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Gas migration properties through a bentonite/argillite interface

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ABSTRACT

Among diversified industrial uses, see for instance Koch [Koch, Appl. Clay Sci., (21) 2002], and following positive *in situ* experiments, compacted bentonite blocks are potential candidates for sealing nuclear waste repositories, thanks to their swelling ability in a wet environment.

As requested by Andra (French Agency for Nuclear Waste Management) and complementarily to *in situ* experiments, an original experimental laboratory set-up was designed in order to reproduce the introduction and swelling of bentonite plugs inside an argillite host rock. Once the argillite/bentonite interface is established, an increase in storage tunnel gas pressure is simulated and the interface gas migration pressure (or *gas critical pressure*) is evaluated. More precisely, a first experimental set-up provides bentonite swelling pressure and kinetics (i.e. mainly hydraulic cut-off, time to reach asymptotic swelling pressure and value of asymptotic swelling pressure) at given initial compaction and saturation rate. This phase is preparatory to reproducing the introduction and subsequent swelling of a bentonite plug inside the argillite host rock, which uses a similar test rig. Experimental results of water permeability and gas critical pressure are provided for MX80 compacted bentonite associated to Bure Callovo-Oxfordian argillite.

1. Introduction

High-level and long-lived nuclear waste storage on the long term has become a key issue for industrial countries, so that it has been extensively investigated for the last twenty-five years. In particular, clayey geological formations situated away from reputed seismic areas have been selected for their potentially high impermeability and stability. In France, Andra (National Agency for Nuclear Waste Management) is investigating a deep underground storage solution (Lebon and Mouroux, 1999) inside a Callovo-Oxfordian argillite lying in the East, in both Haute-Marne and Meuse French Départements. This formation welcomes a full-scale underground research laboratory still under construction (Fouché et al., 2004).

The deep-seated nuclear waste repository generally consists of a main access tunnel drilled in clayey (or crystalline) rock, in which disposal pits are aligned (Komine, 2004; Montes-H et al., 2005), each of them containing a waste canister. Wastes are vitrified (or cemented, or bituminized, see Mügler et al., 2006), confined in a copper/steel canister before being surrounded by pre-compacted swelling clay blocks in the disposal pit (Lloret et al., 2003; Martin and Barcala, 2005; Victoria Villar and Lloret, 2007), the whole system thereby forming an engineered barrier to potential radionuclide leakage through the host rock. Pre-compacted swelling clay is also considered for global repository tunnel sealing. Over time, such a clay should absorb surrounding groundwater and swell, closing up any construction clearance (Horseman et al., 1999). This effect is known as *buffering*. Several engineered compacted clays are considered (Lloret et al., 2003). A large majority of them are made up from naturally swelling sodium bentonites, and are composed in majority of montmorillonite (Mitchell, 1993; Neaman et al., 2003; Andra, 2005). In particular, MX80 bentonite, as used in this study, is composed of ca. 90% montmorillonite (Horseman-99).

For performance and safety assessment purposes, varied damage and failure scenarios are investigated. In particular, humid corrosion of copper/steel canisters may occur on the long term, due to groundwater seepage through the engineered barrier. Coupled to radioactive waste decay and radiolysis of water, humid corrosion may induce hydrogen gas production (Horseman et al., 1999). Gradually, hydrogen gas pressure could increase notably, first, inside disposal pits, at the interface between waste metal canisters and clay plugs, and, subsequently, inside the repository tunnel. Whenever gas pressure reaches groundwater pressure, the capillary threshold for gas entry into the clay is reached, whereby hydrogen gas leakage may occur through the repository sealing plug. Horseman et al. (1999) present evidence that this eventuality is much less probable than that of the creation of micro-cracks and fracturing of the pre-compacted sealing clay due to gas pressure increase, so that, finally, H_2 gas (and later, radionuclides) may leak from the repository.

Gas migration through bulk compacted sealing clays has been extensively investigated. As reported by Gallé (2000); Hildenbrand et al. (2002), gas migration mechanisms involve the creation and

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propagation of preferential pathways throughout the water-saturated clay. Depending on the gas pressure value, these pathways may be unstable and lead to intermittent gas flow. Nevertheless, it is generally admitted that gas convective transport in fully water-saturated clay is possible whenever gas pressure marginally exceeds the sum of clay swelling pressure and interstitial water pressure (Horseman et al., 1999; Gallé, 2000; Hildenbrand et al., 2002). For instance (Horseman et al., 1999), for a fully water-saturated bentonite of initial 1.6 g cm^{-3} dry density, the expected swelling pressure is 7 MPa. When such bentonite is subjected to 5 MPa interstitial water pressure, which corresponds *in situ* to 500 m depth, gas migration is expected at a pressure slightly above 12 MPa. Similar prediction is provided by Gallé (2000) for Fo-Ca clay. Horseman et al. (1999) also points out the potential importance to gas migration properties of the boundary conditions for a swelling clay. Indeed, gas migration through a compacted swelling clay is generally accommodated by global dilation. Yet, no investigation has been made to our knowledge as to the influence of oedometric boundary conditions upon the gas migration properties of swelling clays. Finally, Gallé (2000) proposes a model for gas migration at the Fo-Ca clay/metal overpack interface, which predicts an order of magnitude similar to gas breakthrough pressure: the gas migration phenomenon may not be neglected for long-term disposal safety assessment.

Complementarily, our study aims at characterising the gas migration conditions through the bentonite/host rock *interface*, by using laboratory experiments. An originality of this work is that it reproduces in the laboratory 1) the *in situ* positioning of an unsaturated bentonite plug inside a brittle clayey rock (namely argillite), 2) the bentonite swelling, followed by 3) a gas pressure increase. Although each test may last more than a month, this experiment provides additional information upon swelling, transport properties and gas interface migration to those obtained during similar *in situ* experiments (Van Geet et al., 2007). All along the study, argillite swelling is assumed negligible compared to bentonite swelling.

2. Experimental methodology

All tests are performed in a temperature-controlled room at 22 °C. Two different versions of the experimental set-up are proposed. Both are based upon former set-ups designed in the laboratory for low permeability measurements of bulk materials (Skoczylas and Henry, 1995; Loosveldt et al., 2002; Davy et al., 2007). These consist of placing a specimen inside a triaxial cell and subject it to both confining pressure and interstitial liquid or gas flow, see Fig. 1. For all tests, a 12 MPa confining pressure is used. This reproduces the *in situ* lithostatic pressure at approximately 500 m depth, given that argillite density is evaluated at 2.4 (Wright, 2001).

One set-up version aims at characterising bentonite swelling kinetics and pressure by surrounding initially unsaturated bentonite with an instrumented and calibrated tube (phase 1). Several versions of the tube ductile material (aluminium, Plexiglas™ or aluminium–Plexiglas™) are tested in order to retain the most sensitive and reliable one. The second set-up version reproduces the *in situ* swelling conditions. A cautious swelling procedure is devised and validated, which reproduces the adequate positioning of a bentonite plug inside the argillite host tube while avoiding argillite failure. The bentonite–argillite specimen (or sample) is referred to as a *mock-up*, since it represents a reduced-scale tunnel seal. After bentonite swelling, water permeability of the mock-up is derived from indirect fluid flow rate measurements (or *pulse test technique*), see Skoczylas (1996); Meziani and Skoczylas (1999); Loosveldt et al. (2002); Davy et al. (2007), and gas critical pressure at the bentonite/argillite interface is determined (phase 2).

2.1. Materials

All tests are performed on argillite provided by Andra from a single plug MSE 00707 cored at a height of 555 to 556 m at the Bure underground laboratory site. The water pressure of 4 MPa imposed in our experiments is therefore a lower bound value of the *in situ* water column pressure. Bentonite plugs are made in our laboratory from MX80 powder provided by Eurogeomat-Consulting company (Orléans, France) (Hökmark et al., 2007). Its average grain size, evaluated using a Beckman-Coulter™ LS230 laser granulometer, is 264±2 µm.

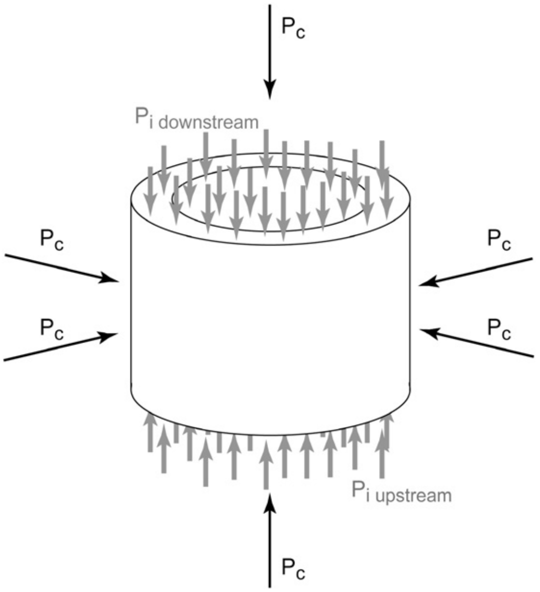

Fig. 1. Triaxial loading sustained by an argillite/bentonite interface specimen (resp. by a ductile material/bentonite specimen).

2.2. Mock-up design

The mock-up geometry and dimensions are chosen according to various constraints, with proper representativeness as a main focus. First, a circular cylindrical geometry ensures uniform loading conditions in a triaxial cell. The triaxial cell is chosen to reproduce surrounding rock hydrostatic loading upon the sample. Besides, bentonite tunnel seals have this overall geometry (Lloret et al., 2003; Komine, 2004). Existing laboratory experience upon argillite confinement involves 65 mm diameter (or alternately 37 mm diameter) circular cylindrical specimens, which guarantee material representativeness relatively to their micro-structure scale (this scale is fixed at 10 µm in Abou-Chakra Guéry et al., 2008). Consequently, the geometry chosen for the interface specimen is such that the argillite host rock corresponds to a tube of 65 mm outer diameter, 40 mm inner diameter, and 50 mm height. Four argillite tubes have been cored for this study, using a purposely-built diamond circular cylindrical double saw, with extra precautions aimed at avoiding argillite shattering during the process. After coring, the tube top and bottom surfaces are ground in order to guarantee proper planeity and parallelism, see the result in Fig. 3(b). Tube n.4 has displayed one single major macro-crack which resulted in splitting the tube in two parts (of about (1/4)th and (3/4)th of the total tube volume). We have chosen to seal it with a thin layer of PC12 glue (usually used to glue strain gauges) and to use the tube to make sample n.4.

Bentonite is a circular cylindrical plug placed inside the argillite tube, see Fig. 1. In order to reproduce *in situ* conditions, Andra requested that the bentonite plug should have an initial of 3 to 4% clearance inside the tube inner volume, see Section 2.2.1 for details.

For bentonite swelling characterisation, a mock-up of identical dimensions is chosen. Yet, the tube is made of a ductile material less prone to failure than argillite; it is either made of aluminium (two tubes have been made for this study), of Plexiglas™ (one tube), or of a