

Background

In Belgium, Boom Clay is studied as a reference host formation for geological disposal of radioactive waste. The current reference design of the engineered barrier system ('supercontainer design') plans to use a considerable amount of cementitious materials as construction material, buffer and backfill. Interactions between the alkaline pore fluids from the concrete engineered barriers with the Boom Clay may change the retention properties of the Boom Clay in the vicinity of the interface with the engineered barriers.

Objectives

The objective of this work is to assess the extent of an alkaline disturbed zone in the Boom Clay for a period up to 10^5 years using reactive-diffusion model simulations. The sensitivity of different model parameters and of (major) model assumptions regarding the Boom Clay mineralogy and the choice of the secondary phases is assessed

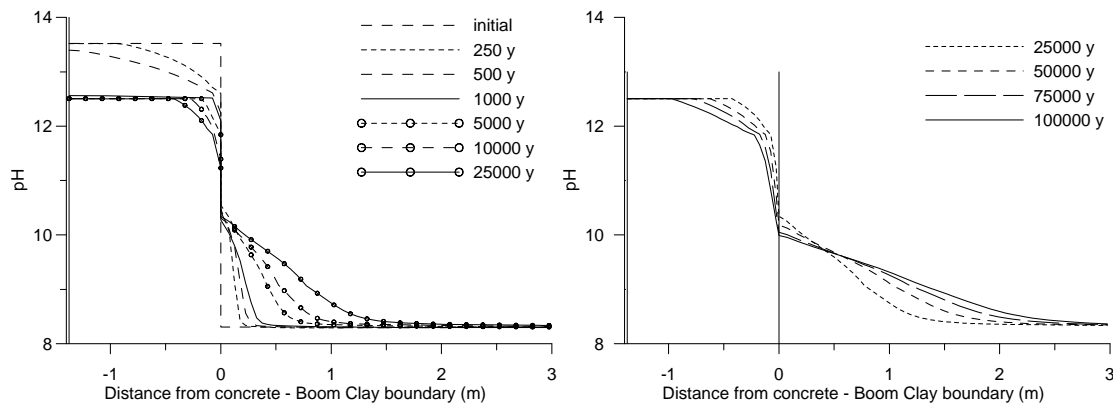
Principal results

The diffusion of an alkaline plume is simulated for a period up to 10^5 years with the PHREEQC-2.12 geochemical code (Parkhurst and Appelo, 1999) using the *lnl.dat* database in that code. To account for the mass balance and variations in the composition of the alkaline plume with time, the concrete material in the near field is explicitly included in the model. The concrete material is located between 0.22 m and 1.62 m from the centre of radial simulation geometry. The steel components in the supercontainer design are ignored, and an initial homogeneous concrete material was assumed.

A reference model is defined including five minerals for the Boom Clay (quartz, kaolinite, illite, Na-montmorillonite, and calcite), a pH-independent (clay minerals) and dependent (organic matter) cation exchange complex and surface acidity reactions on the illite and Na-montmorillonite (after Bradbury et al., 2005). All mineralogical reactions are assumed to be in equilibrium. In addition to the reference case, other simulations assess the sensitivity of the capacities of the exchange and surface sites, the initial amount of primary minerals (plus and minus 25%) and the diffusion coefficient in the Boom Clay (five times smaller and larger). In addition, some alternative model formulations for the Boom Clay mineralogy and the choice of the secondary phases are tested.

In all these simulations, there is no feedback of mineral precipitation and dissolution to changes in porosity and, subsequent, diffusion. For example, clogging of the pore space in the concrete due to calcite precipitation will certainly have an effect on the diffusion in and out the concrete. Nevertheless, the current simulations will give an idea of the possible extent of the alkaline plume perturbation in the Boom Clay.

The figure shows the pH evolution in the reference model. Within the first 1000 years, the Na- and K-oxides in the concrete are depleted resulting in a pH decrease from 13.5 to 12.5. Portlandite is completely depleted up to 0.3 and 1.0 m from the concrete – Boom Clay interface after 25 000 and 10^5 years, respectively (pH is smaller than 12.5). At the concrete – Boom Clay interface, a substantial amount of calcite is precipitated (data not shown). Overall, the disturbance of the Boom Clay by the diffusion of an alkaline plume is limited up to 0.4 – 1.2 m from the concrete – Boom Clay boundary after 25 000 years (for different model formulations) and up to 2.0 – 2.5 m after 10^5 years (for the reference case). After 25 000 years, the volume occupied by minerals is increased by a maximum of 15 percent for the reference case (taken into account parameter uncertainty) and of 25 percent for the different model formulations.



pH evolution near the concrete – Boom Clay boundary for the reference model (vertical line indicates the concrete – Boom Clay interface)

Parameters defining the capacities of the cation exchange complexes or the surface acidity sites have no effect on the extent of changes in pH. The initial amount of the primary minerals has a slightly larger effect, especially kaolinite. The most crucial parameter is the diffusion coefficient. Although this parameter is relatively well defined for non-disturbed Boom Clay, it is still an uncertain parameter with respect to the altered Boom Clay by an alkaline plume. The pore diffusion coefficient depends on the porosity of the porous medium and the alkaline plume may change the porosity. However, these effects were not taken into account in the present simulations.

The selection of mineral reactions and sequence is a crucial factor for assessing the spatial extent of the alkaline plume perturbation. Decreasing (e.g., neglecting exchange and surface acidity reactions or allowing primary minerals only to dissolve) or increasing (adding dolomite-dis as a primary mineral) the total buffer capacity in the model increase or decrease the spatial extent of the disturbed zone, respectively. An alkaline plume disturbed zone is most limited if the partial pressure of CO₂ in the Boom Clay is buffered by an assemblage of minerals as proposed by De Craen et al. (2004). In that case, the disturbed zone is limited to 0.4 m after 25 000 y.

Future developments

The conceptual models for the concrete and Boom Clay will be further developed, including e.g. solid-solutions for cement phases as the CSH-phases or the role of the organic matter in pH buffering in the Boom Clay. Additional process to include are the feedback of mineralogical changes on porosity and, consequently, on the diffusion coefficient and the effect of temperature increases and decreases on the cement mineralogy and the interactions between cementitious and Boom Clay pore waters.

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Main references

- Wang, L., Jacques, D., and De Cannière, P. (2007), "Effects of an alkaline plume on the Boom Clay as a potential host formation for geological disposal of radioactive waste", First full draft- v1.1 (for external review), External Report, SCK•CEN-ER-28, October, 2007, Mol, Belgium
- Jacques, D., and L. Wang, 2008, "Modelling the long term interaction of cementitious clay water with Boom Clay". Phys. Chem. Earth (submitted).
- Bradbury, M.H., B. Baeyens, H. Geckeis, and Th. Rabung, 2005, "Sorption of Eu(III)/Cm(III) on Ca-montmorillonite and Na-illite. Part 2: Surface complexation modelling". Geochim. Cosmochim. Acta, 69:5403-5412.
- De Craen, M., L. Wang, M. Van Geet, and H. Moors, 2004, "Geochemistry of Boom Clay pore water at the Mol site". SCK•CEN-BLG-990, 179 p.

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