

Rapid Determination of Actinides in 1g Concrete and Brick Samples

Summary of Method U, Pu, Np, Am and Cm are separated and concentrated from 1 gram samples of concrete and brick. Samples are fused with NaOH at 600°C in zirconium crucibles. The fusion cakes are dissolved in water, transferred to 250mL centrifuge tubes and precipitated sequentially with iron-hydrous titanium oxide and lanthanum fluoride to facilitate matrix removal. Actinides are separated on stacked 2mL cartridges of Eichrom TEVA, TRU and DGA resins. Actinides are measured by alpha spectrometry following CeF₃ microprecipitation onto Eichrom Resolve® Filters. Chemical yields of tracers ranged from 79-98% for ²³⁶Pu, 77-90% for ²⁴³Am, and 72-81% for ²³²U. Measured values typically agreed to within 10% of reference values. Sample preparation for batches of 12 samples can be completed by a single operator in <8 hours. Alpha spectrometry count times will depend on detection limit and data quality objectives.

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

Figure 1. Sample Preparation

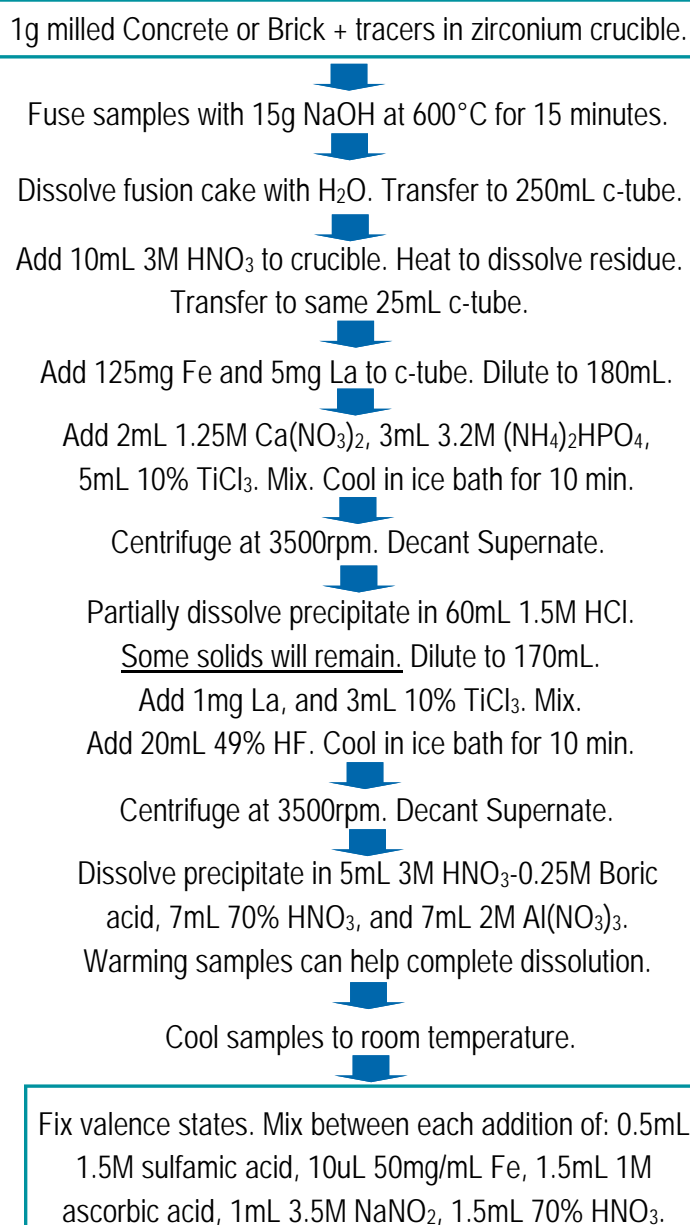



Figure 2. Actinide Separation on TEVA - TRU - DGA* and Source Preparation

<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. (13) Rinse DGA cartridge sequentially with: -5mL 4M HCl -5mL 1M HNO₃ -15mL 0.05M HNO₃ (14) Strip Am and Cm from DGA with 10mL 0.25M HCl. Add 0.2mL 30% H₂O₂.</p>	<p>(23) Rinse sample tube with 5mL DI water and add to filter. (24) Rinse filter funnel with 3mL DI water and 2mL 100% ethanol. (25) Draw vacuum until filter is dry.</p>
<p>(6) Rinse TEVA cartridge with: -10mL 3M HNO₃ -20mL 9M HCl -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>(15) Rinse TRU cartridge with 15mL 4M HCl-0.2M HF-0.002M TiCl₃. (16) Rinse TRU cartridge with 10mL 8M HNO₃. (17) Strip U from TRU with 15mL of 0.1M ammonium bioxalate. (18) Add 0.5mL 10% TiCl₃.</p>	<p>(26) Remove filter from funnel assembly and mount filter on stainless steel planchet with 2-sided tape.</p>	
<p>(9) Rinse DGA cartridge with 10mL 0.1M HNO₃. (10) Place TRU cartridge above DGA. (11) Strip Am/Cm from TRU onto DGA with 15mL 4M HCl.</p>	<p>(19) Add 50ug Ce carrier to all samples. Mix well. Add 1mL 49% HF. Mix well. Wait 15-20 minutes. (20) Set up Resolve[®] Filter Funnel on vacuum box. (21) Wet filter with 3mL 80% ethanol followed by 3mL DI water. (22) Filter sample.</p>	<p>(27) Dry filter under heat lamp for 3-5 minutes. (28) Measure actinides by alpha spectrometry.</p>	

*Adding 50uL 30% H₂O₂ to the tube rinse can improve Uranium recoveries and decontamination in the Pu/Np fraction.

Method Performance

Analyte	Replicates	Tracer	Tracer % Yield	Analyte Reference (mBq/g)	Analyte Measured (mBq/g)	% Bias
²³⁹ Pu	5	²³⁶ Pu	90 ± 7	18.0	18 ± 2	0.0
²³⁸ Pu	5	²³⁶ Pu	90 ± 7	14.8	15 ± 2	1.4
²³⁷ Np	5	²³⁶ Pu	90 ± 7	37.0	33 ± 1	-11
²⁴¹ Am	5	²⁴³ Am	85 ± 6	25.4	24 ± 1	-5.5
²⁴⁴ Cm	5	²⁴³ Am	85 ± 6	35.0	35 ± 2	0.0
²³⁸ U	5	²³² U	77 ± 3	29.6	31 ± 3	4.7
²³⁴ U	5	²³² U	77 ± 3	28.4	26 ± 4	-8.5

References

1) Sherrod L. Maxwell, Brian K. Culligan, Angel Kelsey-Wall, Patrick J. Shaw, "Rapid radiochemical method for determination of actinides in emergency concrete and brick samples," *Analytica Chimica Acta*, 701(1), 112-118 (2011).