

Rapid Determination of Np/Pu in 20-75g Soil Samples (ICP-MS)

Summary of Method Plutonium and Neptunium are separated and concentrated from 20-75 gram soil samples. Samples are leached with HNO₃ and HCl. The leachates are evaporated to dryness, and sequential precipitations with Fe/Ti-hydroxide and LaF₃ facilitate matrix removal. Pu-Np are separated on 2mL cartridges of Eichrom TEVA and DGA resins. Pu-Np are measured by ICP-MS. Chemical yields of the ²⁴²Pu tracer were 87±4%, 75±6%, and 70±3% for 20, 50 and 75g samples, respectively. Measured values for ²³⁹Pu agreed to within 1% of reference values, while ²³⁷Np agreed to within 15%. Decontamination factors of >10⁶ were achieved for Pu over U (²³⁸U-H can interfere with the measurement of ²³⁹Pu by ICP-MS). Sample preparation for batches of 12 samples can be completed by a single operator in <8 hours.

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)
²⁴²Pu tracer La carrier (10mg/mL)
 Deionized Water 2M Al(NO₃)₃
 10% (w:w) TiCl₃ HNO₃ (70%)
 HCl (37%) NH₄OH (28% HN₃ or 56% NH₄OH)
 HF (49%) or NaF Boric acid
 NaNO₂ Sulfamic Acid
 Ascorbic Acid Hydroxylamine Hydrochloride

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)
 600mL Glass beakers
 50mL and 250mL Centrifuge Tubes
 ICP-MS system
 Centrifuge
 Hot Plate
 Analytical Balance
 Vacuum Pump

Figure 1. Sample Preparation

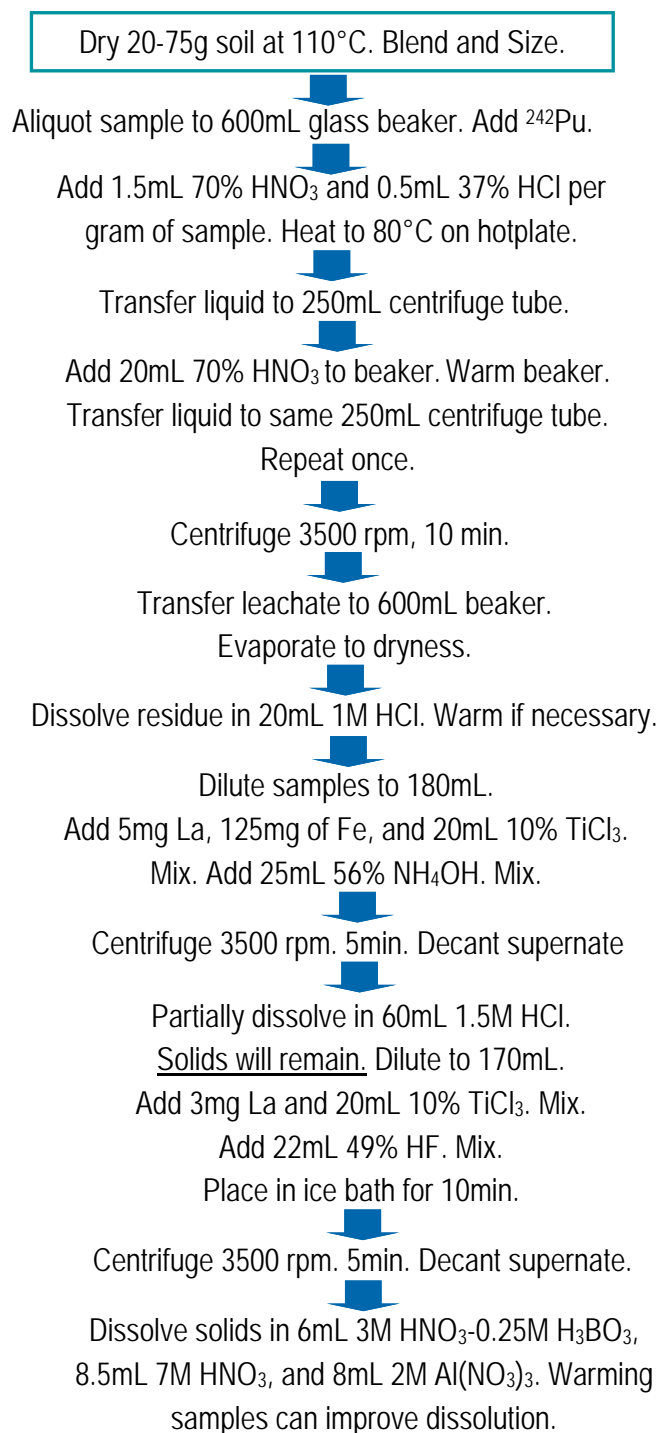




Figure 2. Actinide Separation on TEVA - DGA

<p>Cool samples to room temp. Fix valence by adding: (mix between steps) -0.5mL 1.5M sulfamic acid -40uL 50mg/mL Fe carrier -1.5mL 1M ascorbic acid (Wait 3 min) -1mL 3.5M NaNO₂</p>	<p>(8) Rinse TEVA with 5mL 3M HNO₃. (9) Strip Np from TEVA with 14mL 0.25M HCl-0.005M HF-0.01M Hydroxylamine hydrochloride.</p>	
<p>(1) Precondition 2mL TEVA, cartridges with 5mL 8M HNO₃. (2) Load Sample. (3) Rinse centrifuge tube with 5mL 6M HNO₃. Add to TEVA.* (4) Rinse cartridges with: -30mL 3M HNO₃ -15mL 9M HCl (Th removal)</p>	<p>(10) Rinse DGA with: -5mL 8M HNO₃ -20mL 0.1M HNO₃ -10mL 0.05M HNO₃. (11) Strip Pu from DGA with 11mL 0.02M HCl-0.005M HF-0.01M Hydroxylamine hydrochloride.</p>	
<p>(5) Add 2mL DGA cartridge below TEVA.** (6) Strip Pu from TEVA onto DGA with 15mL 3M HNO₃-0.1M Ascorbic Acid-0.02M Fe²⁺. (7) Separate TEVA and DGA.</p>	<p>(12) Measure ²³⁷Np and Pu by ICP-MS.</p>	
		

*Adding 50uL of 30% H₂O₂ to the 6M HNO₃ tube rinse can further improve uranium decontamination factors.

**Placing a 1mL UTEVA cartridge between TEVA and DGA can provide additional decontamination from uranium.

References

- 1) Sherrod L. Maxwell, Brian K. Culligan, Gary W. Noyes, "Rapid separation method for ²³⁷Np and Pu isotopes in large soil samples," *Applied Radiation and Isotopes*, 69(7), 917-925 (2011).
- 2) Sherrod L. Maxwell, Brian K. Culligan, Vernon D. Jones, Sheldon T. Nichols, Gary W. Noyes, Maureen A. Bernard, "Rapid Determination of ²³⁷Np and Plutonium Isotopes by ICP-MS and Alpha Spectrometry," *Health Physics*, 101(2), 180-186 (2011).