Contribution ID: **33** Type: **Oral**

Redox chemistry, solubility and hydrolysis of Np: XAFS contribution to thermodynamics

Tuesday 20 May 2014 12:10 (35 minutes)

Actinide aqueous chemistry is a relevant, multifold and challenging research field of inorganic chemistry. In the context of nuclear waste disposal, actinides arise as potentially relevant dose contributors in the long term. Because of their specific electronic configuration, several oxidation states of actinides (+III to +VII) can exist in aqueous solution. This imposes a differential chemical behaviour as a function of the boundary redox conditions, which is of special concern for the safety case of repositories for nuclear waste disposal. In this framework, it is essential to continuously reduce experimental and systematic uncertainties, fill existing gaps in thermodynamic databases and improve knowledge and available data for more complex and relevant (geo)chemical systems.
This presentation focuses on neptunium as a relevant and challenging case of actinide science and good example of the experimental and conceptual approaches employed at KIT–INE for investigating actinide aquatic chemistry and thermodynamics. Based upon experimental studies characterizing neptunium redox transformation processes, separate series of solubility experiments were performed involving Np(IV), Np(V), Np(VI) and Np(VII) in dilute to concentrated saline systems. Investigations with other actinides (Th, U, Pu) are discussed in order to support the interpretation of the Np studies. In this context, XAFS techniques are envisaged as key tools complementing solubility experiments and contributing to gain insight on the chemical models governing the investigated systems. Selected examples will highlight the existing synergies between solubility studies and XAFS techniques in the development of thermodynamic models:
- Np(IV) and Th(IV) solubility and hydrolysis in CaCl2 solutions
- Np(V) solubility in NaCl solutions: formation of ternary Na–Np(V)–OH solid phases
- Redox chemistry of Np(V/VI) under near-neutral to hyperalkaline pH conditions
- Np(VI) solubility in alkaline NaCl media: analogies with U(VI) and Pu(VI)
- Formation of Np(VII) under hyperalkaline oxidizing conditions

All studies included in this presentation aim at deriving comprehensive thermodynamic models of the investigated systems, which can be implemented in thermodynamic databases for geochemical model calculations.

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Session Classification: Solution and Coordination Chemistry of the Actinides

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