

Reactive transport modelling of the interaction between a high pH plume and a fractured marl

J. M. Soler

Paul Scherrer Institut, Waste Management Laboratory, CH-5232
Villigen PSI, Switzerland

In the context of the proposed low and intermediate level radioactive waste repository at Wellenberg (Switzerland), calculations simulating the interaction between hyperalkaline solutions and a fractured marl, at 25°C, have been performed. The aim of these calculations is to evaluate the possible effects of mineral dissolution and precipitation on porosity and permeability changes in such a fractured marl, and their impact on repository performance. Solute transport and chemical reaction are considered in both a high permeability zone (fracture), where advection is important, and the wall rock, where diffusion is the dominant transport mechanism. The mineral reactions are promoted by the interaction between hyperalkaline solutions derived from the degradation of cement (a major component of the engineered barrier system in the repository) and the host rock.

The model

A modified version of the GIMRT software package (Steeffel and Yabusaki, 1996) has been used for the reactive transport calculations. Both diffusive/dispersive and advective solute transport are taken into

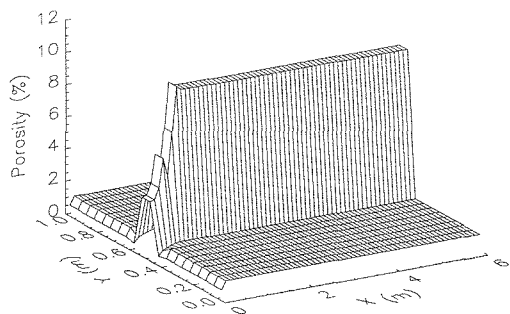


FIG. 1. Porosity distribution in the system at $t = 10000$ y. The high permeability zone (fracture) crosses the domain from left to right. Notice the reduction of porosity at the inlet boundary ($x = 0$), and especially at the fracture inlet.

account in the calculations. Mineral reactions are described by kinetic rate laws. The primary minerals that make up the rock in the simulations are calcite, dolomite, quartz, and muscovite (as a surrogate for illite, illite/smectite, and chlorite). The reaction rates for the primary minerals are based on experimentally-determined rates published in the literature, and geometric considerations regarding mineral surface areas. Relatively fast rates for the secondary minerals have been used, so the results resemble the local equilibrium solution for these minerals, at the time scales of interest (thousands to tens of thousands of years).

The composition of the solutions entering the domain is derived from modelling studies of the degradation of cement under the conditions at the proposed underground repository at Wellenberg (Neall, 1994). These are high pH solutions (pH 12.5–13.5) at equilibrium with calcite and under-

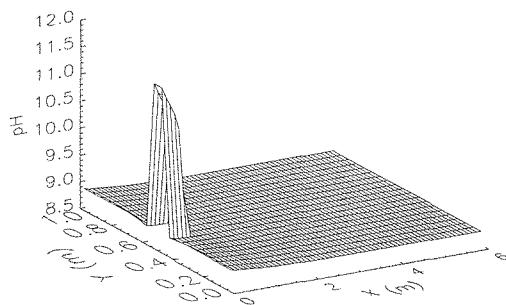


FIG. 2. pH at $t = 10000$ y. The pH diminishes from 12.5 to a value about 8.8 in a distance of less than 1 m along the fracture. Notice also that since the discretization of the domain is based on a cell-centered finite difference scheme, the pH on the left-hand-side of the plot is less than 12.5 (the pH of the incoming solution). The first node in the x direction corresponds to the center of the first cell, so the fluid has already changed its initial composition, due to chemical reaction, at that point in space.

TABLE 1. Initial mineralogy and porosity of the marl, as vol%, for both the wall rock and the high permeability zone (fracture)

	Wall rock	Fracture
Calcite	50	45
Dolomite	6	5
Quartz	15	14
Muscovite	28	26
Porosity	1	10

saturated with respect to the other primary minerals.

The fluid flow system under consideration is a two-dimensional porous medium (marl, 1% porosity), with a high permeability zone simulating a fracture (10% porosity) crossing the domain. The fracture starts right at the flow inlet boundary, which is equivalent to the assumption of a worst case scenario, with a high conductive feature starting right at the repository. The dimensions of the domain are 6 m per 1 m, and the fracture width is 10 cm (Fig. 1). The boundary conditions of the flow domain are no flow conditions at the top and bottom boundaries, and fixed hydraulic heads on the left and right boundaries, so there is an overall regional gradient of 0.4 m/m (Nagra, 1994) driving flow from left to right. At $t = 0$ Darcy velocities are of the order of 10^{-2} m/y along the high permeability zone, and 10^{-5} m/y in the rest of the domain. The fluid flow field is updated during the course of the simulations by solving the equation of conservation of fluid mass with the updated porosities and permeabilities. Permeabilities are updated according to Kozeny's equation.

Results and discussion

Rock and fluid compositions are given in Tables 1 and 2. Initially, the fluid is at equilibrium with the primary minerals in the marl. The incoming solution is at equilibrium with calcite but undersaturated with respect to the other primary minerals.

The dissolution of dolomite at the fracture inlet causes the precipitation of calcite. Quartz and muscovite also dissolve. Regarding the Mg-containing secondary minerals, there is precipitation

Table 2: Composition (molality) of the solution entering the flow domain (incoming) and the initial fluid in the pores of the marl

	Incoming	Initial
pH	12.50	7.50
Al	1.32×10^{-4}	6.50×10^{-9}
Si	1.40×10^{-5}	1.06×10^{-4}
Na	4.61×10^{-1}	4.61×10^{-1}
K	7.10×10^{-4}	7.10×10^{-4}
Ca	3.51×10^{-2}	5.05×10^{-3}
Mg	1.40×10^{-7}	2.65×10^{-4}
Carbonate	9.64×10^{-6}	3.39×10^{-3}
Sulphate	1.30×10^{-5}	1.07×10^{-5}
Cl	4.70×10^{-1}	4.69×10^{-1}

of brucite right at the fracture inlet, and sepiolite a few cm further down the fracture. With time (after a few thousands of years), brucite dissolves and sepiolite is the dominant Mg-containing mineral. Zeolites and tobermorite also precipitate close to the inlet.

The precipitation of calcite and the other secondary minerals causes a reduction of porosity (sealing) at the fracture inlet (Fig. 1). Also, the pH of the solution goes down to less alkaline values (Fig. 2). The reduction of porosity would be highly beneficial for repository performance, since a sealing of the fracture inlet would mean that the solutions coming from the repository and potentially carrying radionuclides in solution would have to flow through low conductive rock before they were able to get to the higher conductive features.

Acknowledgements

Partial financial support by the Swiss National Cooperative for the Disposal of Radioactive Waste (Nagra), and the helpful comments by Andreas Jakob, are gratefully acknowledged.

References

- Nagra (1994) Nagra Technical Report 94-06.
- Neall F. B. (1994) Paul Scherrer Institut Report 94-18.
- Steeffel, C.I. and Yabusaki, S.B. (1996) Pacific Northwest National Laboratory Report PNL-11166.