

Rapid Determination of Actinides in Fecal Samples

Summary of Method Actinides are separated and measured from fecal samples. Fecal samples are muffled and wet ashed prior to fusion with sodium hydroxide. Sequential precipitation steps remove sample matrix prior to actinide separation on 2mL cartridges of Eichrom TEVA, TRU and DGA resins. Actinides are measured by alpha spectrometry following cerium fluoride microprecipitation onto Eichrom Resolve® Filters. Samples can be prepared for measurement in less than 24 hours.

Reagents

TEVA Resin, 2mL Cartridges (Eichrom TE-R50-S)
 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)
²⁴²Pu (or ²³⁶Pu if meas. Np), ²⁴³Am and ²³²U tracers
 Oxalic acid/Ammonium oxalate
 La carrier (10mg/mL) Ce carrier (1mg/mL)
 Deionized Water 1.25M Ca(NO₃)₂
 3.2M (NH₄)₂HPO₄ 2M Al(NO₃)₃
 10% (w:w) TiCl₃ HNO₃ (70%)
 HCl (37%) NaOH
 HF (49%) or NaF Boric acid
 H₂O₂ (30%) NaNO₂
 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
 250mL Ceramic crucibles Hot Plate
 250mL Zirconium crucibles with zirconium lids
 Stainless Steel Planchets with adhesive tape
 Alpha Spectrometry System Vellum paper
 Centrifuge Muffle Furnace
 Analytical Balance 1L Glass Beakers
 Vacuum Pump Heat Lamp

Figure 1. Sample Preparation

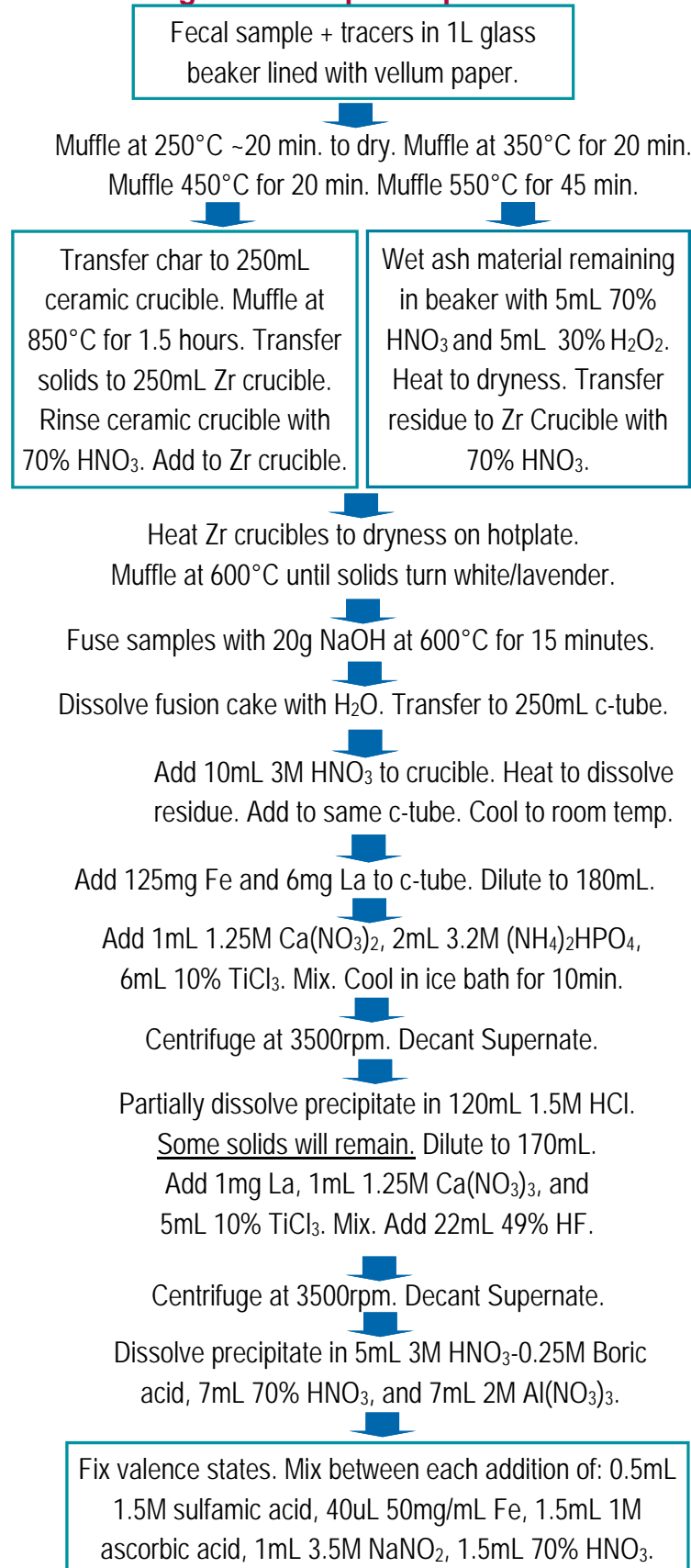

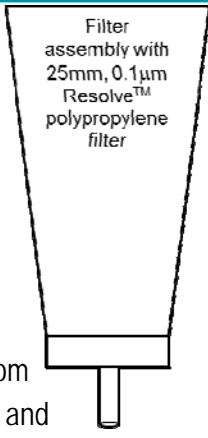
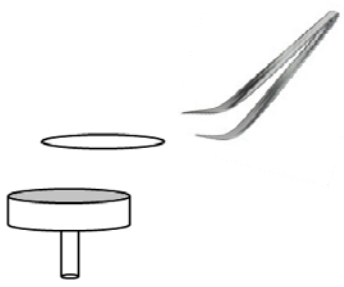


Figure 2. Actinide Separation on TEVA - TRU - DGA* and Source Preparation

<p>(1) Precondition stacked 2mL TEVA, TRU, DGA cartridges with 10mL 3M HNO₃. (2) Load sample solution. (3) Rinse sample tube with 5mL 3M HNO₃. Add tube rinse to cartridges. (4) Rinse cartridges with 10mL 3M HNO₃. (5) Separate TEVA, TRU, and DGA cartridges.</p>		<p>(12) Separate TRU cartridge from DGA cartridge. Set TRU aside for U recovery.</p>	<p>(23) Rinse filter funnel with 3mL DI water and 2mL 100% ethanol. (24) Draw vacuum until filter is dry. (25) Remove filter from funnel assembly and mount filter on stainless steel planchet with 2-sided tape.</p> 
<p>(6) Rinse TEVA cartridge with: -10mL 3M HNO₃ -20mL 9M HCl (Remove Th) -5mL 3M HNO₃ (7) Strip Pu (and Np) from TEVA cartridge with 20mL 0.1M HCl-0.05MHF-0.01M TiCl₃. (8) Add 0.5mL 30% H₂O₂ for Uranium decon. in alpha source preparation.</p>	<p>(13) Rinse DGA cartridge with: -5mL 3M HCl -3mL 1M HNO₃ -15mL 0.05M HNO₃</p>	<p>(14) Strip Am and Cm from DGA with 10mL 0.25M HCl. Add 0.2mL 30% H₂O₂.</p>	<p>(25) Dry filter under heat lamp for 3-5 minutes. (26) Measure actinides by alpha spectrometry.</p> 
<p>(9) Rinse DGA cartridge with 10mL 0.1M HNO₃. (U removal). (10) Place TRU cartridge above DGA. (11) Strip Am/Cm from TRU onto DGA with 15mL 3M HCl at 1-2mL/min.</p>	<p>(15) Rinse TRU cartridge with: -15mL 4M HCl-0.2M HF-2mM TiCl₃ -5mL 8M HNO₃.</p>	<p>(16) Strip U from TRU with 15mL of 0.1M ammonium bioxalate. (17) Add 0.5mL 10% TiCl₃ to U samples.</p>	
	<p>(18) Add 50-100ug Ce carrier to all samples. Mix well. Add 1mL 49% HF. Mix well. Wait 15-20 minutes.</p>	<p>(19) Set up Resolve® Filter Funnel on vacuum box.</p>	
	<p>(20) Wet filter with 3mL 80% ethanol followed by 3mL DI water.</p>	<p>(21) Filter sample. (22) Rinse sample tube with 5mL DI water and add to filter.</p>	

*Adding 50uL 30% H₂O₂ can improve Uranium recoveries and decontamination in Pu(Np) fractions.

Method Performance

Analyte	Samples	Tracer	% Tracer Recovery	Reference (Bq/sample)	Measurement (Bq/sample)	% Bias
^{239/240} Pu	5	²⁴² Pu	95 ± 9	0.085 - 0.204	0.081 - 0.198	-11 to -1.5
²³⁸ Pu	5	²⁴² Pu	95 ± 9	0.066 - 0.156	0.071 - 0.146	-5.3 to 3.0
²⁴¹ Am	5	²⁴³ Am	83 ± 4	0.199 - 0.476	0.201 - 0.464	-11 to 1.0
²³⁸ U	5	²³² U	63 ± 7	0.226 - 0.541	0.196 - 0.592	-9.0 to 2.9
²³⁴ U	5	²³² U	63 ± 7	0.218 - 0.521	0.206 - 0.536	-13 to 9.4

6 hour count time

References

1) Sherrod L. Maxwell, Brian K. Culligan, Jay B. Hutchinson, Ronie B. Spencer "Rapid fusion method for determination of actinides in fecal samples," *J. Radioanal. Nucl. Chem.*, 298(3), 1533-1542 (2013).