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ORAL PRESENTATION - Coupling actinide speciation and thermodynamics: Neptunium(VI) solubility and speciation in alkaline NaCl solutions

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In order to assess the long-term safety of a nuclear waste repository it is essential to derive robust predictions of radionuclide solubility and speciation. Based upon correct and reliable chemical models it is possible to derive comprehensive sets of thermodynamic data and quantitatively analyse radionuclide behaviour for different scenarios. Coupling modern actinide speciation tools and conventional thermodynamic approaches offers important advantages for developing comprehensive quantitative geochemical models. The use of advanced spectroscopic tools in actinide chemistry, one main driving force for aqueous actinide chemistry over the last decade, yields detailed molecular level information on chemical speciation and aqueous and solid phase characteristics. Conventional solubility studies allow deriving robust solubility limits and information on macroscopic thermodynamic properties like solubility products, complex formation constants or ion-interaction parameters. Improved spectroscopic information on actinide speciation will consequently result in improved chemical models and thus contribute to improved thermodynamic descriptions and geochemical model predictions.

In recent studies of Np(VI) solubility in alkaline NaCl solutions we have successfully combined advanced spectroscopic studies with information from solubility studies and chemical thermodynamics. Using XAFS-techniques, we were able to derive stability fields for Np(VI) in aqueous solutions and correlate this to data derived from conventional Eh and pH measurements. Having established the relevance of Np(VI) we have proceeded to perform integral solubility studies in dilute to concentrated NaCl solutions over the entire alkaline pH range. The studies show a distinct dependence of the Np(VI) solubility on pH and ionic strength conditions and indicate significant Np(VI) retention over a large set of chemical boundary conditions. By analysing the solubility data and comparing to the previously investigated chemically analogue U(VI) system, we were able to derive a comprehensive thermodynamic model for the system Np(VI)-Na+-H+-OH−Cl−H2O at 25℃ not available before.

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