Actinides and Rare Earths
Room 320 - Session AC-ThM

Actinides and Rare Earths Early Career

Moderators: Dr. MIles Beaux, LANL, **Dr. Krzysztof Gofryk**, Idaho National Laboratory, **Prof. Itzhak Halevy**, Ben Gurion Uni. Be'er Sheva, **Prof. Dr. Gertrud Zwicknagl**, Technical University Brauschweig

11:00am **AC-ThM-13 Scanning Tunneling Spectroscopy Reveals Extreme Stoichiometric Sensitivity in Actinide Oxides***, **Benjamin Heiner**, Miles Beaux,* Los Alamos National Laboratory

With the recent availability of single crystals, we have characterized the surface of Np2O5 using scanning tunneling spectroscopy (STS) at cryogenic temperatures. In addition to the band gap, two electronic features were observed on the surface: finite electron density in the previously observed band gap and a semimetallic suppression of the density of states at the Fermi Energy. These were unexpected and unpredicted, but using a proven theoretical model, we attribute these behaviors to areas of the crystal surface with variable oxygen vacancies. The model predicts that as oxygen vacancies are added to the calculation, the neptunium 5f orbital density fills in the band gap of pure Np2O5 until the crystal eventually becomes semimetallic. This work contextualizes the characterization of plutonium substoichiometric sesquioxide by STS, which contradicts calculation of plutonium oxides to be semiconductors. That the band gap of Np2O5 collapses into semimetallic behavior leads us to hypothesize that stoichiometric plutonium oxides would also have band gaps, but the electronic structure of actinide oxides are sensitive to even slight deviations to atomic ratios.

LA-UR-26-23359

11:15am **AC-ThM-14 Structural and Magnetism of Lanthanide- and Actinide-based Pyrochlores***, **Binod Rai**, Gia Thinh Tran,* Savannah River National Lab

Pyrochlore oxides are of significant interest because of their structural flexibility, radiation tolerance, and complex magnetic behavior arising from geometric frustration. In lanthanide- and actinide-based pyrochlores, the corner-sharing tetrahedral network promotes competing magnetic interactions that can lead to unconventional magnetic ground states and emergent quantum phenomena. These materials are also promising candidates for nuclear waste immobilization due to their ability to accommodate radionuclides while maintaining structural stability.

In this work, we investigate the structural, magnetic, and thermodynamic properties of the solid-solution series $(Tb_{1-x}Er_x)_2Ti_2O_7$. The end-member compounds exhibit contrasting magnetic behaviors: $Tb2Ti_2O_7$ remains magnetically disordered at very low temperatures despite strong antiferromagnetic interactions, whereas $Er2Ti_2O_7$displays antiferromagnetic ordering. By systematically varying the Tb/Er ratio, we examine how competing anisotropies and exchange interactions influence the evolution of magnetic frustration and phase behavior. Structural characterization is correlated with magnetic susceptibility and heat-capacity measurements to establish composition-dependent trends. In addition, recent results on actinide-based pyrochlores will be presented, highlighting their structural stability and magnetic properties relevant to strongly correlated 5f-electron systems and nuclear materials applications.

11:30am **AC-ThM-15 Role of Surface Oxidation in the Interdiffusion Behavior of Uranium-Niobium Diffusion Couples***, **Michelle Greenough**, Joseph Boro, Adrian Gonzales, W. Preston Cole, Debra Rosas, Erik Oerter, Scott Donald, Tien Roehling, Tae Wook Heo,* Lawrence Livermore National Laboratory

Understanding uranium oxidation behavior under controlled environmental conditions is important for predicting the stability and long-term performance of uranium-based materials. In this study, oxidized pieces of uranium were then incorporated into uranium-niobium diffusion couples to serve as a system for evaluating the impact of oxide chemistry and thickness on uranium-niobium diffusion. Both the oxidized specimens and U-Nb diffusion couples were characterized using Auger electron spectroscopy (AES), X-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM), and electron probe microanalysis (EPMA). These complementary techniques allowed for correlation between oxide characteristics and the nature of the formed diffusion couple. Oxide chemistry and thickness were demonstrated to influence the resulting diffusion material after testing several different relative humidities.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344 and was supported by the LLNL-LDRD Program under Project No. 25-ERD-005.

11:45am **AC-ThM-16 Laboratory-Based X-Ray Absorption Spectroscopy of Actinide Materials***, David Shuh, Alexander Ditter,* Lawrence Berkeley National Laboratory (LBNL)*; **Haisley Windsor**,* Lawrence Berkeley Lab, University of California, Berkeley

Synchrotron radiation spectroscopy of actinide materials has led to marked improvement in the fundamental understanding of their *f*-electron chemistry and physics. Beamlines and endstations with unique spatial resolution, energy resolution, and intensity characteristics have made it possible to determine the chemical and physical properties of a range of actinide materials. The recent development of tunable laboratory-based x-ray instruments with sufficient performance, albeit not fully to the level of synchrotron radiation facility beamlines/endstations, are beginning to provide a reasonable alternative for x-ray measurements of actinide materials. The first results from the LBNL Sigray QuantumLeap x-ray absorption spectroscopy (XAS) spectrometer will be presented.

At the same time as interest and capabilities are growing in laboratory-based actinide XAS, there has been already been great activity in the tender x-ray region for high-resolution studies of actinide materials. As synchrotron radiation light source upgrades get underway at the Advanced Light Source (ALS-U) and at the National Synchrotron Light Source II (NSLS IIU), it is worthwhile to assess and present the possible impact of new, fully-optimized tender x-ray beamline/endstation capabilities on the field of actinide science conducted in energy region with respect to current capabilities.

12:00pm **AC-ThM-17 Scanning Tunneling Spectroscopy of Pu Compound Crystals***, **Miles Beaux**, Benjamin Heiner,* Los Alamos National Laboratory*; Andrew Yost,* Scienta Omicron*; William DeBenedetti, Filip Ronning, Eric Bauer, David Arellano, Derek Prada, Paul Tobash,* Los Alamos National Laboratory

Scanning tunneling spectroscopy (STS) has been performed on a variety of Pu compound crystals including PuB_4, $PuCoGa_5$, and $PuIn_3$, with the results revealing distinct characteristics of semiconductive, semimetallic, and metallic, respectively. Specifically, the STS measurement showed the PuB_4 to have a band gap of approximately 0.042 eV, $PuCoGa_5$ to have no band gap with an electron density of zero at (and only at) the Fermi energy (E_F), and $PuIn_3$ to have no band gap with electron density at E_F. STS measurements were performed at temperatures ranging from 25 Kelvin (above the supercritical transition temperature of $PuCoGa_5$) to room temperature. In addition to the STS results, structural scanning tunneling microscopy (STM) images were successfully obtained for the $PuIn_3$ and $PuCoGa_5$ surfaces. STM images of the $PuIn_3$ surface revealed step edges of approximate multiples of 0.8 nm consistent with a 111-surface orientation. STM images of the $PuCoGa_5$ revealed two distinct Moiré patterns consistent with different faces of the crystal structure.

Author Index

Bold page numbers indicate presenter