



Argillite-cement interaction: the Tournemire experimental platform (France) as an analogue of a deep geological setting

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The alkaline disturbance of clayey rocks in contact with cementitious materials is an important task in safety performance studies of deep geological storage for high-level radioactive wastes in France. In this purpose, the Institute for Radioprotection and Nuclear Safety (IRSN) and the Scientific Research National Center (CNRS) have developed a research program in the Tournemire experimental platform (France). This program, integrated in the TRASSE Research Group, aims to describe the modifications of Toarcian argillites in contact with cementitious materials over various time scales (from a few years to a hundred of years).

The Tournemire experimental platform of IRSN is based on a tunnel built between 1882 and 1886 and several galleries excavated since 2003 through Domerian marls and Toarcian argillites. The tunnel walls were recovered by lime that is yet in contact with the argillites. In 1990 years, exploration boreholes were realized from the basement of the tunnel and then filled with concrete and cement that are presently in contact with the Toarcian argillites for 15-20 years. In 2003, two experimental research galleries were excavated perpendicularly to the tunnel and covered by several centimeters of four concrete formulations. These different engineered analogues accessible in the Tournemire experimental platform are rare examples of contexts presenting various - in nature and age - clayey rock / cementitious materials contacts that could be studied as engineered analogues. This study is completed by lab experiments (diffusion and advection) performed in smaller time and space scale to control some parameters.

This presentation aims to detail the scientific program developed on two engineered analogues contexts (1 and 2) and on diffusion lab samples (3).

(1) In the DM borehole, two disturbed zones were observed at the cement-concrete contact: a P1 matricial disturbance underlined by a continuous zonation from cement-concrete toward the argillite and a P3 disturbance identified along inframillimetric fissures perpendicular to the cement contact and filled with secondary minerals. Once more a zonation was observed on the wall of the fissure according to the distance to the cement-concrete. Macroscopic and microscopic observations displayed several textural and mineralogical drastic changes in the argillite in contact with cement over a distance of 1 cm at the P1 scale and of 1.5 cm for P3 scale. Even if the transformations were less developed within the argillite matrix than for disturbance along fissures, nearly the same mineralogical transformations were observed for the two paths. Beyond the disturbance zones, argillite was again well bedded and similar to a reference sample. $87\text{Sr}/86\text{Sr}$ and $d^{13}\text{C}$ isotopic ratios measured in the carbonated phase of the clay changed also significantly in argillites close to the contact; The disturbance length was more pronounced along P3 than in P1 (see Techer et al. poster in the GMPV13 session). Spatial porosity distribution into argillite and cement were revealed using (^{3}H -PMMA and ^{14}C -PMMA) autoradiograph. Quantification of porosity by the autoradiograph image analysis software argued for disturbances into both materials at the interface: (i) an increase of the porosity into the cement in the first centimetre in contact with clay and (ii) a decreasing of the porosity into the first and half centimetre in the argillite in contact with cement.

(2) In the 2003 galleries of the Tournemire experimental platform, four cores were drilled perpendicularly

to the wall crosscutting the concrete and the argillites over around 1 metre. The argillite/concrete interface was analyzed in four configurations: (i) contact with a Portland concrete containing siliceous ripples (ii) contact with a Portland concrete containing carbonated ripples (iii) contact with a CHF concrete containing siliceous ripples and (iv) contact with a CHF concrete containing carbonated ripples. Preliminary results obtained on the mineralogical (macroscopic and mineralogical observations, XRD measurements) and Sr isotopic analyses of argillites collected close and far from these interfaces are presented.

(3) The diffusion study was designed to provide better understanding of the phenomena that govern diffusion processes during the transient phase between site and alkaline conditions. Experiments involving the use of "through-diffusion" cells were performed to reproduce the effect of an alkaline plume through different types of Tournemire clayey materials. Different kinds of alkaline fluids (fresh concrete (N1) and moderately degraded concrete (N2) synthetic fluids) and sliced rock materials (no fractured, with an opened fracture or with a tectonic fracture filled with calcite) were investigated for the diffusion of both water and major cations. Monitoring of the pH and of the major cations contents (Na, K, Ca, Mg) and investigations of the solids before and after alteration lead to identify numerous alkaline alteration parameters.