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Computational Modeling of Actinide Ions in Aqueous Solution

RAYMOND ATTA-FYNN, Department of Physics, The University of Texas at Arlington — Unraveling the chemical behavior of actinide species is difficult owing to the complex electronic structure of these species, the fact that many of these species can occur in multiple oxidation states, and the difficulties encountered in their experimental studies. First principles dynamical modeling, although computationally costly, allows us to gain rich insights into the behavior of actinide species. In this talk, we present results of the hydration shell structure and x-ray absorption spectra of aqueous actinides in different oxidation states including U(VI), U(V), U(IV), and Cm(III) using relativistic *ab initio* molecular dynamics at 300 K. We also probed the thermodynamics of hydrolysis by calculating the first acidity constant for uranium in all three oxidation states (IV, V, and VI). We predicted, for the first time, that UO_2^+ is a weak acid in solution with a pKa value of 8.5. This result is particularly important since no thermodynamic data are available for hydrolyzed species of U(V). In our most recent work on concentrated Cm(III) solutions, we showed that counter-ions can strengthen or weaken the solvent structure itself rather than just the water coordination number. These new results are better explained in terms of the hydrogen bond lifetimes of the solvents.

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