Uranyl-Chloride Speciation at Hydrothermal Conditions.

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Understanding dissolution, speciation and reactivity of uranium-bearing phases in water-mediated processes at elevated temperature (T) and pressure (P) conditions is critical for tackling a range of issues in nuclear fuel cycle technologies from uranium ore formation to disposal of high-level wastes in a geological repository to development of robust accident-tolerant nuclear fuels. Despite the importance, our knowledge on the interactions of uranium-bearing phases with water at elevated T/P is limited. In this presentation, we will highlight our recent studies on the speciation and stability of uranyl in chloride-bearing aqueous solutions at high T/P using synchrotron X-ray absorption spectroscopy (XAS) and Raman spectroscopy, combined with the hydrothermal diamond anvil technique. To meet the safety requirements at synchrotron facilities, a multi-layer containment was developed to surround the HDAC for in situ XAS experiments. In addition, density functional theory was employed to calculate the Gibbs free energies of various possible aqueous complexes at high T/P. Our results demonstrate that the lowly or neutrally charged uranyl-chloride species become predominant at hydrothermal conditions.