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Cement water injection in an equipped borehole of the Meuse Haute Marne Underground Research Laboratory: monitoring and modeling

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Concrete will be significantly used in the context of deep repository for radioactive wastes for building access structures, galleries as well as vaults and waste packages for Intermediate Level Wastes (ILW). These materials constitute a compromise between properties, technical use and cost. As the repository can be constructed in a low-porosity detrital argillite (Callovo-Oxfordian) in the Mesozoic formations of Eastern part of the Paris Basin (France), and as swelling clays are planned to be used as sealing materials, concretes will be in contact with clayish materials. Due to the geochemical disequilibrium between concrete and clays, chemical reaction can modify both chemical and physical properties of these materials (i.e. mineral phases, diffusion coefficient, et cetera). In order to improve knowledge on the long term behavior of concrete/clays interfaces and the evolution of the properties with time, the MLH experiment has been conducted since July 2010 in the Meuse/Haute-Marne Underground Research Laboratory. The results will be used to calibrate and improve long term predictive modeling.

The MLH experimental set up included two synthetic cement waters (pH 13.5 and 12.5, Table 1), circulation in two separated chambers (75 cm height, diameter 76 mm) in a borehole drilled in the GED gallery. D₂O has been also injected as a tracer. Iodine and bromide have been spike in the synthetic solutions to quantify the anions accessible porosity. In the following these two solutions are called Alc (synthetic pore water with alkali ions) and EqPort (synthetic solution in equilibrium with portlandite). Samplings are performed as a function of times and the sampled solution is then analyzed. Major cations and anions are monitored with time as well as pH and alkalinity. After 265 days the pH has dropped to pH 7.2 for the EqPort solution and 8.6 for the Alcsolution.

Table 1 : Elements concentrations and pH of the two injected solutions. The Callovo-Oxfordian (COX) pore water is also recalled for comparison. Two injections 1 and 2 have been performed.

Elements	Measured concentrations (mol.L ⁻¹)				COX pore water
	Solution 1 (Alc)		Solution 2 (EqPort)		
	Injection 1	Injection 2	Injection 1	Injection 2	
Na	5.9 10 ⁻²	5.8 10 ⁻²	1.0 10 ⁻³	9.9 10 ⁻⁴	5.61 0 ⁻² ± 4 10 ⁻³
K	3.2 10 ⁻¹	3.5 10 ⁻¹			9 10 ⁻⁴ ± 3 10 ⁻⁴
Ca	8.3 10 ⁻⁴	4.3 10 ⁻⁴	1.8 10 ⁻²	1.8 10 ⁻²	7.6 10 ⁻³ ± 1.4 10 ⁻³
Br.	1.2 10 ⁻³	1.0 10 ⁻³			
I			9.3 10 ⁻⁴	8.4 10 ⁻⁴	20-40 10 ⁻⁶
SO4	1.9 10 ⁻³	1.8 10 ⁻³			1.9 10 ⁻⁰² ± 4 10 ⁻³
pH	13.5	13.5	12.5	12.5	7.2 ± 0.2

The two solutions have been renewed after 265 days. In order to reproduce the pH decrease, the major ions changes within the solution a coupled geochemistry transport model has been built using

PHREEQC. The model proposed by Appelo et al.(2008) to obtain the porewater composition of a clay rock by modeling the in- and out-diffusion of anions and cations from an in-situ experiment has been adapted to model to our experiment. The transports parameters (diffusion coefficient, porosity, size of the zone around the borehole disturbed mechanically by drilling and equipment) have been calibrated on the D₂O data. Regarding the setting up of the geochemical reaction two models have been tested. The first one integrates multicomponent diffusion (MCD) and diffuse double layer effects in clays in conjunction with ion exchange and calcite dissolution, whereas in the second one a full mineral assemblage has been taken into account. In this second model MCD was still considered but not diffuse double layer effects in clays. The full mineral assemblages was chosen according to the clay pore water robust model described by Gaucher et al.(2009). The first simplified model duly calculates transport of the K and Na in clay rocks for both EqPort and Alc solutions, indicating that their behavior is driven by the exchange on the clay. Taking into account the full mineral assemblage and allowing precipitation and dissolution of the minerals allow to properly reproduce the Ca profile over time. The quick pH decrease is also described although it does not perfectly match the pH profile. In order to improve this latter point some hypothesis (e.g. kinetic instead local equilibrium) are currently in progress.

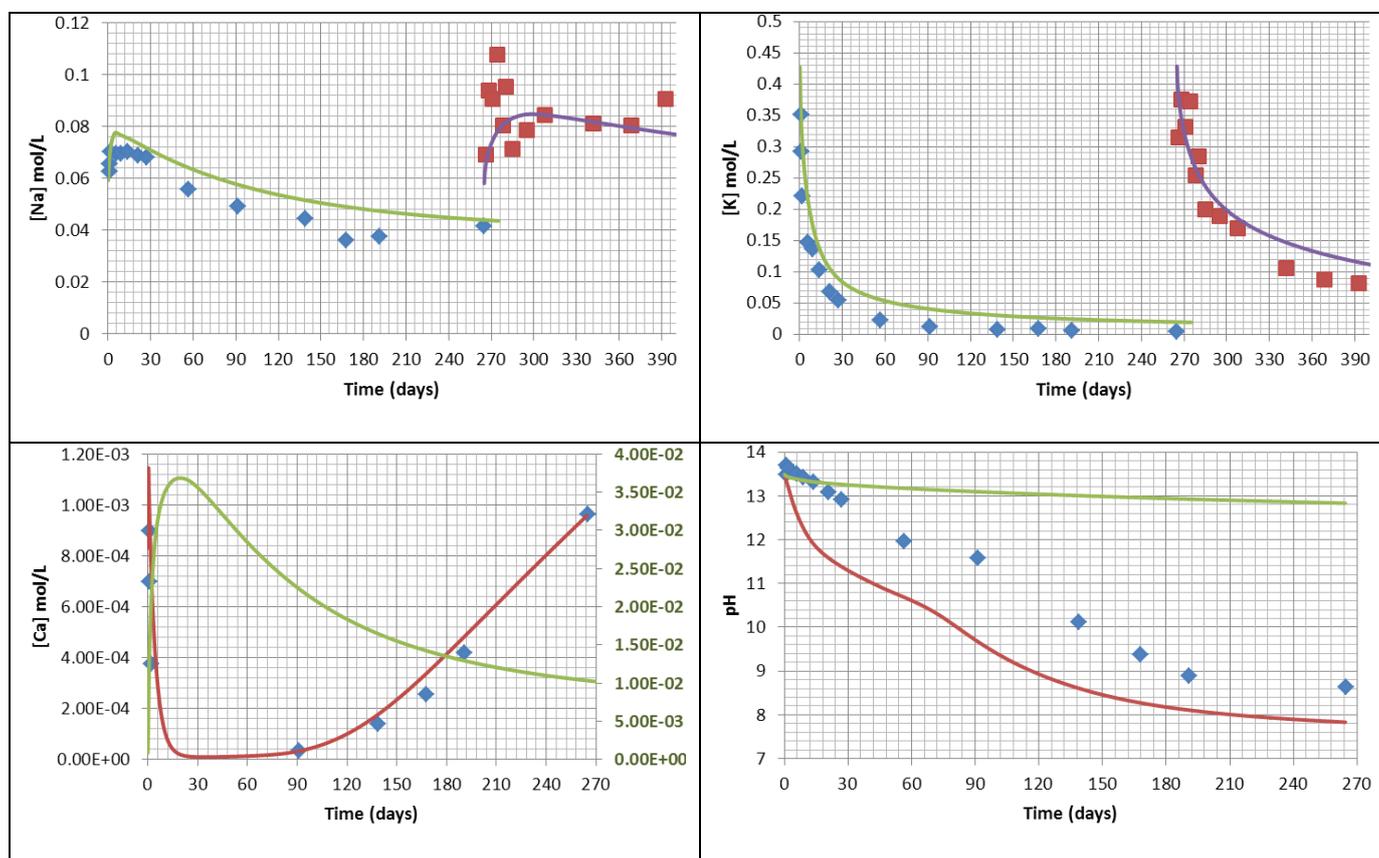


Figure 1 : Na, K, Ca and pH evolution as a function of time for the Al-s solution. Exchange on the clay reproduces the Na and K profile. This simplified model cannot reproduce Ca evolution (green curve and right y axis), whereas a full mineral assemblages (red curve and left y axis) allow a good fit for Ca and overestimate pH drop (red curve).

Appelo, C.A.J., Vinsot, A., Mettler, S., Wechner, S., 2008. Obtaining the porewater composition of a clay rock by modeling the in- and out-diffusion of anions and cations from an in-situ experiment. J Contam Hydrol 101, 67-76.

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