EVALUATION OF OXIDATION OF BOOM CLAY DURING CONSTRUCTION AND OPERATION OF A RADIOACTIVE WASTE REPOSITORY

Background

Within the EC's 6th framework programme, NF-PRO is an Integrated Project on the key-processes and their couplings in the near-field of a repository for the geological disposal of vitrified high-level radioactive waste and spent fuel. For the construction and operation of a geological repository of radioactive waste, the excavation and ventilation of galleries is necessary. As such, the oxidation of the anaerobic host rock, containing pyrite, is unavoidable. This chemical perturbation can affect the favourable host rock characteristics and influence the engineered barrier system.

Objectives

The aim of this study is to evaluate

- the extent of oxidation in the Boom Clay related to excavation of galleries;
- the evolution of the extent of oxidation during ventilation of galleries.

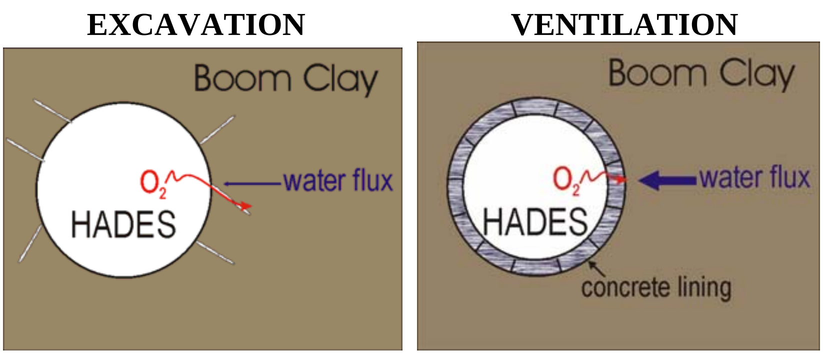
The latter aspect might deliver important input towards design, especially concerning the time to leave galleries open. Some valuable information towards performance assessment can be given as well.

Principal results

Based on experimental and modelling work, a conceptual model is developed. This model explains the oxidation of mainly pyrite within the clay during excavation and ventilation. During excavation, fractures are created in the surrounding host rock to a depth of about 1 m. These are open fractures in contact with air. At that moment an important, instantaneous oxidation of the clay occurs. This oxidation front extends for about 1 m. Within the plastic Boom Clay, sealing of the fractures occurs fast. The oxidation products are then trapped within this first meter around the gallery.

A substantial hydraulic gradient exists around an open gallery and the oxidation products will be redistributed by advective-diffusive transport. Scoping calculations illustrated that the diffusion of the oxidation products further into the undisturbed Boom Clay is negligible even after 50 years and that the majority of the oxidation products are dragged towards the gallery. Consequently, the extent of the perturbed zone remains limited to about 1 m.

During ventilation, the Boom Clay is fully saturated. Thus, the only way to oxidise the clay is by dissolution of oxygen into the pore water and diffusion into the Boom Clay. However, it needs to be reminded that a combined diffusion-advection regime is present. A transport scoping calculation shows that after 20 years of ventilation, an oxygen concentration of about 10^{-6} M is obtained at about 1 m. It should be noted that lower oxygen concentrations would probably already induce oxidation, but on the contrary, the scoping calculation did not take into account the reactivity of Boom Clay with oxygen. Consequently, the extent of 1 m in-diffusion of oxygen into the Boom Clay surrounding the galleries is probably a conservative calculation.

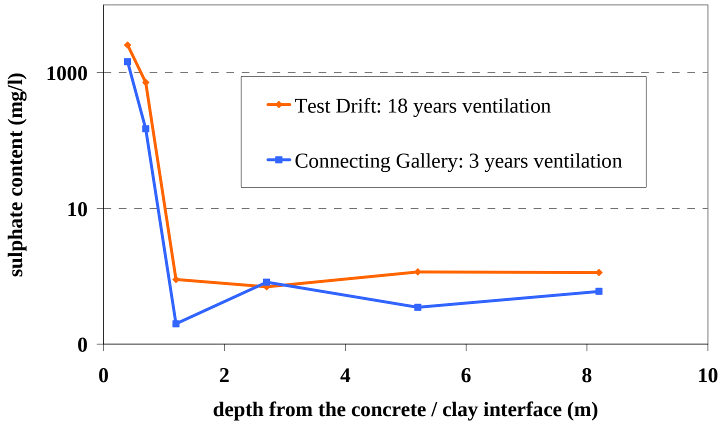


Conceptual model of oxidation around the excavated galleries in Boom Clay. During excavation (left) open fractures are created up to about 1 metre and instantaneous oxidation around the fractures occurs. During ventilation (right) oxygen needs to dissolve in water and diffuse into the Boom Clay, while a large hydraulic gradient is present towards the gallery. Even after 20 years the in-diffusion of oxygen is limited to about 1 metre.

The above mentioned conceptual model is scientifically underpinned with in-situ experiments performed in the underground research laboratory HADES, Mol, Belgium. This laboratory contains a gallery, which was manually excavated in 1987 and has been ventilated during 18 years before installation of the experiment. HADES also contains a newer gallery, industrially excavated in 2002 and only ventilated for 3 years before installation of the experiment. The experiment consists of piezometers in both galleries (old and new) with filters at different depths from the concrete liner / host rock interface. The drillings were the first in HADES to be performed with nitrogen instead of compressed air, in order to avoid any additional oxidation caused by drilling.

The effects of oxidation of Boom Clay are known, based on earlier experimental work and are confirmed within these experiments. Pore water chemistry has a quick response towards oxidation compared to mineralogical changes. The main process occuring is the oxidation of pyrite (FeS_2), resulting in an increase of sulphate in the pore water and a precipitation of iron oxy-hydroxides.

Snapshots of the drilling with compressed nitrogen and installation of multifilter piezometer



Experimental data of sulphate concentrations measured at different filter positions around Test Drift (18 years of ventilation) and Connecting Gallery (3 years of ventilation)

The pore water from the above mentioned in-situ experiment showed an increase in sulphate content for the first two filters located within one metre from the concrete lining / host rock interface. In deeper filters only background concentration of sulphates were measured, indicating the successful use of nitrogen during drilling. Moreover, the pore water in the first metre around the galleries also indicates an increase in major cations and sometimes thiosulphate concentrations, clearly the result of pyrite oxidation. The same observations were made around both galleries (18 years and 3 years of ventilation). Within the galleries, precipitates of several sulphate salts have been analysed, indicating that the water drained towards the gallery is indeed rich in sulphates.

It can be concluded that the extent of the perturbed zone as a result of oxidation is limited and remains limited even during decades of ventilation. The degree of oxidation is most probably correlated with the ventilation time. The change in pore water chemistry is quite pronounced. Oxidation does slightly change the mineralogy of the Boom Clay (mainly the non-clay fraction), but this is not necessarily negative towards radionuclide migration as the neo-formed iron precipitates have a high sorption capacity. Anyhow, the current concept used in performance assessment, namely a reduction of the effective thickness of the clay to account for the perturbed zone around the galleries, is still valid and conservative for the effect of oxidation.

Future work

Future work will be concentrated on the evaluation of the effect of the oxidation products on hydraulic conductivity of the host rock and on the performance of the engineered barrier system. Therefore, the data set on pore water chemistry (especially pH values) and mineralogy will be enlarged and will be incorporated into a reactive transport modelling.

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Main reference

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Van Geet, M., De Craen, M., Weetjens, E., Sillen, X., 2006. Extent of oxidising conditions in the host formation – Experimental data and scoping calculations. SCK•CEN report ER-05.

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