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Measurement of Radio-isotopic Impurities in Purified Solutions of Ra-226 and Ac-227

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Radio-Isotopic Impurities in Purified Solutions of Ra-226 and Ac-227

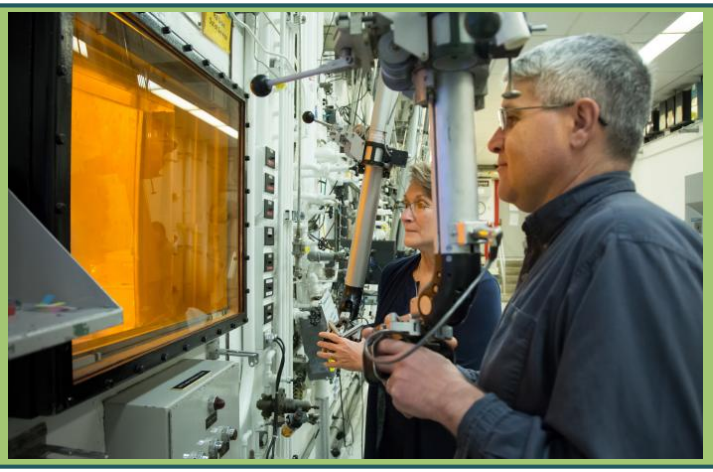
- Production of Ac-227 at ORNL
- Radio-isotopic impurities in Ra-226 & Ac-227
- Method development for detection of Sr-90 & Pb-210
  - Adjustments of separation procedure on Sr resin
  - Develop liquid scintillation protocol for Pb-210
- Method development for detection of Po-210
  - Adjustment of detection methods
  - Implement separation procedure on Sr resin

Production of Ac-227 at ORNL

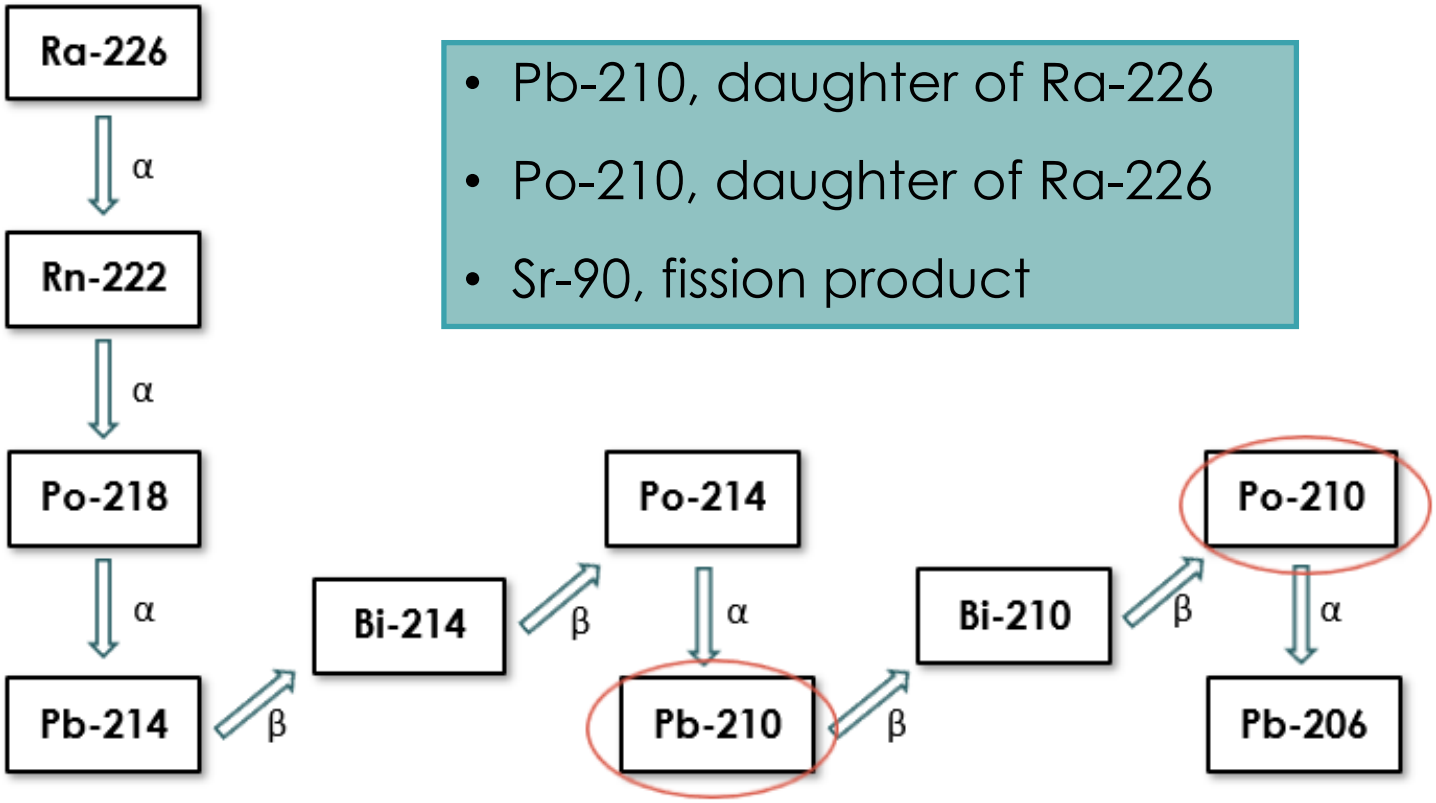
- ORNL has been approved as a qualified supplier of Ac-227 to meet demands of the cancer drug Xofigo®

- Ra-226 targets are prepared from legacy medical devices and then irradiated in the High Flux Isotope Reactor

- The irradiated targets are dissolved and Ac-227 is chemically separated and purified by the Nuclear & Radiochemistry group at ORNL



Radio-Isotopic Impurities

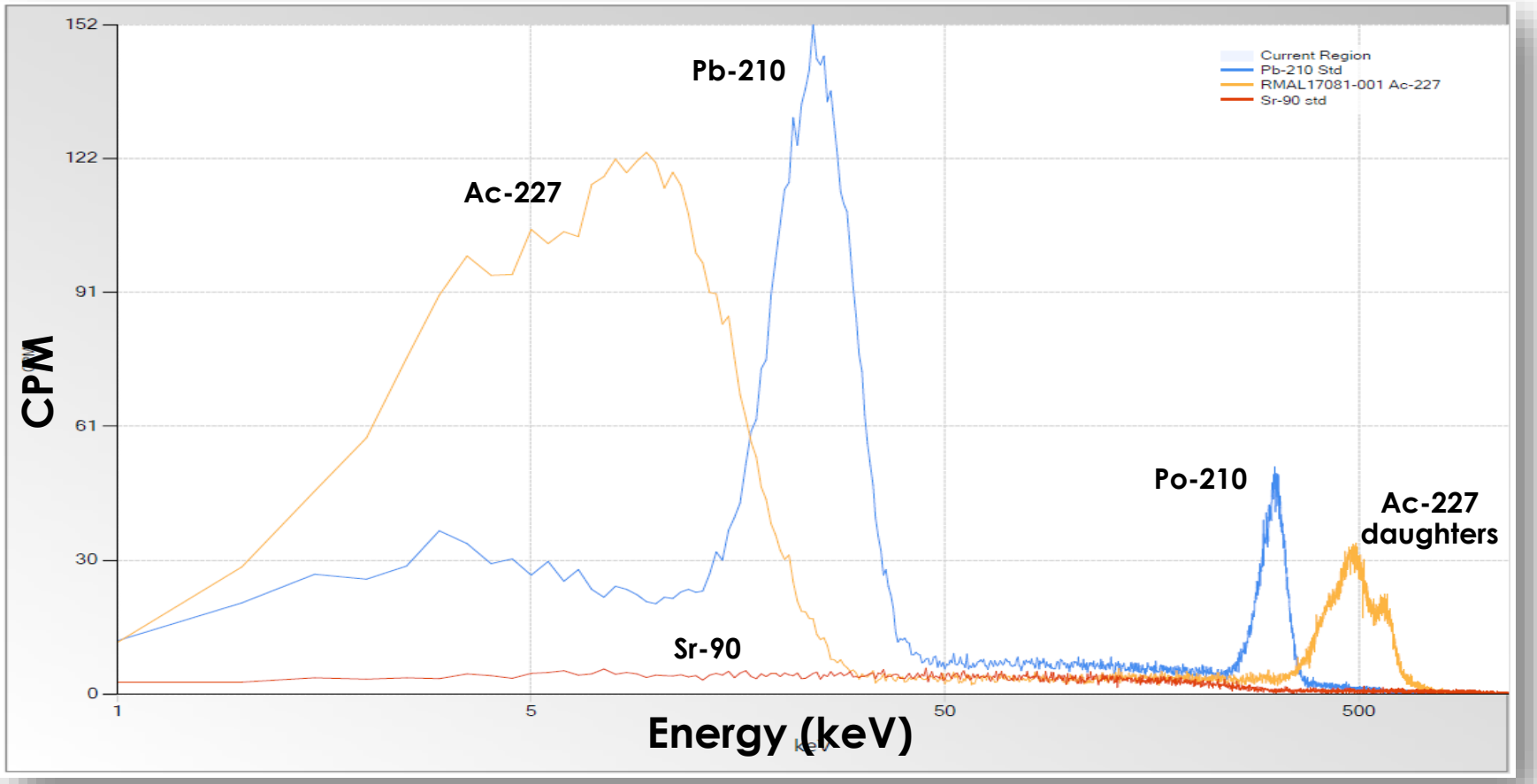


- Pb-210, daughter of Ra-226
- Po-210, daughter of Ra-226
- Sr-90, fission product

- To pass Bayer specifications for the COA, isotopic impurities need to be <0.1% activity of specified isotope (Ac-227 or Ra-226)
- RMAL had to develop and qualify methods for Sr-90, Pb-210 & Po-210

Radio-Isotopic Impurities (cont.)

'Quantify' Pb-210, Sr-90 and Po-210 using liquid scintillation



Separations are needed!

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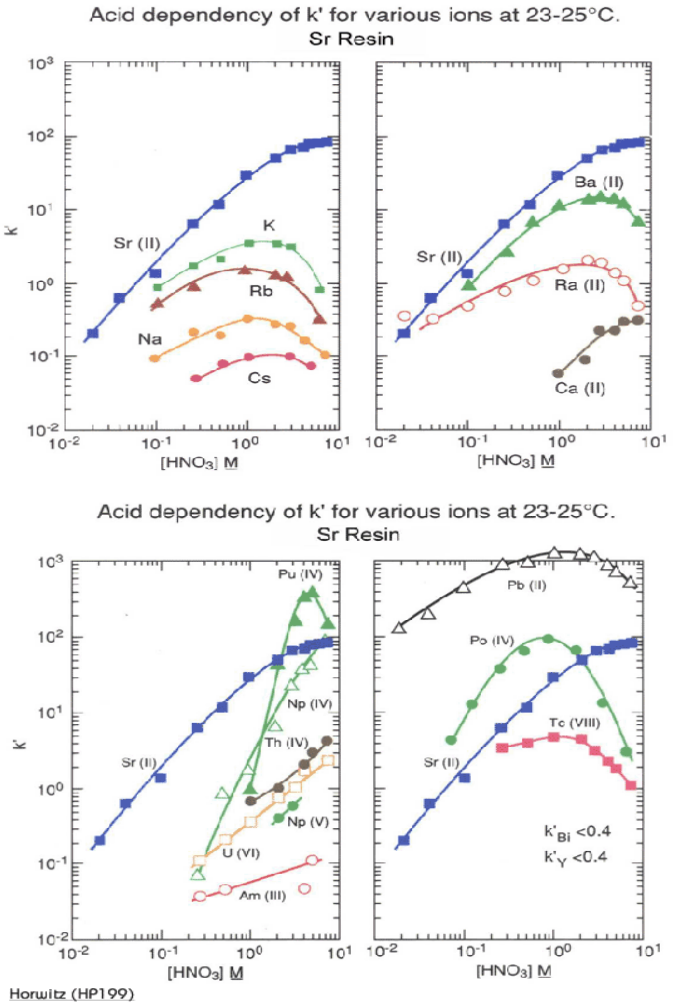
Sr-90 & Pb-210 Initial Separation Procedure

- Modified existing Sr-90 procedure to elute Sr-90 & Pb-210 in one separation

Step	Volume + Reagent
Condition Column	10 mL 8M HNO_3
Sample Load to Column	10 mL 8M HNO_3
Rinse 1	10 mL 8M HNO_3
Rinse 2	5 mL 1M HNO_3
Sr Strip	10 mL 0.05 M HNO_3
Pb Strip	10 mL 0.05M Ammonium Citrate

Sr-90 detection = gas flow proportional counting	Pb-210 detection = liquid scintillation counting



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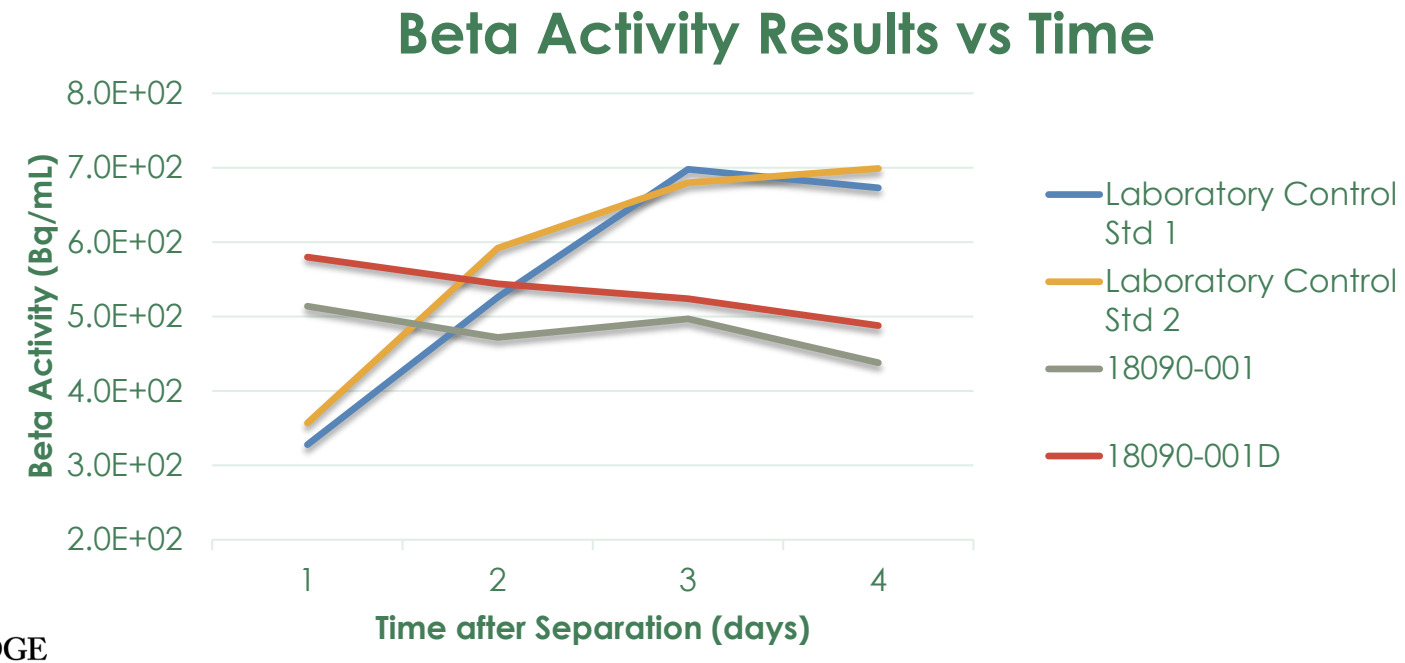
Sr-90 & Pb-210 Initial Results

- Elevated Sr-90 results in a Ra-226 batch
- Recycled radium, larger quantities of Ra-228
- Detected Pb-214 and Bi-214 via gamma spec
  - Short-lived daughters of Ra-226



Modified Sr-90 & Pb-210 Separation Method

Based on communications with Eichrom/Dan McAlister

Step	Volume + Reagent
Condition Column	10 mL 8M HNO_3
Sample Load to Column	10 mL 8M HNO_3
Rinse 1	5 mL 8M HNO_3
Rinse 2	10 mL 3M HNO_3-0.05M oxalic acid
Rinse 3	5 mL 8M HNO_3
Rinse 4	5 mL 1M HNO_3
Sr Strip	10 mL 0.05 M HNO_3
Pb Strip	10 mL 0.05M Ammonium Citrate

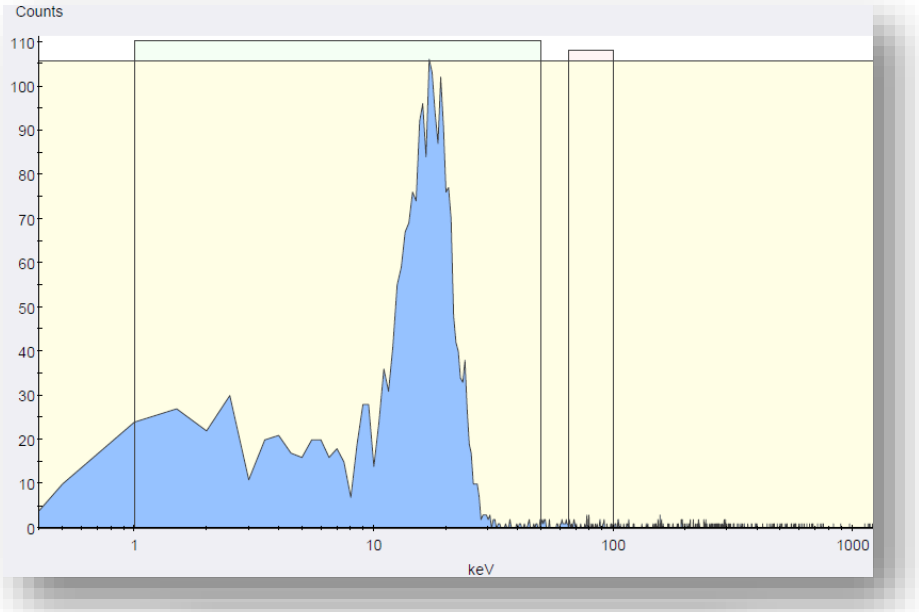
Sr-90 detection = gas flow proportional counting

Pb-210 detection = liquid scintillation counting

→ Separation procedure currently used for samples

Improvements for Pb-210 Detection

- Developed liquid scintillation protocol for Pb-210 opposed to quantifying via Bi-210 ingrowth
  - Experimentally determined Pb-210 efficiency
  - Sample matrix always the same
- Added stable Pb carrier for chemical recovery (0.1mg/mL): ICP-MS



Liquid scintillation analyzer, Quantulus GCT 6220

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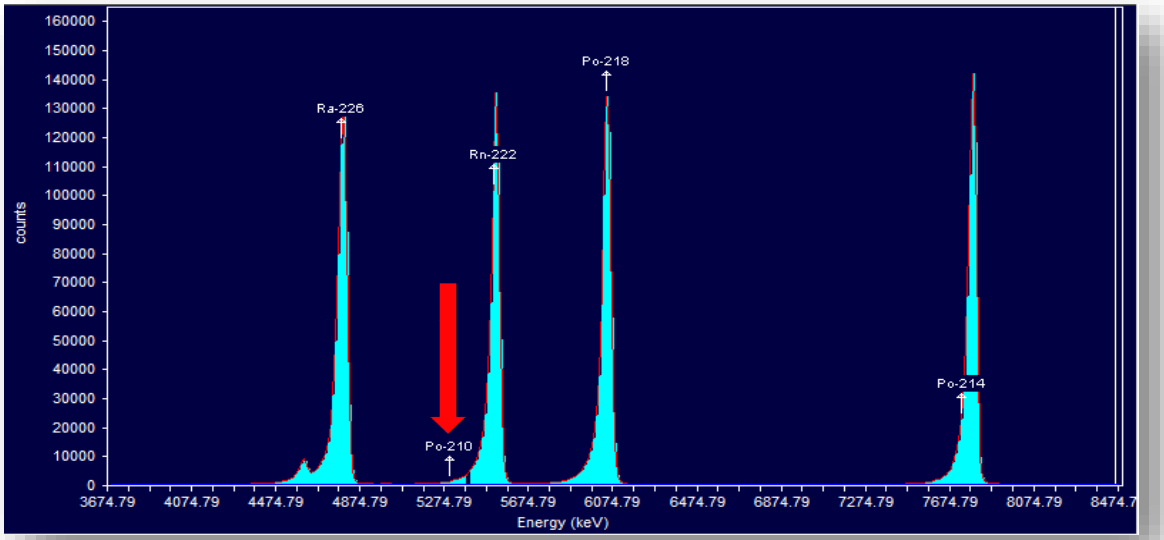
QC Results for Modified Sr-90 and Pb-210 Method

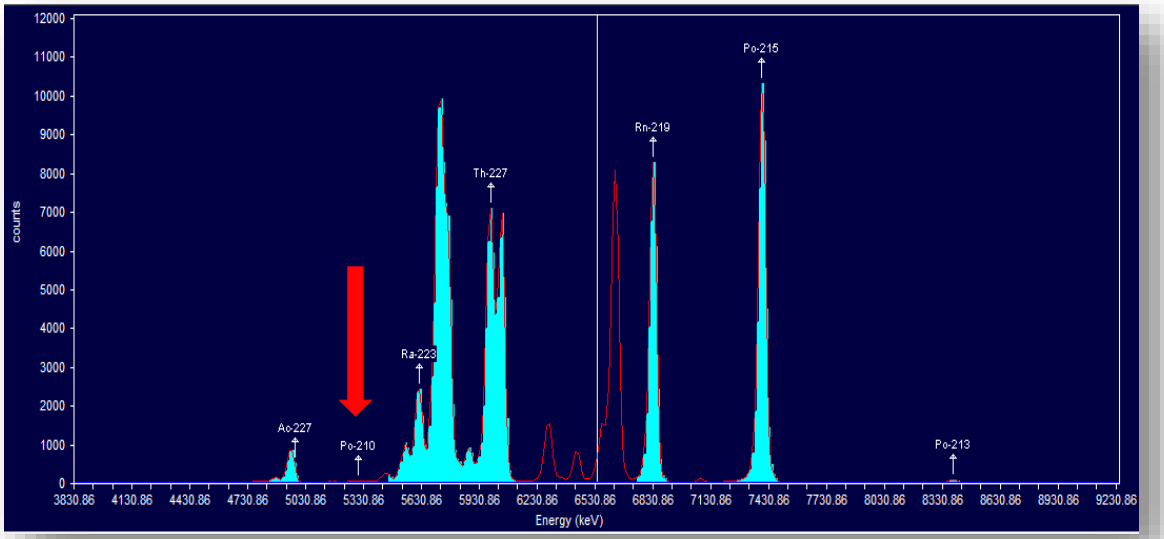
Sample Type	Carrier	% Carrier Recovery	LCS	% LCS Recovery
Ra-226 (N=7)	Stable Sr	88 ± 22	Sr-90	116 ± 12
Ra-226 (N=7)	---	---	Pb-210	76 ± 2
Ac-227/225 (N=9)	Stable Sr	90 ± 16	Sr-90	87 ± 9
Ac-227 (N=3)	Stable Pb	94 ± 4	Pb-210	89 ± 20

- RMAL QAP Requirements
  - Carrier: 40-110% yield
  - LCS: ± 25% deviation

Po-210 Method Development

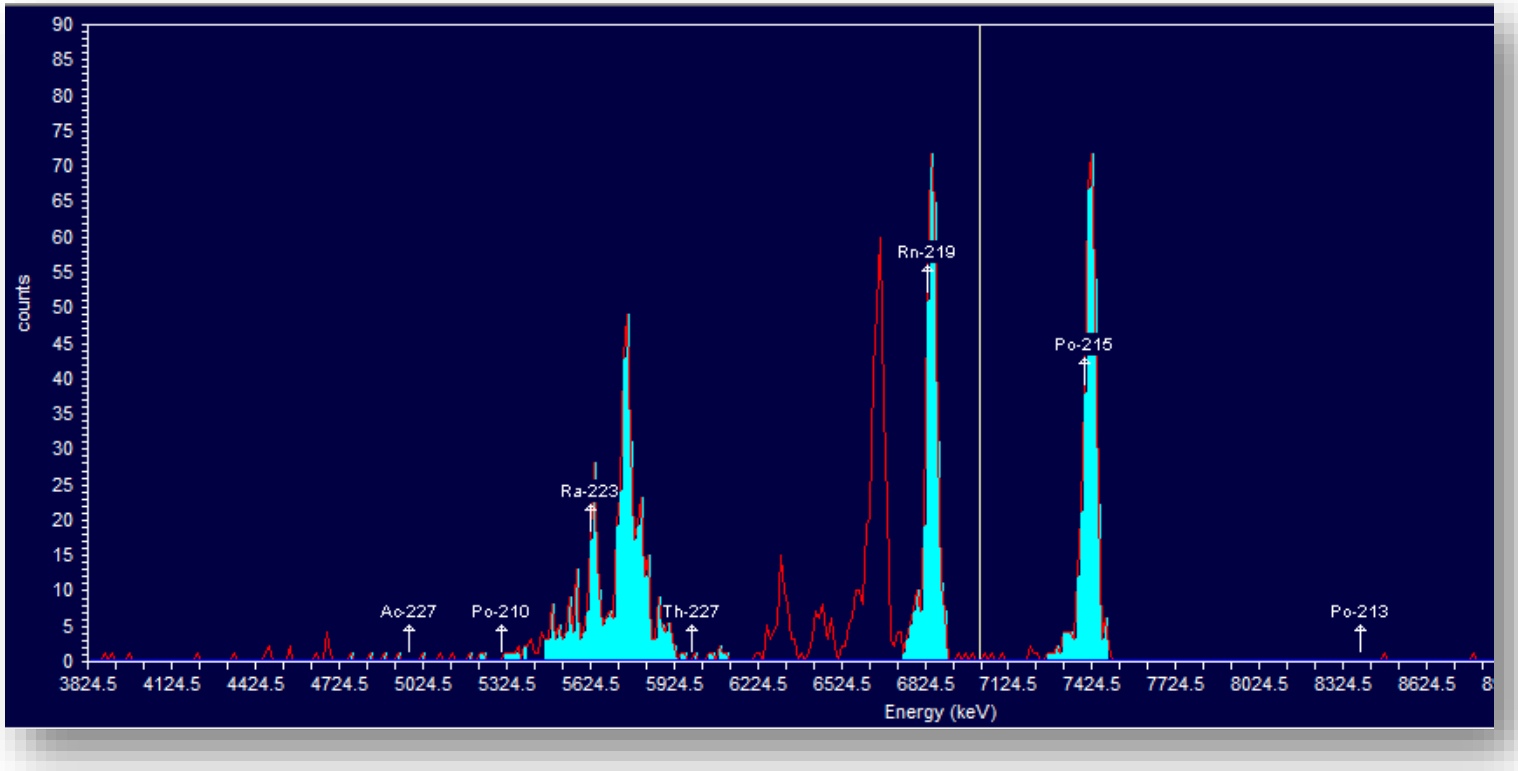
- No method in place
- Desired detection was alpha spectrometry
- Samples were counted as is, no separations





Po-210 Method Development (cont.)

Background spectrum one day after Ac-227 sample count

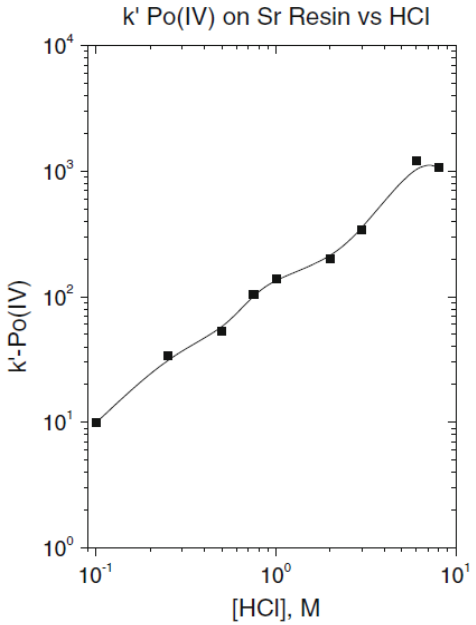


- Background contamination from Rn and daughters
- Alpha recoil contamination

Po-210 Separation Method

- Eichrom procedure for separation of Po-210 on Sr-resin

Step	Volume + Reagent
Condition Column	10 mL 2M HCl
Sample Load to Column	10 mL 2M HCl
Rinse 1	5 mL 2M HCl
Rinse 2	5 mL 2M HCl
Po Strip	5 mL 1M HN03
Po Strip	15 mL 0.1M HNO_3



Maxwell, S. J Radioanal. Nucl Chem (2013) 298:1977-1989

- Detection Method?
  - RMAL was not equipped for auto deposition source prep
  - Return to original requested analysis: **liquid scintillation**

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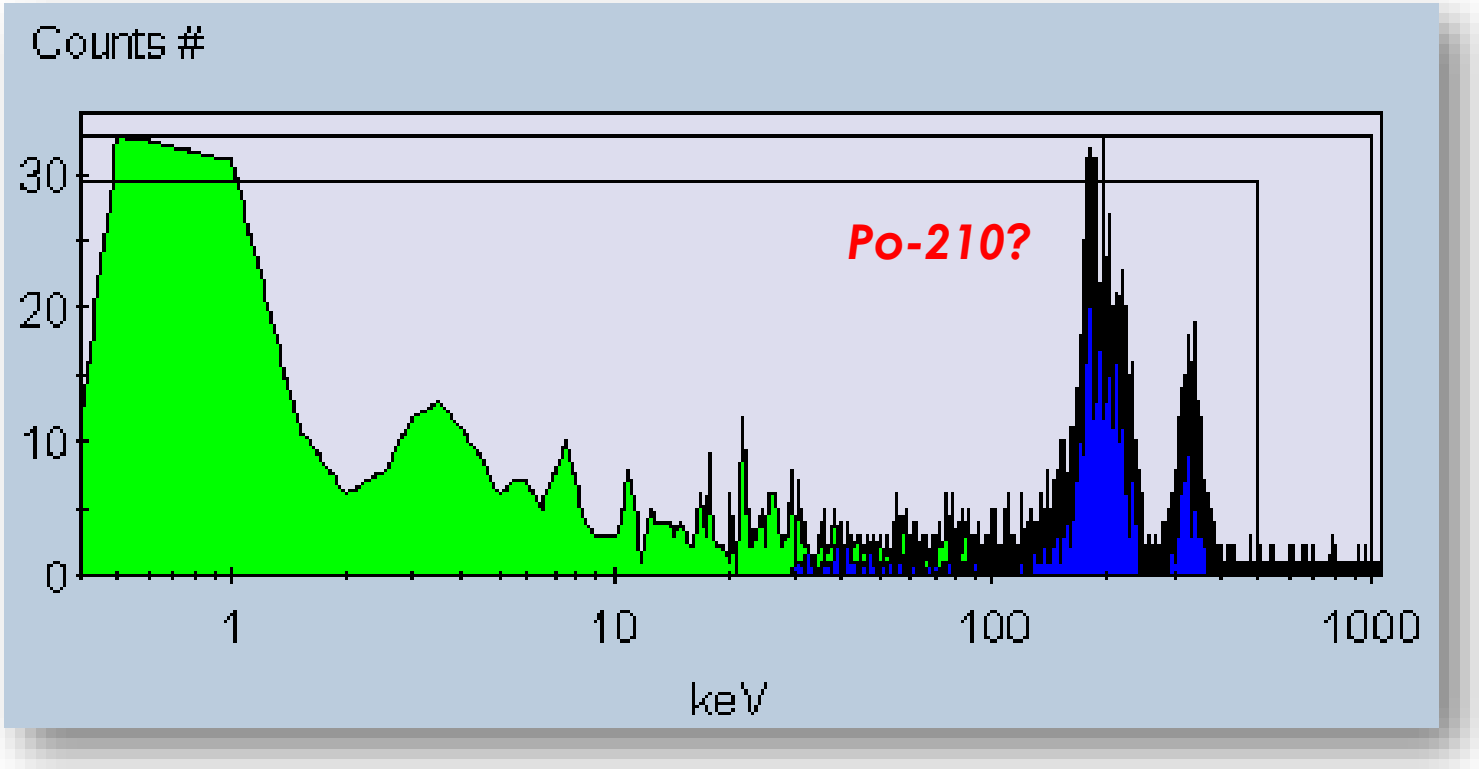
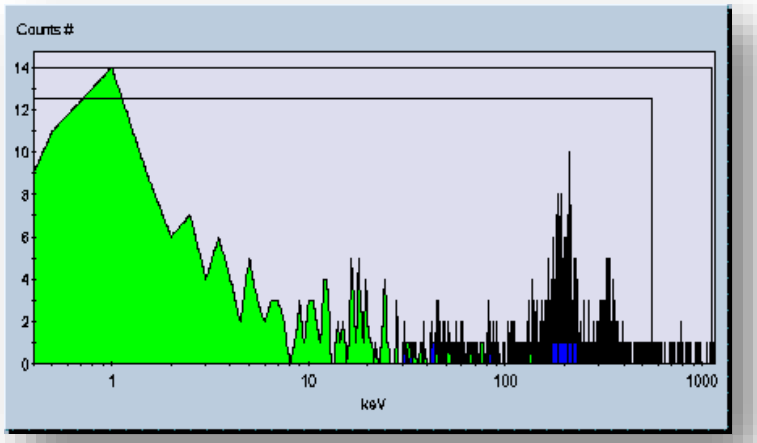
Po-210 By Liquid Scintillation

Po-210 LS spectrum of separated Ra-226



- Alpha/beta discriminator was utilized
- No distinction between alpha peaks

Po-210 By Liquid Scintillation (Cont.)



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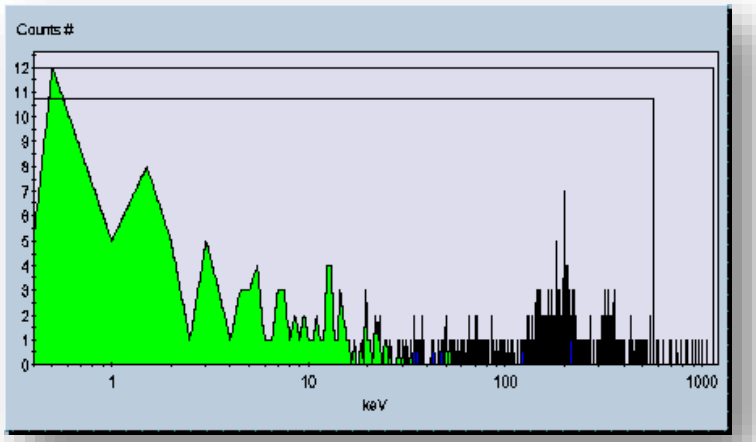
Po-210 LS spectra of separated Ra-226



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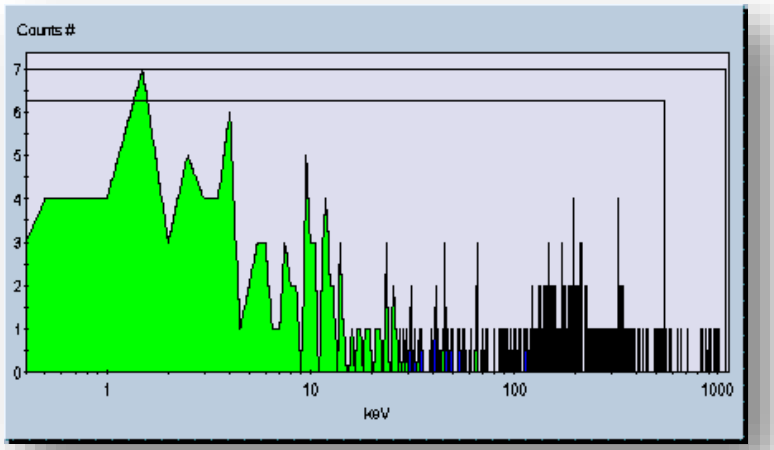

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Calculated ~3.5-6 day half-life

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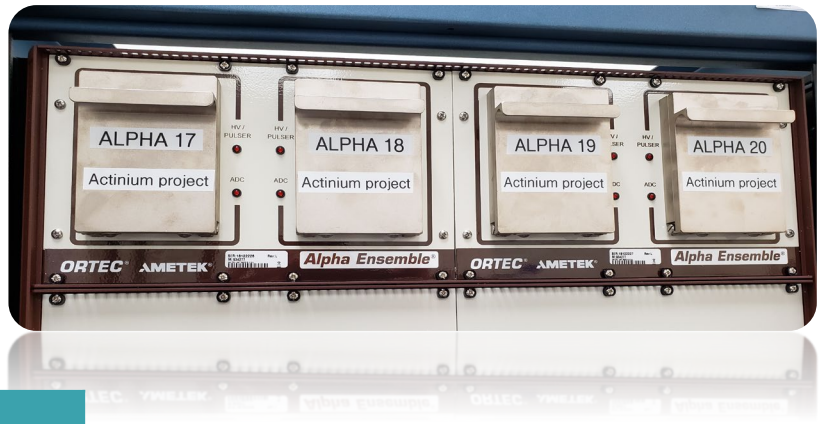
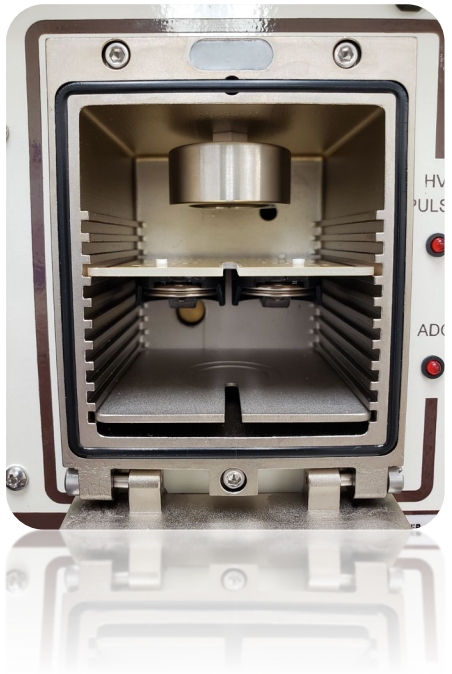
Bismuth Phosphate Microprecipitation

Switch methods, back to detection by alpha spec!

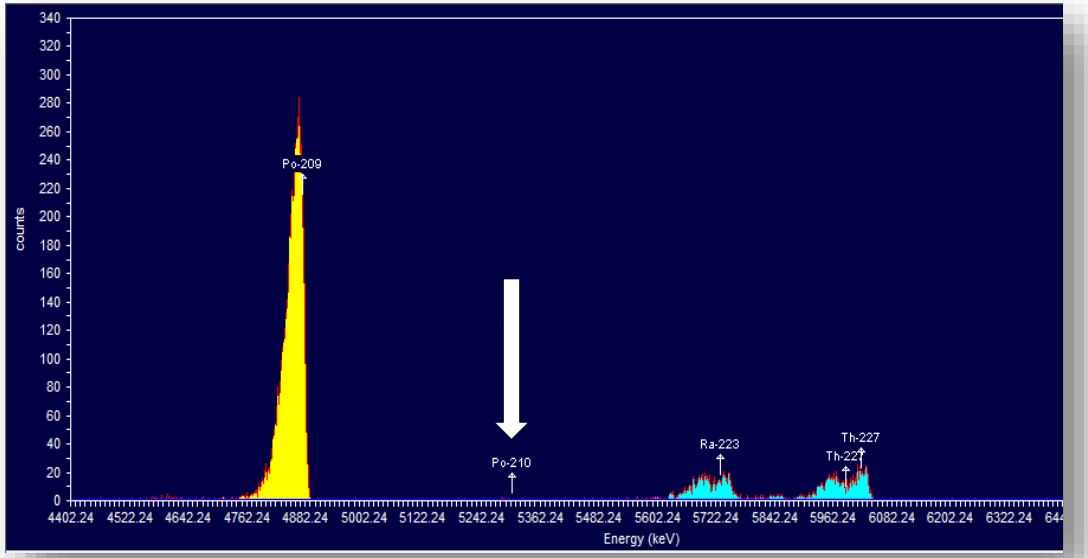


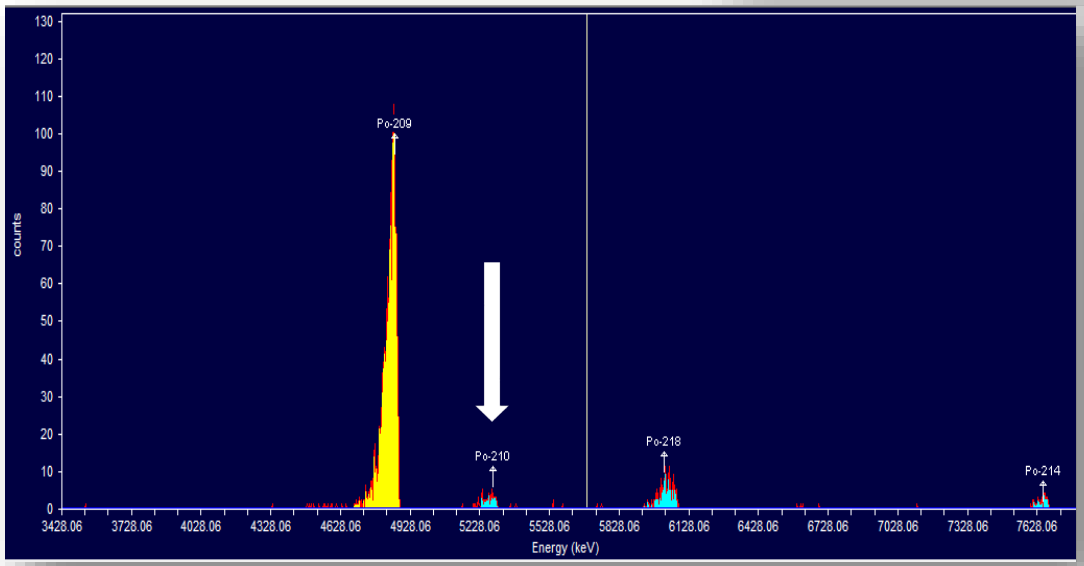
$BiPO_4$ Precipitation Procedure
1) 50 µL of H_20_2
2) 125 µL of bismuth carrier
3) 0.75 mL of $(NH_4)_2HPO_4$
4) 0.6 mL of conc. NH_4OH
5) Mix, wait ~30 minutes
6) Collect precipitate on 0.1 µm resolve filter



Po-210 By Alpha Spec: $BiPO_4$ Microprecipitation

Alpha spectrum of Po-210 post-separation in **Ac-227**





Alpha spectrum of Po-210 post-separation in **Ra-226**

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QC Results for Po-210 Method

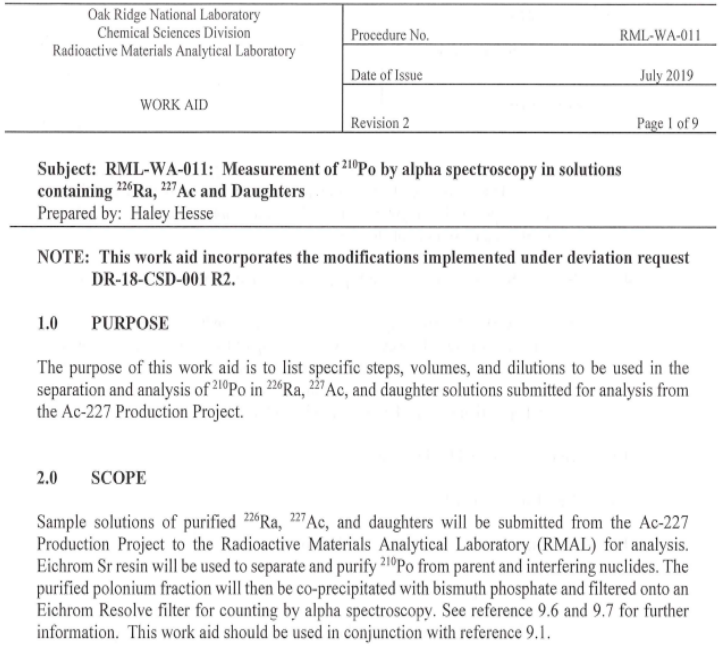
- **RMAL QAP Requirements**
  - **Tracer: 30-110% yield**
  - **Internal std: ± 40% deviation**

Sample Type	Tracer	% Tracer Recovery	Average FWHM
Ra-226 (N=15)	Po-209	67 ± 11	53 ± 13
Ac-227 (N=10)	Po-209	61 ± 13	47 ± 6

Sample Type	Internal Std	% Recovery
Ra-226 (N=3)	Po-210	92 ± 6
Ac-227/225 (N=6)	Po-210	88 ± 20

Summary

- Separations using Sr resin were needed to isolate impurities from high activity samples of Ra-226 & Ac-227
- Improved detection methods for Pb-210 and Po-210
- Passed strict qualifications implemented by Bayer
- Procedures were finalized and accepted
  - RMAL-WA-008
  - RMAL-WA-010
  - RMAL-WA-011



Oak Ridge National Laboratory
Chemical Sciences Division
Radioactive Materials Analytical Laboratory

WORK AID

Procedure No. RML-WA-011
Date of Issue July 2019
Revision 2 Page 1 of 9

Subject: RML-WA-011: Measurement of ^{210}Po by alpha spectroscopy in solutions containing ^{226}Ra, ^{227}Ac and Daughters
Prepared by: Haley Hesse

NOTE: This work aid incorporates the modifications implemented under deviation request DR-18-CSD-001 R2.

1.0 PURPOSE

The purpose of this work aid is to list specific steps, volumes, and dilutions to be used in the separation and analysis of ^{210}Po in ^{226}Ra, ^{227}Ac, and daughter solutions submitted for analysis from the Ac-227 Production Project.

2.0 SCOPE

Sample solutions of purified ^{226}Ra, ^{227}Ac, and daughters will be submitted from the Ac-227 Production Project to the Radioactive Materials Analytical Laboratory (RMAL) for analysis. Eichrom Sr resin will be used to separate and purify ^{210}Po from parent and interfering nuclides. The purified polonium fraction will then be co-precipitated with bismuth phosphate and filtered onto an Eichrom Resolve filter for counting by alpha spectroscopy. See reference 9.6 and 9.7 for further information. This work aid should be used in conjunction with reference 9.1.