

Actinides and Rare Earths Focus Topic Room 202C - Session AC+MI+SA-WeM

Magnetism, Complexity, and Superconductivity in the Actinides and Rare Earths

Moderators: Melissa Denecke, University of Manchester, UK, David Geeson, AWE, James Tobin, UW Oshkosh

8:00am AC+MI+SA-WeM-1 Strong electron-electron Interactions in the Actinides: Using Organometallics to Probe Delocalization Effects, *Corwin Booth*, Lawrence Berkeley National Laboratory **INVITED**

Systems exhibiting strong electron-electron interactions remain at the forefront of inquiry into complex properties of condensed matter systems due to their exciting properties (eg. superconductivity) and their resistance to being understood on a fundamental level. A bottleneck toward a better understanding has been the difficulty of the required many-body calculations for extended solids. Alternatively, calculations on small molecules require fewer and better approximations, potentially offering a better description. Although strong electron-electron interactions are well established in extended solids, recent work on lanthanide organometallic coordination compounds has demonstrated the importance of such interactions, fueled by the propensity for certain 4f orbitals to be partially delocalized. Meanwhile, recent work on the actinides challenges the canonical view that the 5f electrons can bond in the light actinides but are essentially localized in the heavier actinides. A major stumbling block for such work is the paucity of known structures for elements beyond Am in the periodic table.

For the discussion presented here, work on Ce and Yb organometallics will provide context in terms of f-occupancy and in bonding characteristics and the effect on magnetism. The role of strongly electron interactions will be described in terms of configuration interaction (CI) and related calculations.

Occupancy is measured using Ln L_{II}-edge x-ray absorption near-edge structure (XANES) techniques, and local structure (EXAFS) measurements demonstrate the final effect on the bonding at the metal center. Of particular interest is what happens in formally Ce(IV) systems that exhibit strong interactions.

XANES measurements of actinides are more difficult to interpret and will be discussed. The focus will be, however, on EXAFS measurements across the An series in the presence of strongly oxidizing ligands. Chosen ligands include hydroxypyridonone (HOPO), with less oxidizing ligands, such as diethylenetriaminepentaacetic acid (DTPA) used for comparison. Cations include Th, U, Pu, Am, Cm, Bk, and Cf. Discussions will center on nearest-neighbor bond lengths, using DFT calculations as a guide. The surprising role of covalency in the late actinides will be considered, both in terms of the EXAFS results and in terms of the edge shifts.

This work was supported by the U.S. Department of Energy (DOE), Office of Science (OS), Office of Basic Energy Sciences (OBES), under Contract No. DE-AC02-05CH1123.

8:40am AC+MI+SA-WeM-3 Structure and Magnetism of U-based Thin Films and Heterostructures, *Evgeniya Tereshina-Chitrova*, Institute of Physics, Academy of Sciences of the Czech Republic, Czech Republic; *L Havela*, Charles University, Prague, Czech Republic; *T Gouder*, Z Bao, Institute for Transuranium Elements, Germany; *M Dopita*, Charles University, Prague, Czech Republic; *R Caciuffo*, Institute for Transuranium Elements, Germany **INVITED**

Uranium is the basic component of most nuclear fuels. The production of uranium-based films has advantage over bulk materials studies as it allows performing advanced physics and chemistry experiments on small amounts of radioactive material and on its clean and smooth surfaces. Other interesting field is uranium magnetism. Although uranium itself is non-magnetic, uranium compounds display a rich variety of magnetic phenomena intimately related to the variable character of the 5f electron states [1]. Additional degrees of freedom can be used in thin films, in which the reduced dimensionality and structure modifications far exceed the limits imposed by thermodynamics, obeyed in bulk systems. We review the achievements in the field of sputter-deposited films, in which variations of deposition conditions can dramatically suppress crystallinity of the deposited material. The 5f itinerant magnetic systems (as US or UN [2]) react to the low substrate temperatures and high deposition rates by decreasing ordering temperatures and eventually by the loss of U magnetic moments. The strong ferromagnetism of uranium hydride is, on the other hand, almost insensitive, which underlines its local-moment character.

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The possibility to combine films of various materials on the nanostructure scale can also give rise to new functionalities. For example, the exchange bias (EB) effect [3], arising as a result of combination of a ferromagnet biased by exchange interaction at the interface to an antiferromagnet, is particularly interesting if uranium magnetism are involved. The new ingredient, strong spin-orbit interaction, can lead to very strong magnetic anisotropy, which represents an essential parameter. We have been systematically studying films of Fe₃O₄ (ferromagnet) grown using different substrates on the top UO₂, playing the role of biasing antiferromagnet [4]. The resulting high bias field (> 0.2 T) and a proximity effect, in which the high Curie temperature of Fe₃O₄ provides the EB functionality even at temperatures exceeding ordering of UO₂, demonstrate the promising aspects of using actinides in this non-traditional way.

The work is supported by the Czech Science Foundation under the project #18-02344S. Part of the work was supported by "Nano-materials Centre for Advanced Applications," Project No.CZ.02.1.01/0.0/0.0/15_003/0000485, financed by ERDF.

[1] V. Sechovsky, L. Havela, in: Magnetic Materials, K.H.J. Buschow (Ed.), Elsevier, Amsterdam, 1998, Vol. 11, p. 1.

[2] L. Havela et al., JALCOM 408-412, p. 1320 (2006).

[3] W. H. Meiklejohn and C. P. Bean, Phys. Rev. B 102, 1413 (1956).

[4] E.A. Tereshina et al., Appl. Phys. Lett. 105(12),122405 (2014).

9:20am AC+MI+SA-WeM-5 Field Induced Lifshitz Transitions in URu₂Si₂, *E Calegari*, Univ Federale Santa Maria, Brazil; *S Magalhaes*, Universidade Federale Rio Grande do Sul, Brazil; *Peter Riseborough*, Temple University **INVITED**

We report calculations on an unusual phase of the Under-screened Anderson Lattice (UAL) model, the so called spin-dependent inter-orbital density wave that has been proposed as describing the "Hidden Order" (HO) phase of URu₂Si₂.

We determine the effects of an applied magnetic field. Since the order parameter describes an ordering in the x-y plane, the electronic properties of the system are anisotropic below the critical temperature THO. We show that the magnetic susceptibility becomes anisotropic below THO.

Furthermore, for fields applied along a spontaneously chosen hard axis, THO decreases towards zero and that the HO transition changes from second order to first order at a large value of the magnetic field. Also, we find that the system undergoes a cascade of field-induced Lifshitz transitions and also show how these properties originate from the dependence of the quasi-particle bands on the orientation of the applied field. The good qualitative agreement with experimental findings provides strong support for the proposed description of the HO phase as a spin-dependent inter-orbital density wave phase.

11:00am AC+MI+SA-WeM-10 New Form of Uranium Hydride - UH₂, *Ladislav Havela*, *M Paukov*, *M Dopita*, *L Horak*, *P Minarik*, *M Divis*, *I Turek*, Charles University, Prague, Czech Republic; *D Legut*, VSB-Technical University of Ostrava, Czech Republic; *T Gouder*, *A Seibert*, *F Huber*, European Commission - Joint Research Centre; *E Tereshina-Chitrova*, Institute of Physics, Academy of Sciences of the Czech Republic, Czech Republic

Most of f-elements form with hydrogen both di- and trihydrides. Actinide and rare-earth dihydrides occur, as a rule, in the CaF₂ structure type. Uranium represents an exception, only UH₃ is present in the binary phase diagram. It exists in two different structure types. The metastable form α-UH₃ forms in the Cr₃Si structure type, which is in fact bcc U lattice filled with hydrogen. The stable form β-UH₃ has a larger cubic cell with two different U sites. Both forms are ferromagnets with the total U moment of ≈ 1 μ_B/U and the Curie temperature T_c ≈ 165 K. We have recently synthesized UH₃ thin films using a reactive sputter deposition. XRD analysis indicated the β-UH₃ structure, modified by a pronounced (001) texture and compressive residual strains imposed by the deposition dynamics. Magnetization measurements proved T_c = 165 K.

The sputter deposition on a cooled substrate (T = 170 K) using Si wafer the crystal structure turned different. The deposited material is undoubtedly cubic, of the fcc type, and the lattice parameter a = 5.3598 ± 0.0014 Å is very close to that of PuH₂ (a = 5.359 Å) and NpH_{2+x} (a = 5.343-5.355 Å). Hence we can assume that UH₂ in the fluorite structure has been formed. The key role in stabilization plays likely the effect of substrate (Si has a = 5.431 Å) in combination with low temperature deposition. The UH₂ film was subsequently subjected to magnetization measurements, which indicated a ferromagnetic ground state with T_c ≈ 125 K. This is lower than in the UH₃ phases, although the U-U spacing in UH₂ should be higher, 3.78

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Å, than in both UH₃ phases (3.31 and 3.60 Å for β- and α-UH₃, respectively). This fact points to the U-U interaction being more important than the U-U spacing. The ferromagnetic state is also the ground state obtained from ab-initio calculations. Scalar relativistic calculations (LDA) for experimental lattice parameter yield the spin moment $\mu_s = 2.0 \mu_B/U$. LDA+U (U = 2.25 eV) gives the equilibrium lattice parameter $a = 539.9 \text{ \AA}$, i.e. 0.7% larger than the experimental one, the ferromagnetic ground state with (111) easy-magnetization direction and the magnetic anisotropy energy $E_a = 9 \text{ meV}$. The total moment $0.45 \mu_B/U$ consists of $2.59 \mu_s$ and $-3.04 \mu_L$.

This work was supported by the Czech Science Foundation under the grant No. 18-02344S. The work at JRC Karlsruhe was supported by the European FP7 TALISMAN project, under contract with the European Commission. Part of the work was supported by the project "Nanomaterials centre for advanced applications", Project No. CZ.02.1.01/0.0/0.0/15_003/0000485, financed by ERDF.

11:20am AC+MI+SA-WeM-11 Tuning of Electronic Properties of U- and RE-Metallic Systems by H Absorption, *Silvie Maskova*, Charles University, Prague, Czech Republic; *K Miliyanchuk*, Ivan Franko National University of Lviv, Lviv, Ukraine; *A Kolomiets*, Lviv Polytechnic National University, Lviv, Ukraine; *L Havela*, Charles University, Prague, Czech Republic

The sensitivity of the interactions in the intermetallic systems to modification of the crystal structure makes the experimental techniques involving alteration of the atomic arrangement especially important. Various studies under compression are well-known examples of such methods. From this point of view hydrogenation can be treated as a complementary technique that provides „negative“ pressure. Hydrides can be defined as compounds for which the hydrogen absorption leads to the modifications of the crystal structure, such as pure lattice expansion or the formation of a new structure. As a result, the new compounds (hydrides) exhibit qualitatively new physical properties and such modifications provide us with additional information on the peculiarities of interatomic interactions in the initial compounds.

As an example, we will compare the impact of H absorption on U- and RE-compounds using A₂T₂X (A = Rare-Earth (RE) or actinide, T = transition metal, X = p-metal) compounds crystallizing in the tetragonal Mo₂FeB₂ structure type (space group *P4/mbm*). U₂T₂X interact with H₂ only at high pressure ($\approx 100 \text{ bar}$) reaching 2 H/f.u. The H absorption produces a lattice expansion (lower than 10 %), while the tetragonal structure is preserved. Higher H concentrations, which can be achieved in some RE₂T₂X compounds (up to 8 H atoms/f.u.), lead to amorphization or structure symmetry changes (with volume expansion exceeding 20 %), imposed by a minimum H-H distance requirement.

Magnetic properties of U-compounds strongly depend on the U-U distances. Hydrogen intrusion modifies the lattice by expanding it without changing the crystal-structure type leading to a 5f band narrowing. As a consequence, doping of U intermetallics by interstitial hydrogen leads to stronger magnetic properties. On the other hand, the hydrogen absorption has opposite effect on magnetic properties of RE₂T₂X compounds. For RE compounds, hydrogenation affects mainly the inter-site exchange interaction, which is weakened presumably by reducing the concentration of conduction electrons, responsible for the RKKY interaction.

11:40am AC+MI+SA-WeM-12 Magnetic Structures of U_nRhIn_{3n+2} Materials, *Attila Bartha*, *M Klicpera*, Charles University, Prague, Czech Republic; *P Cermak*, Forschungszentrum Juelich GmbH, Germany; *B Ouladdiaf*, Institute Laue-Langevin, France; *J Custers*, Charles University, Prague, Czech Republic
In the past decade, U-compounds crystallizing in the HoCoGa₅-type structure (*P4/mmm*), frequently referred to as 115, have been in the focus of attention in experimental and theoretical research. Vigorous activities have been motivated by the high superconducting transition temperature of $T_c = 8.7 \text{ K}$ in PuRhGa₅ [1] and $T_c = 18.5 \text{ K}$ in PuCoGa₅ [2]. No further superconductivity has been reported in neither U-115 nor in the closely related U₂TGa₈ compounds (T = transition metal). However, interesting magnetic properties have been observed: neutron scattering experiments revealed that UNiGa₅ exhibits the G-type antiferromagnetic (AFM) phase, while UPdGa₅ and UPtGa₅ exhibit the A-type AFM state. Note that G-type indicates a 3D Néel state, while A-type refers to a layered AF structure where spins align FM in the *ab* plane and AFM along the *c* axis [3]. The difference in the two magnetic structures is significant since it implies a sign change of the nearest-neighbor (NN) interaction.

Here we report on the magnetic structures of URhIn₅ and U₂RhIn₈, two new members of the U_nT_mX_{3n+2m} (X=In,Ga) family of compounds [4]. URhIn₅ displays AFM order below $T_N = 98 \text{ K}$. The observed increase of the resistivity for current parallel [100], [110] and [001] are reminiscent to a spin-density

wave (SDW) type of transition with the gap opening first along the [001] direction [5]. U₂RhIn₈ enters the AFM state at $T_N = 117 \text{ K}$. No increase in resistivity in the vicinity of T_N is found which would hint to a SDW gap opening. Neutron diffraction experiments on URhIn₅ were performed at the Heinz Maier-Leibnitz Zentrum (MLZ) in Garching using the triple axis spectrometer PANDA. Single crystals with accumulated mass of 10 mg where glued on an Al-plate. Our results confirmed the magnetic propagation vector $k=(1/2,1/2,1/2)$ predicted by NMR experiments [6] and a magnetic moment of $1.65 \mu_B/U^{3+}$. The neutron study on U₂RhIn₈ has been conducted at ILL, Grenoble using D10 on only one single crystal with $m \approx 1 \text{ mg}$. Analysis revealed a propagation vector $k=(1/2,1/2,0)$ and an ordered moment of $1.7 \mu_B/U^{3+}$. UIn₃, URhIn₅ and U₂RhIn₈ all show G-type AFM phase. While the *c*-axis parameter differs significantly the *a* lattice parameter equals 4.601 \AA , 4.621 \AA and 4.6056 \AA respectively, being a change of less than 1% pointing to the fact that the NN coupling is important for the type of magnetic structure.

[1] F. Wastin *et al.*, J.Phys.Condens.Matter **15**, S2279 (2003)

[2] J.L. Sarrao *et al.*, Nature (London) **420**, 297 (2002)

[3] T. Hotta, Phys.Rev. B **70**, 054405 (2004)

[4] A. Bartha *et al.*, J.Magn.Magn.Mater. **381**, 310 (2015)

[5] A. Bartha *et al.*, Acta Phys.Pol. A **127**, 339 (2015)

[6] H. Sakai *et al.* Phys.Rev. B **88**, 045123 (2013)

12:00pm AC+MI+SA-WeM-13 Insights into the Magnetic Dead Layer in La_{0.7}Sr_{0.3}MnO₃ Thin Films from Temperature, Magnetic Field and Thickness Dependence of their Magnetization, *Navid Mottaghi*, *M Seehra*, *R Trappen*, *S Kumari*, *C Huang*, *S Yousefi*, *G Cabrera*, *A Romero*, *M Holcomb*, West Virginia University

Detailed dc magnetization (*M*) measurements of a 7.6 nm La_{0.7}Sr_{0.3}MnO₃ thin film samples is investigated. The sample was fabricated by pulsed laser deposition. Zero-field-cooled (ZFC) *M* vs. applied field (*H*) cooled down to $T = 5 \text{ K}$ reveal the presence of negative remanent magnetization (NRM) as well as in ZFC *M* vs. temperature (*T*) measurements in $H = 50 \text{ Oe}$ and 100 Oe . ZFC and FC (field-cooled) protocols are used to determine the blocking temperature T_B in different *H*. Isothermal hysteresis loops at different *T* are used to determine the temperature dependence of saturation magnetization (M_s), remanence (M_R) and coercivity H_c . The M_s vs. *T* data are fit to the Bloch law, $M_s(T) = M_0(1 - BT^{3/2})$, showing a good fit for $T < 100 \text{ K}$ and yielding the nearest-neighbor exchange constant $J/k_B \approx 18 \text{ K}$. The variations of T_B vs. *H* and H_c vs. *T* are well described by the model often used for randomly oriented magnetic nanoparticles with magnetic domain diameter $\approx 9 \text{ nm}$ present in the dead-layer of thickness $d = 1.4 \text{ nm}$. Finally, the data available from literature on the thickness (*D*) variation of Curie temperature (T_c) and M_s of LSMO films grown under 200, 150, and 0.38 mTorr pressures of O₂ are analyzed in terms of the finite-size scaling, with M_s vs. *D* data fit to $M_s(D) = M_s(b)(1-d/D)$ yielding the dead layer thickness $d = 1.1 \text{ nm}$, 1.4 nm and 2.4 nm respectively.

Actinides and Rare Earths Focus Topic Room 202C - Session AC+AS+SA-WeA

Chemistry and Physics of the Actinides and Rare Earths

Moderators: Krzysztof Gofryk, Idaho National Laboratory, Ladislav Havela, Charles University, Czech Republic, David Shuh, Lawrence Berkeley National Laboratory

2:20pm **AC+AS+SA-WeA-1 New Directions in f-Block Separations Chemistry based on Metal and Ligand Redox Activity**, *Eric Schelter*, *A McSkimming*, University of Pennsylvania; *J Su*, Los Alamos National Laboratory; *T Cheisson*, *H Fang*, University of Pennsylvania; *L Moreau*, Lawrence Berkeley National Laboratory, Berkeley; *B Cole*, *B Manor*, *M Gau*, *P Carroll*, University of Pennsylvania; *E Batista*, *P Yang*, Los Alamos National Laboratory; *C Booth*, Lawrence Berkeley National Laboratory; *Y Qiao*, *J Bogart*, University of Pennsylvania

INVITED

Chemical separations of mixtures of cations in the context of nuclear waste processing for lanthanide/actinide mixtures and ore processing of rare earth elements remain some of the most challenging to achieve selectivity.

We have been developing new or improved methods to address these challenges using redox chemistry. For example, by changing ligand characteristics it may be possible to attain unusual oxidation states, in the case of actinides, that would enable selective separations. This talk will include discussion of some of our efforts to manifest chemical distinctiveness in actinide redox chemistry and the associated electronic structures of such complexes. In a parallel effort, we have been developing separations chemistry for rare earth cations based on kinetic resolution.

Kinetic resolution has only rarely been applied in metal separations chemistry and represents an interesting new direction. Using a redox active ligand, a predictive system for inter-rare earth partitioning and separations will be described.

3:00pm **AC+AS+SA-WeA-3 Bond Distance Variations for Lanthanide and Actinide Compounds and its Implication**, *Tsuyoshi Yaita*, Japan Atomic Energy Agency, Japan; *S Suzuki*, *T Kobayashi*, *H Shiwaku*, Materials Sciences Research Center, Japan Atomic Energy Agency, Japan

INVITED

Lanthanide and actinide are f-electron filling series and the properties of the series on their chemical behavior are very similar if valence states are same, while an electronic configuration of the inner shell for each element is slightly different compared to the similarity in size, and the value of special elements such as Nd and Dy used as neodymium magnet is high in the use of high-tech products. Regarding actinides series, radiotoxic Am is paid for attention in the geological disposal of radioactive waste and the R&D for partitioning and transmutation technique is performed. Accordingly, intra-series separation for lanthanide and actinide with similarity in chemical behavior would be quite important for the recovery of noble metal used as high-tech parts and treatment of high-level waste prior to geological disposal, resulting that the development of useful separation system could improve economic efficiency and reduction of environmental load.

On these backgrounds, we focus on the intra-series separation for lanthanide and actinide, especially, the relationship between variation of separation efficiency in the series and systematics of structure and electronic structure, and then, we try to propose new separation concept. Especially, in this talk, we talk about the variation of hydration or complex bond distances for trivalent actinide and lanthanide series based on X-ray crystallography and EXAFS and the interpretation based on the SX-XAS/XES and theoretical calculation.

4:20pm **AC+AS+SA-WeA-7 Spectroscopic Studies of Trivalent Actinide Coordination**, *Benjamin Stein*, *M Kerlin*, *A Morgenstern*, *E Batista*, *S Bone*, *S Cary*, Los Alamos National Laboratory; *J Lezama Pacheco*, SLAC National Accelerator Laboratory; *S Kozimor*, *P Yang*, Los Alamos National Laboratory

INVITED

Radioisotopes have a rich history in medicine, with their use dating back to the earliest studies of radioactivity. Only recently, however, have α -particle emitting radionuclides been considered for medical applications. Targeted alpha therapy utilizes the unique properties of α -emitting radionuclides to selectively kill cancer cells, with the short range of α -particles causing minimal collateral damage to nearby healthy cells. Actinium-225 (^{225}Ac) has been identified by the Department of Energy Isotope Program Long Range plan as an isotope of high national interest for targeted alpha therapy, due to its favorable half-life (10 days) and 4 α -emissions in the decay chain. However, if the ^{225}Ac is not securely bound to the targeting vector this

effectiveness results in very high toxicity to off-target (i.e. healthy) cells. Due to the high radioactivity and limited supply of all actinium isotopes, very little fundamental chemistry is known about this elusive element. Utilizing the unique radiological facilities at Los Alamos, we have been able to use microscopic amounts (~ 30 micrograms) of the longer-lived isotope actinium-227 (half-life of 22 years) for chemical studies. Utilizing this isotope, we have developed handling and containment techniques to perform "classic" spectroscopic and chemical studies in support of developing actinium chelates to advance the use of ^{225}Ac in targeted alpha therapy. During these studies of actinium coordination chemistry we have also made comparisons with the more "traditional" trivalent actinides, in particular americium and curium. We will discuss our latest EXAFS, NMR, and computational results on these difficult to handle elements by presenting a comparison of acetate and phosphonate binding, and how this informs chelator development.

5:00pm **AC+AS+SA-WeA-9 Speciation of Rare Earth Elements in Coal Harvesting Byproducts**, *Xu Feng*, *M Council-Troche*, *J Morris*, *A Noble*, *R Yoon*, Virginia Polytechnic Institute and State University

Rare earth elements (REEs) are critical for the development of renewable energy resources, national security, and advanced manufacturing. With the recent closure of the rare earth mine in California, the U.S. relies entirely on foreign imports mainly from China, which poses serious economical and national security concerns. According to a study commissioned by the National Energy Technology Laboratory (NETL), the U.S. coal and coal byproducts contain ~ 11 million metric tons of recoverable REEs, only a small fraction of which could satisfy the domestic need [1].

Recent USGS studies showed that the REEs in U.S. coals are preferentially partitioned to clay minerals [1], suggesting that the clay byproducts may be a major source of the critical materials. Ion-adsorbed REEs in clay appear to exist as two distinct forms: (1) ionic species adsorbed by coulombic attraction which is thought to be the primary form in REE-adsorbed clay deposits in South China, and (2) colloidal REEs formed by hydrolysis, each requiring a unique extraction strategy. However, the conditions under which the ion-exchange clays were formed in the U.S. coals and accompanying mineral matter may be different from those for the South China ion-adsorption clays, and the specific speciation of REEs in U.S. coal materials is currently unknown. It is, therefore, critical to study the fundamental mechanisms by which REEs are adsorbed on clay minerals in aqueous media to develop effective targeted extraction strategies.

In this work, X-ray photoelectron spectroscopy (XPS) was used to characterize the chemical state of representative light and heavy rare earth elements on artificial REE-adsorbed clay samples. Characteristic REE $3d_{5/2}$ features of the artificial clay sample, including the peak position of the two multiplet-split components, the magnitude of the multiplet splitting and the intensity ratio of each multiplet-split component, were compared to those of the high-purity REE standards such as $\text{REE}(\text{OH})_3$, REE_2O_3 and REECl_3 to provide insight into the identification of REE speciation on the artificial sample. XPS results suggest that $\text{REE}(\text{OH})_3$ is the primary REE species on the artificial REE-adsorbed clays. Furthermore, X-ray Adsorption Spectroscopy (XAS) was used to probe the speciation of representative light and heavy REEs in natural coal and coal byproduct samples by comparing the oxidation states and specific bonding environments to those of REE standard materials.

[1] Bryan, R. C., D. Richers, H.T. Andersen, and T. Gray, "Assessment of Rare Earth Elemental Contents in Select United States Coal Basins," Document No: 114-910178-100-REP-R001-00, January 2015.

5:20pm **AC+AS+SA-WeA-10 Exotic Electronic Properties of Strongly Correlated Compounds NpPd_3 and PuPd_3** , *Krzysztof Gofryk*, Idaho National Laboratory; *J Griveau*, *E Colineau*, Institute for Transuranium Elements; *K McEwen*, University College London; *W Nellis*, Harvard University; *J Smith*, Los Alamos National Laboratory

Actinides are characterized by the coexistence of localized and itinerant (delocalized) $5f$ -states near the Fermi energy. This dual nature of the $5f$ -electrons leads to many complex phenomena that are observed in these strongly correlated materials, spanning magnetic ordering, heavy-fermion ground state, unconventional superconductivity, and/or "non-Fermi liquid" state. The electronic properties of the strongly correlated electron systems are related to the formation, near the Fermi level, of a narrow band with large density of states and in spite of intensive theoretical and experimental efforts their nature is still not well understood. This behavior is well emphasized in $An\text{Pd}_3$ (An -U, Np, Pu) system. UPd_3 crystallizes in the hexagonal crystal structure and shows four phase transitions below 7.8 K, attributed to a succession of antiferroquadrupolar orderings of the

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uranium ions localized on the quasi-cubic sites of the *dhcp* structure. Depending on a heat treatment, NpPd_3 crystallizes in hexagonal and cubic crystal structures. The hexagonal NpPd_3 (*h*- NpPd_3) exhibits two transitions at 30 and 10 K. It has been suggested that the low temperature transition might be due to ordinary antiferromagnetic ordering while the high temperature one might be caused by a quadrupolar order. The cubic NpPd_3 (*c*- NpPd_3) orders antiferromagnetically below 52 K and the magnetic and transport measurements suggests that the transition is first order. PuPd_3 crystallizes in the cubic structure and shows an antiferromagnetic order below 24 K. To explore the influence of electronic correlations on the physical properties in the *ANpD₃* system, here we present our detailed magnetic, thermodynamic, and transport studies of NpPd_3 and PuPd_3 . We show that all results obtained present characteristic behaviors of 4f- and 5f-electron strongly correlated materials. The magnitude and overall temperature dependence of the electrical resistivity, magnetoresistivity, Hall and Seebeck effect, and heat capacity of NpPd_3 and PuPd_3 are archetypal of materials with Kondo interactions. Our measurements also reveal an unusual magnetic ordering in *c*- NpPd_3 . At T_N , the specific heat exhibits an extremely large peak [as large as 1000 J/(mol K)] and the magnetic susceptibility shows a clear jump. The transport properties of *c*- NpPd_3 indicate a dramatic reconstruction of the electronic structure at the Néel temperature, probably accompanied by a large change in the Fermi surface topology, which shows up as pronounced anomalies at this temperature in the electrical resistivity, the magnetoresistivity, and the Seebeck and the Hall coefficient. We will discuss implications of these results.

Biomaterial Interfaces Division Room 104B - Session BI+AC+AS+HC+NS+SS+TF-WeA

Current and Future Stars of the AVS Symposium II

Moderator: David Boris, U.S. Naval Research Laboratory

2:20pm **BI+AC+AS+HC+NS+SS+TF-WeA-1 Medard W. Welch Award Lecture: A Surface Scientist's Journey from Small Molecules to Biomolecules and Biomaterials, David G. Castner¹**, University of Washington

INVITED

Surface science plays an important role in a wide range of research and development areas such catalysis, biomaterials, microelectronics, clean energy and corrosion. The toolbox of surface scientist allows us to easily move across research topics and make significant impacts in both industrial and academic settings. The typical surface scientist is an expert in multiple techniques, surface modification, sample preparation/handling and instrumentation. We have all benefited from the significant and numerous advances that have occurred in the past 40 years in terms of improved instrumentation, introduction of new techniques and development of sophisticated data analysis methods, which has allowed us to perform detailed analysis of increasing complex samples. For example, comprehensive analysis of surfaces and surface immobilized molecules with modern surface science instrumentation provides an unprecedented level of detail about the immobilization process and the structure of the immobilized molecules. Results from x-ray photoelectron spectroscopy (XPS), time-of-flight secondary ion mass spectrometry (ToF-SIMS), near edge x-ray absorption fine structure (NEXAFS), surface plasmon resonance (SPR) and quartz-crystal microbalance with dissipation (QCM-D) biosensing, atomic force microscopy, and sum frequency generation (SFG) vibrational spectroscopy combined with computation methods such as molecular dynamic (MD) and Monte Carlo (MC) simulations provide important information about surface chemistry and structure. However, even with the advances that have been achieved with these powerful surface science techniques, there still remain many significant challenges for surface scientist. These include characterizing the surface chemistry and structure of nanoparticles, determining the atomic level structure of complex molecules bound to surfaces, 3D imaging of samples, and improved sample preparation methods that maintain materials in a relevant state when using ultra-high vacuum based analysis techniques.

This talk will discuss my adventures as a surface scientist starting from chemisorption and reactivity studies of small molecules on single crystal surfaces followed by industrial catalysis research and eventually moving to biomedical surface analysis with side ventures into areas such as clean energy. It has been an exciting journey and I will use it to provide some

examples of the multidisciplinary nature of surface science as well as discuss opportunities for addressing future challenges.

3:00pm **BI+AC+AS+HC+NS+SS+TF-WeA-3 Invited Talk-Future Stars of AVS Session: Making, Studying, and Designing Hierarchically Structured Soft Materials, Keith A. Brown²**, Boston University

Nature realizes extraordinary materials by structuring them precisely from the molecular scale to the macroscopic scale. While nature may have perfected this process over evolutionary time scales, synthetically recapitulating this level of control is tremendously difficult due to the large number of length scales involved and our limited knowledge of interactions between these scales. Faced with the daunting challenge of synthetically realizing soft hierarchical materials, we have adopted a three pronged strategy to: (1) make structures with control from the molecular scale to the macroscopic scale by directing bottom-up growth processes with top-down control, (2) learn how the properties of such materials emerge from their hierarchical structure, and (3) design the best performing structures using novel approaches borrowed from machine learning and autonomous research. In this talk, we will report recent progress in this complementary areas of making, studying, and designing hierarchical materials. In particular, we will focus on two major classes of materials, nanoparticle-based structures where the assembly and organization of particles leads to emergent mechanical properties at the bulk scale and polymer-based materials where we are connecting the synthesis, patterning, and properties of polymer structures across scales. In addition to lessons about the fundamental properties of hierarchically organized soft matter, we will highlight the synergies possible when combining, synthesis, detailed characterization, and advances in materials design.

3:20pm **BI+AC+AS+HC+NS+SS+TF-WeA-4 Invited Talk-Future Stars of AVS Session: Vapor Phase Infiltration for Transforming Polymers into Hybrid Materials: Processing Kinetics and Applications, Mark Losego³**, Georgia Institute of Technology

Vapor phase infiltration (VPI) is an emerging processing technology for infusing polymers with inorganic constituents to create new organic-inorganic hybrid materials with novel chemical, electrical, optical, and/or physical properties. These new hybrid materials have demonstrated applications including chemical separations, photovoltaics, and microelectronics patterning. This talk will focus on our development of a fundamental VPI processing kinetics phenomenology to create a pathway for rational design of material composition and structure. By measuring VPI compositional profiles as a function of space or time and temperature, we can extract fundamental energy barriers for the sorption, diffusion, and reaction processes and delineate amongst different rate limiting steps. In our materials development, we often find that partial infiltration of a polymer film, fiber, or foam is sufficient to impart desired properties; so rational design of the infiltration kinetics can enable desired performance without waste in processing time or materials. Here, we will demonstrate several examples including our work to create chemically insoluble polymers and membranes. We find, for example, that infiltration depths of about 0.75 microns are sufficient to yield PMMA chemically insoluble in organic solvents regardless of whether it is in a thin film geometry or a macroscopic plexiglass object of centimeters in dimension. In PIM-1 membranes used for chemical separations, we find that we can achieve > 30 wt% inorganic loading with a single infiltration exposure. After infiltration, these membranes become stable in new separations solvents that previously swelled and/or dissolved the polymer.

(Submitted for the Future Stars of the AVS Symposium.)

4:20pm **BI+AC+AS+HC+NS+SS+TF-WeA-7 Invited Talk-Future Stars of AVS Session: Surface Preparation Methods for the Selective Oxidation of Ethanol to Acetaldehyde over $\text{TiO}_2/\text{Au}(111)$, Ashleigh Baber⁴, D Boyle, J Wilke, V Lam, D Schlosser**, James Madison University

Obtaining a molecular-level understanding of the reaction of alcohols with heterogeneous model catalysts is critical for improving industrial catalytic processes, such as the production of hydrogen from alcohols. The use of reducible oxides provides a source of oxygen on $\text{Au}(111)$ for the reaction of ethanol, which is easily regenerated in the presence of an oxygen background. The redox chemistry of small alcohols, including methanol and propanol, has been studied on $\text{Au}(111)$ supported TiO_2 nanoparticles, yet the active site for the chemistry has not yet been elucidated. Depending on the surface preparation conditions, $\text{Au}(111)$ supported TiO_2 nanoparticles

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³ Future Stars of the AVS

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¹ Medard W. Welch Award Winner

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react with small alcohols to form either reduced and oxidized products. The desire to selectivity form oxidized or reduced products merits an investigation of alcohol reactivity over differently prepared TiO₂/Au(111) surfaces. In this work, a systematic study of ethanol reactivity over several TiO₂/Au(111) surfaces elucidates the effect of surface conditions on the selectivity of the reaction between ethanol and TiO₂/Au(111). The reactivity of the surface for ethanol oxidation was altered by controlling the oxidation state of TiO_x (x<2). Atomic force microscopy (AFM) provides information regarding the structure of the Au(111) supported TiO₂ nanoparticles and ultrahigh vacuum temperature programmed desorption (TPD) monitors the selectivity of the reaction between ethanol and TiO₂/Au(111). The presence of TiO₂ nanoparticles on Au(111), ~25 nm in diameter, led to the catalytic conversion of ethanol to acetaldehyde at temperatures greater than 400 K. Low coverages of fully oxidized TiO₂ nanoparticles on Au(111) are active for the selective oxidation of ethanol to form acetaldehyde.

4:40pm BI+AC+AS+HC+NS+SS+TF-WeA-8 Invited Talk-Future Stars of AVS Session: Single Atom Catalysis: An Atomic-Scale View, Gareth Parkinson¹, TU Wien, Austria

Single-atom catalysis is a rapidly emerging area of research that aims to maximize the efficient usage of precious metals through "single atom" active sites. Although catalytic activity has been demonstrated for several single-atom catalyst systems, an inability to accurately characterize the catalyst based on single atom active sites ensures that the field remains controversial, and little is really known about how a single atom adsorbed on a metal oxide support catalyzes a chemical reaction. In this lecture, I will describe how we are addressing the crucial issues of stability and reaction mechanism using a surface science approach. The work is based on the magnetite (001) surface, which exhibits an unusual reconstruction based on subsurface cation vacancies.

The surface stabilizes ordered arrays of metal adatoms (of almost any variety) with a nearest neighbor distance of 0.84 nm to unprecedented temperatures as high as 700 K. Crucially, because the geometry of the adatoms is uniform and precisely known, reactivity experiments are performed on a well-defined model system, and theoretical calculations can be performed to shed light on the mechanisms at work. Several examples of our recent work will be used to illustrate the trends discovered to date, including how strong CO adsorption destabilizes Pd and Pt adatoms leading to rapid sintering, and how extraction of lattice oxygen from the metal-oxide is central to catalytic activity in the CO oxidation reaction.

5:00pm BI+AC+AS+HC+NS+SS+TF-WeA-9 Invited Talk-Future Stars of AVS Session: Multimodal Chemical and Functional Imaging of Nanoscale Transformations Away from Equilibrium, Olga Ovchinnikova², Oak Ridge National Laboratory

The key to advancing energy materials is to understand and control the structure and chemistry at interfaces. While much of the dynamic chemistry can be studied on macro-scale systems, there is a lack of means to localize chemical measurements and correlate them to nanoscale structure of the material. Through a unique merger of advanced scanning probe microscopy and mass spectrometry techniques rooted in innovative data processing and control algorithms, we are now able to understand the interplay between chemical and physical functionality at the fundamental length using multimodal chemical imaging. This multimodal imaging transcends existing techniques by providing nanoscale structural imaging with simultaneous quantitative nanomechanical properties and quantitative chemical analysis. In this talk I will discuss how we have developed and used this capability to visualize dynamic material transformations at interfaces, to correlate these changes with chemical composition, and to distil key performance-centric material parameters. One exciting capability is that the AFM can be used to drive materials away from equilibrium at the nanoscale with highly localized electric fields. This allows field confinement effects on localized chemistry in materials to be locally probed, especially at interfaces. This in turn yields direct information on key energy related questions such as electron and ion motion distribution and transport at and between interfaces. We have applied this approach to the study of systems and processes that underlie energy capture, conversion and storage, including photovoltaics and oxide ferroelectrics, which have historically eluded comprehensive understanding of the mechanisms behind the spatially heterogeneous interfacial

chemistry and its link to material performance. Overall, I will focus on ways to unlock the mystery of active interface formation through intertwining data analytics, nanoscale elemental and molecular characterization, with imaging; to better grasp the physical properties of materials and the mechanistic physics-chemistry interplay behind their properties.

5:20pm BI+AC+AS+HC+NS+SS+TF-WeA-10 Invited Talk-Future Stars of AVS Session: Expanding the Structural Toolkit to Characterize Heavy Actinide Complexes, Rebecca Abergel³, Lawrence Berkeley Lab, University of California, Berkeley; G Deblonde, A Mueller, P Ercius, Lawrence Berkeley National Laboratory; A Minor, Lawrence Berkeley Lab, University of California, Berkeley; C Booth, W de Jong, Lawrence Berkeley National Laboratory; R Strong, Fred Hutchinson Cancer Research Center

Structural characterization of actinide elements from actinium to einsteinium can be a challenging task due to the high radioactivity and limited availability of some of the isotopes of interest. However, significant work is needed to address a certain lack of understanding of the fundamental bonding interactions between those metal centers and selective ligands. Such understanding presents a rich set of scientific challenges and is critical to a number of applied problems including the development of new separation strategies for the nuclear fuel cycle, the need for decontamination after a nuclear accident or the use of radioisotopes for new cancer treatments. Our studies utilize luminescence sensitization, UV-Visible, X-ray absorption, and X-ray diffraction spectroscopic techniques as well as transmission electron microscopy and electron energy loss spectroscopy to investigate specific heavy actinide coordination features. Using simple inorganic complexes but also strong hard oxygen-donor ligands as well as more elaborate higher molecular weight protein assemblies allows the differentiation of heavy actinide species even when limited to minute amounts of materials. Innovative structural characterization approaches based on X-ray absorption, X-ray diffraction and electron microscopy that were applied to series of isostructural systems and used to derive coordination trends in the later 5f-element sequence will be discussed.

5:40pm BI+AC+AS+HC+NS+SS+TF-WeA-11 Invited Talk-Future Stars of AVS Session: Trends in Adsorbate Interactions with Bimetal Surfaces, Liney Arnadottir⁴, L Sprowl, Oregon State University

Surface chemistry plays an important role in a large range of applications and technologies, such as catalysis and electrocatalysis, device fabrication through film growth, and degradations through oxide formation, carbonization, and corrosion. Bimetal surfaces are of increasing interest for single atom catalysis and corrosion resistance of alloys. Previous studies have shown correlations between adsorbate interactions and d-band shifts for different metals as well as for alloying effects of metal sandwich structures, but for mixed metal surfaces the nature of the adsorption site can change (ensemble effect) as well as the number of electrons in the surface layer which effects the d-band center. Here we explore correlations of adsorbate interactions with mixed metal surfaces through density functional theory calculations of adsorbate interactions with different facets, (100) and (111), of pure Ni and eleven Ni-based surface alloys as well as Ni in different host metal. We find that the addition of surface alloying atom has little effect on the binding of CO and C but C binding varies for different facets. On the other hand, O binding is highly dependent on the alloying element due to ensemble effect. This has an interesting effect on the predicted reaction energies of CO dissociation which is endothermic on the (111) facet and exothermic on the (100) facet governed by C interactions but the changes in the reaction energy within each facet are mostly governed by the ensemble effect on O adsorption. The relationship between the formation energy of the mixed metal surface and adsorbate interactions will also be discussed.

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² Future Stars of the AVS

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³ Future Stars of the AVS

⁴ Future Stars of the AVS

Actinides and Rare Earths Focus Topic Room 202C - Session AC+AS+SA-ThM

Nuclear Power, Forensics, and Other Applications

Moderator: Ladislav Havela, Charles University, Czech Republic

8:00am **AC+AS+SA-ThM-1 Electron Microscopy in Nuclear Forensics, Edgar Buck, D Reilly, J Schwantes, J Soltis, T Meadows, D Meier, J Corbey,** Pacific Northwest National Laboratory **INVITED**

Recent advances in electron microscopy both in terms of spatial resolution, sensitivity, and the ability to perform in-situ chemistry experiments, present enormous opportunities to the nuclear forensic field. Extraction of specific particles of interest with dual-beam SEM-FIB instruments can be readily applied plutonium-contaminated materials. Furthermore, other developments in microscopy have enabled in-situ monitoring of interfacial processes and 3D tomographic views of specimens and phases. Several new advances in technology have enabled great advances and potential for nuclear science including nuclear forensics. These include dual-beam ion-electron systems for precise sample isolation and preparation, the electromagnetic lens aberration-corrector, high-count capacity x-ray detector systems, faster digital cameras, high performance electron backscattered diffraction systems in combination with 3D visualization tools, application of cryoTEM methods to material science, as well as micro-fabricated in-situ cells that enable direct observation of chemical and electrochemical processes in the EM. The application of these technologies to nuclear forensics will be discussed.

8:40am **AC+AS+SA-ThM-3 New Frontiers with Fission Track Analysis and TOF-SIMS Techniques, Itzhak Halevy,** Nrcn Israel, Israel; *R Radus,* Ben Gurion University, Israel; *S Maskova,* Charles University, Prague, Czech Republic; *A Kogan, S Samuha, D Gridchin, E Grinberg, E Boblil, N Haikin,* IAEK-NRCN, Israel; *I Orion,* Ben-Gurion University -Negev, Israel; *A Weiss,* Faculty of Engineering, Bar-Ilan University, Israel **INVITED**

Illicit trafficking of radioactive materials is known to exist from the early days of radioactive era. The nuclear forensics deals with recognizing the materials and processes of the radioactive industry. The properties of the materials can give a hint about the source of material and its original use.

The most common radioactive material involved in illicit trafficking is uranium. Uranium is a common natural element which can be found everywhere. The cosmogenic uranium is well known and defined. The natural enrichment of uranium is varying within a small range around 0.72 % and is indicative to its source. The enrichment of the anthropogenic uranium can vary much depending on the purpose and use of the material.

Different enrichments are known for individual nuclear power plants, research reactors and military uses.

Measuring the uranium properties can indicate its enrichment, presence of other elements or impurities and can help in finding its attribution, namely its origin.

To learn more about the history of found material accurate isotopic measurements are needed.

The ratio between ^{230}Th and ^{234}U can give a good estimation of how much time passed from the last chemical cleaning of the material. This technique is called radio-chronometry or age dating.

We developed new Lexan detectors with much better signal to noise ratio to improve sensitivity and reduce the false alarm.

New Automated software can recognize the fission track (FT) automatically and give the parameters of the track, like: roundness, intensity, number of tracks and color histogram. In that program we can add ROI (region of interest) or to cancel a false positive FT identification.

Analytical equipment like TOF-SIMS (Time Of Flight - Secondary Ion Mass Spectrometry) and ICP-MS (Inductively Coupled Plasma - Mass Spectrometer located at the Geophysical survey Israel - GSI) together with new FTA software give new frontiers to the nuclear forensic research .

A microscope equipped with TOF-SIMS gives the ability to measure quantitatively, the ratio between the different isotopes and molecules. Using the scanning electron microscope, we are able to choose the desired particle which can be then analyzed using the TOF-SIMS. TOF-SIMS is a technique capable to distinguish not only between elements but also different isotopes. Knowing the ratio between the isotopes is very important as it can help us to classify the material looking for its possible origin. Morphology and depth profile than give other insight to the

processes that the sample went through. Nano manipulators can extract the particle and transfer it to a different technique.

9:20am **AC+AS+SA-ThM-5 Predictive Nuclear Forensics: Fundamental Frameworks to Fill Missing Pieces, Jenifer Shafer, M Koehl, A Baldwin, D Wu,** Colorado School of Mines; *R Rundberg,* Los Alamos National Laboratory; *M Servis,* Washington State University; *T Kawano,* Los Alamos National Laboratory **INVITED**

Understanding the origin of nuclear forensic signatures provides the benefit of understanding how these signatures can be compromised and provides a framework to predict signatures that might arise under various conditions. The ability to predict signatures is particularly useful for the nuclear forensics community since only a limited number of samples exist. Frequently access to these samples can be further constrained due to classification boundaries. This talk provides two examples of how fundamental chemical and physical phenomena can be leveraged to understand signature origins, thus enabling a more robust nuclear forensics capability. The first study focuses on understanding how organic phase aggregation chemistry in the PUREX process can dictate trace metal, such as fission or corrosion product, partitioning patterns. By understanding how trace metals partition, information regarding the processing history, including the reprocessing site, could be ascertained. Trace metal partitioning patterns were studied by producing radiotracers in the USGS 1 MW TRIGA reactor. The influence of extractant aggregation on trace metal partitioning was then assessed using a combination of diffusion NMR spectroscopy and small angle neutron scattering. The second study focuses on understanding how cumulative fission product yields can describe the incident neutron energy. Fission yield curves of uranium-235 have a decrease in valley radionuclide production when the incident neutron energy is in the epithermal energy regime. This decrease in valley radionuclide production seems tied to the excitation of the uranium-236 to the 3- spin state. The octupole deformation of the 3- spin state enables more asymmetric fission than typically encountered with fast or thermal neutrons and thus suggests the structure of the excited uranium-236 compound nucleus could be, in part, responsible for cumulative fission product yields. These two studies highlight how fundamental science enables signature development.

11:00am **AC+AS+SA-ThM-10 Soft X-ray Synchrotron Radiation Spectromicroscopy Studies of Radioactive Materials, David Shuh,** Lawrence Berkeley National Laboratory; *A Altman,* Lawrence Berkeley National Laboratory and UC Berkeley; *D Kilcoyne, S Minasian, J Pacold, D Smiles, T Tylliszczak, D Vine,* Lawrence Berkeley National Laboratory; *L He, J Harp, M Meyer,* Idaho National Laboratory; *C Degueldre,* University of Lancaster, Switzerland

Soft X-ray synchrotron radiation methodologies are being developed and employed at the Advanced Light Source (ALS) of Lawrence Berkeley National Laboratory (LBNL) to elucidate the electronic structure of radioactive and actinide materials. Results from these investigations have begun to provide improved fundamental knowledge that can be used as a scientific basis for the enhanced design of actinide materials, complexes, ligands, and the overall understanding of actinide materials. The experimental developments at the ALS have centered on studies of radioactive materials with the soft X-ray scanning transmission X-ray microscope (STXM) at Beamline 11.0.2 for spatially-resolved near-edge X-ray absorption spectroscopy (XAS). The spectromicroscopy capabilities of the STXM provide the means to determine the speciation and composition in a range of actinide materials, particularly those of technological and environmental interest with spatial resolution that can reach to the true nanoscale. A particular emphasis has been on the use of light atom (B, C, N, O, F, Na, Mg, Al, Si) ligand K-edge XAS technique to determine the electronic structure characteristics in an array of unique and relevant materials. Furthermore, there are a host of additional electron energy level thresholds (such as the L-edges of the transition metals, the M-edges of the lanthanides, and others) that can be probed by near-edge XAS in the soft X-rays.

Recently, STXM spectromicroscopy studies have been extended to focused ion beam (FIB) prepared radioactive and irradiated material specimens in collaboration with Idaho National Laboratory. Future scientific developments and applications of soft X-ray spectromicroscopy investigations utilizing ptychography and in-operando methodologies will be discussed.

Thursday Morning, October 25, 2018

11:20am **AC+AS+SA-ThM-11 Comparison of the Oxidation Rates for Alpha Versus Delta Plutonium by X-ray Photoelectron Spectroscopy**, *Art Nelson, S Donald, D Roberts, W McLean*, Lawrence Livermore National Laboratory

X-ray photoemission spectroscopy (XPS) was used to characterize differences in the oxidation rates for polycrystalline α -Pu versus δ -Pu related to variations in grain size and surface electronic structure. The evolution of the Pu 4f core-level chemical shift as a function of oxygen exposure at ambient temperature was quantified in oxidation profiles. In addition, the X-ray excited Pu NOO Auger line-shapes were combined with the chemical shift of the Pu 4f_{7/2} photoelectron line that defines the Auger parameter and results in a reliable method for determining oxidation states independent of binding energy calibration. The oxidation profiles reveal that α -Pu oxidizes faster than δ -Pu, both resulting in the PuO₂/Pu₂O₃/Pu oxide structure. This data was used to produce chemical state (Wagner) plot for select plutonium oxides.

This work was performed under the auspices of the U.S. Dept. of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

11:40am **AC+AS+SA-ThM-12 A Single-Stage AMS Detector for Secondary Ion Mass Spectrometry and its Applications to Nuclear Materials Analyses**, *David Willingham, E Groopman, K Grabowski*, U.S. Naval Research Laboratory; *L Sangely*, International Atomic Energy Agency; *A Meshik, O Pravdivtseva*, Washington University in St. Louis; *D Weisz, K Knight*, Lawrence Livermore National Laboratory

Secondary ion mass spectrometry (SIMS) has long been applied to the analysis of isotopic heterogeneities in nuclear materials. Few other methodologies can compete with the ability of SIMS to measure the isotopic composition of nuclear materials with high accuracy and precision with micrometer spatial resolution. Like many other mass spectrometry techniques, however, the presence of molecular isobaric interferences at any given mass-to-charge complicate interpretation of SIMS measurements. Uranium isotopes, for example, can be valuable and informative markers for the process of enriching uranium for nuclear fuel for the production of nuclear energy. In addition to the major uranium isotopes (²³⁸U and ²³⁵U), the minor isotope ²³⁶U is of interest because it is an indication for the presence in the sample of uranium reprocessed from spent nuclear fuel. The resolving power needed to separate ²³⁶U from the molecule ²³⁵U¹H, however, is greater than 38,000 - far beyond most the mass resolution of commercially available SIMS instruments. One solution to this problem is to use a Single-Stage Accelerator Mass Spectrometer (SSAMS) to accelerate secondary ions to a high enough energy (300keV) to enable molecular dissociation within a stripper gas while retaining good transmission, followed by SIMS-based detection. At the U.S. Naval Research Laboratory, we have developed the Naval Ultra-Trace Isotope Laboratory's Universal Spectrometer (NAUTILUS) to achieve this goal. Using the NAUTILUS, we accomplish molecule-free isotopic analysis of nuclear materials without sacrificing the benefits of SIMS. In this work, we demonstrate the broad applications of the NAUTILUS to areas of nuclear materials analyses including uranium bearing particle analysis for nuclear Safeguards, analysis of the Oklo natural nuclear reactor, and analysis of uranium doped silicate glasses as working reference analogs for nuclear fallout materials. The NAUTILUS represents a new era in SIMS analyses of complex materials with specific application to nuclear materials and general application to the SIMS community as a whole.

12:00pm **AC+AS+SA-ThM-13 Physicochemical Properties of Ag in Annealed ZrN/SiC/Ag Heterostructures Used to Simulate TRISO Nuclear Fuels**, *Jeff Terry, M Warren, R Seibert*, Illinois Institute of Technology

Silicon carbide (SiC) is used as a supportive and protective barrier in the cladding of tristructural-isotropic (TRISO) nuclear fuel particles. Previous studies both of surrogate surfaces and irradiated fuel have shown that the fission product silver (Ag) exhibits transport into and sometimes through the SiC barrier with temperatures above 500 °C. This silver release can cause safety concerns for maintenance workers due to plate-out on in-reactor components. Although an exact diffusion mechanism for Ag in SiC is unknown, a solution is needed to prevent this effect. The use of a ZrN protective coating may mitigate Ag transport and potential release. This study examines the transport of Ag in SiC through the use of surrogate multilayered thin-films. Thin films of subsequent layers of SiC, Ag, SiC, and ZrN deposited by pulsed-laser ablation deposition (PLD) under a range of annealing temperatures up to 1200 °C are studied. After heating, X-ray photoemission spectroscopy (XPS), scanning electron microscopy (SEM),

and energy-dispersive X-ray spectroscopy (EDS) was used to examine the surface of the ZrN. Initial results show that Ag does not readily diffuse through the ZrN layer like it does through SiC. The results and implications of this study will be discussed.

Actinides and Rare Earths Focus Topic

Room 202C - Session AC-ThA

Early Career Scientists

Moderators: Tomasz Durakiewicz, National Science Foundation, David Shuh, Lawrence Berkeley National Laboratory

2:20pm AC-ThA-1 Complexation, Characterization and Separation of the Lanthanides and Actinides: Shedding Light to Subtle Differences within the f-element Series, *Gauthier Deblonde, C Booth*, Lawrence Berkeley National Laboratory; *M Kelley, J Su, E Batista, P Yang*, Los Alamos National Laboratory; *A Müller, P Ercius, A Minor, R Abergel*, Lawrence Berkeley National Laboratory

INVITED

From the nuclear fuel cycles to the therapeutic use of radioisotopes for cancer diagnostics and treatment, the solution chemistry of lanthanides and actinides has become increasingly relevant to a number of applied problems. Understanding the fundamental bonding interactions of selective metal assemblies and the intrinsic differences between f-elements presents a rich set of scientific challenges and is critical to the development of highly efficient separation reagents and new actinide- or lanthanide-based therapeutics.

Our approach to these challenges uses a combination of techniques (EXAFS, liquid-liquid extraction, protein crystallization, UV-vis, fluorescence, DFT...) to characterize f-block aqueous complexes with highly selective and bio-inspired chelators or with more classical aminocarboxylate ligands. With the goal of always minimizing the amount of radioactive material needed and the worker's radiation exposure, we are also investigating the use of TEM spectroscopy to characterize inorganic salts of heavy actinides (BkCl₃, CfCl₃...) while using only a few nanograms of actinides.

Our journey into the chemistry of the f-elements, and especially that of Am, Cm, Bk, and Cf, led us to capture subtle difference within the trivalent actinide series. A broad study on the aminocarboxylate complexes of Am³⁺, Cm³⁺, Bk³⁺, and Cf³⁺ by EXAFS spectroscopy revealed an unexpected change in speciation between the Cf chelates and its Am, Cm, and Bk analogues. Similarly, the study of the chelation of the lanthanide and actinide cations by some bio-inspired chelators (ex: siderophore derivatives), led us to develop versatile and highly efficient liquid-liquid extraction processes for the purification radioisotopes.

3:00pm AC-ThA-3 Improving the Understanding of Actinides Through Spectroscopy, *Samantha Cary, J Su*, Los Alamos National Laboratory; *S Galley, T Albrecht-Schmitt*, Florida State University; *E Batista, M Ferrier, S Kozimor, V Mocko, B Scott, B Stein*, Los Alamos National Laboratory; *F White*, Florida State University; *P Yang*, Los Alamos National Laboratory

INVITED

Understanding the fundamental aspects of bonding is important in predicting an element's behavior, unfortunately when it comes to the actinides, there is little known. Acquiring a better understanding of these elements will affect a number of different areas including nuclear forensics, national security, and nuclear fuel cycles. Here we will describe the preparation of M(S₂CNET₂)₃(X) (M^{III} = Nd, Sm, Eu, Gd, Am, Cm, and Cf; X = n-heterocyclic aromatic ligands) and compare their structure, X-ray absorption spectroscopy, and electronic structure calculations.

4:00pm AC-ThA-6 Structural Chemistry of M(IV) (M = Ce, Th, and U) Complexes Isolated from Aqueous Solution, *Karah Knope*, Georgetown University

INVITED

Understanding actinide metal ion speciation and reactivity is of great strategic and scientific importance, and relates to a number of areas ranging from waste management to separations chemistries. Speciation depends on oxidation state, and our knowledge of the structural and energetic properties of tetravalent actinide complexes is relatively limited in comparison to the higher valent oxidation states. This lack of structural and chemical information has resulted in large discrepancies in thermodynamic data, significant challenges in process chemistry, and unanticipated behaviors in environmental systems. As a means of filling this knowledge gap, we have been examining the solid-state structural chemistry of tetravalent Ce, Th, and U complexes obtained from aqueous solutions with an eye towards understanding the directing effects of inner and outer coordination sphere interactions. Moreover, using spectroscopic and X-ray scattering techniques, we have been probing the correlation between the precipitated phases and the solution phase species. Presented

here will be an overview of recent efforts to elucidate the effects of both inner- and outer- coordination sphere interactions on the structural chemistry of tetravalent metal ion (Ce, Th, and U) complexes. How the synthetic conditions, identity of the counter-ions, and nature of complexing ligands affect the speciation and reactivity of the Ce(IV), Th(IV)- and U(IV)-building units will be discussed.

4:40pm AC-ThA-8 Hundess, Coherence and Magnetism in URu₂Si₂- and USb₂-family Materials, *L. Andrew Wray, L Miao, H He*, New York University; *S Ran*, University of Maryland, College Park; *N Butch*, NIST / Umd; *J Denlinger, Y Chuang*, Advanced Light Source, Lawrence Berkeley National Laboratory

INVITED

The uranium compounds URu₂Si₂ and USb₂ present fascinating low temperature phase diagrams, and are focal points of long-standing debates regarding how the crossover between strong correlations and electronic itinerancy should be conceptualized and evaluated. It has recently been found that uranium O-edge resonant X-ray spectroscopies can help to image this multi-natured wavefunction by providing a fingerprint of the f-electron atomic multiplet states. I will present a systematic O-edge spectroscopic characterization of URu₂Si₂ and USb₂ as a function of doping, and show that these data align well with a "Hund's metal" picture for both compounds. Distinct differences in the degree of "Hundness" (same-atom alignment of electron magnetic moments) as a function of chemical composition are found to underlie important features of the low temperature phase diagrams, such as the transition from a "hidden order" phase to antiferromagnetism, and the loss of a low temperature coherence feature in transport measurements. Based on these results, I will propose that developing a more quantitative experimental characterization of Hundness in many-body wavefunctions is of fundamental importance to the broader goal of understanding the phase diagrams in metallic systems with non-trivial local moment physics.

Thursday Evening Poster Sessions, October 25, 2018

Actinides and Rare Earths Focus Topic

Room Hall B - Session AC-ThP

Actinides and Rare Earths Poster Session

Moderators: David Shuh, Lawrence Berkeley National Laboratory, James Tobin, UW Oshkosh

AC-ThP-1 Upconversion Photoluminescence Efficiency Dependence of Yb ions in Gd_{0.91-x}NbO₄: Yb₃₊, Er_{0.09}, S Yi, Seung Gon Lee, Silla University, Republic of Korea

Gd_(0.91-x)NbO₄: Er³⁺_{0.09}, Yb³⁺_x (x= 0.03, 0.06, 0.09, 0.12 and 0.15) phosphors were synthesized by the facile solid state reaction method. Yb³⁺ concentrations were changed from 0.03 to 0.15 mol for the fixed Er³⁺ concentration at 0.09 mol. The particle size of phosphors was around 180 ~ 350 nm and shape were angular oval observed by scanning electron microscopy. The crystalline structures of the phosphors were investigated by X-ray diffraction. The photoluminescence emission based on the green emissions near 528 and 551 nm and red emissions near 657 and 675 nm were observed and the highest emission intensity occurred for the sample Yb_{0.15}Er_{0.09}. Also, under the 980 nm excitation, Er³⁺, Yb³⁺ co-doped GdNbO₄ phosphors appeared the up-conversion emission based on the green emission near 535 nm and 556 nm radiated by ²H_{11/2} → ⁴I_{15/2} and ⁴S_{3/2} → ⁴I_{15/2} transitions and red emission about 657 nm and 675 nm radiated by ⁴F_{9/2} → ⁴I_{15/2} transition, which assigned to the intra 4f transitions of Er³⁺ ions.

AC-ThP-2 Luminescence Characteristics of (Gd_{0.85-x}Yb_{0.15})NbO₄:Er³⁺ Phosphors, S Yi, DongGyu Lee, Silla University, Republic of Korea

Gd_(0.85-x)NbO₄: Yb³⁺_{0.15}, Er³⁺_x (x= 0.03, 0.06, 0.09, 0.12 and 0.15) phosphors were synthesized by the solid state reaction method. Er³⁺ concentrations were changed from 0.03 to 0.15 mol for the fixed Yb³⁺ concentration at 0.15 mol. The crystalline structures of the phosphors were investigated by X-ray diffraction. The particle size of phosphors was around 140 ~ 320 nm and shape were angular oval observed by scanning electron microscopy. The photoluminescence emission based on the blue emission near 471 nm, green emission near 596 nm and red emission near 621 nm were observed and the highest emission intensity occurred for the sample Yb_{0.15}Er_{0.09}. Also, under the 980 nm excitation, Er³⁺, Yb³⁺ co-doped GdNbO₄ phosphors appeared the up-conversion emission based on the green emission near 535 nm and 556 nm radiated by ²H_{11/2} → ⁴I_{15/2} and ⁴S_{3/2} → ⁴I_{15/2} transitions and red emission about 657 nm and 675 nm radiated by ⁴F_{9/2} → ⁴I_{15/2} transition, which assigned to the intra 4f transitions of Er³⁺ ions.

AC-ThP-3 Exploring the Electronic Structure of Molecular Lanthanide Complexes in the +2 Oxidation State Using Photoelectron Spectroscopy, Daniel Huh, J Bruce, J Hemminger, W Evans, University of California, Irvine

Recent advances in rare-earth metal reduction chemistry have led to the isolation of a new series of Ln(II) complexes. For Ln = Y, La, Ce, Nd, Gd, Tb, Dy, Ho, Er, and Lu, reduction of 4fⁿ (C₅H₄SiMe₃)₃Ln^{III} complexes generates [(C₅H₄SiMe₃)₃Ln^{II}]⁻ products that exhibit unusual 4fⁿ5d¹ mixed-principal quantum number electron configurations. X-ray photoelectron spectroscopy (XPS) has been employed to examine and compare these mixed-principal quantum number electronic structures with those of traditional Ln(II) complexes that have 4fⁿ⁺¹ configurations. In this work, X-ray and ultraviolet photoelectron spectroscopy have been used to examine the electronic structure of [K(2.2.2-cryptand)][(C₅H₄SiMe₃)₃Ln^{II}] (Ln = Eu, Gd, Tb) where Gd(II) and Tb(II) have previously been shown to have non-traditional 4f⁷5d¹ and 4f⁸5d¹ electronic configurations, respectively, and where Eu(II) has been shown to have a traditional 4f⁷ electronic configuration.

AC-ThP-4 Effects of Cerium Content on Local Structure in U_{1-x}Ce_xO₂ Solid Solution, H Cao, Shanghai Institute of Applied Physics, Chinese Academy of Science, China; Yuying Huang, Shanghai Institute of Applied Physics, Chinese Academy of Science, China

Uranium-cerium mixed oxides U_{1-x}Ce_xO₂ (x=0.05, 0.20, 0.35, 0.50, 0.65, 0.80) were prepared by co-precipitation method. X-ray diffraction data shows that the U_{1-x}Ce_xO₂ samples are solid solutions with lattice parameters following Vegard's law. The local structures of U_{1-x}Ce_xO₂ were studied by X-ray absorption fine structure spectroscopy at both U L₃ and Ce K edges. To avoid the interference from L₂ edge in Ce edge XAFS data, Ce K edge XAFS was used to obtain more accurate local structure of Ce. XAFS data shows that there is local structure disorder in U_{1-x}Ce_xO₂ solid solution. The effects of Ce content on the local structure disorder in U_{1-x}Ce_xO₂ solid

solution were studied. For a cerium content less or equal to about 35%, the disorder increases with the addition of cerium in the U_{1-x}Ce_xO₂ solid solution. When the cerium content increases to more than 50% the disorder of the sample decreases rapidly.

AC-ThP-5 Magnetism of the (Nd,R)₂Fe₁₄B - H system with R = Er and Tm, I Tereshina, Lev Ivanov, M.V. Lomonosov Moscow State University, Russian Federation; D Gorbunov, Helmholtz-Zentrum Dresden-Rossendorf, Germany; M Paukov, Charles University, Prague, Czech Republic; E Tereshina-Chitrova, Institute of Physics, Academy of Sciences of the Czech Republic, Czech Republic; M Doerr, Technische Universität Dresden, Germany; L Havela, Charles University, Prague, Czech Republic; A Andreev, Institute of Physics ASCR, Czech Republic

Since the discovery in 1984, the R₂Fe₁₄B (R is a rare earth) compounds received a considerable scientific attention due to their hard magnetic properties [1,2]. Fundamental characteristics of the best permanent magnet Nd₂Fe₁₄B are known to be highly sensitive to the atomic substitutions and absorbed light atoms such as hydrogen. In the present work, we studied a combined influence of substitutions of Er and Tm for Nd and hydrogen absorption on the behavior of magnetization in magnetic fields up to 60 T. All studies were conducted on free powder samples at 2 K.

It is found that the substitution in the rare earth sublattice decreases the saturation magnetization as a result of ferrimagnetic ordering of magnetic moments of heavy rare earths with respect to the moments of Nd and Fe. However, under sufficiently strong magnetic fields the magnetic moments rotate and in the ideal case, the field-induced ferromagnetic state is observed. This phenomenon is directly connected to the strength of the inter-sublattice exchange interactions (Fe and Nd, Er, Tm sublattices).

In the parent materials (Nd_{0.5}Er_{0.5})₂Fe₁₄B and (Nd_{0.5}Tm_{0.5})₂Fe₁₄B in fields up to 60 T no increase of the magnetization was observed. Hydriding of the compounds up to the maximum possible hydrogen content 5.5 at.H/f.u. allows us to observe a forced-ferromagnetic state in the (Nd_{0.5}Tm_{0.5})₂Fe₁₄BH_{5.5} compound. The transition from ferri- to the ferromagnetic state occurs gradually: it begins at the 35 T field and finishes at 55 T. For the compound (Nd_{0.5}Er_{0.5})₂Fe₁₄BH_{5.5} we also observe a magnetization increase. So that, hydrogenation is found to weaken the intersublattice exchange interaction in these three-sublattice materials.

This work is performed with financial support of the grant of Russian Scientific Foundation (project № 18-13-00135). We acknowledge the support of HLD at HZDR (member of the European Magnetic Field Laboratory) and the Materials Growth and Measurement Laboratory (<https://mgml.eu> [<https://mgml.eu/>]).

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Actinides and Rare Earths Focus Topic Room 202C - Session AC+MI+SA-FrM

Actinide and Rare Earth Theory and Related Measurements

Moderators: Paul S. Bagus, University of North Texas, David Shuh, Lawrence Berkeley National Laboratory

8:20am **AC+MI+SA-FrM-1 Periodic Boundary Condition and Embedded Cluster DFT Calculations of Water Adsorption on AnO_2 ($\text{An} = \text{U}, \text{Pu}$) Surfaces**, *Nikolas Kaltsoyannis*, University of Manchester, UK, United Kingdom of Great Britain and Northern Ireland **INVITED**

Over half of the World's stockpile of civil plutonium (c. 126 tonnes) is stored at Sellafield in the UK as PuO_2 powder in sealed steel cans. There is evidence of gas generation in some of these cans. Many routes to gas production have been suggested, several of which involve complex, interconnected and poorly understood $\text{PuO}_2/\text{H}_2\text{O}$ interactions.

We have an ongoing project to study computationally the interaction of AnO_2 ($\text{An} = \text{U}, \text{Pu}$) surfaces with water. Standard periodic boundary condition (PBC) implementations of DFT using generalized gradient approximation (GGA) functionals can fail to reproduce key features of actinide solids, e.g. predicting metallic properties in systems known to be insulating. This failure stems from incorrect description of the strongly correlated 5f electrons, which are overly delocalized by the GGA, and the standard solution to this problem is to correct the GGA functionals with an onsite Coulomb repulsion term known as the Hubbard U . An alternative solution is to employ hybrid DFT, in which some of the exact exchange energy of Hartree-Fock theory is incorporated into the Hamiltonian. Such functionals typically produce more localized 5f electrons, and recover insulator behavior. They are, however, extremely expensive to employ in PBC calculations, and hence are rarely used. We have therefore sought a model which allows the routine use of hybrid DFT in AnO_2 /water systems, and have adopted the periodic electrostatic embedded cluster method (PEECM), in which a quantum mechanically treated cluster is embedded in an infinite array of point charges. We treat a cluster of AnO_2 and adsorbing water molecules using hybrid DFT (PBE0) whilst the long-range electrostatic interactions with the bulk are modelled *via* embedding in point charges.

In this presentation, I shall describe the results of both PBC and PEECM studies of the interactions of water with both stoichiometric and reduced (oxygen vacancy) {111}, {110} and {100} surfaces of UO_2 and PuO_2 . The geometries and energetics of single and multiple layers of water will be presented, together with our calculations of water desorption temperatures, from which we propose an alternative interpretation of experimental data.

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9:00am **AC+MI+SA-FrM-3 Understanding the Role of Oxidation States on the Chemistry of Actinides through Integration of Theory and Experiment**, *Wibe de Jong, J Gibson*, Lawrence Berkeley National Laboratory; *R Abergel*, Lawrence Berkeley Lab, University of California, Berkeley **INVITED**
Fundamental insights into the role of 5f electrons in the redox chemistry of actinides is essential for predicting the fate and controlling the behaviour of nuclear materials in the ecosystem, as well as for developing new advanced applications in energy, medicine and forensics. In recent years computational chemistry has become a key pillar of multidisciplinary actinide research, advancing the fundamental understanding of actinide chemistry through strong synergies between experiment and simulations. We will present some recent multidisciplinary gas-phase thermochemistry and spectroscopy efforts focused on understanding the role of metal-ligand interactions on the oxidation states of actinides across the series.

9:40am **AC+MI+SA-FrM-5 An Experimentalist's Viewpoint: The Tremendous Strengths and Occasional Weaknesses of Actinide Cluster Calculations**, *James G. Tobin*, University of Wisconsin-Oshkosh

Over the course of the last several years, cluster calculations have been used in a myriad of ways to analyze spectroscopic results from actinide systems and gain insight into the electronic structure of these actinide systems. For example, the calculated 6d Unoccupied Density of States (UDOS) in Uranium Tetrafluoride and Uranium Diode were probed using U

L3 (2p) X-ray Absorption Near Edge Structure (XANES) as well as U N7 (4d) X-ray Absorption Spectroscopy (XAS). [1-3] Cluster calculations have also been used successfully to study the development from atomic to bulk electronic structure in Pu [4,5] and issues associated with the 2p Occupied Density of States (ODOS) in Uranium Tetrafluoride. [6, 7] However, there now appear to be some interesting discrepancies between what has been observed experimentally and the predictions of cluster theory. In particular, the F 1s XAS of Uranium Tetrafluoride [8] and the U 4d XAS branching ratio (BR) predictions for oxidized uranium. [1] These issues will be addressed in the talk.

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10:00am **AC+MI+SA-FrM-6 Ligand and Metal XAS Edges in Heavy Metal Compounds**, *Paul S. Bagus*, University of North Texas; *C Nelin*, Consultant

In previous work, [1] it has been shown that the magnitude and character of the covalent mixing in representative lanthanide and actinide oxides depends strongly on the nominal oxidation state of the oxide. In the present work, ab initio theoretical wavefunctions, WF, have been determined and have been used to obtain the energies and dipole intensities for different Near Edge X-Ray Absorption (XAS) Fine Structure, NEXAFS edges. The theoretical NEXAFS spectra have been examined in order to determine the extent to which the features of these edges can be used to infer the character and extent of the covalent bonding. An important and novel feature of the theory is that the angular momentum coupling of the open shell electrons is taken into account and the multiplets, both resolved and unresolved, associated with the various NEXAFS features are identified. Furthermore, different sets of variationally optimized orbitals are used for the initial, ground state, and the final, excited state, configurations. Thus, the relaxation and screening in response to the core-hole are taken into account. The systems examined are Ce(III) and Ce(IV) in CeO_2 and U(IV), U(V), and U(VI) in UO_2 . For these systems, cation p and d edges are used to distinguish excitations to the frontier nf and (n+1)d orbitals, especially since it has been shown that both of these cation orbitals have significant covalent mixing with the ligands. The O K-edge NEXAFS is presented and the relative excitation energies and intensities into the cation nf and (n+1)d orbitals are compared to the covalent character of the different systems. The WFs are solutions of the Dirac-Coulomb Hamiltonian. [2] Comparisons are made with experiment to demonstrate the accuracy of the theoretical treatments used.

Support for this work by the Geosciences Research Program, Office of Basic Energy Sciences, U.S. DOE, is acknowledged.

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10:20am **AC+MI+SA-FrM-7 Thermal Expansion and Conductivity of Th and Ac from First Principles Calculations**, *Dominik Legut, L Kyvala*, VSB-Technical University of Ostrava, Czech Republic; *U Wdowik*, Pedagogical University, Poland

We investigate electronic structure, lattice dynamics, and thermophysical properties of elemental thorium and made a prediction for actinium as

here the experimental data are scarce. The results were obtained within density function calculations. Recently, Actinium (Ac) shows excellent potential in radiotherapy and Thorium (Th) appears to be a suitable substitution for uranium in novel nuclear fuels. Therefore, we determined the effect of spin-orbit interaction (SOI) for the electronic, elastic, phonon and thermodynamic properties of number of Ac phases (hcp, bcc, fcc, and sc). Similarly, in the case of Th in addition to SOI we investigated also localized versus itinerant behavior of the f electrons. Therefore we applied so-called Hubbard parameter U in the range of 0.5-2 eV for the Th 5f electrons. Next, we calculated the elastic tensor by the linear-response method as implemented in VASP code [1] to determine elastic constants (Cij). Subsequently, the criteria of stability based on Cij were evaluated. Next, we calculated vibrational properties using the direct force-constant method as implemented in the PHONOPY code [2]. Actinium phase transition is discussed within the quasi-harmonic approximation under temperature and pressure to draw different conclusion than in Ref. [3]. We advance our knowledge to predict the thermal conductivity Using the Boltzman transport code [4] we address contribution to the thermal conductivity not only from phonons but also from free electrons. Our determined mechanical and thermodynamical properties of Th show very good agreement with experimental data and give us a high confidence to be able to predict similar properties of Ac and to explain the highest know thermal conductivity of Th among actinides.

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Acknowledgement

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10:40am **AC+MI+SA-FrM-8 XANES Investigation into the Electronic Structure of Ce Coordination Complexes, Liane Moreau, C Booth, Lawrence Berkeley National Laboratory; Y Qiao, E Schelter, University of Pennsylvania**

Cerium is of particular interest among the lanthanide elements for its stability in both the Ce(III) and Ce(IV) oxidation states. This provides unique redox properties, making Ce a promising candidate for advanced energy materials. To this end, chemistries to control the Ce oxidation state have been widely developed. More interestingly, Ce-ligand bonding has in some cases shown intermediate behavior between Ce(III) and Ce(IV), which is indicative of a multiconfigurational ground state. This phenomenon results in low-energy ligand-to-metal charge transfer in addition to its magnetic behavior, such as temperature independent paramagnetism. In order to understand the effects of Ce electronic structure on compound chemical and physical properties, it is imperative to develop methods to accurately probe the fractional occupancy between $4f^1$ and $4f^0$ configurations.

Ce L₃ edge X-ray absorption near edge structure (XANES) measurements provide detailed information concerning the density of states from varying Ce-ligand bonding configurations. In particular, XANES spectra of Ce(IV) imido, Ce(III/IV) guanidinate-amide and Ce(IV) anilide complexes are presented and related to their specific coordination chemistry. From XANES, lanthanide-ligand bonding covalency is explored. We also consider the effects of varying the alkali metal species in metal-capped Ce(IV) imido. Results from the Ce(IV) complexes exhibit characteristic double white line absorption, indicative of significant 4f orbital participation in metal-ligand bonding. The extent to which covalency is observed is highly dependent on the specific ligand chemistry. In combination with DFT and magnetism results, XANES is used to study the effects of such chemistries on the orbital configurations and relate them to the observed novel properties of Ce coordination complexes.

11:00am **AC+MI+SA-FrM-9 Structure and Properties of Reactively Deposited Uranium Hydride Coatings Studied by the X-ray Scattering Methods, Milan Dopita, L Havela, L Horák, E Chitrova, Charles University, Prague, Czech Republic; D Legut, VSB-Technical University of Ostrava, Czech Republic; M Cieslar, Charles University, Prague, Czech Republic; Z Matěj, MAX-IV, Lund, Sweden**

A series of Uranium hydride coatings was prepared using a reactive sputtering method. The samples were sputtered with different Mo doping levels as well as at different hydrogen pressure and various substrate temperature. Fused silica and Si single-crystals were used as substrates for deposited coatings. The structure and the real structure (i.e. the deviations of the structure from the ideal 3D crystal periodicity) of the samples were investigated using the x-ray scattering methods. The x-ray reflectivity yielded the information on the coatings thickness, roughness and electron density. The key results of our work were the determination of the phase composition and its evolution, the description of the coherently diffracting domains - crystallite size and defects distributions, the description of the residual stress, strains and preferred orientation of crystallites - textures and its depth profiles in the coatings.

The sample sputtered on the fused silica substrates crystallized in the β -UH₃ structure (stable uranium hydride phase) with the stress free lattice parameter $a = 0.6703 \pm 0.0004$ nm. The coating exhibited strong 001 preferred orientation of crystallites and compressive residual stress $\sigma = -5.1 \pm 0.1$ GPa. Additionally, the coating revealed a bimodal distribution of crystallite sizes with two components having the mean crystallite size $D_1 = 200$ nm and $D_2 = 3.6$ nm with volume fractions of 60:40, respectively, and huge microstrain in both components. Contrary, the coating sputtered on cooled Si single-crystal substrate showed totally different behavior. It did not form nor the stable β -UH₃ neither the meta-stable α -UH₃ structure. The measured diffraction pattern revealed the fluorite fcc structure, analogous to e.g. PuH₂. This is highly interesting result, since to our best knowledge such form of uranium hydride has not been so far reported in the literature. The polycrystalline coating exhibited tilted *hhh* texture, stress free lattice parameter $a = 0.53598 \pm 0.00014$ nm, compressive residual stress $\sigma = -1.54 \pm 0.09$ GPa and mean crystallite size of 82 nm. Besides, the coating contains pronounced microstrain caused by the crystal lattice defects and significant fraction of the stacking faults. The results of the x-ray scattering studies were correlated with data obtained from HRTEM. Additionally, the possibility of formation of uranium hydride with fcc fluorite structure was confirmed by the ab-initio theoretical calculations (LDA+U).

This work was supported by the project "Nanomaterials centre for advanced applications", Project No. CZ.02.1.01/0.0/0.0/15_003/0000485, financed by ERDF.

11:20am **AC+MI+SA-FrM-10 Ligand Induced Shape Transformation of Thorium Dioxide Nanocrystals, Gaoxue Wang, E Batista, P Yang, Los Alamos National Laboratory**

Nanocrystals (NCs) with size and shape dependent properties are a thriving research field. Remarkable progress has been made in the controlled synthesis of NCs of stable elements in the past two decades; however, the knowledge of the NCs of actinide compounds has been considerably limited due the difficulties in handling them both experimentally and theoretically. Actinide compounds, especially actinide oxides, play a critical role in many stages of the nuclear fuel cycle. Recently, a non-aqueous surfactant assisted approach has been developed for the synthesis of actinide oxide NCs with different morphologies, but an understanding of its control factors is still missing to date. Herein we present a comprehensive study on the low index surfaces of thorium dioxide (ThO₂) and their interactions with relevant surfactant ligands using density functional calculations. A systematic picture on the thermodynamic stability of ThO₂ NCs of different sizes and shapes is obtained employing empirical models based on the calculated surface energies. It is found that bare ThO₂ NCs prefer the octahedral shape terminated by (111) surfaces. Oleic acid displays selective adsorption on the (110) surface, leading to the shape transformation from octahedrons to nanorods. Other ligands such as acetylacetone, oleylamine, and trioctylphosphine oxide do not modify the equilibrium shape of ThO₂ NCs. This work provides atomic level insights into the anisotropic growth of ThO₂ NCs that was recently observed in experiments, and thus may contribute to the controlled synthesis of actinide oxide NCs with well-defined size and shape for future applications.

Friday Morning, October 26, 2018

11:40am AC+MI+SA-FrM-11 Perspectives on the Synthesis, Characterization and Applications of Upconversion and Downconversion Nanomaterials, *Martin Ntwaaborwa*, University of the Witwatersrand, South Africa

Luminescent nanomaterials incorporating one or more active ions (usually lanthanides) have many uses today in applications such as information displays, light emitting diodes, theft prevention, advertising, medical imaging, photodynamic therapy and photovoltaic cells. In the past few decades, there has been an upsurge of research interest in the synthesis of luminescence down-conversion and up-conversion nanomaterials with different particle morphologies, and their characterization using different optical techniques. The use of lanthanide ions to convert photons either by down-conversion or up-conversion process to different and more useful wavelengths is an interesting research phenomenon for a wide range of applications. For example, the use of down-conversion and up-conversion nanomaterials to improve the power conversion efficiency of crystalline silicon solar cells and for treatment of tumour cells, respectively, have been demonstrated. While efficient down-conversion has been demonstrated from a combination of many rare-earths ions, the most efficient up-conversion has been demonstrated from only a few combination of rare-earths ions such as erbium (Er^{3+}) – ytterbium (Yb^{3+}), thulium (Tm^{3+}) – Yb^{3+} , and holium (Ho^{3+}) – Yb^{3+} incorporated in different host lattices with low phonon energies. Different mechanisms of luminescence down-conversion and up-conversion will be discussed. In addition, a few selected methods used to synthesise luminescence down-conversion and up-conversion nanomaterials will be discussed.

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