Extraction behavior of ^{93m}Mo and ¹⁷⁹W from HF / HCl into Aliquat336 as model experiments for seaborgium (Sg)

A. Mitsukai, *1,*2 A. Toyoshima, *2 Y. Komori, *3 S. Yano, *3 H. Haba, *3 M. Asai, *2 K. Tsukada, *2 T.K. Sato, *2

and Y. Nagame^{*1,*2}

Chemical studies of superheavy elements with atomic numbers $Z \ge 104$ provide not only crucial and challenging opportunities to advance our understanding of the properties of matter at the limits of existence but also elucidate the influence of relativistic effects on atomic electrons and the architecture of the farthest of the periodic table of elements.^{1,2)} These elements are all artificial and synthesized at accelerators using nuclear reactions of heavy-ion beams with heavy element target materials. As both the half lives and cross sections of these nuclides are rapidly decreasing with increasing Z, they are usually available in quantities of only a few atoms or often one atom at a time.

Recently, we started studying the aqueous chemistry of element 106, seaborgium (Sg), to clarify its complex formation and redox reactions in aqueous phases. In this work, the solvent extraction behavior of molybdenum (Mo) and tungsten (W), which are lighter homologs of Sg, from a HF/HCl mixed solution into toluene with an amine extractant Aliquat336 is studied by a batch wise method to elucidate the complex formation of these homologs.

At the RIKEN AVF cyclotron, 93mMo and 179W were produced in the ${}^{93}Nb(d, 2n){}^{93m}Mo$ and ${}^{nat}Ta(d, xn){}^{179}W$ reactions, respectively. The reaction products were transported to the laboratory by a gas-jet method and deposited on a small Naflon® sheet. The deposited products were dissolved with 700 µL of HF/HCl and the solution was collected in a plastic tube. Then the same volume of Aliquat336/toluene solution was added to this HF/HCl solution. The mixture was shaken for 60 s. (In a separate experiment, we confirmed that the chemical equilibrium of this reaction was accomplished within 60 s in the present system.)³⁾ After centrifugation, the aqueous and organic solutions were separately taken in plastic tubes for γ -ray spectrometry with a Ge detector. The distribution ratio (D)is defined as the ratio of radioactivity per unit volume between two phase as $D = (A_{\text{org}} / V_{\text{org}}) / (A_{\text{aq}} / V_{\text{aq}})$, where $A_{\rm org}$ and $A_{\rm aq}$ denote radioactivities in the organic and aqueous phases, respectively, and V_{org} and V_{aq} represent the volumes of the organic and aqueous phases, respectively.

In the batch experiments, the concentration of HF was varied from 10^{-4} to 5 M, while those of HCl and Aliquat336 were fixed to be 1 M and 0.2 M, respectively. In Fig. 1, variations of the *D* values of Mo and W are shown as a function of HF concentration, [HF]. The *D* values of both elements start to increase steeply at approximately

 $[HF] = 10^{-2}$ M, indicating that anionic fluoride complexes are formed beyond that concentration. Above $[HF] = 10^{-1}$ M, on the_other hand, the *D* values of Mo and W decrease with increasing [HF]. This is due to the increase of the HF₂⁻ concentration; the extraction of HF₂⁻ is competed with that of anionic fluoride complexes of Mo and W. Under the present conditions, we clearly observed the formation of anionic fluoride complexes of Mo and W. In the next step, the extraction behavior of Mo and W will be investigated while varying the Aliquat336 concentration in the HF/HCl solution to obtain information on chemical forms of the anionic fluoride complexes of Mo and W



Fig. 1. Variation of the D values of Mo and W as a function of [HF].

References

- 1) A. Türler, V. Pershina, Chem. Rev. 113, 1237 (2013).
- M. Schädel, D. Schaughnessy (eds.): The Chemistry of Superheavy Elements, 2nd edition (Springer, Berlin, 2014).
- 3) A. Mitsukai, Master's Thesis (2017) (unpublished).

^{*1} Graduate School of Science and Technology, Ibaraki University

^{*2} Advanced Science Research Center, Japan Atomic Energy Agency

^{*3} RIKEN Nishina Center