

Selective Separation of Technetium-99 from Aqueous Matrix

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INTRODUCTION

Technetium-99 (⁹⁹Tc) is one of the byproducts of the ²³⁵U and ²³⁹Pu fission in nuclear reactors and is a major component in nuclear waste (Uchida and Tagami, 1997). The ⁹⁹Tc has a half-life of 2.11×10^5 y. Hence, the monitoring of ⁹⁹Tc activity in the environmental samples is essential. Techniques used to measure ⁹⁹Tc in environmental samples include both radiometric (e.g., liquid scintillation counter, gas flow GM counter) and non-radiometric (e.g., inductively coupled plasma mass spectrometer, ICP-MS, accelerator mass spectrometry) approaches. ICP-MS has some advantages among the technique due to its high sample throughput, better selectivity, and commercial availability (Guerin et al., 2016). Rhenium (Re) is often used as the non-isotopic tracer during ⁹⁹Tc measurement by ICP-MS. In the current work, two solid-phase extraction (SPE) materials (AnaLig Tc-01 and TRU) were evaluated for the selective separation of ⁹⁹Tc (using Re as a chemical analog) from the aqueous matrices.

MATERIALS AND METHODS

NexION 300S ICP-MS (PerkinElmer, Waltham, MA) was used for elemental analysis. SPE treatment was performed with a GL-SPE vacuum manifold (GL Sciences, Tokyo, Japan). Milli-Q water purification system (Merck KGaA, Darmstadt, Germany) was used to produce ultrapure water (UPW).

Standard solutions (1000 mg L⁻¹) of Re (SPEX CertiPrep, Metuchen, NJ), molybdenum (Mo) (Nacalai Tesque, Kyoto, Japan), and ruthenium (Sigma-Aldrich, St. Louis, MO) was used to prepare working solutions. Solution pHs were adjusted using either HNO₃ or NaOH (1 mol L⁻¹). SPE materials, namely AnaLig Tc-01 (TC-01) and TRU, packed in 3 mL columns, as received from GL Sciences (Tokyo, Japan), were used. The SPE systems are evaluated for separating ions from the aqueous matrix at different steps: cleaning, conditioning, sample loading, washing, and elution.

RESULTS AND DISCUSSION

The retention behavior of the SPEs at varying sample solution pHs (1~9) has been studied (Figure 1a). Tc-01 resin column showed $\geq 99\%$ Re retention at the entire studied pHs, whereas TRU resin showed $\geq 99\%$ at pHs < 2 only. A flow rate of ~ 0.5 mL min⁻¹ was maintained during sample loading. Ultrapure water and HNO₃ solutions of different concentrations (1 to 4 mol L⁻¹) were used to check Re recovery from SPEs (Figure 1b). A quantitative Re recovery from Tc01 was observed with 3 mol L⁻¹ HNO₃, while 2 mol L⁻¹ HNO₃ did the same for TRU. HCl and H₂SO₄ solutions (3 mol L⁻¹) were also used to check the distinguishing impact on Re elution from Tc01 due to acid variants, and it was observed as negligible.

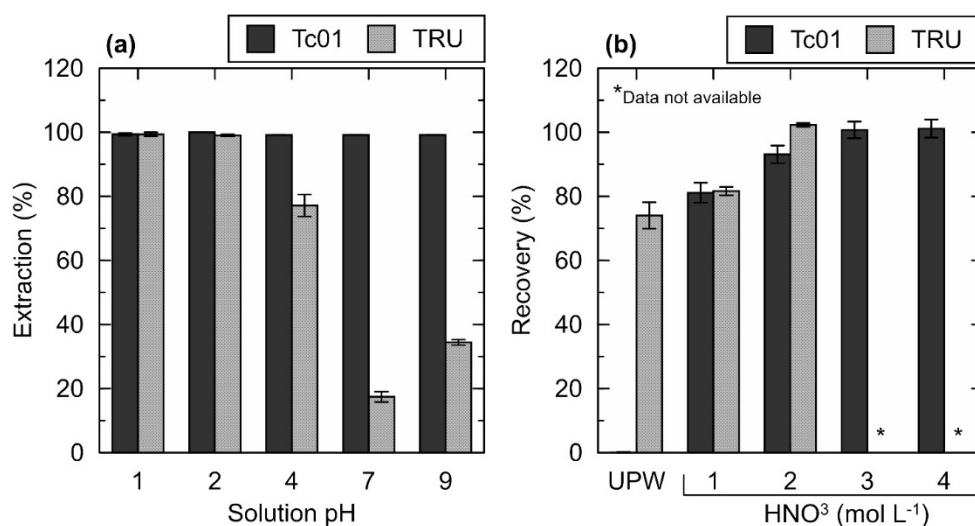


Figure 1: (a) Re-extraction at different pH conditions, (b) Re-recovery with different eluents.

Mo and Ru create molecular and isobaric interferences during ⁹⁹Tc measurement by ICP-MS. The separation factor of Re and interfering elements (Mo and Ru) are shown in Table 2. Tc01 did not retain Ru at all, while the separation selectivity of Tc01 towards Mo was also not quantitative. On the contrary, TRU resin effectively minimized Mo and Ru's impact during the separation of Re from the aqueous matrix.

Table 2 Separation factor (SF) of Re vs. matrix ion Mo and Ru

SPEs →	Tc01					TRU	
pH →	1	2	4	7	9	1	2
SF _{Re/Mo}	31.78	0	0.83	0.95	1.15	9461	*
SF _{Re/Ru}	*	*	*	*	*	*	5437

* Not retained in the column, i.e., separation factor is high.

CONCLUSION

Two different SPEs (Tc01 and TRU) were evaluated for Re separation from the aqueous matrix, and Re has been used as a chemical analog to ⁹⁹Tc. Both SPEs showed the capability of selective Re separation of Re in the presence of interfering elements (Mo and Ru), even though the performance was varied with the sample solution pH, eluent-types, or eluent concentration.

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