

## Actinides and Rare Earths Focus Topic Room 202C - Session AC+AS+SA-ThM

### Nuclear Power, Forensics, and Other Applications

**Moderator:** Ladislav Havela, Charles University, Prague, Czech Republic

8:00am **AC+AS+SA-ThM1 Electron Microscopy in Nuclear Forensics**, *Edgar Buck, D.R. Reilly, J.M. Schwantes, J.A. Soltis, T.Q. Meadows, D.A. Meier, J.F. Corbey*, Pacific Northwest National Laboratory **INVITED**

Recent advances in electron microscopy both in terms of spatial resolution, sensitivity, and the ability to perform in-situ chemistry experiments, present enormous opportunities to the nuclear forensic field. Extraction of specific particles of interest with dual-beam SEM-FIB instruments can be readily applied plutonium-contaminated materials. Furthermore, other developments in microscopy have enabled in-situ monitoring of interfacial processes and 3D tomographic views of specimens and phases. Several new advances in technology have enabled great advances and potential for nuclear science including nuclear forensics. These include dual-beam ion-electron systems for precise sample isolation and preparation, the electromagnetic lens aberration-corrector, high-count capacity x-ray detector systems, faster digital cameras, high performance electron backscattered diffraction systems in combination with 3D visualization tools, application of cryoTEM methods to material science, as well as micro-fabricated in-situ cells that enable direct observation of chemical and electrochemical processes in the EM. The application of these technologies to nuclear forensics will be discussed.

8:40am **AC+AS+SA-ThM3 New Frontiers with Fission Track Analysis and TOF-SIMS Techniques**, *Itzhak Halevy*, Nrcn Israel; *R. Radus*, Ben Gurion University, Israel; *S. Maskova*, Charles University, Prague, Czech Republic; *A. Kogan, S. Samuha, D. Gridchin, E. Grinberg, E. Boblil, N. Haikin*, IAEC-NRCN, Israel; *I. Orion*, Ben-Gurion University -Negev, Israel; *A. Weiss*, Faculty of Engineering, Bar-Ilan University, Israel **INVITED**

Illicit trafficking of radioactive materials is known to exist from the early days of radioactive era. The nuclear forensics deals with recognizing the materials and processes of the radioactive industry. The properties of the materials can give a hint about the source of material and its original use.

The most common radioactive material involved in illicit trafficking is uranium. Uranium is a common natural element which can be found everywhere. The cosmogenic uranium is well known and defined. The natural enrichment of uranium is varying within a small range around 0.72 % and is indicative to its source. The enrichment of the anthropogenic uranium can vary much depending on the purpose and use of the material.

Different enrichments are known for individual nuclear power plants, research reactors and military uses.

Measuring the uranium properties can indicate its enrichment, presence of other elements or impurities and can help in finding its attribution, namely its origin.

To learn more about the history of found material accurate isotopic measurements are needed.

The ratio between  $^{230}\text{Th}$  and  $^{234}\text{U}$  can give a good estimation of how much time passed from the last chemical cleaning of the material. This technique is called radio-chronometry or age dating.

We developed new Lexan detectors with much better signal to noise ratio to improve sensitivity and reduce the false alarm.

New Automated software can recognize the fission track (FT) automatically and give the parameters of the track, like: roundness, intensity, number of tracks and color histogram. In that program we can add ROI (region of interest) or to cancel a false positive FT identification.

Analytical equipment like TOF-SIMS (Time Of Flight - Secondary Ion Mass Spectrometry) and ICP-MS (Inductively Coupled Plasma - Mass Spectrometer located at the Geophysical survey Israel - GSI) together with new FTA software give new frontiers to the nuclear forensic research .

A microscope equipped with TOF-SIMS gives the ability to measure quantitatively, the ratio between the different isotopes and molecules. Using the scanning electron microscope, we are able to choose the desired particle which can be then analyzed using the TOF-SIMS. TOF-SIMS is a technique capable to distinguish not only between elements but also different isotopes. Knowing the ratio between the isotopes is very important as it can help us to classify the material looking for its possible origin. Morphology and depth profile than give other insight to the

processes that the sample went through. Nano manipulators can extract the particle and transfer it to a different technique.

9:20am **AC+AS+SA-ThM5 Predictive Nuclear Forensics: Fundamental Frameworks to Fill Missing Pieces**, *Jenifer Shafer, M. Koehl, A. Baldwin, D. Wu*, Colorado School of Mines; *R. Rundberg*, Los Alamos National Laboratory; *M. Servis*, Washington State University; *T. Kawano*, Los Alamos National Laboratory **INVITED**

Understanding the origin of nuclear forensic signatures provides the benefit of understanding how these signatures can be compromised and provides a framework to predict signatures that might arise under various conditions. The ability to predict signatures is particularly useful for the nuclear forensics community since only a limited number of samples exist. Frequently access to these samples can be further constrained due to classification boundaries. This talk provides two examples of how fundamental chemical and physical phenomena can be leveraged to understand signature origins, thus enabling a more robust nuclear forensics capability. The first study focuses on understanding how organic phase aggregation chemistry in the PUREX process can dictate trace metal, such as fission or corrosion product, partitioning patterns. By understanding how trace metals partition, information regarding the processing history, including the reprocessing site, could be ascertained. Trace metal partitioning patterns were studied by producing radiotracers in the USGS 1 MW TRIGA reactor. The influence of extractant aggregation on trace metal partitioning was then assessed using a combination of diffusion NMR spectroscopy and small angle neutron scattering. The second study focuses on understanding how cumulative fission product yields can describe the incident neutron energy. Fission yield curves of uranium-235 have a decrease in valley radionuclide production when the incident neutron energy is in the epithermal energy regime. This decrease in valley radionuclide production seems tied to the excitation of the uranium-236 to the 3- spin state. The octupole deformation of the 3- spin state enables more asymmetric fission than typically encountered with fast or thermal neutrons and thus suggests the structure of the excited uranium-236 compound nucleus could be, in part, responsible for cumulative fission product yields. These two studies highlight how fundamental science enables signature development.

11:00am **AC+AS+SA-ThM10 Soft X-ray Synchrotron Radiation Spectromicroscopy Studies of Radioactive Materials**, *David Shuh*, Lawrence Berkeley National Laboratory; *A. Altman*, Lawrence Berkeley National Laboratory and UC Berkeley; *A.L.D. Kilcoyne, S.G. Minasian, J.I. Pacold, D.E. Smiles, T. Tylliszczak, D. Vine*, Lawrence Berkeley National Laboratory; *L. He, J. Harp, M. Meyer*, Idaho National Laboratory; *C. Degueldre*, University of Lancaster, Switzerland

Soft X-ray synchrotron radiation methodologies are being developed and employed at the Advanced Light Source (ALS) of Lawrence Berkeley National Laboratory (LBNL) to elucidate the electronic structure of radioactive and actinide materials. Results from these investigations have begun to provide improved fundamental knowledge that can be used as a scientific basis for the enhanced design of actinide materials, complexes, ligands, and the overall understanding of actinide materials. The experimental developments at the ALS have centered on studies of radioactive materials with the soft X-ray scanning transmission X-ray microscope (STXM) at Beamline 11.0.2 for spatially-resolved near-edge X-ray absorption spectroscopy (XAS). The spectromicroscopy capabilities of the STXM provide the means to determine the speciation and composition in a range of actinide materials, particularly those of technological and environmental interest with spatial resolution that can reach to the true nanoscale. A particular emphasis has been on the use of light atom (B, C, N, O, F, Na, Mg, Al, Si) ligand K-edge XAS technique to determine the electronic structure characteristics in an array of unique and relevant materials. Furthermore, there are a host of additional electron energy level thresholds (such as the L-edges of the transition metals, the M-edges of the lanthanides, and others) that can be probed by near-edge XAS in the soft X-rays.

Recently, STXM spectromicroscopy studies have been extended to focused ion beam (FIB) prepared radioactive and irradiated material specimens in collaboration with Idaho National Laboratory. Future scientific developments and applications of soft X-ray spectromicroscopy investigations utilizing ptychography and in-operando methodologies will be discussed.

# Thursday Morning, October 25, 2018

11:20am **AC+AS+SA-ThM11 Comparison of the Oxidation Rates for Alpha Versus Delta Plutonium by X-ray Photoelectron Spectroscopy**, *Art Nelson, S.B. Donald, D.J. Roberts, W. McLean*, Lawrence Livermore National Laboratory

X-ray photoemission spectroscopy (XPS) was used to characterize differences in the oxidation rates for polycrystalline  $\alpha$ -Pu versus  $\delta$ -Pu related to variations in grain size and surface electronic structure. The evolution of the Pu 4f core-level chemical shift as a function of oxygen exposure at ambient temperature was quantified in oxidation profiles. In addition, the X-ray excited Pu NOO Auger line-shapes were combined with the chemical shift of the Pu 4f<sub>7/2</sub> photoelectron line that defines the Auger parameter and results in a reliable method for determining oxidation states independent of binding energy calibration. The oxidation profiles reveal that  $\alpha$ -Pu oxidizes faster than  $\delta$ -Pu, both resulting in the PuO<sub>2</sub>/Pu<sub>2</sub>O<sub>3</sub>/Pu oxide structure. This data was used to produce a chemical state (Wagner) plot for select plutonium oxides.

This work was performed under the auspices of the U.S. Dept. of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

11:40am **AC+AS+SA-ThM12 A Single-Stage AMS Detector for Secondary Ion Mass Spectrometry and its Applications to Nuclear Materials Analyses**, *David Willingham, E.E. Groopman, K.S. Grabowski*, U.S. Naval Research Laboratory; *L. Sangely*, International Atomic Energy Agency; *A.P. Meshik, O.V. Pravdivtseva*, Washington University in St. Louis; *D.G. Weisz, K.B. Knight*, Lawrence Livermore National Laboratory

Secondary ion mass spectrometry (SIMS) has long been applied to the analysis of isotopic heterogeneities in nuclear materials. Few other methodologies can compete with the ability of SIMS to measure the isotopic composition of nuclear materials with high accuracy and precision with micrometer spatial resolution. Like many other mass spectrometry techniques, however, the presence of molecular isobaric interferences at any given mass-to-charge complicate interpretation of SIMS measurements. Uranium isotopes, for example, can be valuable and informative markers for the process of enriching uranium for nuclear fuel for the production of nuclear energy. In addition to the major uranium isotopes (<sup>238</sup>U and <sup>235</sup>U), the minor isotope <sup>236</sup>U is of interest because it is an indication for the presence in the sample of uranium reprocessed from spent nuclear fuel. The resolving power needed to separate <sup>236</sup>U from the molecule <sup>235</sup>U<sup>1</sup>H, however, is greater than 38,000 - far beyond most the mass resolution of commercially available SIMS instruments. One solution to this problem is to use a Single-Stage Accelerator Mass Spectrometer (SSAMS) to accelerate secondary ions to a high enough energy (300keV) to enable molecular dissociation within a stripper gas while retaining good transmission, followed by SIMS-based detection. At the U.S. Naval Research Laboratory, we have developed the Naval Ultra-Trace Isotope Laboratory's Universal Spectrometer (NAUTILUS) to achieve this goal. Using the NAUTILUS, we accomplish molecule-free isotopic analysis of nuclear materials without sacrificing the benefits of SIMS. In this work, we demonstrate the broad applications of the NAUTILUS to areas of nuclear materials analyses including uranium bearing particle analysis for nuclear Safeguards, analysis of the Oklo natural nuclear reactor, and analysis of uranium doped silicate glasses as working reference analogs for nuclear fallout materials. The NAUTILUS represents a new era in SIMS analyses of complex materials with specific application to nuclear materials and general application to the SIMS community as a whole.

12:00pm **AC+AS+SA-ThM13 Physicochemical Properties of Ag in Annealed ZrN/SiC/Ag Heterostructures Used to Simulate TRISO Nuclear Fuels**, *Jeff Terry, M. Warren, R. Seibert*, Illinois Institute of Technology

Silicon carbide (SiC) is used as a supportive and protective barrier in the cladding of tristructural-isotropic (TRISO) nuclear fuel particles. Previous studies both of surrogate surfaces and irradiated fuel have shown that the fission product silver (Ag) exhibits transport into and sometimes through the SiC barrier with temperatures above 500 °C. This silver release can cause safety concerns for maintenance workers due to plate-out on in-reactor components. Although an exact diffusion mechanism for Ag in SiC is unknown, a solution is needed to prevent this effect. The use of a ZrN protective coating may mitigate Ag transport and potential release. This study examines the transport of Ag in SiC through the use of surrogate multilayered thin-films. Thin films of subsequent layers of SiC, Ag, SiC, and ZrN deposited by pulsed-laser ablation deposition (PLD) under a range of annealing temperatures up to 1200 °C are studied. After heating, X-ray

photoemission spectroscopy (XPS), scanning electron microscopy (SEM), and energy-dispersive X-ray spectroscopy (EDS) was used to examine the surface of the ZrN. Initial results show that Ag does not readily diffuse through the ZrN layer like it does through SiC. The results and implications of this study will be discussed.

## Author Index

### Bold page numbers indicate presenter

— A —

Altman, A.: AC+AS+SA-ThM10, 1

— B —

Baldwin, A.: AC+AS+SA-ThM5, 1

Boblil, E.: AC+AS+SA-ThM3, 1

Buck, E.C.: AC+AS+SA-ThM1, 1

— C —

Corbey, J.F.: AC+AS+SA-ThM1, 1

— D —

Degueldre, C.: AC+AS+SA-ThM10, 1

Donald, S.B.: AC+AS+SA-ThM11, 2

— G —

Grabowski, K.S.: AC+AS+SA-ThM12, 2

Gridchin, D.: AC+AS+SA-ThM3, 1

Grinberg, E.: AC+AS+SA-ThM3, 1

Groopman, E.E.: AC+AS+SA-ThM12, 2

— H —

Haikin, N.: AC+AS+SA-ThM3, 1

Halevy, I.: AC+AS+SA-ThM3, 1

Harp, J.: AC+AS+SA-ThM10, 1

He, L.: AC+AS+SA-ThM10, 1

— K —

Kawano, T.: AC+AS+SA-ThM5, 1

Kilcoyne, A.L.D.: AC+AS+SA-ThM10, 1

Knight, K.B.: AC+AS+SA-ThM12, 2

Koehl, M.: AC+AS+SA-ThM5, 1

Kogan, A.: AC+AS+SA-ThM3, 1

— M —

Maskova, S.: AC+AS+SA-ThM3, 1

McLean, W.: AC+AS+SA-ThM11, 2

Meadows, T.Q.: AC+AS+SA-ThM1, 1

Meier, D.A.: AC+AS+SA-ThM1, 1

Meshik, A.P.: AC+AS+SA-ThM12, 2

Meyer, M.: AC+AS+SA-ThM10, 1

Minasian, S.G.: AC+AS+SA-ThM10, 1

— N —

Nelson, A.J.: AC+AS+SA-ThM11, 2

— O —

Orion, I.: AC+AS+SA-ThM3, 1

— P —

Pacold, J.I.: AC+AS+SA-ThM10, 1

Pravdivtseva, O.V.: AC+AS+SA-ThM12, 2

— R —

Radus, R.: AC+AS+SA-ThM3, 1

Reilly, D.R.: AC+AS+SA-ThM1, 1

Roberts, D.J.: AC+AS+SA-ThM11, 2

Rundberg, R.: AC+AS+SA-ThM5, 1

— S —

Samuha, S.: AC+AS+SA-ThM3, 1

Sangely, L.: AC+AS+SA-ThM12, 2

Schwantes, J.M.: AC+AS+SA-ThM1, 1

Seibert, R.: AC+AS+SA-ThM13, 2

Servis, M.: AC+AS+SA-ThM5, 1

Shafer, J.: AC+AS+SA-ThM5, 1

Shuh, D.K.: AC+AS+SA-ThM10, 1

Smiles, D.E.: AC+AS+SA-ThM10, 1

Soltis, J.A.: AC+AS+SA-ThM1, 1

— T —

Terry, J.: AC+AS+SA-ThM13, 2

Tyliszczak, T.: AC+AS+SA-ThM10, 1

— V —

Vine, D.: AC+AS+SA-ThM10, 1

— W —

Warren, M.: AC+AS+SA-ThM13, 2

Weiss, A.: AC+AS+SA-ThM3, 1

Weisz, D.G.: AC+AS+SA-ThM12, 2

Willingham, D.: AC+AS+SA-ThM12, 2

Wu, D.: AC+AS+SA-ThM5, 1