Using Column Chromatography

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Astatine-211 (^{211}At) is a nuclide expected to be applied to targeted alpha therapy (TAT). In order to apply ^{211}At for TAT, production of carrier-free astatine is required to prevent unexpected reactions. The main production method of ^{211}At is ^{209}Bi(α, 2n)^{211}At reaction, which used natural bismuth (Bi) for the target. The irradiated targets are purified by dry distillation in many facilities[1)]. Depending on the conditions during vaporization of ^{211}At, however, the yield of ^{211}At can reduce greatly. Although solvent extraction, one of the other separation methods of ^{211}At, is simple method, aqueous solution is contaminated with the organic solvent after back extraction. Thus, the separation method has to further improve or to develop other approach (for example, ^{211}Rn/^{211}At generator system[2,3)]). In this study, we chose column chromatography as a separation method of astatine. This method can be expected high yield of ^{211}At with simple operation.

We produced ^{210}At at Cyclotron and Radioisotope Center (CYRIC), or ^{211}At at Nishina-Center for Accelerator-Based Science, RIKEN. Bismuth oxide (Bi_2O_3) pellet was used for target (~180 mg). The target was irradiated with 50-MeV α particles (100 particle nA) at CYRIC, and 29-MeV α particles (250 particle nA) at RIKEN AVF cyclotron. Quantification of ^{210}At and ^{211}At was all performed using γ-spectroscopy. The activity of ^{210}At and ^{211}At were determined from peaks of 245-keV and 687-keV γ-rays, respectively. The irradiated Bi_2O_3 target was dissolved in 2 mL of 4 mol/L hydrochloric acid (HCl) containing 1 mol/L sodium hydrogen sulfite ($NaHSO_3$). This solution was added a 6 mL of 0.84 mol/L EDTA·2Na aqueous solution (stock solution).

We tried column chromatography experiments using strong anion exchange resin (Muromac© 1X8 100-200 mesh, Muromachi Chemical Co., Ltd.), activated carbon (CNovel®

MH-00, Toyo Tanso Co., Ltd.), and weak anion exchange resin (3-aminopropylsilica gel, Tokyo Chemical Industry Co., Ltd.). In the cases of strong anion exchange resin and activated carbon, almost all At was trapped on the columns. Trapped At was not eluted by concentrated HCl. However, At on the activated carbon column was eluted by 10 M NaOH solution. Therefore, based on Scheme 1, we drawn an elution curve of At in activated carbon. In the case of weak anion exchange resin, almost of all At was not trapped on the column (Scheme 2 and Fig. 2).

$NaHSO_3$ was added as a reducing agent in the stock solution, which was pH 1. Iodine, one of the homologous of astatine, becomes iodide ion (I^-) if there are enough HSO_3^- [4]. Under the condition of pH 1, the chemical form of astatine can be At^-, At^+, or AtO^+.[5, 6] It is known that most of the HSO_3^- ions at pH 1 are SO_3^{2-}. The redox potential of At^- and SO_3^{2-} are +0.35 V and –0.07 V, respectively[7,8]. From the above, it can be inferred that chemical species of astatine was At^- in the stock solution.

As the result of activated carbon column chromatography, the 85% of charged At^- was eluted by 10 column volumes of 10 M NaOH solution. It is suggested that At is oxidized to AtO(OH) at pH 14 which is the condition of the eluent.[5] This result suggested that AtO(OH) do not adsorbed on activated carbon. We found a simple method to separate At in high yield (~86%). However, the solution of At was a strong alkaline. Therefore, using this solution is impossible for biological research. In the case of weak anion exchange column chromatography, Cl^- in the stock solution may have inhibited At trapping. In the absence of any anions, almost all of I^- adsorb on weak anion exchange column.[9] It is necessary to dissolve the irradiated Bi target under conditions not including anions which may inhibit adsorption of astatine to the resin.

In the future, we will consider other dissolution methods and column chromatography of other adsorbents.

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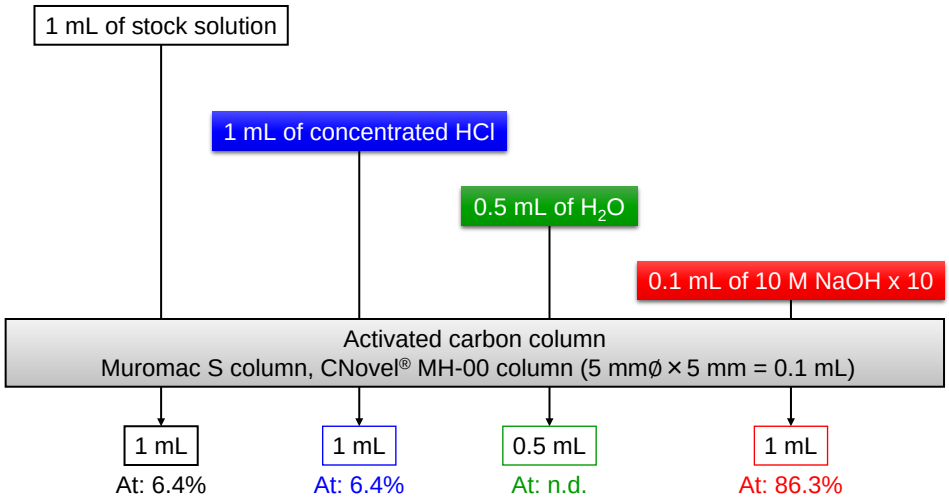
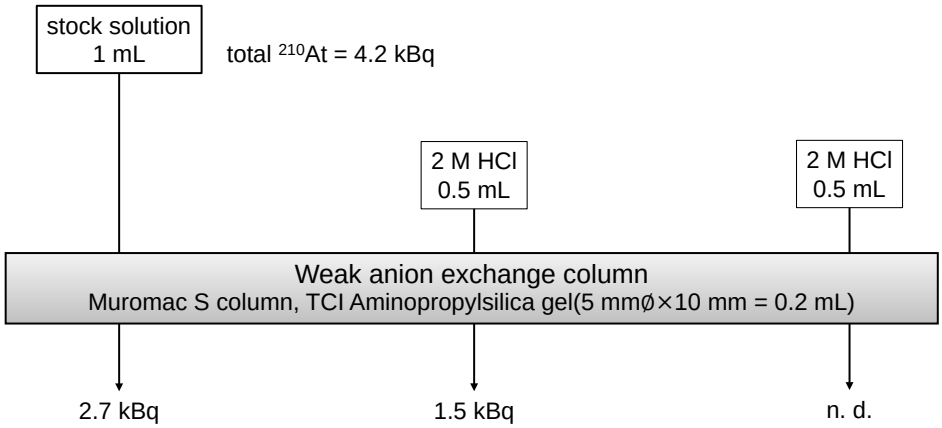


Scheme 1. Separation method of At using column chromatography (adsorbent: activated carbon)



Scheme 2. Separation method of At using column chromatography (adsorbent: weak anion exchange resin)

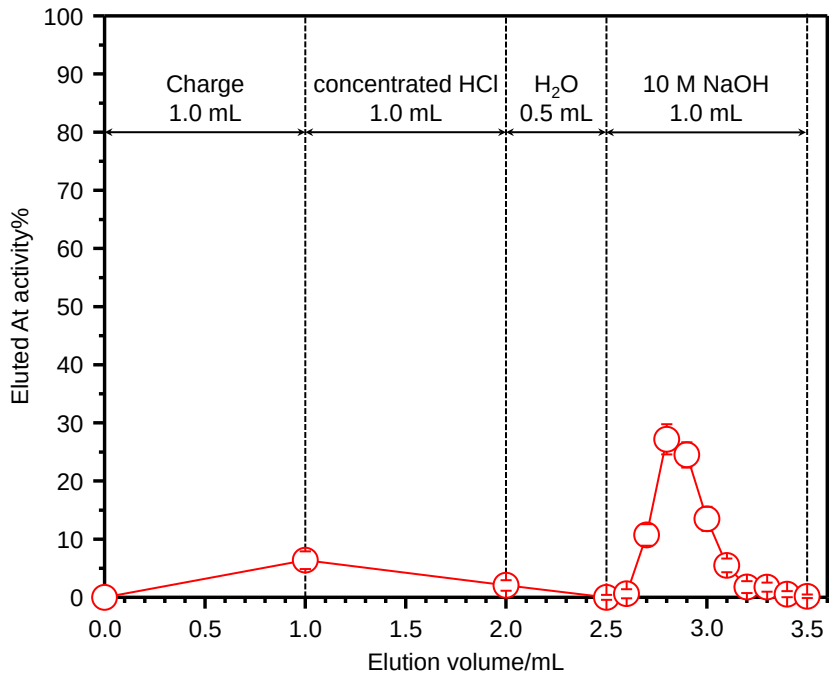


Figure 1. Elution curve of At from activated carbon column

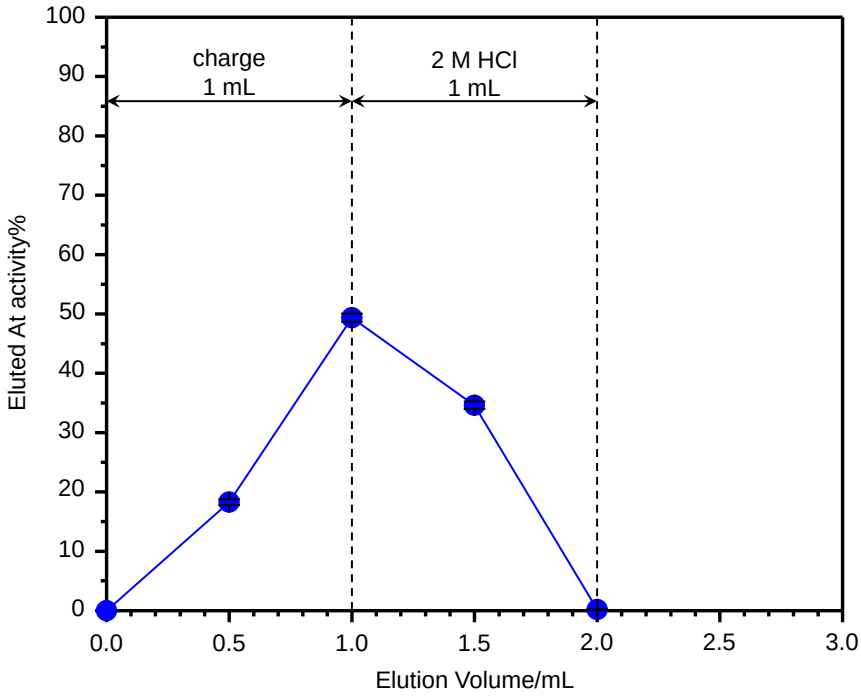


Figure 2. Elution curve of At from weak anion exchange column