Friday Morning, November 13, 2026

Actinides and Rare Earths
Room 320 - Session AC-FrM

Actinides and Rare Earths Science

Moderators: Dr. Krzysztof Gofryk, Idaho National Laboratory, **Prof. Dr. Ladislav Havela**, Charles University, Czech Republic, **Dr. Paul Roussel**, AWE, **Dr. David Shuh**, Lawrence Berkeley National Laboratory

8:15am **AC-FrM-1 Environmental Radioactivity at the Department of Energy's Hanford Site: Sources and Sinks**, ***Carolyn Pearce***, Pacific Northwest National Laboratory; *Jay LaVerne,* University of Notre Dame; *Hilary Emerson, Amanda Lawter, James James Szecsody, Rob Mackley, Xin Zhang, Zheming Wang, Kevin Rosso, Gregory Schenter,* Pacific Northwest National Laboratory; *Linda Young,* Argonne National Laboratory; *Thomas Orlando,* Georgia Institute of Technology; *Aurora Clark,* University of Utah; *Xiaosong Li,* University of Washington; *Lynn Francesconi,* Hunter College, City University of New York **INVITED**

Plutonium production for the U.S. weapons program at the Department of Energy's Hanford Site created one of the world's largest environmental remediation challenges. Cleanup of 200 million liters of radioactive and chemically complex waste stored in 177 underground tanks will take decades and cost billions of dollars. Addressing the sources and sinks of environmental radioactivity requires: (i) characterization of radioactive tank waste chemistry and (ii) understanding contaminant transport and reactivity in the subsurface. The Ion Dynamics in Radioactive Environments and Materials (IDREAM) Energy Frontier Research Center provides mechanistic insights into precipitation and dissolution processes central to Hanford tank waste treatment. Hanford sludge contains water-insoluble aluminum oxides/ hydroxides that influence waste processing. Atomic force microscopy (AFM) showed that gibbsite dissolution in alkaline solutions proceeds through release of aluminate dimers rather than classical monomer detachment mechanisms. Gibbsite can also selectively adsorb or incorporate rare earth elements in tank waste. IDREAM developed an AFM integrated with an X-ray source to directly observe radiolytically driven interfacial reactions with unprecedented spatial resolution. These studies revealed how ionizing radiation, adsorbed water, and surface impurities alter mineral reactivity under highly alkaline conditions. Adsorbed carboxylates or chromium on boehmite nanoplates significantly slowed dissolution despite incomplete surface coverage. In parallel with tank waste retrieval, legacy disposal to cribs and trenches during plutonium separations operations produced vadose zone contamination and groundwater plumes that continue to impact the environment. Key groundwater contaminants include technetium-99, present as highly mobile pertechnetate (TcO_4^-), and uranium. Current remediation strategies include treatment of perched water zones and permeable reactive barriers. Technologies under evaluation include tin apatite particles, sulfur-modified iron particles, bismuth subnitrate particles, calcium polysulfide, and polyphosphate amendments. Proposed immobilization mechanisms include reduction, adsorption, incorporation into mineral phases, and in situ precipitation/coating reactions. Their effectiveness for sequestration of Tc-99 and uranium was evaluated through batch and column studies using Hanford sediments. Advances in understanding radioactive waste chemistry, radiolytic interfacial processes, and contaminant immobilization mechanisms are critical for reducing environmental risks associated with Hanford waste retrieval, treatment, and disposal.

8:45am **AC-FrM-3 Reaching the Monolayer Limit in a van der Waals Actinide Magnet**, ***Priscila Rosa***, Colorado State University **INVITED**

The discovery of local-moment magnetism in van der Waals (vdW) semiconductors down to the single-layer limit has led to a paradigm shift in the understanding of two-dimensional (2D) magnets and unleashed their potential for applications in microelectronic and optoelectronic devices. The incorporation of strong electronic and magnetic correlations in 2D vdW metals remains a sought-after platform not only to enable control of emergent quantum phases, such as superconductivity, but also to achieve more theoretically tractable microscopic models of complex materials. To date, however, there is limited success in the discovery of such metallic vdW platforms, and *f*-electron monolayers remain out of reach. In this talk, I will discuss experimental and theoretical studies of β-UTe_3, an actinide ferromagnet that can be exfoliated to the monolayer limit. A sizable electronic specific heat coefficient provides the hallmark of strong correlations. Remarkably, β-UTe_3 remains ferromagnetic in the half-unit-cell limit with an enhanced ordering temperature of 35 K, a factor of two larger than its bulk counterpart. Time permitting, I will also share recent spectroscopic insights into this system. Our work establishes β-UTe_3 as a novel materials platform for investigating and modeling correlated behavior in the monolayer limit and opens numerous avenues for quantum control with, e.g., strain engineering.

9:15am **AC-FrM-5 Theory of High-Resolution X-Ray Absorption and Resonant Inelastic X-Ray Scattering: Focus on Intermediate Valency**, ***Jindrich Kolorenc***, Institute of Physics (FZU), Czech Academy of Sciences, Prague, Czechia **INVITED**

I will discuss modeling of the high-energy-resolution x-ray absorption and the resonant inelastic x-ray scattering in actinide compounds by means of the charge-transfer multiplet theory, that is, using the Anderson impurity model constructed on the basis of first-principles electronic-structure calculations. As one of the applications, I will analyze uranium L_3 and M_4 x-ray absorption spectra taken on UPd_2Cd_{20}. These spectra indicate that the uranium 5f shell undergoes valence fluctuations in this compound [1].

The regime of valence fluctuations, known from intermetallics based on the anomalous rare-earth elements (such as Ce, Yb, Sm, Eu), has not yet been identified by microscopic techniques in any uranium compound. It was only suggested in a few of them on the basis of thermodynamic properties (such as the magnetic susceptibility) and on their similarity to the properties of the rare-earth valence fluctuators [2]. Such arguments, however, are hardly unequivocal. The x-ray absorption spectroscopy certainly provides a more direct probe of the 5f-shell behavior. Although UPd_2Cd_{20} has only one type of uranium site in its primitive cell, the absorption spectra obtained in the high-energy-resolution fluorescence-detection (HERFD) mode display two edges, each of which can be associated with a different valence state, $5f^2$ and $5f^3$. The shape of the L_3 spectrum is well approximated by two shifted copies of the uranium 6d density of states obtained in density-functional calculations, the shape of the M_4 spectrum is well approximated by a combination of UCl_3 ($5f^3$) and UCl_4 ($5f^2$) spectra reported previously [3]. This points to the $5f^2 \leftrightarrow 5f^3$ fluctuations on a timescale longer than the characteristic time of the spectroscopy technique. Unlike 4f systems, in which such fluctuations typically lead to a destruction of magnetism due to one of the involved $4f^n$ configurations being non-magnetic [4], the $5f^2$ and $5f^3$ states are both magnetic, allowing for a magnetic ordering in the fluctuating state—UPd_2Cd_{20}, in particular, is antiferromagnetic.

1. Y. Hirose *et al*., *Experimental observation of valence fluctuations by high-resolution x-ray absorption spectroscopy in UPd_2Cd_{20}*, Phys. Rev. B (2026), DOI: 10.1103/g9y8-7lq9
2. R. Troć, *The possibility of the mixed valence state in the uranium intermetallic compounds: $UCoGa_5$, U_2Ru_2Sn and U_2RuGa_8. The scaling properties*, J. Alloys Compd. **442**, 34 (2007).
3. C. L. Silva *at al.*, *On the origin of low-valent uranium oxidation state*, Nat. Commun. **15**, 6861 (2024).
4. D. I. Khomskiĭ, *The problem of intermediate valency*, Sov. Phys. Usp. **22**, 879 (1979).

9:45am **AC-FrM-7 Electronic Structure Studies of Tetravalent Actinides by Solid State NMR Spectroscopy**, ***Herman Cho***, *Khyati Anand,* Pacific Northwest National Laboratory; *Sejun Park,* Los Alamos National Laboratory; *Khusboo Rana, Eric Walter,* Pacific Northwest National Laboratory **INVITED**

The magnetic properties of tetravalent actinide centers differ markedly within the actinide row and with isoelectronic counterparts from the lanthanide row. Notable examples include the temperature independent paramagnetism of PuO_2,[1,2] exotic forms of multipolar order in UO_2 and NpO_2,[3,4] and the existence of multiple magnetic transitions in UF_4 and NpF_4.[5] These studies have provided definitive evidence that the assignment of the valence electrons of actinides to 5f orbitals oversimplifies the underlying electronic structure. With magnetic resonance spectroscopic techniques we have shown that local magnetic and electric fields at ligand sites and the metal itself can be measured in the solid state with better than part per thousand resolution. We apply this approach in systematic studies of electronic structure in an isomorphous series of An(IV) compounds including the tetrafluorides (Fig. 1)[6,7] and the dioxides (Fig. 2).[8] Plutonium, which is positioned in the 5f row between the light actinides -- characterized by itinerant valence electrons -- and higher *Z* elements that have valence electrons found to be in localized (atom-like) states, presents examples of both types of behavior.

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8. Rana, K.; Anand, K.; Park, S.; Sinkov, S. I.; Sweet, L. E; Cho, H. *Inorg. Chem*. **2025**, *64*, 21791-21795.

10:30am **AC-FrM-10 High-Resolution X-ray Spectroscopic Studies on Protactinium and Actinium***, **Jacob Branson**, Bianca Schacherl, Kiara Maurer, Tim Prüßmann, Kathy Dardenne,* Karlsruhe Institute of Technology, Institute for Nuclear Waste Disposal, Germany*; Rikard Malmbeck, Olaf Walter,* European Commission, Joint Research Center, Karlsruhe, Germany*; Tonya Vitova,* Karlsruhe Institute of Technology, Institute for Nuclear Waste Disposal, Germany **INVITED**

Despite occurring in nature, the chemistry of protactinium remains relatively unstudied due to its low abundance, radioactivity, and difficulty of isolation. However, existing studies on protactinium reveal chemical behavior and electronic structure that is unique even amongst the actinides, and this behavior has been attributed to the relatively large radial extent of the Pa 5f orbitals in tandem with their similar energy to the Pa 6d orbitals. In this work, the nature of the involvement of the Pa 5f and 6d orbitals is probed using high-resolution X-ray spectroscopy (HR-XANES) and resonant inelastic X-ray scattering (RIXS) experiments at the Pa $M_{3,4}$-edges, which have been demonstrated to be valuable probes of actinide electronic structure. For these studies, K_2PaF_7 and $Pa_2O_5{\cdot}xH_2O$ were synthesized in the solid state and characterized using Pa L_3-edge EXAFS. The Pa $M_{3,4}$-edge HR-XANES and RIXS experiments on these samples were conducted using two different scattering geometries and reveal dramatic differences in Pa 5f and 6d molecular orbital energetics and bond covalency between these coordination environments. We relate these differences to the competing influences of orbital energy and orbital overlap. These results form a basis of understanding for the electronic structure and chemistry of more complex Pa systems and demonstrate the high potential of the Pa 5f and 6d orbitals to participate in bonding interactions. The first experimental results on Ac M_3-edge HR-XANES of Ac-DOTA, relevant for understanding the bonding properties of Ac in radiopharmaceuticals, will also be presented and discussed. All experiments were conducted at the ACT end station of the CAT-ACT beamline at the KIT Light Source in Karlsruhe, Germany.

11:00am **AC-FrM-12 Manufacturing to Performance: Advanced Characterization of Nuclear Fuels***, **Robert Harrison**, Jonathan Morgan,* University of Manchester, UK*; Angus Wylie,* Massachusetts Institute of Technology*; Samira Bostanchi, Chris Green, David Pearmain,* Lucideon, UK*; Dave Goddard,* UKNNL, UK*; Mike Short,* Massachusetts Institute of Technology **INVITED**

Development of novel manufacturing techniques and advanced nuclear fuel materials offers significant economic and safety benefits for current and future reactor systems. However, linking processing to in-pile performance requires correlative advanced characterisation across multiple length scales. This work presents two case studies demonstrating this approach.

Firstly, field-assisted sintering (FAS) techniques, including spark plasma sintering (SPS) and flash sintering (FS), are being developed for UO_2 and mixed uranium–plutonium oxide (MOx) fuels. These methods significantly reduce sintering temperatures and processing times compared to conventional routes. However, understanding the impact of these rapid sintering processes on microstructure and chemical homogeneity—particularly plutonium distribution—is critical for fuel performance and reprocessability. A bespoke uranium-active FS system has been optimised (873-1173 K), achieving >95% theoretical density and ~5µm grain sizes, representing ~50% reductions in temperature and cycle time for UO_2. Extension to MOx surrogate systems (CeO_2-doped UO_2) revealed significant heterogeneity, including Ce-rich inclusions and degraded pellet quality. To resolve this, a correlative characterisation framework combining XRD, SEM-EDS, EPMA and Raman microscopy is employed to probe phase composition, chemical distribution and local defect chemistry. This multi-modal approach provides insight into the formation of heterogeneous regions and their relationship to processing conditions and pellet quality.

Secondly, uranium nitride (UN) is studied as an advanced and accident tolerant fuel. Ion irradiation (Ar, up to 100 DPA at 573 K) is used as a surrogate for neutron damage. Post-irradiation examination integrates XRD and TEM-EELS to characterise defect evolution, including dislocation loops, lattice swelling and gas bubble formation with Transient grating spectroscopy (TGS) to measure near-surface thermal diffusivity. This revealed a significant reduction of thermal conductivity at doses >10 DPA, directly correlated with nanoscale defect populations. In-situ annealing using TGS and TEM shows recovery of thermal transport above ~773 K, linked to point defect recombination. Overall, this work demonstrates how correlative, multi-technique characterisation links manufacturing, defect evolution and functional performance, supporting predictive understanding and accelerated development of advanced nuclear fuels.

11:30am **AC-FrM-14 Cooperative Phenomena in Locally Non-Centrosymmetric Heavy-Systems and Raman Effect in Cecosi***, **Gertrud Zwicknagl**,* TU Braunschweig, Germany

Heavy-fermion systems with magnetic ions in non-centrosymmetric sites have gained high attention during the past years. A prominent example is CeRh2As2 which exhibits a complex phase diagram with many unusual phases [1,2]. Theoretical discussions mainly focused on the role of the Rashba effect, i. e., on the consequences of local inversion symmetry breaking on the spins of the heavy quasiparticles [3].

In the present contribution, I will discuss consequences of local inversion symmetry breaking on the orbital degrees of freedom, i. e., the possible hybridization of orbitals with different parities. The inter-site interaction resulting from local Ce 4f-Ce 5d hybridization and d-d- hopping can lead to new cooperative phenomena like the lattice instability in CeCoSi [4].

I will discuss the consequences of these cooperative phenomena for the Raman spectra of CeCoSi [5].

[1] S. Khim et al, Science **373**, 1012 (2021)

[2] D. Hafner et al, Phys. Rev. **X 12**, 011023 (2022)

[3] see e. g. B. K. Nally and P. M. R. Brydon, New J. Phys. **26**, 093015 (2024) and references therein

[4] Takeshi Matsumura et al., Phys. Rev. **B 113**, 014415 (2026)

[5] Owen Moulding et al., arXiv: 2505.03249

11:45am **AC-FrM-15 Statistical Enhancement of Fissile Isotope Discrimination via Superposition of Fission-Track Clusters***, Rami Babayew, Yaacov Yehuda-Zada,* Nuclear Research Center Negev, Israel*; Galit Bar,* Soreq Nuclear Research Center, Israel*; Noam Elgad,* Nuclear Research Center Negev, Israel*; Danny Dayan,* Ben Gurion University Be'er Sheva, Israel*; Jan Lorincik,* Centre Řež, Czech Republic*; Itzhak Orion,* Ben Gurion University Be'er Sheva, Israel*; Shay Dadon,* Nuclear Research Center Negev, Israel*; Aryeh Weiss,* Bar Ilan University, Israel*; Galit Katarivas Levy, **Itzhak Halevy**,* Ben Gurion University Be'er Sheva, Israel

Accurate fissile isotope identification using Fission Track Analysis (FTA) is fundamentally limited by stochastic variations in individual fission-fragment trajectories and by the finite statistics obtainable from single-cluster measurements. While recent advances in automated image processing and synthetic-data-driven reconstruction have improved the robustness of FTA, isotopic discrimination between fissile materials with similar fission-fragment distributions remains challenging.

In this work, we propose a novel statistical enhancement framework based on superposition of reconstructed fission-track clusters originating from the same fissile grain. The method combines multiple realizations of track topologies into a unified statistical representation, effectively increasing counting statistics without increasing detector area or irradiation complexity.

Monte Carlo simulations using Geant4 were performed for ^{235}U, ^{233}U, and ^{229}Th embedded in aerogel-assisted LEXAN® solid-state nuclear track detectors (SSNTDs). Synthetic microscope images were generated and processed using an automated reconstruction pipeline to extract track morphology, cluster centroids, and Real Flight Path (RFP) distributions.

Statistical analysis demonstrated that the proposed superposition framework reduces stochastic uncertainty approximately according to: $\sigma_{eff} = 1/\sqrt{N}$

where N is the number of superposed cluster realizations. For example, superposition of N = 9 realizations improves discrimination between ^{235}U and ^{229}Th from ~3σ to ~9σ, and between ^{235}U and ^{233}U from ~1σ to ~3σ,

consistent with the expected √N scaling. More distinct isotope pairs exhibited even stronger enhancement, exceeding 5σ significance for modest superposition levels.

The proposed methodology transforms FTA from a primarily single-event morphological analysis into an ensemble-based statistical fingerprinting technique. This approach provides a scalable pathway toward improved isotopic sensitivity, automated nuclear forensic analysis, and enhanced safeguards verification, particularly for isotopes with partially overlapping fission-fragment signatures.

Future work will focus on experimental validation under neutron irradiation conditions and implementation of physics-informed alignment algorithms for real detector datasets.

Author Index

Bold page numbers indicate presenter