AN-1435-10

Rapid Determination of Np/Pu in 20-50g Soil Samples

Summary of Method Plutonium and Neptunium are separated and concentrated from 20-50 gram soil samples. Samples are leached with HNO_3 and HCl. The leachates are evaporated to dryness, and sequential precipitations with Fe/Ti-hydroxide and LaF_3 facilitate matrix removal. Pu-Np are separated on 2mL cartridges of Eichrom TEVA resin. Pu-Np are measured by alpha spectrometry following CeF_3 microprecipitation onto Eichrom Resolve® Filters. Chemical yields of the ^{236}Pu tracer ranged from 82-96%. 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.

Reagents

TEVA Resin, 2mL Cartridges (Eichrom TE-R50-S)
Iron carrier (50mg/mL Fe, as ferric iron nitrate)

^{236}Pu tracer	NH_4OH (28% NH_3 or 56% NH_4OH)
La carrier (10mg/mL)	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)
600mL Glass beakers
Stainless Steel Planchets with adhesive tape
Alpha Spectrometry System
50mL and 250mL Centrifuge Tubes
Centrifuge
Heat Lamp
Hot Plate
Analytical Balance
Vacuum Pump

Figure 1. Sample Preparation

Dry 20-50g soil at 110°C. Blend and Size.

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Aliquot sample to 600mL glass beaker. Add ^{236}Pu.

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Add 1.5mL 70% HNO_3 and 0.5mL 37% HCl per gram of sample. Heat to 80°C on hotplate.

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Transfer liquid to 250mL centrifuge tube.

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Add 20mL 70% HNO_3 to beaker. Warm beaker. Transfer liquid to same 250mL centrifuge tube. Repeat once.

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Centrifuge 3500 rpm, 10 min.

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Transfer leachate to 600mL beaker.
Evaporate to dryness.

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Dissolve residue in 20mL 1M HCl. Warm if necessary.

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Dilute samples to 180mL.
Add 5mg La, 125mg of Fe, and 20mL 10% $TiCl_3$.
Mix. Add 25mL 56% NH_4OH. Mix.

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Centrifuge 3500 rpm. 5min. Decant supernate

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Partially dissolve in 60mL 1.5M HCl.
Solids will remain. Dilute to 170mL.
Add 3mg La and 20mL 10% $TiCl_3$. Mix.
Add 22mL 49% HF. Mix.
Place in ice bath for 10min.

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Centrifuge 3500 rpm. 5min. Decant supernate.

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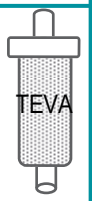
Dissolve solids in 6mL 3M HNO_3-0.25M H_3BO_3, 8.5mL 7M HNO_3, and 8mL 2M $Al(NO_3)_3$. Warming samples can improve dissolution.

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

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 $NaNO_2$

(1) Precondition 2mL TEVA, cartridges with 5mL 8M HNO_3.



(2) Load Sample.

(3) Rinse centrifuge tube with 5mL 6M HNO_3.*
Add to TEVA.

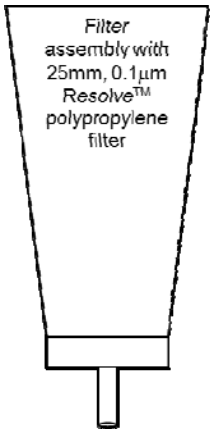
(4) Rinse cartridges with:
-15mL 3M HNO_3
-20mL 9M HCl
-5mL 3M HNO_3.

(5) Strip Pu/Np from TEVA with 20mL 0.1M HCl-0.05M HF-0.03M $TiCl_3$.

(6) Add 0.5mL 30% H_2O_2 to each sample for additional uranium decontamination during CeF_3 ppt.

(7) 50ug Ce carrier. Mix.
Add 1mL 49% HF. Mix.
Wait 15-20 minutes.

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



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

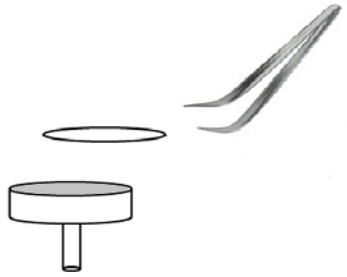
(10) Filter sample.

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

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

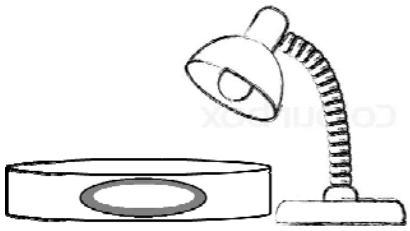
(13) Draw vacuum until filter is dry.

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



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

(16) Measure actinides by alpha spectrometry.



*Adding 50uL 30% H_2O_2 to the tube rinse can improve Uranium decontamination.

Sample, g	replicates	^{236}Pu Tracer % Yield	^{239}Pu Reference (mBq)	^{239}Pu Measured (mBq)	^{239}Pu %Bias	^{238}Pu Reference (mBq)	^{238}Pu Measured (mBq)	^{238}Pu %Bias	^{237}Np Reference (mBq)	^{237}Np Measured (mBq)	^{237}Np %Bias
20	6	89±6	116.3	118±7	1.5	63.2	67±4	6.0	37.0	39±4	5.4
20	6	96±7	1.69	2.1±0.4	24	25.3	25±2	-1.2	37.0	35±2	-5.4
30	6	82±6	116.3	121±5	4.0	63.2	68±5	7.6	37.0	39±4	5.4
50	6	88±5	116.3	114±3	-2.0	63.2	64±2	1.3	37.0	21±11	-43

References

1) Sherrod L. Maxwell, Brian K. Culligan, Gary W. Noyes, "Rapid separation method for ^{237}Np and Pu isotopes in large soil samples," *Applied Radiation and Isotopes,* 69(7), 917-925 (2011).