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Exploring known and new high oxidation states in gas-phase complexes of F-elements

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Gas-phase nitrate complexes $M(O)(NO_3)_{n-1}^{—}$ are produced by elimination of NO_2 from $M(NO_3)_n^{—}$, where M is a lanthanide (Ln = Ce, Pr, Nd, Tb) or actinyl (AnO_2 = UO_2, NpO_2, PuO_2). The product has either a $M\text{-}O^{\cdot}$ oxyl or M=O oxo bond, with respectively no change or an increase in M oxidation state. The reactivity of the oxyl/oxo towards hydrogen abstraction from donor gases to convert to M-OH reveals stabilities of Ln^{III} vs. Ln^{IV} and An^{VI} vs. An^{VII}. Formation of $Ln(O)(NO_3)_3^{—}$ from $Ln^{III}(NO_3)_4^{—}$ was expected to oxidize to Ln^{IV} for Ln = Ce and Pr based on Ln^{3+} ionization energies and $Ln^{3+/4+}$ oxidation potentials. The actual result that Pr^{III} is evidently more stable towards oxidation than Tb^{III} is in apparent discord with ionization energies but in accord with oxidation potentials, which reflects that bond formation is not well represented by atomic or solution properties. Formation of $AnO_2(O)(NO)_2^{—}$ from $An^{VI}O_2(NO_3)_3^{—}$ evidently results in oxidation to Np^{VII} and intermediate $Pu^{VI/VII}$, which reveals decreasing stability of the heptavalent oxidation state beyond Np. The utility of this approach for probing known and new high oxidation states is being extended to other f-elements.

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Gas-phase hydrogen abstraction and oxygen exchange reactions with lanthanide and actinide $M(O)(NO_3)_3^-$ complexes

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Fundamental gas-phase reactions of lanthanide and actinide complexes better define the chemistry of these elements. Gas-phase studies provide uniquely detailed information on essential chemical properties that influence oxidation state accessibility and reactivity; characteristics often not directly evident from condensed-phase reactions. $M(O)(NO_3)_3^-$ complexes have a metal-oxygen bond that can allow the metal center to exhibit an oxidation state of +III for an oxyl ($M\text{-}O^{\cdot}$), +IV for an oxo (M=O), or intermediate +III/IV for a hybrid oxyl/oxo. For metals with accessible +III and +IV oxidation states—e.g., M = Ce, Pr, Nd, Tb, Np, Pu, Am and Cm—the nature of the complex elucidates +III/IV relative stabilities and reactivities. Reactions (1a), (1b) and (2), in which the hydroxide oxidation states are considered well-defined, are being studied for lanthanides and actinides, by experiment and theory, to provide insights into the reactants, products and intermediates.

$M(O)(NO_3)_3^- + H_2O \text{-->} M^{III}(OH)(NO_3)_3^- + OH$ (1a)

--> $M^{IV}(OH)_2(NO_3)_3^-$ (1b)
$M(O)(NO_3)_3^- + H_2{}^{18}O \rightarrow M({}^{18}O)(NO_3)_3^- + H_2O$ (2)

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Exploration of electronic structures of heavy actinides utilizing dipicolinic acid derivatives

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Trivalent lanthanides and actinides have similar chemical and physical characteristics which pose challenges for advancement of the nuclear fuel cycle, waste remediation, and long-term storage. The separation of trivalent lanthanides from trivalent actinides has importance, however the separation of americium from curium is crucial to change the methods of the nuclear fuel cycle. To tackle the intra-series separations, the exploration of the fundamental chemistry of the heavier actinides (Am-Cf) must be understood. Periodic trends of the actinide series have not been established due to the limited availability and hazard of working with transuranics. Recent studies have explored ligand systems with the trivalent actinides from Am-Cf and found increasing 5f orbital participation in bonding with the 2p ligand orbitals resulting in covalent bonding. Dipicolinic acid (DPA) is a well-studied ligand and useful for probing the fundamental properties of the heavier actinides. Due to the open functionalization points on the DPA pyridine ring, the ligand can be modified to change its electronic structure. Various DPA derivatives were synthesized with modifications to the para position to study how electronic manipulation of the DPA pyridine impacts interactions with heavier actinides. The derivatives were chosen for their electron withdrawing or donating character in the para position include the chloro, bromo, hydroxymethyl, methyl piperazine, morpholine, and trimethylamine substituents. Partitioning studies have been performed with Eu-152 as a model for heavier actinides, Am-Cf. This study has been extended into the heavier actinides to determine separation factors and thermodynamic properties of complexation for each metal. Coupled with computational methods, studies revealed stronger bonding energy with the heavier actinides with substituents with more electron donating character.

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Survey of radioactive content in American negative ion wristbands via simple analysis

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The use of radioactive material to make dubious health claims has a long storied history in the United States yet, while the days of patent medicine are long behind us, pseudoscience still flourishes in the american market place. The latest craze in

radioactive quackery is the "negative ion" wristband. These wristbands have numerous unsupported health claims that are said to be responsible from the negative ions they emitt. While the United States Nuclear Regulatory Committe (USNRC) has known about these bands since 2014, very little information exists about their dosage and nuclide identity. This work attempts to fill in the gap by analyzing seven commercially available "negative ion" bands with a Geiger-Müller counter and gamma-ray spectrometer. Three out of the seven bands contained Th-232 with dosages of 0.95 ± 0.04 to 2.45 ± 0.09 μSv/h.

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Characterization of thiolate self assembled monolayers on gold coated silicon chips for future detection of radioactive isotopes

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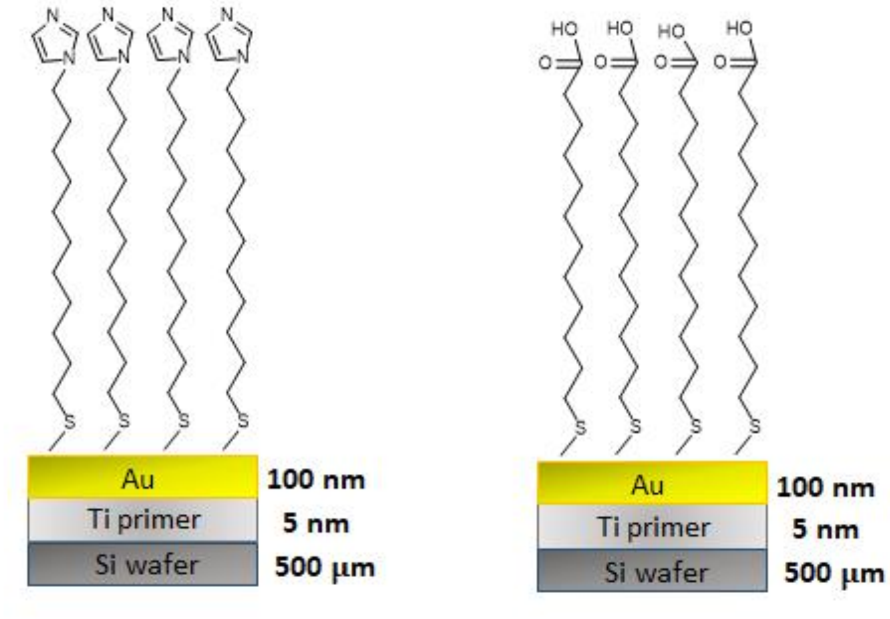
During the last decade, a new approach to the studying of the chemical properties of superheavy elements was developed. It implies coating silicon detectors with gold and, subsequently, determining the standard adsorption enthalpy of superheavy elements on gold surfaces. Element 113, Nh, showed an enhanced reactivity towards gold, which disabled the proper chemical characterization of this element. As a result, a need for new chemically modified gold-coated silicon detectors has emerged. Detectors of such kind will selectively bind the atoms of interest but with weaker interactions.

Meitnerium is expected to be a member of Group IX of the periodic table, and new chemical research is needed to confirm this classification. Initially, the self-assembly of commercially available thiols has been studied on gold-coated silicon chips that mimic functionalized detectors. The main purpose of such devices is to capture radioactive isotopes of iridium and rhodium (meitnerium’s lighter homologs) during online cyclotron-based experiments.

The functionalized chips were characterized via atomic force microscopy, cluster secondary-ion mass spectrometry, X-ray photoelectron spectroscopy, ellipsometry, and neutron activation analysis**.** The dependency of metal adsorption on hydrochloric acid concentration is planned to be studied in the upcoming months.

This talk will include the newest online and offline results of the performed experiments.



Schematic diagrams of the substrates investigated in this study.

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Comparative study of clay materials for the sequestration of technetium

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Nuclear waste repository designs require implementation of backfill buffer materials for containment of radionuclides. Technetium, a fission product environmentally mobile as TcO_4^-, is of concern given the long-lived nature of ^{99}Tc. Commercially available functionalized clays have previously been investigated for the immobilization of Tc under conditions found at the Hanford Site (Washington State, U.S.A). The current work focuses on intercalation of clay with metal, alkylammonium and thiol compounds for the design of potential backfill buffer materials for Tc sequestration. The comparison of synthesized materials and commercially available products will be discussed.

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Optical spectra and proposed crystal structure of ε-UO_3

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By calcining U_3O_8 in O_3(g), ε-UO_3 was synthesized and then identified using powder X-ray diffraction. Infrared spectra collected for ε-UO_3 are in good agreement with previously published results, and low energy (600–200 cm^{-1}) infrared data are provided. For the first time, Raman spectra collected for ε-UO_3 using both 785 and 532 nm excitation wavelengths are presented, and a novel method for analyzing spectral similarity and sample homogeneity is described. A plausible structure for ε-UO_3 was determined via Rietveld refinement of powder X-ray diffraction data and is triclinic, *P-1*, with $a = 4.01$, $b = 3.85$, $c = 4.18$ Å, $\alpha = 98.22$, $\beta = 90.33$, and $\gamma = 120.44°$ ($R_{wp} = 9.00\%$).

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Improving efficacy of a portable LIBS device through implementation of machine learning regression methods

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Ensemble regression techniques and support vector machines (SVMs) were implemented to analyze optical emission spectra of laser-induced plasma from plutonium alloy samples. Predictive regression models were built using the aforementioned techniques to determine quantity of trace Ni and Fe in the Pu matrix from spectral emission intensities. These machine learning based models significantly outperform traditional chemometric models used for analytical spectroscopy, such as artificial neural networks (ANN) and principal components analysis (PCA). The ensemble and SVM regressions yielded limits of detection (LoDs) in the 50-70 parts-per-million (ppm) range for Ni and Fe, an order of magnitude improvement over ANN and PCA models. The results of this study indicate that machine learning paradigms seldom used in analytical spectroscopy could potentially yield promising solutions to complex spectroscopic problems, particularly for analysis of actinide elements. When used in tandem with a portable laser-induced breakdown spectroscopy (LIBS) device, these methods can provide instantaneous, *in-situ* quantification of trace elements in plutonium alloys.

NUCL

High resolution actinide speciation using tensioned metastable fluid detector (TMFD)

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Numerous alpha-emitting isotopes emit alpha particles with energies not resolvable (<20 keV) with solid-state alpha spectrometers. Radiochemical separations followed by

alpha spectrometry can be used to resolve isotopes with overlapping alpha energies such as Pu-238&Am-241, Np-237&U-234 and Cm-242&Cf-252. Mass spectrometry is required to resolve elements with isotopes with overlapping energies such as U-235&U-236, U-233&U-234, Pu-239&Pu-240, and Cm-243&Cm-244. No field deployable systems exist capable of resolving any of these pairs with unresolvable energies. In the nuclear forensics world, there is value in having the capability in the field of measuring any of these pairs. The goal of this project was to establish whether a device based on Tensioned Metastable Fluid Detector (TMFD) technology developed by Purdue University can solve problems such as these. Additionally, the device has the potential to measure low levels of actinides with detection limits superior to those of solid-state alpha spectrometry. Those detection limits combined with the potential high-resolution capabilities could allow this device to assay numerous shorter-lived alpha-emitting isotopes to levels lower than that achievable by mass spectrometry. The results of this research to date will be discussed.

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Additive manufacturing of acrylate and polysiloxane based multi-material scintillator systems

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Additive manufacturing (AM) is a novel method for the rapid fabrication of organic scintillator materials, which are typically made through a lengthy bulk polymerization process. Despite the advantages conferred through a speedy fabrication, scintillators fabricated by AM typically suffer from reduced light output when compared to their commercial counterparts. There are also few applications proposed to take advantage of the unique scintillator geometries afforded by AM. Architected multimaterial scintillator systems (AMSS) are a new class of scintillator systems that utilize additive manufacturing to create multimaterial parts with architectures designed to encode properties of radiation interacting with the scintillator. Microscopic zones of alternating scintillator dyes are sensitive to the recoil tracks of gamma and neutron radiation, allowing for particle discrimination and directional detection. Smooth multimaterial gradients can enable precise position resolution. These capabilities require development of various AM scintillator feedstocks that optimize light output and printability. In this research, various acrylate- and polysiloxane-based materials are developed for AMSS fabrication by stereolithography (SLA) and direct ink-write (DIW) methods, respectively. Both approaches replace poly(vinyl toluene) as the organic matrix in traditional scintillator systems, with various approaches in each material subset used to maximize light output, transparency, and energy transfer while maintaining printability. Characterization of print parameters is crucial; critical dose and cure depth in SLA is highly affected by competition for photons in photopolymerizable resins with similarly absorbing scintillation dyes and photoinitiators, while controlling thixotropy and

shear-thinning properties are crucial for geometric fidelity in DIW. Control and optimization of material parameters related to scintillation and printing are then partnered with various *in-situ* and post-print techniques to create multimaterial parts, including infilling of interstitial spaces, co-printing, and active-mixing printing.

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Studies of uranium hexafluoride interactions with organic self-assembled monolayer surfaces in a low-to-high vacuum environment.

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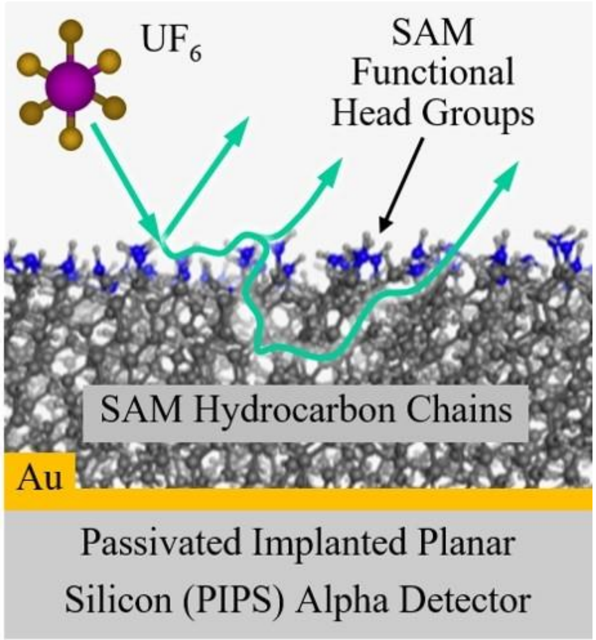
We summarize a computational and experimental collaboration between the University of Memphis and Argonne National Laboratory to study UF_6 interactions on self-assembled monolayer (SAM) surfaces. The program aim was to investigate whether Passivated Implanted Planar Silicon (PIPS) alpha particle detectors could be functionalized to allow direct detection of UF_6 enrichment ratios from process streams. It is possible that SAMs on gold could be used as effective passivation layers for PIPS detectors used in UF_6 detection, if judicious choice of functional group in the organic layer is made. Fluorinated surfaces (long chain perfluorocarbon SAMs and AlF_3) are the best choices of examined surfaces. Beyond the technical determination, much fundamental chemistry is revealed about UF_6 reactions with organic surfaces. Computational efforts include quantum chemical calculations to screen potential functional groups, molecular dynamics simulations of UF_6 interacting with various SAMs, and alpha spectrometry (Monte Carlo) simulations to establish that sufficient signal ratios could be achieved with functionalized PIPS detectors. Analytical intermolecular potential energy surfaces were developed between UF_6 and the best of >40 candidate groups. Experiments required constructing a custom exposure chamber operating from atmosphere to high-vacuum pressures. SAM surfaces are installed within the chamber, and UF_6 pressure is controlled above the surface. Surface changes from UF_6 exposure are tracked with Reflection-Absorption InfraRed Spectroscopy (RAIRS) and Quartz Crystal Microbalance (QCM). Results of exposure experiments reveal details about the kinetics and mechanism of UF_6 reactions with hydrocarbon, fluorocarbon, and amine-functionalized organic surfaces. Functionalized PIPS detectors were also used in studying interactions of UF_6 with AlF_3 surfaces.



Possible UF_6 interactions with functionalized organic surfaces

NUCL

Using microfluidics to study spent fuel corrosion chemistry

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Spent fuel storage in engineered barrier system (EBS) presents a multi-faceted technical challenge. The oxidation of uranium oxide (UO_2) spent fuel could happen from radiolysis of water in a reducing environment in the EBS. The EBS environment is expected to limit oxidation through the production of H_2 gas from anoxic corrosion of iron. The mixed potential model (MPM) is developed to describe the complex processes of spent fuel corrosion. Measurements of electrochemical and diffusion processes that take place during oxidative dissolution of the spent UO_2 fuel are crucial to validate the MPM. We have developed versatile microfluidic electrochemical cells that incorporate UO_2 particles as the working electrode (WE) to study the interfacial corrosion phenomena at the nanoscale using multiple imaging techniques. A thin layer of proton exchange Nafion membrane is fabricated and used to protect the UO_2 particles. During microfluidic electrochemical cell development and performance validation, the UO_2 analogue cerium oxide (CeO_2) particles have been used to reduce nuclear waste generation. Multiple techniques have been demonstrated to deposit or attach nano or micrometer-sized particles as WE and platinum wires as the counter electrode (CE) and reference electrode (RE), respectively. Device reproducibility is verified. The same is true with the microfluidic device electrochemical performance reproducibility. A

multitude of imaging techniques, including atomic force microscopy (AFM), scanning electron microscopy (SEM), SEM/Focused Ion Beam (SEM/FIB), transmission electron microscopy (TEM), and time-of-flight secondary ion mass spectrometry (ToF-SIMS), are used to characterize the pristine and corroded electrode surfaces. Our findings show that microfluidic devices can be used to study UO_2 spent fuel chemistry. This new approach offers the advantage of reduced alpha effects, low consumption of nuclear materials, and transferrability to multimodal characterization platforms, yet maintaining the sensitivity and selectivity of UO_2 redox electrochemistry as in the bulk approach. We anticipate that this new solution will expand in operando electrospectroscopy and microscopy for nuclear material research.

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To what extent does francium follow group 1 periodic trends?

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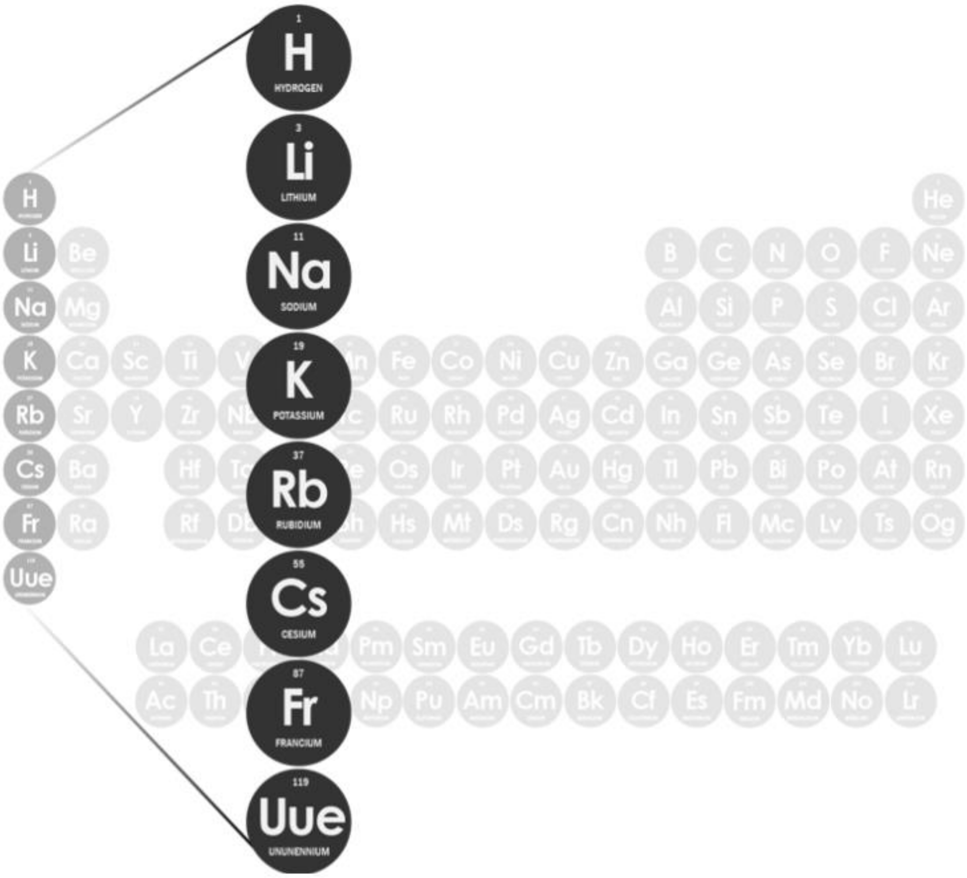
While periodic trends have been used to approximate the reactivity of francium, the largest group 1 element (Z = 87), there is little experimental data to confirm its theoretical properties. The chemistry of francium is largely unexplored due to the element’s short half-life across all isotopes and the time required to reach thermodynamic equilibrium. The experiments presented here use separated Fr-223 (t1/2 = 21.8 min) isolated from a pure Ac-227 source at Lawrence Berkeley National Laboratory, while previous aqueous Fr experiments have been performed with Fr-221 (t1/2 = 5 min) produced from Ac-225 in situ. (Haverlock, 2003). Understanding francium’s chemical behavior may aid in calculating the properties of the undiscovered element 119, which is expected to be the next Group 1 element. Therefore, the solution thermodynamic properties of francium were investigated and compared to those of cesium, the second largest known Group 1 element. Calixarenes and crown ethers, experimentally shown to be effective cesium chelators, were used to perform solvent extractions with Fr-223 and Cs-137 and relative binding affinities were evaluated. We are investigating the unsubstituted molecules to probe the extent to which chelation is size-dependent; future work investigating the impact of increasingly complex moieties is expected.



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Producing isotopes with mA beam currents: The Applications of the IsoDAR cyclotron

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The IsoDAR cyclotron is a high current accelerator which will be capable of delivering a 10 mA proton beam at 60 MeV. While originally intended for a neutrino experiment, the high current produced by the cyclotron makes it a great candidate for producing large quantities of rare medical isotopes and high power target development.The cyclotron accelerates H2+, rather than H- like most commercially available cyclotrons. This gives advantages by lowering space charge as well as allowing the beam to be easily split into multiple beams with adjustable power. With only minor tuning, the IsoDAR cyclotron is also able to accelerate other ions with a charge to mass ratio of 2 to energies of 60 MeV/amu. Many of these have commercially available ion sources, such as alpha particles, and C6 +. Using these additional beam types opens up a path to even more interesting isotope production capabilities.

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Exploration of Pu and Am extraction studies with TTA/Xylene

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Separations chemistry is an essential area of research that support numerous missions at Los Alamos National Laboratory. The focus of this effort seeks to improve and understand a fundamental chemistry that will support the 30 pit per year mission as well as numerous other mission in the Plutonium Facility. The studies of the complexation of 2-thenoyltrifluoroacetonate, TTA, with plutonium and americium are on-going. The distribution coefficients over a wide range of acid and ligand concentrations will be determined using alpha spectrometry and gamma spectroscopy. Batch studies are being carried out contacting ligand and
americium and/or plutonium to determine the best conditions under which to separate the two using this system. These studies will support additional and future studies to replace this system with a more green chemistry approach that will maintain, or improve, the separation efficiency to support these critical efforts. All studies are carried out at room temperature or approximately 25 degrees C.

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^{18}F-SuFEx radiochemistry in hydrous organic solvent systems

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Recent progress in the application of sulfur (VI) fluoride exchange (SuFEx) chemistry has introduced fluorosulfuryl functionality that is being avidly exploited in the pharmaceutical and materials science fields. However, SuFEx typically requires the use of gaseous sulfuryl fluoride (SO_2F_2), which limits its translation to ^{18}F-radiochemistry. Due to the growing interest in the development of ^{18}F-labeled tracers as a versatile tool in positron emission tomography (PET) imaging, an efficient method of ^{18}F incorporation to achieve the desired chemical functionality offers new possibilities for image-guided drug discovery. It is well-known that the nucleophilicity of ^{18}F is adversely affected by hydration, so the time-consuming process of azeotropic-drying is routinely employed to remove water during radiofluorination. Owing to the short half-live ($t_{1/2}$ = 109.7 min.) of ^{18}F, reducing the radiolabeling time would be extremely beneficial for downstream imaging studies. To this end, we have developed a method for introducing ^{18}F ions to sulfonyl cores in a hydrous organic solvent system that removes the necessity for the

azeotropic-drying routine. Various aryl [^{18}F]fluorosulfates and sulfamoyl [^{18}F]fluorides were isotopically activated with cyclotron-produced ^{18}F ions in moderate-to-good radiochemical yields. Unlike aryl [^{18}F]fluorosulfates that can be accessed through their corresponding phenols, producing sulfamoyl [^{18}F]fluorides through their corresponding amines is directly inaccessible. Hence, ^{18}F/^{19}F isotopic exchange was employed to produce sulfamoyl [^{18}F]fluorides in a hydrous organic solvent. This labeling protocol provides an efficient route for incorporating ^{18}F into sulfur-oxygen and sulfur-nitrogen frameworks, thereby obviating the need for tedious azeotropic dying for rapid radiofluorination. This radiofluorosulfurylation approach in hydrous organic media provides an expedient way to access new ^{18}F-labeled SuFEx compounds for potential use in PET imaging.

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Immobilization of copper hexacyanoferrate in polyethyleneimine-reduced graphene oxide membrane for effective removal of cesium ions

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Due to the concerns of the potential release of radioactive materials, rapid and complete decontamination techniques for radionuclides are required to reduce the environmental damage and minimize the amount of waste volume. In this work, we demonstrate a novel method to fabricate a composite membrane, composed of potassium copper hexacyanoferrate (KCuHCF) and graphene-based support, for the efficient removal of cesium ions. KCuHCF has been extensively used for cesium removal due to its high selectivity for cesium, but it is required to fix the KCuHCF particles to supporting material for the actual use, since they are highly dispersible in aqueous condition and have small particle size. There were attempts to enlarge the adsorbent size by immobilizing in polymer beads, but they have been tested mainly in batch conditions. Thus, for rapid treatment of wastewater containing cesium ions, we introduce an adsorptive membrane fabricated by in-situ growth of KCuHCF in a graphene-based membrane. The supporting membrane was first prepared by grafting polyethyleneimine (PEI) to the graphene oxide to provide chelating functional groups for subsequent synthesis of KCuHCF. Copper precursor of KCuHCF, was first coordinated in the amine groups of the composite membrane, and then KCuHCF particles were formed by the subsequent reaction with the $[Fe(CN)_6]^{4-}$. As a result, the KCuHCF nanoparticles were uniformly immobilized in the PEI-rGO membrane, and the composite exhibited high water flux (> 500 L m^{-2} h^{-1}) and selective adsorption performance for cesium. Based on the rapid treatment and selective removal properties for cesium ions, it is expected that this adsorptive membrane could be a promising alternative to conventional membrane processes or an adsorption column in the field of radionuclide decontamination.

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Insight into the nature of hydrogen bonding interaction in Purex process stream

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Throughout the nuclear industry, PUREX process is commmonly used for the separation and purification of U and Pu from the nitric acid solution obtained by dissolving irradiated spent fuel. Tributyl phosphate (TBP) is the extractant used in the PUREX process and is known to extract nitric acid and fission products also. Therefore it becomes important to understand the nature of interactions occuring in TBP-HNO_3-H_2O system which facilitates nitric acid extaction into the organic phase. Technetium-99, a high yield fission product is also known to be extraction as a hydrogen bonded complex of pertechnetic acid. In this context, density functional theory calculations were used to understand the complexation behaviour of TBP with water. nitric acid and pertechnetic acid. Geometry optimizations were performed by employing BP86 density functional in conjunction with the triple- ζdef2-TZVP basis sets. The resulting stationary points are characterized as energy minima by performing harmonic vibrational frequencies at the same level. The resolution-of-identity (RI) approximation and Grimme type dispersion correction with Becke-Johnson damping (D3BJ) were applied throughout the calculations.All calculations were performed with ORCA version 4.2.1 quantum chemistry package and NBO version 6.0. An “atoms in molecule” (AIM) analysis has been performed to evaluate the electron density, Laplacian of electron density, delocalization indices and energy density at bond critical points to assess the strength of hydrogen bonding interactions. The calculations were performed using AIMAll package.

TBP-HNO_3 hydrogen bonded complex

TBP-H_2O hydrogen bonded complex

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Electrochemical bioremediation of uranium (VI) using *Geobacter sulfurreducens* on the boron-doped diamond electrode surface

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The development of nuclear science and technology has led to increasing nuclear waste containing uranium. The contamination associated with nuclear activity, especially uranium, is a significant concern for human and environmental health. Due to the high toxicity, radioactivity, and extensive half-life of uranium, it is necessary to find a way to remove it from the environment. One alternative to solve this problem is bioremediation. With this technique, microorganisms can be used to reduce, eliminate, contain, or transform hazardous waste like radionuclides. This research proposes using the bacteria *Geobacter sulfurreducens* to reduce uranium (VI) (U(VI)) to uranium (IV) (U(IV)). The experiment consists in the *G. sulfurreducens'* immobilization using the boron-doped diamond electrodes (BDD) as an electron donor by electrochemical processes. Taking advantage of the bacteria's microbial metabolism process and using U (VI) as an electron acceptor will reduce the ion to U (IV). First, a potential of -500mV (vs. Ag/AgCl) will be applied to immobilize the *G. sulfurreducens'* to the BDD electrode surface. Second, with the application of a potential between -500mV to -700mV (vs. Ag/AgCl), it will expect to reduce the uranium ion and remove it from the media. Preliminary studies of the behavior of BDD electrodes in the presence of U(VI) had been carried out to determine the electrochemical conditions at which the substrate works with this ion. The results found with these studies will show how *G. sulfurreducens'* will improve U(VI) bioremediation through electrochemical processes.

NUCL

Boron loaded organic glass scintillators

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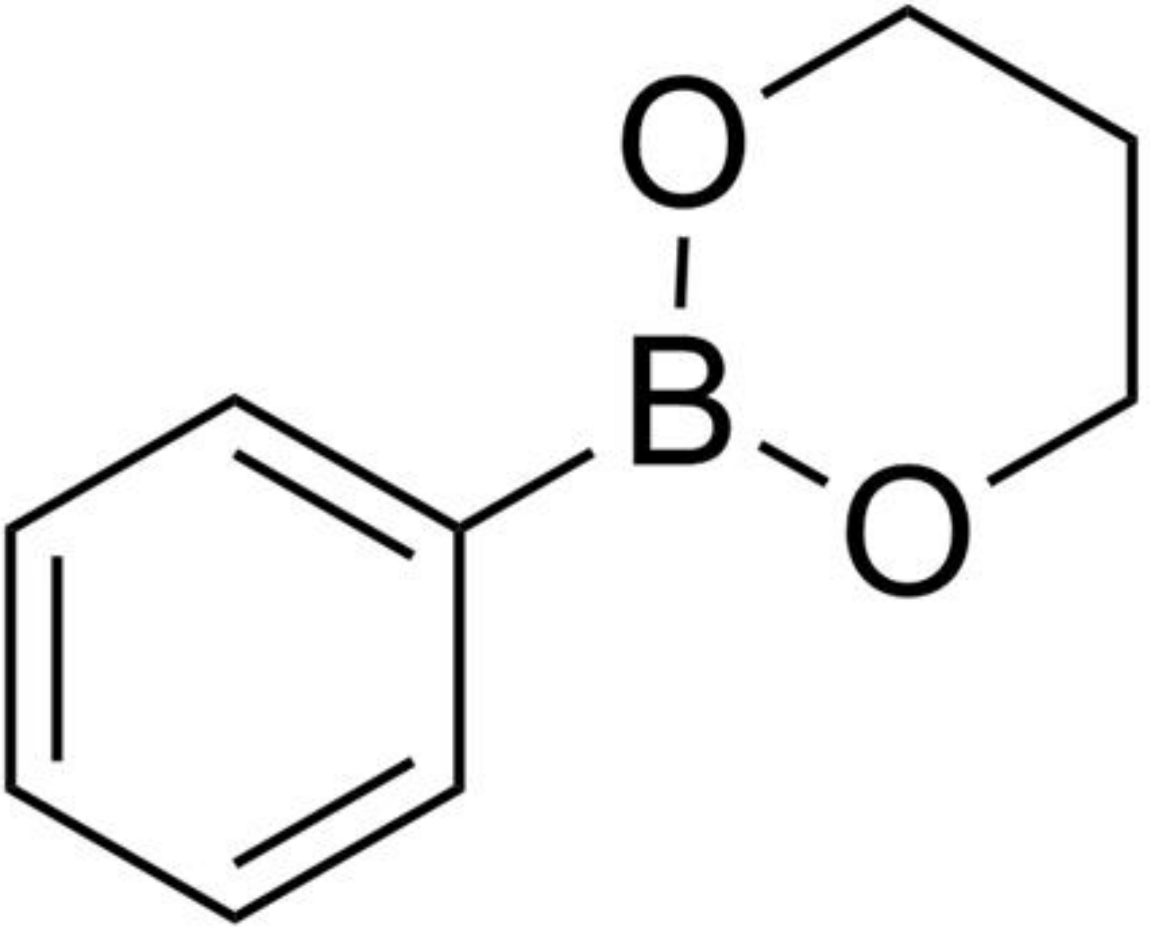
Herein we report the progress towards an organic glass scintillator with fast and thermal neutron sensitivity providing “triple” pulse shape discrimination (PSD) through the inclusion of a boron-incorporated aromatic molecule. The commercially available molecule 2-(p-tolyl)-1,3,2-dioxaborinane (TDB) can be readily synthesized in one step using inexpensive materials and incorporated into the organic glass scintillator at 20%

by weight or 0.25% ^{10}B by mass. In addition, we demonstrate that TDB can be easily scaled up and formulated into organic glass scintillator samples to produce a thermal neutron capture signal with a light yield equivalent to 120.4 ± 3.7 keVee, which is the highest value reported in the literature to date. TDB also modulates the physical hardness of the organic glass scintillator, enabling the casting into more challenging mold materials for high throughput fabrication such as direct casting into pixelated arrays.



NUCL

Selective extraction of organic compounds from radioactive solutions via in-solution solid-phase extraction

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Savannah River National Laboratory facilities characterize radioactively contaminated solutions for an array of compounds, chemicals, and nuclides. In nuclear facilities, organic compound analyses have traditionally been performed via liquid-phase solvent extraction- as hexane and methylene chloride selectively extract organic compounds while leaving active nuclides behind. When considering modern extraction techniques, care must be taken to avoid mechanisms that could concentrate nuclear material and exponentially increase analyst dose rate unnecessarily. Three forms of solid-phase extraction were tested at SRNL for their application to organic extractions from solutions containing radioactive materials: solid-phase disk extractors, solid-phase microextractions (SPME), and stir-bar sorptive extraction (SBSE). The three techniques were compared on dose-rate amplification, method runtime, and analytical parameters such as selectivity, repeatability, limits of detection/quantitation, and accuracy. Application of solid-phase extraction to the unique challenges faced in nuclear laboratories yielded analytical benefits, but care must be taken to avoid unnecessary dose increase, cross-lab contamination, and generation of additional nuclear waste.

NUCL

State of practice and emerging technologies in nuclear forensic science: A readout from the 6th collaborative materials exercise of the nuclear forensics international technical working group

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The ITWG is an informal association of nuclear forensic practitioners, created in 1996 following a G-8 Summit in Ottawa, Canada. Today, with the participation of international experts representing more than 50 countries and multinational organizations, the ITWG has become the clearing house for developing and promoting nuclear forensic “best practices” worldwide. Many of these “best practices” are developed through the execution of Collaborative Materials Exercises (CMXs) that require participating laboratories to analyze special nuclear materials under realistic seizure scenarios. Past exercises have involved the nuclear forensic analysis of reactor and weapons grade

plutonium fluorides and oxides and depleted, natural, low-enriched, and high-enriched uranium oxides and metals. Several exercises have also incorporated “traditional” forensic evidence requiring examination (e.g., fingerprints, tool markings, fibers) that were comingled or contaminated with radiological materials. The latest and largest of this exercise series, CMX-6, was completed in June of 2019 and included participating laboratories from 22 countries and multinational organizations and 15 law enforcement agencies. Highlights from CMX-6 will be presented, along with a summary of the current state-of-practice and emerging technologies in nuclear forensics as demonstrated over 20+ year history of CMXs.

NUCL

Nuclear forensics research at Texas A&M University

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This talk will give an overview of nuclear forensics research at Texas A&M University, including major research goals, facilities, and future plans. In recent years, our work has focused on developing a methodology for identify the reactor type, burnup, and time since irradiation of uranium irradiated in reactors that are common outside the United States. Using a combination of MCNP6 simulations of each reactor type and radioanalytical measurements, a maximum-likelihood method has been developed that has been able to correctly quantify the properties of two different irradiated samples of UO_2 irradiated in pseudo-fast and thermal neutron environments. An additional future irradiation is also planned. We have also conducted a forensic analysis of a highly enriched sampled of ^{239}Pu. This talk will discuss the most recent results and plans for future experimental and computational work.

NUCL

Nuclear forensic analysis to confirm the pedigree of U metal cubes allegedly recovered from Nazi Germany’s nuclear program

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Experimental methods for determining the last chemical process age and geological origin of uranium material is of value to the nuclear nonproliferation and safeguards community. Rare Earth Element (REE) patterns are often used to trace the geological origin of materials sourced from ore bodies. Radiochronometry, a common method used for determining process age of uranium bearing material, involves measuring the in grown ^{230}Th produced from the decay of ^{234}U. An additional uranium decay product pair radiochronometer that is less often employed involves measuring the in grown ^{231}Pa produced from the decay of ^{235}U. Applying more than one radiochronometer to unknown materials is good practice, since concordant results from these chronometers provide a level of confidence that the underlying assumptions of radiochronometry have been met. However, doing so adds to the level of effort required to perform the analysis. An experimental method has been developed that can simultaneously separate Th and Pa from a dissolved uranium metal sample efficiently to provide two independent measures of the uranium material age. This same method can also separate trace level REEs that are characteristic of the geological origin of the uranium. Here we test and demonstrate this experimental method to a set of historic samples allegedly recovered from Nazi Germany's nuclear program during World War, commonly referred to as *Heisenberg Cubes*. Results to date are presented comparing the two chronometers and rare earth element analysis.

NUCL

Characterization of non-critical weapon accident debris

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United States has maintained an advanced nuclear forensics capability to identify the origin, source, and history of nuclear devices in case of a nuclear terrorism attack and other kinds of nuclear detonations. Accidents involving nuclear weapons, always pose a significant risk of allowing particles composed of nuclear materials to enter the environment. While remediation has been conducted numerous times, fuel particles still remain in the soil, providing scientist's samples that can be used to analyze how the fuel changed over time and move through the environment. These particles often differ in characteristics and can be of greatly varying sizes; some are large enough to see with the naked eye and others are so small that they pose a substantial inhalation risk. This work highlights development of a specialized particle mounting technique as well as utilizing NDE techniques such as gamma spectroscopy X-Ray Fluorescence (XRF) and Scanning Electron Microscopy/Energy Dispersive X-Ray Spectroscopy (SEM/EDS) to characterize the non-critical nuclear weapon accident debris of sub-100 micron sizes, which underwent both morphological and elemental characterization. Morphological trends indicated particles across the four evaluated size distributions had similar circularity, and while three major particle classes present based on shape, angularity, and surface features were observed. Elemental analysis indicated the presence of uranium in all the actinide bearing particles and identified trends of other major and trace elements within these particles.

NUCL

Exercise of conventional forensic capabilities within a special nuclear material handling facility

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In 2012, the FBI Laboratory working with Los Alamos National Laboratory began developing a capability to process conventional forensic evidence from items consisting of special nuclear material. While conventional forensic evidence (fingerprints, hairs, fibers, photography, etc.) are commonly collected from items submitted to the FBI Laboratory for examination, until recently there was no way to collect these from items containing significant quantities of special nuclear material within a laboratory setting. In 2020, just prior to the pandemic, forensic examiners from the FBI Laboratory and special nuclear material handlers from Los Alamos participated in an exercise at Los Alamos National Laboratory to demonstrate this capability. During this exercise, using mock special nuclear material items, fingerprints were developed, trace evidence was collected, and photographs were taken under the restrictive processes and procedures required for work around and with special nuclear materials and in a manner consistent with the requirements of the FBI Laboratory.

NUCL

X-ray molecular signatures of UO_2 chemical processing for nuclear forensic science

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Theft, diversion, and trafficking of nuclear materials is an ongoing problem that poses significant threats to U.S. and international security. Uranium oxides are used as fuel material in reactors and are the most common illicitly trafficked form of uranium. Currently, there are various ways to fabricate UO_2 fuel. Many of the multiple synthesis steps involved can be sources of diverted illicit material, yet each provides chemical signatures that could be used to deduce processing and alteration history. However, while baseline efforts have focused on bulk phase unit cell identification, impurity quantification, and isotope systematics to constrain point of origin, molecular signatures that can provide nuanced and highly valuable information on detailed chemical processing remain underdeveloped. There is an ongoing and urgent need for nuanced signatures and new methods that can further constrain forensic problems. The molecular structures of impurities, present in source material or introduced at successive fuel fabrication steps provide potentially valuable signatures of synthesis methods. Here, we demonstrate the applicability of X-Ray Absorption Spectroscopy (XAS) to decode molecular structures of impurity and host elements in U materials. We

will focus on the molecular signatures of both U and impurities that can be extracted with XAS. We will show the potential of using a combination of XRF microprobe imaging with XAS, as applied to samples from the 5th Collaborative Materials Exercise (CMX) of the ITWG, to determine differences in synthesis processing history and conditions. These results show the value of XAS-determined molecular signatures to infer the process history and will further advance our ability to understand processes by which interdicted materials were fabricated and the facilities of origin.

NUCL

X-ray absorption spectroscopy of plutonium oxides for nuclear forensics

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X-ray absorption spectroscopy (XAS) is an established method for examining the local structure of Pu in PuO_2 materials. The variability in the spectra of PuO_2 reported by several recent studies has been a topic of considerable interest and may be due to a number of factors (e.g. particle size, stoichiometry, and disorder). Our work explores the relationships between XAS spectra and PuO_2 sample history to elucidate possible nuclear forensics signatures. We also utilize XRF microprobe imaging in combination with XAS to provide rapid, nondestructive detection of particles that is augmented by the element-specific molecular information (e.g. oxidation state, local structure) of XAS. Due to the high sensitivity of XAS, the collection of local structure information for impurity or alloying elements such as Ga is also possible. Here, we report the local structure signatures of PuO_2 formed *via* several ambient temperature corrosion or high temperature oxidation routes of Pu metal using both bulk and microprobe methods. Our results indicate that high temperature Pu^0 oxidation and exposure of metal to moisture produce PuO_2 with notable differences in Pu local structure. Additionally, XRF microprobe imaging in combination with XAS provides insight into the distribution and speciation of Ga for different oxidation routes of Pu^0.

NUCL

Tender micro-XRF and full-field microscopy for nuclear safeguards particle analysis

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The current method for preparing particles for nuclear safeguards particle analysis involves dissolving particles in a polymer thin film that is neutron-irradiated to enable fission track location, then removing these particles for electron microscopy and thermal ionization mass spectrometry. The overall process involves a great deal of intensive, manual particle handling and is more time consuming than desired. A collaborative effort between Lawrence Berkeley National Lab (LBNL), Pacific Northwest National Lab (PNNL), and Brookhaven National Lab (BNL) seeks to remedy several of these issues by using a suite of x-ray techniques to perform this analysis without removing those particles from the polymer thin films. Recent experiments performed at the Advanced Light Source (ALS) at LBNL in both the tender x-ray and hard x-ray regimes have made progress towards these objectives. The first effort has focused on evaluating and developing tender x-ray micro-XRF techniques at ALS Beamline 10.3.2 utilizing a specialized sample chamber and specifically improving the XRF capability for lighter elements. The second has exploited the unique optical characteristics of Beamline. 8.3.2 to develop new methodologies for rapid particle location through full-field x-ray microscopy.

NUCL

Application of micro-HERFD-XAS spectrometer to nuclear forensics

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Proliferation of nuclear weapons programs, terrorism, and the potential for theft and diversion of special nuclear materials (i.e. enriched uranium and plutonium) pose grave threats to national security. Detecting released materials and forensically determining not only its phase identity, isotope composition, but also its chemical processing history is crucial to establishing likely points of origin. While established baseline methods are often effective, microscale synchrotron radiation-based x-ray fluorescence (SR-XRF) chemical analyses can provide an additional complementary layer of signature recognition and classification capabilities for micro-particle analysis of nuclear materials. SR-XRF is well suited to forensic analyses of small particles because it is rapid, non-destructive, highly sensitive, has good spatial resolution, and can provide chemical information on the elements that are present when combined with x-ray absorption spectroscopy (XAS).

However, the conventional XAS capability in the near edge region as commonly implemented is often inadequate for the sensitivity required for forensics applications. A higher detail of spectroscopic information is required. The particular power of a newly developed x-ray microprobe beamline at the Stanford Synchrotron Radiation Lightsource at the SLAC National Acceleratory Laboratory will be presented, that combines micro SR-XRF and XAS, integrated with a high energy resolution fluorescence detector crystal analyzer. The combination of these techniques with the highly energy sensitive detector, provides a higher information content of the spectral data. This allows for the rapid location and imaging of particles and provides non-destructive analysis that gives chemical and structural information, as well as the distribution of phases across different particles at the micro-scale. We have used this instrument to develop detailed chemical signatures to provide links between the determination of molecular composition, morphological distribution, and nuclear fuel cycle processes.

NUCL

Additive manufacturing of surrogate debris reference materials

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Reproducible realistic surrogate debris reference materials (SDRMs) are needed to test and validate analysis techniques intended to identify, quantify, and interpret major, minor, and trace elemental and isotopic constituents of radioactive debris from a nuclear detonation. Solid-phase SDRMs with precise elemental and isotopic compositions in desired proportions are needed to fully assess analysis process timelines and accuracy, and to evaluate the utility of in-situ microanalytical techniques. We are developing methodology to fabricate solid-phase SDRMS and microanalytical standards using additive manufacturing techniques. Target analytes were doped into sub-micron sized particles during their synthesis, then particles were washed and suspended in ethanol. The particle suspensions were formed into three-dimensional solid samples (~2 by 2 cm squares with ~0.5 cm thickness) via electrophoretic deposition. The green-body deposits were then calcined and sintered in a controlled atmosphere to form densified glassy reference materials. Trace-element reference materials having a bulk silica matrix were developed by synthesizing feedstock particles in a modified Stöber reaction. The incorporation of over 50 trace elements into Stöber particles and their retention during thermal treatment were studied. Syntheses of trace-element doped iron-oxide, aluminum-oxide, and tri-calcium phosphate particles are under development to enable silicate reference materials with customizable bulk and trace elemental compositions and desired spatial distribution with µm resolution.

NUCL

Benchmarking thermal approximations for actinide compounds

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Density-functional theory (DFT) is a powerful tool for predicting the stability of nuclear waste forms. However, many waste forms are metastable and synthesized at high temperatures. Therefore, the 0K nature of DFT calculations can potentially incorrectly predict the stability of synthesized waste forms with respect to competing products. Adding vibrational effects via phonon calculations can improve prediction accuracy but may not be practical for the typical crystal sizes. The Debye method approximates thermal affects via a cell volume and energy relationship, which is orders of magnitudes cheaper to compute than harmonic and quasi-harmonic phonon calculations. This study benchmarks the Debye approximation against harmonic and quasi-harmonic calculations for several small actinide oxides and salts as well as synthesized actinide salt inclusion complexes, demonstrating the methods effectiveness.

NUCL

Characterization of thermophysical and structural properties of trivalent f-element chloride salts with a polarizable ion model

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Advanced nuclear fuel cycles employing molten salts incorporate online pyroprocessing methods for molten fuels and coolants, but lack the chemical knowledge required to manage chemical and neutronic melt properties as well as ensure nuclear security of the systems. Prior to implementation of these online processing techniques, a thorough understanding of salt phase diagrams and coordination environments must be developed. The capability of experimental investigations are limited by the corrosive nature of halide salts, the high temperatures of systems (> 450 °C), and access to relevant radioisotopes. Such challenges necessitate the development of models for predicting salt properties and behavior, especially in the presence of multiple solutes and other contaminants.

This work presents the implementation of both rigid and polarizable force fields for modeling lanthanide and actinide chloride salts. The polarizable force field adapts a previously parameterized point dipole model to a core-shell polarization scheme. The trivalent f-elements cerium and uranium are studied in chloride media at 1100 - 1200 K. Evaluation of structural and transport properties is completed with classical molecular dynamics. Simulation data is supplemented by *ab initio* calculations and experimental data. Preliminary results of the rigid ion model suggest strong ionic interactions resulting in reduced values for diffusion coefficients and enhanced long-range ordering of ion clusters, potentially leading to the development of polymeric networks in the salt. These

networks may include point, edge, and face sharing structures. Rigid model simulations predict M^{3+}-Cl^- coordination numbers of 7.8 and 8.2 for U^{3+} and Ce^{3+}, respectively. The strength of ionic interactions is reduced by the introduction of polarizability to the system.

NUCL

Predicting phase stability of potential actinide-bearing hollandite waste forms using first-principles calculations

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Fission product, cesium-137 with a half-life of approximately 30 years, has been well known as problematic for waste forms. Titanate-based hollandites have long been considered effective phases for sequestering cesium, having the formula $A_{1.33}(Ti^{4+},B)_8O_{16}$, where A is an alkali/alkaline earth element and B is a metal. Due to the beta decay of ^{137}Cs, hollandites must be energetically stable not only to immobilize cesium, but also to remain stable when it is replaced by its decay product, barium. In this study, we have investigated the structural stability of actinide (U, Np, and Pu)-bearing hollandites, that is, when actinides co-exist at B-sites with titanium, to allow their consideration as both a cesium/barium and actinide waste form. The predicted formation enthalpies from density functional theory have provided insight into the stability of these hollandites as a function of composition which are being used to target synthesis efforts.

NUCL

Infrared and Raman spectroscopy of the cerium (III) and uranium (III) chloride species in eutectic mixtures from *Ab initio* molecular dynamics

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Molten salt reactors offer a variety of performance characteristics that benefit the design of the next generation of nuclear reactors for energy production. The new reactor design eliminates the production and disposal of solid fuel, has increased efficiency, and benefits from safer operation methods. To rationally design new fuel compositions and create non-proliferation monitoring capabilities, the speciation within the fuel must be understood and characterization signatures should be developed. Within this work, *ab initio* molecular dynamic (AIMD) simulations are used in the study of cerium (III) and uranium (III) chloride species in LiCl-KCl eutectic mixtures in temperature ranges of 900-1200 K. The local structure was evaluated using radial distribution functions and

network theory while the dynamical behavior was determined using diffusion coefficients, both were compared against prior molecular dynamics and experimental work to confirm the predictability of the methods used. Spectroscopic signatures of the molten salts, both IR and Raman, specifically the sensitivity to different local environments and molecular species were identified.

NUCL

Solute transport mechanisms in liquid/liquid extraction

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Liquid/liquid extraction (LLE) is a standard method of solute separation and purification in the fields of chemistry, engineering, and industry, where a solute distributes from one liquid phase to another based on the favorability of solvation free energies between the two phases. Understanding the mechanism by which simple solute like water, and highly organized ion-pair adducts selectively partition into two immiscible liquids has recently gained attention to enable controlled molecular extraction and design strategies. The experimental elucidation of the solute transport mechanisms face difficulties associated with the probation of biphasic fluidic interfaces and to characterize ion speciation, complexation, and adsorption phenomenon that occurs within the instantaneously evolving, dynamic, and heterogeneous regions of the liquid/liquid interface. The extraction mechanisms of metal ions in solvent extraction involve variations in ion-solvent-extractant adduct formation at the capillary wavefronts and perturbations in free energy of the adduct as it passes from the interface to the organic phase. Moreover, the extraction of water is characterized to occur via the formation of specific surface morphologies like the protrusions, water-fingers, and hinges depending on the choice of extractants and system compositions, via unconstrained molecular dynamics simulations. The presentation will describe the recent developments that are elucidating solute transport mechanisms, for example - water extraction by amphiphilic extractant like tri-butyl phosphate that involves protrusion formation, and octanol transport of water via molecular hinges. Of specific interest is the extent to which the mechanisms that transport water across a phase boundary are extensible to larger solutes like ion-pairs or metal-ligand (ML) complexes under equilibrium conditions.

NUCL

First steps in the chemical evolution of the universe

Michael Wiescher, *wiescher.1@nd.edu. Physics, University of Notre Dame, Notre Dame, Indiana, United States*

The chemical evolution of the universe is dictated by nucleosynthesis processes in multiple generations of stars as the cauldrons of the cosmos. The first generation of stars burn on the ashes of the Big Bang, the primordial abundances of hydrogen, helium

and a bit of lithium. The chemical analysis of the eldest observed stars, however shows a pronounced abundance distribution between carbon, oxygen to to calcium. The reaction chains bridging the mass gap of instability at A=5 and A=8 isotopes, rely on the alpha cluster configuration of light nuclei. These first steps in the chemical evolution of the universe will be discussed on the basis of new experimental data, which provide new insight in the complex and highly dynamic nucleosynthesis environment of first stars.

NUCL

Nuclear physics and the two-neutron star merger

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The neutron star merger event detected in August 2017 along with over 70 electromagnetic transients revealed a spectacular realization for humanity. The fact that this merger of such enormous explosive power shows evidence of r-process nucleosynthesis. There have been other gravitational wave detections resulting from two-neutron star mergers by the LIGO and VIRGO collaborations, however the GW170817 event remains Synthesis of heavy elements all the way to the rare-earth region of the chart of nuclides. The electromagnetic signatures for elemental synthesis went from the visible to the Infrared as it reached the rare earth metals. While it is expected that the process continued to the actinide region of nuclei, direct evidence is missing. Questions regarding the path to the actinides, the potential path to the super-heavy nuclei, and the role of fission remain unanswered.

This talk will focus on the possible, the probable, and the observable.

NUCL

The impact parameter and isospin dependence of the disappearance of flow

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The disappearance of collective transverse flow can be related to the nuclear equation of state and in-medium nucleon-nucleon scattering. We will present experimental and theoretical results for Ar+Sc, Ar+Al, Zn+Al, ^{56}Fe+^{56}Fe, ^{58}Ni+^{58}Ni, and ^{58}Mn+^{58}Fe collisions at incident energies ranging from 35 to 155 AMeV. By comparing experimental results for the balance energy to predictions from transport models, we show that the nuclear mean field description must include momentum dependence. The balance energy is shown to depend on the in-medium cross sections. A model-independent verification of this dependence was done experimentally by varying the N/Z of the system while holding all other parameters constant. The balance energy changes with the isospin of the system in qualitative agreement with a transport model incorporating isospin-dependent nucleon-nucleon scatter and a mean field.

NUCL

Current status of the density dependence of the symmetry energy

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The large imbalance in the neutron and proton densities in very neutron-rich systems highlight the role of the nuclear symmetry energy so that it governs many aspects of neutron stars and their mergers as well as the reaction dynamics of heavy ion collisions. Extracting the density dependence of the symmetry energy therefore constitutes an important scientific objective. In this talk I will show the experimentally determined symmetry energy density functions over a range of density from 0.25-1.5r_0 where r_0 is the normal nuclear matter density, I will discuss the effects of recent PREXII ^{208}Pb skin measurements and the symmetry energy constraints recently obtained from studying the charged pion spectral yield ratios at 1.5 r_0 and their implications on neutron star properties.

NUCL

Recent advances in our understanding of multi-nucleon transfer reactions in heavy nuclei

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Multi-nucleon transfer (MNT) reactions are thought to be useful paths for synthesizing new neutron-rich heavy nuclei and as possible paths for synthesizing nuclei near the N=126 shell closure(of interest for studies of r-process nucleosynthesis. We have performed a number of measurements, using Gammasphere, of the yields of various projectile-like fragments (PLFs) and target-like fragments (TLFs) in the reaction of ^{204}Hg with ^{208}Pb, ^{136}Xe + ^{198}Pt, and ^{204}Hg with ^{198}Pt. We have compared our experimental results with predictions of the GRAZING and GRAZING-F models, the di-nuclear systems (DNS) model, the improved quantum molecular dynamics (InQMD) model , the Langevin model of Zagrebaev and Greiner and time-dependent Hartree-Fock (TDHF) calculations. We also report on recent efforts to study the ^{197}Au + ^{192}Os reaction.

NUCL

Evolution of heavy-ion reaction studies: Historical perspective

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Nuclear chemists have been a key part of the study of nuclear reactions ever since the discovery of fission. As the energies of accelerators increased and their capabilities started to include the acceleration of ever more complex nuclei, the field of heavy-ion reaction mechanisms began to flourish, starting with heavy ion fusion-fission measurements. These then evolved to deep inelastic reactions were the two heavy nuclei never really fuse but reseparate after exchanging charge and mass. As energy increased further to what is termed intermediate reactions and fragmentation of the interacting nuclei, a nuclei phase diagram emerged. This culminates in the creation of a new state of matter termed the quark-gluon plasma in very high energy collisions of heavy ions. Here the constituent protons and neutrons have dissolved into their component quarks and gluons. One of the current frontiers is the study of this matter and identifying the phase transition leading to the quark-gluon state. Recent results from both RHIC at Brookhaven National Lab and the CERN LHC will be presented.

NUCL

30 years of participation of undergraduates in nuclear research at UTEP

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Undergraduate students began participating in research at the University of Texas at El Paso over thirty years ago; many of these studies have been in nuclear science, and some in collaboration with the REU program of the TAMU Cyclotron. Several students have received awards and fellowships, and many have pursued graduate degrees and are now professionals in the field. In this talk I present a summary of these activities.

NUCL

From graduate school to a career at a national laboratory

***August Keksis**, august1909@hotmail.com. Chemistry, LANL, Los Alamos National Laboratory, Los Alamos, NM, US, academic/govt, Los Alamos, New Mexico, United States*

I will discuss my work as a graduate student with Sherry Yennello and how those experiences prepared me for my current areas of research at a national laboratory. Two of the main areas that will be discussed are archival and nuclear data analysis. Also instrumental to my career was learning how to network and work on diverse projects to varying degrees, which Sherry helped pave the way for my success by providing opportunities throughout my graduate studies.

NUCL

Higher education as a means to diversity

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The demand for non-academic technical expertise in the sciences and in the national security enterprise is greater than ever. The technological lead that the US has experienced over the last few decades has narrowed with peer nations. It is now more critical than ever to encourage individuals pursuing higher education to consider and prepare for careers outside of the traditional academic pipeline. This talk will explore the importance of academic institutions in developing a diverse and holistic educational experience that encourages both classic academic achievements along with complementary skill development that cultivates a meaningful and fulfilling career.

NUCL

Scale up of ^{225}Ac production at Los Alamos National Laboratory

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The Isotope Production Facility (IPF) at Los Alamos National Laboratory is configured to maximize production of high purity and high specific-activity isotopes using an intermediate proton beam generated at the Los Alamos Neutron Science Center (LANSCE) accelerator. With a high current (up to 275 mA) beam of 100 MeV protons incident on a water-cooled stack of three targets, IPF is well suited to produce a variety of radioisotopes for medical, industrial and research applications. Recent focus has been on direct production of ^{225}Ac via irradiation of thorium targets as part of a Tri-Lab collaboration with Oak Ridge and Brookhaven National Laboratories. Progress towards Ci-scale production will be described, as well as efforts to increase availability of ^{134}Ce as an analogue of ^{225}Ac for imaging via positron emission tomography (PET).

NUCL

Advancing research in Texas through experiments in medical isotope science

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Alpha emitting radionuclides with medically relevant half-lives are interesting for treatment of tumors and other diseases because they deposit large amounts of energy close to the location of the radioisotope. Some isotopes of interest are available via low energy protons or reactor routes, but some of the most promising isotopes are inaccessible via these conventional production pathways. Production and supply of these unique isotopes is difficult due to the limited number of facilities with production capabilities, including targetry, accelerated heavy-ion beams of appropriate composition and energy, and radiochemistry expertise to isolate components of interest. The Cyclotron Institute of Texas A&M is capable of providing charged particle beams over a broad range of masses and energies. Many medical isotopes of interest are within reach of the accelerator. A program is being developed to explore production of alpha emitters with medically relevant half-lives. In particular, the 88" cyclotron provides a beam of ^{4}He @ 7.2 MeV/u at an intensity of 10 puA, sufficient to produce ~30 mCi of ^{211}At. Astatine-211 has been produced multiple times, reliability of this process is being improved. Future directions for isotope production at Texas A&M will be discussed.

NUCL

Production and purification of Sc radionuclides from Ti metal and oxide targets

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The use of new radiopharmaceutical agents in nuclear medicine allows for patient-specific strategies that leverages diagnostic techniques to provide effective targeted therapy. This relies on the use of isotope pairs for the development of imaging and therapeutic radionuclides and agents with similar chemistry and pharmacokinetic profiles. In particular, our group has developed cyclotron targetry, purification strategies and radiochemistry for radioactive first row transition metals including ^{43}Sc and ^{47}Sc. The goal of this work is to develop purification strategies and radiochemistry for the development of agents that incorporate the matched-pair radioisotopes of Sc which would guarantee identical chemistries and pharmacological profiles for imaging and therapy studies

The radiometal isotopes of scandium have been of interest for the creation of theranostic strategies using diagnostic and therapeutic radiopharmaceuticals with identical structures. Our group has focused on the use of titanium metal and titanium oxide targets to optimize a robust strategy for the production of the diagnostic isotope, ^{43}Sc and the therapeutic isotope ^{47}Sc via the p,alpha reactions on titanium targets. Recent work has resulted in optimized, robust separation processes with high yields and specific activity. Preliminary studies also provide the groundwork for the development of table complexes with these isotopes for the development of ^{43}Sc and ^{47}Sc radiopharmaceuticals for PET imaging and targeted therapy.

NUCL

Production and imaging of non-standard nuclides for diagnostics and therapy

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Beyond, the traditional PET nuclides such as 11C and 18F, a number of potential PET nuclide can be produced with a medical cyclotron. This talk will review the current status of production of non-standard Positron Emitting nuclides at Washington University, mainly for use in Positron Emission Tomography (PET). Production of nuclides such as 64Cu, 76Br, 89Zr and 124I will be discussed. Utilization of these nuclides is applied to a variety of medical problems for diagnostics and aims at imaging receptors on cancerous cells. Due to the decay characteristics, image quality will varied dependdn on posoton rangeand concimittant emission of additional gamma rays. The imaging capabilities of such nuclides and some applications will be discussed. In the context of radiopharmaceutical therapy (RPT), a radionuclide is systemically delivered in the circulation with the goal of targeting specific receptors or markers on the tumors and delivering radiation to cancer cells while minimizing radiation exposure to normal tissues. Many of radio-therapy nuclides have excellent therapeutic potential with however poor imaging characteristics. This is especially the case of the potent alpha-emitting nuclides such as 223Ra. This limitation has led to the development of therapeutic pairs, where an imaging nuclide is used in conjunction with a therapy nuclide to provide both imaging and therapy potential.

NUCL

Searching for toroidal high-spin isomers in the 7-α breakup of ^{28}Si using FAUST

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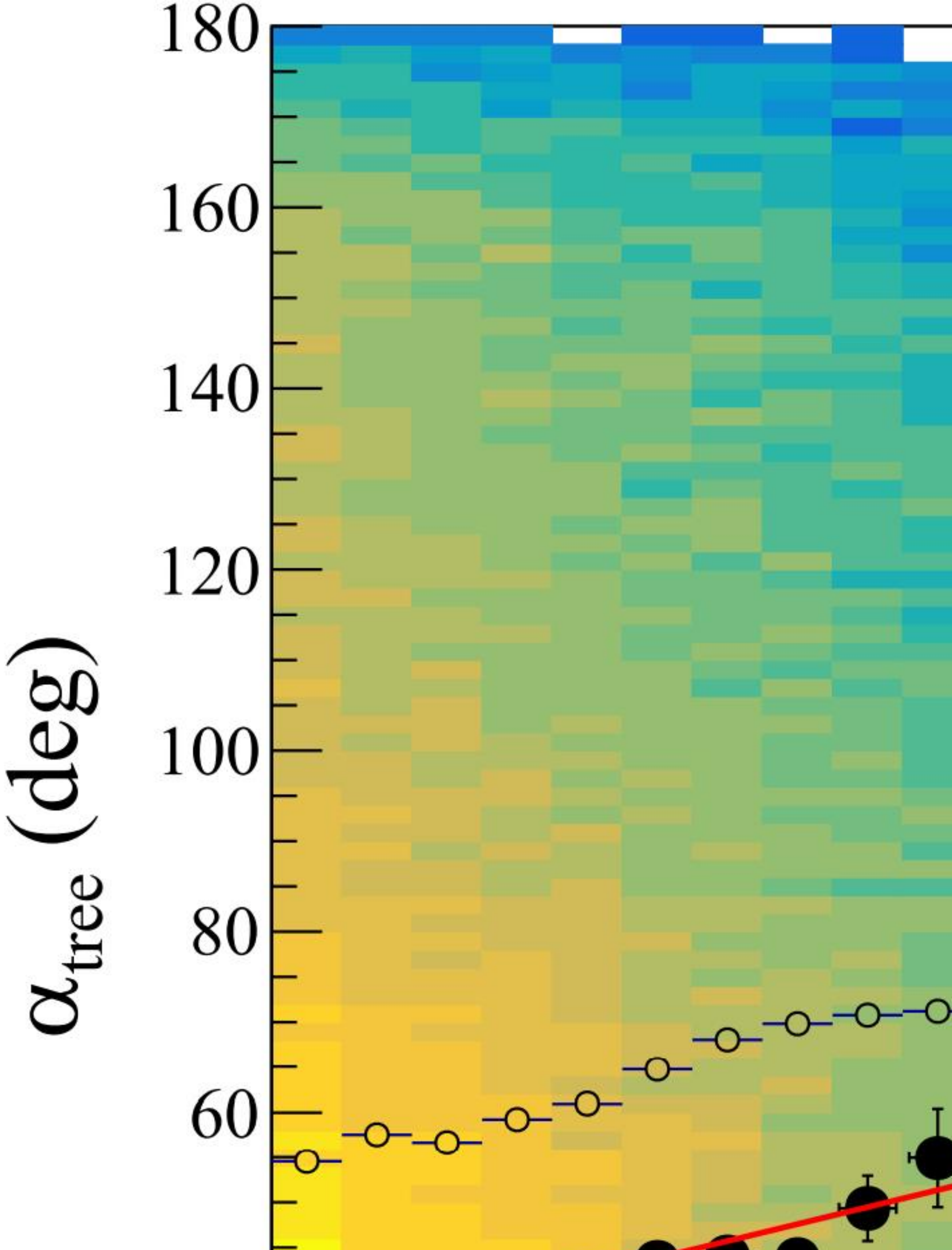
Ground state stable nuclei typically have spherical geometries, however given excitation energy and/or angular momentum, they may exhibit exotic shapes and form clusters of highly stable ^{4}He nuclei (alpha particles) within their bulk. It is predicted that such clustering can promote the production of angular-momentum stabilized toroidal nuclei. By studying the 7-α particle disassembly of ^{28}Si + ^{12}C at 35 MeV/nucleon, a prior experiment performed on the NIMROD detector array observed evidence of high excitation energy peaks in the same range as predicted from toroidal high-spin isomer states. However, the angular resolution of NIMROD is limited by the size of the individual silicon detectors, leading to uncertainties when reconstructing the excitation energy of the fragmenting source. The FAUST detector array uses dual-axis duo-lateral position sensitive silicon detectors capable of sub-1mm position resolution. A precision measurement using FAUST will allow for these peaks to be resolved and provide insight to the α -clustered structure and exotic deformation of nuclei. Experimental design, execution, and preliminary results from this measurement will be discussed.

NUCL

Time dependency of angular alignment in simulated heavy-ion collisions

***Bryan Harvey**, bmharvey@tamu.edu, Mike Youngs, Alan B. McIntosh, Sherry J. Yennello. Texas A&M University System, College Station, Texas, United States*

The Nuclear Equation of State (NEOS) guides the dynamics of heavy-ion collisions and can thus be studied through these dynamics. Neutron-proton equilibration is one process which has been used to study the density dependence of the symmetry energy term in the NEOS. Experimental analysis of the process requires theoretical input regarding the time dependency of the angular alignment, α, an angle used to assess the angle of rotation of a projectile-like fragment after separating from a target nucleus. This study utilizes molecular dynamics simulations of ^{70}Zn + ^{70}Zn with collision energies of 35 and 45 MeV/nucleon to study this correlation. An algorithm is proposed which searches through simulated events and identifies the excited projectile-like fragment (PLF*) after its separates from the target and determines its lifetime, Δt. It also determines the fragments that the PLF* breaks apart into and determines their angular alignment. This technique gives an opportunity to determine how the average contribution of dynamic yield, $<\alpha>_{dyn,}$ evolves with PLF* lifetime. A roughly linear relationship was determined, indicating a correlation with magnitude consistent with classically determined values used for prior experimental results.



180
160
140
120
100
80
60
α_{tree} (deg)

NUCL

Thermodynamics, dynamics, and equilibration

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The nuclear equation of state emerges from the microscopic interactions between nucleons and impacts the properties of finite nuclei (such as the nuclear caloric curve), the properties of neutron stars from crust to core, and nucleosynthetic pathways in the cosmos. By colliding heavy ions in the laboratory, we create nuclear systems far from equilibrium and observe the equilibration process. We investigate how the collision dynamics affects and is affected by neutron-proton equilibration. In particular, through correlations between observables in a wide range of exit channels, we show how the neutron-proton equilibration process can be visualized, using experimental data, like a movie with a frame rate of greater than 2x10^22 frames per second.

NUCL

Nuclear stress test

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Normal materials fail, and “break”, under load, tensile, torque, or sheer stresses. Nuclear matter is extremely well bound if the nucleon density and ratio of neutrons to protons (the asymmetry) are in well-defined regions. Most of the work in nuclear science today is defining how the strong binding devolves (near optimal density) when the asymmetry is not. Many other groups are focusing on when the neutrons are in far excess. This is justified by both astrophysical considerations and the fact that nuclei can tolerate great neutron excesses. We have focused on when protons are in great excess. From a basic nuclear perspective, it is this excess that generates a peak in the binding energy curve, drives fission, and even alpha decay for nuclides above Pb and neutron-deficient rare earths. Our focus has been on the lightest nuclei where this stress induces proton decay. Using invariant-mass techniques we have discovered five new isotopes (11,12O, 13F, 17Na and 18Mg), dozens of new states and refined the properties of many more. This talk will provide highlights of this nuclear stress testing.

NUCL

Using DAPPER to measure photon strength functions via the Oslo Method

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An accurate description of neutron capture is important for modeling stellar lifecycles and can give insight into the production of the elements. The photon strength function (PSF), which describes the likelihood of certain nuclear transitions, is necessary in understanding the neutron capture process. The gamma rays from the decay of ^{60}Fe, an s-process isotope, are seen in the interstellar medium and can constrain stellar evolution models. In addition, an observed enhancement in the PSF at low excitation energies for ^{56}Fe and ^{57}Fe has implications for the reaction rates of neutron-rich isotope production in r-process nucleosynthesis. The Oslo Method was designed to extract both the PSF and the nuclear level density from particle – γ coincidences. For the purpose of measuring the PSF of iron isotopes, the Detector Array for Photons, Protons, and Exotic Residues (DAPPER) has been constructed. The design of the DAPPER array and preliminary results of the analysis of the ^{57}Fe(d,pγ)^{58}Fe reaction via the Oslo Method will be discussed.

NUCL

Photon strength function of ^{60}Fe with DAPPER using the forward method

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^{60}Fe is seen in a variety of places on earth and in space even though it only has a half-life of 2.64 million years. Clearly ^{60}Fe must be produced recently in the galaxy. Since ^{60}Fe has a clear electromagnetic signature associated with it, it can be used to help model the stellar evolution of stars. Improved knowledge of the production reaction, ^{59}Fe(n,γ)^{60}Fe, is needed in order to gain greater insight into these astrophysical observations. The photon strength function (PSF) is important in describing photon emission probabilities and thus it is important in describing this reaction. For some nuclei, experiments have shown an enhancement in the PSF at low energy. This “up-bend” could have a large effect on r-process nucleosynthesis. Experiments have shown an up-bend in both ^{56}Fe and ^{57}Fe nuclei. The PSF of ^{60}Fe will be probed using a ^{59}Fe(d,pγ)^{60}Fe reaction by utilizing the new detector array called DAPPER (Detector Array for Photons, Protons, and Exotic Residues). The Forward analysis method compares modeled cascades with observed to extract the PSF that best fits the experimental results. Preliminary results on the measurement of ^{58}Fe’s photon strength function will be discussed.

NUCL

Investigating alpha cluster structure of ground state nuclei using the NIMROD detector array

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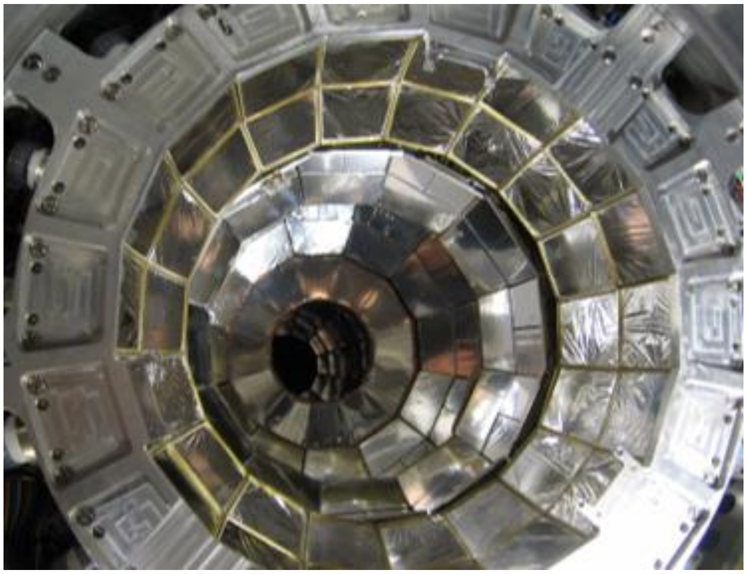
Alpha clusterization is the phenomenon where a certain number of ^{4}He particles can describe the structure of nuclei. At certain excitation energies, these alpha cluster structures tend to dominate as a result of the large binding energy and high excitation energy of ^{4}He. At such excitation energies, the nucleus can be described as a low density, gas-like state consisting of a discrete number of alpha clusters. An example of such a state is the Hoyle state in ^{12}C, which can decay into three alpha particles. However, this type of cluster structure does not exclusively exist in excited states of nuclei. Models calculating the density distributions of alpha conjugate nuclei indicate that some degree of alpha cluster structure coexists with a dominate mean-field type structure at the ground state level and other low-lying states. Such models also suggest that the energy levels of the alpha conjugate nuclei are sensitive to the degree of mixing between the mean-field structure and cluster structure at these states. In order to measure the degree of clusterization at the ground state, we ran a quasifree knockout reaction with a 60 MeV/u ^{4}He beam on various light alpha-conjugate targets. This experiment was performed using the NIMROD detector array (see Figure) so that the detection of quasifree angle pairs is maximized. A general discussion on the experimental search for alpha cluster structure present at the ground state of various alpha conjugate nuclei is presented.



Inside the NIMROD detector array at the Cyclotron Institue at Texas A&M University.

NUCL

Fusion for neutron-rich nuclei around the N=20 and N=28 shell closures

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Fusion in neutron-rich environments is presently a topic of considerable interest. Experiments for an isotopic chain allow systematic exploration of the dependence of fusion on neutron number. In addition, the role of unpaired nucleons on fusion can be studied by measurement of the fusion cross-section near closed shells. Measurements of the fusion cross-section for ^{39,45,47}K+^{16}O and ^{36,44}Ar+^{16}O for E/A = 2-3 MeV/A were conducted at NSCL's ReA3 facility. The measured fusion cross-sections allow to study fusion away from the closed N=20 and N=28 shells and explore the role of the unpaired proton. Details of the experimental technique using E-TOF will be shown. Results yielding the experimental fusion excitation functions and comparison to theoretical models will also be presented.

NUCL

Fusion of neutron-rich light nuclei: The march to the neutron drip-line

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The recent discovery of neutron star mergers as a major nucleosynthetic site emphasizes the importance of nuclear reactions in neutron-rich environments. As fusion of two nuclei involves the interplay of the repulsive Coulomb and attractive nuclear potentials, by examining fusion for an isotopic chain one largely probes the neutron density distribution and how that density distribution evolves as the two nuclei approach and overlap. By examining the average fusion cross-section at energies above the fusion barrier for $^{12-15}$C + ^{12}C and $^{16-19}$O + ^{12}C the impact of neutron excess on fusion dynamics is probed. The experimental results indicate that the fusion cross-section associated with dynamics increases with increasing neutron excess more rapidly than simply the increase in size. Calculations with a time-dependent Hartree-Fock model fail to decribe the observed trend. Proposed experiments to extend these measurements towards the drip line are discussed.

NUCL

Astatine extraction into ketone solvents: Exploring the metal character of At(III)

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Interest in α-emitting radionuclides for applications in Targeted Alpha Therapy (TAT) continues to grow. With its relatively short half-life (~7.2 h) and its rather simple decay scheme, At-211 is one of the most advantageous nuclides for this purpose. However, study of astatine chemistry has been limited due to the practical difficulties of having no long-lived isotopes to investigate. The K150 cyclotron at Texas A&M is capable of producing α-particle beams necessary to make At-211 via the Bi-209 (α,*2n*) reaction. The availability of the short-lived species at Texas A&M provides an avenue to achieve systematic analysis of its chemical properties. Recent findings have shown solvents containing ketone functionality have a strong affinity for At, presumably as trivalent AtO^+, which leads to selective extraction of the At directly out of nitric acid. These findings have stemmed investigation to better understand the interactions occurring between the AtO^+ molecular cation and the ketones, which will be discussed in detail.

NUCL

Exploring the chemistry of Astatine through the thermodynamics of liquid-liquid extraction systems

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Astatine-211 is an isotope that possesses a short half-life of 7.2 hours, a high linear energy transfer, and a quantitative decay by α-emission (41.80% immediate, 58.20% ec). As such, ^{211}At is one of a small number of α-emitting radionuclides whose decay characteristics are well-suited for use in Targeted Alpha Therapy (TAT), especially of metastatic cancers. Moreover, the chemistry of astatine is perhaps the most poorly understood of all naturally occurring elements on the planet, due to its extreme unavailability. Since production quantities limit extracted astatine concentrations to trace amounts, characterization of the element and its species requires thorough understanding of the solvent systems used in its extraction. Studies at Texas A&M are ongoing to understand the thermodynamics of promising solvent systems that preferentially extract ^{211}At out of nitric acid, with an eye also towards understanding astatine speciation and interactions.

NUCL

Non-conventional solvents for astatine and bismuth separation

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Fundamental chemistry of astatine is one of the important topics for Targeted Alpha Therapy which considers ^{211}At to be one of the most promising radionuclides. This element does not have any stable isotopes and as a result radiochemical methods must be applied. Relatively short ~7.2 h half-life and convenient ~1 barn cross section of ^{209}Bi(α, 2n)^{211}At reaction at ~29 MeV beam energy require fast and efficient separation procedure performed at cyclotron facilities. Liquid phase based chemistry usually starts with a target material dissolution in nitric acid, non-favorable conversion to hydrochloric medium, and liquid-liquid extraction into a series of conventional organic solvents. Our group has developed a chemical procedure allowing for efficient extraction of astatine directly from nitric media. However, to the best of our knowledge, there are no studies on astatine behavior in the presence of non-conventional solvents, namely deep eutectic mixtures and ionic liquids. Moreover, the bismuth behavior under the same conditions also has been studied. Thus, consistency and minimization of a dose on personnel dictate development of automated systems to safely perform desired chemical manipulations. Taking into account that our group successfully converted liquid-liquid extraction method to a compact extraction chromatography column, the automation of such a system becomes feasible. These new results and details will be presented.

NUCL

Advancements in Bi targetry for ^{211}at production

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Astatine-211 is an interesting isotope for targeted alpha therapy (TAT) cancer research given its short half-life and simple decay pathway. It is most often produced via the ^{209}Bi(α,2n)^{211}At using a 28.8 MeV alpha beam, which limits the number of potential production facilities to those with a sufficiently energetic and intense alpha beam. The Texas A&M Cyclotron Institute has recently joined the DOE Isotope Program's University Isotope Network, the goal of which is to develop production of this important isotope throughout the country. In order to maximize the production of astatine, it is important that the bismuth targets are optimized for production and well characterized. Astatine-211 is only formed in the first 70 μm of the bismuth, so reducing excess bismuth in beam should significantly help with both the production and separation of

astatine. Building on target-making methods from other institutions, Texas A&M has produced and analyzed targets made via several different methods to study the consistency and durability of the targets.

NUCL

From heavy ion reactions to ion beam applications: New career options in nuclear chemistry

***Graham F. Peaslee**, gpeaslee@nd.edu. Department of Physics, University of Notre Dame, Notre Dame, Indiana, United States*

The field of radio and nuclear chemistry is broad and lies at the intersection between inorganic chemistry, nuclear physics and even materials science in many respects. After a career in fundamental studies in heavy ion reactions, my research program has taken a turn towards both applied nuclear science and environmental science. Still using particle accelerators, it is the applications of nuclear measurement techniqes to questions of societal interest that marks the most significant change in this research, and it is possible to be productive in both fundamental science and applied science - especially in radio and nuclear chemistry. An update on our isotope harvesting efforts together with the applied ion beam analysis efforts for environmental fate and transport of an entire class of chemicals of concern (per- and poly-fluorinated alkyl sustances) will be provided. Future opportunities in these interdisciplinary area for a new generation of radio and nuclear chemists will be discussed as well.

NUCL

Molecular Imaging of HIV persistence: Tools for a cure?

***Henry F. VanBrocklin**, henry.vanbrocklin@ucsf.edu, Timothy Henrich. University of California San Francisco, San Francisco, California, United States*

A major challenge to the eradication of human immunodeficiency virus (HIV) is the presence of HIV infected cells that persist in patients on suppressive antiretroviral therapy (ART). The infected cells reside in anatomical and lymphoid tissues outside of the peripheral circulation that are difficult to access for routine sampling and analysis. Additionally, the anatomical distribution and tissue burden of HIV infection reservoirs is not well known. The objective of this work is to develop PET molecular imaging agents that home to the HIV infected cells to visualize and characterize the whole-body HIV burden. VRC01, a broadly neutralizing antibody developed by the NIH Vaccine Research Center as a potential anti-HIV therapy, targets the CD4 binding sites of the HIV-1 external envelope protein gp120, and has been shown to be safely administered in early phase clinical trials. VRC01, modified with a pendant deferoxamine (DFO) was radiolabeled with positron-emitting zirconium-89 (78h half-life). Under FDA IND and institutional IRB approval, ^{89}Zr-VRC01 was evaluated in viremic and ART suppressed patients and healthy volunteers. Significantly greater ^{89}Zr-VRC01 uptake was noted in

inguinal and axial lymph nodes, nasal-associated lymphoid tissue (NALT) and bone marrow in HIV infected participants. Significant differences were seen in the inguinal lymph nodes and NALT between ART-suppressed patients and healthy controls. Inguinal lymph node biopsy demonstrated the correlation of ^{89}Zr-VRC01 uptake with p24 protein expression, a marker of HIV infection. Overall, these data indicate that HIV envelope-specific PET imaging with ^{89}Zr-VRC01 was able to visualize HIV burden in viremic and ART-suppressed HIV patients. This uptake correlated with HIV protein expression. Thus, HIV-targeted PET imaging has the potential to localize and quantify anatomical HIV reservoirs providing an opportunity to support development of HIV curative strategies.

NUCL

Investigation of signatures of short-range correlations in intermediate energy heavy ion collisions

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Short Range Correlations (SRCs) result from fluctuations when nucleons form pairs with high relative momentum and small center of mass momentum for short periods of time. SRCs are an important aspect in the investigation of large relative momentum and short distance properties of nuclear wave functions. Electron scattering experiments suggest that about 20% of nucleons in nuclei belong to SRC pairs. Particle scattering experiments at high energies also indicate effects of SRCs. In addition, signatures are predicted in collisions of intermediate energy heavy ions. Analysis of energy spectra of light particles from violent collisions in intermediate energy heavy ion reactions will be presented in a search for signatures of SRCs at these energies.

NUCL

Scientists creating equity

***Roxanne Springer**, rps@phy.duke.edu. Physics, Duke University, Durham, North Carolina, United States*

I've known Prof. Yennello for decades, mostly in the context of her achievements and leadership in the American Physical Society. I would like to share with you what I have watched her accomplish, and what I have learned from her about being an effective agent for change. The point of this talk is to demonstrate that we can each learn to implement her techniques and create the culture in our community that will allow science and scientists to thrive. That includes "patiently and persistently" obtaining data to measure equity and inclusion and how it is (or is not) changing under the influence of new policies and programs; persuading institutions to implement best practices and codes of conduct, along with consequences for violating them; and being a personal advocate and "upstander" at every opportunity.

NUCL

Sherry Yennello: Growing a strong, diverse workforce

***Allena Opper**, aopper@nsf.gov. National Science Foundation, National Science Foundation, Alexandria, VA, US, govt, Alexandria, Virginia, United States*

In addition to her many accomplishments in nuclear chemistry, Sherry Yennello's career includes strategic and effective steps to improve the training and mentoring of undergraduate students and to increase diversity in the STEM workforce. This talk will review some of that work and also include some observations of her impact on the field.

NUCL

Synthesizing elements with the weak r process

***Jolie A. Cizewski**, cizewski@physics.rutgers.edu. Rutgers The State University of New Jersey, New Brunswick, New Jersey, United States*

The rapid (r) neutron capture process is responsible for the synthesis of about half of the nuclei heavier than iron. The astrophysical site(s) of this process continue to be an open question. In particular, the synthesis of the elements with atomic mass 80 (near the neutron N=50 shell closure) likely require more than one scenario, often described as a "weak" r process. In 2014 Surman and colleagues [1] analyzed the impact of unknown neutron capture rates in reproducing observed "weak" r-process abundances. Near shell closures, neutron capture could proceed via two processes: direct (semi-direct) (DSD) and via the population and decay of a compound nucleus (CN). While the nuclei important for weak r-process nucleosynthesis are unstable, they can be accelerated to enable reaction studies to inform neutron capture.

Direct neutron capture (and the semi-direct process that proceeds via the giant dipole resonance) has a sensitive dependence on specific low-spin neutron single-particle excitation energies and spectroscopic strengths that can be deduced from neutron transfer reactions such as (d,p). The Oak Ridge Rutgers University Barrel Array (ORRUBA) collaboration has informed the DSD capture on several key A≈80 nuclei. More recently, ORRUBA has been coupled to large arrays of gamma-ray detectors, including GRETINA, to inform CN neutron capture via (d,pγ) reactions.

The present talk will summarize the efforts to inform neutron capture on A≈80 nuclei important to understand weak r-process nucleosynthesis and prospects for informing the competition between DSD and CN capture including with the Facility for Rare Isotope Beams.

NUCL

Isospin effects on the nuclear equation of state: Diffusion and migration

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In this contribution we investigate the nuclear dynamics and the nuclear equation of state (NEOS) by a detailed study of heavy-ion collisions at intermediate energies. Indeed, we explore the isospin transport phenomena occurring during the collision between the projectile and target having different isospin contents. In particular, two observables are investigated the isospin diffusion and isospin migration. The former is related to the nucleon exchange process between the projectile and target; the latest is related to the neutron migration towards the low density region in the neck formed at mid-rapidity. Both observables provide an important information on the density dependence of the symmetry energy term of the NEOS.
We will report on the experiments ^{40,48}Ca+^{40,48}Ca at E/A=35 MeV performed at GANIL, where we coupled the VAMOS spectrometer with the 4π INDRA detector. The use of the high acceptance spectrometer is to measure the isotopic and velocity distributions of the projectile like fragment (PLF) with high resolution, and INDRA is used, to estimate the impact parameter and excitation energy by measuring all charged products emitted in coincidence with PLF.
From these experiments we obtain a set of data that for the first time measure different isospin sensitive observables in the same reaction. In particular a direct detection of the PLF residue in coincidence with an extraction of information about the N/Z of mid-rapidity. The set of data is open to comparison to all transport models engaged to link data to the symmetry energy.

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Prospecting the nuclear landscape

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Following in the footsteps of the early pioneers of nuclear chemistry, such as Irene Curie, one of the contributions of Professor Yennello has been the search for new isotopes of the elements and the use of unusual forms of nuclear matter to understand nuclear chemistry. This talk will review where we stand on the exploration of the nuclear landscape and some of the gems we have extracted from its exploration. Currently, a bit more than 3,000 isotopes have been discovered, but there may be more than 10,000 that could exist in nature. A major advance will come from the new Facility for Rare Isotope Beams, FRIB, which may have the possibility to dramatically expand our knowledge of the limits. With this information, we will have a benchmark to test nuclear models and search for new phenomena that occur in weakly-bound nuclides.

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Radioactive ion beam developments at the cyclotron institute: Texas A&M University

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Cyclotron Institute at Texas A&M University started a project to develop reacceleration of radioactive ions using the two operational cyclotrons and a Charge Breeding ECR ion source. The radioactive ions are produced primarily via (p,n) reactions using the well-known IGISOL technique. The reaction products are transported into a Charge Breeder ECR ion source where their charge state is boosted from 1+ to higher, depending on the product of interest and the operational state of the Charge Breeder ECR ion source. The transport of the products and the injection into the ion source are very important for the efficiency of charge breeding. Presentation of the entire project, the new injection technique, experimental results, and future plans will be discussed.

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COACh for women scientists and engineers: Research results equity in the graduate experiences and career plans in chemistry

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Recent events prompted scientists in the United States and throughout the world to consider how systematic racism affects the scientific enterprise. This paper provides evidence of inequities related to race-ethnicity and gender in graduate school experiences and career plans of PhD students in the top 100 ranked departments in one STEM discipline, chemistry. Mixed-model regression analyses were used to examine factors that might moderate these differences. The results show that graduate students who identified as a member of a racial/ethnic group traditionally underrepresented in chemistry (URM) were significantly less likely than other students to report that their financial support was sufficient to meet their needs. They were also less likely to report having supportive relationships with peers and post-docs. Women, and especially URM women, were significantly less likely to report supportive relationships with advisors. Despite their more negative experiences in graduate school, students who identified as URM expressed greater commitment to finishing their degree and staying in the field. When there was at least one faculty member within their departments who also identified as URM they were also more likely than other students to aspire to a university professorship with an emphasis on research. Men were significantly more likely than women to express strong commitment to finishing the PhD and remaining in chemistry, but this difference was stronger in top ranked departments. Men were also more likely than women to aspire to a professorship with an emphasis on research, and this difference remained when individual and departmental-level variables were controlled.

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Investigating the O-Na anticorrelation through ^{20}Ne(p,γ)^{21}Na at LENA2

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Globular Clusters (GCs) were among the first structures to form in the galaxy and are important for understanding galactic evolution. Within each studied cluster, there exists a measurable oxygen-sodium (O-Na) anticorrelation. Although the exact mechanisms by which this becomes present are still unclear, the neon-sodium (NeNa) cycle has been suggested to contribute to this anomaly in intermediate-mass, thermally pulsing asymptotic giant branch (TP-AGB) stars within the GC. The first and least understood reaction in this cycle is ^{20}Ne(p,γ)^{21}Na. This reaction sets the rate at which the rest of the cycle continues and can impact the O-Na abundances. However, the γ-rays emitted in the reaction have energies that lie directly within the environmental background. Therefore, to improve cross section measurements for this reaction, a position-sensitive, planar Ge detector is currently being developed at the Low Energy Nuclear Astrophysics 2 facility to significantly reduce background events within the experiment.

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Indirect constraints on neutron capture cross sections for nucleosynthesis and stewardship science

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Understanding the nucleosynthesis of heavy elements continues to be a key component of nuclear science. Numerous stellar processes contribute to the nucleosynthesis of elements heavier than iron including the rapid (r), slow (s), and intermediate (i) neutron capture processes. The stellar conditions vary for each process as does the range of

nuclei produced. Accurate prediction of final abundances requires many pieces of nuclear data including neutron-capture cross sections. Direct neutron capture cross section measurements are not feasible since both the neutron and rare isotopes in questions are unstable and cannot be made into targets. Therefore, indirect techniques are necessary to constrain the neutron capture cross section on short-lived nuclei. One such indirect technique is the β-Oslo method in which the nuclear level density (NLD) and γ-strength function (γSF) are extracted following the β-decay of a neutron-rich parent and are used in a statistical reaction model to constrain the neutron capture cross section. The β-Oslo technique has been applied to extract the neutron capture cross sections of many neutron-rich nuclei from A ~50-100. In addition, the same technique and, in some cases, the same nuclei are important for stockpile stewardship applications. In this work, a description of the β-Oslo method and its application to select nuclei will be presented as well as the prospect for future measurements.

NUCL

Graduate education

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Some retrospectives about graduate education 50 years later will be shared -- with emphasis on today's honoree.

NUCL

Nuclear chemistry: Fundamental science impacting society

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Award talk for Glenn T. Seaborg Award for Nuclear Chemistry.

NUCL

High performance computing: Advances and challenges in modeling rare earth elements and actinides

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Applying High Performance Computing (HPC) has been essential to advances in modeling systems containing Rare Earth Elements (REEs) and actinides. Predictive capabilities with sufficient accuracy and robustness to model large scale systems have faced limitations from an imbalanced hardware-software ecosystem. This presentation will include a historical perspective of HPC evolution since the 1990s, and HPC's integration in modeling REE- and actinide-containing compounds relevant to nuclear and radiochemistry. An overview of challenges and advances in HPC applications in chemical modeling relevant to nuclear and radiochemistry, and extensions to data science and artificial intelligence will be presented.

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Numerical methods and benchmarking across scales, precisions, and hardware platforms

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For many decades now, HPC served scientific community in a variety of ways that advanced the computational simulation capability based on models that not so long ago remained theoretical. Measuring the progress made by the high-end hardware has become its own field with a vibrant community of performance engineers exchanging the practical experiences and even building theory of performance models that allow the users to gain a deeper comprehension of the multi-million dollar machines. With transition to data science and great demand for deep models for statistical inference, the need for accurate measurement of hardware capability has remained strong. With comparative performance studies that inform the decision makers ranging from purchasing to software implementation and deployment, this talk will offer a glimpse of how scientifically important numerical methods can serve to give insightful information into the potential and the drawbacks of the modern hardware platforms. The talk will feature description of algorithms, historical perspective on the HPC hardware, and results from a variety of systems from relevant benchmarks.

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A database approach to predicting trans uranic waste forms

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Density-functional theory (DFT) provides an efficient way to screen potential trans-uranic (TRU) waste forms. To this end, the Center for Hierarchical Waste form Materials (CHWM) has created a structural database using known parent structures to generate hypothetical TRU crystal structures. The calculated formation enthalpies for the candidates are then compared to those of competing reaction products, following the methodology of the Open Quantum Materials Database (OQMD). The results provide relative stabilities that are used to speculate formation probability. Our calculations

using this approach have led to the discovery of new relatively stable structures, encouraging potential adoption across a wide span of applications.

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Machine learning-accelerated design for advanced nuclear waste form development

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The design of advanced nuclear waste forms needs to optimize the composition of the waste form for enhanced performance. Complex ceramics with various elements occupying multiple crystallographic sites can have an enormous number of possible compositions as a solid solution. To fully utilize the structural flexibility and compositional complexity of some ceramic phases and to search for materials with optimal performance, an extensive search may be required by experiments using the trial-and-error approach. Data-intensive computational techniques such as machine learning (ML) may be able to tackle such a problem. In combination with first-principles modeling, ML can be employed to screen and evaluate a large number of potential new compositions and their properties. Apatite and hollandite were investigated as model systems for the immobilization of radioactive iodine and cesium. Artificial neural network, a supervised ML approach based on statistical principles, was utilized to explore a large number of compositions of apatite- and hollandite-structured materials. Based on the crystal chemistry of the incorporation site, a series of new compositions were predicted to be possible for iodine and cesium incorporations in apatite and hollandite. For iodoapatite ($A_5(XO_4)_3I$), a combination of A-sites with Ag^+, K^+, Sr^{2+}, Pb^{2+}, Ba^{2+}, and Cs^+, and X sites with Mn^{5+}, As^{5+}, Cr^{5+}, V^{5+}, Mo^{5+}, Si^{4+}, Ge^{4+}, and Re^{7+} was predicted to be promising iodoapatite compositions. For Cs-hollandite ($A_2B_8O_{16}$), a number of previously unknown Cs-hollandite compositions were predicted to be possible, including $M^{4+} = Zr^{4+}$ and Sn^{4+} at the B-site and Ti-based hollandite.

NUCL

Dynamic simulation of plutonium processing

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Studies of the processing of PuO2 have been performed over many years due to its role in nuclear fuel and waste storage. Further studies have been performed to develop models for the synthesis of lab-scale PuO2. The PreCalc project combines known and new models for use in modeling plant-scale precipitation. The precipitation module simulates plutonium oxalate nucleation and growth using known processing parameters including molecular weight, concentration, and time. These parameters are correlated to

the primary homogeneous nucleation rate and size independent growth rate to predict the Pu-oxalate particle size distribution after precipitation. This multi-stage software framework also utilizes a data-driven calcination model to predict the PuO2 product characteristics after calcination. A number of processing conditions were identified to control the specific surface area and mean particle size of the final product. Using SRNL calcination results a data-driven model was developed to correlate process data to the specific surface area (SSA) and mean particle size (PS). By combining the two model frameworks the PreCalc software provides prediction of the full PuO2 product characteristics after both precipitation and calcination processes. The results presented here are meant to provide insight into the current efforts in software development and illustrate future capabilities.