Twenty-Second Symposium on Separation Science and Technology for Energy Applications

November 3-6, 2025
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830

SEPARATION SCIENCE & TECHNOLOGY ★ ★ FOR ENERGY APPLICATIONS ★ ★

Prepared by
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Welcome to the 22nd Symposium on Separation Science and Technology for Energy Applications

The organizers of the 22nd Symposium on Separation Science and Technology for Energy Applications are pleased to welcome you to this year's conference. We restarted this symposium in Knoxville, where it was originally organized in the mid 1980's two years ago and are excited to reestablish its regular schedule. For this 22nd edition, we chose to organize it at Oak Ridge National Laboratory. Due to unforeseen challenges, we have decided to provide an hybrid option for the oral presentations. Some of the talks will be presented virtually. For those of you who are able to attend in person, we hope you will get the opportunity to tour the Spallation Neutron Source. In the future, we will attempt to move it back to an area closer to the Great Smoky Mountains so that we may continue calling by its "Gatlinburg meeting" nickname. We have a number of excellent papers scheduled for presentation in both the oral and poster sessions, and encourage active participation in the discussions of technical papers and posters. We also hope that all the participants will take advantage of the opportunity this conference provides for interchanges of ideas and information both in the formal sessions and informal discussions outside the conference. And finally, a big thank you to our generous sponsors.

If there is anything we can do to make your stay here more comfortable or productive, feel free to call on us. We will help in any way we can.

Thank you for attending, and we hope you enjoy the symposium.

Lætitia H. Delmau
Chair of the 22nd Symposium on Separation Science and Technology for Energy Applications

Organizing Committee

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Symposium Agenda

Monday, November 3, 2025

Badging and Registration	7:30 – 8:00 a.m.
Welcome and Introduction	7:55 – 8:00 a.m.
Session I: Plenary Session	8:00 a.m. – 12:00 p.m.
Lunch (on your own)	12:00 – 2:00 p.m.
Session II: Membrane/Sorption/Ion Exchange Separations	2:00 - 4:50 p.m.
Session III: Lightning Talks, Poster Session, and Reception	5:00 – 8:00 p.m.

Tuesday, November 4, 2025

Badging and Registration	7:30 – 8:15 a.m.
Session V: Solvent Extraction	8:15 – 11:30 a.m.
Lunch (on your own)	11:30 a.m.
Afternoon Free for individual discussions	
Session VI: Medical Isotopes / Novel Separations	6:00 – 7:45 p.m.

Wednesday, November 5, 2025

Badging and Registration	7:30 – 8:15 a.m.
Session VII: Novel Separations	8:15 – 11:30 a.m.
Lunch (on your own)	11:30 a.m. – 1:30 p.m.
Session VIII: Nuclear Fuel Cycle Separations	1:30 – 4:45 p.m.
Banquet Reception	6:00 – 8:00 p.m.

Thursday, November 6, 2025

Badging and Registration	7:30 – 8:15 a.m.
Technical Session IX: DOE Applications	8:15 – 11:30 a.m.
Adjourn	11:30 a.m.

Conference Sponsorship

The conference organizing committee would like to thank the following companies and organizations for their sponsorship of the conference this year.

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Based in Janesville, Wisconsin, SHINE deploys its safe, cost-effective and environmentally friendly fusion technology in a stepwise approach. Its systems are used to inspect industrial components in aerospace, defense, energy and other sectors. SHINE's proprietary medical isotope production processes create non-carrier-added lutetium-177 and will soon create molybdenum-99 via an aqueous subcritical fusion/fission hybrid system. As a next step, SHINE plans to scale its fission product separation and fusion technologies to help solve one of energy's toughest hurdles by recycling spent nuclear fuel and transmuting targeted isotopes. Through a purpose-driven and phased approach, SHINE ultimately aims to generate fusion power to deliver clean, abundant energy that could transform life on Earth. Want to learn more about SHINE?

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TrisKem International develops, produces and markets the reagents used for radioactivity analysis in environmental, medical, bioassay and waste matrices. TrisKem International markets radionuclide separation solutions and applications for nuclear medicine and radiopharmacy. Our main products line of extraction chromatographic resins which allow the separation of radionuclides and elements from various samples and matrices (environmental, medical and bioassay samples, irradiated targets as well as waste samples). Our products are a standard technology in radiochemistry used by international accredited laboratories and monitoring agencies.
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The organizers would like to thank the Isotope Science and Enrichment Directorate at Oak Ridge National Laboratory for supporting the symposium.

Plenary Session

Chair: Lætitia Delmau (Oak Ridge National Laboratory)

8:00 a.m.	Separation Needs for the US Isotope Supply Chain **Jeremy Busby**	*Oak Ridge National Laboratory*
8:45 a.m.	Practical Considerations in the Production of Rare-Earth Elements **David Harvey**	*MP Materials*
9:30 a.m.	Open Questions and Opportunities in Solvent Extraction and Beyond **Bruce Moyer**	*Oak Ridge National Laboratory*
10:15 a.m.	Break	
10:30 a.m.	Transforming Nuclear Waste Management – SHINE's Integrated UNF Recycling and Transmutation Strategy **Abdul Momen**	*SHINE Technologies*
11:15 a.m.	Separations for Radioisotope Recovery and Environmental Remediation **Paul Sylvester**	*Energy Solutions*
12:00 p.m.	Lunch	

Notes

Technical Session I
November 3, 2025

Plenary Session

Presentation Abstracts

Notes

Separation Needs for the US Isotope Supply Chain

Jeremy Busby

Oak Ridge National Laboratory, Oak Ridge, TN 37831

The plenary talk will highlight current and future separation needs for the Isotope Science and Enrichment Directorate. Challenges (and the attempts to overcome them) along with successes for the production of stable isotopes and radioisotopes will be presented.

Practical Considerations in the Production of Rare-Earth Elements

David Harvey[1], *and Joe Pascoe*[2]

[1]Process Development, MP Materials, Lafayette, IN 47909
[2]Process Development, MP Materials, Mountain Pass, CA 92366

MP Materials has been on the forefront of rare-earth production within the United States. Our Mountain Pass location is the largest rare-earth mine and refining operation in the Western Hemisphere. Our experience in the separation and purification of rare earth has made us uniquely aware of the numerous chemical and operation complexities that are inherent to the process. While solvent extraction has been the primary separation technique industrially for the past several decades, there still exist a variety of possible improvements to the process which could enhance productivity and reduce costs significantly. This talk aims to identify the most prominent pain points for current techniques for the separation and purification of rare earth elements, as well as outline criteria necessary for the implementation of any new separation technology that could replace current industrial practice. Crud, low capacity, temperature considerations, low selectivity between rare earth, and difficult to separate impurities all limit productivity in the manufacture of high-quality rare-earth oxide. The creation of new extractants or other technologies should be aware of these considerations when they are being developed in order to be usable in an industrial setting.

Fig. 1 An aerial view of Mountain Pass, photographed in 2024.

Open Questions and Opportunities in Solvent Extraction and Beyond

Bruce Moyer

Oak Ridge National Laboratory, Oak Ridge, TN 37831-6119

Solvent extraction is widely considered a "mature" technology. Is it therefore at a plateau, destined for incremental advances? Already by the 1980s, some practitioners argued that new extractants were no longer needed: applications could be successfully developed, they said, with existing reagents and equipment. Compelling advantages of solvent extraction among available separation techniques include selectivity, high throughput, ease of handling liquid streams, simple equipment, remote operability, and scalability from exotic single-atom experiments to tens of thousands of tonnes per year. Enabled by these advantages, applications appeared in chemical analysis by the 1930s, giving way to nuclear applications such as PUREX in the 1950s, closely followed soon after by hydrometallurgical triumphs such as copper recovery from oxidic ores. But new needs constantly arise, and yesterday's extractants and equipment no longer suffice. For example, growing demands for technologies for nuclear fuel recycle, recovery of critical materials, and recycle of electronics do not find ready-to-use chemistry and engineering. Not surprisingly, attempts to adapt old reagents and equipment have been leading to inefficient and uneconomical approaches.

In generic terms, technology needs, for example, include targeted selectivity from ever-more complex multicomponent mixtures, ability to concentrate from dilute solutions, and environmental friendliness. Such needs seem, on the one hand, timeless but, if anything, have become even more urgent. Fundamental advances, though, are astonishing, taking advantage of powerful new instrumental techniques and user facilities, but old questions identified in early textbooks still stand. These questions include understanding of solvation, aggregation, solution structure, principles of extractant design, interfacial structure, kinetics, fluid dynamics, and equipment design. New questions and opportunities also arise from "big data" and how to exploit its escalating ability and potential.

Beyond solvent extraction, the field of separation science and technology is similarly albeit more broadly challenged. In fact, as a unit operation in a laboratory or plant environment, solvent extraction depends on upstream methods to prepare its feed streams and then must deliver manageable streams to downstream processes. Feed streams often originate from solid materials such as ores, wastes, and end-of-life devices. Thus, questions of sorting, liberation, and leaching bear upon the nature of the separation task presented to solvent extraction. Downstream processes impose stringent requirements for concentration and purity along with environmental limitations. The need for reduced emissions from solvent extraction, both in air and liquid streams, must be recognized and improved upon for greater applicability of solvent extraction. In conclusion, despite the development of solvent extraction as a powerful separation technique and its numerous triumphs, old questions still stand as challenges along with new questions, all demanding new solutions ultimately leading to new applications.

Acknowledgment
This research was Supported by the U.S. Department of Energy, Office of Science, *Office of Basic Energy Sciences*, Separation Science program and Materials Chemistry program under Award Number DE-SC00ERKCG21.

Transforming Nuclear Waste Management—SHINE's Integrated UNF Recycling and Transmutation Strategy

Abdul Momen, Ross Radel

SHINE Technologies, Janesville WI, USA

SHINE Technologies is pioneering a transformative approach to nuclear waste management through its integrated recycling and transmutation initiative. Leveraging its experience in medical isotope production and 10 CFR Part 50 facility licensing, SHINE is developing a pilot-scale UNF recycling facility that combines advanced aqueous reprocessing with strategic isotope recovery.

At the heart of SHINE's process is a liquid-liquid extraction system that begins with voloxidation followed by the CoDCon and ALSEP separation steps. This flowsheet enables the recovery of valuable isotopes—including uranium, plutonium, strontium-90, and noble metals—for commercial use, while isolating minor actinides and other long-lived isotopes for transmutation and reducing the volume and radiotoxicity of final waste streams. The REDUCE method (Recover Elements – Destroy Undesirables – Create Energy), recently funded under ARPA-E's NEWTON program, further enhances this strategy by coupling molten salt targets with external neutron sources to transmute residual waste into less hazardous forms.

SHINE is actively exploring advanced voloxidation and uranium crystallization technologies as part of its broader R&D strategy to optimize UNF recycling. Recent lab-scale studies have demonstrated that up to 90% of uranium can be selectively crystallized from LWR post-voloxidation feed, significantly reducing the volume and complexity of downstream separation processes such as CoDCon and ALSEP. SHINE has developed standard voloxidation methods for DU to U_3O_8 at kilogram scale for its Mo-99 production facility. SHINE intends to incorporate crystallization and voloxidation into its pilot facility if feasibility and cost-effectiveness are confirmed. Alternatively, these technologies may be deployed in a next-generation plant to further reduce waste volumes and improve economic performance.

SHINE's commercialization roadmap emphasizes cost-effective facility design, safeguards-by-design licensing, and co-location of fuel fabrication and disposal capabilities. Through strategic partnerships with national laboratories and industry leaders, SHINE is advancing the technical readiness of key recycling components and positioning UNF recycling as a viable, scalable solution to the nation's nuclear waste challenge.

Separations for Radioisotope Recovery and Environmental Remediation

Paul Sylvester

Energy*Solutions*, 121 W. Trade Street, Suite 2700, Charlotte, NC 28202

Over the last quarter of a century, my career has involved the use of separations techniques for the recovery of radioactive isotopes from irradiated targets, the development of radioisotope generators for medical applications, drinking water treatment, liquid nuclear effluent remediation and the recovery of valuable isotopes from nuclear wastes. The scale has varied from laboratory processes to large scale systems capable of treating thousands of gallons of liquid per minute. This has required familiarity with many different techniques, including solvent extraction, ion exchange, coagulation/filtration and adsorption and a knowledge of the advantages and drawbacks of each approach. Successes and failures over the years have demonstrated that there is no universally successful approach to solving a problem and many factors can influence what technique or techniques are selected.

This presentation will detail some of the separations projects and includes descriptions of the following:

- Sr-90/Y-90 separations to generate ultra-pure Y-90 for radiopharmaceuticals
- Uranium and radium removal from drinking water
- Cooling water treatment at Fukushima nuclear power plant, Japan
- Fuel Element Debris (FED) treatment at Bradwell nuclear power plant, England
- Isotope recovery from nuclear wastes

Each of these projects had their own specific challenges requiring innovative thinking, process development and teamwork to develop robust solutions that satisfied regulatory and environmental criteria.

Notes

November 3, 2025
Monday Afternoon

Technical Session II
SNS, Iran Thomas Auditorium

Membranes/Sorption/Ion Exchange Separations

Chair: Shalina Metzger (Oak Ridge National Laboratory)

2:00 p.m. Removal of Calcium and Silica from Simulated Blowdown Water Using CO_2-Assisted Magnesium Electrocoagulation
Swapnamoy Dutta *Oak Ridge National Laboratory*

2:25 p.m. Alkaline-Side Recovery and Purification of Uranium by Ion-Exchange
Joel Castillo *Pacific Northwest National Laboratory*

2:50 p.m. Loading and ion-Exchange Behavior of Zirconium Phosphate Nanoplatelets
Adrianna Orsi *University of Alabama - Birmingham*

3:15 p.m. Break

3:35 p.m. Electrochemically Enhanced Critical Materials Separations in 2D COF-MXene Membranes
Julia Chung *Lawrence Livermore National Laboratory*

4:00 p.m. Analytical Capabilities in the Radioactive Materials Analytical Laboratory
Shalina Metzger *Oak Ridge National Laboratory*

4:25 p.m. Processing Elusive f-Elements at ORNL: ^{147}Pm, ^{249}Bk, ^{254}Es, and ^{257}Fm
Cristian Celis-Barros *Oak Ridge National Laboratory*

Notes

Technical Session II
November 3, 2025

Membranes / Sorption / Ion Exchange Separations

Presentation Abstracts

Notes

Removal of Calcium and Silica from Simulated Blowdown Water Using CO_2-Assisted Magnesium Electrocoagulation

Swapnamoy Dutta;[1,2] *Gyoung Jang.;*[1] *Jong Keum;*[3] *Jeffery Einkauf;*[4] *Rejaun Nelson;*[5] *Erika Yamazaki;*[6] *Marta Hatzell, M.;*[6] *David Jassby;*[5] *Costas Tsouris*[1*]

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[2]The Bredesen Center for Interdisciplinary Research and Graduate Education, The University of Tennessee, Knoxville, TN 37996, USA
[3]Center for Nanophase Materials Sciences, ORNL, Oak Ridge, TN 37831, USA
[4]Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA
[5]Department of Civil and Environmental Engineering, University of California, Los Angeles, California 90095, USA
[6]George W. Woodruff School of Mechanical Engineering, Georgia Institute of Technology, Atlanta, Georgia 30332, USA

Blowdown (BD) operations in power generation industries use substantial amounts of water to maintain energy-efficient and cost-effective operations. Due to recirculation, the concentration of salt and contaminants in BD water increases significantly. Due to its low chemical usage, high process efficacy, cost-effectiveness, and environmental benefits, electrocoagulation (EC) is used in this study to treat BD water to make it suitable for discharge into surface water. Magnesium (Mg) has emerged as a novel electrode material for EC to meet discharging standards (30 mg L^{-1}). Magnesium alloy (AZ31B) electrodes successfully removed ~91% silica at a charge loading of 5400 C/L but only 31% calcium due to insufficient availability of carbonate ions to interact with calcium. Introducing carbon-dioxide (CO_2) at various concentrations such as simulated natural-gas fired flue gas containing 5% CO_2, simulated coal fired flue gas containing 14% CO_2, and pure CO_2 in the EC process facilitated the formation of carbonate ions, which enhanced pollutant removal. CO_2 addition during EC led to the formation of carbonate ions, which reacted with calcium and magnesium ions to generate calcium and magnesium carbonate precipitates. CO_2 facilitated calcium removal at a rate of ~70%, more than two times higher than the removal rate without CO_2, while maintaining a high silica removal. Formation of calcium carbonate and calcium magnesium carbonate complex ($Ca_xMg_{(1-x)}CO_3$) was found through XRD and SEM-EDS analyses in solid flocs of EC with CO_2 addition. Raman spectroscopy was used in situ with EC to investigate the benefits of CO_2 addition, revealing efficient interaction between Ca and Mg ions with carbonate ions to yield higher calcium removal. Absence of carbonate peaks in EC with CO_2 addition indicated efficient interaction between Ca and Mg ions with carbonate ions to form $CaCO_3$ and $Ca_{0.936}Mg_{0.064}(CO_3)$, which led to higher calcium removal. This study demonstrated that CO_2-assisted Mg-EC can effectively remove calcium and silica ions from industrial wastewater.

Alkaline-Side Recovery and Purification of Uranium by Ion Exchange

Joel Castillo, Tatiana G. Levitskaia, Valerie J. Munoz, Avalon B. Tarbet-Mendoza, Elizabeth M. Garcia, Amy L. Speelman, Nathan P. Bessen, Gregg J. Lumetta, Sergey I. Sinkov, and Gabriel B. Hall

[1]Nuclear Chemistry & Engineering, Pacific Northwest National Laboratory, Richland, WA 99354

Over the last few years, the use of alkaline guanidinium carbonate (GC) media was investigated as a means for recovering uranium (U) from commercial used nuclear fuel (UNF) [1]. This method requires advanced voloxidation using NO_2 as the oxidant, where the fuel matrix is converted from tetravalent UO_2 to hexavalent ε-UO_3 prior to dissolution. This eliminates the need for an oxidizing agent such as H_2O_2 [2]. Recently, ion exchange chromatography using a strongly basic anion exchange resin was identified as a promising method to recover and purify U(VI) from GC solutions [3]. This work describes IX column purification of U(VI) from GC solutions prepared from ε-UO_3 doped with transuranic elements (TRU), lanthanides (Ln), and/or cesium (Cs).

The obtained AmberSep™ 21K XLT resin was initially converted to carbonate form and used for subsequent U(VI) purification. Initial batch testing demonstrated favorable U(VI) uptake with fast kinetics, a Langmuir monolayer homogeneous adsorption mode, and a large uptake capacity for U(VI) in 1 M GC matrix (76.7 ± 1.6 mg U per g of hydrated resin). Six column breakthrough experiments were performed using feed solutions prepared by the dissolution of UO_3 doped with Cs, Eu, Ce, Am, Pu, and/or Np. U(VI) behaved reproducibly in all column experiments demonstrating nearly identical S-shaped breakthrough profiles **(Fig. 1)** indicative of strong interaction with the resin and consistent breakthrough points at ~5.0 – 5.9 bed volumes (BV) achieving full column loading at ~18 – 20 BV. All of the tested dopants showed a lack of retention on the column, within experimental uncertainty. These results indicate high selectivity of the AmberSep™ 21K XLT resin in the carbonate form for U(VI) over TRU and selected fission products in the dynamic column configuration and suggest that anion exchange chromatography is a viable option for achieving effective recovery and purification of U from other elements following alkaline dissolution and/or crystallization.

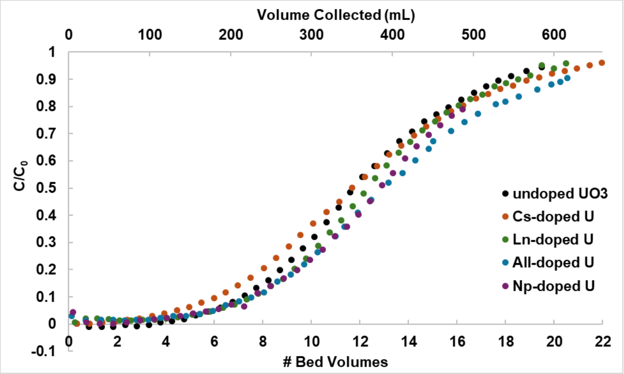

Fig. 1 Breakthrough profile for U(VI) with various dopants.

References

[1] Bessen, N. P. et al. *Dissolution behavior of used nuclear fuel components in carbonate/guanidine/hydrogen peroxide media,* PNNL-36318; Pacific Northwest National Laboratory, Richland, WA, 2024.

[2] Peruski, K. M. et al. *Characterization of powders from NO_2 voloxidation studies*, ORNL/TM-2023/2936; Oak Ridge National Laboratory, Oak Ridge, TN, 2023.

[3] Levitskaia, T. G. et al. *Report on investigation of alkaline-side separation schemes*, PNNL-36311; Pacific Northwest National Laboratory, Richland, WA, 2024.

Loading and Ion Exchange Behavior of Zirconium Phosphate Nanoplatelets

Adrianna L Orsi, *Brahamanage Don Imansha Madhushan'*, *and Jonathan D. Burns*

Department of Chemistry, University of Alabama at Birmingham, Birmingham, AL 35205

A study of the ion exchange behavior of α-zirconium phosphate (α-ZrP) nanoplatelets has been performed. α-ZrP is a layered ion exchange material with two exchangeable protons per unit formula that can be replaced with other positively charged ions. This property and others provide the potential for it to be a radionuclide vehicle, with promising medical applications, both on the diagnostic and therapeutic sides of treatment. For this investigation, α-ZrP was synthesized and converted to the potassium phase (K-ZrP) by reaction with potassium hydroxide. This conversion increased the interlayer distance of the particles, allowing for more rapid ion exchange. The ion exchange properties of the K-ZrP have been studied with a variety of positively charged metal ions in different combinations. When Sc^{3+} and Cr^{3+} were contacted with K-ZrP at pH 1, there was a selective uptake of Sc^{3+} over Cr^{3+}. This selectivity indicates that K-ZrP also has potential as a material for separations. Details of α-ZrP synthesis, conversion to the K-ZrP, and ion exchange will be discussed.

Electrochemically enhanced critical materials separations in 2D COF-MXene membranes

Julia Chung[1], *Felix Gboyero*[2], *John O. Hoberg*[2], *Teng Zhang*[3], *Yury Gogotsi*[3], *and Alex Noy*[14]

[1]Materials Science Division, Lawrence Livermore National Laboratory, Livermore, CA 94550
[2]Department of Chemistry, University of Wyoming, Laramie, WY 82071
[3]Department of Chemistry, Drexel University, Philadelphia, PA 19104
[4]Department of Chemistry, University of California at Merced, Merced, CA 95343

The separation of rare earth elements (REE) is a technical scientific challenge with significant geopolitical implications. Although mining activities take place worldwide, more than 90% of the global supply is processed in China due to the environmental burden of the separations process. To support US domestic processing aligned with national energy and environmental goals, I present here a reusable bench-scale technology that can bind REEs with minimal energetic input. This platform integrates extreme confinement within titanium carbide nanochannels with chemical recognition through covalent organic frameworks to bind REEs. An applied electrochemical potential further modulates adsorption kinetics for precision separations without the use of harsh reagents (Fig. 1). This coupling of nanoconfinement, molecular recognition, and electrochemical control offers a domestically viable route towards REE processing and separations.

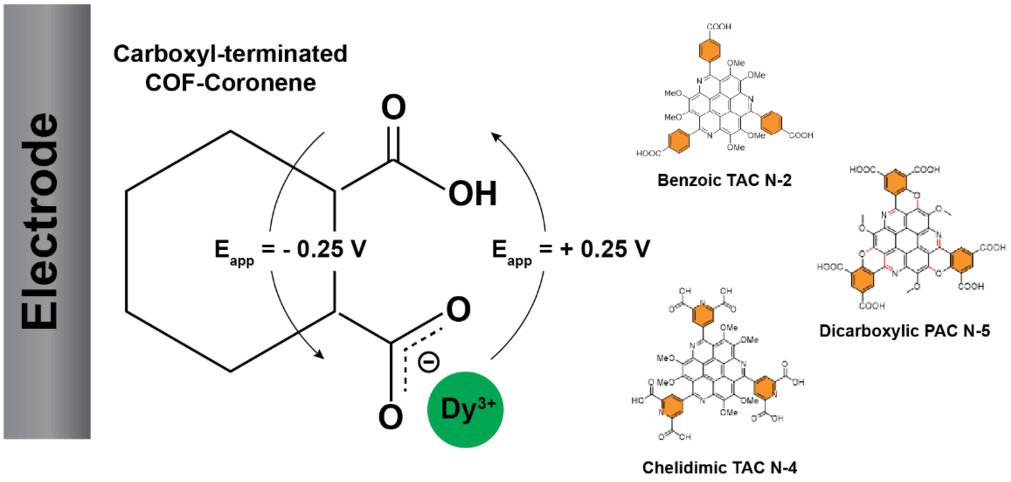

Fig. 1 Schematic of proposed electrochemically-gated adsorption switching.

Analytical Capabilities in the Radioactive Materials Analytical Laboratory

Shalina C. Metzger, *Marc R. Chattin, Haley H. Wightman, Kelly M. McKamey, Selena M. Gilland, Taylor M. Weilert, Adam J. Martin, and Tamara J. Keever*

Radioactive Materials Analytical Laboratory, Oak Ridge National Laboratory, Oak Ridge, TN 37831

The Radioactive Materials Analytical Lab (RMAL) houses a specialized nuclear analytical chemistry group that applies advanced measurement systems and analytical procedures in support of ORNL's science and technology mission. RMAL provides leadership in analytical sciences for the detailed characterizations of materials and chemical processing for the production and distribution of anthropogenic isotopes, detection of radioactive isotopes and EPA-regulated characteristics in the environment, and fission and activation products in bulk irradiated materials related to nuclear fuel. With a highly skilled technical and collaborative workforce, RMAL works to develop new analytical methodologies to solve complex issues and produce quality analytical results in support of big nuclear science projects.

Processing of Elusive f-elements at ORNL: Pm-147, Bk-249, Es-254, and Fm-257

Cristian Celis-Barros[1], *April Miller*[1], *Ashley Harvey*[1], *Nathan Sims*[2], *and Lætitia Delmau*[1]

[1]Radioisotope Science and Technology Division Oak Ridge National Laboratory, Oak Ridge, TN 37830
[2]Isotope Processing and Manufacturing Division, Oak Ridge National Laboratory, Oak Ridge, TN 37830

As the primary producer of rare f-element isotopes for the Western world, Oak Ridge National Laboratory (ORNL) plays a critical role in industrial applications and fundamental research. The lab's main transplutonium product, ^{252}Cf, is used as a neutron source in oil exploration, in initiation of nuclear reactors, in power generation, for detection devices, and in medical research. Additionally, the subproducts ^{249}Bk, ^{254}Es, and ^{257}Fm are in high demand for performing fundamental studies and discovering superheavy elements. Current purification methods rely heavily on established knowledge of these elements, whose chemistry—unlike that of earlier-series members—is predominantly governed by the trivalent oxidation state. Additionally, ORNL recovers another unique f-element isotope, ^{147}Pm, purified from fission products to address a global shortage. The need for a stable supply and more efficient processes has led to the development of improved chemical processing techniques. This work highlights the importance of combining fundamental and applied chemistry to advance current purification processes.

November 3, 2025
Monday Evening

Technical Session III-IV
5:00 – 8:00 p.m.

Lightning Talks, Poster Presentation, and Reception

Chair: Lætitia H. Delmau (Oak Ridge National Laboratory)

1 **Krishna P. Gnyawali**, *Tennessee Technological University*, TODGA-Inspired, Amide Functionalized Bis-1,2,4-Triazinyl-2,6-Pyridine as a Complexant for Minor Actinide Extraction from Lanthanides

2 **Konstantinos Kavallieratos**, *Florida International University*, Lipophilic mono- and bis-Sulfonamidophenol Ligands as f-Element Chelators for High-Level Waste Separations under Alkaline Conditions

3 **Kenneth O. Klutse**, *Oak Ridge National Laboratory*, Crystal Clear: Insights into Rare Earth Separation via a Tunable Ligand

4 **Nathan Sims**, *Oak Ridge National Laboratory*, Recovery and Purification of Bk-249 from the Cf-252 Campaign

5 **Ashley Harvey**, *Oak Ridge National Laboratory*, Processing of Es-253/254 and Fm-257 from the Cf-252 Campaign

6 **April Miller**, *Oak Ridge National Laboratory*, Pm-147 Processing from Fission Products

7 **Daniel G. Racz,** *University of Tennessee-Knoxville*, Improved Adjacent Lanthanide Separation with Diglycolamide Resins and Neutral Aqueous Complexants

8 **Sara E. Speetjens**, *Pacific Northwest National Laboratory*, Robust Lithium Recovery from Lithium-Aluminum Matrices: A Scalable Ion Exchange Approach

9 **Amanda Morgan**, *Indiana University*, Multi-Ion Complexes and Competition from Bulky BArF^- Anions for Chloride Binding in Ion Pairing Conditions

10 **Luigi Gutierrez-Cruz**, *Indiana University*, Sequence-defined Triazolophane Macrocycles: Toward the Binding of ReO_4^- for Dilute Liquid-Liquid Extraction

11 **Lakshi M. Thilakarathne**, *Tennessee Technological University,* Development of Advanced Complexant Frameworks and Solvent-Impregnated Resins for Sustainable Separation of Trivalent Actinides from Lanthanides in Spent Nuclear Fuel

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20 **Samantha Mills**, *Oak Ridge National Laboratory,* Analytical Methods in Support of the Separation and Purification of Pm-147 in FY26

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Notes

Technical Session III-IV
November 3, 2025

Poster Session

Presentation Abstracts

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TODGA-Inspired, Amide Functionalized Bis-1,2,4-Triazinyl-2,6-Pyridine as a Complexant for Minor Actinide Extraction from Lanthanides

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The selective separation of trivalent actinides (An^{3+}) from chemically similar lanthanides (Ln^{3+}) remains a critical challenge in nuclear waste management. Since the introduction of bis-1,2,4-triazinyl-2,6-pyridine (BTP) complexants by Kolarik in 1999, extensive efforts have been devoted to optimizing extractants for liquid–liquid extraction. Here, a streamlined synthetic approach is presented for a new class of amide-functionalized BTP complexants via an alkylation–condensation methodology. Incorporation of amide groups along the aromatic backbone significantly enhances solubility in industrially relevant diluents such as Exxal-8, 1-octanol, and toluene, while simultaneously improving extraction performance. This complexant exhibits high distribution ratios, rapid equilibrations (30 min), and excellent selectivity for $^{241}Am^{3+}$ over $^{154\text{-}152}Eu^{3+}$. Decomplexation studies confirm efficient An back-extraction and demonstrated the reusability of complexants in biphasic separation system. This presentation will focus on the design and synthesis of the complexant, equilibrium study, the influence of acid and complexant concentrations, and decomplexation studies, emphasizing importance for advanced nuclear waste management.

Lipophilic mono- and bis-Sulfonamidophenol Ligands as f-Element Chelators for High-Level Waste Separations under Alkaline Conditions

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Our group has identified lipophilic mono- and bis- *o*-sulfonamidophenols as promising f-element chelators, for Ln(III) and An(III) extraction under alkaline conditions, thus showing potential for applications in High Level Waste (HLW) separations. These ligands are effective chelators due to the electron-rich O- and N- donor sites generated upon deprotonation. In earlier studies, we demonstrated up to 80% single-cycle extraction and recovery of Sm(III), used as a surrogate for Am(III), by disulfonamides from 0.1 - 1.0 M NaOH into chlorinated solvents,[1] and more recently we have expanded this work into lipophilic *o*-sulfonamidophenol ligands that show compatibility with the CSSX solvent in use at the Savannah River Site, in an effort towards a combined Cs(I)/Actinide extraction system. Building on these findings, we now report the synthesis and characterization of five new bis-sulfonamidophenol derivatives, expanding the structural diversity of this chelator class (**Figure1**), as well as further extraction studies with *o*-sulfonamidophenol ligands with the CSSX solvent system.

Extraction studies with *o*-sulfonamidophenols were conducted using either **mm1** (N-(5-(tert-butyl)-2-hydroxyphenyl)-2-ethylhexane-1-sulfonamide) or **mm2** (N-(5-(tert-butyl)-2-hydroxyphenyl)-3-methylbutane-

1-sulfonamide) in the BOBCalixC6-based CSSX solvent system (0.007 M BOBCalixC6, 0.75M Cs-7SB, 0.003 M trioctylamine in 2.0 M NaOH).[2] Contacts were performed at a 1:1 organic: aqueous phase volume ratio (2.0 ml each) containing 20.0 mM of *o*-sulfonamidophenol and 2.0 mM of Sm(III) and/or Cs(I), with phases contacted for 24 h on a rotating wheel (55 rpm) and stripped with 0.1 M HNO_3. Aqueous phases were analyzed with ICP-MS after centrifuge-assisted phase separation. Results showed 34% and 49 % single-cycle recoveries of Sm(III) for **mm1** and **mm2** respectively, with Cs(I) recoveries at 13% and 15%, respectively.

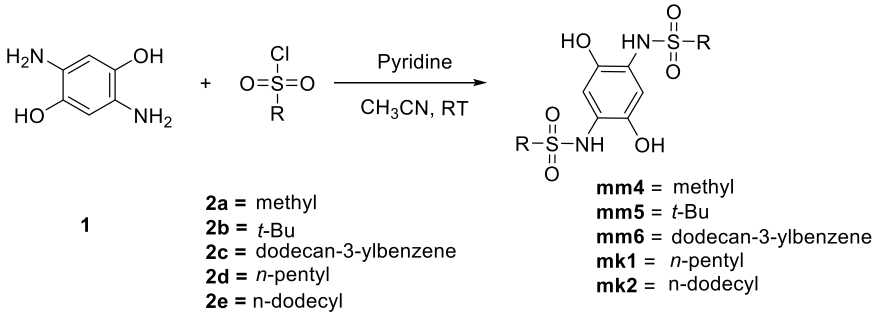

Figure 1. Synthesis of bis(sulfonamidophenol) derivatives

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Crystal Clear: Insights into Rare Earth Separation via a Tunable Ligand

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Rare Earth Elements (REE), with their unique electronic transitions, pronounced magnetic properties, high thermal stability, and versatile catalytic activity, are critical components of technologies ranging from batteries and automobiles to wind turbines and nuclear reactors. As technological innovation accelerates, the demand for REEs continues to rise. However, their naturally low ore deposit grades make cost-effective and environmentally sustainable mining particularly challenging, underscoring the urgent need for efficient recovery and recycling strategies for end-of-life products. To address this need, we have developed preorganized multidentate pincer ligands capable of selectively separating specific REEs through crystallization. Furthermore, we have found that, by fine-tuning separation parameters (e.g., solvent, anions, additives), we can shift ligand selectivity across the Rare Earth series, thus providing an additional level of control over the REE separation process beyond ligand design. Ongoing experiments and characterizations are focused on evaluating the separation efficiency, recovery yield, and degree of differentiation achievable under a diverse range of conditions.

Recovery and Purification of Bk-249 from the Cf-252 Campaign

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Berkelium-249 (Bk-249) is produced and processed at Oak Ridge National Laboratory (ORNL) as a subproduct in the californium-252 (Cf-252) campaigns.[1] The Cf-252 program is the second largest after the production of plutonium-238 (Pu-238) at ORNL. Once the bulk of the Cf-252 is recovered in hot cells, the adjacent heavy actinide subproducts can be further isolated and purified in glove boxes because they are present in significantly smaller quantities. These isotopes are utilized for fundamental research and for the discovery of superheavy elements, including tennessine (Ts), which was synthesized in 2016 by bombarding Bk-249 with Ca-48 ions.[2] From a chemical standpoint, Bk-249 is the only heavy actinide with a relatively accessible tetravalent oxidation state. This key feature enables efficient separation from its redox analogue, cerium, and from the adjacent actinide, californium. Process improvements are continually being evaluated to achieve high radiochemical purity—by removing other radioisotopic impurities—and to eliminate non-radioactive contaminants such as trace transition metals and organic materials. Herein, we present a summary of our efforts to efficiently recover and purify Bk-249 from the latest Cf-252 campaign.

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Processing of Es-253/254 and Fm-257 from the Cf-252 Campaign

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The production of transcurium elements at Oak Ridge National Laboratory (ORNL) is closely tied to the vision of Glenn T. Seaborg. Seaborg advocated for the establishment of facilities capable of producing and processing large quantities of heavy actinides following their discovery in the late 1940s and early 1950s. Over the course of 81 campaigns, ORNL has continued to produce and process these rare actinides as part of the Californium-252 (Cf-252) program for the Department of Energy Isotope Program (DOE IP). Einsteinium (Es) and fermium (Fm) are among the heaviest elements that can be produced by neutron capture and are generated only in sub-microgram quantities. Moreover, the limited understanding of their chemical properties further complicates their isolation and purification. Herein, we present our efforts and challenges toward isolating Es-253/254 from Cf-252 and Fm-257 within the context of the latest Cf-252 campaign at ORNL.

Pm-147 Processing from Fission Products

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The production of promethium-147 (Pm-147) at Oak Ridge National Laboratory (ORNL) addresses the need identified by the Department of Energy Isotope Program (DOE-IP) to reestablish domestic production of this isotope and bridge the gap between supply and customer demand worldwide. Pm-147 ($t_{1/2}$ = 2.62 y) is a low-energy beta emitter with minimal gamma radiation, primarily used in industry for measuring the thickness of thin films, but also in beta-voltaic batteries, and radioisotope thermoelectric generators. Since 2020, ORNL has recovered Pm-147 from Pu-238 fission products, ranging from a few curies (Ci) to over a hundred in a year. This continuous improvement is the result of constant efforts towards bridging the gap between fundamental and applied chemistry. Furthermore, as the demand for this isotope increases, ORNL has identified a new pathway to produce Pm-147 by separating the fission products of Cf-252. This new route and improved process guarantees a continuous supply of this valuable isotope between Pu-238 campaigns. This work showcases some of the mayor achievements and challenges experienced during our latest Pm-147 campaigns.

Improved Adjacent Lanthanide Separation with Diglycolamide Resins and Neutral Aqueous Complexants

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The United States lacks a robust domestic supply for many critical lanthanides (Lns), unable to meet the growing demand for these essential elements in nuclear medicine, clean energy technologies, and space exploration. In nuclear medicine, isotopes such as ^{152,153}Eu, ^{161}Tb, ^{161}Dy, ^{177}Lu, play a pivotal role in targeted radiotherapy, diagnostic imaging, and theragnostics, due to their unique nuclear, chemical, and physical properties that enable precise treatment and detection of disease. The production of many of these isotopes relies on irradiating an element one atomic number below the target isotope—for example, neutron irradiation of ^{176}Yb to produce ^{177}Lu [1]. However, one of the formidable challenges for separating neighboring or adjacent Lns is due to their very similar ionic radii and predominant +3 oxidation state [2, 3]. While there are existing industrial-scale processes for separating Lns [2], they lack the necessary selectivity for efficient, rapid, and cost-effective methods for medical isotope production. This study investigates a solid-liquid separation technique using diglycolamide (DGA) resin, a lipophilic neutral ligand, as a stationary phase, combined with neutral water-soluble complexants as back-extracting agents to separate adjacent lanthanide pairs.

The complexant employed includes dioxaoctanediamide (DOODA), as this ligand demonstrates a higher affinity for lighter lanthanides, contrasting with the behavior of DGA's performance. Two DGA resins were evaluated: commercially available N,N,N',N'-tetraoctyl diglycolamide (TODGA) and a novel resin based on 2,2'-oxybis(N-octyl-N-(3,5,5-trimethylhexyl)acetamide) (SDGA). Extending the concept of a two-ligand separation strategy for Ln separation that has been demonstrated in liquid-liquid systems [4], we aim to enhance the separation of adjacent Lns while employing neutral lipophilic and hydrophilic ligands in a continuous, closed loop separation system. This work focuses on maximizing the separation of specific lanthanide pairs—Eu-Gd, Tb-Dy, Yb-Lu, and Gd-Tb—by varying conditions of lanthanide concentration, nitrate concentrations, and aqueous ligand concentration using a single column run.

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Robust Lithium Recovery from Lithium-Aluminum Matrices: A Scalable Ion Exchange Approach

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The efficient separation of lithium (Li) from lithium-aluminum matrices is essential for evaluating Li isotopics from irradiated materials, which is key to advancing nuclear applications for ceramic materials. To enable high precision isotopic measurements, tritium must first be removed to render samples radiologically manageable, while preserving the unfractionated ^{6}Li/^{7}Li ratio through quantitative recovery of Li, as even a small loss of Li to the columns will likely lead to isotopic fractionation. Here we report the first application of a strong macroporous cation exchange resin (AG MP-50) column scheme to nuclear materials for this purpose using a method modified from Zhu et al. 2020. Using a 0.5 x 5 cm column configuration, lithium recovery consistently approached 100%. Across all tests, the absence of leaching or contamination during extended washing (up to 70 resin bed volumes) demonstrated the resin's reliability in retaining lithium and ensured robust, reproducible isotopic measurements. Lithium isotopic measurements by thermal ionization mass spectrometry (TIMS) demonstrated no fractionation between ^{6}Li and ^{7}Li. This separation approach provides a reliable, high-yield pathway for tritium decontamination of lithium-aluminate matrices prior to isotopic analysis. Its successful demonstration on nuclear materials for the first time establishes a new, validated method for preparing TPBAR samples for high-precision lithium isotopic characterization.

These findings affirm the process's reliability and suitability for applications requiring precise lithium recovery and isotopic stability. Future efforts will refine tolerances for tritium decontamination to further validate the method's effectiveness.

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Multi-Ion Complexes and Competition from Bulky BArF⁻ Anions for Chloride Binding in Ion Pairing Conditions

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Cationic charges have long been used to enhance anion binding. Embedding charge introduces strong ion pairing for target anions but also for off-target ions, ultimately generating a mixture of multi-ion species that are hard to identify and quantify. While many sidestep this problem using polar solvents and weakly-coordinating ions, these approaches exclude a substantial cross section of conditions found in applications spanning recognition, assembly, separations, templation, and catalysis. To confront this complexity, we study anion binding to a cationic receptor featuring low shape complementarity in a low-polarity solvent to maximize ionic interactions. We prepared the receptor as a salt of the weakly-coordinating tetrakis[3,5-bis(trifluoromethyl)phenyl]borate (BArF⁻) anion and studied binding of small chloride (Cl^-) and medium sized iodide (I^-) anions. Surprisingly, use of the bulky BArF⁻ anion does not suppress ion pairing interactions, with 65% of the receptor being paired at 0.5 mM in dichloromethane. We observe multi-ion receptor-Cl^- complexes (2:1, 1:1, 1:2), reinforcing the complexity that emerges when working in low-polarity media. We reveal the dependence of affinity on anion charge density and size, and that bulky BArF⁻ counter anions compete for chloride binding. These studies reveal the non-innocence of BArF⁻ anions, and strategies to quantify multi-ion species.

Synthesis of Sequence-defined Triazolophane Macrocycle Toward the Binding of ReO_4^- for Dilute Liquid-Liquid Extraction

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Anions are integral components of the chemical cycles that govern natural and human systems. For instance, nuclear power production generates complex aqueous media containing the radioactive anion pertechnetate, amongst other radionuclides, that needs to be separated and safely immobilized. For this purpose, liquid-liquid extraction is a useful separation technology that benefits from using synthetic receptors for the selective capture of pertechnetate at low concentrations in competitive solutions. To address this need, we designed a receptor in the form of a rigid triazolophane macrocycle bearing an elliptical cavity lined with CH bond donors to selectively bind pertechnetate. The macrocycle was obtained using a sequence of reactions involving nitration, benzylamine condensation, Sn(II) reduction, azidation of 1,8-naphthalic anhydride, and installation of two alkynes in the 2 and 6 positions of pyridine. The key CH donors are introduced by building block selection with the electron deficient naphthalimides and using Cu(I)-catalyzed azide–alkyne cycloaddition chemistry to install polar 1,2,3-triazole units. The sequence of naphthalimide, pyridine and triazole units inscribe alternating dipoles around the macrocycle to promote cooperative high fidelity 2:1 sandwich-like dimer complexes. These complexes display remarkable femtomolar scale sensitivities ($\beta 2 \sim 10^{14}$ M^{-1}, CD_2Cl_2) toward the pertechnetate surrogate, perrhenate. We will present the synthesis and preliminary binding studies. These findings highlight the potential towards liquid-liquid extraction of radioactive pertechnetate from competitive media.

Development of Advanced Complexant Frameworks and Solvent-Impregnated Resins for Sustainable Separation of Trivalent Actinides from Lanthanides in Spent Nuclear Fuel.

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Nuclear energy is renowned for producing carbon-free power with minimal atmospheric pollution. However, the highly radioactive waste generated from spent nuclear fuel (SNF), particularly due to the presence of minor actinides, poses a significant challenge to sustainable and effective management of SNF. The selective separation of trivalent minor actinides (An^{3+}) from chemically similar lanthanides (Ln^{3+}) remains one of the most pressing issues in the nuclear fuel cycle. Previously, bis-1,2,4-triazinyl-2,6-pyridines (BTPs) have been successfully applied in liquid-liquid extraction (SANEX-type), providing a foundation for the development of new complexant frameworks, such as bis-1,2,4-triazinyl-2,2'-bipyridines (BTBPs), bis-1,2,4-triazinyl-1,10-phenanthrolines (BTPhens), and terpyridine-based bis-1,2,4-triazine, 3,3'-[2,2':6',2''-terpyridine]-6,6''-bipyridines (TerPy-BTPs) as a key new strategy aimed at improving solubility, selectivity, and overall separation efficiency. While these advances enhance liquid - liquid extraction performance, challenges still exist, including the use of volatile organic diluents, secondary waste generation, and limited scalability. To address these limitations, research has focused on solvent-impregnated resins as a greener, and more scalable alternative, with current studies on BTP-based resins and future directions expanding to advanced complexants, including BTBPs, BTPhens and TerPy-BTPs. Collectively, this research integrates complementary projects at various stages, including the synthesis and optimization of BTBP, BTPhen, and TerPy-BTP complexants for liquid–liquid extraction, along with the development and evaluation of BTP-based resins for selective separations, current results and future directions will be presented.

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Progress Towards Synthesis of Unsymmetric Pyridyl-1,2,4-Triazine Complexant Scaffolds Via Tandem Metal-Catalyzed Borylation/Cross-Coupling For Application to Minor Actinide Separation

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The selective separation of minor actinides from lanthanides in spent nuclear fuel (SNF) is a key challenge in nuclear waste management. Nitrogen-rich bis-1,2,4-triazinylpyridines (BTPs) are effective soft N-donor ligands for this purpose, but widespread application has been hindered by poor solubility, hydrolytic instability, and synthetic challenges. A modular synthetic strategy is being developed to access unsymmetric pyridyl-1,2,4-triazine scaffolds through tandem metal-catalyzed borylation and Suzuki–Miyaura cross-coupling. Selective triflation of 3-hydroxy-1,2,4-triazines provides key intermediates, which are subjected to undergo Pd-catalyzed heteroaryl coupling. Current efforts have focused on optimizing Buchwald-type ligands (XPhos, RuPhos, DavePhos, XantPhos, SPhos), bases, and solvents. In parallel, Kumada and Negishi cross-coupling strategies are being investigated as complementary routes for pyridyl introduction. Initial studies confirm that pyridylmagnesium and pyridylzinc intermediates can participate in Pd-catalyzed couplings, though stability and conversion remain strongly condition-dependent. This presentation will highlight progress towards the synthesis of unsymmetric pyridyl-1,2,4-triazine complexant scaffolds using tandem borylation/cross-coupling strategies and discuss prospective application for minor actinide separation.

Synthesis of Aqueous Bis-lactam-1, 10-phenanthroline for Lanthanide Separation

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The escalating demand for lanthanides has driven the development of structurally precise, electronically tuned small-molecule ligands capable of selectively coordinating f-block elements by resolving subtle differences in ionic radii and coordination behavior across the lanthanide series. Our group here at Oak Ridge National

Laboratory has spent the past several years advancing this objective by developing synthetic strategies and molecular design principles that afford ligands with preferential affinity for specific subsets of lanthanides, thereby enabling the targeted separation of light and heavy lanthanide ions. Building on these efforts, we now report the synthesis of a water-soluble bis-lactam-1,10-phenanthroline ligand that embodies this design philosophy – coupling architectural rigidity with modular arrangement of donor atoms to achieve selective coordination. The route centers on a convergent strategy culminating in a late-stage Rh(III)-catalyzed C–H annulation, which forges a lactam directly onto the phenanthroline framework. This transformation proceeds via carbamate-directed C–H activation, alkyne insertion, and a concerted metalation-deprotonation pathway under redox-neutral conditions. The resulting tetradentate N, O-donor-based motif offers a preorganized coordination environment with high aqueous solubility, optimized for size-selective binding across the lanthanide series. The aqueous solubility derives in part from the polar functional groups introduced through strategic selection of alkyne coupling partner, which incorporates hydrophilic substituents into the ligand framework.

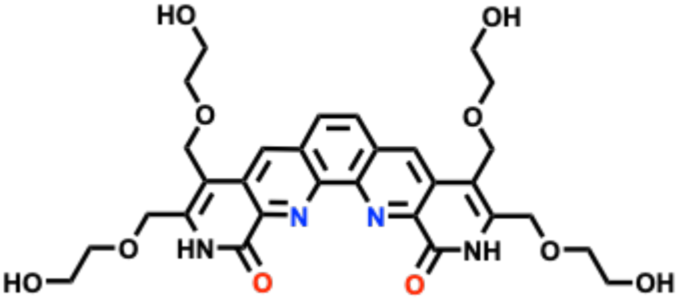

Figure 1: Chemical structure of aqueous bis-lactam-1,10-phenanthroline

Progress Towards the Hofmann Rearrangement of Heteroaryl Carboxamides to Afford Heteroaryl Amino Complexant Synthons

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Nuclear fuel is an extremely advantageous energy source. However, one of the drawbacks includes the waste produced, spent nuclear fuel (SNF). Current and past research focuses on developing soft Lewis basic complexants to assist in the separation of actinides and lanthanides via the SANEX process. Successful development of soft Lewis basic complexants that separate actinides from lanthanides has been accomplished, but work continues in hopes of making these complexants conveniently transferable to industry. The separation of the actinides from lanthanides would greatly decrease the radioactivity of the SNF and allow for easier storage. One of the goals of this specific project is to obtain a heteroaryl amine from a heteroaryl carboxamide via the Hofmann rearrangement. The precursor has been synthesized via the Suzuki-Miyaura cross coupling and was reacted via the Hofmann rearrangement to afford the heteroaryl amine. The current synthetic route was established after several attempts of placing amino groups on N-heteroarenes via the Minisci reaction proved to be unsuccessful. Experimental conditions and mitigation strategies for the

Hofmann rearrangement will be discussed. The overall goal of this project is to obtain the amine to facilitate construction of advanced complexants.

Zirconium Tetrachloride ($ZrCl_4$) Recovery Utilizing Advanced Chlorination, Sedimentation, and Microfiltration

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To enhance the sustainability of nuclear energy, we need new separations to recover materials from spent nuclear fuel and enable the continuous use of these materials in a closed nuclear fuel cycle. Cladding comprises 25% of nuclear fuel by mass, making it a worthwhile target for research on used fuel recycling. Several zirconium-rich alloys are commonly used as cladding, and a variety of methods for recovering this Zr have been proposed, including advanced chlorination. Advanced chlorination readily transforms solid Zircaloy cladding into a suspension of chlorinated metal (especially, $ZrCl_4$) particles in a chlorinating solvent, commonly thionyl chloride or sulfur monochloride (S_2Cl_2). Performing advanced chlorination at scale in a cost-effective manner requires the continuous recovery of the chlorinating solvent so that it can be returned to the reactor and reused. Recycling the solvent is difficult due to substantial amounts of $ZrCl_4$ and other particles, which can cause processes failures and shutdowns by clogging or abrading auxiliary equipment.

We are investigating a new strategy to facilitate mechanically simple, continuous recovery of both zirconium particles and chlorinating solvents during advanced chlorination of Zircaloy cladding. This strategy uses gravitational settling to remove larger particles from the suspension; finer particles are recovered by passing the solvent through a microfiltration membrane at low-velocity and in an upward flow direction. This minimizes the exposure of the membrane surface to particles that could foul it. In addition, the modest pressure (< 70 kPa) required to drive solvent flux through the microfiltration membrane is provided by elevating the fluid level in the reactor above the membrane module so that gravitational head at feed side of the membrane is sufficient, obviating the need for a pump.

Here, we present preliminary results from our bench-scale study of this strategy. We dissolved Zircaloy-4 in S_2Cl_2, characterized the resulting suspension and particles, and measured membrane separation performance using a commercial polytetrafluoroethylene (PTFE) microfiltration membrane. We measured settling velocities of the particles and used Stokes Law together with these data to gain insight into particle sizes. Particle morphology and composition were analyzed via optical microscopy, scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), and micro-Raman microscopy. A custom membrane testing cell was designed and fabricated to mimic membrane separation conditions at full-scale. We used this cell to measure the flux of both pure S_2Cl_2 and suspensions of Zircaloy-4 dissolved in S_2Cl_2 through the membrane with 10 psi (69 kPa) of driving force. The accumulation of particulate on the membrane during these experiments was also assessed via SEM and EDS. Overall, these data indicate strong potential for the combined sedimentation-microfiltration approach to enable continuous advanced chlorination of Zircaloy cladding and enhance the sustainability of the nuclear fuel cycle.

Acknowledgement: This research was supported by an appointment to the Oak Ridge National Laboratory Graduates in Engineering and Materials Science Internships (GEM) sponsored by the U.S. Department of Energy (DOE) and administered by the Oak Ridge Institute for Science and Education, as well as the Office of Nuclear Energy's (DOE-NE) Material Recovery and Waste Form Development Campaign under the Nuclear Fuel Cycle and Supply Chain (NFCSC) Program.

Separation and Recovery of Rare Earth Elements from EV magnets using Membrane Solvent Extraction System

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Rare earth elements (REEs) particularly dysprosium (Dy), praseodymium (Pr), and neodymium (Nd) have drawn an urgent attention worldwide and have become strategically critical for the security of global economy, energy, transportation, and communications. Majority of REEs are imported and are not domestically produced in United States. This puts the entire supply chain of REEs at a large geopolitical risk. On the other hand, widespread applications of REEs in different devices such as portable electronic and especially hybrid/electric vehicles (EV) generate millions of tons of end-of-life (EOL) waste containing metallic contaminants. Recycling REEs from EOL EV magnets can help in mitigating the supply side risk of REEs and can also address the environmental challenges arising due to EV waste containing metallic contaminants. In this work, we report the separation and recovery of high purity REEs form multiple EoL EV magnets with varying composition using scalable, energy-efficient, and environmentally friendly membrane solvent extraction (MSX) system. In MSX, extractant is immobilized inside the pores of hollow fibers and it interacts selectively with the target elements by making a complex and thus facilitates the separation of the target elements. In the first step of MSX, REEs (Dy, Nd, and Pr) are separated from non-REEs (Iron, Copper, Zinc, and Boron etc.) with >99.5 wt.% purity and >95% recovery using diglycolamide as an extractant. Thereafter, in second step, heavy REE (Dy) is separated from light REEs (Nd and Pr) using phosphonic ion based extractant with >99.9 wt.% purity and >95% recovery. The purity of the recovered product was further confirmed with additional characterization such as SEM, EDS, and XRD. Overall, MSX technology provides a potential pathway for industrial deployment to domestically produce of REEs in United States.

Keywords: Rare earth elements, EV magnets, Scalable process, Energy-efficient, MSX

Novel Membrane Solvent Extraction Process for Recovering High-Purity Rare Earth Elements from US Mine Tailings

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Mining feedstocks often contain rare earth elements (REEs) such as neodymium (Nd), cerium (Ce), dysprosium (Dy), and praseodymium (Pr). These REEs are essential for advanced electronic technologies and US national defense, yet domestic production remains limited while China continues to dominate global supply. This dependence presents significant geopolitical and economic risks due to potential supply chain disruptions.

In this work, we demonstrate the separation and recovery of high-purity REEs from U.S. iron mine tailings using a scalable, energy-efficient, and environmentally friendly membrane solvent extraction (MSX) process. In the MSX system, the extractant is immobilized within the pores of hollow fibers, where it selectively complexes with target REEs to facilitate their separation. Starting with feed material containing 2.4 wt.% REEs, a stepwise leaching process enriched the concentration to 12 wt.% for subsequent separations. In the first separation stage, heavy REEs (Dy and Y) were recovered from non-REE impurities (e.g., Ca, Fe) with > 88 wt.% purity, while most remaining impurities were phosphorus-based. In the second stage, light REEs (La, Ce, and Nd) were separated with > 97 wt.% purity. These high-purity recoveries were achieved through the novel process design and optimized separation process conditions.

Keywords: Rare earth elements, mine tailings, membrane solvent extraction

Combined Electrocoagulation and Activated Carbon Treatment of Farm Wastewater

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Farm wastewater contains high levels of pollutants such as chemical oxygen demand (COD), total organic carbon (TOC), total nitrogen (TN), and other chemical contaminants that can cause substantial environmental threats. This study focuses on using iron electrocoagulation (EC) in treating farm wastewater and exploring the possibility of reclaiming it to use as irrigation water and livestock feed water. Significant removal of COD, TOC, turbidity, and ions including calcium, sulphur, and silica was achieved by the EC process. Specifically, more than 95% of TOC and COD removal was achieved by using a charge loading of ~37,500 C/L in a five-electrode (2-anode, 3-cathode) EC configuration followed by ultrafiltration. Electrochemically generated iron hydroxide from the sacrificial electrodes produced flocs through adsorption and coagulation of organic and inorganic pollutants during the EC process, leading to high removal efficiency of pollutants. SEM-EDS and FTIR characterization of produced flocs and foam provided evidence of ion removal including calcium, sulphur, and silica. Conversion of nitrogen to ammonia through intermediate conversion of nitrate/nitrite was another important finding. Additionally, the simultaneous Fenton reaction occurring during the Fe-EC process facilitated successful inactivation/removal of microorganisms (>99.9% of bacteria). Activated carbon treatment performed on post EC treated water effectively reduced COD (to ~10 ppm), TOC (to ~10 ppm), color, and odor. Thus, the combined approach of EC, ultrafiltration, and adsorption by activated carbon led to effective treatment of the highly polluted dairy-farm water.

Mechanistic Mass Transfer Model for AI-Driven Design of Membrane Solvent Extraction System

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Membrane solvent extraction (MSX) offers key advantages over conventional mixer–settler SX for critical mineral recovery, including continuous operation, reduced solvent loss, and enhanced phase stability. However, industrial adoption has been hindered by the absence of predictive tools that couple interfacial chemistry, transport resistances, and module-scale design. Existing empirical or lumped-parameter models cannot generalize across extractants, pH regimes, or module geometries—limiting their usefulness for process scaling or AI-driven optimization.

This work presents a mechanistic mass-transfer formulation suitable for direct integration into a physics-informed AI design framework. The model decomposes transport resistances across the feed boundary layer, membrane pores, and strip side, while capturing asymmetric partitioning arising from pH-dependent complexation equilibria and extractant stoichiometry (e.g. D2EHPA and CYANEX 272). By explicitly retaining these physical constraints, the formulation enables hybrid coupling to machine-learning layers that can learn non-idealities (e.g., shear-dependent effective diffusivities, extractant aging, membrane wetting dynamics) without sacrificing extrapolation accuracy.

Embedding this mechanistic model as the “physics backbone” within an AI surrogate enables a new predictive design tool that can:

- Recommend optimal membrane area and module configuration for a given chemistry

- Predict the operating time of MSX for a given chemistry

This hybrid mechanistic+AI architecture creates a pathway to rational MSX system design—supporting module scaling, parameter tuning, and autonomous process optimization for critical mineral recycling.

Analytical Methods in Support of the Separation and Purification of Pm-147

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In FY26, the Oak Ridge National Laboratory (ORNL) is pursuing full scale production of plutonium-238 through the irradiation of aluminum-doped neptunium oxide (NpO_2) pellets in ORNL's High Flux Isotope Reactor (HFIR) and Idaho National Laboratory's (INL) Advanced Test Reactor (ATR). The chemical processing of the irradiated targets is completed at the Radiochemical Engineering Development Center (REDC) at ORNL. The Pu-238 Production Program is a Department of Energy (DOE) program focused on the domestic production of Pu-238 to fuel radioisotope power systems for NASA space programs. One of the byproducts of the plutonium-238 production program is promethium-147, a rare isotope used in radioisotope batteries for satellites and space probes, and to accurately measure the thickness of very thin polymer films.

The Transuranium Analytical Laboratory (TAL) leads the analytical effort at ORNL in support of all phases of the Pu-238 Production Program, the separation and purification of Pm-147 from the fission product waste solutions. The TAL provides quick-turn analysis on process samples, in most cases providing same day service, and high precision characterization of Pm-147 product. The TAL utilizes proportional counting, alpha spectrometry, liquid scintillation, gamma spectrometry, and ICP-MS determinations for process and product sample analysis. Pm-147 production is moving to full-scale production as well with three campaigns scheduled for FY26. An overview of the analytical methodology employed for the characterization of Pm-147 will be presented.

Investigating the Separation and Determination of Fluoride in a High Actinide Matrix

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Legacy Neptunium Oxide material from Savannah River National Laboratory dating from 1986 is used in the assembly of aluminum clad Neptunium-237 target rods at the Radiochemical Engineering Development Center for use by the Plutonium-238 program at Oak Ridge National Laboratory. After examination of the analysis report performed on the original material in 1986, it was discovered to contain approximately 80 ug/g of fluoride. The possibility that the fluoride present in the neptunium oxide has been concentrated in the waste and dissolution tanks is a potential problem in that in a highly acidified environment the fluoride would be converted into hydrofluoric acid. If formed, hydrofluoric acid will likely cause corrosion to any containment vessel not specifically designed to resist it. With limited to no ability to visually assess the storage tanks, or the lines leading from them, it is imperative that the fluoride content be quantitatively determined. Different avenues were and are being explored to definitively quantify this potential fluoride separately from its high

neptunium matrix, or in solution. During the scope of this investigation methods including ion chromatography, ion specific electrodes, and colorimetry were tested for their accuracy, and alternate methods considered. Complicating factors for analysis were the high acid molarity of the solutions and the high activity due to the concentration of neptunium, additionally the lack of homogeneity in the original solution presented its own challenges. After extensive testing it was concluded that given available equipment, anion exchange ion chromatography still represented the best technique for fluoride quantification without column separation, after establishing acid molarity and neptunium concentration guidelines to minimize impact on results and allow for acceptable detection limits. Further investigation into other analytical methods, including the installation of a Combustion Ion Chromatography instrument (CIC), as well as the use of Nuclear Magnetic Resonance (NMR) are being considered.

November 4, 2025 Technical Session V
Tuesday Morning SNS, Iran Thomas Auditorium

Solvent Extraction

Chair: Santa Jansone-Popova (Oak Ridge National Laboratory)

8:15 a.m. Understanding the Kinetics of Lanthanide(III) Extraction from Nitric Acid Media by TODGA with and without Phase Modifiers
Wyatt S. Nobley *Colorado School of Mines*

8:40 a.m. Partitioning of Np and Tc in the Direct Extraction Process Using DEHiBA Solvent
Peter Tkac *Argonne National Laboratory*

9:05 a.m. Investigation of Rare Earth Solvent Extraction Separation from Monazite Feed
Tatiana Levitskaia *Pacific Northwest National Laboratory*

9:30 a.m. Solvent Extraction Flowsheet Optimization of a Pm-147 Surrogate in N,N,N',N'-Tetraoctyl Diglycolamide Using the Argonne Model for Universal Solvent Extraction (AMUSE) Code
Clarice Phelps *Oak Ridge National Laboratory*

9:55 a.m. Break

10:15 a.m. Direct Extraction of Uranium Oxides with Monoamide Ligands
Amy Speelman *Pacific Northwest National Laboratory*

10:40 a.m. Extraction and Light-Driven Release of Superhydrophilic Oxoanions Using Amphiphilic Iminoguanadine Ligands
Nabarupa Bhattacharjee *Oak Ridge National Laboratory*

11:05 a.m. Characterization of f-Element Complexes with Hydroxypyridinone Chelators for Redox Stabilization and Extraction
Marlom Martinez *Florida International University*

11:30 a.m. Lunch

Afternoon Free for individual discussions

Notes

Technical Session V
November 4, 2025

Solvent Extraction

Presentation Abstracts

Notes

Understanding the Kinetics of Lanthanide(III) Extraction from Nitric Acid Media by TODGA with and without Phase Modifiers

Wyatt S. Nobley[1]*, Johnathan J. Klein*[1]*, and Mark P. Jensen*[1,2]

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Continued development of modern technologies and a push toward renewable energy have placed a critical mineral tag on several of the lanthanide elements. Their unique properties are highly sought after, but their 4*f*-block chemical similarity makes trivalent lanthanides difficult to separate from one another. Solvent extraction is a valuable technique used to separate lanthanide cations, and one of the most promising extractants is *N,N,N',N'*-tetra(*n*-octyl)diglycolamide (TODGA). TODGA/alkane solvents have a propensity to undergo third phase formation, limiting their usefulness, but implementing phase modifiers (PMs) into the organic phase has proven to be an effective way to prevent phase-splitting under relevant conditions. PMs are amphiphilic, interfacially active, and often added in far greater concentrations than TODGA, yet their fundamental chemical impacts on TODGA-based lanthanide(III) solvent extraction are not well understood.

This work focusses on our findings regarding the equilibrium and kinetic aspects of the unmodified TODGA/*n*-dodecane extraction system, which provide a greater understanding of the bulk complexation and the interfacial mass transfer mechanism. Solvent extraction and *operando* UV/Vis spectroscopy were used to investigate the system thoroughly. The equilibrium was interpreted using an activity-based thermodynamic model, and the rate determining steps were identified in both directions of mass transfer. The mechanism of Nd(III) extraction was determined to follow a complex, interfacial two-step consecutive reactions mechanism involving multiple TODGA molecules, nitrate anions, and nitric acid adducts.

Furthermore, we explored the effects of adding two commonly used PMs, *N,N*-di(*n*-hexyl)octanamide (DHOA) and tri(*n*-butyl)phosphate (TBP), to the organic phase. While DHOA was determined to be essentially non-interacting, TBP was incorporated into the extracted complex and significantly altered the contribution of TODGA and nitrate species in the rate determining processes at the interface. Not only were the extraction and stripping mechanisms altered, but the rate of mass transfer was greatly affected by TBP as a modifier, relative to the unmodified phase. These findings highlight the ramifications of phase modification beyond the intended purpose of preventing third phase formation. This work provides a large step forward in our understanding of phase modification in trivalent lanthanide extractions by TODGA.

Partitioning of Np and Tc in the Direct Extraction Process Using DEHiBA Solvent

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The use of DEHiBA (N,N-di-(2-ethylhexyl)isobutyramide) solvent in the GANEX-1 (Group Actinide Extraction) process involves selectively separating uranium from dissolved used nuclear fuel. This requires an initial dissolution of the used fuel in an acidic aqueous phase, followed by contact with the DEHiBA solvent to extract U(VI). In the typical GANEX-1 process, Np(VI) and Tc co-extract with U(VI), necessitating additional purification steps to obtain a pure uranium stream.

In contrast, the direct extraction approach involves dissolving oxidized used fuel directly into the DEHiBA solvent without the need for an aqueous phase. This direct extraction method could potentially simplify the process by eliminating the need for initial acid dissolution and reducing the generation of secondary waste streams. To minimize radiation degradation of the organic solvent, which can reduce its extraction efficiency and selectivity, the dissolution process should be quick, ideally taking only minutes. To address these concerns, it's important to enhance the dissolution kinetics by optimizing the voloxidation conditions and solvent composition, ensuring efficient and selective dissolution for the recovery of uranium.

In this work, the focus was on the direct extraction approach following advanced voloxidation of UO_2 material containing Np, Tc, and other fission product surrogates using NO_2 gas. Experimental results discussing the partitioning behavior of Np, Tc, and selected fission products during direct extraction with varying DEHiBA concentrations at different uranium loading will be presented.

Investigation of Rare Earth Solvent Extraction Separation from Monazite Feed

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Separations of the rare earth element (REE) remain an outstanding challenge, accounting for the major cost in producing purified individual REEs from ores. The current technology for industrial REE separations utilizes solvent extraction with phosphorus-based acidic extractants, which are highly effective in partitioning of the bulk REE into organic aliphatic diluent but suffers from insufficient selectivity to discriminate individual REE. This results in a complex multistage process notorious for its excessive chemical consumption, wastewater effluents, and hundreds of processing steps required to produce individual purified REEs.

Alternative separation approach is currently under development where first bulk REE are extracted into the organic phase using a neutral diglycolamide (DGA) extractant followed by the selective scrubbing of individual or adjacent pairs of REE. The selectivity is achieved via modulation of the salt concentration in the scrub and addition of the aqueous REE dioxaoctanediamide complexant (DOODA). We also investigate whether a combination of neutral DGA and organophosphorus acidic extractant could offer additional selectivity modality through pH variation by manipulating acidity of the aqueous scrub. We demonstrated that the organic solvent comprised of (2-ethylhexyl)phosphonic acid mono-2-ethylhexyl ester (HEH(EHP) and DGA in the aliphatic diluent tolerates high concentration of REEs. High loading of this extraction phase has been achieved when using the feed material derived from the monazite ore. The aqueous buffer was used to mediate the ion exchange of HEH(EHP), allowing complete extraction of REEs from aqueous to organic. Our current research is focused of the development of the sequential stripping of the light REE accomplished by either varying the salt content, DOODA concentration, and/or acidity of the aqueous scrub solutions.

Solvent Extraction Flowsheet Optimization of a ^{147}Pm surrogate in N,N,N',N'-Tetraoctyl Diglycolamide Using the Argonne Model for Universal Solvent Extraction (AMUSE) Code

Clarice E. Phelps[1,2], *Zachary C. Odom*[3], *Kiera Blevins*[3]

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Promethium-147 (^{147}Pm) is a low beta-emitting isotope (E_β, avg = 62 keV) that is used for many applications such as radioisotope thermoelectric generators (RTGs) and thickness measurement gauges. Irradiation of neodymium-146 (^{146}Nd) in the High Flux Isotope Reactor at Oak Ridge National Laboratory offers an alternative production route for ^{147}Pm via the ^{146}Nd(n,γ)^{147}Nd ($t_{1/2}$ = 10.98 days, $\beta^- \rightarrow {}^{147}$Pm)[1] reaction, which could supplement the current methods of Pm recovery from plutonium-238 (^{238}Pu) raffinate.[2] Post irradiation removal of the bulk Nd would be required to purify the Pm from the irradiated target before column chromatography separations. Previous work to optimize the solvent extraction process was performed by studying the distribution coefficients (K_d) in various organic extractants using a samarium (Sm) surrogate. Like most adjacent lanthanides, Pm shares similar atomic properties with Sm, such as a +3 valence state and similar atomic radii (Pm: 2.62 Å, Sm: 2.59 Å).[3] Therefore, estimated K_d values for Pm were based on data obtained for Nd and Sm K_d values, with Pm K_d values currently being determined experimentally. However, additional data would need to be obtained for K_d values under various conditions to determine the ideal solvent extraction parameters between the adjacent lanthanide species. For this work, N,N,N',N'-tetraoctyl diglycolamide (TODGA), which has been shown to be effective in adjacent lanthanide separations,[4] was used to model and optimize adjacent lanthanide extraction behavior at temperatures from 20°C to 40°C for Nd and Sm. Using the Argonne Model for Universal Solvent Extraction (AMUSE), the behavior of the ^{147}Pm and ^{147}Nd in the solvent extraction process can be predicted using the determined K_d values of natural Nd and the Pm surrogate, Sm. By varying the AMUSE code parameters to reflect solvent extraction conditions, the percent recovery and purity of the ^{147}Pm can be determined.

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Direct Extraction of Uranium Oxides with Monoamide Ligands

Amy L. Speelman[1]*, Nathan P. Bessen*[1]*, Daria Boglaienko*[1]*, Laura E. Maggos*[2]*, Candido Pereira*[2]*, Gregg J. Lumetta*[1]*,*
and Gabriel B. Hall[1]

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During conventional reprocessing, used nuclear fuel is dissolved in hot nitric acid and the desired components are recovered using solvent extraction. Significant process intensification could potentially be achieved by instead directly dissolving voloxidized uranium oxides into a process solvent. This approach offers several potential advantages over conventional reprocessing, including reducing reprocessing plant footprint by combining the dissolution and extraction operations, performing dissolution under milder conditions less corrosive towards plant infrastructure, and reducing the generation of secondary nuclear waste. In this presentation, the direct extraction of ε-UO$_3$ simfuels by monoamide ligands, primarily 1.5 M *N,N*-di(2-ethylhexyl)isobutyramide (DEHiBA) in *n*-dodecane, will be discussed. Distribution ratios of metals between nitric acid solution and loaded organic phases generated via direct extraction will be presented and compared with those from conventional forward solvent extraction. These distribution ratios were used to design a flowsheet to scrub and strip the loaded organic phase while generating lower volumes of high-level waste and reducing the HNO$_3$ requirements compared to a traditional solvent extraction flowsheet. Additional experiments examining the impact of organic phase nitric acid concentration, reaction time, ligand concentration, and monoamide ligand identity on direct extraction of simufels will also be presented.

Extraction and Light-driven Release of Superhydrophilic Oxyanions Using Amphiphilic Iminoguanidine Ligand

*Nabarupa Bhattacharjee, Radu Custelcean, and Jeffrey D. Einkauf**

Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37830

Superhydrophilic oxyanions (SO$_4^{2-}$, SeO$_4^{2-}$, ReO$_4^-$, ^{99}TcO$_4^-$) are extremely difficult to separate from aqueous solutions owing to their high free energies of hydration. Nevertheless, their targeted separations are a crucial part in desalination, oil production, and in the processing of legacy nuclear waste. Various high-affinity synthetic receptors have been reported to achieve selective oxyanion separation through molecular recognition; however, relatively fewer can overcome the Hofmeister bias to liquid-liquid extraction (LLE) of these ions, especially from increasingly complex and dilute mixtures. Achieving high affinity and selectivity often leads to difficulties in releasing the captured ion without generating dilute waste streams and large amounts of secondary wastes. Herein, we address this issue by designing an amphiphilic, photoactive iminoguanidine ligand (SGPyDIG) appended with long alkyl chains. This ligand can be readily synthesized as a chloride salt in one step and its high solubility in non-polar solvents (1,2-dichloroethane) make it a desirable candidate for LLE. SGPyDIG can facilitate selective (~80-90 %) removal of oxyanions from aqueous solution over chloride. The ligand binds and extracts non-radioactive divalent anions like SeO$_4^{2-}$ as well as radioactive monovalent anions such as ^{99}TcO$_4^-$ with high efficiency. This remarkable extraction efficiency may be attributed to a combination of two factors: (1) oxyanion recognition through hydrogen bond interaction of the guanidinium core of the ligand and (2) through formation of large aggregates in the non-polar solvent. Additionally, when stimulus in the form of light is used, a triggered release (~89 %) of the captured ion is achieved. This work establishes a sustainable liquid-liquid separation system for challenging superhydrophilic oxyanions, which in turn can help inspire future advances in critical separations.

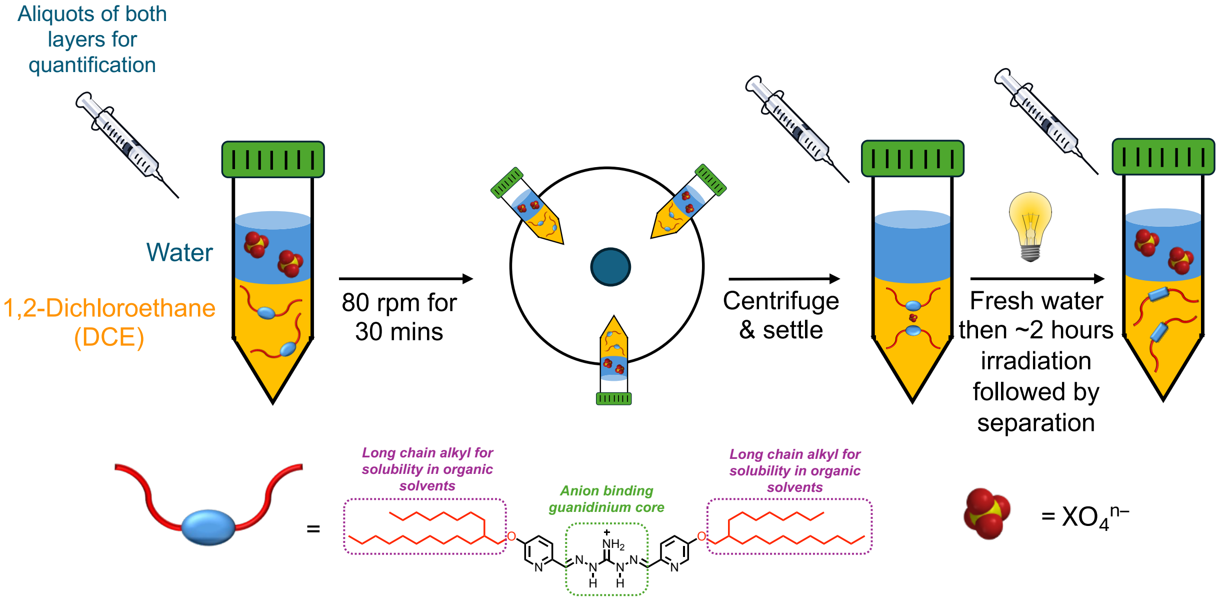

Fig. 1 Workflow of extraction and release of oxyanions using Supergreasy PyDIG ligand.

Characterization of *f*-Element Complexes with Hydroxypyridinone Derived Chelators for Redox Stabilization and Extraction

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With recent developments in today's world, it is as important as ever to develop and continuously innovate processes for recycling components of used nuclear material from a variety of sources including research and energy generation. This can maximize options for storage and/or disposal in geological sites. The separation of fuel elements can be facilitated through solvent extraction using ligands that are designed to bind specific elements or elemental groups. In the body, actinides accumulate in the liver and overtime, can remain in the long bones. This presents opportunities to develop bio-inspired ligands that strongly coordinate *f*-elements. These include hydroxypyridinone (HOPO), which mimics siderophores found in the liver as well as phosphates that are contained within the structure of bones. Inspired by these functional groups, we have developed ligands that include both a 1,2-HOPO functional group and a phosphonic acid. These multidentate ligands are capable of binding *f*-elements through one or both functional groups. Structural characterization of the ligands by NMR, FT-IR, and elemental analysis will be presented. Binding studies, spectroscopic studies, and electrochemical studies with lanthanides and uranium, will be presented to evaluate the binding strength, stoichiometry, and catalytic properties of different HOPO-derived ligand complexes. These properties highlight the impact these ligands may have on existing processes for used fuel reprocessing.

November 4, 2025
Tuesday Evening

Technical Session VI
SNS, Iran Thomas Auditorium

Medical Isotopes/Novel Separations

Chair: Subhamay Pramanik (Oak Ridge National Laboratory)
Co-Chair: Jeff Einkauf (Oak Ridge National Laboratory)

6:00 p.m.	Advanced Separations for Scalable, High Specific-Activity, No Carrier-Added Lutetium-177 Production **MD. A. Momen**	*Shine Technologies*
6:25 p.m.	Small-Scale ^{225}Ac Production from Photonuclear Transmutation of ^{226}Ra Targets **Cory Hawkins**	*NorthStar Medical Radioisotopes*
6:50 p.m.	From Architecture to Selectivity: Computational Insights into Lanthanide-Ligand Complexation **Anton Pozdeev**	*Vanderbilt University*
7:15 p.m.	Ligand Design for Rare Earth Separations: Crafting Selectivity with Precision **Subhamay Pramanik**	*Oak Ridge National Laboratory*

Notes

Technical Session VI
November 4, 2025

Medical Isotopes / Novel Separations

Presentation Abstracts

Notes

Advancing Separations for Scalable, High-Specific Activity, No-Carrier-Added Lutetium-177 Production

Abdul Momen, Marek Piechowicz, Ross Radel

SHINE Technologies, Janesville WI, USA

The expanding field of theranostics has created an urgent, global demand for high-specific activity no-carrier-added (n.c.a.) Lutetium-177 (Lu-177).

SHINE Technologies addresses this need with the Cassiopeia facility, one of the largest n.c.a. Lu-177 production facilities in North America. At Cassieopeia, enriched Yb-176 targets are produced and sent to the Missouri University Research Reactor (MURR) for irradiation. Upon receipt, targets are assayed and processed to separate the Lu-177.

The purification relies on an integrated multi-stage HPLC system. The initial step involves dissolving the irradiated target and preparing the solution by complexing the Yb/Lu with a DOTA-based chelator. The core separation is then achieved through a serial system featuring two stages of HPLC followed by a final ion exchange (IX). The HPLC stages are critical for the Yb/Lu separation, utilizing dual UV and CZT detection for real-time collection of the Lu-177 product and ensuring strict control over separation from bulk impurities like Yb, transitional metal, and other lanthanides.

A significant processing challenge that was observed and managed through the course of process development is radiolysis, where Lu-177 β^- decay leads to dechelation and product loss. SHINE has implemented a novel strategy to overcome this by introducing sacrificial radiolytic scavengers. Specifically, lanthanum nitrate is added in the first HPLC stage, and sodium ascorbate is used in the second HPLC stage. These additives protect the Lu-177-chelator complex ensuring high yields throughout the separation. The process concludes with an ion exchange step, which effectively removes residual metal cations before the final, highly pure Lu-177 product is eluted with high-concentration HCl.

This commercial process demonstrates separation science innovation required to meet growing global market demand for high-specific activity therapeutic radioisotopes, with the Cassiopeia facility designed to achieve a maximum output of approximately 2000 patient doses each week.

From Architecture to Selectivity:
Computational Insights into Lanthanide–Ligand Complexation

Anton S. Pozdeev[1,2]*, Alexander S. Ivanov*[3]*, Santa Jansone-Popova*[1]*, and De-en Jiang*[2]

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[3]Department of Nuclear Engineering, The University of Tennessee, Knoxville, TN, 37996

The efficient separation of lanthanide elements remains a persistent challenge due to their nearly identical ionic radii and chemical properties, motivating intensive theoretical and experimental research. In this work, we explore strategies for improving Ln selectivity through systematic theoretical modifications of ligand structures. Our study covers a broad range of ligands with diverse structural features—varying numbers of donor heteroatoms (3–5), different degrees of preorganization, and distinct N/O donor ratios — to uncover how these parameters influence complexation behavior.

To ensure a comprehensive assessment, we developed a unified theoretical framework capable of describing both classical 1:1 metal–ligand complexes and multi-ligand systems (1:2 for bulkier ligands and 1:3 for smaller ones) with an example in Fig. 1.

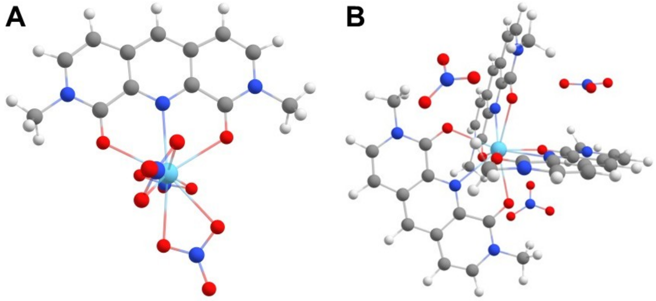

Fig. 1 Examples of (A) 1:1 complexation model and (B) 1:3 multi-ligand complexation model

We perform a global conformer search for 1:2–1:3 complexes at the GFN2-xTB level of theory and construct 1:1 complexes by reproducing reported crystal structures. All structures were optimized and verified by harmonic frequency analysis at the TPSS0/def2-SVP level, using large-core relativistic pseudopotentials for lanthanide atoms.

Our analysis reveals that increasing the number of heteroatoms significantly enhances selectivity, provided that the overall ligand architecture remains structurally consistent. Preorganization acts as a fine-tuning factor, further improving selectivity and thermodynamic stability of complexes. In contrast, the coordination of additional ligand molecules (1:2 or 1:3 complexes) does not introduce substantial changes in selectivity trends, indicating that ligand efficiency can be reliably evaluated based on 1:1 complexation models.

By elucidating the electronic and structural origins of these effects, this study provides an interpretation of ligand design principles and offers valuable guidelines for developing more selective extractants for lanthanide separation.

Small-Scale ^{225}Ac Production from Photonuclear Transmutation of ^{226}Ra Targets

Cory A Hawkins, Miguel T. Gonzalez, and Daniel J. De Vries

NorthStar Medical Radioisotopes, LLC, Beloit, WI 53211

Actinium-225 is becoming more widely available from a variety of production methods.[1-3] As more and diverse suppliers enter the Ac-225 active pharmaceutical ingredient (API) market, the greater the need for demonstrating adequate quantification of radionuclidic impurities. NorthStar has developed a process for production and purification of Ac-225 by photonuclear transmutation of Ra-226 targets and aqueous chemical separations. As part of small-scale production runs, results from in-process sample analysis and purified product quality control testing have provided insights supporting process improvements and analytical method development.

Targets comprise Ra-226 salt deposits in metal capsules irradiated by 40-MeV electron accelerators employing high-Z convertors producing high-intensity bremsstrahlung. A Ra-226(γ,n)Ra-225 reaction is to be optimized for commercial Ac-225 production. Due to (γ, 2n), (γ, 3n), and (n,γ) side-reactions, Ra-224, Ra-223, and Ra-227 are co-produced. The chemical process involves target dissolution and column purification cycles, yielding Ra-225 generator sources that are harvested periodically. Given the variety of radionuclides and chemical elements in their decay chains, the chemical process is designed to efficiently remove the non-radium minor components, purifying the radium solution.

Various other elements (e.g., Ac, Ln, Pb, Bi, Th) are removed in source generation with significant retention on the columns. After the end of irradiation, Ra-227 quickly decays to the 22-year half-life Ac-227, which is partitioned away from the radium-bearing sources during their generation. Regulatory implications for Ac-227 in the USA include; a mandatory spill reporting limit of only 2 nCi, financial assurance requirements for possession over 10 μCi, and an annual limit on intake (ALI) of only 0.4 nCi.[1] Some EU member countries have indicated that a separation factor of at least 10^8 (Ac-227/Ac-225) may be required. Similarly stringent requirements also exist for Ra-223,224,226 and a variety of other important potential impurities, including Pb-210,212 and Po-210. In this presentation we provide an overview of the production and chemical separations processes, concentrating on how the desire to demonstrate the highest standards of radionuclidic purity (RNP) drives development.

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Ligand Design for Rare Earth Separation: Crafting Selectivity with Precision

Subhamay Pramanik,[1] Ilja Popovs,[1] Alexander Ivanov,[1] Jeffrey Einkauf,[1] Marcy Lamb,[1] De-en Jiang,[2]
and Santa Jansone-Popova[1]

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[2]Chemical and Biomolecular Engineering Department, Vanderbilt University, Nashville, TN, US, 37235

With the rapid advancement of modern technologies, lanthanides—commonly known as rare earth elements (REEs)—have become critical to the United States economy, clean energy development, and national security. However, the efficient separation of these chemically similar elements remains a significant challenge due to their nearly identical properties and the limitations of current extraction methods. Additionally, conventional separation processes often pose environmental concerns, underscoring the urgent need for innovative, sustainable, and selective separation strategies [1]. For instance, separation of crucial radioactive Promethium (^{147}Pm, $\tau_{1/2}$ = 2.62 years) from nuclear fuel waste is a grueling challenge. We recently discovered diglycolamide (DGA) ligand for chelating ^{147}Pm in aqueous media. [2] We also designed and synthesized multidentate preorganized ligands for the selective separation of lanthanides. This work emphasizes the significant implications resulting from modifying the donor group configuration within an N, O-based tetradentate ligand and the changes in the solvation environment of Ln ions in the process of separating Lns, with a unique ability to achieve peak selectivity in the light, medium, and heavy Ln region. [3] The structural rigidity of the bis-lactam-1,10-phenanthroline (BLPhen) ligand enforces size-based selectivity, displaying an exceptional affinity for Lns having larger ionic radii, such as La. Modifying the ligand by eliminating one preorganization element (bis-lactam bipyridine) results in the fast formation of BLBPy complexes with light Lns. However, the peak selectivity shifts towards middle Ln (Sm) after reaching equilibrium, resulting in time-resolved separation. However, the change in extraction mechanism is observed at high nitric acid concentrations, leading to the formation and preferential extraction of anionic heavy Ln ensemble, $[Ln(NO_3)_5]^{2-}$, that self-assemble with ligands that have undergone protonation forming intricate supramolecular architectures. We also have designed preorganized multidentate pincer ligands that enable selective separation of specific REEs chlorides through crystallization. Notably, by fine-tuning separation parameters (e.g., solvent, anions, additives), selectivity can be shifted across the REE series, offering an additional layer of control over the separation process. Recently, we have been focusing on the development of environmentally friendly electrochemical separation of trivalent lanthanides using novel redox-active DGA ligands. This presentation will highlight our recent results in unraveling the synthesis, coordination, and selective methods to separate REEs.

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November 5, 2025 Technical Session VII
Wednesday Morning SNS, Iran Thomas Auditorium

Novel Separations

Chair: Abdul Momen (SHINE Technologies)

8:15 a.m. Rare Earth Ion Transport in Small Diameter Metallic and Semiconducting Carbon Nanotube Porins
Aleksandr Noy *Lawrence Livermore National Laboratory*

8:40 a.m. Removal of Superhydrophilic Oxyanions from Ultradilute Aqueous Environments and of CO_2 from Air using Light-Responsive Guanidine Receptors
Radu Custelcean *Oak Ridge National Laboratory*

9:05 a.m. Small Molecule Chelating Extractants for Select Economically Critical Metals
Stephen Long *National Energy Technology Laboratory*

9:30 a.m. Transuranic Trapping Inside Guanidinium-Uranyl-Carbonate Double Salt
Peter Zalupski *Idaho National Laboratory*

9:55 a.m. Break

10:15 a.m. Advancing Minor Actinide Extraction *via* Alkoxy-Functionalized BTPs
Jesse Carrick *Tennessee Technological University*

10:40 a.m. Chemoinformatics-Guided Design of MA Extractants – Integrating Fluorinated Solvents and Machine Learning
Masahiko Nakase *Institute of Science Tokyo*

11:05 a.m. Redox Behavior of Eu(III) Using TODGA-Functionalized Carbon Paste Electrodes
Melanie Guerra *University of Massachusetts – Lowell*

11:30 a.m. Lunch

Notes

Technical Session VII
November 5, 2025

Novel Separations

Presentation Abstracts

Notes

Rare Eart Ion Transport in Small Diameter Metallic and Semiconducting Carbon Nanotube Porins

Jobaer Abdullah, Yuhao Li, Yu Chen, Margaret Berrens, Julai Chung, Tuan Anh Pham, Aleksandr Noy

Materials Science Division, Lawrence Livermore National Laboratory, Livermore, CA 94550

Rare earth elements have been powering the key technologies shaping our societies, yet their production and availability are severely constrained by the lack of clean and efficient separation approaches. In this work we investigated the selectivity and efficiency of REE^{3+} ions transport through the inner pores of species-pure sub-1-nm diameter carbon nanotube porins (CNTPs) [1]. We report that REE^{3+} ions enter these extremely narrow pores and translocate through them with transport rates that are only an order of magnitude lower than that of K^{+} ions. CNTPs channels also exhibited statistically-significant selectivity between light and heavy REE ions. Surprisingly, semiconducting CNTP channels translocated REE ions consistently faster than the metallic CNTPs of the same diameter and also exhibited statistically significant discrimination between heavy REE ions. Density functional theory (DFT) simulations examined the microscopic mechanism of REE^{3+} ion entry into CNTPs and reveal a series of events at the CNT channel entrance that enable the ions to translocate through the CNT channel.

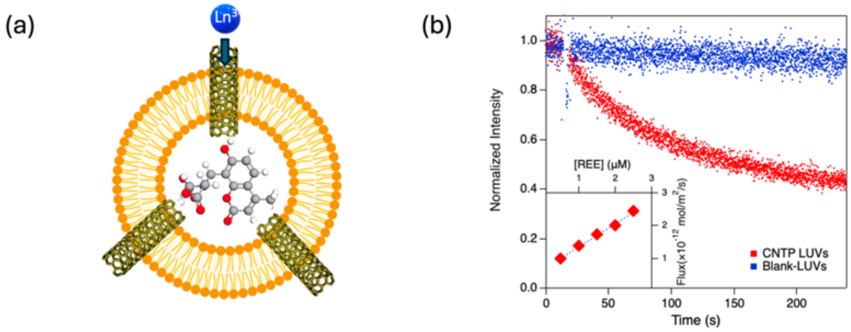

Fig. 1. a. Schematics of the REE ion transport measurement showing a vesicle with carbon nanotube porins inserted into the lipid shell. The vesicle contains REE-sensitive CB dye. **b**. Representative kinetic traces of CB dye fluorescence recorded after vesicles with and without CNTPs were exposed to Eu^{3+} concentration gradient of 2.5 mM. Inset: Plot of the Eu^{3+} ion flux as a function of REE concentration gradient.

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Removal of Superhydrophilic Oxyanions from Ultradilute Aqueous Environments and of CO_2 from Air using Light-Responsive Guanidine Receptors

Radu Custelcean, Jeffrey Einkauf, and Nabarupa Bhattacharjee

Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831

Selective separation of superhydrophilic oxyanions (e.g., SO_4^{2-}, SeO_4^{2-}, CrO_4^{2-}) from aqueous solutions is extremely difficult due to the exceedingly high dehydration penalties associated with their removal from water. Furthermore, many of these anions are often found in very dilute concentrations or are subject to stringent regulatory aqueous concentration limits. For instance, the EPA requirements for chromate or selenate concentrations in drinking water or aquatic environments is in the ppb range. On the other hand, carbonate, another environmentally relevant anion tied to the natural carbon cycle, is under equilibrium with atmospheric CO_2 and thus typically present in the ppm concentration range in natural waters.

Responding to the challenge of separating these oxyanions from ultra-dilute and competitive aqueous solutions requires designing receptors that possess not only exceptional selectivity, but also extraordinary affinity so that the target anionic species may be concentrated from very large volumes of feed streams. Conversely, receptors with very strong binding affinities, lead, in turn, to challenges in achieving effective release (stripping) of the bound species. Traditionally, ion stripping has involved changes in chemical gradients such as concentration, ionic strength, or pH swings, which typically generate dilute streams of the separated ions and large amounts of secondary wastes.

Recent research in our group has focused on a new class of iminoguanidine anion receptors readily synthesized in one step and high yields from commercially available aldehydes or ketones, and amino- or diamino-guanidinium precursors. These receptors selectively recognize oxyanions like sulfate, selenate, chromate, or carbonate through bidentate hydrogen bonds, and remove them efficiently from water via crystallization, liquid-liquid extraction, or ion exchange. The anion is then easily released by a pH swing, which deprotonates the guanidinium groups, thereby neutralizing their anion-binding abilities. Furthermore, photoisomerization of the iminoguanidinium groups from E to Z configurations can release the anions using only energy inputs, leading to greener and more sustainable anion-separation cycles.

This research was supported by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences, Chemical Sciences, Geosciences, and Biosciences Division.

Small Molecule Chelating Extractants for Select Economically Critical Metals

Stephen M. Long[1], Scott E. Crawford[2], Wissam A. Saidi[2], Sean M. Nations[2], Lauren Burrows[2],
Tyler Bridges[2,3], Michael Schulz[3]

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The production of economically critical metals has increased significantly since the start of the 21st century due to a combination of ever-increasing demand and global supply chain vulnerabilities. The selective recovery of metals from both primary and secondary resources presents a significant challenge due to the high chemical similarity of metals that are adjacent on the periodic table and high concentrations of non-target metals relative to target metals. The use of small molecule chelating extractants in solvent extraction has emerged as a promising technology for separating economically critical metals due to its effectiveness at low concentrations of metal ions and capability to be custom designed for rigidity, size, or functional group to influence affinity for specific metals and enhance selectivity. However, experimental trial and error for chelator design remains a time consuming and costly challenge. Here, we present a tandem modelling-experiment approach to accelerate chelator development and evaluation. The combination of computational insight into chelator-metal binding affinities, chemical synthesis, validation of the model using isothermal titration calorimetry, and, finally, evaluation in metal extraction processes mitigates experimental trial and error and will be discussed in detail in this presentation using several exemplary chelators.

Transuranic Trapping inside Guanidinium-Uranyl-Carbonate Double Salt

Peter R. Zalupski, Travis S. Grimes, and Dean R. Peterman

Radiochemical Separations and Radiation Science Department,
Idaho National Laboratory, Idaho Falls, ID 83404

The primary goal of alkaline-side dissolution flowsheet is the recovery of uranium using crystallization. The intent is to dissolve a voloxidized used nuclear fuel in hot carbonate-rich media and induce crystal formation upon cooling. This chemistry relies on the limited solubility of the double salts of uranyl tris-carbonato complexes when balanced by monovalent cations such as guanidinium. Studies have shown that relative to actinyl double salts of alkali metals, the guanidinium analogs are more soluble, presumably due to the hydrogen donating ability of those ions.[1] In carbonate solutions, hydrogen bonds can stabilize the $[UO_2(CO_3)_3]^{4-}$ ions, increasing their aqueous solubility. The geometry, solubility and solution thermodynamics of such double salt compounds are not exclusive to uranyl, however, as other actinides capable of forming the dioxoactinyl ions behave similarly.[2] Accordingly, even partial dissolution of transuranic component of used nuclear fuel in alkaline media may challenge the bulk uranium recovery when neptunium and plutonium co-crystallize in the guanidinium uranyl tris-carbonate crystal lattices. This co-crystallization behavior has been demonstrated for U-Np and U-Pu, but not for U-Am in guanidine carbonate. The isolated crystals from the U-Np dissolver liquor contained a 96:4 weight distribution. The U-Pu crystalline material showed a 89:11 U:Pu weight composition. Americium was found to crystallize discretely from U, presumably due to its pentavalent oxidation state, supporting different coordination geometry.
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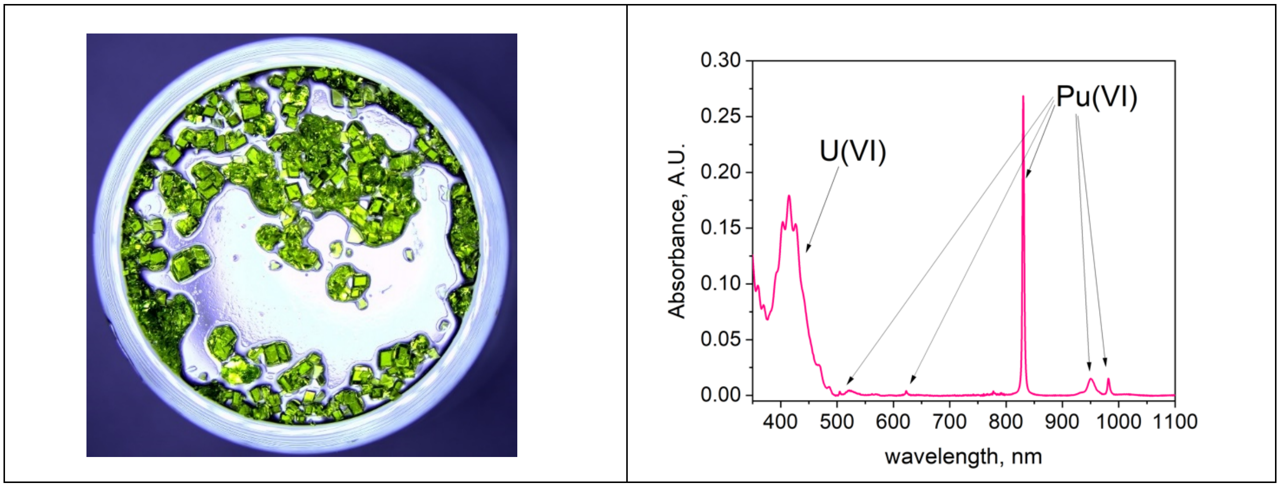

Fig. 1 (left) Picture of crystalline solid isolated from the U-Pu mixture in guanidine carbonate. (right) Absorption spectrum of the dissolved solid after UO_2^{2+} - PuO_2^{2+} co-crystallization. Dissolution media: 2 mol·L^{-1} HNO_3.

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Advancing Minor Actinide Extraction via Alkoxy-Functionalized BTPs

Krishna P. Gnyawali,[1] *Lakshi M. Thilakarathne,*[1] *Dale D. Ensor,*[1] *David A. Dixon,*[2] *Stephen P. Mezyk*[3]*,*
and Jesse D. Carrick[1]

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Sustainability of the nuclear fuel cycle can be improved with viable approaches to selectively extracting the minor actinides from the lanthanide fission products contained in spent nuclear fuel. Designing separation systems to preferentially associate with actinides over lanthanides is not trivial. Similar physical properties including oxidation state, cationic radius, and hardness render this extraction a grand challenge in separation science. Although, a perfect separations system has yet to be disclosed, extensive progress in this area over the last two decades has been made since Kolarik’s disclosure in 1999 that bis-1,2,4-triazinyl-2,6-pyridine (BTP) complexants show preference for the minor actinides.

Work in this laboratory has focused on designing, developing, and evaluating soft-Lewis basic, BTP donor complexants which are soluble in hydrocarbon diluents, have robust stability to strongly acidic environments, and accentuate metal-complexant covalency of the actinides. Recent results have demonstrated the potential that 3,3’-alkoxy-functionalized-BTPs in the context of symmetric,[1] and unsymmetric complexants[2] for ^{241}Am and ^{244}Cm extraction.[3] The addition of extended carbon chains at the 3,3’-positions of the benzil confer unique, and improved extraction performance properties, beyond 3,3’-methoxy-BTP disclosed in 2018, including solubility in isooctanol, octanol, and even octanol:kerosene[4] in certain cases, hydrolytic stability in 6M $HNO_{3(aq)}$, as well as utility for multiple extractions with the same complexant with similar results.

Complexant design, extraction, decomplexation, recycling, and preliminary radiolytic degradation data of current BTPs of interest will be presented.

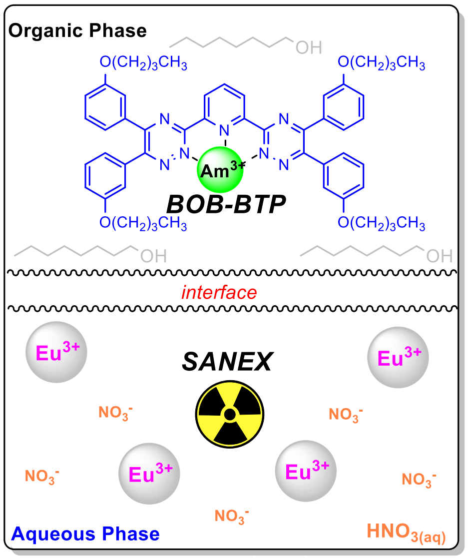

Fig. 1. Complexant-based SANEX

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Chemoinformatics-Guided Design of MA Extractants
- Integrating Fluorinated Solvents and Machine Learning

Masahiko Nakase[1,2], *Takahito Nishihara*[1,2], *and Tomohiro Okamura*[1,2]

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To advance minor actinide (MA) separation and process development, we have pursued not only the design of extraction agents but also the incorporation of complexing agents and the exploration of solvents. In particular, aiming to reduce waste liquid and enhance process safety, we have applied machine learning techniques to the development of extraction systems based on fluorinated solvents. Among the many potential solvent candidates, we have pursued the development of fluorinated solvents, considering them most suitable for MA extraction due to their various advantages and compatibility with the subsequent processes we envision. As part of this effort, we developed the AACE program, which enables regression model construction, experimental design, and extrapolative transfer learning [1]. Given the high cost of acquiring training data in the Actinide chemistry field, we leveraged existing databases, notably the International Database on Extractant Ligands (IDEaL), compiled in reports such as the OECD/NEA Nuclear Science Committee (NSC) Expert Group on Fuel Recycling and Waste Technology (EGFRW)'s State-of-the-Art Report on Nuclear Fuel Cycle Chemistry. IDEaL contains unstructured data for 439 MA extractants, including molecular formulas, molecular weights, partition coefficients, separation factors, and associated processes. This data was aggregated into MongoDB and preprocessed for machine learning, requiring extensive data wrangling due to its mixed numerical and textual formats. Some of the chemoinformatics attempts were made. We constructed a chemoinformatics scheme to explore extractant side-chain structures, focusing on diglycolamide (DGA) derivatives suitable for fluorinated solvents. However, the limited availability of DGA-specific data in existing databases poses challenges for regression analysis and model generalization. To address this, we implemented a clustering-based approach that identifies structurally similar extractants and utilizes their associated extraction data for training [2]. Preliminary results suggest that side chains with approximately a straight-chain side chain with a carbon number of 4 are meant to be favorable for fluorinated solvent compatibility, although solubility effects related to chain length have yet to be fully integrated. Extractants registered in the IDEaL were classified into acidic and neutral extractants, and those with characteristic structures, and regression models were developed accordingly. The result showed a higher accuracy in the distribution ratio of Am for classification- based modeling compared to using all data. To enable the unexpected discovery of high-performance or unique extractants, it remains desirable to construct models capable of predicting across structurally diverse candidates within a unified framework.

Acknowledgement
This study is supported by the MEXT Innovative Nuclear Research and Development Program Grant Number JPMXD0221459189, and the GIMRT Program of the Institute for Materials Research, Tohoku University (Proposal No. 202412-IRKAC-0010, 202311-IRKAC-0001, 202211-IRKAC-0037, 202412-RDKGE-0029, 202311-RDKGE-0002,202211-RDKGE-0010).

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Redox Behavior of Eu(III) Using TODGA-Functionalized Carbon Paste Electrodes

Melanie Guerra[1] *and Erin R. Bertelsen*[2]

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Functionalized carbon materials are promising platforms for radiochemical separations and electrochemical interrogation of lanthanides. In this work, tetra-*n*-octyl diglycolamide (TODGA) was physisorbed on conductive carbon black (Vulcan XC-72) and incorporated into carbon paste electrodes (CPEs) for the one-electron reduction of Eu(III) to Eu(II). Nitrogen sorption analysis indicated that TODGA functionalization reduced the accessible surface area and pore size distribution, as determined by Brunauer–Emmett–Teller (BET) theory and the Barrett–Joyner–Halenda (BJH) method, respectively. Electrochemical studies using cyclic voltammetry (CV) and differential pulse voltammetry (DPV) revealed two distinct Eu(III)/Eu(II) redox processes, corresponding to unbound and TODGA-coordinated Eu species. The TODGA-bound species exhibited a shift in the redox couple to more negative potentials, consistent with stabilization of the trivalent oxidation state. These results demonstrate that TODGA-modified carbon electrodes alter the redox behavior of Eu species, providing a reproducible platform for electrochemical studies of lanthanide systems.

November 5, 2025
Wednesday Afternoon

Technical Session VIII
SNS, Iran Thomas Auditorium

Nuclear Fuel Cycle Separations

Chair: Gabriel Hall (Pacific Northwest National Laboratory)
Co-Chair: Peter Tkac (Argonne National Laboratory)

1:30 p.m. Effect of Radiolytic HNO_2 and DBP on the Extraction of Np in CoDCon
Gabriel Flores *Argonne National Laboratory*

1:55 p.m. Separation of TRISO Particles from bulk graphite
Gabriel Hall *Pacific Northwest National Laboratory*

2:20 p.m. Experimental Studies and Molecular Modeling of the Physicochemical Properties of Pure Monoamide Extractants
Clovis Poulin-Ponnelle *University of Montpellier, France*

2:45 p.m. Dissolution Characteristics of Mixed Oxide Fuel in Alniflex
Jarrod Gogolski *Savannah River National Laboratory*

3:10 p.m. Break

3:30 p.m. Np(V)/Np(VI) Equilibria and Kinetics in Advanced Nuclear Fuel Recycling
David Bettinardi *Argonne National Laboratory*

3:55 p.m. Correlating Radiolytic, Non-Radioactive Radical, and Computational Methods for Monoamide Degradation
Julia Brumaghim *Clemson University*

4:20 p.m. PUMAS – From Recycling to Multicycling: A Monoamide-based Solvent Extraction Process for C-Recovery of Uranium and Plutonium from Spent UOX and MOX Fuel
Loic Daronnat *Orano Recycling*

6:00 p.m. Banquet Reception at the Double Tree Hotel
Conference Banquet, Student Poster Awards, and After-Dinner Presentation

Notes

Technical Session VIII
November 5, 2025

Nuclear Fuel Cycle Separations

Presentation Abstracts

Notes

Effect of radiolytic HNO_2 and DBP on the Extraction of Np in CoDCon

Gabriel A. Flores, David J. Bettinardi, Laura E. Maggos, Candido Pereira, and Peter Tkac

Chemical and Fuel Cycles Technologies Division, Argonne National Laboratory, Lemont, IL 60439

CoDCon is a tributylphosphate (TBP) based process that co-extracts uranium (U) and plutonium (Pu). In acidic media, Np is predominately Np(V) but disproportionates into Np(IV) and Np(VI). Because Np(V) is not readily extractible by TBP but Np(IV) and Np(VI) are, Np is partitioned between the aqueous and organic streams in nuclear reprocessing. Nitrous acid (HNO_2) and dibutylphosphate (DBP) are radiolytic products of nitric acid (HNO_3) and TBP respectively which have an effect on the extraction behavior of Np. In this work, the effect of nitrous acid (HNO_2) and DBP on the extraction of Np in CoDCon was investigated for the purpose of finding conditions where 99% of Np can be co-extracted with U. HNO_2 was found to increase the amount of total Np extracted in a batch extraction by increasing the oxidation kinetics of Np(V) to Np(VI). DBP is a stronger extractant than TBP as it can form more stable complexes with metal cations. The presence of DBP in the organic phase increased the distribution ratio of total Np but had the expected consequence of co-extracting transition metals such as zirconium (Zr) and palladium (Pd). Although the increased presence of DBP increased Np extraction, it has a negative impact on the purity of the U, Np, Pu product and should be minimized.

Separation of TRISO particles from bulk graphite

Gabriel B. Hall,[1] *Jonathan B. Wierschke,*[2] *Jenna Metera,*[1] *Parker Okabe,*[3] *Lucas Boron-Brenner,*[1] *Lauren Richmond,*[1]
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TRi-structural ISOtropic (TRISO) fuel is proposed for use in multiple advanced nuclear reactor designs including the Westinghouse eVinci, X-energy XE-100, Kairos KP-FHR, and Project Pele among others. TRISO fuel is particularly well encapsulated. It is comprised of ~500 µm fissile kernels which are encapsulated by porous graphite, SiC, and pyrolytic graphite for form a ~ 1 mm TRISO particle. Tens of thousand TRISO particles are then impregnated into a bulk graphite matrix to form either spherical pebbles or cylindrical compacts. The bulk graphite can comprise up to 95% of the TRISO fuel.

The extensive encapsulation of the fuel offers many potential advantages during operation including neutron moderation, high thermal stability, chemical inertness, and potential for on-line refueling. However, this also makes the fuel meat extremely difficult to access for scrap recovery in the event of manufacturing defects, or for recycling of valuable actinides after irradiation. Both scrap recovery and recycling may be more economically advantageous for TRISO than conventional light water reactor fuel due to the use of High Assay Low Enriched Uranium (HALEU) in TRISO fuel. There may also be economic advantages to separating the

TRISO particles from the bulk graphite if a strategy of direct disposal in a deep geological repository is implemented.

The inherent presence of 14C prevents removing the carbon through bulk oxidation to CO_2 as any generated 14CO_2 would need to be recaptured adding to the volume of waste. Thus, a method is needed to deconsolidate the TRISO compacts/pebbles as well as to physically separate the bulk graphite from the TRISO particles. Further this needs to be accomplished without fracturing of the TRISO particles to the point they release fission products. To this end Pacific Northwest National Laboratory has been examining electrochemical methods for the deconsolidation of TRISO compacts followed by separation of the TRISO particles from the bulk carbon via elutriation.

Experimental Studies and Molecular Modeling of the Physico-chemical Properties of Pure Monoamides Extractants

Clovis Poulin-Ponnelle, Abderrazak Masmoudi, Dominique Guillaumont, and Philippe Guilbaud

CEA, DES, ISEC, DMRC, Univ Montpellier, Marcoule, 30207 Bagnols sur Cèze, France

For several years now, research groups around the world have been looking for alternative processes to the PUREX process to treat spent nuclear fuel.[1] PUREX uses tributyl phosphate (TBP) as selective extractant to recover the reusable elements. Molecules from the monoamide family appear to be the most promising candidates as extracting agents to develop a new process.[2] They allow complete incineration thanks to their chemical composition, which consists solely of C, H, O and N atoms. They also enable U and Pu to be separated during the partitioning step, without the redox step of reducing Pu(IV) to Pu(III) required in the case of TBP.

Although monoamides have been widely studied as extractants, their intrinsic physico-chemical properties such as viscosity, solubility, density and surface tension are not well understood. A better understanding of these intrinsic properties would lead to a better understanding of the extraction solvent, which is often a monoamide diluted in a diluent such as hydrogenated tetrapropylene (TPH), and therefore better control of the system on an industrial scale. For this reason, in our work, we focus on investigating the physico-chemical properties of monoamides extractants as pure *i.e.* without diluent. This is done by coupling molecular modeling using molecular dynamics simulations[3] and the COSMO-RS approach[4] with experimental measurements to determine the viscosity, solubility and density of a large number of monoamide molecules. Our results have shown that molecular modelling reproduces experimental data qualitatively, and that the intrinsic properties of monoamides molecules can be modulated by modifying the alkyl chain lengths and branching. We have also found that external parameters such as extraction of nitric acid, U-loading and temperature have a major influence on these properties. In parallel, the speciation of monoamide-uranium and plutonium complexes formed in pure extractants was studied. It highlighted that in a pure extractant, the speciation is slightly different from that observed in the presence of a diluent.

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Dissolution Characteristics of Mixed Oxide Fuel in Alniflex

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Separation Sciences and Engineering, Savannah River National Laboratory, Aiken, SC 29808

Uranium-zirconium (U-Zr) fuel has been used for various past and prospective advanced reactors. Dissolution of U-Zr fuel, to recover the uranium, can be done safely with excess HF to preferentially dissolve the intermetallic U-Zr_2 that can form during U-Zr fuel fabrication. While excess HF will readily corrode ubiquitous stainless steel dissolvers, the Alniflex dissolvent (2 M HF, 1 M HNO_3, 1 M $Al(NO_3)_3$, and 0.1 M $K_2Cr_2O_7$) proposed in the 1960s offers a potential dissolution pathway of U-Zr fuel in existing stainless steel dissolver vessels [1]. Previous work lowered the $Al(NO_3)_3$ concentration from 1 to 0.5 M to improve the solubility range of AlF_3 so that multiple charges, or bundles of U-Zr fuel, can be dissolved in a single solution [2]. An additional consideration is the solubility of transuranics as the burnup of U-Zr fuel can vary and no previous data has been recorded on this subject. Therefore, mixed oxide pellets (MOX) containing approximately 71.7% U, 15.3% Pu, 1.1% Am, and 0.05% Np were used to ascertain bounding solubility characteristics of the actinides within Alniflex and Alniflex-based solutions over a range of temperatures (Table 1).

Table 1: Initial solution conditions for MOX dissolution runs

Exp. #	**Main Temp. Range (℃)**	**HF**	**HNO_3**	**$Al(NO_3)_3$**	**$Cr(NO_3)_3$**	**$K_2Cr_2O_7$**
				(M)		
1	22 - 90	2.0	1.0	0.5	NA	0.100
2	50 - 90	2.0	1.0	0.5	NA	NA
3	70 - 90	2.0	2.0	0.5	NA	0.100
4	22 - 90	2.0	1.0	0.5	0.201	NA

NA-Not Applicable

Metal coupons of Al-6061, Zircaloy-2, and stainless steel were added to help simulate initial dissolution or corrosion of the bundle, cladding, and dissolver vessel, respectively. As noted in Table 1, the Alniflex dissolvent was adjusted from run to run to help isolate other dissolution characteristics of the MOX pellet, such as using Cr(III) instead of Cr(VI). While none of the changes to the dissolvent seemed to affect transuranic solubility, the MOX dissolution trends appreciably changed. Additional experimental results will be discussed.

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Np(V)/Np(VI) Equilibria and Kinetics in Advanced Nuclear Fuel Recycling

David J Bettinardi, Gabriel Flores, Laura Maggos, Candido Pereira, Peter Tkac

Chemical and Fuel Cycle Technologies, Argonne National Laboratory, Lemont, IL 60439

One of the challenges in implementing the co-decontamination (CoDCon) chemical flowsheet for obtaining a mixed U/Pu/Np nitrate product from dissolved used nuclear fuel is reliably directing all (99%+) Np to the product stream. Np chemistry is challenging because it is kinetically and thermodynamically sensitive to its redox environment. In dissolved fuel, the redox environment can vary widely depending on factors like acid concentration, fuel composition, burnup, and cooldown time. Np(V) is extracted very poorly by tri-butyl phosphate, so understanding the processing conditions that promote the conversion of Np(V) is key.

We evaluated methods to promote Np(V)→Np(VI) oxidation in situ, including using higher HNO_3 and the presence of other redox active species. We also consider their downstream effects to devise a modified CoDCon flowsheet that achieves 99%+ removal of Np. This talk will cover our experimental results where UV-Vis-NIR spectroscopy was used to measure and quantify Np(IV,V,VI) equilibria in 1-6 M HNO_3, 0.1-50 mM HNO_2. Kinetic data were collected and fit to chemical rate equations. We then developed a model to numerically solve the time-dependent concentrations on Np during a multi-stage separation (i.e., centrifugal contactor bank). The model was validated against experimental data. Finally, our rate equations were implemented into dynamic- AMUSE (dyAMUSE) software for complete flowsheet development.

Our results show that the rate of interconversion between Np(V)-Np(VI) increases with HNO_3 and HNO_2. However, the relative proportion of Np(V) at equilibrium also increases with HNO_2. This means that there is an optimal concentration of HNO_2 that can be determined for a given flowsheet, with consideration to the specific operating parameters used (flowrates, holdup volumes, etc.).

Likewise, given a specific fuel type, it is possible to determine the optimal operating parameters for Np removal. These methods will be used to calculate and verify a CoDCon flowsheet using a fuel simulant on a bank of 3D-printed, chemically-resistant annular centrifugal contactors.

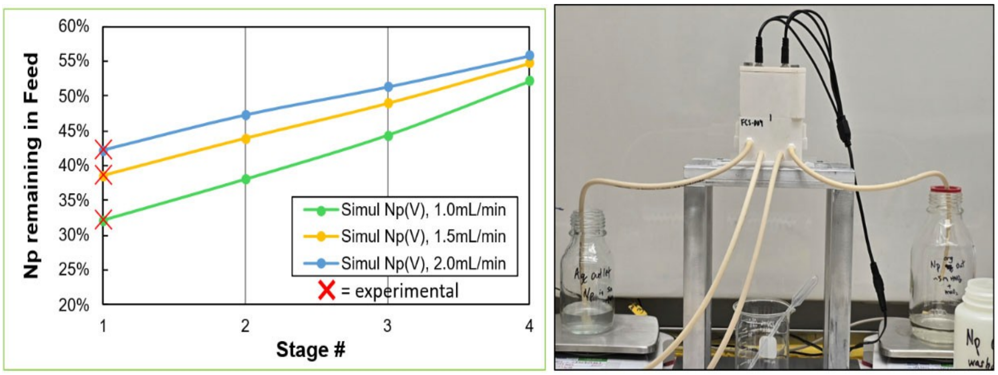

Fig. 1 *Experimental vs. simulated Np(V) remaining in the CoDCon Feed as a function of stage number in a 3D-printed four-stage mini centrifugal contactor. Right: Photograph of the 3D-printed four-stage contactor used in the experiment.*

Correlating Radiolytic, Non-Radioactive Radical, and Computational Methods for Monoamide Degradation

Julia L. Brumaghim, Brandon G. Wackerle, Madison Vicente, Fatema Tuz Zohara, and Modi Wetzler

Department of Chemistry, Clemson University, Clemson, SC 29634-0973, USA

The PUREX (Pu and U Reduction Extraction) process uses TBP as the extractant to recover Pu and U from nuclear waste, but the presence of phosphorus makes waste incineration difficult, and TBP degradation products can lead to third-phase formation. Dialkyl amides such as *N*,*N*-di-2-ethylhexyl-isobutyramide (DEH*i*BA) and *N*,*N*-di-2-ethylhexyl-butyramide (DEHBA) have been studied as alternatives to tributyl phosphate (TBP). Monoamides are more readily incinerable than TBP and their carboxylic acid and amine degradation products prevent third-phase formation because of their water-solubility. The gold standard for examining radiolytic stability is gamma radiolysis, but it is low-throughput and high-cost. We developed a non-radioactive radical assay to screen complexant stability and have examined correlations between monoamide degradation products formed in organic-only conditions using this assay and with gamma radiolysis.[1] In this radical assay, azohydroperoxide forms hydroxyl radical and *tert*-butyl radical upon heating in toluene as the organic solvent. Using gas chromatography mass spectrometry (GC-MS), we determined that the same degradation products, including amine, secondary amide, amide-H_2, and solvent-adduct products, are formed in the radical assay and gamma radiolysis of the monoamides in toluene. Bibenzyl, formed from two tolyl radicals, is also observed under both radical assay and gamma radiolysis conditions. The dose-responsiveness of bibenzyl formation allows us to establish a correspondence between the two methods and semi-quantitatively predict the radiolytic stability of these monoamide complexants. The smaller monoamides such as DBBA, D*i*BBA, DB*i*BA, and D*i*B*i*BA, form fewer total degradation products than DEHBA and DEH*i*BA under both radiolytic and radical assay conditions. Computational studies of monoamide radical stability indicate that the lowest-energy radicals form in proximity to the amide functional group, but the energy differences between radical formation sites are relatively small. The large number of observed degradation products corroborate these computational results and suggest that identifying the degradation products, not just following degradation of the parent complexant, may be critical for understanding monoamide separations on industrial scales.

PUMAS – From Recycling to Multirecycling: A Monoamide-Based Solvent Extraction Process for Co-Recovery of Uranium and Plutonium from Spent UOX and MOX Fuels

Loïc Daronnat[1]*, Solenne Michaud*[1]*, Pierre Sarrat*[1]*, Sylvain Costenoble*[1]*, Cécile Marie*[1]*, Xavier Heres*[1]*, Fabien Lengrand*[1]*, Manuel Miguirditchian*[1]*, Steve Jan*[1]*, Ana Gil Martin*[2]*, Laure Diaz*[3] *and Christian Sorel*[1]

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To address the challenges of plutonium multirecycling in future nuclear fuel cycles, the PUMAS (Plutonium Uranium MonoAmide Separation) process is currently under development at the ISEC institute at CEA Marcoule. This novel liquid-liquid solvent extraction system offers a redox-free and CHON-compliant alternative to the conventional PUREX process, specifically designed for the co- recovery of Uranium and Plutonium from spent UOX and high Pu-content MOX fuels.

PUMAS relies on monoamide extractants, structurally optimized to ensure high distribution ratios, excellent U(VI)/Pu(IV) selectivity, and robust behavior in nitric acid media. Modifications of alkyl chain length and branching have been investigated to enhance phase stability, prevent third-phase formation, and maintain manageable viscosity. Separation of uranium and plutonium is achieved by controlling the acidity of the aqueous phase, avoiding the need for redox chemistry and simplifying the process.

A detailed physicochemical model has been developed and implemented in the PAREX+ simulation platform to guide the process. The PUMAS process was validated through extensive experimental campaigns involving spent UOX and MOX fuel solutions. Mixer-settler tests, performed in hot cells in ATALANTE facility at CEA Marcoule, confirmed uranium and plutonium recovery yields above 99.6%, excellent decontamination factors and a high product purity.

From a technological point, the PUMAS process has reached a level of maturity above TRL 3, with ingoing work aiming to scale up the process toward industrial production of the monoamide followed by industrial demonstration.

Strategically, PUMAS supports the French national policy toward closing the nuclear fuel cycle. It aligns with long-term goals to reduce natural uranium dependency, stabilize plutonium inventories, and minimize final waste volumes. As such, PUMAS represents a key building block in the evolution from monorecycling to full multirecycling

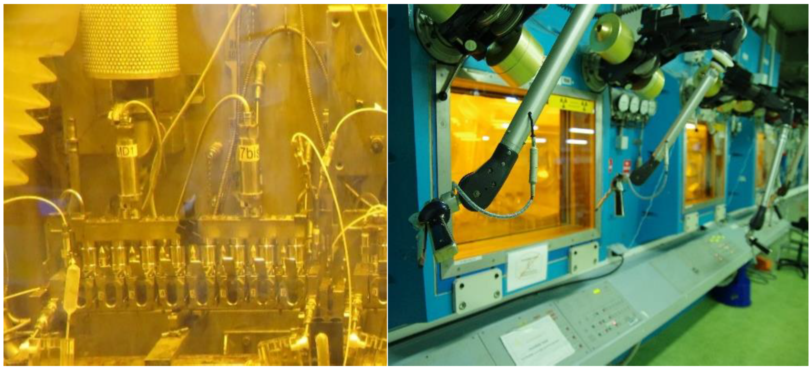

Fig. 1 *Hot cells in ATALANTE facility at CEA Marcoule*

Banquet Presentation

Ernest Lord Rutherford: How a Potato Digger from New Zealand Changed the World

Dale D. Ensor
Professor Emeritus
Tennessee Technological University

Ernest Lord Rutherford (1871–1937) is often remembered as the brilliant physicist who discovered the atom's nuclear structure, but behind the discoveries was a human figure who was curious, kind-hearted, and exhibited a disciplined common sense. This talk invites both scientists and non-scientists alike into the world of the man who discovered alpha and beta radiation, formulated the theory of radioactive decay, and tutored seven future Nobel prize winners. Just as important as his scientific triumphs were his unique qualities as a mentor, colleague, and friend. Through stories about his initial struggles at Cambridge, his time at McGill University, his research group at the University of Manchester, and his return to the laboratory at Cambridge, a more complete picture of the father of nuclear physics as both a Nobel Prize winning scientist and a warm, unforgettable human being will emerge. Our world has been forever changed by Lord Rutherford and his insights into the structure of the atom.

Dr. Dale D. Ensor is an Emeritus Professor of Chemistry at Tennessee Technological University. He received his PhD. from Florida State University in Inorganic and Radiochemistry under Dr. Greg R. Choppin in 1977. He has been a visiting scientist at Oak Ridge National Laboratory, Argonne National Laboratory, and Los Alamos National Laboratory and is author and co-author of over 50 publications on lanthanide and actinide chemistry. Since joining the faculty at Tennessee Technological University in 1978, has directed the independent research of over 75 undergraduate students and 31 graduate students in chemistry. Dr. Ensor retired from active research in 2016 but maintains passion for the history of nuclear and radiochemistry.

November 6, 2025
Thursday Morning

Technical Session IX
SNS, Iran Thomas Auditorium

DOE Applications

Chair: Thomas Shehee (Savannah River National Laboratory)
Co-Chair: Jarrod Gogolski (Savannah River National Laboratory)

8:15 a.m. Rethinking Rare Earth Element Separations: Harnessing Selectivity at the Leaching Stage
Lesta Fletcher *Oak Ridge National Laboratory*

8:40 a.m. CLEANEX Solvent Extraction Process for Californium-252 Production: Replacement of HDEHP by TODGA
Lætitia Delmau *Oak Ridge National Laboratory*

9:05 a.m. Development of an Am/Bi Separation After Oxidation of Am(III) using $NaBiO_3$
Jennifer Pyles *University of Alabama at Birmingham*

9:30 a.m. Progress for HALEU Production at the Savannah River Site
Thomas Shehee *Savannah River National Laboratory*

9:55 a.m. Break

10:15 a.m. High-Pressure Ion Chromatographic Chemical Separations for Inductively-Coupled Plasma Mass Spectrometry Assay at Oak Ridge National Laboratory
Tamara Keever *Oak Ridge National Laboratory*

10:40 a.m. Orphaned Fuels: Adopting a Domestic Nuclear Fuel Cycle
Richard Mayes *Oak Ridge National Laboratory*

11:05 a.m. Next-Generation Caustic-Side Solvent Extraction Process for Cesium Separation from Hanford Tank Wastes Containing High Potassium and Complexant Concentrations
Bruce Moyer *Oak Ridge National Laboratory*

11:30 a.m. Concluding Remarks

Notes

Technical Session IX
November 6, 2025

DOE Applications

Presentation Abstracts

Notes

Rethinking Rare Earth Element Separations: Harnessing Selectivity at the Leaching Stage

Lesta S. Fletcher[1], *Aubrey D. Lemer*[1], *Zhenqi Zhao*[2], *Md Faizul Islam*[1], *Dustin D. Youmans*[3], *Aaron Donovan*[3], *Long Qi*[2], *Nikki A. Thiele*[1]

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Domestic separation of rare earth elements (REEs) is one of the most pressing scientific challenges in the United States. The first industrial step in REE separations is non-selective leaching of a feedstock using a strong acid, while the majority of research efforts focus on post- leaching separation techniques such as solvent extraction or selective crystallization. Selective leaching using strategically designed leaching agents, or chelators, remains an underexplored yet potentially transformative approach to REE separations. In this presentation, we share our selective leaching results from a complex rare earth hydroxide sample derived from authentic domestic mining waste, demonstrating how leaching selectivity can be effectively tuned through chelator design. Furthermore, our research highlights how core principles of coordination chemistry can be harnessed to enhance industrial processes such as hydrometallurgical leaching, offering a pathway to more efficient and economical recovery of REEs.

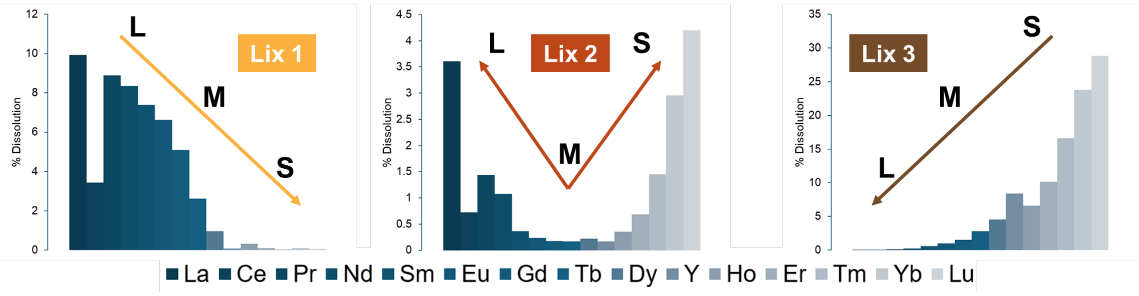

Fig. 1 Results from selective leaching studies of a rare earth hydroxide sample derived from authentic domestic mine waste illustrating distinct size selective patterns of large (L), medium (M), and small (S) REEs using three different lixiviants (Lix 1-3). Conditions: 20 mM lixiviant, 25 mg loading of feedstock (~75 mM total RE), 25 °C, pH ~8.

CLEANEX Solvent Extraction Process for Californium-252 Production: Replacement of HDEHP by TODGA

Lætitia H. Delmau, Emory D. Collins, Julie G. Ezold, Samantha K. Schrell, and Dennis E. Benker

Radioisotope Science and Technology Division
Oak Ridge National Laboratory, Oak Ridge, TN 37830

Originally based on the cation exchanger di(2-ethyl)hexyl phosphoric acid (HDEHP), the solvent extraction process used to produce californium-252 (CLEANEX process) has been modified to allow the use of the neutral ligand tetraoctyl diglycolamide (TODGA). Using a neutral ligand selective for trivalent actinides and lanthanides means that the extraction can be performed at high acidity, which naturally follows the dissolution of the irradiated curium targets. When using HDEHP, the feed must be adjusted down to a concentration of free acid less than 0.05 M when typically starting from a concentration greater than 6 M. The acid boil-off and acid adjustment are tedious and require multiple analyses. Moreover, the solvent then requires high acidity stripping, and the large amount of hydrochloric acid then needs to be evaporated for the subsequent processing step (separation of lanthanides from americium/curium and transcurium elements) based on high lithium chloride/low hydrochloric acid concentrations. In contrast, a neutral ligand can be stripped with a low molarity acid, which makes the evaporation much easier. The effort to demonstrate the feasibility of this process started 10 years ago with micro amounts of curium and californium. Over the years, the amount increased, and a proof of concept was performed in 2023 with about 5% of the californium campaign. The results were adequate, and the current campaign used this approach a few weeks ago. Results from the early tests through the most recent run will be presented, along with successes and lessons learned.

Development of an Am/Bi Separation after Oxidation of Am(III) using $NaBiO_3$

Jennifer M. Pyles[1]*, Adrianna L. Orsi*[1]*, Jeffery D. Einkauf*[2]*, Luke R. Sadergaski*[3]*, Lætitia H. Delmau*[3]*, Bruce A. Moyer*[2]
and Jonathan D. Burns[1]

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To support the implementation of nuclear power into a sustainable energy strategy, our research has been evaluating the utility of the CoXTL Concept for over a decade. The CoXTL Concept aims to address the challenge of used nuclear fuel (UNF) recycling in a single-step separation utilizing crystallization. The ideal method for UNF recycling should maximize the recovery of the usable materials in the UNF, minimize the impact of waste bound for a geological repository, maintain nonproliferation standards, and have the ability to scale the process for a diverse fleet of reactor fuels.[1] The CoXTL Concept, based on hexavalent actinide co-crystallization, has shown promise as an alternative approach to nuclear fuel reprocessing by utilizing hexavalent transuranic elements (U-Am) as dioxo actinyl molecular ions, AnO_2^{2+}, recovered in a uranyl nitrate hexahydrate (UNH) crystal lattice.[2, 3] For the CoXTL Concept to be adopted, the challenge to effectively and efficiently oxidize Am to the hexavalent state must be addressed due to the high redox potential of ~1.75 V. Sodium bismuthate ($NaBiO_3$) has served as the benchmark for the generation of Am(VI) in acidic solution, [4] where our research specifically focuses on a HNO_3 matrix. [5, 6] Several optimization studies of the Am(III)/Am(VI) oxidation were conducted, which have created a need to reprocess the Am(III) for future experimentation by removing the Bi from the solution.

A method to recover the Am(III) from the oxidation studies with NaBiO3 is under development, utilizing Ce as a surrogate for Am. Mass ratios of Ce:Bi from 1:1 up to 1:100 were evaluated and analyzed by ICP-OES, focusing on maintaining the HNO_3 matrix. Current progress on these separations will be discussed for both TK200 and TK201 resins. The results demonstrated >90% purity and recovery of both Ce and Bi.

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Progress for HALEU Production at the Savannah River Site

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High-Assay Low-Enriched Uranium (HALEU) fuels are being developed to support the replacement of Highly Enriched Uranium (HEU) fuels used in U.S. High-Performance Research Reactors (USHPRR) as well as advanced nuclear power reactor designs. The projected demand for HALEU fuel far exceeds the supply and studies are underway to assess various options to partially mitigate the potential short supply. For almost a decade, the H-Canyon facility at the Savannah River Site (SRS) produced Low Enriched Uranium (LEU) containing 4.95% U-235 from the reprocessing of highly enriched foreign and domestic research reactor fuel for the Tennessee Valley Authority's (TVA) commercial power reactor market. The production of LEU at the H-Canyon facility can be readily transitioned to produce 19.75% HALEU solutions from the current separated inventory of purified HEU solutions in H-Canyon storage.

The Savannah River National Laboratory (SRNL) has supported this effort through characterization of a storage tank containing the current separated inventory of purified uranium solutions available at SRS. Through work with a potential fuel fabrication facility, specifications for the HALEU fuel solutions have been defined as well as the methods necessary to achieve the minimum limits of detection. Select methods of analyses will be presented for the consolidated tank inventory. Downblending will utilize a natural uranium stock solution that is part of the SRS inventory.

High-Pressure Ion Chromatography chemical separations for Inductively-Coupled Plasma Mass Spectrometry assay at Oak Ridge National Laboratory

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Mass spectrometry is a powerful analytical technique widely used for the precise measurement and characterization of metals in diverse materials. However, when applied to radiochemical isotopic analyses, significant challenges arise due to molecular and isobaric interferences. Overcoming these interferences is essential for generating accurate and reliable data.

At Oak Ridge National Laboratory's Radioactive Materials Analytical Laboratory (RMAL), researchers have developed and validated advanced methods that integrate high-pressure ion chromatography (HPIC) with inductively-coupled plasma mass spectrometry (ICP-MS). This combined HPIC–ICP-MS approach enables real-time, automated separation and analysis of radioactive and stable isotopes, offering substantial efficiency gains over traditional manual ion-exchange column techniques.

The HPIC–ICP-MS method effectively mitigates many complex interference issues that complicate isotopic analyses of lanthanides and actinides. In particular, it enables the precise determination of neodymium, uranium, and plutonium isotopes, which are critical for accurate spent nuclear fuel burnup calculations. Beyond this, the developed separation methodology also provides versatility, allowing simultaneous and precise analyses of actinides such as americium, curium, californium, and plutonium within a single analytical sequence with high precision. This advancement streamlines workflows, improves analytical accuracy, and enhances safety by minimizing manual handling of radioactive materials due to its automated nature.

Orphaned Fuels: Adopting a Domestic Nuclear Fuel Cycle

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The United States has a storied history in the development and testing of a diverse set of nuclear fuels for varying reactor applications. Unfortunately, these development activities have significantly outpaced strategy and implementation of final disposal pathways that include recycle. This combination of events has led to the production of orphaned fuels that do not have a clear disposal pathway. As the push to recycle spent nuclear fuel increases, the need to address final disposal pathways for new fuels and orphaned fuels will only increase. FuelOx, the gas-phase oxidation of used nuclear fuel, has the potential to accept a wide variety of fuel types while producing a single output product. This would allow the integration of a wide variety of fuel forms into a generalized fuel recycling program. In this talk we will discuss strategies and updates to FuelOx that enable the recycling of orphaned nuclear fuel.

This work was supported by the US Department of Energy Office of Nuclear Energy Materials Recovery and Waste Form Development Campaign.

Next-Generation Caustic-Side Solvent Extraction Process for Cesium Separation from Hanford Tank Wastes Containing High Potassium and Complexant Concentrations

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Results described in this presentation demonstrate the versatility of the Next-Generation Caustic-Side Solvent Extraction process (NG-CSSX) to remove cesium from challenging legacy alkaline tank wastes containing high potassium and complexant concentrations as stored at the Hanford site.[1] Based on the ability of calix[4]arene crown-6 ethers to selectively bind Cs^+ ion, the NG-CSSX process and its forerunner, the CSSX process,[2] have together proven to be effective in treating millions of gallons of the salt wastes stored at the Savannah River Site (SRS) since 2008. By comparison, Hanford wastes have proven more challenging in being more varied in composition, especially in containing elevated concentrations of complexants and potassium. Previous tests showed that the presence of complexants in some of the Hanford wastes fortunately would not likely affect cesium removal by the CSSX process, but competitive potassium loading of the calixarene-crown extractant would weaken extraction of Cs^+ ion by effectively switching the mechanism to cation-exchange:

$$Cs^+_{aq} + [Calix(K)]^+X^-_{org} \rightleftharpoons [Calix(Cs)]^+X^-_{org} + K^+_{aq} \qquad (1)$$

where X^- is a co-extracted anion, mainly nitrate or hydroxide equivalents. In the present work, the Cs^+ and K^+ concentrations were followed in both phases in extraction, scrubbing, and stripping under NG-CSSX conditions. The Cs/K separation factor averaged 164 ± 11 in extraction from five complex simulated tank wastes, including four different Hanford variants and the average SRS waste simulant. Elevated complexant concentrations in two of the Hanford wastes gave no detectable effect, but increasing potassium concentrations up to 1 M effectively loaded the solvent more than 90%. Cesium distribution ratios (D_{Cs}) accordingly approached inverse dependence on aqueous potassium concentration (Eq. 1) to a minimum D_{Cs} of 8 at 1 M potassium concentration. Three scrubs with 25 mM NaOH removed >99.8% of the loaded potassium, allowing effective stripping with 10 mM boric acid. From the results, a 23-stage preliminary flowsheet was designed, delivering for the worst case a Cs decontamination factor of 15,000 with a Cs concentration factor of 30 and throughput of 20 gpm.

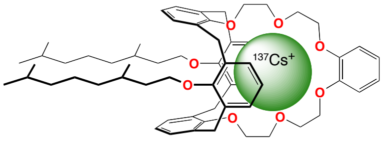

Figure. Calix[4]arene-crown-6 ether known as MaxCalix shown binding Cs^+ ion in its cavity subtended by six O atoms and two phenyl rings. The formulation of the NG-CSSX solvent used in this work consists of 0.05 M MaxCalix, 0.75 M Cs-7SB modifier, and 0.003 M *N*,*N*',*N*''-tri-*iso*-decylguanidine modifier (TiDG).

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Thank you for your participation in this conference.
We hope you have enjoyed it and wish you a safe return home.
See you in two years!