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Standard Revision. Change bars denote changes.

1.0 PURPOSE

The method is designed to analyze for Pu, Np, U, Th, Am/Cm isotopes in 1-2 gram emergency or routine soil samples per CTF via alpha spectrometry. It provides rapid total dissolution of samples and efficient removal of alpha interferences prior to counting by alpha spectrometry. Strontium–89/90 is also separated and counted by gas proportional counting.

2.0 SCOPE

Samples are fused at 600C using sodium hydroxide in zirconium crucibles. An iron hydroxide precipitation is performed. After acidification of the precipitate, a lanthanum fluoride precipitation is done to further eliminate the sample matrix. The lanthanum fluoride precipitate is redissolved in nitric acid, boric acid and aluminum nitrate. A column separation using TEVA, TRU and DGA Resins are applied to separate the actinides into three fractions: plutonium-neptunium, uranium and americium/curium. Pu-242 (or Pu-236 if Np-237 is measured), Th-229, Am-243, U-232 are used as tracers to determine yield. Some isotopes of plutonium can also be reported. Th-228, if present as a daughter of U-232 tracer, will interfere with Th-228 analysis. “Self-cleaning” U-232 tracer with Th-228 removed is required if thorium isotopes are separated and measured with uranium. Sr Resin is used to separate Sr-89/90 for measurement by beta counting.

This procedure shall provide instructions for Environmental Bioassay Laboratory (EBL) personnel.

3.0 PRECAUTIONS/LIMITATIONS

3.1 Safety

1. Comply with Standing Radiological Work Permits (SRWPs), as applicable.
2. Comply with L3.23-40000, EBL General Safety Rules, for additional Personal Protective Equipment (PPE) requirements and handling requirements for use of concentrated acids.
3. All personnel using this procedure shall follow posted/approved site, department, and facility safety rules, and any additional requirements which may be included in these instructions.
4. Hydrofluoric acid (HF) is extremely corrosive and is a bone seeker. Burns may appear hours after the initial contact. HF acid has been classified as being a hazardous chemical (blue dot) strictly. Handle concentrations of HF acid greater than 1 molar (M) in a containment unit. The fumes of the acid are irritating to the eyes and respiratory tract.
5. When acid contacts skin and/or eyes, immediately shower with large quantities of water for 15 minutes and immediately notify First Line Manager (FLM). Respond to HF exposure per Manual 4Q, Procedure 111, “First Response Measures for HF Skin Contact” and BAL-ADMIN-0016, “BAL Spill Management Program”.
6. HF solutions attack (dissolves) glass, therefore, use polypropylene, polyethylene, teflon, or platinum containers. If HF < 1M, glass can be used if water is added to flask first to dilute the HF.
7. Wear heat resistant apron with sleeves, heat resistant gloves, face shield, goggles, and use tongs when loading and removing crucibles from the hot furnace.
8. Addition of reagents and tracers to sample aliquots shall be performed in a laboratory chemical fume hood.
9. Soil samples larger than approximately 2 grams should be analyzed using L3.23-10037 (unless directed otherwise per CTF), where larger samples are digested first using nitric acid and hydrofluoric acid to remove silica prior to the fusion.

3.2 Waste Precautions

1. All cartridges, columns, or other material that comes in contact with radioactive standards and spikes shall be disposed of in low level waste.
2. All liquids shall be drained from all materials before disposal.

3.3 Responsibilities

Laboratory Analysts - responsible for the execution of this procedure. The analysts must read, understand, and follow each step in this procedure. They must assess if an operation is safe to continue and know the proper channels to follow to stop work, when appropriate. They are responsible for chemical, sample, and waste labeling in compliance with the Hazard Communication Standard. Waste streams must be characterized and disposed of properly from a regulatory and environmental perspective.

Laboratory personnel - should follow the QA/QC protocol for all analytical operations and reagent preparations. Expiration dates of raw materials and prepared reagents must be observed and adhered to.

All M&TE instrumentation used in the analysis and reagent preparation must be calibrated, and the calibration documented according to the established frequency.

Resident - contact the B-Area Laboratory Chemical Coordinator before placing any chemical or radionuclide (internal/external sources included) in any Environmental Bioassay Laboratory (EBL) Facility. Chemicals or radionuclides that are disposed (in waste or by drain), spilled, dissipated (evaporated) or removed from the EBL (735-B/735-1B) facility in any manner must be subtracted from CMS by the end of each shift, except for full to empty.

Label chemical and waste, and update CMS in compliance with BAL-ADMIN-0012, BAL-ADMIN-0005 for low level waste, BAL-ADMIN-0010 for Green-Is-Clean waste, BAL-ADMIN-0007 for hazardous waste and BAL-ADMIN-0011 for hazardous waste.

CTF - may change sample aliquots, reagents, volumes, concentrations, flow rates, and the lab apparatus based on sample needs.

4.0 PREREQUISITES

4.1 Equipment

- Analytical balance - 0.0001 gram sensitivity
- Analytical balance - 0.01 gram sensitivity
- Beakers, 100 mL Teflon
- Centrifuge, 6 place rotor
- Centrifuge tubes, 225-mL and 50 mL graduated plastic
- Columns (empty luer tip, CC-10-M) and 12 mL reservoirs (CC-06-M), Image Molding, Denver, Co, or equivalent
- Connectors for vacuum box, PFA 5/32"x 1/4" Heavywall tubing, Natural, Ref P/N 00070EE, cut to 1 inch , Cole Parmer or equivalent
- Connector tips (yellow) for vacuum box extraction system such as Andwin Scientific CEN 4000 (250-100 uL) or equivalent

NOTE : *Zirconium crucibles are rinsed well with warm nitric acid as part of the cleaning process to remove any alkaline residue.*

- Zirconium crucibles, 250 mL (P/N 10-0250LF), with lids (P/N 10-025C), Metal Technology, Inc, Albany, OR, 800-394-9979.
- Filters - 25-mm, 0.1 micron polypropylene Eichrom Resolve Filters or equivalent

4.1 Equipment, Cont.

- Filter apparatus - Gelman apparatus (0.1 micron - 25 mm filters with polysulfone base, polysulfone screen, and 50-mL polysulfone funnel) and multi-port vacuum manifold
- Furnace
- Graduated cylinders - 1000-mL or less, plastic
- Infrared heat lamp
- Pipettes, calibrated, electronic pipetors or fixed sizes, as needed
- Plastic disks, 1 1/4"
- Heat resistant apron with sleeves
- Heat resistant gloves
- Hot plate
- Face shield and goggles
- Tongs
- Sample carrier (planchets and sample tower can be utilized)
- Tweezers

4.2 Reagents

- Actinide standards for laboratory control samples depending on which actinides are measured or as directed by CTF:
  - 0.5 mL ~ 2pCi/mL Pu-238
  - 0.5 mL ~ 2pCi/mL Am-241
  - 0.5 mL ~ 2pCi/mL Cm-244
  - 0.5 mL ~ 2pCi/mL U-235
  - 0.2 mL ~ 4pCi/mL Th-230
  - 0.5 mL ~ 2pCi/mL Np-237
  - 1 ml ~ 80 pCi/mL Sr-90
- Actinide tracers depending on which actinides are measured or as directed by CTF:
  - Uranium-232 tracer solution, 0.5 mL ~4 to 20 pCi/mL
  - Plutonium-236 tracer solution, 0.5 mL ~2 to10 pCi/mL
  - Plutonium-242 tracer solution, 1 mL~2 to10 pCi/mL
  - Americium-243 tracer solution, 1 mL ~2 to10 pCi/mL
  - Thorium-229 tracer solution, 1 mL ~1.5 to10 pCi/mL

NOTE 1: *All references to "water" refer to deionized or distilled water.*

NOTE 2: *CTF may direct removal of any uranium impurities in the aluminum nitrate using a UTEVA Resin separation as needed.*

- Ammonium hydrogen phosphate [$(NH_4)_2HPO_4$] solution, 3.2M: DISSOLVE 106 grams of $(NH_4)_2HPO_4$ in 200 mL of water, HEAT gently to dissolve, and DILUTE to 250 mL with water.
- Aluminum nitrate [$Al(NO_3)_3$] solution, 2M: 750 grams of aluminum nitrate [$Al(NO_3)_3$] $9H_2O$] shall be added to 600 mL of water and diluted to 1 liter with water.
- Ammonium bioxalate ($NH_4HC_2O_4$) solution, 0.1M: 6.31 grams of oxalic acid [$H_2C_2O_4$. $2H_2O$] and 7.11 grams of ammonium oxalate [$(NH_4)_2C_2O \bullet H_2O$] shall be dissolved in 900 mL of water, filter, and dilute to 1 liter with water.

4.2 Reagents, Cont.

- Ascorbic acid ($C_6H_8O_6$) solution, 1.5M: 132.1 grams of ascorbic acid ($C_6H_8O_6$) shall be dissolved in 250 mL of water; heat gently to dissolve and diluted to 500 mL with water (expiration date 30 days after preparation).
- Barium nitrate solution, 10%: 50.0 grams of barium nitrate shall be dissolved in ~300 mL of water; heat gently to dissolve, and diluted to 500 mL. CTF may direct use of an equivalent amount of barium chloride instead of barium nitrate.
- Cerium carrier, 0.5 mg Ce/mL: 0.155 g cerium (III) nitrate hexahydrate [$Ce(NO_3)_3 \cdot 6H_2O$] shall be dissolved in 50 mL water, and diluted to 100 mL with water.
- Calcium nitrate [$Ca(NO_3)_2$], 1.25M: DISSOLVE 147 grams of $Ca(NO_3)_2 \cdot 4H_2O$ in ~300 mL of water and DILUTE to 500 mL with water (~50 mg Ca/mL).
- Ethanol (C2H5OH), 95%: Use directly as purchased if already 95%. Typically, 95% is purchased.
- For pure (100%) ethanol: 95 mL ethanol shall be added to 5 mL water.
- Hydrochloric acid, 12M - concentrated hydrochloric acid (sp gr 1.19)
- Hydrochloric acid solution, 9M: 750 mL of concentrated HCl (sp gr 1.19) shall be added to 100 mL of water to dilute to 1 liter with water.
- Hydrochloric acid solution, 3M: 250 mL of concentrated HCl (sp gr 1.19) shall be added to 500 mL of water to dilute to 1 liter with water.
- Hydrochloric acid solution, 2M: 167 mL of concentrated HCl (sp gr 1.19) shall be added to 500 mL of water to dilute to 1 liter with water.
- Hydrochloric acid solution, 0.25M: 20.8 mL of concentrated HCl (sp gr 1.19) shall be added to 500 mL of water to dilute to 1 liter with water.
- Hydrochloric acid (0.1M) - Hydrofluoric acid (0.05M) solution: 1.8 mL concentrated HF and 8.3 mL concentrated HCl (sp gr 1.19) shall be added to 500 mL of water. Dilute to 1 liter with water, and mix well.
- Hydrochloric acid (0.01M) solution: 0.83 mL concentrated HCl (sp gr 1.19) shall be added to 500 mL of water. Dilute to 1 liter with water, and mix well.
- Hydrochloric acid (4M) - Hydrofluoric acid (0.2M) solution: 7.14 mL concentrated HF shall be added to 1000 mL 4M HCl, and mix well (3.57 mL concentrated HF for 500 mL volume).
- Hydrochloric acid (1.5M) – 125 mL of concentrated HCl (sp.gr.1.19) shall be added to ~ 800 mL of water and diluted to 100 mL with water.

NOTE: *Prepare Hydrochloric acid (.1M) - Hydrofluoric acid (0.05M)- titanium chloride (0.01M) freshly within 30 minutes of use.*

- Hydrochloric acid (0.1M) - Hydrofluoric acid (0.05M) - titanium chloride (0.01M) solution: 2.5 mL of 20 wt% titanium chloride shall be added per 250 mL of Hydrochloric acid (0.1M) - Hydrofluoric acid (0.05M) solution, and mix well. (For alternate volumes, prepare 1 mL 10 wt% titanium chloride for each 100 mL of hydrochloric acid (0.1M) - Hydrofluoric acid (0.05M) solution.
- Hydrogen peroxide (H_2O_2), 30%
- Hydrofluoric acid, 28M - concentrated hydrofluoric acid (sp gr 1.2)
- Iron carrier (50 mg/mL), 181 grams ferric nitrate dissolved in 500 mL with water.
- Iron carrier (5 mg/mL), 18.1 grams ferric nitrate dissolved in 500 mL with water.

4.2 Reagents, Cont.

- Lanthanum carrier, 0.5 mg La/mL: 0.78 g lanthanum (III) nitrate hexahydrate [$La(NO_3)_3 . 6H_2O$]shall be dissolved in 300 mL water, and diluted to 500 mL with water.
- Nitric acid, 15.8M - concentrated nitric acid (sp gr 1.42)
- Nitric acid solution, 3M: 190 mL of concentrated HNO_3 (sp gr 1.42) shall be added to 800 mL of water, and diluted to 1 liter with water.
- Nitric acid solution (3M)-oxalic acid (0.05M) ADD 191 mL of concentrated HNO_3 and 6.3 grams of oxalic acid dihydrate to 800 mL of water and dilute to 1 liter with water.
- Nitric acid–boric acid solution, 3M-0.25M: 15.4g of boric acid, 190 mL of concentrated HNO_3 (sp gr 1.42) shall be added to 500 mL of water, heat to dissolve, and diluted to 1 liter with water.
- Nitric acid solution, 8M: 510mL of concentrated HNO_3 (sp gr 1.42) shall be added to 400 mL of water, and diluted to 1 liter with water.
- Nitric acid solution, 7M: 446mL of concentrated HNO_3 (sp gr 1.42) shall be added to 400 mL of water, and diluted to 1 liter with water.
- Nitric acid solution, 6M: 378mL of concentrated HNO_3 (sp gr 1.42) shall be added to 400 mL of water, and diluted to 1 liter with water.
- Nitric acid solution, l M, 63 mL of concentrated HNO_3 shall be added to ~ 800 mL of water and diluted to 1 liter with water.
- Nitric acid solution, 0.1M, 3.16 mL of concentrated HNO_3 shall be added to 497mL of water to prepare 500 mL.
- Nitric acid solution, 0.05 M, 3.16 mL of concentrated HNO_3 shall be added to 997mL of water to prepare 1 liter.

NOTE: *New Sodium nitrite, 3.5 M, shall be prepared daily.*

- Sodium nitrite, 3.5 M: 6.1 grams of sodium nitrite shall be dissolved in 25 mL of water.
- Sr carrier, ~20 mg/mL (standardized)
- Sr Resin, 1mL and 2 mL cartridges, 0.7 g, 50-100 um particle size resin
- Sr-90 standard, ~80 pCi/mL
- Sulfamic acid, 1.5 M: 73 grams sulfamic acid shall be dissolved in 400 mL of water, and diluted to 500 mL with water.
- Titanium (III) chloride, [TiCl3]: 10 wt% solution in 20-30 wt% hydrochloric acid (available from Aldrich Chemical Corporation, Milwaukee, WS). DO NOT use if solids present when volume is very low in bottle.
- TEVA resin Prepackaged 2 mL cartridge, 0.7 g, 50-100 um particle size resin (available from: EiChrom Technologies, Inc., Tel: 630-963-0320)
- TRU resin Prepackaged 2 mL cartridge, 0.7 g, 50-100 um particle size resin
- DGA-N resin Prepackaged 2 mL cartridge, 0.7 g, 50-100 um particle size resin
- Sodium hydroxide pellets

5.0 PERFORMANCE

5.1 Sample Preparation

NOTE: *All of the following information pertains to this section:*

- *CTF may direct changing reagent concentrations, volumes, beaker sizes, etc. as needed depending on sample matrix.*
- *Tracers are added to an empty crucible to prepare the blank. Tracers and LCS standards are added to an empty crucible to prepare the LCS.*
- *Soil samples received by the EM sample collections group and dried, sieved and blended prior to analysis.*
- *Steps that can be performed in advance for efficiency such as preparing columns, labeling tubes, etc. may be done prior to chronological listing in sections of this procedure.*
- *CTF may direct chilling of water and 0.01 M HCl prior to use in order to reduce the time needed to cool the samples in the ice bath.*

1. **ADD** 1 gram of soil sample (or alternate amount per CTF) with appropriate tracers to a 250 mL zirconium crucible.

NOTE: *For actinide reagent blanks, an empty crucible is used. For Sr-89/90 only, sea-washed sand or an empty crucible containing 1ml 1.25M calcium nitrate may be used.*

2. **PREPARE** one duplicate, one blank and one LCS for each set of 20 or less samples.

NOTE: *CTF may direct use of different level tracers as needed for emergency soil samples.*

3. **PIPET** each volumes below (or per CTF) of each tracer solutions below into each beaker depending on which analyses are required:

Pu:	1 mL Pu-242 tracer
Pu/Np or Np only:	0.5 mL mL Pu-236 tracer
U:	0.5 mL U-232 tracer
Am/Cm:	1 mL Am-243 tracer
Th	1 mLTh-229 tracer
Sr-89/90:	0.3 mL strontium carrier (20 mg/mL Sr carrier)

4. **PIPET** LCS spikes to "LCS" crucible as instructed by CTF, **AND**

   RECORD spike volume, type and activity on batch analysis sheet.

NOTE: *LCS volumes may be increased per CTF as needed for emergency samples.*

5. **USE** aliquots of the following LCS spikes unless directed otherwise by CTF:

Pu:	0.5 mL Pu-238 standard
Pu/Np or Np only:	0.5 mL Pu-238 and 0.5mL Np-237 standards
Am:	0.5 mL Am-241 standard
Am/Cm:	0.5 mL Am-241 and 0.5 ml Cm-244 standards
U:	0.5 mL U-235 standard
Th-230	0.2 mL Th-230 standard
Sr-90	1 mL Sr-90 standard

NOTE: *A matrix spike sample is not required for the actinides, but is needed for Sr-89/90 analysis.*

6. **IF** Sr-89/90 is analyzed, **THEN**

   PIPET 1 mL of Sr-90 standard also into a matrix spike crucible.

7. **DRY** crucibles on medium heat on hot plate (using KIMFILL boards).

8. **REMOVE** and **ALLOW** crucibles to cool.

9. **ADD** 15 grams of sodium hydroxide (or alternate amount per CTF) to each crucible.

5.1 Sample Preparation, Cont.

!WARNING!: HEAT RESISTANT APRON WITH SLEEVES, HEAT RESISTANT GLOVES, FACE SHIELD, GOGGLES SHALL BE WORN, AND TONGS SHALL BE USED WHEN WORKING WITH HOT FURNACE.

10. **COVER** with lid, **AND**

    FUSE for approximately 10 minutes at 600C.

11. **REMOVE** hot crucibles from furnace very carefully using tongs, **AND**

    TRANSFER to hood. (typically using metal pans with Kimfill boards).

12. **ADD** approximately 25-50 mL of water to crucible about 10 minutes after removing crucibles from furnace, **AND**

    HEAT on hotplate to loosen/dissolve solids

13. **IF** necessary for dissolution, **THEN**

    ADD more water, **AND**

    WARM water as needed on a hotplate.

14. **PIPET** 2.5 mL (or alternate volume per CTF) of iron carrier (50 mg/mL) into a clean, labeled 225 mL centrifuge tube for each sample.

15. **PIPET** 8 mL (or alternate volume per CTF) of 0.5 mg La/mL to each tube.

16. **TRANSFER** each sample to a 225 mL centrifuge tube water, **THEN**

    RINSE crucibles well with water, **AND**

    TRANSFER rinses to each beaker.

17. **DILUTE** to approximately 180 mL with water.

18. **COOL** 225 mL centrifuge tubes in an ice bath to approximately room temperature.

NOTE: *For High Ca Soils, the CTF may direct that less Ca be added.*

19. **ADD** 1.25M Ca $(NO_3)_2$ and $(NH_4)_2HPO_4$ to each sample as follows:

    - For actinides only add 1ml 1.25M Ca $(NO_3)_2$ and 3 ml 3.2M $(NH_4)_2HPO_4$
    - For actinides and Sr-89/90; add 3ml .25 Ca $(NO_3)_2$ and 5 ml 3.2M $(NH_4)_2HPO_4$
    - For Sr-89/90 only; add 3ml 1.25M Ca $(NO_3)_2$ and 5 ml 3.2M $(NH_4)_2HPO_4$

NOTE: *The $TiCl_3$ needs to be mixed well to ensure effective uranium reduction.*

20. **PIPET** 5 mL of 20 wt% $TiCl_3$ into each tube, **AND**

    CAP and **MIX** immediately.

21. **COOL** 225 mL centrifuge tubes in an ice bath for approximately 10 minutes.

22. **CENTRIFUGE** tubes for 6 minutes at 3500 rpm.

23. **POUR** off the supernate liquid, **AND**

    DISCARD to waste.

5.1 Sample Preparation, Cont.

24. **ADD** 1.5M HCl to each tube to redissolve each sample in a total of 60 mL.
25. **CAP** and **SHAKE** each tube to dissolve solids as well as possible.
26. **DILUTE** each tube to ~170 mL with 0.01M HCl.
27. **CAP** and **MIX**.
28. **PIPET** 2 mL (or alternate volume per CTF) of 0.5 mg La/mL into each tube.
29. **PIPET** 1 mL 1.25M Ca $(NO_3)_2$ into each tube.

NOTE: *For High Ca Soils, the CTF may direct that no additional Ca be added.*

30. **PIPET** 3 mL of 20 wt% $TiCl_3$ into each tube (or alternate volume per CTF).
31. **CAP** and **MIX** each tube to dissolve solids as well as possible.

!WARNING!: USE EXTREME CARE WHEN HANDLING HF.

32. **ADD** 22 mL of concentrated HF into each tube.
33. **CAP** and **MIX**.
34. **PLACE** tubes to set in an ice bath for ~10-15 minutes to get the tubes very cold.
35. **CENTRIFUGE** for ~10 minutes at 3500 rpm or as needed.
36. **POUR OFF** supernate liquid, **AND**
    DISCARD to waste.
37. **IF** only Sr-89/90 is required (no actinides), **THEN**

    REDISSOLVE the precipitate in 5 ml concentrated HNO_3 5ml 3 ml HNO_3-0.25M boric acid 5 ml 2M $Al(NO_3)_3$, **AND**

    PROCEED TO Step 5.1.44.
38. **IF NOT, THEN**

    PROCEED TO Step 5.1.40.
39. **PIPET** 5 mL of warm 3M HNO_3 - 0.25M boric acid into each tube.
40. **CAP**, **MIX** and **TRANSFER** contents of the tube into a labeled 100 mL teflon beaker.
41. **PIPET** 6 mL of 7M nitric acid into each tube, **AND**

    TRANSFER rinse to beaker.
42. **PIPET** 7 mL of 2M aluminum nitrate acid into each tube, **AND**

    TRANSFER rinse to beaker.
43. **SWIRL** each beaker and warm on hot plate for a few minutes until warm to dissolve well (but do not overheat and evaporate).
44. **REMOVE** each beaker, **AND**

    ALLOW each beaker to cool to room temperature.

5.2 TEVA-TRU Separations

NOTE: *All of the following information must be read and understood:.*

- *A 3-column separation using TEVA, TRU, and DGA Resin is applied to separate the actinides into three fractions: Pu-Np on TEVA, U and Am/Cm using DGA and TRU. The CTF may change column load, rinse and strip reagent volumes and concentrations based on sample needs. Flow rates given in procedure are important. Do not exceed these flow rates unless directed by CTF.*
- *To avoid sample cross-contamination, keep vacuum boxes and lids very clean. The acid volumes should be added in the exact amounts specified.*
- *If only Pu, Np or Th analysis is required, only TEVA Resin is used and the separation using TRU and DGA Resin does not have to be performed.*
- *The CTF may direct that reagent volumes used in column work may be added by pouring rather than pipetting when accurate amounts can be added using volume marks on column reservoirs.*

5.2.1 Column Loading

1. **IF** more than a small amount of solid residue remains, **THEN**

   CENTRIFUGE samples in 50 ml tubes @3500 rpm for ~6 minutes to remove solids.
2. **TRANSFER** solutions to beakers, **AND**

   RINSE solids with 3 mL 3M HNO_3.
3. **CENTRIFUGE** samples @3500 rpm for ~6 minutes to remove rinsed solids, **AND**

   TRANSFER rinse to beakers.
4. **IF** Sr-89/90 is analyzed alone (no actinides), **THEN**

   PROCEED to Sr column separation in Section 5.4.
5. **PIPET** 0.5 mL 1.5M sulfamic acid to each solution in the sample load solution from Section 5.1, **AND**

   SWIRL to mix.
6. **IF** Np-237 is analyzed, **THEN**

   PIPET 0.2 mL of 5 mg/mL iron carrier into each beaker.
7. **IF** Np-237 is NOT analyzed, **DO NOT ADD** any iron carrier.
8. **PIPET** 1.25 mL of 1.5M ascorbic acid to each solution, swirling to mix.
9. **ALLOW** beakers to stand 3 minutes.
10. **PIPET** 1 mL of 3.5 M sodium nitrite into each solution, **AND**

    SWIRL to mix.
11. **PIPET** 1.5 mL of concentrated HNO_3 into each solution, **AND**

    SWIRL to mix.
12. **PLACE** one TEVA on top, one TRU cartridge then one DGA cartridge for each sample on the vacuum box using yellow connector tips on the bottom (or cartridges as needed), **AND**

    EMPTY columns with 12 mL reservoirs at the top.
13. **PLACE** a waste tube below each column or use a new labeled tube if Sr-89/90 is analyzed.

5.2.1 Column Loading, Cont.

14. **PIPET** 5 mL of 3M HNO_3 into the top reservoir to condition resin, **AND**

    ALLOW the cartridges to drain using vacuum at 1 drop per second.

15. **TRANSFER** each sample solution into each column, **AND**

    ALLOW the cartridge to drain using vacuum at ~1 drop per second.

16. **PIPET** 5 mL of 6M HNO_3 to rinse each beaker (warming if necessary), **AND**

    TRANSFER into the top of the appropriate column.

17. **ALLOW** the cartridge to drain at ~ 2 drops per second.

NOTE: *The load and rinse solution are collected to perform Sr-89/90 analysis.*

18. **TURN OFF** the vacuum, **AND**

    TRANSFER the rinse solution to a labeled 250 mL beaker and evaporate to - 15 mL to prepare for Sr – 89/90 separation in Section 5.4.

19. **PIPET** 10 mL of 3M HNO_3 directly into each column, **AND**

    ALLOW the cartridge to drain at ~ 2 drops per second.

20. **TURN OFF** the vacuum, **AND**

    TRANSFER the rinse solution to a labeled 250 mL beaker and evaporate to ~15-20 mL to prepare for Sr-89/90 separation in Section 5.4.

21. **DISCONNECT** the TEVA, TRU and DGA cartridges, **AND**

    PROCESS the TEVA and TRU / DGA cartridges on separate boxes from this point on.

22. **MOVE** the DGA cartridges to a separate vacuum box, **SET** the TRU cartridges aside for later use, **AND**

    PLACE the new columns and reservoirs above the DGA cartridges.

5.2.2 TEVA Cartridge

1. **IF** any solids are in the column above the TEVA cartridge, **THEN**

   REPLACE the column and reservoir
2. **ENSURE** a waste tube is below each column.
3. **PIPET** 10 mL of 3M HNO_3 directly into each column, **AND**

   ALLOW the cartridge to drain at ~ 2 drops per second.

5.2.3 Th Removal-TEVA Cartridge

NOTE: *9M HCl is added to remove thorium interference even when thorium is not collected and measured.*

1. **ALLOW** each volume of 9M HCl added to drain, **THEN**

   ADD the next volume of this rinse to the reservoir.
2. **IF** thorium is NOT analyzed, **THEN**

   PLACE waste tube below each TEVA column, **AND**

   PROCEED TO Step 5.2.3.4.
3. **IF** thorium is analyzed, **THEN**

   A. **ENSURE** clean, plastic tubes are labeled with sample number and "Th" and are below each column.

   B. **PLACE** new column, reservoirs, and yellow tips on each cartridge.
4. **PIPET** 10 mL of 9M HCl into each TEVA column reservoir, **AND**

   ALLOW cartridge to drain using vacuum at ~1-2 drops per second.
5. **PIPET** 10 mL of 9M HCl into each TEVA column reservoir, **AND**

   ALLOW cartridge to drain using vacuum at ~1-2 drops per second.
6. **TURN OFF** the vacuum, **AND**

   REMOVE the tubes.
7. **IF** Th is NOT analyzed, **THEN**

   DISCARD rinse solution, **AND**

   PROCEED to Section 5.2.4.
8. **IF** Th is analyzed, **THEN**

   DILUTE solutions in each tube to 45 mL with water, **AND**

   PROCEED to Section 5.3.

5.2.4 Np-Pu Removal-TEVA Cartridge

NOTE: *The 5 mL 3M HNO_3 rinse may be collected with the 9M HCL thorium removal rinse if there is enough room in the tube and Th is not being analyzed.*

1. **PIPET** 5 mL of 3M HNO_3 into each TEVA column to remove any residual extractant, **AND**

   ALLOW cartridge to drain (slowly) into a waste tube using vacuum at ~2 drops per second.

2. **TURN OFF** the vacuum.

3. **REMOVE** tubes, **AND**

   DISCARD rinse solution.

4. **PLACE** a clean, labeled plastic tube below each cartridge.

5. **PLACE** new reservoirs and yellow tips on each TEVA cartridge, **AND**

   PREPARE freshly made 0.1M HCl – 0.05 M HF - 0.01M $TiCl_3$ by adding 2.5 mL of 20% titanium chloride per 250 mL of 0.1M HCl - .0.05M HF. (1mL $TiCl_3$ per 100 mL).

6. **ADD** 20 mL of 0.1M HCl-0.05M HF-0.01M $TiCl_3$ into each column reservoir to strip plutonium and/or neptunium, **AND**

   ALLOW cartridge to drain using vacuum at ~1-2 drops per second.

7. **TURN OFF** the vacuum, **AND**

   REMOVE tubes.

8. **SET** tubes aside for cerium fluoride filter preparation in Section 5.3.

5.2.5 U-Am Separation-DGA and TRU Cartridges

NOTE: *If only Pu, Np analysis is required, TEVA Resin is used and the separation using DGA and TRU Resin does not have to be performed.*

1. **PLACE** the DGA cartridges on the vacuum box.
2. **PLACE** new columns and reservoirs and yellow tips on each DGA cartridge.
3. **ENSURE** lid is clean.~
4. **PLACE** a labeled tube is below each cartridge.
5. **PIPET** 10 mL (or alternate volume per CTF) of 0.1M HNO_3 directly into each DGA column.
6. **ALLOW** to drain using vacuum at approximately 1-2 drops per second.
7. **TURN OFF** the vacuum, **AND**

   REMOVE the tubes.
8. **IF** Sr-89/90 is analyzed, **THEN**

   TRANSFER rinse solution to the labeled 250 mL beaker from Step 5.2.1.16, rinsing tubes with ~ 3 mL of 3M HNO_3, **AND**

   EVAPORATE rinse solutions to dryness for Sr-89/90 separation in Section 5.4.
9. **IF** Sr-89/90 is NOT analyzed, **THEN**

   DISCARD rinse solution.
10. **PLACE** the TRU cartridges above each DGA cartridge.
11. **ENSURE** a waste tube is below each column.
12. **PIPET** 15 mL of 3M HCl into each column to strip any Am/Cm from the TRU Resin onto the DGA resin.
13. **ALLOW** to drain at ~1drop per second using vacuum.
14. **TURN OFF** the vacuum, **AND**

    SET aside the TRU cartridges.
15. **ENSURE** a waste tube is below each column.
16. **ADD** 5 mL of 3M HCl into each column, **AND**

    ALLOW cartridge to drain using vacuum at ~1-2 drops per second.
17. **ADD** 3 mL 1M HNO_3 into each column, **AND**

    ALLOW cartridge to drain using vacuum at ~2 drops per second.
18. **ADD** 15 mL 0.10M HNO_3 (or volume per CTF) into each column, **AND**

    ALLOW cartridge to drain using vacuum at ~1-2 drops per second.
19. **TURN OFF** the vacuum, **THEN**

    REMOVE tubes, **AND**

    DISCARD rinse to trade waste drain.
20. **PLACE** new columns and reservoirs and yellow tips on each cartridge.
21. **PLACE** a clean, plastic tube labeled with sample number and "Am" below each DGA cartridge.

5.2.5 U-Am Separation-TRU Cartridge, Cont.

22. **ADD** 10 mL of 0.25M HCl directly into each DGA column.
23. **ALLOW** the HCl strip solution to drain through each cartridge using vacuum at ~1 drop per second or slower.
24. **TURN OFF** the vacuum.
25. **IF** Am/Cm is analyzed, **THEN**

    SET tubes aside for cerium fluoride filter preparation in Section 5.3.
26. **IF** Am is NOT analyzed, **THEN**

    DISCARD the Am-Cm strip solutions to liquid waste.
27. **IF** the uranium fraction is NOT to be analyzed, **THEN**

    PROCEED to Section 5.3.
28. **PLACE** waste tubes in the vacuum box below each cartridge.
29. **PLACE** new columns and reservoirs and yellow tips on each TRU cartridge, **AND**

    PREPARE freshly made 4M HCl – 0.2 M HF - 0.002M $TiCl_3$ by adding 0.6 mL of 20% titanium chloride per 300 mL of 4M HCl - .2M HF. (0.2mL $TiCl_3$ per 100 mL).
30. **PIPET** 12 mL of 4M HCl - 0.20 M HF - 0.002M $TiCl_3$ to the TRU cartridge to strip any residual thorium, **AND**

    ALLOW the cartridge to drain using vacuum at 1-2 drops per second.
31. **PIPET** 5 mL (or alternate volume per CTF) of 8M HNO_3 directly into each column, **AND**

    ALLOW the cartridge to drain at ~ 2 drops per second.
32. **TURN OFF** the vacuum, **AND**

    DISCARD rinse solution.
33. **PLACE** new yellow tips below each cartridge.
34. **ENSURE** clean, plastic tubes are labeled with sample number and "U" and are below each column.
35. **ENSURE** lid is clean.
36. **ADD** 15 mL of 0.1M ammonium bioxalate to elute uranium from each TRU column.
37. **ALLOW** the cartridge to drain using vacuum at ~1-2 drops per second.
38. **SET** the "U" tubes aside for cerium fluoride precipitation.

5.3 Cerium Fluoride Precipitation

1. **PIPET** 0.1 mL of cerium carrier to tubes containing final fractions for uranium, americium/curium, and plutonium/neptunium.
2. **SWIRL** to mix thoroughly.

NOTE: *For U levels>10 pCi: in tube, use 0.15 ml cerium carrier.*

3. **PIPET** 0.5 mL of titanium chloride to uranium fractions only.
4. **SWIRL** to mix thoroughly.
5. **PIPET** 0.5 mL of 30 wt% hydrogen peroxide to Pu/Np samples only.
6. **SWIRL** to mix thoroughly.

5.3 Cerium Fluoride Precipitation, Cont.

!WARNING!: USE EXTREME CARE WHEN HANDLING HF.

7. **PIPET** 1 mL of concentrated HF to each tube for Pu/Np , and U samples only.
8. **PIPET** 1mL of concentrated HF to each Am/Cm sample only.
9. **PIPET** 5 mL of concentrated HF to each Th sample only (where 9M HCl was diluted to 45 mL with water).
10. **CAP** and **SWIRL** to mix well.
11. **ALLOW** the solutions set for at least 15 minutes before filtering.
12. **SET UP** a 0.1 micron, 25-mm membrane filter, glossy side down, on a Gelman filter apparatus with polysulfone screen and 50-mL polysulfone funnel.
13. **PLACE** the appropriate number of filter funnels onto the vacuum manifold.
14. **IF** some manifold ports are NOT needed, **THEN**

    TURN OFF the vacuum to the unused ports.
15. **ADD** a few drops of 95% ethanol to wet each filter, **AND**

    APPLY vacuum.
16. **ENSURE** NO leaks are along the sides.
17. **ADD** 2 - 3 mL of water to each filter while vacuum is being applied (typically 10-15 inches of mercury).
18. **FILTER** each sample slowly.
19. **RINSE** plastic sample tubes with 5 mL of water, **AND**

    TRANSFER the rinse to the filter apparatus.
20. **RINSE** each filter with 5 mL of water, rinsing the walls of the filter funnel.

NOTE: *This ethanol rinse should require less than 10 drops of ethanol*.

21. **WASH** each filter with enough 95% ethanol to displace the water.
22. **REMOVE** filters, **THEN**

    DRY under heat lamps for ~ 5 minutes or more.
23. **ENSURE** filters are completely dry.
24. **IF** the filter funnels are to be used more than once per batch, **THEN**

    RINSE with water before the next use.
25. **WRITE** the sample tracking number on the back of a self-adhesive plastic disk with indelible ink for each sample.
26. **MOUNT** each filter on the appropriate plastic disk.
27. **ENSURE** the filter is NOT wrinkled and is CENTERED on the plastic disk.
28. **LOG** onto SQL*LIMS, **AND**

    ENTER the appropriate preparation information, **AND**

    PRINT the SQL*LIMS Worklist.

5.3 Cerium Fluoride Precipitation, Cont.

29. **PLACE** the sample disks in a sample carrier so that the disks do not contact one another so that each disk is in a planchet inside a planchet holder, which is in turn placed in a sample tower.

NOTE: *CTF may direct that disks may be leached in 15ml warm 3 ml HNO_3 – 0.25M boric acid and reprocessed through columns to remove interference as needed.*

30. **TRANSPORT** the sample carrier to the counting room, **AND**

COUNT by alpha spectrometry using L3.21-10005.

5.4 Strontium Separation

1. **TRANSFER** the evaporated load and rinse solutions from Section 5.2 to 50 mL centrifuge tubes.

RINSE beaker with ~ 3 – 5 mL of 8M HNO_3 **AND** add rinse to centrifuge tubes.

DILUTE to ~25 mL with 8M HNO_3

2. **TRANSFER** the solutions to 50 mL centrifuge tubes and **CENTRIFUGE** samples @3500 rpm for ~6 minutes to remove residual solids.

3. **TRANSFER** solutions to beakers

AND rinse solids with 3 mL 8M HNO_3.

4. **CENTRIFUGE** samples @3500 rpm for ~6 minutes to remove rinsed solids and **TRANSFER** rinse to beakers.

5. **ENSURE** Sr Resin cartridges (2 mL + 1 mL) are on vacuum box for each sample.

6. **USE** yellow connector tips (with heavy wall tubing connectors inserted) on bottom and empty columns with 12mL reservoirs at top.

7. **PLACE** waste tube below each column.

12. **PIPET** 5 mL of 8M HNO_3 into the column reservoir to condition resin, **AND**

ALLOW cartridges to drain using vacuum at 1 drop per second.

13. **LOAD** each Sr sample solution onto Sr cartridges (2 mL+1 mL), **AND**

ALLOW cartridges to drain using vacuum at 1 drop per second.

14. **ADD** 5 mL of 8M HNO_3 to rinse each beaker.

15. **TRANSFER** into column reservoir, **AND**

ALLOW cartridges to drain at ~ 1 drop per second.

16. **PIPET** 10 mL of 8M HNO_3 into each column, **AND**

ALLOW to drain at ~ 2 drops per second.

17. **PIPET** 5 mLs of 3M HNO_3-0.05M oxalic acid into each column, **AND**

ALLOW to drain at ~ 1-2 drops per second.

18. **PIPET** 10 mLs of 8M HNO_3 into each column, **AND**

ALLOW to drain at ~ 2 drops per second.

19. **RECORD** end time of last rinse to nearest 15 minutes as start time of yttrium in-growth.

20. **TURN** vacuum off, **AND**

DISCARD rinse solution.

5.4 Strontium Separation, Con't

NOTE: *CTF may direct that small aliquots of this solution may be mounted instead of 15 ml for higher level samples.*

21. **ELUTE** strontium with 15 mL of 0.05N HNO_3 into a clean, labeled tube at ~ 1 drop per second.

5.5 Strontium Mounting

1. **LABEL** clean muffled planchets in numerical order using a paint marker.
2. **WEIGH** a clean muffled planchet on an analytical balance accurate to the nearest tenth of a milligram, **AND**

   RECORD tare weight.
3. **ADD** sample in small portions to planchet on a hotplate, being careful not to overflow sample on planchet.
4. **RINSE** beaker or tube with ~2 mL of 0.05N HNO_3, **AND**

   TRANSFER to planchet.
5. **EVAPORATE** to dryness, **THEN**

   HEAT on hotplate for ~15 minutes longer.
6. **REMOVE** planchets **AND**

   ALLOW planchet to cool.
7. **RE-WEIGH** planchets, **AND**

   RECORD final gross weight on Analysis Request Sheet.
8. **CALCULATE** gravimetric recovery manually, or if available, by computer program or in LIMS as directed by CTF or supervision.
9. **IF** carrier recovery is *greater than* 110%, **THEN**

   REHEAT planchet on hot plate to ensure dryness.
10. **REMOVE** planchet, **AND**

    ALLOW planchet to cool.
11. **RE-WEIGH** carrier recovery.
12. **RECORD** planchet holder number on batch sheet.
13. **PLACE** planchet containing the sample in an *upright position* on top of planchet holder.
14. **PLACE** set of holders for batch in a planchet carrier.
15. **ENTER** the tare weights, gross weights and separation time into SQL*LIMS, **AND**

    PRINT the SQL*LIMS worklist.
16. **SEND** samples to the Count Room along with the labels and batch sheet.
17. **PROVIDE** count room with the printed SQL*LIMS worklist contaning the following information:
    - Date and start time of yttrium in-growth
    - Aliquots of samples used
    - Name, activity value, date of assay, and aliquot(s) of spike used
    - Strontium gravimetric recovery (for samples containing standardized strontium carrier)

5.6 Calculation of Gravimetric Recovery

CALCULATE gravimetric recovery based on final weight of strontium nitrate (the expected residual compound) on planchet.

NOTE: *The weight ratio of strontium in $Sr(NO_3)_2$ is 0.41404.*

% Recovery = 100 * [net $Sr(NO_3)_2$ g / Sr carrier added g] * 0.41404
where net $Sr(NO_3)_2$ = Gross weight – tare weight

Examples:
- 100 * [0.0145g / 0.0065 g] * 0.41404 = 92.36%

6.0 POST PERFORMANCE ACTIVITIES

6.1 Waste Handling and Disposal

1. **DISPOSE** of solid rad low-level waste per BAL-ADMIN-0005.
2. **DISPOSE** of Green-Is-Clean per BAL-ADMIN-0010.
3. **DISPOSE** of liquid waste per procedure BAL-ADMIN-0013.
4. **IF** disposal method is unknown, **THEN**

   CONSULT ECA and GCO before disposing of liquid or solid.
5. **OBTAIN** GCO approval before disposing of soil in rad low-level waste.

6.2 Waste Minimization

NOTE: *Do not dispose of soil or soil mixtures in 735-B drains. Contact the GCO and ECA if soil disposal is required.*

1. **CHARACTERIZE** and **DISPOSE** of waste streams properly from a regulatory and environmental perspective.
2. **LABEL** and **DISPOSE** radioactive waste per BAL-ADMIN-0005 and BAL-ADMIN-0013 for drain disposal of liquids.

7.0 RECORDS

Final reports and raw data shall be maintained per the Site Retention Schedule Matrix, WSRC-EM-96-00023, and Manual 1B, MRP 3.3l, Records Management.

8.0 REFERENCES

- AHA LAB-2549
- L3.21-10005, Operating Instructions For The Alpha Spectrometry System
- L3.23-40000, EBL General Safety Rules
- L3.23-6000, Preparation of Nitric Acid Solution
- BAL-ADMIN-0013, Drain System Guidelines
- BAL-ADMIN-0005, Low Level Waste
- BAL-ADMIN-0010, Green-Is-Clean
- BAL-ADMIN-0012, BAL Chemical Management System Guidelines
- BAL-ADM-0016, BAL Spill Management Program
- Horwitz, E.P. et al., 1993. Separation and preconcentration of actinides from acidic media by extraction chromatography. Analytica Chimica Acta, 281, 361-372
- Horwitz, E.P. et al., 1992. Separation and preconcentration of uranium from acidic media by extraction chromatography. Analytica Chimica Acta, 266, 25-37
- Horwitz, E. Philip; Dietz, M.L.; Chiarizia, R., Diamond; H., Maxwell, S.L.; Nelson, M.R. 1995, Analytica Chimica Acta, vol. 310, 63-78
- Maxwell III, S.L.; Fauth, D.J. , 2000, Radioactivity and Radiochemistry, vol. 11, No 3, 28-24
- Maxwell III, S.L. Rapid Actinide Analysis for Large Soil Samples, 50th Annual Radiochemical and Radiobioassay Measurements Conference, Cincinnati, OH, Nov -2004
- Radioanalytical Methods in Interdisciplinary Research: Fundamentals in Cutting Edge Applications: "Chapter 18: Rapid Actinide Column Extraction Methods for Bioassay Samples", American Chemical Society Symposium
- Manual 4Q, Procedure 111, “First Response Measures for Hydrogen Fluoride (HF) Skin Contact”

9.0 ATTACHMENTS

Appendix 1 Derivation of Sr-89/90 Equations with Y-90 in-growth (8 Pages)

Appendix 1 Sr-89/90 Equations with Y-90 in-growth to differentiate Sr-89 and Sr-90
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These calcuations are based on the normal equations for radioactive decay known as the Bateman Equations. The Bateman Equations are the set of coupled differential equations that express the amounts of reactants and products as a function of time as these species are undergoing both production and loss by nuclear reaction and radioactive decay.

Some primary references used in the development of these calculations are:

Derivation of Sr-89/90 Equations with Y-90 In-growth, SRS document, OBU-OLS-2004-00041
25 October 2004, Brian K. Culligan

Chase and Rabinowitz, **Principles and Radioisotope Methodology**, 3rd Edition, Burgess Publishing, 1967.

B. C. Henderson, *Technical Overview of Strontium Counting with Gas Flow Proportional Counters*, ESH-EM-20011002, 29 June 2001.

W. B. Bowman, et. al., "Radiochemical Determination or Sr-89 and Sr-90: A Comparison of Methods Based on Error Analysis", *Health Physics*, Pergamon Press, 1976, Vol. 31 (December), pp. 495-500.

ASTM Method D 5811-95, Standard Test Method for Sr-90 in Water

ASTM Method D 5811-00, Standard Test Method for Sr-90 in Water

EPA-600 / 4-76-011, March 1976, *Measurement of Sr-89 and Sr-90 in Environmental Waters, A Tentative Reference Method.*

John R. Taylor, **An Introduction to Error Analysis**, 2nd Ed., University Science Books, 1997.

Derivation:
From the general Bateman Equation for a Parent-Daughter relationship (resulting in Secular equilibrium):

$$N_2 = N_{2o}e^{-\lambda 2\Delta t} + [(N_{1o}\lambda_1)/(\lambda_2-\lambda_1)](e^{-\lambda 1\Delta t} - e^{-\lambda 2\Delta t}) \quad [1]$$

Where:

N_1 = Atoms of Sr-90 at time t
N_2 = Atoms of Y-90 at time t
λ_1 = Decay constant Sr-90, min^{-1}
λ_2 = Decay constant Y-90, min^{-1}
N_{1o} = Initial # of Sr-90 atoms
N_{2o} = Initial # of Y-90 atoms
Δt = Elapsed Time (min)

At separation (t = 0), there is no Y-90 present ($N_{2o}e^{-\lambda 2\Delta t} \rightarrow 0$) and Equation [1] reduces to:

$$N_2 = [(N_{1o}\lambda_1)/(\lambda_2-\lambda_1)](e^{-\lambda 1\Delta t} - e^{-\lambda 2\Delta t}) \quad [2]$$

Let A = the decay rate (activity) of a radioactive species so that:

$$A = \lambda N$$

And,

$$N_1 = A_1/\lambda_1, \qquad N_2 = A_2/\lambda_2 \quad [3]$$

Where:

A_1 = Decay rate (activity) of Sr-90, dpm
A_2 = Decay rate (activity) of Y-90, dpm

Appendix 1 Sr-89/90 Equations with Y-90 in-growth to differentiate Sr-89 and Sr-90
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Substitute [3] into [2]:

$$A_2/\lambda_2 = [(A_{1o}\lambda_1/\lambda_1)/(\lambda_2-\lambda_1)](\, e^{-\lambda 1\Delta t} - e^{-\lambda 2\Delta t})$$

Canceling λ_1/λ_1 and solving for A_2:

$$A_2 = [(A_{1o}\lambda_2)/(\lambda_2-\lambda_1)](\, e^{-\lambda 1\Delta t} - e^{-\lambda 2\Delta t}) \qquad [4]$$

Since $(\lambda_2-\lambda_1) \approx \lambda_2$:

$$A_2 = A_{1o}(\, e^{-\lambda 1\Delta t} - e^{-\lambda 2\Delta t})$$

For a small Δt after separation (recall exponent rule $a^0 = 1$):

$$(e^{-\lambda 1\Delta t} - e^{-\lambda 2\Delta t}) \approx 1 - e^{-\lambda 2\Delta t}$$

Thus:

$$A_2 = A_{1o}(1 - e^{-\lambda 2\Delta t}) \qquad [5]$$

The observed Count Rate (CR, in counts per minute, cpm) can be expressed as:

$CR_1 = E_{Sr90}A_1$ for Sr-90
$CR_2 = E_{Y90}A_2$ for Y-90

Where:

E_{Sr90} = counting efficiency for Sr-90
E_{Y90} = counting efficiency for Y-90

Combining these two equations with [5] (since $A_1 = CR_1/E_{Sr90}$ and $A_2 = CR_2/E_{Y90}$) gives:

$$CR_2/E_{Y90} = (CR_1/E_{Sr90})(1 - e^{-\lambda 2\Delta t})$$

The TOTAL Count Rate of any sample at time t is defined as:

$$CR_T = CR_1 + CR_2 = E_{Sr90}A_1 + E_{Y90}A_2$$

And, using [5]:

$$CR_T = E_{Sr90}A_1 + E_{Y90}A_1(1 - e^{-\lambda 2\Delta t})$$

Recall that $CR_1 = E_{Sr90}A_1$ (and $A_1 = CR_1/E_{Sr90}$). Substituting for $E_{Sr90}A_1$ and A_1 yields:

$$CR_T = CR_1 + (E_{Y90})(CR_1/E_{Sr90})(1 - e^{-\lambda 2\Delta t})$$

Factoring CR_1 gives the observed count rate for Sr-90 + Y-90:

$$CR_T = [1 + (E_{Y90}/E_{Sr90})(1 - e^{-\lambda 2\Delta t})]CR_1$$

Add counts due to the presence of any Sr-89, independent of Sr-90 and Y-90 gives:

$$CR_3 = CR_{3o}(e^{-\lambda 3\Delta t})$$

Where:

λ_3 = Decay constant for Sr-89, min^{-1}
CR_{3o} = Initial count rate of Sr-89, cpm

thus,

$$CR_T = [1 + (E_{Y90}/E_{Sr90})(1 - e^{-\lambda 2\Delta t})]CR_1 + (e^{-\lambda 3\Delta t})\, CR_3 \qquad [6]$$

Appendix 1 Sr-89/90 Equations with Y-90 in-growth to differentiate Sr-89 and Sr-90
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This linear equation in two unknowns can be solved using simultaneous equations. To evaluate this equation, the sample must be counted at two widely separated times to ensure sufficient Y-90 in-growth (approximately 7 to 14 days). The OXFORD LB5100 will provide the net, background and crosstalk corrected count rate ($INST_{cps}$) for each of these two counts.

First set:

$[1+ (E_{Y90}/E_{Sr90})(1- e^{-\lambda 2\Delta t})]$ = D1 or D2; Sr-90 decay and Y-90 in-growth factors at times t1 and t2
$(e^{-\lambda 3\Delta t})$ = B1 or B2; Sr-89 decay factors at t1 and t2
CR_1 = X, cpm Sr-90, net
CR_3 = Z, cpm Sr-89, net
CR_T = C1 or C2; observed net count rates, cpm at t1 and t2, where ($C1 = INST1_{cps}*60$, $C2 = INST2_{cps}*60$)

and,

INST1cps = First Count instrument result in cps
INST2cps = Second Count instrument result in cps

Then set up the two equations:

$$C1 = D1X + B1Z$$
$$C2 = D2X + B2Z$$

First, solve C1 for Z:

$$Z = (C1-D1X)/B1 = \text{Sr-89 cpm, net} \qquad [7]$$

Then substitute for Z in C2:

$$C2 = D2X + B2(C1-D1X)/B1$$

And solve for X:

$$C2B1 = D2B1X + B2C1 – D1B2X$$

$$C2B1 – B2C1 = X(D2B1 – D1B2)$$

$$X = (C2B1 – B2C1)/(D2B1 – D1B2) = \text{Sr-90 cpm, net} \qquad [8]$$

Calculate X (cpm Sr-90, net) from the results of the two counts then calculate Z (cpm Sr-89, net) using X (from [8]) in equation [7].

Appendix 1 Sr-89/90 Equations with Y-90 in-growth to differentiate Sr-89 and Sr-90
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Special Case: No Sr-89 present
Assuming there is no Sr-89 present in the sample (based on its half-life of 50.5 days), the total count rate from Sr-90 and Y-90 in-growth is:

$$CR_T = [1+ (E_{Y90}/ESr90)(1- e^{-\lambda 2\Delta t})]CR_1 \quad [9]$$

When Δt is large (> 14 days, approaching secular equilibrium):

$$e^{-\lambda 2\Delta t} \rightarrow 0$$

and

$$CR_T = [1+ (E_{Y90}/ESr90)(1- 0)]CR_1$$

$$CR_T = [1+ (E_{Y90}/ESr90)]CR_1 \quad [10]$$

According to B. C. Henderson, a value for E_{Y90}/E_{Sr90}, of 1.145, was determined empirically and is used in the software, yielding:

$$CR_T = [1+ (1.145)]CR_1$$

$$CR_T = (2.145)CR_1$$

The uncertainty in this empirical value for E_{Y90}/E_{Sr90} is not determined herein or used in subsequent error propagation.

If the counting efficiencies of Sr-90 and Y-90 are equal then $E_{Sr90} = E_{Y90}$ and Equation [10] would reduce to the expected value for the Sr-90 / Y-90 pair in secular equilibrium:

$$CR_T = 2CR_1$$

Immediately after separation only Sr-90 is present and decaying ($\Delta t = 0$). Equation [9] reduces to:

$$CR_T = CR_1$$

Appendix 1 Sr-89/90 Equations with Y-90 in-growth to differentiate Sr-89 and Sr-90
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Final Equations to determine Sr-90 and Sr-89 in pCi/g:

To calculate the concentration of the strontium isotopes from experimental data using the decay and in-growth equations derived above, terms for background current, chemical recovery, and volume of sample taken have been incorporated into the final equations:

$$Sr90\ (pCi/g) = (X_{net})/(2.22RVE_{Sr90}) \quad [11]$$

$$Sr89\ (pCi/g) = (Z_{net})/(2.22RVE_{Sr89}) \quad [12]$$

Where:

X = net Sr-90 cpm as determined above from simultaneous equations
Z = net Sr-89 cpm as determined above from simultaneous equations
R = fractional chemical recovery (gravimetric or radiometric)
W = Sample weight, grams
E_{Sr90} = Counting Efficiency for Sr-90
E_{Sr89} = Counting Efficiency for Sr-89
The constant 2.22 has units of dpm/pCi

If no Sr-89 is present, use single count to measure CR_T and determine Sr-90 (CR_1) from:

$$CR_T = [1+ (E_{Y90}/E_{Sr90})(1- e^{-\lambda 2\Delta t})]CR_1$$

And set CR_1 = X and use equation [11] to calculate Sr-90 in pCi/g.

Appendix 1 Sr-89/90 Equations with Y-90 in-growth to differentiate Sr-89 and Sr-90
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ERROR PROPAGATION:

The general formula for propagation of a function of multiple independent random variables:

$\gamma = f(x,y,z)$, $\sigma_\gamma = [(\delta f/\delta x)^2\sigma_x^2 + (\delta f/\delta y)^2\sigma_y^2 + (\delta f/\delta z)^2\sigma_z^2]^{1/2}$ [13]

where $\delta f/\delta x$ is the partial derivative of f with respect to x holding all other variables constant.

Defined Constants and Variables:

Constants:

λ_{Sr90} = 4.5445 E-8 min^{-1}
λ_{Y90} = 1.8023 E-4 min^{-1}
λ_{Sr89} = 9.53 E-6 min^{-1}

Variables (Independent and Random):

Recovery	σ_R	=	$\pm$ 5% relative	(1σ)
Weight	σ_V	=	$\pm$ 1% relative	(1σ)
Efficiency	σ_E	=	$\pm$ 5% relative	(1σ)
Sample error (cpm),	σ_X	=	$\pm (X_{s,\,gross}/t_s)^{1/2}$	(1σ)
Background error (cpm),	σ_b	=	$\pm (b/t_b)^{1/2}$	(1σ)

(Sigma for Sample and Background Error are derived from standard counting statistics functions).

The total Function to be propagated incorporates (X_{gross} – b) to account for a background (b) error term:

Sr90 (pCi/g) = $(X_{gross}-b)/(2.22RVE_{Sr90})$

Letting ($X_{gross} = X_{net} + b$), where X_{net} (cpm) is determined in equation [8] above, and the background, b is provided by the instrument (b must be multiplied by 60 to convert cps to cpm).

The Total Propagated Uncertainty (TPU) can be determined by taking the partial derivative of the function

$f = (X_{gross} - b)/(2.22RVE_{Sr90})$

for each of the variables, R, W, E, X, and b:

$\delta f/\delta R = -(1/R^2)(X-b)/(2.22VE_{Sr90}) = -(1/R)$(Sr90 pCi/g)
$\delta f/\delta W = -(1/W^2)(X-b)/(2.22RE_{Sr90}) = -(1/W)$(Sr90 pCi/g)
$\delta f/\delta E = -(1/E_{Sr90}^2)(X-b)/(2.22RW) = -(1/E_{Sr90})$(Sr90 pCi/g)
$\delta f/\delta X = 1/(2.22RWE_{Sr90})$
$\delta f/\delta b = -1/(2.22RWE_{Sr90})$

The total propagated error, using equation [13] is:

$$\sigma_{TPU} = [(\delta f/\delta R)^2\sigma_R^2 + (\delta f/\delta W)^2\sigma_W^2 + (\delta f/\delta E)^2\sigma_E^2 + (\delta f/\delta X)^2\sigma_X^2 + (\delta f/\delta b)^2\sigma_b^2]^{1/2}$$

This equation can be simulated using an Excel® spreadsheet to determine the TPU for various values of R, σ_R, W, σ_V, E, σ_E, X, σ_x. b, and σ_b.

Test Equations (Hand Calculation vs. Spreadsheet, ✔ indicates match):

Set					
	R	=	0.8	$\pm$ 0.04	(5% at 1 sigma)
	W	=	0.5g	$\pm$ 0.005g	(1% at 1 sigma)
	E	=	0.5	$\pm$ 0.025	(5% at 1 sigma)
	$X_{cpm\ gross\ Sr90}$	=	10 cpm	$\pm (X/t_s)^{0.5}$	
	b_{cpm}	=	2 cpm	$\pm (b/t_b)^{0.5}$	
	t_s	=	20 min.	$\pm$ negligible uncertainty	
	t_b	=	20 min.	$\pm$ negligible uncertainty	

Appendix 1 Sr-89/90 Equations with Y-90 in-growth to differentiate Sr-89 and Sr-90
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Sr90 pCi/L = (10-2)/(2.22*0.8*0.5*0.5)
= 18.02 pCi/g ✔

MDC = $[2.71 + 4.65((b(t_b))^{1/2}] / 2.22*R*W*E*t_s$
= 3.617 pCi/g ✔

$$\sigma_{TPU} = [(\delta f/\delta R)^2\sigma_R^2 + (\delta f/\delta W)^2\sigma_W^2 + (\delta f/\delta E)^2\sigma_E^2 + (\delta f/\delta X)^2\sigma_X^2 + (\delta f/\delta b)^2\sigma_b^2]^{1/2}$$

$$\sigma_{TPU} = [(0.8116) + (0.0325) + (0.8116) + (2.5363) + (.5073)]^{1/2}$$

$\sigma_{TPU} = \pm 2.1678$ pCi/g ✔

The σ_X value used in this error propagation does not include any uncertainty contributed by correcting the total counts of Sr-90, X, for Y-90 in-growth or Y-90 counting efficiency before calculating the Sr-90 concentration. This contribution is negligible when the Δt since separation is small, i.e., when the correction for Y-90 in-growth is insignificant. This uncertainty increases as Δt increases. Uncertainty contributed by assigning any fraction of the total activity to Sr-89 is also ignored in this error propagation, until it is added in the last step of the Total Uncertainty calculation.

Single Count Sr-90 with Y-90 In-growth:

Assuming only Sr-90 $\rightarrow$ Y-90 and single efficiency (i.e., no significant difference between E_{Sr90} and E_{Y90}):

$$\% \text{ Y In-growth} = 100\,(1 - e^{-\lambda Y90 \Delta t})$$

where Δt is the time since separation (in minutes) and λ_{Y90} = 1.8023 E-4 min^{-1}. If Δt is near 0, then $e^{-\lambda Y90 \Delta t} \rightarrow 1$ and % Y90 In-growth is insignificant. This is the ideal situation but may not, at times, be practical. An Excel® spreadsheet can be constructed to determine the % In-growth provided the time since separation is known.

Example:

Δt	Hand	Spreadsheet	Decay Calculator
240 min	4.233%	4.233%	4.232%

The small (< 0.02%) difference between the hand / spreadsheet calculation and the Decay Calculator (Desktop Software Application) is due to slight differences in the decay constant used in the calculation.

Appendix 1 Sr-89/90 Equations with Y-90 in-growth to differentiate Sr-89 and Sr-90
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At any time after separation, the activity due to Y-90 is given by:

$$A_{Y90} = (A_{Sr90})(\%Y_{in}/100)$$

Total Activity follows:

$$A_T = A_{Sr90} + A_{Y90}$$

$$A_T = A_{Sr90} + (A_{Sr90})(\%Y_{in}/100)$$

$$A_T = A_{Sr90} [\, 1 + (\%Y_{in}/100)]$$

At any time after separation:

$$A_{Sr90} = A_T / [\, 1 + (\%Y_{in}/100)]$$

It is a simple matter to measure the total activity, the Δt, and, assuming no Sr-89 or other β^- emitters, calculate the Sr-90 fraction of the total activity.

Option: Calculate the TPU by adding errors in quadrature:

$$\sigma_{quad,\%} = 100*[(\sigma_R/R)^2 + (\sigma_V/W)^2 + (\sigma_E/E)^2 + (\%CE/100)^2]^{1/2}$$

See spreadsheet for calculation based on the above.

To get the 2 sigma error, simply take the 1 sigma error as calculated in the spreadsheet and multiply by 2. The $\sigma_{TPU,\%}$ for the propagated function gives values equal to the % error as the $\sigma_{quad,\%}$.