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Probing Orbital Bonding and Surface Interactions of Hydrolyzed Actinide Oxides using Synchrotron X-ray, Vibrational Spectroscopy, and Electron Microscopy

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Dr. Vahid Majidi, Laboratory Director, Savannah River National Laboratory

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Introduction

Actinide dioxides (MO_2, M = Th, U, Np, Pu, Am, Cm) are the simplest actinide compounds and often used as long term storage materials.[1-4] They can provide fundamental insights into 5f/6d orbital bonding and electronic structure. Scientists have been trying to understand the role of 4f, 5f, and 6d electrons in covalent bonding for decades.[5-8] Most hydrolysis studies of actinide materials involve uranium and uranium oxides due to their relevance for nuclear fuel and waste.[9-12] Very few hydrolysis studies, if any, involve thorium, neptunium, and transplutonium dioxides or mixed actinides.[3,4,13-15] Actinide surface speciation can be investigated in great detail with spectroscopic techniques such as EXAFS, vibrational spectroscopy, or electron micrsocopy.[16] Actinide mineral-water interface reactions have been thoroughly summarized for non-oxide actinides;[16] however, the reaction mechanisms provide useful ideas into the incorporation of water into the actinide lattice as a result of hydrolysis. Hydrolysis will have an incremental effect on the crystalline lattice allowing for models to predict the next changes to the electronic states.

Long term understanding of stability of actinide dioxides in the environment is important for future storage. Second, this work needs the facilities to synthesize the actinide dioxides. SRNL has those facilities and the knowledge. Third, several analytical methods that can handle radioactive materials are needed to perform various analytical measurements to develop kinetic model of hydrolysis for actinide dioxides and mixed actinide dioxides. The proposed work is submitted for the Heavy Element Thrust Area in FY20 call for basic energy sciences.

Several questions will be answered by characterization of hydrolysis of actinide dioxides, leading to a better understanding of nuclear waste forms and alternative waste forms. What is the contribution of f-electrons vs. d-electrons to the bonding, and how does that change with hydrolysis? How can the calculation parameters (pseudopotentials etc.) be changed to match the experimental data? How quickly does the hydrolysis at different relative humidity change the particles? Do the particles undergo oxidation or reduction? **Different spectroscopic, microscopic techniques, and theory will be used together to form a**

complete picture of the hydrolysis effect on the electronic states and 5f/6d orbitals used in bonding, and the kinetic rate of hydrolysis.
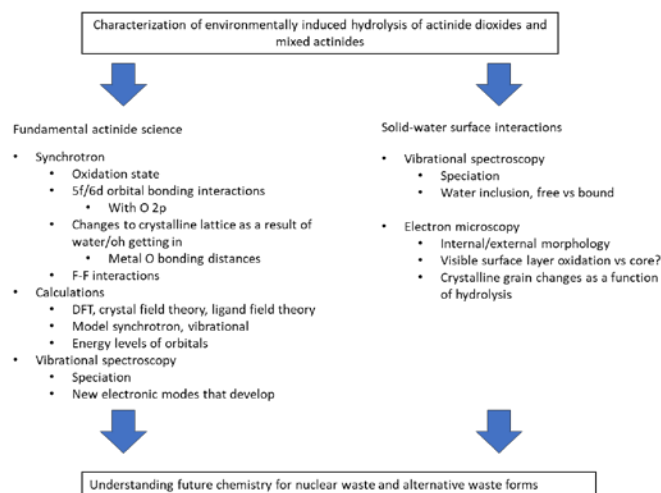

Figure 1 Schematic tying characterization of environmentally induced hydrolysis of actinide dioxides affects fundamental science and surface-water interactions to understand nuclear waste forms

Fundamental actinide science
Synchrotron X-ray techniques will be used to characterize the crystalline lattice, bonding structure, electronic transitions, and 5f/6d orbital bonding interactions. Synchrotron techniques can probe the atomistic positions and near neighbor interactions more effectively than any other experimental techniques. Combining synchrotron and computational calculations, scientists are better able to probe how the orbitals are interacting, and their individual contributions to bonding. In the case of actinide materials, do they possess more covalent character or more ionic character? What is the contribution of the 5f/6d orbitals as compared to 2p orbitals, and how does that affect the bonding character? Synchrotron radiation is needed to explore fundamental actinide science.

Fluorescence microprobe will be the first measurement of every sample. This will be useful in creating a map of the sample in containment to identify where particles are located. There are distinct locations for the L-edges of fluorescence above 12 keV for each actinide, and M-edges below 4 keV that may not be visible. XANES at the L_3 edge will provide insight into the oxidation state changes as the particles are hydrolyzed, while also probing the 6d states. As water is added to the particles, oxidation on the metal center could occur through redox reactions. For uranium, this would be higher oxides of $UO_2(OH)_2 \cdot xH_2O$ before U_3O_8. Haschke et al had observed PuO_{2+x} (x ≤ 0.27) phase containing Pu(VI), formed from adsorbed water at 25°C.[3] Korzhavyi et al using theory, suggests the formation of PuO_{2+x} is an endothermic process with water, and that it really forms from the products of the radiolysis of water.[17,18] This role is important for better understanding long term waste products.[17,18] With the long term presence of water vapor, it is expected that the actinides will stabilize at the most common oxidation state. EXAFS measurements will determine the bonding distances between actinide and oxygen atoms, as well as contraction of expansion of the lattice. EXAFS can also be used to determine speciation as the particle hydrolyze. The M-edge of the actinides is particularly useful for XANES and HERFD. The M-edge will directly probe the 5f orbitals

and determine their occupancy and energy levels and be more sensitive to oxidation state change. STXM at the oxygen K-edge will probe the 2p orbital bonding contribution in the actinide system. Calculations discussed below are of importance in the interpretation of synchrotron data and the determination of density of states of the valance orbitals, and their contribution to binding, whether covalent or ionic.

Computational DFT, crystal field theory, and ligand field theory calculations will need to be performed to understand the role the f-electrons play in bonding and the energies of the orbitals as a result of hydrolysis.[19]. DFT calculations will be used to fit the experimental data. The updated DFT model will then be used to calculate the orbital energies using crystal field theory and the 5f/6dorbital interactions with 2p oxygen using ligand field theory. Crystal field theory and ligand field theory will help to interpret the vibrational spectroscopy and predict new vibrational bands as they appear.

Raman spectroscopy is well adapted for the investigation of nuclear materials due to insitu analysis and a small sample size.[20,21] Raman spectroscopy is becoming an essential technique for the study of nuclear materials, owing to its multiple advantages with respect to other traditional techniques[22] ThO_2 Raman spectroscopy seems to be less studied with a publication in 2016 of varying compounds.[23] More recent interest has focus on Raman spectroscopy of UO_2-ThO_2 mixed oxides.[24] UO_2 and the higher oxides are the most often studied compounds of uranium, due to their relevance in the nuclear industry.[25-27] Allen et al was able to measure four uranium oxides by Raman spectroscopy.[25] As is the case with most uranium compounds, laser-induced sample heating can create problems in characterization. Most recent investigation explored temperature dependence on the UO_2 spectra.[22] Elorrieta et al found that by heat UO_2 up to 600°C, the T_{2g} and the 2LO bands have shifts to lower wavenumbers.[22]

Several authors have used Raman spectroscopy to investigate PuO_2.[28-31] Begun et al was the first to publish the Raman spectra and compared the T_{2g} band of different isostructural compounds such as UO_2, ThO_2, and AmO_2. SRNL has recently characterized PuO_2 electronic modes using multiple wavelength lasers.[31] Work from SRNL shows the importance of using multiple wavelengths on a sample (*Figure 2*). Varying the wavelength allows Raman spectroscopy to probe varying depths of PuO_2 particulates, with 325 nm being more surface analysis, and 785 nm being deep penetration.[32] Different wavelengths are also used to characterize emerging bands, and to reduce fluorescence from changes to the crystalline lattice.
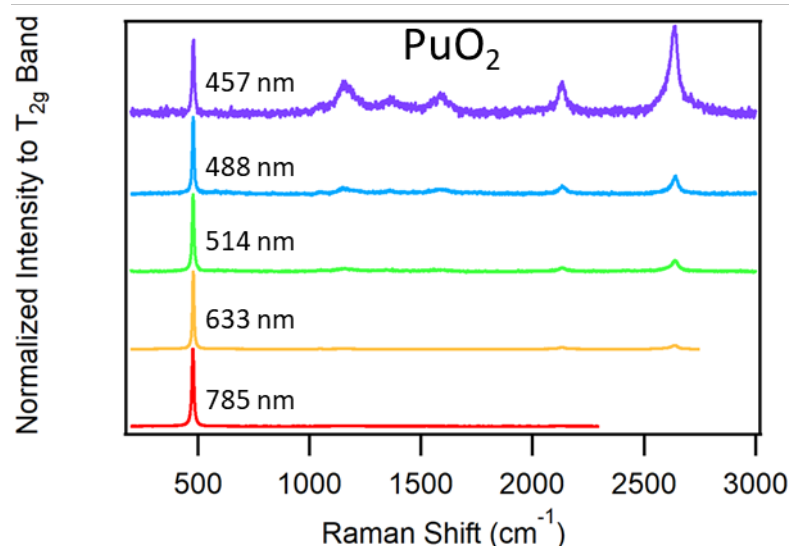
Figure 2 Spectra of PuO$_2$ Raman spectroscopy showing the effects of wavelength dependent resonance Raman.[32]

Unlike UO$_2$, some Raman mode assignments of neptunium dioxide are uncharacterized.[21] Interestingly, Naji et.al investigated the T$_{2g}$ band of NpO$_2$ that shows asymmetry at the low part of the band and was observed by other previously.[21,28,33] Further investigation with crystal field theory revealed that this is a secondary mode which is a signature of the NpO$_2$ substructure.[21]

Naji et al has also investigated AmO$_2$ with specially designed sample holders.[20] They were able to mount the sample on an SEM stub with carbon sticky tape. The stub was attached to a plexiglass rod that fit into a plexiglass cylinder with a lock ring. At the top of the cylinder was a quartz window to allow measurements via Raman lasers. While he was able to measure the T$_{2g}$ band of AmO$_2$, the signal to noise was insufficient for complete characterization of all the Raman bands.

Hydrolysis studies
To understand long term nuclear waste forms, an understanding of actinide materials interaction with water vapor is needed. Water vapor and temperature are usually more important than the presence of liquid water, as liquid water can more easily carry materials and create contamination. The water vapor could cause redox reactions and therefore change the speciation and oxidations state of the materials both internal and external.[16] A change in oxidation state could change the mobility of materials. Investigations at the solid-water interface will be performed by vibrational spectroscopy and electron microscopy to elucidate speciation, internal and external morphological changes, and an investigation of surface boundaries. Additional details such as oxidation state will come from synchrotron XANES data.

Most hydrolysis studies of actinide materials involve uranium and uranium oxides due to their relevance for nuclear fuel and waste.[9-12] Very few hydrolysis studies, if any, involve thorium, neptunium, and transplutonium dioxides or mixed actinides.[3,4,13-15] Actinide surface speciation can be investigated in great detail with spectroscopic techniques such as EXAFS, vibrational spectroscopy, or electron micrsocopy.[16] Actinide mineral-water interface reactions have been thoroughly summarized for non-oxide actinides;[16] however, the reaction mechanisms provide useful ideas into the incorporation of water into the actinide lattice as a result of hydrolysis.

Recent work at SRNL has described the hydrolysis of UF4 by Raman, IR, and electron microscopy.[34] TEM/EDS of hydrolyzed UF$_4$ developed fluorine deficient, oxygen rich pores in the interior of the particle (Figure 3A-B). The surface morphology of the particle had a smoothing of the surface and a visible defect allowing water vapor to enter the interior of the particle(Figure 3C). For higher relative humidity, an oxide shell formed, with empty space between the shell and a core of UF$_4$(Figure 3D). The hydrolysis was measured insitu by Raman spectroscopy (*Figure 4* left) demonstrate UF$_4$ bands still present during oxide formation. TEM/EELS analysis of other UF$_4$ hydrolyzed particulates revealed visible layers with increased oxygen content nearest the surface, confirming the oxide shell (*Figure 4* right).

SEM images will be taken of the particles to investigate surface morphology changes and EDS/EELS can be direct probes of the surface layers. A selection of particles will have their interior exposed by using focused ion beam milling techniques. Ion milling allows investigation of the interior of the particle to investigate the presence of pores and thinning of samples needed for TEM/EELS measurements. TEM measurements on the interior of the particle are a way to determine phases of crystalline materials. The phases can help determine speciation changes to the interior of the particle compared to the surface

edge. EELS measurements are much the same as EXAFS measurements, and is a way to determine atoms in the crystalline lattice. Electron microscopy is a technique to observe and measure changes to individual particles and will be useful in determining and mitigating expected changes for future nuclear waste storage.
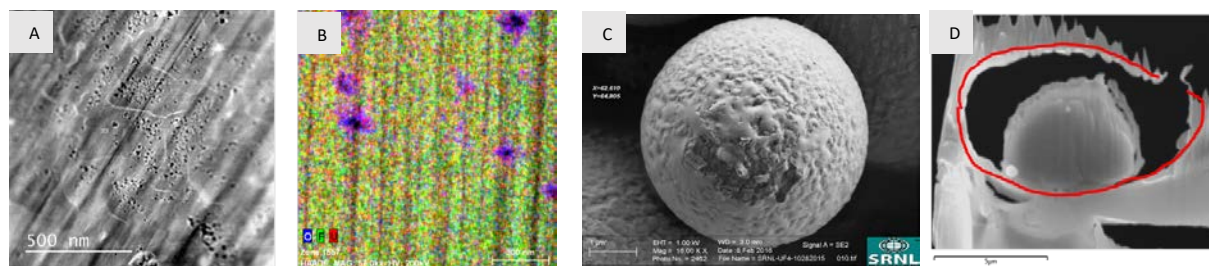

Figure 3 Combination of TEM images and EDS mapping where; A) TEM micrograph showing pores within the interior of particle exposed after FIB processing. B) EDS mapping where pores show reduced fluorine content relative to the bulk material. C) SEM image of hydrolyzed particle at 85% humidity showing the breakdown of the shell where water vapor is starting to penetrate D) FIB image of hydrolyzed particle at 85% RH. Red lines show particle surface
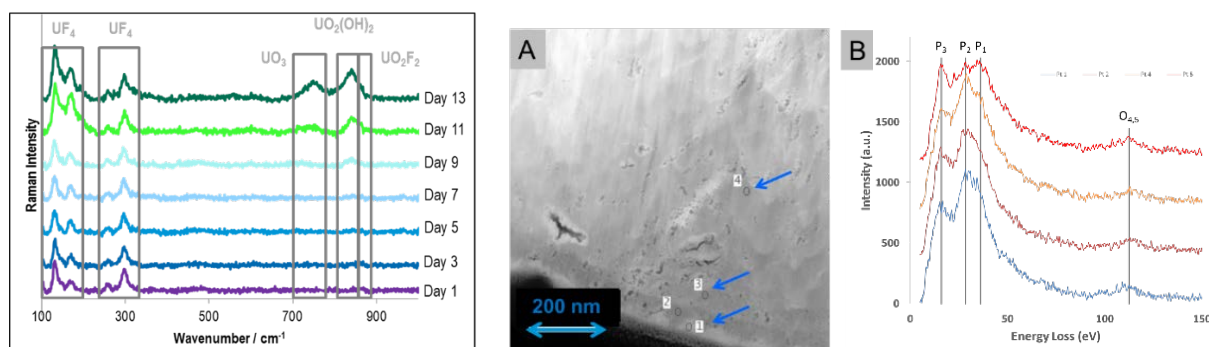

Figure 4 *Left:* In-situ Raman spectra of UF_4 hydrolysis at 85% relative humidity. The uranium oxides are part of the shell and UF_4 is in the core of the particle. Right: (a) HAADF STEM image (b) EELS spectra collected near the surface at position 1-4. The low loss regions show the U absorption edge peaks for P and O-type electronic transition states.

Vibrational spectroscopy, mentioned above, will be used to identify changes in the speciation of materials both at the surface and through penetration of the laser in the materials. Raman spectroscopy can identify the bulk speciation, while infrared can indicate the presence and location of water due to OH stretching and bending modes.

Research Plan
Through previous research and legacy activities, SRNL has the facilities and knowledge to process many transactinide materials for research and development. SRNL also has many of the materials needed for

this project on site and sufficient quantities. Synthesis of actinide dioxide (MO_2, M = Th, Np, Pu, Am, Cm) materials will take place at SRNL using negative pressure gloveboxes and published synthesis routes. The materials will be stored in salt-bath controlled relative humidity chambers, and aliquots removed every 180 days for characterization and analysis. It is expected that higher relative humidity should change the particles (chemically and physically) faster.

Experiments to be performed involve synchrotron radiation, vibrational spectroscopy, electron microscopy, and theoretical calculations. Hard and tender X-ray synchrotron experiments will take place at the National Synchrotron Light Source II at Brookhaven National Laboratory. The hard X-ray techniques to be used at NSLS-II will be fluorescence microprobe to locate the particle, L_3 edge XANES, and EXAFS. The L_3 edge XANES will give information about the contribution of the 6d orbitals to bonding and hints about oxidation state of the material. EXAFS will give information about the size of the crystalline lattice, and the distance and speciation of neighboring atoms. The tender X-rays will be the M_5 edge XANES and are used to measure the contribution of the 5f orbitals. The M_5 edge is also more sensitive to oxidation state changes than the L_3 edge of the hard X-rays in characterizing oxidation or reduction of the metal center. Scanning Transmission X-ray Microscopy (STXM) synchrotron experiments will take place at the Advanced Light Source at Lawrence Berkeley National Laboratory. Samples for STXM will be prepared by electron microscopy particle milling, to minimize radiation. STXM will be used to measure the K-edge of the oxygen atoms and the contribution to bonding of the 2p orbital. Theoretical calculations will take place at SRNL using the HPC center to simulate and match the synchrotron data. Vibrational spectroscopy will take place at SRNL's laser laboratory, equipped with different laser excitation wavelengths and techniques such as Raman and infrared spectroscopy. Vibrational spectroscopy will characterize speciation on both the surface and interior of the particles dependent on laser excitation penetration. There are also new bands expected to increase in intensity as the hydrolysis continues, especially at the HOH bending (~1600 cm^{-1}) and OH stretching regions (~3200 cm^{-1}). The speciation of the materials is an important part in establishing progeny from hydrolysis and developing a kinetic hydrolysis model. Crystal field calculations will help interpret the presence of new Raman bands and their association with the T_{2g} band. Electron microscopy will be performed via subcontract with EXpressLO LLC. EXpressLO are experts in ex-situ lift out and SEM/TEM characterization. They will also prepare samples for STXM as mentioned earlier. Electron microscopy will focus on describing the surface morphology and particle size as a function of hydrolysis time and relative humidity. Focused ion beam (FIB) milling of the particles will reveal their interior grain structure and possibly the presence of pores, indication of water inside the material. Near the edge of the interior of the particle, visible surface layers are expected to form which could be the presence of oxidized actinide oxide materials. SEM/TEM/EDS characterization of the layers and pores could reveal oxygen rich areas compared to the rest of the particle that should contain more actinides.

The first year will involve 5 tasks to get the project started on the correct path. First, will be a number of administrative tasks. Safety documents will need to be prepared, evaluated, and approved. During safety documentation evaluation, a subcontract will be prepared for EXpressLO LLC for electron microscopy experiments discussed above. SRNL designed relative humidity salt baths will be procured via rapid prototype. In consultation with radiological protection and the lead laser spectroscopist, this project will develop new vibrational spectroscopy holders for transplutonium materials. A double containment cell with quartz windows are already approved for plutonium. The new cells will be of similar shape and size but made of bismuth to shield the operators from radiation. A third task for year 1 will be a meeting with

beamline scientists to discuss goals and objectives of the project, and meeting with each beamline's radiological protection. This is used to establish working relationships with scientists and radiological protection. These meetings will also be used to discuss general user proposal requirements and tips for having successful proposals. Task four is the beginning of theoretical calculations to repeat published results and provide a starting basis for this work. Calculations will also be performed to predict t = 0 samples synchrotron and crystal field theory. A final task will be the synthesis, characterization, and storage of actinide dioxide materials using published procedures in the last quarter. The materials will be characterized by powder X-ray diffraction to determine purity, and if further processing is needed. An aliquot will be removed and stored in low humidity chambers until analysis can be performed. The rest of the actinide dioxides will be stored in salt-bath controlled relative humidity chambers purchased in task #1 with hygrometers. The relative humidity will be set at

Year 2 has five tasks that will be accomplished. First is the characterization of T = 0 samples via vibrational spectroscopy and electron microscopy. The characterization of T = 0 samples will be the basis of comparison for all subsequent sampling. The next task is the development and acceptance of general user proposals for synchrotron experiments. Task three is the removal of T=180 and T=360 day samples, and their subsequent characterization by vibrational spectroscopy and electron microscopy. Vibrational spectroscopy will investigate speciation changes, new Raman bands, and bands located at the HOH bending or the OH stretching region. Electron microscopy will explore changes to morphology, both internal and external. Task four is the first synchrotron experiments of the t=0 and t=180 day samples and analysis. These will only involve the hard and tender X-rays of the NSLS-II. If there are interesting or unexpected results, a STXM sample will be prepared via electron microscopy for characterization and analysis. Task five is theoretical calculations to explain any new Raman bands, and to fit synchrotron experimental data. The contribution of the 5f/6d orbitals will also be calculated.

Year three also has 5 tasks that need to be accomplished. Task one, is the successful synthesis and characterization of mixed actinide dioxides (U/Pu, U/Am, etc.). Mixed systems are more realistic to long term storage waste forms. They are also unique in that 5f electrons should mix if the metals are in within close proximity. This would bring unique spectroscopy and is theorized that there could be mixed oxidation state systems. The mixed actinide dioxides will first be characterized by powder XRD for purity. The second task involves the removal of aliquots of single actinide dioxides (t =540 and t=720), and t =180 mixed actinide dioxide materials for characterization via electron microscopy and vibrational spectroscopy. This task will investigate speciation changes, and morphological changes, such as the presence of pores, or visible surface layers. Task three is the synchrotron characterization of the t=360, t=540 days single actinide dioxides and t=0 t=180 mixed actinide dioxides. If there are interesting or unexpected results, a STXM sample will be prepared via electron microscopy for characterization and analysis. Task five is theoretical calculations to explain any new Raman bands, and to fit synchrotron experimental data.

Year 4 will be a continuation of year 3 tasks. Vibrational spectroscopy and electron microscopy will be performed on t=900 and t=1080 days single actinide dioxide samples, and for t=360 and t=540 mixed actinide dioxide samples. Another task is the synchrotron characterization of t=720 and t=900 single actinide dioxides and t=360 and t=540 mixed actinide dioxides. The last task will involve the theoretical calculations to fit synchrotron data and determine orbital contributions.

The final year tasks will include final synchrotron measurements as needed. Final theoretical calculations will be performed throughout the year as well as final interpretation of vibrational spectroscopy and synchrotron experimental data. A final report for the project will be produced. Materials will be stored or disposed as transuranic waste as SRNL policy dictates.

Appendix 1 Biographical Sketch

Education and Training

Post-doctoral Research

Savannah River National Laboratory 2015-2016

Oak Ridge National Laboratory 2014-2015

Education

Ph.D. – Auburn University, Organic Chemistry, 2014

M.S – Auburn University, Organic Chemistry, 2012

B.S. – University of Tampa, Chemistry 2007

Research and Professional Experience

Senior Scientist – Savannah River National Laboratory – Work in non-proliferation technology section, in the national security division. Have worked on various NA-22 and LDRD projects involving trace measurements of nuclear materials using electron microscopy and vibrational spectroscopy. NA-241 project for production of actinide containing particulates as reference materials for SIMS measurements for the IAEA. LDRD for the measurement of PuO_1 at the Advanced Photon Source at Argonne National Laboratory.

Post-Doctoral – Savannah River National Laboratory – Have worked on various NA-22 and LDRD projects from trace measurements of nuclear materials using electron microscopy and vibrational spectroscopy.

Post-doctoral – Oak Ridge National Laboratory – Work in Chemical Sciences Division for Bruce Moyer/Laetitia Delmau. Project involved the synthesis of calix-arenes with arm ligands attached to extract uranyl compounds. By binding with the ligands, the uranyl bond should stretch, and this was thought to cause the oxygen atoms to become more negative and allow for hydrogen bonding with the calix-arene.

Graduate School – Auburn University – Worked for Anne Gorden. Performed organic synthesis for various quinoxaline Schiff base ligands. Used the ligands for the UV-Vis and fluorescent detection of uranyl, copper, and cobalt in aqueous solutions.

Publications

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2. M.A. DeVore II; M.S. Wellons; R.J. Smith; M.E. Summer; L.D. Trowbridge; H.L Jennings, "Synthesis and Characterization of UF_5 from Reduced UF_6 Deposits" SRNL-STI-2016-00733 December 2016
3. M. Wellons, M. DeVore II, E. Villa-Aleman, T. Dourradi, "Characterization of Environmentally Induced Chemical Transformations of Uranium Tetrafluoride: Project #LDRD-2016-00031" LDRD External Report Summary
4. M. Wellons, M. DeVore II, M. Summer, "3D Characterization of Micron Particulates" – LDRD External Report Summary

5. Gorden, A.E.V., DeVore II, M. A., Maynard, B., "Coordination Chemistry with *f*-Element Complexes for an Improved Understanding of Factors That Contribute to Extraction Selectivity." *Inorganic Chemistry* **2013,** 52, 3445

Synergistic Activities

Secretary, Savannah River Local Section of American Chemical Society, 2019 – present

Member, American Chemical Society, 2008 – present

Member, Materials Research Society, 2015 – present

Collaborator and Co-Authors

Savannah River National Laboratory

Dr. Matthew S. Wellons, Dr. Eliel Villa-Aleman, Dr. Wendy Kuhne, Dr. Michael Bronikowski, Dr. Joseph Mannion, Dr. Amanda Houk, Dr. Jacob Venzie, Dr. Maria Kriz, Dr. Thomas Shehee, Dr. Lindsay Roy

Argonne National Laboratory

Dr. Jeffrey Fortner, Dr. Matthew Newville, Dr. Antonio Lanzirotti

Clemson University

Dr. Brian Powell, Dr. Lindsay Schuller-Nickels

Other Collaborators

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Dr. Lucille Giannuzzi, EXpressLO LLC

Graduate Advisor

Dr. Anne Gorden, Auburn University

Post-Doctoral Advisors

Matthew S. Wellons, Savannah River National Laboratory

Laetitia Delmau and Bruce Moyer, Oak Ridge National Laboratory

Appendix 2: Current and Pending Support

Current Support

NA-241 – Generation and Characterization of a Uranium Particulate Reference Material

Total = $745K

Start – August 2018

Effort – 8-10 months

The International Atomic Energy Agency (IAEA) has requested uranium reference particles of specific isotopic compositions for proficiency swipe testing applications. The request includes micron sized particulates, of four different compositions, and identified as priority #1 or #2 respectively. However, there are no readily available uranium containing particulate test materials with both the required particle size distributions and isotopic compositions. SRNL has recently demonstrated the ability to generate viable uranium reference particulates of controlled size and has developed the capability to tailor their isotopic composition. SRNL proposes to continue production efforts begun in FY18 to meet IAEA particulate reference materials needs via a multifaceted approach. This includes completing the synthesis of priority #1 particulates begun in FY18, their characterization, and packaging in a form suitable for use by the IAEA. This work will be complemented with laboratory studies to assess potential multi milligram particulate production and collection methods. Priority #2 isotopic composition requirements are anticipated to be finalization in FY19; SRNL will procure starting materials needed, preform the necessary chemistry to mix and generate the chemical feedstocks required, and begin particle production. Lastly, SRNL will initiate efforts to generate mixed actinide particulate reference materials capable of addressing future IAEA needs.

SRNL LDRD – Synchrotron Based Microstructural Characterization Method Development for Pu Oxides

Total = $60K

Start – October 2018

End – September 2019

Effort – 6 months

Advanced microstructural techniques (Synchrotron, SEM, TEM) were used to probe the internal structure of plutonium oxide particles as part of the development of new forensic signatures. The objective was to characterize plutonium oxide samples, synthesized by various routes, to develop an understanding between process conditions and particle morphology.

Pending Support

NA-241 - Generation and Characterization of a Uranium Particulate Reference Material

Total - $700K

Start – October 2019

Effort – 6-8 months

The International Atomic Energy Agency (IAEA) has requested uranium reference particles of specific isotopic compositions for proficiency swipe testing applications. The request includes micron sized particulates, of four different compositions, and identified as priority #1 or #2 respectively. However, there are no readily available uranium containing particulate test materials with both the required particle size distributions and isotopic compositions. SRNL has recently demonstrated the ability to generate viable uranium reference particulates of controlled size and has developed the capability to tailor their isotopic composition This work will be complemented with laboratory studies to assess potential multi milligram particulate production and collection methods. Priority #2 isotopic composition requirements are anticipated to be finalization in FY19; SRNL will procure starting materials needed, preform the necessary chemistry to mix and generate the chemical feedstocks required, and begin particle production. Lastly, SRNL will initiate efforts to generate mixed actinide particulate reference materials capable of addressing future IAEA needs.

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Appendix 4: Facilities and Other Resources

Laser Spectroscopy Laboratory

- Raman Spectroscopy (Lasers available range from ultraviolet to infrared)
- Advanced Raman Spectroscopy (Stimulated Raman Spectroscopy, Coherent Anti-Stokes Raman Spectroscopy, etc.)
- Infrared Spectroscopy (Attenuated Total Reflectance, Reflectance using Microscope, Photoacoustic)

Additional Laboratory Equipment

- Femtosecond, Picosecond, and CW Lasers
- Spectrometers and Spectrographs (Spectral response range from ultraviolet to infrared)
- Microscopes can be coupled with lasers and spectrographs
- Oscilloscopes

CW Lasers

244 nm (Class 4) Argon
- Power: 0.025 W (Diameter: 1.1 mm)
257 nm (Class 4) Argon
- Power: 0.05 W (Diameter: 1.1 mm)
325 nm (Class 3B) HeCd
- Power: 0.015 W (Diameter: 1.2 mm)
414 nm (Class 3B) Diode
- Power: 0.018 W (Diameter: 2.2 mm)
457 nm (Class 4) Argon
- Power: 0.045 W (Diameter: 1.1 mm)
488 nm (Class 3B) Argon
- Power: 0.130 W (Diameter: 0.72 mm)
488 nm (Class 4) Argon
- Power: 0.35 W (Diameter: 1.1 mm)
514 nm (Class 4) Argon
- Power: 0.4 W (Diameter: 1.1 mm)
532 nm (Class 3B) Diode
- Power: 0.020 W (Diameter: 1.2 mm)
532 nm (Class 4) Diode
- Power: 6 W (Diameter: 2.25 mm)
532 nm (Class 4) Diode
- Power: 10 W (Diameter: 2.25 mm)
561 nm (Class 3B) Diode
- Power: 0.050 W (Diameter: 0.2 mm)
633 nm (Class 3B) HeNe
- Power: 0.005 W (Diameter: 0.59 mm)
633 nm (Class 3B) HeNe
- Power: 0.015 W (Diameter: 0.75 mm)
633 nm (Class 3B) HeNe
- Power: 0.017 W (Diameter: 0.96 mm)
633 nm (Class 3B) HeNe
- Power: 0.038 W (Diameter: 1.25 mm)
635 nm (Class 3R) Diode
- Power: 0.0045 W (Diameter: 3 mm)
662-679 nm (Class 3B) Diode
- Power: 0.015 W (Diameter: 2.7 mm)
670 nm (Class 3B) Diode
- Power: 0.5 W (Diameter: 3 mm)
780 nm (Class 3B) Diode
- Power: 0.050 W (Diameter: 3 mm)
785 nm (Class 3B) Diode
- Power: 0.095 W (Diameter: 2.5 mm)
785 nm (Class 3B) Diode
- Power: 0.4 W (Diameter: 2.6 mm)
785 nm (Class 3B) Diode
- Power: 0.5 W (Diameter: 1 mm)
788 nm (Class 4) Diode
- Power: 1.5 W (Diameter: 3 mm)

1064 nm (Class 3B) Diode
- Power: 0.050 W (Diameter: 1 mm)
1064 nm (Class 4) Diode
- Power: 1 W (Diameter: 1.47 mm)
1064 nm (Class 4) Diode
- Power: 1.421 W (Diameter: 2 mm)
10600 nm (Class 4) CO2
- Power: 40 W (Diameter: 3.5 mm)
10600 nm (Class 4) CO2
- Power: 125 W (Diameter: 4.4 mm)

Pulsed Lasers

200-600 nm (Class 4) Crystal Tuning
- Energy: 40, 50, 24, and 20 mJ/pulse (Diameter: 5 mm)
- Pulse Duration: 10 ns (10 Hz)
235-2600 and 4000-16000 nm with 800 nm laser pumping the OPA (Class 4) Ti:Sapphire
- Power: 0.5 W (Diameter: 6 mm)
- Pulse Duration: 32.5 fs (1000 Hz)
266, 355, 532, and 1064 nm (Class 4) Nd:YAG
- Energy: 10, 15, 30, and 50 mJ/pulse (Diameter: 3 mm)
- Pulse Duration: 7 ns (15 Hz)
337 nm (Class 4) Nitrogen
- Power: 0.0024 W (Diameter: 3 mm)
- Pulse Duration: 3 ns (20 Hz)
337 and 400-700 nm (Class 1: Embedded Class 4) Nitrogen
- Power: 0.0024 W (Diameter: 3 mm)
- Pulse Duration: 3 ns (20 Hz)
355, 532, and 1064 nm (Class 4) Nd:YAG
- Energy: 5.9, 5.65, 15.7 mJ/pulse (Diameter: 10 mm)
- Pulse Duration: 9 ns (10 Hz)
355, 532, and 1064 nm (Class 4) Nd:YAG
- Energy: 180, 400, and 600 mJ/pulse (Diameter: 6.4 mm)
- Pulse Duration: 10 ns (10 Hz)
355, 532, and 1064 nm (Class 4) Nd:YAG
- Energy: 275, 135, 60, and 30 mJ/pulse (Diameter: 1 mm)
- Pulse Duration: 10 ns (14 Hz)
360-700 nm (Class 4) Dye
- Energy: 132 mJ/pulse (Diameter: 6 mm)
- Pulse Duration: 10 ns (10 Hz)
360-700 nm (Class 4) Dye
- Energy: 400 mJ/pulse (Diameter: 6 mm)
- Pulse Duration: 10 ns (10 Hz)
532 and 1064 nm (Class 4) Nd:YAG
- Power: 4.65 and 3.3 W
- Pulse Duration: 5 ns (10 Hz)
720-1000 nm (Class 4) Ti:Sapphire
- Power: 1.4 W (Diameter: 0.8 mm)
- Pulse Duration: 200 fs (76 MHz)
720-1000 nm (Class 4) Ti:Sapphire
- Power: 1.4 W (Diameter: 0.8 mm)
- Pulse Duration: 3 ps (76 MHz)
800 nm (Class 4) Ti:Sapphire
- Power: 7.45 W (Diameter: 11.0 mm)
- Pulse Duration: 32.5 fs (1000 Hz)
1064 nm (Class 4) Q-Switch
- Energy: 100 uJ/pulse (Diameter: 2 mm)
- Pulse Duration: 10 ns (10000 Hz)
1319 nm (Class 4) Q-Switch
- Power: 25 mW (Diameter: 1.5 mm)
- Pulse Duration: 10 ns (5000 Hz)

High Performance Computing – SRNL

General Application

Bowtie2 – Bowtie 2 is an ultrafast and memory-efficient tool for aligning sequencing reads to long reference sequences

Gaussian – Latest in the Gaussian series of electronic structure programs

HDF – Data model, library, and file format for storing and managing data

HP-MPI – High performance implementation of the MPI standard for HP servers and workstations

MPICH – implementation of the Message Processing Interface standard that supports clusters

OCTAVE – Open source alternative for MATLAB. Numerical computing environment and programming language.

OpenMPI – Open MPI is an open source, freely available implementation of both the MPI-1 and MPI-2 documents

Paraview – Open-source, multi-platform data analysis and visualization application

Python – Dynamic, modular programming language with easy-to-understand syntax

R – Statistical computing and graphics software environment

RSEM – RSEM is a software package for estimating gene and isoform expression levels from RNA-Seq data

SAMtools – SAM (Sequence Alignment/Map) format is a generic format for storing large nucleotide sequence alignments

Solve – Automated crystallographic structure solution for MIR, SAD, and MAD

Szip – General purpose lossless compression program

Tophat- TopHat is a program that aligns RNA-Seq reads to a genome in order to identify exon-exon splice junctions

VisIt – Parallel visualization and graphical analysis tool for viewing scientific data

License Based

Abaqus – Abaqus is a suite of general purpose nonlinear finite element analysis (FEA) tools

ADF – ADF package is software for first-principles electronic structure calculations

Comsol – Finite element analysis and solver software package for various physics and engineering applications

Fluent – Comprehensive CFD (computational fluid dynamics) analysis tool

Gambit – CFD Preprocessor used to create geometry models and meshes for simulations in FLUENT

Medea – Predicts materials properties using simulations

PGI Compilers – Portland Group (PGI) Fortran, C, and C++ compilers and program development tools

Porflow – Solves problems involving transiet or steady state fluid flow, salinity and mass transport in porous media

Tecplot – Engineering plotting software with extensive XY, 2D and 3D capabilities

Tecplot 360 – Numerical simulation and CFD visualization software

RSICC Controlled

Dragon – Lattice Cell Code System

MCNP – General-purpose Monte Carlo N–Particle code

Windows

Aspen – Engineering plotting software with extensive XY, 2D and 3D capabilities

PGI Compilers – Portland Group (PGI) Fortran, C, and C++ compilers and program development tools

HPC Nodes
Disk Space: 125TB
Cluster: 988 processors, 81 compute nodes
Cluster Nodes
8 Westmere-EP nodes with 164GB of memory
2 6-core Intel Xeon CPUX5690
12 cores/node
164GB memory

12 Magny-Cours nodes with 12GB of memory
1 8-core AMD Opteron Processor 6140
8 cores/node
12GB memory
12 Westmere-EP nodes with 76GB of memory
2 6-core Intel Xeon CPUX5690
12 cores/node
76GB memory
10 Westmere-EP nodes with 127GB of memory
2 6-core Intel Xeon CPUX5675
12 cores/node
127GB memory
6 Gainestown nodes with 24GB of memory
2 4-core Intel Xeon CPUE5520
8 cores/node
24GB memory
12 Istanbu nodes with 32GB of memory
2 6-core AMD Opteron Processor 2435
12 cores/node
32GB memory
12 Abu Dhabi nodes with 64GB of memory
2 12-core AMD Opteron Processor 6344
24 cores/node
64GB memory
1 Shanghai nodes with 256GB of memory
4 4-core AMD Opteron Processor 8384
16 cores/node
64GB memory

Radiological Laboratory

Equipped with gloveboxes and radiological hoods for material manipulation, storage, and handling.

Machine Shop – SRNL – They are able to make a number of holders as needed.

Rapid Prototype Laboratory – SRNL – Plastic holders as needed for synchrotron work.

Appendix 5: Equipment

Glovebox in radiological laboratory in nuclear facility

Appendix 6: Data Management Plan

Data from synchrotron experiments will be stored on 2 external hard drives (primary and backup). The data will also be on a shared drive at SRNL.

Data from vibrational spectroscopy will be stored on the laboratory instrument, 2 external hard drives (primary and backup), and a shared drive at SRNL.

Data from electron microscopy will be stored at the collaborators institution, and 2 external hard drives (primary and backup).

Data from computational work will be stored on the HPC system, and 2 external hard drives (primary and backup).

Data will be disseminated as requested by DOE-BES and at conferences and presentations.