Solubility of Ra in a radioactive repository environment

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Background

The mobility of ^{226}Ra, an important radionuclide in high-level radioactive waste, will mainly depend on solubility controls within the repository. In safety analyses, solubilities are generally calculated for pure radionuclide phases (e.g. Nagra, 1994). However, geochemical evidence indicates that many radionuclides are incorporated as trace elements in suitable host phases. Considering pure phase solubility conservatively overestimates radionuclide concentrations and consequently their mobility. Radium is known to form solid solutions with sulfates, particularly with barite. By applying solid solution thermodynamics we show that incorporation of radium in Ba-rich sulfates will strongly reduce Ra equilibrium concentrations in the aqueous phase.

Discussion of results

Re-evaluation of laboratory investigations of radium incorporation in barite (Germann, 1921; Doerner and Hoskins, 1925) with the methods of modern solid solution thermodynamics, indicates that $RaSO_4$ and $BaSO_4$ mix nearly ideally. We therefore adopted the ideal solid solution model for our subsequent calculations.

The Swiss concept for a high-level repository foresees emplacement of the waste in tunnels sealed with compacted bentonite. Bentonite contains about 100-300 ppm of Ba and Sr, which are assumed to interact with Ra and form sulfate solid solutions via recrystallisation. Field data also indicate saturation of the groundwater with gypsum, celestite and barite.

Our calculations show that Ra solubility decreases from 1×10^{-7} to 2×10^{-11} M when equilibrium with a binary ideal $(Ba,Ra)SO_4$ solid solution is considered instead of pure $RaSO_4$, in spite of the tiny amounts of barite involved. Considering formation of a quaternary $(Ba,Sr,Ca,Ra)SO_4$ solid solution further decreases the Ra equilibrium concentration to 8×10^{-12} M.

Conclusions

Compared to pure $RaSO_4$ solubility, solid solution formation with even small amounts of barite will reduce dissolved Ra by 3-4 orders of magnitude in the present repository design. This substantially reduces calculated fluxes to the geosphere and improves repository performance.

References

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Novel approaches to the cultivation of marine sediment bacteria

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Only a small percentage of the prokaryotes present in marine sediments can be cultured in traditional media. Most sediment bacteria are classified as 'unculturable', although they have thrived in their natural environments. Our approach involves some new aspects designed to increase the cultivation success. These include

(1) Use of non-selective media, which allow a large variety of microorganisms to grow. These media contain mixtures of monomers, polymers, refractile substrates or marine sediment extract.

(2) Application of low substrate concentrations (100 µM or less).

(3) Addition of particles (e.g. FeS precipitates) to liquid media.

(4) Promotion of syntrophic interactions by addition of background bacteria to MPN series.

In our media the bacteria cannot grow to high density. Therefore growth analysis has to make use of sensitive detection techniques, as analyis of radiotracers (e.g., formation of ^{35}S-sulfide from radiolabelled sulfate) or counting of grown bacteria after staining with fluorescent DNA stains, FISH or other molecular biological techniques. First results of cultivation experiments with sediments from 5000-year-old North Sea sediments, up to 300 000-year-old Mediterranean sediments and sediments obtained during ODP Leg 201 from the Equatorial Pacific and Peru Margin with an age of several million years will be discussed.