Anion-exchange behaviour of Nb, Ta and Pa in H_2SO_4

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Studies on the behavior of the group-4 elements Zr and Hf, which are homologous elements of Rf, have been conducted in H_2SO_4.[1] It was found that the ion-exchange reaction of Zr and Hf is slow in H_2SO_4 and cannot reach equilibrium in current superheavy element studies. On the other hand, there are not many researches on the group-5 elements in H_2SO_4. Similarly to the group-4 elements, the ion-exchange reaction of the group-5 elements Nb and Ta, and their pseudo homologue Pa may be slow. To elucidate the behaviour of Db in H_2SO_4, it is necessary to clarify the behaviour of Nb, Ta and Pa in H_2SO_4.

^{95}Nb ($T_{1/2}$ = 35.0 d) and ^{179}Ta ($T_{1/2}$ = 1.82 y) were produced in the ^{nat}Zr(d, xn) and ^{nat}Hf(d, xn) reaction, respectively, by bombarding metallic ^{nat}Zr and ^{nat}Hf foils with a 24-MeV deuteron beam using the RIKEN AVF cyclotron.

Each target material was dissolved in 0.2 mL of concentrated HF solution in a PTFE beaker. After evaporation to dryness, the residue was dissolved with 0.2 mL of 1 M HF and then dried up. The residue was dissolved with 0.3 mL of 1 M HF and fed onto a chromatographic column filled with the anion-exchange resin (BIO RAD AG 1 × 8 100–200 mesh, F^- form 5 mm *i.d.* × 45 mm). After washing the column with 2.5 mL of 1 M HF, Zr and Hf were eluted from the column with 5 mL of the mixed solution of 9 M HCl/0.004 M HF. Then, Nb was eluted with 5 M HNO_3/0.2 M HF, while Ta was eluted with 4 M HCl.[2] ^{233}Pa ($T_{1/2}$ = 27.0 d) was chemically separated from ^{237}Np ($T_{1/2} = 2.144\times10^6$ y). ^{237}Np was dissolved with 0.2 mL of 9 M HCl, and fed onto the chromatographic column filled with the 100–150 mesh of the TK400 resin. Np was eluted with 20 mL of 9 M HCl, then Pa was eluted subsequently with 1.8 mL of 1 M HCl. The solution was evaporated to dryness to fume out HCl. Then, the residue was dissolved with pure water and evaporated to dryness 3 times. Then, radio tracers were stocked in a polypropylene vessel in 1 M H_2SO_4. For the batch experiments with low H_2SO_4 concentrations, the radiotracer was diluted to 0.2 M H_2SO_4 with pure water.

The anion-exchange resin used was the strongly basic anion exchanger, MCI GEL CA08Y supplied by Mitsubishi Chemical Corporation. The resin was washed with 2.0 M NaOH and 2.0 M H_2SO_4 alternatively 3 times to replace Cl^- ion in CA08Y with HSO_4^-. Finally, the resin was washed with distilled water and dried up to a constant weight at 70°C in an oven.[1]

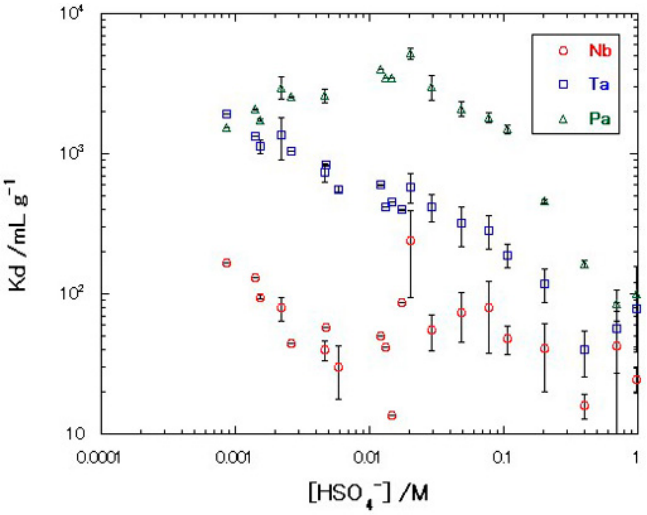

Fig. 1. Variation of the K_d values of ^{95}Nb, ^{179}Ta and ^{233}Pa on the anion-exchanger CA08Y as a function of HSO_4^- concentration [HSO_4^-].

We determined the distribution coefficient (K_d) values of Nb, Ta, and Pa on CA08Y in H_2SO_4 at various concentrations. 5–25 mg of the resin and 3 mL of H_2SO_4 solutions with 50 μL of the radiotracer were placed in a polystyrene centrifuge tube and shaken for 120 min at 22°C. After centrifugation, a 1 mL of the solution was pipetted and subjected to γ-ray spectrometry with a Ge detector The same treatment was conducted without the resin to determine the reference radioactivity. The radioactivity on the resin was determined by subtracting the solution radioactivity from the reference activity.

Figure 1 shows a variation of the K_d values of Nb, Ta, and Pa as a function of [HSO_4^-]. Especially in the case of Nb, the K_d values varies greatly. Therefore, it's necessary to check the reproducibility of the data.

It was reported that the ion-exchange reaction of the group-4 elements is slow.[1] However, we found that the reaction of the group-5 elements reach the equilibrium faster, within 20 s.

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