eichrom[®]

Rapid Determination of Actinides in 1g Concrete and Brick Samples

AN-1432-10

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%)
HCI (37%) NaOH
HF (49%) or NaF Boric acid

Denatured ethanol Sulfamic Acid

Ascorbic Acid

 H_2O_2 (30%)

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

NaNO₂

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

1g milled Concrete or Brick + tracers in zirconium crucible.

Fuse samples with 15g NaOH at 600°C for 15 minutes.

Dissolve fusion cake with H₂O. Transfer to 250mL c-tube.

Add 10mL 3M HNO_3 to crucible. Heat to dissolve residue. Transfer to same 25mL c-tube.

Add 125mg Fe and 5mg Lato c-tube. Dilute to 180mL.

Add 2mL 1.25M Ca(NO₃)₂, 3mL 3.2M (NH₄)₂HPO₄, 5mL 10% TiCl₃. Mix. Cool in ice bath for 10 min.

Centrifuge at 3500rpm. Decant Supernate.

Partially dissolve precipitate in 60mL 1.5M HCl. Some solids will remain. Dilute to 170mL.

Add 1mg La, and 3mL 10% TiCl₃. Mix.

Add 20mL 49% HF. Cool in ice bath for 10 min.

Centrifuge at 3500rpm. Decant Supernate.

Dissolve precipitate in 5mL 3M HNO₃-0.25M Boric acid, 7mL 70% HNO₃, and 7mL 2M Al(NO₃)₃. Warming samples can help complete dissolution.

Cool samples to room temperature.

Fix valence states. Mix between each addition of: 0.5mL 1.5M sulfamic acid, 10uL 50mg/mL Fe, 1.5mL 1M ascorbic acid, 1mL 3.5M NaNO₂, 1.5mL 70% HNO₃.

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

(1) Precondition stacked 2mL (23) Rinse sample tube with 5mL DI (12) Separate TRU cartridge from DGA TEVA, TRU, DGA cartridges water and add to filter. cartridge. Set TRU aside for U recovery. TEVA with 10mL 3M HNO₃. (13) Rinse DGA cartridge sequentially (24) Rinse filter (2) Load sample solution. assembly with 25mm, 0.1µm -5mL 4M HCI with: funnel with 3mL (3) Rinse sample tube Resolve IM polypropylene filter -5mL 1M HNO₃ DI water and 2mL with 5mL 3M HNO₃.* Add TRU -15mL 0.05M HNO₃ 100% ethanol. tube rinse to cartridges. (14) Strip Am and Cm from DGA with (25) Draw vacuum (4) Rinse cartridges with 10mL 0.25M HCI. Add 0.2mL 30% H₂O₂ until filter is dry. 10mL 3M HNO₃. DGA (15) Rinse TRU cartridge with 15mL (5) Separate TEVA, TRU, (26) Remove filter from 4M HCI-0.2M HF-0.002M TiCl₃ . and DGA cartridges. funnel assembly and (16) Rinse TRU cartridge with 10mL (6) Rinse TEVA cartridge with: mount filter on stainless 8M HNO₃. -10mL 3M HNO₃ steel planchet with 2-sided tape. -20mL 9M HCI (17) Strip U from TRU with 15mL of -5mL 3M HNO₃ 0.1M ammonium bioxalate. (7) Strip Pu (and Np) from TEVA (18) Add 0.5mL 10% TiCl₃. cartridge with 20mL 0.1M HCI-(19) Add 50ug Ce carrier to all samples. 0.05MHF-0.01M TiCl₃. Mix well. Add 1mL 49% HF. Mix well. (8) Add 0.5mL 30% H₂O₂ for Uranium Wait 15-20 minutes. decon. in alpha source preparation. (9) Rinse DGA cartridge with 10mL (20) Set up Resolve® Filter Funnel on

*Adding 50uL 30% H_2O_2 to the tube rinse can improve Uranium recoveries and decontamination in the Pu/Np fraction.

(21) Wet filter with 3mL 80% ethanol

followed by 3mL DI water.

vacuum box.

(22) Filter sample.

Method Performance

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

References

0.1M HNO₃.

(10) Place TRU cartridge above DGA.

(11) Strip Am/Cm from TRU onto

DGA with 15mL 4M HCI.

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).

(27) Dry filter under heat lamp for

(28) Measure actinides by alpha

3-5 minutes.

spectrometry.