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Interaction processes at the concrete-bentonite interface after 13 years of FEBEX-Plug operation. Part II: Bentonite contact

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ABSTRACT

The *in situ* FEBEX experiment performed at the URL in Grimsel (Swizerland) was dismantled after 18 years of operation. Interface samples between bentonite and a shotcreted concrete plug that was constructed in a second operational phase have been studied after 13 years of interaction. Mineralogical and geochemical characterization of samples have been performed by XRD, SEM-EDX, TG and FTIR techniques in addition to determinations of major ions by chemical analysis of aqueous extracts, $\delta^{18}O$ and $\delta^{13}C$ stable isotopes both in concrete paste and bentonite, and exchangeable cations in bentonite.

Low mineralogical alteration impact was observed in bentonite that is only affected by a few millimeters. A large accumulation of Mg was observed at the bentonite side of the interface precipitating as silicates in various forms. In addition, heterogeneous carbonation was observed at the interface, but mostly affecting the concrete side.

Migration of aqueous species occurred, being the most relevant the diffusion of chloride and sulfate from bentonite to concrete, in agreement with Part I of this study. Chloride advanced more into the concrete, while sulfates reacted to form ettringite, which has an evident alteration impact at the very interface (<0.5 mm rim) within the concrete.

The ionic mobility has also redistributed the exchangeable cations in bentonite, increasing the content in Ca^{2+} and Na^{+}, compensated by a decrease in Mg^{2+}.

The results presented in this paper complement those presented in Part I, focusing on the alteration of concrete by the bentonite and the granite groundwater.

1. Introduction

A broad consensus exists in the scientific community that deep geological repositories (DGR) are actually the best solution for safe long-term storage of high-level radioactive waste (HLRW) (U.S. DoE, 2014; NEA-EC, 2003). The waste would be isolated from the biosphere by a system of multiple engineered and natural barriers. Deep geological disposal of HLRW would be technically feasible according to numerous in-situ experiments performed for more than three decades in underground research laboratories (URL) (Blechschmidt and Vomvoris, 2010).

URLs are essential to provide scientific and technical information and practical experience that are needed for the design and construction of disposal facilities and, most importantly, for safety assessment. Current URL are focussing on various long-term experiments under simulated repository conditions, full-scale demonstration and verification experiments of the disposal system, prototype repository tests and confidence building tests (Wang, 2014). The main difference between these facilities consist of the type of host rock (clay, salt or crystalline rocks) which, indeed, condition the whole system of engineered barriers. Examples of URL emplaced in crystalline rocks are found in Äspö Hard Rock Laboratory (in southeast Sweden), Olkiluoto (in western Finland), Mizunami Underground Research Laboratory (in central Japan) or Grimsel (in the central area massif of the Swiss Alps).

The FEBEX project consisted of an *in situ* full-scale simulation of

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a HLRW disposal facility performed in a drift excavated in the northern zone of the URL at the Grimsel Test Site (GTS). The GTS, located at an elevation of 1725 m in the Swiss Alps, is characteristic for a granitic host rock. The drift had a diameter of 2.28 m and an initial length of 70.4 m.

The FEBEX project was based on the Spanish disposal reference concept for disposal of HLRW in crystalline rock. Two carbon steel heaters that simulated the waste canisters were placed horizontally in the drift, located within a steel liner installed concentrically with the drift and surrounded by compacted bentonite blocks that were slowly hydrated in a natural way with groundwater from the granite (Fig. 1A). A brief description and scheme of the project is also summarized in Part I of this study, and reported in this special issue (Alonso et al., 2016). The heaters were maintained at a constant temperature of 100 °C on their surface. A concrete plug was applied to seal the drift. The operational stage started in 1997. In 2002, after five years of operation, the heater closer to the concrete plug was switched off and dismantled, along with the surrounding bentonite and measuring instrumentation in the same section of the drift (Villar and Lloret, 2007; Villar et al., 2012). A concrete plug was constructed by a shotcrete technology to seal the remaining part of the experiment and new sensors were installed in the buffer. A second operational phase started in 2002, with the configuration shown in Fig. 1B, and the experiment was definitively dismantled in 2015 (more details in http://www.grimsel.com/gts-phase-vi/febex-dp/febex-dp-introduction).

Physical and chemical compatibility between the different barriers and components of the DGR is a mandatory requirement for its construction and operation. The FEBEX experiment is contributing to investigate and quantify the evolution of the DGR in terms of thermo-hydro-mechanical and thermo-hydro-geochemical behaviour, which is derived from the interaction among the different barriers, and is a major issue in the safety evaluation of any DGR.

Compacted bentonite will be used in the DGR because its advantageous properties in terms of plasticity, high swelling capacity, low hydraulic conductivity and high capability for retarding the radionuclides mobility, which makes it essential as barrier system. However, alkaline leachates from the concrete used as plug (Ordinary Portland Cement based, OPC-based) interact with the bentonite and can have a detrimental effect on the main properties of bentonite. The thermal impact at the concrete plug-bentonite interface is low. The range of temperatures considered for this case are between 15 and 35 °C (Villar et al., 2008, 2012).

A variety of processes have been identified in granite groundwater-bentonite-concrete scenarios, being the most relevant as follows:

1) The carbonation of concrete at the concrete-bentonite interface, favoured by the dissolution of portlandite and precipitation of carbon species: from groundwater, bentonite porewater and those migrated in response to chemical gradients. Precipitation of calcite rims have been observed at the interface of an OPC cement paste in contact with a Callovo-Oxfordian argillite (Dauzères et al., 2014), while calcite, aragonite and gypsum have been observed either at the concrete hydration surface or at the interface of concrete and bentonite (Turrero et al., 2011). Precipitation of calcite at the interface produces a net increase in volume leading to pore space clogging which could temporarily isolate the concrete and bentonite system (Jenni et al., 2014). In the long term, calcite will compete with calcium silicates hydrates (C-S-H) evolution. Some of these processes have been summarized by Bildstein and Claret (2015).
2) Incorporation of Al in C-S-H. C-A-S-H phases have been identified at FEBEX bentonite-CEM I and lime mortar interfaces at different time scales, from several months up to 7 years (Fernández et al., 2016), consistent with narrow compositional types with 0.2–0.3 Al/(Al+Si) and ~0.8 Ca/Si ratios (Jackson et al., 2013). Structural interaction, either by intercalation or association of montmorillonite and C-A-S-H may cause hindrance of montmorillonite swelling.
3) Sulfate and chloride migrate into concrete. Chloride can diffuse from compacted bentonite and react with cement minerals to form Friedel's salt from AFm phases or absorb in C-S-H phases (Baur et al., 2004; Florea and Brouwers, 2012; Yuan et al., 2009). Simultaneously, sulfate can react with dissolved portlandite from concrete and dissolved Al from montmorillonite to form ettringite (Cuevas et al., 2016) which, in realistic conditions, produce porosity changes in concrete (Dauzères et al., 2014) that can be relevant for the geochemical evolution of the barriers system.
4) Mg perturbation and redistribution of exchangeable cations in montmorillonite. High pH cements are a source of Na^+, K^+ and Ca^{2+} that diffuse from the concrete porewater into the bentonite. Montmorillonite selective dissolution and K^+ ion exchange can lead to apparent illitization processes (Bildstein and Claret, 2015; Cuevas et al., 2016; Gaucher and Blanc, 2006), followed by precipitation of secondary phases like C-(A)-S-H phases. Decrease of Mg and increase of Ca in both, the bentonite porewater and the exchangeable cation population, have been observed to occur in FEBEX compacted bentonite

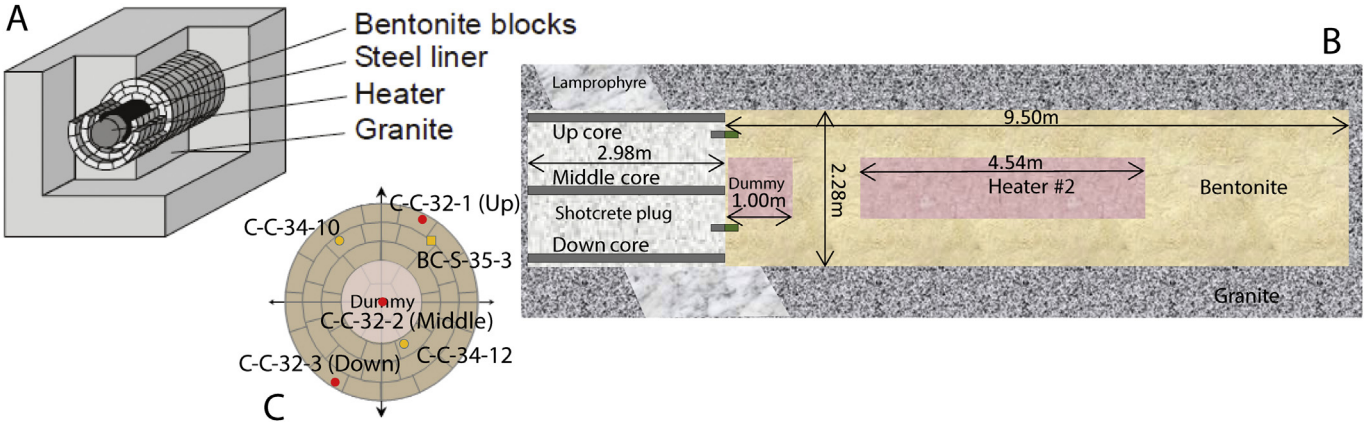

Fig. 1. A: Scheme of the engineered barrier experiment installation (FEBEX-e). B: lateral view of the FEBEX-e installation and location of the samples evaluated in this paper and in the Part I of the study (Alonso et al., 2016, this issue). C: frontal view of the FEBEX-e installation and location of the samples evaluated in this study.