

Rapid Determination of Actinides in Emergency Water Samples

Summary of Method Uranium, Plutonium and Americium-Curium are separated and concentrated from up to 400mL water samples using calcium phosphate precipitation. The precipitate is dissolved in nitric acid and aluminum nitrate. Actinides are separated from matrix impurities and potentially interfering radionuclides in the sample using 2mL cartridges of Eichrom TEVA and TRU Resins. Actinides are measured by alpha spectrometry following source preparation by cerium fluoride microprecipitation onto Eichrom Resolve® Filters. Chemical yields are determined by recovery of ^{232}U , ^{243}Am , and ^{242}Pu (or ^{236}Pu , if measuring ^{237}Np) tracers. Typical chemical recoveries are >90%. A single operator can complete the separation method for batches of 12-24 samples in as little as 4-5 hours.

Reagents

TEVA Resin, 2mL Cartridges (Eichrom TE-R50-S)
 TRU Resin, 2mL Cartridges (Eichrom TR-R50-S)
 Ammonium Hydroxide (listed as 28% NH_3 or 56% NH_4OH)
 Nitric Acid (70%)
 Hydrochloric Acid (37%)
 Hydrofluoric Acid (49%) or Sodium Fluoride
 Deionized Water
 Iron Carrier (50mg/mL)
 Cerium Carrier (1mg/mL)
 1.25M $\text{Ca}(\text{NO}_3)_2$
 3.2M $(\text{NH}_4)_2\text{HPO}_4$
 2M $\text{Al}(\text{NO}_3)_3$
 10% (w:w) TiCl_3
 ^{242}Pu (or ^{236}Pu if meas. Np), ^{243}Am and ^{232}U tracers
 Oxalic acid/Ammonium oxalate
 Sulfamic Acid
 Ascorbic Acid
 Sodium Nitrite
 Denatured Ethanol

Equipment

Vacuum Box (Eichrom AR-24-BOX or AR-12-BOX)
 Cartridge Reservoir, 20mL (Eichrom AR-200-RV20)
 Inner Support Tubes-PE (Eichrom AR-1000-TUBE-PE)
 Yellow Outer Tips (Eichrom AR-1000-OT)
 Resolve Filters in Funnel (Eichrom RF-DF25-25PP01)
 50mL and 250mL Centrifuge Tubes
 Centrifuge
 Analytical Balance
 Alpha Spectrometry System
 Vacuum Pump

Figure 1. Sample Preparation

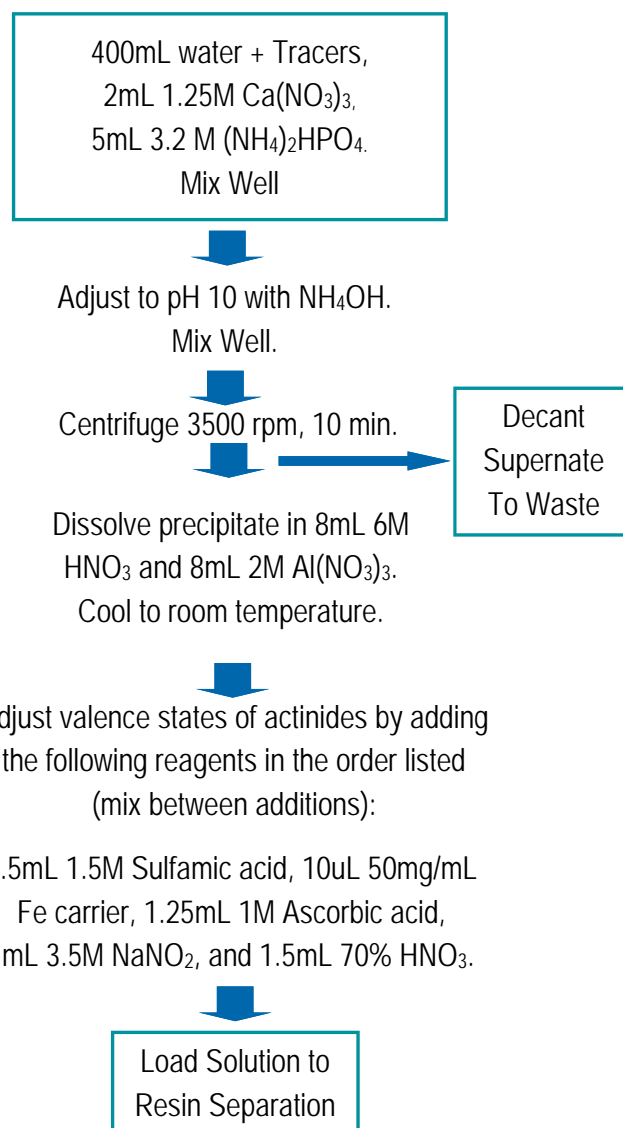

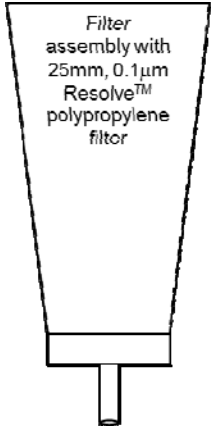

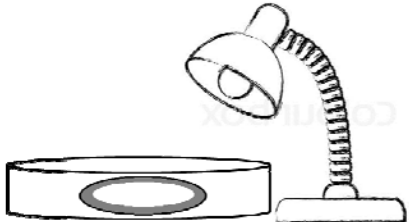


Figure 2. Actinide Separation on TEVA - TRU*

<p>(1) Precondition stacked 2mL TEVA-TRU with 10mL 3M HNO₃.</p> <p>(2) Load sample solution.</p> <p>(3) Rinse sample tube with 5mL 6M HNO₃. Add tube rinse to cartridges.**</p> <p>(4) Rinse cartridges with 5mL 3M HNO₃.</p> <p>(5) Separate TEVA and TRU cartridges.</p>		<p>(11) Add 0.5mL 30% H₂O₂ to Pu and 0.2mL 30% H₂O₂ to Am/Cm samples for additional U decon. during CeF₃ ppt.</p> <p>(12) Add 0.5mL of 10% TiCl₃ to each U sample for CeF₃ ppt.</p> <p>(13) Add 50-100ug Ce carrier to all samples. Mix well. Add 1mL 49% HF. Mix well. Wait 15-20 minutes.</p>	<p>(19) Draw vacuum until filter is dry.</p> <p>(20) Remove filter from funnel assembly and mount filter on stainless steel planchet with 2-sided tape.</p>
<p>(6) Rinse TEVA cartridge with: -15mL 3M HNO₃ -20mL 9M HCl (remove Th) -5mL 3M HNO₃</p> <p>(7) Strip Pu(Np) from TEVA with 20mL 0.1M HCl-0.05MHF-0.01M TiCl₃.</p>	<p>(14) Set up Resolve® Filter Funnel on vacuum box.</p> <p>(15) Wet filter with 3mL 80% ethanol followed by 3mL DI water.</p>		
<p>(8) Strip Am/Cm from TRU with 15mL 4M HCl. Dilute to 30mL prior to CeF₃ ppt.</p> <p>(9) Rinse TRU with 15mL 4M HCl-0.2M HF. (Th removal)</p> <p>(10) Strip U from TRU with 15mL 0.1M ammonium bioxalate.</p>	<p>(16) Filter sample.</p> <p>(17) Rinse sample tube with 5mL DI water and add to filter.</p> <p>(18) Rinse filter funnel with 3mL DI water and 2mL 100% ethanol.</p>	<p>(21) Dry filter under heat lamp for 3-5 minutes.</p> <p>(22) Measure actinides by alpha spectrometry.</p> 	

*Strontium may also be measured by adding a 2mL Sr Resin Cartridge below DGA and following the separation scheme in Eichrom application note AN-1411, "Rapid Determination of Sr in Emergency Water Samples."

**Adding 50uL of 30% H₂O₂ to the 6M HNO₃ tube rinse can help improve U recoveries and decontamination in the Pu/Np fraction.

References

- 1) Sherrod L. Maxwell, Brian K. Culligan, "Rapid separation method for emergency water and urine samples," *J. Radioanal. Nucl. Chem.*, 279(3), 901-907 (2009).