# Re-visiting Hydrothermal Speciation of Uranium.

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Hydrothermal processes define formation controls of many types of uranium ore deposits. Understanding these processes is also essential for safe operation of nuclear reactors, and for assessment of risks associated with disposal of nuclear waste to geological repositories. Complexation of Uranium in aqueous solutions is currently pretty well mapped at ambient conditions. In contrast, when it comes to hydrothermal conditions, experimental information for most of the U-bearing species is extremely sparse, and existing thermodynamic models attempting to predict its behavior in hydrothermal systems have largely relied on extrapolations from low-temperature data.

To fill this knowledge gap, we have undertaken a research program aimed at experimental determination of the stability of the main aqueous species of U at hydrothermal conditions. Here we present the first sets of data on the speciation of U(IV) and U(VI) in chloride and sulfate-bearing solutions, obtained at temperatures up to 350 ºC. Our data suggest predominance of neutrally charged species at elevated temperatures and aqueous conditions (ligand concentrations). The results also suggest significant increases in the contribution of U(IV) to the balance of dissolved U at temperatures above 250 ºC. In contrast to the common understanding of aqueous chemistry of U, it was found that at these temperatures solubility of U in acidic solutions containing NaCl (sea water) in some cases can be higher at reducing conditions than that at oxidizing conditions. Similarly, it has been demonstrated that the stability of U(IV)-sulfate complexes at $T > 200$ ºC is several orders of magnitude greater than that predicted based on extrapolations from ambient temperature data and can even reach ppm levels in solutions having near-neutral pH and 0.1 m total concentrations of sulfate.