Anion and cation exchanges of Zr, Hf, and Th in H_2SO_4 for chemical study of Rf

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Clarifying the chemical properties of superheavy elements with atomic number $Z \geq 104$ is an intriguing and important subject. These elements are produced at accelerators using heavy-ion-induced nuclear reactions. The production rates of these elements are low, and their half-lives are short ($T_{1/2} \leq 1$ min). Thus, chemical studies on these elements are conducted on a single-atom basis.[1)]

The solution chemistry of superheavy elements has been studied mainly for element 104, Rf. In these studies, experiments on Rf and homologous elements have been carried out under the same conditions, but for even homologous elements, equilibrated data were obtained in few conditions. Particularly, it was reported that the chemical reaction kinetics between Zr and Hf are different in H_2SO_4.[2)] Thus, the observation of equilibration and the equilibrated distribution data are very important to characterize the sulfate complex formation of Rf. In this work, we performed anion and cation exchange of Zr, Hf, and Th by using ion-exchange resin and fiber to search the rapid reaction system for the chemical study of Rf.

We produced ^{88}Zr and ^{175}Hf in the ^{89}Y$(d,3n)$ and $^{\mathrm{nat}}$Lu$(d,xn)^{175}$Hf reactions (nat = natural isotopic abundance), respectively, by using the RIKEN AVF cyclotron. ^{234}Th was obtained as an α-decay daughter nuclide of ^{238}U. These radiotracers were purified by an anion-exchange method.

The ion-exchange experiments of Zr, Hf, and Th were performed by the batch method using 0.1–36.3 mg of the ion exchanger (anion-exchange resin (Mitsubishi Chemical, MCI GEL AC08Y), anion-exchange fiber (Nichibi, IEF-BrA-SA), cation-exchange resin (Mitsubishi Chemical, MCI GEL CK08Y), or cation-exchange fiber (Nichibi, IEF-MR-SC)) and 0.25 mL of 0.12–1.0 M H_2SO_4. We also performed control experiments without the resin. The distribution ratio (Q_d) was obtained according to $Q_\mathrm{d} = (A_\mathrm{c} - A_\mathrm{s})V/A_\mathrm{s}w$, where A_c and A_s denote the radioactivities in the control and extracted solutions, respectively; V is the volume of the solution; and w is the weight of the ion exchanger.

The dependence of the Q_d values of Zr, Hf, and Th on the shaking time is shown in Figs. 1(a), (b), and (c), respectively. When using the anion-exchange resin, the Q_d values of Zr, Hf, and Th are constant in the entire time range studied. When using the anion-exchange fiber, the Q_d values of Zr, Hf, and Th become constant after 1 h, 2 h, and 10 s for Zr, Hf, and Th, respectively. When using the cation-exchange resin, the Q_d values become constant after 10 min for Zr and Hf and after 10 s for Th. When using the cation-exchange fiber, the Q_d values of

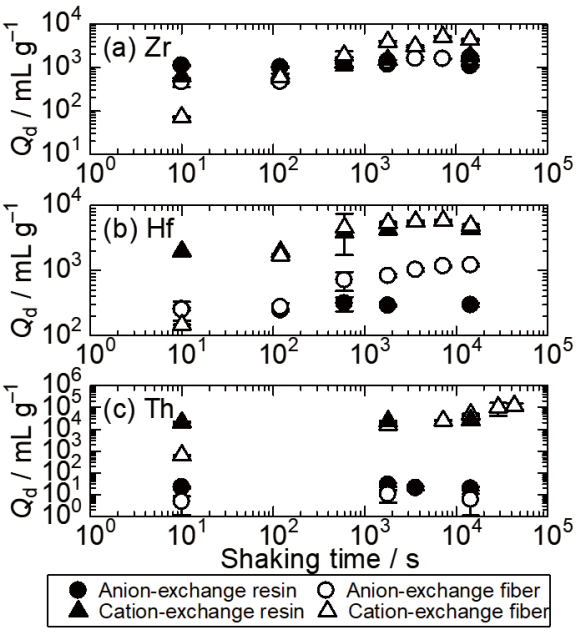

Fig. 1. Variations of the Q_d values of (a) ^{88}Zr, (b) ^{175}Hf, and (c) ^{234}Th on the shaking time in 0.1 M H_2SO_4.

Zr, Hf, and Th become constant after 2 h, 10 min, and 4 h, respectively. These results suggest that the anion-exchange reaction reaches equilibrium within 10 s when using the anion- exchange resin, and this reaction system would be applicable for ^{261}Rf ($T_{1/2}$ = 68 s). On the other hand, the time required to reach equilibria in the ion-exchange reaction with the other ion exchangers used in this work are relatively long compared to that with the short-lived ^{261}Rf. We found that the time required to reach the equilibrium with the ion-exchange resin is shorter than that with the fiber for both anion and cation exchange. We concluded that the ion-exchange fibers are unavailable for experiments with the short-lived ^{261}Rf.

We also studied the variations of the distribution coefficients (Q_d values in equilibrium condition) of Zr, Hf, and Th on the anion-exchange resin to obtain comparison data and to determine the appropriate experimental condition for Rf experiment. We would like to perform a similar experiment on Rf experiments by using AMBER,[3)] to observe the equilibration of the anion-exchange reaction, and to investigate the chemical properties of the sulfate complex of Rf.

References

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