

Separation of ^{121m}Te from antimony without redox reaction

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Radioactive tellurium isotopes, such as ^{129}Te , ^{131}Te , and ^{132}Te , were released in the Fukushima Dai-ichi nuclear accident and are regarded as significant for the evaluation of exposure dose during 2011. Owing to their short half-life, radioactive materials derived from the accident have already decayed out; thus, the evaluation requires some laboratory experiments. In experiments where radio nuclides are added to plant and animal samples, carrier-free and high-purity tracers should be employed to avoid undesirable chemical effects. The production of a suitable tracer had been investigated in previous reports where radioactive tellurium generated by the bombardment of Sb_2O_3 with deuterium at an energy of 24 MeV was purified through a redox reaction.^{1,2)} However, this method generates toxic chlorine gas. More moderate methods, therefore, should be developed.

We found that tellurium is separated as a cation from anionic antimony through anion exchange chromatography. Chemical species of both elements were calculated under several conditions, following which tellurium was separated using the calculation results.

Tellurium and antimony exist as chloro-complex anions in a high-concentration HCl solution. With increasing pH, Te(IV) is converted into TeOOH^+ , while Sb(III) forms a precipitate. Through this precipitation, a large part of tellurium is co-precipitated. We considered that tellurium can be purified with careful control of the pH and HCl concentration, and we investigated the optimal conditions using Medusa-Hydra chemical equilibrium software for Windows.³⁾ The calculation results are shown below. Te(IV) was converted into TeOOH^+ at $\text{pH} > -0.2$ and $[\text{HCl}] < 1.6 \text{ M}$. Sb(III) remained as a solution at $\text{pH} < -0.5$ and $[\text{HCl}] > 3.2 \text{ M}$ with $[\text{Sb}] = 200 \text{ mM}$, which is equivalent to 900 mg Sb_2O_3 dissolved in 30 mL HCl and to a concentration derived from ordinary production at one time. At this antimony concentration, tellurium cannot be purified. Based on the fact that the solubility of antimony depends on its concentration, the maximum concentration of antimony at $\text{pH} = -0.2$ without precipitation was calculated to be 7 mM. In order to ensure that the purification was conducted without precipitation, we applied a concentration of 5 mM Sb.

An irradiated Sb_2O_3 pellet was dissolved in concentrated HCl, following which insoluble antimony was removed by centrifugation. The supernatant was diluted with H_2O to 2.4 M HCl and 5 mM Sb solution. The solution was loaded onto anion exchange resin (AG1x8,

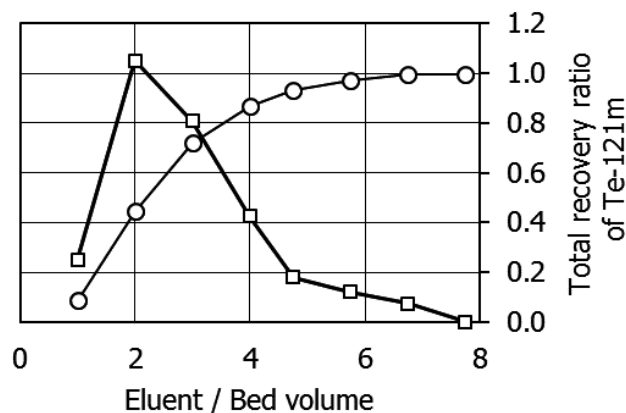


Fig. 1. Elution (□) and total recovery ratio (○) of tellurium from antimony (5 mM) with 1.6 M HCl.

Cl-form, 100–200 mesh, 10 mL), following which Te-121m was eluted with 1.6 M HCl.

Figure 1 shows the elution curve of Te-121m and the total recovery ratio of Te-121m. The total recovery ratio of Te-121m reached almost 100% with five bed volume eluent, while antimony was adsorbed onto the anion exchange resin without elution.

In this research, we investigated the purification of tellurium from matrix antimony with pH regulation. Its performance was satisfactory for a low antimony concentration. Further investigations are required to optimize the separation method for higher concentrations.

References

- 1) T. Kubota *et al.*, RIKEN Accel. Prog. Rep. **50**, 254 (2017).
- 2) T. Kubota *et al.*, RIKEN Accel. Prog. Rep. **52**, 205 (2019).
- 3) I. Puigdomenech, Chemical equilibrium diagrams: MEDUSA, (2010), <https://www.kth.se/che/medusa/downloads-1.386254> (accessed 2020-07-20).

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