

Determination of ^{225}Ac in Geological Samples

Summary of Method Soil or rock samples are pulverized to <1mm and dissolved, either by acid digestion or sodium hydroxide fusion. ^{225}Ac ($t_{1/2} = 10$ days) is separated from matrix ions using a ferric hydroxide precipitation. Following dissolution in 4M HCl, ^{225}Ac is separated from radiometric impurities using 2 mL cartridges of TRU and DGA, Normal resin. ^{225}Ac is prepared for measurement using a CeF_3 or LaF_3 microprecipitation on Resolve[®] Filters. Chemical recovery of actinium can be traced using ^{227}Ac (alpha spectrometry) or stable La (ICP-MS or ICP-AES). ^{225}Ac may be measured by alpha spectrometry (5.54-5.83 MeV) or gamma spectrometry (via its ^{221}Fr daughter, 218 keV, 11.44%). The alpha emission from the ^{227}Ac tracer (4.71-4.90 MeV) only occurs in 1.38% of decays. Therefore, use of ^{227}Ac tracer may be more efficient by measuring its ^{227}Th (5.59-6.04 MeV) or ^{223}Ra (5.24-5.87 MeV) daughters after a period of ingrowth and decay of ^{225}Ac . The mass of La that can be added to use a yield tracer must be minimized (55 μg) to prevent degradation of the alpha spectra through self-absorption. However, this amount of La should be sufficient to measure via ICP-MS or ICP-AES.

Reagents

DGA Resin, 2 mL Cartridges (Eichrom DN-R50-S)
 TRU Resin, 2 mL cartridges (Eichrom TR-R50-S)
 Iron Carrier (50 mg/mL Fe, as ferric nitrate)
 Cerium carrier (10 mg/mL)
 ^{227}Ac tracer or Lanthanum Carrier (10 mg/mL)
 Hydrofluoric Acid (49%) or Sodium Fluoride
 Boric acid HNO_3 (70%) HCl (37%)
 NaOH Deionized Water H_2O_2 (30%)

Equipment

Vacuum Box (Eichrom AR-24-BOX or AR-12-BOX)
 Cartridge Reservoir, 20 mL (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)
 50 mL and 250 mL Centrifuge Tubes
 Alpha Spectrometry System
 Ball mill grinder or equivalent
 Centrifuge Vacuum Pump
 Heat Lamp Analytical Balance

Fusion Option

250 mL Zirconium crucibles with zirconium lids
 Muffle Furnace

Digestion Option

Hot Plate Teflon Beakers

Sample Preparation

0.25-50g Soil or Rock

Pulverize to <1mm.

Aliquot Sample.

Add ^{227}Ac tracer or 55 μg La.

Acid Digestion Option

Digest in Teflon beaker on hotplate with 2:1 conc. HNO_3 :HF to near dryness.

Digest in Teflon beaker on hotplate with conc. HNO_3 + Boric Acid.

Dissolve Residue in 4M HCl + 0.25M Boric acid. If solids remain. Repeat digestion. Proceed to ferric hydroxide precipitation.

Fusion Option

In Zr crucible. Add 10-15g NaOH

Muffle at 600°C for 15-30 minutes.

Cool. Dissolve fusion cake with 50mL water.

Heat as necessary. Rinse crucible with 50mL 4M HCl.

Proceed to ferric hydroxide precipitation.

Ferric Hydroxide Precipitation

Transfer sample to 250mL centrifuge tube.

Dilute to 150mL with water.

Add 25mg Fe carrier. Mix well.

Centrifuge 2500 rpm for 10 minutes.

Decant Supernate.

Rinse ppt with 50mL water. Centrifuge.

Decant Supernate.

Dissolve precipitate with 10mL conc. HNO_3 .

Dilute to 25mL.

Ac Separation on TRU-DGA

(1) Precondition 2mL TRU and DGA with 10mL 6M HNO₃.

(2) Load sample solution.

(3) Rinse sample tube with 5mL 6M HNO₃. Add tube rinse to TRU-DGA.

(4) Rinse TRU-DGA with 10mL 3M HNO₃.

(5) Rinse TRU-DGA with 15mL 8M HCl (transfers La/Ac to DGA).

(6) Discard TRU. Rinse DGA with 20mL 0.5M HNO₃.

(7) Strip Ac from DGA with 20 mL 2M HCl. (2M HCl is used to achieve additional decontamination from Th.)

*If using stable La as the yield tracer, remove 2 mL aliquot and dilute to 10 mL for recovery measurement by ICP-MS or ICP-AES.



(7) Add 0.5mL 30% H₂O₂ to samples.

(8) Add 50ug Ce carrier** to samples.

Mix well. Add 1mL 49% HF.

Mix well. Wait 15-20 minutes.

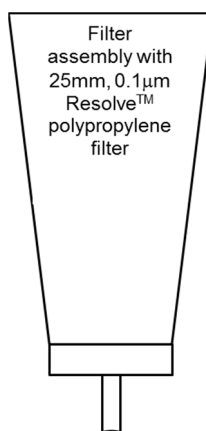
**If stable La is used as the yield tracer, omit the addition of 50 ug of Ce. The La will serve as the carrier for the rare earth fluoride micro-ppt.

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

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

(11) Filter sample.

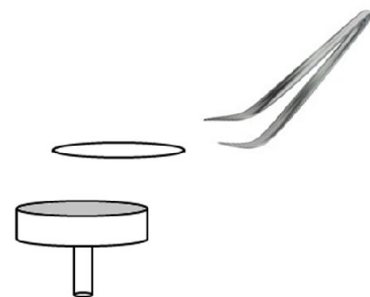
(12) Rinse sample tube with 5mL DI water and add to filter.



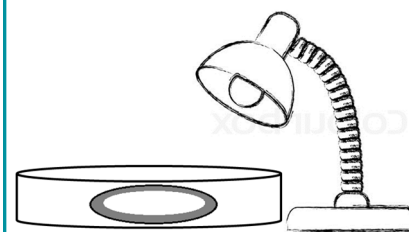
(13) Rinse filter funnel with 3mL DI water and 2mL 100% ethanol.

(14) Draw vacuum until filter is dry.

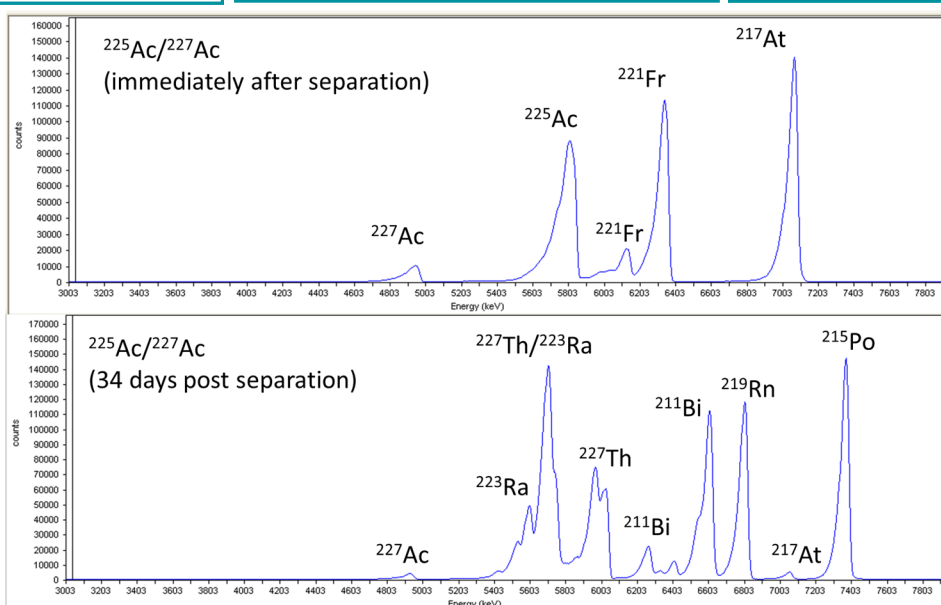
(15) Remove filter from funnel assembly and mount filter on stainless steel planchet with 2-sided tape.



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



(17) Measure actinides by alpha spectrometry.



References

1) H. Dulaiova, K.W.W. Sims, M.A. Charette, J. Prytulak, J.S. Blusztajn "A new method for the determination of actinium-227 in geological samples," *J. Radioanal. Nucl. Chem.*, 296, 279-283 (2013).