Online anion-exchange experiment of Zr in H_2SO_4 for the chemical study of Rf in H_2SO_4

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The chemical properties of superheavy elements (SHEs) with atomic number $Z \geq 104$ are expected to deviate from the periodicity of their lighter homologues in the periodic table. Thus, chemical studies on SHEs are interesting. These elements are produced at accelerators by 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, the chemical studies of SHEs are conducted on a single-atom basis. Therefore, performing, the chemical experiment of SHEs is difficult. Thus far, the chemical properties of SHEs have been discussed by comparing them with those of their homologous elements.[1)]

In the solution chemistry of SHEs, ion exchange and solvent extraction are often performed.[1)] To study the sulfate complexation of Rf, we plan to perform anion-exchange experiments of Rf along with its homologous elements. By using the batch-type solid-liquid extraction apparatus called AMBER[2)], we measured the K_d values of Rf in 0.060–0.46 M H_2SO_4.[3,4)] In our previous study, to obtain the comparison data for Rf, we performed an online anion-exchange experiment of Zr and Hf by using AMBER.[5)] However, the K_d values of Zr had large errors, because the radioactivity of ^{85}Zr was very low. In this work, we re-examined the online anion-exchange behavior of ^{89m}Zr with higher radioactivities.

^{89m}Zr was produced in the ^{89}Y$(d, 2n)^{89m}$Zr reaction by using the K70 AVF cyclotron at RIKEN. The nuclear reaction products were transported to a chemistry room by a He/KCl gas-jet system. The transported products were deposited on the collection site of the AMBER's dissolution equipment for 1 min and were then dissolved with 0.23 mL of 0.060, 0.11, 0.26, 0.30, 0.46, and 0.72 M H_2SO_4. The solution sample was injected into a chemical reaction container containing 1.4–3.8 mg of an anion-exchange resin (Mitsubishi Chemical Co., MCI GEL CA08Y). After shaking the container with a shaker for 10, 30, and 90 s, only the solution phase was discharged from the container by using a PTFE filter with compressed air. The discharged solution was collected in a PP tube, and was assayed by γ-ray spectroscopy. We also performed control experiments with 10, 30, and 90 s shaking without the resin to determine the standard radioactivity of the solution sample. The K_d values were determined from the following equation:

$$K_d = A_r V_s / A_s w_r = (A_c - A_s) V_s / A_s w_r \quad (1)$$

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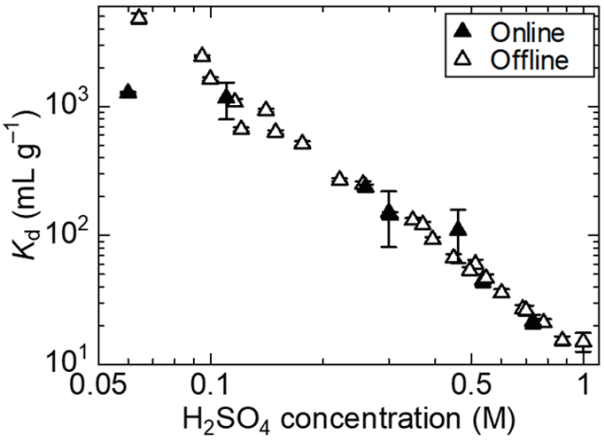

Fig. 1. The K_d values of Zr in anion exchange as a function of H_2SO_4 concentration.

where A_r, A_s, and A_c are radioactivities of the resin, the solution, and the control solution, respectively; V_s is the volume (mL) of the solution; and wr is the mass of the dry resin (g).

Figure 1 shows the H_2SO_4-concentration dependence of the K_d values of Zr in the online and offline experiments. The K_d values of Zr in the online experiment are in agreement with those in the offline experiment within the error range in 0.11–0.72 M H_2SO_4. This indicates that the K_d values of Zr in equilibrium are obtained in the online experiment at these H_2SO_4 concentrations. However, at 0.060 M H_2SO_4 the K_d value of Zr in the online experiment was lower than that in the offline experiment. A probable reason for this disagreement is that the K_d value at this low concentration is too high to be determined with the AMBER system. Another possibility is that the chemical reaction time is not sufficient for the anion-exchange equilibrium. Therefore, it is important to directly investigate the reaction time dependence of the anion-exchange behavior of Rf to confirm the anion-exchange reaction equilibrium of Rf.

From this work, we obtained the comparison data for Rf. We will discuss the sulfate complexation of Rf based on the anion-exchange behavior of Rf, Zr, Hf, and Th in a separate study.[4)]

References

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