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Title:	A Report on the Activities, Publications, and Pending Research of DHS/DOD Sponsored Post-doctoral Research Associate at Los Alamos National Laboratory
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A Report on the Activities, Publications, and Pending Research of a DHS/DOD Sponsored Post-doctoral Research Associate at Los Alamos National Laboratory

Floyd Stanley and Lav Tandon

1. Introduction:

Since beginning at Los Alamos National Laboratory in February of 2012, I have been working as a DHS./DNDO Postdoctoral Research Associate under the mentorship of Lav Tandon and Khalil Spencer (NA-22 and mass spectrometry). The focus of my efforts, in addition to pursuing needed training and qualifications, has been the application of various instrumental approaches (e.g. Thermal Ionization Mass Spectrometry; TIMS) to a range of systems of interest in materials characterization and nuclear forensics. Research to be pursued in the coming months shall include the continued use of such approaches to advance current methods for: *modified total evaporation, monitoring critical minor isotope systems,* and *chronometry*. Each of the above points will be discussed below.

2. Credentials and Training:

Various aspects of on-site and professional training have been completed to date. In addition to gaining significant exposure to lab/building specific policies, I have completed sessions dedicated to CPR, nuclear materials handling, and other on-the-job training relevant to maintaining a safe working environment (e.g. working in fume hoods and with high voltage instrumentation). Furthermore, an L-level security clearance reinstatement has been completed and a Q-level upgrade has been initiated.

Immediate goals in this area include the completion of all on-the-job training so that work can be carried out fully independently, continuing to develop an understanding of LANL infrastructure and resources, and identifying potential contributions that can be made within my research group.

3. Research and Development Overview:

3.1 Modified Total Evaporation: Classical total evaporation [1] enables rapid, highly-precise analysis of actinides in TIMS by volatilizing an entire sample while integrating the signals for each isotope. This approach essentially allows for the elimination of fractionation effects during the evaporation process that alter resulting isotope ratios throughout the run; fluctuations in resulting isotopic ratios as a function of fractionation effects are illustrated in Figure 1 for NBL CRM U-500. However, the classical total evaporation does not fully account for modern instrumentation improvements and is not optimized for the determination of minor actinide isotope ratios (e.g. $^{234}U/^{238}U$ and $^{236}U/^{238}U$).

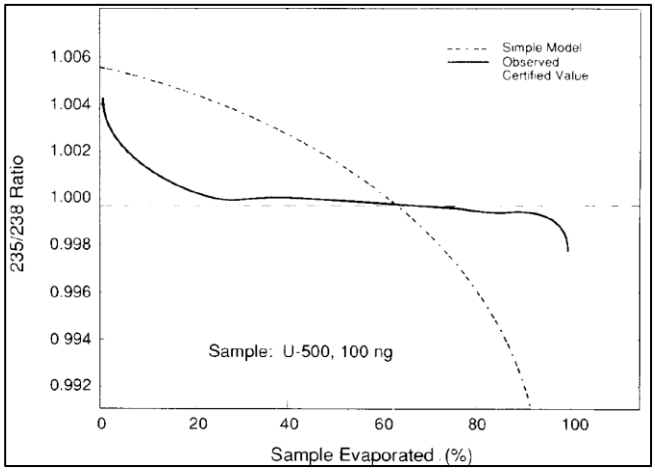

Figure 1. Theoretical and observed, by TIMS, fractionation for NBL CRM U-500. Adapted from Reference 1.

Significant effort will dedicated to establishing state-of-the-art total evaporation methods that capitalize on the strengths of currently available instrumentation while providing greater accuracy for minor isotope measurements. These goals will be met

through updated programming designed to incorporate additional peak centering, peak tailing, baseline, inter-detector correction features.

3.2 Minor Isotope Analysis: The analysis of minor actinide isotopes is essential in materials characterization, safeguards, and non-proliferation. These species (e.g. ^{236}U, ^{233}U or ^{244}Pu) can serve as key indicators of material processing and potentially illicit or undeclared activities. [2] The analysis of these constituents, however, may be complicated by the large range of compositions possible for materials ranging from natural to highly processed. For example, $^{236}U/^{238}U$ ratios can vary from 10^{-14} for natural materials up to 10^{-2} for reprocessed material, making updated, robust instrumental approaches a necessity in modern nuclear materials research. [3]

Efforts to enhance minor isotopic analysis procedures beyond the existing methods [1,4] will be partially joined with the work described above. This research will ultimately consider various isotopic systems to develop updated methods capable of dramatically reducing current levels of uncertainly associated with the measurement of minor isotope ratios.

3.3 Chronometry: The decay of actinide isotopes can provide various, unique chronometric relationships of value in determining the amount of time elapsed since a nuclear material's last purification (See Figure 1). [5] Such age values are critical parameters in identifying a material's source and processing, as well as potentially involved parties. Furthermore, this information is critical in verifying international compliance with proposed treaties (e.g. Fissile Material Cut-off Treaty).

The goal of the chronometry research, in general, is to further the current ability to age-date nuclear materials of varied provenance and purpose. This work will rely on the development and application of high-performance TIMS methods, in conjunction with state-of-the-art instrument programming, sample preparation and ionization techniques. In addition to exploiting known, valuable chronometric relationships (e.g. $^{234}U/^{230}Th$, $^{235}U/^{231}Pa$), novel strategies for determining material purification-related biases will also be examined for relatively "young" materials. [6]

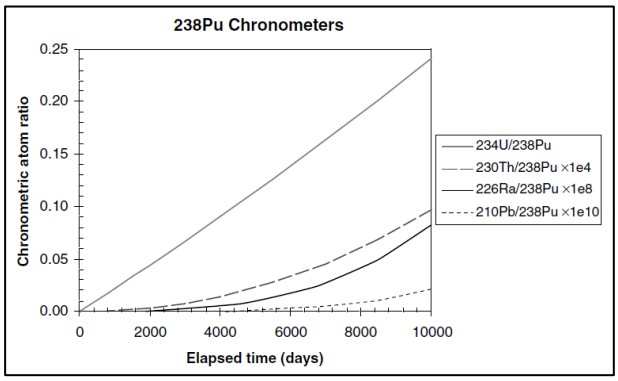

Figure 2. Isotope ratios of the ^{238}Pu decay chain as a function of time elapsed since parent material purification. Adapted from Reference 5.

4. Conference Representation:

1. F.E. Stanley, S.E. Glover, A.M. Stalcup, H.B. Spitz, D. Beals. Alpha Spectrometric Evaluation of SRM-995 as a Uranium/Thorium Double Tracer System for Age-Dating Uranium Materials. Poster Presentation; Methods and Applications of Radioanalytical Chemistry; March 25-30, 2012. Kailua-Kona, HI.

2. F.E. Stanley. The Application of Thermal Ionization Mass Spectrometry in Various Investigations. Oral Presentation; NTNFC Annual Academic-Laboratory Collaboration and Program Review Meeting; June 4-8, 2012. Oak Ridge, TN.

5. Relevant Publications:

1. F.E. Stanley, A.M. Stalcup, H.B. Spitz. A Brief Introduction to Analytical Methods in Nuclear Forensics. *Trends in Analytical Chemistry* (2012) In Progress.

2. F.E. Stanley, S.E. Glover, A.M. Stalcup, H.B. Spitz, D. Beals. Alpha Spectrometric Evaluation of SRM-995 as a Potential Uranium/Thorium Double Tracer System for Age-Dating Uranium Materials. *J. Rad. Nuc. Chem.* (2012) Submitted.

6. References:

1. E.L. Callis, R.M. Abernathey. *Int. J. Mass Spect. Ion Proc.* (1991) 103:93.

2. M. Magara, T. Sakakibara, S. Kurosawa, M. Takahashi, S. Sakurai, Y. Hanzawa, F. Esaka, K. Watanabe, S. Usuda. *J. Nuc. Sci. Tech.* (2002) 39: 308.

3. S. Richter, H. Kuhn, Y. Aregbe, M. Hedberg, J. Horta-Domenech, K. Mayer, E. Zuleger, S. Burger, S. Boulyga, A. Kopf, J. Poths, K. Mathew. *J. Anal. At. Spectrom.* (2011) 26: 550.

4. S. Richter, S.A. Goldberg. *Int. J. Mass Spec.* (2003) 229: 181.

5. K.J. Moody, I.D. Hutcheon, P.M. Grant. Nuclear Forensic Analysis. CRC Press Taylor and Francis Group, Boca Raton FL, 2005.

6. Z. Varga, M. Wallenius, K. Mayer, E. Hrnecek. *J. Rad. Nuc. Chem.* (2011) 290: 485.