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Procedia
Earth and Planetary Science**Water Rock Interaction [WRI 14]****Insights on desaturation processes based
on the chemistry of seepage water from boreholes
in the Callovo-Oxfordian argillaceous rock**A. Vinsot^{a*}, Y. Linard^a, M. Lundy^a, S. Necib^a, S. Wechner^b^aAndra, Meuse/Haute-Marne Center, RD960, F-55290 Bure, France^bHydroisotop GmbH, D-85301 Schweitenkirchen, Germany**Abstract**

Seepage waters from boreholes were collected in the Meuse/Haute-Marne Underground Research Laboratory (NE France) in the Callovo-Oxfordian clay-rich host rock. Over the first months after the drilling of the boreholes, these waters were saltier than the waters collected later on, even in the boreholes that had been drilled without oxygen. These chemical composition variations give insights on the volume of rock affected by the loss of water around the boreholes. This data will help to characterize the desaturation processes in such a rock and the consequences with respect to the composition of the water that will fill the underground works after their closure.

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1. Introduction

The Callovo-Oxfordian argillaceous rock in the eastern Paris Basin has been studied since 2004 in the Meuse/Haute-Marne Underground Research Laboratory (MHM URL) built by Andra, the French National Radioactive Waste Management Agency [1]. At the end of 2012, more than one km of 5 to 6 m diameter drifts has been excavated, about ten 70-cm-horizontal micro-tunnels have been sunk and several hundred boreholes have been cored in the URL. Since their excavation, drifts and micro-tunnels have been in contact with the URL ventilation atmosphere. Most of the boreholes were cored with air, whereas a few boreholes were drilled with dry nitrogen or argon. In all of these underground works, gas interactions between the rock pore-water and the gas phase have taken place at the rock surfaces that have

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been exposed to the gas phase due to excavation. Two of the interacting gases play a major role on the rock surface mineralogy and the pore water composition: water vapor and oxygen. At each surface that has been exposed to the gas phase, water vapor fluxes go from the rock to the gas phase which is under-saturated with respect to water. The water loss induces an increase in the solute concentrations in the pore water at the rock surface. At the same time, oxygen from the air reacts with the reduced species in the rock at each exposed rock surface, except in the boreholes drilled with an anoxic gas and closed just after the drilling phase.

In the framework of the feasibility study of radioactive waste underground disposal in such a rock, there is a series of questions concerning the consequences of these processes on the disposal evolution.

The consequences of these processes on the rock properties around the drifts were studied with the help of *in situ* experiments such as the ventilation experiment (VE) in the Mont Terri Rock Laboratory (Zheng *et al.* 2008 [2]). When they modeled the VE experiment, Zheng *et al.* took into account both multiphase flow and multicomponent reactive transport. Their calculated results were compared to pore-water compositions inferred from measurements on core samples taken from around the ventilated micro-tunnel. They concluded that the impact of these processes on the rock porosity was negligible. Nevertheless, they showed that concentrations of several dissolved species had increased at the tunnel wall due to water loss and oxidation reactions.

In the MHM URL, liquid water production observed in several boreholes makes it possible to confirm this effect at borehole scale. Moreover, the boreholes that were drilled with argon or nitrogen made it possible to observe only the effects of the rock desaturation (without oxidation) on the water composition.

This paper presents the composition of such waters and how this data could contribute in describing the desaturated zone surrounding the underground works.

2. Chemistry of borehole waters

The data presented below come from 3 boreholes cored in drifts located at a depth of -490 m in the MHM URL (Table 1). In each of these boreholes, equipment including an inflatable packer isolates a test interval at the far end of the borehole. The setup of the borehole PAC1002 is described in details in [3]. The dimensions and setups of the PAC1002 and POX1201 boreholes are identical. The test intervals of these 2 boreholes were filled with pure argon after equipment installation. The test interval of the borehole MCO1202 was filled with synthetic pore water and a gas mixture of N₂ and 1% CO₂. The pressure in all of the test intervals was set at a value of a few bars from the beginning. The pore pressure surrounding the test intervals is larger than 30 bars. So, the difference between these pressures is greater than 25 bars. As a consequence, in spite of the low value of rock hydraulic conductivity ($< 10^{-12}$ m.s⁻¹), water flows from the rock into the test intervals. The measured water production flow rate in the 3 boreholes was close to 1 L/month over the first year after their installation and after an initial transition time period for MCO1202. The area of the rock wall in the test interval is close to 1 m² in the 3 boreholes. The water production flow rate was between 30 and 40 mL/day/m² in the 3 boreholes.

Table 1. Characteristics of the boreholes.

Borehole	Total length (m)	Diameter (mm)	Orientation	Coring gas	Test interval length (m)	Filling fluid
PAC1002	15	76.3	Vertical upward	Nitrogen	5	Argon
POX1201	15	76.3	Vertical upward	Argon	5	Argon
MCO1202	12	350	Vertical downward	Nitrogen	0.65	Synthetic pore water + gas (N ₂ and 1% CO ₂)

Figure 1 (a, b) presents the concentration evolution of the two ions which have the highest concentrations in the seepage water collected in the PAC1002 and POX1201 boreholes over 800 days for Na^+ and over more than 1700 days for Cl^- . (After 800 days, O_2 was introduced into the test intervals in both boreholes and modified the seepage water Na^+ concentration. These oxidation effects are not addressed here).

Results obtained on both boreholes are similar. After a time period of about 300 days, the Na^+ concentration became almost stable. Over the first 300 days, the concentration of this species decreased. The Cl^- concentration showed variations over the five-year monitoring period. Nevertheless, the largest Cl^- concentration values were observed during the first 300 days. The largest concentration values were 1.2 to 1.3 times larger than the center value of concentrations measured after 300 days. Indeed, the water sample with the largest solute concentrations from the boreholes PAC1002 and POX1201 has a composition similar to that of the water sample taken after 300 days that would have lost about one fifth of its water by evaporation.

The test interval of the borehole MCO1202 was partly filled with 17.1 L of synthetic pore-water. The total volume available for fluids in this test interval was about 45 L. The composition of the synthetic pore-water was similar to the PAC1002 water compositions ($\text{Cl}^- \approx 38 \text{ mmol/L}$). Over the first 200 days of the experiment, the water volume increased by around 4.2 L in the test interval due to the inflow of seepage water. The Cl^- concentration at the end of this time period was 77 mmol/L (Fig. 1c). Then, between day 200 and day 700, the Cl^- concentration decreased in the water of the borehole MCO1202.

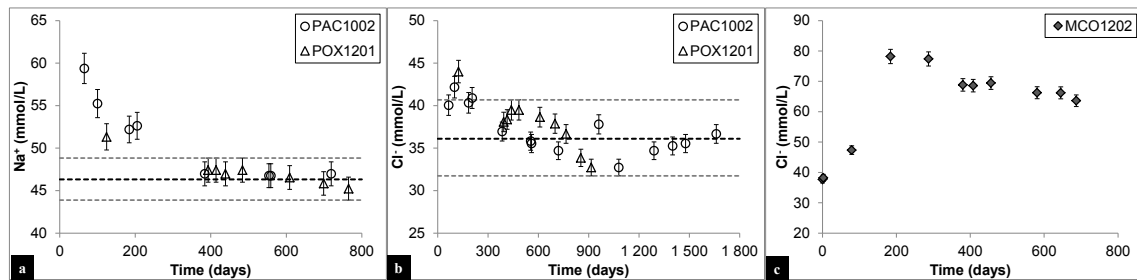


Fig. 1. (a) and (b): Na^+ and Cl^- concentrations measured in the seepage water of PAC1002 and POX1201. The dashed lines represent the limits and the center of the whole concentration range after 300 days. Initial time is the end of the drilling phase. (c): Cl^- concentration measured in the water of MCO1202, which was partly filled with synthetic water from the beginning.

3. Discussion

Based on the data gathered from the PAC1002 and POX1201 boreholes, one can estimate the total quantity of Na^+ and Cl^- in the seepage water volume collected over the first 300 days, which is in excess with respect to the same volume of pristine pore water. This quantity is obtained by integrating the curves presented on Fig. 1 (a, b) over the first 300 days, down to the center value after 300 days, and by considering a water flow rate of 40 mL/day over this time period. Based on this quantity, we can deduce that there had been a water loss ranging from 1.4 to 1.8 L in the rock surrounding the test intervals.

Regarding the evolution of the Cl^- concentration in the borehole MCO1202 before day 200 (Fig. 1c), the 17.1 L of added synthetic pore water should have diluted the pore water from the rock which had lost water around the test interval; on the other hand, the 4.2 L of seepage water should include pore water which had also lost water. After day 200, the decrease of the Cl^- concentration should be due to the inflow

of pristine pore water and to the diffusion of Cl^- from the borehole water to the pore water in the rock surrounding the test interval. This hypothesis was tested successfully with the flow and transport model and the diffusion parameters used in [4]. Based on the Cl^- data collected before day 200 and on the hypotheses presented above, we can deduce that there had been a water loss ranging from 18 to 24 L in the rock surrounding the test interval of the borehole MCO1202.

Finally, it comes from these results that the volume of evaporated water was 1.6 ± 0.2 L in the case of the 76.3 mm-diameter boreholes, and 21 ± 3 L for the 350 mm-diameter borehole, whereas the areas of the test interval rock wall were similar (close to 1 m^2) in the 3 boreholes.

This observation gives an insight regarding the open fractures surrounding the boreholes. Indeed, the evaporated water likely came from the fracture surfaces that were exposed to the gas phase. The geometry of the fractured zones surrounding various underground works in the URL was described by Armand *et al.* [5]. Figure 2 illustrates this geometry for a drift parallel to the horizontal major stress. Armand *et al.* showed that the same geometry was observed at various scales in this orientation: around rock bolts (5 cm-diameters), micro-tunnels (70 cm-diameters) and drifts.

We propose that there is a relationship between the surface area of the fractures in the traction zone (where the fractures are open to the gas phase) described in [5] and the volume of evaporated water. This link is qualitative at this stage. Multiphase flow calculations should help to precise this link.

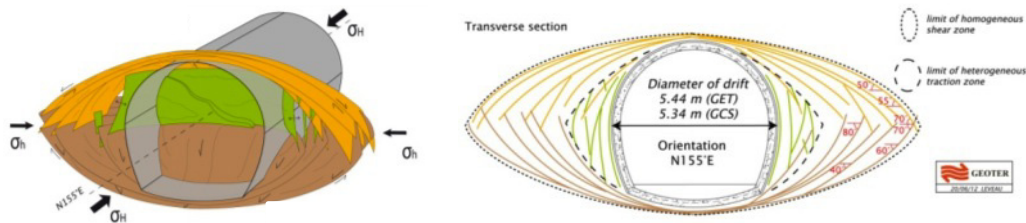


Fig. 2. Conceptual model of the induced fracture network around a drift parallel to the horizontal major stress (from [5]).

In the forthcoming years, efforts will be put on sampling and analyzing the first seepage waters from the micro-tunnels or from the drifts. This data should help to complete the preliminary investigations presented here and to improve the prediction on the composition of the water that should interact with repository materials.

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