

## Anion-exchange Behavior of Rf in HCl and HNO<sub>3</sub> Solutions

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A short review is given on our current anion-exchange studies of element 104, rutherfordium (Rf), in HCl and HNO<sub>3</sub> solutions. The distribution coefficients of the Rf homologues Zr, Hf, and Th(IV), and Pu(IV) on an anion-exchange resin were measured in 1.0–11.5 M HCl and 1.1–13.1 M HNO<sub>3</sub> with a batch method using the radiotracers <sup>88</sup>Zr, <sup>175</sup>Hf, <sup>234</sup>Th, and <sup>237</sup>Pu. In experiments for the short-lived <sup>261</sup>Rf, the isotopes <sup>85</sup>Zr, <sup>169</sup>Hf, and <sup>261</sup>Rf were produced in the <sup>18</sup>O-induced reactions on <sup>nat</sup>Ge, <sup>nat</sup>Gd, and <sup>248</sup>Cm targets, respectively, and their anion-exchange behavior in 4.0–11.5 M HCl and 8.0 M HNO<sub>3</sub> was investigated using the Automated Ion-exchange separation apparatus coupled with the Detection system for Alpha spectroscopy (AIDA). In the HCl system, the percent adsorption of Rf on the anion-exchange resin increases steeply with increasing HCl concentration from 7.0 M to 11.5 M. This adsorption behavior is similar to that of the group-4 elements Zr and Hf, and is quite different from that of the pseudo-homologue Th(IV). The percent adsorption decreases in the order Rf > Zr > Hf. In the HNO<sub>3</sub> system, Rf behaves like Zr and Hf in 8.0 M HNO<sub>3</sub> but not like Pu(IV) and Th(IV), implying the formation of cationic or neutral species.

### 1. Introduction

Chemical behavior of Rf in hydrochloric acid solutions has been investigated to extract information on chloride complexation and hydrolysis.<sup>1–5</sup> Hulet et al.<sup>1</sup> compared the formation of anionic chloride complexes of Rf to those of Hf, Cm, and Fm by testing their relative absorption from 12 M HCl onto a column containing trioctylmethylammonium chloride (Aliquat-336). Czerwinski et al.<sup>2</sup> extracted Rf into an organic phase of 1.0 M and 0.1 M triisooctylamine (TIOA) in benzene from 12 M HCl. Both the authors found that the chloride complexation of Rf is stronger than that of the trivalent group-3 elements and is similar to that of the group-4 elements Zr and Hf. On the other hand, extraction behavior of Zr, Hf, Th, Pu, and Rf into 0.25 M tributylphosphate (TBP) in benzene was examined by Czerwinski et al.<sup>3</sup> in 8–12 M HCl. The results showed that extraction for these elements increases or remains high as a function of HCl concentration and that the extractability increases in the order of Hf < Rf < Zr. Bilewicz et al.<sup>4</sup> studied adsorption behavior of Zr, Hf, Th, and Rf on glass supports coated with cobalt ferrocyanide, and interpreted the result such that the hydrolysis trend increases in the order Th < Zr ≈ Hf < Rf. Both the experiments<sup>3,4</sup> have been criticized for some experimental deficiencies<sup>6</sup> and may not have revealed a true order in the group-4 elements. Recently, a chromatographic separation of Zr, Hf, and Rf in the 8 M HCl-TBP system was performed by Günther et al.<sup>5</sup> using micro columns containing TBP on an inert support. The

extraction order Hf < Rf < Zr was obtained.

An anion-exchange experiment on Rf allows us to study the formation of chloride complexes of Rf in more detail and to verify the previous results<sup>1–5</sup> from different viewpoints. The distribution coefficients ( $K_d$ ) of Zr and Hf on an anion-exchange resin in HCl solutions were measured by Huffman et al.<sup>7</sup> At <7 M HCl, Zr and Hf are present as cationic or neutral species, and their negligible adsorption was observed on anion-exchange resin. At higher concentrations, anionic species are formed and adsorbed with high  $K_d$  values. Under these conditions, the  $K_d$  values of Zr are higher than those of Hf. A good separation between Zr and Hf was accomplished at 9 M HCl using an anion-exchange column.<sup>7</sup>

Chemical behavior of Rf in pure nitric acid solutions has not been investigated yet. In HNO<sub>3</sub> solutions, the variation of the  $K_d$  values of Zr and Hf on an anion-exchange resin is expected to be quite different from that of Th(IV) and Pu(IV),<sup>8</sup> though the  $K_d$  measurements for Zr and Hf in Reference 8 are incomplete.

In the present study, we first measured the  $K_d$  values of Zr, Hf, Th, and Pu on the anion-exchange resin CA08Y in 1.0–11.5 M HCl and in 1.1–13.1 M HNO<sub>3</sub> using carrier-free radiotracers of <sup>88</sup>Zr, <sup>175</sup>Hf, <sup>234</sup>Th, and <sup>237</sup>Pu in order to find a suitable experimental condition for <sup>261</sup>Rf. The Automated Ion-exchange separation apparatus coupled with the Detection system for Alpha spectroscopy (AIDA) was developed to perform fast and repetitive ion exchange chromatography of <sup>261</sup>Rf. The isotopes <sup>85</sup>Zr, <sup>169</sup>Hf, and <sup>261</sup>Rf were produced in the <sup>nat</sup>Ge(<sup>18</sup>O, xn), <sup>nat</sup>Gd(<sup>18</sup>O, xn), and <sup>248</sup>Cm(<sup>18</sup>O, 5n) reactions, respectively, and their anion-exchange behavior was successfully investigated with AIDA in 4.0–11.5 M HCl and 8.0 M HNO<sub>3</sub>.

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## 2. Experimental

**2.1. Batch Experiments.** The carrier-free radiotracers  $^{88}\text{Zr}$ ,  $^{175}\text{Hf}$ , and  $^{237}\text{Pu}$  were produced in the  $^{89}\text{Y}(p, 2n)$ ,  $^{175}\text{Lu}(p, n)$ , and  $^{237}\text{Np}(p, n)$  reactions, respectively, using the JAERI tandem accelerator.  $^{234}\text{Th}$  was chemically separated from commercial  $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  as a daughter of  $^{238}\text{U}$ . These tracers were stored in polypropylene vessels as 6.0 M HCl or 8.0 M  $\text{HNO}_3$ .

The anion-exchange resin MCI GEL CA08Y with a particle size of about 20  $\mu\text{m}$  supplied by Mitsubishi Chemical Corporation was dried to a constant weight at 80 °C in a vacuum oven. 10–200 mg of CA08Y and 3 mL of a desired solution containing 50  $\mu\text{L}$  of the tracer solution were taken in a polypropylene tube, and shaken for 30 min at 20 °C. After centrifugation, 1 mL of the aqueous phase was pipetted into a polyethylene tube and subjected to  $\gamma$ -ray spectrometry with a Ge detector. As a standard, 50  $\mu\text{L}$  of the tracer solution was diluted to 1 mL with 6.0 M HCl or 8.0 M  $\text{HNO}_3$  in another polyethylene tube.

### 2.2. On-line Experiments

**2.2.1. Production of  $^{261}\text{Rf}$ ,  $^{85}\text{Zr}$ , and  $^{169}\text{Hf}$ .** The isotope  $^{261}\text{Rf}$  was produced by the  $^{248}\text{Cm}(^{18}\text{O}, 5n)$  reaction using a 94 MeV  $^{18}\text{O}^{6+}$  beam from the JAERI tandem accelerator. The  $^{248}\text{Cm}$  target of 610  $\mu\text{g}/\text{cm}^2$  thickness and 5 mm diameter was prepared by electrodeposition of  $\text{Cm}(\text{NO}_3)_3$  in 2-propanol onto a 2.4 mg/cm<sup>2</sup> thick beryllium backing foil. The target contained Gd (39.3%-enriched  $^{152}\text{Gd}$ ) of 36  $\mu\text{g}/\text{cm}^2$  thickness to produce Hf isotopes simultaneously and to monitor behavior of Hf in the identical experimental condition as  $^{261}\text{Rf}$ . The 108.5-MeV  $^{18}\text{O}$  beam passed through a Havar vacuum window (2.0 mg/cm<sup>2</sup>), helium cooling gas (0.09 mg/cm<sup>2</sup>), the beryllium target backing, and finally entered the target material at 94 MeV. At this energy, the excitation function for the  $^{248}\text{Cm}(^{18}\text{O}, 5n)^{261}\text{Rf}$  reaction exhibits a maximum of 13 nb.<sup>9</sup> The beam intensities were 200–300 particle nA.

In order to investigate behavior of Zr and Hf simultaneously,  $^{85}\text{Zr}$  and  $^{169}\text{Hf}$  were produced from a target consisting of  $^{nat}\text{Ge}$  and  $^{nat}\text{Gd}$  by the  $^{nat}\text{Ge}(^{18}\text{O}, xn)$  and  $^{nat}\text{Gd}(^{18}\text{O}, xn)$  reactions, respectively. The  $^{nat}\text{Gd}$  target of 370  $\mu\text{g}/\text{cm}^2$  thickness and 5 mm diameter was electrodeposited on a 2.7 mg/cm<sup>2</sup> thick beryllium backing foil, and then on the resulting Gd target the Ge target of 660  $\mu\text{g}/\text{cm}^2$  thickness was deposited by evaporation in vacuum.

Reaction products recoiling out of the target were stopped in helium gas (~1 bar), attached to KCl aerosols generated by sublimation of KCl powder at 620–650 °C, and continuously transported through a Teflon capillary (2.0 mm inner diameter, 20 m long) to a chemistry room. The transport efficiency of  $^{261}\text{Rf}$  was estimated to be 33%.<sup>9</sup>

**2.2.2. On-line Anion Exchange Chromatography.** The on-line anion exchange chromatography of  $^{261}\text{Rf}$ ,  $^{85}\text{Zr}$ , and  $^{169}\text{Hf}$  was performed using AIDA.<sup>10</sup> The chromatography unit of AIDA was almost the same as that of the Automated Rapid Chemistry Apparatus (ARCA) developed by Schädel et al.<sup>11</sup> The reaction products transported by the He/KCl gas-jet system were deposited on the collection site of AIDA. After the deposition for 125 s, the collection site was moved on top of the micro column where  $^{261}\text{Rf}$  and  $^{169}\text{Hf}$  were dissolved with 170  $\mu\text{L}$  of conc. HCl (11.5 M) and fed into the column at a flow rate of 1.0 mL/min. On the average,  $6.56 \pm 0.31$  mg of the anion-exchange resin CA08Y were filled in each 7 mm long column of 1.6 mm inner diameter. The effluent was collected on a tantalum disk as Fraction 1 and was evaporated to dryness with hot He gas and a halogen heat lamp. The remaining Rf and Hf in the column were eluted with 230  $\mu\text{L}$  of 4.0 M HCl at 1.1 mL/min. This effluent was collected on another Ta disk and evaporated to dryness as Fraction 2. In the experiments at 4.0, 7.0, 8.5, 9.0, and 9.5 M HCl, Rf and Hf were first fed into the column with 170  $\mu\text{L}$  of conc. HCl and the effluent was discarded. Then, 290  $\mu\text{L}$  of 4.0–9.5 M HCl was pumped to the column and the ef-

fluent was collected on the Ta disk as Fraction 1. The remaining Rf and Hf were eluted with 250  $\mu\text{L}$  of 4.0 M HCl as Fraction 2.

In the  $\text{HNO}_3$  experiment, Rf and Hf, dissolved in 190  $\mu\text{L}$  of 8.0 M  $\text{HNO}_3$ , were fed into the column at 1.2 mL/min and the effluent was collected as Fraction 1. Then, 220  $\mu\text{L}$  of 4.0 M HCl was used as Fraction 2.

Each pair of Ta disks, Fractions 1 and 2, were automatically transferred to  $\alpha$ -spectrometry station of eight 600 mm<sup>2</sup> passivated ion-implanted planar silicon (PIPS) detectors. Every detector had a 40% counting efficiency and its  $\alpha$ -particle energy resolution was about 80 keV FWHM. All events were registered event-by-event. After the  $\alpha$  measurement, the 493 keV  $\gamma$  ray of  $^{169}\text{Hf}$  was monitored by Ge detectors for every third or fourth pair of samples to determine the distribution of Hf and its chemical yield. The average chemical yield of Hf was about 60% including the deposition efficiency of aerosols.

The experiments with  $^{85}\text{Zr}$  and  $^{169}\text{Hf}$ , produced from the Ge/Gd target, were performed in 4.0–11.5 M HCl and in 8.0 M  $\text{HNO}_3$  under the same experimental conditions as those with  $^{261}\text{Rf}$ . The effluents were collected in polyethylene tubes and were investigated by  $\gamma$ -ray spectrometry.

## 3. Results and Discussion

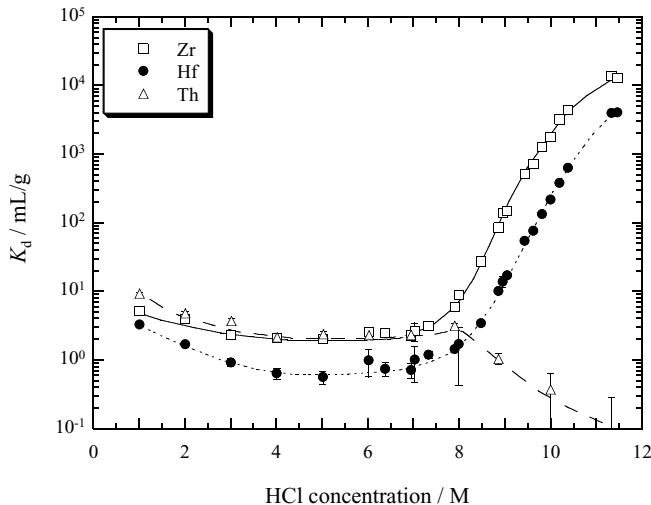
**3.1. Batch Experiments.** The distribution coefficient  $K_d$  in units of mL/g was calculated using the following equation:

$$K_d = \frac{A_r V_s}{A_s W_r}, \quad (1)$$

where  $A_r$  and  $A_s$  are the activities in the resin and the solution, respectively.  $V_s$  is the volume of the acid solution [mL] and  $W_r$  is the weight of the resin [g]. The variation of the  $K_d$  values on CA08Y for Zr, Hf, and Th(IV) is shown in Figure 1 as a function of HCl concentration. In the range of 1.0–7.0 M, the  $K_d$  values of these three elements decrease slightly with increasing HCl concentration and become almost constant, about 2 mL/g for Zr and Th and about 0.6 mL/g for Hf. Here, Zr and Hf are expected to be present as cationic and/or neutral species, i.e.,  $[\text{M}(\text{OH})_2\text{Cl}]^+$ ,  $\text{M}(\text{OH})_2\text{Cl}_2$ , and  $\text{M}(\text{OH})\text{Cl}_3$  ( $\text{M}=\text{Zr}, \text{Hf}$ ).<sup>12</sup> At higher concentrations, the  $K_d$  values of both Zr and Hf increase steeply with increasing HCl concentration, indicating that anionic species such as  $[\text{M}(\text{OH})\text{Cl}_5]^{2-}$  and  $[\text{MCl}_6]^{2-}$  are formed. On the other hand, the  $K_d$  value of Th decreases with increasing HCl concentration above 8 M; no anionic chloride species are known for Th. The  $K_d$  values of Zr are higher than those of Hf at all the concentrations studied. It was found that the separation factor  $K_d(\text{Zr})/K_d(\text{Hf})$  shows a maximum of 10 at 9.5 M HCl, as reported by Huffman et al.,<sup>7</sup> and that the  $K_d$  value of Th (0.46 mL/g) at 9.5 M HCl is also quite different from those of Zr (650 mL/g) and Hf (60 mL/g). An anion-exchange experiment of Rf at 9.5 M HCl can be considered appropriate to find a difference in the chloride complexation from its homologous elements.

In Figure 2, the variation of the  $K_d$  values on CA08Y for Zr, Hf, Th(IV), and Pu(IV) is shown as a function of  $\text{HNO}_3$  concentration. The  $K_d$  values of Zr and Hf decrease gradually with  $\text{HNO}_3$  concentration and they are almost identical. On the other hand, those of Th and Pu increase with increasing  $\text{HNO}_3$  concentration and attain a maximum around 8 M  $\text{HNO}_3$ . The  $K_d$  values of Th and Pu at 8 M are more than one order of magnitude higher than those of Zr and Hf, reflecting the difference in the complex structures; Th and Pu are expected to form anionic complexes  $[\text{M}(\text{NO}_3)_6]^{2-}$ , while Zr and Hf form neutral or cationic ones. Thus, the anion-exchange experiment of Rf was performed at 8 M  $\text{HNO}_3$ .

**3.2. On-line HCl Experiments.** From ion-exchange experiments performed 245, 366, 400, 395, 328, and 159 times at 11.5, 9.5, 9.0, 8.5, 7.0, and 4.0 M HCl, respectively, a total of 186  $\alpha$  events from the 78-s  $^{261}\text{Rf}$  (8.28 MeV)<sup>13</sup> and its daugh-



**Figure 1.** Variation of the distribution coefficient ( $K_d$ ) for Zr, Hf, and Th(IV) on the anion-exchange resin CA08Y as a function of HCl concentration. The associated errors were evaluated from counting statistics. See text for other details.

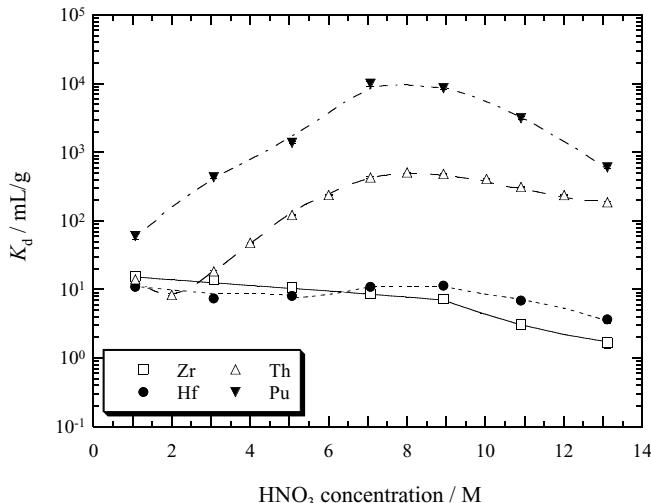
ter 25-s  $^{257}\text{No}$  (8.22, 8.27, 8.32 MeV)<sup>13</sup> were registered in the energy range of 8.00–8.36 MeV, including 35 time-correlated  $\alpha$  pairs. The event ratio between  $\alpha$  singles and  $\alpha$ - $\alpha$  correlations, estimated from the 40% counting efficiency of the detector, is 3 : 1, which is consistent with the observed event ratio of 116 (= 186 – 35 × 2) : 35 within the counting statistics.

From the activities  $A_1$  and  $A_2$  observed in Fractions 1 and 2, respectively, the percent adsorption (%ads) on CA08Y was calculated according to the following equation:

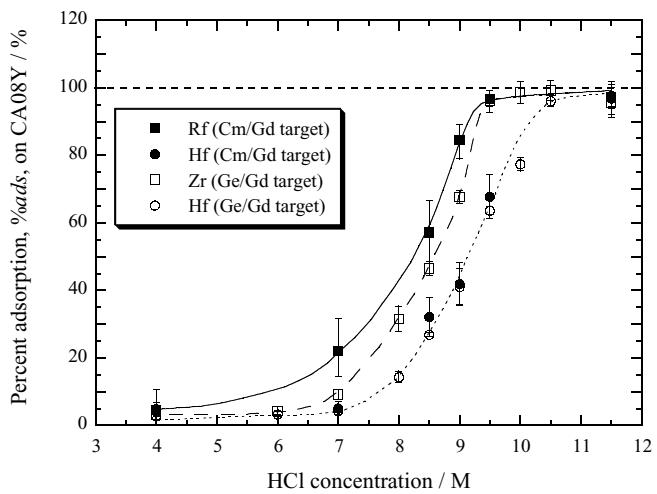
$$\%ads = \frac{100A_2}{A_1 + A_2}. \quad (2)$$

The contribution of  $^{257}\text{No}$  formed from  $^{261}\text{Rf}$  was taken into account based on the %ads values for No determined in the separate experiments using  $^{255}\text{No}$  produced in the  $^{248}\text{Cm}(^{12}\text{C}, 5n)$  reaction.<sup>14</sup> The %ads value of  $97^{+3}_{-6}\%$  at 11.5 M HCl indicates that Rf forms anionic chloride complexes which are strongly adsorbed on the anion-exchange resin. This is consistent with the previous findings in the extraction experiments into Aliquat-336 and TIOA<sup>1,2</sup> as mentioned in sect. 1. In addition, the %ads values of  $^{85}\text{Zr}$  and  $^{169}\text{Hf}$  from 11.5 M are  $96 \pm 2\%$  and  $98 \pm 4\%$ , respectively. These high adsorptions indicate that the loading of these elements onto the column with 170  $\mu\text{L}$  of 11.5 M HCl was completely performed in the experiments at 4.0–9.5 M HCl.

The variation of the %ads values on CA08Y is shown as a function of HCl concentration in Figure 3. The data for  $^{261}\text{Rf}$  and  $^{169}\text{Hf}$  obtained from the  $^{248}\text{Cm}/^{152}\text{Gd}$  target are shown by closed



**Figure 2.** Variation of the distribution coefficient ( $K_d$ ) for Zr, Hf, Th(IV), and Pu(IV) on CA08Y as a function of HNO<sub>3</sub> concentration.



**Figure 3.** Variation of the percent adsorption (%ads) for Zr, Hf, and Rf on CA08Y as a function of HCl concentration. The associated errors for  $^{261}\text{Rf}$  were evaluated from counting statistics and those for  $^{169}\text{Hf}$  and  $^{85}\text{Zr}$  were standard deviations in the replicate experiments. See text for other details.

squares and closed circles, respectively. Those for  $^{85}\text{Zr}$  and  $^{169}\text{Hf}$  from the  $^{nat}\text{Ge}/^{nat}\text{Gd}$  target are indicated by open squares and open circles, respectively. The data of Hf from both targets agree well. The %ads values of Rf increase rapidly with increasing HCl concentration from 7.0 M to 9.5 M, indicating that anionic chloride complexes such as  $[\text{Rf}(\text{OH})\text{Cl}_5]^{2-}$  or  $[\text{RfCl}_6]^{2-}$  are formed. This adsorption behavior of Rf is very similar to that of the group-4 homologues Zr and Hf, and is quite different from that of the pseudo-homologue Th as expected from the low  $K_d$  values in Figure 1. It is interesting to note that the adsorption order is Rf > Zr > Hf, especially at 9.0 M HCl. This may indicate that the chloride complexing strength in group-4 elements decreases in the order of Rf > Zr > Hf. The %ads values of Zr, Hf, and Rf at 4.0 M HCl are  $3.0 \pm 0.3\%$ ,  $3.0 \pm 0.3\%$ , and  $4.5^{+6.3}_{-3.2}\%$ , respectively. These values are low enough for these elements to be stripped from the anion-exchange column.

**3.3. On-line HNO<sub>3</sub> Experiments.** From anion exchange experiments for 8.0 M HNO<sub>3</sub> performed 217 times, a total of 20  $\alpha$  events from  $^{261}\text{Rf}$  and  $^{257}\text{No}$  were registered in the energy range of 8.00–8.36 MeV, including 5 time-correlated  $\alpha$  pairs. The %ads values of  $^{261}\text{Rf}$  and  $^{169}\text{Hf}$  obtained from the Cm/Gd target on CA08Y with 190  $\mu\text{L}$  of 8.0 M HNO<sub>3</sub> are  $13^{+11}_{-7}\%$  and  $12 \pm 7\%$ , respectively. The values for  $^{85}\text{Zr}$  and  $^{169}\text{Hf}$  from the Ge/Gd target are  $5.5 \pm 1.3\%$  and  $2.9 \pm 1.7\%$ , respectively. In order to determine the %ads value of Th, the tracer solution of  $^{234}\text{Th}$  was pipetted and evaporated to dryness on the collection site of AIDA, and the ion exchange of  $^{234}\text{Th}$  was performed under the same experimental condition as those for  $^{261}\text{Rf}$ . The %ads value of  $^{234}\text{Th}$  at 8.0 M HNO<sub>3</sub> was found to be  $99 \pm 3\%$ . More than 80% of Zr, Hf, and Rf were eluted from the column with 190  $\mu\text{L}$  of 8 M HNO<sub>3</sub>, while Th was strongly adsorbed. This indicates that Rf forms cationic or neutral species in 8.0 M HNO<sub>3</sub> like Zr and Hf but not like Th(IV) and Pu(IV).

The structures of Zr, Hf, and Th complexes in the aqueous systems are quite important for discussion of the results of ion-exchange experiments and also of theoretical molecular orbital calculations. Recently, we measured X-ray Absorption Fine Structure (XAFS) spectra of Zr and Hf in 1.0–11.5 M HCl and 8.0 M HNO<sub>3</sub> at the High Energy Accelerator Research Organization Photon Factory (KEK-PF). In addition, the relativistic density functional calculations for the hexachloride- and hexanitrate-complexes of Zr, Hf, Th, and Rf are under study in our group.<sup>15,16</sup> These results will be reported elsewhere.

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