EFRC: MATERIALS SCIENCE OF ACTINIDES (MSA)

UPDATED: AUGUST 2016

AWARDS: $18.5M (August 2009 – July 2014); $11.6M (August 2014 – July 2018)
WEBSITES: http://science.energy.gov/bes/efrc/centers/msa/; http://msa-efrc.com
TEAM: **University of Notre Dame (Lead):** Peter C. Burns (Director), Jeremy Fein, Amy Hixon, Edward Maginn; **Stanford University**: Rodney C. Ewing, Wendy Mao; **University of California, Davis**: Alexandra Navrotsky, William H. Casey, Mark Asta; **University of Tennessee:** Maik Lang; **University of Minnesota:** Laura Gagliardi; **University of Akron:** Tianbo Liu; **Oregon State University:** May Nyman; **George Washington University:** Christopher Cahill; **Los Alamos National Laboratory:** Albert Migliori

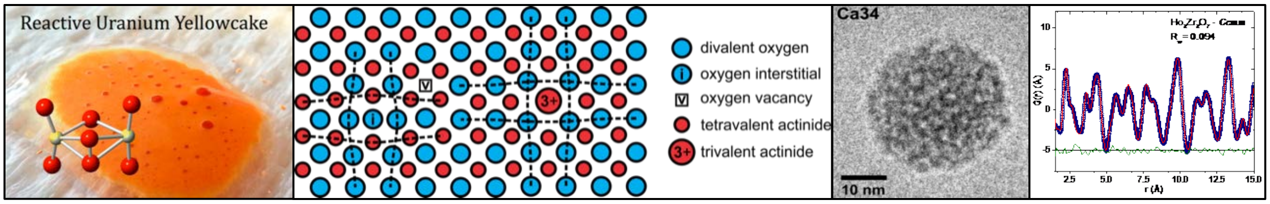
SCIENTIFIC MISSION AND APPROACH

The mission of the MSA is to conduct collaborative, multidisciplinary, novel and transformative research on actinide materials emphasizing actinide ceramic, metallic, hybrid, and nanoscale materials; effectively integrate experimental and computational approaches; and solve research questions that are critical to the energy future of the nation. Workforce development is a fundamental and inherent goal of this university-based center. The center consists of three themes:

1) **Nanoscale Cage Clusters**: Develop and understand the unique properties of cage clusters built from actinyl ions to exert nanoscale control of actinide materials.
2) **Complex Ceramic and Metallic Materials**: Develop studies of the thermochemistry of actinide materials of importance in the fuel cycle that expand knowledge of actinide chemistry and physics.
3) **Materials Under Extreme Environments:** Study the response of actinide materials to combinations of extreme conditions of temperature, pressure, chemical environments, and high-radiation fields.

SELECTED SCIENTIFIC ACCOMPLISHMENTS

- Developed a family of several dozen self-assembling uranyl cage clusters with diameters from 1.5 to 4.0 nm containing as many as 124 uranium atoms, as well as an understanding of their properties, formation mechanisms, and potential applications in nuclear material processing.
- Energetic radiation can cause dramatic changes in actinide materials, degrading their performance in nuclear energy systems. Advanced synchrotron techniques showed that the redox behavior of the actinide governs radiation tolerance. Limiting the redox activity of actinide materials through control of composition or microstructure can reduce swelling and microstrain.
- In high temperatures or highly ionizing radiation, complex oxides partially lose their crystallinity, which was assumed to occur by a random mixing of their atomic constituents. Neutron scattering experiments revealed that the atoms are not randomly arranged, but only appear so when sampling over longer length scales. Discrepancies may arise when extrapolating the materials structure from the microscale to the atomic scale (or vice versa), which has significant implications for modelling properties and degradation phenomena in actinide materials.
- Uranium dioxide is an important nuclear fuel but it builds up impurities of other oxides as fission products during and after use. A combination of calculations and experimental measurements provided the energetics and structural features of these substitutions, enabling better understanding of the behavior of fuel rods in a reactor and when stored as waste.
- Uranyl peroxide solids and clusters have been shown to be widespread in the nuclear fuel cycle. Their thermodynamic properties have been determined to constrain their formation and decomposition conditions and their potential impact on uranium transport in the environment.



MSA research, from left: reaction of uranium yellowcake with water, releasing O_2 gas, due to compound U_2O_7; cation reduction causes structural distortion in the actinide oxide lattice that are identical to that caused by Frenkel defects; cryogenic transmission electron microscope image of a blackberry formed by the aggregation of U60 clusters in water; Neutron scattering data revealing the local structure of disordered $Ho_2Zr_2O_7$.

IMPACT

- **Conference Symposiums Organized by MSA**: American Chemical Society (ACS) Symposium Fall 2013 (Indianapolis). ACS Symposium Spring 2011 (Anaheim). ACS Symposium Fall 2009 (Washington, D.C.).
- **Selected Awards**: **Ewing**: Roebling Medal of the Mineralogical Society of America, International Mineralogical Association Medal. **Burns**: Peacock Medal of the Mineralogical Association of Canada. **Navrotsky**: American Ceramic Society Kingery Medal, Geochemical Society Goldschmidt Medal. **Migliori**: Joseph F. Keithley Award for Advances in Measurement Science. **Casey**: Geochemical Society Patterson Medal.
- EFRC research concerning pressurization of drums of uranium yellowcake revealed the presence of an amorphous and reactive uranium peroxide that was structurally characterized. This material may release oxygen gas, potentially causing pressurization of drums of yellowcake during storage or transport that could cause an accident. This research has been used by **Uranium One** to prevent accidents involving rapid depressurization of yellowcake drums. http://www.uranium1.com/

PUBLICATIONS AND INTELLECTUAL PROPERTY

As of May 2016, MSA had published 272 peer-reviewed publications cited over 4,200 times and filed 4 disclosures and 4 US patent applications. The following is a selection of impactful papers:

- Burns, P.C., Ewing, R.C., Navrotsky, A. Nuclear fuel in a reactor accident. *Science* **335**, 1184-1188, DOI:10.1126/science.1211285 (2012). [**101 citations**]
- Qiu, J., Burns, P.C. Clusters of actinides with oxide, peroxide, or hydroxide bridges. *Chemical Reviews* **113**, 1097-1120, DOI:10.1021/cr300159x (2013). [**82 citations**]
- Tracy, C.L. *et al.* Redox response of actinide materials to highly ionizing radiation. *Nature Communication* **6**, 6133, DOI:10.1038/ncomms7133 (2015). [**12 citations**]
- Vlaisavljevich, B., Gagliardi, L., Burns, P.C. Understanding the structure and formation of uranyl peroxide nanoclusters by quantum chemical calculations. *Journal of the American Chemical Society* **132**, 14503-14508, DOI:10.1021/ja104964x (2010). [**63 citations**]
- Shamblin, J. *et al.* Probing disorder in isometric pyrochlore and related complex oxides. *Nature Materials* **15**, 507-511, DOI:10.1038/nmat4581 (2016). [**8 citation**]
- Lang, M. et al. Characterization of ion-induced radiation effects in nuclear materials using synchrotron X-ray techniques. *Journal of Materials Research* **40**, 1366-1379, DOI:10.1557/jmr.2015.6 (2015). [**3 citation**]
- Guo *et al.* Thermodynamics of formation of coffinite. *Proceedings of the National Academy of Sciences of the United States of America* **112**, 6551-6555, DOI:10.1073/pnas.1507441112 (2015). [**12 citations**]
- Odoh, S.O. *et al.* Structure and reactivity of X-ray amorphous uranyl peroxide, U_2O_7. *Inorganic Chemistry* **55**, 3541-3546, DOI:10.1021/acs.inorgchem.6b00017 (2016). [**3 citation**]
- Rai, N. *et al.* Force field development for actinyl ions via quantum mechanical calculations: An approach to account for many body solvation effects. *Journal of Physical Chemistry B* **116**, 10885-10897, DOI:10.1021/jp3028275 (2012). [**19 citations**]
- Wang, S. *et al.* NDTB-1: A supertetrahedral cationic framework that removes TcO_4^- from solution. *Angewandte Chemie International Edition* **49**, 1057-1060, DOI:10.1002/anie.200906397 (2010). [**87 citations**]