

Rapid Determination of ^{90}Sr in up to 40 Liter Seawater Samples

Summary of Method Yttrium-90, the daughter product of ^{90}Sr decay, is separated and concentrated from up to 40L samples of seawater. A ferric hydroxide precipitate enhanced with 10mg of lanthanum and 1mg of yttrium concentrates ^{90}Y , while rejecting much of the salt content of the seawater sample. A second precipitation with lanthanum fluoride removes additional matrix ions. Yttrium is separated from potentially interfering radionuclides in the sample, including rare earths such as ^{138}La and $^{139/144}\text{Ce}$, using a 2mL cartridge of Eichrom DGA Resin. ^{90}Y is measured on a low background gas flow proportional counter following cerium fluoride microprecipitation onto an Eichrom Resolve® Filter. Chemical yield of stable yttrium is determined by ICP-MS or ICP-AES. Average chemical recovery of yttrium is $84 \pm 7\%$ for 40L samples. Measured values of ^{90}Sr (^{90}Y) agree to within 5% of reference values, with two hour count times. The minimum detectable activity for ^{90}Sr for 40L samples with a two hour count time is 0.35mBq/L. The average time to complete the method is 8 hours. While standard methods targeting Sr are limited by the ~8mg/L native Sr content in seawater, targeting ^{90}Y directly allows for the efficient processing of very large seawater samples to achieve very low minimum detectable activities. However, interference by the fission product ^{91}Y ($t_{1/2} = 58.51$ days) precludes application of this method for the measurement of ^{90}Sr (^{90}Y) immediately following a radiological incident involving the release of un-aged nuclear fuel or fission products.

Reagents

DGA Resin, Normal 2mL Cartridges (Eichrom DN-R50-S)
 Nitric Acid (70%)
 Hydrochloric Acid (37%)
 Hydrofluoric Acid (49%) or Sodium Fluoride
 Ammonium Hydroxide (listed as 28% NH_3 or 56% NH_4OH)
 Deionized Water
 Iron Carrier (50mg/mL Fe, as ferric nitrate)
 Yttrium and Cerium Carriers (1mg/mL)
 Lanthanum Carrier (10mg/mL)
 1.25M $\text{Ca}(\text{NO}_3)_2$ 2M $\text{Al}(\text{NO}_3)_3$
 ^{90}Sr standard Boric 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)
 50mL and 500mL Centrifuge Tubes
 Centrifuge
 Gas Flow Proportional Counter
 Analytical Balance
 Vacuum Pump
 Heat Lamp

Figure 1. Sample Preparation

Up to 40L Sample of Seawater.
 Acidify to pH 2 with 37% HCl.
 Add 1mg Yttrium carrier.

Add 10mg La carrier. Add 50mg Fe carrier per liter of sample. Mix Well.

Adjust to pH 9 with 56% NH_4OH . Mix.
 Allow precipitate to settle.

Decant supernate until ~2L remains.

Transfer remaining supernate and precipitate to 500mL centrifuge tubes. Centrifuge 3000rpm for 10 minutes. Decant supernate. Repeat until entire sample centrifuged.

Wash precipitate with 100mL water.
 Centrifuge. Decant supernate.

Dissolve precipitate in 100mL 1.5M HCl.
 Add 75mg Ca and 50mL 49% HF. Mix.
 Wait 15 minutes. Centrifuge. Decant supernate.

Dissolve precipitate in 10mL 3M HNO_3 -
 0.25M Boric acid, 10mL 70% HNO_3 ,
 and 10mL 2M $\text{Al}(\text{NO}_3)_3$.

Load Solution for Sr separation.

Figure 2. Yttrium Separation on DGA and CeF₃ Microprecipitation

(1) Precondition DGA Resin with 5mL 8M HNO₃.

(2) Load sample at 1-2mL/min.

(3) Rinse sample tube with 5mL 8M HNO₃.

(4) Add tube rinse to DGA Resin. Elute at 1-2mL/min.

(5) Rinse DGA Resin sequentially with:

- 15 mL 8M HNO₃ (Ca, Sr, Pb)
- 20mL 0.05M HNO₃ (La, Ce, Sr, U)
- 15mL 3M HNO₃-0.25M HF (U, Th)
- 10mL 3M HCl (Ca, La, Pb)

(6) Dispose of (1) to (5) as waste.

(7) Strip Y with 20mL 0.25M HCl at 1mL/min.

(8) Remove 0.1-1.0mL aliquot for stable Y recovery by ICP-MS or ICP-AES. Dilute aliquot as appropriate.

(9) To remaining sample:

- Add 100ug Ce carrier.
- Mix well.
- Add 2mL 49% HF.
- Mix well. Wait 15-20 minutes.

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

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

(12) Filter sample.

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

(14) Rinse filter funnel with 3mL DI water.

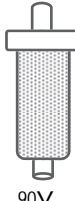
(15) Rinse filter funnel with 1-2mL 100% ethanol.

(16) Draw vacuum until filter is dry.

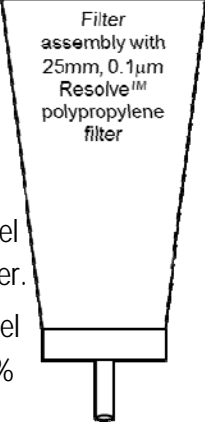
(17) Remove filter from funnel assembly and mount filter on stainless steel planchet with adhesive tape.

(18) Dry filter under heat lamp for 10-15 minutes.

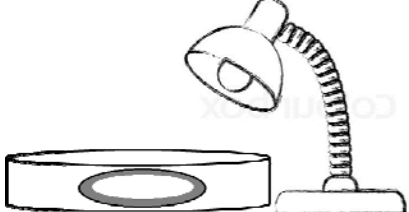
(19) Measure ⁹⁰Y on low background gas flow proportional counter.



2mL DGA Cartridge
⁹⁰Y



Filter assembly with 25mm, 0.1µm Resolve™ polypropylene filter



Method Performance 10-40L Spike Seawater Samples

Sample Volume, L	% Recovery Y carrier	⁹⁰ Sr (mBq/L) Reference	⁹⁰ Sr (mBq/L) Measured	% Bias
10	85.5	296	310	4.7
20	89.2	28.2	28.1	-0.4
30	72.3	18.8	18.5	-1.6
40	87.6	14.1	13.7	-2.8
40	86.5	14.1	13.9	-1.4

MDA for 40L sample = 0.35 mBq/L for 2 hour count time

MDA for 40L sample = 0.20 mBq/L for 8 hour count time

References

1) Sherrod L. Maxwell, Brian K. Culligan, Jay B. Hutchinson, Robin C. Utsey, Daniel R. McAlister, "Rapid determination of ⁹⁰Sr in seawater samples," *J. Radioanal. Nucl. Chem.*, 303, 709-717 (2015).