NUCL 3803471

Shifting structural and vibrational properties of UO_3 phases under pressure

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Nuclear fuel cycle materials, including the ubiquitous uranium trioxide phases, may be exposed to elevated pressures during their life cycle. Therefore, understanding the behavior of these materials under pressure is critical. Raman and infrared spectroscopy can reveal details about the changing lattice dynamics under pressurized conditions; complementary computational investigations can assist in the interpretation of experimental results at the atomic level. Here, the high-pressure (up to 45 GPa) behavior of four phases of UO_3 was predicted using density functional theory and detailed structural and vibrational examination of the results revealed unique changes in the lattice dynamics by phase.

We predicted that β and γ phases have an anisotropic response to pressure, and α and δ were expected to change isotropically. At each pressure studied, the vibrational modes of each pressurized structure were predicted using density functional perturbation theory. Examination of the phonon eigenvectors allowed us to completely assign all modes and identify the structural origins underpinning some unexpected softening of several phonon modes. We also aggregated our assignments by unique uranium coordination environments across all phases studied.

NUCL 3808945

Direct analysis of uranium on cotton swipes by microextraction-ICP-MS

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Environmental sampling is a major tool used by the International Atomic Energy Association (IAEA) to detect undeclared nuclear material. After collecting the samples, they are sent to the IAEA Network of Analytical Laboratories for bulk and particle analysis. Traditional analytical methods result in highly sensitive and accurate measurements but require lengthy sample preparation. Directly sampling the surface of an environmental sample would cut out most of the sample preparation, leading to much shorter analysis time. A method to directly extract uranium and plutonium from the surface of an environmental sample was developed using a commercial off-the-shelf microextraction probe, which lowers onto the surface of a sample and forms a seal. A solvent is pumped through the probe head onto the surface of the sample, and then is delivered into the nebulizer of an inductively coupled plasma-mass spectrometer (ICP-

MS). This method has been used to measure major and minor isotope ratios of uranium and plutonium solutions deposited onto swipes and solid particulates of various uranium compounds.

NUCL 3809353

Imaging based approaches for understanding antigen distribution in vaccine formulations

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The efficacy of vaccines is depending upon antigen persistence at the site of injection and trafficking to lymphoid organs. Adjuvants are used in vaccines to help potentiate and direct the resulting immune response, partly by modulating the pharmacokinetic profile of the co-formulated antigen. Despite widespread use, surprisingly little is known about the absorption and distribution of antigens from emulsion-adjuvanted vaccines. To address this issue, the model antigen ovalbumin (OVA) was radiolabeled with zirconium-89 and admixed with clinically-relevant oil-in-water (MF-59, AS03) and water-in-oil (IFA) emulsion adjuvants. These formulations, along with a free antigen control, were then injected subcutaneously into the dorsal flank of 8-10 week old female C57Bl/6 mice and serial PET/CT scans were then acquired along with biodistributions over a 10 day period. Quantitative analysis of the injection site found that both AS03 and MF-59 significantly enhanced the rate of antigen adsorption compared to the free antigen control. IFA on the other hand formed an antigen depot. Uptake into the lymph nodes and other distant organs (i.e., spleen) was pronounced for the oil-in-water cohorts. We have continued this PET-based approach with other clinically relevant adjuvant system, with other clinically relevnt adjuvant systems, such as alums and cationic liposomes, to further help identify pharmacokinetic differences between the adjuvant classes to aid rational vaccine design and formulation in the future.

NUCL 3811436

Hydrolysis of the uranium trioxide phases: Applications of photothermal spectroscopy

***Nicholas Kaitschuck**[1], nick.kaitschuck@austin.utexas.edu, Tanya Hutter[2], Sheldon Landsberger[1], Andrew Miskowiec[3], Tyler L. Spano[3]. (1) Nuclear and Radiation Engineering, University of Texas at Austin, Austin, Texas, United States(2) Mechanical Engineering, University of Texas at Austin, Austin, Texas, United States(3) Nuclear Nonproliferation, Oak Ridge National Laboratory, Oak Ridge, Tennessee, United States*

Uranium trioxide polymorphs appear in various stages of the nuclear fuel cycle and are therefore of interest to the fields of nuclear forensics and security. The formation and time-evolution of these phases is complex and thus requires systematic investigation. A comprehensive understanding of the distinct structures of UO_3 polymorphs and the resulting influence on hydrolysis could be used to determine the provenance and process history of a sample. A survey of the literature indicates the existence of up to seven unique phases and one amorphous phase. This work aims to clarify and consolidate information that has been presented on UO_3 over the last century, as well as explore gaps in understanding which still exist. The evolution of these phases in humid environments presents itself as a clear candidate for further inspection. To set a baseline prior to our environmental aging studies, three of the UO_3 phases have been characterized by Raman and IR spectroscopy using the novel photothermal spectroscopy from Photothermal Spectroscopy Corp. These data are compared with recently published spectra to validate this innovative spectroscopic technique as an appropriate methodology for our hydrolysis experiments.

NUCL 3811436

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NUCL 3813899 - Withdrawn

NUCL 3814310

Technetium and iodine immobilization by organic-inorganic functionalized clay material

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Nuclear waste repository designs require backfill material capable of immobilizing radionuclides. Technetium-99 and iodine-129 are fission products of concern for nuclear waste management; they are present as anionic species (TcO_4^-, I^-, and IO_3^-) in the environment. This work focuses on the development of novel clay materials for the sequestration of anionic Tc and I. We synthesized clays with a combination of quaternary amines and Fe or Zr as an inorganic moiety, characterized the material with XRD, FTIR, and zeta potential measurements, and tested the performance in Tc or I retention using batch tracer experiments. The immobilization of Tc and I by clay functionalized with both inorganic and organic molecules will be discussed and compared to conventional inorganic or organic functionalization.

NUCL 3814404

Purification and measurement of $^{135}Cs/^{137}Cs$ in nuclear debris

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The 135 and 137 mass chains are produced in greater than 6% of the thermal fissions from ^{235}U and ^{239}Pu. Both of these decay chains result in the long half-life cesium isotopes ^{137}Cs (30yr) and ^{135}Cs ($2.3x10^6$ yr). However, due to the differences in the independent yield of ^{137}Cs and ^{135}Cs as well as the inherent volatility and varied half-lives of the cesium precursors (Sb, Te, I, Xe) the ratio of ^{135}Cs to ^{137}Cs is dependent on both the time after fission event and temperature at which the sample condensed. Therefore, the ratio of $^{135}Cs/^{137}Cs$ offers a novel method to investigate the formation conditions of a piece of nuclear debris and how it may be influenced by conditions directly after detonation.

Until recently, measurements of ^{135}Cs were not feasible as its long half-life and pure beta emitting decay make it an especially difficult measurement by radiometric counting. However, advances in thermal ionization mass spectrometry (TIMS) have made the analysis of cesium isotopes achievable. I will present novel methods developed at LANL to separate cesium and analyze it by TIMS.

In this talk I will first discuss the development of a method for the purification of cesium from a variety of sample matrices and load masses. I will focus on our efforts to remove ionization inhibitors such as rubidium during sample purification. Comparison of our results to previously published data will also be discussed.

The second portion of the presentation will focus on how $^{135}Cs/^{137}Cs$ measurements can be applied to analysis of nuclear debris. Because debris from an above ground nuclear

event comes in many different forms, (glassy rock, aerodynamic beads, etc.) all of which are formed at different times, temperatures, and distances from the detonation site, variations in the $^{135}Cs/^{137}Cs$ratios between sample types offer insights into nuclear debris formation but also into the broader question of how to use such values for treaty monitoring, safeguards, and forensics applications. We will show high precision TIMS results on debris from the Trinity nuclear test and discuss how these results influence our understanding of how nuclear debris is formed, as well as relating them to broader nuclear forensics and treaty monitoring missions. LA-UR-22-30853

NUCL 3814642

Research on the thermal stability of swaging & drawing SS/Zr/SS in LOCA situation as innovation ATF cladding

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After the Fukushima nuclear accident in 2011, PWR (Pressurized Water Reactor) is scheduled to be treated with verification technology applied with ATF (Accident tolerant fuels) technology until 2028. Among nuclear power plant materials, nuclear fuel tubes must have resistance to accident situations as well as pressurized and high-temperature environments. In order to delay the accident of the Zr-alloy tube in the LOCA (loss-of-coolant accident) situation, research on surface coating and composition change is in progress. However, since Zr-alloy is used as the base material, it is very difficult to provide dramatic changes to overcome the nuclear accident. This Korean research team shows excellent thermal stability in the LOCA simulation situation for the swaging-drawing technology, which is the core technology to which the ATF technology is applied. In addition, SS-Zr-SS tube (SS316L/Zr-alloy/SS316L) exactly match the specifications of commercial Zr-alloy tubes for nuclear fuel and has the advantage of being a room temperature and mass production process. A swaging-drawing of an ultra-thin SS tube of about 70 micrometers was performed inside and outside, and thermal stability evaluation was performed in the LOCA simulation situation at 1,200°C. Oxidation was inhibited by the formation of Zr-nitride in the interfacial environment of SS and Zr. In theory, it was also confirmed that there was no decrease in the power generation output of a Fe-based alloy having a high neutron absorption cross-sectional area. In conclusion, the innovative swaging-drawing triple tube is expected to be an ATF application technology candidate for PWR, which has not only thermal stability but also accident resistance in the LOCA simulation situation.

NUCL 3815038

Nuclear chemistry summer school: A surprising new journey towards a rewarding career

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When I was a junior in college I was working in an analytical chemistry group anticipating pursuing my graduate degree in the field. A few years later, and I am now a Ph.D. candidate in nuclear chemistry at Texas A&M University. How did my career path change so dramatically in such a short amount of time? While looking for a summer job, I came across an advertisement for the Nuclear Chemistry Summer School (NCSS) funded by the DOE. Although I knew nothing about the subject or any previous participants, I was very intrigued to try something new and exciting so I applied. A few months later, I was a part of NCSS's Class of 2019. Although the coursework was quite intensive throughout the summer, the connections we made with each other and all of the visiting scientists were equally as valuable. The fall of my senior year, I completely changed research groups to begin my journey down a new path and began applying to many nuclear chemistry graduate programs. Through visiting with the amazing scientists at NCSS and hearing about their research activities, I had a general understanding of what I might be interested in when visiting these programs and preparing my applications. I then ultimately decided to pursue nuclear chemistry for graduate school under the guidance of Dr. Sherry Yennello at Texas A&M University, whom I met during NCSS. Since beginning this new chapter, I have made numerous, invaluable connections within the community. I am also making my own scientific contributions by pursuing research in isotope production for the advancement of cancer treatments using targeted alpha therapy (TAT). This new journey, although sudden, has been extremely rewarding thus far, and I look forward to my future career, inspired by the mentorship of Trish Baisden during my time at NCSS.

NUCL 3815041

From 2019 NCSS to functionalized detector surfaces at Texas A&M University

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Attending Nuclear Chemistry Summer School (NCSS) in the summer of 2019 has been one of the defining moments in determining the momentum of my research career. I was exposed to the possibility of going to NCSS through my internship at Los Alamos National Laboratory (LANL). After spending the first half of my summer at LANL, I flew to San Jose, where I met several individuals who would end up profoundly influencing my career, including Dr. Trish Baisden. Attending NCSS provided me with a technical education, tours of world class nuclear science facilities, and practical advice on how to have a successful career in nuclear science. Ultimately, the experience I had at NCSS solidified my decision to continue my career in nuclear science, and pivoted the direction of it. Based on the information that NCSS provided on nuclear chemistry programs, and some of the professors I met at the school, I ultimately decided to attend Texas A&M University, working under Prof. Charles M. Folden III. At Texas A&M, my

work has consisted of using functionalized self assembled monolayers (SAMS) to functionalize gold coated silicon detector surfaces for the detection of lighter homologues of super heavy elements. This talk will discuss my path toward studying nuclear chemistry and how NCSS has impacted my career.

NUCL 3815551

Pretargeting with alpha emitters: Delivering ^{225}Ac and ^{212}Pb to the tumor

David Bauer[1]*, bauerd1@mskcc.org, Roberto Degregorio*[1]*, Alexa Michel*[1]*, Spencer Kaminsky*[1]*, Mengshi Li*[2]*, Michael K. Schultz*[2,3]*, Jason S. Lewis*[1,4,5]*. (1) Radiology, Memorial Sloan Kettering Cancer Center, New York, New York, United States(2) Viewpoint Molecular Targeting, Inc., Coralville, Iowa, United States(3) Radiology-Division of Nuclear Medicine, The University of Iowa Healthcare, Iowa City, Iowa, United States(4) Program in Pharmacology, Weill Cornell Medicine, New York, New York, United States(5) Radiology, Weill Cornell Medicine, New York, New York, United States*

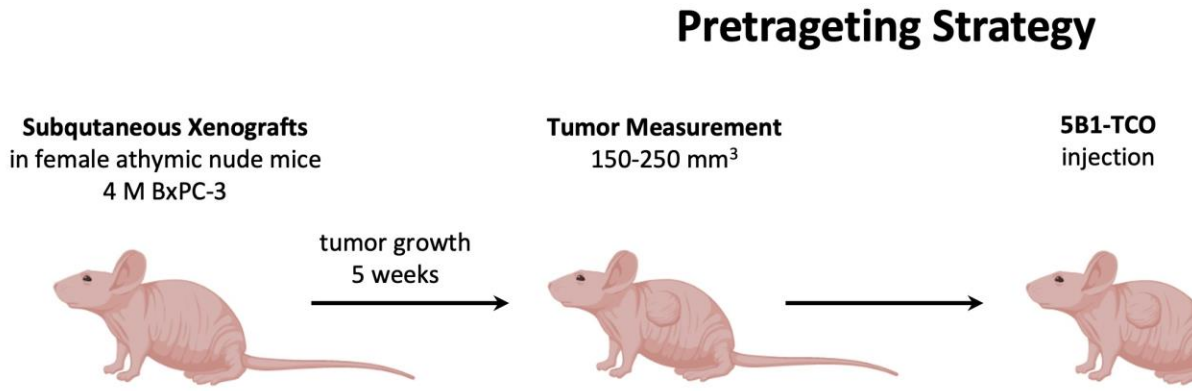
Background: Pretargeting is a fantastic strategy for targeted radionuclide therapy in cancer, benefiting from an antibody's excellent affinity toward its target and the rapid clearance of small molecules. One kinetically and radiochemically outstanding approach is the bioorthogonal reaction between radiolabeled tetrazines (TZ) and antibody-conjugated trans-cyclooctenes (TCO). In this study, the Tz unit was conjugated to various chelators, e.g., DOTA, TCMC, and macropa, which can be labeled with alpha-emitting radiometals like ^{225}Ac ($t_{1/2}$ = 9.9 d) or the in vivo alpha-generator ^{212}Pb ($t_{1/2}$ = 10.6 h).

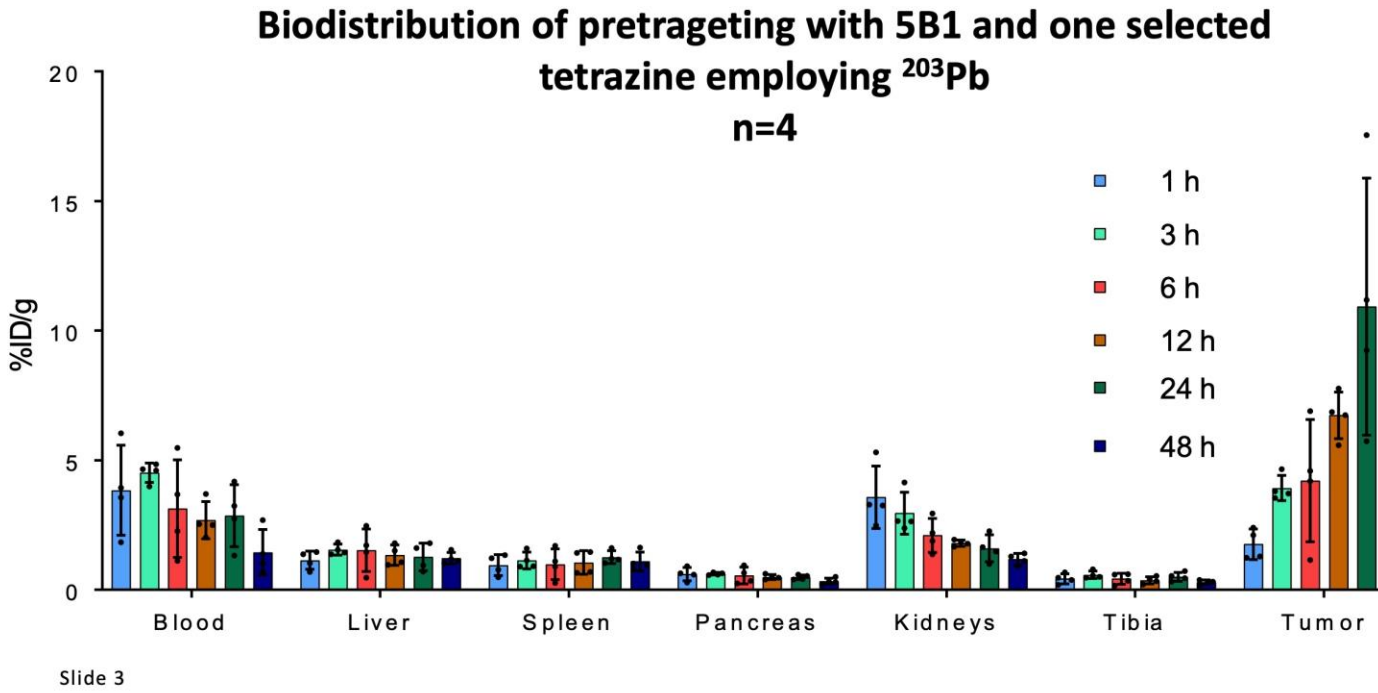
Aim: This study aimed to investigate the radiolabeled Tz-radiotracer in vitro and in vivo for targeted alpha-particle therapy. Tumor uptake and clearance profiles were determined in a xenograft mouse model. We focused on elucidating the fate of the radioactive progeny and potential radiotoxicity.

Methods: Five Tz-conjugates were synthesized using different chelators. Radiolabeling was performed with $^{203/212}$Pb and ^{225}Ac. The in vitro stability of the Tz chelates was tested in human serum via radio iTLC and HPLC. In vivo pilot studies were performed with ^{212}Pb, and studies using ^{225}Ac-labeled Tzs are in preparation. For the pretargeting studies, the TCO-conjugated 5B1 antibody was employed. The clinically evaluated 5B1 antibody targets the carbohydrate antigen 19.9, which is upregulated, e.g., in pancreatic ductal adenocarcinoma. The radiotracers were evaluated in a nude mouse model, subcutaneously xenografted with the pancreatic BxPC-3 cancer cell line. A three-day interval between pretargeting the tumor and injecting the radiolabeled Tzs was chosen.

Results and Outlook: This study investigates the pretargeting approach for ^{212}Pb and ^{225}Ac. The first preclinical studies showed promising biodistribution for the selected Tz-conjugates. The ongoing therapy study with ^{212}Pb revealed a significant therapeutic effect. This effect will be compared to the 225Ac-labeled radiotracer in the following

studies. With this comparison, we hope to determine the difference in therapeutic efficiency and radiotoxicity between these two alpha emitters.





Slide 3

NUCL 3816360

Learning never ends after the nuclear chemistry summer school

***Rebecca Lewis**, beckylewis17@gmail.com. Zeno Power, Washington, District of Columbia, United States*

The Nuclear Chemistry Summer School has been teaching students about this lesser-known discipline for a long time. It has been a decade since I attended the San Jose program, and the foundation I built that summer is still supporting my career in nuclear science. My pathway after NCSS zig-zagged from nuclear medicine, to PhD research in nuclear astrophysics, to federal employment supporting US nuclear defense R&D, and finally to my current position at a nuclear technology start-up. Through it all, I have leaned on what I learned and the connections I made in San Jose. I have utilized my network for support and to learn about new job opportunities; I have opened the textbooks I received more than I ever expected to find useful information; and I have been inspired to share my excitement with young students in the hopes that will choose nuclear science as a career path.
I have greatly benefited from the mentorship of both of the awardees that we are honoring this year. I was in the first NCSS class that Trish Baisden came back to teach in 2012, and returned the following year to TA with her. I first met Paul Mantica when he came to San Jose to recruit new grad students to Michigan State University, and he was on my committee when I received my PhD. For the past 10 years, I have been able to learn from both of them, and I look forward to hearing more about their impact on others like me.

NUCL 3816631

Measuring trace impurities in uranium by ICP-MS and ICP-OES after matrix separation by solvent extraction with TBP

***Luke R. Sadergaski**, sadergaskilr@ornl.gov, Benjamin T. Manard, Laetitia H. Delmau. Oak Ridge National Laboratory, Oak Ridge, Tennessee, United States*

An analytical procedure was developed for determining trace elements relative to uranium(U) in quality control–certified reference samples (i.e., CRM-124-1 and CUP-2) by inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma optical emission spectroscopy (OES), after the extraction of U from nitric acid into 30% tributyl phosphate diluted in dodecane or kerosene. The separation eliminated isobaric and polyatomic interferences in ICP-MS and spectral interferences in ICP-OES related to U, which allowed direct analysis of the raffinate stream. The process was validated against UTEVA column separations. Extraction and stripping tests were used to determine distribution ratios and decontamination factors for nearly 67 elements.

NUCL 3816685

Nuclear and radiochemistry in support of assured security of nuclear materials

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States(2) Institute for Nuclear Chemistry, University of Tennessee, Knoxville, Tennessee, United States(3) Global Security and Strategic Partnerships, Y-12 National Security Complex, Oak Ridge, Tennessee, United States

Securing nuclear materials remains a global challenge, and is a critically important technical area in which nuclear and radiochemistry plays an significant role. It spans the range from understanding the signatures of nuclear material origins and process history for meeting international safeguards obligations to rapidly providing technical analysis in the event of material that is outside of regulatory control, even up to preparedness for a nuclear explosion. Universities play a critical role in creating the skilled scientific and engineering community that can sustainably address these challenges though both research and education of the future members of that community.

This is not a new need. The same challenge of meeting the future workforce needs led to the creation of the DOE/ACS Summer School in Nuclear and Radiochemistry in 1984, and the dedicated work of Drs. Baisden and Mantica that are being honored by this symposium.

This presentation reviews research and education efforts addressing this continuing challenge at the University of Tennessee (UT) as well as related partnerships and ongoing efforts.

NUCL 3816720

Career impact of the national nuclear chemistry summer school

Grace M. Arntz[1]*, gmarntz@gmail.com, Allen G. Oliver*[2]*, Peter C. Burns*[1,2]*. (1) Civil and Environmental Engineering and Earth Science, University of Notre Dame, Notre Dame, Indiana, United States(2) Chemistry and Biochemistry, University of Notre Dame, Notre Dame, Indiana, United States*

The National Nuclear Chemistry Summer School (NNCSS) has served as a pipeline to allow undergraduate students to receive teaching and training in nuclear chemistry that is often unavailable at the undergraduate level. This talk will focus on my experience as a student in the NNCSS and how that has shaped my career. Under the guidance of Dr. Trish Baisden, I was introduced to the breadth of the nuclear field and given the foundational knowledge necessary to pursue a career in actinide chemistry. The professional connections I forged at the NNCSS facilitated my summer internship at the University of Notre Dame with Dr. Peter C. Burns, which ultimately lead to me becoming a PhD student with Dr. Burns. The NNCSS launched my career in nuclear chemistry and the network provided by the school is a continued asset to my professional development.

NUCL 3816826 - Withdrawn

NUCL 3816904

Harnessing radionuclide imaging to investigate the *in vivo* behaviour of metal-based drugs

*George Firth[1], Jana Kim[1], Karlijn Codee Van-Der Schilden[2], Peter J. Sadler[3], Philip J. Blower[1], **Cinzia Imberti**[3,4], cinzia.imberti@warwick.ac.uk. (1) Imaging Chemistry and Biology, King's College London, London, London, United Kingdom(2) Nuclear Research and Consultancy Group, Petten, Netherlands(3) Chemistry, University of Warwick, Coventry, West Midlands, United Kingdom(4) Radiology, Memorial Sloan Kettering Cancer Center, New York, New York, United States*

The platinum agent cisplatin is the pillar of ovarian cancer chemotherapy, but its clinical utility is limited by the onset of resistance. Reduced platinum accumulation in tumours has been proposed as a hallmark for platinum resistance. In turn, a correlation between copper trafficking and cisplatin accumulation in cancer has been repeatedly reported based on in vitro cell studies.
In this study, we set out to use radionuclide imaging as a tool to untangle the link between platinum resistance and cisplatin and copper trafficking in vivo, in ovarian cancer models with different sensitivity to cisplatin. ^{195m}Pt-cisplatin was first utilised in vivo to quantify accumulation in cisplatin-sensitive A2780 ovarian cancer xenografts in comparison to its cisplatin-resistant counterpart A2780cisR. SPECT imaging and ex vivo biodistribution showed rapid renal excretion and very sparse tissue accumulation of ^{195m}Pt-cisplatin, but confirmed that at 2 h post injection cisplatin-sensitive A2780 xenografts accumulate significantly more ^{195m}Pt-cisplatin (%ID/g=1.6±0.1) than A2780cisR (%ID/g=0.7±0.1).
Dynamic PET studies were then performed on the same models and time points using ^{64}Cu as a proxy for endogenous copper. In cisplatin-naïve mice, significantly higher ^{64}Cu accumulation was observed in sensitive A2780 tumours (%ID/g=7.0±0.7) compared to A2780cisR (%ID/g=3.0±0.3), suggesting that copper mirrors cisplatin behaviour in vivo. Most interestingly, when the same animals were imaged again two days later with a second dose of ^{64}Cu, immediately after cisplatin treatment (4 mg/kg), no difference in ^{64}Cu tumour accumulation was observed for the two models with copper uptake in A2780 tumours falling to the same level of A2780cisR, indicating inhibition of ^{64}Cu uptake by cisplatin treatment in the cisplatin sensitive model. Ex vivo analysis of the same tumour tissues by laser-ablation ICP-MS confirmed that, similar to what was found for ^{195m}Pt-cisplatin imaging, cisplatin resistant A2780cisR xenografts accumulated less cisplatin in vivo.
Overall, this work shows that cisplatin and copper accumulation in the A2780/A2780cisR models are deeply interconnected, and suggests that cisplatin may compete for the same entry machinery as copper in cisplatin-sensitive A2780 xenografts. This work also show-cases the power of radionuclide imaging as a tool to investigate the pharmacology of new and established drugs in an in vivo setting.

NUCL 3816904

Harnessing radionuclide imaging to investigate the *in vivo* behaviour of metal-based drugs

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NUCL 3817122

Analytic gradients for spinor-based relativistic coupled-cluster methods and geometry optimization for heavy-element-containing molecules

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We report an implementation of analytic gradients for spinor-based relativistic coupled-cluster singles and doubles with a non-iterative triples correction [CCSD(T)] method. Combined with exact two-component theory to treat relativistic effects, this analytic gradient technique enables geometry optimization and vibrational frequency calculations for heavy-element-containing molecules with accurate treatments of both electron-correlation and relativistic effects. Benchmark calculations of equilibrium structures for molecules containing early actinide elements, together with a study of the infrared spectra for these molecules with anharmonic contributions taken into account are presented to demonstrate the accuracy and applicability of the new computational techniques.

NUCL 3817559

Novel radioimmunoconjugate targeting delta-like ligand 3 for small-cell lung cancer imaging *in vivo.*

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Objectives

Small cell lung cancer (SCLC) is an exceptionally radiosensitive tumor, with selective cell surface expression of the inhibitory Notch ligand Delta-like ligand 3 (DLL3). The selective expression of surface DLL3 on cancer cells presents a target for a variety of diagnostic and therapeutic strategies, including radioimmunoconjugates. The goal of this project is to develop a next-generation DLL3-targeting radioimmunoconjugate for both imaging and therapy.

Methods

In collaboration with the Tri-Institutional Therapeutics Discovery Institute, we screened and assessed >100 antibodies for their binding affinities and internalization rates. Due to pending intellectual property, the specific clones cannot be disclosed. Herein, we present the top-performing clone (mAb1) for imaging purposes of DLL3 expressing SCLC tumors.

The mAb1 was conjugated with the bifunctional chelate p-isothiocyanatobenzyl-desferrioxamine (DFO-Bz-NCS) in a 10-fold molar excess. The conjugated antibody, DFO-mAb1, was then radiolabeled with Zirconium-89 (^{89}Zr), and iTLCs were performed on the final solution to ensure high radiochemical purity.

The *in vitro* and *in vivo* studies were performed on the NCI-H82 cell line that was selected as a DLL3-medium positive cell line and was derived from the pleural fluid of a SCLC patient. Single tumor subcutaneous xenografts were performed in nude mice. Tumor-bearing animals were injected with [^{89}Zr]Zr -DFO-mAb1 (~ 30 µg and 120 µCi / mouse) via tail vein. Immuno-positron emission tomography (immunoPET) was

acquired during 24 h-120 h post-injection. A terminal biodistribution study was also performed.

Results

The cell binding assay showed good binding affinity of the [^{89}Zr]Zr -DFO-mAb1 to H82 tumor cells (>20%). *In vitro* serum stability assays presented no issues with the radioimmunoconjugate. Furthermore, the pilot imaging study showed high tumor uptake of the radioimmunoconjugate (~ 20 % ID/g). Most importantly, the [^{89}Zr]Zr -DFO-mAb1 showed low uptake in non-tumor organs, with a good delineation of the tumor to background.

The *in vivo* biodistribution study, performed 120 h post-injection, confirmed high tumor uptake (~ 20 % ID/g). The uptake of the radioimmunoconjugate in all non-tumor organs was less than 5 % ID/g.

Conclusions

This study shows the potential of utilizing a novel radioimmunoconjugate as a diagnostic tracer, prognostic biomarker, and a predictive biomarker in conjunction with DLL3-targeted therapies.

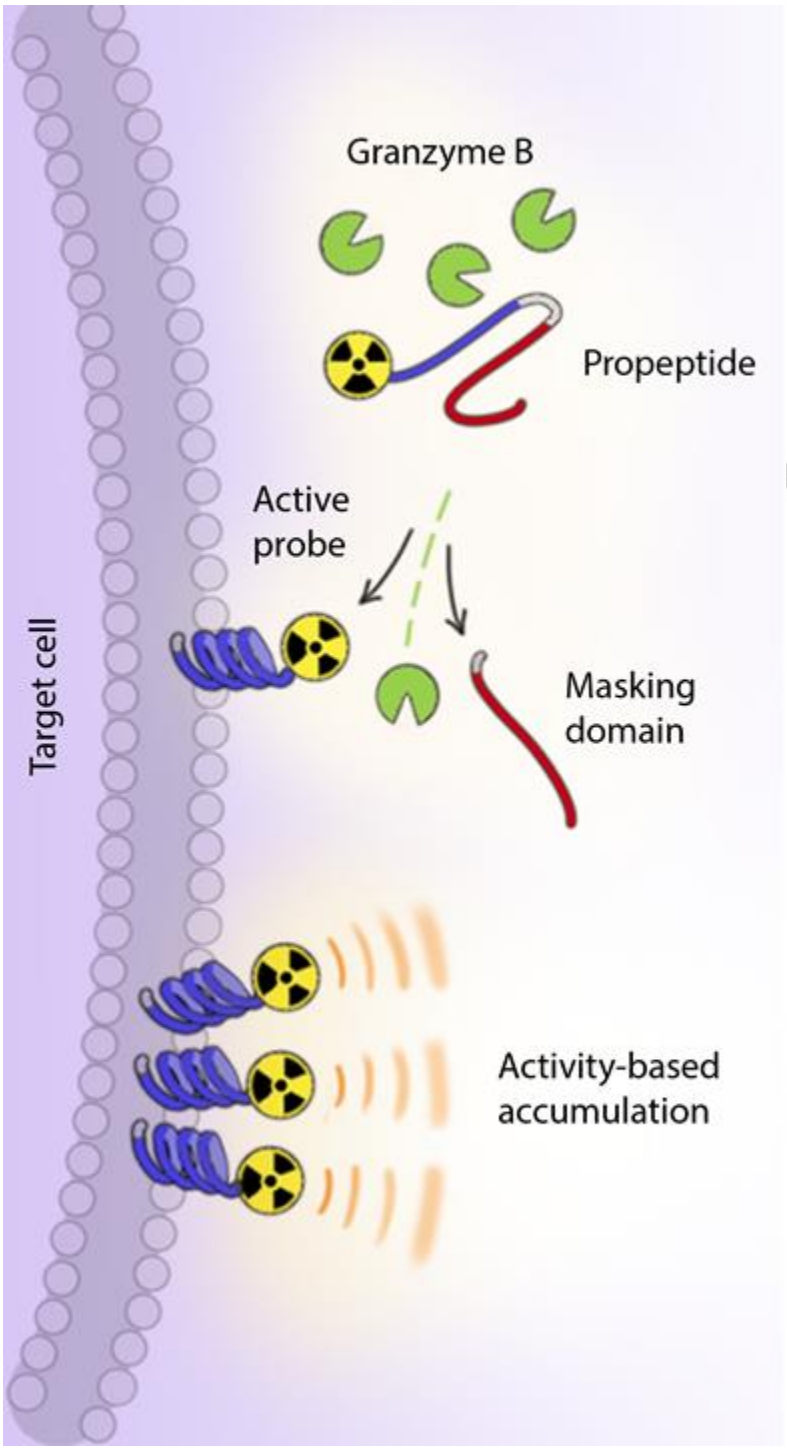
NUCL 3817713

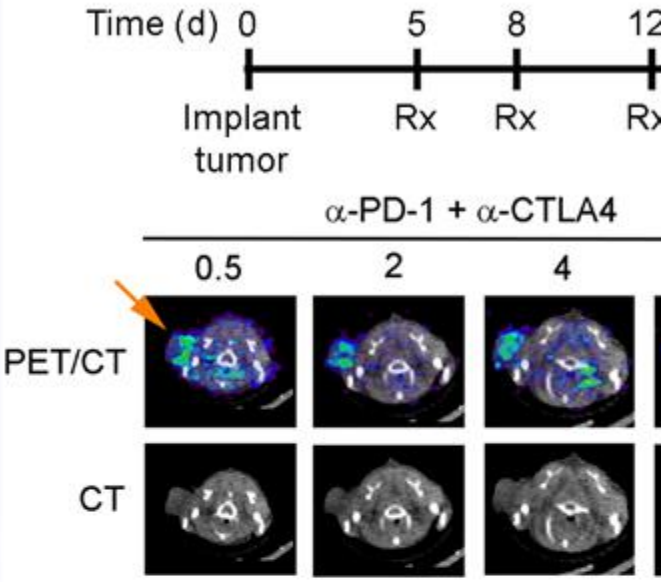
Maximizing tumor responses to targeted radiotherapies with conditionally activated membrane binding probes

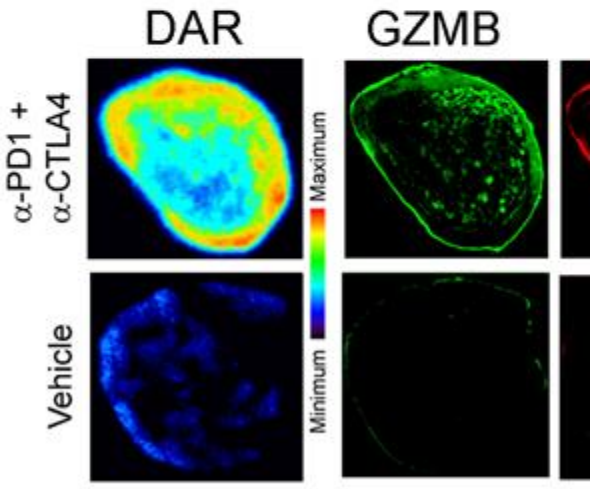
Garima Arvikar[1], Apurva Pandey[1], Ning Zhao[1], Conner Bardine[2], Charles S. Craik[2], ***Michael J. Evans****[1], michael.evans@ucsf.edu. (1) Radiology and Biomedical Imaging, University of California San Francisco, San Francisco, California, United States(2) Pharmaceutical Chemistry, University of California San Francisco, San Francisco, California, United States*

The recent FDA approvals (Lutathera, Azedra, Pluvicto) and the swell of promising experimental agents in clinical trials underscore the surging enthusiasm to investigate molecularly targeted radiotherapy (TRT) as a treatment modality for cancers. However, tumor responses to TRTs are often transient and/or variable among patients. We hypothesize that alternative delivery strategies to radiolabeled small molecule or biologic ligands may confer more durable tumor responses, as radioligands are undermined by heterogeneous target expression among tumors, dissociation or degradation of ligand/receptor complexes, and incomplete target saturation due to low mass doses and infrequent repeat dosing. We have approached this challenge by developing a new class of radiopharmaceuticals termed “restricted interaction peptides” (RIPs) which are linear and unstructured low molecular weight peptides that are internally cleaved by a tumor endoprotease of interest to unmask a radiolabeled, helical membrane binding peptide. Once liberated, the radiolabeled helical peptide immediately and irreversibly attaches to a nearby phospholipid membrane in the tumor. Using PET, we have found that RIPs may have several properties advantageous for TRT, including catalytic amplification of tumor uptake and long persistence of the radioisotope in tumors due to the stability of the peptide/lipid membrane interaction. Thus, RIPs offer an unusual combination of the desirable safety profile characteristic of a low MW RLT with

a high tumoral uptake more typical of a large MW TRT. Our initial experience with RIPs labeled with Cu-64/67, Lu-177, or Ac-225 have shown profound antitumor effects at well tolerated doses. Clinical translation of a ^{64}Cu-labeled RIP targeting human granzyme B is currently underway to assess the safety, pharmacokinetics, and dosimetry of this new drug class.







NUCL 3817884

Development of chemical procedures for isotope harvesting at FRIB: ^{172}Hf and ^{73}As/^{73}Se

***Kelly Kmak**, knkmak@berkeley.edu, John D. Despotopulos, Nicholas Scielzo. Lawrence Livermore National Laboratory, Livermore, California, United States*

With the recent commissioning of the Facility for Rare Ion Beams (FRIB), the development of chemical procedures for harvesting radioisotopes that will be produced at this facility is of interest for many areas of research including astrophysics, stockpile stewardship and medical isotope production. This talk with address on-going R&D efforts at LLNL to develop chemical procedures for harvesting radioisotopes from aqueous and solid materials at FRIB. Solid harvesting will largely focus on long-lived radioisotopes that accumulate in materials such as beam stops or mass slits during operation. One such isotope is ^{172}Hf ($t_{1/2}$ = 1.9 years), which is created in the W-alloy beam stops used at the NSCL and FRIB. Hafnium-172 is the parent isotope of ^{172}Lu ($t_{1/2}$ = 6.7 d), which is of interest for many research applications, including perturbed angle correlation (PAC) studies, a radiochemical diagnostic for stockpile stewardship and, potentially, as a medical radioisotope. A procedure for the separation of trace hafnium from a bulk tungsten alloy (454 g) was established using tracer isotopes (^{175}Hf, ^{88}Zr, ^{173}Lu, ^{88}Y). The procedure has a high yield for Hf (90 ± 8%) with a Hf/W separation factor >10^9 and a purity that meets or exceeds medical isotope standards for all elements except Fe, which was present as an impurity in the W-alloy. Zirconium follows hafnium quantitatively in this procedure; there was no detectable rare earth elements in the final sample. Further research efforts focus on the aqueous harvesting of ^{73}Se and its daughter ^{73}As, which is of interest for nuclear data studies as well as a tracer isotope for radiopharmaceutical research. The preliminary results for a procedure for the isolation, and subsequent separation, of these elements from an aqueous system will be discussed.

NUCL 3818557

Harnessing PET to track micro- and nanoplastic pollutants noninvasively in mice

***Outi Keinanen**[1,2,3], Omk2009@med.cornell.edu. (1) Hunter College, New York, New York, United States(2) Helsingin yliopisto, Helsinki, Uusimaa, Finland(3) Memorial Sloan Kettering Cancer Center, New York, New York, United States*

Micro- and nanoplastic pollution in the environment increases at an alarming rate, and humans are continuously exposed to and consume them. The scientific and medical communities have become increasingly wary of the dangers posed to human health by chronic exposure to microplastics (< 5 mm diameter) and nanoplastics (< 100 nm

diameter). To accurately evaluate the health effects of these pollutants, it is essential to determine their pharmacokinetic behavior in a mammalian model system. Molecular imaging has provided accurate, sensitive, and non-invasive tool to study variety of compounds ranging from small molecules to microparticles. In this presentation, I will show how positron emission tomography (PET) can be an effective tool to study the *in vivo* behavior of plastic pollutants. One of the key factors influencing the biodistribution of plastic particles is the administration route. I will share our results with three different routes of administration – intravenous, pulmonary, and oral. I will also discuss the differences in *in vivo* behavior of micro- and nano-sized plastic particles.

NUCL 3818663

Biphasic isothermal titration calorimetry (ITC) as a tool to develop new polymeric extractants

Connor M. Gallagher[1,2]*, CGallagher@vt.edu, Michael D. Schulz*[1,2]*. (1) Chemistry, Virginia Polytechnic Institute and State University, Blacksburg, Virginia, United States(2) Macromolecules Inovation Institute, Virginia Polytechnic Institute and State University, Blacksburg, Virginia, United States*

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

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NUCL 3819047 - Withdrawn

NUCL 3819246

Influence of lanthanide identity on ^{1}H NMR within a metallacrown framework

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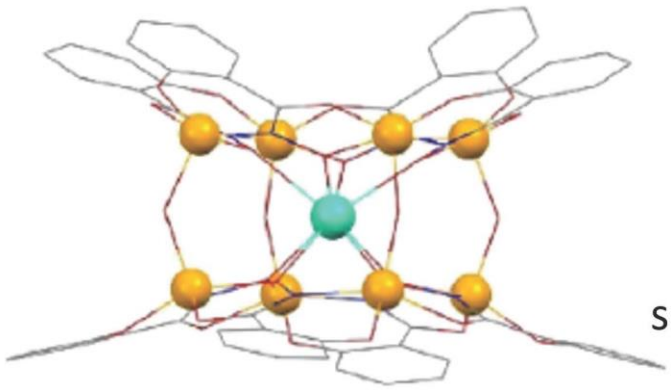
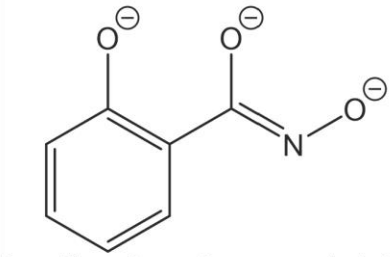
Lanthanide-containing metallacrowns (MCs), are a class of metallamacrocycles structurally analogous to crown ethers, often of interest for magnetic or luminescent applications. Solution characterization of these self-assembled complexes is less well-defined when compared to understanding of solid state properties. Using $LnGa_8(shi)_8(OH)_4$ (Ln = Pr^{III} - Yb^{III}, and Y^{III}) in DMSO-*d_6* and $LnCu_5(pheHA)_5$ (Ln = Nd^{III}, Sm^{III}, Eu^{III}) in aqueous solution as examples, nuclear magnetic resonance (NMR) was used to assess factors such as solution stability and the magnetic impact of the lanthanide on individual proton resonances by analysis of lanthanide induced chemical shift, T_1 relaxation times, and diffusion (DOSY). In MCs where the metal of the macrocycle ring is also paramagnetic, the paramagnetic shift in NMR is dominated by the influence of these transition metal centers, but when the ring metal is diamagnetic, significant contribution from the lanthanide may be characterized. Typically, NMR resonances that had T_1 greater than the diffusion delay defined for the DOSY

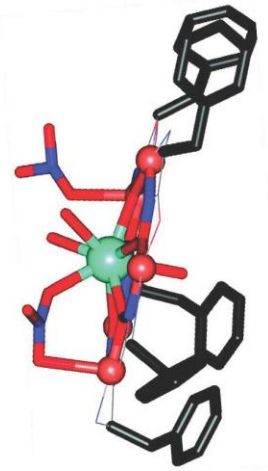
experiment could be observed with the pulsed gradient sequence (excepting broad peaks). Such investigations could have implications for magnetic resonance imaging, molecular sensing, or next generation data storage.



$LnGa_8(shi)_8(OH)_4$



salicylhydroximate (shi)



$LnCu_5(pheHA)_5$

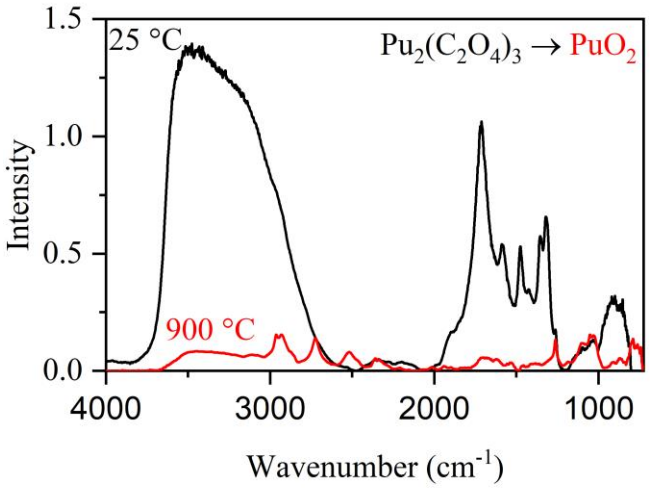
NUCL 3819321

Infrared spectral signatures from the thermal decomposition of plutonium oxalates

Eliel Villa-Aleman[3]*, eliel.villa-aleman@srnl.doe.gov, Jason R. Darvin*[1]*, Samuel Uba*[1]*, Jonathan H. Christian*[2]*, Don D. Dick*[1]*, Bryan J. Foley*[2]*. (1) Nonproliferation & Safeguards, Savannah River National Laboratory, Aiken, South Carolina, United States(2) Chemical Processing, Savannah River National Laboratory, Aiken, South Carolina, United States(3) Global Security, Savannah River National Laboratory, Aiken, South Carolina, United States*

The Savannah River National Laboratory (SRNL) has been conducting nuclear forensics research using a suite of spectroscopic techniques. Our recent efforts at applying diffuse reflectance infrared Fourier-transform spectroscopy (DRIFTS) as a tool for monitoring plutonium oxalate decomposition has shown early success in the identification of the Pu oxalate signatures in the infrared spectrum of PuO_2. The following presentation discusses the thermal decomposition of $Pu_2(C_2O_4)_3$ and $Pu(C_2O_4)_2$ and the evolution of infrared bands observed as calcination temperature increases from 25 °C to 900 °C. Interestingly, the decomposition of Pu (III) and Pu (IV) oxalate occur at different temperatures even though both oxalates produce similar molecular intermediate species (i.e., plutonium carbonate and CO_2) during calcination en route to PuO_2. Spectroscopic analysis of the resultant PuO_2 showed that certain

vibrational bands appeared at different frequencies depending on whether the oxalate precursor was $Pu_2(C_2O_4)_3$ or $Pu(C_2O_4)_2$. This suggests that DRIFTS may be used for determining the identification of Pu precursors in the PuO_2 spectrum and providing additional insight into the history of an interdicted quantity of material.



NUCL 3819541

Synthesis of cerium trifluoride and plutonium trifluoride from ionic liquids

Elodia Ciprian[1]*, eciprian@nd.edu, Jonathan H. Christian*[1,2]*, Bryan J. Foley*[2]*, Eliel Villa-Aleman*[3]*, Amy E. Hixon*[1]*. (1) Civil Environmental Engineering and Earth Sciences, University of Notre Dame, Notre Dame, Indiana, United States(2) Chemical Processing Section, Savannah River National Laboratory, Aiken, South Carolina, United States(3) Global Security Directorate, Savannah River National Laboratory, Aiken, South Carolina, United States*

Actinide fluorides are important nuclear fuel cycle materials (e.g., UF_4 and PuF_4 are common intermediates in the production of uranium and plutonium metal), yet much remains to be learned about the fundamental properties of these materials, including the effects of aging on their chemical and physical properties. Such studies are challenging because standard production techniques involve the manipulation of large quantities of hydrogen fluoride at high temperatures and there are radiological hazards associated with handling actinide materials. In contrast, the synthesis of lanthanide fluorides from ionic liquids and lanthanide dioxide is well established and provides an appealing alternative route to the production of actinide fluorides. This work explores an approach in which the ionic liquids are used to fluorinate lanthanide and plutonium oxalates (e.g., $Ce_2(C_2O_4)_3{\cdot}9H_2O$ and $Pu_2(C_2O_4)_3{\cdot}9H_2O$) to produce anhydrous lanthanide and or actinide fluorides (e.g., LnF_3, AnF_3). Powder X-ray diffraction, electron microscopy,

thermogravimetric analysis, and vibrational spectroscopy were used to characterize the resulting materials. This alternative synthesis and characterization of actinide fluorides will provide value in the areas of nuclear forensics and nuclear fuel processing.

NUCL 3819771

Award Address (Glenn T. Seaborg Award for Nuclear Chemistry sponsored by the ACS Division of Nuclear Chemistry and Technology). Jason S. Lewis, PhD

***Jason S. Lewis**, lewisj2@mskcc.org. Radiology, Memorial Sloan Kettering Cancer Center, New York, New York, United States*

Dr. Lewis' research interests are focused on the development of new PET radiopharmaceuticals for the diagnosis and treatment of cancer. His work incorporates F-18, C-11, and nonstandard nuclide radiopharmaceutical development, with an emphasis on cancer detection and therapy, in both adults and children. The Lewis Lab works on the development of small molecules, radiolabeled peptides, antibodies and nanoparticles that target the overexpression of receptors and antigens on tumors as well as imaging changes in the tumor microenvironment associated with malignancy.

One area of emphasis for the Lewis Lab has been centered around the remarkable specificity and selectivity of antibodies for cancer biomarkers have made immunoglobulins some of the most flexible and adaptable tools in modern medicine. For therapeutic purposes, a wide range of non-labeled antibodies has now entered the clinic. Antibody-based PET and SPECT imaging agents are not far behind. For example, an array of ^{89}Zr-labeled radioimmunoconjugates has shown significant promise in both preclinical and clinical studies. Zirconium-89 has a number of distinct advantages which make it ideal for ImmunoPET including that the radioactive half-life of 78.4 h matches closely the extend times required for optimum biodistribution of intact mAbs. This Seaborg award presentation will review the current state-of-the-art on the use of radiometals with antibody constructs.

NUCL 3819848

Visualizing the implications of senescence in pancreatic cancer with ImmunoPET

***Edwin C. Pratt**[1], pratte@mskcc.org, Riccardo Mezzadra[4], Amanda Kulick[7], Spencer Kaminsky[1], Scott W. Lowe[4,5,6], Jason S. Lewis[1,2,3]. (1) Radiology, Memorial Sloan Kettering Cancer Center, New York, New York, United States(2) Pharmacology, Weill Cornell Medicine, New York, New York, United States(3) Radiology, Weill Cornell Medicine, New York, New York, United States(4) Cancer Biology and Genetics Program, Memorial Sloan Kettering Cancer Center, New York, New York, United States(5) Howard Hughes Medical Institute - Cold Spring Harbor Laboratory, Cold Spring Harbor, New York, United States(6) Geoffrey Beene Cancer Research Center, Memorial Sloan Kettering Cancer Center, New York, New York, United States(7) Tumor*

Assessment Core, Memorial Sloan Kettering Cancer Center, New York, New York, United States

Molecular imaging with antibody-based positron emission tomography (ImmunoPET) has yielded highly specific agents to quantify abundance and location of numerous antigens.
Many molecular imaging agents target a molecular feature present on a particular cancer cell, and senescent pancreatic cancer represents a new hallmark that needs new and specific imaging tools. Furthermore, this work looks to identify new cancer antigens that occur in response to chemotherapy, possibly as an early sign of resistance. Studies have linked several tumorigenic and pro-metastatic factors to senescence and the Senescence Associated Secretory Profile (SASP). This talk will cover how chemically induced senescence with targeted small molecules such as trametinib and palbociclib or with gemcitabine chemotherapy alone can lead to senescent pancreatic tumors with an altered tumor microenvironment. ImmunoPET imaging of the membrane bound antigen uPAR (urokinase plasminogen activated receptor) shows an enrichment of uPAR in chemotherapy induced senescence treated pancreatic cancer, but alone is not a sole marker of senescence. The challenges and implications of imaging shed antigens such as vascular endothelial growth factor (VEGF) and interleukin 6 (IL6), known to be part of SASP, will also be discussed. This talk will also highlight pharmacologic and theranostic combinations that could further target senescent pancreatic cancer. Jason Lewis has pioneered the use of ImmunoPET imaging, identifying theranostic targets across several cancer types and has ushered pretargeting with click chemistry alongside many pharmacological and chemical approaches to improve theranostics. As a mentee of Jason's, he has given us the freedom to grow scientifically yet prepare us to be the leaders of tomorrow in molecular imaging and he is well deserved to receive this Seaborg Award.

NUCL 3820227

Curium, neodymium, and cerium oxide synthesis and characterization: Curium source term impacts on heavy element production

Connor J. Parker[1]*, parkercj@ornl.gov, Samantha K. Cary*[1]*, Ida M. DiMucci*[2]*, Stosh A. Kozimor*[2]*. (1) Oak Ridge National Laboratory, Oak Ridge, Tennessee, United States(2) Los Alamos National Laboratory, Los Alamos, New Mexico, United States*

Curium oxides are stored for extensive periods before their incorporation into target fabrication processes, likely degrading during this time. This work investigates the characterization of Cm oxides and its stable surrogates—Nd and Ce—during these storage periods. Oak Ridge National Laboratory manages Cm for transcurium heavy element production, primarily the irradiation of Cm isotopes to produce ^{252}Cf, Bk, Es, and Fm. Curium oxides are the feedstock material for the target fabrication process, but minimal characterization has been performed on these heavy oxides, creating a knowledge gap in both the literature and in heavy element production. With the supply of heavy curium dwindling, improving our understanding of the structure and bonding of

Cm oxide systems and how they age over time will improve efficiency in ^{252}Cf and heavy element production and aid in our improved long-term storage of this material. Initial studies of these systems have focused on Nd and Ce as surrogates to inform the Cm experiments. Under a variety of differing conditions, oxides and sulfate materials have been synthesized via resin calcination and analyzed by a host of techniques. Scanning electron microscopy imaging and electron dispersive x-ray spectroscopy provides insights into calcined material homogeneity, particle size distributions, microsphere morphology, and the presence of oxysulfate phases in residual materials that have calcined insufficiently. Thermogravimetric analysis informs heat transfer and reaction modeling performed in COMSOL Multiphysics to optimize furnace operation to sufficiently calcine material and degrade carbon and sulfur in resin materials. Additionally, x-ray absorption spectroscopy has informed the bonding environment of the Cm, Ce, and Nd oxides to the traits observed in the physical characterization of calcined materials. Ultimately, these analyses elucidate the role of feedstock characteristics in how the material degrades over time, while ongoing aging studies take place and can be compared with freshly produced feed material.

NUCL 3820249 - Withdrawn

NUCL 3820482

Condensed phase reaction dynamics of the hydrolysis of the metal hexafluorides UF_6 and MoF_6 by cryogenic layering on a diamond substrate

Abigail Waldron[1]*, abigail.waldron@srnl.doe.gov, Louis McNamara*[1]*, Michael Thomas*[1]*, Patrick O'Rourke*[1]*, Eliel Villa-Aleman*[1]*, Alicia Fessler*[1]*, Darrell Simmons*[2]*. (1) Savannah River National Laboratory, Aiken, South Carolina, United States(2) Oak Ridge National Laboratory, Oak Ridge, Tennessee, United States*

Uranium hexafluoride (UF_6) is highly utilized material feedstock for uranium enrichment processes and reacts readily with atmospheric humidity. The mechanism for this spontaneous hydrolysis reaction is poorly understood. If released into the atmosphere, UF_6 reacts with water to form chemically toxic and radioactive reaction products, uranyl fluoride (UO_2F_2) and HF gas. Historical UO_2F_2 particulate research performed at Oak Ridge National Laboratory has shown the end state of the chemical reaction to be dependent on the amount of water present in the atmosphere. Here we have used a cryogenic layering technique to study the reaction kinetics of the hydrolysis of the metal hexafluorides MoF_6 and UF_6. Metal hexafluoride gas and air were sequentially layered on a diamond substrate kept at liquid nitrogen temperature (~-193 °C) and then allowed to warm and react. This was achieved using a custom designed cryogenic cell using a copper cold finger. Reaction progress was monitored via FTIR over several hours while allowing the substrate top warm up to room temperature and substrate temperatures were recorded. Products were characterized via UV/Vis, XRF, SEM and FTIR.

NUCL 3820564

Density functional study of binding mechanism of rare-earth elements with bis(ethylhexyl)amido diethylenetriaminepentaacetic acid

***Timothy Quainoo**, gw3083@wayne.edu, Zhenfei Liu. Chemistry, Wayne State University College of Liberal Arts and Sciences, Detroit, Michigan, United States*

Bis(ethylhexyl)amido diethylenetriaminepentaacetic acid, denoted as modified DTPA here, has recently emerged as an outstanding ligand for extracting rare-earth elements on a solid-phase media, while the microscopic mechanism for the binding between rare-earth elements and the modified DTPA is still unknown. In this work, we fill this knowledge gap by employing first-principles density functional theory to study the binding mechanism of rare-earth elements with the modified DTPA at different pH values. We first compute the pKa values of the modified DTPA and study the binding between hydrated cations of rare-earth elements and the dominating modified DTPA species at different pH values. Our calculations of the Gibbs free energy explain the qualitative trends observed experimentally in the extraction of rare-earth elements. Our study provides insight into the thermodynamics of binding and separation of rare-earth elements using such materials and guides the future design of ligands for the extraction of rare-earth elements.

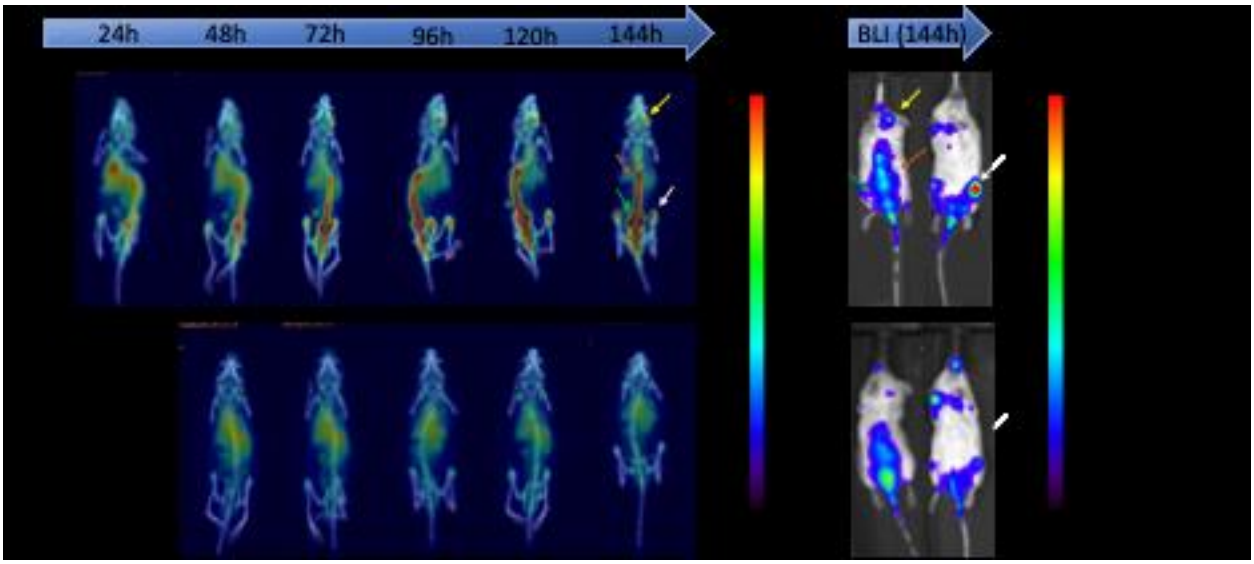
NUCL 3820638

Towards the optimal PET tracer for multiple myeloma imaging

***Natalia Herrero Alvarez**, herreron@mskcc.org, Alexa Michel, Tara Viray, Jason S. Lewis. Radiology, Memorial Sloan Kettering Cancer Center, New York, New York, United States*

Multiple myeloma (MM) represents the second most common hematological malignancy, accounting for 2% of all cancers and 19% of hematological tumors.[1] While novel therapeutic approaches and treatments have been successfully introduced in recent years, MM remains incurable largely due to suboptimal means of measuring disease burden.[2] Only sensitive detection allows for early intervention, directly associated with progression-free and overall survival. Furthermore, detection of minimal residual disease (MRD) following therapy is crucial as most patients eventually relapse. Thus, a sensitive method is urgently needed.[3]
In this context, molecular maging plays a pivotal role. More specifically, PET has proven to be a powerful tool for these unmet clinical needs.[4]We therefore seek to optimize targeted PET imaging as a noninvasive mean to detect, localize, and quantify MM lesions.
Our *working hypothesis* is that finding the optimal pair of PET agent/tumor biomarker will allow us to determine tumor burden, predict the therapy effectiveness, monitor treatment response and rule out MRD. Ultimately, these agents could be extended to the targeted radiotherapy of this malignancy.
Consequently, we have evaluated different biomarkers, known to be overexpressed in MM cells relative to normal cell populations, and developed antibody, antibody fragment

and peptide-based selective imaging agents.
We have recently reported the synthesis and preclinical evaluation of [^{89}Zr]Zr-DFO-isatuximab, the latest FDA-approved CD38-targeting antibody, for the noninvasive CD38-targeted imaging of MM.[6] Here, we present its *in vitro* and *in vivo* evaluation in different models of disease.
Our findings show the potential of [^{89}Zr]Zr-DFO-isatuximab as a selective immunoPET agent for detection of MM lesions, and therefore, the possibility to further develop a platform for targeted radiotherapy. Additionally, modulation of the pharmacokinetics of the radioimmunoconjugate platform results in the modification of imaging times and radiation doses which could hold important clinical implications.



***In vivo* evaluation of ^{89}Zr-DFO-Isatuximab in a disseminated model of MM.** MIP images at 144h in control and block cohort.

NUCL 3820638

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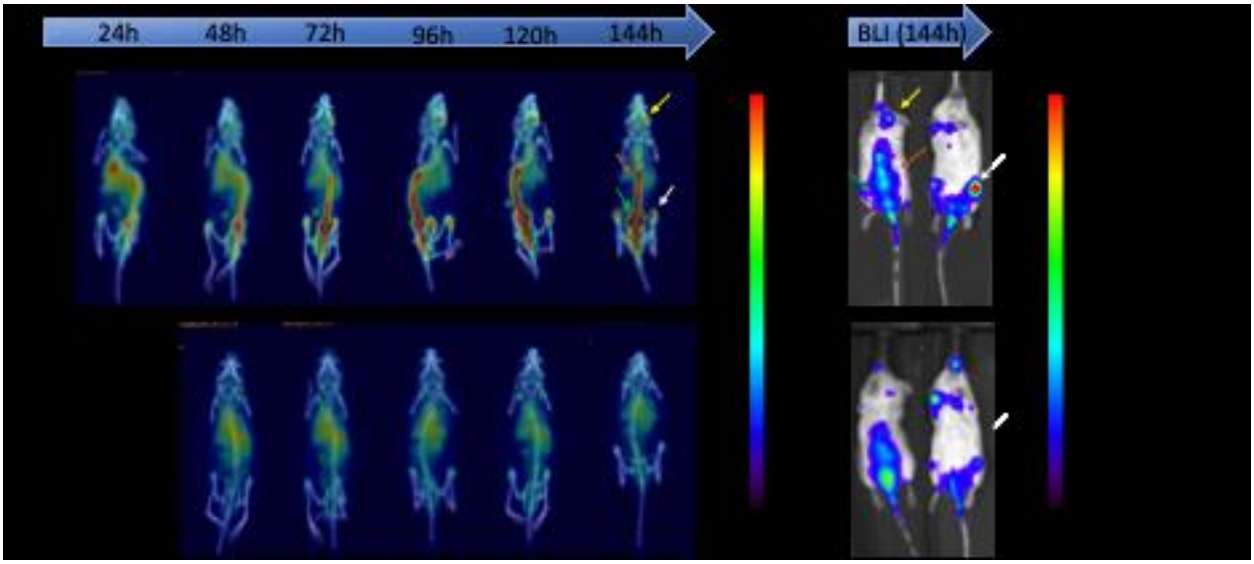
Thus, a sensitive method is urgently needed.[3]
In this context, molecular maging plays a pivotal role. More specifically, PET has proven to be a powerful tool for these unmet clinical needs.[4]We therefore seek to optimize targeted PET imaging as a noninvasive mean to detect, localize, and quantify MM lesions.
Our *working hypothesis* is that finding the optimal pair of PET agent/tumor biomarker will allow us to determine tumor burden, predict the therapy effectiveness, monitor treatment response and rule out MRD. Ultimately, these agents could be extended to the targeted radiotherapy of this malignancy.
Consequently, we have evaluated different biomarkers, known to be overexpressed in MM cells relative to normal cell populations, and developed antibody, antibody fragment and peptide-based selective imaging agents.
We have recently reported the synthesis and preclinical evaluation of [^{89}Zr]Zr-DFO-isatuximab, the latest FDA-approved CD38-targeting antibody, for the noninvasive CD38-targeted imaging of MM.[6] Here, we present its *in vitro* and *in vivo* evaluation in different models of disease.
Our findings show the potential of [^{89}Zr]Zr-DFO-isatuximab as a selective immunoPET agent for detection of MM lesions, and therefore, the possibility to further develop a platform for targeted radiotherapy. Additionally, modulation of the pharmacokinetics of the radioimmunoconjugate platform results in the modification of imaging times and radiation doses which could hold important clinical implications.



***In vivo* evaluation of ^{89}Zr-DFO-Isatuximab in a disseminated model of MM.** MIP images at 144h in control and block cohort.

NUCL 3820677 - Withdrawn

NUCL 3820739

Harnessing copper-free click chemistry for site-specific bioconjugation

Brian M. Zeglis[1,2,3]*, bz102@hunter.cuny.edu. (1) Chemistry, Hunter College, New York, New York, United States(2) Radiology, Memorial Sloan Kettering Cancer Center, New York, New York, United States(3) Radiology, Weill Cornell Medicine, New York, New York, United States*

The development of novel approaches to the site-specific bioconjugation of radioimmunoconjugates is essential as these well-defined and homogeneous probes have repeatedly exhibited improved *in vivo* performance in preclinical models of disease compared to analogues synthesized using traditional, stochastic methods. In this talk, I will describe our efforts to harness the strain-promoted azide-alkyne cycloaddition (SPAAC) reaction for the creation of two different strategies for site-specific bioconjugation. In the first, a pair of enzymes — including a promiscuous galactosyltrasnferase — are employed alongside the SPAAC reaction to site-specifically modify the heavy chain glycans of immunoglobulins. I will discuss the development of this methodology, our exploration into how this bioconjugation strategy modulates the interaction between radioimmunoconjugates and the immune system, and our ongoing first-in-human clinical trial focused on immunoPET with a ^{89}Zr-labeled variant of pertuzumab synthesized using this approach. In the second, the unique selectivity of perfluorophenyl esters for a pair of lysine residues within the IgG is exploited to install azide-functionalized synthons that can then be modified via the SPAAC ligation with cyclooctyne-bearing cargoes. I will describe our recent use of this approach to synthesize ^{89}Zr-labeled radioimmunoconjugates as well as future directions for this technology.

NUCL 3820776

Expanding the toolbox of radiometals for oncologic imaging and therapy

Suzanne E. Lapi*, lapi@uab.edu. Radiology, The University of Alabama at Birmingham Heersink School of Medicine, Birmingham, Alabama, United States*

The integration of targeted molecular imaging and therapeutic radiopharmaceuticals into clinical care has had a significant impact on the diagnosis and treatment of cancer patients. For example, the recent FDA approvals of new radiopharmaceuticals for imaging and therapy of patients diagnosed with neuroendocrine tumors or prostate cancer illustrate this momentum of the nuclear medicine field. New isotopes and the development of corresponding radiochemistry are critical to a continued expansion of these impactful techniques. In particular, theranostic strategies where similar or identical radiopharmaceuticals are used for imaging and therapy can benefit from the availability of elementally matched pairs of isotopes such as ^{64}Cu and ^{67}Cu. Recent advances in cyclotron targetry and radiochemistry has enabled the development of additional pairs including ^{43}Sc/^{47}Sc and ^{203}Pb/^{212}Pb. ^{43}Sc (suitable for PET imaging) and ^{47}Sc (suitable for therapy) can be produced via proton bombardment of Ti targets, purified using ion

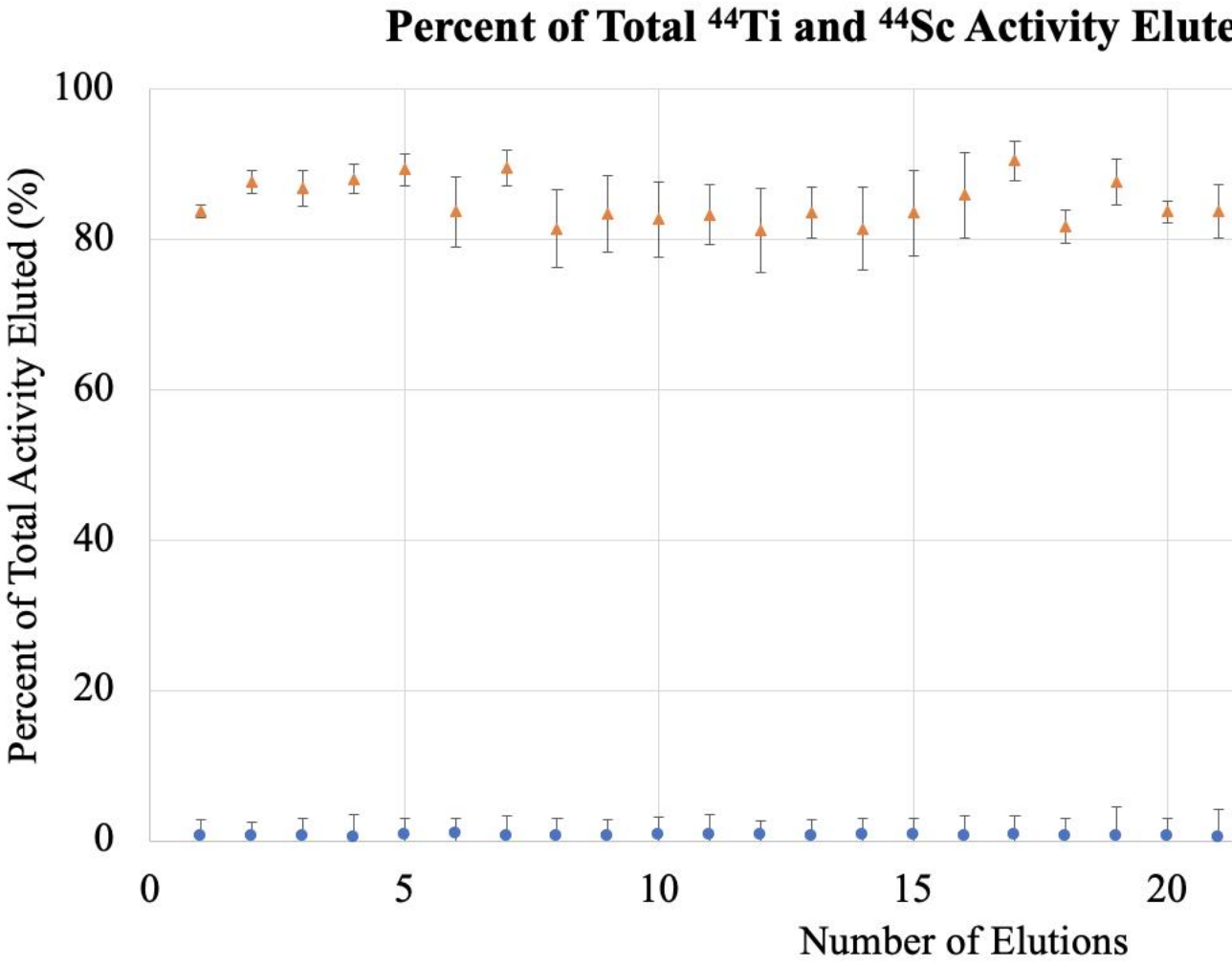
chromatography and complexed with a variety of chelators for radiopharmaceutical development. ^{203}Pb (suitable for SPECT imaging) can be produced from proton bombardment of thallium targets whereas ^{212}Pb is available through a generator system. Recent collaborative results from both radiochemistry and imaging studies will be presented.

NUCL 3820780

Evaluation of a SnO_2-based ^{44}Ti/^{44}Sc generator for medical applications

Christine E. Schmidt[1,2], *cschmidt1@gradcenter.cuny.edu, Jennifer A. Shusterman*[3,4], *Melissa A. Deri*[1,2]*. (1) Ph.D. Program in Chemistry, CUNY The Graduate Center, New York, New York, United States(2) Department of Chemistry, Lehman College, CUNY, Bronx, New York, United States(3) Department of Chemistry, Hunter College, New York, New York, United States(4) Nuclear and Chemical Sciences Division, Lawrence Livermore National Laboratory, Livermore, California, United States*

^{44}Sc has been gaining interest within the field of nuclear medicine as a potential radionuclide for positron emission tomography due to its relatively short half-life ($t_{1/2}$ = 3.97 h) and high positron branching ratio (94.3%). One convenient way to independently produce ^{44}Sc without the need for an on-site cyclotron is by the decay of the long-lived ^{44}Ti parent ($t_{1/2}$ = 59.1 y) via a ^{44}Ti/^{44}Sc generator. However, current separation methods exhibit ^{44}Ti breakthrough during elution, potentially due to inadequate binding to the resin. This work focuses on the synthesis and use of tin dioxide (SnO_2), a robust inorganic-based resin, as the stationary phase for a ^{44}Ti/^{44}Sc generator. The sorption behavior of ^{44}Ti/^{44}Sc was tested on SnO_2 with varying acids, concentrations, and times. Preliminary batch study results showed relatively high ^{44}Ti retention to the resin at lower acid concentrations. Two pilot generators were evaluated for almost a year, demonstrating >80% ^{44}Sc elution yields and <1% ^{44}Ti breakthrough in nitric acid concentrations between 4-5 M. Based on capacity studies, an upscaled generator system is currently under development for further evaluation of the SnO_2 resin and the efficacy of the eluted ^{44}Sc for radiolabeling. The results will be discussed.



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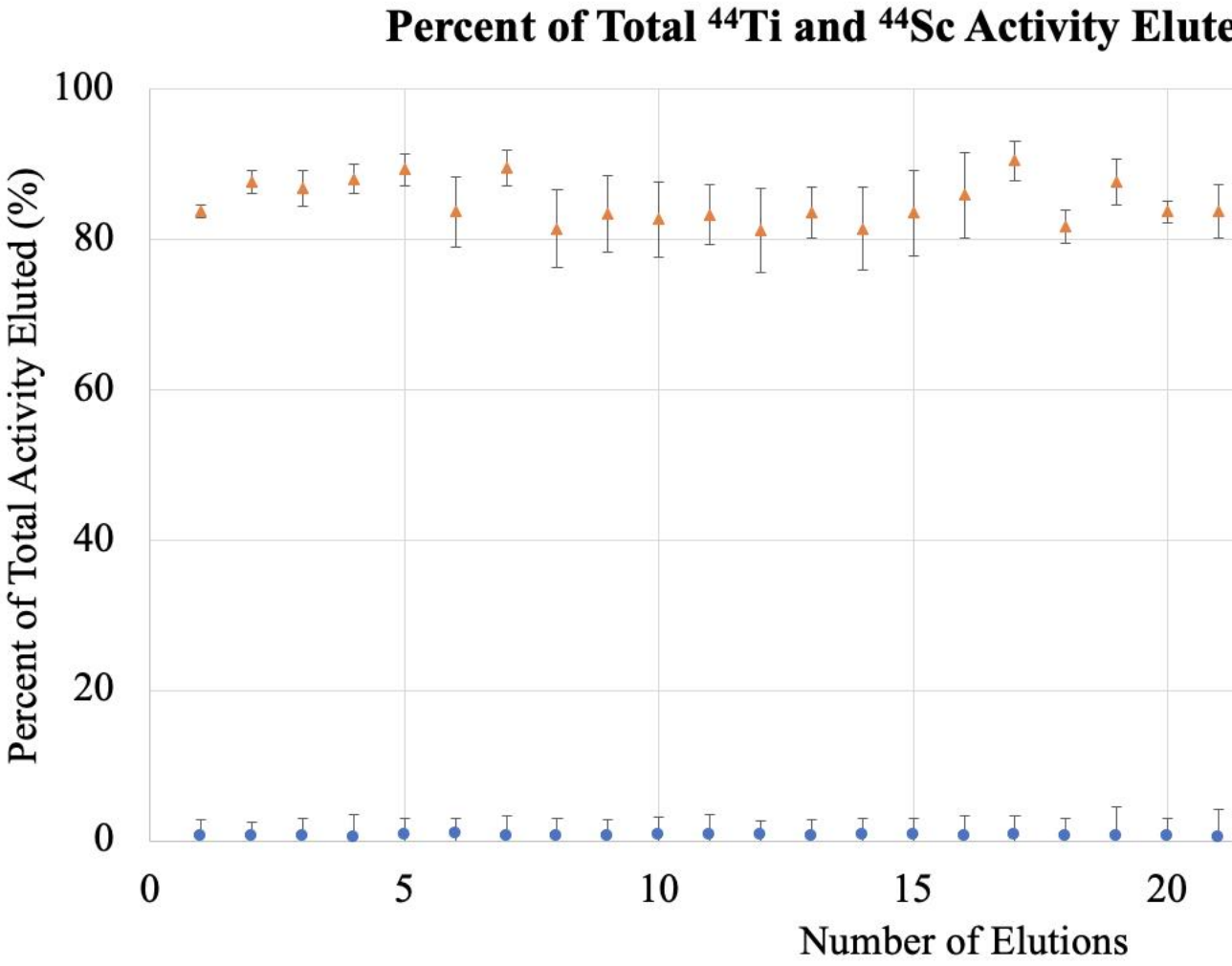

NUCL 3820990

Imaging tumor biology: Lessons learned in the Lewis Lab

Patricia Ribeiro Pereira, *ribeiropereirap@wustl.edu. Radiology, Washington University in St Louis, St Louis, Missouri, United States*

Cancer is a dynamic and heterogeneous disease that requires techniques able to assess biological processes in the entire tumor tissue and in real time. One non-invasive imaging technique with high potential for clinical translation is immunoPET. ImmunoPET combines positron-emitting radioisotopes with an antibody's ability to bind to a target expressed in high levels in the tumor cells. Dr. Lewis, Glenn T. Seaborg Awardee, is a leader in the field of immunoPET, and it was under his mentorship that I learned how to use this technique to understand biological processes of tumor resistance. This is now a major research area in my laboratory, and in this talk honoring Dr. Lewis, I will share two major lessons learned in the last few years:

1. Membrane target availability affects immunoPET and tumor response to antibody therapy. Receptors are not always available at the cell membrane of cancer cells for antibody binding because they can be internalized through endocytosis. We have developed pharmacologic approaches that modulate these processes in ways that enhance therapeutic efficacy. More specifically, we demonstrated that cholesterol-depleting drugs (statins) enhance membrane receptor availability and improve antibody efficacy. Our preclinical findings provided justification for ongoing clinical investigations of antibody drugs combined with statins.

2. Endocytic processes can be modulated to allow for pretargeted therapy of internalizing membrane receptors. Antibodies bearing long-lived therapeutic radioisotopes present an unwanted clinical complication: unnecessarily high radiation dose rates to healthy tissues. Pretargeted approaches—the antibody and a radiolabeled small molecule administered separately combine within the body—offer a potential alternative to the use of radiolabeled antibodies. However, this methodology depends on the presence of the target at the cell membrane. In exploring the role of endocytosis in receptor stability at the cell membrane, we pharmacologically anchored receptors on the cell surface to outperform previous attempts of pretargeting internalizing receptors.

NUCL 3821214

Insights into the structural ambiguity of $An(C_2O_4)_2{\cdot}6H_2O$ sheets (An = Th(IV), U(IV), Np(IV), Pu(IV))

*Kirstin Sockwell[1], Teagan F. Sweet[2], **Brodie Barth**[1], bbarth@nd.edu, Nicole DiBlasi[1], Jennifer E. Szymanowski[1], Ginger Sigmon[1], Allen G. Oliver[2], Peter C. Burns[1,2], Amy E. Hixon[1]. (1) Civil and Environmental Engineering and Earth Sciences, University of Notre Dame, Notre Dame, Indiana, United States(2) Chemistry and Biochemistry, University of Notre Dame, Notre Dame, Indiana, United States*

Oxalates are frequently used as precipitating agents for tri- and tetravalent actinides in the nuclear fuel cycle. For example, plutonium has been purified by the precipitation of plutonium(IV) oxalate for over 60 years, and a common route for the production of ThO_2 involves the calcination of thorium(IV) oxalate. Despite this, the crystal structures of $Pu(C_2O_4)_2{\cdot}6H_2O$ and $Th(C_2O_4)_2{\cdot}6H_2O$ and other relevant compounds to the above processes, have yet to be reported. The structures of their isomorphs, $Np(C_2O_4)_2{\cdot}6H_2O$

and $U(C_2O_4)_2{\cdot}6H_2O$, were reported in 1997 and 2008, respectively, and subsequent work relating to the Pu and Th structures assumed isomorphism with the previously reported U and Np structures. In this work, four actinide(IV) oxalates, $An(C_2O_4)_2{\cdot}6H_2O$ (An = Th(IV), U(IV), Np(IV), Pu(IV)), were synthesized and characterized. The structures of the Th and Pu oxalates are reported for the first time. Additionally, the structures of the U and Np oxalates were re-evaluated along with the Th and Pu compounds, and the data collected within this work revealed insights regarding the coordination of waters to the metal centers as well as a change in orientation of the coordinated oxalates.

NUCL 3821938

Solid-phase isotope harvesting: Harvesting radioisotopes from heavy-ion beam irradiated tungsten beam-blocker

Samridhi Satija[1,2]*, samridhi0417@gmail.com, Chloe Kleinfeldt*[1,2]*, Vladyslav S. Bodnar*[1,2]*, Katharina Domnanich*[1,2]*, Chirag Vyas*[1,2]*, Scott D. Essenmacher*[1,2]*, Greg Severin*[1,2]*. (1) Facility for Rare Isotope Beams, East Lansing, Michigan, United States(2) Chemistry, Michigan State University, East Lansing, Michigan, United States*

The interest in ‘Isotope harvesting’ has peaked because of the tremendous quantities of radioisotopes produced during routine operation of heavy ion-beam facilities such as the Facility for Rare Isotope Beams (FRIB) and its recently decommissioned predecessor National Superconducting Cyclotron Laboratory (NSCL). This presents an opportunity to harvest the long-lived radioisotopes that have wide ranging applications in areas such as nuclear medicine, stockpile stewardship, and material science to name a few, when they get deposited or produced at multiple sites throughout the accelerator and beamline components. By the time these accelerator and beamline components are removed from operation, often considerable activities will have built up. Techniques to recover the by-product radioisotopes from these components is called ‘solid-phase isotope harvesting’ or ‘Isotope mining’ and can help to meet the increasing demand for radioisotopes in a variety of fields. One such decommissioned component is the tungsten beam-blocker from the NSCL that acted as the beam-dump for the primary heavy-ion beams while NSCL was in operation. This talk will elucidate the radioanalytical separation techniques and methodologies used in the extraction and purification of the embedded radioisotopes from the tungsten beam-blocker and their consequent use in Perturbed Angular Correlation Spectroscopy (PAC-Spectroscopy) measurements.
Establishing proof-of-concept procedures for solid-phase isotope harvesting using this decommissioned tungsten beam-blocker is necessary in preparation for the large-scale isotope harvesting at FRIB.

NUCL 3821938

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Establishing proof-of-concept procedures for solid-phase isotope harvesting using this decommissioned tungsten beam-blocker is necessary in preparation for the large-scale isotope harvesting at FRIB.

NUCL 3821968

Monitoring the gas phase reaction dynamics of MoF6 by femtosecond pump-probe IR spectroscopy

Louis E. McNamara[1]*, Louis.mcnamara@srnl.doe.gov, Abigail Waldron*[1]*, Michael Thomas*[1]*, Darrell Simmons*[2]*, Patrick O'Rourke*[1]*, Eliel Villa-Aleman*[1]*, Alicia Fessler*[1]*. (1) Savannah River National Laboratory, Aiken, South Carolina, United States(2) Oak Ridge National Laboratory, Oak Ridge, Tennessee, United States*

Uranium hexafluoride (UF_6) is highly utilized material feedstock for uranium enrichment processes and reacts readily with atmospheric humidity. The reaction intermediates of the associated hydrolysis reaction are debated in the literature. Here we have used molybdenum hexafluoride as a non-radioactive substitute for uranium to study the gas phase hydrolysis of metal hexafluorides. The reaction kinetics of molybdenum hexafluoride (MoF_6) are interrogated using femtosecond pump-probe spectroscopy. Samples are cryogenically deposited on the surface of a diamond substrate kept at liquid nitrogen temperatures (~-193 °C) to form inert layers which are then desorbed by a femtosecond IR laser pump pulse. The desorbed molecules are allowed to react, and

the reaction progress is monitored by a femtosecond infrared probe pulse. The resulting reaction species and kinetics are compared to theoretical values and previously monitored condensed phase reaction intermediates.

NUCL 3823900

Effect of $CeCl_3$ molten salt corrosion as analogous urnium-chlroride for Ni-based structural material in MSR

***Min Gue Lee**, acdc5640@gachon.ac.kr, JISU NA, JeongHye Jo, UNHO LEE, Young Soo Yoon. Gachon University, Seongnam, Korea (the Republic of)*

The whole world needs investment in renewable energy to achieve a Net-Zero policy. Nuclear power generation, which is one of the low-carbon power generation sources, is negatively accepted due to the problem of the disposal method of spent nuclear fuel. Due to the recent nuclear accident Fukushima nuclear accident in 2011, safety is prioritized over economic feasibility in energy generation. Among them, MSR can fundamentally prevent serious accidents caused by loss of coolant through immediate solidification of molten salt. Also, high-temperature corrosion resistance and creep properties of structural materials are essential for the long-term operation of MSR. Instead of UCl_3, which is nuclear fuel, we conducted a simulation experiment with $CeCl_3$ having the same trivalent component and crystal structure (hexagonal). The structural material used a Ni-based alloy and coexists in the isometric line connecting the eutectic point and composite of $CeCl_3$ vertex in the phase diagram (0.453 NaCl-0.207 KCl-0.34 $CeCl_3$) for a composition with a melting point of 900 K, which is scheduled for the actual operating temperature. ClNaKCe salts (Na-K-Ce, 30.7%-13.4%-55.9% or 52.0%-29.0%-19.0%) of different compositions of NaCl, KCl, and $CeCl_3$ were used, and each salt had the same melting point. Candidate salts are pre-treated with H_2O and O_2 10 ppm or less atmosphere to prevent further chemical corrosion, followed by corrosion immersion at 650°C for 96 hours. In this study, through optical analysis, changes in grain and microstructure according to various ratios of cerium (III) chloride were confirmed through corrosion behavior and depth of the cross-section. We aim to accumulate new data in areas where direct utilization of UCl_3 is difficult by confirming the corrosion behavior according to the amount of $CeCl_3$ added. In conclusion, the experimental results of adding nuclear fuel salt to cooling salts suggest various structural material candidates and methods for the corrosive immersion simulation of uranium nuclear materials.

NUCL 3824042 - Withdrawn

NUCL 3824341

Thirty-eight years of the nuclear chemistry summer school: A tribute to Trish Baisden

*Jeffrey C. Bryan[7], Cathy S. Cutler[6], Melissa A. Deri[5], Melody Esfandiari[3], Nicholas E. Esker[2], **Lynn C. Francesconi**[1], Lfrances@hunter.cuny.edu, Victor Maraschin[4], Vanessa Sanders[6], Annalise L. Van Wyngarden[2]. (1) Chemistry, Hunter College, New York, New York, United States(3) Chemistry, San Jose State University, San Jose, California, United States(5) Natural and Social Science, Lehman College of CUNY Division of Natural and Social Science, Bronx, New York, United States(6) Medical Isotope Research and Production, Brookhaven National Laboratory, Upton, New York, United States(7) Chemistry, University of Wisconsin LaCrosse, La Crosse, Wisconsin, United States*

In 1977, recognizing the decline in educational opportunities in nuclear and radiochemistry and the concomitant lack of undergraduate student exposure in the field, the NUCL division of the ACS determined that increasing opportunities for graduate studies was paramount. This could be achieved by supporting new faculty and by increasing the interest of undergraduates in the field. Dr. Patricia A. Baisden, Trish, then of Lawrence Livermore National Laboratory, LLNL, conceived the idea of an undergraduate summer school in nuclear chemistry. In 1981, Trish singularly lobbied and engaged the NUCL division and NAS/NRC in this enterprise. San Jose State University (SJSU) was chosen as the site for the Nuclear Chemistry Summer School (NCSS) to take advantage of their Nuclear Science Facility and experienced nuclear faculty. SJSU could offer college credit, affordable student housing and was near National Laboratories. LLNL defrayed some costs. Trish obtained funding from the Department of Energy. The first NCSS was held at SJSU in 1984. In 1989, a second NCSS was established at Brookhaven National Laboratory. Since they were first initiated in 1984, the NCSS has successfully introduced 862 undergraduates to nuclear chemistry and radiochemistry. Of this group, over 20%, and, over the past 8 years, over 40%, have chosen careers in nuclear science. Many of these individuals influence the next generations of students to enter the field. Trish has taken on many roles in the NCSS over the years. In 2013, she retired from a stellar 37-year National Laboratory career that culminated with her as the Deputy Program Manager for the Inertial Confinement Fusion Program. She had a weekend to transition to the position of lead instructor for the NCSS at SJSU. COVID did not deter the NCSS; Trish and the team pulled together an on-line program. Trish’s commitment to the students does not end after the conclusion of the NCSS. She worked tirelessly to help the students earn fellowships, participate in summer research, and gain admission to graduate school. She is an inspirational and effective educator and life-coach. The DOE and ACS NCSS continue to play a major role in ensuring a pipeline of outstanding students enter the field. As she steps away from the rigors of the NCSS, Trish leaves a legacy of nuclear chemists and radiochemists who are and are becoming leaders in their fields.

NUCL 3824366

Inorganic chemist’s adventures in radiometals before and during the era of Jason Lewis

***Lynn C. Francesconi**, Lfrances@hunter.cuny.edu. Chemistry, Hunter College, New York, New York, United States*

As an inorganic chemist, research on technetium-99m, ^{99m}Tc, almost four decades ago, was an exciting opportunity to examine a new transition metal. In fact, ^{99m}Tc was one of the first radiometals where the contributions of inorganic chemists were instrumental to development of new radiopharmaceuticals. Multidisciplinary teams developed ^{99m}Tc coordination compounds for SPECT imaging of the heart, brain and renal system. These ^{99m} Tc species were characterized and chemical speciation determined using the tools of the inorganic chemist. Research on the inorganic chemistry of technetium benchmarked studies of future radiometals and provided best practices for understanding the speciation of radiometals *in vitro* and *in vivo*. Four decades later, radiometals have exploded on the scene for diagnosis, therapy and theranostics. PET imaging and radiotherapy are important fields of investigation and clinical utility. Chelators tailored to the chemistry of specific radiometals has become an important area of study. Since he was a graduate student, Jason Lewis has been at the forefront of development of new radiometal probes and applications of radiometals for PET imaging and radiotherapy. Moreover, in addition to his achievements in nuclear medicine, Jason has mentored other scientists to achieve successes in these fields. As a testament to his accomplishments as a scientist, mentor and collaborator, Jason is honored with the Seaborg award.

NUCL 3824508

Feasibility of early fuel cycle taggant incorporation for intentional forensics

***Tyler L. Spano**, spanotl@ornl.gov, Tash Ulrich, Toya Beiswenger, Rodney Hunt, Ashley Shields. Oak Ridge National Laboratory, Oak Ridge, Tennessee, United States*

To develop strategies for incorporating taggants into oxide fuels and understand how taggant candidates persist through early fuel cycle processes, synthetic procedures have been developed to produce intentionally tagged early fuel cycle intermediates. Towards this end, metal nitrate solutions are introduced to aqueous uranyl nitrate (UN) to produce tagged UN. From the tagged UN, uranyl peroxide tetrahydrate and dihydrate were prepared, after which α-, β-, and amorphous-UO_3 were synthesized. Structural influences of taggant incorporation are investigated using powder X-ray diffraction and Raman spectroscopy to provide insight into crystallographic modifications resulting from addition of tags to these early fuel cycle materials and elucidate the chemical form of tags introduced at these stages. The possibility of segregation of taggant species into discrete phases within uranium matrices is investigated using scanning electron microscopy–energy dispersive X-ray spectroscopy and Raman spectroscopic mapping with principal component analysis. Finally, production and analysis of tagged U_3O_8 as a precursor to UO_2 powders and compacts will proceed after confirmation of taggant incorporation in earlier intermediates. Results from this study will inform strategies for optimizing taggant incorporation in UO_2.

NUCL 3824796

Hydrophobic ternary mixtures of pharmaceutical and food grade reagents: Characterization in indium extraction from aqueous hydrochloric acid media

Sofie Allison[1,2], *sofie.allison@gmail.com*, *Evgeny Tereshatov*[2], *Charles M. Folden*[3,2]. *(1) Chemistry, Mount St. Mary's University, Emmitsburg, Maryland, United States(2) Cyclotron Institute, Texas A&M University, College Station, Texas, United States(3) Chemistry, Texas A&M University, College Station, Texas, United States*

Environmentally friendly and cost-efficient ways to extract rare metals from aqueous phase solutions are desirable for safety and economical reasons. New hydrophobic mixtures made of common pharmaceutical reagents were introduced to form low transition temperature mixtures (LTTMs). LTTMs are liquids in which no true melting point (a first order phase transition) is observed, but the system instead has only a glass transition point, which is a second order phase transition. As the system cools, the mixture becomes glassy instead of crystalline. Indium is one of the near critical metals for the US and European economies. Understanding of its behavior in the presence of LTTMs is important because it can provide an opportunity to develop a new non-toxic and selective chemical system for metal extraction not only for liquid-liquid but also for gas-solid interactions. The latter is the only path to study chemical properties of superheavy elements ($Z \geq 112$). These elements are produced in nuclear fusion reaction, usually induced by ^{48}Ca projectiles. They are studied in comparison with their light homologs and indium is the light homolog of nihonium ($Z = 113$). Thus, the project is devoted to developing procedures to study chemical properties of superheavy elements. Using different ratios and combinations of five chosen organic reagents (ibuprofen, lidocaine, menthol, methyl anthranilate, and Proton Sponge^{TM}), four of which are actively used pharmaceuticals and food grade ingredients, we have produced and characterized LTTMs by studying their ability to extract indium from hydrochloric acid medium. The medical radioisotope indium-111 was utilized to test the extraction efficacy. This carrier-free radionuclide was transferred from the aqueous to the organic phase and its partitioning was estimated by means of distribution ratio values, which are the ratio of the analyte concentration in organic to aqueous phases.

NUCL 3824796

Hydrophobic ternary mixtures of pharmaceutical and food grade reagents: Characterization in indium extraction from aqueous hydrochloric acid media

Sofie Allison[1,2], *sofie.allison@gmail.com*, *Evgeny Tereshatov*[2], *Charles M. Folden*[3,2]. *(1) Chemistry, Mount St. Mary's University, Emmitsburg, Maryland, United States(2) Cyclotron Institute, Texas A&M University, College Station, Texas, United States(3) Chemistry, Texas A&M University, College Station, Texas, United States*

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NUCL 3824869

Microscopic characterization of Pu-bearing compounds with diffuse reflectance spectroscopy

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Retrospective determination of the process history, age, and storage conditions of interdicted nuclear materials is a key goal in the fields of nuclear forensics and nuclear nonproliferation. Savannah River National Laboratory (SRNL) has been actively engaged in research efforts to answer those key questions. We recently added diffuse reflectance spectroscopy (DRS) in the visible and short-wave infrared (SWIR) spectral regions to our arsenal of spectroscopic tools. Presented here are diffuse reflectance spectra in the SWIR for $^{239}Pu(C_2O_4)_2$ and $^{239}Pu_2(C_2O_4)_3$ and their thermal degradation product $^{239}PuO_2$ at calcination temperatures ranging from 25 °C to 900 °C. An electronic band at 1432 nm was found to be an indicator of crystallinity in PuO_2. Ancillary Raman measurements will be presented as corroborating evidence. Principal component analysis (PCA) of the spectra, in the SWIR, was able to successfully rank the samples based on calcination temperature. It is shown that information pertaining to the process history of $^{239}PuO_2$ can be obtained through the combination of DRS and PCA. Diffuse reflectance spectra of aged $^{240}PuO_2$ produced at SRNL and $^{239}PuO_2$<span style="font-

size:10.8333px"> samples obtained from Pacific Northwest National Laboratory will be compared qualitatively and for PCA model performance.

NUCL 3824924

Route to chemical accuracy for computational uranium thermochemistry

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We present benchmark spinor-based relativistic coupled-cluster calculations for the ionization energies of the uranium atom, the uranium monoxide molecule (UO), and the uranium dioxide (UO_2) molecule, for the bond dissociation energies of UO and UO_2, and for the atomization energy of the uranium hexafluoride (UF_6) molecule. The accuracy of these calculations in the treatments of relativistic, electron-correlation, and basis-set effects is analyzed. The intrinsic convergence of the computed results and the favorable comparison with the experimental values demonstrate the unique applicability of the spinor representation of quantum-chemical methods to open-shell uranium-containing atomic and molecular species with uranium oxidation states ranging from U(0) to U(VI).

NUCL 3825187

Targeting cytokines for imaging cancer and inflammation

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Cytokines exert pleiotropic influences during innate and adaptive immune responses. Inteferon-γ is a hallmark of T helper 1 signaling and plays a key role in stimulating immune pathways towards an anti-tumor response. Interleukin-12 is a pro-inflammatory cytokine secreted primarily by antigen presenting cells in response to microbial pathogens. Interleukin-23 is an effector molecule that is critical for promoting inflammation in various autoimmune conditions, especially in inflammatory bowel disease. Applications to detect and image these cytokines via immunoPET imaging to monitor immune response as well as in inflammatory settings will be presented.

NUCL 3825260

Radical assays used as a screening tool for determining the radiolytic stability of monoamide complexants

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The PUREX (Pu and U Reduction Extraction) process commonly uses tributyl phosphate (TBP) as the extractant to separate out uranium and plutonium from nuclear waste. Using TBP results in third-phase formation and makes waste disposal more difficult because the presence of phosphorus in TBP complicates incineration. To avoid these drawbacks, *N,N*-dialkyl amides (thereafter, monoamides) have been studied as alternatives to TBP. Monoamides are more readily incinerated than TBP and their amine and carboxylic acid degradation products are water-soluble, preventing third-phase formation. Although monoamides are promising complexants, they must also be radiolytically stable to allow scale-up of the separation process. γ-Radiolysis is the gold standard technique for predicting radiolytic degradation products and kinetics, but it is also a low-throughput, high-cost process, so studies of monoamide radiolytic stability are limited. To help eliminate the bottleneck, we are developing non-radioactive radical assays as a potential screening tool for use in conjunction with radiolysis. Upon heating in toluene, azohydroperoxide forms hydroxyl and *tert*-butyl radicals that degrade monoamides, including process-relevant DEHBA and DEHiBA. We have identified correspondence of the same degradation products from the radical assay and γ-radiolysis of the monoamides in toluene, including amine, secondary amide, amide - H_2, and solvent-adduct products. We are working to extend the qualitative agreement of products to quantitative predictions of degradation product ratios and dose-dependence. Based on these correlations, these radical assays show promise as a screening tool to quickly examine the potential radiolytic stability of monoamide complexants before γ-radiolysis studies.

NUCL 3825623 - Withdrawn

NUCL 3826172

Radioligand therapy: The past, the present, and a (possible) future

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Over the last 10 years, radioligand therapy (RLT) has transformed from a nice application to a boardly applied, promising method for controlling tumor growth. Particularly in heavily pretreated and otherwise resistant patient populations - often with significant metastatic burden - RLT has proven to be remarkably successful. With several durgs now being on the market, and others following suit, we will look at the technology's historical background, path to success and potential future directions.

NUCL 3826298

Ligand-induced formation of actinyl-actinyl interactions in neptunium

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High-valent, early actinide elements (U/Np/Pu) are known to form into the unique actinyl cation, $[An(V/VI)O_2]^{+/2+}$ under oxidizing conditions. For neptunium, the pentavalent oxidation state is the most stable, where the $[NpO_2]^+$ structure is well-observed in both solid-state and solution chemistry. The $5f^2$ electronic structure of Np(V) adds increased electron density to the metal center compared to its U(VI) and Np(VI) counterparts, leading to weakened actinyl-oxo bonds and activation of the oxo groups to engage in intermolecular interactions. Actinyl-Actinyl Interactions (AAIs) are a distinct intermolecular interaction which occurs when the oxo group of one neptunyl moiety coordinates along the equatorial plane of another. This interaction can lead to the formation of neptunyl dimers often classified into either T-shape or diamond-shape (D-shape) bonding motifs. The T-shape dimer is prevalent in solution and the solid-state, where formation of this structure is expected to be concentration driven (≥0.1 M). Only a handful of D-shape dimers have been observed in the solid-state often in the presence of carboxylate ligands bridging the neptunyl units in a μ_2 fashion. In this work, Density Functional Theory (DFT) was used to explore the formation of the T-shape and D-shape dimers in solution. Thermodynamic analysis of the dimer formation energies showed a shift in the favorability upon inclusion of a series of different aliphatic and aromatic carboxylate ligands. A frequency analysis between the T- and D-shape structures also revealed activation of new $Np{=}O_{yl}$ vibrational bands upon dimerization where intensity and peak positions may be used to classify specific dimer types. Results from this work were directly compared to experimental Raman studies of Np(V)/carboxylate solutions, and for the first time show the presence of AAIs in low Np(V) concentrations.

NUCL 3826345

Research progress and possibilities in PET imaging for pediatric applications

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With the advent of more sensitive PET scanners in recent years, reductions in scan times and injected activity have made clinical imaging research more amenable to pediatric patients in spite of unique challenges with patient motion and sedation for many younger children. Several PET tracers that are well validated in the adult population have never been translated to pediatric use, and facilitating their application in this setting is a major goal of our group. An overview of our recent efforts addressing PET imaging in catastrophic pediatric diseases will be shared. In addition to the synthesis optimization of known tracers, we have tackled synthesis of a C-11-labeled drug candidate for the treatment of PKAN, a rare pediatric neurodegenerative disease. Development of this novel tracer allowed imaging that confirmed concerns about corneal toxicity for investigators developing the drug.

NUCL 3826369

PET imaging isotopes as surrogates for targeted alpha emitting radiotherapeutics

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Advances in molecular imaging and radionuclide therapy have led to a renaissance in the field of nuclear medicine, giving birth to the new field of theranostics. One definition of theranostics is radiotherapeutic/ diagnostic isotope exchange in the same molecule to provide an imaging agent that may be applied for pharmacokinetics and dosimetry. Actinium-225 (9.9d half-life with multiple alpha and beta emissions), among other alpha emitters (including Th-229, Bi-213, Pb-212, Tb-149), has emerged as a promising isotope for targeted alpha therapy. While positron emitting zirconium-89 has been used as a PET imaging surrogate for these isotopes, its utility is limited in that the chelate for zirconium, DFO, is structurally different from those that bind the alpha emitters. This may potentially introduce disparities in in vivo distribution of the diagnostic relative to the radiotherapeutic. There are several PET imaging radioisotopes (La-132, La-133, Ce-134, Nd-140) being explored that form stable chelates in the same molecules as actinium and other radiotherapeutic isotopes. Recently, we have evaluated Ce-134 (3.2d half-life) that decays to positron-emitting La-134 (6 m half-life) as a surrogate for Ac-225. As this isotope pair is in secular equilibrium, molecules labeled with Ce-134 are in vitro and in vivo generators of La-134. We have examined the chelation properties of Ce-134 and in vivo imaging characteristics of Ce-134, Ce-134 chelates and Ce-134 conjugated molecules. Ce/La-134 is a promising imaging surrogate for Ac-225 radiotherapeutics.

NUCL 3826425

Electrospray ionization tandem mass spectrometry with collision induced dissociation: Probing uranyl-bridging ligands

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Our group has been interested in studying uranyl peroxide nanoclusters (UPCs) since their discovery in 2005 to study property-size-composition-stability relationships. We have now utilized electrospray ionization mass spectrometry (ESI-MS) with collision-induced dissociation (CID) to probe clusters bridging ligands through fragment ion products. The interpretation has revealed clusters transfer into the gas phase through electrochemistry at the electrospray source, considering the fragment ions do not equate to the expected mass to charge of monomeric units if we were to assume the initial known solid-state chemistry. We have utilized the CID technique on nanoclusters U_{24}, $[(UO_2)_{24}(O_2)_{24}(OH)_{24}]^{24-}$, U_{28}, $[(UO_2)_{28}(O_2)_{42}]^{28-}$, $U_{24}Py_{12}$, $[(UO_2)_{24}(O_2)_{24}(P_2O_7)_{12}]^{48-}$, U_{60}, $[(UO_2)_{60}(O_2)_{60}(OH)_{60}]^{60-}$, and $U_{60}Ox_{30}$, $[(UO_2)_{60}(O_2)_{60}(C_2O_4)_{30}]^{60-}$. Gas phase reactions complicate the ultimate interpretation, yet nanoclusters with pyrophosphate

and oxalate bridging ligands can easily be distinguished by the fragmentation pattern. The technique and interpretation will allow us to probe UPCs speciation in an aqueous solution as a function of time to study stability relationships amongst nanoclusters.

NUCL 3826425

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NUCL 3826633

Chemical corrosion mechanism of nickel-based structural materials for molten salt reactor according to addition chlorine species

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The 4th generation nuclear power plant is not only eco-friendly with Net-Zero, but also an extremely safe power generation system. Next-generation nuclear power plants operate for at least 10 years, and the design of passive operation to minimize human error is the main focus. The selection of salt, a coolant, is an important challenge for the natural circulation operation of a nuclear reactor for a very long period. Candidate salts are $_xNaCl\text{-}_{(1-x)}UCl_3$ or $_xKCl\text{-}_{(1-x)}UCl_3$ that maximizes U (uranium) enrichment. When using ClNaK (NaCl-KCl mixed salt) with UCl_3, the operating temperature range is 600~750°C,

and white hydrogen production is possible at the same time as power production. The structural material is a Ni-based super alloy, which suppresses chemical corrosion by designing the most stable metal ion species by Gibbs free energy. Ni-alloy was immersed in ClNaK eutectic salt at 800°C for 48 hours to prove the thermodynamic theory. The corrosion acceleration test provides residual energy at a temperature about 150°C higher than the eutectic point, and about 6.0 times acceleration time condition is confirmed through the Arrhenius equation. Corrosion tests were performed according to ion size and the number of chlorine ions by adding various chlorine-based salts (LiCl, $MgCl_2$, $CaCl_2$, and $AlCl_3$) to prove chemical corrosion. It was possible to predict the effect of the addition of UCl_3 in MSR on the chemical corrosion results. Consequently, we propose structural materials and operating conditions suitable for MSR, types of chlorine-based salts, and conditions for corrosion protection through the results of chemical corrosion research.

NUCL 3826871

Detailed investigations of solvent effects for ligands containing phosphorus functional groups employing ^{31}P NMR

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^{31}P NMR spectroscopy is uniquely suited as an analytical tool because it is highly informative about the structure and molecular environment surrounding the phosphorus atom. Numerous molecules utilized across a broad range of disciplines rely upon ligands containing a complexing phosphorus functional group. In particular, nuclear extraction technologies use phosphorus containing ligands to complex actinide and lanthanide metal ions in complex mixed media solvent environments. Despite the potential usefulness of ^{31}P NMR for studying these systems little is known about the effect of solvent on the chemical shift of ligands containing phosphorus as most studies are situational with respect to both ligand and solvent composition. In order to quantify these effects, we have investigated the influence of solvent polarity on the ^{31}P NMR chemical shift of four phosphorus ligands used in nuclear fuel separation processes. The ^{31}P NMR signals were correlated with empirical solvent parameters to assess solvent-ligand interactions that provide a simple and rapid method for quantifying both solvent electronic and steric effects on metal ligand complexes in applied solvent systems relevant to nuclear extraction technologies.

NUCL 3826985

Characterization, reformulation, and the exploration of the therapeutic benefit of an ^{90}Y-based liquid brachytherapy agent

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BetaBrach^{TM}, a beta-minus (β^-) emitting agent formulated as a liquid suspension of [^{90}Y]yttrium hydroxycarbonate microparticles for intratumoral administration, releases high energy β^- particles into diseased tissue with minimal radiotoxic dose to surrounding tissues. The current radiosynthetic method uses high specific activity [^{90}Y]YCl_3 sourced from a ^{90}Sr/^{90}Y generator. In this presentation, we will report the physical and chemical characterization of non-radioactive and decayed microparticles along with the reformulation of the radiosynthesis to use a low specific activity [^{90}Y]YCl_3 reagent prepared from the ^{89}Y(n,γ)^{90}Y nuclear reaction at the University of Missouri Research Reactor. In addition, we will present results from a particle migration study, which revealed high retention of the radiomicroparticles at the site of administration, as well as a brachytherapy study, which demonstrated a significant therapeutic effect in tumor-bearing mice.

NUCL 3827148

Celebrating 50 years of high energy proton production of isotopes at Brookhaven National Laboratory

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In the 1970's the Brookhaven Linac Isotope Producer (BLIP) came online to accept protons from the 200 MeV Linac that synergistically supports multiple programs including the isotope production program, the Nasa Space Radiation Laboratory (RHIC) as well as the Relativistic Hadron Ion Collider (RHIC). In 2022 BNL celebrated 50 years of isotope production largely supporting medical applications. In addition to the BLIP the isotope program operates the Radionuclide Research and Production Laboratory (RRPL) which contains laboratories and hot cells for processing targets irradiated at the BLIP for external customers as well as internal research. New All Inclusive Production (AP) hot cells have are being brought online to aid in the processing of Ac-225. The BLIP allows the production of isotopes from 200 MeV and down in energies using stacked target arrays that allows for multiple isotope production. High energy accelerators play a critical role in supplying radionuclides. They continue to be upgraded to further production yields by installing beam rastering systems that have

allowed higher intensities and thus higher production yields. Demand for isotopes that can be produced by these systems have also increased. Linear accelerators such as the one at Brookhaven National Laboratory when operating at maximum proton energy of 200 MeV can have simultaneous production of several medically relevant isotopes. Among those are Ac-225 (T1/2=10.0 d), Ce-134 (T1/2=76 h), Pd-103 (T1/2= 17 days) (Se-72/As-72 (T1/2=26 h), Sr-82/Rb-82 (T1/2=1.26 min) and Ti-44/Sc-44 (T1/2=3.97 h). Discussion of recent facility enhancement and production of these novel radionuclides will be presented

NUCL 3827433

Uncovering the intricacies of immunoPET

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ImmunoPET has emerged as a powerful tool to noninvasively characterize the in vivo biodistribution of antibody-based therapeutics as well as to uncover and/or validate complex biology in the treatment of tumors with immunotherapies. Given the volume of preclinical research being done using immunoPET combined with the development of antibody-based radiotheranostic agents, we focused on investigating the non-isotopic aspects that govern the outcomes from the use of this exciting molecular imaging platform.
Foremost, we learned that not all antibodies are created equal. Different isotypes of antibodies generated against the same molecular target (antigen) can yield different in vivo biodistribution profiles. Native IgG4 antibodies are prone to Fab arm exchange and their Fab fragments can be eliminated via the kidneys. Furthermore, afucosylated IgG1 antibodies show high accretion in the liver, bones, and lymph nodes of mice whereas the aglycosylated version of the same IgG1 showed low non-tumoral uptake. Next, we learned that the degree of labeling and the conjugation chemistry used to append the chelator to the antibody impacts the in vivo distribution of the radiolabeled antibody and its metabolites. Radioimmunoconjugates with a high degree of labeling lose their ability to bind their cognate target antigen and end up forming hydrophobic immunocomplexes that get rapidly cleared via the liver. Lastly, we learned how the biological host – i.e., different strains of immunodeficient mice – influence outcomes in immunoPET studies. Low doses of human(ized) IgG1 radioimmunoconjugates get taken up rapidly in the liver, spleen, and bones of highly immunodeficient mice strains such as NOD-SCID-Gamma due to unoccupied Fc receptors expressed on several immune effector cells in these tissues. Thus, tumors implanted in NSG mice had significantly lower uptake of antibody relative to the same tumor implanted in nude mice. Ultimately, from the perspective of what constitutes a druggable molecular target amenable to imaging by immunoPET, we demonstrated that targets such as Delta-Like Ligand 3 (DLL3), which have low tumor cell-surface expression can indeed be imaged by immunoPET since their expression on the surface of healthy cells is absent.
Collectively, over the years, we have described aspects pertaining to the reagent –

radioimmunoconjugate, the biological host, and the molecular target that can have a major influence on outcomes from preclinical immunoPET studies.

NUCL 3827770

VLA-4 targeted ^{177}Lu(III), ^{161}Tb(III), and ^{89}Zr(IV) theranostic agents for imaging and treating melanoma

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Very late antigen-4 (VLA-4) is a transmembrane noncovalent heterodimer that is overexpressed in melanoma tumors and is associated with tumor growth, angiogenesis, and metastasis by promoting cancer cell adhesion and migration. We are developing conjugates of the ligand LLP2A which targets VLA-4 for PET imaging and radiopharmaceutical therapy. Here we present an ongoing structure-activity study to evaluate changing the bifunctional chelator from the standard DOTA to Lumi804, an octadentate, macrocyclic chelator based on four 1-hydroxypyridin-2-one (1,2-HOPO) coordinating units. We also investigated how incorporating an albumin binding agent (4-(*p*-iodophenyl)butyric acid, pIBA) affects biodistribution and therapeutic efficacy. The Lumi804 chelator forms very stable complexes with mid-to-late lanthanides including Tb(III) and Lu(III), and it also binds Zr(IV) and Th(IV) exceptionally well. PEG4-LLP2A and PEG4-pIBA-LLP2A were conjugated with DOTAGA or Lumi804, and then radiolabeled with Lu-177, Tb-161, or Zr-89 (Lumi804 only). All Lumi804-LLP2A conjugates were radiolabeled at room temperature in under 30 min, whereas the DOTAGA- conjugates required heating to at least 70 °C. Antigen binding affinity of ^{89}Zr-, ^{177}Lu-, and ^{161}Tb-Lumi804-LLP2A for VLA-4 showed high affinity (low nM) in VLA-4-positive B16F10 mouse melanoma cells. Preliminary biodistribution data show comparable tumor targeting with improved non-tumor clearance for the Lumi804 agents, demonstrating that this new chelator may improve target selectivity in applications with many theranostic radionuclides.

NUCL 3828027

Spectroscopic and liquid/liquid extraction studies of 3,3’-dibutyloxy BTP toward minor actinide separations

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Development of a more sustainable nuclear fuel cycle continues to generate interest with research focused on ligand-based extractions of minor actinides from lanthanides contained within spent nuclear fuel. The post-PUREX raffinate, containing fission products that include both trivalent actinides and lanthanides, poses a significant challenge in closing the nuclear fuel cycle due to radiotoxicity, residual heat load, and long half-life of the transuranics, such as americium. The difficulty of ligand-based separations of actinides from lanthanides includes the similarities of ionic radius and coordination chemistry between the metal and organic ligand, along with stability of the ligand under acidic conditions. Solubility of the ligand in preferred diluents also presents challenges. Our group has focused on synthesizing ideal ligands for Am/Eu separations as a model system. Current work includes the investigation of complexation and separation of a newly synthesized ligand class, 3,3'-dialkyloxy-phenyl-bis-1,2,4-triazinyl-2,6-pyridine. The complexation chemistry of 3,3'dibutyloxy-BTP with trivalent lanthanide complexes has been studied using spectrophotometric methods and separation assays have been performed under a broad range of conditions to assess preliminary efficacy. Biphasic solvent extraction studies for the separation of ^{241}Am from ^{152}Eu, as well as spectrophotometric titrations toward understanding metal-ligand complexation, will be disseminated.

NUCL 3828055

Density functional theory analysis of ThX_4 (X=F-At)

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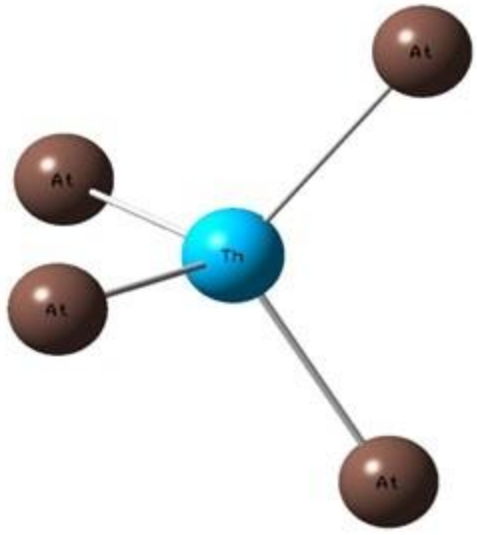
Thorium tetrahalides up to $ThAt_4$ have been studied for bonding, frontier molecular orbitals and molecular surface properties. The molecules have been optimized by meta-GGA density functional TPSS and some of their properties were calculated by using the Gaussian quantum chemical program package. The optimized geometry of $ThAt_4$ is given in Fig.1. All the structures have T_d symmetry. The wave function analysis of the optimized structures have been analayzed using Multiwfn. As a result of molecular surface analysis, it is concluded that variance, molecular polarity index and sphericity decreases with an increase in the atomic number, Z, of the halogen atom. Conversely, the charge of Th decreases with the atomic number of halogen atom as inferred by the electronegativity trend. After molecular orbital decomposition analysis, the highest contributions to HOMO come from halogens, since the LUMO is almost formed from s and f orbitals of Th. The HOMO-LUMO energy gap decreases with the atomic number of halogens. The decrease stems mostly from an increase in HOMO values. Additionally, the Wiberg bond order under NAO basis is found to gradually increases with Z of halogen. The other bond order values (MBO and FBO) exhibits a similar trend. It can be said that the number of the shared electrons generally increase with Z of the halogen.



NUCL 3828062

Probing trivalent actinide/lanthanide covalency through mixed O/S-donor imidazole thione ligands

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Covalency of An(III) and Ln(III) can be explored using ligands containing soft donor atoms such as sulfur and selenium, whose orbitals should overlap more extensively with the radially extended 5f orbitals of actinides compared to the 4f orbitals of lanthanides. Combining these soft atoms into a mixed-donor ligand with hard, negatively charged oxygen atoms will provide strongly chelating ligands to test actinide/lanthanide covalency. Understanding this covalency may be key in determining how ligands can effectively separate An(III) and Ln(III) for purposes such as nuclear waste management, since the ionic radii and charges of these trivalent metals are nearly identical.

We have recently developed a synthetic route toward previously inaccessible diacetate/dipropionate imidazole thiones and selones, which also enable investigation of the importance of pocket size and arm flexibility in the coordination. Mass spectrometry studies demonstrate coordination of these mixed O/S-donor ligands with Nd(III) in 1:1 and 2:1 ligand-to-metal stoichiometries. Indeed, aqueous phase extraction studies with Eu-152 from 1M HDEHP in heptane demonstrate stronger effective binding of our mixed O/S-donor ligands to Eu-152 than is observed for 2,5-thiophenedicarboxylic acid (TPA). Ongoing work exploring the extraction affinity of these ligands for Pu(III) is quantifying

the selectivity between An(III) and Ln(III) and benchmark the strength of interaction of the *f*-block metal with the chalcogenic donor atom. Establishing this difference in the binding and extracting ability between the ligands will be important in demonstrating their utility in probing actinide covalency.

NUCL 3828070

Synthesis and reactivity of uranyl superoxide and its persistence in aqueous solutions

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Radioactive decay associated with spent nuclear fuel in presence of moisture produces an array of reactive oxygen species affecting the surface chemistry of $U(IV)O_2$-based fuels. Such surface reactivity leads to oxidation of U(IV) and results in $U(VI)O_2^{2+}$ alteration phases in form of uranyl peroxide and uranyl carbonate materials. The superoxide anion and the hydroxide radical are among most reactive products that are formed under such radiolytic conditions, but the interaction between these reactive oxygen species and actinides has not been extensively studied. We report isolation of U(VI) mixed peroxide/superoxide solid phase and explore its reactivity under ambient conditions. Single crystal and powder X-ray spectroscopy, vibrational spectroscopy, absorption spectroscopy, magnetic susceptibility, and electron paramagnetic spectroscopy have all been employed to confirm the existence of uranyl superoxide adduct, as well as showcase the reactivity of such adduct both in the solid-state and in solution.

NUCL 3828073

Synthesis and electrochemical characterization of uranium salophen complexes

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Understanding and controlling actinide redox chemistry plays a central role in the reprocessing of used nuclear fuel, where precise oxidation state control is required. This is in part because the actinides (U, Np, Pu, Am, and Cm) exist in oxidation states from +3 to +6 in used fuel recycling conditions. These oxidation states can be generated through redox processes with radiolysis products. This variety also presents separations challenges where oxidation states such as 5+ (AnO_2^+) are difficult to coordinate and can be unstable or reactive. To improve our understanding of actinides in the hexavalent and pentavalent oxidation states we have undertaken electrochemical studies with the schiff base ligand salophen. Uranyl(VI) complexion with salophen is used to not only better understand the properties controlling the stability of UO_2^+, but also to act as a surrogate for AmO_2^+. Cyclic voltammograms of the UO_2^{2+} Salophen complex in different

organic solvents feature differences in the reversibility of the U(V/VI) couple. Exposure to light and oxygen also result in different complexes, suggesting O_2 can become involved in the coordination.

NUCL 3828918

Impact of the nuclear chemistry summer school at Michigan State University

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The Michigan State University nuclear chemistry program includes studies of radiochemistry, nuclear reactions and structure, astrophysics, and the nuclear equation of state. The strength of the graduate program is dependent on the overall quality of its graduate students. For many years, even decades, the ACS Nuclear Chemistry Summer School (NCSS) has identified the brightest students across the nation and brought them to two locations to expose them to nuclear science through intense coursework coupled with hands-on applications in a laboratory setting under the strong leadership of Trish Baisden and Paul Mantica. The NCSS has proved to be an excellent venue to recruit high-achieving future graduate students that have gone on to graduate programs such as ours and then on to successful careers. I will discuss the impact that the ACS NCSS has had the Michigan State University program and I am glad to acknowledge the effort put forward by Trish and Paul as leaders and the many instructors that have kept it running. We look forward to continued success of the school and the development of high quality students that it attracts that will be necessary for success at FRIB.

NUCL 3828928

TB, a generator of special nuclear “material”

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When bright, interested undergraduates come in contact with Trish Baisden they often become activated and the result is nuclear chemists. Trish Baisden has been acting as a generator of this very special nuclear material for many years. Trish was involved in starting the ACS/DNCT Nuclear Chemistry Summer School in the early 1980’s. After several years of persuasive discussions - with anyone who couldn’t avoid being cornered - the first school was held in 1984. In the history of the NUCL division it is stated that “Some very capable and dedicated members were responsible for its success, but Patricia Baisden’s extra-ordinary leadership and effort make her a true NUCL Heroine”. In 2013, thirty years after instigating the summer school, Trish became the Principle Instructor for the SJSU site. While 120 students have been transmuted by

a summer with Trish, nearly 750 more have benefitted from Trish's activity. We are all fortunate that the excitation energy is high and the halflife is long for Baisden so as to produce a plethora of nuclear chemists.

NUCL 3829881

Recovery of Cf-252 from campaign rework solutions via tailored radiochemical separations

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Californium-252 has been recovered from campaign rework solutions using DGA resin and tailored elution sequences, providing a potential source of ^{252}Cf for wire production. Californium campaigns are conducted at Oak Ridge National Laboratory every other year to produce nominally 50–100 mg of this valuable isotope. Produced from the irradiation of curium targets in the High Flux Isotope Reactor, californium requires several processing steps to be obtained in a purity adequate for wire fabrication. Because of the number of processes, rework solutions are unavoidably generated and commonly set aside for the next campaign. For the 2021 ^{252}Cf campaign, decision was made to harvest the californium present in rework solutions. Surrogate tests were first carried out with lanthanides to determine a range of acceptable conditions, and these tests were followed by glove box tests involving either a very small amount of actual rework solution or surrogate feed spiked with actual rework solution. Results from the small-scale tests done inside the hot cells allowed us to refine the conditions for full-scale runs. This endeavor yielded the recovery of 9 mg of ^{252}Cf, which is more than 10% of the original amount present in the irradiated targets.

NUCL 3848846

Career in radiopharmaceutical sciences from the 1984 summer school in nuclear chemistry

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As a grateful alumnus of the 1984 Summer School in Nuclear Chemistry at San Jose State University, my fulfilling career in radiopharmaceutical chemistry is due in large part to the vision of Dr. Trish Baisden. Trish recognized in the 1970's that the workforce in nuclear chemistry and radiochemistry was aging, with few formal training programs and a lack of incoming investigators to take up the mantle from the pioneers of the Manhattan project. She and her colleagues appealed to the Department of Energy to fund a summer school to formally train undergraduates in nuclear chemistry and

radiochemistry, areas of chemistry that are typically neglected in the undergraduate curriculum. As a student in the inaugural class, I was in awe of the instructors, the luminaries we were able to meet with in small groups and share meals with, and the opportunities that were laid out for us. She encouraged me to do my PhD research with Greg Choppin at FSU and spend the summer prior to starting graduate school with Jim Sullivan at Argonne National Lab, where I was introduced to the field of radiopharmaceutical chemistry through Jim's collaborators. In this symposium, I will present a brief overview of my 30+ year career in radiopharmaceutical chemistry. It is gratifying that this field has grown exponentially, particularly in the past decade due to FDA approval of several new PET imaging and theranostic agents. This growth requires a major push for additional training programs, which will also be discussed.

NUCL 3854458

Kinetics of rare earth element oxide dissolution in organic solutions

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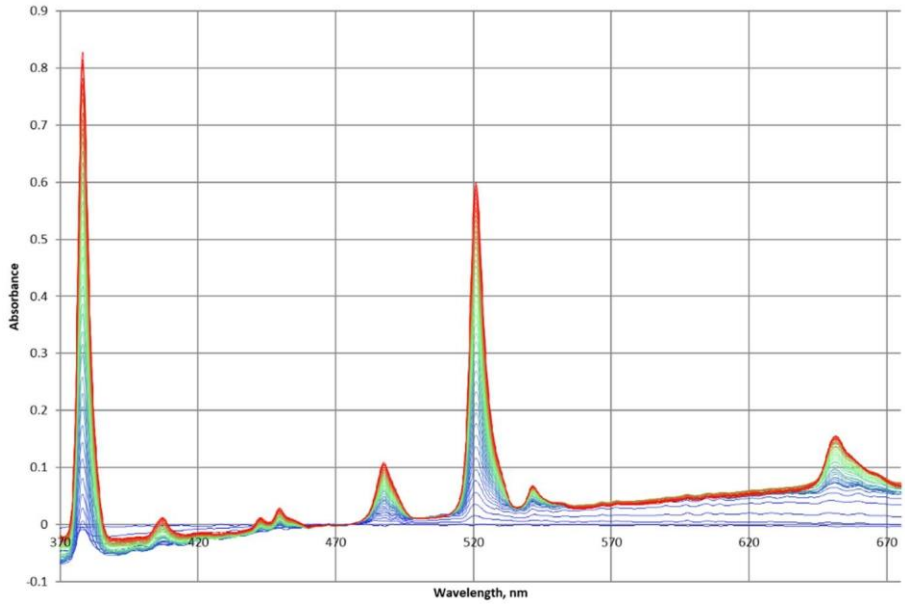
A potentially more efficient method to reprocess used nuclear fuel (UNF), instead of using a massive reprocessing facility, is to combine the dissolution of UNF in the organic solution (containing an extractant like tributyl phosphate-TBP) with two cycles of solvent extraction. This hybrid process would dissolve the fission products (mainly the rare earth elements) and actinides directly into the extractant. While the dissolution of uranium nitrates and oxides into TBP have been demonstrated, it is uncertain how well the other elements in UNF will dissolve. The rare earth elements are of particular interest because they comprise many of the fission products and can be surrogates for modeling the dissolution kinetics of the trivalent actinides. We studied the kinetics of several lanthanide oxides in TBP (pre-equilibrated with nitric acid) containing organic solutions using UV-visible absorbance, Raman, and x-ray absorbance spectroscopies. Trivalent elements are not readily extracted by TBP; however, by increasing the nitrate concentration the lanthanides become more soluble in the organic phase. Interestingly, it appears that the lanthanide oxides
convert to their nitrate form and then bind to the TBP. As can be seen in the following figure with erbium oxide dissolving in 30 wt % TBP over seven hours using in situ visible spectroscopy. The spectral peaks are more like the lanthanide nitrate compounds rather than the oxides. Our preliminary studies indicate that the lighter lanthanides will dissolve faster than the heavier actinides.



NUCL 3854464 - Withdrawn

NUCL 3854465

High performance computing: Advances and challenges in modeling rare Earth elements and actinides in 2023

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Computational science applications to model rare earth elements (REEs), lanthanides, and actinides are essential to address needs in national and nuclear security. Computationally-driven findings are critical in many applications, including to advance radiotherapeutics design, optimization of separations in REE production and purification technologies, and radiological waste management.

High performance computing (HPC) advances in recent decades have enabled a multitude of solutions to challenges in global needs, including weather predictions, green energy, medical therapeutics, and materials science. A large contribution has emerged from applications of artificial intelligence (AI) – including machine learning (ML) and deep learning. Having entered the exascale era of computing, software

technologies to model REEs and actinides face challenges due to an imbalanced hardware-software ecosystem that presents limitations to obtain accurate predictions of REE- and actinide-containing compounds at large scale. Challenges in scalability, performance, and memory limitations restrict efficient modeling in nuclear- and radiochemical applications.

This presentation will address a historical perspective of HPC resources development since the 1990s, current tools to model REEs and actinides, computational protocols for preferential binding in radiochemical separations, and applications of AI/ML in binding selectivity. Additionally, current contributions to exascale efforts in multidisciplinary work involving co-design will be highlighted.

NUCL 3854472 - Withdrawn

NUCL 3854476

Symposium on diversity, equity, and inclusion in nuclear science and technology

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During recent decades, nuclear and radiochemistry fields have suffered a workforce shortage. This challenge is exacerbated by a current workforce in the field lacking diversity. Although some diversity challenges may be approached through optimizing recruiting efforts, lack of representation of diverse demographics has highly negative effects in retaining underrepresented minorities. Increasing representation of underrepresented minorities in the workforce is critical to attract students to pursue nuclear and radiochemistry in undergraduate and graduate school. This session will facilitate a conversation for the community through a panel led by experts in radiochemistry, and diversity, equity, inclusion, and belonging (DEIB).

NUCL 3854477

Facilitating advances in nuclear and radiochemistry through computational science

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Advances in High Performance Computing (HPC) have accelerated the ability of computational sciences to predict unknown chemical and physical properties, and to

complement and inform experimental research. However, as HPC becomes both more powerful and more user-friendly, and traditional experimental groups expand to perform their own computational modeling, it is important for the nuclear and radiochemical community to evaluate research needs and expand HPC training opportunities. This demands establishing synergistic efforts to find solutions to challenges, and optimize opportunities at the intersection of experimental and computational efforts. This session will facilitate a conversation for the community through a panel led by experts in radiochemistry, computational chemistry, HPC, scientific computing, and materials science. Cutting-edge HPC offers the nuclear and radiochemical community the opportunity to ask new, bigger, more complex questions than ever before and cross-disciplinary collaboration between HPC applications and traditional experiments will drive the next generation of advances in the field.

NUCL 3861425

Isotope separation by liquid centrifuge

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A general method of separating isotopes by centrifuging liquid form of isotopes is developed. This technique can be applied to the majority of elements and does not require gasification of isotopes. The method has been applied to Ca, Mo, O and Li with single-stage selectivities of 1.046-1.067 per unit neutron difference (e.g., 1.434 in $^{40}Ca/^{48}Ca$, 1.134 in $^{16}O/^{18}O$), which are better than or comparable to various conventional methods. Modeling was established to understand the process, which is in good agreement with experimental results. The scalability of the technique has also been demonstrated by performing a three-stage enrichment of ^{48}Ca.

NUCL 3868402

Machine learning trained algorithms aid the design of macromolecular ligands with predetermined selectivity for rare earth binding

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Several macromolecules show reverse size selectivity, binding to large, light REs (Rare Earth) rather than the typical small, heavy REs. This desirable quality could lead to selectivity-tuned ligands that are selective for particular REs. However, metal-ligand binding is still an elusive property to predict computationally, particularly when considering the REs due to their similar reactivity. While methods to compute stability constants to describe binding have been previously published, the size of the

macromolecular systems makes computing many different systems too expensive, even with DFT (Density Functional Theory). To provide fast and accurate results to experimental collaborators, an approach that uses LOGKPREDICT, a message passing neural net trained algorithm, coupled with HostDesigner, molecular design software, we show how new ligands with predetermined selectivity may be created using computer-aided design.

NUCL 3868407

Comparison of RIXS cuts and direct XAS excitations

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For high resolution X-Ray Adsorption Spectroscopy, XAS, described as HR-XANES, the XAS is taken as a slice of the Resonant Inelastic X-Ray Scattering, RIXS map; see, for example, Refs. [1-2]. There is, however, the concern that the RIXS slice may contain features not present in the direct excitation to the core-excited states as modeled with XAS. This is because there are overlaps between different portions of the RIXS features due to the broadening of the RIXS measurements.3 In this work, we make a direct comparison of slices of theoretically generated RIXS maps with the direct theoretical XAS calculations. The wavefunctions for the initial, intermediate, and final states where the core-hole is filled are obtained as fully relativistic ab initio solutions of Dirac Hartree-Fock and many body configuration interaction solutions.4 The intensities for the XAS and the RIXS spectra are computed from many-body dipole matrix elements between the initial and intermediate, for XAS and RIXS, and the intermediate to final. for RIXS. This is the first time that such a direct comparison, with the same level of theoretical treatment for the XAS and RIXS has been made. The comparison will be made for selected edges of Neptunyl compounds.