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Coupled reactive transport modelling of the international Long-Term Cement Studies project experiment and implications for radioactive waste disposal

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ABSTRACT

Cementitious materials are commonly included as encapsulants and backfill or barrier materials in geological disposal facilities for radioactive wastes. An understanding of the long-term behaviour of cement-based materials is therefore required to provide confidence in their safety functions. Reactive-transport models have been produced of an *in-situ* experiment carried out at the Grimsel Test Site in Switzerland, as part of the collaborative Long-Term Cement Studies (LCS) Project, called "LCS Experiment 2". The experiment involved the emplacement of hardened Portland cement through a borehole into a fully-saturated, water-conducting shear zone (fracture) in granite for a period of six years, to explore cement leaching/degradation and the potential interaction of highly-alkaline fluids with the rock. Reactive transport models of the cement present in the experiment illustrate that it is possible to simulate the behaviour of Portland cement interacting with a low ionic strength groundwater, namely leaching of the cement (dissolution of primary solids, especially portlandite) and precipitation of carbonate minerals and secondary aluminosilicates, using a complex 3D model geometry. However, the model results highlight uncertainties surrounding cement solid dissolution rates and rates of secondary mineral formation, both of which could be explored in future research. The study illustrates the importance of modelling large-scale experimental systems which, along with natural/industrial analogue data, can be used to build confidence in the long-term behaviour of engineered barriers in radioactive waste disposal systems.

1. Introduction

1.1. Background

Cementitious materials are commonly included as encapsulants and backfill/barrier materials in repository concepts for the geological disposal of radioactive wastes (e.g. Gribi et al., 2008; SKB, 2011; Bamforth et al., 2012; RWM, 2016a). An understanding of the long-term behaviour of cement-based materials and their potential interactions with other barrier materials (including host rock) is therefore required to provide confidence in their safety functions within an environmental safety case for a given radioactive waste repository. The safety function of cementitious materials varies depending on their use and repository concept. Regarding cementitious backfill, for example, safety functions may include: protection of waste containers via alkaline pore fluids (which limit corrosion rates and reduce the potential for microbially-induced corrosion) and, in the case of higher-strength rock, offering some protection from rock falls; limiting the release and transport of contaminants (by providing high pH conditions to limit solubilities of key radionuclides, and presenting a substrate for sorption); and providing a gas-permeable medium to prevent over-pressurisation as gases are released from wastes (RWM, 2016b). The changes in hydraulic properties of cementitious backfill materials over time will also affect contaminant transport after container failure. Therefore, the potential implications of cement aging and degradation need to be understood.

The long-term behaviour and properties of materials proposed for use in radioactive waste repositories are established through a combination of smaller-scale laboratory and larger-scale *in-situ* experiments, supplemented by data from natural and industrial ("engineered")

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analogues. Each of the systems studied through these means provides supporting data at different temporal and spatial scales for the development of models of long-term evolution for both engineered and natural barriers (Savage et al., 2011). Not only do *in-situ* experiments provide the potential to investigate material behaviour over scale-lengths greater than those possible in the laboratory, but they also provide greater realism where interactions with the disposal facility host rock are of paramount importance. For example, the host rock may have significant impacts on geochemical parameters such as pH, Eh, $p\mathrm{CO}_2$ and dissolved silica activity, and for a clay-based buffer, may exert the dominant control over these parameters rather than the engineered barrier system (EBS) itself (e.g. Savage et al., 1999; Gaucher et al., 2010; Suzuki-Muresan et al., 2011). *In-situ* experiments do, however, pose challenges, in that the experimental regime may be difficult to fully characterise without disturbing prevailing conditions.

In many radioactive waste disposal concepts proposed internationally, cementitious materials play a large role, whether this is in the form of grouts used to seal fractures, or cement and concrete used in tunnel linings, structural components, backfill, plugs, containers for certain waste types and so on (e.g. SKB, 2011; RWM, 2016a). In relation to these cementitious materials, the constraints placed upon the repository by the host rock may be over-arching in terms of defining not only the longevity of such materials and the amount and style of alteration products, but also the potential magnitude of any acceptable perturbation of the geochemical conditions in the geosphere itself. For example, this includes the definition of any so-called "alkaline disturbed zone" (e.g. Baker et al., 2002), both at the small-scale around grouted fractures, and at the large-scale, around a repository with cementitious wastes and backfill. The nature and mechanisms of mineralogical alteration potentially associated with alkaline plumes have been extensively researched in recent decades (for example, Braney et al., 1993; Soler, 2003; Watson et al., 2013) and this subject has been reviewed in detail elsewhere (e.g. Gaucher and Blanc, 2006; and Savage, 2011). Such interactions are now largely understood, although the spatial and temporal scales of alteration (including the blocking or "clogging" of fractures or channelling of flow) appear somewhat uncertain, and some analogue data suggest clogging could occur (Savage, 2011) and attempts have been made to simulate this phenomenon (e.g. Watson et al., 2016).

Here, we describe the background, data input, and results of reactive transport modelling of an *in-situ* experiment carried out at the Grimsel Test Site in Switzerland as part of the Long-Term Cement Studies (LCS) Project, called "LCS Experiment 2". This project was a collaboration between JAEA (Japan), Nagra (Switzerland), Posiva (Finland), RWM (UK) and SKB (Sweden), with the overall aim of increasing understanding of high-pH cement interaction effects in the repository near field and geosphere, in order to make confident, robust and safety-relevant predictions of future system behaviour, irrespective of host rock, EBS and waste type.

LCS Experiment 2 involved emplacement of hardened cementitious material, through a borehole, into a fully-saturated, water-conducting shear zone (fracture). Observation boreholes allowed passive sample collection to monitor the extent and composition of the high-pH plume. At the end of the experiment in late 2015, the emplacement borehole was overcored and mineralogical analysis subsequently undertaken to determine the impact of the high-pH plume on the surrounding host rock.

Modelling by the LCS teams was undertaken concurrently with the experiment. Models were constructed that examined the leaching and degradation of the cement source in the experiment, as well as water flow in fractured "host rock", and the interaction of hyperalkaline cement leachate with the rock. In this paper, we present the results of detailed modelling of the cement source undertaken by the UK LCS team. Early modelling was conducted "blind", using only data related to the setting up of the experiment, e.g. physical dimensions, initial chemical compositions (of rock, cement and groundwater), transport parameters, and results of tracer tests conducted prior to the experiment, but not data produced by the experiment. As data became available, additional model cases were developed, partly for practical reasons, because although the groundwater composition in the observation boreholes was monitored in "real time", detailed information on mineralogical changes could only be obtained once the experiment was concluded and the borehole overcored for analysis, Also, there are numerous other merits in blind modelling exercises, such as: testing the general conceptual model of the system as a whole; exploring the robustness and predictive power of numerical models without "over-fitting"; providing a "bigger picture" of similar systems rather than the minute details of one particular experiment which may be subject to experimental artefacts; and providing insight into expected experiment behaviour to inform sampling.

During the preparation of this paper, modelling of the experiment by one of the other LCS modelling teams has also been published (Chaparro et al., 2017); however, the approach taken here is somewhat different, in that models are presented that include more complex spatial discretisation of the experiment, with different conceptual models and different values used for model input parameters. The differences between the two modelling approaches and implications for future work are briefly outlined in Section 4.

1.2. Description of LCS experiment 2

The *in-situ* "LCS Experiment 2" involved pre-hardened Portland cement that was placed in emplacement/injection borehole LCS08.003, which intersects fracture F16 at the Grimsel Test Site (Fig. 1). Boreholes LCS07.002 and LCS08.001 intersect the fracture further along the flow path and were used to monitor responses and retrieve fluid samples (the "observation" and "extraction" boreholes, Fig. 1). The observation borehole was just over half a metre from the emplacement borehole, and the extraction borehole is similarly just over half a metre away from the observation borehole. Thus, the distance between emplacement and extraction boreholes is a little over a metre.

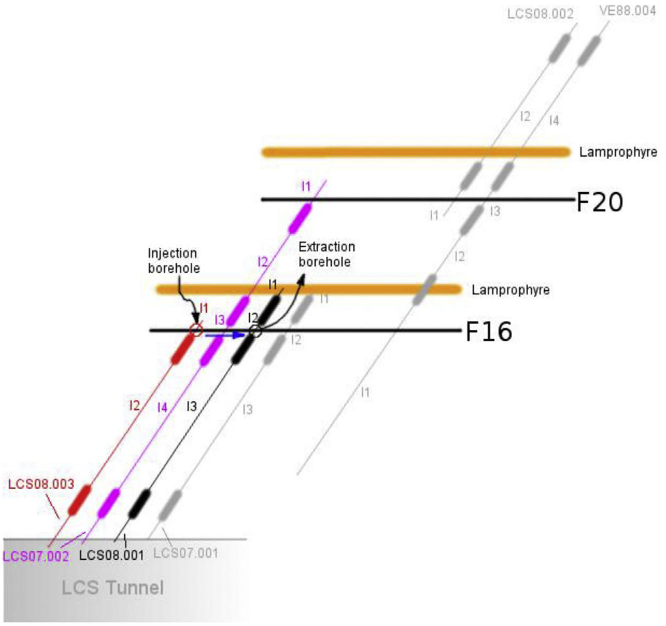

Fig. 1. Schematic diagram of the fractures at Grimsel and boreholes intersecting them. For LCS Experiment 2, the cement sample was emplaced in borehole LCS08.003 at interval I1 (Fracture F16); the alkaline plume was monitored via borehole LCS07.002 and samples were retrieved from borehole LCS08.001. The diagram also indicates the location of bands of lamprophyre (ultrapotassic igneous rocks) that are present in the Grimsel geology (courtesy of Nagra).