

SIMULATION OF THE DEGRADATION OF A CONCRETE/CLAY INTERFACE: INFLUENCE OF TEMPERATURE, UNSATURATED CONDITIONS AND POROSITY VARIATIONS

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Introduction

For long-lived intermediate-level radioactive waste, the use of concrete as engineering barrier and Callovian-Oxfordian clay as geological barrier at a depth of 500 m is considered in the French disposal concept (ANDRA, 2005). Upon emplacement, initially unsaturated concrete is expected to experience coupled processes involving heating, re-saturation with groundwater from the clay formation, gas exchanges and geochemical reactions. After an early period of re-saturation, solute transport is supposed to be diffusion-controlled because of the extremely low permeability of the two media. These coupled processes may lead to changes in the porosity of the concrete or clay barriers. In the present paper, a fully coupled Thermo-Hydro-Chemical (THC) response of a two-phase (gas and solution) mass-transfer model was evaluated and tested by a sensitivity analysis. This study is an extension of a previous model applied to an isothermal and fully saturated concrete/clay interface (Burnol et al, 2005); it investigated the coupled effect of temperature and unsaturated conditions assuming no production of H₂(g). The system was simulated for a 2000-year period, which covers the most predominant thermal perturbation.

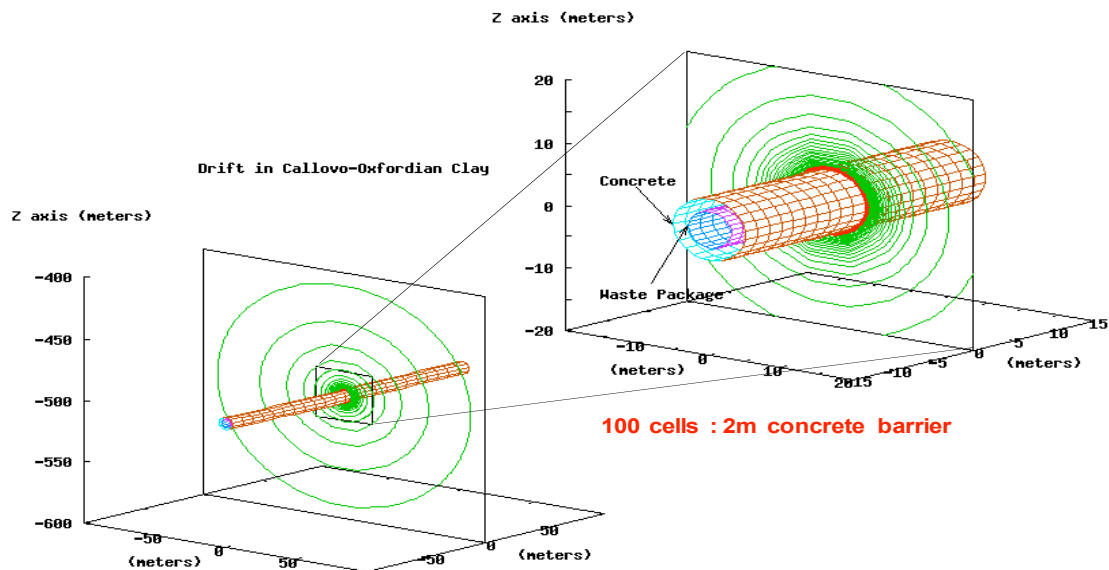


Figure 1: 1D radial grid used by the simulation of degradation of the concrete/clay interface around long-lived intermediate-level radioactive waste in a deep repository in clay (500m)

Model description

Simulations were performed using the two-phase non-isothermal reactive transport code TOUGHREACT (Xu et al., 2001). This code was developed by introducing reactive geochemistry into the framework of TOUGH2 V2 (Pruess et al., 1999). The modeled system represents a simplified 1D radial geometry which includes waste canister packages with a radius of 3 m, surrounded by a 2-m wide concrete barrier and by a 100-m portion of Callovian-Oxfordian clay (Figure 1). The dimensioning, the

power of the canisters at the time of deposition (460 W after 10 years), the thermal properties of the engineered barrier were factors that controlled the temperature. The initial conditions in the concrete barrier were assumed to be 50°C temperature, 1 atm pressure and 70% gas saturation. In clay, the initial conditions were assumed to be 25°C, 5 MPa (representing hydrostatic conditions at a disposal depth of 500 m) and fully water saturated, which were the same as the boundary conditions. Both constitutive relationships (capillary pressure-saturation and relative permeability-saturation) for the two-phase flow were supposed to follow the Van Genuchten/Mualem formulations.

The chemical system includes 12 aqueous components, 32 minerals and 1 gas. All the dissolution and precipitation reactions were assumed to proceed at thermodynamic equilibrium because of the considered large time scale. Inclusion of kinetics rate constants in the modeling was also studied in an other presentation (Marty et al., Lille 2007). Fixed geochemical conditions were imposed at the outer model boundary. The coupled transport and geochemical equations were solved using the Sequential Non-Iterative method. The base-case simulation was then perturbed by changing key parameters values or by switching on/off a key process in order to evaluate the uncertainty of predictions.

Base-case results and uncertainty analysis

A maximum temperature around 70°C at the canister-concrete interface was reached about 10 years after waste emplacement, this result is consistent with the ANDRA concept for B5.2 waste. At the concrete-clay interface, the maximum temperature was about 65°C. After 2000 years, the simulated temperatures were less than 30°C in all the modeled system.

The clay pore water will be driven to the concrete under both hydraulic pressure and capillary pressure gradients. This re-saturation time depended on many parameters (permeability, capillary pressure of concrete/clay) and affected the possibility of gas phase diffusive transport. During the early period of re-saturation of concrete, the high CO₂ partial pressure in surrounding clay pore has been redistributed by the gas phase transport. The high pH of concrete caused the solubilization of CO₂(g), which caused in turn the following carbonation of the 2-m wide concrete barrier by dissolution of portlandite and precipitation of calcite: $\text{CO}_3^{2-} + \text{Ca}(\text{OH})_2(\text{s}) \rightarrow \text{CaCO}_3(\text{s}) + 2 \text{OH}^-$. The process of degassing of CO₂(g) and therefore the carbonation process were only slightly increased by the heating due to the waste packages. The simulation showed also that this carbonation in the concrete will be limited by the re-saturation.

The major porosity change occurred in response to the dissolution/precipitation processes at the concrete-clay interface. There was even an occlusion of the porosity, i.e. a volume fraction change equal to the porosity. A part of the change in porosity was caused for example by the precipitation of high molar volume phases (like torbermorite). Further reactive transport relationships were investigated and a set of reactive transport parameters and their uncertainties was proposed.

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