



2016 LLNL Nuclear Forensics Summer Program

Glenn T. Seaborg Institute
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Physical and Life Sciences Directorate
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Sponsors:
National Technical Nuclear Forensics Center, Domestic
Nuclear Detection Office, Department of Homeland
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Administration under Contract DE-
AC52-07NA27344.
LLNL-TR-709693



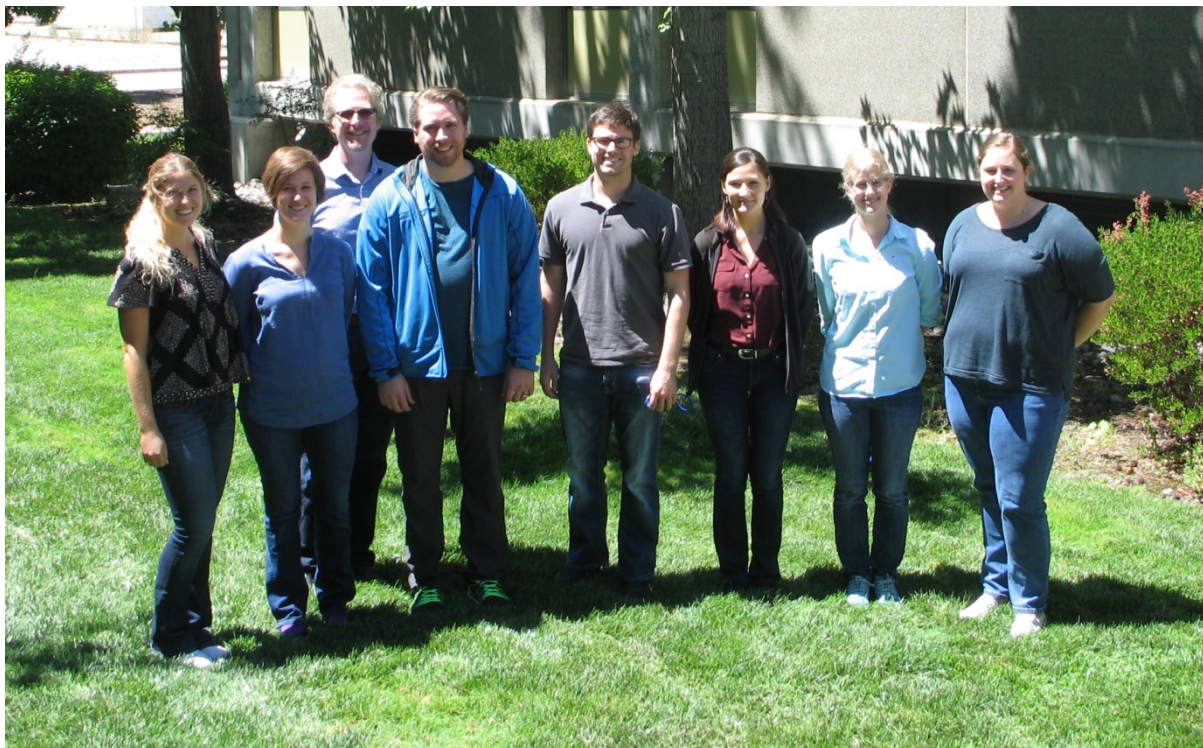
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Auspices

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA273.

2016 Nuclear Forensics Summer Program



Mavrik Zavarin, Director, Glenn T. Seaborg Institute (back) with 2016 Nuclear Forensics Summer Program students (left to right) Carina Edelman, Kelly McHugh, John “Jack” Goodell, Austin Carter, Marie Kirkegaard (NFGFP), Ellen Monzo (NFUSP), Mary Kathryn “Katie” Hoffman, and Nathan Veale (not pictured).

The Lawrence Livermore National Laboratory (LLNL) Nuclear Forensics Summer Program is designed to give graduate students an opportunity to come to LLNL for 8–10 weeks for a hands-on research experience. Students conduct research under the supervision of a staff scientist, attend a weekly lecture series, interact with other students, and present their work in poster format at the end of the program. Students also have the opportunity to meet staff scientists one-on-one, participate in LLNL facility tours (e.g., the National Ignition Facility and Center for Accelerator Mass Spectrometry), and gain a better understanding of the various science programs at LLNL.

Currently titled the Nuclear Forensics Summer Program (NFSP), this program began 17 years ago as the Actinide Sciences Summer Program. The program is run within the Glenn T. Seaborg Institute in the Physical and Life Sciences Directorate at LLNL. The goal of the NFSP is to facilitate the training of the next generation of nuclear scientists and engineers to solve critical national security problems in the field of nuclear forensics. Students are selected from the fields of physics, chemistry, geology, mathematics, nuclear engineering, chemical engineering and environmental sciences. Students engage in research projects in the disciplines of actinide chemistry, radiochemistry, isotopic analysis, computation, radiation detection, and nuclear engineering in order to strengthen the “pipeline” for future scientific disciplines critical to DHS (DNDO).

The NFSP is highly competitive with over 50 applicants for the 6-8 slots available. Additional students funded through paid internships from NNSA, DHS and DOE are invited to participate in the summer lecture series and poster symposium. This year, the NFSP hosted students from 7 universities (See Table 1) across the US (Figure 1). This year’s NFSP students conducted

research on such diverse topics as Monte Carlo N-particle (MCNP) modeling of the Vast Area Detection for Experimental Radiochemistry (VADER) diagnostic instrument at the National Ignition Facility (NIF), plutonium reaction kinetics on mineral surfaces, MCNP modeling of irradiation and gamma-ray spectra of metal foils for nuclear forensics applications, characterization of nuclear test debris using Nano-Secondary Ion Mass Spectrometry (NanoSIMS) and Scanning Electron Microscopy (SEM), evaluation of uranium radiochronometry methods using Multi-Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICP-MS), application of Resonance Ionization Mass Spectrometry (RIMS) to nuclear forensics, and application of stable isotope and noble gas mass spectrometry (NGMS) in earth systems (see Table 2 for poster titles). Graduate students are invited to return for a second year at their mentor's discretion (two 2015 students returned in 2016). Continuation of research collaboration between graduate student, faculty advisor, and LLNL mentors is strongly encouraged. In many cases, NFSP research evolves into a significant component of the students' graduate theses.

In addition to hands-on training, students attend a weekly lecture series on topics applicable to the field of nuclear forensics (see Table 3). Speakers are selected to represent the breadth of expertise that is required for nuclear forensics research. Speakers discuss the importance of their work in the context of national and international nuclear forensics.

Graduate and undergraduate students on fellowships such as the Nuclear Forensics Graduate Fellowship are invited to join our summer program. They usually come for 8-9 weeks and can return the following summer or stay throughout the year depending on their research needs. This year we had 1 Nuclear Forensic Undergraduate Scholarship program student and 2 Nuclear Forensic Graduate Fellowship program students join the summer program (Table 1). In addition, five students funded by other nuclear science fellowships participated in the summer program activities.

As part of our effort to build a "pipeline" for next generation nuclear forensics scientists, we host students who are participating in the DOE-sponsored "Summer School in Radiochemistry" held at San Jose State University (SJSU). The SJSU summer students come to LLNL to meet our summer students, discuss nuclear forensics research opportunities at LLNL, and tour LLNL's state-of-the-art facilities. The SJSU summer students are strongly encouraged to apply to LLNL's nuclear forensics program (e.g. Ellen Monzo and Marie Kierkegaard are both SJSU summer student graduates).

Our summer program is providing a nuclear forensics pipeline of top-quality students from universities across the U.S. Since 2002, 30-40% have returned to conduct their graduate research at LLNL. In addition:

- 17 became postdoctoral fellows at LLNL
- 6 became postdoctoral fellows at other national labs
- 13 were hired as career scientists at LLNL
- 4 were hired as career scientists at other national labs
- 4 were hired as faculty in the area of nuclear forensics/radiochemistry/nuclear science
- 4 were hired at other government institutions

A big factor in the success of this program is the dedication of the staff scientists who volunteer to mentor the summer students. Three of those mentors are, in fact, past recipients of NTNFC fellowships and are now helping to grow the next generation of nuclear forensics scientists. In

FY16, funding from NTNFC's Graduate Mentor Assistance Program (GMAP) helped to support the time required to mentor summer interns. The GMAP allows staff scientists to develop summer projects for their students, oversee necessary safety training, and dedicate time to helping the interns maximize their productivity and scientific potential. Posters summarizing each student's research were presented at our Laboratory Student Poster Day and are included at the end of this report.

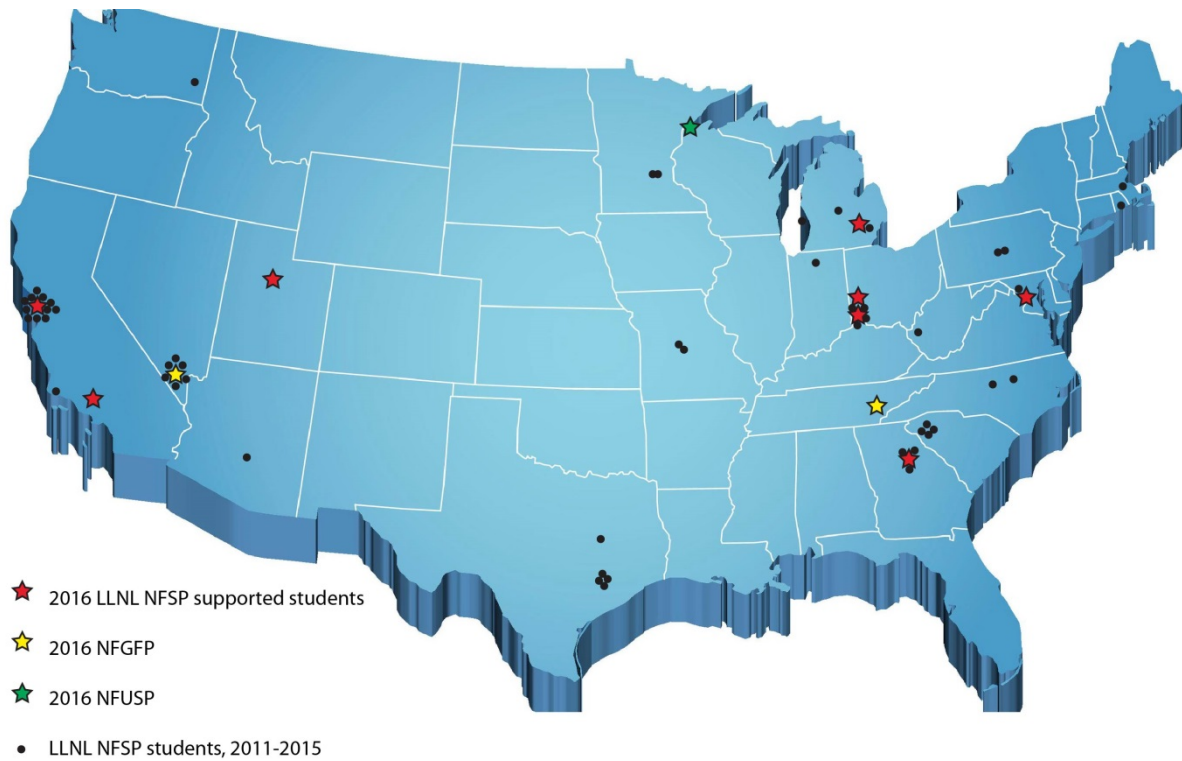


Figure 1. LLNL's 2016 Nuclear Forensics Summer Program participants are selected from across the United States.

Table 1. 2016 Nuclear Forensics Summer Program Students

Student	Major	University	Year
Austin Carter	Chemical Engineering	Brigham Young University, Provo UT^a	Graduate
Carina Edelman	Geology	University of California, Santa Barbara CA	Graduate
John “Jack” Goodell	Nuclear Chemistry	University of Maryland, College Park, MD	Graduate
Mary Kathryn “Katie” Hoffman	Chemistry	University of Cincinnati, OH	Graduate
Kelly McHugh	Geology	Miami University, Oxford OH	Graduate
Rodrigo Tapia	Chemistry	University of Georgia, Athens GA	Undergraduate
Nathan Veale	Geology	California State University East Bay, Hayward, CA	Graduate
Marie Kirkegaard^b	Engineering Science	University of Tennessee, Knoxville TN	Graduate
Jeffrey Rolfes^b	Radiochemistry	University of Nevada, Las Vegas NV	Graduate
Ellen Monzo^c	Biochemistry	University of Minnesota, Duluth MN	Undergraduate

^a Now at University of Michigan, Ann Arbor MI^b Nuclear Forensics Graduate Fellow^c Nuclear Forensics Undergraduate Fellow

Table 2. 2016 Student Projects and Mentors

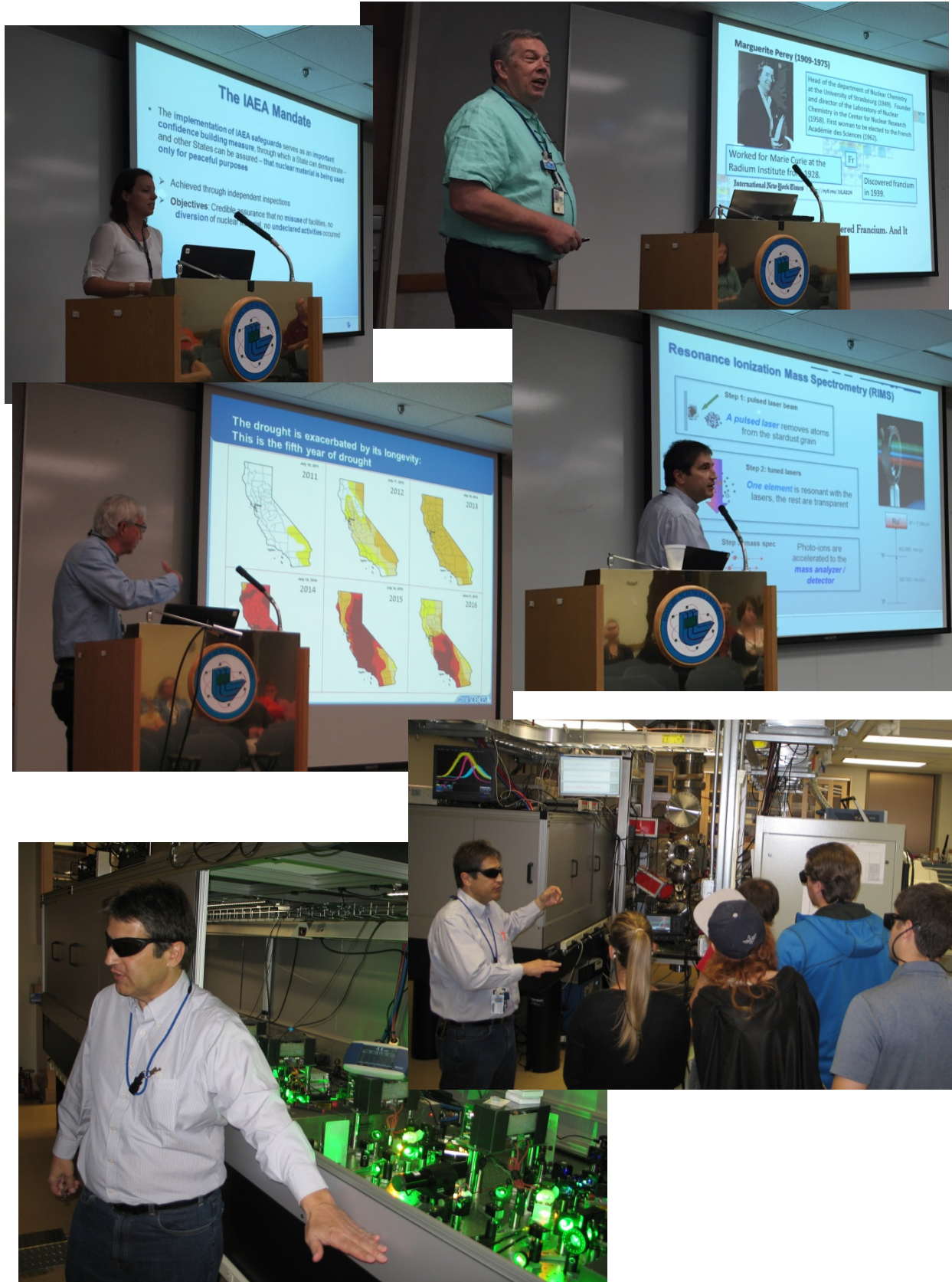
Student	Mentor	Project Poster Title
Austin Carter	Narek Gharibyan/Lucile Dauffy	Monte Carlo Modeling of Vast Area Detection for Experimental Radiochemistry (VADER) NIF Diagnostic Instrument
Carina Edelman	James Begg	Experimental Pu Sorption and Reduction on Clays: Predicting Environmental Behavior
John “Jack” Goodell	Bryan Bandong/Christine Egnatuk/Steven Padgett	Modeling Neutron Irradiation and Gamma Emission Spectra of Activation Foils for Nuclear Forensics Applications
Mary Kathryn “Katie” Hoffman	Ruth Kips/Chad Durrant ^b /Mike Kristo	Characterization of Cesium-Bearing Soils from the Marshall Islands using Micro-Analytical Techniques
Kelly McHugh	Amy Gaffney/Kerri Schorzman	Th-U and Pa-U Model Ages of Bulk Uranium Radiochronometry Standards
Rodrigo Tapia	Brett Isselhardt ^b	Modeling Sputtering and Ion Optics for Resonance Ionization Mass Spectrometry (RIMS)
Nathan Veale	Ate Visser/Brad Esser	Hydrology of the SPRUCE Climate Change Experiment
Marie Kirkegaard ^c	Mavrik Zavarin/Brenda Rubenstein ^a	Auxiliary-Field Quantum Monte Carlo for UO_2^{2+}
Ellen Monzo ^d	Tashi Parsons-Moss ^b / Kim Knight	Optimization of Uranium Molecular Deposition for Alpha Counting Sources

^a Now assistant professor, Brown University, Providence RI^b DHS NTNFC graduate fellowship and/or post-doctoral fellowship recipients^c Nuclear Forensics Graduate Fellow^d Nuclear Forensics Undergraduate Fellow

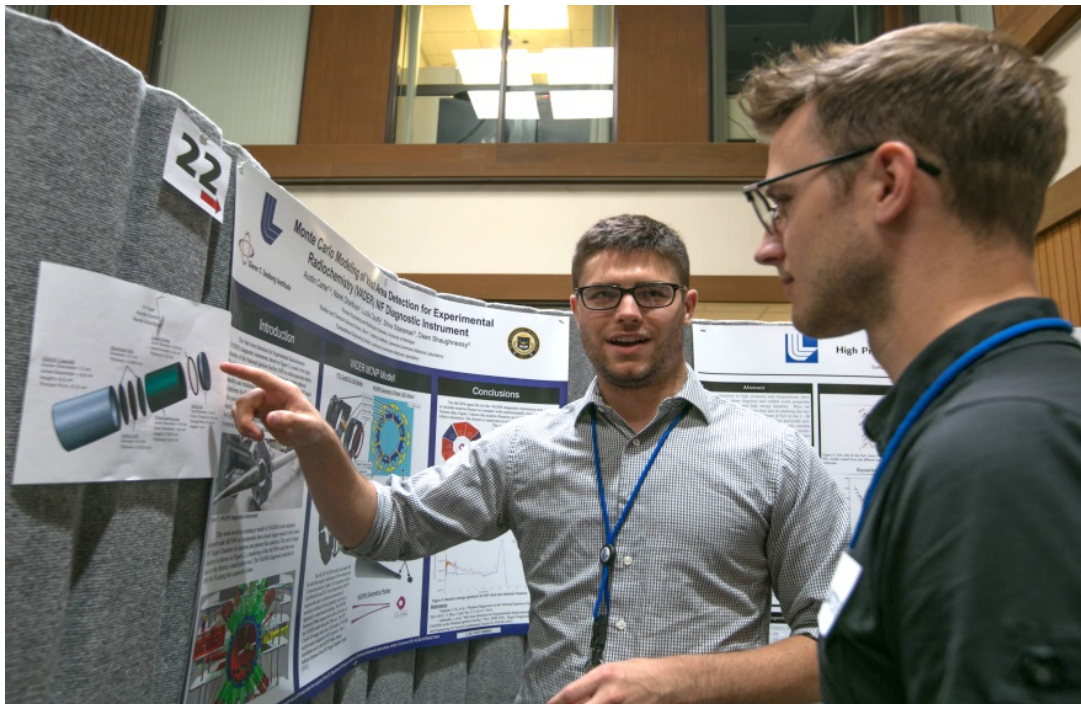
Table 3. 2016 Nuclear Forensics Summer Program Seminar Schedule

Date	Speaker	Topic
6/16/16	Naomi Marks <i>Staff Scientist, Chemical & Isotopic Signatures Group, Nuclear and Chemical Sciences Division</i>	Don't Believe Everything You Think: Forensic Signatures from the Front End of the Uranium Fuel Cycle
6/23/16	Dawn Shaughnessy <i>Group Leader, Experimental Nuclear and Radiochemistry, Nuclear & Chemical Sciences Division</i>	Nuclear Science Research with Dynamic High Energy Density Plasmas at NIF
7/6/16	Brad Esser <i>Group Leader, Environmental Radiochemistry, Nuclear & Chemical Sciences Division</i>	Characterizing California Groundwater with Isotopes: Applications to the Drought and Climate Change
7/15/16	Ruth Kips <i>Staff Scientist, Chemical & Isotopic Signatures Group Nuclear and Chemical Sciences Division</i>	Life of a Nuclear Safeguards Inspector
	Brett Isselhardt <i>Staff Scientist, Chemical & Isotopic Signatures Group, Nuclear & Chemical Sciences Division</i>	Developing Resonance Ionization Mass Spectrometry Analysis for Nuclear Forensics
7/21/16	Ross Williams <i>Staff Scientist, Chemical & Isotopic Signatures Group, Nuclear and Chemical Sciences Division</i>	Radiochronometry: A Historical Perspective
7/28/16	Mike Savina <i>Staff Scientist, Chemical & Isotopic Signatures Group, Nuclear and Chemical Sciences Division</i>	Starry Messengers: Stardust Grains Deliver Stellar News to Earth
8/5/16	Mavrik Zavarin <i>Director, Glenn T. Seaborg Institute, Physical and Life Sciences Directorate</i>	Closeout

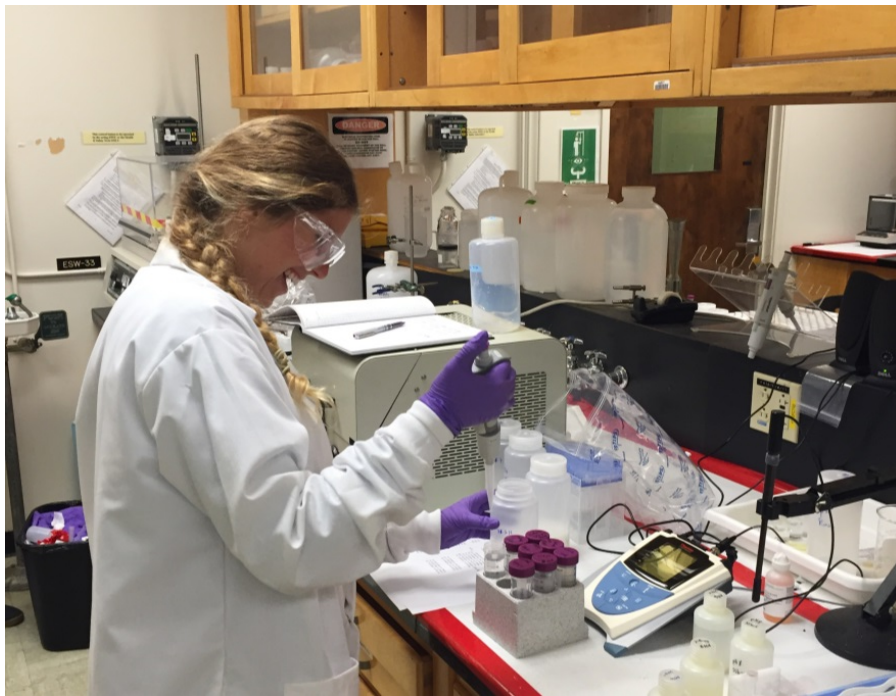
2016 Lectures and Tours



2016 Nuclear Forensics Summer Program students



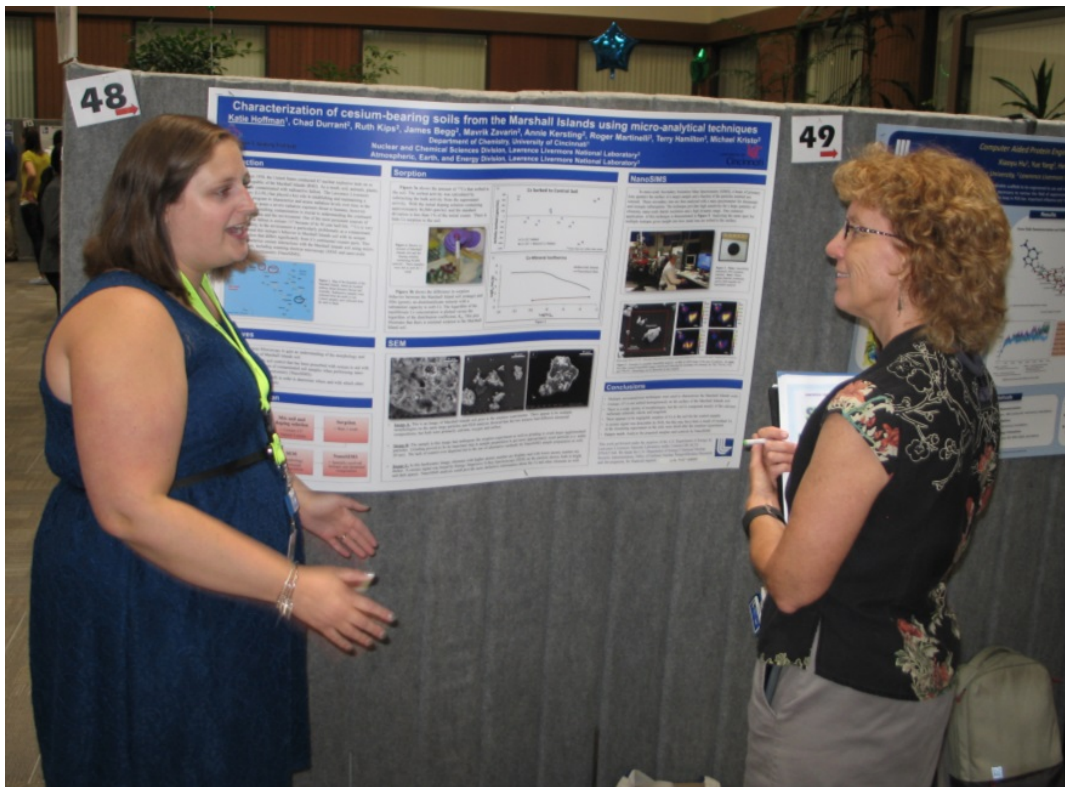
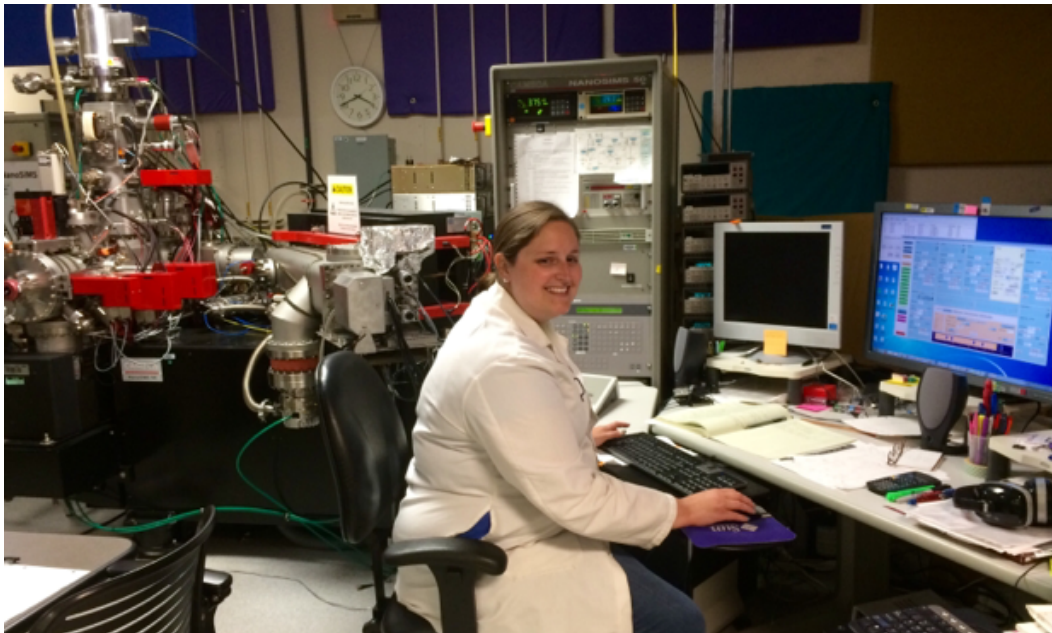
Austin Carter



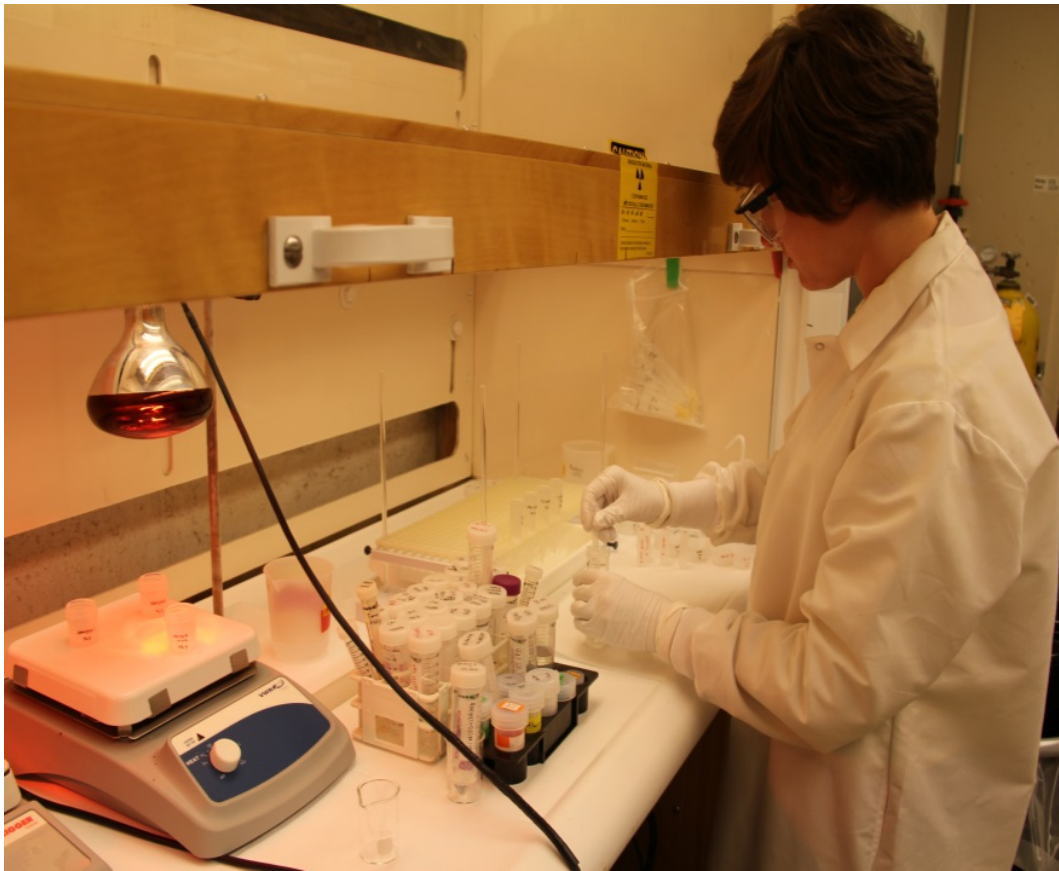
Carina Edelman



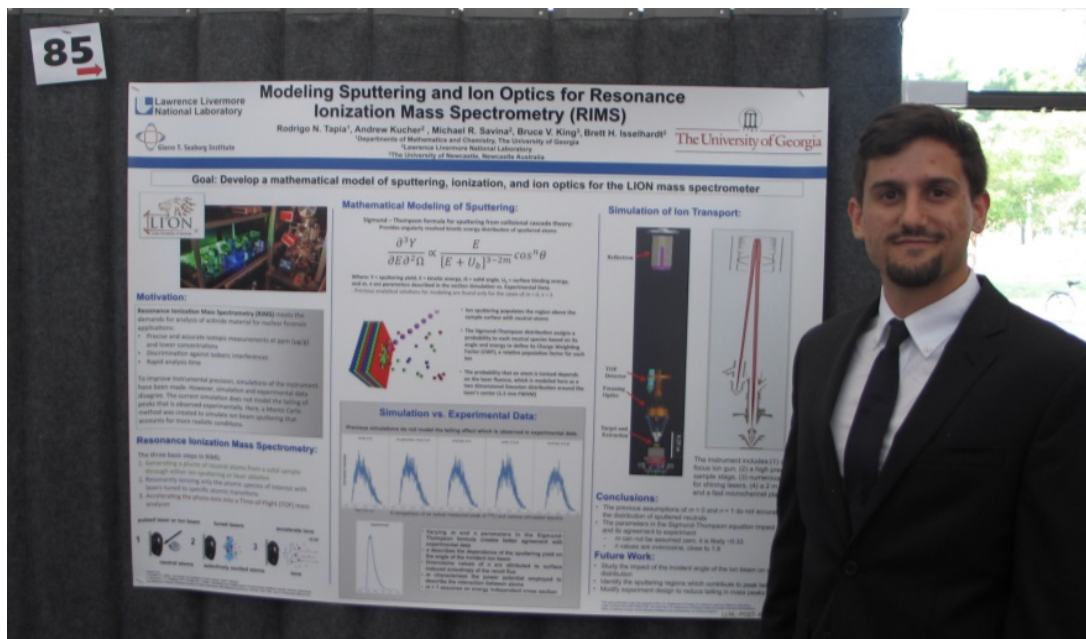
Jack Goodell



Katie Hoffman



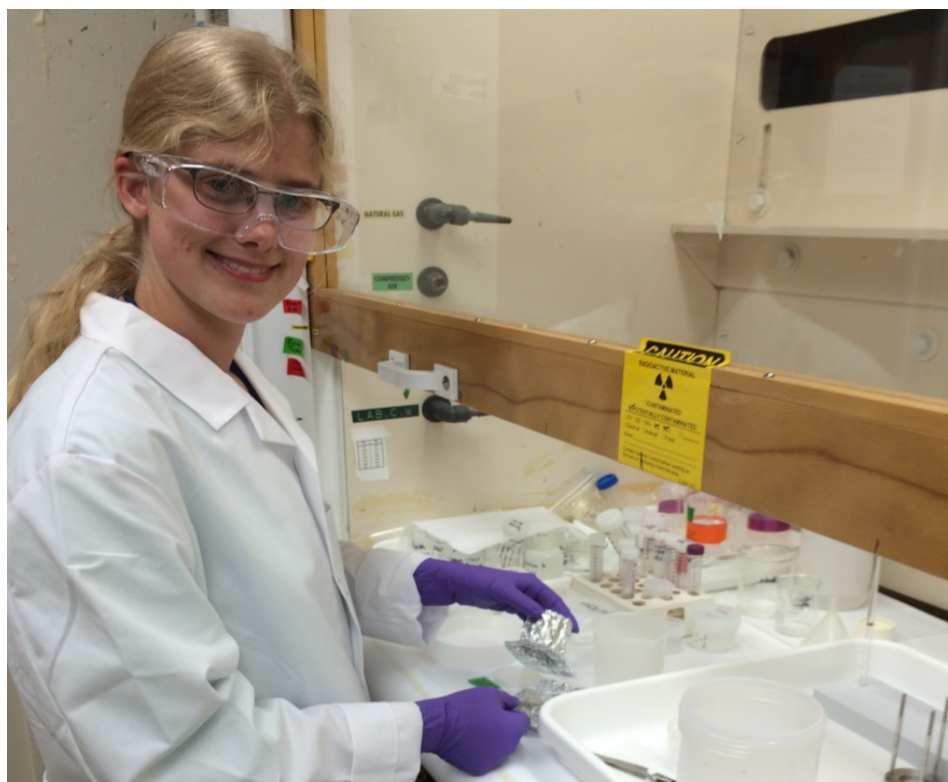
Kelly McHugh



Rodrigo Tapia

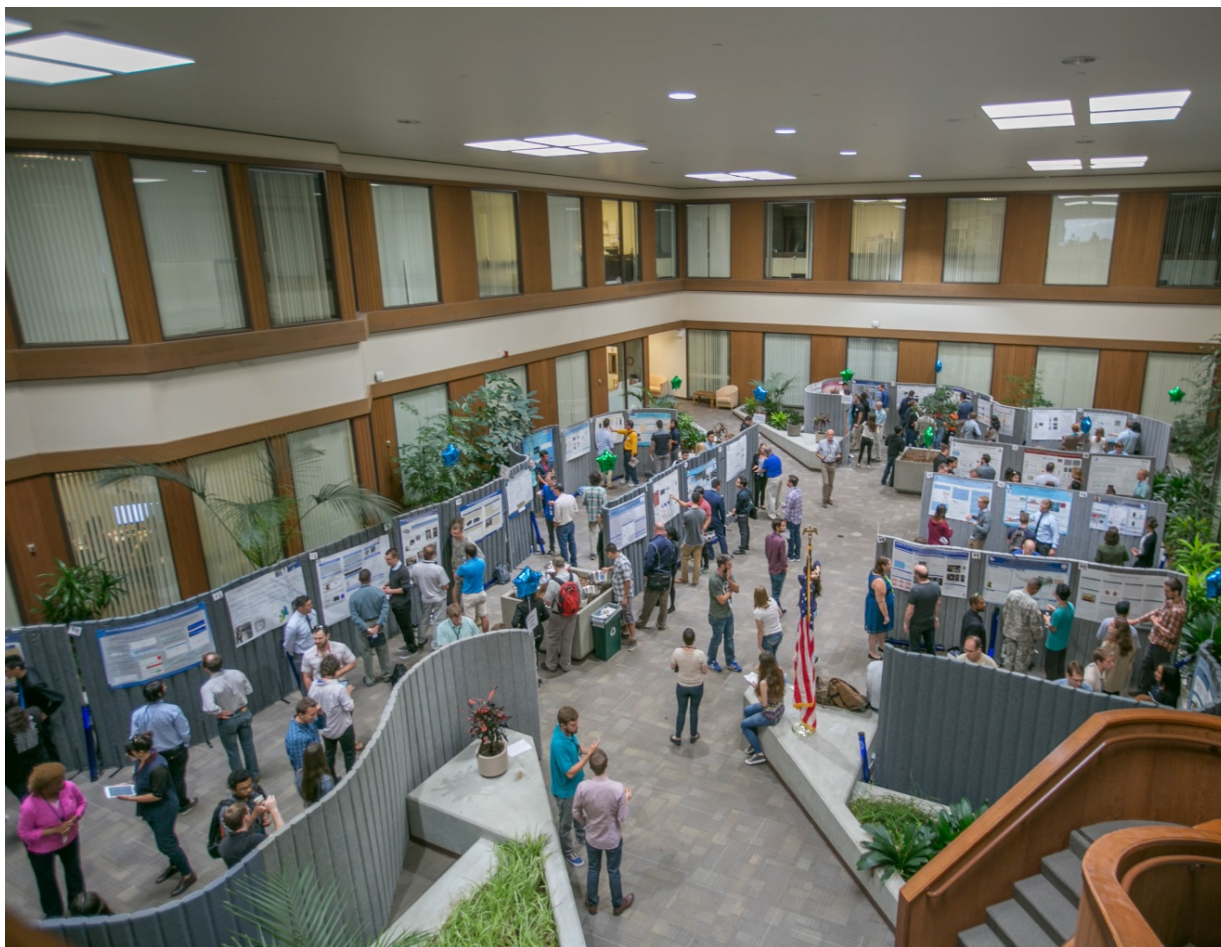


Nathan Veale



Ellen Monzo

2016 LLNL Summer Poster Session



2016 Nuclear Forensics Summer Program posters



Glenn T. Seaborg Institute

Monte Carlo Modeling of Vast Area Detection for Experimental Radiochemistry (VADER) NIF Diagnostic Instrument

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²Nuclear and Chemical Sciences Division, Glenn T. Seaborg Institute, Lawrence Livermore National Laboratory

³Computational Engineering Division, Lawrence Livermore National Laboratory



Introduction

The Vast Area Detection for Experimental Radiochemistry (VADER) diagnostic instrument, shown in Figure 1, is used in the target chamber of the National Ignition Facility (NIF) to collect post-shot debris. The debris includes radionuclides produced through neutron induced reactions on materials fielded as part of the target assembly. The NIF target assembly and traditional locations inside the target chamber can be utilized as platforms for fielding materials and measuring cross sections of interest to the post-detonation nuclear forensic community. In support of this research, understanding of the neutron environment in the NIF chamber from high yield deuterium-tritium shots is necessary for the evaluation of the neutron spectra at various locations.



Figure 1: VADER diagnostic instrument

This work involves creating a model of VADER in the radiation transport code MCNP6 to incorporate into a much larger model of the entire NIF Target Chamber for neutron and photon flux analysis. The entire Target Chamber is shown in Figure 2, a rendering of the MCNP6 code that was done in the Moritz visualization tool. The VADER diagnostic attaches to DDM 90-78 along the equatorial plane.

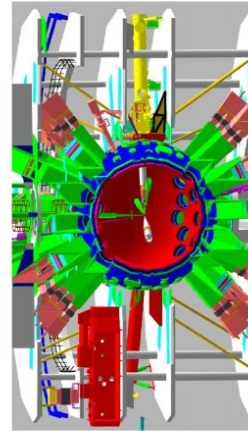
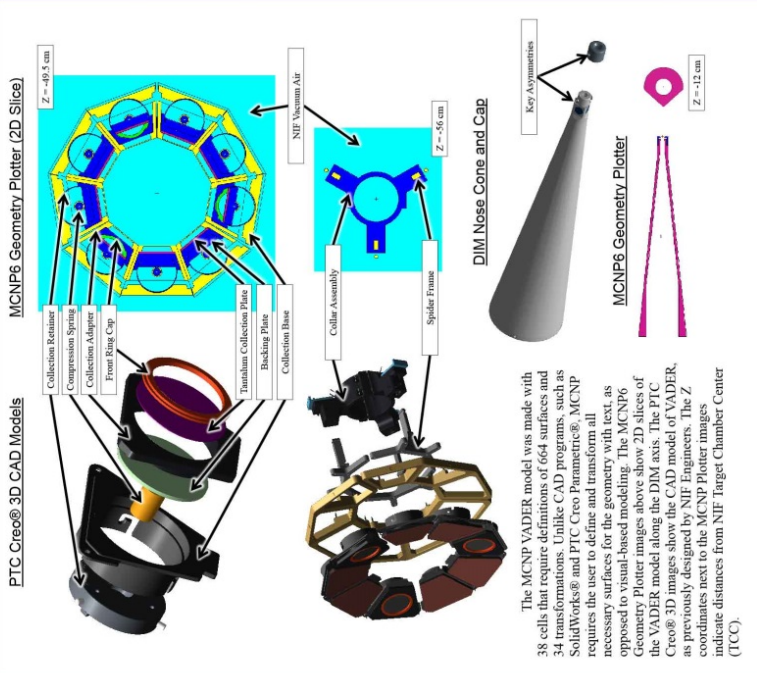


Figure 2: MCNP6 model of NIF Target Chamber

VADER MCNP Model



Conclusions

The MCNP6 input file for the VADER diagnostic instrument was used to calculate neutron fluence to compare with experimentally measured forensic data. Figure 3 shows the neutron fluences on VADER and their relative intensities. The fluence is analyzed on each of the tamalium collectors.

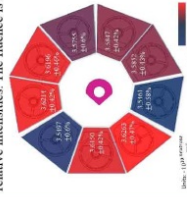


Figure 3: Neutron fluence through VADER

Simulations were performed to demonstrate the change in the neutron fluence from the source to the position of the collectors. Figure 4 shows a comparison of the shape of the source neutron spectrum to that of the neutron fluence that passes through the detector at 9.5°. Both maintain a strong peak at 14 MeV, which is the result of the deuterium and tritium fusion reaction. The detector fluence spectrum significantly deviates from the source spectrum in neutron energies lower than 14 MeV. As the neutrons collide with matter in the target chamber, they scatter to lower energies (where scatter probability increases with lower energy neutrons), which causes this deviation.

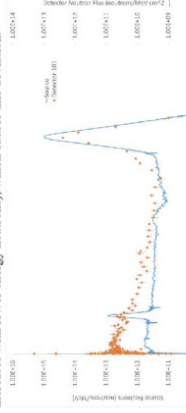


Figure 4: Neutron energy spectrum for NIF shot and detector fluence

Yemmas, C.B., et al., "Nuclear Diagnostics at the National Ignition Facility, 2013-2015," J. Phys.: Conf. Ser. 717, 012117 2016
Gallagher, J., et al., "Vast Area Detection for Experimental Radiochemistry (VADER) at the National Ignition Facility," Proc. SPIE 9591, 95910G, 2015
and Engineering for Bacterial Containment Facility 14, 95910G, 2015

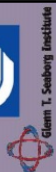
This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Experimental Pu sorption and reduction on clays: predicting environmental behavior

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²University of California, Santa Barbara



Prepared by Lawrence Livermore National Laboratory under contract number DE-AC02-09OR21400

Motivation

Globally ~2000 MT of Pu have been released to the environment. Therefore understanding plutonium transport is of vital importance. Mineral colloids have been shown to be an important transport vehicle for Pu. Pu-mineral colloid interaction studies will help develop future transport models.

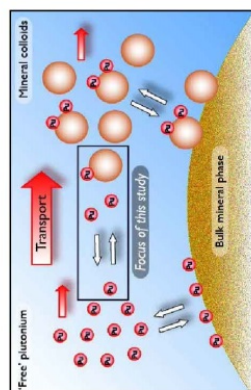


Fig. 1 Pu transport in the environment. This study focuses on the sorption of aqueous Pu to montmorillonite.

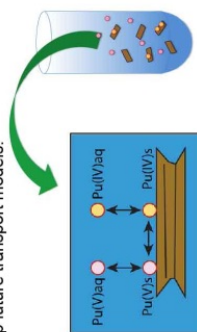


Fig. 2 Aqueous Pu(V) sorbs to the mineral particle surface and then is reduced to Pu(IV). There is ongoing debate as to what drives this reduction.

Experimental Setup

Goal: Conduct laboratory experiments to monitor plutonium sorption to clays with different compositions. Study Pu sorption in the presence of environmentally relevant mineral colloids to help develop a conceptual model for Pu transport in the subsurface environment.

1. Samples of 3 different montmorillonite clays were adjusted to pH 4, 6, and 8 spiked with 4.87 E-08 plutonium²³⁹ (V) periodically
2. Aliquots of each sample were taken
3. The supernatant solution was collected and the counts of Pu were measured via liquid scintillation counting (LSC)
4. The Fe content of each clay was measured using UV/Vis spectroscopy

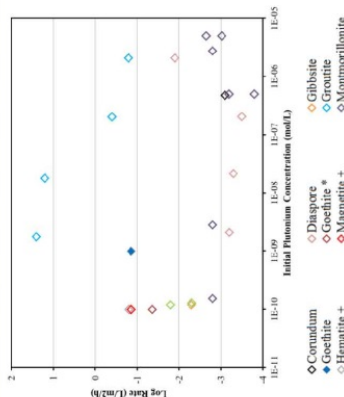


Fig. 3 Sorption rate constants for mineral phases with high Fe/Mn are greatest due to rapid reduction of Pu(V).

Results

Swy-1

Stx-1

Ssb-1

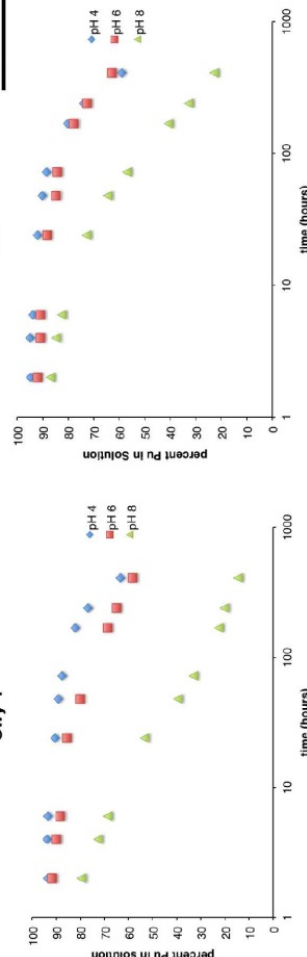


Fig. 4 Pu sorption for Swy-1 clay with 2.9 E-06 mol/g Fe

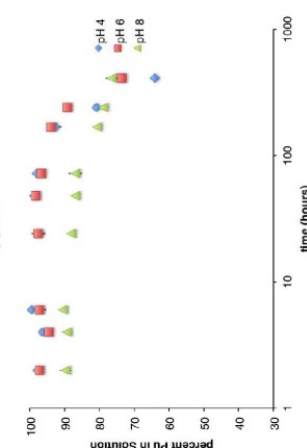


Fig. 5 Pu sorption for Stx-1 clay with 6.6 E-07 mol/g Fe

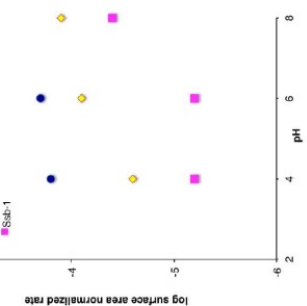


Fig. 6 Pu sorption for Ssb-1 clay with 1.2 E-07 mol/g Fe

Fig. 7 Sorption rate constants

Conclusions

- ◆ Percent plutonium in solution decreased for each of the three clays in the 17 day (408 hours) experiment.
- ◆ The Pu in solution decreased with increasing pH for each of the clays.
- ◆ Swy-1 montmorillonite had the highest total iron content, and showed the largest decrease of Pu in solution with the fastest sorption rate constants. The rate constant at pH 8 is an order of magnitude greater than those at pH 4 and 6.
- ◆ Ssb-1 montmorillonite showed a similar trend of decreased Pu in solution with slightly lower rate constants than Swy-1.
- ◆ Ssb-1 montmorillonite had the lowest total iron content, and showed little change of Pu in solution. Ssb-1 had the slowest sorption rates of all three clays, however the rate at pH 8 is significantly greater than those at pH 4 and 6.

Future Work

- ◆ Future work might mimic this experiment using these clays with the iron extracted to determine whether that affects the sorption rate of the Pu.
- ◆ Additionally, neptunium sorption experiments would provide insight to surface reduction processes, as Np(V) is not as readily reduced on the mineral surface as Pu(V).



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Modeling Neutron Irradiation and Gamma Emission Spectra of Activation Foils for Nuclear Forensics Applications

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¹ University of Maryland – Department of Chemistry & Biochemistry

² Lawrence Livermore National Laboratory – Nuclear & Chemical Sciences Division, Physical & Life Sciences Directorate

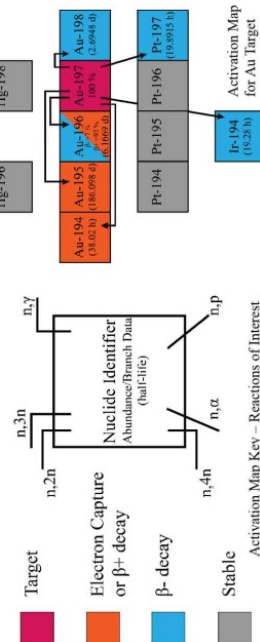


INTRODUCTION: In a world of ever tightening budgets and shrinking access to facilities and materials, simulations play a crucial role. Having a model which accurately depicts experimental setups or real world scenarios allows for multiple “experiments” to be performed at no cost. However, the value of the simulation is dependent on how well it mimics real world geometries and interactions – a simple basic model is often of little benefit. Therefore, simulations need to be as detailed as possible to be considered helpful for planning future experiments. This is especially true for nuclear forensics, as many potential situations are difficult or impossible to observe experimentally.

This work focuses on establishing a start-to-finish simulation of the neutron irradiation of metal foils and subsequent gamma spectroscopy of the activation products. By doing so, a process is provided which allows researchers to build new diagnostic tools for nuclear forensics and then test these tools in experiments, where possible.

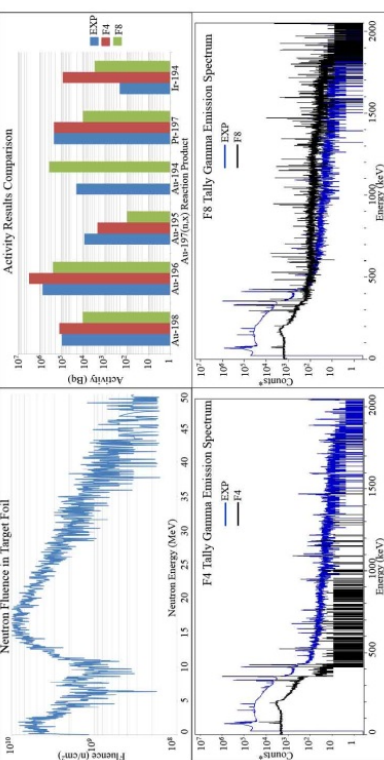
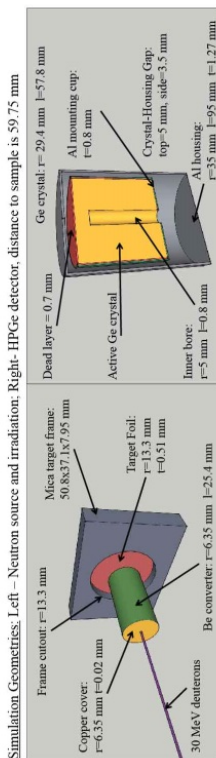
OBJECTIVES:

- Accurately represent the geometry of the irradiation setup at the Crocker Nuclear Laboratory Cyclotron
- Simulate irradiation of metal foils; identify activation products for the reactions of interest
- Accurately represent the geometry of a typical HPGe detector for gamma spectroscopy
- Simulate gamma spectroscopy measurements based on the activation results
- Compare simulated and experimental gamma spectra

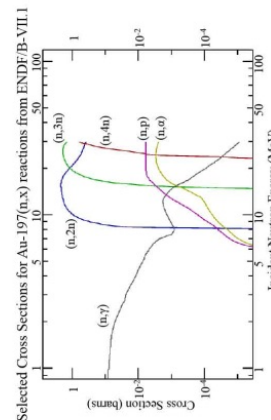


SIMULATION DETAILS:

- Using MCNP6.1 radiation transport code
- Separate simulations for irradiation and gamma spectroscopy steps
- Identified 30 MeV deuteron source for neutron production
- Neutrons produced through Be-9(d,n)B-10 reaction
- Calculate simulated activities using 2 methods
- Tally reaction rates using data only (F4 tally) – ENDF/B-VII-1
- Tally residual product nuclei using data and physics models (F8 tally)
- Using CEM03.03 and LAQSM03.03 physics models



*Simulated and experimental counts are normalized to enable qualitative comparison



Characterization of cesium-bearing soils from the Marshall Islands using micro-analytical techniques

Katie Hoffman¹, Chad Durrant², Ruth Kips², James Begg², Mavrik Zavarin², Annie Kersting², Roger Martinelli³, Terry Hamilton³, Michael Kristo²

¹Department of Chemistry, University of Cincinnati¹
²Nuclear and Chemical Sciences Division, Lawrence Livermore National Laboratory²
³Atmospheric, Earth, and Energy Division, Lawrence Livermore National Laboratory³



Introduction

From 1945 to 1958, the United States conducted 66 nuclear explosive tests on or near atolls in the Republic of the Marshall Islands (RMI). As a result, soil, animals, plants, and marine life were contaminated with radioactive fallout. The Lawrence Livermore National Laboratory (LLNL) has played a key role in establishing and maintaining a radiological survey program to characterize and assess radiation levels over time in the RMI. Fallout no longer poses a severe radiation exposure threat to humans, however characterization of the resulting contamination is crucial to understanding the continued health effects for residents and the environment. One of the most persistent sources of radiation from nuclear fallout is cesium-137 because of its 30 year half-life. ¹³⁷Cs is very soluble and its mobility in the environment is particularly problematic as a contaminant. We seek to understand this isotope's behavior in Marshall Islands soil with its unique elemental composition that differs significantly from its continental counterpart. This study seeks to characterize cesium interactions with the Marshall Islands soil using micro-analytical techniques, including scanning electron microscopy (SEM) and nano-scale Secondary Ion Mass Spectrometry (NanoSIMS).

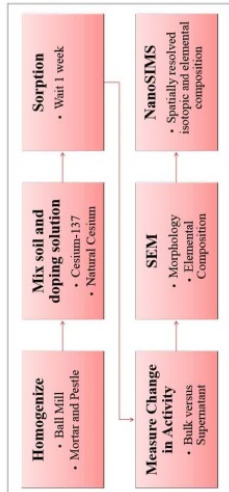


Figure 1. Map of the Republic of the Marshall Islands showing the location of the nuclear test sites and the location of the Marshall Islands in the Pacific Ocean.

Objectives

- Use Scanning Electron Microscopy to gain an understanding of the morphology and elemental composition of Marshall Islands soil.
- Prepare a Marshall Islands soil control that has been presorbed with cesium to aid with identification and quantitation of contaminated soil samples when performing nano-scale Secondary Ion Mass Spectrometry (NanoSIMS).
- Use spatially resolved NanoSIMS in order to determine where and with which other elements cesium sorbs on the soils.

Experimental Plan



Sorption

Figure 3a shows the amount of ¹³⁷Cs that sorbed to the soil. The sorbed activity was calculated by subtracting the bulk activity from the supernatant activity. With the initial doping solution containing approximately 40,000 cpm/mL and the standard deviation is less than 1% of the initial counts. There is little Cs sorption to the soil.



Figure 3b shows the difference in sorption behavior between the Marshall Islands soil (orange) and illite (green), an aluminosilicate mineral with a substantial capacity to sorb Cs. The logarithm of the equilibrium Cs concentration is plotted versus the logarithm of the distribution coefficient, K_d . This plot illustrates that there is minimal sorption to the Marshall Islands soil.

SEM

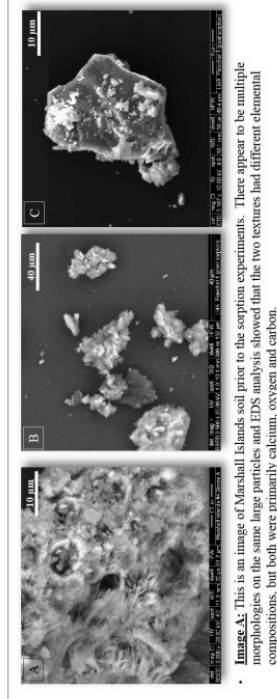


Image A: This is an image of Marshall Islands soil prior to the sorption experiments. There appear to be multiple morphologies on the same large particles and EDS analysis showed that the two textures had different elemental compositions, but both were primarily calcium, oxygen and carbon.

Image B: The sample in this image had undergone the sorption experiment as well as grinding to crush larger agglomerated particles. Grinding proved to be an important step in sample preparation to get more appropriately sized particles (i.e. under 20 um). The lack of control over dispersal led to the use of alternative methods for NanoSIMS sample preparation as well.

Image C: In this backscatter image, elements with higher atomic number are brighter and with lower atomic number are darker. A cesium signal was found by Energy Dispersive X-Ray Spectroscopy (EDS) on the particle shown, both in bright and dark spaces. NanoSIMS analysis could provide more definitive information about the Cs and other elements as well.

NanoSIMS

In nano-scale Secondary Ionization Mass Spectrometry (SIMS), a beam of primary ions spatters the surface of a target sample and a fraction of the particles emitted are ionized. These secondary ions are then analyzed with a mass spectrometer for elemental and isotopic information. The technique provides high sensitivity for a large quantity of elements, nano-scale lateral resolution and a high dynamic range. One common application of this technique is demonstrated in Figure 5. Analyzing the same spot for multiple isotopes gives insight into how metal ions are sorbed to the surface.

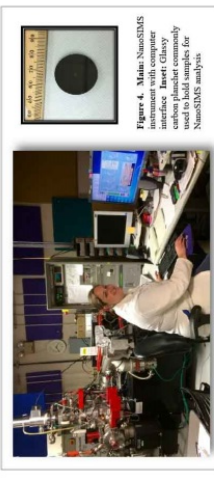
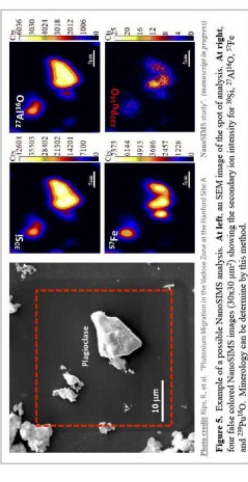


Figure 4. NanoSIMS instrument with computer interface. Heart Glassy used to hold samples for NanoSIMS analysis.



Conclusions

- Multiple microanalytical techniques were used to characterize the Marshall Islands soils
- Cesium-137 is not sorbed homogeneously on the surface of the Marshall Islands soil
- There is a wide variety of morphologies, but the soil is composed mostly of the calcium carbonate minerals, calcite and aragonite
- There appears to be negligible sorption of Cs to the soil for the control sample
- A cesium signal was detectable by EDS, but this may have been a result of residual Cs in the remaining supernatant as the soils were dried after the sorption experiment.
- **Future work:** Analyze the prepared samples and controls by NanoSIMS

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. We thank the U.S. Department of Energy's National Nuclear Security Administration, Office of Defense Nuclear Nonproliferation Research and Development, for financial support.

Th-U and Pa-U Model Ages of Bulk Uranium Radiochronometry Standards

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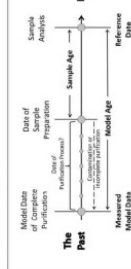
² Lawrence Livermore National Laboratory, Nuclear and Chemical Sciences Division, Glenn T. Seaborg Institute, Livermore, CA



Introduction

Many characteristics of a nuclear material can aid in the positive identification of its origin and history. One of these attributes is its model age (or calculated date of purification) which is a signature unique to each batch of material produced. Analysis of certified radiochronometry standards provides assurance of the efficacy of dating methods. This study evaluates the methods used at LLNL for ²³³Pa-²³⁵U and ²³⁰Th-²³⁴U radiochronometry. These methods have been used to analyze three commercially available uranium radiochronometry certified reference materials (CRMs): CRM 125A, CRM U630, and IRMM-1000. CRM 125A and CRM U630 were pre-existing isotopic standards that were certified based on their Th-U composition (with unknown purification dates); whereas IRMM-1000 was created to be a U-Th radiochronometry standard (prepared on a known date). A comparison of historical LLNL model ages to those newly acquired in July 2016 illustrates the validity of the methods used, the reproducibility of LLNL's results, and apparent discrepancies with certified model purification dates. Continual analysis of these materials verifies LLNL's ability to analyze unknown samples in nuclear forensics investigations and generates confidence in international, interlaboratory measurement comparisons.

Model Ages



Model ages are those calculated from the measured daughter-parent ratio. These represent the purification age if all of the assumptions below are true:

- (1) All daughter products where removed during initial purification.
- (2) The system has remained closed since purification.
- (3) Half-lives used to calculate ages are accurate.
- (4) Analytical methods produce accurate results.

Calculating model ages:

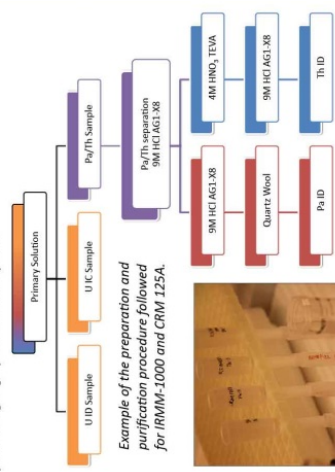
$$t = \frac{1}{\lambda_1 - \lambda_2} \ln \left[1 + \frac{R(\lambda_1 - \lambda_2)}{\lambda_1} \right]$$

Where t = age of the material;
R = daughter/parent atomic ratio;
and λ = decay constants of parent (1) and daughter (2).

References
New Brunswick Laboratory (2013). Certificate of Analysis CRM 125A Uranium (UO₂) Isotope and Radiochronometric Standard.
New Brunswick Laboratory (2013). Certificate of Analysis CRM U630 Uranium (UO₂) Isotope and Radiochronometric Standard.
Wondolowski et al. (2016). IRMM-1000A and IRMM-1000B: uranium reference materials certified for the production date based on the ²³³Pa/²³⁵U and ²³⁰Th/²³⁴U ratios. J. Radioanal. Nucl. Chem. 308: 105-111. DOI: 10.1007/s10967-015-0488-9
Williams et al. (2015). The LLNL Model Age Dating Method: A Review of Age Dating for Nuclear Forensics. International Conference on Advances in Nuclear Forensics: Countering the Evolving Threat of Nuclear and Other Radioactive Material out of Regulatory Control. LLNL-CONF-650509.
LLNL-CONF-650509

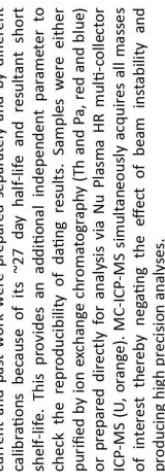
Methods

In order to verify historical results, a new primary solution of CRM 125A was made from a piece of unmodified fuel pellet as an independent evaluation of previous dissolutions. Previously prepared primary solutions were used for IRMM-1000 and CRM U630. Sample aliquots were taken from the primary solutions for U isotopic composition and U, Th, and Pa isotope dilution analyses (spiked with ²³³U, ²²⁹Th, and ²³³Pa, respectively). ²³³Pa spikes for current and past work were prepared separately and by different calibrations because of its ~27 day half-life and resultant short shelf-life. This provides an additional independent parameter to check the reproducibility of dating results. Samples were either purified by ion exchange chromatography (Th and Pa, red and blue) or prepared directly for analysis via Nu Plasma HR multi-collector ICP-MS (U, orange). MC-ICP-MS simultaneously acquires all masses of interest thereby negating the effect of beam instability and producing high precision analyses.

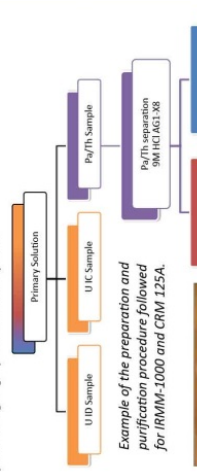


Age Dating Results

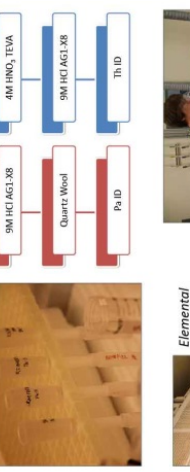
Sample ID	Reference Date (years before)	Model Uncertainty (days)	Expanded Uncertainty (days)	Certified Date (years before)
CRM 125A-1	5-Jul-16	22.00	0.19	29-Jun-04
CRM 125A-2	5-Jul-16	22.00	0.19	29-Jun-04
IRMM-1000	4-Jul-12	4.00	0.06	4-Jul-12
CRM U630 #1	20-Jul-16	27.75	0.17	10-Oct-88
CRM U630 #2	20-Jul-16	27.75	0.17	10-Oct-88



CRM 125A: 4.1% ²³⁵U (NBL, UO₂ Fuel Pellet)
LLNL Th-U and Pa-U model ages are consistently calculated to be older (~1.5 - 7 months) than the certified model age.



CRM U630: 63.4% ²³⁵U (NBL, U₃O₈)
LLNL Th-U analyses repeatedly produce model ages that are ~5 - 7 months older than the certified model age while Pa-U ages are in agreement.



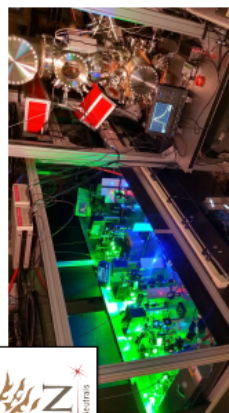
IRMM-1000: 3.6% ²³⁵U (IRMM, uranyl nitrate)
Th-U ages agree well with the known age. However, Pa-U ages are ~2 - 4 months older than the known age. These results indicate that Th was effectively removed while Pa remained residual during purification.

Summary

- Newly acquired model ages for three radiochronometry standards are in agreement with LLNL's historically reproducible values, verifying the methods used at LLNL.
- Discrepancies exist between certified model ages and measured ages depending on the standard and daughter-parent pair; however, IRMM-1000 Th-U ages are in agreement with its known production age.
- These observations highlight the importance of age-dating assumptions.

Modeling Sputtering and Ion Optics for Resonance Ionization Mass Spectrometry (RIMS)

Goal: Develop a mathematical model of sputtering, ionization, and ion optics for the LION mass spectrometer



Motivation:

Resonance Ionization Mass Spectrometry (RIMS) meets the demands for analysis of actinide material for nuclear forensic applications:

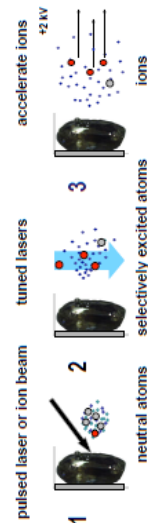
- Precise and accurate isotopic measurements at ppm ($\mu\text{g/g}$) and lower concentrations
- Discrimination against isobaric interferences
- Rapid analysis time

To improve instrumental precision, simulations of the instrument have been made. However, simulation and experimental data disagree. The current simulation does not model the tailing of peaks that is observed experimentally. Here, a Monte Carlo method was created to simulate ion beam sputtering that accounts for more realistic conditions.

Resonance Ionization Mass Spectrometry:

The three basic steps in RIMS:

1. Generating a plume of neutral atoms from a solid sample through either ion sputtering or laser ablation
2. Resonantly ionizing only the atomic species of interest with lasers tuned to specific atomic transitions
3. Accelerating the photo-ions into a Time-of-Flight (TOF) mass analyzer



References:

Greene, H. (1999). *Low-energy ion irradiation of solid surfaces*. Berlin: Springer.

Verwey, I. V., Chakrany, W. P., & Patin, M. J. (2004). Calculating time-of-flight spectra of post-ionized sputtered neutrals. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 219-220, 1061-1067. doi:10.1016/j.nimb.2004.01.211

Mathematical Modeling of Sputtering:

Sigmund – Thompson formula for sputtering from collisional cascade theory:

Provides angularly resolved kinetic energy distribution of sputtered atoms

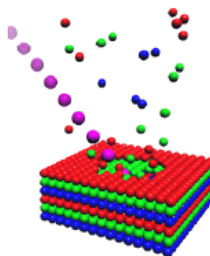
$$\frac{\partial^3 Y}{\partial E \partial^2 \Omega} \propto \frac{E}{[E + U_b]^{3-2m}} \cos^n \theta$$

Where: Y = sputtering yield, E = kinetic energy, Ω = solid angle, U_b = surface binding energy, m , n are parameters described in the section Simulation vs. Experimental Data.

Previous analytical solutions for modeling are found only for the cases of $m = 0$, $n = 1$

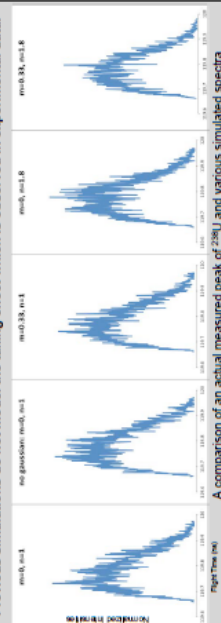
1. **Formal definition:** A formal definition is a precise, unambiguous statement that defines a term or concept. It typically follows a specific structure, often using the format: "X is a Y that Z." For example, "A triangle is a polygon with three sides." Formal definitions are essential for clarity and consistency in technical writing, legal documents, and scientific research.

- Ion sputtering populates the region above the sample surface with neutral atoms
- The Sigmund-Thompson distribution assigns a probability to each neutral species based on its angle and energy to define its Charge Weighting Factor (CWF), a relative population factor for each ion
- The probability that an atom is ionized depends on the laser fluence, which is modeled here as a two dimensional Gaussian distribution around the center of mass (1.5 mm FWHM)



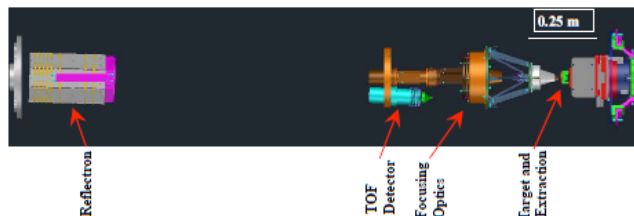
Simulation vs. Experimental Data:

Previous simulations do not model the tailing effect which is observed in experimental data.

[illegible]

- Varying m and n parameters in the Sigmund-Thompson formula creates better agreement with experimental data
- n describes the dependence of the sputtering yield on the angle of the incident ion beam
- Overcosine values of n are attributed to surface induced anisotropy of the recoil flux
- m characterizes the power potential employed to describe the interaction between atoms
- $m = 1$ assumes an energy independent cross section

Simulation of Ion Transport:



The instrument includes: (1) a fine focus ion gun, (2) a high precision sample stage, (3) numerous viewports for shining lasers, (4) a 2 m flight tube, and a fast microchannel plate detector

Conclusions:

- The previous assumptions of $m = 0$ and $n = 1$ do not accurately model the distribution of spattered neutrals
- The parameters in the Sigmund-Thompson equation impact peak shape and its agreement to experiment
 - m can not be assumed zero, it is likely -0.33
 - n values are overcosine, close to 1.8

Future Work:

- Study the impact of the incident angle of the ion beam on sputtering distribution
- Identify the sputtering regions which contribute to peak tails
- Modify experiment design to reduce tailing in mass peaks





Hydrology of the SPRUCE Climate Change Experiment

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¹California State University East Bay, Hayward, Department of Earth and Environmental Sciences

²Nuclear and Chemical Sciences Division, Lawrence Livermore National Lab

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Overview

The Spruce and Peatland Responses Under Climatic and Environmental Change (SPRUCE), conducted by Oak Ridge National Laboratory, experiment seeks to understand the impact of climate change upon the USDA Forest Service Marcell Experimental Forest, located approximately 300m north of Grand Rapids, MN. In order to respond to the potential CO₂ sequestration are used to study the potential ecosystem responses to future climates.

This ecosystem, which is located at the margin of the boreal Peatland forest in Northern Minnesota, is considered especially vulnerable to the effects of climate change because of feedbacks associated with the high carbon content of the ecosystem.



Aerial view of the SPRUCE site enclosures.

Our work focused on characterizing the hydrology of the enclosures; specifically, measuring vertical conductivity and analyzing isotopic tracers (H_2 , ^{18}O , 3H) to understand the transport of dissolved organic carbon (DOC) flux through the upper three meters. The peat itself is two to three meters deep, overlying glacial till and a sandy aquifer.

Variations of stable isotopes (H_2 , ^{18}O) of water can reveal seasonal recharge of precipitation. Tritium (3H) is a radioactive isotope with a 12.32 year half-life and its concentration relates to the residence time of peat pore water.

Carbon cycling through the saturated and unsaturated zones can help us understand how methane producing bacteria will respond to an influx of greenhouse gases.

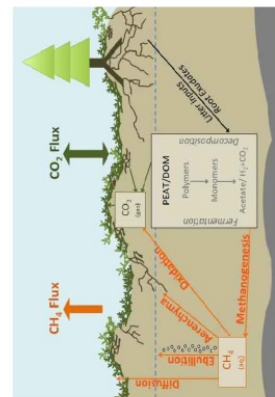


Figure: Diagram of carbon cycling at the SPRUCE site.

Source: McFarlane, K., LLNL.

Methods

Field Sampling



Samples were collected across a two year timespan from wells at several depths from enclosures with ambient (plot S6), +4.5C (plot S13) and +9C (plot S16) temperature forcing

Stable Isotope Analysis



Both precipitation and peat pore water samples were analyzed for $\delta^{18}O$ and δ^3H by cavity ring down laser isotope analysis at CSU East Bay.

Sensors & Pump Tests



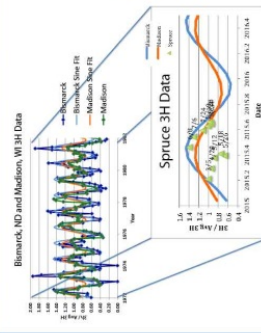
Water level monitoring methods were developed using simple pressure sensors connected to an Arduino circuit board. Physical depth measurements were also taken with beep tape at regular intervals.

Noble Gas Mass Spectrometry



Groundwater samples were analyzed for tritium by NGMS at LLNL. The instrument is able to detect small amounts of both tritium and tritogenic helium-3.

Tritium Results

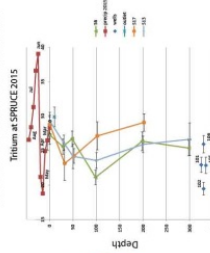


Tritium data collected at SPRUCE were plotted against data from Ottawa and Bismark, archived in the IAEA's GNIP database. Since tritium data follow a seasonal pattern in temperate climates, these two sites were volume weighted and fit to sine curves.

Tritium data match the fitted curves well in the summer months. Lower values in spring could be due to depleted storm waters from the Gulf of Mexico.

The tritium concentrations are variable with depth. The graph to the right plots tritium concentrations with depth as enclosures S17 and S13. An overlay of precipitation values by month is also included.

Activities decrease with depth up to 50cm and 100cm for the three plots, which is to be expected as tritium decays to helium-3. The cause of the increase at two meters is not entirely clear.



Summary

- Characterizing the hydrology helps to understand the rates of recharge to the aquifer, as well as how water moves within the peat. This has important implications for carbon cycling and DOC effluxes.
- Analysis of $\delta^{18}O$ and δ^3H shows the SPRUCE site receives the majority of its recharge in the spring and early summer months and 3H data show relatively young water at shallow depths with typical tritium decay profiles.
- The relatively high tritium activities found in deeper samples present an anomaly, suggesting lateral mixing to avoid low conductivity layers, or some other hydrologic mechanism.
- Future Work
 - More stable isotope samples would help determine if the trend of recharge of isotopically heavy spring precipitation continues.
 - More closely spaced tritium data are needed to understand the hydrologic mechanisms occurring at depths greater than ~1.5 meters.

Prepared at LLNL under Contract DE-AC52-07NA27344 (LLNL-POST-698596)

Auxiliary-Field Quantum Monte Carlo for UO_2^{2+}

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We have attempted to use Auxiliary-Field Quantum Monte Carlo to calculate the energy and geometry of the uranyl ion. While this technique holds promise as a scalable computational method for handling large systems like the actinides, further work is necessary to determine how it may best be applied.

Motivation

Characterizing compounds containing the uranyl ion, UO_2^{2+} , is of interest for both security and environmental reasons. However, obtaining experimental data can often be a challenge. This makes computational studies of these compounds very important.

The large number of electrons in these systems limits the level of theory that can be applied. Quantum Monte Carlo is a highly scalable technique and may achieve more accurate results with a lower computational cost.



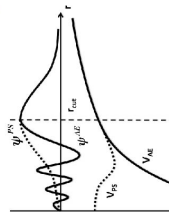
Computational Chemistry 101

The Schrödinger equation cannot be solved exactly for systems with multiple electrons. Instead, approximate methods must be employed to solve for the ground state energy, such as Mean Field Theory (MFT). In MFT, each electron interacts with an average of the other electrons. Many methods have been developed to better describe this interaction between electrons, but these methods are increasingly computationally expensive.

Auxiliary Field Quantum Monte Carlo (AFQMC) is a different way of solving for the ground state energy. Instead of trying to better describe electron interaction with complicated integrals, we sample over many different auxiliary fields to capture field fluctuations.

Effective Core Potentials

With 92 electrons, uranium is difficult to deal with computationally. Rather than treating all electrons explicitly, core electrons are replaced with an effective core potential (ECP). The more electrons that are treated explicitly, the more accurate (but expensive) the calculation becomes.



AFQMC Theory

The Hamiltonian for a multi-electron system,

$$\hat{H} = -\frac{\hbar^2}{2m} \sum_{m=1}^M \nabla_m^2 + \sum_{m=1}^M V_{ext}(r_m) + \sum_{m<n}^M V_{int}(r_m - r_n),$$

can be broken into one-body and two-body elements,

$$\hat{H} = \hat{H}_1 + \hat{H}_2.$$

In AFQMC, this Hamiltonian is used to propagate a trial wave function, $|\psi_T\rangle$, through imaginary time to converge on the ground state, $|\psi_0\rangle$. This propagation is done iteratively.

$$|\psi_0\rangle \propto \lim_{\tau \rightarrow \infty} e^{-\tau \hat{H}} |\psi_T\rangle$$

$$|\psi_{n+1}\rangle \propto e^{-\Delta\tau \hat{H}} |\psi_n\rangle$$

The Hubbard-Stratonovich transformation is used to write the two-body elements of the Hamiltonian as one-body interactions with a set of random external auxiliary fields. This gives

$$e^{-\Delta\tau \hat{H}} = \int d\mathbf{x} p(\mathbf{x}) B(\mathbf{x}),$$

where $p(\mathbf{x})$ is the probability density function (PDF), and $B(\mathbf{x})$ is the propagator.

In each time step, an auxiliary field configuration, \mathbf{x} , is sampled from the PDF and used to construct the propagator. The wave function is deconstructed into "walkers", which are propagated to generate a new wave function. This is continued until convergence is reached.

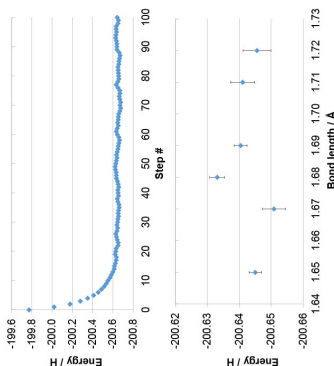
U-O Benchmarks

Before running AFQMC simulations, the U-O bond length in various uranyl compounds was benchmarked using a variety of different conventional computational chemistry methods. Results for UO_2^{2+} , in A_h , are shown below.

ECP	DFT-LDA	DFT-B3LYP	MP2	CCSD
60 e ⁻	1.698	1.696	1.719	1.679
78 e ⁻	1.693	1.682	1.708	1.688

Preliminary Results

AFQMC simulations were run at a series of different uranyl bond lengths. A single Slater determinant was used as the trial wave function. A sample simulation and summary of the calculated energy vs. bond length are shown below.



Future Work

Our preliminary results are inconclusive and this method must be further investigated. It is likely that using a single Slater determinant to describe the trial wave function is inadequate for this complicated system. Simulations using multiple determinants are ongoing. In addition, results may improve with the use of smaller ECPs. Thus, despite our results, this technique still shows promise as a method of calculating actinide properties.

Reference

S. Zhang et al. (1997). *Phys. Rev. B*, 55, 7464.

Optimization of Uranium Molecular Deposition for Alpha Counting Sources

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¹University of Minnesota Duluth, ²Lawrence Livermore National Laboratory

Introduction

Precision measurements of the ^{234}U , ^{235}U , and ^{238}U decay constants (λ) are critical for nuclear forensic analysis and geochronology. The $\lambda_{\text{U-234}}$, $\lambda_{\text{U-235}}$ and $\lambda_{\text{U-238}}$ are used for U-Pb chronometry and to calibrate geologically important decay constants.^{1,2} The ^{234}U half life is important for $^{237}\text{Pu}/^{234}\text{U}$ chronometry applicable to both nuclear forensic and geologic time scales.³ Lawrence Livermore National Laboratory (LLNL) is leading an effort to re-determine $\lambda_{\text{U-234}}$, $\lambda_{\text{U-235}}$ and $\lambda_{\text{U-238}}$.

^{234}Th and ^{235}U are being determined via direct alpha counting experiments using a low geometry alpha counter and an $\alpha\text{-}\gamma$ coincidence counter. These experiments require homogenous sources with well-constrained geometries and atomic concentrations. This poster presents research to characterize and optimize source production methods.

Methods

Molecular deposition was chosen as the best source preparation method to investigate because it is a quantitative process resulting in thin, uniform layers of material.³ The type of platinum anode and cathode polishing were targeted for optimization because they have been shown to improve source homogeneity.³ Teflon cells were tested because they should resist uranium sorption and increase plasma yield. All experiments used deuterated uranium (DU).

Figure 1 displays three circular samples and a ruler for scale. From left to right: a thin wire solid disc, a mesh, and a platinum electrode. The ruler indicates the diameter of the samples is approximately 1.25 cm.

From left: Thin wire solid disc, mesh, and thick wire solid disc electrodes.

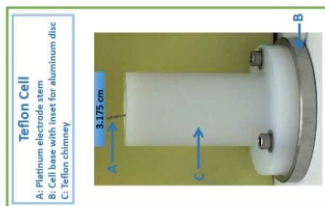
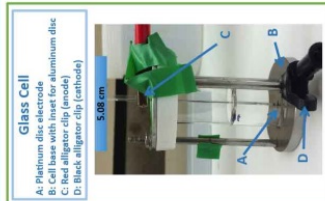
In the first series of experiments, plates were created to test source homogeneity. Plates (parameters at right) were produced in five steps:

- Dry down DU from 2M HNO_3 solution
- Fume with 50 μL 16M HNO_3 2X to destroy organics
- Dissolve DU in 50 μL H_2O
- Quantitatively transfer DU to plating cell using 12 mL isopropyl alcohol
- Plate DU out of solution

A second set of experiments tested different methods of polishing the aluminum plates prior to deposition. Plates were electropolished using 4.14 mL HClO_4 and 7.86 mL acetic anhydride at 113 mA, 6 V³, or chemically polished using 5.200 mL H_2O_2 , 2.44 mL HNO_3 , 31.090 mL H_3PO_4 , and 2.488 mL acetic acid.

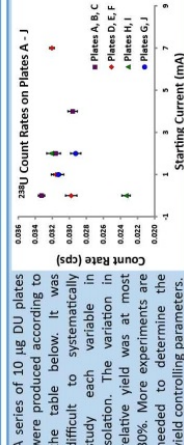
Plates were characterized by a combination of alpha counting, autoradiography, and scanning electron microscopy (SEM). Activity distribution in the sample was imaged by autoradiography using a GE Typhoon FLA 7000 scanner and SR-type phosphor screen image plates scanned at 626 v after a 22 hour exposure, and processed using ImageJ. SEM images were acquired using an FEI Inspec F SEM equipped with an Everhart-Thornley secondary-electron detector.

Plating Cells



Molecular Deposition Plates

A series of 10 µg DUJ plates were shown according to the above protocol. The results are shown in Figure 1. The results are difficult to systematically compare each variable in isolation. The variation in the total yield was at most 10%. More experiments are required to determine the yield controlling parameters.

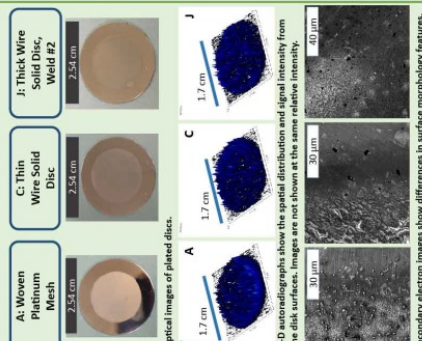


Molecular Deposition Plating Parameters

Plate	A	B	C	D	E	F	G	H	I	J
Type of Cell	PTFA Teflon	Glass	Glass	Glass	Glass	Glass	PTFA Teflon	PTFA Teflon	Glass	
Platinum Anode	Mesh	Thin wire	Thin wire	Thin wire	Thin wire	Thin wire	Mesh	Mesh	Thin wire	Thin wire
Agitation of Solution	None	None	None	None	None	None	None	None	None	None
Starting Electrode Height Above Aluminum (mm)	-3	-3	-3	2.4	5.5	5.7	4.9	-3	-3	-5
Yes electrode height varied?	No	No	No	Yes	No	Yes	No	No	No	No
starting current (mA)	4	2	<1	7	1	<1	1	<1	2	2
rising Current (mA)	3	2	<1	5	<1	1	1	<1	1	1
rising Time (minutes)	90	90	86	90	90	90	90	90	90	90

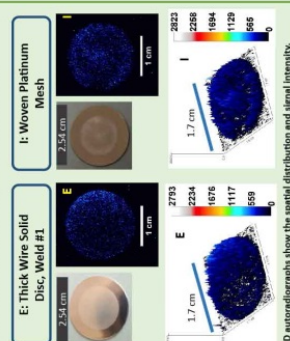
Electrode Type and Homogeneity

Of the three platinum electrodes tested, the solid platinum disc attached to a thick wire (J) produced the most homogeneous plate (see below). The solid disc had the most uniform platinum distribution and the thick wire prevented electrode tilting.



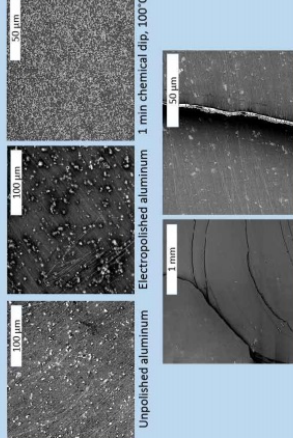
Tilt Effects on Activity Distribution

on-parallel orientation of the discs relative to the Al anode. The Pt mesh electrode (below, Plate E) resulted in more DU on the portions of the plate closest to the Pt electrode. The DU was distributed most heavily on areas directly below the thick wire frame of the Pt mesh electrode and less on the areas covered by the thin mesh (below, Plate I).



Comparison of Polishing Approaches

Secondary electron images of the electropolished and 1 minute chemically polished plates reveal signs of etching and pitting, which is non-ideal for deposition.



The 30 second, 23°C chemically polished plate shows the effects of aluminum flaking. Left: Flaking edges. Right: Close up of flaked edge.

Conclusions

- The amount of DU deposited varied by up to 30 %.
- A solid platinum disc anode with a thick shaft produced the most homogeneous uranium sources.
- Electrode tilt and platinum electrode location influenced where the DU was concentrated on the source.
- Future work will include:
- Plating five copies of a 10 µg DU plate with a thick wire platinum disc electrode to verify reproducibility
 - Optimize fine polishing of aluminum plates
 - Characterize self-absorption behavior of plates with >10 µg DU
 - Experiments to determine the yield controlling parameters

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