Geophysical Research Abstracts,
Vol. 11, EGU2009-10103, 2009
EGU General Assembly 2009

Sr, C and O isotopes as markers of alkaline disturbances in the Toarcian argillites of the Tournemire experimental platform (France). Case of a 15-years old engineered analogue.

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In France, the concept of a geological disposal of high-activity and long-period nuclear wastes requires the use of concrete and cement-bearing materials as building structures or as waste containment packages, in conjunction with clayey barriers (e.g., compacted bentonite as an engineered barrier and/or argillite-type rocks as a geological barrier). Hydrolysis of cementitious phases is however known to produce hyper-alkaline pore fluids with pH ranging from 10 to 13.5 that will be in disequilibrium with the geological setting environment (argillite pore-water pH around 8). The disturbance of clayey rocks in contact with such materials is thus an important task in safety assessment studies of deep geological storage. This concerns the knowledge of mineral / solution paths but also the spatial extent of the alkaline plume into the clayey material.

Experimental and modelling approaches were performed this last decade to answer these questions. In addition to these approaches, natural or engineered contexts in which a clayey formation has been in contact with cementitious materials can be considered as analogues of a deep geological storage for the study of argillite /cement interaction. Such contexts can be found in the IRSN Tournemire experimental platform in Aveyron (France). This platform is based on a tunnel, excavated between 1882 and 1886 through Domerian marls and Toarcian argillites, which is dedicated since 1990 to multidisciplinary research programs. In the frameworks of these programs, exploration boreholes were realized from the basement of the tunnel in the 1990 years. The boreholes were then filled with concrete and cement that are presently in contact with the Toarcian argillites for 15-20 years.

One of this borehole – DM borehole – was overcored in 2005 in order to collect the Toarcian argillites in contact with the cement and the concrete. Mineralogical, petrographic and microstructural analyses have argued for a clear disturbance of the Toarcian argillites close to the cement-concrete contact. These data are detailed in another session of this meeting (Techer et al., ERE6 session). In order to precise the spatial extent of the disturbances and to discuss the nature of the responsible fluids, a systematic chemical and isotopic study was performed focusing on the Sr, C and O isotopes known to be very good markers of alkaline fluids percolation (Fourcade et al. 2006).
Four studied levels were selected according to their location on the DM overcore and the nature of the cementitious material in contact to the argillites: -155 cm level (DM155) and -180 cm level (DM180) where argillites were in contact with a 1.5 to 3 cm thick concrete; -245 cm level (DM245) and -300 cm level (DM300) where the cementitious material was represented by a fine grained Portland cement 2 to 5 cm thick. Samples of the cementitious materials were collected at each level. Argillites were sampled perpendicularly to the cement-concrete contact with a continuous sampling every 2 to 5 mm millimeters (P1). Micro-fissures developed perpendicularly to the cement-concrete contact were opened and infilling secondary minerals were collected by scrap as a function of the distance to the cement-concrete (P3).

Along P1 and P3, no significant variation of the carbonates d18O isotopic values was observed in the argillites.

On the opposite d13C and 87Sr/86Sr values of these minerals changed significantly in the direct contact with the cement-concrete over a distance of 15 to 25 mm. Beyond this distance, argillites were again well bedded and showed values similar to those measured in a reference sample. Negative d13C values measured in the disturbed zone and close to those encountered in the cement and the concrete argued for a perturbation induced by an alkaline solution. 87Sr/86Sr isotopic ratios showed a progressive evolution in the disturbed zone, with increasing trends towards the cementitious material. Thanks to complementary Sr elementary contents measurements, these intermediate data were explained by the precipitation of secondary carbonates from fluids generated by the cement and/or the concrete alteration. The imprint of these secondary phases, and thus the spatial extent of the alkaline fluids into the clayey material were observed over 25 mm along P3 (along discontinuities) and over 15 mm along P1 (in the matrix).

References

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Techer et al., this meeting, ERE6 session