

Center for Actinide Science & Technology (CAST)
EFRC Director: Thomas E. Albrecht-Schoenartz
Lead Institution: Florida State University
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Mission Statement: *To advance our understanding of how electronic structure and bonding control the properties of radioactive materials. This knowledge will aid in the development of nuclear technologies that enhance energy security, address nuclear legacy issues and environmental concerns, and foster the next generation of nuclear scientists.*

Cultivating new technologies that address the challenges associated with nuclear waste generated during the Cold War requires an advanced, science-based understanding of *f*-element (lanthanide and actinide) behavior both in solution and in the solid state. The Center for Actinide Science & Technology (CAST) brings together scientists with *f*-element expertise from Florida State University, the National High Magnetic Field Laboratory, Florida International University, Los Alamos National Laboratory, Lawrence Berkeley National Laboratory, Purdue University, the University of Pennsylvania, the University of Manitoba, and SUNY Buffalo to create synergy and cross-fertilization between chemists, physicists, materials scientists, and theoreticians. We are developing unique expertise in synthesis, characterization, and theory in order to create new materials that provide unprecedented chemical selectivity for specific radionuclides. An important aspect of this research is developing experimentally-verified theoretical methods for understanding complex chemistry and physics. These advancements in experiment and theory will expand our knowledge of actinide materials under normal and extreme conditions. In addition to the practical applications of this research, an improved understanding of these enigmatic elements will expand the boundaries of the periodic table.

Synthesis of Solid-State and Molecular Materials. The actinide materials and molecular systems that are the focus of CAST are being prepared using the unique synthetic capabilities at FSU, LANL, and LBNL. A host of physical property measurements on these materials are taking place at the NHMFL. This allows us to probe properties under a variety of extreme conditions including high magnetic fields (the highest in the world), temperatures, and pressures. Similarly, the synthesis of novel actinide materials and molecules is being lead by FSU, LANL, LBNL, and other partner institutions. The preparative efforts include the design, synthesis, and optimization of new and existing ligands needed for new transuranium separations. The ligand design and complexation studies with lanthanides, thorium, and uranium primarily occurs at the University of Pennsylvania and Purdue University, and is then transferred to institutions with transuranium laboratories. The capabilities of FSU and LANL to work with macroscopic quantities of transuranium elements, such as plutonium, for basic research purposes are almost unique. As such, CAST is focused on directly probing materials of interest such as plutonium solids and americium coordination complexes as opposed to using surrogates for these elements that often provide an incomplete or sometimes even inaccurate predictions for real radioactive molecules and materials. These materials are fully characterized using all available structural and spectroscopic methods that one would normally employ to understand non-radioactive materials. Thus, some of CAST's experimental efforts are devoted to developing safe methods for performing these measurements at user facilities. Obtaining the highest quality experimental data is essential not just to our experimental understanding, but also provides critical data for benchmarking new theoretical methods.

Electronic Structure Theory. There are no broadly available, reliable, and predictive electronic structure approaches for strongly-correlated materials. We are expanding our approaches to employ high-accuracy electronic structure methods for predicting the properties of actinide compounds. Results from these calculations are validated by comparison with experimental results including those from X-ray absorption

spectroscopy and magnetic susceptibility measurements. Of particular importance is the fact that actinide elements possess electrons that are travelling at significant fractions of the speed of light. This causes their mass to increase, the shape of orbitals to change, and reorganization of the orbital energies. Historical computational methods have not been up to the task of tackling the large number of electrons and the large number of orbitals involved in bonding in actinide coordination complexes or materials. Moreover, the importance of relativistic effects in these systems has also been neglected. Today methods exist that can tackle both large numbers of electrons and the effects of relativity, but even these modern methods utilize approximations that decrease the accuracy of these calculations. CAST theoreticians are working directly with experimentalists to obtain the data needed to benchmark true, four-component, Dirac-Fock calculations on actinide molecules and materials. These efforts represent the state-of-the-art and are being led by researchers at LANL, SUNY-Buffalo, FSU, and the University of Manitoba.

Solution Chemistry. The solution chemistry of actinides is complex because these metal ions possess high coordination numbers, rapid ligand exchange kinetics, and there is a lack of even a rudimentary understanding of the origin of ligand preferences by actinide ions. In general, it is not currently understood whether enthalpic or entropic factors control the selectivity of separations processes. To solve these issues, efforts are underway in CAST that focus on quantifying *f*-element-ligand interactions both experimentally and computationally. These data are then used to fine-tune these interactions and achieve maximize selectivity. In addition to these more classical studies, CAST researchers are also advancing photochemical separations that capitalize on the narrow line-width of the *f*-element absorption features to selectively transform ligands and enact separations simply using light from LEDs. Additionally, researchers at LANL, Purdue, UPenn, FSU, and FIU are utilizing X-ray spectroscopic methods to probe the electronic structure and speciation relevant to these separations processes. These spectroscopic measurements allow for a deeper understanding of actinide bonding and are used to benchmark the theoretical and simulation efforts.

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