Rapid Determination of Actinides in Emergency Urine Samples

AN-1412-10

Summary of Method Uranium, Plutonium, and Americium-Curium are separated and concentrated from 100mL urine samples using calcium phosphate precipitation. The precipitate is dissolved in HNO_3-H_2O_2 and wet ashed to destroy residual organic material. The wet-ashed residue is dissolved in nitric acid and aluminum nitrate. Actinides are separated from matrix impurities and potentially interfering radionuclides in the sample using 2mL cartridges of Eichrom TEVA and TRU Resins. Actinides are measured by alpha spectrometry following source preparation by cerium fluoride microprecipitation onto Eichrom Resolve® Filters. Chemical yields are determined by recovery of ^{232}U, ^{243}Am, and ^{242}Pu (or ^{236}Pu, if measuring ^{237}Np) tracers. Typical chemical recoveries are >90%. A single operator can complete the separation method for batches of 12-24 samples in as little as 4-5 hours.

Reagents

TEVA Resin, 2mL Cartridges (Eichrom TE-R50-S)
TRU Resin, 2mL Cartridges (Eichrom TR-R50-S)
Ammonium Hydroxide (listed as 28% NH_3 or 56% NH_4OH)
^{242}Pu (or ^{236}Pu if meas. Np), ^{243}Am and ^{232}U tracers
Oxalic acid/Ammonium oxalate
Hydrofluoric Acid (49%) or Sodium Fluoride

Nitric Acid (70%)	Hydrochloric Acid (37%)
Hydrogen Peroxide (30%)	Deionized Water
Iron Carrier (50mg/mL)	Cerium Carrier (1mg/mL)
1.25M $Ca(NO_3)_2$	3.2M $(NH_4)_2HPO_4$
2M $Al(NO_3)_3$	10% (w:w) $TiCl_3$
Sulfamic Acid	Ascorbic Acid
Sodium Nitrite	Denatured Ethanol

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
Hot Plate
Analytical Balance
250mL Glass Beakers
Alpha Spectrometry System
Vacuum Pump
Heat Lamp

Figure 1. Sample Preparation

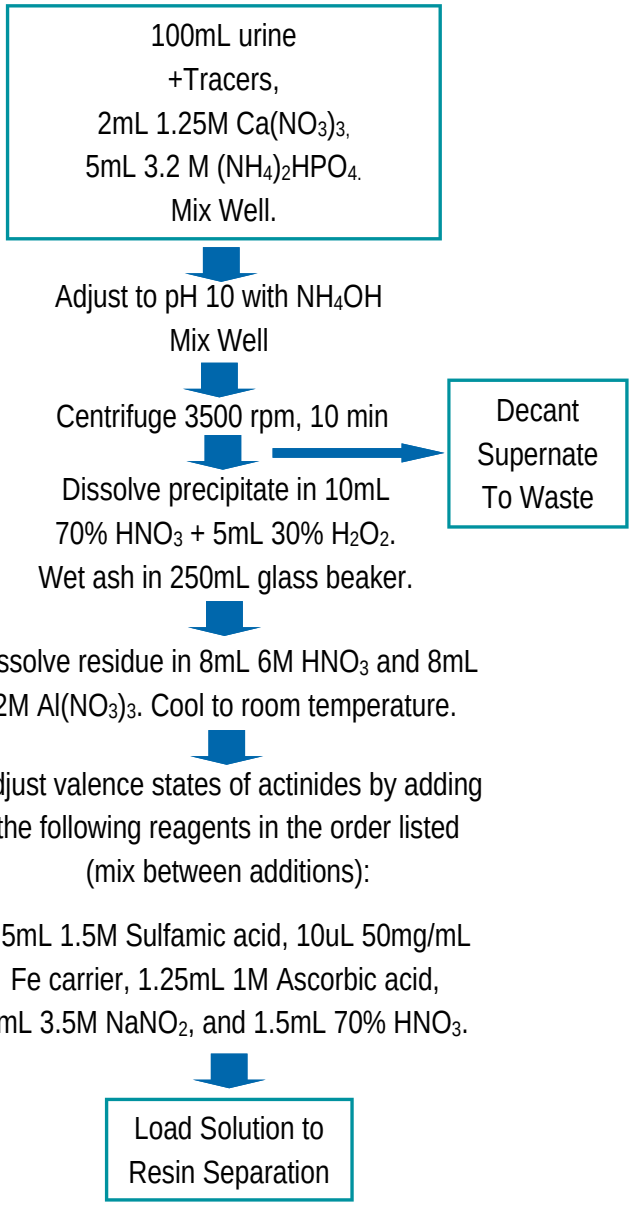
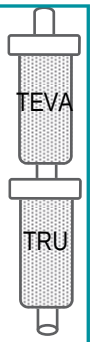


Figure 2. Actinide Separation on TEVA - TRU*

(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 $TiCl_3$.

(8) Strip Am/Cm from TRU with 15mL 4M HCl. Dilute to 30mL prior to CeF_3 ppt.

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

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

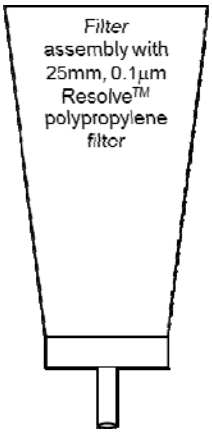
(11) Add 0.5mL 30% H_2O_2 to Pu and 0.2mL 30% H_2O_2 to Am/Cm samples for additional U decon. during CeF_3 ppt.

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

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

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

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



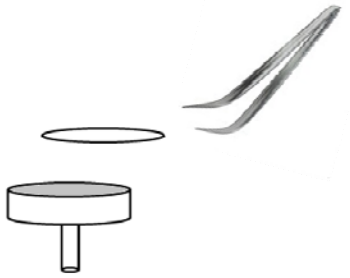
(16) Filter sample.

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

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

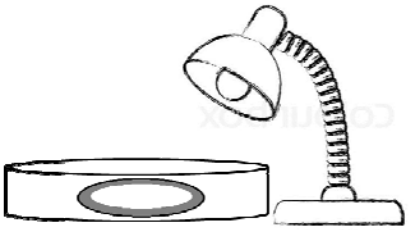
(19) Draw vacuum until filter is dry.

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



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

(22) Measure actinides by alpha spectrometry.



*Strontium may also be measured by adding a 2mL Sr Resin Cartridge below DGA and following the separation scheme in Eichrom application note AN-1410, "Rapid Determination of Sr in Emergency Urine Samples."

**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, "Rapid separation method for emergency water and urine samples," *J. Radioanal. Nucl. Chem., 279(3), 901-907* (2009).