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Title: Annual Report on the Activities and Publications of the DHS-DNDO-NTNFC-Sponsored Post-doctoral Fellow at Los Alamos National Laboratory

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Annual Report on the Activities and Publications of the DHS-DNDO-NTNFC-Sponsored Post-doctoral Fellow at Los Alamos National Laboratory

April, 2011 – April 1, 2012

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A. Introduction

This report is a summary update on the projects Magen Coleman is working on as a post-doctoral fellow funded by DHS. These research projects are designed to explore different radioanalytical facets of nuclear forensics, including destructive and non-destructive analysis, chronometry measurements and modeling, spectral analysis, and actinide separations. Method development and instrumental analysis are integral parts of these projects, along with strong radiochemical knowledge.

The current projects discussed here include method development for ^{232}U analysis, the assembly of a gamma mapping system, the testing of neutron bubble detectors for neutron spectrometry, and continued work on the chronometry of plutonium and uranium materials. This report documents the work that has been performed since April 2011.

B. Experience and Training

Magen's experience and training for this year including learning analytical tools required for nuclear forensics analysis, nuclear material handling, conducting experiments, preparing reports and literature searches.

C. U-232 analysis by gamma spectroscopy and alpha spectroscopy

Uranium-232 is an important isotope for nuclear forensics due to its ability to shed light on the irradiation history of the sample. Several methods exist for the analysis of ^{232}U in uranium samples, particularly using gamma spectrometry or separations followed by alpha spectroscopy. A series of uranium samples (metals, oxides, and fluoride) with independent chemistries and ^{232}U values determined using various methods at external laboratories are being subjected to both methods to provide results to evaluate comparative accuracy and precision.

Alpha spectrometry offers the benefits of being a direct measurement of the ^{232}U content in the sample, though more intense chemistry and sample preparation is involved before the sample can be counted. Gamma spectrometry is a technique that offers shorter count times and minimal sample preparation, but is an indirect method that relies on measuring the progeny of ^{232}U and requires that the sample be in equilibrium with respect to those progeny. Both the alpha spectrometry and gamma method were developed in collaboration with other groups and national laboratories.

Figures 1 and 2 show the experimental design for both alpha counting and gamma counting. For the alpha spectrometry method, blanks and a uranium sample DHS/DNDO and FBI PT sample from 2010 campaign were run along with each set of samples through the full procedure to ensure the quality

of the data. The gamma spectrometry procedure was performed both on a detector open to background and on a detector inside of an Ortec HPLBS1F lead shield.

Figure 1: Experimental Plan for Gamma Spectrometry for U-232 Measurements

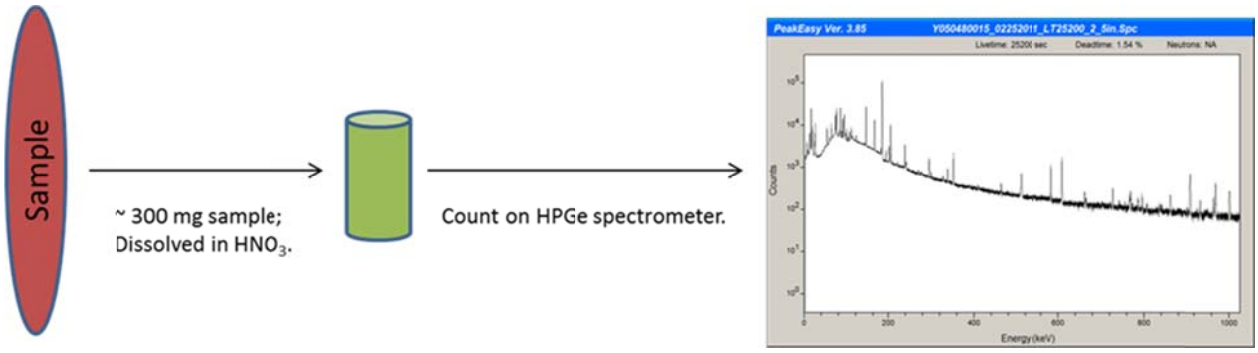
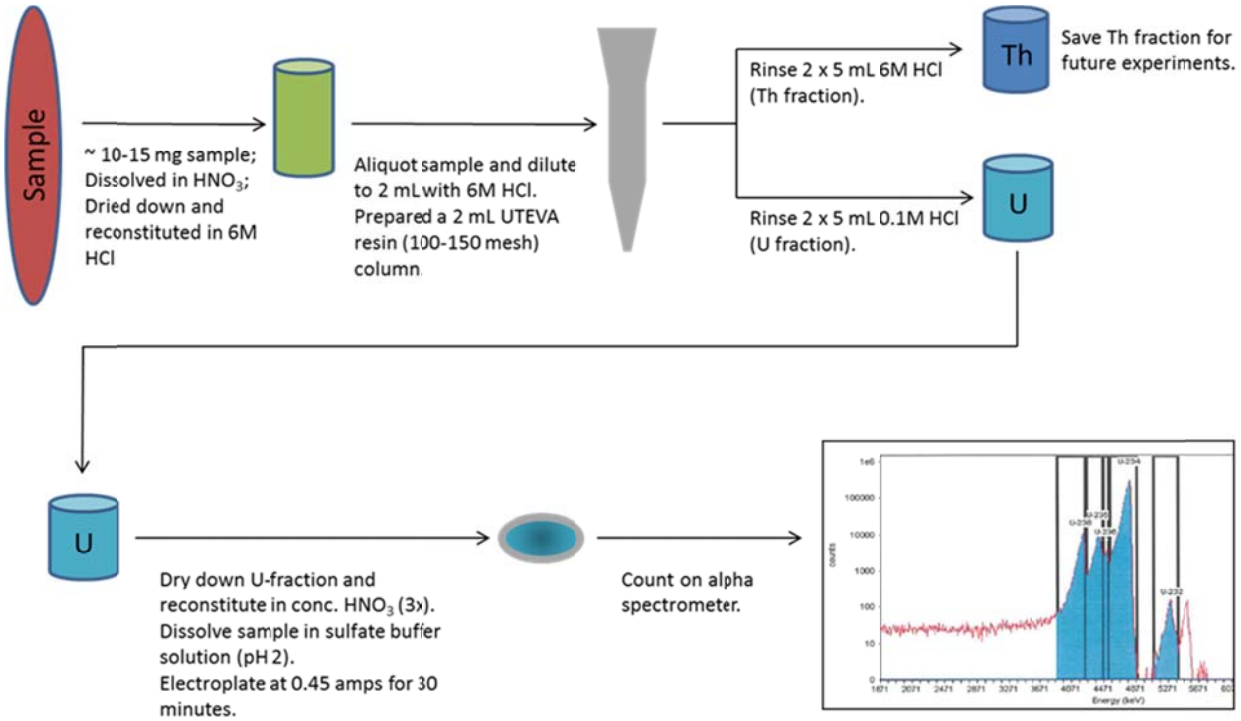


Figure 2: Experimental Plan for Separations and Alpha Spectrometry for U-232 Measurements



Since ^{232}U is present at extremely low concentrations in the samples, contamination control is critical.

These methods are currently being validated using NIST traceable standards, independent analytical methods (alpha and gamma spectrometry) and using uranium methodology samples as standards. Principles of ISO GUM were also applied for uncertainty analysis of the results.
Results from these experiments are shown in the following tables. Table 1 shows the results from the gamma spectrometry method with the Ortec lead shield in place. In nuclear facilities background peaks can offer significant challenging for determination of U-232 by gamma spectrometry. Results from the alpha spectrometry method are presented in Table 3. A graphical representation of the comparison of these results is shown in Figure 3.

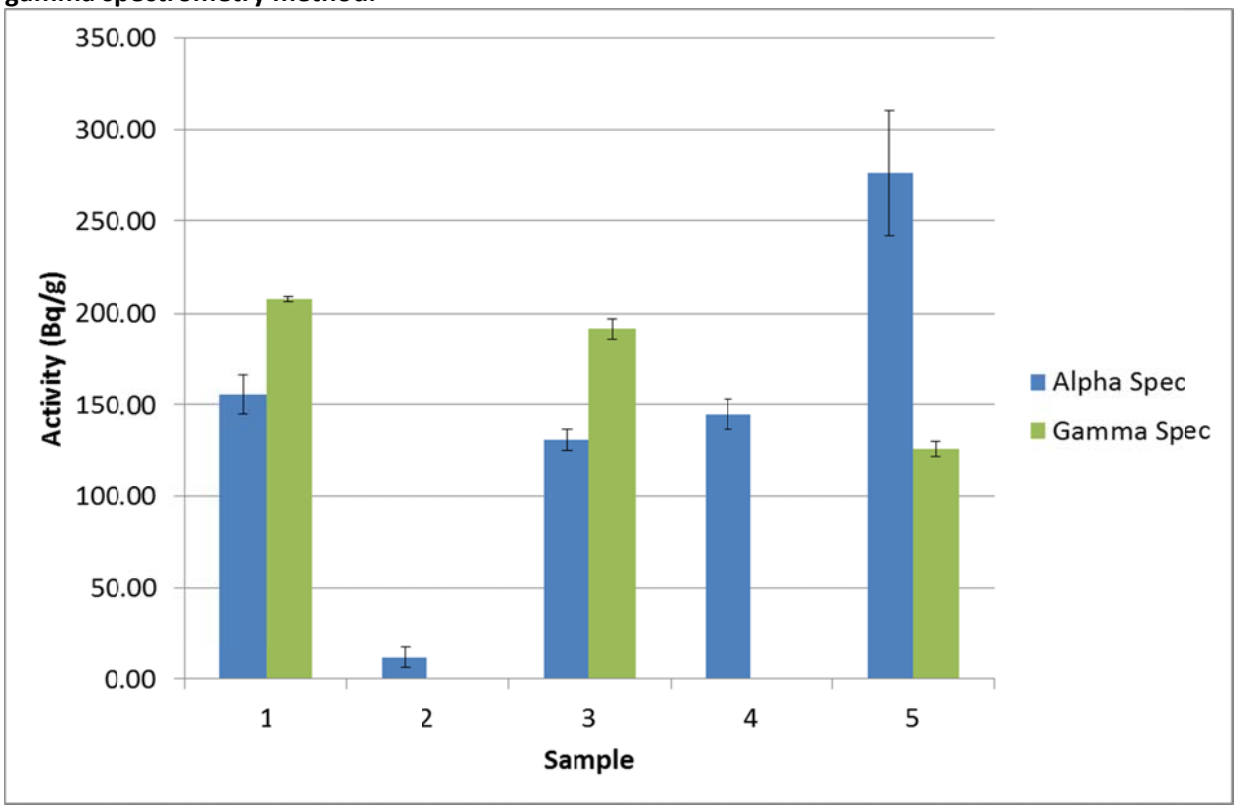
Table 1: Results from the gamma spectrometry method, for a subset of samples counted on a detector inside the lead shield.

Sample	Average ^{232}U (Bq/g U)	n	% RSD	Uncertainties for individual samples
1	208.00	3	0.75	1.6 – 1.7 %
3	191.56	3	3.01	1.6 – 1.7 %
5	126.20	3	3.22	1.0 – 1.8 %

Table 2: Results from the alpha spectrometry method.

Sample	Average ^{232}U (Bq/g U)	n	% RSD	Uncertainties for individual samples
QC	149.05	8	5.07	4-6 %
1	155.46	8	6.81	6.5-8%
2	11.65	4	46.90	11-37%
3	131.11	5	4.52	5.5-6.5%
4	144.94	4	5.61	6-9%
5	276.05	4	12.45	5.5-7.5%

Figure 3: Comparison of results from the alpha spectrometry method and the two variations of the gamma spectrometry method.



Both the alpha spectrometry method and the improved gamma spectrometry method have minimum detection limits of about 10 Bq/g. Testing of additional samples and further data analysis and additional method validation studies are in progress.

D. Two-dimensional scanner for radioactive samples

The first step in conducting a nuclear forensic examination of a sealed container is typically an external gamma spectrometry measurement to determine the presence, identity, and quantity of gamma-emitting nuclides within. To provide more information about the identity and location of the gamma-emitting nuclides within a sealed container, a two-dimensional gamma mapping system is being built using high-Z shielding, a two-dimensional linear stage, and a high-purity germanium (HPGe) detector. This system can provide complimentary information to traditional radiography about the nuclear materials in a sealed container. Integrating control of the two axes of movement with the HPGe detector and acquisition system allows for high-confidence identification of the gamma-emitting nuclides and their location within the sample. The ultimate aim is to create a map of sealed object nuclear material down to a one micron level. These measurements are critical from not only from a nuclear forensics perspective, but also from a health and safety perspective. Knowing where the radioactive material is located within a sealed object can help in determining safe handling measures and for contamination control.

As of writing this report, the scanning system has been assembled, the stage and HPGE detector have been programed to work together to scan an object, and the system is being tested. Figure 4 shows a photograph of the assembled system.

Figure 4: Photograph of the assembled 2-dimensional gamma mapping system.

E. Neutron Spectrometry using BTI Bubble Detectors

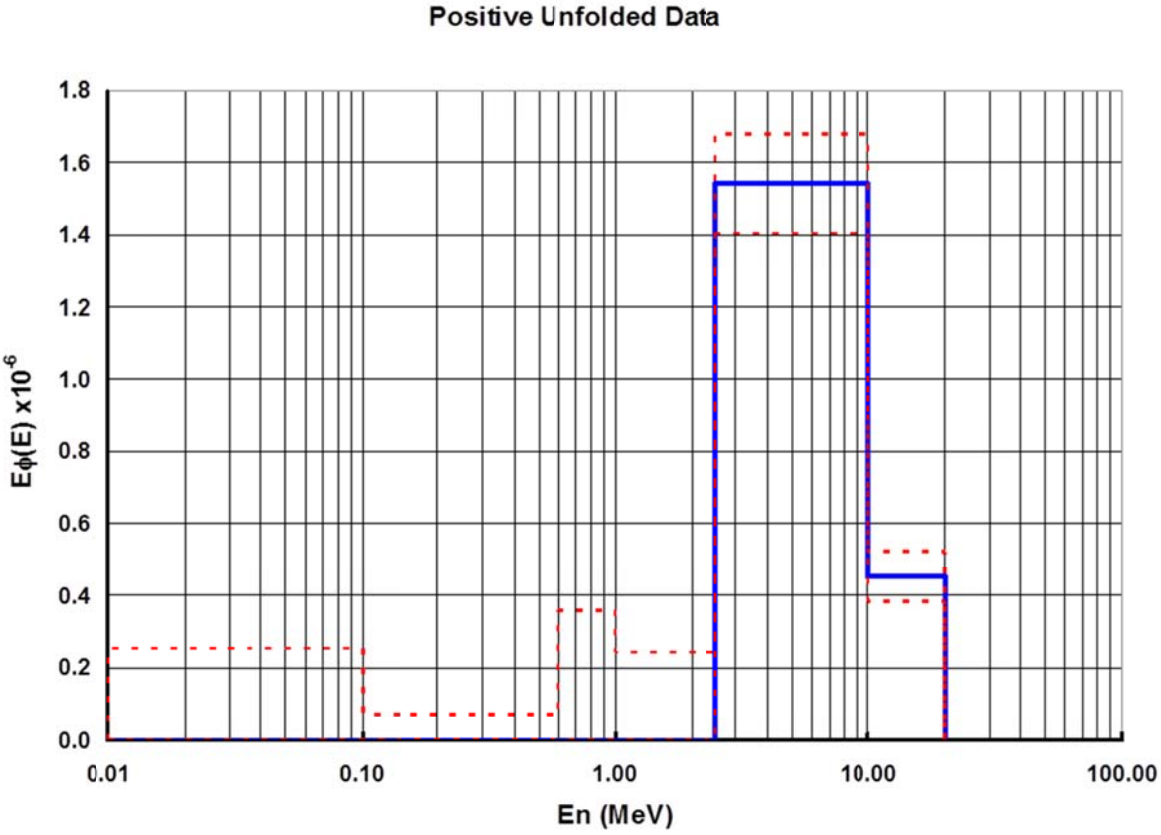
Historically, plutonium beryllium (PuBe) sources have been used in a number of applications: gauging devices, well logging, reactor start up, power production operations, medical uses, and research. PuBe neutron sources are forensically important due to the fact that they contain material that could be used in a conventional or improvised nuclear device.

These sources use the (α,n) reaction to produce neutrons. The resulting neutron energy spectrum and neutron flux/fluence are dependent on the chemistries of the source, the material form (chunk vs. powder), and the design of the source, including the layers of containment. Thus, the neutron energy spectrum and flux/fluence can provide some insight into the original purpose of the neutron source.

Neutron detectors made by Bubble Technology Industries (BTI) were recently acquired and applied to evaluate the PuBe sources for the DHS/DNDO funded for Foreign Materials/Material Characterization project for this FY. These bubble detectors are based on the interaction of an emitted neutron with tiny droplets of a superheated liquid suspended in a clear polymer gel. When the neutron hits the droplet, the energy transfer causes the droplet to vaporize, leaving behind a small bubble in the gel. The number of bubbles formed in each tube is proportional to the neutron fluence of the source. Each tube is calibrated to represent a portion of the neutron energy spectrum, and the use of a series of tubes allows for the calculation of the total neutron fluence as well as a low-resolution energy spectrum for the PuBe source. Therefore, this technology provides not only the energy distribution of neutrons but also the flux/fluence of each source. The flux/fluence result addresses the basic question of the intended use of these sources as the flux/fluence of neutron sources varies with application. For this

experiment, six sets of bubble detectors were placed at 5 inches from the PuBe source, M96, and exposed for a total of 3 hours. The detectors were then counted using the associated digital imager and software package. The results showed a neutron fluence of 2.45 x 106 n/cm^2 over the course of the 3 exposure hours. This gives a neutron flux of 226 n cm^{-2} s^{-1}. The graphical results of the neutron spectrum obtained from the data is shown in Figure 3. This spectrum shows an average neutron energy between 2.5 and 10 MeV, with neutrons of those energies accounting for 83% of the total neutron emissions. The experiments were repeated and the results were reproducible.

Figure 5: Resulting neutron spectrum from the use of the BTI neutron bubble detectors. The blue line represents the neutron spectrum, while the red, dashed lines represent the uncertainty.



Work is continuing on using these bubble detectors on additional neutron sources, including an AmBe source which is used by the local radiation protection team for calibrating neutron dosimeters. Other methods for determining the neutron spectrum from these sources, particularly using activation foils, similar to methods used in nuclear reactors, are also being evaluated to acquire more detailed spectra.

F. Chronometry of Plutonium and Uranium

Chronometry is an important capability for any nuclear forensics laboratory. Chronometry of nuclear material is essentially determining the time elapsed since the last chemical separation of the material. This works by using the known decay rates for the radioactive parents and the known amount of progeny that has grown in. This project works on validating methodologies for performing chronometry measurements in C-AAC for plutonium. Methods are being developed for chronometry of

uranium materials. The chronometry project is part of a larger study in collaboration with other scientists at LANL as well as international collaborators.

Initially, models were created using the Bateman equation to study the decay of plutonium and uranium materials and to determine the best parent/daughter pairs for chronometry studies. The plutonium model was tested using data for a series of measurement control materials and dates of last separation were determined for all of the Exchange metals using this model. Overall, the calculated ages were very close to the documented dates of separation and/or manufacture. In addition, dates calculated by multiple chronometers are very close, showing consistency in the results (Table 4). Figure 6 shows a plot of the model for $^{235}U/^{239}Pu$ with the ages for control metal D plotted with error bars. In this figure, non-decay-corrected data was used to calculate an age since last separation. Thus, for each year that a control sample was analyzed, the material was a little older. Table 4 shows the year since last separation for this sample by multiple chronometer pairs.

Figure 6: Plot of the model for $^{235}U/^{239}Pu$ with the ages for LANL's Plutonium Exchange Metal D

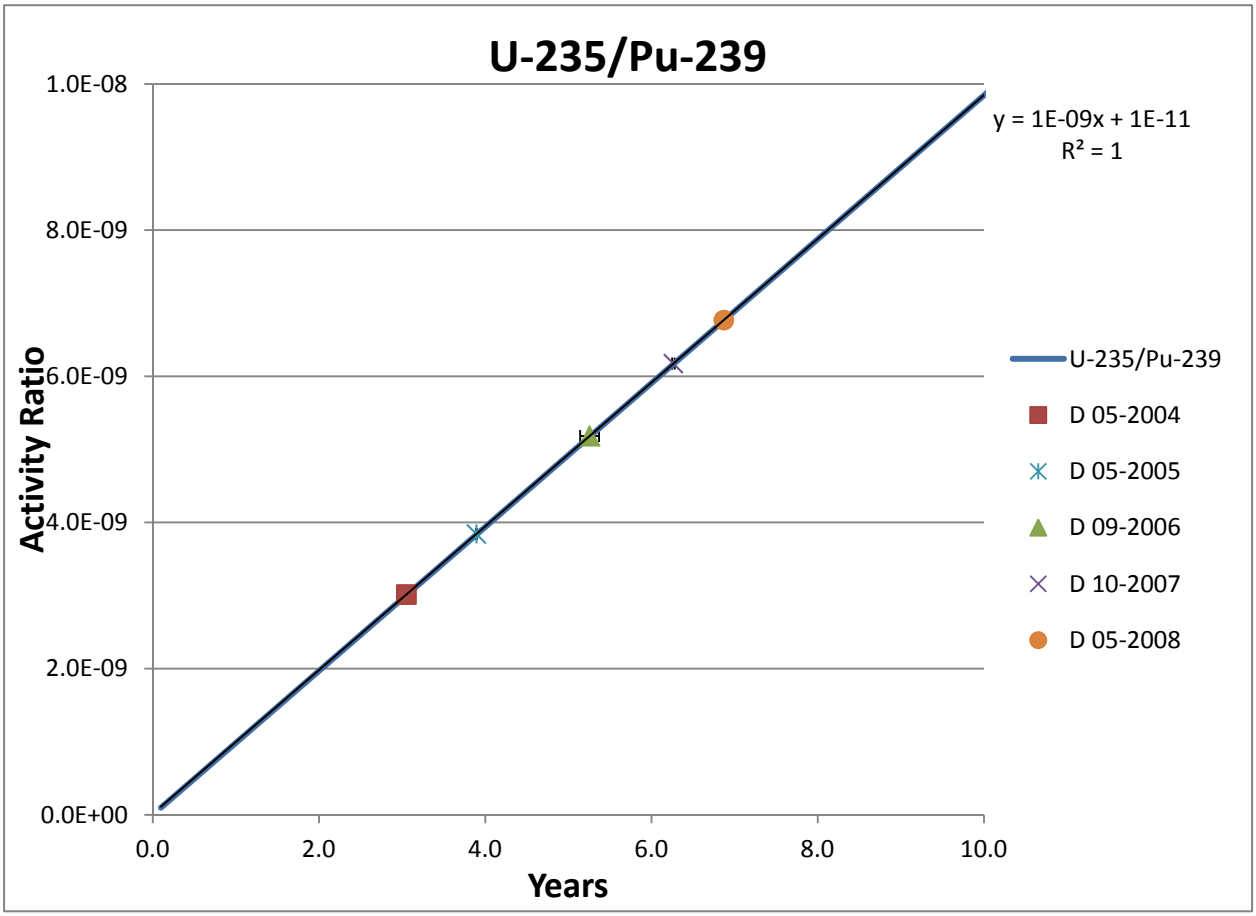


Table 4: Age since last separation calculated using various daughter/parent pairs

Sample (Date analyzed)	$^{241}Am/^{241}Pu$			$^{235}U/^{239}Pu$			$^{236}U/^{240}Pu$		
	Age (y) ± 1σ		Year (month)	Age (y) ± 1σ		Year (month)	Age (y) ± 1σ		Year (month)
D (5/04)	3.31	0.14	2001 (1)	3.05	± 0.02	2001 (4)	3.04	±0.02	2001 (4)
D (5/05)	4.28	0.12	2001 (1)	3.89	± 0.00	2001 (6)	3.88	±0.01	2001 (6)
D (9/06)	5.61	0.20	2001 (1)	5.26	± 0.11	2001 (5)	5.26	±0.11	2001 (5)
D (10/07)	6.56	0.25	2001 (3)	6.26	± 0.01	2001 (6)	6.26	±0.02	2001 (6)
D (5/08)	7.15	0.42	2001 (3)	6.87	± 0.05	2001 (6)	6.87	±0.05	2001 (6)

Actual last separation date: June 2001

Work on the chronometry of uranium is currently ongoing. This work is being performed in several parts, including 1) improvement of the separation and purification procedures for uranium, thorium, and protactinium; 2) testing of radiochemical methods for the quantification of the uranium, thorium, and protactinium isotopes of interest; and 3) improvement of the mass spectrometry techniques for quantification of the uranium and thorium isotopes of interest. The project is currently focusing on the first two parts of this project, while Floyd Stanley, a second postdoc, will be working on the first and third. Initially, work will be focused on the $^{230}Th/^{234}U$ chronometer pair, and method development for improved uranium/thorium separations is currently in progress. One method involving the use of UTEVA resin from Eichrom is being tested with tracers to determine decontamination factors and to evaluate the usefulness of this method for this application. In addition, the project will be evaluating the use of both alpha and gamma spectrometry, both individually and in combination, as detection methods for ^{234}U, ^{230}Th, ^{235}U, and ^{231}Pa, the latter two used as an alternative chronometry ratio to $^{230}Th/^{234}U$.

G. Future Plans

In the near future, it is hoped that these projects will be taken to completion and papers reporting the results will be written and published in peer-reviewed journals. In addition to the projects described here, other nuclear-forensics projects will be worked on, including measuring femtocurie-levels of ^{241}Am by gamma spectrometry.

Magen also plans on continuing to attend conferences related to nuclear forensics and hopes to continue to interact with nuclear forensics experts both here at LANL, from other national laboratories, and from the international community. She will also continue working with the experts here in order to continue developing her skills as a radiochemist and actinide analytical chemist in the field of nuclear forensics.

H. Acknowledgements

In addition to the funding from DHS-DNDO-NTNFC, we would also like to acknowledge the assistance and expertise provided by the members of the Actinide Analytical Chemistry (C-AAC) group here at LANL and Pam Thompson of AWE Aldermaston. We would also like to thank the Seaborg Institute for additional funding through their Postdoctoral Fellow program.

I. Reports, Publications, and Presentations

Internal Reports

Porterfield, D.; Coleman, M.; Myers, S.; Decker, D.; Martinez, P.; Tandon, L. "Gamma Spectrometry Analysis of Fission Products in Japan Soils," LA-CP-12-00419.

Kuhn, K.; Coleman, M.; Porterfield, D.; Myers, S.; Tandon, L. "Foreign Material Characterization & Database Development – Non-Destructive Analysis Report", LA-CP-12-00453, April 2012

Coleman, M.; Walker, L.; Tandon, L.; Wong, A. "A Revised Radioactivity Decay Calculation Program for Plutonium Materials", LA-UR-11-02480.

Kuhn, K.; Coleman, M.; Porterfield, D.; Myers, S.; Tandon, L. "Foreign Material Characterization & Database Development – Non-Destructive Analysis Report", LA-CP-11-00532, April 2011

Coleman, M.; Hahn, T.; Eglin, J.; George, G.; Slemmons, A.; Wannigman, D.; Wong, A. "Evaluation of Personnel Protective Clothing Materials for Routine Analytical Chemistry Operations in the Radiological Areas", LA-CP-11-00352, March 2011.

External Publications

Coleman, M.; Bond, E.; Moody, W.; Tandon, L. "The Analysis of Uranium-232: Comparison of radiochemical techniques and an improved method by alpha spectrometry." Submitted to Journal of Radioanalytical and Nuclear Chemistry as part of the MARC IX proceedings. LA-UR-12-20186.

Presentations

M.E. Coleman, E. Bond, L. Tandon. "Analysis of U-232 in Uranium," International Conference on Methods and Applications of Radioanalytical Chemistry (MARC) IX, March 25-30, 2012.

L. Tandon, C. Ruggiero, M. Coleman. "Non-Destructive Analysis." Presented at the IAEA Nuclear Foresnics Methodologies training course, held at PNNL, February 27-March 6, 2012. LA-UR-11-04486

M.E. Coleman, D.R. Porterfield, L. Tandon. "Development of a 2D System for Mapping Gamma-Emitting Radionuclides in Support of Forensic Examination," 242nd American Chemical Society National Meeting and Exposition, Denver, CO, August 28 – September 1, 2011. Talk presented August 29, 2011.

M.E. Coleman, L. Tandon. "Nuclear Forensics at Los Alamos National Laboratory," 2011 DHS-DNDO-NTNFC Annual Program Review, April 11-15, 2011, LA-UR-11-01940.

J. Meetings, Conferences, and Other Travel

Past Travel from April 2011 to the Present

April 11-15, 2011. DHS-DNDO-NTNFC Annual Program Review, held at SRNL. Presented a talk on the nuclear forensics research capabilities and educational opportunities at LANL.

August 28-September 1, 2011. 242nd American Chemical Societry National Meeting and Exposition, Denver, CO. Presented research on the two-dimensional gamma mapping system.

October 31-November 4, 2011. IAEA Nuclear Forensics Methodologies training course, dry run meeting. Took part in a dry-run meeting that helped put together the final course to be given to international participants.

February 27-March 1, 2012. IAEA Nuclear Forensics Methodologies Training Course. Presented a talk on non-destructive analytical methods, other than alpha and gamma spectrometry. Also participated in assisting with demonstrations and hands-on activities.

March 25-30, 2012. International Conference on Methods and Applications of Radioanalytical Chemistry (MARC) IX. Presented research on measuring ^{232}U in uranium by alpha and gamma spectrometry.

Future Travel

May 13-19, 2012. Scoping meeting for JOWOG 29 US/UK Staff exchange, as well as work on JOWOG 22, AWE Aldermaston, UK. Will be joining George Brooks and Hugh Selby for this initial meeting, as well as meeting others through Lav Tandon's participation in JOWOG 22.

June 4-8, 2012. DHS-DNDO-NTNFC Annual Program Review, held at Y12. Will be presenting nuclear forensics research.