Geochemically based source term assessment for the Asse salt mine – modelling and experimental results

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Safe disposal of radioactive waste relies on multi-barrier systems. Part of these systems are engineered barriers which provide plugging of the openings and chemical buffering in the case of groundwater (brine) intrusion. The source term for radionuclide release is based on models, which consider reactions in the specific geochemical milieu defined by the composition of the solution, host rock, waste form, container, buffer and backfill materials. Corrosion of the waste changes the solution compositions, resulting in significant changes of radionuclide solubilities (Metz et al., 2002).

Low- and intermediate-level radioactive waste was emplaced in the Asse salt mine from 1972 to 1976. The application of buffer and backfill materials is discussed for closure of the mine. The selection of these materials is based on geochemical modelling taking into account the corrosion of cemented waste forms, degradation of organic matter and the presence of different types of complexing ligands in salt brine. The modelling led to the conclusion that only cement, $Mg(OH)_2$-based material and crushed salt should be used as buffer and backfill materials. According to the modelling results, a Mg-based buffer material provides a long-term stable and robust geochemical milieu. The radionuclide source term of each emplacement room was calculated using the actual inventories for the studied actinides and fission products. The calculated thermodynamic solubilities of Th, Pu and U are lower than their inventory based concentrations. Maximum concentrations of fission products and other actinides are limited by their inventories.

Laboratory experiments were undertaken to verify the modelling results. A good agreement was found for experimental and calculated results regarding corrosion of cemented waste forms and Mg-based buffer in the relevant salt brines. Moreover, the experiments demonstrate the functionality of the Mg-based material to buffer the *pH* and to limit the carbonate concentration in solution, which may increase due to organic matter degradation. The carbonate concentration constraints concentration of actinides by complexation. As an example, when Pu(IV)-carbonate solution was added to brines in the presence of Mg-based buffer, the observed [Pu] was below 10^{-10} mol (kg H_2O)$^{-1}$. In brines without the Mg-based buffer, [Pu] was significantly higher due to carbonate complexation.

Reference:

Metz, V., Kienzler, B. and Schüßler, W (2002): J. Contam. Hydrol. (in press).

SHRIMP RG Measurements of ^{87}Sr/^{86}Sr in Granitoid Calcites; Implications for Calcite Petrogenesis

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Strontium isotope ratios in calcite from granitoid rocks were determined with SHRIMP RG: The Rio Blanco Quartz Diorite (Tertiary) from Puerto Rico, Granodiorite of Arch Rock (Cretaceous) from Yosemite, California, Panola Granodiorite (Missippian/Pennsylvanian) from Panola, Georgia , and Silver Plume Granite (Precambrian) from Loch Vale, Colorado. Calcite content of the granitoids does not correlate with whole rock CaO concentration. Calcite concentrations range from 320 mg kg $^{-1}$ to 3320 mg kg$^{-1.}$ These granitoids have been previously described in White et al. (1999a and 1999b).

Calcite occurs in several morphologies within the granitoids: as disseminated interstitial grains, as fracture fillings and as replacement of plagioclase feldspars. In general the ^{87}Sr/^{86}Sr ratio of interstitial calcite is similar to that of the primary K-feldspar and plagioclase feldspar of the granitoids which was previously determined by TIMS.

^{87}Sr/^{86}Sr ratios were not determined for calcites with high Rb. The Rb in these calcites are likely due to the inclusion of micaceous phases. Calcite that has a ^{87}Sr/^{86}Sr ratio similar to that of the primary feldspar grains may indicate that calcite crystallized early in the history of the granitoid. Calcite with a ^{87}Sr/^{86}Sr ratio higher than the primary feldspar is likely of later origin.

References

White, A.F., Bullen, T.D., Vivit, D. V., Schulz, M.S., and Clow, D.W., (1999), Geochimica et Cosmochimica Acta, 63, 13/14.

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