AN-1433-10

Rapid Determination of Actinides in Emergency Air Filter Samples

Summary of Method U, Pu, Np, Am and Cm are separated and concentrated from air filters. Samples are digested in Teflon beakers once with HNO_3-H_2O_2-HF and then several times with HNO_3-H_2O_2. After evaporating to dryness from HNO_3-H_3BO_3 to complex any residual fluoride, actinides are valence adjusted and separated on stacked 2mL cartridges of Eichrom TEVA and TRU resins. Actinides are measured by alpha spectrometry following CeF_3 microprecipitation onto Eichrom Resolve® Filters. Chemical yields of tracers averaged from 94±12% for ^{242}Pu, 87±6% for ^{243}Am, and 67±32% for ^{232}U. Poor ^{232}U recoveries in some samples were traced to insufficient mass of Ce carrier in the source preparation step. Recovery of ^{232}U improved upon increasing to 100ug of Ce carrier. 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)
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
Ce carrier (1mg/mL)

Deionized water	2M $Al(NO_3)_3$
10% (w:w) $TiCl_3$	HNO_3 (70%)
HCl (37%)	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 Centrifuge Tubes
Centrifuge
Heat Lamp
Hot Plate
Analytical Balance
250mL Teflon beakers
Stainless Steel Planchets with adhesive tape
Alpha Spectrometry System
Vacuum Pump

Figure 1. Sample Preparation

Air Filter + tracers in Teflon beaker.

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Wet ash to dryness with 3mL 70% HNO_3, 3mL 30% H_2O_2 and 5mL 49% HF.

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Add 3mL 3M HNO_3 –0.25M H_3BO_3. Evaporate to dryness.

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Dissolve residue in 6mL 6M HNO_3 and 6mL 2M $Al(NO_3)_3$.

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Cool samples to room temperature.

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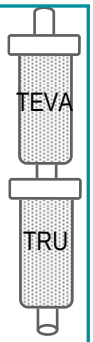
Fix valence states. Mix between each addition of: 0.5mL 1.5M sulfamic acid, 10uL 50mg/mL Fe, 1.5mL 1M ascorbic acid, 1mL 3.5M $NaNO_2$.

Figure 2. Actinide Separation on TEVA - TRU*

(1) Precondition stacked 2mL TEVA-TRU with 10mL 3M HNO_3.



(2) Load sample solution.

(3) Rinse sample tube with 5mL 6M HNO_3. Add tube rinse to cartridges.**

(4) Rinse cartridges with 5mL 3M HNO_3.

(5) Separate TEVA and TRU cartridges.

(6) Rinse TEVA cartridge with:
- -15mL 3M HNO_3
- -20mL 9M HCl(remove Th)
- -5mL 3M HNO_3

(7) Strip Pu(Np) from TEVA with 20mL 0.1M HCl-0.05MHF-0.01M $TiCl_3$. Add 0.5mL 30% H_2O_2 for additional U decon. during CeF_3

(8) Strip Am/Cm from TRU with 15mL 4M HCl. Dilute to 30mL and add 0.2mL 30% H_2O_2.

(9) Rinse TRU with 15mL 4M HCl-0.2M HF. (Th removal)

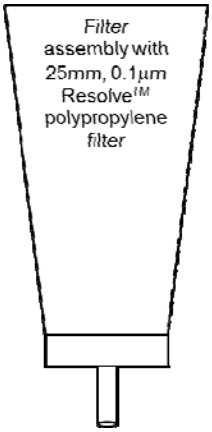
(10) Strip U from TRU with 15mL 0.1M ammonium bioxalate.

(11) Add 0.5mL of 10% $TiCl_3$ to each U sample for CeF_3 ppt.

(12) Add 50-100ug Ce carrier to all samples. Mix well. Add 1mL 49% HF. Mix well. Wait 15-20 minutes.

(13) Set up Resolve® Filter Funnel on vacuum box.

(14) Wet filter with 3mL 80% ethanol followed by 3mL DI water.



(15) Filter sample.

(16) Rinse sample tube with 5mL DI water and add to filter.

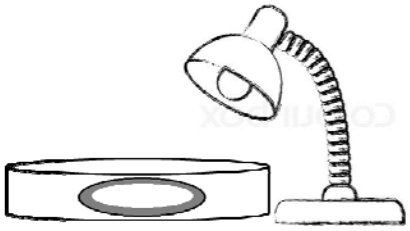
(17) Rinse filter funnel with 3mL DI water and 2mL 100% ethanol.

(18) Draw vacuum until filter is dry.

(19) Remove filter from funnel assembly and mount filter on stainless steel planchet with 2-sided tape.

(20) Dry filter under heat lamp for 3-5 minutes.

(21) Measure actinides by alpha spectrometry.



*$^{89/90}$Sr can also be measured by placing a 2mL Sr Resin cartridge below DGA and following the separation scheme in application note AN-1434

**Adding 50uL of 30% H_2O_2 to the 6M HNO_3 tube rinse can help improve U recoveries and decontamination in the Pu/Np fraction.

References

1) Sherrod L. Maxwell, Brian K. Culligan, Gary W. Noyes, "Rapid separation method for actinides in emergency air filter samples," *Applied Radiation and Isotopes,* 68(12), 2125-2131 (2010).