AN-1807-11

Alpha Spectrometry Source Preparation: Cerium Hydroxide Microprecipitation

Summary of Method Cerium hydroxide microprecipitation is an alternative to rare earth fluoride microprecipitation and electrodeposition for alpha spectrometry source preparation, providing adequate alpha peak resolution for most analytical applications, while greatly reducing the time for sample preparation relative to electrodeposition. Alpha spectrometry sources can often be prepared directly from the eluate used to recover the actinide fraction from the chromatographic column used to separate the actinides from the sample matrix and potentially interfering nuclides, eliminating the numerous evaporation and digestion steps normally required for electrodeposition, reducing the alpha spectrometry source preparation time from 3-8 hours to 30-60 minutes, and eliminating the emission of corrosive acid fumes through the laboratory fume hood vents.

Cerium hydroxide is an alternative to rare earth fluoride microprecipitation for labs looking to avoid the use of HF. Cerium hydroxide precipitates will nearly quantitatively carry actinides in all oxidation states from mineral acid solutions, but **will not work from bioxalate or other complexing agents**. Additional U decontamination of Th, Np/Pu and Am/Cm samples achieved by the rare earth fluoride precipitation (AN-1805) will not occur using the cerium hydroxide precipitation. The cerium hydroxide precipitate has a yellow color, providing visual confirmation of the collection of the precipitate on the Resolve Filter and easy identification of the surface of the filter containing the precipitate.

Cerium carrier, hydrogen peroxide and a pH indicator are added to each sample fraction from the appropriate separation method. After mixing to distribute the carrier, ammonium hydroxide is added to adjust the pH. The optimal pH and the appropriate pH indicator will depend on the actinide metal ion being collected. U and Th show the highest recovery from pH 5-7, utilizing the bromocresol purple pH indicator. However, U and Th recoveries do not decrease dramatically if the pH is increased to 8-10. Am and Pu/Np recoveries are most consistent utilizing thymol blue, with a color change from pH 8-10. The higher pH range is important to ensure high recoveries of Am. Since Pu and Np are often measured together, with a single ^{236}Pu yield tracer, it is important that their recoveries are very similar. The pH of 8-10 is important to ensure similar recoveries of Pu and Np. At lower or higher pH, Np recovery can diverge significantly from Pu. [2]

Eichrom's Resolve Filters (RF-DF25-25PE01) are manufactured specifically for alpha spectrometry source preparation. The manufacture and quality control procedures ensure a uniform surface for the collection of the rare earth fluoride precipitate, reducing self attenuation of the alpha emissions, which can degrade peak resolution. Other filter membranes may not be suitable for alpha source preparation or may require the addition of substrate to achieve adequate resolution.

Sources prepared by micropreciptitation precipitation and mounted to stainless steel planchets with double-sided tape or glue typically sit closer to the detector in alpha spectrometry systems than electrodeposition sources. The difference in distance from the source to the detector can lead to a 5-10% higher efficiency for the measurement of microprecipitation sources. Since most laboratories calibrate their alpha spectrometry systems with electrodeposited sources, the efficiency difference must be considered when determining the absolute recovery of the chemical yield tracers.

Reagents

Cerium Carrier (10 mg/mL)
Deionized Water
30% Hydrogen Peroxide (H_2O_2)
Denatured Ethanol
Ammonium Hydroxide (NH_4OH)
Bromocresol Purple or Thymol Blue

Equipment

Vacuum Box (Eichrom AR-24-BOX or AR-12-BOX)
Yellow Outer Tips (Eichrom AR-1000-OT)
Resolve Filters in Funnel (Eichrom RF-DF25-25PE01)
Stainless Steel planchets with two sided tape (A.F Murphy part no F-2-C)
Alpha Spectrometry System
Heat Lamp
50 mL Centrifuge Tubes
Vacuum Pump

Figure 1. Cerium Hydroxide Alpha Spectrometry Source Preparation*

Uranium Samples

(1) Obtain a purified sample of U in a 50 mL centrifuge tube using an appropriate separation method. Samples are typically in 10-20 mL of 1M HCl.

(2) Add 25-50 μg of Ce carrier, 0.2 mL of 30% H_2O_2 and bromocresol purple. Mix.

(3) Adjust to pH 5-7 (blue/purple color). Mix well. Proceed to step (4).

Thorium Samples

(1) Obtain a purified sample of Th in a 50 mL centrifuge tube using an appropriate separation method. Samples are typically in 20 mL of 6-9M HCl.

(2) Add 25-50 μg of Ce carrier, 0.2 mL of 30% H_2O_2 and bromocresol purple. Mix.

(3) Adjust to pH 5-7 (blue/purple color). Mix well. Proceed to step (4).

Pu/Np Samples

(1) Obtain a purified sample of Pu/Np in a 50 mL centrifuge tube using an appropriate separation method. Samples are typically in 15-20 mL of dilute HCl-HF with a reducing agent.

(2) Add 25-50 μg of Ce carrier, 0.2 mL of 30% H_2O_2 and thymol blue. Mix.

(3) Adjust to pH 8-10 (light blue color). Mix well. Proceed to step (4).

Am/Cm, An(III), and Ln(III) Samples

(1) Obtain a purified sample of Am/Cm, An(III) or Ln(III) in a 50 mL centrifuge tube using an appropriate separation method. Samples are typically in 15-20 mL of 0.1-4M HCl. Samples with high native rare earth content will require removal of rare earths using TEVA-SCN (AN-1806).

(2) Add 25-50 μg of Ce carrier, 0.2 mL of 30% H_2O_2 and thymol blue. Mix.

(3) Adjust to pH 8-10 (light blue color). Mix well. Proceed to step (4).

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

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

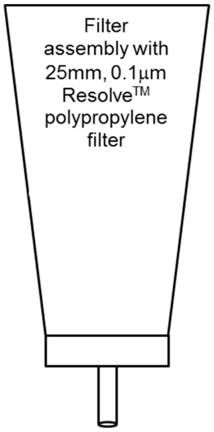
(6) Filter sample.

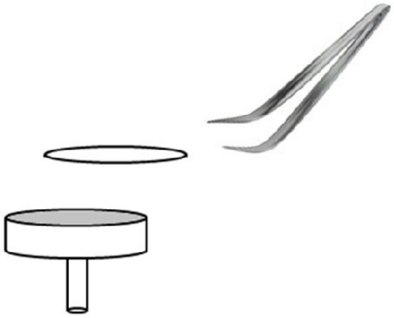
(7) Rinse sample tube with 5 mL DI water. Add to filter.

(8) Rinse funnel with 3 mL DI water and 2 mL ethanol.

(9) Draw vacuum until filter is dry.

(10) Remove filter from funnel. Mount filter on stainless steel planchet with 2-sided tape.**





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

(12) Measure actinides by alpha spectrometry.

***Results in Table 1 are for typical sample matrix from separation method. Performance with other matrices or volumes should be verified prior to implementation.**

***Some users prefer to dry the filters before mounting. With the polyethylene Resolve Filters®, this can lead to curling, making the filters more difficult to mount. Mounting the filters prior to drying is recommended.**

Typical Performance of $Ce(OH)_4$ Microprecipitation onto Eichrom Resolve Filters

Nuclide	pH	μg Ce	Matrix	Yield	Resolution (FWHM)
^{230}Th	5-7	25	20 mL 9M HCl	95-99%	25-35 keV
$^{238/234}$U	5-7	25	20 mL 1M HCl	93-97%	25-35 keV
^{237}Np	8-10	25	20 mL 0.15M HCl-0.05M KF-0.04M Rongalite	89-93%	25-35 keV
^{239}Pu	8-10	25	20 mL 0.15M HCl-0.05M KF-0.04M Rongalite	86-92%	25-35 keV
^{241}Am	8-10	25	15 mL 4M HCl	91-95%	25-35 keV

References

1) Claude W. Sill, “Precipitation of Actinides as Fluorides or Hydroxides for High-Resolution Alpha Spectrometry,” Nuclear and Chemical Waste Management, 7, 201-215 (1987).

2) Hiromu Kurosaki, Rebbeca J. Mueller, Susan B. Lambert, Govind R. Rao, “Alternate method of source preparation for alpha spectrometry: no electrodeposition, no hydrofluoric acid,” *J. Radioanal. Nucl. Chem.*, 311, 323-329 (2017).