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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

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A. Introduction

This report is a summary of the projects Jung Rim is working on as a DHS postdoctoral fellow at Los Alamos National Laboratory. These research projects are designed to explore different radioanalytical methods to support nuclear forensics applications. The current projects discussed here include development of alpha spectroscopy method for $^{240/239}$Pu isotopic ratio measurement, non-destructive uranium assay method using gamma spectroscopy, and ^{236}U non-destructive uranium analysis using FRAM code. This report documents the work that has been performed since the start of the postdoctoral appointment.

B. Experience and Training

Jung has completed various on-site and professional training since the past FY. These include extensive On-the-Job Trainings to work at a nuclear facility and learning analytical tools for nuclear forensics analysis. He attended a LANL FRAM class to learn about gamma spectral analysis in July 2104.

He had opportunities participate in planning of CMX exercise and analyzed samples for first 34 hours and 1 week study. Also, participated in several surveillances and audits related to ISO17025 accreditation. He learnt a bit on exercising operational BSAP capabilities.

Jung also had assisted in writing several white papers and proposals.

C. $^{240/239}$Pu isotopic ratio measurement using alpha spectroscopy

Alpha spectroscopy is an extremely important tool in nuclear radiation measurement. It is faster and more cost effective than mass spectrometer measurement. It is capable of accurately measuring the alpha activity of a sample and has sufficient energy resolution to provide isotopic information. Commercially available alpha detectors have a maximum resolution of 8 to 9 keV FWHM. This resolution may not be sufficient to distinguish peaks in certain alpha spectra. In a typical plutonium measurement, alpha spectroscopy is capable of measuring ^{242}Pu and ^{238}Pu, but not ^{239}Pu and ^{240}Pu. These two major isotopes in plutonium have alpha particle energies located within a range of 65 keV as shown in Table 1. Therefore, the peaks for ^{239}Pu and ^{240}Pu show significant overlap and appear as a highly-convoluted peak in a typical silicon detector. As a result, only $^{239+240}$Pu activity is typically reported for alpha spectrometry analyses. One way to overcome the physical limitation of the silicon alpha detector is to mathematically process the data to deconvolute and fit the peaks. Fitting each peak in an alpha spectrum with a representative mathematical function is often the most accurate method to measure a peak area.

Table 1 - ^{239}Pu and ^{240}Pu decay energies and intensities

	Energy (keV)	Emission Fraction
^{239}Pu	5155.5	0.732 ± 0.007
^{239}Pu	5142.8	0.151 ± 0.002
^{239}Pu	5104.7	0.106 ± 0.013
^{240}Pu	5168.2	0.735 ± 0.004
^{240}Pu	5123.7	0.2639 ± 0.0021

For this experiment, alpha spectra fitting algorithm developed by Bortels was used []. This function is Gaussian based and has exponential component to describe the tailing. The following equation is Bortels' equation with a single tailing component,

$$f(x) = \sum_{i=1}^{n} \frac{h_i \sigma \sqrt{2\pi}}{2\tau} exp\left(\frac{x-\mu_i}{\tau} + \frac{\sigma^2}{2\tau^2}\right) erfc\left[\frac{1}{\sqrt{2}}\left(\frac{x-\mu_i}{\sigma} + \frac{\sigma}{\tau}\right)\right]$$

where n is number of peaks, μ is the mean, σ is the standard deviation, τ is the distortion, and h is the height of a peak[26]. For a given source and detector system, the standard deviation and the distortion are the same for all peaks.

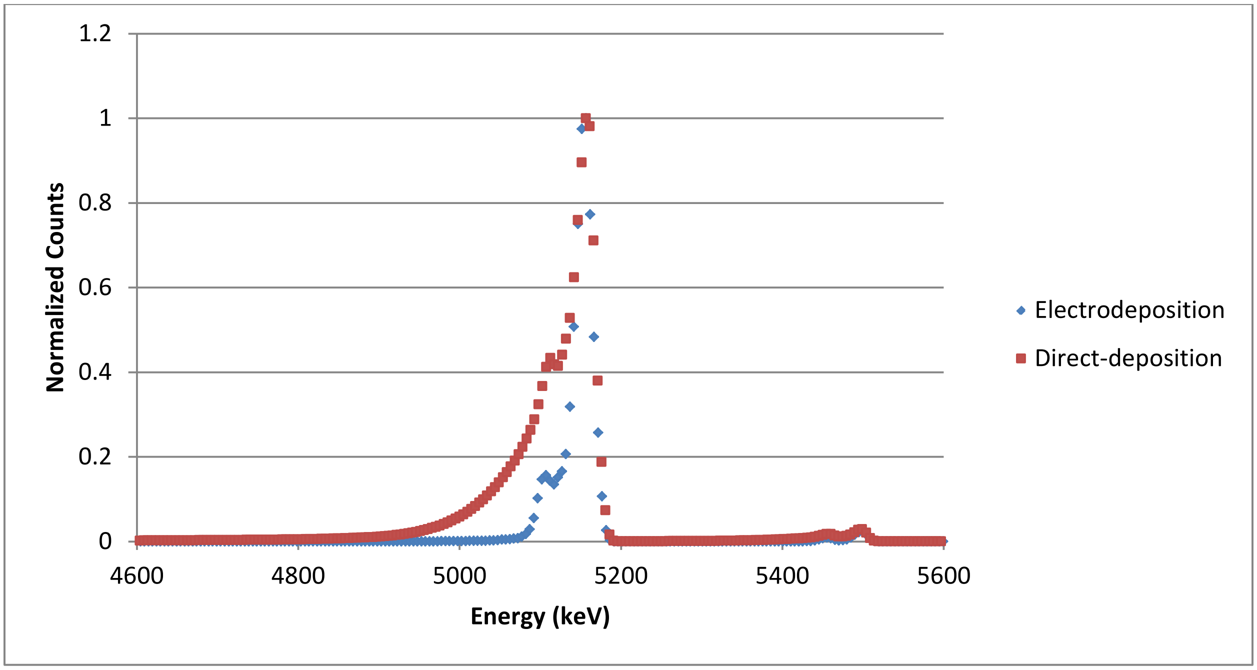

Figure 1 - Electrodeposition and direct-deposition sample alpha spectra comparison

In this experiment, plutonium samples were prepared by two distinctive methods: direct-deposition and electrodeposition. The direct-deposition is a simpler method to prepare samples but it produces thick depositions, which result in degraded spectra due to sample attenuation. Electrodeposition is a process of using an electrochemical cell to deposit a coating of a chemical species onto a metallic source. This technique produces the thin and uniform layers of actinides required to minimize the degradation in spectral resolution caused by alpha particles attenuation within a source. Spectral resolutions of samples prepared by direct-deposition and electrodeposition are compared in Figure 1.

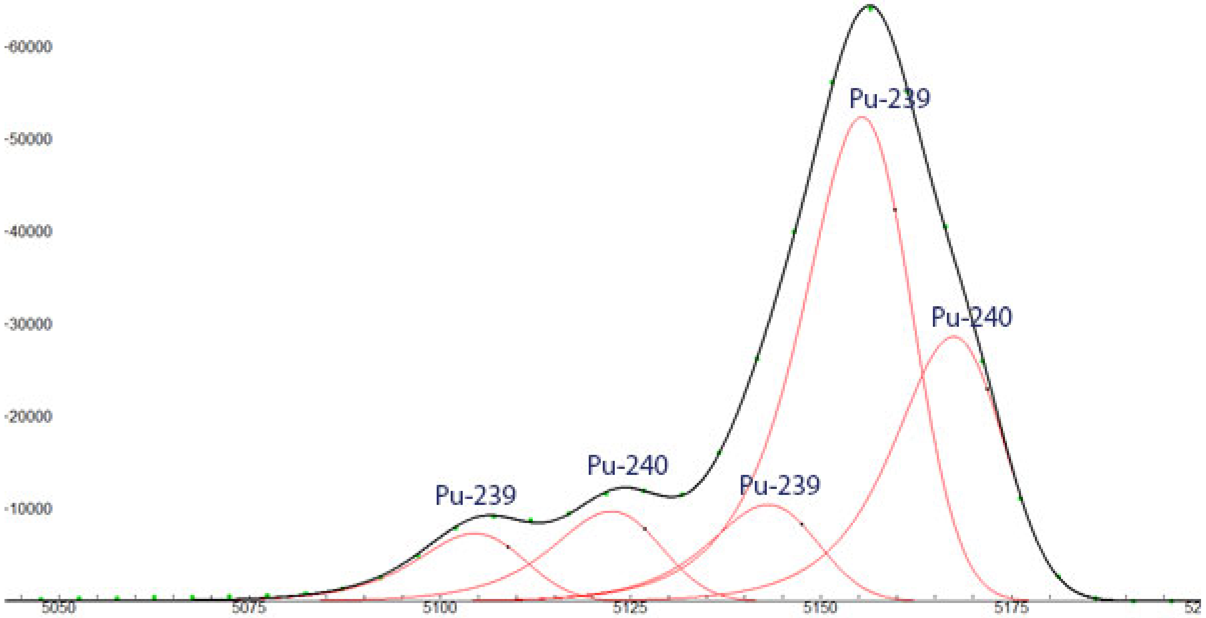

Figure 2 - ^{239}Pu and ^{240}Pu peaks deconvoluted and fitted with Bortel’s alpha equation

Samples prepared with both methods were evaluated with Bortels’ fitting function. ^{238}Pu peaks at 5456.3 and 5499.03 keV were first fitted with the function because these peaks are free of any interference. These ^{238}Pu fitting terms were then used to fit ^{239}Pu and ^{240}Pu peaks. The fitting parameters were the representation of the detector response to the individual sample and should be physically consistent for all the peaks within a single spectrum. For the direct-deposited samples, the tailing of spectra was too pronounce and hindered from effectively analyzing the data using a fitting scheme. For electrodeposited samples, ^{239}Pu and ^{240}Pu were effectively deconvoluted and fitted using Bortels’ function as shown in Figure 2. The electrodeposition and fitting scheme was tested using NBL CRM-136, 137 and 138. The $^{240/239}$Pu isotopic ratios calculated from the alpha spectra were compared to the certified values and listed in Table 2. The certified isotopic ratios all fall within the uncertainties of the measurements. The alpha fitting technique can be also used to measure $^{238/239}$Pu ratio independent from TIMS measurement, which will significantly reduce time required for the measurement and more cost effective.

Table 2 - $^{240/239}$Pu atomic ratio measured with alpha spectroscopy and fitting algorithm compared to certified values

	$^{240/239}$Pu ratio from alpha spec (%)	$^{240/239}$Pu certification (%)
CRM-136	14.9 ± 2.4	14.500 ± 0.018
CRM-137	25.1 ± 1.7	24.080 ± 0.029
CRM-138	9.13 ± 0.97	8.614 ± 0.011

D. Assessing effectiveness of FRAM code in measuring ^{236}U from gamma-ray spectra

The commercially available FRAM software package provides a very effective means to process gamma-ray spectra to obtain plutonium or uranium isotopic information, and it is used globally for international nuclear safeguard applications. ^{236}U gamma-ray energies are not easily measured with HPGe. Due to this limitation, FRAM has a built-in correlation function to estimate ^{236}U concentration. This particular function was derived from the large number of uranium samples previously examined. The correlation equation uses ^{235}U, ^{238}U and ^{228}Th concentration to estimate the ^{236}U mass fraction. However, in many cases, ^{228}Th concentration in uranium sample is low and is ignored in ^{236}U correlation calculation. This makes ^{236}U estimation solely dependent on ^{235}U and ^{238}U, which has very little physical basis in estimation ^{236}U concentration in sample.

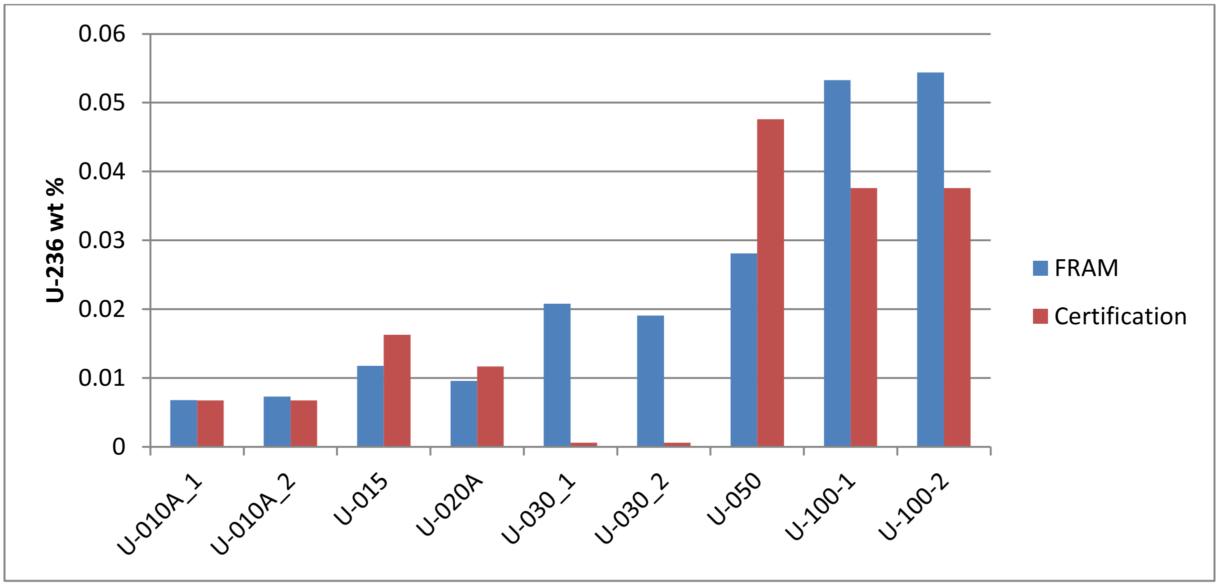

Figure 3 – U-236 weight percent comparison between FRAM results and certification from U-010A through U-100.

In test with NBL uranium series standards, FRAM was accurate in estimating U-236 only at limited occasions. In several instances, the estimations were significantly over or under the certified values as shown in Figure 3 and 4. There were no trends that can be derived from the result and the FRAM code may not be suitable for estimating ^{236}U from gamma-ray spectra data. The code was designed and used with large quantity samples and estimating ^{236}U for small samples may not be adequate. The best practice will be forcing FRAM to only calculate ^{236}U concentration by the actual measurement and disable the correlation function.

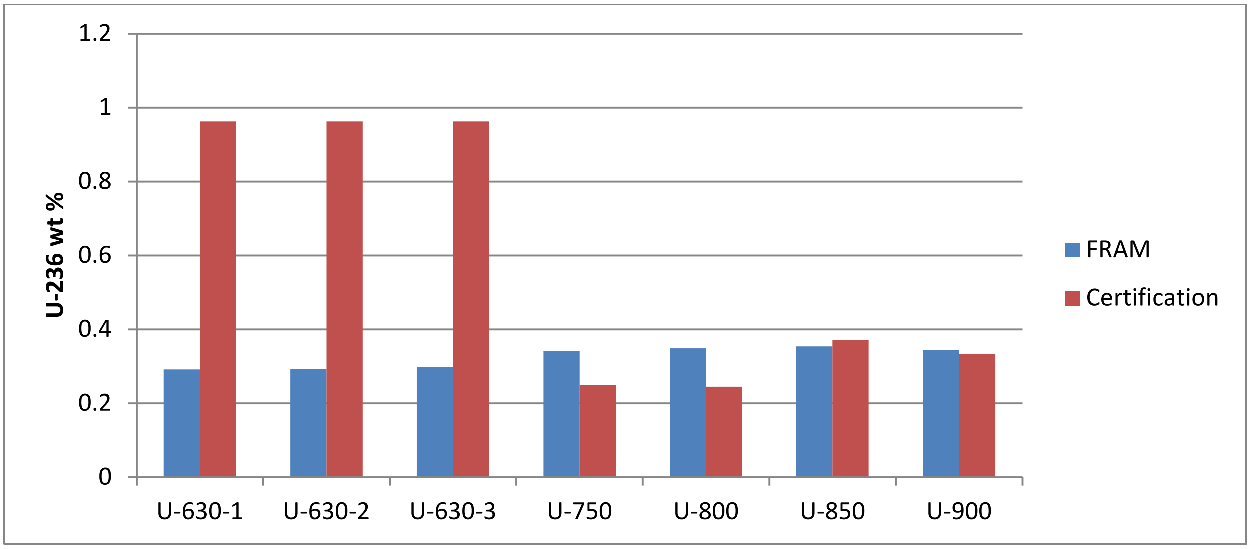

Figure 4 – U-236 weight percent comparison between FRAM results and certification from U-0630 through U-900.

E. Uranium assay using gamma spectroscopy

A uranium assay is important for nuclear forensic analysis. A routine was established to non-destructively perform uranium assay using a HPGe gamma detector and a software package. This method is highly flexible and does not require geometrically identical standards to calibrate the HPGe system. Once a sample is counted using a detector system, data analysis was performed using Ortec GammaVision and Angle software. These computer programs were used to convert gamma-ray spectroscopy data into mass for ^{234}U, ^{235}U, and ^{238}U. It has to be noted that ^{236}U cannot be measured with gamma spectroscopy at a low concentration. All three samples analyzed showed no measurable ^{236}U with the HPGe gamma detector.

The background was first subtracted from sample gamma spectra before any analysis. A NIST traceable Ho-166m calibration source was used for energy and efficiency calibration in GammaVision. The reference efficiency calibration curve generated using Ho-166m source was imported to Angle and used to produce a geometry corrected efficiency curve for each sample. These geometry corrected efficiency curves were then applied to an appropriate spectrum to convert counts to activity in Bq. The activity is then converted to mass of uranium. The efficiency curve generated by neither GammaVision nor Angle assigns uncertainties to its values. For the GUM compliant uncertainty calculation, 10% uncertainty was assigned to efficiencies used in this calculation.

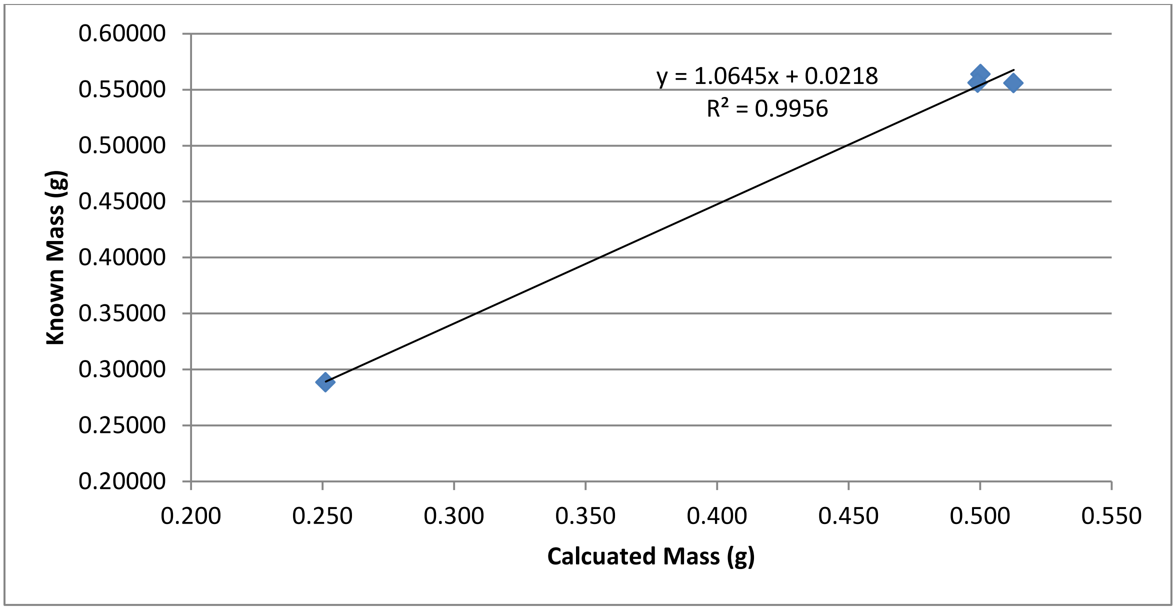

Figure 5 - Sample bias correction plot generated using NBL uranium CRMs

Four NBL CRM QCs were run along with three unknown samples. These QCs showed a tendency for uranium mass to be lower than the known mass. To correct for the bias, the QC mass data plot was fitted with linear regression as shown in Figure 5, and correction terms were applied to the samples. The bias corrected values were compared to values measured using Davies & Gray titration and both measurements were within the uncertainties of each other as shown in Table 3.

Table 3 – Uranium assay measurement comparison between NDA and Davies & Gray titration

	Uncorrected		Corrected		Davies & Gray
	U mass (g)	U assay (g U /g sample)	U mass (g)	U assay (g U /g sample)	U assay (g U /g sample)
Sample-1	1.90 ± 0.19	0.80 ± 0.10	2.04 ± 0.28	0.86 ± 0.12	0.8817 ± 0.0007
Sample-2	1.90 ± 0.19	0.81 ± 0.10	2.05 ± 0.28	0.86 ± 0.12	0.8816 ± 0.0007
Sample-3	2.40 ± 0.23	0.81 ± 0.10	2.58 ± 0.35	0.87 ± 0.12	0.8718 ± 0.0007

F. Future Plans

Jung will continue to perform alpha sample analysis work. Once plutonium work is completed, uranium samples will be analyzed. This will involve measuring ^{232}U concentration from the bulk uranium samples.

G. Acknowledgements

We gratefully acknowledge the support from the National Technical Nuclear Forensics Center at the U.S. Department of Homeland Security's Domestic Nuclear Detection Office. In addition, we would also like to acknowledge the assistance and expertise provided by the members of the Actinide Analytical Chemistry (C-AAC) group at LANL.

H. Reports, Publications, and Presentations

Presentations

1. Rim J, Doyle J, Byerly B, Tandon L (2014) LANL R&D: All Current Efforts to Support NTNF.

2. Rim J (2015) Uranium Assay using HPGe Detector System.

I. Meetings, Conferences, and Other Travel

Past Travel from April 2014 to the Present

Attended Bulk SNM Analysis Program (BSAP) Technical Analysis Working Group (TAWG) meeting at LLNL, October 28-30, 2014

Future Travel

April 11-17, 2015 MARC

Jung Rim, Jamie Doyle, Lav Tandon, and Donivan R. Porterfield; Developing Gum Compliant Uncertainty Approach for FRAM

Jung Rim, Lav Tandon, and Donivan R. Porterfield; Determination of the Pu-240/Pu-239 Isotopic Ratio Using an Alpha Spectrum Peak Fitting Algorithm