

## Actinides and Rare Earths Focus Topic

### Room A215 - Session AC+AS+LS-TuA

#### Forensics, Science and Processing for Nuclear Energy

**Moderators:** Paul S. Bagus, University of North Texas, Tomasz Durakiewicz, National Science Foundation, David Geeson, AWE

2:20pm **AC+AS+LS-TuA-1 Helium Implantation Studies in Metals and Ceramics for Nuclear Energy Applications, Microstructure and Properties**, *Peter Hosemann, M Baloach, S Stevenson, A Scott*, University of California, Berkeley; *Y Yang*, Lawrence Berkeley Lab, University of California, Berkeley

**INVITED**

Helium damage in materials is a concern for fission and fusion materials. The development of Helium bubbles in a solid can lead to fundamental property changes ranging from embrittlement to surface blistering. Especially shallow ion beam implantation is of interest to the fusion community since the surface degradation of materials is one of the main materials degradation mechanism. This work features a rapid Helium implantation and screening method based on nanometer precise helium implantation using the helium ion beam microscope (HIM). This tool allows multiple doses in the same grain with subsequent rapid materials examination. We evaluate single and polycrystalline SiC, W, Cu, and Ti in this work and find interesting channelling phenomena which have been fit to modelling results from the literature. Further we evaluate mechanical and microstructural property changes using TEM, nanoindentation, AFM and micropillar compression testing on these alloys.

3:00pm **AC+AS+LS-TuA-3 Origin of Element Selectivity during Solvent Extraction of Rare Earths: Studies of Model Interfaces using Synchrotron Radiation**, *M Miller, Y Liang, H Li, M Chu, S Yoo*, Northwestern University; *W Bu*, University of Chicago; *M Olvera de la Cruz, Pulak Dutta*, Northwestern University

**INVITED**

Solvent extraction, commonly used to remove lanthanides from aqueous solutions, is known to preferentially extract heavier lanthanides. To understand the underlying nanoscale mechanisms, we studied a model system: extractant monolayers floating on an aqueous solution containing lanthanide ions. X-ray fluorescence near Total Reflection (XFNTR) was used to detect interfacial cations, using synchrotron radiation at the ChemMatCARS facility, Advanced Photon Source.

We found that the interfacial density of adsorbed cations increases very sharply as the solution concentration is increased, reminiscent of a first-order phase transition. The threshold concentration is an order of magnitude lower for erbium ( $Z=68$ ) compared to neodymium ( $Z=68$ ). When the solution contains both ions equally, only erbium is found at the extractant interface, and the bulk erbium concentration must be decreased by one order of magnitude relative to neodymium before both species appear equally at the interface.

These results indicate that element selectivity during liquid-liquid extraction occurs at the extractant-laden interface. Electrostatic effects arising from the dielectric mismatch, ionic correlations, and the sizes of the cations can be used to theoretically model the observed behaviors.

4:20pm **AC+AS+LS-TuA-7 Analysis of Aged of Uranium Particles via X-ray Xpectromicroscopy**, *Andrew Duffin, J Ward*, Pacific Northwest National Laboratory

**INVITED**

Uranyl fluoride ( $\text{UO}_2\text{F}_2$ ) particles are a common by-product when uranium hexafluoride ( $\text{UF}_6$ ), a volatile form of uranium used in industry, is released into air. Uranyl fluoride particles continue to react in the environment, eventually moving towards uranium oxide. Understanding the chemical changes uranyl fluoride takes is important as the chemical state affects how this contaminant is transported in the environment. We employed scanning transmission X-ray microscopy (STXM) to map the chemical changes that occur in single particles of uranyl fluoride, aged under controlled conditions. We have shown that ligand K-edge X-ray absorption spectroscopy can fingerprint uranium chemical species at the nm spatial scale and we have used this specificity to gain insight into the transformation of uranyl fluoride to uranium oxide.

5:00pm **AC+AS+LS-TuA-9 Heat Transfer and Phase Stability of Early Actinides and Actinide Compounds**, *Dominik Legut, L Kývála, U Wdowik, G Jaglo, P Piekarczyk*, Technical University of Ostrava, Ostrava, Czechia; *L Havela*, Charles University, Prague, Czechia

**INVITED**

Actinides and especially their carbides as prospective nuclear fuel materials for the generation IV reactors were investigated using the density

functional theory. We demonstrate that their electronic, magnetic, elastic, and thermal properties can be at present well described if the spin-orbit interaction and partial delocalization 5f electrons is properly included in the computational approaches. One can well reproduce not only basic electronic structure but also elastic constants, phonon dispersions, and their density of states, provided by XPS, UPS, BIS, and inelastic neutron scattering data [1-4]. The localization of the 5f electrons could be captured using a moderate value of the on-site Coulomb interaction parameter. The case studies include a realistic description of the ground-state properties of elemental metals as Th, U and their monocarbides ThC and UC. The realistic description of the electronic structure and lattice dynamics (phonons) explains much higher thermal expansion and heat capacity in pure actinides (as Th) comparing with respective actinide monocarbides. The modelling also gives an insight up to which temperature the heat transport depends on lattice vibrations and where the electron transport starts to dominate. We can identify the vibration modes influencing the heat transport most. Carbides are compared with actinide oxides, where the major role is played by optical phonon branches [3]. The phase stability and elastic properties have been explored in a cascade of uranium hydrides [5], for which the requirement to reproduce the ferromagnetic moments gives an important feedback for the choice of on-site Coulomb U and Hund's exchange J.

1. U. D. Wdowik, P. Piekarczyk, D. Legut, and G. Jaglo, Phys. Rev. B 94, 054303 (2016).

2. L. Kývála and D. Legut (submitted to PRB April 2019)

3. P. Maldonado, L. Paolasini, P. M. Oppeneer, T. R. Forrest, A. Prodi, N. Magnani, A. Bosak, G. H. Lander, and R. Caciuffo, Phys. Rev. B 93, 144301 (2016).

4. Y. Yun, D. Legut and P. M. Oppeneer, J. Nucl. Mat. 426, 109 (2012).

5. L. Havela, M. Paukov, M. Dopita, L. Horak, D. Drozdenko, M. Divis, I. Turek, D. Legut, L. Kývála, T. Gouder, A. Seibert, F. Huber, Inorg. Chem. 57, 14727 (2018).

5:40pm **AC+AS+LS-TuA-11 Reactivity of Potential TRISO Fuel Barrier Layers (SiC and ZrN) with H<sub>2</sub>O Probed with Ambient Pressure Photoelectron Spectroscopy**, *Jeff Terry, M Warren*, Illinois Institute of Technology; *R Addou, G Herman*, Oregon State University

While the use of TRISO fuels has been long postulated within High Temperature Gas Reactors, another potential use for the TRISO fuels is as an accident tolerant fuel in Light Water Reactors (LWRs). Before TRISO fuels can be used in LWRs, the corrosion properties of the different layers of TRISO fuels must be well understood. Photoelectron Spectroscopy (PES) has long been utilized to study the oxidation behavior of materials due to its sensitivity to both element and chemical state. The problem with PES has been that it has historically been a technique that required Ultrahigh Vacuum conditions for measurements. This made it difficult to study corrosion in situ. New instruments have expanded the capabilities of PES. It is now possible to measure photoemission spectra at ambient pressure. We have measured the in situ corrosion of a SiC layer grown as a TRISO simulant at a pressure of 1 mbar of H<sub>2</sub>O at temperatures up to 500 C using an ambient pressure photoemission system. We see no oxidation of the SiC layer by water at temperatures up to 350 C. Above 350 C, the SiC begins to oxidize with the formation of SiO<sub>2</sub>. In contrast, ZrN reacts at a pressure of 1 mbar of H<sub>2</sub>O at room temperature. As the temperature increases, the ZrN layer is completely converted to ZrO<sub>2</sub>. In the TRISO fuel, the barrier layer is surrounded by pyrolytic carbon. We model the protective ability of the outer carbon layer by making multilayers SiC/C and ZrN/C and measuring these under the same conditions. We find that a 2 nm carbon layer prevents the underlying barrier layers from reacting with water.

## Author Index

### Bold page numbers indicate presenter

— A —

Addou, R: AC+AS+LS-TuA-11, **1**

— B —

Balooch, M: AC+AS+LS-TuA-1, **1**

Bu, W: AC+AS+LS-TuA-3, **1**

— C —

Chu, M: AC+AS+LS-TuA-3, **1**

— D —

Duffin, A: AC+AS+LS-TuA-7, **1**

Dutta, P: AC+AS+LS-TuA-3, **1**

— H —

Havela, L: AC+AS+LS-TuA-9, **1**

Herman, G: AC+AS+LS-TuA-11, **1**

Hosemann, P: AC+AS+LS-TuA-1, **1**

— J —

Jaglo, G: AC+AS+LS-TuA-9, **1**

— K —

Kývala, L: AC+AS+LS-TuA-9, **1**

— L —

Legut, D: AC+AS+LS-TuA-9, **1**

Li, H: AC+AS+LS-TuA-3, **1**

Liang, Y: AC+AS+LS-TuA-3, **1**

— M —

Miller, M: AC+AS+LS-TuA-3, **1**

— O —

Olvera de la Cruz, M: AC+AS+LS-TuA-3, **1**

— P —

Piekarz, P: AC+AS+LS-TuA-9, **1**

— S —

Scott, A: AC+AS+LS-TuA-1, **1**

Stevenson, S: AC+AS+LS-TuA-1, **1**

— T —

Terry, J: AC+AS+LS-TuA-11, **1**

— W —

Ward, J: AC+AS+LS-TuA-7, **1**

Warren, M: AC+AS+LS-TuA-11, **1**

Wdowik, U: AC+AS+LS-TuA-9, **1**

— Y —

Yang, Y: AC+AS+LS-TuA-1, **1**

Yoo, S: AC+AS+LS-TuA-3, **1**