NUCL 4282707

Using spectroscopy to further investigate uranyl bonding in ammonium diuranate

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The crystal structure and chemical formula of ammonium diuranate (ADU), general formula (xUO_3●yNH_3●zH_2O), is known to vary as a function of the synthetic conditions used to prepare the material. We prepared ADU through an ammonium hydroxide (NH_4OH) precipitation route to form $3UO_3$●NH_3●$5H_2O$, which was verified by powder X-ray diffraction and thermogravimetric analysis. Our interest in doing so, was to determine the exact nature of the uranyl (UO_2^{2+}) metal-coordination environment, and the identity of the ligands and non-coordinated species. We have therefore examined the pentahydrate form of ADU using photoluminescent (PL) spectroscopy to study the coordination environment and, moreover, determine if unexpected species like hydroxide or ammonium are present. The PL analysis revealed two distinct emissive components in the blue (440 nm) and green (535 nm) regions. The blue-centered emission is surprising because coordinated water is not sufficiently reducing to cause this behavior. Interestingly, cooling the specimen to 77 K suppressed the blue-centered emission, implying a ligand and/or temperature dependence. This behavior was studied further via the intentional dehydration of $3UO_3$●NH_3●$5H_2O$ by heat treatment to form a dehydrated species, presumably $3UO_3$●NH_3●$1.5H_2O$, because the hydration state of ADU is known to causes changes in the UO_2^{2+} coordination environment. Reexamination by PL revealed the loss of the green emissive component at ca. 550 nm, and a notable increase in the intensity in the blue region at ca. 450 nm. This suggests the loss of water caused a redistribution of reducing ligands to the equatorial binding sites on the metal center. Presented here will be our investigation of the uranyl bonding environment and composition of ADU and the associated spectroscopic and structural data.

NUCL 4285449

Spectral sleuthing of transition metal signatures in hydrothermally synthesized uranium dioxide

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Radiological dispersion devices (RDDs) pose an increasing threat highlighting the need for forensic methodologies to trace radioactive materials back to their geological and processing origins. Unlike highly regulated nuclear weapon materials, radioactive sources used in industry, medicine, and research are relatively accessible, posing significant security risks. Identifying reliable forensic signatures, specifically elemental impurities within uranium dioxide, can substantially enhance capabilities to trace and

mitigate the use of illicit nuclear materials.
This study focuses on developing an accurate pre-detonation chemical fingerprinting method for uranium dioxide synthesized via a one-step hydrothermal reaction from uranyl nitrate. Hydrothermal synthesis is chosen due to its reproducibility, control over product morphology, and practical relevance in simulating UO_2 materials that might be encountered in illicit scenarios.
To investigate the provenance indicators, this study examines the deliberate incorporation of transition metals chromium, iron, and nickel into UO_2. Transition metals have a documented persistence through extensive uranium processing and enrichment stages, providing potentially robust forensic signatures. Incorporation is attempted via the addition of transition metal salt solutions directly into the hydrothermal reaction mixture and solid-state incorporation through controlled melting. These methods are systematically compared to assess the degree and uniformity of transition metal integration within UO_2 matrices.
Characterization is done with X-ray diffraction (XRD), Raman spectroscopy, and scanning electron microscopy coupled with energy dispersive spectroscopy (SEM-EDS). Like tracing subtle clues in a molecular mystery, each metal leaves a distinct signature that is revealed through meticulous spectral interrogation.
This pre-detonation forensic characterization framework establishes clear elemental and spectroscopic signatures correlating to specific synthesis and processing conditions. While this investigation currently presents only pre-detonation data, these findings provide the foundational knowledge required for future post-detonation analyses. Ultimately, these chemical fingerprints can support forensic investigations aimed at tracing the origins and processing histories of radioactive materials utilized in potential terrorist scenarios involving RDDs.

NUCL 4291828

Separation and purification of praseodymium and neodymium using ligand assisted displacement cation exchange methods for the purification of accelerator produced Nd-140

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Isotope pairings such as Nd-140/Pr-140 are coveted for their “theragnostic” capabilities, where Nd-140 provides therapeutic potential in the form of Auger elections, and Pr-140, its daughter, provides diagnostic value via Positron Emission Tomography (PET) imaging. Accessing this theragnostic pair via accelerator production involves proton bombardment of ^{nat}*Pr-141. Thus, these radionuclides must be purified from accelerator target materials before being used for healing purposes. Separation of the lanthanide elements, particularly adjacent elements like Pr and Nd, proves difficult because of their nearly identical chemical behavior. Given that the lanthanide elements commonly prefer the 3+ oxidation state, separation strategies rely on exploiting their slight differences in*

ionic radii. Ligand-assisted displacement column chromatography takes advantage of the competitive interactions between lanthanides, organic extractants, and solid support resins necessary to finely separate these elements. We demonstrated that the pH of EDTA solutions used for the MP-50 cation column elution was critical for successful debulking of Pr from the solution containing 500 mg of lanthanides ($[Pr^{3+}]:[Nd^{3+}]$ = 11.5 to 1). In the initial studies 90% Nd was recovered with up to 7% purity. This approach represents the first step in chemical separation in the process of Nd-140 production from Pr-141 targets with proton beam.

NUCL 4299517

Evaluation of sulfamic acid as an EPR dosimeter for in vivo quality control in HDR brachytherapy

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*Ensuring accurate and safe dose delivery is crucial in **high-dose-rate (HDR) brachytherapy** to minimize the risks of underdosing or overdosing. In vivo dosimetry plays a vital role in **radiation safety** and **nuclear medicine.** Developing a reliable, precise, and sensitive dosimetric system is essential for improving treatment quality and patient safety.*

*In this study, sulfamic acid was investigated as a potential dosimetric material for **EPR (Electron Paramagnetic Resonance) dosimetry**, a **physical chemistry technique** known for its accuracy and non-destructive properties. Sulfamic acid samples weighing **80±1 mg** were irradiated using a **0.355 MeV iridium-192 source (Nucletron microSelectron)** at the Ryad Oncologia Clinic in Morocco to assess its dosimetric performance in HDR brachytherapy. The samples were exposed to radiation doses ranging from **5 to 50 Gy** to establish a dose-response curve, and a dose of **40 Gy** was delivered to different dosimeters to map the dose distribution.*

*EPR spectroscopy was employed to analyze the irradiated samples, using **peak-to-peak measurements** for dose estimation. The results demonstrated that the **Sulfamic Acid/EPR system provided dose estimations comparable to the treatment planning system (TPS) calculations**, confirming its potential as a promising **quality control tool in HDR brachytherapy.***

*This work also contributes to advancements in **radiation technology and environmental safety** by improving dosimetry techniques that enhance precision in cancer treatments.*

NUCL 4300615

Combinatorial approach for measuring phase transition behavior of multicomponent salt systems

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Active fission products such as lanthanide metals, alkaline earth metals and some actinides from the used nuclear fuel (UNF) get accumulated in LiCl-KCl eutectic salt electrolyte during the electrorefining operation of pyroprocessing technology. The accumulation of fission products results in the formation of a molten multicomponent system. Lanthanides are of particular concern as they tend to form soluble chlorides ($LnCl_3$*, etc.) that adversely affect the physical and thermodynamic properties of the molten salt electrolyte. The acquaintance with the melting temperature of the multicomponent system, and solubility of lanthanide compounds in the LiCl – KCl eutectic salt is crucial for the effective purification of an electrolyte to reuse it again.*
In this work, melting temperatures of single component salt and the binary LiCl – KCl salt system at various concentrations were measured using thermal ramping analysis coupled with first order derivative approach. The phase transition temperatures of the binary salt system were accurately determined through this novel approach, which was then further implemented to analyze the ternary $NdCl_3$ */LiCl – KCl eutectic salt system with* $NdCl_3$ *between 7.0 and 18.0 mol. %. This work emphasizes phase transition data for relatively higher concentrations of* $NdCl_3$ *(7.0 – 18.0 mole%) in LiCl – KCl eutectic salt in comparison to previous work. Furthermore, the solubility of* $NdCl_3$ *in LiCl – KCl eutectic salt was attempted to measure using an inductively coupled plasma mass spectrometry (ICPMS) technique over the temperature range of 500.0 – 700.0°C. In addition, an industrial endoscope was used to confirm the physical state of the salt systems during the experimentation. The melting temperature, solubility, and visual observation findings would provide a comprehensive understanding of the multicomponent chloride salt systems at elevated temperatures.*

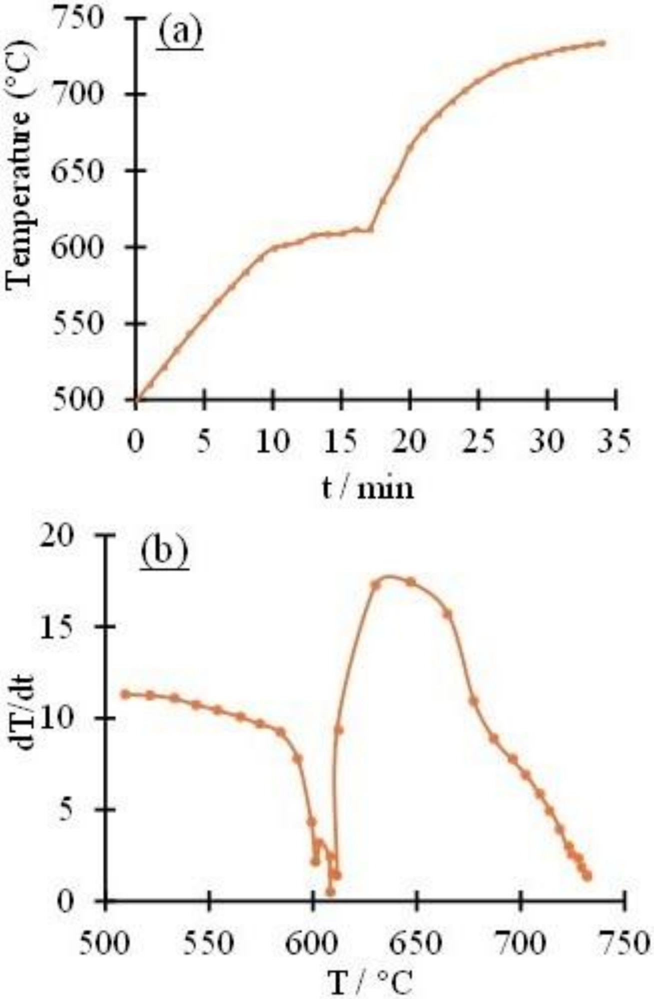

Fig. 1. (a) Pure LiCl temperature profile (b) first order derivative

NUCL 4301291

Uranium- and thorium- containing anions studied by photoelectron spectroscopy

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Experimental anion photoelectron spectroscopy (aPES) is a powerful technique for probing the neutral electronic structure of uranium- and thorium- containing small molecules and ligated complexes by photodetaching their anionic precursors. The interpretation of such experimental spectra is essential for advancing high-level computational methodologies, as well as understanding molecular anionic species relevant to nuclear technology and actinide separation science. In particular, the diffuse 5f orbitals, strong spin-orbit coupling, and sheer number of electrons in heavy elements like uranium and thorium result in complex bonding motifs, posing a significant challenge for electronic structure calculations. In this presentation, actinide micro cluster anions (e.g., Th_{2-7}^- and U_{3-8}^-), small molecular anions (e.g., UF^-, $US_{1,2}^-$, $U(BH)_2^-$, and $U_3O_8^-$ (yellowcake)) , as well as ligated uranyl complex anions (e.g., $UO_2(OAc)_{2,3}^-$) have been generated in the gas phase, photodetached, and their experimental spectra corroborated computationally. In addition, the molecular structure of actinide dioxide cluster anions have been mapped. Overall, the systems in this presentation contribute to a deeper understanding of actinide bonding, with implications for nuclear chemistry and actinide separation science.

NUCL 4301755

Redox reaction of iron under γ irradiation of iron oxide particle aqueous dispersion

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Many studies about the radiolysis of water in the presence of metal oxides have been reported. The presence of Al_2O_3 particles increased the production of molecular hydrogen. Iron oxide particles were also studied to generate H_2 gas. The researchers measured the amount of H_2 molecules. They characterized the iron oxide particles by temperature-programmed desorption, diffuse reflectance infrared Fourier transform *spectroscopy, scanning electron microscopy, Raman spectroscopy, X-ray photoelectron spectroscopy, and X-ray diffraction. However, they did not investigate the redox reaction of iron under γ irradiation of iron oxide particle aqueous dispersion. In this study, we measured the ratio of Fe^{II}/Fe^{III} of iron oxide particles on γ irradiation by the o-phenanthroline spectrophotometric method.*

Iron oxide particles were prepared using the co-precipitation method with $FeCl_2$ and $FeCl_3$. The particles were fractionated with nylon mesh (pore size 46 μm) and filter papers (pore size 6, 3, 1 μm). The iron concentrations of iron oxide particle aqueous dispersions were adjusted to 1-150 mg/mL. The irradiation experiments were carried out for 1 hour under an argon atmosphere at the research laboratory for quantum beam science at Osaka University. The amount of hydrogen produced from a 10-mL dispersion at dose 1.04 kGy was measured by GC with a pulsed discharge detector. The divalent iron concentration of test solutions was determined by the o-phenanthroline spectrophotometric method. The test solutions were divided into two portions. One portion was measured directly to determine Fe^{II}, and another was measured after Fe^{III} was reduced to Fe^{II} with hydroxylamine. As a result, iron oxide particles preferred reduction of Fe^{III} to Fe^{II} under high Fe concentration. On the other hand, the particle size did not bring a significant difference.

NUCL 4306835

Effects of oxidative and reducing flow gases on the combustion synthesis of uranium oxides

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Using bench-scale experimental setups recently established at Oak Ridge National Laboratory (ORNL), we explore the effects of flow gas choice on the combustion synthesis of uranium oxides. Solution combustion synthesis (SCS) is a unique method that offers rapid, easily scaled, and energy-efficient means to produce bulk uranium oxides. Recent work at ORNL has explored the use of SCS in incorporating taggants into uranium oxides for intentional forensics research. Exploring the effects of varying oxidative and reducing flow gases allows for optimization of such systems and subsequent effects on produced materials. Here, we present results from said flow gas investigations, including effects on oxide products and improved understanding of the underlying combustion process. Based on these findings, we also provide results on the bulk scale production of uranium oxides for the formation of an analogue fuel pellet and the incorporation of nickel taggants through combustion using a reductive atmosphere.

NUCL 4308224

Novel spent fuel reprocessing method utilizing solid state actinide fluoridization and antisolvent recrystallization

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Nuclear power is the most economical and sustainable method to meet the growing baseload global energy demand while limiting fossil fuels. Although nuclear reactor fuel is not inherently renewable, it can be recycled repeatedly, offering a pathway for enhanced clean energy production while limiting environmental impact from mining. While other countries have enacted closed-cycle technologies, the United States has yet to implement these systems due to proliferation concerns and secondary waste management difficulties. Due to its history as a weapons production technique, wide-spread reimplementation of PUREX reprocessing raises significant concerns about nuclear proliferation, stunting commercial reprocessing work in the US and other countries. Two-phase reprocessing techniques like PUREX are wasteful, generating substantial quantities of highly acidic radioactive waste and using unstable organic reagents.

This study proposes a novel approach to bulk actinide separation through a gas-mediated solid-state reaction of spent actinide oxide fuel with ammonium bifluoride. The low-temperature reaction produces actinide fluoride salts that are unusually soluble in a saturated ammonium fluoride solution. Further purification and recovery of the actinide salts is accomplished by antisolvent recrystallization. Notably, this process does not rely on the use of hazardous high-temperature gaseous hydrogen fluoride or strong acids. Additionally, preliminary results have shown that the recycling process can be completed within a practical timeframe, and with minimal additional purification steps, thereby offering a promising alternative for future nuclear fuel reprocessing.

NUCL 4308983

Pyridine-based covalent triazine framework for lanthanide-actinide separation

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Covalent triazine frameworks (CTFs) are a porous organic polymer, cross-linked by the chemically stable triazine ring. Despite their rich chemistry, robustness, and high porosity, CTFs remain largely unexplored as a sorbent for metal ion separations. Here, a dicyanopyridine-based CTF is characterized as an ion-exchanger and optimized for the aqueous chromatographic separation of lanthanides from actinides.

NUCL 4309537

Neural network-based effect detection applied to plutonium oxide morphology

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Statistical effect detection of sample processing parameters on particle morphology is an important goal of the material sciences. Current advancements in the field of computer vision have found that models trained on large sets of particle micrographs—or even everyday images—produce quantitative representation that effectively assist in downstream classification of particle characteristics. However, there remains a need for statistically rigorous methods to detect effects on these high-dimensional encodings to determine if a model's representations are robust enough to make relevant classifications. Prior work demonstrated success using non-parametric approaches such as classifier 2-sample tests (C2STs) and permutation-based high-dimensional distance tests to detect effects like strike order, oxalic feed type, and calcination temperature on encodings of synthesized Pu(III) oxalates. This work introduces a hybrid approach leveraging both the sensitivity of C2STs and precision of permutation distance tests. High-dimensional representations are generated by sending scanning electron micrographs of plutonium oxide (PuO_2) through a ResNet-50 model pretrained on the ImageNet dataset, enabling the training of 2-sample classifiers on these representations to detect processing parameter effects such as precipitation routes (III oxalate, IV oxalate, and non-oxalate routes) and calcination temperatures (450°C, 650°C, 950°C). Successfully detected effects will undergo further analysis using distance-based tests, serving as a more conservative approach to demonstrating the capability of neural network representations to capture variation in morphology associated with processing techniques. We will test the extent to which these approaches can achieve low data (single particle) classifications. Finally, a sensitivity analysis of these methods to particle size distributions and image preprocessing techniques will be presented.

NUCL 4309610

Adaptation of a reactor-type discrimination methodology to americium samples

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Nuclear forensics historically focused on both post-detonation analyses, and the U (and Pu) fuel cycles, with less attention paid to byproducts not integral to these. However, ^{241,243}Am are both widely proliferated nuclear materials used extensively in civilian industries, which creates opportunities for theft/diversion towards nuclear terrorism. Both isotopes would be attractive for radiological dispersive device attacks and are vulnerable to non-state and state actors. As a result of these risks, Am deserves more attention across the fields of both nuclear forensics and nuclear nonproliferation.

Previously, our group created an MCMP modeling library-based forensic method for reactor-type discrimination to assign a Pu sample's source reactor type, burnup, and time since irradiation. Groups of intra-elemental isotope ratios of actinides and fission

products, including $^{239-242}$Pu, ^{133,134,137}Cs, ^{153,154}Eu, and ^{149,150,152}Sm, can be characteristic of irradiation history and are resistant to past chemical purifications, and measured values are compared against the modeled library. Purified Pu samples representative of Pu reprocessing were created from irradiated uranium pellets using a benchtop PUREX separation scheme and then were analyzed for their relevant isotope ratios. For stable fission product isotopes, this requires an α-HIB column chromatography separation prior to ICP-MS measurement as a final chemical procedure.

The current work focuses on adapting this process from samples of Pu to samples of its daughter, Am. Two separate sample routes have been considered. Firstly, because trace forensic signatures in Am fractions could be lower in concentration than in Pu fractions, methods for the extraction of the elements listed above have been tested. An extraction based on a modified TODGA ligand extraction scheme from literature is being tested against an LN resin column separation scheme to optimize for extraction efficiency and speed. Secondly, a benchtop Am production scheme, analogous to benchtop PUREX for Pu production, is being developed to produce Am samples of known history. Use of 'known' samples will allow direct comparison to previous Pu work and will provide data on the effects of Am production processes to the forensic signatures present in parent Pu samples. This talk will discuss the most recent results and future plans.

NUCL 4309660

Evaluation of thiol-based solid-phase extraction materials for ^{73}Se/^{73}As isotope harvesting at the Facility for Rare Ion Beams

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Radioactive arsenic (radioarsenic) isotopes have garnered significant interest due to their applications in nuclear medicine, biology, national security, and environmental studies. Various methods exist for producing radioarsenic isotopes, including traditional reactor-based and particle irradiation approaches. Recently, isotope harvesting techniques at the Facility for Rare Ion Beams (FRIB) have been explored for aqueous harvesting of ^{73}Se and its daughter isotope, ^{73}As, which is of particular interest for nuclear data studies. Both traditional production methods and harvesting processes require chemical separations to purify radioarsenic from the parent, target material and/or any co-produced radioisotopes. While several solid-phase methods for arsenic and selenium separations have been developed, many rely on complex oxidation state control or highly acidic conditions to achieve effective separation. This study presents preliminary results on novel solid-phase materials designed to enable uptake at significantly lower acidity and eliminate the need for intricate redox chemistry. Specifically, the performance of three thiol-based resins is evaluated: (1) thiophenol-polystyrene, (2) thiourea-silica, and (3) alkyl thiol-silica. The evaluation focuses on uptake characteristics, including distribution coefficients (D_W) and kinetics, as well as

column separation behavior for ^{75}Se and ^{73}As in nitric and hydrochloric acid solutions. The application of these materials for ^{72}Se/^{72}As medical isotope generators is also explored through ligand stability and repeated elution studies.

NUCL 4309714 - Withdrawn

NUCL 4310902

Evaluation of free radicals in uranium ore concentrates during thermal preparation of yellowcake

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Uranium ore concentrates (UOCs) are important intermediate phases encountered in the processing of uranium ore into nuclear fuel. The U(VI) peroxide studtite ($[(UO_2)(O_2)(H_2O)_2]\bullet 2H_2O$) is a common UOC encountered in the production of relevant uranium oxide materials. Processing begins with heating studtite, to form the dehydrated phase, metastudtite ($[(UO_2)(O_2)(H_2O)_2]$), between 22 – 100°C, which is followed by an orange amorphous phase above 150°C with a stoichiometry often reported as U_2O_7. As the amorphous phase is heated above 400°C, the solid recrystallizes into an α-UO_3 phase, and finally U_3O_8 above 600°C. Recent work has shown the presence of superoxide ($O_2^{\bullet -}$) radicals in studtite and metastudtite and observed increasing $O_2^{\bullet -}$ concentration proportional to the specific activity and age of a sample, indicating the potential for free radical composition to be used for nuclear forensic applications of uranium ores. While characterization of UOCs and their thermal decomposition products has been studied for nuclear forensic application, namely with a focus on morphology, there are no systematic studies that have probed for free radical species within UOCs during their thermal processing. Here we evaluated changes in the radical speciation present in studtite isolated using three different methods and at different points during the thermal decomposition to U_3O_8. Studtite was synthesized using a standard precipitation reaction and isolated using three different methods: (1) centrifugation with drying in a desiccator, (2) vacuum filtration, and (3) vacuum filtration with a water wash. Thermal decomposition was evaluated using thermogravimetric analysis (TGA) and powder X-ray diffraction (PXRD). Electron paramagnetic resonance (EPR) spectroscopy was used to evaluate differences in the composition of radical species in these decomposition products. Overall, we observe differences in free radical signatures for studtite produced using different isolation methods in both low temperature and higher temperature regimes that suggest EPR spectral signatures can provide information about processing of these materials.

NUCL 4311992

Machine learning potential development for advanced oxide fuels

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Molecular dynamics simulations provide valuable insights into the microstructural evolution of nuclear materials under in-reactor conditions. However, the accuracy of these simulations depends on the quality of a potential energy surface (PES) describing the forces acting on atoms in the fuel lattice. While these PESs are typically computed using density functional theory (DFT), strong f-electron correlations in actinide-based materials lead to significant increases in computational complexity. Computational expense also limits the system size that can feasibly be modeled using DFT. This is especially detrimental for modeling doped oxide fuels, whose microstructure can only be precisely captured using larger unit cells. Machine learning potentials (MLPs) present an alternative to expensive and size-constrained DFT calculations, combining the efficiency of empirically-fitted potentials with the accuracy of electronic structure calculations. After training a neural network on a subset of reference data, MLPs achieve DFT-level accuracy at a fraction of the computational cost. We report the use of higher-order equivariant message-passing neural networks to fit a series of MLPs for uranium oxide fuels, including fission-gas bearing systems. A DFT+U training dataset is first generated. We then benchmark our potentials against experimental data, semi-empirical potentials, and recently-reported MLPs. Finding that our MLPs outperform many of these potentials and reproduce experimental data with unprecedented accuracy, we calculate thermodynamic and kinetic parameters necessary to simulate the microstructural evolution of nuclear fuel. Additionally, we implement methods to accelerate MLP development and minimize the quantity of expensive reference calculations necessary for sufficient training. These methods include 1) fine-tuning "foundational" models that are pre-trained for applications across the periodic table and 2) employing optimization procedures such as stochastic weight averaging. This work seeks to extend atomic-scale approaches to grain- and pellet-scale simulations. Ultimately, we aim to enable highly-accurate mechanistic fuel performance models reflective of novel material behavior for next-generation reactors.

NUCL 4312581

Molten salt synthesis of uranium nitride

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Uranium mononitride (UN) is a promising fuel for Generation IV nuclear reactors due to its superior thermal conductivity, low heat capacity, and high fissile density compared to conventional oxide fuels. However, industrial scale synthesis methods such as carbothermic reduction require prolonged synthesis times at high temperatures (>1500

°C) under a continuous flow of nitrogen gas which leads to high energy consumption, carbon impurities, and excess enriched ^{15}N consumption. Carbon impurities increase fuel porosity which can lead to reduced thermal conductivity, worsened fuel swelling, and cladding failure. Alternative synthesis methods have been explored including gas-solid reactions, arc-melting, and direct ammonolysis, resulting in remnant impurities, difficult scalability, and still require long synthesis times at high temperatures.

This study reports a novel molten salt synthesis (MSS) method to produce UN. By addressing the limitations of conventional synthesis techniques, this work seeks to advance the development of high-purity UN fuels for advanced nuclear energy systems. Preliminary results using this method have shown the successful formation of UN from 700 °C to 900 °C in addition to the formation of a secondary phase, $LiUN_2$. The ratio of UN to $LiUN_2$ has shown to increase as the reaction temperature decreases, suggesting UN formation favors lower synthesis temperatures. This method exhibits high N atom efficiency and improved purity than current industrial-scale methods.

NUCL 4312594

Superoxide as a key player in radical generation within alkaline actinide systems

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The study of actinides in alkaline solutions is important for understanding highly alkaline radioactive waste management. In such environments, radioactive decay produces reactive oxygen species like peroxide, superoxide, and hydroxyl radicals. However, actinide behavior under these extreme conditions is not well understood. In this study, we synthesize and characterize a previously reported uranyl(VI) complex along with a novel iso-structural neptunyl(VI) complex: $Li_{18}[AnO_2(OH)_4][AnO_2(O_2)_3]_4{\bullet}x(H_2O)$ (An = U or Np). The structure of the newly identified phase is determined via single-crystal X-ray diffraction, while powder X-ray diffraction confirms the formation of the previously reported uranyl phase. Raman spectroscopy reveals a red shift of 20 cm^{-1} and 16 cm^{-1} in the actinyl stretching modes for $[AnO_2(OH)_4]^{2-}$ and $[AnO_2(O_2)_3]^{4-}$, respectively, in the neptunyl complex compared to the uranyl complex, indicating a weakening of the actinyl bond in neptunyl. Electron Paramagnetic Resonance (EPR) spectroscopy was used to study oxygen-centered radicals. Solid-state EPR detected an axial feature associated with superoxide. In solution-phase experiments, superoxide radicals appeared immediately after adding 5,5-dimethyl-1-pyrroline N-oxide (DMPO), followed by hydroxyl radical signals after 10 minutes. By 30 minutes, the dominant signal corresponded to the DMPO-O-DMPO dimer, indicating persistent hydroxyl radical activity. Density Functional Theory (DFT) calculations suggest that hydroxyl radicals form through superoxide reacting with hydroxide ions.These findings underscore the role of superoxide in radical chemistry within alkaline actinide systems. The study confirms superoxide presence in actinyl peroxides and its contribution to radical generation, offering new insights into actinide behavior in highly alkaline radioactive waste.

NUCL 4315198

Transforming nuclear material laboratory analysis for nuclear forensics to support rapid processing

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To address the need for accelerated nuclear material analysis and staff development for nuclear forensics, Pacific Northwest National Laboratory (PNNL) launched the Nuclear Forensics Transformational Innovation (NFTI) LDRD (Laboratory Directed Research and Development) Initiative, a five-year research effort. NFTI is focused on fundamentally changing the laboratory nuclear material analysis process by exploiting microsampling and microanalysis techniques while autonomously optimizing the new process with data analysis tools that assist forensics experts. The current, very robust pre- and post-detonation nuclear forensics process relies on analyzing bulk samples. NFTI is exploring selecting microsamples within a bulk sample to dramatically reduce unwanted background which has the potential to greatly streamline analytical methods and support process automation. Research is also focused on developing a chemistry model that can learn from early-time data to optimize processing steps and utilizing AI/ML-assisted data analysis to signal when data are ready to be reviewed and reported. An overview of the research, staff development, and partnerships planned for both pre- and post-detonation nuclear material analysis for this new PNNL LDRD initiative will be presented.

NUCL 4316501

Separation of adjacent lanthanides with diglycolamide resin and neutral aqueous complexants

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The United States faces a significant shortage of critical lanthanide isotopes (e.g., $^{149/155}$Eu, ^{153}Gd, ^{161}Tb, ^{161}Dy, ^{176}Yb, ^{177}Lu) and lacks a domestic supply chain to meet the growing demand for these essential metals. Lanthanides exhibit favorable nuclear and chemical characteristics that are crucial for applications in medicine, clean energy technologies, and space exploration. These applications highlight the need for efficient separation techniques to ensure a reliable supply; however, separating adjacent lanthanides is a considerable challenge due to their similar ionic radii and common +3 oxidation state. This work focuses on separating specific lanthanide pairs—Eu–Gd, Tb–

Dy, Yb–Lu, and Gd–Tb—by varying conditions of lanthanide concentration, nitrate and chloride concentrations, and aqueous ligand concentration, showcasing the potential of this method to improve adjacent lanthanide separation. This study investigates a solid–liquid separation technique using diglycolamide (DGA)-based resin as a stationary phase, combined with neutral water-soluble complexants as back-extracting agents to separate adjacent lanthanide pairs. The hydrophilic complexants used in this study include: dioxaoctanediamide (DOODA) and bis-lactam-1,10-phenanthroline (BLPhen); since these ligands demonstrate higher affinity for lighter lanthanides, contrasting with the behavior of DGA's performance. Additionally, water-soluble DGAs 2,2'-oxybis(N,N-bis(2-hydroxyethyl)acetamide) ($_{aq}$DGA-1) and 2,2'-oxybis(1-(pyrrolidin-1-yl)ethan-1-one) ($_{aq}$DGA-2) were tested to explore alternatives for yielding an optimal, efficient lanthanide separation. Four DGA resins were evaluated: commercially available N,N,N',N'-tetraoctyl diglycolamide (TODGA) and three novel resins based on N,N-didodecyl-2-((1-hexyl,2-oxopiperidin-3-yl)oxy)acetamide (SDGA), 2,2'-oxybis(N-octyl-N-(3,5,5-trimethylhexyl)acetamide) (DGA-13), and 2,2'-oxybis(N-(4-butyldecyl)-N-octylacetamide) (DGA-14). The "tug-of-war" between lipophilic and hydrophilic ligands is expected to enhance the separation efficiency, offering an improved approach to near-lanthanide separation. Our results highlight previously unexplored and relatively understudied chemical characteristics that can be leveraged to fine-tune separation parameters for enhanced recovery and purity of precious radiometals.

NUCL 4316536

TODGA-inspired, synthesis of amide functionalized bis-1,2,4-triazinyl-2,6-pyridines as complexants for minor actinide extraction from lanthanides

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The selective separation of trivalent actinides from complex spent nuclear fuel metrices remains a significant challenge due to the chemical similarities between actinides and lanthanides. Since the introduction of bis-1,2,4-triazinyl-2,6-pyridine (BTP) complexants by Kolarik in 1999, the development of an optimized, scalable complexant for actinide-lanthanide separation has been an ongoing pursuit. Here, a streamlined synthetic approach is presented for amide-functionalized BTP complexant inspired by TODGA, utilizing an alkylation-condensation methodology to enhance solubility and extraction efficiency. The incorporation of amide groups along the aromatic backbone significantly improves solubility in organic solvents such as Exxal-8, 1-octanol, and toluene, facilitating their practical application in liquid-liquid extraction. This TODGA-inspired, amide-functionalized BTP complexant exhibits remarkable selectivity for ^{241}Am^{3+} over $^{252\text{-}254}$Eu^{3+} under simulated spent nuclear fuel conditions, achieving high distribution ratios and separation factors. Furthermore, decomplexation studies confirm the efficient recovery of extracted actinides, demonstrating the recyclability of these complexants in biphasic separation systems. This presentation will highlight key aspects of complexant design, synthetic preparation, equilibrium study, acid and complexant concentrations

study, as well as decomplexation, underscoring the potential of this novel complexant for advanced nuclear waste management strategies.

NUCL 4317761

Umpolung C-H (Radio)fluorination of hydroxamide derivatives via strained Aziridinone intermediates enabled by N-O scission

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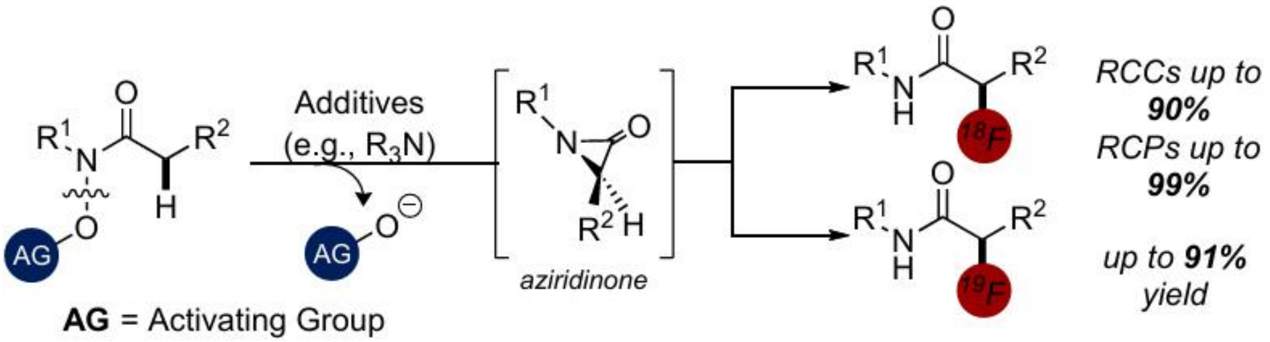
***Objectives:** The Scott lab recently described a zinc-mediated labeling reaction that furnishes metabolically robust tertiary α-radiofluoroamides in excellent radiochemical conversions (RCC), yields (RCY), and purities (RCP).[1] However, the α-haloamide precursors employed in this reaction have limited synthetic accessibility and are inefficiently labeled using primary and secondary halides. To address these limitations, our group aims to radiolabel amides using more readily accessible N-sulfonyloxyamide precursors via umpolung C-H activation and potentially extend the scope to include other nucleophiles.*
***Method:** N-sulfonyloxyamides precursors are synthesized via amide coupling between corresponding carboxylic acid and hydroxylamine and then sulfonylation with mesyl chloride. Screening reactions were undertaken with 100 µL cyclotron produced radiofluoride, 10 µmol precursor and various bases in MeCN. RCC, identity and RCP analyses were conducted with radio-thin-layer-chromatography and radio-high-performance liquid chromatography.*
***Results:** N-sulfonyloxyamides were able to undergo radiofluorination under newly optimized radiolabeling conditions in good RCCs (up to 90%) and excellent radiochemical purities (>99%). Optimizations have also extended this reaction to regular ^{19}F, unlocking synthetically challenging amide reference standards that otherwise require harsh conditions for direct preparations.[2] Mechanistically, reaction outcomes using electronically modified hydroxamide precursors, in conjunction with previous reports, point to a ^{19}F-fluorination mechanism that proceeds via strained α-lactam, which may undergo heterolysis to the corresponding aza-oxyallyl cation before nucleophilic trapping.[3]*
***Conclusion:** Augmented (radio)fluorination systems are described for preparing α-fluorinated amides via umpolung C-H activation. Key reaction optimizations have enabled the preparation of α-fluoroamides with both ^{18}F and ^{19}F fluoride sources.*



- **Approach***: Scission of N-O Bonds for Concerted C-H Activation/Cyclization*
- **Result:** *Efficient Incorporation of Nucleophiles, Including Fluorine-18*
- **Application:** *Radiolabeling of (Pre)Clinical Agents for PET Imaging*

NUCL 4318179

EPR and computational study on the radiation-induced transformations in the nuclear fuel cycle

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Extraction separation plays a crucial role in the nuclear fuel cycle by utilizing specific organic ligands designed to selectively recover fuel components and radioactive fission products from waste solutions. Additionally, supplementary compounds are employed to regulate process conditions, including pH and prevent third-phase formation. Among the chemical classes commonly utilized are phosphoric acid esters (e.g., tributyl phosphate [TBP], dioctyl phosphate [DOP]), phosphine oxides (e.g., octyl(phenyl)-N,N-diisobutylcarbamoylmethylphosphine oxide [CMPO]), phosphonates (e.g., dibutyl((diethylamino)carbonyl)phosphonate [DBDECP]), diglycolamides (e.g., N,N,N',N'-tetraoctyl diglycolamide [TODGA]), N,N-dialkyl amides (e.g., N,N-bis(2-ethylhexyl)butanamide [DEHBA], di-2-ethylhexylisobutyramide [DEHiBA]), carboxylic acids (e.g., oxalic acid [OA], lactic acid [LA], citric acid [CA]), and acetohydroxamic acid (AHA).

Considering these ligands are exposed to ionizing radiation, their radiolytic stability becomes a critical factor affecting extraction efficiency and solvent longevity. In this study, we present a comprehensive investigation of radiation-induced transformations of these ligands in both organic and aqueous phases using electron paramagnetic resonance (EPR) spectroscopy and quantum chemical calculations. The radiation-chemical yield of paramagnetic species in neat TBP exposed to X-rays was *quantitatively assessed using the spin trapping method, although structural insights provided by this method remain limited. To clarify radical structures formed upon irradiation, we conducted EPR studies at 77 K on electron-beam irradiated ligands. In all cases, carbon-centered radicals were identified. Subsequent thermal annealing induced conformational relaxations without further chemical transformations.*

In the aqueous phase, the radiolytic fate of ligands is influenced by reactions with water radiolysis products, particularly hydroxyl radicals. Using continuous-flow EPR spectroscopy, we investigated these reactions with selected organic acids, enabling the determination of radical structures as well as their bimolecular decay kinetics.

In conclusion, the combined EPR and computational approaches provide crucial mechanistic insights into radiation-induced ligand degradation, offering a robust framework to optimize ligand selection and improve the durability and performance of extraction systems in nuclear waste reprocessing

NUCL 4318979

Radiation Induced surface modification of Gibbsite in the alkaline environment

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Aluminum (oxy)hydroxide minerals, boehmite (AlOOH) and gibbsite (Al(OH)3), formed through the corrosion of aluminum from spent nuclear fuel cladding, present a significant challenge in managing highly radioactive waste stored in underground tanks at the Hanford site, WA. Understanding the chemical and physical transformations of gibbsite under extreme tank waste conditions is crucial for effective waste management. Exposure to radiation, primarily beta particles and gamma rays from strontium-90 and cesium-137, along with the presence of salts and ions, creates a complex, multicomponent chemical system within the highly alkaline tank environment.

Previous studies investigated the radiolytic species generated from irradiating gibbsite synthesized using different precursors and examined the effects of Cr and Fe doping to assess interactions with salts and ions present in the tank. This study focuses on radiation-induced surface modifications of gibbsite in a highly alkaline NaOH environment. Gibbsite nanoparticles were hydrothermally synthesized using aluminum chloride (AlCl3) as a precursor, forming nanoplatelets with an average size of ~280 nm. These particles were dispersed in a 2M NaOH solution at a concentration of 50 mg/mL and subsequently irradiated.

Unirradiated and irradiated samples were characterized using DRIFTS, XPS, Raman spectroscopy, solid-state UV-Vis, zeta potential, and EPR measurements. DRIFTS revealed additional peaks corresponding to structural hydroxyl groups, indicating radiation-induced surface modifications. Raman spectroscopy showed a broad peak in the irradiated samples, further confirming additional functionalization on the gibbsite surface.

This study demonstrates that gibbsite undergoes surface modification under radiation in

the presence of NaOH, leading to hydroxyl group functionalization. The interaction of NaOH with gibbsite is driven by the formation of radicals and active sites induced by gamma radiation, as previously reported. These findings provide insights into the chemical transformations of gibbsite under extreme tank waste conditions, contributing to a deeper understanding of radioactive waste behavior and potential mitigation strategies.

NUCL 4320526

Effect of fission products on the thermophysical properties of actinide containing chloride fuel salts

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Chloride salts are potential fuel solvents and coolants of Molten salt reactors (MSR's), a proposed generation IV advanced nuclear reactor class. These reactors, denoted Molten Chloride Fast Reactors (MCFR's), contain a primary loop of fissile material containing fuel salt which is pumped through the reactor core. Chloride salts, alternatively from commonly studied fluoride molten salts, are desired due to their lower supply costs and high actinide solubility. However, as fission results in elements across the periodic table, normal reactor conditions can cause alterations in the fuel salt redox potential, composition, and thermophysical properties. By measuring the thermophysical properties of a candidate chloride fuel salt such as $NaCl-UCl_3$, before and after the addition of fission products added in a "kitchen sink" method, the propagated uncertainty in properties caused by fission can be quantified. This work will show the thermophysical properties of eutectic $NaCl-UCl_3$ including melting point, thermal expansivity, density, and viscosity before and after the addition of fission product salts and compare the values to the relative tolerances and reactor sensitivities for reactor operation and safety analysis.

NUCL 4321243

Radiosynthesis and preclinical evaluation of [^{11}C]fentanyl in rodents using positron emission tomography

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Background: *In 2022, the United States reported 107,941 drug overdose deaths, corresponding to an age-adjusted mortality rate of 32.6 per 100,000 population. While fentanyl remains a potent analgesic in medical practice, it is also a significant factor in cases of opioid use and overdoses nationwide. Despite the widespread use and misuse of fentanyl, there is no in vivo imaging probe for pharmacokinetics research. Herein, we*

synthesized [^{11}C]fentanyl and report its preliminary brain study in rodents using positron emission tomography (PET).

Methods: *[^{11}C]fentanyl was synthesized in two steps: 1) preparation of [^{11}C]propionyl chloride, 2) amidation of 4-anilino-N-phenethylpiperidine (4-ANPP). [^{11}C]propionyl chloride was prepared using an "in-loop" method for [^{11}C]carboxylation, where [^{11}C]CO_2 in a stream of helium was reacted with ethylmagnesium bromide. Phthaloyl chloride was then used as a chlorinating agent to produce [^{11}C]propionyl chloride, which was subsequently purified by distillation. Various tertiary amines and solvents were tested for rapid amidation in the radiosynthesis of [^{11}C]fentanyl.*

Results: *The total synthesis time from the end of bombardment (EOB) was approximately 42 minutes, achieving high radiochemical purity (>99%), decay-corrected radiochemical yield of 10.4 ± 5.7% (n = 5), and high specific activity (44.7 ± 22.1 Ci/μmol, n = 5) at EOB. Diisopropylethylamine in chloroform provided the highest radiochemical yield for the [^{11}C]amidation step (62.5 ± 11.2%, n = 3). Baseline PET imaging revealed that IV [^{11}C]fentanyl exhibited rapid brain uptake followed by fast clearance, with a maximum standardized uptake value (SUV_{max}) of 2.71, a time to reach maximum uptake (T_{max}) of 1.7 minutes, and a half-life ($T_{1/2}$) of 4.7 minutes (n = 3).*

Conclusion: *2-step radiosynthesis was optimized to provide a [^{11}C]fentanyl production method showing high specific activity and radiochemical purity. PET study for fentanyl's pharmacokinetics is under way in rodents and non-human primates.*

NUCL 4321991

Search for nuclear stopping using bremsstrahlung photons emitted from ultra-central Pb+Pb collision at the Large Hadron Collider (CERN)

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Abstract

***Background**: Ultra-relativistic heavy ion collisions at the Large Hadron Collider (LHC) recreate conditions akin to the early universe. In these collisions, bremsstrahlung photons are emitted due to the extreme deceleration (nuclear stopping) of near-light-speed charged particles. Studying these photons is crucial for understanding energy loss mechanisms, quark-gluon plasma formation, and the thermodynamics of nuclear matter—key to both high-energy physics and nuclear chemistry.*

***Purpose**: This work estimates bremsstrahlung photon energy from lead-lead collisions at 5.36 TeV using 2024 LHC data captured by the Compact Muon Solenoid (CMS) Zero Degree Calorimeter (ZDC). While ZDC detection of such photons was previously unverified, this study confirms their observation, offering new insights into nuclear stopping and gluon saturation.*

***Methods**: Using 2024 CMS heavy-ion data, we simulated bremsstrahlung radiation via Monte Carlo (inverse sampling) and analyzed ZDC photon capture efficiency by correlating shower polar and azimuthal angles. Ultra-central collisions were selected to maximize photon yield.*

***Results & Conclusion**: Experimental and simulated data show strong agreement,*

*confirming detector reliability. Despite the ZDC's 140m distance from the collision point, it captures **43 ± 3%** of photons, with an average bremsstrahlung energy of **2 TeV**. This marks the first observation of bremsstrahlung photons in the ZDC, a groundbreaking finding. The study enhances nuclear chemistry by modeling energy dissipation and isotope production in extreme conditions. Research is ongoing to further explore these dynamics.*

NUCL 4325386

Evaluation of total body distribution of [18F]Olaparib and [11C]Niraparib using the penn PET Explorer: Total-body PET of two clinical PARP inhibitors

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Poly(adenosine diphosphate ribose) polymerase-1 (PARP-1) is a protein implicated in DNA damage repair and is often overexpressed in ovarian/breast cancer. Several PARP-inhibitors (PARPi) have been deployed as chemotherapeutics such as Niraparib (Zejula, GSK) and Olaparib (Lynparza, AstraZeneca & Merck). Radiolabeling of PARPi offers a means to evaluate their target engagement in both the central and peripheral nervous systems. This can expedite the development of PARPi for treating cancers beyond ovarian/breast. [^{18}F]Olaparib and [^{11}C]Niraparib were radiolabeled and their biodistribution were evaluated using total-body (TB) PET on the PENN PET Explorer. This study was conducted according to GSK and University of Pennsylvania's policies on the Care, Welfare and Treatment of Animals and reviewed by the Institutional Animal Care and Use Committee at UPENN. Total-body distribution of [^{18}F]Olaparib (n=4) and [^{11}C]Niraparib (n=5) were evaluated using a 90-minute PET scan in a healthy rhesus macaque model. Time-activity curves were generated for the central and peripheral nervous system. Venous blood was sampled prior to radiotracer injection and at 5-60 minutes post-injection. HPLC analysis was used to determine the parent fraction of each radiotracer. Linear fixed-effects model of response (AUC) of radiotracers, phase of crossover design, their two-way interaction along with baseline bodyweight, and dose per tissue were used for statistical analysis. [^{18}F]Olaparib was isolated in > 95% RCP and with a molar activity of 8.2 ± 5.3 Ci/µmol at EOS with a radiochemical yield of 10%. [^{11}C]Niraparib was isolated in > 95% RCP with a molar activity was greater than 3.0 Ci/µmol at end of EOB with an average radiochemical yield of 5%. Both radiotracers demonstrated >90% parent fraction at 5 minutes post-injection. [^{18}F]Olaparib maintains a higher parent fraction than [^{11}C]Niraparib after 60 minutes. Both radiotracers are metabolized via the hepatobiliary system. [^{11}C]Niraparib demonstrates significantly

higher brain uptake and brain retention than [^{18}F]Olaparib. [^{18}F]Olaparib showed higher uptake in kidney (P-value=0.0591) and spleen (P-value=0.0115). This demonstrates that these tracers may be useful in guiding therapeutic development of CNS-specific PARPi. Total-body PET can distinguish between the differing routes of metabolism between the two PARP-inhibitors. Furthermore, these two PET tracers can be used to quantify target engagement of novel PARPi in vivo.

NUCL 4325719

Photomediated Radiohalogenation of aryl thianthrenium salts using

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Intro

Radiohalides feature in many diagnostic and therapeutic small organic imaging agents used for precision medicine-guided treatments in nuclear medicine. Berger et al. recently demonstrated the feasibility of photo mediated halogenations of complex arenes using metal mediators with thianthrenium leaving groups. This sparked our interest in translating this method for radiohalogenation using ^{125}I, ^{77}Br and ^{211}At for applications in oncology radiotherapeutics. In this presentation, we disclose a highly efficient method for radiohalogenation of electronically diverse thianthrenium salts using ^{77}Br, ^{125}I, and ^{211}At in excellent radiochemical yields.

Method

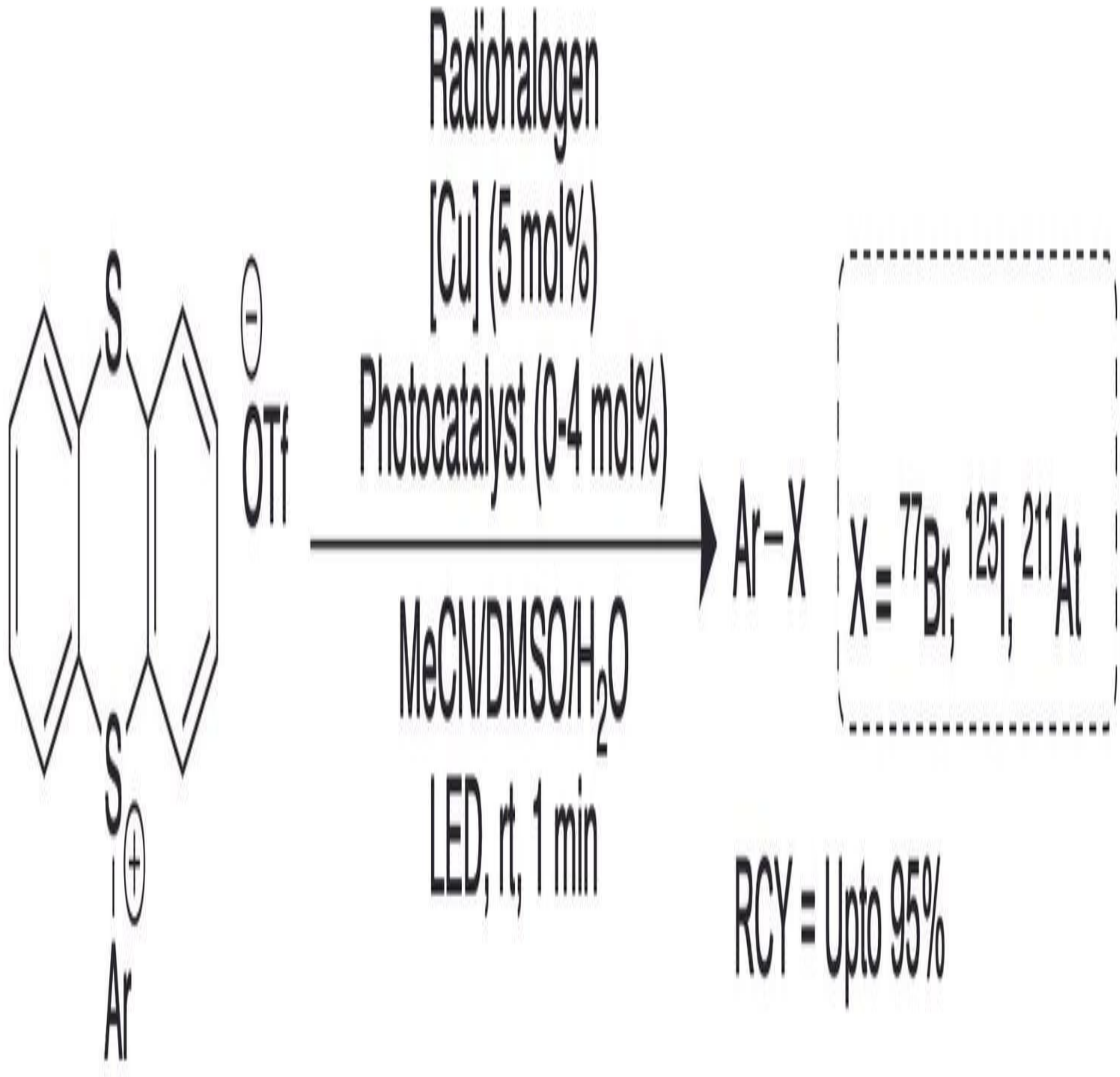
^{211}At was produced onsite using a JSW BC3015 cyclotron via the α-particle bombardment of ^{209}Bi. ^{125}I was purchased from Perkin Elmer/Revvity, and ^{77}Br was obtained from the University of Wisconsin. In a typical radiolabeling experiment, radiohalide, transition-metal mediator (Cu(I)), and photocatalyst (Ru or Ir) were added to the thianthrenium precursor and irradiated with LED photoreactor at RT.

Results

Systematic optimizations conducted to explore several reaction factors: the wavelength of visible LED light, the impact of inert Ar atmosphere, the influence of photo catalysts and compatible reaction time with half-life of radionuclides. The radionuclides ^{77}Br, ^{125}I and ^{211}At, were successfully incorporated via photomediated ipso-substitution of a thianthrenium salts in excellent yields with both ruthenium and iridium photocatalysts. In case of ^{211}At, removing the Ir/Ru photocatalyst boosted the conversion of the radioastatination, illustrating the difference in chemical properties and reaction pathways between these radiohalides. The reaction was well tolerant of air and moisture. Overall, the final optimized reaction conditions lend themselves to the expedient incorporation of radionuclides into small organic molecules, improving access to radiomedicines used to treat various cancers.

Conclusion

In conclusion, we describe new methodologies for the photomediated radiohalogenation of aryl thianthrenium salts using ^{77}Br, ^{125}I, and ^{211}At. These reactions proceed rapidly under mild conditions and efficiently furnish labeled bioactive molecules used in oncological radiotherapy.



NUCL 4325867

Environmental aging of ceria nanoparticles with different morphologies

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The characterization of nuclear materials under controlled conditions promotes a better understanding of their chemistry, insight into their stability, and the use of morphological signatures during a nuclear forensics investigation. While previous work demonstrates that the morphology of bulk PuO_2 is affected by environmental aging (e.g., relative humidity, temperature, type of containment), it is unclear how the initial morphology of the material might affect long-term observations. In this work, ceria (CeO_2) nanoparticles were synthesized via a hydrothermal process to produce rod, cube, and polyhedral morphologies. Differing morphologies were produced via two approaches in which the molar ratio of hydroxide to cerium was held constant at 100:1. In the first approach, the molar ratios of urea to cerium (4:1, 6:1, 10:1) were varied. The second approach introduced a surfactant, cetyltrimethylammonium bromide (CTAB), while holding the ratio of urea to cerium constant at 4:1. Vibrational spectroscopy, X-ray diffraction, and electron microscopy were used to characterize these initial materials before aging as a function of time, temperature (25 °C versus 45 °C) and relative humidity (ambient, 40 %, and 80 %) to determine whether any forensic signatures exist. Future work will focus on expanding these synthesis and environmental aging approaches to plutonium dioxide nanoparticles. Elucidation of these forensic signatures will improve the current database by considering the material scale and morphologically-induced changes.

NUCL 4325972

Are structural hallmarks reliable indicators of the nature of soft-donor interactions with uranium?

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Quantum chemical methods can be used to understand how soft-donor ligands engage in bonding interactions that lead to structural changes in uranium complexes. Our group has leveraged density functional theory (DFT) and multireference methods (CASPT2) to explore interactions with soft-donor ligands across diverse ligand frameworks. We studied the dramatic change in molecular geometry observed for a uranium(V) oxo complex. When oxygen-based ligand are used, the expected axial oxo structure is obtained; however, when the ligand is modified to include thiophenolate groups, the oxo appears in the equatorial position. By invoking the concept of inverse trans influence (ITI), we can understand the different behavior with soft-donor sulfur groups compared to the analogous oxo-donating ligands. The classic structural hallmark was not observed for an ITI despite the change in oxo orientation. However, DFT showed that steric effects limit the ability of the U-S distance to shorten and that this structural change is obtained on a model system with less steric hindrance. Moreover, the ligand selected also includes a mesitylene group. Prior DFT calculations assigned a δ-bonding interaction in the complex with oxygen-based ligands supporting the measured EPR parameters. Revisiting this complex with higher-level methods showed an orbital interaction between the arene π orbitals and the ligand, but it is a σ-type interaction. Likewise, in the complex with thiophenolate ligands, no δ-bonding interaction is

obtained. EPR parameters computed with second-order multireference methods (SO-XMS-CASPT2) are in good agreement with the experiment, emphasizing once more the importance of making such comparisons in assigning ground state electronic structure. Similar challenges with using structural hallmarks for actinide-arene bonds for terminal arenes have also been shown by us and will be included for comparison.

NUCL 4326119

Spectroscopic analysis and leaching behavior of alternate technetium species from legacy nuclear wastes

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Technetium-99 (Tc) is a long-lived, radionuclide of interest in legacy nuclear wastes that is highly mobile in the environment when in the +7 oxidation state as the pertechnetate ion, $[TcO_4]$. As such, most of the research on Tc in nuclear waste treatment has been on $[TcO_4]$. However, there are non-pertechnetate species that have been identified within nuclear wastes (Tc with oxidations states < +7) and these species are understudied with respect to their behavior in nuclear waste treatment processes (e.g. pre-treatments, immobilization etc.). Herein, we report the first example of characterizing and tracking the chemical behavior of non-pertechnetate before, during, and after immobilization in simulated grouted/cementitious waste forms. Tests were performed using actual nuclear waste samples and a synthetic Tc(I) complex spiked into simulated waste. The solidified grout/concrete samples were subjected to leach tests and subsequently characterized by NMR, EPR and X-ray absorption spectroscopies, including a novel L-edge XANES measurement for Tc-99.

NUCL 4326744

Using biphasic isothermal titration calorimetry to develop new polymeric extractants

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f-Block elements are critical to the technological and energy infrastructure that underpin our modern and future world; however, their production and purification rely on economically and environmentally costly solvent extraction processes. Polymeric extractants are an emerging class of materials that can improve or supplant these methods. We use isothermal titration calorimetry (ITC), a technique widely used in the study of bio(macro)molecule binding, to directly determine the impact of polymer structure on the underlying binding thermodynamics and to develop rational design principles based on structure-property relationships. Motivated by the need for new, *organic soluble polymer extractants, recent work has focused on developing a new method for biphasic ITC that has the potential to determine the entire thermodynamic profile of an extraction in a singular experiment. Using the well-studied model system of* Eu^{3+} *extraction by di-(2-ethylhexyl) phosphoric acid (DEHPA), we have been able to reliably determine the* ΔG_{extr} *in a single experiment rather than a more laborious determination of the distribution coefficient. Using a modular polymer approach to eliminate batch to batch variations, we synthesize polymeric analogues of DEHPA that allow us to modify the hydrophobicity and number of exchangeable protons and study their binding using ITC. In total, this work directly compares key thermodynamic parameters between small–molecule and polymeric extractants and will enable the rational development of new polymer extractants.*

NUCL 4327045

Microfluidic separation platform for post-detonation nuclear forensic applications

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A microfluidic chemistry platform is under development to support post-detonation nuclear forensics. The microfluidic platform is designed to be compact, use a minimum amount of reagents, and utilize deployable technologies to support in-field use. The platform has been shown to separate U and Pu from realistic matrices using robust chemistry. After chemical separation, integrated UV-VIS, gamma spectrometry, and alpha spectrometry systems are used to perform online measurements of specific U and Pu isotope ratios. This platform is also being tested as a rapid separation chemistry front-end for ICP-MS analysis of U and Pu isotope ratios from post-detonation samples.

NUCL 4327220

Effects of second sphere interactions on f-element structure and behavior

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Counterions are ubiquitous in inorganic chemistry, often serving as charge balancing species. However recent work has shown that these second sphere interactions can affect not only solubility, but modulate catalytic activity, shift redox behavior, and impact magnetic performance. For the actinides, in particular, understanding the effects of counterions is critical in separations, nuclear waste management, and environmental transport, where control over actinide coordination chemistry is essential for predicting and manipulating chemical behavior. Motivated by this recognition, our group has looked to understand the impact of second sphere counter-cations, including alkali metal ions and N-H heterocycles, on the structural chemistry, spectroscopic properties,

and overall chemical behavior of the early actinides, Th-Pu. Presented will be an overview of the effects of counterions on actinide structural chemistry and reactivity in various ligand systems.

NUCL 4327461 - Withdrawn

NUCL 4327573

Federated AI architectures for uncertainty-quantified neutron activation analysis: enhancing nuclear forensic capabilities through multi-reactor learning

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Modern nuclear forensics has established techniques like neutron activation analysis (NAA) as a gold standard for non-destructive material characterization. However, variations in reactor conditions, data availability constraints, and the need for enhanced analytical accuracy create challenges for nuclear material attribution. This work proposes a federated AI framework to improve NAA's reliability across different reactor environments while maintaining data security and sovereignty.

A Bayesian U-Net model is introduced to enable reactor-agnostic spectral analysis; designed to process gamma spectra from multiple reactor types while accounting for neutron flux variations. Additionally, this approach explores integrating prompt gamma activation analysis (PGAA) and instrumental NAA (INAA) through physics-informed spectral fusion, with the potential to reduce actinide detection false positives. To address data limitations, conditional generative adversarial networks (GANs) trained on simulated neutron flux profiles are proposed for generating synthetic spectra to enhance training datasets while preserving NAA's intrinsic accuracy.

Also, by integrating AI-driven spectral analysis with established NAA methodologies, this framework advances nuclear forensic capabilities, ensuring compliance with INFCIRC/908 verification protocols for uranium particulate analysis. The interdisciplinary approach also shows the possible potential of AI in enhancing the precision and reliability of nuclear forensic investigations, bridging the gap between traditional methodologies and modern computational advancements.

It is important to note that this study presents a methodological framework and proof-of-concept for applying federated AI to nuclear forensics. The proposed techniques align with international nuclear security objectives and could significantly enhance material characterization efficiency. Future work will focus on validation through experimental implementation and benchmarking against existing forensic methodologies.

NUCL 4327724

Decting and characterizaing charged particles using CR39 during electrochemical co-deposition processes

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Low-energy nuclear reactions (LENR) have long been a subject of scientific interest due to their potential implications for clean energy generation and nuclear physics. Among various experimental approaches, the electrochemical co-deposition (co-dep) method pioneered by SPAWAR Systems Center San Diego has shown promising evidence of LENR effects, including the emission of charged particles and energetic neutrons. We recently conducted electrochemical co-deposition experiments inspired by the SPAWAR studies, in which Pd is co-deposited onto an Au substrate in heavy water. This process results in a highly fractal Pd deposit with vacancies, facilitating rapid deuterium loading. Both high D/Pd loading and vacancy formation are considered essential for initiating LENR. A notable observation in our experiments is the emission of MeV charged particles, a phenomenon indicative of LENR that remains unexplained by conventional theories. Given the low production rate of charged particles, CR-39 track detectors are particularly well-suited for long-duration measurements in these low-count-rate conditions. Our results display track patterns on CR-39 similar to those previously reported in SPAWAR studies. Additionally, we observed triple tracks on the CR-39 detector, providing evidence of energetic neutron emission. These tracks are consistent with C-12 disintegration caused by neutrons with energies exceeding ~9 MeV. Reproducibility has been a persistent challenge in LENR research, with the need for robust and accessible "lab rat" experiments. Our findings demonstrate that the SPAWAR co-deposition experiment offers a reproducible platform for investigating LENR phenomena, contributing to the ongoing exploration of anomalous nuclear effects. Further investigation of the properties of Pd co-deposits and the establishment of clearer correlations between their structural characteristics and observed LENR signatures will advance the fundamental understanding of LENR and create opportunities for potential applications.

NUCL 4328173

Spectroscopic signature development for An(IV) oxalates

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The field of nuclear forensics aims to use material signatures to identify the age and provenance of unknown nuclear materials. Plutonium oxalates are prevalent species in the nuclear fuel cycle and common starting materials for weapons manufacturing, though these materials themselves are understudied. Pu(III) and (IV) oxalate species are known to possess unexplained reactivity and changes in color depending on age

and production method, despite crystallographic data indicating consistent structures. Recent studies have shown the ability of uranium to stabilize hydroxyl and superoxide radicals in solid-state complexes and clusters, which highlight the possibility for radiolysis-induced free radical formation and stabilization of free radicals in plutonium materials as well. We are broadly interested in free-radical signature identification and development, and here we will describe initial efforts focused on oxalate systems. In order to safely and efficiently work with plutonium in an academic laboratory, method development and optimization with a surrogate cation is required, inspiring our initial work with uranium(IV) oxalates. By synthesizing and characterizing uranium(IV) oxalate species with known Pu(IV) analogues, we can identify potential material signatures related to sample age, activity, preparatory methods, and storage conditions. This work describes the synthesis and characterization of U(IV) oxalate species using vibrational spectroscopies and electron paramagnetic resonance, the role of isotopic compositions in driving spectroscopic signatures, and will highlight preliminary results from our studies focused on identifying free-radical based material signatures in plutonium systems as well.

NUCL 4342879

Influence of strike order on the morphological features of U_3O_8

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Nuclear forensics remains critical in addressing threats associated with illicit and malicious use of radioactive materials, necessitating continuous advancements in the characterization of uranium ore concentrates (UOCs). In this study, we investigate the influence of precipitation strike order, direct and reverse strike, on the morphological features of U_3O_8 derived from four synthetic routes. [Ammonium Uranyl Carbonate (AUC), Ammonium Diuranate (ADU), Uranyl Peroxide (UO_4), Magnesium Diurante (MDU)] Strike order conditions influence local supersaturation, nucleation, and growth, thereby affecting the crystallization kinetics and final morphology as captured by powder X-ray diffraction (p-XRD) and scanning electron microscopy (SEM).

Morphological signatures of SEM images were analyzed using a convolutional neural network (CNN) architecture based on a ResNet34 backbone for transfer learning, with images processed into “spot” locations that combine multiple magnifications and detector modes to enhance feature extraction. Machine learning classification was performed to differentiate between synthesis routes, capturing subtle variations in product morphology. While most synthetic pathways displayed marked differences in phase distribution and crystallinity between strike orders, the magnesium diuranate (MDU) route exhibited only minor morphological differences as determined by CNN analysis.

These findings indicate that the final morphology between direct and reverse strike UOCs are synthetic route dependent that produce distinct signatures under both *addition orders, suggesting a robustness to strike order variations that may have implications for forensic attribution and process monitoring.*

NUCL 4343070

Separations for nuclear forensics analysis

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Nuclear forensic analysis requires precise elemental separations to quantify sample composition accurately. Traditional separation techniques, such as liquid-liquid extraction and chromatography, were primarily developed for the civilian nuclear fuel cycle. However, transition metals present in post-detonation debris can interfere with the separation and subsequent analyses of the materials. This study aims to enhance rapid separation processes by investigating the extraction behavior of activation products using commercially available extractants. We focused on the extraction of Fe(III) by tri-n-butyl phosphate (TBP) in n-dodecane and Co(II) by tetraoctyl diglycolamide (TODGA) in toluene, from nitrate and chloride media. This highly collaborative work combines solvent extraction data, neutron activation analyses, potentiometric titrations, EXAFS and XANES measurements, and computational science (molecular modeling and artificial intelligence) to understand of the separation systems at a molecular level.

NUCL 4344166

PCET chemistry of pentavalent Np and Pu Imidophopshorane complexes

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A series of isostructural mid-actinide (M = U, Np, Pu) complexes which feature divergent proton-coupled electron transfer (PCET) reactivities at the $M^{5+/4+}$ couple are presented. These studies include the isolation of a pentavalent Np species $[Np^{5+}(NP^{J})_4][B(ArF_5)_4]$, which features the Np^{5+} center in an unprecedented low-coordinate (tetrahedral) environment without support from MLMB interactions such as those found in oxo and imido species. The study of this complex and its electronic structure has allowed us to explore fundamental reaction chemistry across U–Pu. The PCET reactivity of the

isolated Np^{5+} complex is probed on the NMR timescale, while the Pu^{5+} species can be accessed electrochemically and undergoes a PCET reaction at a rate orders of magnitude greater than that of Np^{5+}. These are the first quantitative measurements of the thermodynamic and kinetic parameters of a PCET reaction in molecular transuranic chemistry. Differences in the redox chemistry, reactivity, and electronic structure of these species inform new routes to stabilizing high-valent f-element complexes.

NUCL 4344174

Modeling heterogeneity and bonding behavior in actinide chemistry using XAFS-based approaches

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Characterization using X-rays is of particular importance towards interrogating the structural properties of actinide systems due to 1) high penetration, which enables encapsulation of radioactive samples inside of containment layers to decrease the likelihood of instrument contamination, 2) the ability to measure small quantities of material non-destructively and 3) the broad array of samples that can make use of X-ray techniques to elucidate materials structural properties. In the Moreau group, we are interested not only in utilizing X-ray characterization tools such as X-ray spectroscopy and X-ray scattering to probe actinide-based systems, but also in developing new approaches towards utilizing these techniques and conducting data analysis in previously unexplored ways. In this talk, two different projects will be discussed, which involve new approaches to the application of X-ray spectroscopy and X-ray scattering. The first project involves modeling heterogeneity in UO_2 nanoparticles using EXAFS. Radially dependent models were developed, with varying distributions of changes in disorder and bond contraction. These models were substituted into the EXAFS equation and the best-fit models between particles of different size were compared, to extract particle size-dependent trends in structural properties. Our results accurately predict the nanoparticle size that is required to induce the antiferromagnetic behavior observed in bulk. The second project makes use of anomalous X-ray scattering, along with XAFS, to precisely determine uranyl binding sites from uranyl uptake within metal organic frameworks. Through anomalous scattering, we are able to determine which crystallographic planes are affected by the presence of uranyl, and from XAFS, we are able to determine the local structure of the uranium and compare our results to potential binding sites within the MOF. Each of these studies provide X-ray analysis approaches that can be broadly applicable to nanoparticle and MOF-guest chemistry.

NUCL 4344799

Solid ctate ^{59}Co NMR study of a high-valent Np complex: $[Co(NH_3)_6]_3[NpO_4(OH)_2]_3 \bullet 4H_2O$ and $[Co(NH_3)_6][NpO_4(OH)_2] \bullet 2H_2O$

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The Np(VII) compound with $[Co(NH_3)_6]^{3+}$ *cations was synthesized and structurally examined using powder X-ray diffraction. Multiple phases were observed, consisting of octahedral* $[Co(NH_3)_6]^{3+}$ *cations, discrete tetragonal bipyramidal* $[NpO_4(OH)_2]^{3-}$ *anions, and waters of hydration. Electric field gradient tensors and asymmetry parameters at the Co sites were determined by solid-state* ^{59}Co *nuclear magnetic resonance (NMR) spectroscopy at 7.04 and 11.74 T and compared with values obtained by theoretical calculations. The interpretation and dependence of these parameters, along with* ^{59}Co *chemical shifts and spin-lattice relaxation times* (T_1)*, on the electronic structure of the complex will be discussed. Aging effects were monitored by measuring the* ^{59}Co *spectra and* T_1 *over time. Specifically, aging has a prominent impact on the spectral shape and intensity.*

NUCL 4344804

Luminescence and circularly polarized luminescence of molecular complexes of actinides (Am to Es)

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Circularly polarized luminescence (CPL) is the preferential emission of right- or left-handed circularly polarized light. In the lanthanide series, because of the core-like nature of 4f orbitals, the split between energy levels of 4f-elements is larger through spin-orbit coupling than through crystal field splitting. In 4f-elements, each luminescent transition between spin-orbit coupling term levels is associated with a specific transition type, resulting in different relative CPL strengths. More importantly, CPL spectroscopy allows for better distinction of the individual components resulting from crystal field splitting, though these transitions' selection rules are not well understood.

We are interested in investigating if the principles of CPL observed in 4f-elements are also applicable in 5f-elements and/or how they differ. We will show that by employing appropriate ligands, luminescent molecular complexes of americium, curium, berkelium, californium, and einsteinium can be obtained. Additionally, by utilizing chiral ligands, CPL spectra of transplutonium elements (Am and Cm) can be observed, and quantitative information about their crystal field splitting can be obtained.

NUCL 4344805

Probing the magnetic properties of actinide complexes by paramagnetic NMR

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Paramagnetic nuclear magnetic resonance (pNMR) is a powerful tool for probing electronic structure and magnetic properties in paramagnetic systems. While pNMR has been extensively applied to transition metal and lanthanide complexes, its use in actinide chemistry remains relatively underexplored due to the radioactivity of the transuranide complexes. Understanding the pNMR signatures of actinide compounds requires a rigorous theoretical framework that accounts for spin-orbit coupling, magnetic anisotropy, and covalency, while maintaining close synergy with experimental data. Paramagnetic NMR experiments on actinide complexes are performed by Claude Berthon (CEA Marcoule France). In this work, we apply advanced theoretical methods—based on relativistic quantum chemistry, density functional theory (DFT), and wavefunction-based approaches—to model and interpret the pNMR chemical shifts (^{1}H*,* ^{13}C*,* ^{15}N*,* ^{17}O*,* ^{19}F*) of two series of actinide complexes,* $[An(NO_3)_6]^{2-}$ *and [AnDOTA]L An=U, Np, Pu and L=*H_20*,* F^-*.*

In complexes with a strong anisotropy, it gives access to the anisotropic magnetic susceptibility Δχ. In complexes with a compact coordination sphere, it gives access to the spin density at the nuclei of the ligands, and consequently to the ligand hyperfine coupling constants. The pathway of the spin density from the magnetic 5f orbitals to the nuclei will be discussed.

For the DOTA series, the primary effect of the apical molecule on the magnetic properties is analyzed. The magnetic susceptibility tensor of the [UDOTA]H_20 *complex, calculated from first principles calculation and deduced from pNMR spectroscopy is compared to the torque magnetometry and SQUID experiments performed by the group of Mauro Perfertti (Florence, Italy).*

The interplay between experiment and theory is essential in magnetic properties of actinides: theoretical models rely on experimental data for validation, while experimental results require theoretical interpretation to unravel the underlying electronic and magnetic interactions.

NUCL 4344821

Reactions of transuranic cations with nitric oxide studied by inductively coupled plasma tandem mass spectrometry

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The actinides reside at the edge of the periodic table and test the utility of current chemical theory because several competing electronic structure effects converge. Adequate theoretical models must consider relativistic effects, large spin orbit coupling constants, and many low-lying electronic states to provide a reasonable prediction of actinide chemistry. A central question to address in An research is the extent of 5f orbital participation in the observed chemistry. Gas phase studies offer an ideal route to examine the intrinsic chemical behavior of the actinides because perturbations such as solvent effects are absent, which enables a direct comparison with theoretical results. The primary obstacle in assessing the accuracy of theoretical models of the An is the limited availability of experimental gas phase transuranic values for comparison.

Inductively coupled plasma tandem mass spectrometry was used to study the energy dependence of the reaction An^+ + NO (An^+ = Th, U, Np, Pu, Am). The reaction to form AnN^+ is endothermic and enabled a determination of An^+-N bond dissociation energies, $D_0(An^+\text{-}N)$, which show a strong correlation to the energy required to access the first excited state with two free 6d electrons, $E_p(6d^2)$. A comparison of the rates of the reaction forming AnO^+ to the respective lanthanide analogs indicates that participation of the 5f orbitals may contribute to enhanced An^+ reactivity even though they are not the primary contributors to bonding in the final product. An examination of the potential energy surface provides direct evidence of 5f orbital participation in bonding in the linear intermediate ($NAnO^+$), which lowers the N-O bond activation cost and explains the increased reaction efficiency observed for the An^+.

NUCL 4345521

Career panel in nuclear chemistry

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The nuclear science field offers rich and broad career trajectories ranging from fundamental research in academia to innovative applications in industry and national laboratories. It is often complex for young investigators to navigate the breadth of opportunities available. This panel, presented as part of the "Young Investigators in Nuclear Chemistry and Technology" session, will highlight career pathways in nuclear science by bringing together researchers, leaders and professionals from various sectors. Panelists will share personal experiences on breaking into the field, discuss the interplay between technological advancements and research priorities, and highlight the transferable skills that open doors to a variety of roles. Attendees will gain practical insights into how to identify mentors, seek interdisciplinary collaborations, and align professional goals with strategic growth areas in nuclear chemistry, radiopharmaceuticals, reactor design, radiation safety, and beyond. The interactive discussion will help participants envision how to build fulfilling and impactful careers that address global challenges, from energy security to medical innovation, while advancing the boundaries of nuclear science and technology.

PRES 4278755

Careers in industrial radiochemistry

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This poster will describe experiences of working in industrial radiochemistry. What types of knowledge and skills are needed, what roles and responsibilies are involved, and how does one combine scientific knowledge and material from suppliers/production to create useful products?

PRES 4281516

Peptide-based radiological agent for diagnosis and therapy of multiple myeloma

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Macrocyclic peptides are gaining significant attention by targeting disease-specific markers due to their high in vivo stability, optimal pharmacokinetics, high target affinity and ease of synthesis. Peptides hold promise as theranostics for early disease detection using positron emission tomography (PET) and serve as radiotherapeutics, addressing the need for effective treatments. However, these approaches are not available for multiple myeloma (MM). MM is a plasma cell malignancy originating in the bone marrow where early diagnosis and therapy are crucial for effective disease management. CD38, a receptor uniformly and highly expressed in MM is an ideal theranostic target. Here, we present the development of a first-in-class peptide-based radiotheranostic, [^{68}Ga/^{225}Ac]AJ263, and comprehensively evaluate its utility as a PET imaging agent and therapeutic efficacy in mouse models of MM.

A bicyclic peptide AJ263 was synthesized using solid phase peptide synthesis and characterized by mass spectrometry. AJ263 exhibits K_D of 17.9 nM affinity for human CD38. AJ263 was radiolabeled with gallium-68, a PET radioisotope in a high radiochemical yield and purity. [^{68}Ga]AJ263 shows high binding with MOLP8 and MM1S cells (CD38^{high}) and low binding with U266 cells (CD38^{low}). The specificity of [^{68}Ga]AJ263 for CD38 was validated by the reduced uptake in the presence of 1 μM non-radioactive AJ263 and its correlation with receptor density measured via flow cytometry. PET imaging of mice with MM xenografts showed rapid clearance from normal tissues and high accumulation in MM tumors, producing high-contrast images by 60 minutes. These findings were further validated by CD38 IHC analysis of tumor sections.

Next, AJ263 was labeled with actinium-225, a therapeutic radioisotope. Therapeutic evaluation of [^{225}Ac]AJ263 in MOLP8 tumor model showed significant tumor control and improved median survival, compared to saline-treated controls (25 d vs. 15 d for 18.5 kBq, $P < 0.002$). Safety assessments, including body weight monitoring and H&E

staining of lung and liver tissues, indicated marginal toxicity for [^{225}Ac]AJ263 treatment. The combination of [^{68}Ga]AJ263 for PET and [^{225}Ac]AJ263 for radiotherapy underscore potential to guide personalized radiotherapeutic interventions for MM and other CD38-expressing diseases.

PRES 4285372

Spectroscopic investigation on the electronic features underpinning the variable color of Pu(IV) oxides

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Plutonium dioxide (PuO_2) powder has long been known to vary in color, from light tan to deep olive green, the latter sometimes appearing almost black. The spectroscopic features that underpin this color variation remain underexplored. Using diffuse reflectance spectroscopy, we investigated the optical features that give rise to the apparent color via the analysis of four samples of PuO_2 powder. For each, we collected high-resolution diffuse reflectance spectra which covers the entire UV/Vis/NIR/SWIR region (250−2500 nm). The spectra reveal that both a Pu–O charge transfer transition and superimposed Pu-based 5f–5f transitions contribute to the perceived color. We establish a possible relationship between these optical features and the calcination temperature used to prepare each sample. Yet, controlling for temperature, smaller variations in material color can still be observed between otherwise similar samples. To further explore this behavior, we analyzed three additional PuO_2 samples that were prepared via similar methods. Each specimen displayed similar hues but varied in overall luminance, caused by a notable broadening of the Pu–O charge transfer transition. Chemical, physical and structural parameters that could potentially explain these subtle color variations are presented, including data from X-ray diffraction, thermogravimetric analysis, and vibrational spectroscopies.

PRES 4289829 - Withdrawn

PRES 4290800

Enriched stable isotopes: Precursors to high purity radioisotopes

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The enrichment of a stable, naturally occurring isotope is a critical first step in the production of high purity radioisotopes. Historically, the enrichment of these materials was completed using the calutrons, which was a civilian repurposing of systems from the Manhattan Project.

Renewed efforts are underway for the enrichment of stable isotopes in the United States using modernized techniques.

The high isotopic purity of these materials makes them appropriate for use in energy, medicine, security, and basic research applications. These varied applications utilize a broad array of chemical and physical forms. The development of the required fabrication techniques is driven by lowering material losses and optimizing material recovery for reuse. This presentation will introduce stable isotope enrichment, discuss the fabrication of targets, and their application to the production of radioisotopes.

PRES 4296364

Toward systematizing radiopharmaceutical development and evaluation

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Radiotheranostics have changed how we diagnose and treat human cancers. However, many of the agents being evaluated now are derivatives of molecules specific to targets we have known about for decades. For the modality to gain a firmer foundation, systematic development of novel agents is needed—that is new molecules specific to new targets for new diseases. Novel targets can be identified by integrating next-generation sequencing datasets of patient tumor samples. New biomolecules specific to these targets can exploit increasingly economical discovery (e.g. phage display, high-throughput screens) as well as artificial intelligence platforms (e.g. AlphaFold2) can be integrated in existing radiotheranostics labs to achieve these goals. Using hepatocellular carcinoma, a radiosenstive cancer for which there is both a need for functional imaging and improved treatments, Dr. Escorcia shares work attempting to address these unmet clinical needs from his lab and beyond.

PRES 4301633

Hospital-integrated molten salt reactors: Sources of power and radionuclides

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Molten salt reactors (MSRs) have emerged as a promising technology for energy generation, offering advantages in safety, efficiency, and fuel utilization over traditional nuclear technologies. In addition to their role in power generation, small-scale MSRs present an opportunity for direct implementation in medical facilities, where they could simultaneously supply reliable energy and produce medically useful nuclides on-site, while maintaining minimal waste production.

Hospitals rely on a steady supply of radionuclides for diagnostic imaging, cancer therapy, and other medical applications. Currently, many medically useful nuclides are produced in specialized facilities and must be transported to hospitals, leading to logistical challenges, half-life limitations, and potential supply chain disruptions. A dedicated small-scale MSR operating at a hospital could mitigate these issues by generating nuclides on-site, ensuring consistent availability.

This research explores the feasibility of hospital-integrated MSRs, examining reactor size, fuel composition, and isotope extraction methodologies tailored to medical applications. Operational and safety considerations are also evaluated to ensure compatibility with healthcare environments, maximizing tailored benefits to hospitals. Additional topics in the overlap of nuclear science and medicine, such as how the combination of MSRs and medical facilities would positively impact radiopharmaceutical research, will be briefly discussed.

The integration of MSRs into hospital settings offers several advantages. MSRs operate at high efficiency and provide a stable source of electricity, reducing reliance on conventional power sources and enhancing energy security for medical operations. The operation of a MSR allows for real-time harvesting of short-lived medically useful nuclides, eliminating delays associated with transportation and extending the usability of these isotopes. MSRs minimize waste production while maximizing safety, making them well-suited for deployment in sensitive facilities in populated areas.

The successful deployment of MSRs in hospitals could revolutionize medical nuclide supply chains while providing a reliable source of energy for healthcare facilities. By combining power generation with nuclide production, MSRs could significantly enhance the efficiency and self-sufficiency of modern hospitals, paving the way for a new paradigm in nuclear medicine and energy sustainability.

PRES 4301994

Using radioactive material to evaluate decontamination of contaminated electronics

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Electronic materials are used everywhere and can get easily contaminated by their use in the field/laboratory. With electronic devices getting smaller and smaller, electronic devices become harder to clean when they become contaminated. The goal of this project was use radioactive material to track the effectiveness of cleaning procedure for contaminated electronics. Radioactive potassium bromide (KBr) was used as a model contaminant in four contamination scenarios to gauge the effectiveness of decontamination of contaminated electronic devices. This poster will discuss the *contamination scenarios, the decontamination technique, and effectiveness of the decontamination procedure.*

PRES 4306606

Tracing transition metal signatures in uranium dioxide using a tube furnace: Advancing nuclear forensics for radiological threats

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The ability to trace the provenance of radiological materials is critical for nuclear forensics, particularly in scenarios involving radiological dispersal devices (RDDs). This study focuses on the synthesis and characterization of uranium dioxide (UO_2) doped with transition metal impurities, simulating potential forensic indicators in post-detonation debris. Transition metals such as Cr, Fe, and Ni, known to persist through uranium ore processing, serve as key tracers for determining material origin. UO_2 samples are synthesized via solid-state reaction by melting uranium oxide with transition metal powders utilizing a tube furnace. The elemental incorporation is quantified using inductively coupled plasma mass spectrometry (ICP-MS) while structural and phase analysis is performed using powder X-ray diffraction (pXRD) and Raman spectroscopy to assess metal incorporation and its impact on the UO_2 crystal structure. The findings from these syntheses provide crucial insights into the persistence and transformation of metallic impurities within radiological materials, enhancing forensic capabilities for source attribution in RDD events. By analyzing the differences between pure UO_2 and impurity laced UO_2, this study contributes to improving nuclear forensic methodologies and strengthening national security responses to radiological threats.

PRES 4307909

Structural investigation of UO_3 polymorphs via neutron scattering

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In recent years, significant progress has been made in understanding the structure-property relationships of the uranium trioxide (UO_3) polymorphs. UO_3 crystallizes in at least 6 distinct structural modifications in addition to an amorphous phase. The consistent stoichiometry and variable structure of these materials make the UO_3 polymorphs an ideal system for exploring optical vibrational spectroscopic signatures and the structural origins thereof. The structural diversity of UO_3 can be attributed to variations in precursor materials and synthesis parameters. Three polymorphs (α-, δ-, and ε-UO_3) are of particular importance to both basic actinide science and applied

nuclear fuel cycle chemistry and questions remain regarding the exact details of their crystal structures. To this end, we employ powder neutron diffraction and inelastic neutron scattering techniques to elucidate features in their crystal structures that give rise to unique optical vibrational spectroscopic signatures.

PRES 4309630

Studies on the incorporation of nuclear waste relevant species into studtite

***Zachary Murphy**, zach.murphy0302@gmail.com, Marlena Mutter, Peter Lopez, Vasileios Anagnostopoulos. Department of Chemistry, University of Central Florida, Orlando, Florida, United States*

Studtite ($UO_2O_2 \bullet 4H_2O$) is a uranyl peroxide mineral known to form in nuclear waste environments through the reaction of uranium species with hydrogen peroxide generated by the alpha radiolysis of water. During the precipitation of studtite, other species present in the complex matrix of nuclear waste can be incorporated into the studtite mineral structure, such as fission and decay products. This incorporation may play a critical role in immobilizing environmentally hazardous radionuclides, thereby reducing their mobility and mitigating long-term environmental risks. However, this process also alters the stability and behavior of the uranyl peroxide phase, with potential implications for the evolution of nuclear waste over time.

As such, this work investigated the incorporation of nuclear waste relevant species into the studtite mineral structure. The resulting solid phase was characterized using XRD, SEM-EDS, FTIR, and XPS to assess structural and compositional changes in the mineral. Sequential extractions, coupled with ICP-MS analysis, were utilized to provide insight into the extent and mechanism of incorporation. The findings of this study enhance our understanding of the chemical transformations occurring in nuclear waste over extended timescales, particularly in deep geological repositories. By elucidating the behavior of contaminant incorporation in studtite, this research informs predictive models of radionuclide mobility and contributes to the development of safer, long-term nuclear waste management strategies.

PRES 4309724

Molten salt synthesis efforts towards lanthanide and actinide borides

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The long-term storage of nuclear waste is an ongoing technical and policy challenge. Actinide waste, due to the millions-to-billions of year half-lives, is a particular challenge, as storage must last functionally the rest of the planet’s history. As decarbonization efforts will benefit from the increased implementation of nuclear energy and policy efforts reduce the spread of nuclear weapons in stockpiles worldwide, the way in which

we manage radioactive waste becomes ever more integral in their success. The goal of this work is to establish a repeatable, milligram-scale synthesis of phase-pure lanthanide and actinide borides by exploring reactions between common f-element starting materials and boron sources with varied reductants. The metal hexaboride (MB_6) target phase is of great interest due to its refractory properties, making it particularly useful as a nuclear waste form. Its chemical and thermal stability suggests an ability to withstand long-term irradiation. Historical syntheses of hexaborides either require high temperatures (> 1500^{o}C) or have failed to produce a phase-pure hexaboride product, suggesting metastability. The work presented will address a molten salt synthesis approach, intended to reduce the energy requirement with faster and lower-temperature reactions, preventing the formation of more thermodynamically favored borides. Discussion of trends will focus on evidence from powder x-ray diffraction, thermogravimetric analysis, scanning electron microscopy, and energy dispersive x-ray spectroscopy.

PRES 4310748

Metal doping in uranyl peroxide for enhanced nuclear security

***Marlena Mutter**, ma820491@ucf.edu, Zachary Murphy, Vasileios Anagnostopoulos. Chemistry, University of Central Florida, Orlando, Florida, United States*

As a result of various instances of unauthorized activities involving radioactive materials, a countless number of expensive and drawn-out investigations that frequently remain inconclusive have been initiated. Doping precursor materials of commonly used (and frequently mishandled) nuclear fuels with specific trace amounts of metal taggants could offer a straightforward, cost-effective method of generating intentional signatures and provide an additional tool for investigative work needed to pinpoint the origin of illicit material. In this study, tagged uranyl peroxide tetrahydrate (studtite, $UO_2O_24H_2O$) was synthesized via precipitation from aqueous uranyl nitrate hexahydrate ($UO_2(NO_3)_26H_2O$) solutions containing dissolved metal dopants. Initial incorporation rates of each dopant metal were estimated by comparing their initial concentrations in solution against residual concentrations in the supernatant after precipitation. Structural incorporation and the distribution of dopants within the studtite matrix were evaluated using Raman spectroscopy, X-ray diffraction (XRD) and scanning electron microscopy–energy dispersive X-ray spectroscopy (SEM–EDS). The data collected in this study serves to inform and refine synthetic protocol for incorporating forensic signatures within certain nuclear fuel precursors.

PRES 4312156

Radiotracers in mechanistic studies

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Radiotracers are molecules that have one or more radioactive atoms in their structure, making them detectable through the emitted radiation. They have provided valuable insights across various scientific disciplines, such as chemistry, biology, medicine, and environmental science. Radiotracers act as markers that can track the fate of a specific molecule or process in a system. In this talk, I will focus on their use in investigating chemical reaction mechanisms and biological pathways. Two distinct features make radiotracers particularly attractive for these studies. First, the detection of radioactivity is quantitative and linear. Second, tracking even in very low concentrations is possible due to the unparalleled sensitivity of radioactivity measurement. To create a radiotracer, a radionuclide should be easily incorporated into the compound of interest. Selecting the appropriate radionuclide involves careful consideration of factors such as half-life, type of radiation, chemical properties, biological behavior, availability, cost, and safety. The half-life of the radionuclide dictates how long it can be followed in the experimental system, and nuclear characteristics dictate what kind of detection method should be used. The radiotracer should mimic the behavior of the compound it is tracing. In some studies, it is necessary to create an exact radiolabeled replicate of the compound of interest, such as sucrose labeled with carbon-14 (^{14}C) to study sucrose metabolism. In this case the radiolabeled sucrose will behave exactly as the non-radiolabeled sucrose since the radiolabel is introduced by replacing one of the carbons in the sucrose with radioactive isotope of carbon – ^{14}C. In the simplest form the radiotracer can be just a simple molecule or the radionuclide itself. For instance, $^{42}K^+$ and $^{13}NO_3^-$ have been used to study plant development and nutrient absorption in different growing environments. In some experiments non-isotopic introductions of radionuclide are possible and sometimes they are even preferred due to nuclear characteristics and availability of suitable radionuclides. For example, large (bio)molecules often behave similarly to the original structure after the addition of a small radioactive motif. During this talk, I will go over examples of how radiotracers have provided detailed insights into the mechanisms of chemical and biological processes.

PRES 4312646

Advancing nuclear science through separation techniques

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Separation techniques play a critical role in nuclear science, enabling advancements in fields such as nuclear fuel cycle, nuclear forensics, and isotope production. Liquid-liquid extraction, ion exchange resins, and precipitation methods are widely employed to isolate, purify, and recover actinides, fission and activation products for a range of applications. In the nuclear fuel cycle, these techniques are essential for reprocessing spent fuel, minimizing waste, and ensuring sustainable resource utilization. In nuclear forensics, precise separations support the identification of radioactive signatures, aiding in nonproliferation and security efforts. Additionally, the production of medical and industrial isotopes relies on efficient separation processes to ensure high-purity products for diagnostic and therapeutic use. As the global energy landscape evolves *and the demand for critical isotopes increases, refining these separation methods remains imperative for advancing nuclear technology, ensuring national security, and supporting future innovations. This presentation will discuss the principles of these techniques, examples of their current applications, and the future directions in nuclear science that depend on continued advancements in separation methodologies.*

PRES 4313612

Computational modeling of iron and cobalt separation for ligand optimization

Brittany Stieferman[1]*, brittanystieferman@ucf.edu, Charles C. Peterson*[2]*, Madeleine Forbes*[3]*, Rachel Wood*[3]*, Jonathan Lovett*[4]*, Deanna Nguyen*[4]*, Grayson Spillman*[4]*, Omehvy Rivera Martinez*[4]*, Julian Poston*[4]*, Nathalie A. Wall*[3]*, Deborah A. Penchoff*[1]*. (1) University of Central Florida, Orlando, Florida, United States(2) University of California Los Angeles, Los Angeles, California, United States(3) University of Florida, Gainesville, Florida, United States*

Efficient predictive capabilities are critical for iron (Fe) and cobalt (Co) separation, as these metals play key roles in industrial, environmental, and nuclear applications. Accurate computational modeling of metal-ligand interactions facilitates the optimization and design of selective extractants, reducing the need for costly and labor-intensive experimental studies. This study employs computational modeling to investigate the thermodynamic properties and binding interactions associated with the extraction of Fe(III) with tributyl phosphate (TBP) from hydrochloric acid (HCl) and nitric acid (HNO_3) solution, and the extraction of Co(II) by N,N,N,N Tetraoctyl Diglycolamide (TODGA) from HCl solutions. This work provides valuable insight into the coordination and binding patterns of the first-row transition metals. The computational modeling based on electronic structure theory is employed to predict the structural and thermochemical characteristics of the bonded species, including the Gibbs free energy (ΔG) of the extraction reactions, and data compared to the experimental work on the same systems. These data are used to clarify metal speciation, metal-ligand affinity, and separation efficiency. Evaluations of methods performance will also be addressed. The insights gained from this study aim to contribute to the refinement of computational modeling techniques applied to transition metal separations, thereby increasing accurate predictions for the solvent extraction chemistry of the Fe and Co.

PRES 4313989

Processing signatures in Calcinations of uranium ore concentrates

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Uranium ore concentrates (UOCs) are uranium materials formed during mining, milling, leaching, and precipitation done at the front end of the nuclear fuel cycle. The

predominant UOCs produced in industrial settings are ammonium uranyl carbonate, uranyl peroxide, and ammonium diuranate. Although these processes are well established, underlying chemistry questions still need to be answered about the precipitation reactions that are not well understood. Recent work on the structure of ammonium diuranate has shown that long-held structural assumptions are not true, highlighting a need for a better understanding of the material. In this study, precipitation reactions were conducted under various processing conditions to determine the signatures present for changes in temperature, stir rate, and strike direction. Each precipitate was then split into three batches and calcined to UO_3, U_3O_8, and UO_2. Processing signatures of the calcinations were then studied using spectroscopic and crystallographic techniques. Sample heterogeneity was also examined using a variety of spatially resolved analytical techniques. Additionally, changes in trace elemental composition and uranium isotope ratios of the samples were examined.

PRES 4315186

Ion Dynamics in Radioactive Environments and Materials (IDREAM)

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The scientific mission of the Ion Dynamics in Radioactive Environments and Materials (IDREAM) Energy Frontier Research Center is to master the cascade of radiation chemistry that drives far-from-equilibrium speciation and reactivity in chemically complex environments, linking attosecond timescales to decadal processes. By achieving its scientific mission, IDREAM is providing the technical basis to predict and control speciation and reactivity—this can be leveraged by the Department of Energy (DOE) to support operational decisions for improved efficiency in retrieval, transport, and treatment of millions of gallons of legacy highly radioactive waste.

Plutonium production at DOE's Hanford Site has resulted in the most complex environmental cleanup in history. Accomplishments in the last four years have uniquely positioned IDREAM to fully focus our efforts on how radiation has driven solution speciation and precipitation to yield the complex phase assemblages at the heart of the waste processing challenge. IDREAM is studying the extreme far-from equilibrium conditions that result from the interplay between high ionic strength and radiation-induced metastability across different ***spatiotemporal regimes (STaRs)*** *by targeting*

the following project objectives: (1) Identify the instantaneous, radiation-induced changes to electronic structure that determine reactive species in concentrated electrolytes. (2) Characterize nonequilibrium ensembles and reaction cascades within concentrated alkaline electrolyte ion networks. (3) Determine how radiation drives nucleation, growth, dissolution, and aggregation of particles in concentrated electrolyte suspensions.
By achieving our project objectives over the next four years, we will create a framework to predict behavior during long-term storage of radioactive waste, not just from weapons production and contemporary fission reactors, but also from new coolants and solvents required for Generation IV nuclear reactors and beyond. These objectives will deliver scientific innovations that transcend the current understanding of radiation-driven chemical transformation and will revolutionize the way we analyze and manipulate chemical processes in extreme far-from-equilibrium conditions, allowing us to improve tank waste retrieval and processing efficiency and save the nation billions of dollars.

PRES 4318174

Future of radiopharmaceuticals: From chemistry to the clinic

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Molecular imaging is a powerful technique that can be applied to basic, translational, and clinical research as well as to routine patient care. In particular, Positron Emission Tomography (PET) allows for spatial localization and quantification of biological processes such as metabolism, enzyme activity, cell proliferation, receptor density and cellular transport that are not readily assessed with conventional anatomic imaging techniques. Recent developments in radiochemistry have allowed for a dramatic expansion in radiopharmaceutical development and clinical translation while theranostic strategies with paired imaging and therapeutic compounds are changing the landscape of treatment strategies for many patients. What does the future hold and how can nuclear and radiochemistry enable customized effective treatments in oncology?

Ideal theranostic pairs which include radioisotopes of the same element (isotope pairs) but with different emissions (i.e., one suitable for diagnosis and the other for therapy) are emerging for precision dosimetry. Combination therapies which leverage the impact of targeted radiation on the immune system are being investigated. Finally, new targeted imaging agents which enable insights into therapeutic efficacy for both radioactive and conventional therapies, are moving into clinical trials.

PRES 4318232

Actinide separations: Challenges for today and tomorrow

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As an integral part of discovering and isolating the elements in the bottom line of the periodic table, actinide separations have been developed and refined over time. These developments allowed the Curies to separate uranium from its decay products and discover radioactivity at the turn of the twentieth century, as well as McMillan and Seaborg to isolate neptunium and plutonium for the first time in the 1940s. Today, actinide separations are commonly found in isotope production or as part of the nuclear fuel cycle to process spent fuel. Challenges lie in the isotope purity requirements, process efficiencies, ligand selectivity, kinetics of extraction systems, process conditions, equipment, and proliferation resistance. Recognized as an abundant source of energy free of gas emissions, nuclear energy's main challenge is the waste associated with the spent fuel. Efficient, proliferation-resistant recycling processes that would allow for reusing over 98% of the spent fuel have actinide chemistry at their core. An option envisioned to provide more portable energy is the development of small modular reactors. From fuel generation to spent fuel reprocessing, actinide separations will play a huge part in these reactors. Likewise, actinide separations have a significant role to play in isotope production. Whether producing isotopes for targeted alpha therapies (e.g., ^{225}*Ac,* ^{227}*Ac,* ^{230}*U) or for industry (e.g.,* ^{238}*Pu,* ^{252}*Cf), actinide separations are necessary to achieve the required amounts and purity. And when the waste of these processes contains valuable fission products, their recovery hinges on a series of tailored separations, such as solvent extraction and/or ion exchange, among others. Challenges for tomorrow lie in developing ligands or supports that can be easily disposed of once they can no longer be recycled. Modeling and artificial intelligence can be useful tools to guide such discoveries, but access to facilities where these can be tested is also critical. Examples of actinide separations currently in place and those being developed will be presented, along with an outlook on the next decade.*

PRES 4319646

What is radiochemistry and why/how can you get involved?

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From the electrons (where chemistry happens) to the nucleus (where radioactivity eminates), radiochemistry provides tools that are unique to follow reaction mechanisms, analytical processes, and much more. This opening session will introduce the audience to the "periodic table of the radiochemist" and set you up for today's Presidential Session investigating Nuclear Today & Tomorrow. If you are interested in the phenominal cosmic power for energy generation, environmental concerns for waste, continuing job growth, or challenging chemical questions in medical diagonstics and therapeutics, this Presidential Session is for you.

PRES 4320182

USANCA: Preparing the Army for the nuclear threat

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The U.S. Army Nuclear and Countering Weapons of Mass Destruction Agency (USANCA) increases the Army and Joint Force's lethality and operational survivability by providing Nuclear and Countering Weapons of Mass Destruction (CWMD) expertise and analysis to inform Strategic Guidance, Plans, and Policy. USANCA's six core functions include 1) Integrate nuclear weapon effects into joint operations; 2) Integrate nuclear deterrence, CWMD, and chemical, biological, radiological, and nuclear (CBRN) defense policy, planning, and readiness requirements; 3) Lead the Army CBRN survivability program; 4) Manage the Army Reactor Program; 5) Fulfill proponent functions for the Army Functional Area (FA) 52 (Nuclear & CWMD Officer) and ASI 5H (Nuclear Target Analyst); and 6) Increase interoperability with joint and multinational forces in CBRN environments. USANCA utilizes threat- and scientific-based analysis to modernize the Army against current and emerging threats to enhance survivability of the force. USANCA also deploys Nuclear Employment Augmentation Teams (NEAT) to support Army and Joint Force Commanders during crisis or conflict, focusing on nuclear targeting, planning, and operations. NEAT serves a unique and specialized role in combatant command planning, drawing from the Army's legacy nuclear mission and knowledge of conventional planning and execution. Finally, USANCA publishes the Countering WMD Journal semi-annually. The Countering WMD Journal is a peer-reviewed publication welcoming articles and other content from all U.S. Government agencies and academia involved with CWMD matters.

PRES 4320533

Exploration of bipolar complexants for minor actinide extraction from lanthanides in simulated spent nuclear fuel

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Expansion of carbon-neutral nuclear energy for civilian use is contingent, in part, on the development of strategies for management of spent fuel processing in an environmentally responsible, economically viable, and securely nonproliferative manner. Separation and recycling of the minor actinides from the lanthanides via the partition and transmutation strategy can decrease the volume, radiotoxicity, and heat load of spent nuclear fuel destined for storage in a geologic repository. Extraction of the minor actinides from the neutron-poisoning lanthanides presents numerous challenges as physical properties of the f-elements including ionic radius, hardness, and oxidation state are quite similar.

Work in this laboratory over the last decade has sought to develop chemoselective complexants which preferentially chelate the minor actinides over lanthanides using

liquid-liquid extraction by accentuating covalent metal:complexant interactions with soft-N-Lewis basic donors as an extension of earlier work by Kolarik. Achieving high solubility of polar molecules in nonpolar diluents, selectivity for minor actinides over lanthanides, stability to hydrolytic and radiolytic degradation, and reversibility of the metal:complexant binding event for use of the complexant for sequential extractions are at the forefront of separation method development. Recently, a discovery was made that solubility of C2-symmetric BTP complexants in process-relevant diluents, including isooctanol and kerosene, could be effectively modulated by purposeful installation of additional alkyl groups at selected positions of requisite starting materials. Complexants which display bipolar physical properties are postulated to effectively traverse the organic/aqueous interface to facilitate selective minor actinide extraction. Reversibility of the metal:complexant binding interaction without competitive ligand exchange, leveraging only adjustment of pH has also been developed. Molecular design strategies, separations validation through extraction assays and spectroscopic measurement, as well as computational approaches will be presented.

PRES 4320703 - Withdrawn

PRES 4321310

Isotopic functionalization of sulfur-heteroatom scaffolds with hydrated [^{18}F]fluoride ion

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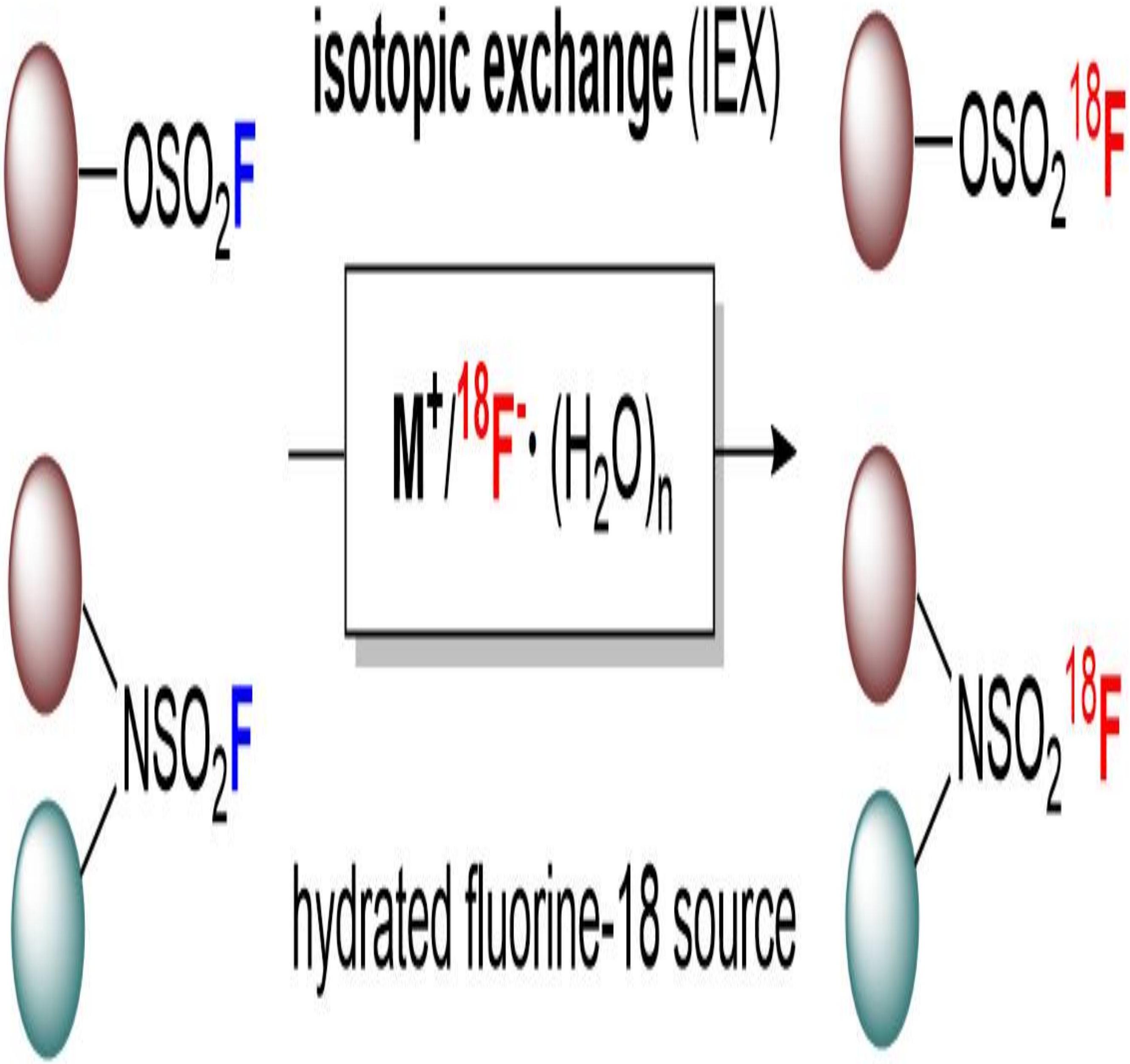
With the growing importance of ^{18}F-labeled radiotracers as effective diagnostic tools for various diseases, there is considerable interest in establishing a rapid, reliable, and robust approach to ^{18}F-radiolabeling for positron emission tomography (PET) in molecular imaging. There is an unmet need for an effective practice to install fluorine-18 into diverse chemical frameworks to functionalize the target compounds for use in image-based drug development. The fluorosulfuryl group, utilized in sulfur(VI) fluoride exchange (SuFEx), serves as a versatile functional moiety that has been extensively explored in both pharmaceutical and materials research. The use of cyclotron-produced [^{18}F]fluoride ion to activate various fluorosulfuryl moieties enhances the new possibilities for innovative approaches in drug discovery. Here, we report a streamlined method for isotopic exchange at the sulfonyl core between ^{19}F and radioactive ^{18}F. To overcome the issues associated with the short physical half-life of ^{18}F (109.7 min), we developed the azeotropic drying-free and cryptand-free radiofluorination by direct use of ^{18}F-eluent from QMA (quaternary methyl ammonium) cartridge. Various aryl [^{18}F]fluorosulfates and sulfamoyl [^{18}F]fluorides were synthesized through ^{19}F/^{18}F isotopic exchange, even with heavily hydrated fluorine-18 source (with water content up to 5% in organic reaction medium). The late-stage isotopic activation provided diverse ^{18}F-labeled aryl fluorosulfates and sulfamoyl fluorides at radiochemical conversion (RCC) up to 92%.

This labeling technique offers a reliable way to incorporate ^{18}F into sulfur-oxygen and sulfur-nitrogen frameworks. It eliminates the need for time-consuming azeotropic drying and simplifies the downstream quality control process without using cryptands. This newly devised method provides a simple and practical means to produce diverse ^{18}F-labeled SuFEx compounds, which holds promise for use in PET imaging.



PRES 4321335

Safety assessment of interim disposal facility in KFUPM area for low- and intermediate-level radioactive waste (LILW) repository using RESRAD-OFFSITE code

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An extensive assessment of the safety of an interim disposal facility within King fahd university for Petroleum and minerals (KFUPM) located in Dhahran, Saudi Arabia for the low and intermediate level radioactive waste, was done with the aid of RESRAD-OFFSITE code and is reported here. Within the boundaries of the repository acceptance criteria, 13 radionuclides and gross alpha were chosen for the evaluation preliminary study. There and in the facility, relevant groundwater was treated as undisturbed and its composition was modelled using the thermodynamic database from the Japan Atomic Energy Agency in combination with the PHREEQC program. Furthermore, the distribution coefficients of the facility elements were derived from the same agency‘s sorption data. All these geochemical parameters were then included alongside other known descriptors of the disposal facility and its site into the RESRAD-OFFSITE model. From the results, the safety assessment stated that the calculated exposure dose would be only 2.6% of the limit of 0.07mSv per year. In addition, changing key geochemical parameters confirmed sensitivity shatters the estimation but the foregone exposure dose remained within the safety limit.

PRES 4322909

Evaluation of americium and curium-ligand complexation for liquid-based separation systems

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In the research area of spent nuclear fuel reprocessing, the difficulties of extracting and separating the minor actinides Am and Cm from each other have been attributed to their similar chemical behaviors. However, the implementation of high-performance computing methods may greatly benefit research into Am/Cm separation based on interpolating data from reported bonds between these minor actinides and chelating ligands. An evaluation of Am- and Cm-containing compounds was conducted to determine preferential binding in liquid-liquid extraction systems. Gibbs free energies of

reaction were calculated with electronic structure methods. The differences in Gibbs free energies of reaction between Am- and Cm-compounds with each ligand are used to determine the predicted efficiency of their use for extracting either actinide from the other. Results from level of theory evaluations, their significant impact on the chosen parameters of the environment and theoretical descriptions of the species, and preferential binding will be presented.

PRES 4323192

ASTM, ISO, and IEEE standards organization update

Donivan R. Porterfield[1,2]*, dporterfield@lanl.gov. (1) Actinide Analytical Chemistry Group, Los Alamos National Laboratory, Los Alamos, New Mexico, United States(2) Subcommittee D19.04 (Water / Methods of Radiochemical Analysis), ASTM International, West Conshohocken, Pennsylvania, United States*

This presentation will report on the status of some voluntary standards development organization efforts (e.g. ASTM International, International Organization for Standardization (ISO), and IEEE) that may be of interest to the radiation measurements and standards community. And as an introduction, I will highlight some differences between these standard development organizations as well as U.S. government policies with respect to standard development organizations.

PRES 4323936

Reversibly cross-linkable hydrogel coating for low-temperature surface decontamination of radioactive cesium via borate–diol ester bonds

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Wide-area surface decontamination is crucial in the event of a sudden release of radioisotopes to the public, such as during nuclear accidents or terrorist attacks. Although various strippable coatings demonstrate effective removal performance in the nuclear industry, they are not ideal for wide-area decontamination in such emergencies. These coatings typically become radioactive waste after use, leading to high disposal costs, and they are often difficult to remove completely from porous surfaces.

Our group has recently developed a series of reversibly cross-linkable complexes between polyvinyl alcohol (PVA) and borate ($B(OH)_4^-$), designed for easy removal from surfaces. This approach aims to reduce radioactive waste treatment costs and enable potential reuse. The PVA–borax-based coatings from previous studies are cost-effective, easy to manufacture, and leave no residual contaminants on porous surfaces after removal. Additionally, borax provides Na^+ ions, which can exchange with Cs^+ ions via ion exchange. However, the effectiveness of such coatings under low-temperature conditions has not been thoroughly investigated.

In this study, we developed a peelable coating with a subzero freezing point to ensure

effective decontamination in cold environments. To achieve this, partially hydrolyzed poly(vinyl acetate) (PHPVAc) and borate were used, with a water/ethanol mixture as the solvent to improve performance at subzero temperatures. In this system, two separate solutions—one containing PHPVAc loaded with an adsorbent and the other containing borate—are simultaneously applied to contaminated surfaces. This forms a viscous coating with a three-dimensional gel network via reversible borate–diol ester bonding, which adheres to the surface and adsorbs Cs-137. The embedded adsorbent enhances the removal efficiency of Cs-137 even at a low temperature of −9 °C. Moreover, the coating can be easily removed by rinsing with water and leaves no residue, even on porous surfaces such as cement, due to the reversible nature of the PVA–borate complexation.

Therefore, this hydrogel system shows strong potential for the decontamination of hazardous contaminants, including radionuclides, from porous surfaces under low-temperature conditions.

PRES 4324947

Pyrrophen and other imine donor ligands in f-element complexes

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Pyrrophen ligands, inspired by six-coordinate macrocyclic ligands, are acyclic compounds featuring six potential donor sites. They are synthesized through the Schiff base condensation of ortho-phenylenediamine or 2,3-diaminopyrazine with pyrrole carboxaldehyde. These ligands can be easily modified by altering the substituents on the aromatic ring or by changing the ester or amide group on the pyrrole moiety. Previously, we reported their effectiveness in selectively coordinating with uranyl, demonstrating high selectivity for uranyl compared to first-row transition metals. In this study, we further explore the functionality of these ligands and their formation of metal complexes.

PRES 4325301

Nuclear Security: The evolving terrain of securing nuclear materials and technology against malicious acts

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The origin of nuclear security predates the Manhattan Project, beginning at the first neutron-induced fission event. The vast amounts of energy released on a per-atom basis could potentially be weaponized, and early nuclear security pracices were

deployed as an ad hoc effort. In practice today, nuclear security typically focuses on threats from non-state actors intent on theft, sabotage, or nuclear terrorism. Safeguards, nonproliferation, counter-proliferation, treaty monitoring, operational assurance, and related information analyses are viewed as separate (and in some cases disconnected) efforts.

Given recent global events, the terrain of nuclear security is changing fast. Multiple conventional and advanced nuclear power stations are under construction globally. The commercialization of space presents new challenges. The Russo-Ukraine war has broken established norms by making civilian nuclear installations strategic and tactical military targets. Long-established norms and security alliances among nuclear allies are under stress. The People's Republic of China is surging toward nuclear weapons parity with both the U.S.A. and Russia.

In this environment, it is necessary to rethink our perspectives around nuclear security to adapt to this new terrain effectively. We explore these looming challenges and assess potential strategies to meet them.

PRES 4325399

Uncovering enrivonmental radiochemistry

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Environmental radiochemistry is an interdisciplinary field that integrates chemistry and biology, both computational and experimental approaches to investigate the origins and fates of radionuclides. Legacy nuclear material production processes have left enduring impacts at sites such as the Hanford Site at Pacific Northwest National Laboratory and the Savannah River Site, where the complexity of waste continues to challenge the capabilities of current processing technologies. This talk will uncover cutting-edge research focused on advanced techniques for the immobilization of critical elements in nuclear waste, including technetium, iodine, cesium, and uranium. Additionally, it will explore innovations that deepen our understanding of environmental radiochemistry and contribute to the development of sustainable solutions for managing radioactive contamination.

PRES 4325487

Nitrates and nitrites removal from nuclear waste simulant by electrolysis with a plate electrolyzer

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Nitrates and nitrites in nuclear waste pose a serious processing and safety risk for nuclear waste disposal by vitrification. Electrochemical reduction provides a promising alternative to conventional treatment methods due to its high efficiency, selectivity, and minimal chemical input. This study investigated the electrochemical reduction of nitrate (NO_3^-*) and nitrite (*NO_2^-*) in simulated nuclear waste solutions (simulant) using a plate electrolyzer assembled with three Ti cathodes and two Ru coated Ti anodes. Key parameters, including electrode composition, current density, pH, and electrolyte conditions, were tested and optimized to maximize nitrate and nitrite removal and gaseous products (including *NH_3* and *N_2*) selectivity. The results demonstrated that the electrodes are robust and efficient, and the electrochemical treatment offers a controllable, scalable, and waste-minimizing approach for nitrate remediation in nuclear waste management. This work shows the potential for electrochemical processes to improve the efficiency and safety of nuclear waste treatment flowsheets.*

PRES 4325517

Cesium isotope determination in envirmonmental samples by TIMS

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*Release of Cs fission products, *^{135}*Cs and *^{137}*Cs, into the environment has been documented throughout the nuclear fuel cycle. The *^{135}*Cs/*^{137}*Cs isotopic ratio can be used to provide insight on various Cs source terms, including nuclear fuel reprocessing, nuclear accidents, and nuclear fallout. We present new *^{135}*Cs/*^{137}*Cs ratio and concentration thermal ionization mass spectrometry (TIMS) data on three standard reference materials: IRMM-426 Wild Berries, SRM 4350b River Sediment, and SRM 4354 Freshwater Lake Sediment. The sample set ranges from low *^{135}*Cs/*^{137}*Cs values observed in nuclear accidents (Chernobyl and Fukushima) to high *^{135}*Cs/ *^{137}*Cs values measured in nuclear fallout (Russell et al., 2015). 10-1000 mg soil aliquots of each reference material were used for *^{135}*Cs/*^{137}*Cs determination. Cs in the sample was chemically separated using a custom AMP-PAN and Eichrom Sr Resin based method based on Dunne et al. (2007) and Snow et al. (2015). TIMS analyses of SRM 4354 and SRM 4350b fractions yielded *^{135}*Cs/*^{137}*Cs ratio values of 3.56 ± 0.46 2SD and 1.94 ± 0.17 2SD, respectively (1/1/2015 reference date). An aliquot of IRMM-426 was spiked with *^{134}*Cs tracer for *^{137}*Cs concentration determination. PNNL TIMS data, 735 ± 15 2SD Bq/kg, shows good agreement with the certificate value, 780 ± 70 *^{137}*Cs Bq/kg (1/1/2009 reference date). A large *^{135}*Cs impurity in our *^{134}*Cs tracer inhibited accurate analysis of the *^{135}*Cs/*^{137}*Cs ratio in IRRM-426. Future analyses of unspiked IRMM-426 aliquots will provide a more accurate *^{135}*Cs/*^{137}*Cs value.*

PRES 4325560

Exploration of alternative anion exchange resins for plutonium separation

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Reillex HPQ is the gold-standard anion exchange resin for the separation of plutonium in nitric acid. HPQ is a methyl-quaternized poly(4-vinylpyridine) resin, crosslinked with 25% divinylbenzene, which retains Pu(IV) with high capacity, rapid kinetics, and radiological hardness. Reillex HPQ is no longer commercially available in research quantities. Therefore, there is a potential impetus to find a commercial-off-the-shelf alternative to Reillex HPQ. Four commercially available resins, Bio Rad AG MP-1, Lewatit MP 500, Reillex 425, and Reillex HP were examined alongside Reillex HPQ for their ability to remove Pu(IV) from nitric acid solutions. Three experiments were conducted to compare resin performance. First, batch Pu(IV) distribution ratios (Kd) of the five resins were measured in 0.35 M, 3.0 M, and 8.0 M HNO3 after a 24-hour contact period. Lewatit MP 500 and Reillex HP exhibited high uptake at 8.0 M HNO3. The latter had no detectable uptake at 0.35 M HNO3, while the former exhibits low uptake (Kd <10^1). Therefore, these two resins were used in a second round of experiments in which distribution ratios in 8.0 M HNO3 were determined after 1-hour of contact to investigate Pu(IV) uptake kinetics. Lewatit MP 500 and Reillex HP had similar distribution ratios after the 1-hour of contact. Reillex HP is a poly(4-vinylpyridine) resin that is highly resistant to radiological degradation compared to quaternary ammonium anion exchange resins, such as Lewatit MP 500. Because of this feature, Reillex HP was selected for the third and final experiment to directly compare the Pu(IV) capacity to Reillex HPQ.

PRES 4325968

Metal-organic frameworks (MOFs) as an emergent technology to remove mercury from nuclear waste tanks

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Thiol functionalized Metal-Organic Frameworks (MOFs) present a promising solution for the removal of mercury from high-level waste tanks, specifically at the Savannah River Site (SRS) where traditional adsorbents have proven ineffective. This study investigates the chemical properties and efficacy of these MOFs in selectively capturing mercury species in various forms and pH conditions. Through the design and structural characterization of isoreticular MOFs, specifically UiO-66-$(SH)_2$ and UiO-67-$(SH)_2$, their mercury uptake and release kinetics, stability under radiolytic and extreme pH conditions, and their adsorption capacities were evaluated. UiO-66-$(SH)_2$ demonstrated a high adsorption capacity of 90% Hg intake at pH 10 within 24 hours, with significant stability in both alkaline and radiolytic environments. These findings indicate that thiol functionalized MOFs with multiple thiol ligands and high porosity could serve as a highly efficient and deployable technology for mercury removal in waste remediation

processes, potentially reducing environmental risks associated with mercury contamination at SRS facilities.

PRES 4326379

Probing the behavior and complexation of technetium under PUREX-like conditions

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One well-used large-scale solvent extraction process utilized for spent nuclear fuel reprocessing is the plutonium uranium reduction extraction (PUREX) process. This process involves an organic phase composed of tributyl phosphate (TBP) in kerosene and an aqueous phase comprised of concentrated nitric acid. In theory, uranium (U) and plutonium (Pu) are extracted into the organic phase via TBP complexation while the fission products remain in the aqueous layer. Pu is then chemically reduced and back-extracted into the aqueous phase. However, it is known that several other elements present in the spent nuclear fuel complicate this process and follow U and Pu into the initial organic phase. This work focuses on the high-yield U-235 fission product technetium (Tc), particularly Tc-99, which has a half-life of 211,000 years. Tc exhibits a wide range of oxidation states (-1 to +7) which result in complicated redox chemistry that is induced by the radiolysis of the high radiation fields of spent nuclear fuel. It is also sensitive to the reductants and oxidants used to modify the oxidation states and extraction behavior of U and Pu. These factors, as well as the presence of dibutyl phosphate (HDBP), the radiolysis-induced degradation product of TBP, result in Tc being extracted into the organic phase in Tc-TBP and Tc-HDBP complexes that are not yet well-determined. Changes in the nitric acid concentration of the aqueous phase additionally influences the Tc-phosphate complexation. Current investigations focus on cataloging the Tc-phosphate complexes for Tc in its most common oxidation states, +7 and +4, utilizing techniques such as slope-analysis, Karl Fischer titrations, and XAS.

PRES 4326467

Using Large Language Models for data extraction with a focus on Isotope separations

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Producing and separating isotopes is a complex process involving multiple steps, such as target design, fabrication, developing the parameters for target irradiation, and proposing and optimizing separations. The success of each step relies on technical

data, which is detailed in scientific literature. With ever increasing availability of new data, locating and systematically organizing relevant information becomes key to remain competitive and efficient in research and development.
To address this issue, we explored the use of Large Language Models (LLMs) as a tool to extract and organize key information from research papers.
In addition to providing a quick summary of a journal article, the extracted data will serve as a training dataset for a new knowledge-based user interface, specially designed to support the specific workflows of isotope production and separation. By incorporating prompt engineering methods to refine how questions or commands are framed, we were able to improve model output and achieve more accurate results.
For this experiment, we conducted a quantitative evaluation using around 20 randomly selected papers and assessed the results for semantic similarity and keyword presence.
The preliminary results suggest that LLMs, guided by well-crafted prompts, can effectively extract critical data on isotope production and separation (i.e., type of resin, target material, and media).
This approach not only accelerates the literature review process but also strengthens the foundation for process optimization and informed decision-making in isotope research and production.

PRES 4326691

On-ramping the fusion economy with kilogram quantities of commercial tritium

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For many reasons, the US has no commercial, domestic tritium production capabilities. The value (2024 \$) of commercial tritium is ~\$33,000,000/kg. A 1 GW_{th} D-T fusion energy plant full power year (FPY) will need more than 55 kgs of tritium/year. These power plants are hoping to breed tritium during operation and the required Tritium Breeding Ratio (TBR) to feed back to the fusion reactor must be > 1.0 (ideally, 1.1 – 1.2). Small uncertainties (~1%) in system TBRs can still lead to changes of over +/- 500 g = per FPY at 1 GW_{th}. Starting a fusion plant with no tritium (using D-D reactions to breed tritium) is not economically viable.
Currently, commercial tritium supplies are produced in heavy-water reactors like the 600 MW, Canada Deuterium Uranium (CANDU) at rates of 0.1 kg / yr.
We propose to investigate the design, development, performance requirements and cost of an accelerator-driven system (ADS) using molten salt (MS) technology as the working material for transmuting used reactor fuel and producing a supply of commercial tritium.
Recycling and transmuting used nuclear fuel (UNF) in an ADS satisfies multiple needs: 1) Long-lived transuranic material is destroyed, thereby improving the acceptance of a UNF repository, 2) Energy is produced by fission (offsetting the power used by the accelerator), and 3) The system is operated in a sub-critical configuration, which improves safety while minimizing criticality constraints.
This ADS+MS concept is well-suited for a commercial tritium production mission and the

US Department of Energy has the requisite experience with handling, processing, storing, and transporting the products.
An ADS+MS facility can achieve TBRs > 20 with current technology, provide kg quantities of tritium annually, decrease the overall cost of construction and operations at a fusion power plant, and lower proliferation risks.

PRES 4326810

Investigation of rhenium chloride speciation in quenched molten salts

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Molten salts reactors (MSRs) are an emerging nuclear reactor technology where the nuclear fuel is dissolved in molten chloride or fluoride salt that acts as the primary heat transfer fluid. While operating at high temperature, the fuel salt composition will change as fission products are produced. One significant challenge to this technology will be removal of fission products to maintain reactor operation. Technetium-99 (Tc-99) is a high-yield product of uranium fission; its extensive redox chemistry and 210,000-year half-life render Tc extremely problematic in nuclear fuel cycles. Understanding Tc-99 speciation in these conditions is maintaining stability of the nuclear fuel cycle. While ongoing radiological lab equipment upgrades prohibit Tc-99 chloride dissolution at high temperatures, we explore speciation of variable oxidation state rhenium chlorides in LiCl-KCl based melts. Rhenium is a non-radioactive surrogate for technetium that offers similar coordination environments and reactivity. Additionally, analogous rhenium studies allow refinement of radiological containment and analytical techniques that will be used in Tc studies. Herein, we present ex-situ x-ray absorption spectroscopy (XAS) studies of the quenched melts to explore oxidation state changes and coordination chemistry of variable oxidation state rhenium chloride compounds in LiCl-KCl based melts. These initial experiments provide the spectroscopic library necessary to identify mixed species produced under high temperature dissolutions.

PRES 4326897

Inspiring the next generation of nuclear scientists through the Nuclear Chemistry summer school and beyond

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Ever since the 1970s, a constant specter in the area of nuclear science has been maintaining an adequate supply of trained experts in the field. Challenges in this arena have historically been the limited facilities and institutions capable of providing this training, the aging workforce, and the increasing national need in the US. Additionally, public perception of nuclear science has often been a hurdle to recruiting students. Increasing educational efforts aimed at young scientists is a core strategy in overcoming these obstacles and ensuring a safe, knowledgeable, and innovative nuclear workforce. The Nuclear Chemistry Summer School (NCSS) was first funded by the Department of Energy in 1984 to address these concerns about the increasing demand for trained nuclear and radiochemists in particular. Its founding goals were to 1) provide fundamental training in nuclear and radiochemistry principles, particularly to physical science and engineering students from institutions that do not have such programs; 2) stimulate interest in physical science and engineering undergraduates in graduate studies in fields founded in nuclear and radiochemistry; and 3) generate awareness and acceptance of nuclear technologies and application in society among participants. Over the last forty years with the ongoing support from the DOE and ACS, the NCSS has followed through on these goals by training over 900 undergraduate students between its two sites at San Jose State University and Brookhaven National Laboratory. The summer school is a six-week intensive program for advanced undergraduate students that includes course lectures, hands-on counting and wet lab experiments, guest seminars, and field trips to local nuclear facilities. The NCSS has had a sizeable impact on the field with many alumni from the summer school going on to become professors, national lab scientists, industry leaders, and more.
This talk will discuss successes and opportunities for the ongoing NCSS program and other growing efforts to increase student engagement with nuclear science.

PRES 4326907

Solvent extraction of europium and gadolinium with a TODGA-TPPO system

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Aqueous reprocessing of used nuclear fuel (UNF) is essential for reducing its long-term radiotoxicity and enhancing the safety and public perception of nuclear energy. In the nuclear fuel cycle, solid spent fuel is reprocessed using a two-phase solvent extraction process. The aqueous phase consists of spent fuel dissolved in concentrated acid, while the organic phase typically comprises an extractant dissolved in a long-chain hydrocarbon solvent. The extractant is designed to possess optimized selectivity and rapid coordination for targeted radionuclides, effectively partitioning them into the organic phase. A diglycolamide (DGA) ligand, N,N,N',N'-tetraoctyl diglycolamide (TODGA), selectively extracts lanthanides and actinides from spent fuel. This study investigates the separation of europium (Eu) and gadolinium (Gd) as surrogates for

americium (Am) and curium (Cm) using a two-phase extraction method with a ligand complex of TODGA and triphenylphosphine oxide (TPPO). Eu and Gd are non-radioactive surrogates that allow the study of solvent extraction mechanisms under safe, controlled laboratory conditions. Previous systems combining TODGA with co-extractants such as tri(n-butyl) phosphate (TBP) or DGA derivatives demonstrated synergistic behavior, improving extraction efficiency. We propose that the TODGA-TPPO combination could enhance extraction efficiency; TPPO acts as a synergist, stabilizing complexes and influencing phase behavior to reduce third-phase formation. The physicochemical properties of Eu and Gd will be analyzed to evaluate the impact of TPPO with TODGA extraction efficiency. This study utilizes an aqueous nitric acid solution and an organic phase of octanol and kerosene, the ligand complexation is hypothesized to improve Eu and Gd separations. Optimal conditions will be identified by varying physicochemical parameters. These findings will provide insights into factors influencing extraction behavior, contributing to solvent extraction advancements and enhancing spent fuel reprocessing by isolating target lanthanides.

PRES 4326940

Vapor diffusion synthesis of α-UO_3

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α-UO_3 is a common material in the nuclear fuel cycle owing to the simplicity of its synthesis using readily available chemical precursors. Therefore, study of its structure-property relationships is of utmost importance due to its propensity to appear in the processing of nuclear materials. The exact crystal structure of this material has been the subject of much debate for decades. Obtaining large, single crystals of α-UO_3 would greatly aid diffraction studies in this discussion, however previous synthesis methods inhibit the production of suitable samples. Current methods involve the rapid addition of liquid hydrogen peroxide to a uranyl nitrate solution to precipitate uranyl peroxide tetrahydrate (studtite). Upon heating, studtite pseudomorphically decomposes to form small α-UO_3 crystals. In this work, we utilize this pseudomorphic decomposition by initially growing large crystals of studtite which retain their morphology during heating to the trioxide phase. Vapor phase hydrogen peroxide is slowly introduced to a solution of uranyl nitrate which allows for the formation of crystals suitable for single crystal diffraction studies. This work demonstrates a novel twist on an established method which could help provide crucial structural information of not only a key nuclear fuel cycle material, but also other materials where poor crystal size hinders structural determination.

PRES 4327338

Comparative study on stress assisted corrosion behavior of stainless steel 316 in a molten NaCl-Na_2SO_4 salt

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Structural performance in molten salt environments is important for materials used in high temperature aerothermal components at marine sites and for novel energy generation such as thermal, solar, and next generation nuclear power systems. Characterization of materials under these operative conditions is critical for understanding their ability to resist corrosion damage. Stainless steel 316 (SS316) is a common material of choice for these high-temperature corrosive environments. SS316 is an austenitic chromium nickel stainless steel with an addition of molybdenum which increases its corrosion resistance and mechanical properties at elevated temperature. The present research aims to characterize corrosion mechanisms and mechanical degradation of stainless steel 316 in Na2O4-NaCl molten salt under biaxial loading conditions and compare the behavior to previous work done on a next-generation high entropy alloy $Al_{0.1}$CrCoFeNi & Alloy 718. Depletion zone depth has been measured as a function of time for these materials at 750°C under biaxial stress imposed using a novel disc bend configuration.

PRES 4327388

Investigation of candidates for reactor produced radioactive materials in support of radiological training exercises

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Bromine-82, Potassium-42 and Copper-64 have been successfully adopted as radioactive surrogates for outdoor large area contamination training. This poster will discuss the investigation of new materials that could supplement potassium bromide (KBr) and copper pellets in radiological dispersal device (RDD) training events to reduce the down time of the training fields and to broaden the toolbox of the RDD surrogates at Idaho National Laboratory.

PRES 4327491

Nuclear survivability for deterrence

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Nuclear survivability sits at the nexus of nuclear science, operational warfighting, medicine, and intelligence. Survivability of humans, materiel, and communications systems is a vital component of deterrence across all domains – it denies adversaries

the benefit of nuclear weapon use by enabling warfighters to fight and win on a nuclear battlefield. The US Army Nuclear and Countering Weapons of Mass Destruction Agency (USANCA) is responsible for ensuring that testing standards and criteria for nuclear survivability are threat-informed, scientifically sound, and operationally relevant. Ongoing efforts to update legacy Cold War requirements, understand and improve human survivability, and prepare for emerging technological advancements help ensure that on the worst day imaginable, U.S. soldiers are able to survive, fight, and win.

PRES 4327698

Understanding radiolysis effects on TcO_4^-/ReO_4^- reduction and exploring reduction in the presence of phosphates

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Using both UV-Light and Gamma rays as irradiation methods, Technethium and its surrogate Rhenium are probed for their products. Using UVC light allows for efficient electron transfer reactions that can mimic the ionizing radiaiton effects of gamma rays. Using both methods, technetium was effectively reduced from its +7 oxidation to +4 in dilute sulfuric acid medium contianing 2-propanol. Next, the introduction of a PUREX relevant organic, tributyl phosphate, allowed for further understanding of how technetium reduction and stability is influenced in the presece of organic contaminents. By using a combination of a homemade UV-irradiatior and Co-60 sources at Brookhaven National Lab many conditions were tested. This reduction was proped using a variety of techniques including ITLC, XAS, UV-Vis, and NMR. Comparisons with Rheium are also done at many points and prove to have differening results.

PRES 4335034

Radioisotope production: Today and tomorrow

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Radioisotopes are produced at nuclear reactors, linear accelerators, cyclotrons, and mined fromvarious places on earth. This discussion will cover the basics of radioisotope production and what types ofimprovements and research opportunities continue to exist for those interested in being a part of this exciting field.

PRES 4335038

Production of isotopes for medical applications and beyond

***Vanessa A. Sanders**, vsndrs1@aol.com, Dmitri G. Medvedev, Cathy S. Cutler. Isotope Research and Production Department, Brookhaven National Laboratory, Upton, New York, United States*

Research in the Isotope Research and Production Department utilizes the 66-220 MeV incident beam Brookhaven Linac Isotope Producer (BLIP) to produce isotopes that can be subsequently used in various applications. The large incident energy range of the BLIP makes it one of a few facilities in the country with this capability allowing for a plethora of isotopes to be produced. The process of producing an isotope of interest from conception to application has several scientific areas of study that researchers focus on. This includes engineering simulations to properly transfer irradiated materials to shielded hot cells, and simulations to determine the optimal beam conditional to produce a given isotope. This is followed by material science studies to find the most advantageous targets. Then the most integral process of purifying the isotope of interest from the target material through rapid chemical separations. Finally preliminary experiments are carried out to ensure the quality of the final product. The departmental production capabilities with a focus on the Positron Emission Tomography (PET) isotope of interest, arsenic-72 (^{72}As), will be discussed.

PRES 4335690

Radiochemical research in nuclear forensics in support of national security mission

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The threat of nuclear materials or devices escaping regulatory control, as well as the risk of a nuclear detonation represent significant concerns. In these high consequence events, nuclear forensics exists as a means to decipher the fundamental questions: What is it? Where did it come from? How did it get there? Within nuclear forensics, there are two domains, framed in the context of a nuclear device, pre-detonation and post-detonation. The US maintains operational and R&D capabilities that work to improve and address this pressing concern across both domains. In this talk, the broader topic of nuclear forensics will be discussed as it pertains to PNNL, focusing on the post detonation domain. We will highlight the efforts of highly diverse multidisciplinary teams, examining separations and radioanalytical chemistry, and emphasize some highly impactful efforts that support the US nuclear forensics capability.

PRES 4336345

Synthesis and structural characterization of ternary actinide fluorides for novel reprocessing methods

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Nuclear power has become increasingly vital in the pursuit of global carbon neutrality and in supporting industries such as artificial intelligence and other high-tech sectors. Many countries have adopted a closed nuclear fuel cycle to enhance the efficiency and sustainability of nuclear energy. However, current nuclear fuel reprocessing methods largely rely on techniques developed in the 1940s for the production of weapons-grade uranium and plutonium. To address modern demands, innovation in this area is needed to develop separation processes that improve safety, increase proliferation resistance, and minimize secondary waste generation.

This work presents a novel methodology for the separation of actinides from a wide range of fission products and fuel cladding elements found in spent nuclear fuel. Additionally, the structural analysis and characterization of the ternary actinide fluoride materials generated by this separation method are discussed. The proposed separation approach employs a gas-mediated, solid-state reaction to synthesize actinide compounds of the form $(NH_4)_4AnF_8$*. Isolation of these compounds is achieved by leveraging their unique solubility in concentrated ammonium fluoride solutions and subsequent precipitation upon the introduction of an antisolvent. The resulting structure of these simple trans-uranium molecular fluorides is characterized by isolated* $[AnF_8]^{4-}$ *antiprisms, which are connected through hydrogen bonding to four ammonium cations. Based on both experimental findings and theoretical considerations, the proposed separation method demonstrates significant potential as an innovative advancement in nuclear reprocessing technology.*

PRES 4336360

Safeguard and nonproliferation considerations for advanced reactors

***Sandra Bogetic**, sbogetic@utk.edu. Nuclear Engineering Department, University of Tennessee, Knoxville, Tennessee, United States*

International safeguards are a set of legally mandated technical measures used by the International Atomic Energy Agency (IAEA) to verify that countries use their nuclear facilities exclusively for peaceful purposes and do not divert materials for nuclear weapons. In contrast, domestic safeguards and security systems address the risk that non-state actors could steal nuclear material or sabotage nuclear facilities. In the United States, the Nuclear Regulatory Commission (NRC) establishes the regulations and standards for domestic safeguards. For international safeguards, however, the IAEA defines the safeguards approach—such as material control and accounting (MC&A)—and implements verification activities (e.g., surveillance and inspections), with domestic regulators (e.g., the NRC) facilitating access.

The deployment of advanced reactors, alternative fuels, and new fuel cycle facilities introduces novel challenges for detecting, securing, and disposing of nuclear materials while maintaining effective safeguards. Verification and security technologies developed *for previous generations of reactors are not directly applicable to these new systems. Furthermore, the fundamental characteristics of many emerging fuel types remain insufficiently understood.*

The early development stage of next-generation nuclear power reactors presents a unique and timely opportunity to integrate safety, security, and safeguards (3S) considerations into the design phase. This includes predicting and measuring fuel compositions during reactor operation, as well as detecting and diagnosing potential material diversion scenarios. This abstract introduces ongoing research conducted at the University of Tennessee, Knoxville (UTK), in collaboration with partner universities and national laboratories, aimed at mitigating these emerging risks.

PRES 4336363

Oxidation chemistry of uranium and its nuclear waste neighbors

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Uranium is one of the most famous, or perhaps infamous, elements. It is the heaviest naturally occurring element, and its dense nucleus draws the core electrons in close, subtly changing the behaviour of its compounds. This has generated many headaches for scientists dealing with nuclear waste. We will show how a better understanding of the electron and oxygen balance is improving our fundamental understanding of its behavior, and allows us to safely extend these studies to its more radioactive neighbors, neptunium and plutonium. The various oxidized forms of these three metal cations engage in a variety of reactions that we need to understand to help us safely deal with our nuclear waste legacy.

PRES 4336527

Accelerating advances in nuclear and radiochemistry with high performance computing and artificial intelligence

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High Performance Computing (HPC) and Artificial Intelligence (AI) have accelerated advances in many areas of research, including nuclear and radiochemistry, enabling both fundamental discoveries and practical applications. Harnessing HPC's ability to manage large simulations of nuclear processes, from intricate reactor models to radiochemical separations, provides unprecedented insight into radiation effects on materials and chemical behavior. Simultaneously, data-driven AI methods offer predictive analytics that streamline experimental design and accelerate the

development of novel extraction agents and radiopharmaceuticals. This combination of computational power and AI-driven algorithms not only optimizes resource use and deepens understanding of radiochemical phenomena, but also paves the way for safer, more efficient nuclear technologies.
This presentation will highlight achievements demonstrating the synergy of HPC and AI, illustrating how these tools collectively boost accuracy, speed up analyses, and guide innovative solutions in nuclear and radiochemical applications. In addition, emerging trends, such as advanced modeling algorithms and rapidly expanding datasets, will be discussed to show how these technologies can further broaden their impact on nuclear and radiochemistry. By spotlighting ongoing research, this presentation will explore how computational strategies are shaping the future of nuclear science and technology.

PRES 4336529

Consortium for nuclear forensics

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For decades, the United States has led the world in reducing the number and role of nuclear weapons and the development of nuclear forensics capabilities that supports this role. However, the current international climate has diminished recent progress towards reducing both the spread of illicit nuclear technology and the number of nuclear arms. Effective, adaptable, and novel nuclear forensics tools and techniques, and the workforce to implement them are therefore necessary to support U.S. national nonproliferation and security capabilities. Recent assessments of the U.S.'s national technical nuclear forensics capability has stressed the importance of maintaining and expanding the U.S. capabilities in nuclear forensics is important in deterring, preventing, and responding to a nuclear attack, not just domestically but also internationally.
Recently, the University of Florida was selected the lead a 16-university and 7 national laboratory Consortium for Nuclear Forensics (CNF). The CNF's overarching theme is the research and development of enabling fundamental science and engineering for the processing, characterization, determination, and quantification of materials for use in nuclear forensics applications, as well as the human capital development to ensure the United States has the means to maintain a robust nuclear forensics capability. Current nonproliferation, forensics, and intelligence needs require not only a firm grasp of the most advanced techniques and technologies, but also the requisite imagination to develop new emerging, transformational technological forensics capabilities. The next paradigm shift in nuclear forensics capabilities will not simply present itself overnight; it will first require initial identification and shepherding of the research and the future scientists and engineers capable of executing the forensics mission. We will highlight several of the current efforts contained within the Consortium, which includes a broad spectrum of collaborative radiochemistry and materials characterization projects.

PRES 4336970

Linking radioelement speciation, workforce development, and global collaboration in radiochemistry for current and future nuclear technology

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The evolving landscape of nuclear science and technology demands a technically proficient, interdisciplinary workforce capable of addressing national and international challenges in energy, medicine, security, and environmental stewardship. Central to this effort is the ability to characterize and interpret radioelements and nuclear materials through speciation, a critical tool in applications such as targeted alpha therapy, nuclear fuel cycle development, environmental monitoring, and the identification of nuclear security signatures.

Speciation studies, which reveal oxidation states, coordination environments, and molecular-scale interactions, provide essential insights into the behavior of radioelements and fission products in complex systems. Sustaining and advancing these capabilities requires an integrated education and training infrastructure that spans the full academic spectrum, from technical programs to graduate education. Universities serve as central platforms for both workforce development and cutting-edge research in radiochemistry and nuclear science. University consortia have proven especially effective in this domain, linking academic institutions with national laboratories to conduct interdisciplinary research and provide specialized training. Studies involving radioelements and nuclear materials at universities are essential components that drive overall success.

This presentation will examine the intersection of radioelement speciation and nuclear workforce development, emphasizing the role of academic institutions and consortia in advancing research and education. Illustrative examples will include applications in nuclear energy, radiopharmaceutical development, and nuclear security, with particular attention to the importance of national and international collaboration in training the next generation of nuclear scientists and technicians. These efforts directly support the current nuclear enterprise while laying the technical and educational foundation for emerging technologies, including advanced reactor systems, sustainable fuel cycles, and novel applications of radioisotopes in medicine, energy, and industry.

PRES 4336973

Nuclear chemistry: Path towards the future

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Whether you are interested in the legacy waste materials from the US nuclear program, the medical possibilities for diagnosing and treating disease, the fundamental chemistry/radiochemistry of elements, the environmental impacts of humans, or the origin of intercepted nuclear material, there are many possibilities about the future of where nuclear and radiochemistry will lead. At the intersection of most of these difficult and exciting problems is having a workforce ready and willing to research, change policies, and promote the best ideas into the next generation. This presentation will focus on current and future uses of nuclear and radiochemistry to solve real-world problems and determining how we can envision chemists and engineers who are trained to work with radioactivity fulfilling the promise of tomorrow's hospitals, energy production, nuclear forensics, environmental protection, and isotope production/nuclide physics.

PRES 4343726

Materials that matter: The role of complex oxides in nuclear innovation

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The growing energy demands worldwide require a mix of reliable and low-carbon energy technologies with nuclear power playing a key role. A summary of recent highlights in nuclear materials chemistry, particularly in the synthesis and performance of advanced nuclear fuels, as well as new insights regarding spent nuclear fuel disposal to increase the safety and efficiency of the nuclear energy sector, will be presented. Innovative wet-chemical synthesis techniques, especially sol-gel methods, are combined with comprehensive materials characterization to deepen our understanding of structure–property relationships in nuclear fuels and waste forms. These insights enable the design of materials with tailored properties, optimized for specific applications across current and next-generation reactor platforms, including LWRs, SMRs, and HTGRs, while suppressing airborne radioactive particles. Several classes of materials will be discussed, including complex oxides for radioisotope power systems and fusion breeder blanket materials. In addition, the systematic understanding of the role of defects as well as dopants such as Mn and Cr in enhancing the performance and safety of high-performance UO_2 fuel candidates will be discussed. Complementary efforts in nuclear waste management research will also be highlighted.

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