Fault and Top Seals
Montpellier 2009

C02

Gas Migration into Low Permeability Callovo-Oxfordian Argillite

J. Talandier* (Andra), F. Skoczylas (LML), B.M. Krooss (RWTH) & M. Prat (IMFT)

SUMMARY

Gas migration into argillaceous formations is an important issue to evaluate the perturbation induced by gas in an underground radioactive waste repository. The mechanisms controlling gas entry and flow into clay media are not fully understood yet and have to be investigated. In that context, Andra and its partners developed an experimental program to characterize the gas behavior of the Callovo-Oxfordian argillite which is the host rock for the French radioactive waste repository. Gas entry pressure and some key parameters such as gas permeability have been measured. We present first the main experimental results obtained on argillite. Then, the analysis of the results leads to discussions about the two-phase flow model used to predict gas migration at the repository scale.

Callovo-Oxfordian argillite located in Eastern France has been retained as a candidate host rock for the disposal of radioactive waste material in deep geological formation. In a radioactive waste repository, several processes will lead to the formation of gas. One of the most important is corrosion of ferrous materials under anoxic conditions and will be at the origin of a large amount of hydrogen. The perturbation induced by gas in such a repository has to be studied and especially its consequences on the host rock.

The understanding of the gas transfer mechanisms in the Callovo-Oxfordian argillite is an essential point when the impact of gases on the repository has to be estimated. The maximum of gas pressure in the repository is clearly dependent on the possibility for the gas to migrate into the large volume offered by the Callovo-Oxfordian argillite. Considering the structure of the argillite, in particular the pore size and connectivity (Andra, 2005), the mechanisms controlling gas entry into and flow within this kind of media are not fully understood yet and have to be investigated.

The gas production will exceed the amount that can be stored by dissolution in the pore water of the argillite (Andra, 2005). Gas will continue to accumulate until its pressure becomes sufficiently large to enter the host rock. Beyond this pressure, a two-phase (gas/water) flow is initiated leading, under the condition of storage (slow gas rate production, gas pressures below the fracture pressure in the order of 12MPa) to a slow and weak desaturation of the argillite.

To measure gas entry pressure in Callovo-Oxfordian argillite, several experiments have been set up including direct measurements (gas injection at a slow rate into a totally saturated sample) and some indirect methods as those proposed by Hildebrand *et al.* (2002). Some of these experiments realised in LML seem to converge to low values of gas entry pressure for argillite between 2 and 4MPa. This is confirmed by some other recent works dedicated to the gas argillite behaviour as in Boulin (2008), or in Yang (2008). At the same time, experimental results obtained by RWTH suggest higher entry pressures as evidenced by the residual pressure differences (ΔP) of 5.3 and 4.75 MPa in Figure 1(a). The continuing convergence of the upstream and downstream pressure curves could indicate that capillary pressure equilibrium (minimum capillary displacement pressure) has not been reached within the duration of the experiment. Alternatively it is attributed to diffusive gas transport across the water-saturated argillite sample.
The influence of some environmental parameters on the gas entry pressure has been studied too, as the dependency with the dynamic of gas injection, the stresses imposed on the sample, some scale effects and the nature of the injected gas. One of the most important difficulties in gas experiments is to be able to reproduce the very slow increasing in gas pressure which happens in the underground repository. For example, the increase due to the corrosion of metal (considering corrosion rates of about 3 to 5 µm/years) could be less than about 1MPa/100 years around vitrified waste canisters (Andra, 2005). The loading of the material in that case should obviously be very different of what could be done in a laboratory especially in terms of mechanical response.

To complete this work, data on further key parameters for gas transfer in porous media have been acquired, such as relative permeabilities to gas and water, water retention curves(figure 1, (b)), and the Klinkenberg effect as function of water saturation.

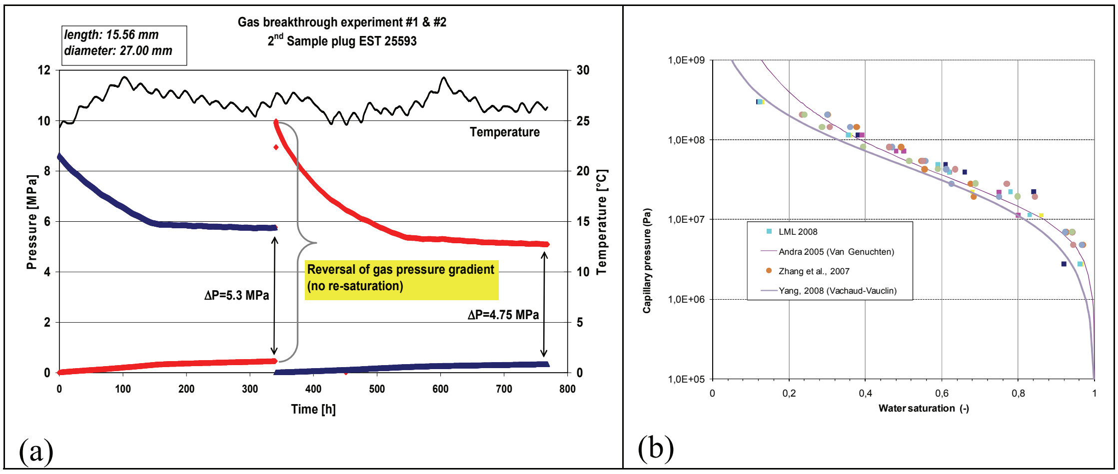

Figure 1 *(a) Experimental results from gas breakthrough measurements on argillite, gas pressure history in upstream and downstream cell compartments; ΔP is the final differential pressure (RWTH). (b) Relationship between capillary pressure and degree of water saturation, comparison of experimental data (LML) and results from literature.*

The low degree of desaturation due to gas in the repository context requires determination of these transfer properties at high values of water saturation (over 90 %). Meanwhile, the capillary number which has been estimated by deducing the velocity from the gas production for a vitrified waste is very low $Ca \approx 10^{-13}$-10^{-14}. These very low values give rise to question about the flow regime that could tend in that case, toward a capillary fingering regime (Lenormand *et al.*, 1988). This involves a critical examination of the validity of the models conventionally used to represent two-phase flow in (coarse-grained) porous media, which are mainly based on the generalised Darcy's law.
One part of this work concerns the potential limits of application of these conventional models. The percolation theory in conjunction with the experimental results has been used to investigate that particular point.

We propose in this paper, a two-step approach with a synthesis of the experimental studies and numerical modelling work to interpret and understand the processes involved in the gas transport in Callovo-Oxfordian argillite. Subsequently, the different results will be analysed to propose a coherent two-phase flow model applicable to predict gas migration at the repository scale.

Andra [2005] Dossier 2005, Référentiel du site Meuse/ Haute-Marne. ANDRA Report n°C.RP.ADS.04.0022.
Boulin P., [2008], Expérimentation et modélisation du transfert d'hydrogène à travers des argiles de centre de stockage de déchets radioactifs, Thèse de Doctorat de l'INPG Grenoble.
Hildenbrand A., Schloemer S., Krooss B. M. [2002] Gas breakthrough experiments on fine-grained sedimentary rocks, *Geofluids,* **2**, 3-23.
Lenormand, R., Touboul, E., Zarcone,C., [1988], Numerical models and experiments on immiscible displacements in porous media, *J.Fluid Mech*., 189, pp;165-187.
Yang D., [2008] Caractérisation par la mesure de perméabilité au gaz de l'endommagement mécanique et hydrique dans l'EDZ des argilites du Callovo-Oxfordien, Thèse de Doctorat de l'Ecole des Mines de Paris.
Zhang C.L., Rothfuchs T. (2007a) – TIMODAZ, Laboratory experiments on the THM behaviour of clay rocks, FP6-036449.