

Radiochemistry and Nuclear Chemistry Focus Areas

Ken Czerwinski
Department of Chemistry
University of Nevada - Las Vegas

NSSC3 Kickoff Meeting and Advisory Board Review
April 19-20, 2022

Research focus area combines foundations behavior of radioelements and radionuclides. Studies utilized compound synthesis, resulting chemical behavior, species identification, and role of radioactive decay

Chemical Synthesis and Speciation: Link fundamental speciation and physicochemical properties, initial focus on halide species

- Continue efforts on metal-organic frameworks to examine specific photophysical signatures

Advanced Spectroscopic Techniques: Develop rapid analytical methods using molecular nuclear forensics

- Synchrotron-based methods and advanced mass spectrometry

Chemical Separations: Apply radionuclide speciation to development of new separation methodologies; new ligand systems and microfluidic techniques

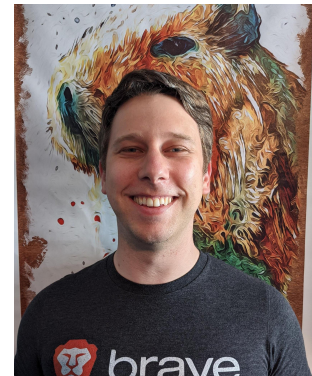
Isotope Based Reactivity: Isotope fractionation; nuclear field shift effect expected to be evident in higher Z elements

Key Personnel



George Washington University:
Prof. Cahill, Benjamin Walusiak,
LANL: Alice Smith

- Consortium member on Walusiak thesis committee

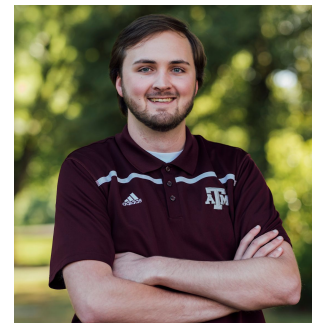


University of Tennessee:
Professor Hall, Ben Jordan,
ORNL: George Chan



Texas A & M:
Prof. Folden, Jenna Garcia, Isaac Haynes,
LANL: John Auxier and Iain May

- Isaac Haynes will participate in 2022 Keepin Nonproliferation Summer School



UC Berkeley: Profs. Arnold and Abergel,
Mark Straub

LBNL: Stephan Minasian,
Post-doc Gaiser,
SNL: D. Farley and E. Uribe

- Inclusion of SNL, continuing synthesis studies



University of Nevada, Las Vegas:
Profs. Poineau, Czerwinski, Gelis, Koury,
Hatchett

Students: Nicholas Chicchetti, Harry
Jang, Josephine Libero, Liuba Williams,
James Louis-Jean, Katherine Luebke

- Collaborations with LBNL, LLNL, LANL, NNSA
- Leveraging consortium for additional funding from MSTC



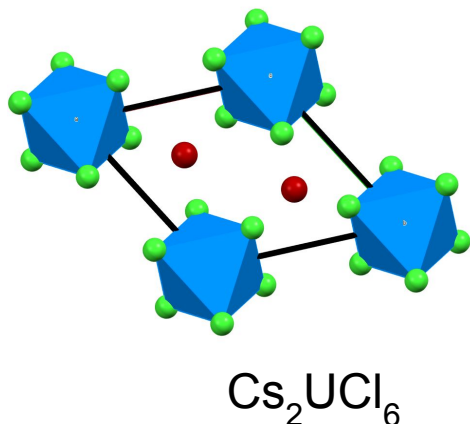
Range of laboratory partners in focus area with overlap of research areas

- LBNL: Natural collaboration with UCB, exploring compound synthesis and spectroscopy. NSSC brings new compounds and actinide species under investigation
- LLNL: Studies on molten salt chemistry of actinides for separation and purification. NSSC adds capabilities in synthesis area to collaboration
- SNL: Expanding focus area effort with another national laboratory, advanced spectroscopic method, NSSC bring evaluation of additional isotopes (^{231}Pa , ^{227}Ac)
- LANL: Multiple collaborations; (GWU) Molten salt chemistry, evaluate Cs_2UCl_6 , 2022 Seaborg Institute Graduate research program; (TAMU) Nuclear forensics on Am, radiochronometry, extension of effort within NSSC; (UNLV) TIMS of fission products, developing lower limit of detection
- ORNL: UTK interaction, Examine of UF_6 spectroscopy. Combines material available at ORNL with UTK spectroscopic capability



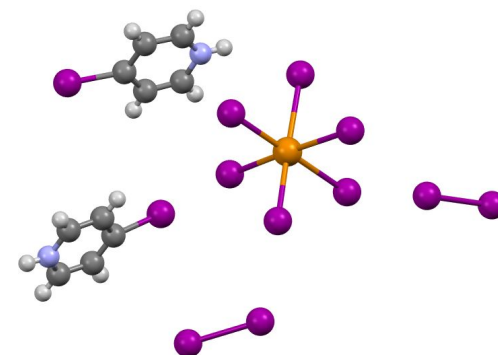
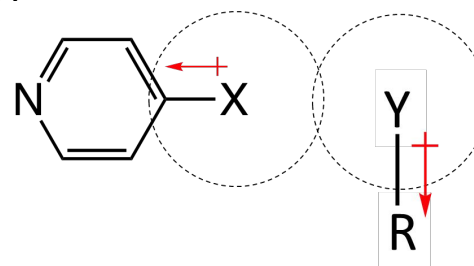
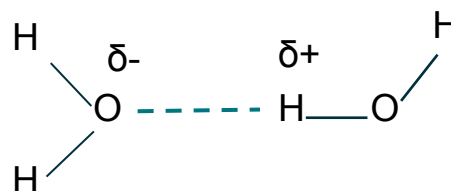
Non-Covalent Interactions Tel_6 Species

Proposed Efforts at LANL

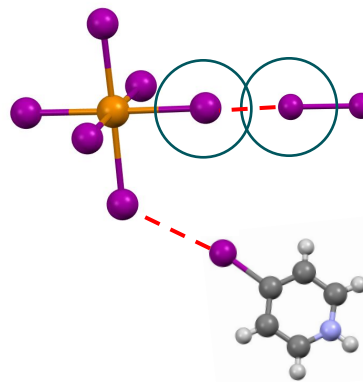


- Potentially important for molten salt reactor or reprocessing
- Interface of fission product with fuel species
- Examine with Neutron diffraction, Differential scanning calorimetry

Hydrogen Bonding



Organic-Inorganic Hybrid



$(\text{IPy})_2[\text{Tel}_6] \cdot \text{I}_2$

Non-covalent interactions play a major role in determining material properties (band gap, frontier orbital landscape)

Americium ($Z = 95$) has widespread commercial and industrial uses. It also has multiple long-lived isotopes (^{241}Am , $^{242\text{m}}\text{Am}$, ^{243}Am). These are fissionable isotopes that are considered alternate nuclear materials.



Americium has potential for use within a nuclear dispersive device and/or a fission weapon, with no current forensic methods that focus on americium-based samples.

Project: Establish a Nuclear Forensic Method for Americium

- Radiochronometry via a combination of α and γ spectrometry.
- Trace contaminant determination via column chromatography & mass spectrometry.
- Collaboration with Texas A&M Nuclear Engineering for reactor-type determination via fission product traces.

Connections to NNSA mission

- Students receive technical training on radiochemical techniques.
- Project began from collaboration with Isaac's NSSC mentor, Dr. John Auxier, at Los Alamos National Laboratory.
- Isaac will work under Dr. Iain May in the NSSC Keepin Nonproliferation Summer program for summer 2022.

This work aims to bring awareness to ^{226}Ra pigments and paints potential usage in nuclear terrorism attacks.

Radium pigments and paints

- Used extensively in the early 1900's
- IAEA states potential usage in RDDs

Nuclear forensic analyses

- Build forensic signature database for ^{226}Ra
- Radiochronometry and trace metal analysis



Laboratory connections

Jenna has previously participated in the 2021 NSSC Keepin program at LANL under Dr. Evelyn Bond focusing on nuclear forensics. She plans to return to LANL this summer with Dr. Bond under the Seaborg Fellowship

Project connection to NNSA missions

Students receive technical training using radiochemical techniques for nuclear forensic applications ultimately to reduce the threat of radiological terrorism around the world

“The NSSC fellows and affiliates of the UNLV Radiochemistry Program are performing research to advance the science of radioelements for nuclear security and nuclear energy applications.”

Harry Jang (Fellow) Preparation uranium microspheres as targets for isotopes production and alternative nuclear fuels. He developed a new method for the preparation of uranium fluoride microsphere.

Academic Advisor: **F. Poineau**

Lab interaction : **LANL-2021 summer internship**

(mentor: **A. Pugmire**)

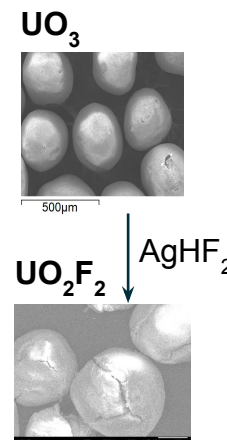
Accomplishment: **Review-article**

(UNLV/LLNL/PNNL) on U metal preparation

Synthetic diversity in the preparation of metallic uranium

Harry Jang¹, James Louis-Jean¹, Bradley Childs², Kiel Holliday³, Dallas Reilly³, Matthew Athon³, Kenneth Czerwinski¹, David Hatchett¹ and Frederic Poineau¹

¹Department of Chemistry and Biochemistry, University of Nevada, Las Vegas, NV, USA
²Materials Science Division, Lawrence Livermore National Laboratory, Livermore, CA, USA
³Pacific Northwest National Laboratory, Richland, WA, USA
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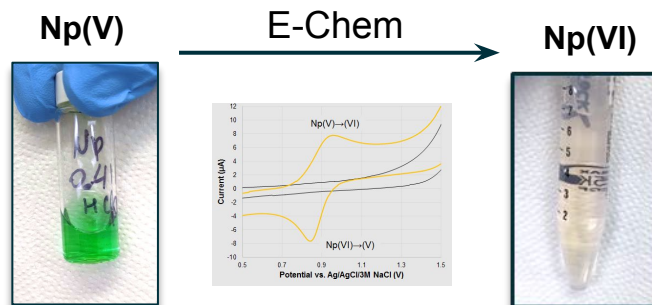


“The reaction between UO₃ and AgHF₂ at 200 °C yield UO₂F₂ microspheres with minimal swelling.”

Nicholas Cicchetti (Fellow) Behavior of Np and Pu in concentrated base using electrochemical and spectroscopic methods.

Academic Advisor: **A. Gelis**

Lab interaction : **LBNL-2022 summer internship (mentor: D. Shuh)**



“The electro-oxidation of Np(V) in HClO₄ at 1.3 V produces Np(VI).”

Josephine Libero (Affiliate) New actinide-transition metal alloys to assess properties Provides data to monitor burn-up for safeguard applications

Academic Advisor: **D. Koury**

Lab interaction : **LLNL-2022 summer Internship**

Liuba Williams (Affiliate) Using arc-welder techniques to prepare realistic urban melt glass for characterization.

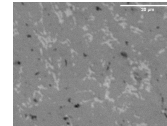
Academic Advisor: **D. Koury**

Lab Interaction : **Nevada National Security Site-MSTS-2022 summer internship**

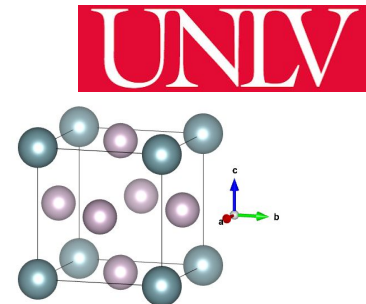
Logan Smith (Affiliate) is developing methods for the separation of radionuclides (i.e., ^{90}Y and ^{225}Ac).

Academic Advisor: **A. Gelis**

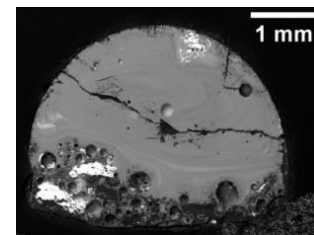
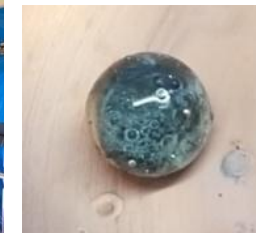
Lab interaction : **LNLL-2022 summer internship**
(mentor: **M. Mitchell**)



UTc_3

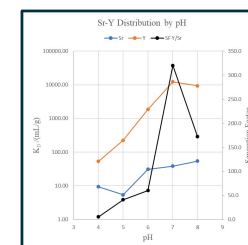


“A new alloy, UTc_3 , was prepared from arc-melting of Tc and U and analyzed by SEM, XRD and theoretical methods.”



(Left) 10%Fe glass bead after arc melting.
(Right) SEM cross-section of 10% Fe_3O_4 bead.

“Arc-melting is used prepare glass samples which composition and morphology are compared to Trinitite samples. “

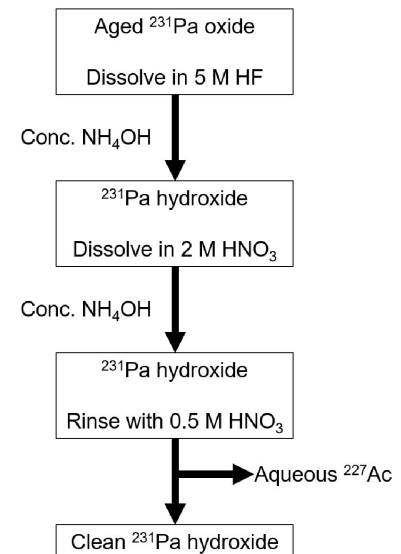
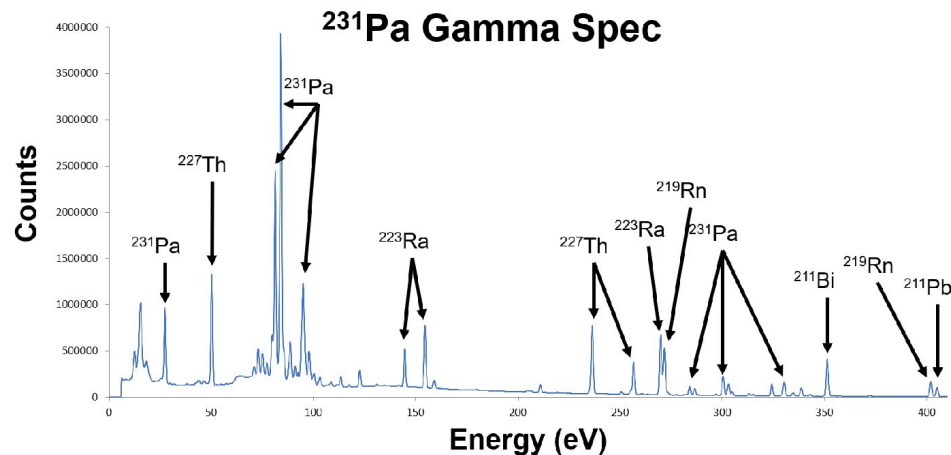
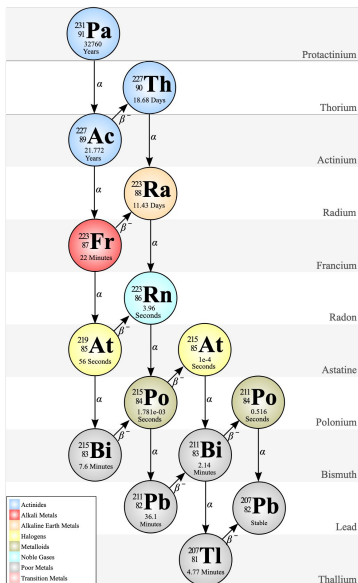


“ ZrO_2 columns are used to efficiently separate Sr-90/Y-90”

Laying the groundwork for Pu/U and Th/Pa separations technologies that will be relevant for online reprocessing of fissile materials in molten salt directly applicable to Molten Salt Reactor (MSR) development



- Purification of legacy ^{231}Pa materials followed by sample spikes in various salt environments for characterization, collection of ^{227}Ac
- Collaboration with SNL
- LIBS development, NMR, Raman spectroscopy





Kevin Glennon

- Kevin was funded by NSSC2 for his research in nuclear forensics. Mentor: Evelyn Bond, LANL.
- His thesis ([link](#)) culminated with a forensics analysis of plutonium containing numerous signatures consistent with production during the Manhattan Project. ([link](#))
- Kevin is now a postdoctoral researcher at LLNL.



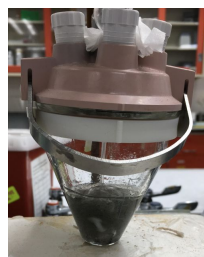
Mariah Ramirez

- Mariah was funded by NSSC2 for her research in nuclear safeguards. Mentor: Rene Sanchez, LANL.
- Her thesis ([link](#)) focused on developing safeguards for ^{237}Np .
- Mariah is now an engineer with IB3, a nuclear engineering firm in Oak Ridge, Tennessee.

Academic Advisor: **D. Hatchett**
 Lab Mentor: **Robert Rundberg (LANL, CN-R)**
 Standing: **Graduate Student**
2019 Seaborg Research Fellowship at LANL
Electrochemical Recovery of Uranium using Ionic Liquids

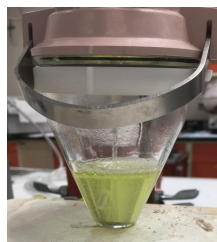


Katherine Luebke



U_3O_8

Dissolution



Electro-deposition



UO_2

LANL Staff Scientist-2, June 2022

Academic Advisor: **F. Poineau**
 Lab Mentor: **Jeremy Inglis (LANL)**
 Standing: **Graduate Student**
2019-2020 African American Partnership Program at LANL
2018 -2019 Seaborg Research Fellowship-7 articles as 1st author
2020 DOE Innovations in Nuclear Technology R&D Awards
*Revisiting the Chemistry of Uranium and Rhenium Fluorides:
 Implication to Fuel Cycle and Nuclear Forensics Science.*

Post-doctoral researcher at SRNL, starting June 2022



James Louis-Jean

Acknowledgements



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