Glenn T. Seaborg Center Seminar

Actinide Speciation: A comparison of correlations in solution and in the solid state

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Similar to dissolved metal ions throughout the periodic table, tetravalent actinides in aqueous solutions have a propensity to hydrolyze. For many years, the observed changes in physical properties that accompany hydrolysis have been attributed to the formation of amorphous oxyhydroxides that, upon aging, lose waters to form metal oxides that are seen to precipitate. Despite the widespread impact of hydrolysis on actinide solution chemistry, an understanding of the mechanistic details and chemical products of these reactions has remained elusive, in part because of the lack of structural probes that can be applied to longer-range correlations in solution. We have approached this problem by combining advances in x-ray structural techniques to probe the atomic level structures of actinide ions and their hydrolysis products both in solution and in the solid state. By comparing the results from single-crystal structural studies with x-ray scattering data obtained from the mother liquors that produced the crystals, we are beginning to understand in more detail the simple hydrolysis and aging of tetravalent actinide ions. The overall picture that is emerging is somewhat different to that found in standard introductory textbooks and is able to better explain the wide ranging, often intractable, chemistries associated with dissolved metal ions in aqueous solution. This work is supported by the U.S. DOE, OBES-Chemical Sciences, under contract W-31-109-ENG-38 and by the University of Notre Dame Environmental and Molecular Sciences Institute.

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