2014 LLNL Nuclear Forensics Summer Program

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

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Sponsors:
National Technical Nuclear Forensics Center, Domestic Nuclear Detection Office, Department of Homeland Security
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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 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, ongoing science at LLNL.

Currently called the Nuclear Forensics Summer Program, this program began 14 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 students participate in conducting research at LLNL. 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 8–12 slots available. Students also come on paid internships from NNSA, DHS. Students come highly recommended from universities all over the country. For example, this year we hosted students from 11 different universities. (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, environmental radiochemistry, heavy element separations chemistry, radiation detector physics development, nuclear chemistry, and scintillator materials development (see Table 2.) Graduate students are invited to return for a second year at their mentor’s discretion. We encourage continuation of research collaboration between graduate student, faculty advisor, and LLNL 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 the national community. 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 3 Nuclear Forensics Fellows, 2 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 (FY14 held at University of Missouri, MO). Staff scientists lecture to the undergraduate students in their field of expertise.

We use our summer program to help develop a successful pipeline of top-quality students from universities across the U.S. Since 2002, 30-40% have returned to conduct their graduate research at LLNL:
- 14 became postdoctoral fellows at LLNL.
- 6 became postdoctoral fellows at other national labs.
- 9 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 FY14, 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. Once again, one of our student’s poster won the ‘best poster’ award out of about 250 students posters that were judged.
Table 1. Summer Students

<table>
<thead>
<tr>
<th>Student</th>
<th>Major</th>
<th>University</th>
<th>Year</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oluwatomi Akindele</td>
<td>Nuclear Engineering</td>
<td>University of California, Berkeley</td>
<td>Grad</td>
</tr>
<tr>
<td>Richard Beck</td>
<td>Nuclear and Radiological Engineering</td>
<td>Texas A&amp;M University</td>
<td>Grad</td>
</tr>
<tr>
<td>Andrew Conant**</td>
<td>Nuclear and Radiological Engineering</td>
<td>George Institute of Technology</td>
<td>Undergrad</td>
</tr>
<tr>
<td>Kelly Daniel</td>
<td>Forensic Science</td>
<td>Marshall University</td>
<td>Grad</td>
</tr>
<tr>
<td>Megan Deeds</td>
<td>Chemistry</td>
<td>University of Cincinnati</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>Connor Hilton**</td>
<td>Geochemistry</td>
<td>Brown University</td>
<td>Undergrad</td>
</tr>
<tr>
<td>Corey Keith</td>
<td>Nuclear Engineering</td>
<td>Texas A&amp;M University</td>
<td>Grad</td>
</tr>
<tr>
<td>Kelly Kmak**</td>
<td>Chemistry</td>
<td>University of California, Berkeley</td>
<td>Undergrad</td>
</tr>
<tr>
<td>Laurence Lewis</td>
<td>Nuclear Engineering</td>
<td>University of California, Berkeley</td>
<td>Grad</td>
</tr>
<tr>
<td>Chris Prokop</td>
<td>Nuclear Chemistry</td>
<td>Michigan State University</td>
<td>Grad</td>
</tr>
<tr>
<td>Andrew Renshaw</td>
<td>Geology</td>
<td>California State University, East Bay</td>
<td>Grad</td>
</tr>
<tr>
<td>Quinn Shollenberger**</td>
<td>Chemistry</td>
<td>Arizona State University</td>
<td>Undergrad</td>
</tr>
<tr>
<td>Rodrigo Tapia**</td>
<td>Mathematics and Chemistry</td>
<td>University of Georgia</td>
<td>Undergrad</td>
</tr>
</tbody>
</table>

*= Nuclear Forensics Graduate Fellows  **= Nuclear Forensics Undergraduate Intern
Table 2.  Student Projects and Mentors

<table>
<thead>
<tr>
<th>Student</th>
<th>Mentor</th>
<th>Project</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oluwatomi Akindele</td>
<td>Bryan Bandong, Kevin Roberts</td>
<td>Simulated Neutron Activation of Metals for Post Detonation Nuclear Forensics</td>
</tr>
<tr>
<td>Richard Beck</td>
<td>Nicholas Scielzo</td>
<td>$\beta$ -Delayed $\alpha$ Angular Correlation in Mass 8 Systems</td>
</tr>
<tr>
<td>Andrew Conant</td>
<td>Martin Robel</td>
<td>Reactor Modeling of Pu and Cs Isotope Ratios in Pressurized Water Reactor Fuel Assemblies</td>
</tr>
<tr>
<td>Kelly Daniel</td>
<td>Mike Kristo</td>
<td>Evaluating Methods for Removing Radioactive Contamination from Traditional Forensic Evidence: Moths</td>
</tr>
<tr>
<td>Megan Deeds</td>
<td>Natalia Zaitzeva</td>
<td>Exploring Polymerization Techniques for PSD Plastic Scintillators</td>
</tr>
<tr>
<td>Marc Fitzgerald**</td>
<td>Kim Knight</td>
<td>Understanding Plutonium in Fallout Formation</td>
</tr>
<tr>
<td>Connor Hilton*</td>
<td>Ross Williams</td>
<td>Local and Global Nuclear Fallout Contributions to a Soil Profile</td>
</tr>
<tr>
<td>Corey Keith</td>
<td>Bryan Bandong, Kevin Roberts</td>
<td>High-Energy Neutron Foil Activation for Davis Cals</td>
</tr>
<tr>
<td>Kelly Kmak</td>
<td>Dawn Shaughnessy</td>
<td>Separations and Nuclear Counting for Research Projects on National Ignition Facility (NIF) Radiochemistry and Nuclear Forensics Sample Production</td>
</tr>
<tr>
<td>Laurence Lewis</td>
<td>Ian Hutcheon</td>
<td>SIMS Analysis of Aerodynamic Fallout</td>
</tr>
<tr>
<td>Chris Prokop</td>
<td>Dawn Shaughnessy</td>
<td>Perform Isotope Production and Nuclear Counting Experiments in Support of Nuclear Forensics</td>
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<tr>
<td>Andrew Renshaw</td>
<td>Brad Esser</td>
<td>Investigation of Radium Analytical Methods for Groundwater with Complex Matrices</td>
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<tr>
<td>Quinn Shollenberger</td>
<td>Greg Brennecka</td>
<td>Examining the Gadolinium Isotopic Signatures of Nuclear Material</td>
</tr>
<tr>
<td>Rodrigo Tapia**</td>
<td>Gary Eppich, Sarah Roberts</td>
<td>Comparison of Historical Fallout with Representative Soil Samples by XRD and ICP-MS</td>
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* = Nuclear Forensics Undergraduate Intern  **= Nuclear Forensics Graduate Fellows
Table 3. Seminar Schedule

<table>
<thead>
<tr>
<th>Date</th>
<th>Speaker</th>
<th>Topic</th>
</tr>
</thead>
<tbody>
<tr>
<td>6/12/14</td>
<td>Kim Knight</td>
<td>Analytical Chemistry: Applications in Nuclear Forensics</td>
</tr>
<tr>
<td></td>
<td>Staff Scientist, Chemical &amp; Isotopic Signatures Group, Chemical Sciences Division</td>
<td></td>
</tr>
<tr>
<td>6/26/14</td>
<td>James Begg</td>
<td>Water, Rocks and Microbes: The Biogeochemistry of Radionuclides</td>
</tr>
<tr>
<td></td>
<td>Staff Scientist, Environmental Radiochemistry Group, Chemical Sciences Division</td>
<td></td>
</tr>
<tr>
<td>7/1/14</td>
<td>Dawn Shaughnessy</td>
<td>Radiochemistry and Nuclear Physics Measurements at the National Ignition Facility</td>
</tr>
<tr>
<td></td>
<td>Group Leader, Experimental Nuclear and Radiochemistry, Chemical Sciences Division</td>
<td></td>
</tr>
<tr>
<td>7/8/14</td>
<td>Jean Moran</td>
<td>Studying the Effects of Climate Change on Water Resources Using Isotopes</td>
</tr>
<tr>
<td></td>
<td>Associate Professor</td>
<td></td>
</tr>
<tr>
<td></td>
<td>California State University East Bay, Department of Earth and Environmental Sciences</td>
<td></td>
</tr>
<tr>
<td>7/16/14</td>
<td>Amy Gaffney, Brett Isselhardt</td>
<td>Chronometry of Geologic and Nuclear Materials</td>
</tr>
<tr>
<td></td>
<td>Staff Scientist, Chemical &amp; Isotopic Signatures Group, Chemical Sciences Division</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Staff Scientist, Chemical &amp; Isotopic Signatures Group, Chemical Sciences Division</td>
<td></td>
</tr>
<tr>
<td>7/24/14</td>
<td>Mark Stoyer</td>
<td>Nuclear Forensics at LLNL</td>
</tr>
<tr>
<td></td>
<td>Group Leader, Experimental Nuclear Physics, Physics Division</td>
<td></td>
</tr>
<tr>
<td>7/31/14</td>
<td>Mona Dreicer</td>
<td>Treaty Monitoring and Verification</td>
</tr>
<tr>
<td></td>
<td>Deputy Program Director for Nuclear and Domestic Security and Deputy Director of the Center for Global Security Research (CGSR)</td>
<td></td>
</tr>
<tr>
<td>8/8/14</td>
<td>Annie Kersting</td>
<td>Closing out the Program</td>
</tr>
<tr>
<td></td>
<td>Director, Glenn T. Seaborg Institute, Physical and Life Sciences Directorate</td>
<td></td>
</tr>
</tbody>
</table>
Simulated Neutron Activation of Metals for Post Detonation Nuclear Forensics

Tomi A. Akindel1, Corey C. Keith2, Kevin E. Roberts1, Robert J. Haslett1, Tzu-Fang Wang3, and Bryan B. Bandong 3

1University of California, Berkeley, Department of Nuclear Engineering
2Texas A&M University, Department of Nuclear Engineering
3Lawrence Livermore National Laboratory, Chemical Science Division

Objective: Determine observable and distinguishable signatures of activation products from thermonuclear and fission neutron capture reactions

Introduction:
Thermonuclear and fission reactions yield different energy neutrons. Evaluating the differences in how these neutrons interact with matter are useful for nuclear forensics diagnostics. Neutron activation with 2-3 and 14 MeV neutrons on 10 elemental, structural metals was simulated for the UC Davis 76” cyclotron’s deuteron beam.

Theory:
Thermonuclear reaction:
\[ ^2\text{H} + ^3\text{H} \rightarrow ^4\text{He} + \text{n} \quad E_n = 14.1 \text{ MeV} \]
Fission reaction:
\[ \text{n} + ^{235}\text{U} \rightarrow \text{A} + \text{B} + \text{xn} \quad E_n = 2-3 \text{ MeV} \]

Methods:
A deuteron beam incident on Be can be used to create spallation neutrons.

Nickel Activation:
The following activation products were created through neutron bombardment on Nickel. Because each reaction has an energy threshold, the production of each isotope is strongly dependent on the neutron energy.

Results:
Spectra resulting from the counting of irradiated material were created for both the 3 and 14 MeV cases with cooling times ranging from 12 to 72 hours in 12 hour intervals. These spectra were compared at different energies and cooling times. The 14 MeV irradiation consistently showed the production of more activation species. To better quantify the differences in observed gamma ray lines, the ratio of the most intense gamma ray lines were plotted for the 6 observed cooling times. The lines chosen were taken from different activation products to avoid redundant results.

Conclusions:
The same analysis was also done for Al, Ti, Fe, Cu, Ir, W, Au, and Pb. The results clearly show observable differences in 2-3 and 14 MeV spectra. Although these simulations showed predictions on expected spectra, it could not predict the formation of isotopes in their metastable state. From this work the priority of sample counting order was also determined. This work is to be performed at the UC Davis 76” cyclotron. Experimental data can then be compared to the simulated results.

References:
**β-Delayed α Angular Correlation in Mass 8 Systems**

**Richard Beck**

**Lawrence Livermore National Laboratory**


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### Introduction

**THEORY**

- β-Decay spectrum determined by phase space, kinematic correction, and nuclear structure effects
- Kinematic correction well understood
- Weak interaction term describes the correlation between the direction the beta particle (electron) is emitted and the directions the two alpha particles are emitted

### Setup

**SAMPLE INJECTION**

- Lithium-8 is produced using 7Li(d,2p) reaction. Lithium-7 beam reacted with Deuterium to form Lithium-8
- Lithium-8 ions stopped, bunched, and purified using Helium gas and electric fields
- Ions held nearly at rest in 1 mm² volume for accurate measurement using an ion trap

### MEASUREMENT

- Detectors: Four 64x64x1 mm³ with 32x32 silicon strips laid perpendicular to each other in front of plastic scintillators
- Measurements give the direction and energy of both alpha and beta particles

### Data

**SPECTRA**

- 3x10⁶ β-α coincidences in the decay of Lithium-8 collected with apparatus

### Simulation vs. Observed

**SIMULATION**

- Simulation reproduces data very well
- Impact of kinematic and weak interaction terms still requires careful comparison
- To interpret data, a simulation of the β decay and the apparatus in GEANT4 physics modeling software was developed to compare to data
- Allows us determination of kinematic and weak interaction terms

**UNEXPECTED EFFECT**

- As the energy of the beta particle increases, there is some disagreement between data and simulation between 90⁰ and 120⁰
- Simulation needs re-investigating and refining in order to uncover source of discrepancy
- Will need to be investigated in Boron-8 data

### Future Goals

**BORON-8 MEASUREMENTS**

- Boron-8 data collected, will soon be compared to both simulation and the Lithium-8 data
- Boron-8 has similar decay chain as Lithium-8, except β decay mechanism is positron-emission (β⁺) instead of electron-emission (β⁻)
- Mirror decay switches the signs on certain mathematical terms, crucial for isolating the magnitude of said terms
- Will then be able to determine the nuclear recoil terms experimentally
- Require 12.7 million events for 5% uncertainty, and 80 million events for 2% uncertainty

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*This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contracts DE-AC52-07NA27344, Lawrence Livermore National Security, LLC.*

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Lawrence Livermore National Laboratory
Reactor Modeling of Pu and Cs Isotope Ratios in Pressurized Water Reactor Fuel Assemblies

Andrew Conant, Martin Robel, Anna Erickson

1Lawrence Livermore National Laboratory, Physical and Life Sciences Directorate, Chemical Sciences Division
2Georgia Institute of Technology, Department of Nuclear and Radiological Engineering

Overview:

- Pu-239 is produced in a reactor via the neutron capture of Pu-238, but some is lost through the fission of Pu-240. Pu-240 comes from the neutron capture of Pu-239.

- Cs-137 is a fission product with ~0.2% of the cumulative fission yield for the thermal fission of Pu-239. Cs-137 is primarily produced from the decay of Sr-90. However, most of the Sr-90 is transmuted to Y-90 (~2.5 MeV).

This makes the Pu-239/Cs-137 isotope ratio sensitive to reactor operating conditions.

Results:

Plutonium

- (Parameter)

- Fuel Enrichment

- Axial Position (cm)

- Moderator Density

- 240Pu/239Pu

- 239Pu Enrichment:

- Axial Position (cm)

- Density (g/cm^3)

- Axial Position (cm)

- Light Water

- Heavy Water

Cesium

- (Parameter)

- Axial Position (cm)

- Moderator Type

- 137Cs/135Cs

- Axial Position (cm)

- Density (g/cm^3)

- Axial Position (cm)

- Light Water

- Heavy Water

Conclusion:

- Pu-239 and Sr-90 both decrease with increasing enrichment. Increasing the enrichment shifts the neutron spectrum to a higher energy, resulting in more captures of Pu-240.

Future Work:

- Model with a deterministic code (e.g., SCALE 6.1)
- Test other reactor core and operation conditions (reactor power, shutdown frequency, core power level, etc.)

References:

Evaluating Methods for Removing Radioactive Contamination from Traditional Forensic Evidence: Moths


*Marshall University Forensic Science Center, 1409 Forensic Science Drive, Huntington, WV 25701
Lawrence Livermore National Laboratory, 7000 East Ave., Livermore, CA 94550

Overview:

Two goals of Nuclear Forensics:

1. Scientific analysis of nuclear material to determine material properties and origin
2. Traditional forensic analysis of contaminated evidence

Traditional forensic evidence associated with interdicted nuclear material or an attack using a radiological dispersal device (RDD or dirty bomb) may become contaminated by dispersible radioactive material and must either be:

- Analyzed by a laboratory capable of handling dispersible radioactive material
- Decontaminated prior to entering a traditional forensic science laboratory

1 April 2009 — Victoria, Australia

Police carried out a drug raid of an alleged amphetamine laboratory. They unexpectedly found 309 grams of uranium oxide in a storage property. After initial analysis by the Australian Science & Technology Organization (ANSTO), aliquots of the material were sent to Lawrence Livermore National Laboratory (LLNL) for further analysis. While evaluating the sample for chemical analysis, researchers at LLNL found the body and head of a moth (Figure 1).

Entomological study of the moth could prove useful for understanding the history of the material from production to interdiction within Australia, a type of signature referred to as a “route attribution” signature in nuclear forensics. However, entomology labs are unequipped to handle dispersible radioactivity.

The decontamination process must remove enough nuclear material to render the evidence safe without destroying the evidentiary value.

Method:

- Exemplar moths were gathered from northern Colorado
- Post-readily available, COx: much larger (possibly more durable) than the evidence moth
- CUP-2, an uranium ore concentrate, was used to contaminate the moths
- Decontamination method: ultrasound in eleven decontamination solvents
- Microscopic examination

Results:

Microscopic Examination:

Two visual indicators of a successful decontamination:

1. Visibly removes the contamination.
2. Does not affect the appearance of the moth.

Figures 2 and 3 demonstrate a partially successful decontamination.

- Figure 2 shows the furry thorax on a moth after contamination. The visible dusty, green particles are CUP-2. After decontamination with water, some small, particles are still visible.

Figures 4, 5, and 6 demonstrate a completely successful decontamination.

- Figure 4 and 5 show the moth before and after contamination with CUP-2. A solution of 5% Radiowash™ was used to remove the CUP-2 particles. Figure 6 shows the radiowash process showing no significant changes to the appearance of the moth.

Figures 7 demonstrates an unsuccessful decontamination.

- The solvent, a solution of 10% RBSSTM-25 (percentage recommended by the manufacturer), might have left a residue on the moth that resulted in the observed appearance after decontamination at 129 °C. The mass difference data (Table 1) suggests that RBSSTM-25 could be a viable option; however, the moth lost many body parts (legs, part of a wing, and many scales) that could contribute to the mass loss.

Conclusions:

- Solvents found to be qualitatively promising for decontamination include:
  - 5% Radiowash™, 5% Decon 90, Acetone, and 1% Nitric Acid.
- These solvents removed the most mass without damaging the moth.
- Mass difference measurements are imperfect due to:
  - Incomplete decontamination — water left in the moth from an incomplete initial decontamination process, resulting in an artificially high mass.
  - Loss of body parts during ultrasonication — legs and antennae were collected, however, scales were unavoidably lost and might have a significant mass contribution.

Future Steps:

- Complete decontamination of moths for more reliable data — may require the use of suppressor and/or longer decontamination time with storage in a decontactor.
- Sparse samples with known amounts of gamma emitters and determine the amount of decontamination with gamma counting.
- ICP-MS analysis to determine the amount of uranium remaining.
- DNA extraction — can DNA be extracted and separated from the radioactive material for analysis in a traditional forensic science laboratory?

References:


Acknowledgements:

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.
Exploring Polymerization Techniques for PSD Plastic Scintillators

Megan E. Deeds, H. Paul Martinez, Andrew Mabe, Leslie Carman, Andrew Glenn, and Natalia Zaitseva

Lawrence Livermore National Lab, 7700 East Ave. Livermore, CA 94550
University of Cincinnati, 2600 Clifton Ave. Cincinnati, OH 45220

Introduction

Neutron detection is important for the detection of nuclear materials. Scintillation techniques have proven to excel in the detection of neutrons for years. Specifically, organic scintillators show promise due to their ability to detect neutrons. Organic scintillators are able to resolve the difference between neutrons and other ionizing particles as well as between neutrons types due to pulse shape discrimination (PSD).

Plastic scintillators have become increasingly attractive due to their robustness. However, compared to both liquid and crystal scintillators, plastics do not perform as well. Performance of scintillators relies heavily on two factors: light yield (LY) and PSD. It is of importance to improve upon these factors, different polymerization techniques have been explored.

PSD Mechanism

PSD mechanism can be explored via an energy diagram. Excitations due to interactions with differing ionizing particles result in different types of emission, delayed and prompt, which gives rise to the PSD phenomenon. Also included is the initial spectrum of the radiator and gamma emissions.

Data Analysis

These are representations of PSD by integrating the wave function with respect to prompt and delayed light and using figure of merit (FOM). FOM is determined by taking the ratio of prompt of interest and discerning by the sum of full width at half maximum (FWHM) of the corresponding peaks.

Methods

1. Dewaxed under vacuum
2. Polymerized
3. Substituted

Compounds and Samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>Sample Description</th>
<th>30APPO + 1%PFO in PS 10g - 30min</th>
<th>30APPO + 0.5%PFO in PS 10g</th>
<th>30APPO + 1%PFO in PS 10g</th>
<th>30APPO + 0.5%PFO in PS 10g</th>
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<tr>
<td>30APPO</td>
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<tr>
<td>30APPO</td>
<td>30APPO + 1%PFO in PS 10g - 30min</td>
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<td>30APPO + 1%PFO in PS 10g</td>
<td>30APPO + 1%PFO in PS 10g</td>
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</tr>
</tbody>
</table>

Choosing an Effective 2° Dye

After using multiple secondary dyes, the dye with the greatest light yield and FOM is RhB-800. This dye is used as the secondary dye allowing the polymerization method.

Polymerizing Under Varying Conditions

Initial results show that an increase in temperature, in initiator-less samples, improves the FOM. This is not seen in the samples containing an initiator, but the sample with initiator at high temperature turned yellow which caused the LY and the FOM significantly.

Future Work

- Studying additional temperatures will allow for a better understanding of this temperature dependence.
- Use of different dyes and/or initiators that will not yellow in order to increase the light yield and the PSD.

Conclusions

- Polymerization at higher temperatures show promise in increasing PSD with samples excluding initiator.
- Following of plastics is likely to be PPO and/or initiator. Organics tend to yellow when subjected to heat and naturally overtime due to decompositions of the compound.
- Preliminary results reveal that removal of initiator (in styrene) increases LY and PSD. This is possibly due to yellowing and absorption of light that interferes with efficient energy transfer.

References

4. This work was supported by the U.S. DOE, NNMB, Office of Defense Nuclear Nonproliferation, Office of Nonproliferation Research and Development (NPRD). Work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.
Understanding Plutonium in Fallout Formation

Fitzgerald, Marc; Knight, Kim; Ramon, Erick; Czerwinski, Kent; Hutcheon, Ian

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

Abstract

Soil composition is the fundamental parameter controlling plutonium incorporation in fallout.

Recent analyses have shown Trinity fallout to be compositionally heterogeneous: glassy material reflecting a mixture of local soil and bomb debris. This composition diversity provides a wealth of previously overlooked information, including phase-composition, diffusive-convective mixing-transport, and phase-activity associations. My work characterizes heterogeneity in major element chemistry and Pu distribution to understand how the composition of the local environment, represented by soil, affects partitioning of plutonium during fallout formation. Analytical measurements were made using SEM-EDS and autoradiography. Pu major element correlation was understood through principal component analysis. Results show that Pu is correlated with Fe and Ca. These results, augmented by theoretical and experimental work, will inform a model capturing the first-order effects responsible for compositional heterogeneities in fallout.

Numerical Approach

Step 1: Aligning Images
- Aligned Autoradiographs

Step 2: Concatenating Images
- Data Preprocessing

Step 3: PCA Data Reduction
- X = TPT + E (Original Data)
- T = Scores
- P_T = Variable Loadings
- E = Residuals

Analytical Approach

Goal: To understand Pu correlation with major elements
- Ten aerodynamic glasses from Trinity were selected for analysis
- Followed to assess the spherical indentation
- Major element composition characterized using Energy Dispersive X-ray Spectroscopy (EDS-SEM)
- Quantitative spatial mapping of Al, Ca, Fe, Na, Mg, Si, and K
- Autoradiography was used to characterize relative Pu concentrations
- Autoradiography signal intensity is proportional to Pu concentration

Results and Discussion

Fig. 3: Numerical approach to data reduction is performed in three steps:
1. Autoradiography data are aligned and scaled to the resolution of the EDS image.
2. Data are concatenated into a 3-dimensional array. The array is transformed into a correlation matrix by mean centering variables followed by scaling by unit variance.
3. A data array is reduced using PCA, producing a score and loadings plot.

Conclusions

- PCA analysis of compositional data for T12, T13, and T17 shows correlations between Pu, Ca, and Fe.
- Correlation in multiple samples from the same event suggests a common chemical and physical environment.
- Major elemental characteristics of Pu, Ca, and Fe may reflect the reactivity of other elements in the fallout.

References


This research was performed under the Nuclear Sciences Graduate Research Program, which is sponsored by the U.S. Department of Energy. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Lawrence Livermore National Laboratory
Local and Global Nuclear Fallout Contributions to a Soil Profile

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Purpose: Investigate the movement of Pu, U, and Cs deposited from atmospheric weapons testing in a soil profile (0-16 cm) from Current Summit Spring (CSS), NV

Background:
- At the Nevada National Security Site (formerly Nevada Test Site), 97 atmospheric nuclear tests were conducted from 1951-58 and 3 in 1962\textsuperscript{1}
- Most NNSS fallout trajectories went east but some went northeast, over CSS\textsuperscript{2}
- The Limited Test Ban Treaty in 1963 banned further atmospheric testing in the US\textsuperscript{3}
- Over 500 atmospheric tests have been conducted around the world from 1945 until 1980\textsuperscript{4}

Results:

- Noticeable NNSS contribution causing a lower Pu ratio than the accepted global fallout level of 0.18\textsuperscript{5}
- 137Cs fallout from weapons testing is concentrated in the upper 4 cm of soil
- \textsuperscript{239}Pu Concentration (Bq/kg)
- \textsuperscript{239}Pu Activity (Bq/kg)

Conclusions:
- Noticeable atmospheric weapons testing fallout contribution to the top 6 cm layers shown by \textsuperscript{239}Pu, \textsuperscript{137}Cs and \textsuperscript{134}Xe
- Noticeable NNSS weapons testing fallout contribution to the top 6 cm layers shown by \textsuperscript{239}Pu
- Evident leaching of \textsuperscript{234}U into deeper soil, possibly by water infiltration
- Total U concentrations within the range of accepted average levels in soils (23 ppm)

Further Work:
- Repeat Pu analyses to determine reproducibility of the data
- Account for the NPS standard Pu deficit
- Collect 8 new CSS soil samples to measure isotopic signature changes after 15 years
- Determine how U is bound in the soil through more systematical leaches

Citations:
\textsuperscript{1}\textit{http://www.nvda.edsupplied.com/}
\textsuperscript{2}\textit{http://www.nvda.edsupplied.com/}
\textsuperscript{3}\textit{http://www.nvda.edsupplied.com/}
\textsuperscript{4}\textit{http://www.nvda.edsupplied.com/}
\textsuperscript{5}\textit{http://www.nvda.edsupplied.com/}

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

Lawrence Livermore National Laboratory
Relative Sensitivity Factors for Manganese and Chromium in Minerals Measured by Secondary Ion Mass Spectrometry (SIMS)

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Motivation

The $^{54}$Mn/$^{52}$Cr radionuclide is useful for dating ambiguous alteration events in chondrites [1]. Secondary minerals (e.g., carbonates, fayalite) can be measured in situ using SIMS. However, to obtain accurate ages, the Mn/Cr relative sensitivity factors (RSFs) must be determined for the mineral being measured. Historically, San Carlos olivine has been used as a proxy for fayalite and biotite [2], but recent measurements [e.g. 2] have shown that small compositional changes can have a large impact on the RSF.

We address this problem by analyzing a variety of Mn and Cr enriched minerals to determine how mineral composition affects the RSF, and to observe shifts in the Mn-Cr isochron due to changes in RSF.

The relative sensitivity factor:

$$\text{RSF} = \frac{\left(^{54}\text{Mn} / ^{52}\text{Cr}\right)_{\text{SIMS}}}{\left(^{54}\text{Mn} / ^{52}\text{Cr}\right)_{\text{TRUE}}}$$

Varies with:

- Mineral composition
- Instrumental tuning
- Duration of measurement

Analytical Methods

NanoSIMS:
- X-ray source
- Spot sizes 5 × 5 µm raster
- Ions measured: $^{54}$Mn, $^{52}$Cr, $^{53}$Cr, $^{54}$Mn, $^{53}$Mn
- Mass resolving power: 4500
- True value determined by EPMA
- Data reduction using LImage and Excel software

Results and Discussion

Ion implant samples:

- Observed Gaussian distribution of implanted Mn and Cr with depth (Fig. 3)
- Homogeneous lateral distribution of Mn and Cr in conductive samples
- Insulating samples (e.g., carbonates) were poorly implanted (Fig. 3), likely due to charge build-up during the implantation process
- RSFs to be determined after SIMS pits are analyzed with a profilometer

Synthesized minerals:

- RSFs were determined for three samples (Fig. 4)

Anorthite with initial ratio of $^{54}$Mn/$^{52}$Cr $= 3.42 \times 10^{-4}$ would yield a formation age of 3.6 Myr after CAI formation if not corrected for RSF.

RSF-corrected ages:

- RSF 1.6 → 1.2 Myr
- RSF 1.6 → 3.2 Myr
- RSF 0.8 → 4.8 Myr

Figure 8: Sample isochrons to show how RSFs from this study would affect the age of a Mn-Cr measurement data from [4].

Conclusions

- Improper RSFs can shift the Mn-Cr isochron leading to dating errors up to ~4 Myr; therefore, proper manganese-matched standards must be used.
- Work in progress: future work includes using a profilometer to determine the penetration depth of the Mn$^+$ and Cr$^+$ ions in implanted standards. New carbonate implants to be made using a focused ion beam to prevent charging.

References:
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 radioisotope production, and the goal is to model this process and compare to experimental results.

Introduction

Radioanalytical measurements on activation products are conducted by LLNL, LANL, PNNL, and UK’s AWE as part of an inter-laboratory calibration study. The activation species are produced at the 76-inch Cyclotron at UC Davis Crocker Nuclear Laboratory by irradiating various metal foils with spallation neutrons from a Be target bombarded with deuterons. The activation species produced will vary based on experimental parameters (such as beam energy and Be target thickness), and these effects were evaluated. Because the existing data for (d,n) reactions in beryllium, as well as production cross-sections for the foils, are limited in the evaluated energy ranges, we investigated the use of a combination of experimental data and physics models utilizing MCNPX.

Neutron Flux Profile/Depth

The neutron fluence profile of a (d,n) reaction on beryllium is shown. The differential neutron flux at different locations with a 20 MeV beam is also shown.

Setup

The various physics models used to model (d,n) reactions incorporated both Inc and Evaporation physics. Scoping work showed that for 40-MeV deuterons, changes in evaporation models had little impact and so the Bertini⁷ model was used. MCNPX2.7 was used to evaluate neutron transport in the system and reaction rates in the foils. The model of the beryllium target and foil system is shown below.

Neutron Flux

The production pathways in various foils will have high energy dependence. The resulting neutron energy flux from a 20 MeV beam will influence what isotopes are favorably produced and in what ratios.

Activation Species Production

Activation species production for 10 foils (Al, Ti, Fe, Ni, Cu, Ir, W, Pt, Au, and Pb) were evaluated at two different deutron beam energies. Various neutron capture reactions were considered for production, such as (n,α), (n,γ), (n,3n), etc. The results for Ti and Ni foils for the 58 MeV deuterion beam are presented below.

Discussion

Scoping studies were done to evaluate different beryllium thicknesses for a given deutron beam energy. This was used to guide the design of the target for optimum neutron yield, minimized thermalization, and improved beam geometry. The isotope production in foil will be energy dependent, with the many production pathways having varying thresholds. Depending on the target nuclide(s) that is desired to be produced, different energy regimes will be needed.

References:


Lawrence Livermore National Laboratory
SIMS Analysis of Aerodynamic Fallout

Characterizing the distribution of residual fuel in glassy aerodynamic fallout from a uranium-fueled test

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The distribution of residual fuel in 5 aerodynamic fallout glasses was determined using secondary ion mass spectrometry (SIMS) and correlated with bulk features in the glasses using scanning electron microscopy (SEM) and electron probe microanalysis (EPMA).

Overview

Glassy fallout forms when a nuclear device is detonated near the Earth's surface. Soil is melted and vaporized and swept up into the fireball, where it interacts with debris. As molten fallout leaves the fireball, drag forces create a variety of aerodynamically-shaped fallout glasses.

Aerodynamic fallout has been shown to host the majority of the radioactivity and contain elevated concentrations of residual fuel, compared to other types of fallout (Ippich et al., 2014). However, how the fuel distributes itself within individual fallout glasses is poorly understood and may provide information on how debris debris interacted with the environment to form fallout. Therefore, spatially resolved analyses within single samples can possibly elucidate fallout formation mechanisms, temperatures, and timescales.

Results

A range of U isotopic ratios was measured within the samples, spanning nearly three orders of magnitude in the 235U/238U, indicating significant uranium isotopic heterogeneity both between samples and within individual samples (Ippich et al., 2014). Between all five samples, the 234U/238U ratio spans a factor of greater than 500, from 0.02 to 11.8. Within a single sample, U2, the 234U/238U ratio ranges from 0.02 to 0.81, with a secondary peak of nearly 400.

While the samples display a large range of U isotopic ratios, sample U1A is homogeneous with respect to both chemical composition and uranium isotopic ratios, except for the 234U/238U ratio of 11.8 which is much lower than the other two samples. To achieve this degree of homogeneity, these samples may have been held above the melting temperature long enough for the combined effects of mixing and diffusion to effectively homogenize the melts prior to quenching.

Samples

Samples are both compositionally heterogeneous and homogeneous.

Samples U2 and U4 display correlations between the 234U/238U ratios with composition, specifically Ca. In U2, two distinct components are evident: one with a 234U/238U ≈ 1, which is associated with the brighter contrast regions in the backscattered electron image (CaO ≈ 15 wt%), and the other with 0.02 < 234U/238U < 1, which is associated with the darker contrast regions (CaO ≈ 5 wt%). The 234U/238U ratio in sample U4 also displays a bimodal isotopic behavior, but exhibits opposite correlation between the 234U/238U and Ca concentration. Regions enriched in 234U are depleted in Ca (CaO ≈ 2 wt%), while regions depleted in 234U are enriched in Ca (CaO ≈ 15 wt%). These correlations suggest that these glasses incorporated different amounts of unspallated fuel from the device, and then cooled too quickly to fully homogenize.

Future Work

- Examine correlations between 234U/238U and fallout size
- Examine correlations between 234U/238U and distance of collection
- Quantitate relationship between 234U/238U and composition

References


Investigation of Radium Analytical Methods for Groundwater with Complex Matrices

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Introduction
California’s diverse geology is reflected in a wide range of groundwater chemical composition. Understanding the hydrogeochemistry and geochronology of California’s ambient groundwaters is important for identifying the impact of conventional or unconventional oil & gas development on groundwater quality. One potential impact is radionuclide contamination. The production and its produced waters from hydraulic fracturing operations in Pennsylvania are high in radium (up to 17,000 pCi/L; Nelson et al., 2014). Flowback and produced waters are also saline, and Nelson et al. (2014) have shown that EPA protocols for the determination of radium in drinking water underestimate Ra activity in high-salinity produced waters. In order to accurately and efficiently analyze Ra in such samples, we are modifying current protocols for use with high-salinity groundwater.

Matrix Complications
Produced and formation waters associated with hydraulic fracturing are difficult to analyze using clean water methods. Co-precipitation of Ra out of solution is significant due to high concentrations of other constituents. The adjacent table shows the concentrations of major ions in flowback fluid from the Marcellus Shale in Pennsylvania (Nelson et al., 2014).

Experimental Approach
- Analyze the applicability of Liquid Scintillation Counting (LSC) for rapid and robust quantification of alpha-emitting radium isotopes, mainly 226Ra.
- Establish a minimum detectable activity (MDA) using standards and samples of known activity.
- Compare and complement LSC data with RAD7 radon detector and HPGe Gamma Spectroscopy data analysis.
- Analyze groundwater samples from the Salinas Valley utilizing LSC and the RAD7.

Methods
1. **QUANTULUS Liquid Scintillation Spectrometer**
   - Samples placed in LSC vial with 10mL sample overlay by 10mL mineral cocktail. Counted for at least 30 minutes for any counts.

2. **DURRIDGE CO. RAD7**
   - The RAD 8000 accessory for the RAD7 was utilized to analyze totalized 320mL, Salinas Valley samples and a laboratory prepared 226Ra standard.

3. **HPGe Gamma Spectroscopy**
   - LSC ratio with known 226Ra and natural U activities were counted for 1200 minutes. Samples diluted to 1/100th from the face of the detector. Efficiency values are generated through Lablogic software.

LSC Vial Geometry
The LSC vial analysis is 10mL, a sample overlay by 10mL, mineral cocktail. 226Ra decay products 214Pb, 214Bi, 214Po and 210Pb partition into the mineral cocktail, which acts as a scintillator. LSC detects the alpha emission of 226Ra, 214Pb and 214Po. The HPGe Gamma Spectroscopy method includes gamma-ray detection only in the cocktail, depending on analytical configuration.

Conclusions
- LSC is a robust and efficient method for quantifying 226Ra in groundwater.
- 226Ra MDA and recovery is not affected by the presence of uranium or radon in the sample.
- RAD7 gamma-ray detection is effective, and no lead is lost in the sampling process.

Future Work
- Fabricate a synthetic sample that mimics flowback or formation water from a hydraulic fracturing operation, and compare the performance of the methods in high-salinity matrices.
- Further refine regional differences in the radium activity in ambient groundwater in California in order to establish background (mean activity and variability).

Acknowledgments
This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. The Lawrence Livermore National Laboratory is operated by Lawrence Livermore National Security, LLC, for the National Nuclear Security Administration of the U.S. Department of Energy (DOE) under Contract DE-AC52-07NA27344.

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Image credits:
- [Image of analysis equipment and sample vials] ( Glenn T, Seaborg Institute)
- [Image of data graph] (Lawrence Livermore National Laboratory)

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Comparison of historical fallout with representative soil samples by XRD and ICP-MS

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**Goal:** to understand relationships between soil mineral phases and chemical composition in the fallout formation process.

**Results: X-ray Diffraction of Soils**

Soil samples representative of those that formed the fallout were ground to micron size particles and analyzed by x-ray diffraction on Bruker D8 X-ray diffractometer. Samples were scanned from 5 – 75° 2θ. Mineral phases were identified using DIFFRAC.EVA software and powder diffraction file database.

**Results: Fallout Sample Analyses**

Fallout sample compositions are compared to those previously studied of a larger size fraction (~1.85 mm diameter, Eppich et al., 2014. J. Radioanal. Nucl. Ch.). K₂O variation is 18% of total K₂O composition.

**Overview:**

We hypothesize that the chemical composition of nearby surface soils will, to a large extent, define the chemical composition of fallout formed during near-surface nuclear tests. To better understand the relationship between soil and fallout, several analyses were performed.

- The mineralogical composition of the soil samples were determined quantitatively by X-ray diffraction, using the data reduction software TOPAS.
- Fallout samples, of subequal diameter (~1.3 mm) and mass 2170 – 4607 µg, were dissolved using mineral acids, and analyzed by quadrupole inductively coupled plasma mass spectrometry (Q-ICPMS), and isotope dilution mass spectrometry (IDMS) using a multicolonlector-ICPMS (MC-ICPMS) for chemical and uranium isotopic composition.

**Fallout Chemical Composition**

Major Elements in Fallout Samples

<table>
<thead>
<tr>
<th>Element</th>
<th>Weight Percent</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na</td>
<td>0.1</td>
</tr>
<tr>
<td>Mg</td>
<td>0.2</td>
</tr>
<tr>
<td>Al</td>
<td>1.3</td>
</tr>
<tr>
<td>K</td>
<td>0.7</td>
</tr>
<tr>
<td>Ca</td>
<td>2.0</td>
</tr>
<tr>
<td>Fe</td>
<td>0.1</td>
</tr>
</tbody>
</table>

Average weight % of major elements in fallout with average diameter ~1.3 mm, measured by Q-ICPMS. Silicon and oxygen were not measured, but are assumed by difference to constitute ~75 wt. % of the sample.

**Conclusions:**

- Chemical and mineral compositions may define characteristics such as melting point and viscosity which could greatly influence fallout formation.
- Fallout U isotopic composition is suggestive of two-component mixing between naturally-occurring U in soil and highly-enriched, device-derived U.
- The smaller size fraction is significantly (~25%) more concentrated in uranium, similar to the difference in surface/volume ratio (35%).