Effects of Salt Formation on the Chemical Environment of a High-Level Nuclear Waste Repository

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In the absence of mechanical disruption, aqueous corrosion will be the primary degradation process limiting the life of waste packages (WPs) and drip shields (DSs) in the proposed high-level nuclear waste repository at Yucca Mountain (YM), Nevada. In a hot repository setting, evaporation processes would increase the aqueous concentrations of Cl^-, F^-, and other ions and lead to the accumulation of multicomponent salt deposits on DS and WP surfaces. Inorganic salts are generally hygroscopic and will absorb moisture from humid air, generating small volumes of potentially corrosive brines. In this study, thermodynamic modeling was done to evaluate the effect of salt formation on the chemical environment of DSs and WPs at the proposed repository. The deliquescence behavior of salt mixtures was calculated using thermodynamic considerations and compared with that of single salts. Also, the chemical evolution of YM porewaters was evaluated to determine the types of brines and salt mixtures that could form by evaporation processes. The results are consistent with published data that show the deliquescence points of salt mixtures are lower than that of individual salts (Figure 1). Mixtures containing Mg and Ca salts have much lower deliquescence points than pure $NaNO_3$, which is used by the U.S. Department of Energy (DOE) to bound the deliquescence point of salt mixtures. Formation of Mg and Ca salts could lead to earlier initiation of aqueous corrosion than assumed by the DOE in its performance assessment modeling. Such mixtures can be formed by evaporation of waters with compositions similar to some Yucca Mountain porewaters.

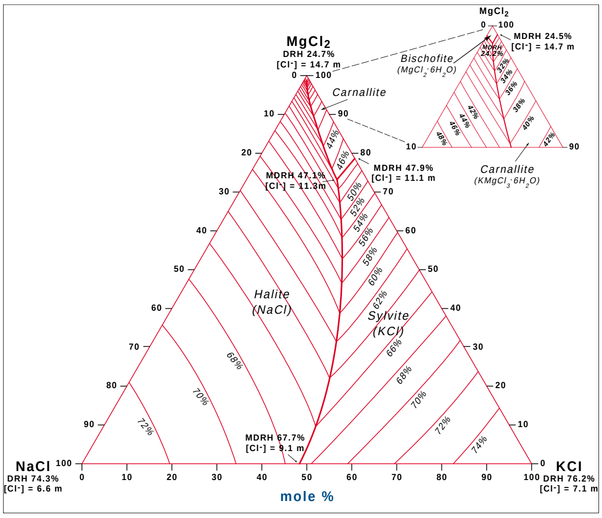

Figure 1. Calculated mutual deliquescence relative humidity, MDRH (%) [contour lines] of salt mixtures in the system $NaCl\text{-}KCl\text{-}MgCl_2$ at 90 °C. The solutions are saturated with the indicated solids.

Geochemistry of groundwater from the Morais Massif (NE.Portugal)

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Objectives

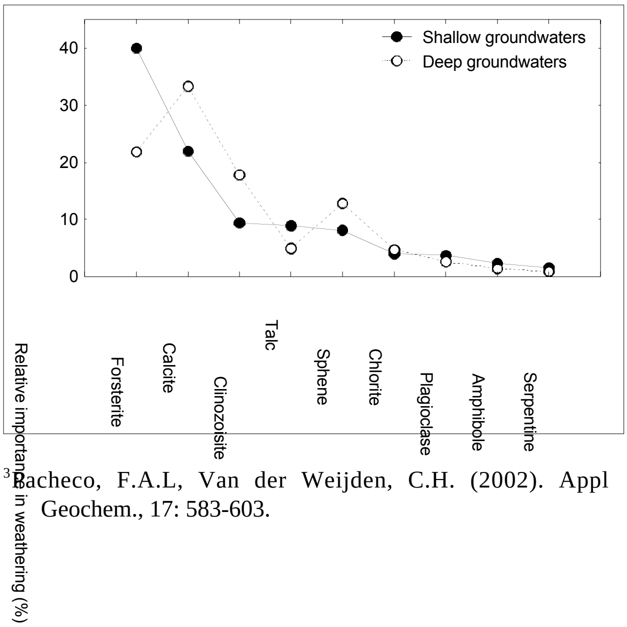
In Spring, Summer and Winter, samples from springs and 60 to 100 m depth wells were collected within amphibolites (AMP) and serpentinites (SER). The main objective is to compare the relative contribution by rock-forming minerals to the chemistries of shallow and deep groundwaters. Analysis of seasonality in the chemistry of springs was a second goal.

Hydrochemistry

Major cation concentrations and alkalinity are a little higher in Summer than in the wet seasons. Alkalinity in springs within AMP is much lower than within SER (≈1.2 and ≈4.8 mM). Within SER, Mg>>>Ca≈Na>K, and within AMP Ca> Mg>Na>>K. Wells are significantly higher in cations and alkalinity than are springs. The average $\log p_{CO2}$ in springs is significantly higher in Summer (≈ -2) than in Spring (≈ -4) and Winter (≈ -2.5). For wells the values are ≈ -2.5. Values >–3.5 (atmosphere) are typical for soil pCO_2 and indicative of a residual weathering potential.

Water-rock interaction

Using a mole-balance model[3], we related the chemical composition of springs with the weathering of rock-forming minerals; minerals were considered to dissolve congruently (calcite) or to alter incongruently (others) into mixtures of smectites, halloysite and Ti/Fe oxides. We now applied this model to deep groundwater. Calcite weathering is more important than olivine weathering, but as calcite is dissolved faster from the weathering profile than olivine the order is reverse (springs). The same holds for sphene and talc (Fig.).



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[3] Pacheco, F.A.L, Van der Weijden, C.H. (2002). Appl Geochem., 17: 583-603.