

# Rapid Determination of Np/Pu/Am in 10-20g Soil Samples

**Summary of Method** Plutonium, Neptunium, and Americium are separated and concentrated from 10 gram soil samples. Samples are fused in zirconium crucibles with 40g NaOH to facilitate complete dissolution. Actinides are separated from matrix using an iron/titanium hydroxide precipitate. A second precipitate with Ca/La-fluoride is used to remove additional matrix, particularly silicates, and decrease the volume of precipitate. Actinides are separated from potential radiometric impurities using 2mL cartridges of TEVA and DGA Resins. Am/Cm fractions may require additional purification using TEVA-NH<sub>4</sub>SCN to remove native rare earths which can degrade alpha spectra. Pu/Np and Am/Cm are prepared for alpha spectrometry measurement via CeF<sub>3</sub> microprecipitation onto Resolve® Filters. To further lower detection limits, two 10g soil aliquots can be fused separately, combined following the Fe/Ti hydroxide precipitate, and then processed through the remaining steps of the method.

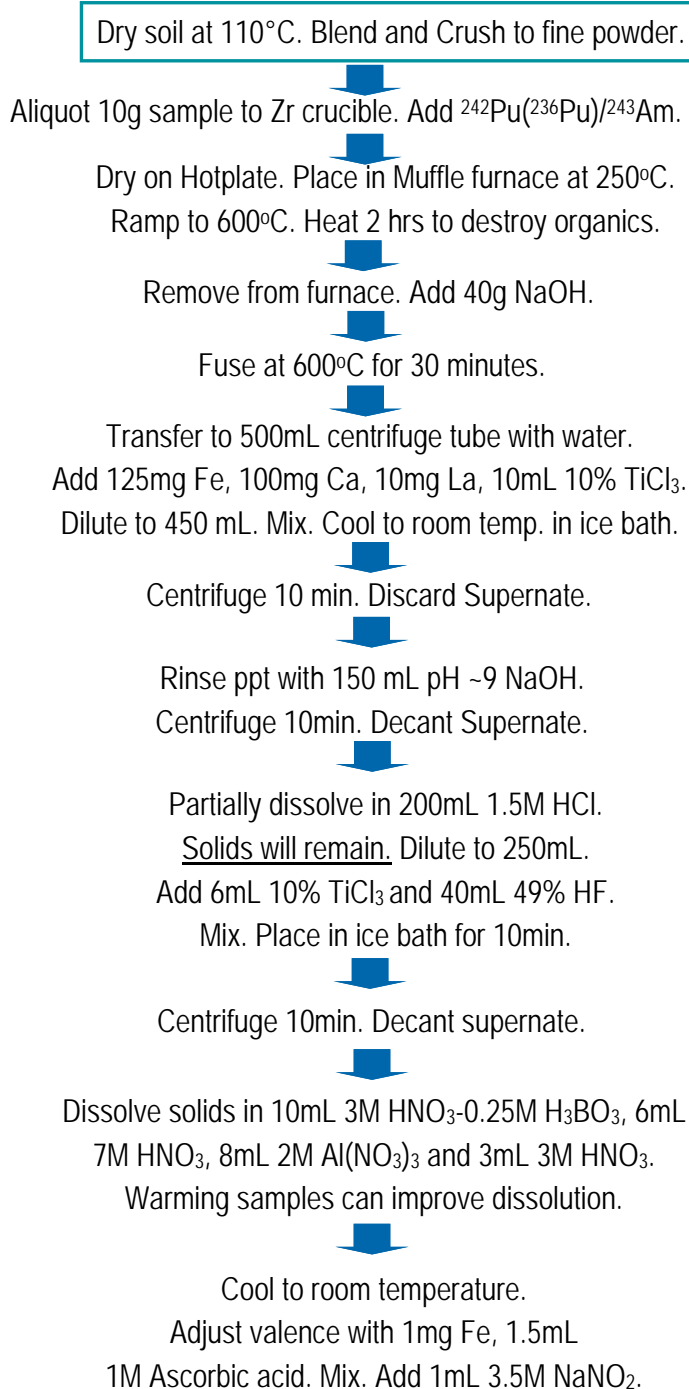
## Reagents

TEVA Resin, 2mL Cartridges (Eichrom TE-R50-S)  
 DGA Resin, Normal, 2mL Cartridges (Eichrom DN-R50-S)  
 Iron carrier (50mg/mL Fe, as ferric iron nitrate)  
<sup>242</sup>Pu (or <sup>236</sup>Pu if Np Measured) tracer  
<sup>243</sup>Am tracer                      La carrier (10mg/mL)  
 Ce Carrier (10mg/mL)        1.25M Ca(NO<sub>3</sub>)<sub>2</sub>  
 Deionized Water                2M Al(NO<sub>3</sub>)<sub>3</sub>  
 10% (w:w) TiCl<sub>3</sub>                HNO<sub>3</sub> (70%)  
 HCl (37%)                         NaOH  
 HF (49%) or NaF                Boric acid  
 NaNO<sub>2</sub>                              Sulfamic Acid  
 Ascorbic Acid                    30% H<sub>2</sub>O<sub>2</sub>  
 NH<sub>4</sub>SCN (rare earth separation)

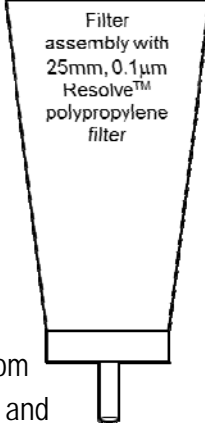
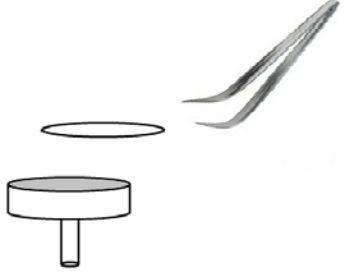
## Equipment

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 with funnel (Eichrom RF-DF25-25PP01)  
 250mL Zirconium Crucibles with lids  
 50mL and 500mL Centrifuge Tubes  
 Alpha Spectrometry System  
 Stainless Steel Planchets with double sided tape  
 Centrifuge                         Hot Plate  
 Analytical Balance                Vacuum Pump  
 Muffle Furnace

**Figure 1. Sample Preparation**



**Figure 2. Actinide Separation on TEVA - DGA**

|   |   |   |
|---|---|---|
| <p>(1) Precondition 2mL TEVA + DGA cartridges with 5mL 8M HNO<sub>3</sub>.</p> <p>(2) Load Sample.</p> <p>(3) Rinse centrifuge tube with 5mL 6M HNO<sub>3</sub>. Add to TEVA + DGA.*</p> <p>(4) Rinse TEVA + DGA with 10mL 3M HNO<sub>3</sub>.</p> <p>(5) Separate TEVA and DGA.</p>          | <p><b>Optional: Rare Earth Removal Steps</b></p> <p>(10) Add 3mL 3M HNO<sub>3</sub> and 3mL 30% H<sub>2</sub>O<sub>2</sub> to Am/Cm fraction.</p> <p>(11) Wet ash on hotplate to dryness.</p> <p>(12) Dissolve in 10mL 1.5M NH<sub>4</sub>SCN.</p> <p>(13) Precondition 2mL TEVA with 5mL 1.5M NH<sub>4</sub>SCN.</p> <p>(14) Load Am/Cm fraction.</p> <p>(15) Rinse beaker with 5mL 1.5M NH<sub>4</sub>SCN. Add to TEVA.</p> <p>(16) Rinse TEVA with 5mL 1.5M NH<sub>4</sub>SCN.</p> <p>(17) Strip Am/Cm with 20mL 1M HCl.</p> | <p>(23) Rinse filter funnel with 3mL DI water and 2mL 100% ethanol.</p> <p>(24) Draw vacuum until filter is dry.</p> <p>(25) Remove filter from funnel assembly and mount filter on stainless steel planchet with 2-sided tape.</p> |
| <p>(6) Rinse TEVA with</p> <ul style="list-style-type: none"> <li>-15mL 3M HNO<sub>3</sub></li> <li>-20mL 9M HCl (Th)</li> <li>-5mL 3M HNO<sub>3</sub></li> </ul> <p>(7) Strip Pu/Np from TEVA with 20mL 0.1M HCl-0.05M HF-0.01M TiCl<sub>3</sub>.</p>  | <p>(18) Add 50ug Ce carrier and 0.5mL 30% H<sub>2</sub>O<sub>2</sub> 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.</p> <p>(22) Rinse sample tube with 5mL DI water and add to filter.</p>   |    |
| <p>(8) Rinse DGA with:</p> <ul style="list-style-type: none"> <li>-10mL 3M HCl (Ca)</li> <li>-3mL 1M HNO<sub>3</sub></li> <li>-15mL 0.1M HNO<sub>3</sub> (La, Ca)</li> <li>-25mL 3M HNO<sub>3</sub>-0.25M HF (Th)</li> <li>-5mL 4M HCl</li> </ul> <p>(9) Strip Am/Cm with 12mL 0.25M HCl.</p> | <p>(26) Dry filter under heat lamp for 3-5 minutes.</p> <p>(27) Measure actinides by alpha spectrometry.</p>  |   |

\*Adding 50uL of 30% H<sub>2</sub>O<sub>2</sub> to the 6M HNO<sub>3</sub> tube rinse can further improve uranium decontamination factors.

**Method Performance**

| Sample   | replicates | analyte               | tracer            | % tracer recovery | mBq/g reference | mBq/g measured | % bias |
|----------|------------|-----------------------|-------------------|-------------------|-----------------|----------------|--------|
| 10g Soil | 10         | <sup>239/240</sup> Pu | <sup>236</sup> Pu | 85 ± 8            | 3.43            | 3.41 ± 0.22    | ± 5    |
| 10g Soil | 6          | <sup>237</sup> Np     | <sup>236</sup> Pu | 82 ± 4            | 3.99            | 4.19 ± 0.16    | ± 6    |
| 10g Soil | 11         | <sup>241</sup> Am     | <sup>243</sup> Am | 89 ± 4            | 2.14            | 2.07 ± 0.16    | ± 6    |

**References**

1) Maxwell, Culligan, Hutchinson, McAlister, "Rapid Fusion Method for the Determination of Pu, Np, and Am in Large Soil Samples," *J. Radioanal. Nucl. Chem.* 305 : 599-608 (2015).