**Combining Structural, Spectroscopic, and Biochemical Tools to Explore Coordination Chemistry Across the Actinide Series**

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From potential contamination of individuals with radioactive fission products after a nuclear accident to the therapeutic use of radioisotopes for cancer diagnostics and treatment, the biological chemistry of actinides is not only relevant to a number of applied problems, it has also revealed some outstanding selectivity and specificity features of natural systems for f-element binding. Understanding the fundamental bonding interactions of these selective metal assemblies presents a rich set of scientific challenges as well as opportunities to design new molecular architectures with specific applications in mind. Our experimental approach uses a combination of structural, spectroscopic, and biochemical studies to characterize the selective binding of f-block metal ions by natural and biomimetic hard oxygen-donor architectures. X-ray diffraction (XRD) and X-ray absorption studies (XAS) will be emphasized as these techniques have allowed for the first direct comparison of analogous small molecule and macromolecular complexes, as well as a pathway to explore the structural chemistry of some of the most exotic natural (AcIII) and synthetic (EsIII) radionuclides in the periodic table.