Reversed-phase chromatography for element 105, Db with Aliquat 336 resin from 2.7 M and 27 M HF solutions

D. Sato,[*1,*2] M. Murakami,[*1] S. Goto,[*1] K. Ooe,[*1] R. Motoyama,[*1] K. Shirai,[*1] R. Yamada,[*1] S. Tsuchiya,[*5] T. Moriyama,[*5] H. Haba,[*2] Y. Komori,[*2] S. Yano,[*2] A. Toyoshima,[*3] A. Mitsukai,[*3] K. Tsukada,[*3] H. Kikunaga,[*4] and H. Kudo[*5]

Elements with atomic number ≥ 104 are called superheavy elements. As aqueous chemical studies for element 105, Db, an anion-exchange experiment was performed in 13.9 M hydrofluoric acid solution.[1] However, the chemical species of Db in HF solution is not still clear. We have been studying a liquid-liquid and a solid-liquid extraction of Nb and Ta which are lighter homologues of Db with Aliquat 336 in solutions of various concentration of HF.[2,3] In the liquid-liquid extraction, univalent anionic complex of Nb and Ta were extracted, and it was implied that the extracted species were $[NbOF_4]^-$ or $[NbF_6]$ and $[TaF_6]^-$.[2] On the other hand, the results of solid-liquid extraction with 52 wt% Aliquat 336 resin lead indicated that the distribution coefficients, K_d, of ^{95g}Nb has the minimum value at 10 M HF, while those of ^{179}Ta gradually decrease with increasing HF concentration.[3] Because the adsorption behaviors of Nb and Ta differed clearly, similarity with the homologues of the character of Db could be clear by measuring the dependence of K_d values of Db on HF concentration. Because the results of Nb and Ta of on-line experiment were good agreement with those of batch experiment in 2.7 M and 27 M HF, it is suggested that the chemical formation of Nb and Ta of on-line experiment is same with that of batch experiment.[3] Therefore, 2.7 M and 27 M HF were decided as the condition of Db experiment. In this study, we performed experiments of a reversed-phase chromatography experiment of Db with 2.7 M and 27 M HF.

The isotope ^{262}Db was produced in the ^{248}Cm(^{19}F, $5n$)^{262}Db reaction at the RIKEN K70 AVF cyclotron. The small amount of ^{nat}Gd was induced in the Cm target in order to monitor the state of the experiment by measuring ^{170}Ta which was produced in the ^{nat}Gd(^{19}F, xn)^{170}Ta reaction. The reaction products transported by the He/KCl gas-jet system were deposited on the collection part in Automated Rapid Chemical Apparatus (ARCA) for 60 s. Then, the products were dissolved in 2.7 M or 27 M HF solution and loaded on a small chromatographic column (1.6 mm $i.d.$ × 7.0 mm height) filled with the 52 wt% Aliquat 336 resin at a flow rate of 1.0 mL/min. The effluent was collected into a Ta disk as a primary fraction. The adsorbed species on the column were stripped with 6 M HNO_3/0.015 M HF solution at a flow rate of 1.0 mL/min. The effluents was collected another Ta disk as a secondary fraction. The both quantity of the primary and the secondary fraction was 150 μL. These Ta disks were heated to dry the solution by halogen lamp, then they were picked up into the automated rapid α/SF detection system, RIDER, by robot arm to measure their radioactivities. The Db experiments were performed for 596 times in 2.7 M HF and for 950 times in 27 M HF. The average chemical yields of ^{170}Ta were about 22% in 2.7 M HF and 18% in 27 M HF, respectively.

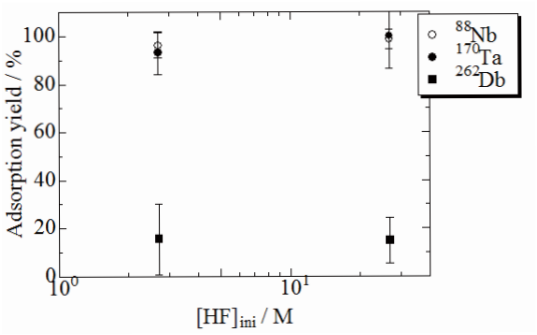

Fig. 1. The dependences of the adsorption yields of Nb, Ta, Db on $[HF]_{ini}$ with 52 wt% Aliquat 336 resin.

From the obtained α-spectra, the number of events corresponding to ^{262}Db (E_α = 8420–8740 keV)[4] in 2.7 M HF were 10 in the 1st fraction and 2 in the 2nd fraction. In 27 M HF, 24 events in 1st fraction and 3 events in 2nd fraction were observed. Considering the contamination, the distribution of decay time and the background, the adsorption yields on the 52 wt% Aliquat 336 resin which were estimated by radioactivity ratio were 15.4% in 2.7 M HF and 14.8% in 27 M HF, respectively. In Fig. 1, it was found that the adsorption yields of Nb and Ta are almost 100% in the both cases of 2.7 M and 27 M HF, while those of Db are very smaller than those of homologues. In addition, the adsorption yields of Db are almost constant to $[HF]_{ini}$. Therefore, if the K_d values of Db are estimated from the adsorption yield, the dependence of K_d values of Db on $[HF]_{ini}$ is also constant. Because the K_d values of Ta are decreased with increasing $[HF]_{ini}$,[3] the extraction behavior of Db would not be like Ta at least. Therefore, it was suggested that the fluoride complex formation of Db is not similar to that of Ta.

*1 Graduate School of Science and Technology, Niigata University
*2 RIKEN Nishina Center
*3 Japan Atomic Energy Agency
*4 Research Center for Electron Photon Science, Tohoku University
*5 Department of Chemistry, Faculty of Science, Niigata University

References

1) K. Tsukada *et al.*, Radiochim. Acta. **97**, 83 (2009).
2) D. Sato *et al.*, RIKEN Accel. Prog. Rep. **47**, 247 (2014).
3) D. Sato *et al.*, JNRS, sorc 61, 3B09 (2017).
4) M. Murakami, Dr. thesis, Niigata Univ. (2015).