

HYDROGEOCHEMICAL INVESTIGATIONS AT THE ANDRA MEUSE/Haute-MARNE UNDERGROUND RESEARCH LABORATORY

A. VINSOT, J. DELAY, H. REBOURS

*Andra, Laboratoire de recherche souterrain de Meuse/Haute-Marne,
RD 960, F-55290 Bure, France*

ABSTRACT

In November 1999 Andra began building an Underground Research Laboratory (URL) in eastern France. The geological formation selected for this laboratory is a 130-meter thick argillaceous rock level. This clay rich layer is located at a 400 to 600 meter depth. To characterize the confining properties of the clay, pore water composition had to be studied. For this purpose an innovative device was designed for gas equilibration and direct sampling of the pore water. The experimental device consists of a vertical ascending borehole with a 5 meter long test interval at its far end in which a gas circulation is established. After a few weeks, due to the hydraulic gradient between the test interval and the rock formation, the water flows freely at a rate of 0.5 to 1.3 liters per month in the borehole and it is sampled. The chemical composition of this water is compared with a theoretical composition deduced from core analyses and thermodynamic modelling.

1 Introduction

The Callovo-Oxfordian formation of the eastern Paris Basin is a 130 m thick clay rich sedimentary sequence. Its water content is around 8 %wt and its hydraulic permeability is below $10^{-11} \text{ m.s}^{-1}$ (10^{-18} m^2). Since 1994, Andra (the French Agency for nuclear waste management) has been studying the feasibility of a high-level long-lived radioactive waste disposal in this formation. The geochemical composition of pore water is one of the elements required to assess the confining properties of this argillaceous rock. Two aspects related to pore water composition are being studied to this effect. The first one concerns the knowledge of the hydrogeochemical mechanisms governing this composition to study the interaction of the water with the various barriers surrounding the waste canisters. The second aspect concerns the distributions of non reactive natural tracer concentrations. Indeed, the interpretation of the observed distributions will help to evaluate the history of solute transfers.

From the outset, the Callovo-Oxfordian pore water composition has been studied through measurements performed on core samples from deep boreholes. A conceptual model of the pore water composition based on thermodynamic equilibrium has been built and a consistent set of data has been obtained on natural tracers such as water stable isotopes, noble gases and chloride [1].

In 2004, in situ direct sampling of the water was carried out for the first time in this type of formation and has been performed continuously since then. For that purpose, two short (15 m long) boreholes were drilled in two drifts at 445 m and 490 m depth (Fig 1). In the open section of these boreholes (test interval), a gas is circulated at a total pressure of about 1.2 bar. Due to the hydraulic head gradient between the test interval and the surrounding rock, the water flowing freely into the interval is pumped out. These two experiments have made it possible to “pump” the pore water at rates between 0.5 and 1.3 L per month for the past two years. This paper presents the design and main results of this innovative experiment.

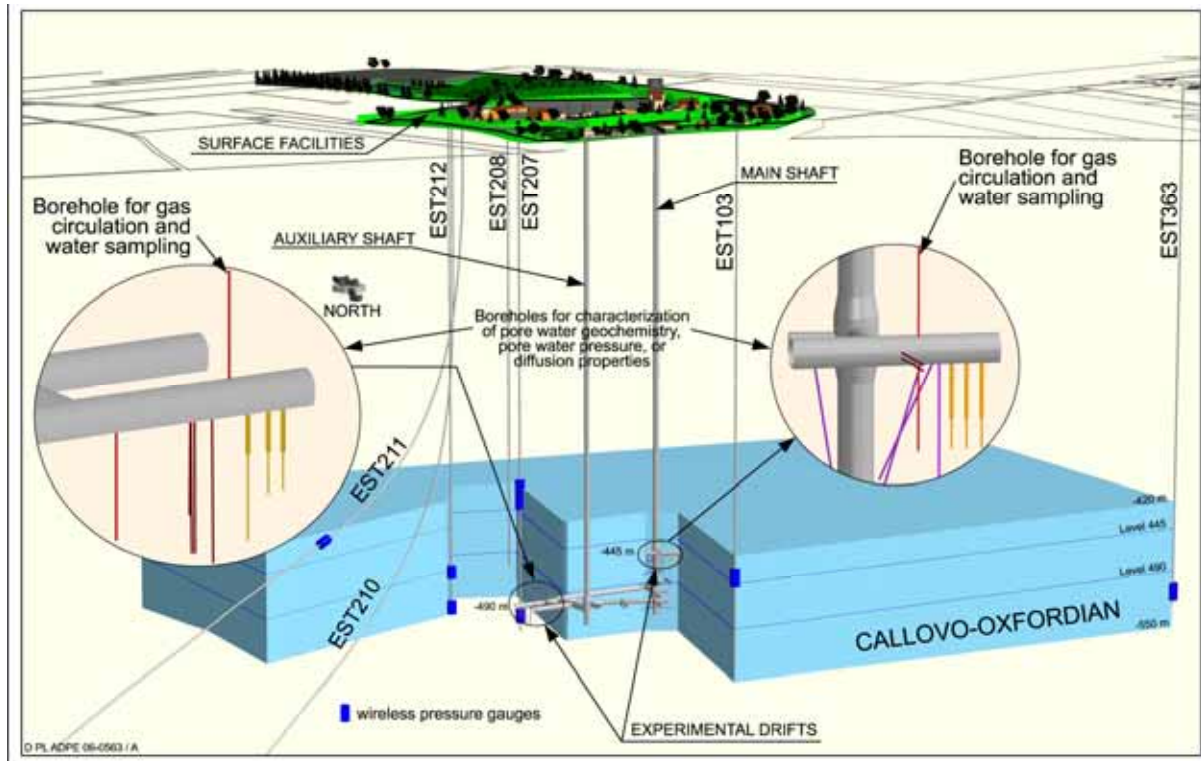


Fig 1. Localization of deep and short boreholes used for Callovo-Oxfordian pore water characterization on the URL site

2 Experimental design

2.1 Drilling phase

The two 15 m long boreholes dedicated to gas circulation and water sampling are vertically ascending. The top 5 meters constitute the test intervals. Taking into account the feedback from the Mont Terri project [2, 3], these boreholes were drilled with nitrogen to avoid rock oxidation. The borehole equipment was installed as quickly as possible after the drilling. Great care was taken to restrict the development of bacterial activity in the boreholes by cleaning the drilling tools and implementing a specific protocol for their handling. The cores provided samples for physical, mineralogical, chemical and microbiological analyses on solids, interstitial fluids and dissolved gases.

2.2 Equipment

The borehole equipment allows the circulation of the gas in contact with the rock in a closed circuit, the sampling of formation water produced in the test interval, and the monitoring of gas and water pressures in the open section. The equipment comprises packers isolating the test interval and 6 gas, water and pressure control lines that link the test interval to the controls in the drift (Fig 2). A slight overpressure of inert gas (nitrogen or argon) was applied to the borehole during the installation of the equipment to prevent oxygen from entering into the test interval. Once the installation completed, the open section and control lines were filled with argon.

The control lines are linked to the so-called “surface” equipment (controls) located in the drift. This equipment consists of pressure gauges connected to a data acquisition system and two modules: a water sampling module and a gas circulation module. The water sampling module allows pumping of the water at a controlled flow rate, online monitoring of the pH, Eh and electrical conductivity of the pumped water and conditioning of the sampled water to avoid contact with the atmosphere. The gas circulation module allows monitoring the evolution of the gas composition due to exchanges with the rock surface of the open section. Monitoring is performed through online infrared spectrometry and gas sampling.

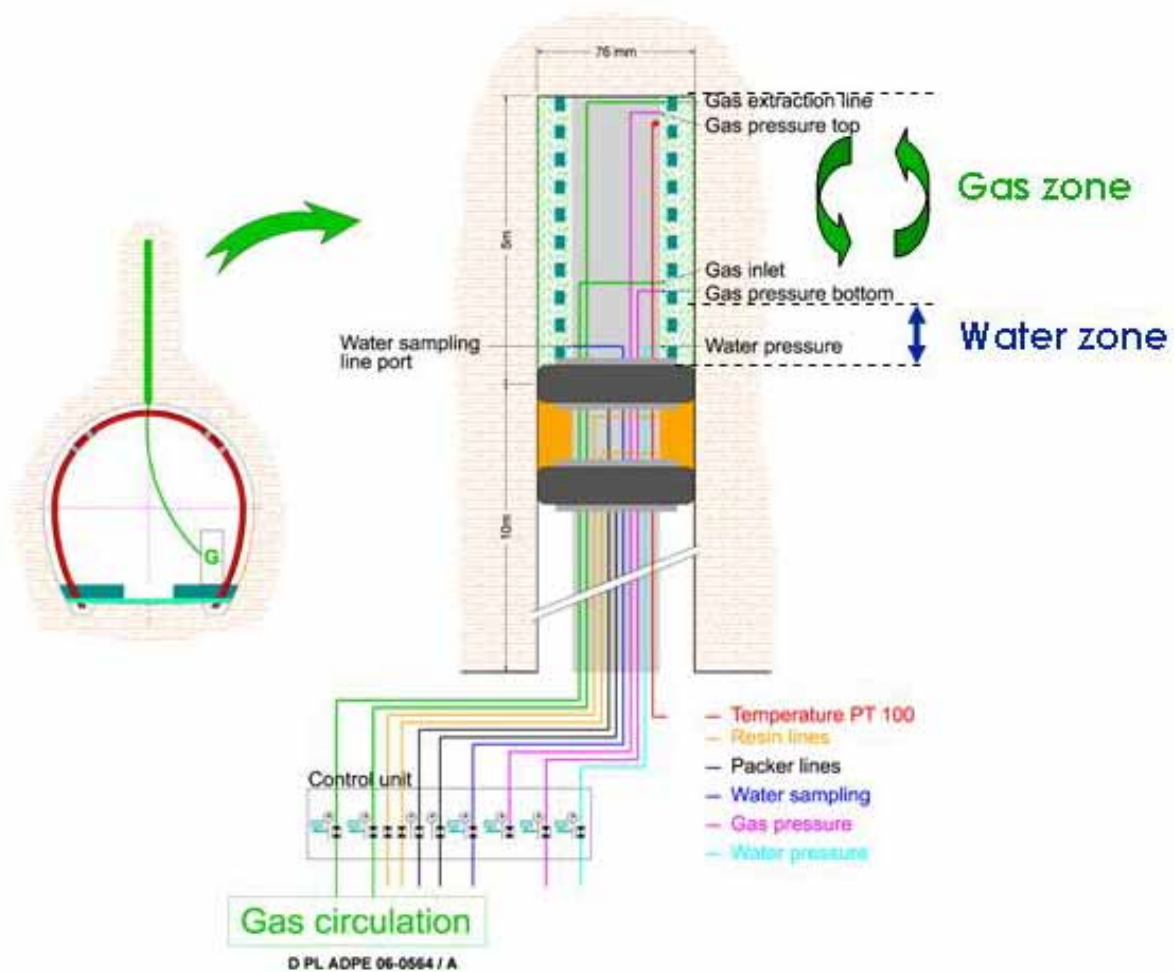


Fig 2. Experimental design for gas circulation and water sampling

2.3 Monitoring and control of the experiment

During the experiment, the water pumping flow rate is adjusted to maintain the water level between 10 and 40 cm at the bottom of the test interval. Above this section, where the water accumulated transiently is pumped out, the gas is circulated in contact with the accumulated water surface and the rock over more than 4.5 m height. Gas pressure was set from the outset and has been maintained throughout between 1.1 and 1.2 bars. These values have induced a high pressure gradient around the test interval.

3 Main results

3.1 Hydraulic properties

The water production in the two boreholes is about 0.5 L per month at -430m (center of the interval of the borehole drilled at -445 m) and 1.3 L per month at -475 m (center of the interval of the borehole drilled at -490 m), respectively. A network of pore pressure gauges measures the natural hydraulic head around the boreholes. Results of calculations obtained through this data show hydraulic permeabilities of about 10^{-13} m/s. These values are coherent with other available measurements at these levels.

3.2 Gas composition

The composition of the circulating gas evolves from pure argon. The main gases are carbon dioxide and methane. The molar fractions of carbon dioxide, methane, and other alkanes up to C5 (pentane), as well as of other gases coming from the rock and its pore water have increased. After more than a year, the gas composition has not stabilized as yet, but the maximum partial pressures observed are close to 2 mbar for both carbon dioxide and methane in the two boreholes.

3.3 Water composition

Fig 3 presents online measurements of pH and Eh carried out on the water pumped in the borehole located in the drift at -490 m (center of the test interval at -475 m). The pH is very stable at 7.3 ± 0.1 . The Eh maximum positive values correspond to calibration phases. Stabilization takes a long time following a calibration phase. Minimum values below -200 mV SHE are considered to be the most representative of the pore water.

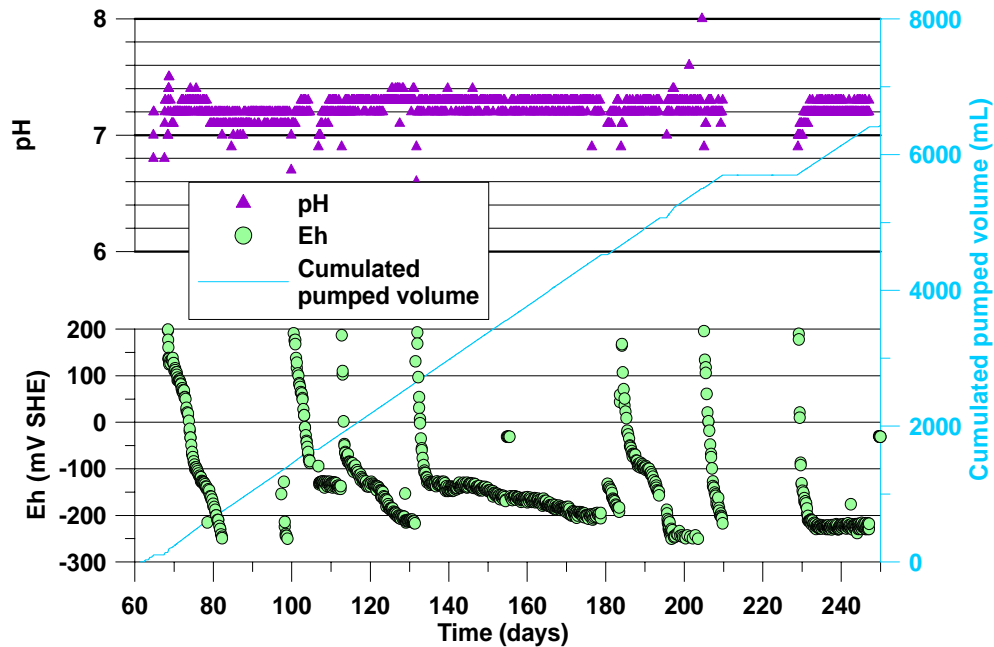


Fig 3. pH and Eh evolution of the pumped water and cumulated volume

Chemical analyses have been performed on the sampled water. Fig 4 shows the results for major species in a Schoeller logarithmic scale diagram. Two of these results (-475 and -430 m) come from the boreholes dedicated to gas circulation and water sampling. Additional analytical results of direct in situ sampling at -490 m were obtained from a borehole dedicated to pore pressure measurements. In this borehole, water inflow was observed for a short period of time following the installation of the multipacker completion. The observed compositions at these three depths are very similar; the main difference concerns chloride concentration. The study of this concentration on cores revealed that it varies slightly with depth [4, 5]. The chloride concentrations observed in the water collected in situ should help refine the values deduced from core measurements and bring new data on the porosities accessible to this species.

The fourth curve of Fig 4 corresponds to the composition worked out through thermodynamic equilibrium modelling before the first in situ collection of water samples. This computation relies on the hypothesis of an equilibrium with the minerals identified in the rock in neofomed states, of reactions of cation exchanges at the surface of the argillaceous minerals and on the average chloride concentration deduced from the core measurements. The constants of these ion exchange reactions were deduced from core tests. [1].

Comparison between the observed compositions and the calculated composition shows a discrepancy below a factor of two for all the species except potassium. For this species an acceptable modification in the model of the potassium and sodium exchange constant leads to the observed concentration.

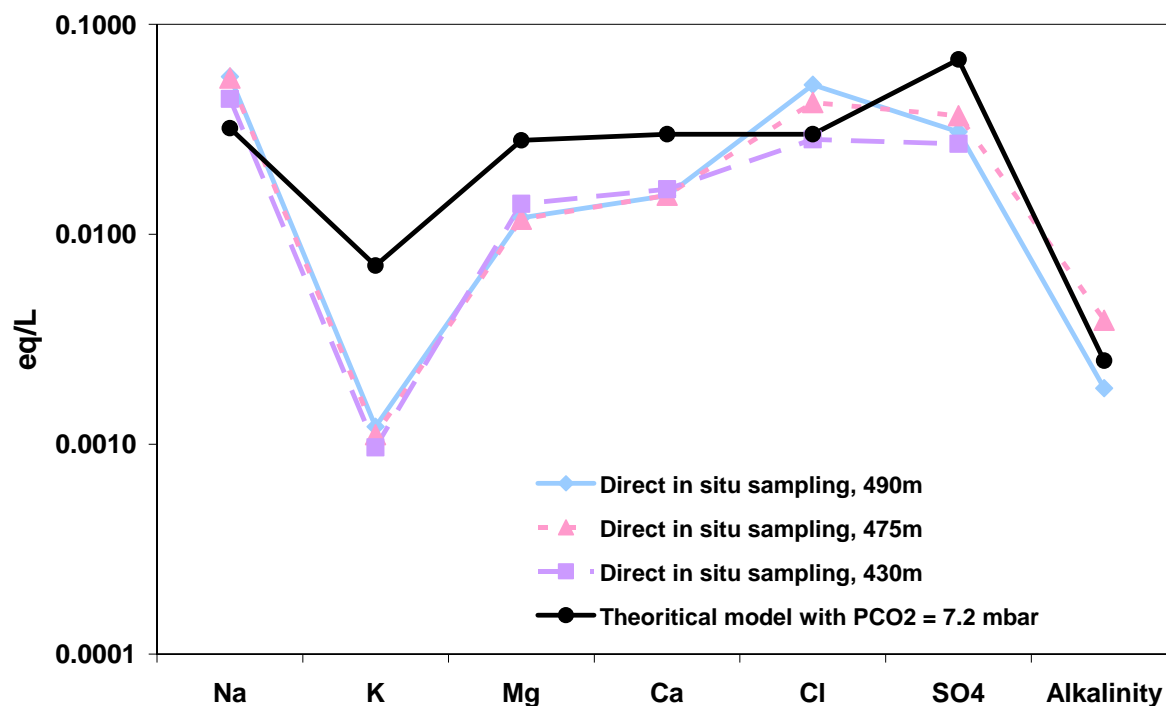


Fig 4. Concentration of major species measured in the pumped water and the composition obtained through thermodynamic modelling

4 Conclusion

The gas circulation and water sampling experiments implemented in the Andra laboratory made it possible and analyse for the first time Callovo-Oxfordian pore water. pH stability over several months demonstrate that the water samples collected through dedicated experiments remain unaltered. The model provided satisfactory prediction for the pore water below a factor 2 for most of the major species. The compositions observed on the water samples should help improve the conceptual model. They confirm the vertical variations of the chloride concentrations and should help refine the results obtained on cores for this species over the entire profile.

5 References

- [1] Andra (2005). Dossier 2005 Argile : Évaluation de la faisabilité du stockage géologique en formation argileuse profonde – Rapport de synthèse, Décembre 2005, Andra, France (available at www.andra.fr).
- [2] Pearson F. J., Arcos D., Bath A., Boisson J. Y., Fernández A. Ma., Gäbler H. E., Gaucher E., Gautschi A., Griffault L., Hernán P., & Waber H. N. (2003). Geochemistry of Water in the Opalinus Clay Formation at the Mont Terri Laboratory. Report of the Federal Office for Water and Geology (Bern, Switzerland), Geology Series, no. 5.
- [3] Vinsot A., Fierz T., Feuillatre L., Blin V. (2006) – PC-C Experiment at Mont Terri – Gas circulation and water sampling – Progress report. Mont Terri Project TN2004-35.
- [4] Lavastre V., Jendrzewski N., Agrinier P., Javoy M. Evrard M. (2005). Chlorine transfer out of a very low permeability clay sequence (Paris Basin, France): ³⁵Cl and ³⁷Cl evidence. *Geochim. Cosmochim. Acta*, **69** (21), 4949–4961.
- [5] Waber H.N., Gimmi T., Mazurek M., Vinsot A. & Gautschi A. (2005). Chloride and Stable Water Isotopes in Pore Water of the Callovo-Oxfordian Shales and Surrounding Formations at the Bure Site, France. International Meeting, March 14–18, 2005, Tours, France. Clays in Natural and engineered Barriers for Radioactive Waste confinement. Abstract Volume 169-170.