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2019 LLNL Nuclear Science and Security Summer Internship Program

M. Zavarin

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Disclaimer

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2019 LLNL Nuclear Science and Security Summer Internship Program

Glenn T. Seaborg Institute
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Sponsors:
Defense Threat Reduction Agency, Department of Defense
Glenn T. Seaborg Institute, Physical and Life Sciences Directorate, Lawrence Livermore National Laboratory

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2019 Nuclear Science and Security Summer Internship Program

2019 Nuclear Science and Security Summer Internship Program students met with DTRA management on August 7, 2019. From left: Silvina DiPietro (Florida International University), Orlando Gomez (Notre Dame University, DTRA intern), Michael Klosterman (University of Utah, DTRA intern), Catherine Apgar (UC Berkeley, DTRA intern), Meena Said (University of Notre Dame, DTRA/Safeguards intern), Frances Zengotita (Florida International University), Jill Rahon (West Point), Jacob Lewis (UC Riverside, DTRA intern), Jacqueline Garcia (Oregon State University, DTRA intern), Amalie Zeitoun (DTRA acting chief of staff), Damon Anderson (UC Berkeley, DTRA intern), Thomas Early (DTRA Human Resources), and Mavrik Zavarin (LLNL Seaborg Institute director).

The Lawrence Livermore National Laboratory (LLNL) Nuclear Science and Security Summer Internship Program (NS^3IP) is designed to give graduate students an opportunity to come to LLNL for 8–10 weeks of hands-on research. Students conduct research under the supervision of a staff scientist, attend a weekly lecture series, interact with other students, and present their work in poster format at the end of the program. Students also have the opportunity to meet staff scientists one-on-one, participate in LLNL facility tours (e.g., the National Ignition Facility and Center for Accelerator Mass Spectrometry), and gain a better understanding of the various science programs at LLNL.

Currently titled the Nuclear Science and Security Summer Internship Program, this program began over 20 years ago as the Actinide Sciences Summer Program. The program is run by the Glenn T. Seaborg Institute in the Physical and Life Sciences Directorate at LLNL. The goal of the NS^3IP is to facilitate the training of next generation nuclear scientists and engineers to solve critical national security problems in the field of nuclear science and nuclear security. Students are selected from the fields of physics, chemistry, geology, mathematics, nuclear engineering, chemical engineering and environmental sciences. Students engage in research projects in the disciplines of actinide chemistry, radiochemistry, isotopic analysis, computation, radiation detection, and nuclear engineering. This Internship Program is supported by the Defense Threat Reduction Agency (DTRA) which enables the Department of Defense and the U.S. Government to prepare for and combat weapons of mass destruction and

improvised threats and to ensure nuclear deterrence. The internship program is intended to strengthen the "pipeline" for future scientific disciplines critical to DTRA and DOE.

The NS^{3}IP is highly competitive, with over 250 applications received in 2019 for the 7-8 available slots. Additional students funded through paid internships and fellowships from NNSA and DOE are invited to participate in the summer lecture series and poster symposium. This year, the NS^{3}IP hosted students from 6 universities (see Table 1) across the United States (Figure 1). The NS^{3}IP students conducted research on such diverse topics as field portable and laboratory radiation detector development, pre- and post-detonation mass spectrometry, nuclear signatures development, and fundamental nuclear physics (see Table 2 for poster titles). Continued research collaboration between the graduate student, faculty advisor, and LLNL mentors is strongly encouraged. In many cases, NS^{3}IP research evolves into a significant component of the students' graduate theses. For example, two graduates of the 2019 NS^{3}IP (Damon Anderson and Orlando Gomez) are continuing their collaboration with LLNL staff and incorporating their summer projects into their PhD research.

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

Graduate and undergraduate students on fellowships such as the DOE Environmental Management traineeship program and Nuclear Safeguards internship program, are invited to join our summer internship activities. This year, LLNL hosted 4 students from Washington State University and Florida International University through nuclear science fellowships and programmatic funding.

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

- 32 interns continued their graduate work at LLNL (9 of which received fellowships)
- 18 became postdoctoral fellows at LLNL
- 7 became postdoctoral fellows at other national labs
- 13 were hired as career scientists at LLNL
- 5 were hired as career scientists at other national labs
- 4 were hired as faculty in the area of nuclear forensics/radiochemistry/nuclear science
- 4 were hired at other government institutions

A big factor in the success of this program is the dedication of the staff scientists (predominantly DTRA funded) who volunteer to mentor the summer students. Four of our 2019 mentors are, in fact, alumni of the Seaborg Institute summer internship programs. The mentors develop summer projects for their students, oversee necessary safety training, and dedicate time to helping the interns and students maximize their productivity and scientific potential. This internship program would not be possible without the mentors' dedication. Posters summarizing the 2019 NS^{3}IP student research were presented at our Laboratory Student Poster Day and are included at the end of this report.

Figure 1. The Seaborg Institute summer interns come from universities from across the United States. Universities associated with the 2019 Nuclear Science and Security Summer Internship program are highlighted with a blue outline.

Table 1. 2019 Nuclear Science and Security Summer Internship Program Students

Student	Major	University	Year
Damon Anderson	Nuclear Engineering	UC Berkeley	Undergradute[1]
Orlando Gomez	Physics	University of Notre Dame	Graduate
Michael Klosterman	Nuclear Engineering	University of Utah	Graduate
Lauren Finney/Ibon	Chemistry	University of Michigan	Graduate
Catherine Apgar	Nuclear Engineering	UC Berkeley	Graduate
Jaqueline Garcia	Health Physics	Oregon State University	Graduate
Jacob Lewis	Materials Science	UC Riverside	Graduate
Meena Said[2]	Nuclear Engineering	University of Notre Dame	Graduate

[1] Accepted to graduate school at the University of Michigan.
[2] Partially supported by the Nuclear Safeguards program.

Table 2. 2019 Nuclear Science and Security Summer Internship Program Student Projects and Mentors

Student	Mentor	Project Poster Title
Damon Anderson	Nerine Cherepy	ASIC Development and Testing For a DTRA-Funded Circuit
Orlando Gomez	Jutta Escher	Refining Nuclear Mass Models using Bayesian Neural Networks
Michael Klosterman	Mike Singleton	Fractionation of Oxygen Isotopes in Uranium Oxides
Lauren Finney/Ibon	David Weisz	Infrared Absorption Spectroscopy to Study Oxide Formation in Laser Produced Plasmas for Nuclear Forensics Applications
Catherine Apgar	Pihong Zhao and Kevin Roberts	Quantifying Uncertainty in Inter-Laboratory Thermal Calibration Exercise
Jaqueline Garcia	Tashi Parsons-Davis	Gamma-Gamma Coincidence Counting for Radioisotope Detection
Jacob Lewis	Brett Isselhardt and Mike Savina	Resonance Ionization Mass Spectrometry applications
Meena Said	Naomi Marks	Measuring Surface Roughness on UO_2 Fuel Pellets for Nuclear Forensics

Table 3. 2019 Nuclear Science and Security Summer Internship Program Seminar Schedule

Date	Speaker	Topic
6/19/19	**Jutta Escher** *Staff Scientist, Nuclear Data & Theory* *Nuclear and Chemical Sciences Division*	**Nuclear Reaction Research for Astrophysics and Lab Applications**
6/26/19	**Nerine Cherepy** *Staff Scientist* *Materials Sciences Division*	**New Scintillators and their Integration into Detector Systems**
7/3/19	**Mike Savina** *Staff Scientist, Chemical & Isotopic Signatures* *Nuclear and Chemical Sciences Division*	**Starry Messengers: Stardust Grains Deliver Stellar News to Earth**
7/10/19	**David Weisz** *Staff Scientist, Chemical & Isotopic Signatures* *Nuclear and Chemical Sciences Division*	**Laser-based spectroscopy to inform post-detonation chemistry**
7/19/19	**Naomi Marks** *Staff Scientist, Chemical & Isotopic Signatures* *Nuclear and Chemical Sciences Division* **SAN JOSE ACS SUMMER STUDENT VISIT**	**Nuclear Forensics Materials Signatures at the front end of the Fuel Cycle**
7/24/19	**Ruth Kips** *Staff Scientist, Chemical & Isotopic Signatures* *Nuclear and Chemical Sciences Division*	**LLNL's International Nuclear Forensics Program**
7/31/19	**Ping Yang** *Los Alamos National Laboratory*	**Predictive Modeling of Actinide Chemistry**
8/9/19	**Mavrik Zavarin** *Director, Glenn T. Seaborg Institute* *Physical and Life Sciences Directorate*	**Close Out**

DTRA Agenda

PHYSICAL AND LIFE SCIENCES DIRECTORATE
GLENN T. SEABORG INSTITUTE

AMALIE ZEITOUN, THOMAS EARLY, JILL RAHON

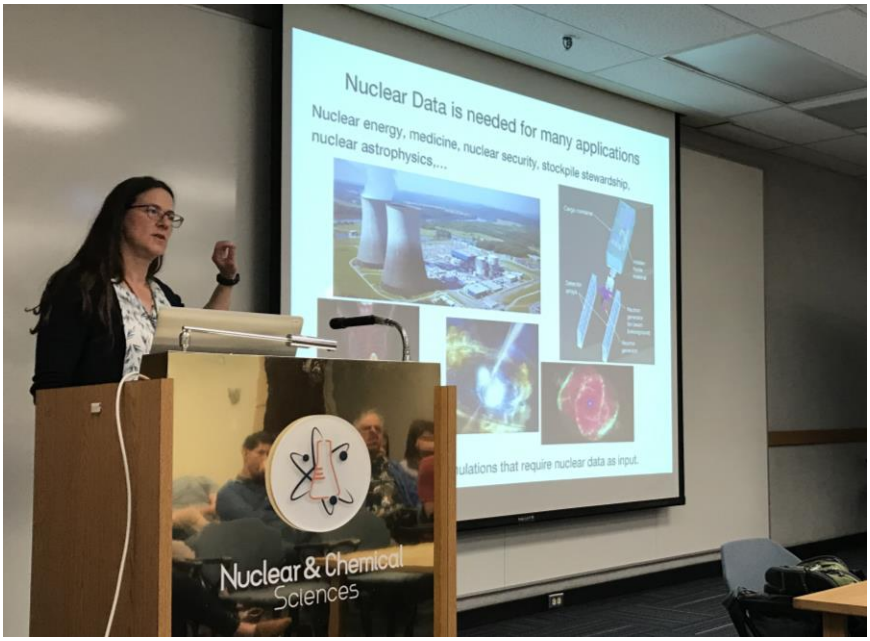
Defense Threat Reduction Agency

AUGUST 7, 2019

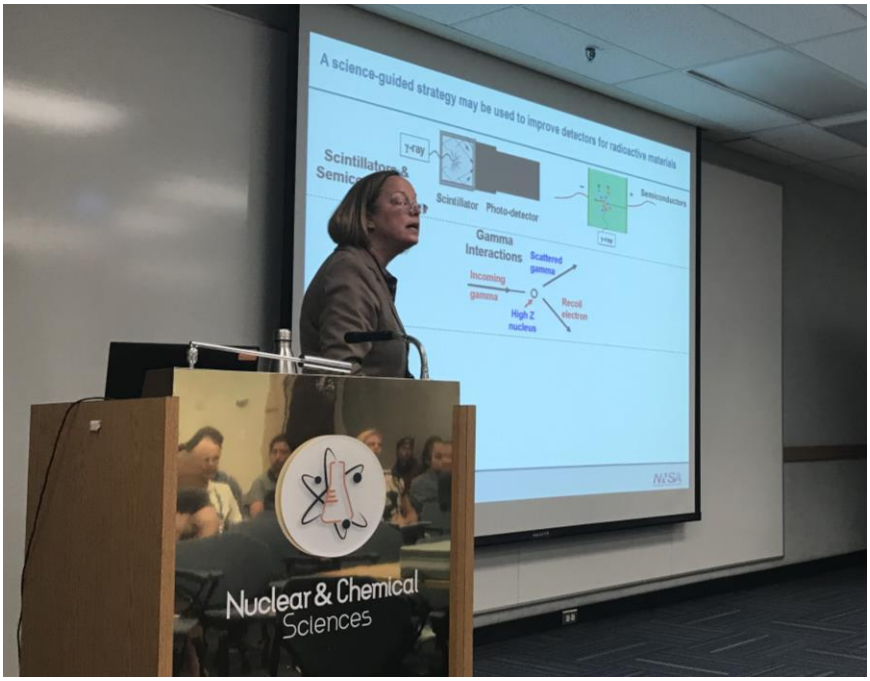
9:45 A.M.	Arrival at West Gate Badge Office	Met by Mavrik Zavarin
10:00 A.M.	Introductions DTRA Summer Interns and Mentors	B151 R1101 Ouija room
11:00 A.M.	SEMINAR Thomas Early, DTRA Mission Overview	B154 R1013 OUO – No Foreign Nationals
12:00 P.M.	LUNCH Mavrik Zavarin	Central Cafe
1:15 P.M.	Lab Tours David Weisz	B151 Lab Tour of Counting Facility (Thomas), Laser Plasma Lab (Weisz), RIMS (Savina)
2:30 P.M.	Student Poster Session David Weisz, Naomi Marks	B543 Attrium
4:00 P.M*	Depart*	

* B151 Ouija room will be reserved until 5pm for followon discussions if needed.

2019 NS^{3}IP in Pictures

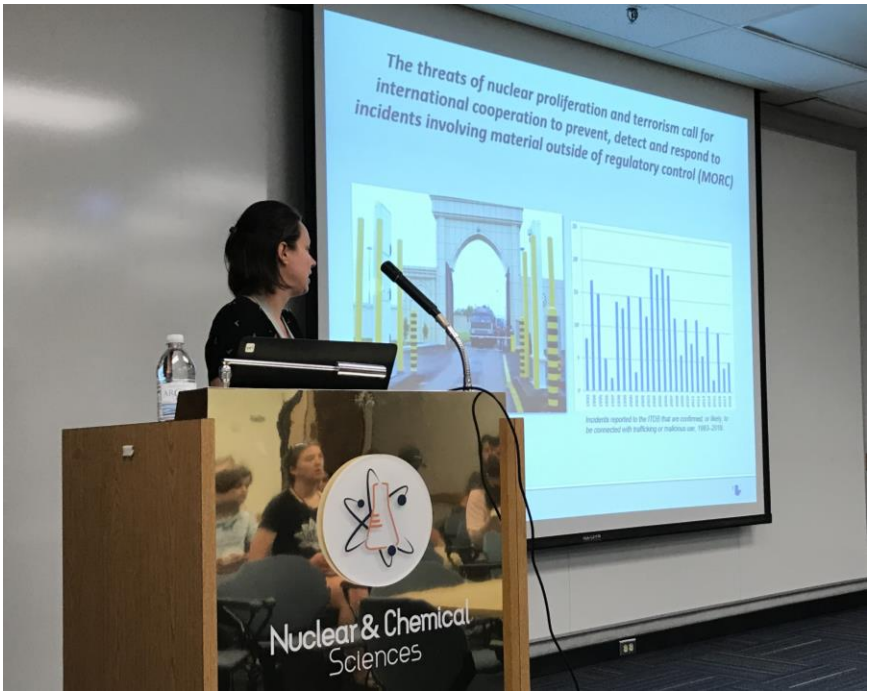


Dr. Jutta Escher describes the role of nuclear theory in nuclear science and security.

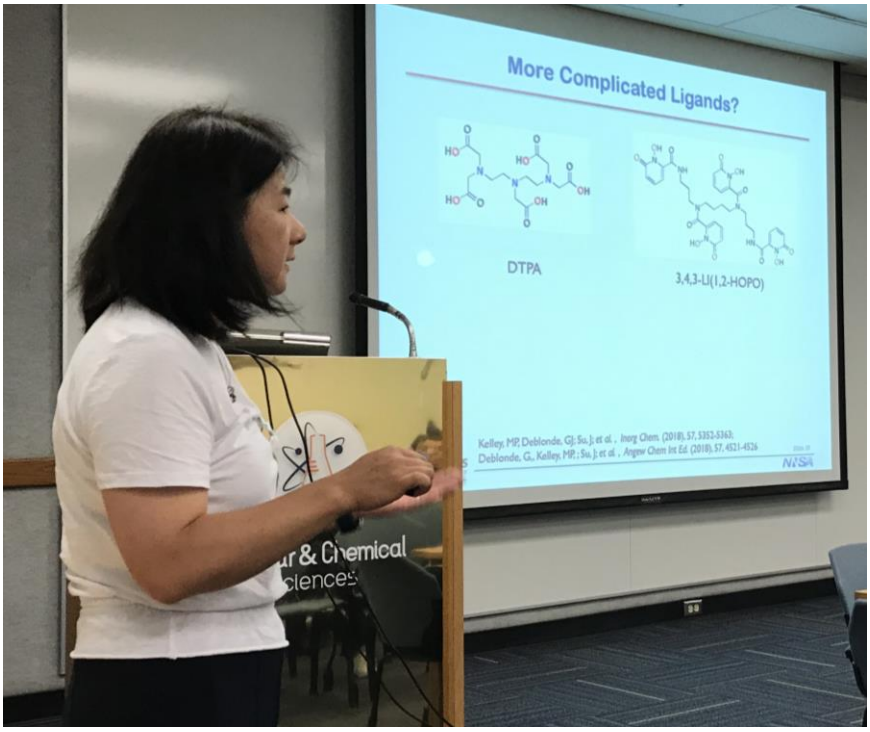


Dr. Nerine Cherepy describing the fundamentals of radiation detection.

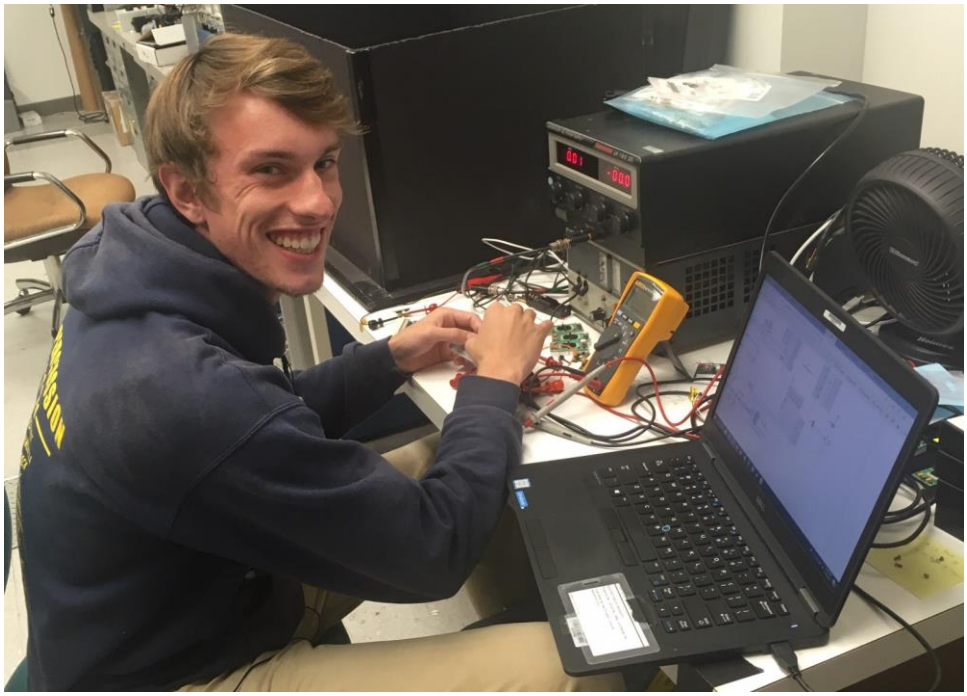
Dr. David Weisz describes the processes controlling formation of nuclear fallout.



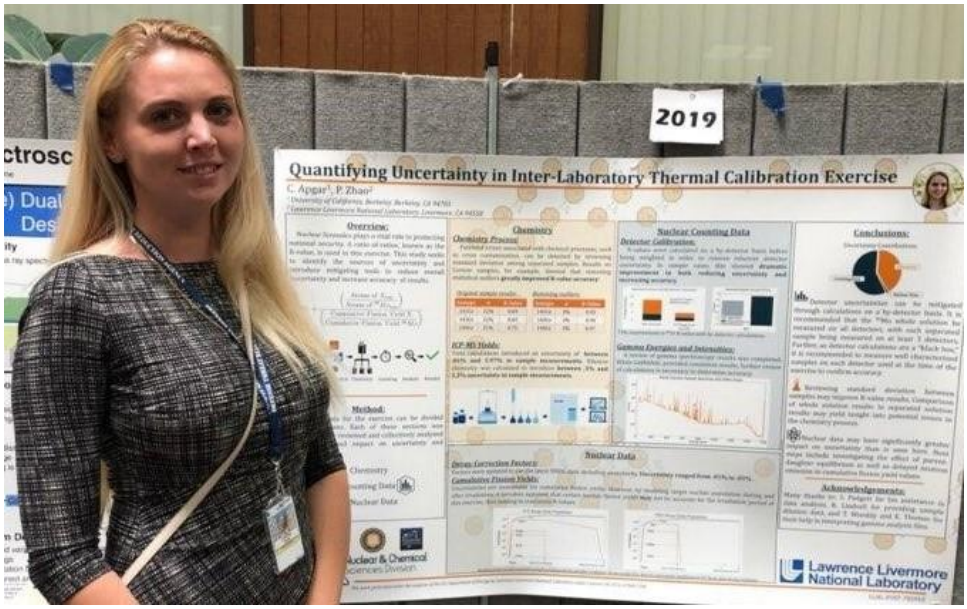
Dr. Ruth Kips presents an overview of international nuclear nonproliferation.



Dr. Ping Yang describes the fundamental chemistry of actinides.

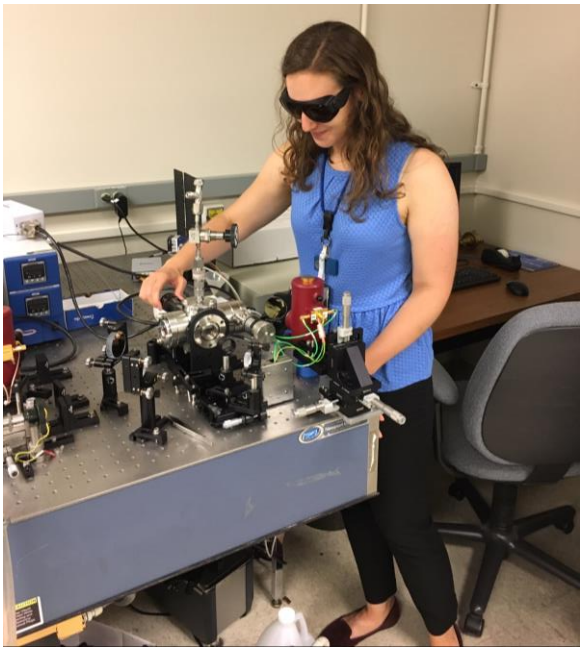


Damon Anderson works on radiation detection circuits.



Catherine Apgar presents research in uncertainty quantification at the poster symposium.

Michael Klosterman works on uranium oxide fluorination and oxygen isotope separation.



Lauren Ibon works on laser spectroscopy under simulated nuclear detonation conditions.

2019 NS^{3}IP Student Posters

LLNL-POST-784318

High Resolution Gamma Ray Spectroscopy

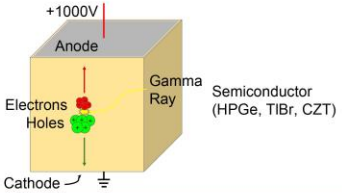
Damon Anderson, Sean O'Neal, Erik Swanberg, Nerine Cherepy, Steve Payne

Glenn T. SEABORG INSTITUTE

Multisource Energy Characterization of Thallium Bromide

Semiconductor Radiation Detectors

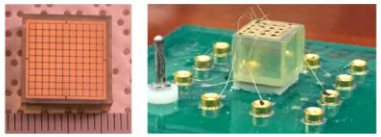
A gamma ray interacts with a biased semiconductor creating charge carriers that subsequently create signals on electrodes.



Pixelated Thallium Bromide

➔Thallium Bromide:
- ◆High Z: (Tl, 81) and (Br, 35)
- ◆High Band Gap: 2.6 eV

➔Pixelation:
- ◆Single polarity charge sensing
- ◆3D spatial resolution

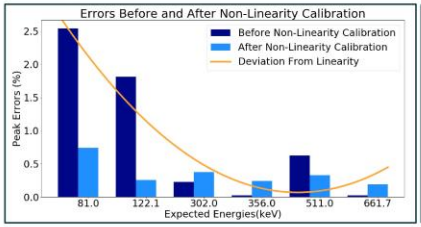


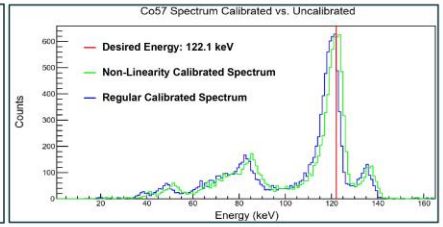
Non-Linearity Characterization

➔Spectra taken for ^{22}Na, ^{57}Co, ^{133}Ba, and ^{137}Cs

➔Measured peaks are plotted against the known peaks to determine linearity

➔Polynomial coefficients are applied to correct peak errors

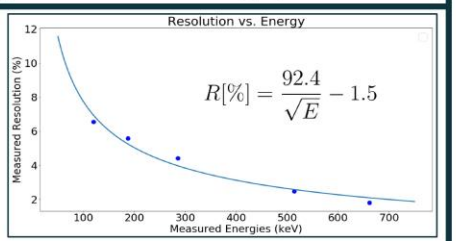




Resolution Characterization

➔Resolution tails off as energy increases as expected

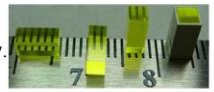
➔Negative offset implies systematic error in measurement
- ◆Limiting energy resolution (E = ∞) must be positive

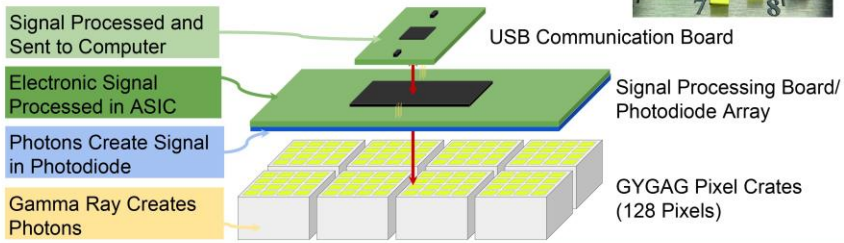


GYGAG (Ce) Dual Module System Design

Single Module Functionality

Pixelated GYGAG provides gamma ray spectroscopy and directionality.

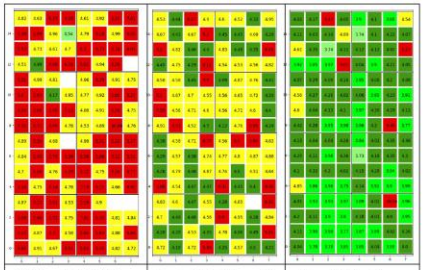




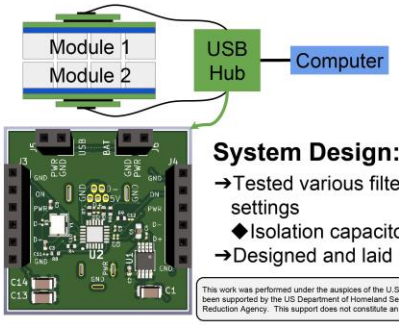
Dual Module Considerations:

➔Electronic Noise:
- ◆Module cross-talk destroys signal
- ◆**Solution:** Aluminum shielding mitigates module cross-talk

➔Temperature:
- ◆On-board linear regulators dissipate heat due to overdriving the voltage
- ◆**Solution**: In module cooling is required



Minimal Shielding; No Cooling	Best Shielding; No Cooling	Best Shielding; Cooled at 45°F
123 Functional Pixels	127 Functional Pixels	128 Functional Pixels
5.14% Resolution	4.76% Resolution	4.29% Resolution



System Design:

➔Tested various filtering configurations and shielding/temperature settings
- ◆Isolation capacitors, power line inductors, grounding connections

➔Designed and laid out PCB implementing the desired configuration

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344 and has been supported by the US Department of Homeland Security, Countering Weapons of Mass Destruction Office. We also appreciate the support from the Defense Threat Reduction Agency. This support does not constitute an expressed or implied endorsement on the part of the Government. LLNL-POST-

Refining Nuclear Mass Models using Bayesian Neural Networks

O.Olivas-Gomez*[1], K.D.Humbird[2], J.E.Escher[2], M.G.Kruse[2]
[1]University of Notre Dame, Notre Dame, IN 46556
[2]Lawrence Livermore National Laboratory, 7000 East Ave, Livermore CA 94550
ogomez@nd.edu

Introduction:

Constraining nuclear masses far from stability is of paramount importance in Nuclear Astro Physics. Nuclear masses far from stability helps us better understand nucleosynthesis in hot stellar environments (r-process) as well as understanding the crustal composition of cold neutron stars. Even with recent advancement in experimental techniques and facilities, many of these nuclei remain beyond experimental reach. Therefore we rely on theoretical mass models and their extrapolations to make predictions about neutron-rich isotopes far from stability. However, many nuclear mass models disagree dramatically once extrapolated [4] (see left fig).

In recent studies, Bayesian Neural Networks (BNN), a machine learning tool, have been shown to improve nuclear mass model predictions as well estimate their uncertainties [1]. Using DJINN, a decision tree to neural network mapping algorithm (see next panel), we develop our own refined nuclear mass models. We compare our models with those in the literature as well as make predictions of the limit of nuclear stability, the neutron drip-line.

Methods:

Using 2,407 experimentally measured masses from the 2016 Atomic Mass Adjustment (AME2016) with $Z, N > 8$, DJINN is trained on the mass residuals

$$Res(Z,N) = M_{AME}(Z,N) - M_{th}(Z,N)$$

Where M_{th} is the model mass prediction. We chose to investigate the microscopic Duflo-Zucker 10 parameter model and the macroscopic Liquid Drop model using the same parameters in [2].

To test the neural network, it is trained on 80% of the data (~1,800 nuclei) and predicts the remaining 20% of data it has not yet seen (see table below).

A drop-out percentage of 5% is used as a Bayesian Approximation to quantify the uncertainties in the model.

	LDM – DJINN	DZ-DJINN
Training Set		
MSD_{det}	2.989	0.583
MSD_{post}	0.691	0.342
Validation Set		
MSD_{det}	2.969	.609
MSD_{post}	0.674	0.415
Entire Set		
MSD_{det}	2.984	0.589
MSD_{post}	0.688	0.358

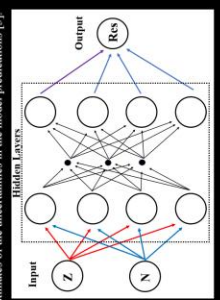
The quality of the BNN is evaluated by comparing the rms of the residuals before and after training.

The BNN residual predictions are compared to the experimental values. A perfect agreement would lie on the dashed red-line which is a visual aid.

Neural Networks & DJINN:

Neural networks trained via supervised learning can discover subtle relationships between variables and are well-suited for complex physical systems. However, it difficult to design and train an accurate model.

Deep Jointly-Informed Neural Networks (DJINN) combines the user-friendly features of tree-based models with the accuracy, flexibility, and scalability of deep neural networks [3]. The algorithm first models the data using decision trees and then produces a network with hidden layers, neurons, and initial weights that reflect the decision tree structure. The neural network is subsequently trained using beak-propagation to optimize predictive performance. The inclusion of drop-out layers work as a Bayesian Approximation allowing statistical estimates of the uncertainties in the model predications [5].



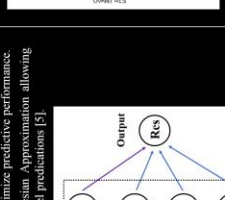
Cartoon depiction of a neural network showing nodes and weight connections.

Results:

With nuclear masses, one application is to calculate neutron separation energies.

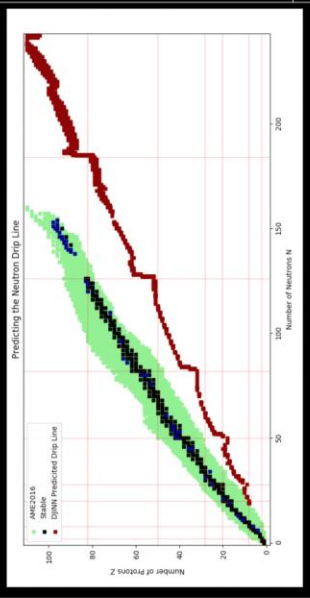
$$S_n(Z,N) = M(Z,N) - M(Z,N-1)$$
$$S_{2n}(Z,N) = M(Z,N) - M(Z,N-2)$$

Of particular interest is when $S_{2n} < 0$ and the nucleus becomes unstable to neutron emission.



The DZ-DJINN model predicts that ^{71}Ca is the last stable isotope along the Z=20 chain

According to DZ-DJINN model, ^{176}Sn is not particle-stable (N = 126 magic number).



The chart of nuclides with black squares stable isotopes observed in nature. The red curve is DZ-DJINN's prediction of the neutron drip line ($S_{2n} < 0$) using the DZ model refined by DJINN. Due to the Drop-Out Bayesian Approximation, an uncertainty can be estimated which gives a thickness to the drip line. Spanning several isotopes among certain Z chains (ex Z = 51 Sb).

Discussion / Future Work

Using DJINN, we can refine current nuclear mass models and help better predict the masses as well as the neutron drip line ($S_{2n} < 0$). Additionally we can estimate the uncertainty in the neural networks by adding drop-out layers.

The uncertainty tends to grows with extrapolation. Thus, the largest uncertainties in the drip line are in the trans-uranium ($Z > 92$) region, where there is relatively few measurements and the extrapolation is farthest.

Other nuclear mass model exists, and generally disagree once they are extrapolated beyond existing data. Using DJINN, each mass model could be refined and compared.

Other mass models to consider:
-FRDM -HFB-19, 21, 27 -MN

Other nuclear statistical parameters may also be predicted using BNN using similar methods.

References

[1] R. Utama and J. Piekarewicz, Phys. Rev C93, 014311 (2016)
[2] R. Utama and J. Piekarewicz, Phys. Rev C96, 044308 (2017)
[3] K.D. Hiumbird et al., IEEE Trans Netw Learn Syst. 1286-1295 (2018)
[4] K. Blaum Phys. Reports 425 (2006)
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LLNL-POST-783119-DRAFT

LLNL-POST-784724

Fractionation of Oxygen Isotopes in Uranium Oxides

Impact of Atmospheric Composition During Calcination

M.R. Klosterman[1], A.L. Deinhart[2], E.J. Oerter[2], L.W. McDonald[1], M.J. Singleton[2]

[1] Civil & Environmental Engineering, Nuclear Engineering Program, University of Utah

[2] Nuclear & Chemical Sciences Division, Lawrence Livermore National Laboratory

THE UNIVERSITY OF UTAH

ABSTRACT

Oxygen isotope ratios in meteoric waters are known to vary as a function of latitude, elevation, and inland distance. As a result, oxygen isotopes have been used in earth sciences for decades as a signature of the origin, formation conditions, and migration of rocks, fluids, organisms, contaminants, and food sources. The purpose of this work is thus to investigate the potential for oxygen isotopes to provide information relevant to the origin, processing history, and movement of uranium materials.

Calculated geographic distribution of δ18O values in meteoric water (Bowen & Wilkinson, 2002).

Uranium oxides are encountered in nearly every stage of the nuclear fuel cycle, from the original ore to the spent fuel. As a first step in this project, a synthetic yellowcake compound, metastudtite, was synthesized by precipitation from a well characterized water source. This material was then calcined at different temperatures, times, and atmospheric conditions to produce oxides of uranium commonly encountered prior to fuel fabrication. The oxygen isotopic composition of all calcined materials was then characterized by extraction of oxygen using High Vacuum Fluorination Systems (HVFS) at both the University of Utah and Lawrence Livermore National Laboratory, followed by mass spectrometric analyses either as CO_2 (Utah) or O_2 (LLNL).

The results of this work show that variation in atmospheric composition during calcination of a uranium oxide has a significant impact on the resulting oxygen isotopic signature of the material. In an inert atmosphere, a lack of oxygen-containing species with which to fractionate leads to the preferential disassociation of ^{16}O and correspondingly higher $\delta^{18}O$ values. In dry air, the calcined oxides undergo a highly temperature-dependent equilibration with atmospheric oxygen, which in this case led to $\delta^{18}O$ values higher than the starting metastudtite. When water vapor depleted in ^{18}O was added to the calcination atmosphere, samples underwent a slower equilibration process that produced highly depleted oxides. The overall range in $\delta^{18}O$ values (> 44‰) measured among samples calcined in different atmospheres highlights the lability of the oxygen isotopic signature during processing.

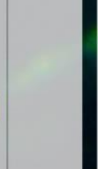
METHODS

Synthesis

Uranyl nitrate hexahydrate ($UO_2(NO_3)_2{\cdot}6H_2O$) was dissolved in previously characterized water (-15.6‰) and subsequently precipitated as studtite ($UO_4{\cdot}4H_2O$) by addition of 30% H_2O_2 (-7.0‰). After rinsing and filtering, the precipitate was dried at 80°C for 24 hours to produce metastudtite ($UO_4{\cdot}2H_2O$).

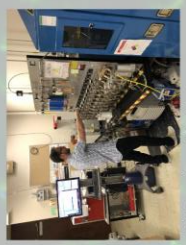
Dry and Inert Calcinations

Samples were loaded into platinum crucibles and calcined for 20 hours in a quartz tube furnace.

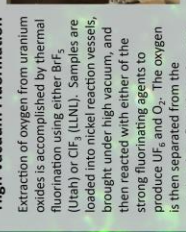


High Vacuum Fluorination

Extraction of oxygen from uranium oxides is accomplished by thermal fluorination using either BrF_5 (Utah) or ClF_3 (LLNL). Samples are loaded into nickel reaction vessels, brought under high vacuum, and then reacted with either of the strong fluorinating agents to produce UF_6 and O_2. The oxygen is then separated from the uranium hexafluoride and excess reagent using liquid nitrogen traps. At Utah, the extracted oxygen is converted into CO_2 by reaction with graphite at 800°C with the aid of a platinum catalyst.



HVFS at the University of Utah



HVFS at Lawrence Livermore National Laboratory

Isotope Ratio Mass Spectrometry

Oxygen isotope ratios are measured offline as CO_2 using a gas chromatograph coupled to a Thermo Finnigan Delta Plus Advantage IRMS (Utah). At LLNL, the fluorination line is coupled directly to an IsoPrime 100 dual inlet IRMS. After purification using liquid nitrogen traps and a heated salt trap, the oxygen is collected on a 10Å molecular sieve before being sent into the IRMS for $^{17}O/^{16}O$ and $^{18}O/^{16}O$ analyses.

Calcinations with Water Vapor

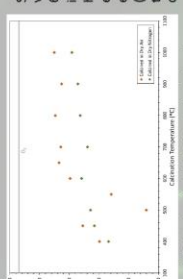
Samples were loaded into nickel capsules and calcined in a TA Discovery Thermogravimetric Analyzer (TGA). Samples were exposed to water vapor (~ 15,000 ppmV) carried by a flow of nitrogen throughout the calcinations. Before each run, the $\delta^{18}O$ value of vapor was measured by coupling the outlet of the TGA to a Picarro L-2130i cavity ring down isotope ratio infrared spectrometry (IRIS) water isotope analyzer.

TGA-IRIS at Lawrence Livermore National Laboratory

RESULTS AND DISCUSSION

Dry and Inert Calcinations

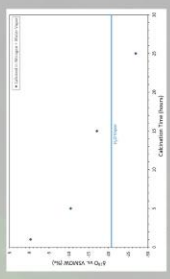
Samples calcined for 20 hours in dry air had $\delta^{18}O$ values that varied significantly from 400°C to 650°C. The transition of Am-UO_3 to α-UO_3 at 500°C is associated with a depletion of ^{18}O. Further heating of the crystalline compound leads to an enrichment of ^{18}O, which may be indicative of equilibrium fractionation with atmospheric oxygen (+23.5‰). In contrast to the highly variable temperature-dependent fractionation, samples calcined in dry air at 700°C between 30 minutes and 20 hours exhibited very little difference (~1‰) in $\delta^{18}O$ values.



Measured $\delta^{18}O$ values of samples calcined for 20 hours in either dry air or nitrogen (100 mL/min gas flow and 10 °C /min ramp rate).

Samples calcined for 20 hours in dry nitrogen showed a minor enrichment in ^{18}O with increasing temperature; however, the transition from amorphous to crystalline phases was not associated with a depletion in ^{18}O. The enrichment in ^{18}O—or more appropriately, the depletion of ^{16}O—likely represents a kinetic fractionation mechanism in which the lighter isotope is preferentially dissociated from the compound and then removed from the system.

Previously measured $\delta^{18}O$ values of samples calcined in a tube furnace at 700°C with a flow of dry air for various durations (Klosterman, 2019, ACS Presentation).



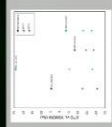
Measured $\delta^{18}O$ values of samples calcined in the TGA at 600°C with a flow of humid nitrogen for various durations.

Humid Calcinations

Samples exposed to water vapor during calcination exhibited a time- and temperature-dependent equilibration process. In contrast to samples calcined in dry air or nitrogen, samples exposed to water vapor (-21‰) showed significant depletion of ^{18}O relative to ^{16}O, with equilibrium $\delta^{18}O$ values lower than that of the vapor. Analysis of samples heated to 600°C for durations between 1 and 25 hours had substantially decreasing $\delta^{18}O$ values, ranging from -0.3‰ to -26.9‰ between the shortest and longest calcination durations, respectively. Under humid conditions, calcination time plays a major role in the ultimate oxygen isotopic composition of a uranium oxide.

FUTURE WORK

Previous analysis of UO_3 and U_3O_8 calcined in natural air in Salt Lake City showed depleted $\delta^{18}O$ values, which suggests a preferential fractionation with water vapor over O_2 when both species are present. Future experiments using controlled atmospheric conditions will help to further understand this process.



Previously measured $\delta^{18}O$ values of different starting materials and their products which were calcined in natural air in Salt Lake City.

THE UNIVERSITY OF UTAH
DEPARTMENT OF CIVIL & ENVIRONMENTAL ENGINEERING

ACKNOWLEDGEMENTS

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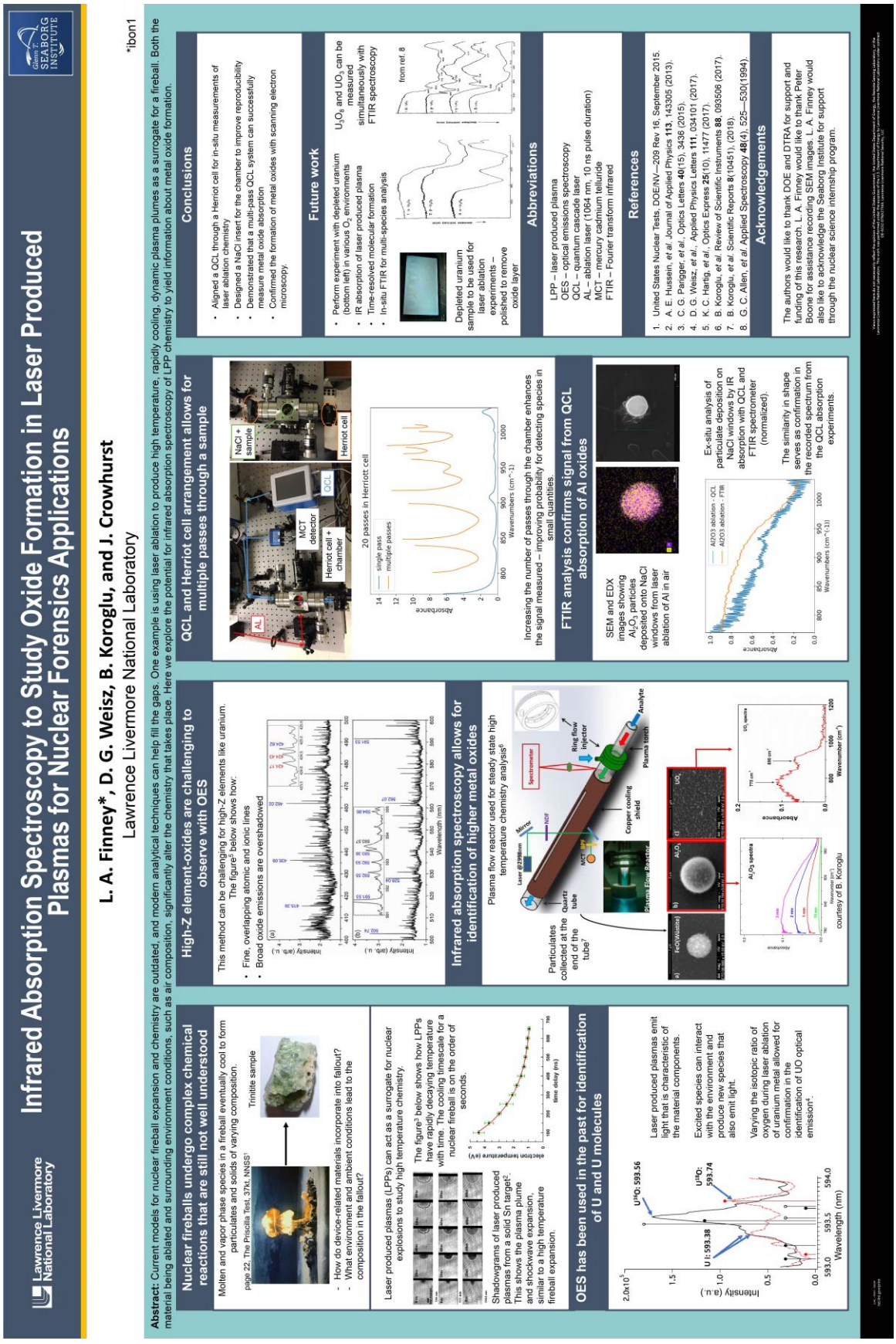
University of Utah Middle Bench Facility for Environmental Research (MBFAR)

LLNL-POST-784357



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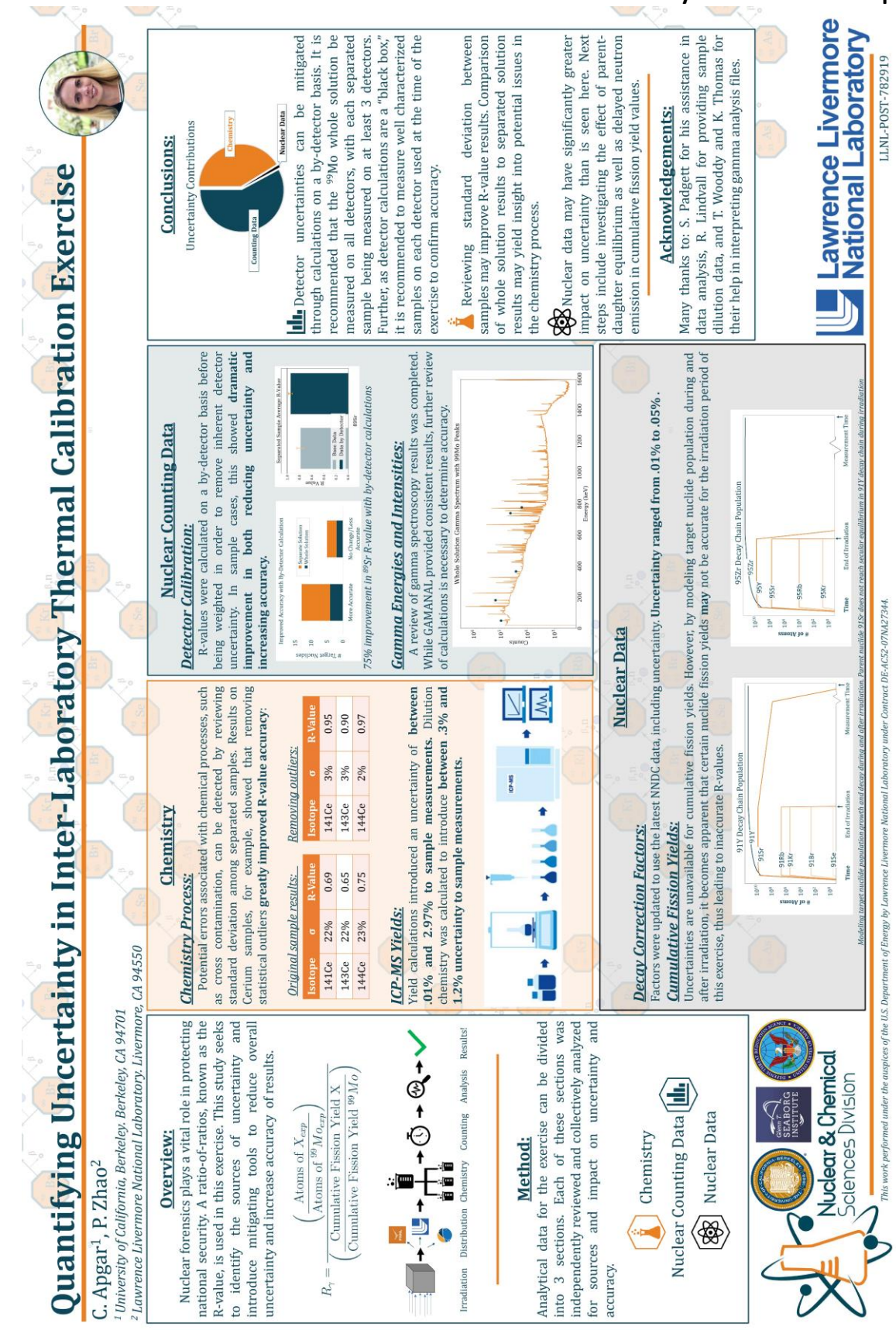


LLNL-POST-782919

Gamma-Gamma Coincidence Counting for Radioisotope Detection

Jacqueline K. Garcia, Tashi Parsons-Davis, Jack Goodell, Keenan Thomas. Stephen Padgett, Nuclear and Chemical Sciences
Lawrence Livermore National Laboratory

Abstract

Background: The purpose of this project is to fully implement gamma-gamma coincidence measurements by developing tools to facilitate rapid assay of activation and fission products for the purpose of early-time nuclear debris analysis. *Methods:* The project is compartmentalized into 4 components: hardware, analysis (and Software), testing/data, and produce documentation. *Set-up:* The detector set-up includes 3 p-type coaxial HPGe detectors with a 70% relative efficiency, BGO Compton suppressors, and CAEN signal-processors/ADC with list-mode data collection. *Results:* The tools developed in this project have demonstrated a significant improvement in the practicality of the system. *Conclusion:* Including automation and conjunction of programming scripts with the hardware system will improve analysis techniques for future work done on radioisotope counting.

Introduction

Post-detonation technical nuclear forensics (TNF) is an intricate and often ambiguous process that requires assay of nuclear debris particles, such as fission products, actinides, and activation products. Assay data is:

- used to assess the fuel type, yield, etc. as quick as possible
- difficult to read due to complex gamma spectra because of high activity
- typically obtained through radiochemical separation which takes time.

The project aims to improve early assay of fission and activation products by eliminating gamma complexity for specific nuclear decay schemes.

This leads to:

- eliminating interference
- identification of weak signatures in high radiation field

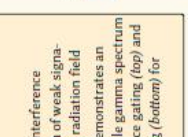
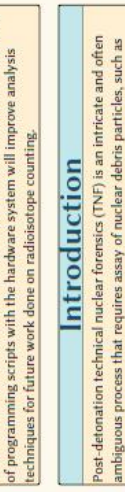


Figure 1: *(left)* demonstrates an early fission sample gamma spectrum with no coincidence gating *(top)* and coincidence gating *(bottom)* for Sb-128 decay.

Methods



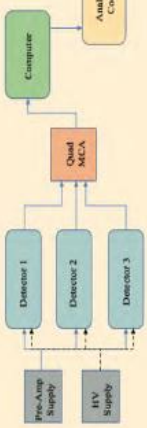
The **test/data** utilizes outputs of the analysis and software component and involves testing the Root scripts, potentially with fission samples.

Experimental Set-up

Figure 2 *above* illustrates the side-looker 3rd detector connected to the other two HPGe detectors allowing for multiple efficiency measurements.

Figure 3 *above* is the final assembled gamma-gamma coincidence, Compton suppression system. Beneath lies the NIM bin with the HV supply.

Detector Hardware Setup



Programming Work-Flow

The *Reduced Files*, in conjunction with the ROOT parser, help narrow down the excessive noise typically seen in fission product counting and help the user quicken the analysis spectra by providing a predetermined list of possible isotopes.

Results

This project demonstrated promising results for the use of **energy** and **time** coincidences to create gated spectra. However, there is a trade-off between coincidence measuring and efficiency, shown in **Table 1**.

Energy (keV)	CH1 eff.	CH2 eff.	CH1Tr2 eff.
1173.23	2.026E-3	1.828E-3	2.580E-5
1332.49	1.774E-3	1.635E-3	2.400E-5

Table 1 *above* demonstrates the lower efficiency encountered in coincidence measuring, thus requiring longer measuring time.

Figure 4a *(top)* is a spectrum of Ag-108m with Cs-137 and Mn-54 collected in singles mode. **Figure 4b** *(bottom)* is the same spectrum collected in coincidence mode with Ch2 triggering on Ch1. Although Compton from Cs-137 and Mn-54 is more present in coincidence mode, the Ag-108m peaks are more prominent.

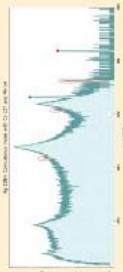
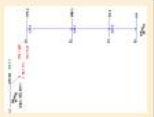


Figure 5 *(right)* demonstrates where the coincidences occur for emitted gamma rays in Ag-108m

Developing this **tool** utilizing Python web parsing and ROOT parsing has proven to improve radio-analysis by greatly reducing analysis time and noise by removing non-coincidence data.

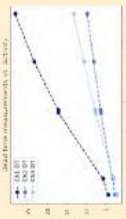


Discussion and Future work

In the immediate future, the next steps are to:

- test the code with fresh samples check for bugs
- find the dead-time limitation *with* coincidence by analyzing dead time effects vs. activity quantity
- alter it for future use (increase its diversity)

Figure 8 on the *right* demonstrates the association between dead time and activity.



In the near future, more dead-time vs. activity calculations will be done to better define this relationship.

The long term goal is to provide quick answers for messy data. This improves analysis time in nuclear forensics and can also help researchers reduce time spent analyzing spectrum data.

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Physical and Life Sciences
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LLNL-POST-784542

Enhancing Resonance Ionization Mass Spectrometry at LLNL: Automated sub-nanosecond timing control to maintain sensitivity and mass resolution

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[2]Materials Science and Engineering Program, University of California, Riverside
[3]Lawrence Livermore National Lab

Abstract

Resonance Ionization Mass Spectrometry (RIMS) offers unparalleled selectivity of species in the determination of isotopic mass from samples of diverse elemental constitution. This property of the technique emerges from the excitation at any given element's characteristic and narrow absorption band to promote a bound electron to a higher energy state. Successive excitations are applied at the appropriate energies such that only the intended elements are ionized and accelerated into a mass spectrometer. The work reported at present is concerned with the requisite tight timing constraints of ablation sources, successive ionization lasers, and signal detectors employed in this technique. A single, unifying graphical user interface software was developed for the control and automation of experimental elements. Improvement of the timings of these sources will serve to enhance the instrument's mass resolution, sensitivity, efficiency, reproducibility, and the operator's ease-of-use.

Introduction

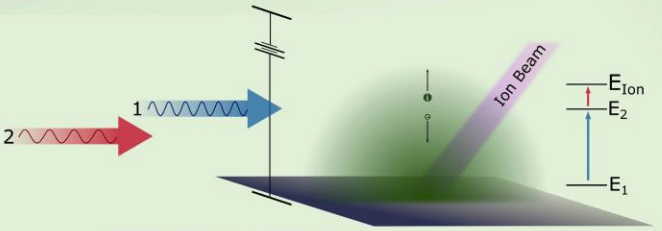
- The RIMS technique is dependent on the tight timing tolerances of experimental components. For instance, if the ion excitation lasers fire prior to or too late after the ion beam ablates the surface of the sample no signal will be detected.
- Programmatic control of components will ensure reliable timings of the equipment.
- Alone, this solution does not address the time evolution of the beam spectral center of gravity inherent in real-world laser systems.
- This will be addressed by continuous monitoring of the beams' spectral center of gravity and programmatically apply slight adjustments to the angle of the Ti:Sapphire lasers' diffraction grating, thus creating an automatic control feedback loop.

Methods

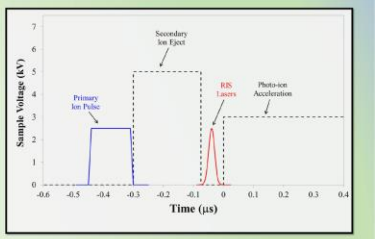
- The equipment used to control the RIMS components are SRS DG645 digital delay generators.
- Automatic communication is established via SCPI commands passed from computer by python 3.7 through ethernet networking.
- A Graphical User Interface (GUI) was designed to provide a unifying control system.
- Periodic monitoring of oscilloscope measurements of beam spectra and the adjustment of timings will address the undesired time evolution of the beams' spectral centers of gravity.

Results

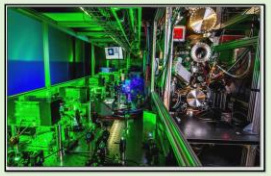
- Programmatic control of the instruments' components has been achieved in a single, unifying GUI.
- The LION instrument's ease-of-use and speed of operation has been improved.
- Still to be completed is the automatic monitoring and control of the lasers' spectral center of gravity.



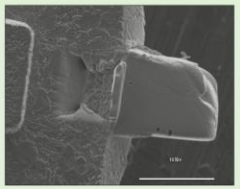
Schematic figure of RIMS technique's interaction with samples. Initially, an ablation source such as an ion beam strikes the sample and sputters off atoms of the sample. Then a series of laser pulses impinge upon the atom cloud to successively promote the electrons of the targeted element.

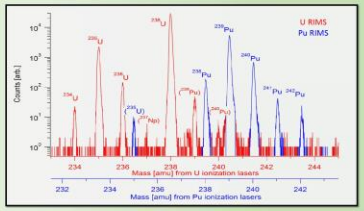


Example timing scheme showing the tight timing requirements in the RIMS technique.

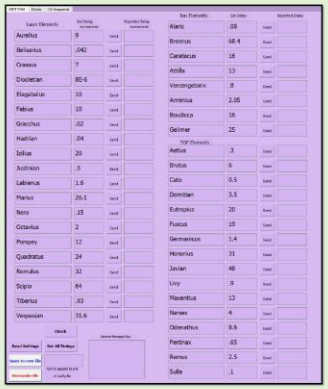


Picture of the LION system with lasers and optical elements to the left and the sample chamber and mass spectrometer on the right.

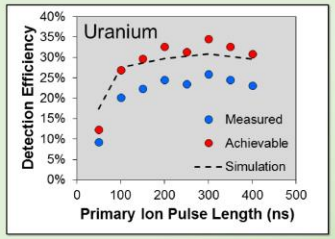




The RIMS technique is highly useful in the field of nuclear science. The extraordinary useful yield allows for the analysis of minute quantities of sample. Additionally, as can be seen for the spent nuclear reactor fuel (SEM pictured left), the technique is capable of attaining simultaneous measurement of isotopes of multiple elements.



Screenshot of the simplified view screen of the control GUI developed in python 3.7 and PyQt5.



Discussion

- The success of the GUI has proven the viability of further automation of this niche technique and could prove to be a vital component in the push towards popularization of the method.
- The future implementation of beam sampling and automatic control could prove an important step in the future full automation of the LION instrument at LLNL.

Measuring Surface Roughness on UO_2 Fuel Pellets for Nuclear Forensics

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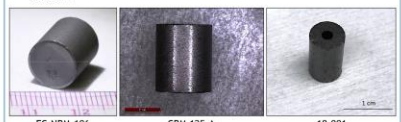
[2]Lawrence Livermore National Laboratory, Livermore, Nuclear and Chemical Sciences Division

INTRODUCTION

Information regarding the bulk composition, macroscopic properties, trace impurities, and isotopic composition of elements is crucial to assemble any practical conclusions from a given sample regarding production location and/or process history (Moody et al., 2014). The enrichment and fabrication of uranium into UO_2 fuel pellets plays a major role within the nuclear fuel cycle. UO_2 fuel pellets undergo grinding procedures to create their cylindrical shape with dimensions set by the production plant's specifications. Previous studies have shown that measuring UO_2 fuel pellet surface roughness via profilometry can indicate production location based on the grinding method (Pajo et al., 2001). There is currently no standard procedure in place to measure surface roughness of UO_2 fuel pellets and still little to suggest it could play a role as a robust forensic signature. This work aims to determine a best practices method for accurately measuring surface roughness to determine fuel pellet provenance.

SAMPLE PREPARATION

- Samples of interest for this method development study include:
  - Natural fuel pellet, intact
  - Fuel pellet piece, 3-4% ^{235}U enrichment
  - Fuel pellet piece, 17% ^{235}U enrichment
- Images of each sample can be found in Figure 1, and physical measurements were acquired using digital calipers, as seen in Table 1.



EC-NRM-106 CRM 125-A 18-001

Figure 1. UO_2 fuel pellets.

Zygo ZeGage Optical Profilometer

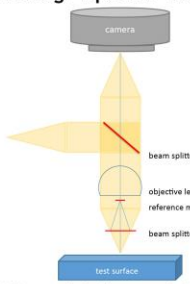


Figure 2. How an optical profilometer works (Adapted from Zygo).

METHOD DEVELOPMENT

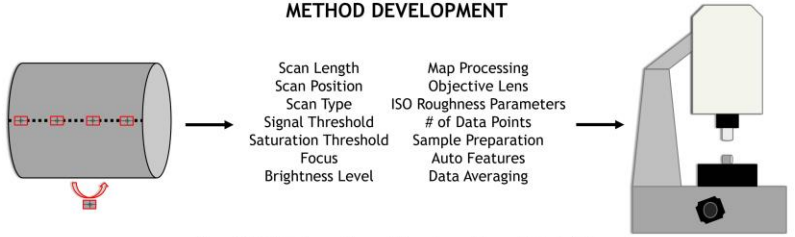
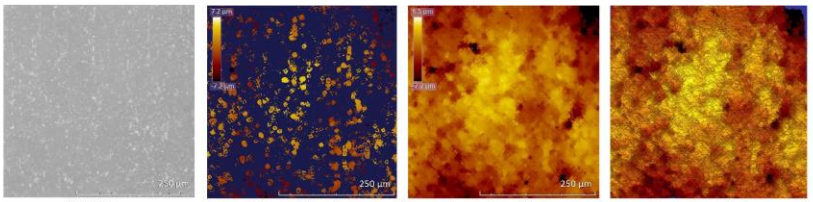


Figure 3. Method development for quantifying surface roughness on UO_2 fuel pellets.



Stage view | Data acquisition view | Processed map, 2D view | Processed map, 3D view

Figure 4. Data acquisition and map processing.

RESULTS

Table 1. Diameters, average areal surface roughness (S_a) and average profile surface roughness (R_a) of UO_2 fuel pellets.

Sample Name	Reactor Type	Diameter (mm)	Hole Diameter (mm)	Height (mm)	S_a (µm)	Std Dev (σ)	R_a (µm)	Std Dev (σ)
EC-NRM-106	1	10.62 ± 0.01	N/A	12.16 ± 0.01	1.68	0.11	1.39	0.17
CRM 125-A[1]	2	8.15 ± 0.08	N/A	9.96 ± 0.17	1.82	0.10	1.82	0.22
18-001[1]	3	5.77 ± 0.02	1.71 ± 0.01	9.01 ± 0.01	2.90	0.34	2.90	0.48

[1]Physical measurements of intact pellets were taken from previous work.

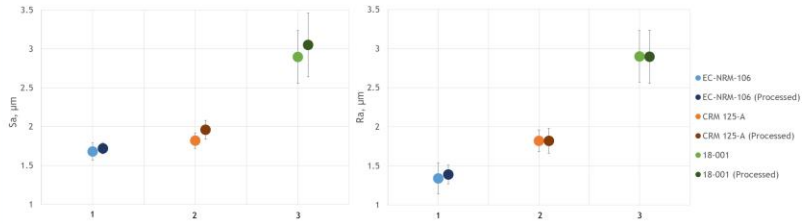


Figure 5. UO_2 fuel pellet surface roughness calculated using Sa values (left) and Ra values (right), as well as post-processing measurements versus raw data values.

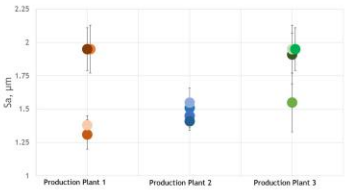


Figure 6. Ongoing fuel pellet measurements.

DISCUSSION

Project Objectives:

- What parameters will be used to determine surface roughness of the material?
- How many analyses are necessary per sample to acquire a sufficient amount of surface roughness data?
- What are the differences between fuel pellets that have undergone grinding versus fuel pellets that have not?
- Is it possible to differentiate provenance through surface roughness measurements using optical profilometry?

Takeaways:

- The traditionally used Ra value for quantifying surface roughness via 2D profile measurements is variable and representative of a small sample set.
- Sa values pertain to the roughness average of a 3D areal view of the surface, providing a more complete picture of the bulk while maintaining constrained uncertainties.
- Parameters such as scan length and measurements per sample were optimized for efficiency and accuracy.
  - 5 regions along the length of the fuel pellet were measured, within a 400 x 400 µm area.
  - For shorter pellets, 5 regions along the cylinder were measured within a 400 x 400 area.
- Fuel pellet fragment from producer 3, which showed signs of no grinding, displayed the highest surface roughness values.

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