Applications of Eichrom Resins to Savannah River Site High Activity Waste Measurements

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Nuclear Measurements Group/Analytical Development Section

Eichrom UGM RRMC 2017
Savannah River Site High Level Waste Flowsheet

- F-Canyon
- SRS Production Reactors
  - No longer in service
- H-Canyon
- DWPF
- Glass Canisters to Yucca Mountain
- SRR Production Reactors
- Waste Receipt & Storage Tanks
- 2F Evaporator System
- Feed Staging Tanks
- Recycle
- 2H Evaporator System
- Feed Staging Tanks
- 3H Evaporator System
- Feed Staging Tanks
- Interim Processes
- ARP
- MCU
- Decontaminated Salt Solution
- Saltstone Processing Facility
- Saltstone Vault
- Spent MST & Cs stream
- Spent MST from ARP & Cs stream from MCU
- Saltstone Vault

We put science to work.™
F and H Tank Farms

Two tank farms
49 waste tanks
  • 22 “old-style” tanks
  • 27 “new-style” tanks

Approximately 37 million gallons of waste

Salt Supernate
- 16.9 Mgal
- 212 MCi
- 7.8E18 Bq

Saltcake
- 16.6 Mgal
- 3.0 Mgal

Sludge
- 185 MCi
- 6.9E18 Bq

Dominant chemical & radiological constituents
- Sodium nitrate
- Sodium nitrite
- Sodium hydroxide
- Sodium aluminate
- Cs-137

- Crystallized sodium nitrate
- Cs-137

- Metal (Fe, Mn, Al) oxides & hydroxides
- Sr-90, actinides
SRS Laboratories Conducting Radiochemical Analyses

Savannah River National Laboratory

Science and Technology Division

Analytical Laboratory Department

National Security Division

Non-Proliferation Technology

Ultra Low Level Radioactive Matrices for Forensics/Non-Proliferation

Analytical Development Section

F&H Lab (24/7)
- High Radioactive Matrices
- H-Canyon/B-Line Process Support
- SRR Process Support
- High Activity Forensic Measurements

SRNS/ESS&H

B Area Lab
- Environmental Radioactive Matrices
- Environmental Monitoring
- Bioassay

Nuclear Measurements Group
- R&D Activities Radiochemistry Support
- Site Process Support for Activities not Covered by ALD Laboratories
- High Activity Forensic Measurements
- Hold-up/NDA in-situ SRS measurements
Nuclear Measurements Organization

Radiochemistry

• 1 PhD Nuclear Chemist
• 2 PhD Nuclear Engineers
• 1 MS Radiochemist
• 1 MS Biochemist
• 4 BS Chemists
• 1 Specialist
• 1 Laboratory Technician

Nuclear field measurements

• 1 PhD Nuclear Chemist
• 1 Specialist
• Support as needed from Radiochemistry Personnel
• In last 12 months lost 2 PhD Nuclear Scientists to retirement
Radiochemistry Preparation Laboratories

• 5 Laboratory Modules
  – 2 Chemical Hoods
  – 3 Gloveboxes
    • Ability to work with up to 400 grams Plutonium
  – 10 Radiological Hoods, 3 Radiobenches
    • Routinely work with samples containing up to $1E+10$ dpm ($1.67E+8$ Bq) alpha and beta
    • Routinely work with samples having up to $10$ mRem/h (0.1 mSv/h) whole body dose @ 30cm
    • Routinely work with samples having up to $2000$ mRem/h (20 mSv/h) contact dose

• SRNL Shielded Cells Facility utilized for initial separation steps of high-activity samples
Nuclear Measurement Counting Instrumentation

- 4 counting rooms contain nuclear measurement instrumentation
- In addition, the radiochemistry group leverages additional ADS instrumentation for radiochemical analyses
  - i.e. ICP-MS, ICP-AES

Beta Spectrometry - Triple, double, single PMT LSC counters, cosmic suppressed LSC, Portable beta spectrometers, conversion electron spectrometer, GFPCs, beta PIPS

Alpha Spectrometry
~100 alpha PIPS + portable alpha spectrometers

Gamma Spectrometry
- 15 shielded spectrometers ranging from planar to coaxial to well HPGe
- 4 automated systems
- Numerous field deployable x-ray and gamma-ray spectrophotometers
- Calibrations generated with NIST traceable standards, Canberra LABSOCS/ISOCS, or with Customized MCNP Models
Neutron Activation Analysis Facilities

- Supports radiological tracer production (i.e. Tc-99m)
- Supports radiochemical separation tracer recovery measurements
  - Iodine, selenium, strontium, samarium
- ~2mg (decayed from ~60mg in 2003) Cf-252 generates ~1E7 n/s/cm² thermal neutron flux
- Pneumatic system allows for repeated irradiations
- System being replaced with an Adelphi D-D neutron generator with ~5E7 n/s/cm² thermal neutron flux
AD Tasked to Conduct Radiological Characterizations on SRS Waste Tanks Slated for Closure

- 6 Tanks Characterized to date
  - Tank 19F in 2009
  - Tank 18F in 2009
  - Tank 5F in 2011
  - Tank 6F in 2011
  - Tank 16H, primary and annulus in 2013
  - Tank 12H in 2015

- Residues are highly radioactive, and vary from Tank to Tank (up to 1.3E9 Bq/g beta, 1.7E7 Bq/g alpha)

- Requested to analyze for trace radionuclides (to as low as 0.37 Bq/g in some cases) in the presence of gross levels of interfering radionuclides

- Large list of analytes large (up to 54 radioisotopes), requested for up to ~40 samples of Tank Waste

- Short turnaround time for the entire effort from development to completion (~5 months)
Tank Closure Campaigns

Tasked to conduct radiological characterizations on SRS Waste Tanks slated for closure
- Waste tanks slated for closure have been mechanically or chemically cleaned
- Residues are highly radioactive, as high as 1.3E9 Bq/g Beta, 1.7E7 Bq/g Alpha
- Required analyses for trace radionuclides (as low as 0.37 Bq/g) in the presence of gross levels of interfering radionuclides
- Large list of analytes requested for numerous samples of Tank Waste (up to 40 in recent campaigns)
  - Cs-137 is the main contributor to whole body dose
  - Sr-90/Y-90 main contributor to Extremity Dose
  - Radiochemical separations run much more efficiently in radiohoods as opposed to the Shielded Cells
Tank 19 & 18  54 Radio-isotopes Requiring Characterization

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Every Waste Tank often has unique challenges even for routine analyses

Target typically to measure down to the 0.37 Bq/g neighborhood, Tank Waste in the 2.5E7 Bq/g activity range

Question becomes how many analytes will actually be present at much higher levels (makes for a much easier analysis), and how many will require procedures to get down to the 0.37 Bq/g levels, and then, can we even do it in this time frame.
54 Radio-isotopes’ Origins

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From Fission Products

From Activation Products

Natural
Fission Product Distribution Over Time

Fraction of Radioactivity

- $^{90}$Sr/Y
- $^{137}$Cs
- $^{147}$Pm
- $^{79}$Se
- $^{135}$Cs
- $^{126}$Sn
- $^{154}$Eu
- $^{137}$Cs
- $^{129}$I
- $^{151}$Sm
- $^{99}$Tc
- $^{107}$Pd

Age of SRS Tank Sludge Waste

20 years
## Activation Products

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- Was the precursor present to be exposed to a neutron flux to generate levels having current activities >0.37 Bq/g?
  - i.e. H-2(n,γ)H-3, or N-14(n,p)C-14 Probably so
  - i.e. Cl-35(n,γ)Cl-36 or Pt-192(n,γ)Pt-193m Probably not
# Looking at Isotopes on the List That Use Eichrom Resins

Isotopes in blue make use of Eichrom Products

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Sr-90/Y-90

Peroxide Fusion Digestion in Shielded Cells

For HLW supernate we do 2 Sr extractions, for HLW sludge we do 1

Eichrom Sr resin Extraction

Neutron Activation Analysis to determine Sr Carrier yields

Liquid Scintillation Counting for Sr-90

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Savannah River National Laboratory
Operated by Savannah River Nuclear Solutions
We put science to work.™
Pu-238, 239, 240, 241, 242, 244

Peroxide Fusion Digestion in Shielded Cells

Thenoyltrifluoroacetone (TTA) solvent-solvent extraction for Pu

For Pu-238, and Pu-239/240, traced with Pu-236

Eichrom TEVA resin Extraction

Eichrom TEVA resin Extraction

ICP-MS for Pu-239, 240, 242, 244

Liquid Scintillation Counting for Pu-241 to Pu Alpha ratio

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Tc-99

Pressurized Mixed Acid Digestion in Shielded Cells

Eichrom TEVA resin Extraction

Eichrom TEVA filter Extraction

3x3 NaI Well gamma spectrometer for Tc-99m

Tc-99m Tracer Generation

Mo-99 + n > Tc-99m

MIIK extraction of Tc-99m tracer

Liquid Scintillation Counting for Tc-99
Th-229, Th-230, Ac-227

- Peroxide Fusion Digestion in Shielded Cells
- Eichrom TEVA resin Extraction
- Cerium Co-Precipitation
- ICP-MS for Th-232 among other things

Occasionally unexpectedly high in Thorium

- We yield from Th-229 or Th-228 tracers or from Th-232 measured from ICP-MS
- Calculate Ac-227 from measured Th-227

Alpha Spectrometry for Th-227, 229 and 230
Am-241, 242m, 243, Cm-242, 243, 244, 245, 248, Cf-249

Peroxide Fusion Digestion in Shielded Cells

Eichrom RE resin extraction of Trivalent actinides in Shielded Cells

Gamma and or Cesium removed gamma analysis for Am-241 or Am-243

2 week cool to let Y-90 decay

Eichrom Ln resin extraction of Trivalent actinides in radiohoods

ICP-MS for Am-241, Cm-244, Cm-245, Cm-247, Cm-248

Alpha Spectrometry for Am-241, Am-243, Cm-242/Am-242m, Cm-244

Gamma Spectrometry for Am-241, Am-243, Cm-243, Cm-245, Cf-249

Alpha Spectrometry for Th-227, 229 and 230
U-232, U-233, U-234, U-235, U-236, U-238

Peroxide Fusion Digestion in Shielded Cells

Eichrom U-TEVA resin Extraction

Cerium Co-Precipitation

ICP-MS for U-235, U-238 among other isotopes

Eichrom U-TEVA resin Extraction

Eichrom U-TEVA resin Extraction

ICP-MS for U-233, U-234, U-236

Alpha Spectrometry for U-232, traced with U-233 or ratioed to U-238 measured by ICP-MS
Pa-231

- Peroxide Fusion Digestion in Shielded Cells
- Samples spiked with Pa-233 (from Np-237) to trace separation
- Cesium removal with AMP
- Eichrom TEVA decontamination step
- Eichrom RE Extraction
- Gamma Analysis for Pa-233 tracer
- ICP-MS for Pa-231
Ni-59/63

Pressurized Mixed Acid Digestion in Shielded Cells

Eichrom Ni resin Extraction

Bio-RAD AMP Strike

Eichrom Ni resin Extraction

Liquid Scintillation Counting for Ni-63

Be windowed Semi-planar HPGe for Ni-59

ICP-AES for Ni Carrier Recovery

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Pt-193m

- Peroxide Fusion Digestion in Shielded Cells
- Pt carrier added for tracing
- Bio-RAD AMP Strike
- Eichrom TEVA resin Extraction
- Bio-RAD AMP Strike
- Eichrom TEVA resin Extraction
- Be-windowed Semi-planar HPGe for Pt-193m
- ICP-AES for Pt Carrier Recovery
Pd-107

Peroxide Fusion Digestion in Shielded Cells

Eichrom Ni resin Extraction

Spike aliquot with stable Pd if measurable stable Pd hasn’t already been measured

Eichrom Ni resin Extraction

ICP-MS for Pd-107 and Pd Carrier Recovery
Pm-147/Sm-151

Peroxide Fusion Digestion in Shielded Cells → Eichrom RE resin Extraction

Eichrom Ln resin Extraction

Liquid Scintillation Counting for Sm-151 and Pm-147

Neutron Activation Analysis to determine Sm Carrier yields
Acknowledgments

I would like to acknowledge the members of the Nuclear Measurement Team

- Robin Young
- Ceci DiPrete
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- Tim Aucott
- Alex Brand
- Travis Deason
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