

Development of a rapid solvent extraction apparatus for the aqueous chemistry of the heaviest elements

Y. Komori,^{*1} H. Haba,^{*1} K. Ooe,^{*2} A. Toyoshima,^{*3} A. Mitsukai,^{*3} M. Murakami,^{*1,*2} D. Sato,^{*2} R. Motoyama,^{*2} S. Yano,^{*1}
K. Watanabe,^{*1} A. Sakaguchi,^{*4} J. Inagaki,^{*4} H. Kikunaga,^{*5} and J. P. Omtvedt^{*6}

To realize the aqueous chemistry studies of Sg ($Z = 106$) and heavier elements, we are developing a continuous and rapid solvent extraction apparatus coupled with the GARIS gas-jet system.¹⁾ This apparatus consists of a continuous dissolution apparatus called as Membrane DeGasser (MDG), Flow Solvent Extractor (FSE), and liquid scintillation detectors. In this work, we developed the MDG and FSE, and conducted online solvent extraction of Tc and Re as homologs of element 107, Bh.

We modified the MDG developed by a research group of Univ. Oslo and JAEA.²⁾ Major modifications were to reduce a dead volume of the apparatus from 90 μL to 23 μL and to simplify the structure of the gas/solution-mixing unit to dissolve shorter-lived nuclides with higher efficiencies at a lower flow rate of 1 mL/min. Further, we fabricated the FSE that consisted of a Teflon capillary and a phase separator by referring to a well-known flow injection analysis technique.³⁾ The inner diameter of the capillary is 0.5 mm. We can easily change the extraction time by varying the capillary length. In the phase separator, organic and aqueous phases are separated from each other with a PTFE membrane filter (ADVANTEC T300A013A) by applying pressure on the outlet of the capillary of the aqueous phase.

We investigated the performance of the MDG using the AVF cyclotron at RIKEN. Short-lived $^{90\text{m,g}}\text{Nb}$ ($T_{1/2} = 18.8$ s, 14.6 h) and $^{178\text{a}}\text{Ta}$ ($T_{1/2} = 2.36$ h) were simultaneously produced in the $^{\text{nat}}\text{Zr}(d,xn)$ and $^{\text{nat}}\text{Hf}(d,xn)$ reactions, respectively. The reaction products were transported by the He/KCl gas-jet system to the MDG in the chemistry laboratory. The reaction products were dissolved with 1 M HF at a flow rate of 1 mL/min. The solution from the MDG was collected for 1 min and was then subjected to γ -ray spectrometry with a Ge detector. The dissolution efficiencies were determined by comparing the radioactivity in the solution with that of the direct catch on a glass fiber filter (ADVANTEC GB-100R). Next, we coupled the MDG and the FSE and performed online solvent extraction of Tc and Re. $^{92,94\text{m}}\text{Tc}$ ($T_{1/2} = 4.25, 293$ min) and ^{181}Re ($T_{1/2} = 20$ h) produced simultaneously in the $^{\text{nat}}\text{Mo}(d,xn)$ and $^{\text{nat}}\text{W}(d,xn)$ reactions, respectively, were transported by the gas-jet to the MDG. They were continuously dissolved with 0.5 M and 1 M HNO_3 , and they were extracted with the FSE into an organic phase of tri-*n*-octylamine (TOA) in toluene. Both aqueous and organic phases from the FSE

were then subjected to the γ -ray spectrometry to determine the distribution ratios (D). We measured the D values of Tc and Re by varying the capillary length from 5 cm to 100 cm to change the extraction time. We also varied a concentration of TOA from 0.005 M to 0.1 M to evaluate an applicable D range with the FSE. These D values were compared with those in the batch extraction that were obtained by 3-min shaking.

The dissolution efficiencies of $^{90\text{m}}\text{Nb}$, $^{90\text{g}}\text{Nb}$, and $^{178\text{a}}\text{Ta}$ with the MDG were 56 ± 2 , 88 ± 6 , and $82 \pm 7\%$, respectively. The efficiency of the short-lived $^{90\text{m}}\text{Nb}$ is smaller than those of the long-lived $^{90\text{g}}\text{Nb}$ and $^{178\text{a}}\text{Ta}$. In the online solvent extraction with the MDG-FSE, a variation of the D values of Tc and Re is shown in Fig. 1 as a function of the capillary length. The extraction equilibrium of Tc and Re was attained with the 40-cm capillary. The time required for the solutions to pass through the 40-cm capillary is calculated as 2.4 s, which is sufficiently short to perform the solvent extraction with $^{265}\text{Sg}^{a,b}$ ($T_{1/2} = 8.5$ s, 14.4 s)⁴⁾ and ^{266}Bh ($T_{1/2} = 10.7$ s).⁵⁾ We also found that the D values of Tc and Re increase with increasing the TOA concentration. The D values with the MDG-FSE were consistent with those in the batch extraction except for Tc at a high TOA concentration of > 0.05 M, where the D values obtained for Tc with the MDG-FSE were slightly smaller than those in the batch extraction. The reason of this discrepancy is under consideration.

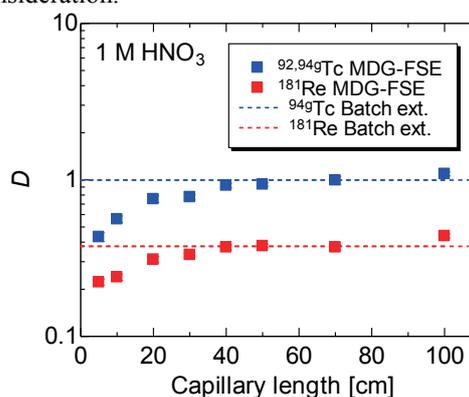


Fig. 1. Variation of the D values of Tc and Re as a function of the capillary length. The concentrations of HNO_3 and TOA are 1 M and 0.01 M, respectively.

References

- 1) H. Haba et al.: Chem. Lett. **38**, 426 (2009).
- 2) K. Ooe et al.: J. Radioanal. Nucl. Chem. **303**, 1317 (2015).
- 3) S. Motomizu: J. Flow Injection Anal. **5**, 71 (1988).
- 4) H. Haba et al.: Phys. Rev. C **85**, 024611 (2012).
- 5) H. Haba et al.: Pacificchem 2015, Honolulu, Hawaii, U.S.A., Dec. INOR 1578, 2015.

*1 RIKEN Nishina Center

*2 Graduate school of Sci. and Technol., Niigata Univ.

*3 JAEA

*4 Univ. Tsukuba

*5 ELPH, Tohoku Univ.

*6 Univ. Oslo