

Effect of borate on An(III/IV/V and VI) solubility in dilute to concentrated NaCl, CaCl₂ and MgCl₂ solutions

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The assessment of the long term safety of a nuclear waste repository requires reliable information about the chemistry and migration behavior of actinides. Boron may be present in the repositories for radioactive waste as a component of the emplaced waste. In repositories in rock-salt formations, boron can further be a component of the intruding brine solutions.

In the present work, the interaction of Nd(III) (as chemical analogue of An(III)), Th(IV), Pu(IV), Np(V) and U(VI) with borate in dilute to concentrated alkaline NaCl, MgCl₂ and CaCl₂ solutions was studied at $22 \pm 2^\circ\text{C}$ by a comprehensive series of solubility experiments complemented with XRD and XPS measurements. No significant increase in Nd(III) solubility occurred in any of the investigated salt systems in the presence of $[\text{B}]_{\text{tot}} \leq 0.4 \text{ M}$, compared to borate-free systems. On the contrary, a significant decrease in Nd(III) concentration was observed at $\text{pH}_c \leq 9$ in NaCl and MgCl₂ systems with $[\text{B}]_{\text{tot}} \geq 0.16 \text{ M}$. This observation, together with a clear change in the slope of the solubility curve and the further confirmation by XPS analyses indicated the transformation of Nd(OH)₃(am) into a so far unknown Nd(III)-borate solid phase. The solubility of Th(OH)₄(am) remains largely unaffected by borate within $7 \leq \text{pH}_c \leq 11$ in dilute to concentrated NaCl and MgCl₂ solutions. Additional experiments with PuO₂(am) under similar conditions are currently under way. No significant increase in the solubility of NpO₂OH(am) in NaCl and MgCl₂ solutions with $0.04 \text{ M} \leq [\text{B}]_{\text{tot}} \leq 0.16 \text{ M}$ is observed in first scoping experiments although minor effects at present cannot be ruled out. Additional experiments are ongoing to further assess the Np(V) and also U(VI) case (solubility studies with UO₃·2H₂O(am) solid phase are being started) in order to establish robust solubility data and analyze possible transformation processes into less soluble borate bearing actinide phases.