

Rapid Determination of Actinides in Soil Samples

Summary of Method Actinides are separated and measured from 1-2g samples of soil. Soil samples are fused in zirconium crucibles with sodium hydroxide. Sequential precipitations remove matrix prior to separation of actinides on 2mL cartridges of Eichrom TRU and DGA resins. Actinides are measured by alpha spectrometry following cerium fluoride microprecipitation onto Eichrom Resolve® Filters. Chemical recoveries averaged $97\pm 9\%$, $96\pm 7\%$, and $91\pm 4\%$, respectively, for ^{242}Pu , ^{243}Am and ^{232}U tracers. Measured values typically agreed to within 3% of reference values. Batches of 12 samples can be prepared for measurement in as little as 4 hours.

Reagents

TRU Resin, 2mL Cartridges (Eichrom TR-R50-S)
 DGA Resin, 2mL Cartridges (Eichrom DN-R50-S)
 Iron carrier (50mg/mL Fe, as ferric iron nitrate)
 ^{242}Pu (or ^{236}Pu if meas. Np), ^{243}Am and ^{232}U tracers
 Oxalic acid/Ammonium oxalate
 La carrier (10mg/mL) Ce carrier (1mg/mL)
 Deionized Water 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 HNO_3 (70%)
 HCl (37%) NaOH
 HF (49%) or NaF Boric acid
 H_2O_2 (30%) NaNO_2
 Denatured ethanol Sulfamic Acid
 Ascorbic Acid

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
 Muffle Furnace
 Analytical Balance
 250mL Zirconium crucibles with zirconium lids
 Stainless Steel Planchets with adhesive tape
 Alpha Spectrometry System
 Vacuum Pump
 Heat Lamp

Figure 1. Sample Preparation

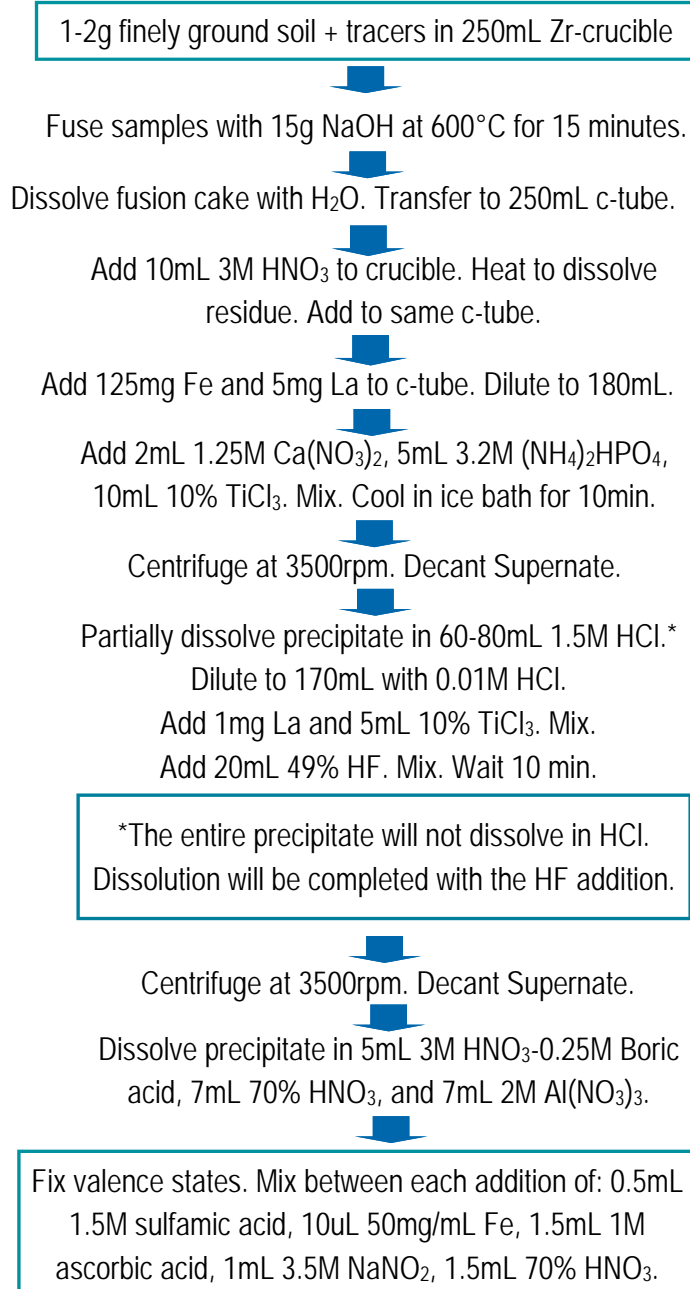

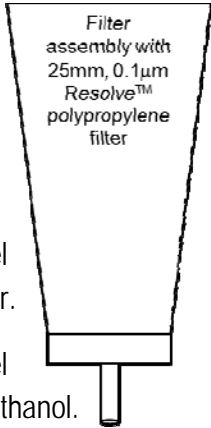
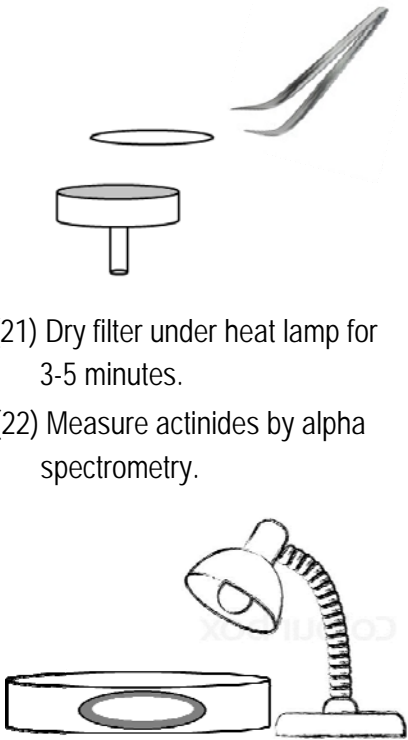


Figure 2. Actinide Separation on TRU/DGA and Source Preparation

<p>(1) Precondition TRU/DGA resin with 5mL 8M HNO₃.</p> <p>(2) Load samples.</p> <p>(3) Rinse sample tube with 5mL 8M HNO₃, and add tube rinse to TRU/DGA.*</p> <p>(4) Rinse TRU/DGA with: -10mL 10M HNO₃ -15mL 4M HCl</p> <p>(5) Separate TRU and DGA.</p> <p>(6) Strip Pu from TRU w/ 15mL 3M HCl-0.02M TiCl₃. Add 0.5mL 30% H₂O₂.</p> <p>(7) Rinse TRU with: -5mL 8M HNO₃ + 50uL 30% H₂O₂ -10mL 4M HCl-0.2M HF -10mL 4M HCl-0.2M HF-2mM TiCl₃ -3mL 8M HNO₃</p> <p>(8) Strip U from TRU with 15mL 0.1M ammonium bioxalate. Add 0.5mL TiCl₃ for CeF₃ ppt.</p> <p>(9) Rinse DGA with: -12mL 3M HCl -20mL 0.05M HNO₃ -12mL 3M HNO₃-0.25M HF</p>		<p>(10) Rinse DGA with 5mL 3M HCl.</p> <p>(11) Strip Am/Cm from DGA with 12mL 0.25M HCl. Add 0.2mL 30% H₂O₂.</p> <p>(12) Add 50-ug Ce carrier to each sample. Mix well. Add 1mL 49% HF. Mix well. Wait 15-20 minutes.</p> <p>(13) Set up Resolve® Filter Funnel on vacuum box.</p> <p>(14) Wet filter with 3mL 80% ethanol followed by 3mL DI water.</p> <p>(15) Filter sample.</p> <p>(16) Rinse sample tube with 5mL DI water and add to filter.</p> <p>(17) Rinse filter funnel with 3mL DI water.</p> <p>(18) Rinse filter funnel with 2mL 100% ethanol.</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 adhesive tape.</p> <p>(21) Dry filter under heat lamp for 3-5 minutes.</p> <p>(22) Measure actinides by alpha spectrometry.</p>	
---	---	--	---	---	--

*Adding 50uL of 30% H₂O₂ to tube rinse can improve U recoveries and decontamination in Pu(Np) fractions.

Method Performance

Analyte	Replicates	Tracer	% Tracer Recovery	Analyte Reference (mBq/g)	Analyte Measured (mBq/g)	% Bias
²³⁹ Pu	7	²⁴² Pu	97 ± 9	98.0	95 ± 3	-3.1
²⁴¹ Am	7	²⁴³ Am	96 ± 7	61.1	59 ± 4	-3.4
²³⁸ U	7	²³² U	91 ± 4	184	183 ± 6	-0.5

16 hour counts

References

1) Sherrod L. Maxwell, Brian K. Culligan, Jay B. Hutchinson, "Rapid determination of actinides and in asphalt samples," *J. Radioanal. Nucl. Chem.*, 299(3), 1891-1901 (2014).