2013 LLNL Nuclear Forensics Summer Program

Glenn T. Seaborg Institute
Lawrence Livermore National Laboratory
Physical and Life Sciences
Livermore, CA 94550

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Sponsors:
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LLNL: Glenn T. Seaborg Institute, Physical and Life Sciences Directorate
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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
The Lawrence Livermore National Laboratory (LLNL) Nuclear Forensics Summer Program is designed to give both undergraduate and 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) to gain a better understanding of the multi-disciplinary, on going science at LLNL.

Currently called the Nuclear Forensics Summer Program, this program began 13 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 Nuclear Forensics Summer Program 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 and have the student experience conducting research at the Lab. We select students who are majoring in 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, computational analysis, radiation detection, and nuclear engineering in order to strengthen the “pipeline” for future scientific disciplines critical to DHS (DNDO), NNSA.

This is a competitive program with over 80 applicants for the 7–10 slots available. Students come highly recommended from universities all over the country. For example, this year we hosted students from Texas A&M, University of Cincinnati, Washington State University, University of Nevada, Las Vegas, and University of Missouri. (See Table 1).
This year’s students conducted research on such diverse topics as actinide (Np, U, Pu) isotopic fingerprinting, statistical modeling in nuclear forensics, actinide analysis for nuclear forensics, environmental radiochemistry, radiation detector materials development, coincidence counting methods, nuclear chemistry, and heavy element separations chemistry (see Table 2.) Graduate students are invited to return for a second year at their mentor’s discretion. We encourage the continuation of research collaboration between graduate student, faculty advisor, and Laboratory scientists.

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 experts from both within LLNL and external to it. Speakers are able to discuss the importance of their work in the context of advances in the field of nuclear forensics.

Graduate and undergraduate students on fellowships such as the Nuclear Forensics Graduate Fellowship are invited into our summer program. They usually come for 12 weeks and can return the following summer or stay throughout the year depending on their research needs. This year we had five Nuclear Forensics Fellows, four in the graduate program and one in the undergraduate program (Table 1, noted by an asterisk).

We also host students who are participating in the DOE-sponsored “Summer School in Radiochemistry” course held at San Jose State University and have recruited from this program. They come for a day, meet our summer students, see the research our students are doing, and tour our facilities. Staff scientists also participate in the Nuclear Forensics Undergraduate Summer Program sponsored by DHS-DNDO (FY13 held at University of Nevada, Las Vegas). This year the students come to LLNL for two days to meet our summer students, learn about their research, meet the staff and learn more about work at a national laboratory.

We use our summer program to create a successful pipeline of top-quality students from universities across the U.S. Since 2002, 42 students have returned and/or conducted their graduate research at LLNL:

- 12 became postdoctoral fellows at LLNL.
- 3 became postdoctoral fellows at other national labs.
- 7 were hired as career scientists at LLNL.
- 3 were hired as career scientists at other national labs.
- 3 were hired as faculty in the area of nuclear forensics/radiochemistry/nuclear science.

A big factor in the success of this program is the dedication of the staff scientists who volunteer to mentor the summer students. In FY13, funding from the Nuclear Forensics Graduate Mentoring Program (sponsor: DNDO) helped to partially support the time staff took to teach the summer interns. Staff scientists were able to take the necessary time to develop an appropriate summer project for their student, oversee necessary safety training, and dedicate more time to helping the interns maximize their productivity and scientific potential.

The posters presented at our Laboratory Student Poster Day are included at the end of this report. Two of our students won ‘best poster’ award out of about 250 poster presentations.
<table>
<thead>
<tr>
<th>Student</th>
<th>Major</th>
<th>University</th>
<th>Year</th>
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<tr>
<td>Marisa Alfanso</td>
<td>Chemistry</td>
<td>Texas A&amp;M University, College Station</td>
<td>Grad</td>
</tr>
<tr>
<td>Lucas Boron-Brenner</td>
<td>Radiochemistry</td>
<td>University of Nevada, Las Vegas</td>
<td>Grad</td>
</tr>
<tr>
<td>Megan Deeds</td>
<td>Chemistry</td>
<td>University of Cincinnati</td>
<td>Grad</td>
</tr>
<tr>
<td>Chad Durrant*</td>
<td>Nuclear Engineering</td>
<td>The Pennsylvania State University</td>
<td>Grad</td>
</tr>
<tr>
<td>Marc Fitzgerald*</td>
<td>Chemistry</td>
<td>University of Nevada, Las Vegas</td>
<td>Grad</td>
</tr>
<tr>
<td>April Gillens*</td>
<td>Environmental Engineering and Earth Sciences</td>
<td>Clemson University</td>
<td>Grad</td>
</tr>
<tr>
<td>Mitch Goshert</td>
<td>Chemistry</td>
<td>University of Missouri</td>
<td>Grad</td>
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<tr>
<td>Patrick Harms</td>
<td>Geology</td>
<td>Cal State University, East Bay</td>
<td>Grad</td>
</tr>
<tr>
<td>Corey Keith</td>
<td>Nuclear Engineering</td>
<td>Texas A&amp;M University, College Station</td>
<td>Grad</td>
</tr>
<tr>
<td>Morgan Kelley</td>
<td>Radiochemistry</td>
<td>Washington State University</td>
<td>Grad</td>
</tr>
<tr>
<td>Jeff Rolfes*</td>
<td>Radiochemistry</td>
<td>University of Nevada, Las Vegas</td>
<td>Grad</td>
</tr>
<tr>
<td>Rodrigo Tapia**</td>
<td>Mathematics and Chemistry</td>
<td>University of Georgia</td>
<td>Undergrad</td>
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*= Nuclear Forensics Graduate Fellows  
**= Nuclear Forensics Undergraduate Intern
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<tr>
<th>Student</th>
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<th>Project</th>
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<td>Marisa Alfano</td>
<td>Dawn Shaughnessy</td>
<td>Analysis of Vanadium Activation Products from NIF Irradiations</td>
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<td>Lucas Boron-Brenner</td>
<td>Gary Eppich</td>
<td>U Isotopic Measurements of Fallout Spherules Using Isotope Dilution Mass Spectrometry</td>
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<td>Chad Durrant*</td>
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<td>Mitch Goshert</td>
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<td>Patrick Harms</td>
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<td>Tritium and Stable Isotope Survey of California Surface Water</td>
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<td>Morgan Kelley</td>
<td>Jennifer Jo Ressler</td>
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<td>Jeff Rolfes*</td>
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<td>Rodrigo Tapia**</td>
<td>Brett Isselhardt</td>
<td>Using Computed Tomography to Non-Destructively Characterize Radioactive Fallout in 3D</td>
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* = Nuclear Forensics Undergraduate Intern  
** = Nuclear Forensics Graduate Fellows
Table 3. Seminar Schedule

<table>
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<tr>
<th>Date</th>
<th>Speaker</th>
<th>Topic</th>
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<tbody>
<tr>
<td>6/20/13</td>
<td>Dawn Shaughnessy&lt;br&gt;Group Leader, Experimental Nuclear and Radiochemistry, Chemical Sciences Division</td>
<td>Superheavy Element Discovery at LLNL</td>
</tr>
<tr>
<td>6/27/13</td>
<td>Mona Dreicer&lt;br&gt;Deputy Program Director for Nuclear and Domestic Security and Deputy Director of the Center for Global Security Research (CGSR)</td>
<td>Treaty Monitoring and Verification</td>
</tr>
<tr>
<td>7/3/13</td>
<td>Arthur Rodgers&lt;br&gt;Scientist, Earth and Energy Division</td>
<td>Prompt Forensics with Speed-of-Sound Data</td>
</tr>
<tr>
<td>7/11/13</td>
<td>Stephan Friedrich&lt;br&gt;Scientist, Physics Division</td>
<td>Superconducting Gamma Spectrometers with Ultra-high Energy Resolution for Nuclear Forensics</td>
</tr>
<tr>
<td>7/18/13</td>
<td>James Begg, Postdoc, Chemical Sciences Division&lt;br&gt;Greg Brennecka, Postdoc, Chemical Sciences Division</td>
<td>Actinides in the Environment&lt;br&gt;Geolocation of Nuclear Materials for Forensic Applications</td>
</tr>
<tr>
<td>7/25/13</td>
<td>Brett Isselhardt, Scientist, Chemical Sciences Division</td>
<td>Case Studies in Nuclear Forensics</td>
</tr>
<tr>
<td>8/1/13</td>
<td>Kim Budil&lt;br&gt;N Program Manager, Nuclear Counterterrorism, Global Security Principal Directorate</td>
<td>A Journey Through the Nuclear Core: A Career at LLNL</td>
</tr>
<tr>
<td>8/8/13</td>
<td>Annie Kersting&lt;br&gt;Director, Glenn T. Seaborg Institute, Physical and Life Sciences Directorate</td>
<td>Closing out the program</td>
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Analysis of Vanadium Activation Products from NIF Irradiations

Marisa Alfonso1,2, Evgeny Tereshatov2, John Despotopoulos1,3, Narek Gharibyan1, Ken Moody1, Dawn Shaughnessy1

1Lawrence Livermore National Laboratory, Chemical Sciences Division, PLS
2Texas A&M University
3University of Nevada – Las Vegas

Background

National Ignition Facility (NIF) shot at LLNL has capabilities of producing the world’s largest neutron flux - 10^20 neutrons per shot. The instantaneous release of neutrons makes it possible to measure nuclear reaction cross sections without having to correct for low-time neutron contributions, an issue with many other neutron sources. Also, NIF can be used to produce radiolabels for nuclear forensics exercises.

192 laser beams

1. Deuterium-tritium fusion creates 14.1 MeV neutrons
2. Low energy neutrons scatter off the remaining deuterium-tritium andadiator
3. Holmium material, mostly gold, is activated and debris is collected
4. Tracer material added to the capsule is activated and collected

To the right is the Diagnostic Insertion Manipulator (DIM) used to position solid radiolabel chemistry (SRC) collectors 50 cm from the hohlraum. Two-inch diameter capacity disks (95.25% 59V, 0.25% 58V) were used during these NIF shots. Vanadium metal is frequently used as a SRC collector due to its high melting temperature (1913°C) and V activation products do not interfere with the gamma detection of the activated gold.

Methods

Literature states that V metal dissolves in nitric acid, aqua regia, and hot hydrofluoric acid. Many different methods were tested. Passivation occurs (B and C) when using nitric acid or aqua regia to dissolve the V disk. Scandium is detected in the passivation product. Picture D shows V passivation product being dissolved by concentrated hydrofluoric acid. A mixture of both concentrated nitric and hydrofluoric acid completely dissolves the V disk (E and F). Picture G shows Sc being precipitated out of solution as ScF3 using Ce carrier. The yield of this method is 41% which may be due to formation of soluble complex such as ScF3 and/or the Ce carrier not homogenously mixing throughout the solution.

By adding the carrier to the solution while dissolving the V disk in concentrated nitric acid, the Sc does not get trapped in the passivation product and the carrier is homogeneously mixed throughout the solution. Fluoride coprecipitation yields, >90%.

Results

Above is a 3 day spectrum taken at Nuclear Counting Facility of the Sc sample after the V disk was dissolved and Sc was separated from the bulk V material. The precipitate is dissolved using 95/5 v/v nitric acid and 2 M hydrochloric acid. For this sample the ratio of 57Co/54Sc produced during the NIF shot is 0.002 ± 0.0006 and the cross section of the 57Co(n,γ)58Co is calculated to be 83 ± 17 barn. A sample from a different shot has a 57Co/54Sc ratio of 0.007 ± 0.0002 and a calculated 57Co(n,γ)58Co cross section of 132 ± 35 barn.

The two measurements agree within error and have a considerable smaller uncertainty than the evaluated cross section data.

Future Work

The assumptions made to simplify the 57Co(n,γ)58Co cross section calculation may not be correct. Assuming that the cross section of the 57Co(n,γ)58Co is the same as the 54V(n,γ)55V reaction, the calculated cross section for 57Co(n,γ)58Co would decrease by 25 barn. A more accurate way to measure both reaction cross section would be to also analyze an enriched V foil after an NIF Irradiation. To the left is a foil holder that can be placed behind a SRC collector during future NIF shots. The foil would also have less bulk material, making sample preparation easier and faster.

Acknowledgement

I would like to thank the Glenn T. Seaborg Institute for giving me this amazing opportunity to spend a summer working at LLNL. Also, I would like to especially thank Evgeny and also Dawn, Ken, Narek, and John for their guidance during this project.

References

URANIUM ISOTOPIC MEASUREMENTS OF FALLOUT SPHERULES USING ISOTOPE DILUTION MASS SPECTROMETRY

Lucas Boron-Brenner¹, Gary Eppich², Kim Knight², and Ralf Sudowe¹

University of Nevada Las Vegas,¹ and Lawrence Livermore National Laboratory²

Goal: To determine uranium concentration and isotopic composition as a function of fallout glass size

INTRODUCTION

Falling glass found during above-ground nuclear tests is primarily composed of materials incorporated from the nearby environment, e.g. surface soil, as well as fission and activation products together with uranium in the test fallout. The isotopes can be used to determine the fallout glass size and isotopic composition. In this study, samples containing fallout were collected from a location on the ground area. Soil with fallout particles were sorted by size, and portions of glassy fallout were physically separated from the soils. These samples were collected from fallout of different sizes, with fallout held at natural isotopic ratio. The isotopic composition of the individual fallout particles is determined by measuring the concentration and isotopic composition of the fallout glass samples.

Sample Preparation

- Soil separation by grain size using a sieve
- Isotopic analysis of fallout glass assisted by optical microscopy

Chemical Digestion

- Digestion of 25% HNO₃ to 1 part H₂O
- Addition of HClO₄ and 3 M HCl producing stock solution

Uranium Separation I

- U-18/16 in exchange with 4 M H₂SO₄
- Column packed using 4 M H₂SO₄, 9 M HCl, and 5 M HCl
- Uranium eluted using 8 M HCl

Sample Spiking

- Addition of 3He to aliquots of stock solution, spike equilibrated

Uranium Separation II

- Acetate ion exchange with 9 M HCl
- Uranium eluted using 9 M HCl

Isotopic Analysis

- Analysis performed using Micromass Isocap MC-ICP-MS

RESULTS

The isotopic composition of individual and composite fallout samples are shown in Panel A and B. These plots demonstrate that the uranium isotopic composition of fallout can be measured through two components mixing between nuclear fuel from the device (approx. 92% U²³⁵) and the natural soil (0.7% U²³⁵). The y-intercept of Panel A represents the U²³⁵/U²³⁴ ratio from the soil and material, accounting for device and material uranium isotopic composition (U²³⁵/U²³⁴ = 0.102). The device U²³⁵/U²³⁴ = 1.909. When not shown, uncertainty bars are smaller than symbols.

Fallout glass samples (including the 0.25-0.3 mm composite) overlap within uncertainty at U²³⁵/U²³⁴ = U²³⁴/U²³⁸ consistent with the two-component mixing model. As in the previous figure, the U²³⁵/U²³⁴ ratio of composite 0.25-0.3 mm is larger. A fraction of glass in the smallest size fraction may come from a nearby above-ground test, contaminating this size fraction with fallout of a different characteristic U²³⁵/U²³⁴ ratio. The U²³⁵/U²³⁴ and U²³⁴/U²³⁸ ratios measured in these samples may be characterized of this particular test.

Considerable isotopic variation is observed, both between individual particles, and between particles of different grain size. The U²³⁵/U²³⁴ ratio of composite samples decreases with increasing grain size, implying that the smallest fallout glass particles contain a larger fraction of uranium from soil relative to the device.
Fundamental Study of Scintillating Materials and their PSD Properties
Megan E. Deeds, H. Paul Martinez, Natalia Zaitseva*, Leslie Carman, Andrew Glenn, and Mareena Robinson
Lawrence Livermore National Lab, 7000 2nd Ave. Livermore, CA 94550

Abstract

Neutron detection is important for the detection of nuclear materials. Scintillation techniques have proven to be useful in the detection of neutrons for years. Specifically, organic scintillators are preferred due to their ability to detect fast neutrons and hence, thermal neutrons. Organic scintillators are able to resolve the differences between neutrons and other ionizing materials and well as between neutrons types due to their unique interaction with matter.

There are three types of organic scintillators including crystal, liquid, and plastic. Though crystal and liquid scintillators show greater resolution, they do have their drawbacks. Crystals are fragile and often expensive to grow while liquids pose environmental concerns due to the toxicity of solvents. More recently, "soft" solvents have been explored for liquid scintillators. "Soft" solvents offer higher flash pluses as well as toxicity. Additionally, plastic detectors have shown promise as they are durable and relatively inexpensive. One goal of this study is to understand how the different organic scintillators perform in a liquid environment. Literature has shown that lithium-6 and boron-10 can achieve thermal neutron detection when added to organic scintillators. This work will utilize two different lithium compounds affected the PSD capabilities of both liquid and plastic scintillators.

The ability to detect neutrons depends on the energy levels of the components of the scintillating materials. Electrons within a molecule are excited due to ionization, and after a period of time these electrons relax by re-emitting energy, usually in the form of light. This is similar to a chain reaction in which a small section of a long chain can be excited, followed by the re-emission of light. Damaged scintillator requires the interaction of triplet states of pairs of excited molecules. Triplet states are responsible for light being absorbed within the scintillating material, before they collide producing additional excited singlet states that relax into the singlet ground states with a lower fluorescence lifetime. The differences between the prompt and delayed luminescence are used for the PSD phenomenon to exist. This process is shown in Figure 1.

The initial spectrum includes the neutron and gamma waveforms, containing both the prompt and delayed light. The pulse with the long fraction of the population is from the excitation produced by gamma interactions, while the pulse with higher portion of delayed light correspond to the neutron-induced excitation. By integrating at different portions of the waveform, a typical PSD signature is constructed. An alternative representation of PSD using figures of merit (POM) is shown in Figure 1. PSD is determined by taking the ratio of the peaks of interest and dividing by the sum of full width half maximum (FWHM) of the corresponding peaks.

Liquid Scintillators

Sodium is used in order to increase the lifetime of fluorescence. The idea is to use the long lifetime of an excited state, rather than an excited state that decays quickly. Sodium borate has been used as a neutron detection material because the absorption band of the excited state of the sodium borate molecule is available for the long life of the fluor that is used. In this work, we use a secondary dye to increase the fluorescence lifetime, resulting in greater separation of neutrons and gammas.

Future Work

Lithium compounds can be used to further discriminate thermal neutrons from fast neutrons. Currently, it is important to determine the compounds with the best PSD performance. In order to do so, multiple lithium compounds must be synthesized. Once synthesized, the compounds performance is tested using thermal and fast neutrons. Once the PSD performance is known, the liquid scintillator, the two compounds will be chosen for further study in plastics.

Conclusions

Studying various devices in a reactor helped to gain information regarding the effectiveness of secondary dyers. The emission spectrum revealed that only four of the eight compounds would be suitable as secondary dyes. This is due to their emission wavelength being equivalent to that of the emission wavelength of FRO. Of the eight dyes studied in the reactor, none of them proved to have more PSD than CFP.

Because a "soft" solvent, shown previously as a liquid scintillator with PSD properties can be used as a secondary dye for FRO, we have concluded that a combination of FRO and a secondary dye could be used to detect thermal neutrons in a reactor. The secondary dye used for FRO should be selected based on its ability to separate the prompt and delayed light.
ENVIRONMENTAL TRANSPORT OF PU: DESORPTION OF PU(IV) IN A MULTI-MINERAL SYSTEM

Chad Durrant1,2, James Begg1, Mavrik Zavarin1, Pihong Zhao1, Annie B. Kersting1
1Glen T. Seaborg Institute, 2Penn State University

Plutonium Transport
- Globally ~2000 MT Pu released to the environment [1].
- Understanding subsurface Pu transport is of vital importance.
- Mineral colloids identified as an important transport vehicle for Pu.
- Study Pu-mineral colloidal interactions to develop transport models.

Troubleshooting
Initially no plutonium was seen diffusing through the Float-A-Lyzer membrane. To determine the cause of this phenomena two initial possibilities were raised:
- Perhaps the high concentration of montmorillonite was causing interference during liquid scintillation counting
- Perhaps the pore size was not large enough for Pu to diffuse through

Results
After determining that a larger pore size Float-A-Lyzer was needed the flotation cells were set up again with the remaining goethite and bentonite aged Pu(IV) solutions.

Experimental Set-up
- Studies conducted at pH 4 and pH 8
- Four different minerals
- Aged with Pu(IV)
- [Pu(IV)] is 3 x 10^-10 M
- 5 mL of aged sample placed in each Float-A-Lyzer
- The bulk mineral montmorillonite
- [mont] is 5 g/L
- Acts as infinite sink to any plutonium that may desorb

Acknowledgements
This work was supported by the Subsurface Bioremediation Research Program of the U.S. Department of Energy's Office of Biological and Environmental Research. Prepared by LLNL under Contract DE-AC52-07NA27344.

Moving Forwards
Testing the selective affinity for adsorption of plutonium in multiple mineral systems.

Lawrence Livermore National Laboratory
Developing Spatially Resolved Analyses of Historical Fallout

Fitzgerald, Marc; Knight, Kim; Stone, Gary; Calderia, Lee; Austin, Kevin; Ramon, Erick; Czerwinski, Ken; Hutcheon, Ian

1. University of Nevada Las Vegas
2. Lawrence Livermore National Laboratory

Overview

Nuclear detonations where fireball-ground-contact occurs generate significant quantities of nuclear fallout. This material is created by the vaporization, melting, and scattering action of the fireball on the soil immediately surrounding the device. Since fallout incorporates radioactive traces of the device and event, this material is important for nuclear forensics study. The Trinity device, exploded July 16th, 1945, provides a comparatively well-characterized system for studying fallout formation. To understand and characterize this material, we conducted analyses using two methods: autoradiography and microscopy. Microscopy techniques included backscatter (BSE) and energy dispersive x-ray (EDX) scanning electron microscopy (SEM) and EDS. These techniques complement each other for the purpose of imaging the spatial deposition of the actinide fuel and/or fission products with respect to major element abundances.

Applying combined spatially resolved measurements, we extracted mixing and formation relationships not accessible through bulk dissolution analysis.

Theory

Physical basis behind imaging/analytical techniques

Method

Analytical Goals

- Characterize major element abundances
- Determine mixtures of activity
- Backscatter and secondary electron (SE) imaging

Major Elements

- Na, Si, Al, Fe, Mg, P, O, Ca, Ti, and K
- 80 elements for alpha/beta
- 12 elements for alpha/hidron

Image and/or for in-situ reconstruction

Preparation

- FESEM, Fallout, beads analyzed
- SEM-EDX, elements were scanned
- SEM-EDX, analysis

Results and Discussion

Fig 1: Combined imaging of activity and SEM-EDX analysis reveals the correlation between areas of high activity and high Cs. Warmer colors correspond to areas of higher activity, cooler colors correspond to areas of lower activity. Color scale overlays SEM-EDX images, but is not normalized between samples.

Fig 2: SEM/BSE and EDS of a single representative particle. For BSE imaging, areas of higher intensities correspond to higher atomic number. For EDS maps, areas of higher intensity correspond to elemental abundance in phases. This sample highlights the excellent contrast between low-radiating phases, L.B. Delbouillier.

Fig 3: Autoradiography shows two distinct modes of spatial activity: surface and volumetric. The left autoradiograph A and C samples demonstrate both extremes (i.e., exclusively surface or volumetric activity), exposing it shows some contribution of the two modes. A autoradiograph exposure is 50 minutes (Tmax for Americium) and C autoradiograph exposure is 15 minutes for Americium. In most cases, fission products appear to track with alpha phases. Despite efforts, some activity was lost through the bias for this high gamma spectroscopy. Lighter tare counting sources and IC's C and Y.

The Way Forward

Spatially resolved measurements inform formation mechanisms

- Activity-phase correlations would be very difficult or impossible to make using conventional bulk analysis techniques
- We are now developing automated high-resolution autoradiography to increase the utility of this technique for inter-sample comparisons. See figure S.
- LA-ICP-MS or SIMS/microprobe analysis will use this autoradiography information to profile the spatial distribution of activities and fission products.
Characterization of TBP using FTIR-ATR and Carbon Stable Isotopes

April R. Gillens, Michael Singleton, and Brian A. Powell
1Environmental Engineering and Earth Sciences, Clemson University, Clemson, SC
2Environmental Radiochemistry Group, Chemical Sciences Division, Physical and Life Sciences Directorate, Lawrence Livermore National Laboratory, Livermore, CA

Research Objectives

The objective of this research is to determine the carbon stable isotope signatures of tributyl phosphate (TBP) and its degradation products after contact with nitric acid and sodium hydroxide for the purpose of identifying if there is a unique carbon isotope signature in nuclear reprocessing and solvent disposal. The degradation products considered in this research are limited to dibutyl phosphate (DBP) and butanol. In order to meet this objective, studies will be conducted involving the following instrumentation:

- Fourier Transform Infrared Spectroscopy with Attenuated Total Reflectance (FTIR-ATR)
- Gas Chromatography Flame Ionization Detection (GC-FID)
- Gas Chromatography Combustion Isotope Ratio Mass Spectrometry (GC-C-IRMS)

TBP Degradation Mechanisms

Acidic Hydrolysis vs. Basic Hydrolysis

\[
(C_{6}H_{10}O)_{3}PO + H_{2}O \rightarrow (C_{6}H_{10}O)_{2}PO(OH) + C_{6}H_{5}OH
\]

Carbon isotope analysis should confirm the degradation mechanisms of TBP in acidic and alkaline conditions. A carbon isotope fractionation is not expected for TBP in alkaline conditions, because the C–O bond would rupture keeping the carbon in the molecule intact. A carbon isotope fractionation is expected for TBP in acidic conditions because of the cleavage of the C–O bond associated with TBP degradation under this condition. Hence, this work will determine whether there is a unique carbon isotope signature for TBP in nuclear reprocessing and disposal by studying the fundamental characteristics of TBP in acidic and basic hydrolysis. The reaction mechanisms for TBP degradation in acidic and alkaline hydrolysis are distinct. Although the two mechanisms proceed differently, the same degradation products are generated in addition to other products specific to the compound used to degrade the TBP molecule. The majority of degradation products resulting from acidic hydrolysis can be found in both aqueous and organic phases while most of the degradation products from alkaline hydrolysis are found in the aqueous phase. The difference in how TBP degrades in acidic and alkaline media could have major implications on the “signature” of the molecule as a result of its use in nuclear reprocessing and solvent disposal.

Acknowledgements

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FTIR-ATR Peak Intensity Ratio Technique

Acidic Hydrolysis vs. Basic Hydrolysis

A. A sample of TBP degraded in 8 M HNO₃ was evaluated using the ratio intensity ratio above. The linear regression developed for a TBP/DBP system substantiated in 8 M HNO₃ was used. Ratios 1 and 2 are more reliable and the results of ratio 3 do not reflect the conditions of the sample. After five weeks of hydrolysis, an average of 68% of DBP is produced according to ratios 1 and 2. These results have not been verified by gas chromatography. B. This figure describes the average amounts of TBP remaining and butanol formed along with their associated uncertainties for a TBP alkaline hydrolysis experiment involving 12.5 M NaOH, which was characterized by the FTIR-ATR technique using a saturated and unloaded TBP/butanol system and gas chromatography.

Carbon stable isotope signatures of TBP

The GCC-ERMS system consists of an Agilent 6890 gas chromatograph coupled with a combustion furnace, which leads to an isotope ratio mass spectrometer. The Agilent 6890 gas chromatograph is equipped with a 30 meter by 0.25 millimeter (inner diameter) HP-5MS (5% Phenyl Methyl Siloxane) Ultra-Torr fused silica capillary column with 0.25 μm film thickness. Samples are introduced using a syringe/pushe injection port set at 250°C. The GC oven is heated using the following program: isothermal for 1 minute, 150°C/min to 280°C, 100°C/min to 200°C, and isothermal for 2 minutes for a runtime of 18.08 minutes. Nitrogen is used as carrier gas through the column with a constant flow rate of 0.8 mL/min and average velocity of 32 cm/sec.
Bonding and Redox Behavior in Actinide Coordination Complexes: A First Principle Study

Mitchell Goshert, Patrick Huang, and Justin Waiensky

Physical and Life Sciences Directorate and Glenn T. Seaborg Institute

University of Missouri, Columbia

Lawrence Livermore National Laboratory

INTRODUCTION – REDOX POTENTIAL

- Oxidation state determination is common by products of nuclear fuel reprocessing.
- What is the long-term mobility and fate?
- Oxidation state determines solubility and consequently the mobility in the environment.
- What are the thermodynamic parameters characterizing the transformation between oxidation states?
- Activates in solution exhibit a large variation in oxidation state and coordination number.
- Contributions due to variations in actinide speciation difficult to disentangle experimentally.
- Calculations of reduction potentials can provide useful insight on the energy required to reduce these actinide species.

SOLVENT MODEL

- Conductor-like Polarizable Continuum Model (CPCM)
- Solvent effects are an important influence on reduction potentials.
- Model solvent as a continuum dielectric.
- Need to assume a conductor-like cavity containing solute.
- CPCM well-established for polar solvents (e.g., water).

THEORETICAL METHODOLOGY

- Hartree-Fock (HF)
- Mean-field approximation: Each electron feels the same average field, effective potential due to the remaining electrons.
- Overestimates electron-electron correlation → qualitative, but not quantitatively accurate.
- Useful as starting point for higher-level theories.

- Density Functional Theory (DFT)
- Mean-field theory (like Hartree-Fock)
- Approximation of electron correlation via model exchange-correlation functional $v_{xc}$.
- We employ the B3LYP approximation for $v_{xc}$.
- Generally improves bond energies and lengths over HF.
- Better-suited for localized d-or f-electrons (minimizes self-interaction error).

CALCULATION OF REDUCTION POTENTIALS

- Free energies are calculated relative to a standard hydrogen electrode.

- $\Delta_{\text{red}} E = E_{\text{red}} - E_{\text{ox}}$
- $E_{\text{red}}$ is the energy of the reduced species
- $E_{\text{ox}}$ is the energy of the oxidized species

- Reduction Potential
  - $\Delta_{\text{red}} E = -\frac{RT}{F} \ln K$
  - $K$ is the equilibrium constant
  - $R$ is the gas constant
  - $T$ is the temperature
  - $F$ is the Faraday constant

- Inclusion of solvent effects yields a ~4.9 eV improvement over the gas phase model.
- Discrepancy between $E^0$ (solid) and exp. likely due to:
  1. Errors in $H_{\text{f,red}}$
  2. Lack of explicit second coordination sphere
- Errors for calculated acts are typically ~ 2-3 eV

- Experimental data for the reduction potential of UO$_2$(H$_2$O)$_6$ is 0.16 eV, meaning our data is reasonable.

CONCLUSIONS – REDOX POTENTIALS

- Benchmark studies have been done on UO$_2$(NO$_3$)$_2$(H$_2$O)$_6$ and VI
- Our model involves an explicit representation of the first coordination sphere plus a continuum dielectric for the solvent environment.
- Solvation effects are essential for description of red. potentials.

STRUCTURES

\[ UO_2(NO_3)_2(H_2O)_6 \rightarrow \text{Bond Lengths} \]

<table>
<thead>
<tr>
<th>Hf (mD)</th>
<th>HB (mD)</th>
<th>B3LYP (mD)</th>
<th>B3LYP (mD)</th>
<th>Exp (mD)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-O</td>
<td>1.71</td>
<td>1.71</td>
<td>1.77</td>
<td>1.76</td>
</tr>
<tr>
<td>O02-</td>
<td>2.56</td>
<td>2.53</td>
<td>2.55</td>
<td>2.52</td>
</tr>
<tr>
<td>U-Opp</td>
<td>2.50</td>
<td>2.53</td>
<td>2.48</td>
<td>2.45</td>
</tr>
</tbody>
</table>

- B3LYP shows an improved U-O bond length over HF to within ~0.01 Å.
- Both HF and B3LYP yield significant shortening of both U-O$_{pp}$ bonds relative to experiment.
- Both HF and B3LYP accurately predict the U-O$_{pp}$ bond length within ~0.02 Å.

- UO$_2$(NO$_3$)$_2$ – Bond Lengths

<table>
<thead>
<tr>
<th>Hf (mD)</th>
<th>HB (mD)</th>
<th>B3LYP (mD)</th>
<th>B3LYP (mD)</th>
<th>Exp (mD)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-O</td>
<td>1.80</td>
<td>1.81</td>
<td>1.84</td>
<td>1.85</td>
</tr>
<tr>
<td>O02-</td>
<td>2.73</td>
<td>2.63</td>
<td>2.71</td>
<td>2.62</td>
</tr>
<tr>
<td>Opp</td>
<td>2.87</td>
<td>2.85</td>
<td>2.85</td>
<td>2.85</td>
</tr>
</tbody>
</table>

- Uranyl V bond lengths show ~0.1 Å increase on average over uranyl VI.

- Geometry optimizations were performed on both the U(PD)(NDIPP)$_2$(benzene) and U(PD)(NMMe)$_2$(right).

- Conclusions – Bonding Behavior

- A successful geometry optimization has been completed on the U(PD)(NDIPP)$_2$ complex with bond lengths within ~0.02 Å accuracy.
- The U(PD)(NMMe)$_2$ optimization has not fully converged but demonstrates an accuracy within ~0.03 Å.

ACKNOWLEDGEMENTS

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Tritium and Stable Isotope Survey of California Surface Water

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2Chemical Sciences Division, Glenn T. Seaborg Institute, Lawrence Livermore National Laboratory
3International Institute for Applied Systems Analysis, Laxenburg, Austria
4Sponsor: 2013 LLNL Nuclear Forensics Summer Program

Introduction

Tritium (H) and the stable isotopes of water (H2O and D2O) are used to examine watershed dynamics, identify water provenance and determine water residence times. These isotopes are extremely useful as a natural tracer because they follow the path of water as part of the water molecule. Stable isotopes in precipitation have a strong spatial signal that indicates the source of the water. Tritium, which is produced cosmogenically in the upper atmosphere, has a strong temporal signal that allows for age dating of water. Large quantities of H2O were created by atmospheric thermonuclear weapon testing between 1953-1963. Current tritium concentrations in precipitation are approaching pre-nuclear levels. The strong topographic gradient in California (Pacific Ocean to Sierra Nevada) results in distinct tritium and stable isotope signatures, making these tracers especially useful in water resource and age dating investigations.

Results

Stable Isotopes

• 69% and 818O are expressed as δ (permil) = [(Rsample - Rstandard) / Rstandard] * 1000, where R is ratio of heavy to light isotopes.
• The Global Meteoric Water Line (GMWL) represents empirical relationship between δD and 818O found in meteoric water.
• Pacific Ocean samples (blue circle) is similar to Vienna Standard Mean Ocean Water (VSMOW; δD = 0 and δ18O = 0).
• Gilmore Slough (also in blue circle) is similar to Pacific due to ocean influence.
• Estuary samples (red circle) show elevation effects: heavier (less negative) 818O and δD than source water (Pacific Ocean). This effect is stronger for hydrogen than oxygen.
• Coastal samples are lighter than ocean water due to preferential evaporation of light isotopes.
• Salinas River samples (green circle) also shows evaporation effects with coastal water as source.
• Inland samples are lighter than coastal samples due to ratio of heavy isotopes.
• Lagoons (purple circle) and some inland samples show deuterium excess from re-evaporation and precipitation of terrestrial water.

Discussion and Future Work

Discussion

• Preliminary tritium data shows possible elevation gradient and reservoir effects.
• Stable isotope results are consistent with the known coastal to inland and low to high salinity gradients.
• Evaporation effects are also apparent from δD and δ18O offset from the GMWL.

Future Work

• Spring sampling - Surface waters will be resampled during the spring run-off period when precipitation signals will be stronger.
• More watersheds - Future sampling will include watersheds farther to the north and south as well as the Sierras.
• Continuous tritium analysis – Because of 21 day accumulation period, not all samples were analyzed in time to be included in these results.
High-Energy Neutron Foil Activation for Davis Cals

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Davis Cals are inter-laboratory measurements and calibration comparisons of “threshold detector products”. High-energy neutron foil activation is used for the isotope production, and the goal is to model this process and compare to experimental results.

Introduction

Radioanalytical measurements on activation species are being performed by LLNL, LANL, and PNNL. The activation species used for the inter-laboratory measurements are produced at the 76-inch Cyclotron at UC Davis Crocker Nuclear Laboratory by irradiating various detector metal foils with high-energy neutrons produced from bombarding a thick beryllium target with 40 MeV deuterons. The existing nuclear data for (d,n) reactions for beryllium, as well as production cross sections for the foils, are very limited in the evaluated energy ranges of the fast neutron spectrum. As a result, it was proposed to use physics models and Monte Carlo codes to simulate isotope production and compare with recent experimental results.

Neutron Flux Profile on Au Foil

To evaluate the differences between physics models and beam width on isotope production, the neutron flux profile on Au foil was evaluated.

Isotope Production

Isotope Production for the gold and titanium foils were evaluated using MCNPX and the ENDF and TENDL cross sections. The activation run was conducted on 12/27/2012.

Setup

Various physics models that use both intranuclear cascade (INC) and evaporation physics were used to model (d,n) reactions. Early results showed that for 40-MeV deuterons, changes in evaporation models had little impact, and so differences between three INC models (CEM03¹, Bentini² and INCL5) were evaluated. MCNPX 2.7 was used to evaluate neutron transport in the system and reaction rates in the foils.

Neutron Flux

Neutron Flux for various INC physics models evaluated for a for a 6.0 ± 1.7 µA beam current.

Discussion

The three INC models all have limits of reliability in the energy range being explored when compared with recent experimental data. The TENDL cross sections for higher energy are in good agreement for the ¹⁹⁸Au, ¹⁹⁶Au, ⁴⁴Sc, ⁴⁴Sc residual cross sections. Further work is needed to assess production pathways and cross sections for ⁴⁴Sc, ⁴⁴Sc, and ¹⁹⁸Au.

References:


This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.
Validation of a Phenomenological Fission Model

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Fission data are very useful for both forensic and basic science applications. Unfortunately, these data often cannot be easily measured experimentally. Therefore, fission models/theories play an important role to fill the gaps left by experiment. We will determine the extent to which the fission model GEF can be trusted.

**GEF: GEneral Fission model**

GEF uses Monte Carlo methods to calculate:
- Element & isotope yield distribution, before and after neutron emission
- Mass chain yields
- Isomer yields
- Angular momentum distribution
- Prompt gammas and neutron spectra
- Total kinetic energy of fragments
- Numerous other fission observables

Uncertainties are determined based on perturbed parameters during iterative calculations.

**First Assessment: GEF vs. Experiment**

Due to larger experimental uncertainties, the Pu239 Thermal data and GEF data match fairly well, while the U235 Thermal data from experiment is often outside of the GEF values.

**Mass-Chain Yields**

In most cases, GEF does an acceptable job of reproducing experimental yields. However, for particular nuclides, the GEF calculations match experimental data mostly thanks to the large uncertainties that GEF calculates.

**Uncertainty Analysis**

χ² values for most nuclides are well below the 95% confidence limit (GEF may be overestimating uncertainties). Edited the GEF code to include an error factor: a factor by which the internal GEF parameters — the parameters perturbed to generate the calculated uncertainty — are multiplied. Involved the effects of varying the error factor. Attempted to decrease the uncertainty without causing χ² to increase to the 95% critical value. In all cases except U235 thermal, the uncertainty calculated by GEF can be reduced.

**Conclusions**

1. For many nuclides, GEF does an acceptable job of reproducing experimental values within experimental error.
2. GEF gives a good starting point for an estimate of experimental yields. However, the type of validation presented here is critical to properly use these estimates and their respective uncertainties.
3. As a phenomenological model, GEF is useful to more fundamental approaches because it helps us understand many important features of fission.

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Development of a Chemical System for Rutherfordium

Jeff Rolfes¹, John Despotopoulos¹,², Roger Henderson², Ralf Sudowe¹
¹University of Nevada, Las Vegas
²Lawrence Livermore National Laboratory

Background

Studies of the chemical properties of the heaviest elements have always been difficult due to the short half-lives and low cross sections involved. To solve this problem, atom-at-a-time methods are used to determine the properties of short-lived isotopes. Extremely fast kinetics for the chemical reactions studied (on the same order as the nuclide's half-life) are required and the system should have potential for automation. One potential technique, extraction chromatography, offers a way of investigating rutherfordium's properties, without the solvent waste generated in liquid-liquid extractions.

A fast systematic method involving the use of a ligand with higher inner group selectivity is necessary to discover more about element 104. Using the homologs, zirconium and hafnium, studies can be performed using liquid-liquid extractions and extraction chromatography. From literature reviews, chelating ethers, tertiary amines, and catechols show promise in extracting group 4 elements. The development of an extraction system for rutherfordium based on these extractions can further elaborate on its properties.

Purpose

While the primary goal of research involving rutherfordium and its homologs is to study superheavy element chemistry, this research and techniques can be directly applied to zirconium separations in post-decommission material as zirconium is a major fissile product. The speed of separations required for superheavy element chemistry is on the order of seconds. SCUREF's Triethylamine (TEA) especially excels in speed accuracy and precision concerning the analysis of nuclear materials.

The surrounding ligands will be all tested, beginning with TEA. All of them have shown promise in the extraction of zirconium and hafnium in literature, but have not been fully examined for their usefulness in fast chemistry.

Physical and Life Sciences Directorate

Preliminary Results

Preliminary data shows a marked separation of zirconium and hafnium. Nijlum's extraction is also included as well since it is a decay product of ⁹²Zr. It is important to note that the ⁹²Zr used has stable carbon added to it, so its results cannot be directly correlated to the chemical behavior of single atoms due to the amount of Zr polyatomic species formed.

Goals

For the rutherfordium work, the new ligands mentioned will be tested using ⁹²Zr, ⁹³Zr, and ⁹³Hf by performing solvent extractions with hydrochloric acid. In addition, the cyclo-18-crown-6 will tested with nitric and sulfuric systems.

If the kinetics are fast, then synthesizing a resin for extraction chromatography and conducting batch studies will be the next step. If any of these extraction system appears show promise, then it will be tested at Texas A&M cyclotron using short-lived isotopes of zirconium and hafnium. Finally, it will be applied to the study of rutherfordium.

Acknowledgements

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The views and conclusions contained in this document are those of the authors and should not be interpreted as necessarily representing the official policies, either expressed or implied, of the U.S. Department of Homeland Security.

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References

Using Computed Tomography to Non-Destructively Characterize Radioactive Fallout in 3D

Rodrigo N. Tapia, B. H. Isselhardt, K. B. Knight, J. D. Sain, I. D. Hutcheon

Overview:
This project is divided into two main objectives: the exploration of compositional analysis by density mapping and the quantification of void spaces within fallout spheres using CT. Fallout glasses are the resulting debris formed from the rapid vaporization and condensation of ground and device materials during a nuclear detonation.

Why: Void Analysis
- Rapid heating and cooling causes vesicles
- Vesicles are formed by gases that do not fully evolve
- Statistical distributions can help describe timeframe of formation

Why: Density Mapping
- CT provides a 3D density map related to X-ray attenuation
- Greater attenuation is displayed as higher intensity
- X-ray attenuation is correlated to average electron density
- Average electron density is a function of density and atomic number
- This correlation has potential to identify compositions of interest

How CT it Works

Results: Density Analysis
- Analysis of Localized Inclusions
- Threshold values are selected above glass intensity
- Background corrections are made and only hotspots remain
- After threshold selection, hotspots are quantitatively analyzed

Results: Void Analysis
- Developed void isolation algorithm:
  1. Isolated volumes of low attenuation
  2. Reduced noise and established thresholds to identify voids
  3. Calculated void volume and statistics

Conclusions:
- We can spatially resolve objects on the order of 10 microns
- Average electron density and X-ray attenuation are correlated
- We can locate high density inclusions in fallout
- Beam hardening does not significantly impact relative intensity
- Distributions of voids may inform time-temperature sample history

Future Steps:
- Compute 3D maps of fallout features
- Perform CT analyses on 5 additional fallout samples
- Analyze a set of 4 standard metals with CT (Ta, Co, In, Al)
- Construct a calibration for density vs. attenuation
- Study the relationship between attenuation and composition in fallout
- Analyze hotspots by scanning electron microscopy

Acknowledgements:
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