Solid-liquid extraction of ^{261}Rf from hydrochloric acid with Aliquat 336 resin

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The chemical properties of transactinide elements (atomic numbers $Z \geq 104$) are considered to differ from those of its homologues because of the increasingly strong relativistic effects on the valence shell electrons of heavy atoms. The transactinide nuclei are produced at accelerators using heavy-ion-induced nuclear reactions. These nuclei have low production rates and short half-lives ($T_{1/2} \leq \sim 1$ min). Therefore, the chemical experiments of transactinides must be carried out rapidly on one-atom-at-a-time basis using nuclear reaction products transported from the target chamber by a He/KCl gas-jet system. The chemical properties of transactinides have so far been investigated mainly by a partition method based on a comparison of their chemical behavior with those of lighter homologous elements.

Solution chemistry experiments have been often conducted on element 104, rutherfordium (Rf). Anion- and cation-exchange experiments of Rf in HF and HF/HNO$_3$ were successfully performed under conditions such that the distribution behaviors in equilibria were observed for the homologues of Rf. Clearly different behavior of Rf from its homologues Zr and Hf was reported.[1] On the other hand, in the cation exchange in H$_2$SO$_4$/HNO$_3$, the K_d values of Hf in the online experiment were in good agreement with those in the offline batch experiment, whereas the K_d values of Zr in the online experiment were not.[2] Therefore, it is important to investigate the time dependence of the distribution behaviors of transactinides to obtain their equilibrated data because the time required to reach distribution equilibrium would be different among homologues. Thus, we developed a batch-type solid-liquid extraction apparatus to evaluate the time dependence of the distribution behavior of Rf and to obtain the equilibrated K_d values.[3] To determine the experimental condition and obtain comparison data for Rf, we also performed solid-liquid extraction of Zr and Hf and Th (pseudo homologue) with a quaternary ammonium chloride, Aliquat 336, from HCl.[4] In this work, we performed an online solid-liquid extraction experiment on ^{261}Rf and ^{169}Hf using the developed apparatus at RIKEN.

The ^{261}Rf ($T_{1/2} = 68$ s) and ^{169}Hf ($T_{1/2} = 3.25$ min) nuclides were produced in the ^{248}Cm(^{18}O,$5n$)^{261}Rf and ^{nat}Gd(^{18}O,xn)^{169}Hf reactions, respectively, by the RIKEN K70 AVF cyclotron. The nuclear reaction products that recoiled out from the target were caught by KCl aerosols in He gas and were transported by the gas-jet system to the chemistry laboratory. In the solid-liquid extraction experiments, the reaction products in the gas-jet were deposited on a collection site on a dissolution apparatus.

Then, the deposited sample was dissolved in about 0.25 mL of 7.9 and 9.0 M HCl. The solution sample entered the chemical reaction container containing the 28 wt% Aliquat 336 resin (1.11–1.62 mg). After shaking the container with a vortex mixer for 10–60 s, only the solution phase was pushed out of the container by compressed air and collected on the Ta dish. Then the solution sample was evaporated to dryness with hot He gas and a halogen lamp to prepare a sample for α-spectrometry. Subsequently, the Ta dish was transferred to a Si PIN photodiode detector, and α-particle measurement was performed. After the measurement, the γ-ray activity of ^{169}Hf was monitored using a Ge detector to determine its K_d value and chemical yield.

In Fig. 1, the K_d values of Zr, Hf, and Th with the 28wt% Aliquat 336 resin[4] are shown as a function of the HCl concentration. The K_d values of Hf in this work were consistent with those in the offline batch experiment (≥ 4 h of shaking), indicating that the K_d values of Hf in equilibria could be obtained using batch-type solid-liquid extraction apparatus. The 24 and 3 α events were observed in the experiments at 7.9 M HCl (38 extractions, chemical yield: 56%) and at 9.0 M HCl (68 extractions, chemical yield: 47%), respectively, in the α energy region of ^{261}Rf and its daughter ^{257}No. The detected α counts for eluent clearly decreased with an increase in the HCl concentration. This suggests that Rf forms anionic chloride complexes with Cl$^-$ ions, similar to the case of Zr and Hf and different from the case of Th. The present results are consistent with the previous report.[5] We will report the K_d values of Rf in the present experiment in the future.
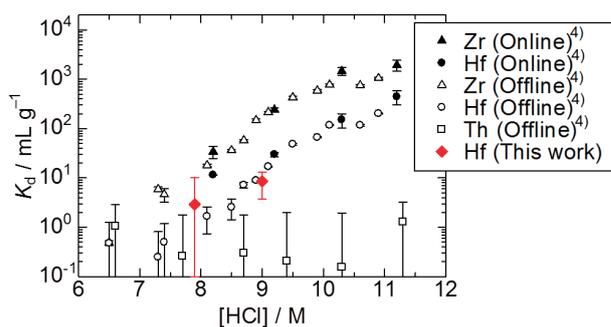

Fig. 1. The K_d values of Zr, Hf, and Th as a function of the HCl concentration with 28 wt% Aliquat 336 resin.

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