Diffusion experiment (^{36}Cl, ^{3}H) across concrete/claystone interface

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Engineered cementitious barrier components juxtaposed to a claystone host rock introduce chemically reactive interfaces, and this may modify transport properties (water, solutes, gas) relevant for the long-term behavior of waste confinement. The CI-D field experiment (diffusion across 10-year-old concrete/claystone interface) carried out at the Mont Terri rock laboratory (Switzerland) aims at directly assessing the impact of an aged cement/claystone interface on diffusive transport of radionuclides, ^{36}Cl and ^{3}H (as HTO).

A small circulation interval was installed in 2018 in a vertical OPC concrete section emplaced in 2007 (Fig. 1), with a closest distance of 8.5 cm from the interface with Opalinus Clay, an indurated claystone of Jurassic age and dedicated host rock for deep storage of radioactive waste in Switzerland. Circulation with a synthetic high-pH fluid (to match aged OPC porewater) started after installation, and radionuclide tracers ^{36}Cl and HTO were added in 2019 ($4 \cdot 10^4$ Bq/L, $2 \cdot 10^5$ Bq/L). The decrease of activity in circulation was monitored by regular sampling and liquid scintillation counting (LSC) analysis. The experiment was stopped in September 2023, after 52.5 month of radionuclide circulation, followed by overcoring and sampling.

Core material was retrieved by drilling parallel to the experiment borehole some distance above and below the experiment, and by overcoring the entire experiment borehole with a large diameter of 350 mm (Fig. 1). A proven stabilization technique was employed whereby fiberglass elements were embedded in epoxy resin in small boreholes drilled parallel to the overcoring direction. This stabilization technique bolted the different concrete and claystone materials together across interfaces.

The overcores from above and below the experiment were drilled with a double-barrel tool of 131 mm diameter, and comprised five well-preserved cores of 3 m length containing Opalinus Clay and intersections of various proportions of concrete of the target vertical pile BCI-6 (Fig. 1). The large-diameter overcore containing the experiment borehole comprised most of the HTO and ^{36}Cl, assumed to be near the experiment borehole. This yielded a 2.15 m long and previously stabilized and extremely well-preserved compound core of 336 mm diameter, and weighing >500 kg.

Sampling of the cored material aimed at obtaining an approximate 3-D distribution of the radionuclide tracers in the porewater and solid of concrete and Opalinus Clay, and specifically across concrete/claystone interfaces to assess the presence or absence of steep activity gradients that would be indicative of a skin-effect. Approximately 250 cuboidal samples were cut for preparing aqueous extracts at a liquid/solid ratio of 1.5, and somewhat fewer samples for determining water contents. Additional samples from the interface region were prepared for a mineralogical and chemical characterization of the interface by electron microscopy/element mapping as done on samples after 5 years of alteration time [1].

Analytical work is on-going. LSC analyses of aqueous extracts are successful near the circulation source for ^{3}H and ^{36}Cl in concrete and adjacent claystone, but further away the much lower detection limit of Accelerator Mass Spectrometry (AMS) is required for ^{36}Cl to trace diffusion profiles across interfaces, requiring extra efforts for chemical purification. Geochemical analysis includes the determination of anions in the aqueous extracts, obtaining chloride and sulfate distribution in concrete and Opalinus Clay, possibly followed by more specialized studies. Measurements will be assembled as a 3-D data grid, followed by visualization, as an input for future radionuclide transport modelling.

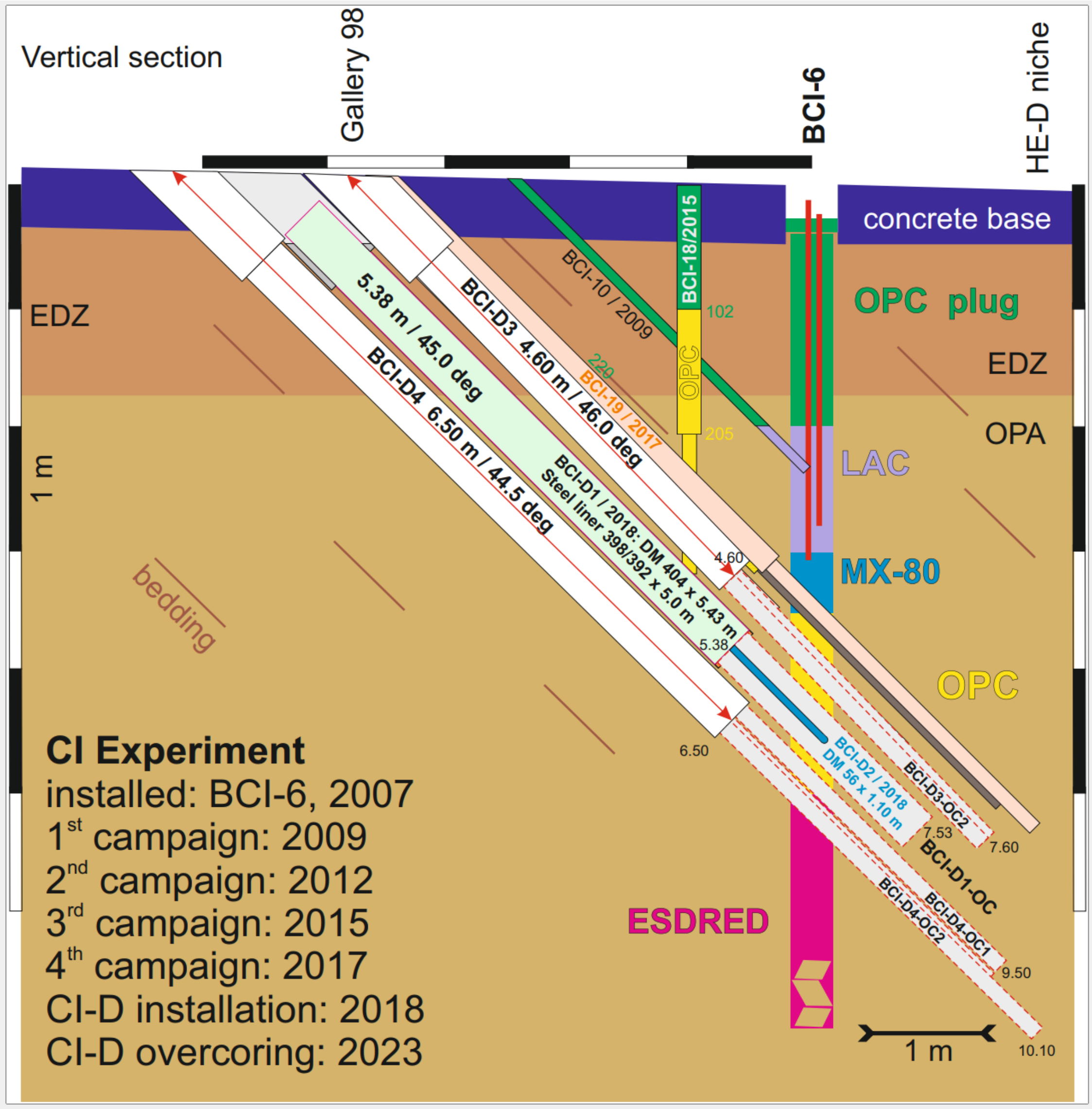

Figure 1: Vertical section containing target borehole BIC-6 and three overcoring boreholes. BCI-D3 and BCI-D4 are two new approach boreholes, and BCI-D1 is the existing approach borehole fitted with a steel liner. BCI-D2 is the experiment borehole (blue, 56 mm diameter) equipped with a double-packer system. The sections stabilized with fiberglass tubes were approximately 2 m long. Overcoring was performed with 131 mm diameter for BCI-D3 (2 cores, 3 m long, light grey) and BCI-D4 (3 cores, 3 m and 3.6 m long). BCI-D1 was overcored with 350 mm diameter (2.15 m long). The borehole mouths of BCI-D4, BCI-D1 and BCI-6 are nearly collinear. BCI-D3 is a bit outside this plane and projected into it.

References

[1] U. Mäder, A. Jenni, C. Lerouge, S. Gaboreau, S. Miyoshi, Y. Kimura, V. Cloet, M. Fukaya, F. Claret, T. Otake, M. Shibata. 5-year chemico-physical evolution of concrete–claystone interfaces, Mont Terri rock laboratory (Switzerland). Swiss Journal of Geosciences, 110, 307-327, 2017.