Tuesday Evening, November 8, 2022

Actinides and Rare Earths Focus Topic
Room Ballroom A - Session AC-TuP

Actinides and Rare Earths Poster Session

AC-TuP-1 The Underlying Simplicity of 5f Unoccupied Electronic Structure*,* ***JG Tobin,*** U. Wisconsin-Oshkosh*; S. Nowak,* SLAC National Accelerator Laboratory*; S. Yu,* LLNL*; P. Roussel,* AWE, UK*; R. Alonso-Mori, T. Kroll, D. Nordlund, T. Weng, D. Sokaras,* SLAC National Accelerator Laboratory

Using a simple empirical model based upon the bremsstrahlung isochromat spectroscopy of elemental Th, it is possible to explain the recent high energy resolution fluorescence detection measurements of UF_4 (n = 2) and UCd_{11} (n = 3) as well as the new inverse photoelectron spectroscopy of Pu_2O_3 (n = 5), where n is the 5f occupation number. A critical issue in this analysis is the assumption that the Th 5f states are essentially empty, which will be confirmed both experimentally and computationally. Thus, for 5f systems, this simple model provides a unified and consistent picture of 5f unoccupied density of states in simple, localized systems, as the 5f occupation varies in the early part of the series, for n = 0, ⅔, 2, 3, and 5. See References 1 and 2 for further detail.

[1] J. G. Tobin, et al., J. Vac. Sci. Tech. A 39, 043205 (2021), https://doi.org/10.1116/6.0001007

[2] J. G. Tobin, et al., J. Vac. Sci. Tech. A 39, 066001 (2021), https://doi.org/10.1116/6.0001315

AC-TuP-4 Epitaxial Actinide Heterostructures: Synthesis and Characterization*,* ***Kevin Vallejo****, B. May, F. Kabir, C. Dennett,* Idaho National Laboratory*; P. Simmonds,* Boise State University*; D. Hurley, K. Gofryk,* Idaho National Laboratory

Actinide-based materials possess unique physics due to the presence of 5*f* electrons. Their study has been mainly focused on their nuclear fuel applications, leaving plenty of fundamental physics aspects open for investigation. The effective examination of the unique quantum phenomena in these materials requires high purity monocrystalline samples. However, thin film synthesis of actinide compounds is particularly underexplored relative to other material systems because of limited source availability and safety regulations due to their radioactive nature. The promises, challenges, and synthesis routes for these actinide-bearing heterostructures is discussed. Molecular beam epitaxy (MBE) presents an attractive avenue for the study of actinide heterostructures because of the high degree of control over dimensionality, strain, and interfaces. Idaho National Laboratory has recently installed an MBE chamber with the specific goal of studying uranium, cesium, and thorium containing compounds. To facilitate deposition of these low vapor-pressure elements, the chamber is outfitted with a quad-pocket electron beam source, several high temperature cells, and a nitrogen plasma source. Additional studies on transition metals with complex oxidation states (Zr, Nb, Mn, Ni, and Cr) will function as surrogates for the actinide-based nitride compounds. These new capabilities will provide unrivaled opportunities for exploration of functional and energy materials with complex electron correlations, together with important experiments for model validation in computational studies.

Actinides and Rare Earths Focus Topic
Room 318 - Session AC+LS+MI-WeM

Magnetism, Electron Correlation, and Superconductivity in the Actinides/Rare Earths

Moderators: James G. Tobin, University of Wisconsin-Oshkosh, **Ladislav Havela**, Charles University, Czech Republic, **Gertrud Zwicknagl**, Technical University Brauschweig, Germany

8:00am **AC+LS+MI-WeM-1 The Relation between Crystal Chemistry and Superconductivity in Actinide-Based Superconductors***, **Eteri Svanidze,*** Max Plank Institute, Dresden, Germany **INVITED**

Crystallographic features play an important role in the physical and chemical properties of a given solid-state material. In particular, actinide-based systems exhibit a wide range of properties – from unconventional superconductivity to peculiar magnetic orders. In this talk, I will highlight some of the old and new actinide-based superconductors, in which a delicate interplay between chemistry and physics is observed. A comprehensive characterization of properties of UBe_{13} has revealed a deep interrelation between the physical and chemical features. Notably, single crystals of this material tend of include many defects which have a dramatic effect on superconducting state [1]. Motivated by this issue, an alternative method of studying intrinsic properties is investigated [2-4]. By creating a micro-scale device, it is possible to measure intrinsic superconductivity of UBe_{13}, whichhas so far remained out of reach [4]. The properties of UBe_{13} are compare to those of other actinide-based superconductors – UTe_2 [5] and $Th_4Be_{33}Pt_{16}$ [6] – in which a strong coupling of lattice and superconducting properties is observed. By studying these systems, it is possible to expand the understanding of crystal chemistry of solid-state materials, while simultaneously providing an insight into which crystallographic parameters impact the physical properties of a given solid-state material.

[1] A. Amon *et al.*, “Tracking aluminium impurities in single crystals of the heavy-fermion superconductor UBe_{13},”*Sci. Rep.***8**, 10654 (2018)

[2] E. Svanidze *et al.*, “Revealing intrinsic properties of UBe_{13}”, *in preparation* (2022)

[3] A. Amon *et al.*, "Interplay of atomic interactions in the intermetallic semiconductor Be_5Pt", *Angew. Chem. Int. Ed.***58**, 2 (2019).

[4] I. Antonyshyn *et al.*, "Micro-scale device - an alternative route for studying the intrinsic properties of solid-state materials: case of semiconducting TaGeIr", *Angew. Chem. Int. Ed.***59**, 2 (2020)

[5] E. Svanidze *et al.*, “Intrinsic crystal structure of UTe_2”, *in preparation* (2022)

[6] P. Kozelj *et al.*, “A noncentrosymmetric cage superconductor $Th_4Be_{33}Pt_{16}$”, *Sci. Rep.***11**, 22352 (2021)

8:40am **AC+LS+MI-WeM-3 Revealing The Beauty of Uranium Compounds:the UMB_4 (M=V, Cr, Fe, Co, Mo, W, Re, Os) and UFe_xSb_2 Cases***, **Antonio Pereira Gonçalves,*** Instituto Superior Técnico, Univ. Lisboa, Portugal **INVITED**

Uranium-based phases with d-metals show a vast and rich variety of physical properties, some of them unique, which are mainly driven by the large 5*f* spin-orbit coupling and the degree of delocalization of the 5*f* states. The understanding of such behaviors is critical for the advance of Solid State Sciences and can lead to the discovery of new phenomena and ground states. In this talk, examples of uranium compounds with unusual properties are presented, the focus being on UMB_4 materials and UFe_xSb_2 alloys.

UMB_4 (M=V, Cr, Fe, Co, Mo, W, Re, Os) compounds are quite notable, crystallizing in the $YCrB_4$-type (for V, Cr, Fe, Co) and $ThMoB_4$ type (for Mo, W, Re, Os) polymorphs. Both structures are closely related, containing the same number of atoms in the unit cell and similar coordination polyhedral, which allows the comparison of their properties as a function of the d-metal. Their electronic structure is influenced by a strong hybridization between the B-2*p* states and both the *d*-metals *d*-states and the uranium (5*f*) valence band states. This avoids the formation of narrow bands with high density of states at the Fermi level, even if the 5*f*-*d* hybridization is weak, i.e. for late d-metals. Hence, the explored UMB_4 compounds are weak Pauli paramagnets, with a density of states at Fermi level of only ~4 times higher than for α-U. Those crystalizing in the $ThMoB_4$ structure show anomalous lattice vibrations, in particular the presence of low-energy Einstein modes, which can lead to superconducting ground states.

UFe_xSb_2 alloys crystallize in the $HfCuSi_2$ type structure and present a remarkable ground state evolution with Fe concentration. $UFe_{0.5}Sb_2$ shows a non-Fermi liquid behavior, with a change from metallic to semiconductor-upon applying magnetic field. In contrast, $UFe_{0.7}Sb_2$ (and, in a minor scale, $UFeSb_2$), shows an increase in electrical resistivity with decreasing temperature under zero magnetic field, but for high magnetic fields a metallic behavior is established. First principles calculations indicated that in $UFeSb_2$ some bands cross the Fermi level, but they are relatively steep and, as a result, the density of states at Fermi level is low, which supports the semi-metallic character of this composition. In the case of $UFe_{0.5}Sb_2$, a higher concentration of bands at the Fermi level, some of them relatively flat, exist, which cause a sudden step-shaped drop of density of states just above Fermi level. The magnetic field application can raise the Fermi level into the pseudo-gap, which is probably the reason for the change from a metallic-like to a semiconductor-like behavior.

11:00am **AC+LS+MI-WeM-10 Uranium Hydrides Revisited***, **Ladislav Havela,*** Charles University, Faculty of Mathematics and Physics, Czechia*; D. Legut,* VSB Technical University Ostrava, Czechia*; J. Kolorenc,* Institute of Physics, Czech Academy of Sciences, Czechia

Uranium hydrides are not only materials important for understanding of fundamental aspects of actinides. They are also relevant for nuclear technologies as well as for specific hydrogen storage tasks, as e.g. storing of tritium in nuclear fusion devices. Electronic structure of uranium hydrides (α- and β-UH_3, UH_2) reflects two contradictory tendencies. One is a charge transfer from U towards H, the other is a stability of the *f* shell. The latter is illustrated also on the case of rare earth hydrides with anomalous rare earth elements, such as Yb or Eu, which surprisingly retain their lower valence state, i.e. with a higher *f*-occupancy. The transfer is thus realized in U by the 6*d* and 7*s* electrons, which become noticeably depleted, but the 5*f* occupancies remain high. The concomitant reduction of the 5*f*-6*d* hybridization contributes, together with the volume expansion, to pronounced ferromagnetism of the U hydrides with Curie temperatures far above 100 K. We compare individual hydrides described by the GGA+*U* calculations, yielding numbers on the charge transfer or spin and orbital moments, with relevant experimental data. Interesting feature obtained from calculations is that the two different U sites in β-UH_3 have the respective magnetizations non-collinear. The same calculations yield also phonon dispersion relations, which are used to determine the temperature dependence of phonon contribution to heat capacity. The agreement in the paramagnetic range is so good that it allows to determine both the magnetic entropy and changes of the Sommerfeld coefficient between the ground state and paramagnetic state, the latter being twice higher. The position of the U hydrides on the map of localization-delocalization dichotomy will be discussed.

The work was supported by the Czech Science Foundation under the grant No. 21-09766S.

11:20am **AC+LS+MI-WeM-11 Towards a Better Understanding of the Rkky Interaction in Ce- and Yb-Based Compounds: Anisotropies from Cef Effects and Fermi Surfaces***, **Gertrud Zwicknagel,*** Institut für Mathematische Physik, Germany*; V. Zevin,* The Racah Institute of Physics, Israel

We calculate the influence of Crystalline Electric Field (CEF) effects and the Fermi surface topology on the indirect Ruderman-Kittel-Kasuya-Yoshida (RKKY) exchange interaction between local 4f moments in Ce- and Yb-compounds. Starting from a periodic Anderson model, we show that the anisotropy of the CEF ground state manifests itself in the anisotropy of the effective exchange coupling constant J(R,R’) between two moments at sites R and R’. If spin-orbit effects among the conduction states can be neglected, the interaction between two Kramers doublets is isotropic in pseudo-spin space. Adopting the Stationary Phase approach, we derive the asymptotic form of the exchange constant for arbitrary Fermi surfaces. We evaluate the anisotropy of the RKKY interaction in tetragonal Ce- and Yb-122 compounds for realistic model bands. The influence of spin-orbit interaction among the conduction electrons is discussed.

11:40am **AC+LS+MI-WeM-12 Mechanically Forced Tuning of Interactions in Tetragonal 221 Intermetallics**, ***Petr Král**, J. Prchal,* Charles University, Czech Republic; *J. Kaštil,* Czech Academy of Sciences, Czech Republic; *D. Daisenberger,* Diamond Light Source, UK; *D. Staško,* Charles University, Czech Republic

Family of R_2T_2X intermetallics adopting the tetragonal Mo_2FeB_2-type crystal structure represents the group of anisotropic systems with potential to exhibit geometrical frustration effects due to the Shastry-Sutherland-like arrangement of *R*-atoms in the aa-planes alternated by usually non-magnetic *T-X* planes along the crystallographic *c*-axis. Crucial for resulting magnetic behavior are the interatomic distances affecting the exchange interactions between individual ions. Especially interesting may be the compounds close to the empirical boundary of crystal structure stability since, based on available data, the lowest observed unit-cell volume among Mo_2FeB_2-type materials is about $V_{\text{f.u.,crit}}$ = 200 Å^3.

The unique tool allowing to act directly on the interatomic distances without changes of chemical composition is mechanical pressure. Employing the high-pressure techniques, it is possible to influence all lattice-related physical properties including e.g. the magnetic interactions and electronic structure. Especially in the strongly anisotropic compounds, the hydrostatic pressure effect depends on the compressibility of individual lattice parameters. This is the case, when the uniaxial pressure may represent the opportunity to act selectively in chosen crystallographic directions and thus to provide the insight into the layout of interactions within the crystal lattice.

Herewith, we are presenting the results of systematical study of pressure impact on the R_2T_2X compounds. Special attention is paid to Ce_2Pd_2In considered to be one of the most interesting systems among R_2T_2X family. It represents the rare example of Ce-based ferromagnet, however the ground state is reached through the antiferromagnetic interphase. High sensitivity of magnetic behavior to hydrostatic pressure has been revealed showing the preference of antiferromagnetic phase at lower pressures followed by the Kondo-like anomaly formation and suppression of magnetic ordering at pressures around 4 GPa. Based on the results of high-pressure XRD experiment, the hydrostatic pressure acts more significantly within the basal plane. Indeed, different response to the uniaxial pressure applied along the *c*-axis was observed.

Compounds with the unit-cell volume closest to $V_{\text{f.u.,crit}}$ belongs to the series R_2Cu_2In. Among them the ferromagnets Dy_2Cu_2In (exhibiting the signs of spin-reorientation at lower temperatures) and Tm_2Cu_2In (with signs of spin-glass behavior) were investigated. Concerning the later one, the pressure was found to support the frustration effects responsible for the spin-glass properties as the unit-cell volume tends to reach the $V_{\text{f.u.,crit}}$.

12:00pm **AC+LS+MI-WeM-13 Effects of O_2 Growth Pressure on the Magnetization of $LaMnO_3$-$SrTiO_3$ Thin Films**, ***Ghadendra Bhandari,*** West Virginia University; *R. Trappen,* University of Waterloo; *N. Mottaghi, M. Holcomb,* West Virginia University

Magnetic properties of epitaxial $LaMnO_3$ thin films grown on $SrTiO_3$ substrate have been studied. Thin films are grown at various oxygen pressure using pulsed laser deposition and deposition has been monitored by reflection high energy electron diffraction (RHEED) to verify layer-by-layer growth. Bulk stoichiometric $LaMnO_3$ exhibits A-type antiferromagnetic order, whereas thin films show a ferromagnetic FM phase. Structure and magnetic properties have been characterized by X-ray diffractometry (XRD), X-ray reflectivity (XRR) and vibration sample magnetometry (VSM). The thin films are showing ferromagnetic (FM) phase. Depth dependent magnetization studied by polarized neutron beam reflectivity found that magnetization is higher near interfacial region and a magnetically dead region lies at the surface. We have observed c-lattice correlated magnetization, which is minimized at 30mTorr O_2 pressure.

Actinides and Rare Earths Focus Topic
Room 318 - Session AC+LS+MI-WeA

Chemistry and Physics of the Actinides/Rare Earths

Moderator: Krzysztof Gofryk, Idaho National Laboratory

2:20pm **AC+LS+MI-WeA-1** ***Novel Preorganized Ligands for Selective and Efficient Separation of f-Elements , Santa Jansone-Popova,*** Oak Ridge National Laboratory **INVITED**

Selective separation of f-elements (lanthanides and actinides) is challenging to realize due to their very similar properties. For example, lanthanides exist predominantly in 3+ oxidation state and their ionic radii decrease across the lanthanide series on average by only 0.01 Angstrom per unit increase in atomic number. Organic molecules (ligands) capable of magnifying these effects and selectively recognizing one element over another are extensively sought after. Implementation of such ligands in separating f-elements would result in substantial cost and energy savings. The neutral organic molecules decorated with at least four donor atoms (oxygens and/or nitrogens), designed to complex with metal ions and that do not undergo conformational change in the process, demonstrate exceptional size selectivity.[1-3] The innate rigidity and size of the cavity in addition to the electronic structure of ligands provide the means to control the selectivity across the trivalent f-element series by design. On the contrary, organic molecules with donor groups that are connected to freely rotating single bonds show high affinity for metal ions that are more Lewis acidic.[4] Superior performance of preorganized ligands in two immiscible solvent system in selectively separating adjacent lanthanides or lanthanides from actinides will be discussed in detail. More specifically, the chemistry of hydrophobic and hydrophilic bis-lactam-1,10-phenanthroline-based ligands having four donor atoms will be presented, focusing on the separation of adjacent lanthanides (e.g., Nd-Pr, Tb-Gd, Ho-Dy) and Americium-Europium pair separation. Additionally, the contrasting performance of rigid ligands decorated with two to four donor atoms will be shown and compared to their non-preorganized counterparts.

[1] Healy, M. R.; Ivanov, A. S.; Karslyan, Y.; Bryantsev, V. S.; Moyer, B. A.; Jansone-Popova, S. *Chem. Eur. J.*, **2019**, *25*, 6326.

[2] Karslyan, Y.; Sloop, F. V.; Delmau, L. H.; Moyer, B. A.; Popovs, I.; Paulenova, A.; Jansone-Popova, S. *RSC Adv.*, **2019**, *9*, 26537.

[3] Jansone-Popova, S.; Ivanov, A. S.; Bryantsev, V. S.; Sloop, F. V.; Custelcean, R.; Popovs, I.; Dekarske, M.; Moyer, B. A. *Inorg. Chem.*, **2017**, *56*, 5911.

[4] Stamberga, D.; Healy, M. R.; Bryantsev, V. S.; Albisser, C.; Karslyan, Y.; Reinhart, B.; Paulenova, A.; Foster, M.; Lyon, K.; Moyer, B. A.; Popovs, I.; Jansone-Popova, S. *Inorg. Chem*. **2020**, *59*, 17620.

3:00pm **AC+LS+MI-WeA-3 Impact of Noncovalent Interactions on Actinide Structural Chemistry*, Karah E. Knope,*** Georgetown University*; J. Wacker,* Lawrence Berkeley National Lab*; M. Shore,* Georgetown University **INVITED**

Noncovalent interactions (NCIs) have received significant attention over the past several years. This interest is fueled by recent examples that have shown that these interactions, which exist beyond the first coordination sphere of a metal ion, can be used to isolate novel structural units, push chemical equilibria, and tune redox behavior. As illustrated by these examples, it has become clear that consideration of NCIs is important for understanding the overall chemical behavior of a metal ion. Taken together with our group's interest in the factors that govern actinide speciation and reactivity, our recent work has thus focused on the solution and solid-state structural chemistry of actinide complexes and clusters that form from acidic aqueous solution in the presence of heterocycles capable of N-H hydrogen bonding interactions. Specifically, we have examined the identity (i.e. composition, charge, coordination number) of actinide structural units that precipitate from monoanionic ligands systems including Br^{1-}, Cl^{1-}, and NO_3^{1-} with N-heterocycles ranging from pyridinium to terpyridinium. While bromide, chloride, and nitrate all form weak relatively weak complexes with the actinides (Th-Pu), clear effects of NCIs are present only in the chloride ligand system with chloride rich (e.g. $[Th(H_2O)_2Cl_6]^{2-}$) to chloride poor (e.g. $[Th(H_2O)_7Cl_2]^{2+}$) complexes having been observed. By contrast, speciation is much more limited in the Br^{1-} and NO_3^{1-} ligand systems. Presented here will be an overview of our experimental and computational efforts to understand the effects of NCIs on phase formation.

4:20pm **AC+LS+MI-WeA-7 XPS Characterization of a Pu-7at.%-Ga Alloy*, Paul Roussel,*** AWE, UK*; K. Graham, S. Hernandez, J. Joyce, T. Venhaus,* Los Alamos National Laboratory

Gallium is often used in small concentrations (<10 at. %) as an alloying element with plutonium to stabilize the face centred cubic phase.Most XPS analyses of these plutonium materials have focused on low concentration alloys (< 4 at. %) where it is difficult to measure the signal and effects of gallium.Here we report the analysis of a higher alloy concentration of $Pu_{0.93}Ga_{0.07}$ in the as received state, during sputter depth profiling and following oxidation either from the addition of Langmuir quantities of oxygen or the in situ environment of the ultra high vacuum system.Quantification of the spectra has been undertaken by comparison of the elastic peak intensity and this has allowed us to test the recently reported relative sensitivity factors for plutonium [1]. The higher alloying concentration has permitted quantification of gallium in both the metal and oxides and, for the first time, the gallium chemical state plot for both alloy and oxide has been determined along with those of plutonium and oxygen.In addition to this we highlight a novel method to quantify the C1s peak which overlaps with the $Pu5p_{1/2}$ photoemission line and demonstrate the effect of gallium on the in situ oxidation rate of this alloy.

1) P. Roussel, A. J. Nelson, Quantitative XPS of plutonium: Evaluation of the Pu4f peak shape, Relative Sensitivity Factors and estimated detection limits, Surf. Interface Anal. https://doi.org/10.1002/sia.7083.

4:40pm **AC+LS+MI-WeA-8 Nanoscale Uranium Oxide: Correlating Colloidal Synthesis Pathways with Structure at the Atomic and Nanometer Length Scale*, Liane Moreau,*** Washington State University

Nanoscale uranium oxide is of interest for the development of advanced nuclear fuels and its importance in the corrosion of conventional fuel pellets and trends in uranium mobility within environmental systems. Knowledge of nanoscale properties of uranium oxide, however, are limited due to 1) a lack of synthetic methods capable of forming monodisperse actinide oxide nanoparticles and 2) the challenges associated with characterization of complex interfaces. Organic-phase colloidal approaches prove promising towards the synthesis of actinide oxide nanoparticles due to precedence for formation of highly monodisperse transition metal oxide nanoparticles, and the capability to synthesize particles in rigorously air- and water-free environments. Three systems will be discussed which exploit colloidal uranium oxide synthesis. The first explores oxidation kinetics in UO_2 nanoparticles. Synthesis is paired with X-ray diffraction and X-ray spectroscopy to compare nanoparticle oxidation trends with those observed in bulk counterparts under controlled oxidative environments. The second traces the formation mechanism of UO_2 nanoflowers and demonstrates the role of solvent interactions in anisotropic growth. The established growth patterns are particularly interesting as anisotropic growth in transition metal counterparts have traditionally depended on surface binding species and trace impurities rather than solvent chemistry. The third system investigates the deposition of uranium oxide onto iron nanoparticle cores, which serves as an analogue to environmental systems where uranium and plutonium have shown to preferentially bind to Fe-based minerals. The high-surface area particles provide a means to investigate the interface in atomistic detail. In describing the aforementioned systems, X-ray focused approaches and characterization method development will be discussed, which prove critical to resolving structural attributes and making connections between synthetic parameters and resulting atomic and nanoscale structure.

5:00pm **AC+LS+MI-WeA-9 Chemical Speciation Mapping of Spent Nuclear Fuel Using Soft X-Ray Spectromicroscopy at the Advanced Light Source*, Alexander Ditter,*** *D. Smiles, D. Lussier,* Lawrence Berkeley National Laboratory (LBNL)*; A. Altman,* Northwestern University*; M. Bachhav, L. He,* Idaho National Laboratory*; M. Mara,* Northwestern University*; S. Minasian,* Lawrence Berkeley National Laboratory (LBNL)*; C. Degueldre,* Lancaster, UK*; D. Shuh,* Lawrence Berkeley National Laboratory (LBNL)

A nuclear reactor is a complex environment, with high temperatures, large temperature gradients and a multitude of fission products in various states. Spent nuclear fuel offers a key window into this environment, but is an important object of study in its own right, particularly with regards to the release of radioisotopes into the environment. The oxidation of spent nuclear fuel is especially important as this can govern the transport and solubility of fission products. To study this process, a focused ion beam (FIB) was used to make thin sections of a low burnup spent fuel pellet at Idaho National Laboratory. These FIB sections were then measured at the O K-edge, U $N_{4,5}$-edges and Ce $M_{4,5}$-edges at the scanning transmission x-ray microscope (STXM) at Advanced Light Source (ALS) Beamline 11.0.2. O K-edge results were analyzed by a modified form of non-negative matrix factorization and revealed two main components, the bulk of the sample which is made up primarily of UO_2, and a thin (est. 8 nm) layer of UO_{2+x} on the surface of the FIB section. This oxidation occurred after FIB sectioning as a thin layer over the entire sample. Cerium measurements showed that the Ce (~ 0.4 wt. %) is at least predominantly in the 3+ oxidation state, though a small contribution of tetravalent cerium cannot be ruled out, representing the first known measurements of Ce oxidation state in spent nuclear fuel. This work lays the foundations for future STXM measurements of FIB sections of spent nuclear fuel which can be expanded to mixed-oxide, uranium nitride, or other advanced fuels.

5:20pm **AC+LS+MI-WeA-10 Structural, Thermodynamics, and the Electronic Properties of Al, Ga, In, and Tl Stabilized δ-Pu***, **Sajib Barman**, S. Hernandez,* Los Alamos National Laboratory

The 5*f* electrons in plutonium show mystifying character contrary to other elements in the periodic table, where there are six solid state phases of plutonium that undergo five solid-state phase transformations in a relatively short temperature range. The low temperature α-phase is brittle and has a monoclinic structure where the high temperature δ-phase is ductile and has an FCC structure. From a metallurgical point of view, the ductile δ-Pu is mostly important due to its favorable elasticity compared to the brittle α-Pu. Therefore, stabilizing δ-Pu at room temperature can be done by alloying with Group IIIA elements (X = Al, Ga, In, and Tl). Nevertheless, the alloyed δ-Pu stability zones highly depend on the alloying content, where the stability zone decreases with increasing atomic number of the Group IIIA elements. In this work, we will present a systematic density functional theory investigation of the thermodynamic stability of alloyed δ-Pu with Group IIIA elements as a function of alloying content (3.125, 6.25, and 9.375 at. %). We have calculated that the δ-Pu-Ga alloys have the highest thermodynamic stability compared to the other Group IIIA alloys, where the stability trend follows Pu-Ga > Pu-Al > Pu-In > Pu-Tl at 0K. After volume optimization, the volume contracts for Pu-Ga and Pu-Al alloys, while the volume expands for Pu-In and Pu-Tl alloys with increasing alloy content. Finally, we will show the electronic structure analysis to discuss the electronic interaction between Pu and the Group IIIA elements within the δ-matrix.

This work was carried out under Los Alamos National Laboratory's LDRD-DR project #20210001DR.

Actinides and Rare Earths Focus Topic
Room 318 - Session AC+AS+LS-ThM

Emerging Topics and Methods in Actinide/Rare Earth Science

Moderators: David Shuh, Lawrence Berkeley National Laboratory, **Alison Pugmire**, LANL, **Paul Roussel**, AWE, UK

8:00am **AC+AS+LS-ThM-1 Nuclear Forensics 2020:A Strategic Inflection Point***, **David Willingham,*** Lawrence Livermore Laboratory **INVITED**

The terrorist attacks of 9/11/01 greatly increased the visibility of nuclear forensics, as policy makers became increasingly concerned about the possibility of well-organized terrorist groups obtaining a nuclear weapon or dirty bomb.The Departments of Defense (DOD), Energy (DOE), Homeland Security (DHS), and State (DOS), as well as the Federal Bureau of Investigation (FBI) and the intelligence community, all play key roles in nuclear forensics. DHS, for example, was founded in November 2002 and included, from the start, a formal nuclear forensics program in its Science & Technology Branch.An August 2007 presidential decision directive established the specific roles these agencies would play and formally established the National Technical Nuclear Forensics Center (NTNFC) within DHS to coordinate planning, integration, assessment, and stewardship of the U.S. government's nuclear forensics capabilities. In 2010, the Nuclear Forensics & Attribution Act established a National Nuclear Forensics Expertise Development Program (NNFEDP) within the NTNFC aimed at "developing and maintaining a vibrant and enduring academic pathway from undergraduate to post-doctorate study in nuclear and geochemical science specialties directly relevant to technical nuclear forensics."National laboratories like LLNL have particularly benefitted from the establishment of postdoctoral fellowships.

In 2021, primary responsibility for nuclear forensics within the US Government transferred from DHS to the National Nuclear Security Administration (NNSA) within DOE with the issuance of National Security Presidential Memorandum 35, National Technical Nuclear Forensics. However, the Nuclear Forensics & Attribution Act of 2007 is still in force, which defines certain roles for DHS, particularly for stewardship of the nuclear forensics' workforce. Starting with the FY21 budget, there have been substantial increases in funding for NNSA for both nuclear forensic operations and R&D. However, the exact configuration of the nuclear forensics expertise development program(s) going forward is still being determined."

This talk will highlight some of the outstanding research conducted by our DHS postdoctoral fellows, including:

1. Development of RIMS as a tool for in situ analyses for nuclear forensics.
2. Improved determination of half-lives and branching ratios important for U rad-chem
3. Exploration of the use of isochrons for age dating of impure samples.
4. Development of new stable isotopic systems as new sources of signatures for nuclear forensics.
5. Development of rapid methods for dissolving solid samples

8:40am **AC+AS+LS-ThM-3 The Non-Integer Occupancy Ground State Hypothesis***, **Miles Beaux,*** Los Alamos National Laboratory **INVITED**

A deeply ingrained and long-standing practice exists for identifying integer orbital occupancy ground state electronic configurations for neutral atoms of the elements. For certain elements, the identification of the electron occupancies of the orbitals in a neutral atom can be a controversial topic, instigating heated debate among scientists. The pedagogy of how atomic structure and the periodic table are initially taught might serve as a driver for this practice. For example, the octet rule is often used to explain the most energetically favorable ionic states, similarities in properties of like-group elements, and the most stable compounds formed by the transfer and sharing of electrons. However, the octet rule is also insufficient to explain the existence, structure, and properties of transition metal elements, including the lanthanides and actinides.

A more fundamental quantum-based understanding of electronic structure provides a firm basis for the overall structure of the periodic table. Specifically, the octet rule is understood as the stability of completely filled *s*-, and *p*-orbitals each having electron capacities of two (l=0; m_l=0; m_s=±½) and six (l=1; m_l=-1,0,+1; m_s=±½), respectively, with each Period, *n*, having n_s and n_p orbitals (except for Period 1, for which no 1*p* orbital exists). Extrapolation of this quantum mechanical underpinning of the Periodic Table explains the existence of the *d*- and *f*-blocks. The filling patterns of the orbitals for elements in the Period Table reveals relative energies of the various orbitals. Deviations from the filling pattern are often explained to varying degrees of satisfaction by the interplay between Hund's rule and the Aufbau principle for near-degenerate states; the stability of empty, half-filled, and completely filled orbitals; and core level screening. As the energy landscape for electron orbitals becomes more crowded for higher Period elements, the potential for near energy degenerate states increases. The potential for quantum superposition of electrons in these near energy states leading to effective non-integer orbital occupancies will be discussed in the context of observed instabilities in actinide and rare earth elements. Experimental and theoretical efforts to investigate this hypothesis for a series of Np, Pu, and Am compounds will also be described.

9:20am **AC+AS+LS-ThM-5 Legacy Plutonium at the Hanford Site***, **Edgar Buck**, D. Reilly, G. Hall, K. Kruska, L. Liu, S. Triphathi, B. McNamara, A. Casella, D. Meier,* Pacific Northwest National Laboratory **INVITED**

The morphological characteristics of plutonium materials may provide information on the processes that were used to create the material; however, understanding of the detailed thermodynamic and kinetic processes needed to predict the evolution of its particle size distribution, crystal habit, and agglomerated state is still evolving. There has been an effort to fill this technical gap using an integrated experimental and modeling approach for formation of plutonium phases, including oxides and oxalates. Several different types of plutonium phases have been observed in wastes at the Hanford site, such as the Z9 crib near the former plutonium finishing plant and plutonium solids found in the SY102 and TX118 tanks. Laboratory experiments have been conducted to probe the formation mechanisms for these materials. By examining the preciptiating plutonium solids using a combination of in-situ optical microscopy (OM), scanning electron microscopy (SEM), cryo-electron microscopy (CryoEM) and in-situ transmission electron microscopy (TEM)., we have been able to demonstrate the occurrence of non-classical crystalline pathways for plutonium particle growth in some instances.

Understanding crystallization pathways in plutonium materials depends on the ability to unravel relationships between the intermediates and final crystalline products at the nanoscale, which is a particular challenge with radioactive materials. However, these powerful new tools of in-situ and cryoEM are providing new insights into the plutonium chemical system. The experimental data is helping to parameterize the computational modeling with the potential to lead to the development of predictive tools for identification.

11:00am **AC+AS+LS-ThM-10 Focused Ion Beam for Spatially Resolved Morphological Analysis of Nuclear Materials***, **Brandon Chung**, S. Donald , D. Rosas, S. Sen-Britain, V. Som, N. Teslich, A. Baker,* Lawrence Livermore National Laboratory*; A. Ditter, D. Shuh,* Lawrence Berkeley National Laboratory

Nuclear forensics requires accurate identification of distinguishing material characteristics of interdicted nuclear materials. Local morphological and chemical variations in nuclear materials are nearly ubiquitous due to the varying provenance, process, and pathways. Conventional analysis using bulk nuclear material provides an overall analytical value, but a typical nuclear material is heterogeneous, requiring highly spatially resolved characterization. Consequently, averaging material characteristics may obscure critical forensic signatures resulting from local chemical and structural variations. We will describe our efforts to strengthen operational and scientific methodologies to employ the focused ion beam-scanning electron microscopy (FIB-SEM) on uranium (U) and plutonium (Pu) materials for direct three-dimensional (3D) morphological analysis or to prepare site-specific material features to obtain spatially resolved characterizations using transmission electron microscopy (TEM) and X-ray synchrotron spectromicroscopy. Our work shows the internal morphological and chemical variations in host nuclear materials provide valuable information about the material's production processes and recent environmental exposures. We identified that internal microscopic features entrapped within bulk nuclear materials contain trace elements or chemical that are attributable to their origin, processing, and environmental exposures.

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11:20am **AC+AS+LS-ThM-11 Studying Combined Influence of Alpha Irradiation and Dissolved Hydrogen on UO-$_2$ Corrosion Using a Microfluidic Electrochemical Cell***, **Jennifer Yao**, B. McNamara, M. O'Hara,* Pacific Northwest National Laboratory*; N. Lahiri,* Pacific Northwest National Lab*; E. Ilton, C. Wang, E. Buck,* Pacific Northwest National Laboratory

It is well accepted concept that α-decay is the most important source of radiation in the spent nuclear fuel (SNF) after 1,000 years [1]. The influence of α-irradiation in the presence of dissolved H_2 on the corrosion of UO_2 can provide important information to assess the impact of the long-term SNF to the storage environment. However, experiments with bulk amount of SNF are expensive owing to the need for shielded hot cell facilities to protect researchers from the intense radiation field. To address this challenge, we employed a novel invention, particle-attached microfluidic electrochemical cell (PAMEC), to investigate UO_2 corrosion under different conditions (e.g., α-irradiation and H_2) at the microscale. Less than 10 µg of UO_2 (containing 1% to 10% $^{233}UO_2$) powder was mixed with polyvinylidene fluoride (PVDF) and carbon black to form the working electrode and included into PAMEC, which ^{233}U was used as alpha source to simulate the"aged" spent fuel [2]. The response of the corrosion potential of a ^{233}U contained UO_2 working electrode to dissolved H_2 in 0.1 M $NaClO_4$ (pH=9.5) will be presented. In addition, the 50nm thick Si_3N_4 detection window on PAMEC allows in situ imaging of the corrosion process using the high-resolution imaging technique, such as scanning electron microscope (SEM) coupled withEnergy-dispersive X-ray spectroscopy (EDS). Furthermore, X-ray photoelectron spectroscopy (XPS) was used to determine the oxidation state of the UO_2 electrode that was exposed to alpha radiation and dissolved H_2. Our work demonstrates the study of combined influences on UO_2 corrosion under the conditions that are known to be present in the long-term SNF environment by employing a microfluidic electrochemical cell. We vision this approach can be widely applied to study the influences of conditions that resemble the practical repository environment on SNF, while with greatly reduced hazardous risk when performing such experiments.

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11:40am **AC+AS+LS-ThM-12 A Model to Extract the Size-Dependent Surface Structure of Actinide Oxide Nanoparticles***, **Shinhyo Bang**, L. Moreau,* Washington State University

Characterization of actinide oxides at the nanoscale presents unique challenges due to their radioactivity, high surface area, and inherent diffraction broadening due to small grain size. Extended x-ray absorption spectroscopy (EXAFS) is an analytical method to investigate atomic-scale structural properties that enables their encapsulation and does not rely on long-range order. There is a limitation that EXAFS only gives the averaged structural information of heterogeneous samples. We aimed to deconvolute EXAFS results to extract the surface coordination environment of UO_2 NPs by proper modeling, and investigate how it evolves with varying sizes (1.4, 4.7, 8 nm). The termination effect was used to quantify the surface terminating species of UO_2 NPs. A higher degree of oxygenation on the surface was observed for 8 nm NPs. EXAFS simulation was implemented to backtrack the surface structure of these NPs. It was observed that the bond contraction due to the surface relaxation effect was localized in a few outermost layers of NPs and the surface disorder of 1.4 and 4.7 nm NPs was significantly enhanced from that of bulk.

Applied Surface Science Division
Room 320 - Session AS+AC+BI+CA+HI-ThM

Unraveling the Composition of Complex Systems with SIMS

Moderators: Steve Consiglio, Tokyo Electron, **Gregory L. Fisher**, Physical Electronics

8:20am **AS+AC+BI+CA+HI-ThM-2 Ex-Situ, Surface and Bulk Investigations of Defluxing Chemistry Effects on Solder Mask***, **J. Elliott Fowler**,* Sandia National Laboratories*; R. Gerhardt,* Georgia Institute of Technology*; J. Ohlhausen, R. Callaway,* Sandia National Laboratories*; M. Watt,* Georgia Institute of Technology*; S. Grosso, S. Rosenberg,* Sandia National Laboratories

Solder masking is an integral process in the manufacture of printed circuit board assemblies (PCBAs) – it serves to protect the unfinished copper traces from environmental effects, define the soldering pads and provide an adherable surface for conformal coating. Liquid photo-imageable (LPI) solder mask is one of the most popular choices for PCBAs due to ideal electrical and physical properties as well as chemical. LPI solder mask is a heterogeneous epoxy acrylate-based matrix, thus its surface chemistry can be very distinct from its overall bulk chemistry. The surface chemistry is of particular interest as it must be compatible with and resistant to a wide range of chemistries and environments which the PCBA will experience during its production and fielding lifetime. For instance, sensitivity to moisture is a well-known issue. Several studies of solder mask moisture absorption reveal that it reaches a saturation of ~1 weight percent in high-humidity environments, and corresponding changes in insulative properties occur. Risks to mask performance have arisen as a result of the use of new flux formulations which require increasingly aggressive aqueous defluxing chemistries for removal. We hypothesize that alkaline defluxing chemistries will significantly modify the surface chemistry of solder mask, making it more vulnerable to moisture and thus deteriorating their insulative properties.

A commercially available LPI solder mask material was prepared on squares of FR4 board and exposed to increasingly alklaine cleaning chemistries: including DI water, a pH neutral and a pH >10 defluxing chemistry. Samples were tested with three complementary surface-sensitive analytical techniques, x-ray photoelectron spectroscopy (XPS), time of flight secondary ion mass spectrometry (TOF-SIMS) and contact angle goniometry. TOF-SIMS results show that both pH neutral and alkaline chemistries alter the surface, removing PDMS and leaving organic residues. Contact angle goniometry shows that the alkaline chemistry makes the board's surface significantly more hydrophilic. XPS results show a decrease in Si at the surface correlating to a decrease in PDMS versus the control, with the largest decrease occurring due to the alkaline chemistry. In all cases, the surface is somewhat recoverable; however, recovery is less pronounced with increasing pH. AC impedance spectroscopy and DC Surface Insulation Resistance (SIR) testing of exposed samples in elevated humidity environments was performed to resolve changes in insulative performance due to observed changes in mask surface chemistry.

SNL is managed and operated by NTESS under DOE NNSA contract DE-NA0003525

8:40am **AS+AC+BI+CA+HI-ThM-3 Unraveling the Composition of Complex Systems with SIMS***, **Birgit Hagenhoff**,* Tascon GmbH, Germany **INVITED**

For more than four decades SIMS has proven to be a valuable tool in academic research as well as for industrial applications. Whereas the first years focussed on understanding the underlying physical processes using mono-elemental samples, the advent of ToF-SIMS (Time-of-Fligt Secondary Ion Mass Spectrometry) instrumentation in the 80s of the last century opened the path to using SIMS as a screening tool for characterising "the unknown", i.e. samples about which hardly anything is known before the analysis, where sample preparation should be as scarce as possible and where the analysis is performed without any chromatographic pre-step to separate out sample compounds.

Over the years, intensive discussions between those developing instruments and those applying them on a daily basis lead to a fruitful and steep learning curve in the SIMS communities. Meanwhile SIMS instruments offer a multitude of operational modes and are capable of analysing as 3D volume pixel by pixel. The lateral resolution has almost reached the physical limit of the collision cascade with values well below 100 nm and a depth resolution in the nm-range can be achieved. The development of cluster ion sources leads to the possibility to also probe organic materials in depth. At the same time data evaluation routines have become more powerful to address the large amount of data (several GByte for a 3D data set).

Generally, users would like to address three main questions: what (identification) is sitting where (localization) and how much is it (quantification)? Whereas the localization problem, as mentioned above, meanwhile is comparatively straightforward, identification and quantification still are challenging. In identification, the parallel presence of many elements and organic compounds in the SIMS spectra still asks for an expert to solve the analytical puzzle. Here, the application of multivariate statistical techniques and, more recently, machine learning approaching offer promising paths into the future. For a reliable quantification, the influence of the SIMS matrix effect on the results has to be taken into account. Here, the availability of suited reference samples plays a key role for closing the gap to quantitative techniques, like e.g. XPS (X-ray Photoelectron Spectroscopy).

The talk will therefore focus on identification and quantification issues including the use of multivariate statistics, MS/MS approaches and the use of reference sample comparing SIMS with the quantitative techniques XPS and LEIS (Low Energy Ion Scattering).

9:20am **AS+AC+BI+CA+HI-ThM-5 Time of Flight Secondary Ion Mass Spectrometry (ToF-SIMS) as a Novel Approach to the Characterization of Coatings and Interfaces of Porous Transport Layers***, **Genevieve Stelmacovich**, M. Walker, J. Foster,* Colorado School of Mines*; D. Cullen ,* Oak Ridge National Laboratory*; A. Paxson ,* Plug Power*; G. Bender, T. Schuler, S. Ware,* National Renewable Energy Laboratory*; S. Pylypenko,* Colorado School of Mines

The United States energy infrastructure aims to move towards the integration of hydrogen energy. As such, the production of reliable hydrogen generation through the optimization of water electrolyzers is imperative. In proton exchange membrane water electrolyzes (PEMWE's), the porous transport layer (PTL) plays an important role. Due to the harsh conditions of the cell, titanium is the current state-of-the-art anode PTL material. However, titanium quickly forms a layer of titanium oxide which significantly decreases conductivity of the PTL and respectively decreases the overall efficiency of the PEMWE system. To mitigate oxide effects, coatings are commonly applied to the PTL.

Focused Ion Beam Scanning Electron Microscopy (FIB-SEM) in conjunction with Scanning Transmission Electron MicroscopyEnergy-dispersive X-ray Spectroscopy (STEM-EDS) analysis is a commonly used technique to characterize PTL materials and PTL coatings, specifically to look at coating homogeneity and unfavorable oxide layer formation. Unfortunately, this approach is both time-consuming and labor intensive. Additionally, STEM-EDS analysis only provides elemental information, so if several oxide layers preside, it can be difficult to differentiate them, and thus lead to a lack of understanding fundamental degradation mechanisms. These technique deficiencies have motivated the development of an alternative approach that allow more efficient characterization of these materials. Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS) is a valuable technique that is commonly used to characterize thin films and buried interfaces. ToF-SIMS can be performed relatively quickly, provides chemical information, and is sensitive to trace elements. This technique also enables visualization of elemental distribution, which provides more detailed surface and interface information. This presentation will demonstrate capabilities of ToF-SIMS for characterization of PTLs comparing results to TEM analysis of cross-sections obtained with FIB-SEM. This study will highlight similarities and differences between the techniques, expand on technique optimization for these morphologically challenging samples, and suggest paths for future investigation moving forward.

9:40am **AS+AC+BI+CA+HI-ThM-6 Construction of Accurate 3D NanoSIMS Depth Profiling Images of Cells in the Presence of Lateral Variations in Sputter Rate***, M. Brunet, B. Gorman, **Mary Kraft**,* University of Illinois Urbana-Champaign

We have developed a strategy for constructing accurate 3D NanoSIMSdepth profiling images of cells in the presence of lateral variations in sputter rate. In this strategy, we use the secondary electrons acquired in parallel with the negative ions during depth profiling to reconstruct the 3D morphology of the cell every time a depth profiling image is acquired. The morphologies created for each raster plane in the depth profile are adjusted to ensure that the height at every pixel decreases with increasing image plane. The resulting reconstructions of the cell's morphology are used to define the z-positions of the voxels in the component-specific 3D NanoSIMS images. We validated this strategy by comparing morphology reconstructions for secondary electron depth profiling images acquired with focused ion beam - secondary electron microscopyand AFMdata acquired from the cell before depth profiling. The shape, curvature, and relative height of the reconstructed morphology agreed well with the AFM data. Depth correction of 3D NanoSIMSdepth profiling data of a metabolically labeled mammalian cell using this strategy improved visualization of the^{18}O-cholesterol and ^{15}N-sphingolipids distributions intransport vesicles and organelle membranes. Accurate 3D NanoSIMS images that show the intracellular distributions of molecules of interest may now be constructed in the presence of variations in sputter rate and the absence of correlated topography data.

11:00am **AS+AC+BI+CA+HI-ThM-10 Innovations in Nuclear Materials Analysis with SIMS***, **Christopher Szakal**,* National Institute of Standards and Technology (NIST) **INVITED**

Global nuclear safeguards efforts, coordinated by the International Atomic Energy Agency (IAEA), require precision measurements to answer questions related to nuclear treaty compliance. Despite those questions seeming relatively simple, and the elemental composition of the target analytes often consisting of just uranium and oxygen, the complex processes employed to answer those questions reveal the importance of subtle nuances to meet analytical objectives. This presentation will explore the complexity of this type of analyte system and how innovations in secondary ion mass spectrometry (SIMS) resulted in a routinely utilized tool to address nuclear safeguards requirements for environmental sampling analysis. The complexity is largely driven by the small amounts of nuclear particle material available for analysis, including challenges for 1) representative sampling of analyte material to represent actual nuclear processes, 2) accuracy and precision of uranium isotopic analyses across a wide dynamic range, and 3) determining when a nuclear process created the analyte material. Precision SIMS measurements provide the opportunity to address these analytical complexities to answer key nuclear safeguards questions, but the presentation will also highlight when limitations are inevitable for which questions can be answered.

11:40am **AS+AC+BI+CA+HI-ThM-12 Understanding Surface Bonding and Molecular Structure with MS/MS Imaging: From Click-Chemistry to Biogenesis***, **Gregory L. Fisher**,* Physical Electronics

A TOF-TOF imaging mass spectrometer allows TOF-SIMS (MS^1) imaging and tandem MS (MS^2) imaging to be achieved in a lossless fashion [1,2]. Secondary ions for MS^1 and MS^2 analysis are produced from the same surface area by a primary ion nanoprobe. Monolayer film samples may be characterized without undesired erosion or degradation; even sub-monolayer 2D films are readily characterized. Kilo-electron volt collision-induced dissociation (keV-CID) enables compositional identification and structural elucidation of precursor ion moieties. This analytical capability has been brought to bear for straightforward molecular identification as well as multifaceted studies involving surface modification, polymers, composites, catalysis, forensic and failure analysis, biology and pharmaceuticals. TOF-SIMS tandem MS imaging was employed to unravel the click-chemistry of sub-monolayer films [3] and shed new light to unlock the mystery of molecular biogenesis [4,5].

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[3] S. Oh, et al, *Chem. Mater*. **32** (2020) 8512-8521.

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12:00pm **AS+AC+BI+CA+HI-ThM-13 Probing Grain Boundary Segregation in 304L Stainless Steel using Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS)***, **James Ohlhausen**, E. Barrick, D. Susan, C. Robino, K. Hattar, J. Herrmann, P. Duran, J. Rodelas,* Sandia National Laboratories

Boron-rich phases in austenitic stainless steels can promote liquation cracking in the heat affected zone during welding. Stainless steels with boron concentrations even as low as 20 wt. ppm are susceptible to grain boundary cracking after heat treatments. The kinetics of phase transformations during heat treatment that generate the crack susceptible microstructure are currently unknown. Time-of-flight Secondary Ion Mass Spectrometry (ToF-SIMS) was used to map Boron at grain boundaries and in the bulk at trace levels for a series of 304L stainless steel coupons that were heat treated across a range of temperatures and cooling rates to investigate these phase transformations.ToF-SIMS and Scanning electron microscopy (SEM) were used to observe the distribution of chromium borides before and after heat treatment. These results will be used to enable quantitative prediction of thermal processing conditions to avoid weld cracking.Sample preparation methods, ToF-SIMS acquisition conditions and data analysis will be discussed.

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