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Editorial: Solubility phenomena in the context of nuclear waste disposal

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Editorial on the Research Topic
Solubility phenomena in the context of nuclear waste disposal

Safety concepts regarding the disposal of nuclear waste in underground repositories generally rely on a combination of engineered and geological barriers that minimize the potential release of radionuclides from the containment-providing rock zone and transport through the biosphere. The presence of water (e.g., groundwater and pore water of repository rocks), however, can alter the engineered barrier system, dissolve radionuclides, and facilitate radionuclide transport that, over millennia, may allow small fractions of water-soluble radionuclides to permeate to the biosphere. Thus, while barrier systems aim to prevent or hinder water from contacting the waste, the possible intrusion of aqueous solutions must be considered for several safety case scenarios impacted by the long-term evolution of a repository. Dissolution, precipitation, and solubility phenomena thus arise as important processes controlling the chemical behavior of radionuclides and other key materials relevant to such repositories and their safety assessment. The solubility and aqueous speciation of radionuclides are of particular interest as they provide upper limits on water-transportable radionuclide concentrations. For many radionuclides, solubility limits are reached only after release from the disposed waste products close to the disposal locations, where the expected maximal concentrations are highest. It is then often the solubility of a secondary phase, precipitated after the dissolution of a primary phase in the waste matrix, that controls the maximum transportable radionuclide concentration close to the disposal location.

Solubility phenomena of radioactive and other gases in water provide important constraints on gas transport and pressure build-up. Moreover, due to their thermodynamic foundation, solubility assessments offer insight into time-independent constraints (e.g., maximum concentrations of radionuclides or gases in a fluid phase) on the evolution of the disposal system. Particularly important in this regard is the geochemical modeling of radionuclide behavior in the engineered barrier system as well as in natural aquatic systems along the transport path of radionuclides to the biosphere. Despite its simplicity, the concept of solubility is difficult to apply in natural water systems as various water constituents will influence solubility, which may vary over the very long time considered in safety assessments of disposal. For instance, the interaction of natural waters with the components of the engineered barrier system surrounding the disposed waste products will influence solution pH, redox potentials, or ligand concentrations. Information obtained from studies evaluating radionuclide behavior provides valuable inputs that appropriately constrain safety analyses of nuclear waste disposal.

In this series of articles, solubility phenomena related to different domains and key components in the context of nuclear waste disposal are presented. Several of these contributions provide insight into the chemical, thermodynamic, and (SIT, Pitzer) activity models describing the investigated systems. This emphasizes the usefulness of solubility studies for the determination of thermodynamic properties, which can be implemented in thermodynamic databases (e.g., NEA-TDB, THEREDA, ThermoChimie, JAEA-TDB, or PSI-Nagra, among others) and further used in geochemical calculations of relevance for nuclear waste disposal.

Three contributions are dedicated to the solubility of key radionuclides, i.e., Pu, U, and ^{99}Tc. Cho et al. investigate the dissolution of PuO_2(cr) in natural waters under atmospheric conditions at T = 25°C and 60°C. By comparing experimental results with geochemical calculations, the authors explain the observed solubility behavior based on the oxidative dissolution of PuO_2(am, hyd), highlighting the key role of Pu(IV) colloids. Grambow et al. investigate the formation of UO_2(s) from aqueous solutions containing U(VI) and U(IV). Combining solubility experiments with thorough solid phase characterization and thermodynamic calculations, the authors conclude that their observations can be explained by three main effects: (i) oxidation of UO_2(s) to U_4O_9(s), (ii) the effect of particle size, and/or (iii) the presence of oxygen traces as low as $1{\cdot}10^{-8}$ atm. The mini-review by Singh et al. discusses some key contributions in the literature dealing with the solubility of ^{99}Tc and Re in different waste forms, i.e., glass, cement, ceramic, and geopolymers. The concluding remarks by the authors summarize future challenges that need to be addressed to minimize the solubilization of ^{99}Tc from the designed waste forms in different environments.

Sorel phases of the general formula xMg(OH)$_2$·yMgCl$_2$·zH$_2$O (x-y-z phases) are considered in the construction of geotechnical barriers in repositories in rock-salt geological formations. The comprehensive studies by Pannach et al. and Freyer et al. provide an extensive experimental basis (solubility data, solid phase characterization) to derive the thermodynamic and (Pitzer) activity models for the Sorel phases and $Mg(OH)_2$ in the Na-Mg-Cl-OH-H_2O system. The 3-1-8 Sorel phase and $Mg(OH)_2$ are identified as the stable solid phases, while the 5-1-8 Sorel phase is metastable. Bok et al. critically review the available solubility data of O_2 in water and saline solutions at temperatures up to 373 K. As a main outcome of this exercise, the authors provide a selection of thermodynamic data for dissolved oxygen O_2(aq), a temperature-dependent Henry's law constant, and Pitzer coefficients for the calculation of oxygen solubility in concentrated salt solutions of the system Na-K-H-Ca-Mg-Cl-SO_4-CO_3-PO_4-OH-H_2O(l), together with their validity range in terms of temperature (273–318 K), ionic strength (≤5 mol kg^{-1}) and O_2 partial pressure (≤101.325 kPa).

The last contribution to this Research Topic deals with beryllium as a chemotoxic element expected in specific waste streams of radioactive waste. Using a combination of solubility experiments, solid phase characterization, and molecular dynamics calculations, Cevirim-Papaioannou et al. derive thermodynamic and (SIT) activity models for the solubility and hydrolysis of Be(II) in dilute to concentrated $CaCl_2$ systems. These models are then used to predict the speciation of beryllium in cementitious environments, such as those considered in repository concepts for the disposal of low- and intermediate-level waste.

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Conflict of interest

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