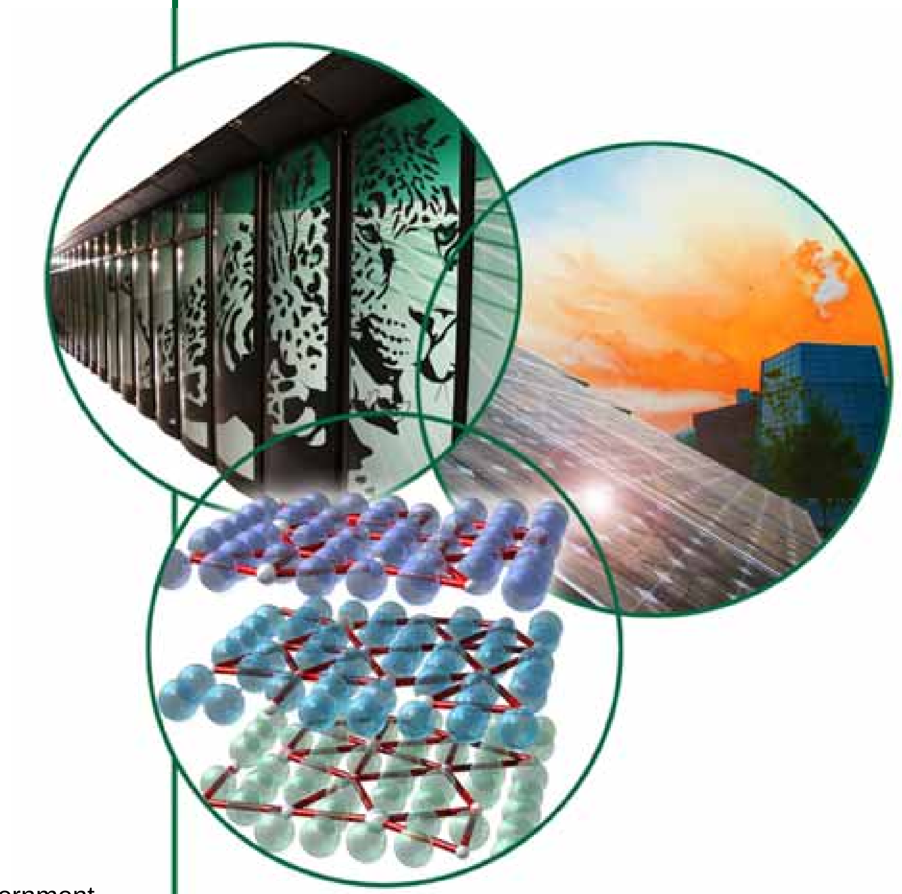
RAPID PREPARATION METHOD FOR THE MEASUREMENT OF TRACE U-232 IN HEU USING RESOLVE FILTERS

Giaquinto, J. M.; Partridge, J. D.

Oak Ridge National Laboratory
Chemical Science Division
Oak Ridge, Tennessee



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Presentation Overview

- Overview of the Expanded HEU Analyses project funded by the Department of Homeland Security (DHS)
- Project goals related to measurement of U-232 content in HEU
- Initial load tests of Resolve™ filters using depleted uranium.
- HEU actinide separation scheme
- Cerium fluoride precipitation procedure used for HEU samples
- Alpha spectrometry PHA measurements
- Merging U-232 alpha measurement into uranium isotopic data measured by MC-ICPMS
- Comparison of results and conclusions
- Acknowledgments

Overview of Expanded HEU Analyses

- Characterization of select analytes for age dating signatures and additional isotopes of interest to expand identifying information associated with HEU sources. HEU samples have a ^{235}U assay of approximately 93%.
  - Determination of ^{237}Np content
  - Determination of ^{232}U content
  - Age since chemical purification of HEU material using $^{234}U/^{230}Th$ chronometer
  - Age since chemical purification of HEU material using $^{235}U/^{231}Pa$ chronometer
  - Age since chemical purification of HEU material using $^{241}Pu/^{241}Am$ chronometer

Overall Project Goal

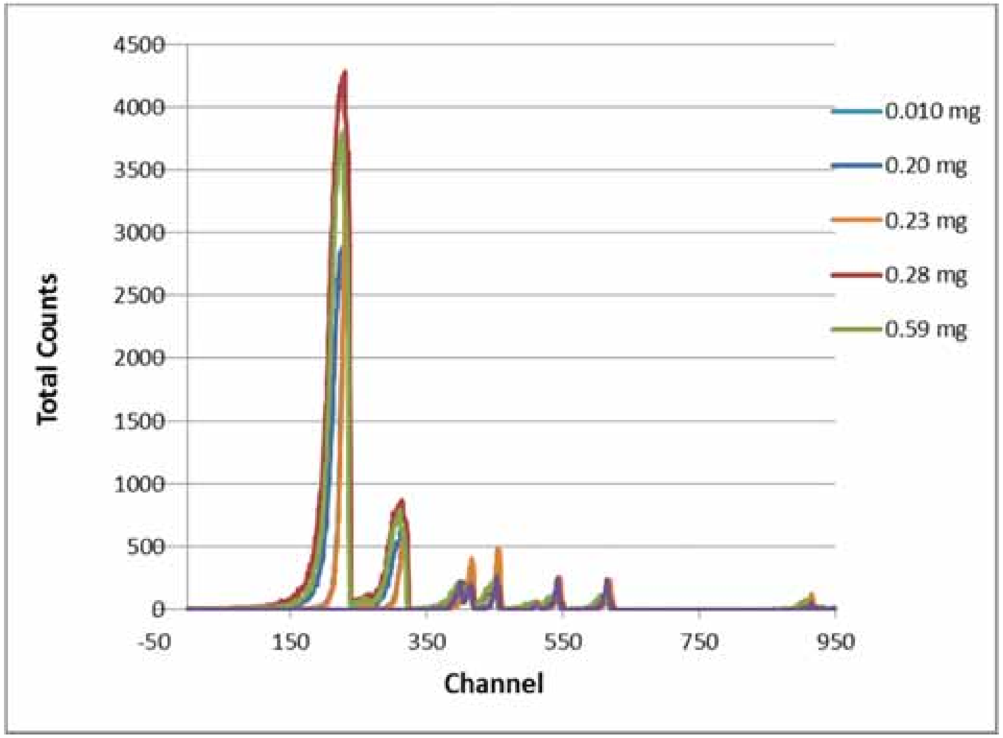
These expanded analyses encompassing age dating and additional nuclear isotopic signatures have the potential to discern discrete samples with analogous isotopic signatures by providing the dates for the last chemical processing of the SNM, as well as, irradiation history of the material.

Project Goals Relating to 232U

- U-232 in HEU is derived from decay of ^{236}Pu, commonly formed in reactors from irradiation of ^{232}Th and ^{235}U. The relative ^{232}U content formed provides an indication of the type of reactor from which the HEU was derived.
- Use solid phase extractions (SPE) (Eichrom resin cartridges) chemistry to purify thorium, uranium, plutonium, and americium from dissolved HEU solutions.
- Perform inter-laboratory comparison of ^{232}U content in HEU samples to demonstrate ORNL's micro-precipitation protocol.

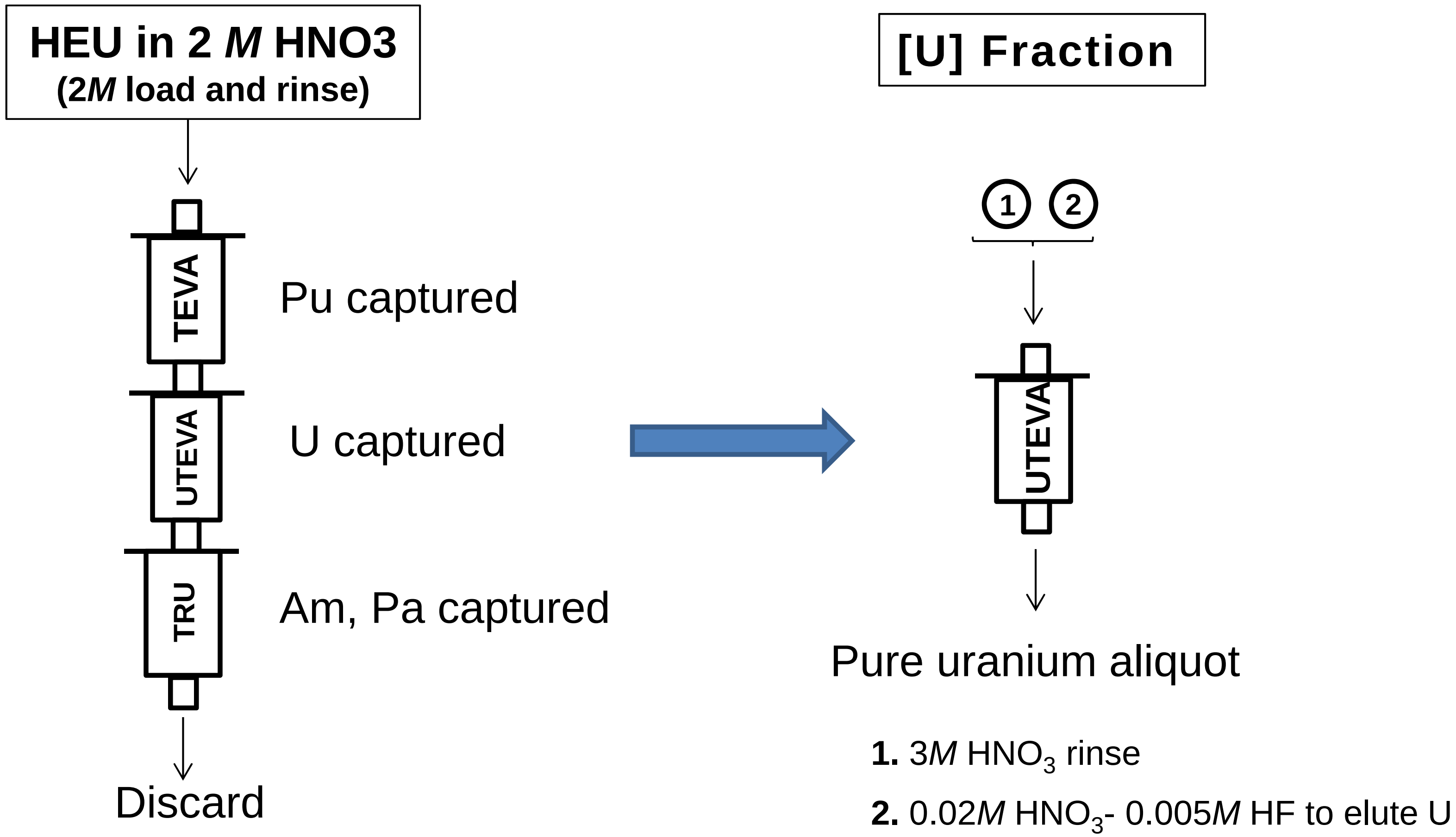
Initial Testing of Various Uranium Loadings

Spectrograph of uranium + daughters alpha peaks for various Resolve™ filter loadings.



Attenuation and peak broadening was evident, but was minimal and did not affect the overall alpha spectra after a 3 day count using a Canberra Alpha Analyst.

HEU Actinide Separation Scheme

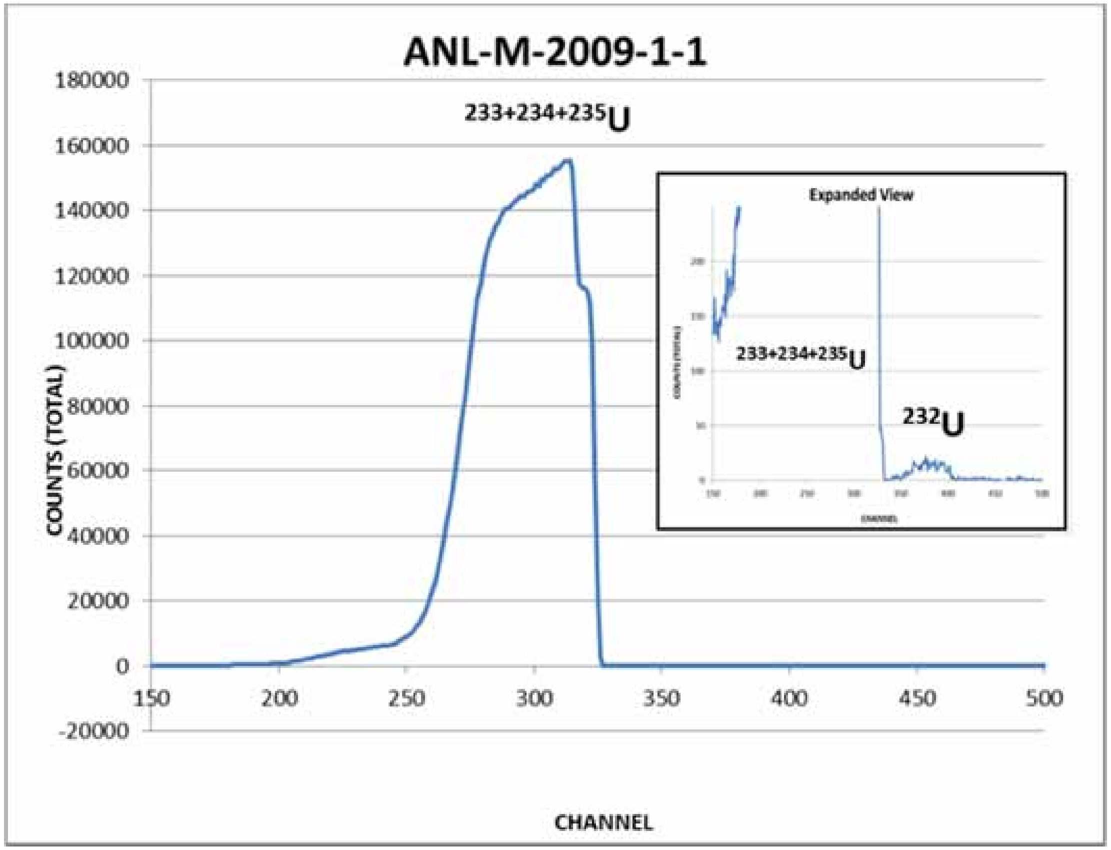


The goal of the separation scheme was to isolate the plutonium on the TEVA cartridge; collect the majority of the uranium (~3 mg) onto UTEVA free from decay daughters; and, finally, capture the americium and protactinium on TRU resin.

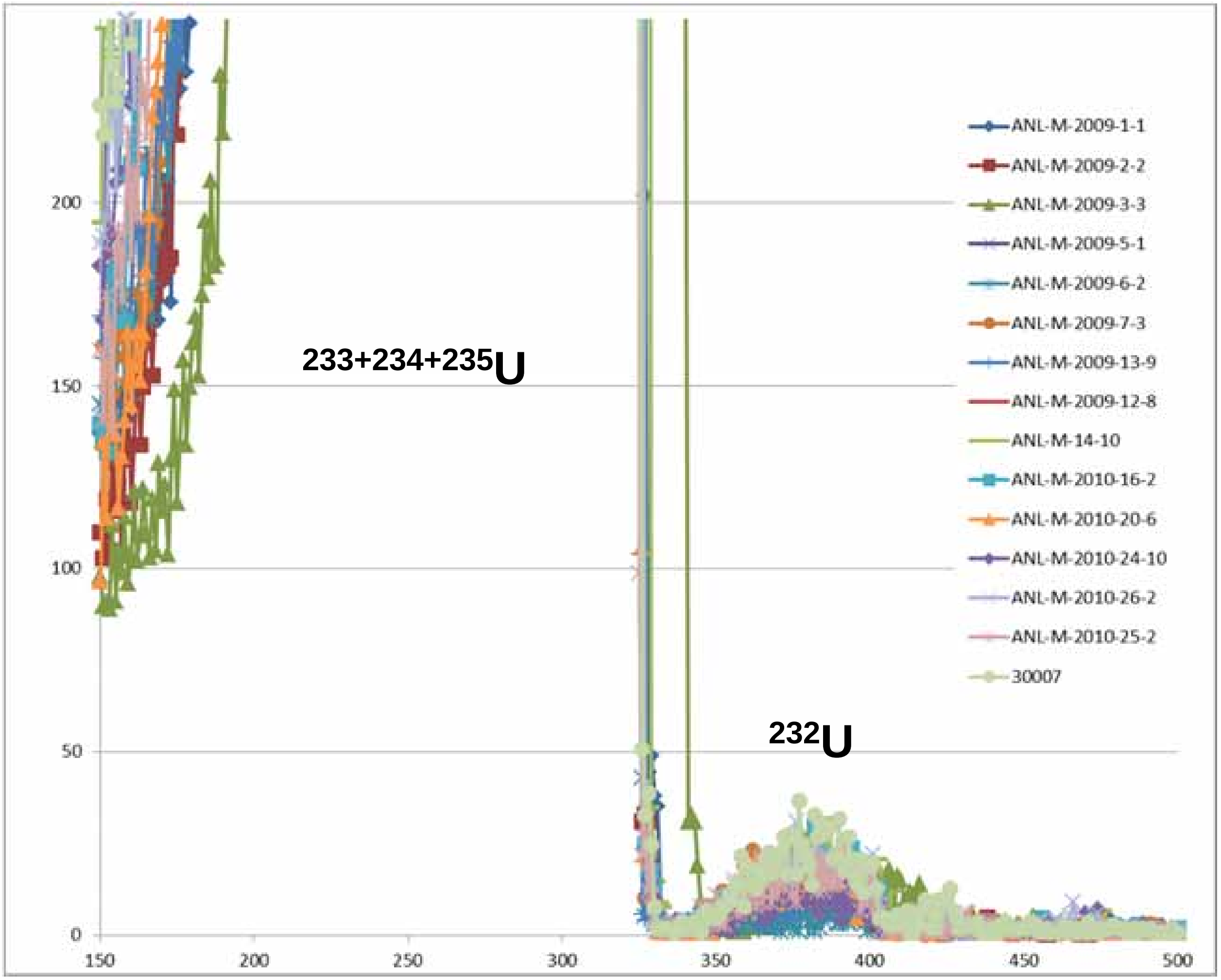
Cerium Fluoride Microprecipitation

- 1 mL of 1.55 mg/mL cerium(III) nitrate hexahydrate carrier added to purified HEU fractions containing ~1 mg uranium.
- 0.5 mL of the titanium chloride solution slowly added to each sample followed by 1.0 mL of concentrated HF.
- Mix then allow the solutions to set for at least 30 min to ensure precipitation.
- Prepare Resolve filters (80% ethanol and water rinse).
- Filter each sample with water rinses.
- Rinse filters with 80% ethanol. Dry under heat lamp.
- Mount the dried filters onto stainless planchets for alpha counting.

Alpha Energy Spectrum of ~1 mg HEU After Micro-precipitation and Collection Onto a Resolve™ Filter



All Alpha Spectrographs (3-5 days counting times)



Merging ^{232}U Alpha With MC-ICPMS Uranium Data

Isotope of Uranium	Specific Activity (Ci/g)	Branching Fraction of Contributing Alpha Energies
^{232}U	22.4	0.998
^{233}U	0.00964	0.992
^{234}U	0.00625	0.998
^{235}U	1.92E-06	0.893

$$C_{U232} = (R_m)\frac{(A_{U233} + A_{U234} + A_{U235})}{(SpA_{U232})(FA_{U232})}$$

where:

C_{U-232} : Concentration of ^{232}U in units of g-U-232/g-U(total)
R_m : Measured alpha peak ratio ^{232}U/(^{233}U + ^{234}U + ^{235}U)
A_{U-233} : Calculated ^{233}U alpha activity contribution per gram U
A_{U-234} : Calculated ^{234}U alpha activity contribution per gram U
A_{U-235} : Calculated ^{235}U alpha activity contribution per gram U
SpA_{U-232}: Specific activity for ^{232}U
FA_{U-232} : Fraction of ^{232}U alpha activity contribution to measured peak

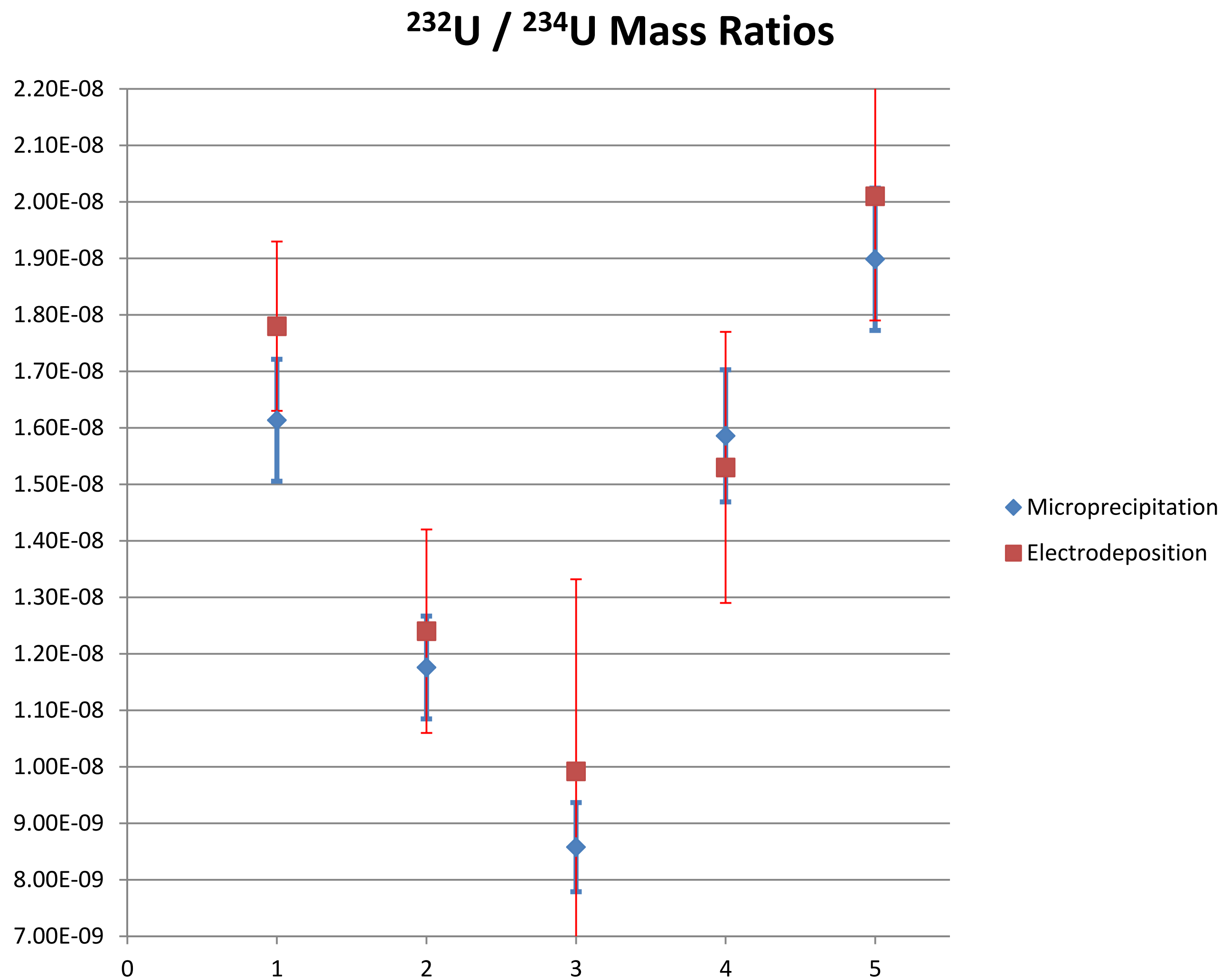
Using ^{xxx}U g / g U by IDMS

Convert to ^{232}U g / g U

Comparison of Results (Table)

	Mass Ratio ^{232}U / ^{234}U				
Sample	HEU 1	HEU 2	HEU 3	HEU 4	HEU 5
Microprecipitation	**1.61E-08**	**1.18E-08**	**8.58E-09**	**1.59E-08**	**1.90E-08**
+/- (k=2)	0.11E-08	0.91E-09	0.79E-09	0.12E-08	0.13E-08
Electrodeposition	**1.78E-08**	**1.24E-08**	**9.92E-09**	**1.53E-08**	**2.01E-08**
+/- (k=2)	0.15E-08	0.18E-08	3.40E-09	0.24E-08	0.22E-08
%D	**-9.8%**	**-5.3%**	**-14.5%**	**3.6%**	**-5.7%**

Comparison of Results (Graph)



$$\sigma_r = \sqrt{\frac{G}{t_G^{\,2}} + \frac{B}{t_B^{\,2}}}$$

2-sigma counting uncertainties calculated using the equation for standard error and sum of squares

Conclusions

- Using the micro-precipitation method,15 HEU aliquots were prepared for counting in 5 hours thus dramatically reducing the preparation time needed to make the alpha plates compared to electrodeposition.
- The collection of micro-precipitates onto Resolve™ filters allows for loading of milligram quantities of uranium onto a plate for detection of low abundance activities over shorter counting times and improved counting statistics without a severe degradation of alpha peak resolutions from high solid content.
- With the ability to load milligram quantities of HEU onto an alpha plate, counting times are dramatically decreased from 20 days to 3 days and improved counting statistics compared to electrodeposition technique.
- EXCELLENT AGREEMENT WITH RESULTS ACQUIRED USING ELECTRODEPOSITION WITH BETTER PRECISION.

Acknowledgements

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