Solid solution – aqueous solution systems and the safety case for nuclear waste disposal

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Groundwater is the principal transport medium facilitating the migration of radionuclides from a deep geological repository for nuclear wastes into the geo-/biosphere. The mobility of radionuclides released from the wastes into the repository near- and far-field is controlled by various processes such as sorption onto minerals and colloids (e.g. by surface complexation or ion exchange), precipitation/ dissolution of solid phases, as well as entrapment in, or solid solution formation with other minerals. However, the aspect of solid-solution thermo-dynamics and the effects of solid-solution formation on radionuclide solubility and mobility are considered only rarely in specific cases in long term safety assessments for nuclear waste repository systems at present.

Here, we present and discuss the development and application of thermodynamic models for solid solution – aqueous solution systems and their relevance to nuclear waste management and safety assessments for deep geological repositories, emphasizing the significance of complementary experimental and state of the art computational approaches (e.g. atomistic modelling). The thermodynmics of mixing of the binary $(Ra,Ba)SO_4$ as well as the ternary $(Ra,Ba,Sr)SO_4$ systems were derived from first principles calculations and verified experimentally in long-term (3 years) batch-type Ra uptake experiments. A detailed characterisation (TOF-SIMS, SEM, FIB/TEM) of the Ra-containing solid solutions regarding chemical homogeneity and microstructure/nano porosity indicate a signifant role of these properties in controlling Ra uptake kinetics.

Using the uptake of Ra by barite as an example, it is demonstrated that an improved mechanistic understanding of solid solution thermodynamics and its consideration in performance assessments leads to a more realistic and scientifically corroborated picture on release, solubility, and subsequent migration of safety relevant radionuclides in the repository near- and far-field.