

Reversed-phase chromatography of Nb and Ta with TBP for conducting a chemical experiment on the 105th element, Db

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Detailed chemical properties of superheavy elements (SHEs) have not been elucidated owing to the difficulty in conducting chemical experiments because of their short half-lives and extremely low production rates. For investigating the chemical properties of the 105th element, Db, we studied the extraction behavior of the group-5 homolog elements Nb and Ta from HF media with tributyl phosphate (TBP), which is used in the industrial separation of Nb and Ta. In our previous study using batchwise solvent extraction,¹⁾ we observed the differences between the ^{95g}Nb and ¹⁷⁹Ta extraction behaviors; the distribution ratios of ¹⁷⁹Ta reach a maximum at the initial HF concentration, [HF]_{ini} = 0.27 M, whereas those of ^{95g}Nb increase with increasing [HF]_{ini}. To examine the applicability of this extraction system to the short-lived ²⁶²Db with $T_{1/2} = 33.8$ s, we performed an on-line reversed-phase chromatographic experiment on Nb and Ta using Automated Rapid Chemistry Apparatus (ARCA).²⁾

The nuclides ^{90g}Nb ($T_{1/2} = 14.6$ h) and ^{178a}Ta ($T_{1/2} = 2.45$ h) were produced via the ^{nat}Zr(*d,xn*) and ^{nat}Hf(*d,xn*) reactions, respectively, with a 24-MeV deuteron beam supplied from the RIKEN AVF cyclotron. The nuclides produced were transported with a He/KCl gas-jet system and were deposited on a collection site in ARCA for 60 s. Then, the products were dissolved in 1–10 M HF solutions and were loaded onto a column (1.6 mm i.d. × 7.0 mm height) filled with a 62-wt% TBP-laden resin, which was prepared in the procedure described in Ref. 3. The flow rate of the eluent was 1.0 mL/min. The effluent fractions were consecutively collected in 7 polypropylene (PP) tubes. The remaining products in the column were stripped with 330 μ L of 10 M HF at a flow rate of 1.0 mL/min, and the effluent was collected in another PP tube. Each fraction was subjected to γ -ray spectrometry with a Ge detector.

Figure 1 shows the elution curves of ^{90g}Nb and ^{178a}Ta in 1.0 M HF as an example, in which correction for a dead volume of ARCA (40 μ L) was made for the effluent volume. ^{90g}Nb was eluted immediately, whereas ^{178a}Ta showed a broad peak around 770 μ L with a small leakage in the first effluent (less than 5%

of the total activity of ^{178a}Ta). Similar elution behavior of ^{90g}Nb was also observed at higher [HF]_{ini}. The peak volume of the elution curves of ^{178a}Ta gradually decreased, corresponding to the decreasing distribution coefficient (K_d) of ^{178a}Ta on the TBP resin. In Fig. 1, the elution curve of ^{178a}Ta was fitted by the Glöckauf formula of chromatography⁴⁾ except for the first leakage fraction. The K_d values are described as $K_d = v_p/m_r$, where v_p and m_r are the peak volume and mass of dry resin (6.31 mg), respectively. The evaluated K_d values of ^{178a}Ta are shown in Fig. 2 together with those of ¹⁷⁹Ta previously obtained by the batch experiments.¹⁾ The good agreement between the on-line and off-line results indicates that the extraction equilibrium is reached in the present column experiment. Recently, we performed an extraction experiment of Db with the studied system.⁵⁾

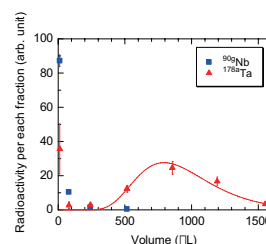


Fig. 1. Elution curves of ^{90g}Nb and ^{178a}Ta in 1.0 M HF on the column of 62 wt% TBP resin.

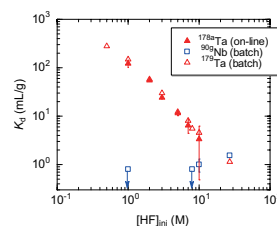


Fig. 2. Variation in K_d values of ^{178a}Ta as a function of [HF]_{ini} (closed symbols). The K_d values of ^{95g}Nb and ¹⁷⁹Ta obtained by the batch experiment¹⁾ are also shown as open symbols.

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References

- 1) S. Tsuto et al.: RIKEN Accel. Prog. Rep. **47**, 270 (2014).
- 2) Y. Nagame et al.: Radiochim. Acta **93**, 519 (2005).
- 3) H. Haba et al.: Radiochim. Acta **95**, 1 (2007).
- 4) E. Glöckauf: Trans. Faraday Soc. **51**, 34 (1955).
- 5) M. Murakami et al.: In this report.