Sealing efficiency of an argillite-bentonite plug subjected to gas pressure, in the context of deep underground radioactive waste storage

FORGE Report D3-08; D3-24, D3-30 and D3-35 – VER.0

<table>
<thead>
<tr>
<th>Name</th>
<th>Organisation</th>
<th>Signature</th>
<th>Date</th>
</tr>
</thead>
<tbody>
<tr>
<td>Compiled</td>
<td>Dr. Jiang Feng Liu; Pr. Frédéric Skoczylas; Dr. Catherine Davy</td>
<td>Ecole Centrale de Lille – LML</td>
<td>4th December 2013</td>
</tr>
<tr>
<td>Verified</td>
<td>RP Shaw</td>
<td>BGS</td>
<td>7th December 2015</td>
</tr>
<tr>
<td>Approved</td>
<td>RP Shaw</td>
<td>BGS</td>
<td>7th December 2015</td>
</tr>
</tbody>
</table>

Keywords
Bentonite; argillite; Callovo-oxfordian, gas

Bibliographical reference
Dr. Jiang Feng Liu; Pr. Frédéric Skoczylas; Dr. Catherine Davy, 2013. Sealing efficiency of an argillite-bentonite plug subjected to gas pressure, in the context of deep underground radioactive waste storage. FORGE Report D3-08; D3-24, D3-30 and D3-35. 112pp.

Euratom 7th Framework Programme Project: FORGE
Fate of repository gases (FORGE)

The multiple barrier concept is the cornerstone of all proposed schemes for underground disposal of radioactive wastes. The concept invokes a series of barriers, both engineered and natural, between the waste and the surface. Achieving this concept is the primary objective of all disposal programmes, from site appraisal and characterisation to repository design and construction. However, the performance of the repository as a whole (waste, buffer, engineering disturbed zone, host rock), and in particular its gas transport properties, are still poorly understood. Issues still to be adequately examined that relate to understanding basic processes include: dilational versus visco-capillary flow mechanisms; long-term integrity of seals, in particular gas flow along contacts; role of the EDZ as a conduit for preferential flow; laboratory to field up-scaling. Understanding gas generation and migration is thus vital in the quantitative assessment of repositories and is the focus of the research in this integrated, multi-disciplinary project. The FORGE project is a pan-European project with links to international radioactive waste management organisations, regulators and academia, specifically designed to tackle the key research issues associated with the generation and movement of repository gasses. Of particular importance are the long-term performance of bentonite buffers, plastic clays, indurated mudrocks and crystalline formations. Further experimental data are required to reduce uncertainty relating to the quantitative treatment of gas in performance assessment. FORGE will address these issues through a series of laboratory and field-scale experiments, including the development of new methods for up-scaling allowing the optimisation of concepts through detailed scenario analysis. The FORGE partners are committed to training and CPD through a broad portfolio of training opportunities and initiatives which form a significant part of the project.

Further details on the FORGE project and its outcomes can be accessed at www.FORGEproject.org.

Contact details:

Dr. Catherine Davy
Maître de conférences HDR/ Lecturer
Ecole Centrale de Lille
Cité Scientifique
CS 20048
F-59651 Villeneuve d'Ascq Cedex
France
E-mail: catherine.davy@ec-lille.fr
Tél: (+33).3.20.33.53.62
Fax: (+33).3.20.33.53.52

Page web (version francaise): http://cdavy.ec-lille.fr/
Webpage (English version): http://cdavy.ec-lille.fr/davyev.html
Profil Viadeo: http://www.viadeo.com/invitation/catherine-a..davy
WP3 FORGE PROJECT

Report corresponding to deliverables n.:
D3-08; D3-24, D3-30 and D3-35

Sealing efficiency of an argillite-bentonite plug subjected
to gas pressure, in the context of deep underground
radioactive waste storage

Dr. Jiang Feng Liu

Pr. Frédéric Skoczylas

Dr. Catherine Davy

Ecole Centrale de Lille – LML, UMR CNRS 8107
Contents
I - Introduction: scientific context.............................................................................................. 5
II – Materials and experimental methods................................................................................... 9
   II.1 Materials and sample preparation.................................................................................. 10
   II.2 Water retention experiments......................................................................................... 11
      II.2.1 Analysis of in situ problem.................................................................................. 11
      II.2.2 Water retention tests under constant volume and free swelling conditions.......... 11
   II.3 Swelling of compacted bentonite (into tube) with gas pressure and water contact...... 15
      II.3.1 Analysis of in situ problem and mock-up description........................................ 15
      II.3.2 Definition of total equilibrium swelling pressure and effective swelling pressure..... 18
   II.4 Gas breakthrough test .................................................................................................... 18
      II.4.1 Why performing the GBT? ................................................................................ 18
      II.4.2 Introduction of experimental method to measure the gas passage....................... 18
      II.4.3 Definition of discontinuous/continuous gas breakthrough.................................. 20
III - Water retention tests under constant volume and free swelling conditions...................... 21
   Introduction .......................................................................................................................... 22
   III.1 Water retention tests under constant volume conditions.............................................. 23
   III.2 Water retention tests under free swelling conditions ................................................... 25
      III.2.1 Water retention tests under free swelling conditions and just after compaction..... 25
      III.2.2 Water retention tests under free swelling conditions (after gas permeability tests)29
   III.3 Comparison of water retention tests under free swelling and constant volume
       conditions (just after compaction).................................................................................. 32
   III.4 Conclusion.................................................................................................................... 34
IV - Sealing ability of partially water-saturated bentonite/sand plugs under the effect of
    confinement .............................................................................................................................. 35
   Introduction .......................................................................................................................... 36
   IV.1 Relative gas permeability – preliminary test results .................................................... 36
   IV.2 Relative gas permeability study – second series of tests............................................. 40
      IV. 2.1 Mass and volume changes.................................................................................. 40
      IV. 2.2 Initial sample gas permeability.......................................................................... 45
      IV. 2.3 Coupled effects of saturation and confining pressure upon gas permeability....... 47
IV.2.4 Dry permeability ............................................................................................................... 53
IV.3 Complementary tests (third test series) ............................................................................... 54
IV.3.1 Mass and volume changes................................................................................................. 54
IV.3.2 Effective gas permeability at given RH ............................................................................ 57
IV.3.3 Dry gas permeability ......................................................................................................... 59
IV.3.4 Pore volume variation of an oven-dried bentonite/sand plug ............................................ 60
IV.4 Conclusion ................................................................................................................... 62

V - Swelling of compacted bentonite/sand plug (into tube) submitted to gas pressure ........... 64

Introduction .......................................................................................................................... 65

V. 1 Bentonite-sand plug swelling without gas pressure .......................................................... 66
   V.1.1 Swelling pressure (SWP) .............................................................................................. 66
   V.1.2 Gas breakthrough test (GBT) ...................................................................................... 68

V.2 Bentonite-sand plug swelling with gas pressure ............................................................. 70
   V.2.1 Effect of gas pressure on the swelling pressure of bentonite-sand plug ..................... 70
   V.2.2 Effect of gas pressure on the gas breakthrough pressure ............................................. 72
   V.2.3 Effect of re-saturation and/or a decrease in gas pressure ............................................ 73
   V.2.4 Effect of gas pressure on plug-tube interface contact pressure ................................. 77

V.3 Effect of sample height upon swelling and GBP ............................................................. 78
   V.3.1 Swelling pressure (SWP) ............................................................................................ 79
   V.3.2 Coupling effect between gas pressure and deformation of bentonite-sand plug .......... 80
   V.3.3 Gas breakthrough test .................................................................................................. 82
   V.3.4 Re-saturation of the sample ......................................................................................... 83

V.4 Conclusion .................................................................................................................... 84

VI - Swelling and GBP of argillite, bentonite and argillite / bentonite interface ............... 86

Introduction .......................................................................................................................... 87

VI.1 Swelling and GBP of bentonite/sand plug without tube (samples D1 and D2) ............... 87
   VI.1.1 Water injection test of plugs D1 and D2 .................................................................... 87
   VI.1.2 Gas breakthrough test of plugs D1 and D2 ............................................................... 89

VI.2 Swelling and GBP of a bentonite-sand plug with a grooved tube (plugs E1 and E2) .... 90
   VI.2.1 Water injection test of plugs E1 and E2 ................................................................. 90
   VI.2.2 Gas breakthrough test of plugs E1 and E2 ............................................................. 93

VI.3 Swelling and GBP of bentonite/sand plug+smooth argillite tube ............................... 96
VI. 3.1 Water injection tests of plugs F1-1, F1-2 and F1-3 ........................................................... 96
VI. 3.2 Gas breakthrough tests of plugs F1-1, F1-2 and F1-3 ....................................................... 97
VI.4 Swelling and GBP of bentonite/sand plug+grooved argillite tube .............................. 99
  VI.4.1 Water injection test of plugs F2-1, F2-2............................................................................. 99
  VI.4.2 Gas breakthrough tests of plugs F2-1, F2-2 ................................................................. 100
  V. 5 Conclusion .................................................................................................................. 103

General Conclusion ................................................................................................................ 105

Reference: ............................................................................................................................... 107

Annex 1 .................................................................................................................................. 110

Annex 2 .................................................................................................................................. 111
I - Introduction: scientific context
The design of long term nuclear waste repositories includes the building of an engineered barrier around the waste containers, which aims to create a “low permeable zone” around them (Komine, 2004; Alonso et al., 2006). Bentonite clay has been chosen by several industrialized countries as a buffer and backfill material to separate radioactive waste from the surrounding host rock. Its main properties are an extremely low permeability, a self-healing ability, low ion transport capacity and high chemical stability, together with high expandability (Kaufhold et al., 2007).

In situ, after water uptake from the host rock, sealing will be obtained due to bentonite swelling. It will fill the space between the buffer material and the disposal pit wall. Meanwhile, formation of gas, mainly hydrogen, due to humid corrosion, degradation of organic matter or water radiolysis, is unavoidable within galleries (Birgersson et al., 2008). During this process, several questions should be answered to understand the sealing efficiency of an argillite-bentonite plug subjected to gas pressure, and more specifically:

![Graph](image)

Figure I.1 Water content (top diagram) and dry density (lower diagram) of a vertical slice of the FEBEX barrier made of bentonite, as measured along six different radial lines from the center of the gallery, at the FEBEX in situ experience (Villar et al., 2005). The initial dry density of the FEBEX bentonite is 1.69-1.7 g/cm$^3$, the initial water content is about 14%.

1- Sealing ability of partially water – saturated bentonite/sand plugs under confinement

For sealing a repository gallery, the clay barrier is constituted by blocks of compacted bentonite arranged on vertical slices, which are put in place with initial construction gaps
(Villar and Lloret, 2006). While fully water-saturated bentonite provides swelling capacity and low permeability. Bentonite/sand mixtures are usually compacted at an intermediate water content \((w \approx 10-15\%)\), and they are progressively wetted by water coming from the host rock formation (Wang et al., 2011; King et al., 2010). It is expected that, during the process of saturation, a significant water saturation gradient will be present between the core and the external surface, itself in contact with the host rock and the underground site water, see Figure I.1 (Villar et al., 2005; Villar and Lloret., 2007) (in the case of the FEBEX in situ experiment). Besides, owing to a high water content, the external part of the massive barrier swells in contact with an extremely stiff host rock, so that it applies a confining pressure to the partially water- saturated core. In this context, it is essential to investigate the sealing ability of the central part of the bentonite/sand barrier under confinement.

2- Effect of gas pressure on water saturation and swelling of bentonite/sand plug

After closing these tunnels, the bentonite/sand plugs will be hydrated by pore water infiltrated from the host rock. In parallel, if the gas generation rate exceeds the flow capacity of dissolved gas by diffusion, or the viscosity capillary flow, gas pressure will increase gradually. One critical scenario to be investigated is that of the effect of such gas pressure upon swelling of the bentonite/sand plugs and sealing efficiency of the disposal pit (e.g. bentonite, argillite and bentonite-argillite interface).

3- The pathway of gas migration: through the argillite, bentonite or argillite-bentonite interface?

After water saturation and swelling of bentonite, a weaker zone is expected as regards gas flow: it could be either the host rock, or the bentonite buffer, or the contact zone between the rock host and bentonite, see Figure I.2. This issue is clearly presented in an in situ experiment initiated by Andra at Bure (East of France). At the laboratory scale, the questions we propose to answer is the following: if gas is injected through a water-saturated mixed plug constituted of a swollen bentonite/sand plug placed within an argillite cylinder (in order to reproduce the tunnel seal), at what pressure and where will this gas flow through (argillite, bentonite/sand mixture or through their interface)?
Figure I.2 Schematization of the possible gas migration pathways in the Opalinus Clay (Marschall et al., 2008).
II – Materials and experimental methods
II.1 Materials and sample preparation

Based on preliminary studies performed by Andra (Bosgiraud, 2004), a single bentonite/sand mix is used throughout this study. It consists of 30%wt TH1000 silica sand and 70%wt GELCLAY WH2 sodic bentonite, taken from a single powder mix provided by the CEA (France), of batch reference RE08015. GELCLAY WH2, also called MX80 WH2 which consists of a pure sodium bentonite from Wyoming (USA). Its average chemical analysis is: 62.90 % SiO₂, 19.70 % Al₂O₃, 4.09 % Fe₂O₃, 2.32 % MgO, 2.32 % Na₂O, 1.28 % CaO, 0.58 % K₂O and 5.86 % loss on ignition. TH1000 sharp silica sand is made of 99.0 % SiO₂ (quartz); its grain size distribution is in the range [0.2-2mm], which is similar to that of GELCLAY WH2. The sand solid density is of 2.65 g/cm³, that of GELCLAY WH2 is of 2.78 g/cm³. More details on the mix characteristics are given in Gatabin (2005).

The target swelling pressure should be obtained for a mixture compacted at a dry density of 1.77g/cm³ with a water content \( w \) of 15.2%. To obtain the adequate water content, before the compaction operation, the bentonite-sand mixture is let in an atmosphere at controlled relative humidity (RH=85%). It is then compacted at 12MPa axial pressure in a steel tube. The resulting sample is 12.5mm/25mm height and 37mm/42.5mm diameter, see Figure II.1.

![Figure II.1 Bentonite/sand plugs obtained after compaction](image-url)
II.2 Water retention experiments

II.2.1 Analysis of in situ problem

In order to assess the safety of the storage system, it is essential to develop appropriate model to predict the long-term behavior of the buffer materials. During the process of long-term storage, the saturation kinetics of the bentonite barrier is one of the most important features. Consequently, in order to model the hydration of the buffer material, it is essential to investigate the water retention property of partially saturated bentonite/sand plug and its permeability. In addition, water infiltration into bentonite was not only dependent on the initial state of the compacted materials but also on the boundary conditions. Therefore, this part focuses on the water retention properties of compacted bentonite/sand plugs under both free swelling and constant volume conditions (Wang et al., 2012; Cui et al., 2008).

II.2.2 Water retention tests under constant volume and free swelling conditions

II.2.2.1 Plexiglass™-aluminium tube and their calibration

Two types of tube have been manufactured, of either 50mm or 25mm height; both types have 42.5 mm internal diameter. The smaller ones are used for the PGZ experiment (see in a next section) where as the higher ones will be used for poro-mechanical testing and some flow tests through Plexiglass™-bentonite interfaces. Each tube has been calibrated in a triaxial cell specially designed for Forge (in fact four cells were manufactured for this project). The tube is placed in the cell and wrapped in a Viton™ jacket, see Figure II.2 ~ Figure II.3. Oil is used to confine the tube up to 12MPa. Therefore gas is injected in the tube at different pressures $P_i$ that simulates the sample swelling. The 4 gauges record the lateral deformation of the tube in order to link the strains to the future swelling pressure due to bentonite, see Figure II.4. Very good linearity and reversibility were obtained for each tube tested.
Figure II.2 Tube in a triaxial cell – first mounting step

Figure II.3 Tube wrapped in a Viton membrane
2.2.2.2 Thermal perturbations (thermal effect): adjustments

It has been documented in some literatures that the change of temperature will induce significant variations in bentonite swelling behavior (Villar and Lloret, 2004; Ishimori and Katsumi, 2012; Ye et al., 2012). According to experimental data, strain gauges are quite sensitive to the fluctuation of temperature. It should be mentioned here that each test, performed in this study, lasts a long time (usually several months). Even if it is carried out in an air-conditioned room, small perturbation of temperature may occur, see Figure II.5 (a). This is sufficient to slightly deviate from the actual results. As a consequence, a reference tube was used, which was not placed inside the triaxial cell but in the room beside it, in order to correct for thermal strains. Figure II.5 (b) shows that the average value of relative strains is as a function of temperature. The observed relationship between temperature and the average strains means that the strains will increase or decrease 23.17 με/d if the temperature changes by one degree. Therefore, it is essential to consider thermal effect when calculating the swelling pressure. However, scarcely any literature published considers about the correction of thermal perturbation.

Figure II.4 Example of 50mm height tube calibration: the average value is used to link the strains to the swelling pressure
2.2.2.3 Experimental procedure and definition of the tests performed

In order to obtain uniformly partially water-saturated bentonite/sand plugs, each was placed in a hermetic chamber at given relative humidity (RH) of 75%, 85%, 92%, or 98%. These relative humidities are provided by various salt solutions. Full water saturation was assumed achieved through mass stabilization at a hermetic chamber at 100% RH (over pure distilled water), while for the constant volume conditions sample was put in a triaxial cell to inject...
water directly to accelerate the saturation process. Within each hermetic chamber, the sample was put right above the water surface, where the actual $RH$ was closest to the required value.

The methodology used was as follow for both tests:

- Samples compaction as described above.
- Weighing of the different samples and pieces of the experimental mounting (tube, plates, etc.).
- Equilibrium at RH=70 (for samples SO1 and SO2), 75, 85, 92, 98 % (until mass stabilization) and then full water saturation in order to obtain saturated mass and to deduce the degree of saturation.
- Some samples were chosen to perform gas permeability tests

For the samples that swelled under constant volume conditions, it had been chosen to make the bentonite/sand plug swell in a tube while the radial deformations were obstructed by the inner surface of the tube and the axial strains were blocked by the use of two porous plates, see Figure II.6.

![Figure II.6 Compacted bentonite/sand plugs swell under free swelling and constant volume conditions.](image)

**II.3 Swelling of compacted bentonite (into tube) with gas pressure and water contact**

**II.3.1 Analysis of *in situ* problem and mock-up description**

The design of the mock-up aims to reproduce the *in situ* situation, where the compacted bentonite plug is placed in the gap between the host rock and concrete structure (or metal). They may also be designed as plugs to constitute and achieve, locally, the sealing systems.
Whatever the destination of these mixtures, there will be a direct contact between bentonite and argillite. Underground water will infiltrate through argillite to the bentonite, and this will lead to the swelling of the bentonite/sand plug. It is expected, following partial desiccation of argillite, due to underground work, that water will be back (a few years) and will be faster than the pressurization in the disposal due to the slow production of hydrogen. As a result, gas pressure would be applied on a partially saturated mixture, see Figure II.7. For the bentonite/sand plugs sketched in this figure, there is a partially saturated layer in contact with an increasingly saturated layer. It is assumed that the material in contact with argillite is fully saturated.

Figure II.7 Schematic diagram of the in situ saturation process with gas

Therefore, an experimental set-up was designed in our laboratory to simulate the in situ situation, see Figure II. 8. Two small Plexiglas™-aluminium tubes were used (Figure II.9). The first tube used for Phase 1 of the test consists in water saturating a bentonite-sand plug with in situ water. Phase 2 begins after the triaxial cell dismounting and re-mounting, with the first fully saturated plug and a second tube+bentonite plug placed just over the first one. Inside this second, upper tube, the bentonite-sand plug is in its initial state (i.e. just after compaction); this second plug is supplied with water by the first one. This procedure is intended to be as realistic as possible. As presented in Figure II. 8, gas pressure is applied at the top of the assembly. We have chosen three possible cases: \( P_g = 0 \) (reference case), \( P_g = 4 \).
MPa or $P_g = 8/6$ MPa maximum value studied. The average *in situ* water pressure is 4MPa. The tube at the top (submitted to gas pressure) is instrumented with strain gauges to record the swelling pressures of the plug (calculated through a calibration test).

Figure II.8 Schematic diagram of the “PGZ” laboratory experimental devices

Figure II.9 One of the small tubes used for the swelling experiment with gas pressure
II.3.2 Definition of total equilibrium swelling pressure and effective swelling pressure

Usually, it takes more than one month for swelling pressure of the upper bentonite-sand plug to stabilize. At stabilization, the pressure, which contains gas pressure, water pressure and contact pressure between bentonite solid matrix and the inner surface of the tube, is called total equilibrium swelling pressure ($P_{\text{total}}$). After stabilization, water and gas pressure are set to zero, the swelling pressure will reach a new equilibrium. This new equilibrium pressure is effective swelling pressure ($P_{\text{eff}}$), which is only due to the bentonite solid matrix acting upon the tube inner surface, in the absence of any pore water pressure and gas pressure.

II.4 Gas breakthrough test

II.4.1 Why performing the GBT?

The main objective of this study is to observe the influence of the gas and its relative pressure (relative to that of water) on the saturation of the material. Several aspects can be taken into account in assessing this influence:

1. The swelling kinetics observed by the evolution of the total pressure.
2. The value of the effective swelling pressure measured after stabilization of the total pressure by stopping water and gas injection: this is the actual swelling pressure.
3. The water saturation of the plug obtained at the end of swelling test.

In fact, the degree of saturation is very difficult to measure because it would necessitate to remove the plug from the tube without loss of material, to weigh and dry it; and on the other hand it is necessary to carry out additional measures for which dismantling the triaxial cell is not necessary. It is more convenient to indirectly assess it by the breakthrough pressure measure. On the first hand it is a valuable tool to evaluate whether the material is fully saturated and, on the second hand, the test designed allows effective gas permeability to be evaluated after breakthrough. The same test can also be analysed to identify the gas pathway: throughout the bulk material or at the plug-tube interface. A partial saturation of bentonite-sand plug would therefore be linked to a lower breakthrough pressure (than for the saturated case).

II.4.2 Introduction of experimental method to measure the gas passage

The methods used to measure gas passage through the sample are summarized in Table II.3 (Thomas et al., 1968; Egermann et al., 2006, Hildenbrand et al., 2002; Horseman et al., 1999).
In our experiment, the step-by-step method was chosen although longer time was needed. The experiment was conducted using a step-by-step method, even though it required more time. Generally, there exist four methods to measure gas passage through a liquid-saturated material.

Indeed, it is allowed to reproduce more closely the in situ case by increasing gas pressure gradually, until the occurrence of breakthrough. Also, this method allows observing different successive fluid flow phases: expulsion of water, intermittent flow and continuous flow, while the observation of these phases is impossible with other methods. As introduced in Chapter II.3.2, swelling test will be stopped when swelling pressure becomes stable. Then, water and gas pressure are set to zero. The aim for this step is to measure the effective swelling pressure. After this pressure is obtained, gas breakthrough test is performed by replacing the lower bentonite-sand plug with an empty tube, see Figure II.10. Gas pressure is injected from the upstream sample side, through the empty tube. Gas detection is conducted in a downstream chamber by both a manometer (accuracy ±1mbar) and a dedicated gas detector (±1µl/sec). The gas pressure increases regularly (by 1MPa steps every one to three days) until continuous gas flow is detected in the other side.

<table>
<thead>
<tr>
<th>Method</th>
<th>Duration</th>
<th>Target value</th>
<th>Accuracy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Step-by-Step Method</td>
<td>long</td>
<td>discontinuous/continuous breakdown pressure</td>
<td>good</td>
</tr>
<tr>
<td>Racking Method</td>
<td>quick</td>
<td>gas entry pressure</td>
<td>good</td>
</tr>
<tr>
<td>Dynamic Method</td>
<td>quick</td>
<td>gas entry pressure</td>
<td>medium/good</td>
</tr>
<tr>
<td>Residual Method</td>
<td>long</td>
<td>snap off pressure (discontinuous breakthrough pressure)</td>
<td>bad</td>
</tr>
</tbody>
</table>
Figure II.10 Schematic diagram of gas breakthrough test: valve IV is opened when gas detection is performed, valve II and valve III are opened during each gas injection step on the upstream side; valve IV is opened to increase gas pressure in the buffer reservoir (while valve III is closed).

II.4.3 Definition of discontinuous/continuous gas breakthrough

As shown in Figure II.10, the specific test operation consists in opening the downstream chamber with valve I, and simultaneously placing the gas detector against the valve opening, in order to detect whether gas is present. If gas is detected during the first few seconds, and then this phenomenon disappears, we call this phenomenon as discontinuous breakthrough, and the relevant gas pressure is discontinuous gas breakthrough pressure ($P_{\text{dis}}$). If we can still detect gas after minutes or hours, we consider this phenomenon as continuous breakthrough, and the relevant gas pressure is continuous gas breakthrough pressure ($P_{\text{con}}$). Meanwhile, the gas permeability ($K_g$) and the rate of increase of downstream gas pressure ($Q_g$) are also utilized to confirm this phenomenon. Besides, this phase also exhibits a significant decrease of upstream gas pressure.
III - Water retention tests under constant volume and free swelling conditions
Introduction

This chapter presents the results of water retention tests of compacted bentonite/sand plugs under constant volume and free swelling conditions. Experimental methodology is detailed in Chapter II.

After initial compaction, each sample undergoes a progressive imbibition starting with RH = 70, then 75, 85, 92, 98, and finally 100% (pure water) for samples SO1 and SO2, or RH = 75, 85, 92, 98 and finally 100% (pure water) for SF1 and SF2 see Table III.1: two samples with the same size (SO1 and SO2) were tested under constant volume conditions, and three sets (samples SF1, SF2 and SF3) under free swelling conditions. The height of sample SF1 is only half of samples SF2 and SF3. Each sample of the series SF3 is subjected to a given relative humidity RH after an initial gas permeability test under variable confinement (up to $P_{c \text{ max}} = 5$ MPa), and all samples were subjected to RH = 100% to determine their complete saturated mass.

Table III.1 Nomenclature of samples and testing boundary conditions

<table>
<thead>
<tr>
<th>Samples</th>
<th>Number</th>
<th>Boundary conditions</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>SO1</td>
<td>1 (H=25mm, D= 42,5mm)</td>
<td>Constant volume Conditions</td>
<td>1) water retention tests are conducted directly after compaction 2) RH = 70% and 75% (SO1 and SO2) or 75% (SF1 and SF2), then 85%, 92%, 98% and finally 100%.</td>
</tr>
<tr>
<td>SO2</td>
<td>1 (H=25mm, D= 42,5mm)</td>
<td>Free swelling conditions</td>
<td></td>
</tr>
<tr>
<td>SF1</td>
<td>1 (H=12,5mm, D= 42,5mm)</td>
<td>Free swelling conditions (water retention tests)</td>
<td>1) water retention tests are conducted after gas permeability test ($K_g$); 2) RH = 75% or 85%, 92%, 98% and all samples RH = 100%.</td>
</tr>
<tr>
<td>SF2</td>
<td>1 (H=25mm, D= 42,5mm)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>SF3</td>
<td>4 (H=12,5mm, D= 37,6mm)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Remark: mass changes of different samples are not identical at RH = 70%: the mass of SO2 at stabilization of RH = 70% is almost the same than that after compaction (it increases by
less than 0.1 g), while sample SO1 has gained more than 0.5% mass (0.7 g), see Figure III.1. It is certainly because some slight differences may have occurred during sample preparation, e.g. dry density, which may lead to differences in the pore microstructures. The dry density could not be measured for each sample as drying leads to irreversible shrinkage and a macro-cracking.

### III.1 Water retention tests under constant volume conditions

Figures III.1 and III.2 show the swelling kinetics of samples SO1 and SO2 under different RH. It is noted that the test duration is quite long, more than 200 days of sample SO1. As shown in Figure III.2, the relative mass variation is calculated with a reference of the initial mass (just after compaction).

![Figure III.1](image-url)

**Figure III.1** Variation of the absolute mass from the mass just after compaction: samples SO1 and SO2.
Figure III.2: Variation of the relative mass from the mass just after compaction: samples SO1 and SO2.

One can see in Figure III.2 that sample mass SO2 is not yet stabilized at RH = 98%. It is nevertheless possible to compare the results for both samples at RH = 92%. We find for each sample a large capacity of water absorption with a speed of stabilization which is very low at higher RH.
As the mass variations of the samples are different at RH = 70%, we chose to plot the evolution of relative mass, taking the mass stabilized at RH = 70% as a reference, see Figure III.3. According to the law of Laplace Kelvin at 20 °C, the higher the RH increases, the more large-diameter pores are filled. Thus, as shown in Fig. III.3, at given RH, it is observed that there are more large-radius pores for sample SO2 than that for sample SO1: in particular, the mass increase at RH = 98% is about 3% for sample SO2 and only 2% for sample SO1. In fact, small changes in the waiting time before compaction (when bentonite powder matures at 85% RH, before compaction) or in the compaction process itself can lead to small changes in the distributions of pore radius. Accordingly, the mass increases at each humidity step are different. However, the total mass variation is quite close for both plugs: 5.86% for sample SO1 vs. 5.79% for sample SO2. This means that the final adsorption capacities (at least at RH=98%) of the two samples are quite close.

III.2 Water retention tests under free swelling conditions

III.2.1 Water retention tests under free swelling conditions and just after compaction

(a) Mass variations under different RH

Figure III.4 shows the evolution of absolute mass of samples SF1 and SF2. We can see that the mass of sample SF1 is not stabilized after 47 days of swelling at 98% RH. However, the
equilibrium time obtained for sample SF2 is about 84 days. The increase in the absolute mass of sample SF1 is lower than for sample SF2, and especially magnified at RH = 92%. This phenomenon can be attributed to the difference of initial mass of the two samples: 46.74 g for sample SF1 against 72.03 g for sample SF2.

Figure III.4 Comparison of the absolute mass: samples SF1 and SF2.

Figure III.5 (a) shows relative mass evolution of the two samples SF1 and SF2 (taking the mass after compaction as a reference). The first and most important observation is that there is no significant difference in terms of the increase of relative mass. For example, the mass increase at RH = 98% is about 8.8% for sample SF1 and 8.9% for sample SF2. Recalling that the height of sample is only half that of sample SF2, this means that scaling effect has little effect on the water absorption capacity of the bentonite/sand mixture.
Figure III.4 Taking the initial mass just after compaction as a reference: (a) comparison of the variation of relative mass of samples SF1 and SF2: RH 75% ~ 98%; (b) variation of relative mass of sample SF2: RH 75 % ~ 100%.

After stabilization at RH = 98%, sample SF2 is put at RH = 100%. It is amazing to see that mass changes of sample SF2 are not stabilized at RH = 100% even after 200 days of swelling, see Figure III.5 (b). Moreover, at RH=100%, it is found that the mass increase is about 17.69%
at the 331
day, which is about two times of the mass increase at RH = 98% (8.9%). This test is still ongoing.

(b) Volume variation under different RH

Figure III.6 and Figure III.7 show the volume evolution (absolute / relative) of sample SF2 during the experimental campaign (measurements are not performed during the preliminary study on sample SF1). Significant variations in volume are measured between the initial state and the mass stabilization at a given RH beyond 75%. 27.15% of increase in volume is measured at RH = 98%, while this value is only 0.69% at RH = 75%, 5.58% at RH = 85% and 10.71% at RH = 92%. It means bentonite/sand mixtures have a very good swelling capacity, especially at higher RH. This is favorable to the sealing efficiency of the disposal pit. More results on volume variations will be presented in the next chapter.

![Figure III.6 Absolute volume variation of sample SF2.](image-url)
Figure III.7 Relative volume variation of sample SF2.

### III.2.2 Water retention tests under free swelling conditions (after gas permeability tests)

This series was used to perform gas permeability tests first (at a confinement $P_{c\ max} = 5$ MPa), and then the samples were put at hermetic chambers to obtain different degree of saturation. This test differs from the previous tests which one sample was hydrated at progressively increasing humidity, while four different samples were put at four hermetic chambers (with different humidity) at the same time.

(a) Changes in mass at different RH

**Figure III.8** shows the relative mass variation of the four samples. It was found that lower RH (e.g. RH = 75% or 85%) has little effect on the mass variation: they contribute only by 0.92% of the mass increase at RH = 73%, and 1.18% at RH = 85%. Moreover, these values are similar to that measured for sample SF2 - not used for gas permeability test. The relative change in mass of sample SF2 (relative to the initial compacted state) is 0.51% at RH = 75% and 1.62% at RH = 85%.

At RH = 92%, mass stabilization of samples of series SF3 takes a little longer time, with 20 ~ 30 days of waiting, similar to sample SF2, see **Figure III.5 (a)**. By contrast, a significantly larger amount of water is absorbed by the sample SF3 at RH = 92% when comparing with corresponding value at RH = 75% (0.92%) or 85% (1.18%): it is 3.03%. This value is slightly smaller than the corresponding value of the sample SF2 (3.73%) at RH = 92%. The last sample of the series SF3 is put at RH = 98% and we found some similarities with the previous tests, both under the constant volume and free swelling conditions. It takes more time for the stabilization of the mass (40 ~ 50 days) and more water is absorbed by the sample (7.9%). It is found that the change in mass at RH = 98% for sample SF3 (7.9%) is smaller than the corresponding values of samples SF1 (8.79%) and SF2 (8.9%). This means the cycle of
loading and unloading during gas permeability test has led to the collapse of some pores and thus caused the decrease in porosity.

Figure III.5 Variation of relative mass of the samples SF3 placed at given RH after gas permeability test up to a maximum confinement $P_c = 5$ MPa.

(b) Changes in volume at different RH

Figure III.9 provides the measurements of volume change of sample SF3 during the experimental campaign. A similar phenomenon, already detected for sample SF2 tested at progressively increasing RH, is found again: the increase in sample’s volume is strongly correlated to the surrounding humidity. For series SF3, the increase of volume at RH = 98% is about 3 times than that of the volume measured at RH = 92%, 7 times than that of the volume measured at RH = 85% and 11 times than that of the volume obtained at RH = 75%.

When comparing with sample SF2, we find that the volume increase of sample SF3 is always smaller, although there is an exception at RH = 75%. The difference between the volume increases of the two series is more and more pronounced with the increase of RH, e.g. 1.18% at RH = 85%, 3.15% at RH = 92% and 4.69% at RH = 98%. As explained above, this difference can be attributed to the loading and unloading during gas permeability test which causes the collapse of some pores of the sample. Another test, which was performed to measure the change in porosity of sample under different confining pressures, will be presented in the next chapter. It allows us to better understand this phenomenon.
Figure III.6 Variation of the relative volume of sample SF3: $\Delta V_{\text{relative}} = (\Delta V_{RH} - \Delta V_{\text{initial}}) / \Delta V_{\text{initial}}$.

Figure III.7 Comparison of the change in the relative volume for samples SF2 and series SF3.

Remarque: in fact, the difference between the two sets of tests lies not only in the additional compaction suffered by the second series. Indeed, it is not clear whether the swelling process following RH = 75% and then 85, 92 and 98% is equivalent to that of mass intake and volume change in the situation where the material is directly put at RH=98%.
III.3 Comparison of water retention tests under free swelling and constant volume conditions (just after compaction)

Different water intakes versus time are shown in Figure III.11, under both constant volume conditions (SO1 and SO2) and free swelling conditions (SF1 and SF2). It can be observed that the boundary conditions have little effect on the saturation kinetics when RH ≤ 85%. This may be explained taking into account that there exist an initial clearance between the plug and the tube, where the initial “constant volume” is in fact “free swelling”. With the increase of RH (RH > 85%), the differences of swelling kinetics are becoming more pronounced, see Figure III.11 and Tableau. III.2. It is found that at higher RH, the swelling kinetics under free swelling conditions is faster than that under constant volume conditions, and that the amount of absorbed water is significantly higher under free swelling conditions. This is due to the large increase of pore volume experienced by the sample during the hydration under free swelling conditions.

![Figure III.8 Comparison of water retention tests under free swelling conditions and under constant volume conditions: changes in the relative mass of samples placed under constant volume conditions (SO1, SO2) and free swelling conditions (SF1, SF2).](image)

The case of RH = 100% is relatively specific. One can clearly recognize that mass intake is really higher when comparing with the corresponding values obtained under other RH. In addition, it can also find that the mass stabilization will be very difficult to achieve. Indeed, as the sample swells, it absorbs water, causing it to swell a little, although this process has a priori reason to stop under the free swelling conditions. In contrast, while the sample’s
volume is confined, the constant volume conditions allow earning less water than that under free swelling conditions. We can see a representation in Figure III.12, from (Komine, 2004).

Table III. 2 Increase of relative mass of sample under different RH and different boundary conditions.

<table>
<thead>
<tr>
<th>RH</th>
<th>No</th>
<th>SO1</th>
<th>SO2</th>
<th>SF1</th>
<th>SF2</th>
<th>minimum</th>
<th>maximum</th>
</tr>
</thead>
<tbody>
<tr>
<td>75%</td>
<td></td>
<td>0,63</td>
<td>0,52</td>
<td>0,00</td>
<td>0,51</td>
<td>0,01</td>
<td>0,63</td>
</tr>
<tr>
<td>85%</td>
<td></td>
<td>2,44</td>
<td>1,92</td>
<td>1,33</td>
<td>1,62</td>
<td>0,30</td>
<td>1,11</td>
</tr>
<tr>
<td>92%</td>
<td></td>
<td>3,74</td>
<td>3,22</td>
<td>2,74</td>
<td>3,73</td>
<td>0,01</td>
<td>1,00</td>
</tr>
<tr>
<td>98%</td>
<td></td>
<td>5,88</td>
<td>5,80</td>
<td>8,79</td>
<td>8,90</td>
<td>2,91</td>
<td>3,10</td>
</tr>
</tbody>
</table>

Figure III.9 Representation of the swelling process of bentonite different boundary conditions: (a) isochoric conditions and (b) vertical displacement permitted (Komine, 2004).
III.4 Conclusion

In this chapter, water retention tests are performed under both constant volume conditions and free swelling conditions (after compaction or after gas permeability test until \( P_{c,\text{max}} = 5 \) MPa). The results show that, at an RH of more than 85% (when individual aggregates of bentonite are completely saturated), the boundary conditions have an effect on the swelling kinetics of the sample. More precisely, at RH> 85%, the swelling kinetics of the sample under free swelling conditions is faster than the sample which swells under constant volume conditions. Moreover, also at RH> 85%, more water is absorbed under free swelling conditions: for example, the mass increase is 8.8% for sample SF2 (free swelling conditions) and only 5.88% for sample SO1 (constant volume conditions). At higher RH (98% and above), and under free swelling conditions, the increase in volume (with respect to the state of just after compaction as reference) is very significant: for example, at RH = 98% it is 22.46% for sample SF3, and 27.15% for sample SF2, which indicates an very good swelling capacity of the bentonite-sand mixture. Lastly, despite a very small change in water content and the volume (in the order of 1%), the cycle of loading - unloading at hydrostatic pressure due to the gas permeability test has an influence on the microstructure of pores of the sample, since water imbibitions at a given RH is smaller than corresponding values of samples SF2. This is interpreted as the collapses of some meso- and macro-pores, which affects the properties of swelling and water retention of the plugs.

For the determination of the retention curve (RH, \( S_w \)), we have found that the samples are are not fully stabilized at RH = 100%, and the saturated mass is not known. Dry mass to be measured after complete saturation, so far we cannot calculate \( S_w \). A few available results were found for SF3, for RH> 70%, see Figure III.13.

![Figure III.13 Relationship between HR and \( S_w \): sample SF3](image-url)
IV - Sealing ability of partially water-saturated bentonite/sand plugs under the effect of confinement
Introduction

This chapter aims to determine whether the central part of the bentonite-sand barrier, when it is only partially saturated with water and confined by the swelling pressure of the saturated plugs (up to about 7.5 MPa, see Chapter V), is permeable to gas or not. We also try to determine, for the partially saturated and strongly confined material, if it suffers a hydraulic cut-off: this corresponds to a measured gas permeability of $10^{-20}$m$^2$ or less, see (Liu et al 2013) and signifies that gas doesn’t pass significantly.

Our whole experimental campaign consists in three successive test series, so that the first series provides preliminary data, while the subsequent series aim at confirming (or not) our first interpretations. The initial state for all our experiments is taken after compaction of the bentonite/sand plugs to given dry density and water content, see Figure IV.1.

Figure IV.1 Experimental procedure followed for the three test series S1, S2 and S3

IV.1 Relative gas permeability – preliminary test results

The main idea for this first experimental campaign was to start from the initial material state, i.e. the state obtained just after compaction. One sample is tested for gas permeability only, right after compaction. Four additional plugs are put each into a different desiccator, at given relative humidity RH: 70, 75, 85 and 92 %. After mass stabilisation, each plug is placed into a triaxial cell, and submitted to a confinement and to gas flow, to assess its effective gas permeability. Effective gas permeability is measured using a quasi-stationary flow method, which principle is detailed in Chen et al. (2009). The average gas pressure of the quasi-stationary flow is of 0.4 to 0.5MPa, while confinement varies between 1.2 and 7.8MPa.
Table IV.1 indicates the results obtained in terms of mass variation and volume change. Dimensional observations are, at this stage, qualitative only, as this test series is the starting point towards more complete investigations, see next two sections. The relative humidity level corresponding to sample equilibrium (i.e. neither mass loss nor gain) is between 75 and 85%RH. Relative mass variations (labeled %mass rel. in the following) are negative for samples S1-1 and S1-2, and positive for samples S1-3 and S1-4, when compared to the initial compacted state, so that on the whole: %mass rel.(S1-4) > %mass rel.(S1-3) > %mass rel.(S1-0) > %mass rel.(S1-2) > %mass rel.(S1-1).

Table IV.1 First test series – mass variation and observed changes in mass intake and dimensions.

<table>
<thead>
<tr>
<th>Sample n.</th>
<th>Initial mass (g)</th>
<th>RH (%)</th>
<th>Stabilized mass (g)</th>
<th>Relative mass variation %mass rel. (% initial mass)</th>
<th>Dimensional observations</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1-0</td>
<td>54.70</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>None - gas permeability only</td>
</tr>
<tr>
<td>S1-1</td>
<td>54.66</td>
<td>70</td>
<td>54.24</td>
<td>-0.77</td>
<td>Shrinkage and important water loss</td>
</tr>
<tr>
<td>S1-2</td>
<td>54.65</td>
<td>75</td>
<td>54.53</td>
<td>-0.22</td>
<td>Shrinkage and slight water loss</td>
</tr>
<tr>
<td>S1-3</td>
<td>54.52</td>
<td>85</td>
<td>54.86</td>
<td>0.62</td>
<td>Swelling and slight water intake</td>
</tr>
<tr>
<td>S1-4</td>
<td>54.67</td>
<td>92</td>
<td>56.17</td>
<td>2.74</td>
<td>Important swelling and water intake</td>
</tr>
</tbody>
</table>

It is recalled here that gravimetric water content \( w(\%) \) is defined, and expressed in mass percentage, as:

\[
w(\%) = \frac{m_{\text{sample}} - m_{\text{dry}}}{m_{\text{dry}}} \times 100 \quad (1)
\]

where \( m_{\text{sample}} \) is sample mass and \( m_{\text{dry}} \) is sample dry mass, so that \((m_{\text{sample}} - m_{\text{dry}})\) is the mass of water contained in the sample. Water saturation level \( S_w \) is defined as the ratio between the volume of pores filled with water \( V_{\text{pores \ (filled \ with \ water)}} \) to the total pore volume \( V_{\text{pores \ (total)}} \), so that:

\[
S_w = \frac{V_{\text{pores \ (filled \ with \ water)}}}{V_{\text{pores \ (total)}}} = \frac{m_{\text{sample}} - m_{\text{dry}}}{m_{\text{saturated}} - m_{\text{dry}}} = \frac{m_{\text{sample}} - m_{\text{dry}}}{\rho_{\text{water}} V_{\text{sample}} \phi} \quad (2)
\]

where \( m_{\text{saturated}} \) is sample water-saturated mass, \( \rho_{\text{water}} \) is water density, \( V_{\text{sample}} \) is sample volume, and \( \phi \) is sample porosity. One should note that while water content is only given...
through current and dry sample masses, $S_w$ also requires determining a fully-water saturated mass, which proves more complicated for swelling materials such as bentonite/sand plugs. In first instance, let assume that all samples have a constant total pore volume (as for cohesive materials), and an identical initial water content (and an identical initial water saturation level) obtained after compaction. Hence, under such assumptions, relative mass variations mean that water contents vary as: $w(S1-4) > w(S1-3) > w(S1-0) > w(S1-2) > w(S1-1)$, or saturation levels $S_w(S1-4) > S_w(S1-3) > S_w(S1-0) > S_w(S1-2) > S_w(S1-1)$. Let now analyze gas transport properties under these assumptions.

Gas permeability results after mass stabilization at given RH are provided in Figure IV.2, for confinements from 1.2 and up to 7.8MPa. When assuming that, at given confinement, water content is the main parameter driving gas transport, these results are not as expected: one does not observe any systematic effect due to the sole changes in water content (or water saturation level). For instance, if $S_w(S1-1) < S_w(S1-2) < S_w(S1-0)$, gas permeability $K_g$ of samples S1-1 and S1-2 should be higher than that of sample S1-0, because S1-1 and S1-2 are less saturated than S1-0. Similarly, gas permeability of sample S1-0 should be higher than that of more saturated sample S1-3, and then than that of S1-4. In fact, such expected result is obtained for S1-0, S1-1 and S1-4 as $K_g(S1-1) > K_g(S1-0) > K_g(S1-4)$, but $K_g(S1-3) > K_g(S1-0) > K_g(S1-2)$, which appears unusual given their respective water saturation levels: $S_w(S1-3) > S_w(S1-0) > S_w(S1-2)$.

At this stage, at given confinement, two possibilities are considered. First, if the assumption of a constant total pore volume holds, the scattering in sample initial properties may drive the variation in $K_g$ from one sample to another. Secondly, and more probably (owing to the caution taken to prepare the samples), the total pore volume of each sample may vary during the tests, and hence, its pore volume accessible to gas. While total pore volume variation is expected under free boundary conditions, an increase in pore volume accessible to gas (together with water intake) is assumed during swelling; similarly, a decrease in pore volume accessible to gas is assumed during shrinkage. This is in good accordance with qualitative ESEM observations of pure MX80 bentonite by Montes-H et al. (2005). Such interpretation is not obvious, since the pore volume of bentonite (both total and accessible to gas) decreases with swelling under more usual oedometric conditions, see Xie et al. (2004) and Alkan et al. (2008).

The consequence for gas transport is a competitive effect between increasing (or decreasing) water intake and swelling (or shrinkage). Indeed, if the latter were true, then: (1) at given total pore volume, greater pore volume filled with water would induce lower gas permeability,
while smaller pore volume filled with water would yield greater $K_g$, as for cohesive geomaterials (Chen et al., 2012); (2) by increasing the total pore volume (and that accessible to gas), swelling would contribute to greater gas permeability, while shrinkage (despite generally leading to micro-cracking) would induce lower gas permeability. The third effect to take into account is confinement, so that any increase in confining pressure is bound to lead to a decrease in effective gas permeability. This effect is observed, as expected, for each sample tested in this series, see Figure IV.3: $K_g$ is a monotonously decreasing function of $P_c$.

More particularly, for sample S1-4, which is subjected to 92% relative humidity, the above interpretation applies, as follows. A RH of 92% is sufficiently high to get a high water saturation level, so that this becomes the predominant effect, and provides a lower $K_g$, despite a strong swelling (which is associated to greater pore volume accessible to gas).

![Effective permeability results vs. confining pressure - at different saturation levels](image)

Figure IV.2 Effective permeability results vs. confining pressure - at different saturation levels

Sample S1-1 loses a significant mass of water, representing -0.77% of its initial value, which hints at a noticeable decrease in its water saturation level. Despite its shrinkage, which reduces the pore volume available for gas flow, gas permeability of S1-1 is higher than that of S1-0, which is in the initial state: this means that the effect of de-saturation is predominant over that of shrinkage.
On the opposite, for samples S1-2 and S1-3, the effect of swelling/shrinkage (and pore volume variation) is predominant over that of saturation level. Sample S1-2 sustains simultaneously a decrease in water saturation (which contributes to an increase in gas permeability $K_g$) and limited shrinkage (responsible for a decrease in pore volume accessible to gas, i.e. in $K_g$), so that, on the whole, its permeability $K_g$ is lower than that of S1-0. For sample S1-3, although one observes an increase in saturation (associated to a decrease in $K_g$), limited swelling (associated to an increase in $K_g$) induces greater $K_g$ than for S1-0.

In order to confirm these antagonist effects more quantitatively, complementary experiments are performed by two supplementary test series, see below.

**IV.2 Relative gas permeability study – second series of tests**

**IV. 2.1 Mass and volume changes**

After compaction, samples of this series are subjected to gas permeability testing up to 5MPa confinement, and then to a given RH ranging from 75 to 98%: for plugs S2-3, S2-4, S2-5 and S2-8 these are, respectively, 98, 92, 85 and 75% RH. Lower RH levels are not studied, as the preliminary study (see Section 3) showed that samples lose water from RH=70% and below. Plug S2-2 was only used in the initial state, after compaction, to test the plug sensitivity to confining pressure values up to 12MPa.
Figure IV.3 Evolution of sample mass vs. time, when the samples are placed at different humidity levels.

Mass evolution from compaction to the atmosphere at fixed RH is given in Figure IV.3. Masses at negative times correspond to the initial compaction state. Masses at day 1 correspond to the initial gas permeability measurement phase.

Figure IV.3 shows that, from one sample to another, there are huge differences in the time required to get a stable mass at given RH. These differences are attributed to the amount of water gained by each sample, which is much greater at higher relative humidity. The longest time for mass stabilisation is obtained with S2-3, which is placed at 98%RH. This is related to the strong swelling, which is observed at this level, see Figure IV.4.

![Graph showing mass and volume variations](image)

Figure IV.4 Mass and volume relative variations due to increasing RH level (before gas permeability testing in partially-saturated conditions), using for reference mass and volume those after the initial gas permeability test.

Table IV.2(a) (below) indicates the main sample characteristics for this second test series. Annex 1 provides all volume change measurements during the experimental campaign. Significant variations in sample volume and mass are measured between the initial compacted state and that after mass stabilization at given RH, and also after drying at 60°C. Indeed, except for S2-8, which loses mass at 75%RH, swelling observed during water absorption of the samples is significant. For example, for S2-3, its volume variation at mass stabilization at RH=98% represents 20.6% of its initial volume (after compaction), see Table IV.2(a). This
means that the actual pore volume is constantly changing during the swelling process due to bentonite/sand deformability (related to water absorption). This is also the case at 100%RH, where sample mass $m_{\text{saturated}}$ reflects the proportion of voids filled with water i.e. the proportion of actual porosity filled with water. Significant sample volume changes also mean that the choice of a reference volume for assessing physical properties (such as density, porosity and water saturation) will require discussion.

Several physical properties of the samples are deduced from the raw data in Table IV.2(a), see Table IV.2(b). Apparent plug density is calculated after compaction as: $\rho = \frac{m_{\text{compact}}}{V_{\text{compact}}}$.

An “apparent” dry density $\rho_{\text{dry}} = \frac{m_{\text{dry}}}{V}$, where $V$ is either $V_{\text{compact}}$ (sample volume after compaction), or $V_{\text{dry}}$ (sample volume in the dry state). It is observed that all samples have a good homogeneity in terms of apparent or dry density. Dry density values are closer when calculated with $V_{\text{compact}}$ rather than with $V_{\text{dry}}$, hinting at greater volume changes after our experiments (and the subsequent drying) than right after compaction.

A discussion about porosity (and further, about water saturation $S_w$) is proposed here, as this property requires a reference volume to be calculated, as:

$$\phi = \frac{(m_{\text{saturated}} - m_{\text{dry}})}{\rho_{\text{water}}} V$$

where $m_{\text{saturated}}$ is sample saturated mass (obtained at stabilization in an hermetic chamber at 100%RH), $m_{\text{dry}}$ is dry mass, $\rho_{\text{water}}$ is water density (1000kg/m$^3$ at 20°C), and $V$ is sample total volume. It is noted that the humidity of 100%RH (used to obtain $m_{\text{saturated}}$) is imposed by placing each sample in a desiccator, over pure distilled water.

In Table IV.2(b), the volume after compaction $V_{\text{compact}}$ (i.e. the initial volume) is chosen as the reference volume $V$ to calculate $\phi$. Therefore, $\phi$ represents an “artificial” porosity rather than an actual one, due to significant sample volume changes from the dry to the so-called fully water saturated state, see Table IV.2(b). Except for sample S2-2, which is significantly more porous and less saturated than the other samples of the series, porosity values are on the order of 47%+/-.7.

Water saturation $S_w$, which is given below, will also be a conventional or artificial saturation, assessed with respect to the same sample volume $V$, as:

$$S_w = \frac{(m_{\text{RH}} - m_{\text{dry}})}{\rho_{\text{water}} \phi V}$$
### Table IV.2(a) Main characteristics of the bentonite/sand plugs of test series 2.

<table>
<thead>
<tr>
<th>N.&amp; RH</th>
<th>$D_{\text{compact}}$ (cm)</th>
<th>$H_{\text{compact}}$ (cm)</th>
<th>$V_{\text{compact}}$ (cm³)</th>
<th>$V_{\text{RH}}$ (cm³)</th>
<th>$V_{\text{dry}}$ (cm³)</th>
<th>$(V_{\text{RH}} - V_{\text{compact}}) / V_{\text{compact}}$ (%)</th>
<th>$m_{\text{compact}}$ (g)</th>
<th>$m_{\text{RH}}$ (g)</th>
<th>$m_{\text{dry}}$ (g)</th>
<th>$m_{\text{saturated}}$ (g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>S2-2</td>
<td>3.70</td>
<td>1.25</td>
<td>13.39</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>27.17</td>
<td>-</td>
<td>23.84</td>
<td>32.59</td>
</tr>
<tr>
<td>S2-3 (98%)</td>
<td>3.70</td>
<td>1.25</td>
<td>13.41</td>
<td>16.18</td>
<td>13.24</td>
<td>20.65</td>
<td>27.33</td>
<td>29.34</td>
<td>24.24</td>
<td>30.12</td>
</tr>
<tr>
<td>S2-4 (92%)</td>
<td>3.70</td>
<td>1.26</td>
<td>13.48</td>
<td>14.28</td>
<td>12.85</td>
<td>5.97</td>
<td>27.29</td>
<td>28.13</td>
<td>23.96</td>
<td>30.10</td>
</tr>
<tr>
<td>S2-5 (85%)</td>
<td>3.70</td>
<td>1.25</td>
<td>13.41</td>
<td>13.66</td>
<td>12.80</td>
<td>1.86</td>
<td>27.25</td>
<td>27.57</td>
<td>23.88</td>
<td>30.72</td>
</tr>
<tr>
<td>S2-8 (75%)</td>
<td>3.70</td>
<td>1.26</td>
<td>13.50</td>
<td>13.62</td>
<td>13.05</td>
<td>0.91</td>
<td>27.46</td>
<td>27.44</td>
<td>23.99</td>
<td>30.56</td>
</tr>
</tbody>
</table>

Notes: $V_{\text{compact}}$ is the sample volume after compaction, $V_{\text{RH}}$ is the volume at mass stabilization at given RH, $V_{\text{dry}}$ is dry sample volume, (measured with a gauge calliper (by D and H)). $m_{\text{compact}}$ is sample mass after compaction, $m_{\text{dry}}$ is dry mass (after stabilization in the oven at 60°C), $m_{\text{RH}}$ is stable mass in the hermetic chamber at given RH, and $m_{\text{saturated}}$ is saturated mass (after stabilization in an hermetic chamber at 100%RH).
Table IV.2(b) Main physical properties of the bentonite/sand plugs of test series 2, deduced from raw data given in Table 2a.

<table>
<thead>
<tr>
<th>N.&amp; RH</th>
<th>$\rho$ (g/cm$^3$)</th>
<th>$\rho$ (g/cm$^3$) with $V_{\text{dry}}$</th>
<th>$\phi$ (%)</th>
<th>$S_w$ initial, after compaction (%)</th>
<th>$S_w$ using masses Eq. (5) (%)</th>
<th>$S_w$ using $V_{RH}$ (%)</th>
<th>$S_w$ using $V_{dry}$ (%)</th>
<th>$S_w$ using $V_{\text{compact}}$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>S2-2</td>
<td>2.03</td>
<td>1.78</td>
<td>-</td>
<td>65.35</td>
<td>38.06</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>S2-3 (98%)</td>
<td>2.04</td>
<td>1.81</td>
<td>1.83</td>
<td>43.84</td>
<td>52.55</td>
<td>86.73</td>
<td>71.89</td>
<td>87.86</td>
</tr>
<tr>
<td>S2-4 (92%)</td>
<td>2.03</td>
<td>1.78</td>
<td>1.86</td>
<td>45.56</td>
<td>54.23</td>
<td>67.92</td>
<td>64.09</td>
<td>71.22</td>
</tr>
<tr>
<td>S2-5 (85%)</td>
<td>2.03</td>
<td>1.78</td>
<td>1.87</td>
<td>51.00</td>
<td>49.27</td>
<td>53.95</td>
<td>52.96</td>
<td>56.52</td>
</tr>
<tr>
<td>S2-8 (75%)</td>
<td>2.03</td>
<td>1.78</td>
<td>1.84</td>
<td>48.68</td>
<td>52.82</td>
<td>52.51</td>
<td>52.04</td>
<td>54.31</td>
</tr>
</tbody>
</table>
V may be chosen as the volume after compaction (initial plug state) $V_{\text{compact}}$, or as $V_{\text{RH}}$ (after stabilization at given RH), or as the dry sample volume $V_{\text{dry}}$. When assessing $S_w$ during the gas transport experiment, the choice of $V_{\text{compact}}$, which is the volume at the start of the tests, as the reference volume is more appropriate than $V_{\text{RH}}$ (obtained during the experiment), or than $V_{\text{dry}}$ (obtained after permeability testing). Eq. (4) is also equivalent to:

$$S_w = \frac{V_{\text{poro}} \cdot (\text{filled} \cdot \text{with} \cdot \text{water})}{V_{\text{poro}} \cdot (\text{total})} = \frac{m_{\text{RH}} - m_{\text{dry}}}{m_{\text{saturated}} - m_{\text{dry}}} \quad (5)$$

Eq. (5) provides identical values for $S_w$ as with $V_{\text{compact}}$ as the reference volume. With this method, $S_w$ ranges from 52.5% (for S2-8, subjected to 75%RH) and up to 86.7% (for S2-3, subjected to 98%RH). It is noted that, while $S_w$ decreases for S2-8 (75%RH) between compaction and after mass stabilization at given RH, $S_w$ increases for all the other samples, subjected to RH from 85 and up to 98%. The case of S2-8 is noticeable, because this sample swells while losing mass (from compaction to mass stabilisation at 75%RH), so that on the whole, its saturation $S_w$ decreases, see Table IV.2(a) and (b). This is attributed to a small de-compaction effect, possibly related to the initial gas permeability measurement phase.

One also observes in Table IV.2(b) that dry density $\rho_{\text{dry}}$ is slightly higher (by 0.01 to 0.04g/cm$^3$) than the targeted one, which is of 1.77. Indeed, it is uneasy to obtain exactly the required humidity for the bentonite/sand mix before its compaction. Nevertheless, this is not an actual issue, as regards the phenomena and properties under study here.

IV. 2.2 Initial sample gas permeability

Results of gas permeability after compaction are provided in Figure IV.5. Except for S2-2, which is only tested in the initial state, confining pressure was limited to 5MPa for the other samples of the series (S2-3, S2-4, S2-5 and S2-8). This aims at limiting microstructure changes before mass stabilization at given RH and subsequent $K_g$ assessment.
Figure IV.5 Initial sample permeability vs. confining pressure.

Table IV.3 Values of gas permeability at the start, maximum confining pressure, and at the end of a confinement cycle, for samples of Series S2 after compaction (before being subjected to given RH or oven-drying).

<table>
<thead>
<tr>
<th>Sample n.</th>
<th>S2-3</th>
<th>S2-4</th>
<th>S2-5</th>
<th>S2-8</th>
<th>S2-2</th>
</tr>
</thead>
<tbody>
<tr>
<td>$K_g$ ($10^{-17}$ m$^2$) at $P_c$=1MPa (start of the confining cycle)</td>
<td>25.3</td>
<td>44.4</td>
<td>36.3</td>
<td>28.3</td>
<td>22.2</td>
</tr>
<tr>
<td>$K_g$ ($10^{-17}$ m$^2$) at $P_c$=5MPa (maximum of the confining cycle)</td>
<td>3.48</td>
<td>9.32</td>
<td>6.38</td>
<td>8.70</td>
<td>4.03</td>
</tr>
<tr>
<td>$K_g$ ($10^{-17}$ m$^2$) at $P_c$=1MPa (end of the confining cycle)</td>
<td>8.2</td>
<td>19.3</td>
<td>14.2</td>
<td>19.5</td>
<td>3.13</td>
</tr>
</tbody>
</table>

It is observed that the scatter on gas permeability values after compaction and at low confinement (1MPa) is relatively limited for this kind of material (which is very sensitive to its initial compaction and water content conditions), with values ranging from $22.2 \times 10^{-17}$ m$^2$ and up to $44.4 \times 10^{-17}$ m$^2$. These values are comparable with those from test series S1, as is the sensitivity to confining pressure – i.e. a decrease in permeability by a factor of 3 to 7 in the range 0~5 MPa confining pressure. It is also noted that, for plug S2-2, which sustains the greatest loading up to $P_c$=12MPa, confining pressure has a major influence on its gas
permeability, which drops down by two orders of magnitude between $P_c=1\text{MPa}$ and $P_c=12\text{MPa}$. This is attributed to a sort of pore collapse, i.e. a loss of pore volume accessible to gas under confinement $P_c$. All samples display such pore closure, and it is irreversible in nature, because no sample regains its initial gas permeability (that before the confining cycle up to 5 or 12MPa). This irreversibility, or hysteresis in $K_g(P_c)$ behaviour, is much more marked for sample S2-2 than for S2-3, S2-4, S2-5 and S2-8, due to a greater confinement amplitude, see Figure IV.5 and Table IV.3.

IV. 2.3 Coupled effects of saturation and confining pressure upon gas permeability

After mass stabilization at given RH, Figure IV.6 to 9 (below) provide the effective permeability of each sample, when confining pressure is varied. These values are compared with the initial gas permeability measurement (after compaction).

![Figure IV.6 Comparison between initial gas permeability (in blue) and gas permeability after stabilisation at 75%RH (in red), for sample S2-8.](image)

Results for sample S2-8 (subjected to 75%RH) are as expected, see Figure IV.6: Point 1 at $P_c=1\text{MPa}$ (at the end of initial permeability test) corresponds to a lower gas permeability than at Point 2 (at $P_c=1\text{MPa}$ but after stabilization at 75%RH). Indeed, as S2-8 has de-saturated slightly at RH=75% (from $S_w=52.8$ to $S_w=52.5$%), its permeability is higher than in the initial state. However, this variation is high: between Point 1 and Point 2, gas permeability is multiplied by a factor of 5. In fact, huge variations in gas permeability are recorded
throughout our experimental campaign on bentonite/sand plugs. For instance, we will see in Sub-section IV 3.4) that gas permeability is three orders of magnitude higher when comparing the initial state and the dry state. This means that there is a very strong effect, upon gas permeability, of drying from 52.8% down to “0”% saturation. Figure IV.6 also shows that the slope of the decrease in permeability is comparable in the initial state and after 75%RH, when confining pressure increases. This is because both saturation states of sample S2-8 are very close. Nevertheless, a strong hysteretic effect is observed upon unloading, so that permeability does not follow the same path during unloading as during loading. This hysteresis occurs mainly from P_c=12MPa down to 5MPa, because, during unloading from P_c=5 to 1MPa, K_g (P_c) still follows a similar slope as during loading. Due to this hysteresis, at the lowest confinement used (P_c=1MPa), gas permeability is irreversibly lowered by the confinement cycle: at P_c=1MPa, K_g decreases by a factor of 8.4 (from 109×10^{-17}m^2 down to 13×10^{-17}m^2). And while K_g (P_c) is almost linear during loading, it is highly non linear in the unloading range (12-5MPa). This is attributed to an irreversible compaction of the sample, with a volume decrease from 13.62cm^3 down to 13.29cm^3, i.e. by 2.4% only.

The case of sample S2-5 is less obvious to analyse: despite an increase in saturation (from S_w=49.3 to 53.9%), gas permeability at point 2 (start of loading after mass stabilization at 85%RH) is higher than that at point 1 (at the end of 1st permeability experiment, before 85%RH), see Figure IV.7. Between Points 1 and 2, due to the placement of S2-5 at 85%RH, an increase by almost 5% of the volume has occurred. The higher gas permeability is attributed to this increase in total volume, which is bound to have brought an increase in pore volume accessible to gas, too.

The confining pressure effect is more sensitive here than for S2-8, as almost three orders of magnitude difference exist between K_g (P_c=1MPa)=73×10^{-17}m^2 and K_g (P_c=12MPa)=0.12×10^{-17}m^2, i.e. between the start of the test and the maximum confinement achieved. This is not a tight state yet, whereby gas passage becomes negligible due to confinement increase, but it is quite spectacular, all the more so as the sample water saturation S_w at the start of this test is 53.9% only.
Figure IV.7 Comparison between initial gas permeability (in blue) and gas permeability after stabilisation at 85%RH (in red), for sample S2-5.

There is also a stronger hysteretic effect during the unloading phase, which occurs now mainly in the range 12-5MPa. As for S2-8, it is associated to sample volume change, from 13.66cm³ (at the start of the second gas permeability test) to 13.25cm³ (at the end of this test), i.e. by 3% only, see Annex 1. This is a clear evidence of the major effect of sample volume variation on its gas permeability. The difference in permeability between 2 and 3 can be clearly attributed to this volume variation.

For sample S2-4 subjected to 92%RH, Figure IV.7 shows that, like for S2-5, at Pc=1MPa, a higher permeability is measured at Point 2 (at mass stabilization at 92%RH) than at Point 1 (at the end of the initial gas permeability test). As saturation Sw is now 67.9%, this fact is attributed to an increase in sample volume, which is by more than 7.5%, see Annex 1: sample volume is of 13.28cm³ at the end of the initial gas permeability test, and of 14.28cm³ at mass stabilization at 92%RH. Between both competitive effects (water intake and sample swelling), the volumetric effect is stronger than the saturation effect at low confining pressure. This confirms our analysis for the first test series.
Sample swelling (and the increase in pore volume accessible to gas) is no longer predominant upon $K_g$ when confining pressure is increased. For instance, at $P_c=5$MPa, the decrease in gas permeability between Point A (before swelling) and Point B (after swelling) is of one order of magnitude: $K_g$(Point A) $= 9.3 \times 10^{-17}$m$^2$ is greater than $K_g$(Point B) $= 0.74 \times 10^{-17}$m$^2$ by a factor of 12.6. If one assumes an equivalent volumetric strain at Point A and Point B (between the initial state and the partially water-saturated state, both at 5MPa confining pressure), the permeability difference is rather attributed to the difference in water saturation. Indeed, water being incompressible, most of the sample volume change (and volumetric strain), due to $P_c$ increase, leads to a direct decrease in pore volume available to gas. More generally, the confining pressure effect is more and more pronounced as the bentonite/sand plug becomes more and more saturated. For sample S2-4, saturated by 67.9% only, this effect is huge: gas permeability decreases by 48000, i.e. by almost five orders of magnitude (from $K_g$($P_c=1$MPa) $= 48 \times 10^{-17}$m$^2$ down to $K_g$($P_c=12$MPa) $= 1 \times 10^{-20}$m$^2$ (loading phase only)).

Finally, the hysteretic behaviour of $K_g$($P_c$) is still marked, and it is greater than for S2-5 and S2-8: hysteresis is stronger and stronger with sample water saturation level. As a consequence of hysteresis, sample volumetric strain (and volume change) is not reversible: after dismounting from the triaxial cell, the sample volume is 13.3cm$^3$, which is 6.8% lower than the 14.28cm$^3$ volume before the second permeability test. This justifies that, at $P_c=1$MPa, gas
permeability at point 3 is smaller by more than two orders of magnitude than permeability at Points 1 and 2.

Figure IV.9 Comparison between initial gas permeability (in blue) and gas permeability after stabilization at 98%RH (in red), for sample S2-3.

For sample S2-3, the effect of confining pressure upon $K_g$ is spectacular, with a decrease by five orders of magnitude when $P_c$ increases from 1 to 12MPa, see Figure IV.9. Due to experimental difficulties, it was not possible to record gas permeability at $P_c=1$MPa during the unloading path.

Sample saturation is 86.7% after mass stabilization at 98%RH, but the increase in volume due to this saturation is of 22.4%: the volume of S2-3 increases from 13.21cm$^3$ to 16.18cm$^3$ at stabilization at 98%RH. The equivalent (but opposite) effects of water saturation increase and swelling (i.e. pore volume increase) are thought to explain why gas permeabilities at Points 1 and 2 are very close. The interpretation of antagonistic effects of water intake and free swelling upon gas transport, as analysed from our preliminary results, are confirmed by this second test series.

In more detail, it is observed that the influence of confining pressure is very high up to 5 MPa (point A). At this stage, gas permeability is of $7.8\times10^{-20}m^2$, whereas it was more than three orders of magnitude at $P_c=1$MPa, with a value of $1.07\times10^{-16}m^2$: this means that sample S2-3 has become virtually impermeable to gas. Further, there is a smaller decrease, yet by two
orders of magnitude, between 5 and 12MPa confinement (when compared to that before \( P_c=5\) MPa). The fact that S2-3 sample is impermeable to gas at \( P_c=5\) MPa certainly means that it is close to 100\% actual water saturation, which is brought by sample compaction under loading. Then, for \( P_c\) above 5MPa, there is a smaller pore volume accessible to gas, which remains to collapse, therefore, leading to less permeability decrease. However, it is considered that from \( P_c=5\) MPa (and more), the bentonite/sand plug is impermeable to gas. It is recalled here that \( P_c=5\) MPa is smaller than the expected in situ swelling pressure of the outer saturated bentonite/sand plugs, so that this result has some significance for sealing purposes: this is an experimental evidence of the possibility for a bentonite-sand structure to become tight to gas, before a full water saturation state is achieved in all of its individual plugs.

In order to highlight the huge effect of confining pressure upon \( K_g\), which is all the more so marked as sample saturation is high, Figure IV.10 (below) plots gas permeability for all the partially water-saturated samples. At \( P_c=12\) MPa, the higher the saturation level \( S_w\), the lower the gas permeability \( K_g\). While sample S2-8 (\( S_w=52.5\%\)) loses two orders of magnitude in \( K_g\) upon loading from \( P_c=1\) MPa to 12MPa, sample S2-3 (\( S_w=86.7\%\)) loses five orders of magnitude in \( K_g\) during the same loading phase. This increased influence of water saturation level is attributed to a decrease in pore volume accessible to gas, which is all the more so critical to gas transport as \( S_w\) is great.

![Figure IV.10](image.png)

Figure IV.10 Comparison of gas permeability after stabilization at given RH, vs. confining pressure, for samples S2-3 (98\%RH), S2-4 (92\%RH), S2-5 (85\%RH) and S2-8 (75\%RH).
IV. 2.4 Dry permeability

Dry permeability values obtained after the second permeability test (in partially water-saturated conditions) are given in Figure IV.11 below.

It is first observed that the dry permeability of the four samples S2-3, S2-4, S2-5 and S2-8 is two (or three) orders of magnitude higher than their initial values after compaction. This is the confirmation that drying between 50% and 0% saturation has a strong influence on gas permeability, as mentioned before. Another aspect is that there seems to remain a sort of memory of previous swelling and shrinkage for the different samples: here, those which were saturated at the higher relative humidities have the higher permeabilities in the dry state. We do not know at this stage if it is an coincidence, or an actual, repetitive, phenomenon: the third test series will contribute to clarify this aspect.

Moreover, the samples, which have sustained the greatest swelling, are also the most sensitive to an increase in confinement i.e. they are more deformable. If confirmed by test series S3, this means that the structure and gas transport behaviour of bentonite/sand plugs is very sensitive to successive drying/imbibition cycles, coupled with confinement/un-confinement cycles.

Figure IV.11 Dry gas permeability vs. confining pressure for all samples of test series S2.
IV.3 Complementary tests (third test series)

A last test series was performed to confirm the main trends, which were observed during the first two series. This series was not subjected to any initial gas permeability measurement (and confinement) prior to free swelling at given RH. Indeed, this was decided in order to avoid any over-compaction of the plugs due to confinement (which is unavoidable with our gas permeability test method), before swelling. One plug only (number S3-9) was tested after the initial compaction, to acquire one reference value for initial gas permeability. Another single sample (S3-14) has been prepared by compaction and oven-drying at 60°C until mass stabilization, in order to assess directly its pore volume accessible to gas under varying confining pressure.

IV. 3.1 Mass and volume changes

Tables 5a and 5b below show the general physical properties of the plugs from this series S3. For this test series, average porosity is of 42.3%±/3.8, which is slightly lower than for test series S2 (φ=47%±/3.8). Dry and apparent densities are comparable for both test series, although they are slightly higher for test series S3 (by 0.05g/cm³ i.e. by 2.4%). Initial saturation levels (after compaction) are also greater for series S3, by more than 10%. After mass stabilization at given RH, as for test series S2, S_w increases with increasing RH, except for S3-10 (75%RH) and S3-11 (85%RH). In this case, initial S_w is greater for S3-10 than for S3-11, so that S3-10 has not stabilized at a lower S_w than S3-11 at 75%RH. Sample S3-10 loses mass during its placement at RH=75%, similarly to S2-8, yet it also shrinks (whereas S2-8 swell). Therefore, results for sample S3-10 will be analysed with caution when comparing with previous test series, and with less saturated S3-11.

A significant shrinkage is noted after mass stabilisation at 11%RH, with -6.7% volume change, while a huge swelling (by 19.8% volume change) occurs at RH=98%.
Table IV.4(a) Main characteristics of the bentonite/sand plugs of test series 3.

<table>
<thead>
<tr>
<th>Plug &amp; RH</th>
<th>D_{compact} (cm)</th>
<th>H_{compact} (cm)</th>
<th>V_{compact} (cm³)</th>
<th>V_{RH} (cm³)</th>
<th>V_{dry} (cm³)</th>
<th>(V_{RH} – V_{compact}) / V_{compact} (%)</th>
<th>m_{compact} (g)</th>
<th>m_{RH} (g)</th>
<th>m_{dry} (g)</th>
<th>m_{saturated} (g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>S3-9 (11%RH)</td>
<td>3.72</td>
<td>2.45</td>
<td>26.62</td>
<td>24.84</td>
<td>24.59</td>
<td>-6.68</td>
<td>54.86</td>
<td>47.84</td>
<td>48.65</td>
<td>58.16</td>
</tr>
<tr>
<td>S3-10 (75%)</td>
<td>3.69</td>
<td>2.46</td>
<td>26.31</td>
<td>26.23</td>
<td>25.13</td>
<td>-0.31</td>
<td>54.95</td>
<td>47.92</td>
<td>54.40</td>
<td>58.25</td>
</tr>
<tr>
<td>S3-11 (85%)</td>
<td>3.69</td>
<td>2.48</td>
<td>26.50</td>
<td>27.04</td>
<td>25.51</td>
<td>2.02</td>
<td>55.00</td>
<td>48.02</td>
<td>55.10</td>
<td>59.77</td>
</tr>
<tr>
<td>S3-12 (92%)</td>
<td>3.69</td>
<td>2.47</td>
<td>26.43</td>
<td>27.95</td>
<td>25.65</td>
<td>5.76</td>
<td>55.00</td>
<td>47.83</td>
<td>56.10</td>
<td>59.18</td>
</tr>
<tr>
<td>S3-13 (98%)</td>
<td>3.70</td>
<td>2.50</td>
<td>26.81</td>
<td>32.11</td>
<td>25.80</td>
<td>19.78</td>
<td>54.95</td>
<td>47.40</td>
<td>58.75</td>
<td>59.77</td>
</tr>
</tbody>
</table>

Notes: $V_{compact}$ is the sample volume after compaction, $V_{RH}$ is the volume at mass stabilization at given RH, $V_{dry}$ is dry sample volume, (measured with a gauge calliper (by D and H)). $m_{compact}$ is sample mass after compaction, $m_{dry}$ is dry mass (after stabilization in the oven at 60°C), $m_{RH}$ is stable mass in the hermetic chamber at given RH, and $m_{saturated}$ is saturated mass (after stabilization in an hermetic chamber at 100%RH).
Table IV.4(b) Main physical properties of the bentonite/sand plugs of test series 3, deduced from raw data given in Table IV.4(a).

<table>
<thead>
<tr>
<th>n. &amp; RH</th>
<th>$\rho$ (g/cm$^3$) with $V_{\text{compact}}$</th>
<th>$\rho$ (g/cm$^3$) with $V_{\text{dry}}$</th>
<th>$\phi$ (%)</th>
<th>$S_w$ initial, after compaction (%)</th>
<th>$S_w$ using masses Eq. (5) (%)</th>
<th>$S_w$ using $V_{\text{RH}}$ (%)</th>
<th>$S_w$ using $V_{\text{dry}}$ (%)</th>
<th>$S_w$ using $V_{\text{compact}}$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>S3-9 (11%RH)</td>
<td>2.06</td>
<td>1.80</td>
<td>1.95</td>
<td>38.77</td>
<td>68.02</td>
<td>7.85</td>
<td>8.41</td>
<td>8.50</td>
</tr>
<tr>
<td>S3-10 (75%)</td>
<td>2.09</td>
<td>1.82</td>
<td>1.91</td>
<td>39.26</td>
<td>68.05</td>
<td>62.73</td>
<td>62.92</td>
<td>65.68</td>
</tr>
<tr>
<td>S3-11 (85%)</td>
<td>2.08</td>
<td>1.81</td>
<td>1.88</td>
<td>44.33</td>
<td>59.40</td>
<td>60.26</td>
<td>59.06</td>
<td>62.60</td>
</tr>
<tr>
<td>S3-12 (92%)</td>
<td>2.08</td>
<td>1.81</td>
<td>1.86</td>
<td>42.95</td>
<td>63.17</td>
<td>72.86</td>
<td>68.90</td>
<td>75.08</td>
</tr>
<tr>
<td>S3-13 (98%)</td>
<td>2.05</td>
<td>1.77</td>
<td>1.84</td>
<td>46.14</td>
<td>61.03</td>
<td>91.75</td>
<td>76.60</td>
<td>95.34</td>
</tr>
</tbody>
</table>
Figure IV.12 shows the saturation (or drying) kinetics and the mass variations for all samples of this series. The same trends, already detected for previous sample series, are confirmed: drying occurs for RH=75% and below, a very low mass increase occurs for RH=85% and mass increases at RH=92 and 98%. Kinetics and relative mass variation are comparable with results given in figures 9 and 10 for test series 2.

IV. 3.2 Effective gas permeability at given RH

Gas permeability results, for all samples stable at given RH%, are analysed from Figure IV.13. The initial state was investigated only for sample S3-9, which was then dried at 11% RH. Higher permeabilities are on the order of $10^{-17}$ m$^2$, which is lower by one to two orders of magnitude when compared to the results obtained for the second test series (see Figure IV.4 and 10). The dry density of S3 samples is slightly higher than that of the second series, which is sufficient to justify lower gas permeability.
As regards the global effects of saturation (and of volumetric variations – see Figure IV.13), there is a complete consistency between these results and those of the second series. Once again, it is observed that gas permeability becomes very low from 5 MPa confining pressure for sample S3-12 (RH=92%) and S3-13 (RH=98%), despite their incomplete saturation: \( S_w(S3-12,RH=92%)=72.8\% \) and \( S_w(S3-13,RH=98%)=91.7\% \). When accounting for their initial difference in gas permeability, these samples follow parallel evolutions when \( P_c \) varies, and these are less sensitive to an increase in confining pressure than in series S2.

For samples S3-10 and S3-11, the competition between change in saturation and volume variations is present, like in test series 2. Sample volume variations are given in Annex 2 for the whole testing process. S3-10 shrinks and loses mass, so that it is more permeable than S3-9 in the initial state: shrinkage brings lower pore volume, whereas water loss increases the pore volume accessible to gas, so that it is the latter effect, which is predominant. S3-11 swells and increases its saturation (mass intake), so that these opposite effects produce a higher gas permeability than in the initial state: swelling bring more pore volume accessible to gas than the amount that water intake fills with water.
IV. 3.3 Dry gas permeability

Dry permeability results are consistent with those measured on S2 series, see Figure IV.14. The order of magnitude is of $10^{-14} \, \text{m}^2$, like for S2 samples, which represents very high values when compared to the initial and partially water-saturated ones.

![Figure IV.14 Dry gas permeability of samples S3-9 to S3-13](image)

One should note that after compaction and drying/imbibition, despite contrasted residual volumes, see Annex 2, dry gas permeability (for both S2 and S3 series) exhibits a low scattering when compared to the initial gas permeability. Observations of a memory effect of RH sustained and drying/saturation upon dry gas permeability (made on S2 series) are no longer true.

Volume variations are presented in Annex 2 for the whole experimental process. As expected, swelling occurs in relation with mass increase, and shrinkage (or decrease in volume) is related to mass decrease, to drying or to compaction due to confining pressure, see also Table 4a. Although it is not observed on dry permeability results, a “memory effect” occurs in terms of volume change for sample S3-13 (98%RH): this sample has sustained an important 20% swelling (with a volume of $32.11 \, \text{cm}^3$), and yet, despite compaction and drying, the residual volume after oven drying (25.75cm$^3$) remains quite close to the initial volume (26.81cm$^3$).
Date: December, 4th, 2013  Deliverables D3-08, D3-24, D3-30 and D3-35

IV. 3.4 Pore volume variation of an oven-dried bentonite/sand plug

A last sample (S3-14) was prepared (compacted, oven-dried at 60°C until mass stabilization and tested) in order to measure its porosity change under confinement.

A dedicated test, similar in its principle to a pycnometric test, was designed using gas injection inside the sample at each step of loading and un-loading (Chen et al., 2012), see Figure IV.16.

![Figure IV.15 Porosity measurement device using gas injection. The sample is mounted in the triaxial cell, and access of gas is permitted on one side only. The buffer reservoir volume is Vo, the volume of the pipes is Vt=(V1+V2).

The sample is in the triaxial cell at given confining pressure Pc. Gas may access the sample on one side, yet it is not allowed to flow out of it (the downstream access valve is closed). Gas is injected from a calibrated reservoir of known volume Vr at a pressure P1, and it is assumed perfect, i.e. it follows the perfect gas law. After gas injection through the sample accessible pore volume, there is an equilibrium at a final pressure Pf such that, in the closed volume of the reservoir, gas pipes and sample pore volume, one gets (from the perfect gas law): P1Vr = Pf (Vr+Vt+Vp). This provides quantification of pore volume Vp, since reservoir volume Vr and pipes volume Vt are known via a preliminary test (which consists in replacing the sample by a non porous one). The manometer used to measure P1 and Pf has an accuracy of 10^-4 MPa.

From Vp data, conventional porosity $\phi$ is calculated using the sample initial volume V: $\phi = (Vp/V)$. Porosity results for sample S3-14 are given in Figure IV.16.
First, it is noted that porosity is of 30.1% at the lowest confinement used ($P_c=1\text{MPa}$): this is lower than porosity values assessed without confinement for all other samples of the S3 series (porosity ranges from 38.7% to 46.1%). This means that this limited confinement is sufficient to already close some porosity. Porosity changes are recorded along confinement/unconfinement cycles, up to 3MPa, 5MPa, 8MPa, 10MPa and finally up to 12MPa, the lower confinement being always of 1MPa. Results in Fig. 22 show a remarkable consistency, with good overlapping of porosity during re-loading with that upon previous unloading. It is noted that from the first confinement cycle, porosity decreases irreversibly, so that it does not reach again the initial value of 30.1%. This experiment, performed on a dried sample so that observed porosity changes may be maximal, shows directly and clearly that porosity accessible to gas decreases irreversibly when confinement is applied to the bentonite/sand plug. In direct relation with former gas permeability experiments, it is noted that porosity decreases linearly up to $P_c=5\text{MPa}$, above which it decreases more slowly. Such difference in behavior with increasing $P_c$ was also observed for $K_g$, see Figure IV 5-10 and 13. This is the signature of two different phases in pore collapse, as directly observed in Figure IV.16, possibly with a homogeneous decrease in pore volume up to 5MPa, and a more localized loss of pore volume above this value. Confirming this interpretation would require further research.
IV.4 Conclusion

This experimental study has aimed at assessing the sealing ability of partially water-saturated bentonite/sand plugs, under increasing hydrostatic stress, in relation with an actual in situ safety issue.

Indeed, in underground radioactive waste storage structures, bentonite/sand arrays of plugs are used to seal the repository galleries. These comprise an arrangement of so-called vertical slices, made of individual bentonite/sand plugs, which form the whole buffer. Similarly to laboratory experiments in constant volume conditions, the bentonite buffer swells in the fixed volume of an underground storage gallery. Yet, unlike small scale samples, plugs located at the rim of each vertical slice swell fully due to underground water coming from the surrounding host rock, while central plugs have less access to water, so that they are bound to be partially saturated for a long time. Therefore, in such context, due to fully swollen plugs at the rim, central bentonite/sand plugs are bound to be confined due to stresses close to 7MPa (which is the swelling pressure of the fully water-saturated plugs). The main question we have aimed at answering here was: are 7MPa confinement high enough to make a partially-saturated bentonite/sand plug fully impermeable to gas?

Our investigation was performed in three successive test series, on bentonite/sand plugs compacted using the same procedure: the first one has provided preliminary results, which have been confirmed by test series S2 and S3, as follows,

- when subjected to relative humidity of 70-75% and below, bentonite/sand plugs shrink and lose mass, whereas these swell and gain mass for RH>70-75%. At 98%RH, swelling is as huge as 20% of the plug initial volume and water saturation reaches 87-92%.

- Due to such huge volume variations, a discussion is proposed on the adequate reference volume to take in order to assess sample porosity and water saturation level. It is shown that the initial plug volume after compaction is a good compromise, which allows to assess a conventional porosity, ranging between 42 and 47%, depending on the test series. Initial water saturation level varies hugely from one test series to the other, by ranging (on average) between 52% for S2 and 64% for S3.

- After mass stabilization at given RH in free boundary conditions, gas permeability results show two opposite effects at given confinement. First, sample swelling is accompanied by an increase in total sample volume and in pore volume accessible to gas, so that this contributes to gas permeability increase. Similarly, sample shrinkage
corresponds to a decrease in sample volume and in pore volume accessible to gas, which promotes gas permeability decrease. Secondly, at given sample volume, water intake fills the pore volume partly, so that gas permeability decreases; at given sample volume, water loss promotes gas permeability increase. These antagonistic effects, brought from the literature, allow to interpret satisfactorily all our gas permeability experiments in partially saturated conditions.

- Tightness to gas is achieved for partially-saturated bentonite/sand plugs under confinement. In particular, at a confinement equivalent to the expected full swelling pressure of 7MPa, gas permeability is lower than $10^{-20}$ m$^2$ for samples initially saturated up to 86-91% only.

After gas permeability in partially water saturated conditions, samples are oven-dried at 60°C until mass stabilization: dry gas permeability is then on the order of $10^{-14}$ m$^2$, which is two to three orders of magnitude greater than the permeability in the partially water-saturated state (whatever the sample, and the saturation state between 50-91%). This testifies of the great ability of bentonite/sand plugs to undergo pore opening upon drying. This completes the observation of a 20% increase in volume upon swelling at 98%RH.
V - Swelling of compacted bentonite/sand plug (into tube) submitted to gas pressure
Introduction

This chapter aims to determine the effect of gas pressure (between 0 and 8 MPa) on the swelling properties and water saturation of compacted bentonite-sand mixture. This is a laboratory study to accompany the test of "PGZ" performed at Bure by Andra, in which pressurized gas is present during the saturation phase of bentonite-sand plugs. The water pressure applied throughout the experimental campaign is 4 MPa, which is close to the in situ water pressure.

The experimental program consists of two, or four, or six steps according to the tested sample, see Figure V.1 and Table V.1. A bentonite/sand plug (just after compaction) was completely saturated with water (phase I) and the saturated plugs was used to supply water to other plugs (by contact), in the presence or absence of gas pressure. For each Plexiglas-aluminum tube (in which the plugs is swollen), the calibration test (Phase II) permits to determine the relationship between the strain gauges (glued at outer surface) and the suffered internal pressure (phase III). Gas breakthrough pressure is the purpose of Phase IV. Thereafter, some samples have not been fully saturated with gas, are re-saturated by direct contact with water (phase V) and then tested again for their breakthrough pressure (Phase VI).

![Figure V.1 Experimental procedure followed by the three test series Ai, Bi and Ci.](image)

Table V.1 shows the nomenclature adopted for the names of the samples according to applied test conditions. For Ai series (A1, A2, A3 and A4), bentonite-sand plug swells without gas pressure: samples A1 and A2 are in direct contact (bottom) with a completely saturated plug, while sample A3 is in direct contact with water. Sample A4 is subjected to the same swelling conditions as sample A3, but its height is two times (50mm instead of 25mm). This allows evaluating a possible scale effect. For series B and C, bentonite/sand plug swells in the presence of both water (in contact with a fully saturated plug) and gas (4/6/8MPa).
Table V.1 Samples nomenclature and tests boundary conditions: water pressure is always 4MPa, while gas pressure is 0/4/6/8MPa. P_g is the downstream gas pressure; P_w is the upstream water pressure.

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>P_g (MPa)</th>
<th>P_w (MPa)</th>
<th>Swelling procedure</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1, A2</td>
<td>0</td>
<td>4</td>
<td>Phase I~ Phase IV</td>
<td>Direct contact with a fully saturated plug</td>
</tr>
<tr>
<td>A3</td>
<td>0</td>
<td>4</td>
<td>Phase II~ Phase IV</td>
<td>Direct contact with 4MPa water(one side)</td>
</tr>
</tbody>
</table>
| A4         | 0         | 4         | Phase II~ Phase IV | 1) H=50mm  
2) direct contact with 4MPa water (two sides) |
| B1, B2, C1, C2 | 4     | 4         | Phase I~ Phase V   | Direct contact with a fully saturated plug |
|            | 8         | 4         | Phase I~ Phase IV  | Direct contact with a fully saturated plug |
|            | 6         | 4         | PhaseIII~ Phase IV | Direct contact with a fully saturated plug |

V.1 Bentonite-sand plug swelling without gas pressure

V.1.1 Swelling pressure (SWP)

As indicated in Table V.2, three samples are saturated without gas pressure (No. A1, A2 and A3). Samples A1 and A2 are saturated by contact with a fully saturated plug, see Figure V1(a), while sample A3 is in direct contact with water, see Figure V1(b). Figure V.2 presents the evolution of swelling pressure of the sample A1~A3 as a function of time. It is clear that, for each plug, two different values of equilibrium swelling pressures are measured. They are, total equilibrium swelling pressure (higher value) and effective swelling pressure (lower value), respectively. As explicated in Chapter II.3.2, effective swelling pressure was obtained after water injection was stopped.

The effective swelling pressures of the three samples obtained are all of the same orders of magnitude, between 7.2MPa and 7.6MPa, which correspond to the target in situ value (between 7MPa and 8MPa), see Table V.2. The differences may be due to small dispersions of materials, saturation and compression, but overall we can be satisfied with the obtained results. The effective swelling pressure obtained for samples A1 and A2 are slightly lower than the relevant value of the sample A3 which is in direct contact with water, it is possible that they are not completely saturated, while the total equilibrium swelling pressures are different for the three samples. Figure V.2 (a) and Figure V.2 (b) can help us to understand the origin of these differences. The water pressure in the top tube for the sample A1 and
sample A2 (case a) varies between 0 and 2 MPa, whereas it varies between 0 and 4 MPa for the sample A3, so it is logical to see a pressure drop lower for the samples A1 and A2, when water pressure is set to 0, than for the sample A3. These pressure drops are quite consistent with the theoretical water pressure in the middle of the tube (1 MPa for the samples A1 and A2- Fig. V.2 (a), and 2 MPa for the sample A3- Figure V.2 (b).

![Figure V.2 (a): distribution of water pressure along the height of the tube: Case A; (b): distribution of water pressure along the height of the tube: Case B.](image)

**Figure V.2** (a): distribution of water pressure along the height of the tube: Case A; (b): distribution of water pressure along the height of the tube: Case B.

![Swelling pressure vs Time](image)

**Figure V.3** Evolution of swelling pressure with time, sample A1 A2 and A3.

Table V.2 summarizes the results of swelling tests of series A, $P_{\text{total}}$ is the total equilibrium swelling pressure, $P_{\text{eff}}$ is the effective swelling pressure, $\Delta P$ is the difference between $P_{\text{total}}$ and $P_{\text{eff}}$. 

Table V.2 summarizes the results of swelling tests of series A, $P_{\text{total}}$ is the total equilibrium swelling pressure, $P_{\text{eff}}$ is the effective swelling pressure, $\Delta P$ is the difference between $P_{\text{total}}$ and $P_{\text{eff}}$. 

67
Meanwhile, we can notice a different swelling kinetic for the two samples A1 and A2, while they are in the same conditions of swelling. This phenomenon can be attributed to the state of the degree of saturation the nether plug. While the lower degree of saturation means it's easier for the water to transfer through the nether plug and permeate into the upper plug, as a result, its swelling speed is quicker, and vice versa. The sample A3, directly contact with water, exhibits a rapid swelling kinetic, which is quite logical.

### V.1.2 Gas breakthrough test (GBT)

As we have mentioned before, gas breakthrough pressure is a very good indicator of the state of saturation of the sample and the tests developed allow evaluating the gas permeability of the material immediately after the passage of gas. Gas breakthrough test, conducted by small increase of gas pressure until detection of continuous gas flow, will play double roles: test the degree of water saturation and gas pathway, i.e. via the interface of bentonite/other material or through the bentonite.

Figure V.5 summarizes the results of gas breakthrough test of the four samples which lead to a discontinuous/continuous gas flow. It can be found that when the bentonite/sand mixture is fully saturated, discontinuous gas flow is detected at $P_g = 3.6\sim4.6$ MPa which is about 50% of the effective swelling pressure. Continuous gas breakthrough pressure through the tube-plug assembly is between 7.1MPa~8.1MPa, which is at least equal to the effective swelling pressure. Meanwhile, from the continuous gas breakthrough pressure, we also recall the value of effective swelling pressure. These results confirm other early tests (performed in our laboratory) and indicate that the continuous gas breakthrough pressure is always the same order of magnitude with the effective swelling pressure and is often a little higher. This conclusion is established on the premise that the sample is fully saturated. We can therefore ask where the gas passes, through the bentonite-sand plug or the interface? This question will be answered in Chapter VI.
Figure V.4 Summary of the gas breakthrough tests of the sample Ai. \( P_{\text{dis}} \) is the discontinuous gas breakthrough pressure; \( P_{\text{con}} \) is the continuous gas breakthrough pressure; \( P_{\text{eff}} \) is the effective swelling pressure.

Figure V.5 Evolution of \( Q_g \) with the upstream gas pressure.

Besides, we can also calculate the gas flow rate \( (Q_g) \) of the downstream gas pressure. Figure V.5 gives an example of the sample A2. The increase of the pressure in the downstream
chamber \((Q_g)\) is measured in MPa/h from temporal data provided by the downstream manometer: we note that while it doesn’t increase significantly when the discontinuous passage is uncertain (it remains in the order of magnitude of \(10^{-4}\) MPa/h), \(Q_g\) increases by one to two orders of magnitude when the continuous passage occurs. In case of malfunction of the argon detector (as we were), the parameter \(Q_g\) provides an independent measurement of discontinuous and continuous gas passage.

**V.2 Bentonite-sand plug swelling with gas pressure**

**V.2.1 Effect of gas pressure on the swelling pressure of bentonite-sand plug**

To perform the test, gas is injected from the top side of the assembly (plug 2 is placed above a completely saturated plug) and water is injected from the bottom side. Water saturation of the plug is performed by contact between the materials and capillary imbibitions. To evaluate the influence of the gas pressure, we will observe the effective swelling pressure and then the gas breakthrough pressure. Two ranges of gas pressure are applied: either 4MPa equal to the water pressure used to water saturation, or extreme value of 8MPa and then 6MPa (for sample C1).

We firstly carried out the tests with 4MPa gas pressure. Figure V.6 shows the effect of this pressure on the swelling kinetic of the plug. It can be found that the swelling kinetics of the two plugs are different. These differences may arise from the difference in the water permeability of the nether plug. There is no effect of gas pressure on the water pressure applied on the lower part of the assembly. We can also observe that the total equilibrium swelling pressures are not the same but, what's more important, the actual pressures (effective swelling pressures are substantially identical. This would tend to prove that, despite a little different of the initial conditions, a final equilibrium occurs with the simultaneous presence of water and gas giving a swelling pressure slightly lower than the plugs of A2 and A3. Remember that the plugs of A2 and A3 are swelling without gas pressure. The effective swelling pressures are a little smaller than the \(P_{eff}\) of the series A which swell without gas pressure, but this difference is only a little. Therefore, a question then arises whether the plugs are still fully saturated even in the presence of 4MPa gas pressure? This will be answered by the following gas breakthrough tests.
The phenomena are very different with a gas pressure of 8MPa. We are able to observe that the gas pressure has a first and immediate effect on the water pressure which is injected on the bottom side of the sample. The injection pressure is fixed at 4MPa but, when gas pressure is applied, the pore fluid pressure, being either gas or liquid is also increased. As a result there is a coupling effect due to the gas pressure. At this step, a choice had to be made that is either letting gas pressure control the water injection pressure or controlling the water pressure by draining the sample in order to maintain it at 4MPa. The first option is chosen because we suppose that the same kind of phenomenon would have taken place in situ. As a result, for the whole test, the water injection pressure is about 7MPa. Figure V.7 shows the evolution of the swelling pressure of the two plugs C1 and C2. We can see that the total equilibrium swelling pressures and effective swelling pressures of the two plugs are very close. Instantaneous increase of the swelling pressure is due to the application of 8MPa gas pressure. Compared with previous results, it can be seen a significant decrease of the effective swelling pressure, which is less than 5MPa. This means that the plug is only partially saturated, otherwise, the swelling pressure would have reached the target value (between 7 and 8 MPa).
V.2.2 Effect of gas pressure on the gas breakthrough pressure

From the previous results, we can see that the effective swelling pressure is affected by the gas pressure. This suggests that the saturation is imperfect and this can be judged by the measured gas breakthrough pressure, see Table V.3 and Figure V.8. It can be noted that the gas breakthrough pressure is much lower than the relevant effective swelling pressure: 1.49MPa ($P_{con}$) vs. 4.80MPa ($P_{eff}$). This can be deemed as the evidence that the sample is not fully saturated, besides we can deduce that gas migrates through the material and not the interface. This is different with the series A, where gas transfer through the interface of tube-plug when continuous gas breakthrough occurring. Meanwhile, it can also be noted that the effective swelling pressure is not sufficient to judge the state of saturation of the plug. For example, the $P_{eff}$ of samples B1 and B2 are 6.93MPa and 6.86MPa, respectively, and this suggest that the plug is fully saturated because the target effective swelling pressure is almost reached, however, the results of gas breakthrough tests negate this hypothesis and shows that the sample is far away from the fully saturated state. For the sample C2, 4.8MPa of the effective swelling pressure indicates that the plug is only partially saturated and this is confirmed by the following gas breakthrough test.
Table V.3 Summary of the swelling tests and gas breakthrough tests of the series B and C.

<table>
<thead>
<tr>
<th>Test No.</th>
<th>Peff (MPa)</th>
<th>Pdis (MPa)</th>
<th>Pcon (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>B1</td>
<td>6.86</td>
<td>1.01</td>
<td>2.16</td>
</tr>
<tr>
<td>B2</td>
<td>4.93</td>
<td>0.6</td>
<td>2.5</td>
</tr>
<tr>
<td>C2</td>
<td>4.8</td>
<td>0.12</td>
<td>1.49</td>
</tr>
</tbody>
</table>

Notes: Pdis of sample C2 is obtained at the first pressure step.

Figure V.8 Comparison of effective swelling pressures and gas breakthrough pressures of samples A2, B2 and C2.

V.2.3 Effect of re-saturation and/or a decrease in gas pressure

The above results clearly show that the plug is difficult to becoming fully saturated due to the presence of gas pressure, at least at 4MPa or more. In situ, due to release of gas after breakthrough, gas pressure in the disposal pit will decrease gradually, and at last the materials might return to the initial situation: Pw=4MPa and Pg=0. In order to simulate this situation, the following tests are performed. For the sample C2, after breakthrough test, we chose to inject gas again, but with 6MPa of gas pressure; while for samples B1 and B2, at the end of the breakthrough tests, we chose to re-saturate the samples again, namely, swelling without gas pressure.
Figure V.9 shows the evolution of swelling pressure of sample C2 as a function of time. Under 6MPa of gas pressure, it can be observed that gas pressure has no influence on the upstream water pressure, which is different with the previous case, e.g., swelling with 8MPa gas pressure. The total equilibrium swelling pressure is about 9.9MPa and the effective swelling pressure is stable at 6.4MPa. $P_{\text{eff}}$ has increased about 1.7MPa, which proves that the sample is a little more saturated when comparing with the former.

![Graph showing swelling pressure evolution](image)

Figure V.9 Evolution of swelling pressure of sample C2 with time (second time): after gas breakthrough test, swelling test is performed again but, this time the $P_g$ is 6MPa not 8MPa. It seems quite logical. The effective swelling pressure is quite close to the $P_{\text{eff}}$ obtained without or with 4MPa gas pressure. Then we start the gas breakthrough test again, and the results are summarized in Table V.4 and Figure V.10. We can see that gas pass through the sample at a very low value, only 0.6MPa, and after first passage, gas migration are accelerated which can be proved by the rate of increase of downstream gas pressure $Q_g$ and gas permeability $K_g$. This is also the evidence that the sample, even though it is re-saturated (by decreasing the gas injection pressure: from 8MPa to 6MPa), is far way form total saturation.
Table V.4 Results of gas breakthrough tests of sample C2: during the swelling period (after first gas breakthrough test), gas injection pressure is 6MPa.

<table>
<thead>
<tr>
<th>$P_{\text{upstream}}$ (MPa)</th>
<th>$P_{\text{downstream}}$ (10$^{-2}$MPa)</th>
<th>$V_{\text{detector}}$ (10$^{-3}$ml/s)</th>
<th>$Q_g$ (MPa/h)</th>
<th>$K_g$ ($10^{-20}$m$^2$)</th>
<th>Passage?</th>
</tr>
</thead>
<tbody>
<tr>
<td>0,6</td>
<td>12,28</td>
<td>6~7</td>
<td>0</td>
<td>0,01</td>
<td>yes (discontinuous)</td>
</tr>
<tr>
<td>1,01</td>
<td>9,32</td>
<td>6~9</td>
<td>0</td>
<td>0,06</td>
<td>4,24~5,67</td>
</tr>
<tr>
<td>2,17</td>
<td>0,7</td>
<td>20</td>
<td>&gt;4</td>
<td>0,09</td>
<td>8,2~18,7</td>
</tr>
<tr>
<td>2,51</td>
<td>3</td>
<td>60~90</td>
<td>&gt;10</td>
<td>0,21</td>
<td>21,6~33,6</td>
</tr>
<tr>
<td>3</td>
<td>1</td>
<td>60~70</td>
<td>&gt;10</td>
<td>1,55</td>
<td>37,9~48,7</td>
</tr>
<tr>
<td>3,5</td>
<td>59,5</td>
<td>600~700</td>
<td>&gt;10</td>
<td>13,75</td>
<td>679~716</td>
</tr>
</tbody>
</table>

Figure V.10 Results of gas breakthrough tests of sample C2.

Samples B1 and B2 are re-saturated by direct injection of water on one side at first and then on both sides. Figure V.11 presents the evolution of swelling pressure of the plug B2 versus time. It can be observed that the initial effective swelling pressure (6.06MPa) is less than the $P_{\text{eff}}$ observed in Figure V.6 (6.86MPa). This phenomenon can be attributed to a slight desaturation of material induced by the previous gas breakthrough test. The differences of total equilibrium swelling pressure, when stopping the injection of water on one side and then on both sides, are consistent with the hypothesis of the linear distribution of water pressure along the height of tube (always coupled with an assumed behavior of Terzaghi type). At the end of
the swelling test, the effective swelling pressure is 7.88MPa, which is slightly higher than the effective swelling pressure of samples A1, A2 and A3. It is a common phenomenon that we have observed in our experiments in which the material is subjected to successive cycles of water pressure and gas pressure. After all of these operations, re-saturation thus leads to a higher effective swelling pressure than the correspond value obtained in first saturation, and this is the sign that the various operations have modified the microstructure of the material and its swelling in the tube (associated with gas overpressure) has certainly changed its dry density. This particular point deserves further investigation. The same phenomenon is also observed for sample B2.

At the end of the test of re-saturation, gas breakthrough test is performed and the results are summarized in Table V.5 and Figure V.12. It can be observed, in this table, a significant gain of the effective swelling pressure and also very significant changes of gas breakthrough pressure, both discontinuous and continuous. For sample B1, the characteristics are similar to sample A3, which has been soaked by direct contact with water. However, the behavior of sample B2 is a little amazing, despite a comparable discontinuous gas breakthrough pressure with the other samples, continuous gas breakthrough pressure is much higher and it is difficult to measure the actual gas breakthrough pressure due to the limit of the device. It is therefore more than 9.8MPa.

Table V.5 Comparison of the swelling pressure and breakthrough pressure before and after re-saturation: samples B1 and B2

---

Figure V.11 Evolution of swelling pressure of samples B1 and B2 with time: re-saturation.
Date: December, 4th, 2013  Deliverables D3-08, D3-24, D3-30 and D3-35

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>P\textsubscript{total}</th>
<th>P\textsubscript{eff}</th>
<th>P\textsubscript{dis}</th>
<th>P\textsubscript{con}</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>MPa</td>
<td>MPa</td>
<td>MPa</td>
<td>MPa</td>
<td></td>
</tr>
<tr>
<td>B1</td>
<td>12.56</td>
<td>6.86</td>
<td>1.01</td>
<td>2.16</td>
<td>First test</td>
</tr>
<tr>
<td></td>
<td>11.42</td>
<td>7.88</td>
<td>3.57</td>
<td>8.53</td>
<td>After resaturation</td>
</tr>
<tr>
<td>B2</td>
<td>10.72</td>
<td>6.93</td>
<td>0.6</td>
<td>2.5</td>
<td>First test</td>
</tr>
<tr>
<td></td>
<td>11.3</td>
<td>7.4</td>
<td>4</td>
<td>&gt;9.8</td>
<td>After resaturation</td>
</tr>
</tbody>
</table>

![Graph](image.png)

Figure V.12 Results of gas breakthrough tests: sample B1.

**V.2.4 Effect of gas pressure on plug-tube interface contact pressure**

During the gas breakthrough test, we find that contact pressure/swelling pressure between the plug and the tube interface is affected by the injected gas pressure. For example, the swelling pressure will augment gradually with the increase of gas pressure. In order to find the relationship between the increasing gas pressure $\Delta P_g$ and the corresponding contact pressure increase $\Delta P_{contact}$, we record the value of the strain gauges during the whole period. When gas pressure $P_g$ is increased, after an immediate response, the contact pressure keeps on increasing until becoming stable and we record this stable contact pressure. Gas pressure is increased by 0.5MPa or 1MPa every time, and corresponding stable contact pressure/swelling pressure is recorded, see Figure V.13. We choose the samples of B1 and B2, before and after re-saturation, two kinds of situations, to analyze. It can be found that the slope of fitting curve $(P_g \Delta P_{contact})$ is between 0.43 and 0.52. Similar phenomena can also be found in Davy et al. (2009). This shouldn't be overlooked, which means that contact pressure/swelling pressure will increases more than 1 MPa when gas pressure is above 2.5 MPa. This coupling effect between gas pressure and contact pressure of plug-tube interface is partially important when
considering the *in situ* gas migration. This phenomenon may be attributed to the coupling effect between the pore fluids (i.e. water pushed by gas) and the bentonite solid skeleton or the linear distribution of gas pressure along the height direction of the tube, just like the distribution of water pressure along the height direction of the tube. Besides, this finding can also supply some useful information to the numerical simulation, e.g., boundary condition. Effect of the upstream gas pressure upon the contact pressure/swelling pressure the plug-tube interface can be generalized as follows:

\[ P_{\text{contact}} = P_{\text{eff}} + (0.43 \sim 0.52) \times P_g \]

Figure V.13 Effect of gas pressure on the plug-tube contact pressure/swelling pressure

Therefore, bentonite swelling pressure can be predicted by the upstream gas pressure during the gas breakthrough test. This relationship is obtained with the two kinds of situations, samples B1 and B2, before and after re-saturation. While for samples C1 and C2, it is difficult to obtain this similar relationship due to lower degree of saturation of the plug which leads to the quick decrease of the upstream gas pressure. Therefore, it is difficult to maintain the upstream gas pressure constant during the gas breakthrough test.

**V. 3 Effect of sample height upon swelling and GBP**

To research effect of the sample height upon the swelling pressure and GBP of bentonite, another test, with the 50mm height of sample, was performed.
V. 3.1 Swelling pressure (SWP)

As shown in Figure V.134, water was firstly injected from the upstream side, and then on the two sides to accelerate the saturation speed. Figure V.15 presents the evolution of swelling pressure of the sample D2 versus time. It can be noticed that the total swelling pressure is about 12.71MPa, while the effective swelling pressure is 7.05MPa. The difference of these two values is 5.66MPa, which is higher than the \( \Delta P_s \) of samples A1 and A2, about 1MPa, and also is larger than the \( \Delta P_s \) of sample A3, about 2MPa. The reason can be found from the Figure V.14 and Figure V.15, which the average water pressure is about 5MPa during the last period: between 10\(^{th}\) and 18\(^{th}\) day. Because water is injected from both sides, this means that the distribution of water pressure along the tube is linear. When water injection is stopped, this pressure will disappear and lead to a decrease of 5MPa of swelling pressure. Another phenomenon can be found that the effective swelling pressure is also approaching the \textit{in situ} swelling pressure, about 7.05MPa. This means that change of height of the sample doesn’t affect its swelling characteristic.

Figure V.14 Swelling process: the height of sample (H=50mm) is two times long of the samples performed in other tests.
V. 3.2 Coupling effect between gas pressure and deformation of bentonite-sand plug

The second part of the test is effectuated to research the coupling effect between gas pressure and the deformation of bentonite-sand plug, which is reflected by the corresponding swelling pressure. The experimental process can be shown in Figure V.16. Gas pressure is firstly injected from the upstream side with 6MPa gas pressure and then from both sides, then gas pressure will decrease to 2MPa. Each step will last several days to let the swelling pressure becoming stable. The second cycle repeats the first cycle, namely, gas pressure is injected firstly with 6MPa from both sides and then with 2 MPa. At last, gas pressure will decrease to 0 to measure its effective swelling pressure.
Figure V.16 Experimental procedure of «gas - water coupling» test for sample A4, indicating the total swelling pressures of each step ($P_{\text{total}}=7.03\,\text{MPa}, 9.93\,\text{MPa}, 13.34\,\text{MPa}, 9.28\,\text{MPa}, 13.36\,\text{MPa}, 9.44\,\text{MPa}, \text{et} 7.15\,\text{MPa})$.

Figure V.17 Results of «gas - water coupling» test: evolution of swelling pressure with time. As shown in Figure V.17, when 6 MPa gas pressure is performed, there is an instantaneous effect on swelling pressure, which increases from 7.03 MPa to 9.93 MPa, i.e., 2.9 MPa. This increase corresponds to the interstitial pressure at the position of the strain gauges, if we suppose a linear distribution of this pressure (from 6 MPa at upstream face to 0 MPa at the
downstream face). This is equal to the increase of the average total stress according to Terzaghi. The swelling pressure is stable at this value until gas pressure is imposed from two sides. Here, if we always suppose a linear distribution of applied pressure, between 6MPa (upstream face) and 6MPa (downstream face), the total increase of the interstitial pressure at the position of strain gauges is equal to 6MPa (3MPa when we apply $P_g=6$MPa at upstream/downstream face only). After two or three days, the swelling pressure increases about 3.3MPa up to 13.34 MPa. This confirms the behavior of Terzaghi again.

The successive unloading of gas pressure confirms this same analyzing of Terzaghi behavior, see Figure V.15 and Figure V.16: for example, when we apply 2MPa gas pressure from two sides of the sample (instead of 6MPa, i.e., a decrease of 4MPa), the total swelling pressure decreases from 13.34 to 9.28 MPa, namely, a decrease of 4.06MPa; when $P_g=6$MPa is performed on both sides, total swelling pressure ($P_{\text{total}}$) is about 13.36MPa instead of 13.34MPa (a very close value) of the previous cycle, then when $P_g=2$MPa is performed on the two sides, $P_{\text{total}}= 9.44$MPa (instead of 9.28MPa of the previous cycle). Finally, at the end of the complete cycle, the effective swelling pressure is about 7.03MPa comparing with 7.15MPa at the beginning of the test. This difference is not significant, giving the dispersion of measuring method by strain gauges. At this stage, the gas pressure, applied to the sample, has no effect on the effective swelling pressure.

V. 3.4 Re-saturation of the sample

At the end of the previous cycle of gas injection, water (4MPa) is injected again to re-saturate the sample, see Figure V.18. Figure V.19 shows the evolution of swelling pressure versus time during the process of re-saturation. It is very interesting to note that between the initial phase and stabilization with water pressure on both sides, the swelling pressure increases about 5.54 MPa, while the water pressure is always stable at 4MPa, which indicates that the increase of swelling pressure is higher than water pressure. This can be attributed to the fact that the bentonite-sand mixture is not fully saturated before re-saturation, thereby generating a little recovery of swelling. This insufficient saturation is due to a small de-saturation caused by the previous «gas - water coupling» test. At the end of re-saturation, the effective swelling pressure is about 7.83 MPa, which is a little higher than the $P_{\text{eff}}$ of samples A1, A2 and A3, subjected to $P_g=0$MPa and with the height of 25mm only. A similar phenomenon has already been found in the re-saturation test of samples B1 and B2, which indicates that various operations have changed the microstructure of the material and its swelling in the tube.
(associated with gas pressure). However, the value of $P_{\text{eff}}$ is still close to the corresponding values of samples A1, A2 and A3 ($H=25\text{mm}$).

Figure V.18 Experimental process of re-saturation ($H=50\text{mm}$).

![Experimental process of re-saturation](image)

Figure V.19 Results of re-saturation test: evolution of swelling pressure versus time.

![Results of re-saturation test](image)

V. 3.3 Gas breakthrough test

Figure V.20 summarizes the gas breakthrough pressures of sample A4 which leads to a discontinuous/continuous gas flow. Similar to A series, discontinuous gas breakthrough pressure is found at about 3.02 MPa, and continuous gas breakthrough pressure is measured at about 8.5 MPa. This means that increase of sample’s height has no significant effect on the sealing efficiency of the bentonite-sand mixture which can be proved by its gas breakthrough...
pressures. Another phenomenon is that continuous gas flow is discovered when gas pressure exceeds the effective swelling pressure, which dovetails nicely with the results of A series. There is still a question proposed before us, namely, how about the gas pathways: through bentonite-sand plug or through the interface? This will be answered in the next chapter.

Figure V.20 Comparison of gas breakthrough pressures between sample A3 (H=25mm) and A4 (50mm).

V. 4 Conclusion

This chapter presents several interesting points about the behavior of bentonite-sand mixture under the effect of gas pressure during its swelling. To assess the impact of the gas pressure on the saturation of the material, we used a device to measure gas breakthrough pressure, which is very sensitive to imperfect saturation of the porous material. The passage of gas through the medium occurs first by a discontinuous (intermittent) flow and then a free and continuous passage which permits measuring a significant permeability of the assembly of plug + Plexiglas-aluminum tube.

We have shown that the presence of gas pressure, during water saturation, plays an important role on the effective swelling pressure. For a gas pressure equal to the water pressure ($P_g = 4$ MPa), this influence is not obvious (slight reduction of the swelling pressure), but when gas pressure is two times of the water pressure ($P_g = 8$ MPa), there is a very significant drop in the
effective swelling pressure. Following the partial swelling ($P_g = 8$ MPa), further testing at a lower pressure ($P_g = 6$ MPa) allows to complete the saturation and to increase the effective pressure.

Breakthrough pressure is a useful tool which permits us to assess the "quality" of the saturation. For the samples whose saturation was proven (A1, A2 and A3, swelling without gas pressure), the continuous breakthrough pressure $P_{con}$ is lightly higher than the actual effective swelling pressure $P_{eff}$: $P_{con}$ is between 7.10 and 8.10 MPa, while $P_{eff}$ is between 7.16 and 7.59 MPa. Otherwise, regardless of the gas pressure (4, 6 or 8 MPa), our tests showed that the saturation is not complete. At $P_g = 6$ and 8 MPa, the sample is almost immediately permeable, proving that the material is far from saturated.

Increasing the length of the sample (it is multiplied by two) has little influence on the properties of swelling and breakthrough pressure. It does not appear that there is a significant scale effect.

Finally, all these tests have allowed us to verify that the hypothesis of a material of Terzaghi (Biot coefficient is equal to 1), but some dispersions of our measuring methods are inefficiently fine to control the coupling effects of fluid-skeleton of this type of material, leading us to remain cautious about the impact of successive cycles - increasing gas pressure, water pressure etc ... This will be the subject of dedicated tests planned for the near future.
VI - Swelling and GBP of argillite, bentonite and argillite / bentonite interface
Introduction

This chapter aims at identifying gas leakage pathway in geologic repositories, e.g. through bentonite/sand plug, argillite or bentonite/argillite interface. In order to answer this question, gas breakthrough pressures (discontinue and continue) and gas leakage rate \(Q_g\) are used to determine the gas migration pathway. The experimental procedure is as follow:

1. Swelling without Plexiglas-aluminium tube (wrapped directly into a Viton™ membrane and placed inside the confining triaxial cell);
2. Swelling in an aluminium tube whose inner surface is grooved, to make the passage of gas more difficult by the presence of obstacles generated by the grooves;

Table VI.1 Definition of tests performed

<table>
<thead>
<tr>
<th>Sample</th>
<th>(P_w) (MPa)</th>
<th>(P_g) (MPa)</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>D1</td>
<td>1.5</td>
<td>0</td>
<td>Bentonite/sand plug swelling without tube</td>
</tr>
<tr>
<td>D2</td>
<td>1.5</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>E1</td>
<td>4</td>
<td>0</td>
<td>Bentonite/sand plug inside a grooved aluminium tube</td>
</tr>
<tr>
<td>E2</td>
<td>4</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>F1-1~F1-3</td>
<td>4</td>
<td>0</td>
<td>Bentonite/sand plug inside an smooth argillite tube</td>
</tr>
<tr>
<td>F2-1, F2-2</td>
<td>4</td>
<td>0</td>
<td>Bentonite/sand plug inside a grooved argillite tube</td>
</tr>
</tbody>
</table>

VI.1 Swelling and GBP of bentonite/sand plug without tube (samples D1 and D2)

VI.1.1 Water injection test of plugs D1 and D2

As shown in Figure VI.1 and Table VI.1, we chose to inject upstream water pressure \(P_w = 1.5\) MPa and with a confinement \(P_c = 2.5\) MPa, so as not to significantly alter the microstructure of the plugs (e.g., dry density), since here they are kept in a Plexiglas-aluminium tube. Based on the previous experimental results, this phase usually lasts at least one month to ensure that the bentonite-sand plug is fully saturated. Here, we use water permeability to judge the state of saturation of the sample, because the tube is not equipped with strain gauges and we cannot
measure the swelling pressure. Subsequently, the triaxial cell is disconnected to water-supplying Gilson\textsuperscript{TM} pump. This phase lasts at least 72h in order to re-equilibrate the internal water pressure to atmospheric pressure, and to check that no gas is present inside the water, as this may disturb the measurements of gas migration. Then confining pressure is increased to 7MPa, which is similar to the \textit{in situ} swelling pressure, and gas migration tests are performed by small increase of gas pressure until 6MPa. If continuous flow is not measured at $P_c = 7$ MPa and $P_g = 6$MPa, we will increase gradually $P_c$ up to 12 MPa (\textit{in situ} lithostatic pressure) and $P_g$ until 11MPa. Compared to the previous tests, it should ensure a good seal between the Viton\textsuperscript{TM} jacket and the sample, which can only be done by increasing the confinement. Besides, confining pressure $P_c$ should be always higher than $P_g$ due to the limit of our device.

Figure VI.1 Experimental process: swelling test and gas breakthrough test.

\textbf{Figure VI.2(a) and (b) presents the results of swelling tests of samples D1 and D2:} evolution of the water volume into the plug and water permeability with time. As shown in the figures, water permeability of the two samples becomes stabilized after about 200 hours of injection. The initial water permeability of the bentonite-sand plug is between $7.62 \times 10^{-18} \sim 1.33 \times 10^{-17} \text{ m}^2$, which is a little smaller than the initial gas permeability of the bentonite-sand plug, about $1.73 \times 10^{-17} \text{ m}^2$ at $P_c = 3$MPa (sample S3-9). Saturation and swelling of the material will cause a progressive decrease of water permeability until a stable value of $2.22 \times 10^{-20} \text{ m}^2$ (Sample D1) or $1.78 \times 10^{-20} \text{ m}^2$ (Sample D2). These values are much lower than the gas permeability in the dry state of the plugs compacted in a similar manner, see Chapter IV: in the dry state, the magnitude of the gas permeability is of $10^{-14} \text{ m}^2$, namely, 6 orders of magnitude higher than the water permeability.

\textbf{Remark:} When applying a gradient of water pressure on a partially water-saturated material, water flow through the pores can be divided into two parts: water flow through the saturated pores $Q_{\text{per}}$ and the imbibitions of capillary flow through the empty pores $Q_{\text{cap}}$ (Liu, 2012). In our calculation, we only consider the effect of the $Q_{\text{per}}$ and overlook the influence of the $Q_{\text{cap}}$. 
Figure VI.2 (a) swelling test of the sample D1; (b) swelling test of the sample D2.

### VI.1.2 Gas breakthrough test of plugs D1 and D2

Subsequently, gas breakthrough tests are executed. As shown in Figure VI.3, we don’t find continuous gas flow for sample D1 during the whole process, but detecting discontinuous gas flow at $P_g=11\text{MPa}$ and $P_{\text{upstream}}=9\text{MPa}$. For sample D2, the results of breakthrough test is a little different: intermittent / discontinuous flow is detected at a value significantly lower than for sample D1, at $P_g=2.12\text{MPa}$.
By contrast, such as for sample D1, we never get the continuous flow. This point coincides with the study of permeability related to the level of saturation and confinement in Chapter IV, where we have found that confinement reduced sharply the gas permeability, which is likely to prevent the continuous gas breakthrough. The discontinuous gas breakthrough pressure of the sample D2 is a little amazing, and we prefer to conclude that the continuous breakthrough pressure can never be less than either the swelling pressure (in the presence of an outer tube), or - here (without tube) - the confining pressure. This test doesn’t allow evaluating the continuous breakthrough pressure, because the tightness of the assembly requires a confinement always higher than the gas injection pressure.

![Graph showing gas breakthrough pressures of samples D1 and D2.](image)

**Figure VI.3 Gas breakthrough pressures of samples D1 and D2.**

**VI.2 Swelling and GBP of a bentonite-sand plug with a grooved tube (plugs E1 and E2)**

**VI. 2.1 Water injection test of plugs E1 and E2**

As presented in Chapter V, gas will migrate through the assembly of tube-bentonite/sand plug, when gas pressure exceeds the effective swelling pressure. At this stage, we cannot conclude about the pathway of gas migration: via the interface of tube – bentonite/sand plug or through bentonite/sand plug. A test was specifically designed, using a tube whose inner surface is grooved (not threaded) to answer this question, see Figure VI.4. Indeed, the presence of
grooves creates a contact zone between tube-plug through which the circulation of gas flow is very difficult (creation of barricade). Once again, it is the measurement of water permeability which allows judging the complete saturation of the plug (no strain gauges on the outer face of the tube).

Figure VI.4 The grooved aluminium tube is used in the experiment: Samples E1 and E2.

The swelling procedure is the same with sample A4, which is in direct contact with water flow with $P_w = 4$MPa and $P_c = 12$MPa: phase I, water is injected only on the upstream side until stabilization of the water permeability, then (phase II), water is injected from both sides. Once the sample is completely saturated, which is judged by the value of the water permeability ($K_w$) and water flow rate ($Q_w$), gas breakthrough test is conducted by small increments of injection until 10-10.5 MPa, see Figure VI.5 (limit of the capacity of argon cylinder).

Figure VI.5 Experimental procedure of the samples E1 and E2: the inner surface of the tube is grooved.
Figure V.6 (a) and (b) show the results of swelling tests: evolution of water volume (enter the bentonite-sand plug) and water permeability versus time. The swelling kinetics of samples E1 and E2 are similar, namely, water permeability becomes stabilized after 40~50 hours water injection. When comparing with samples D1 and D2, the swelling kinetics of sample E1 and E2 are more rapid. The differences may be due to the different water injection pressure: 1.5MPa vs. 4MPa. The higher water injection pressure accelerates the swelling kinetic of sample, similar phenomena are also found by Liu (2012).

The initial water permeability of sample E1 is $1.72 \times 10^{-16} \text{ m}^2$, which is quite higher when comparing with the initial water permeability of samples D1 and D2 ($7.62 \times 10^{-18} \sim 1.33 \times 10^{-17} \text{ m}^2$): in fact, an initial clearance exists between the plug and the inner face of the tube, which increases the capacity of initial water transfer. Therefore, the initial water permeability $K_{\text{ini-w}}$ is only indicative. Water flows through the tube-plug interface, and a few minutes later, we observe the hydraulic cut-off, which can be attributed to the increase of the saturation of the plug and the beginning of the sealing of the interface (due to swelling of the plug), see Figure VI.6 (a). Water permeability decreases quickly to $3.09 \times 10^{-19} \text{ m}^2$ and continues to decline until it reaches a stable value of $1.35 \times 10^{-20} \text{ m}^2$, which is slightly lower than the stable values of samples D1 and D2 ($2.22 \times 10^{-20} \text{ m}^2$ and $1.78 \times 10^{-20} \text{ m}^2$, respectively). As these samples (E1, D1 and D2) are complete saturated, the explanation for this observation can only due to the difference of the confining pressure: $P_c = 2.5 \sim 3 \text{ MPa}$ (samples D1 and D2) and $P_c = 7 \sim 8\text{ MPa}$ (samples E1 and E2). At these values, the confinement therefore has some limited influence on the water permeability of the samples. For sample E2, hydraulic cut-off also occurs in a few minutes, see Figure VI.6 (b). The water permeability is measured after the occurrence of this hydraulic cut-off. It decreases rapidly, until it stabilizes at a value of about $7.5 \times 10^{-21} \text{ m}^2$, lower than that of E1 (D1 and D2).
Figure VI.6 Results of swelling tests: (a) sample E1, (b) sample E2.

VI. 2.2 Gas breakthrough test of plugs E1 and E2

Figure VI.7 and Table VI.2 summarize the results of gas breakthrough tests of samples E1 and E2. For sample E1, discontinuous gas flow is detected at $P_g = 4$–5MPa, while this value is about 10MPa for sample E2. Similar phenomena, already detected for previous sample series, are confirmed: discontinuous gas flow is not stable and reproducible, i.e. the discontinuous gas breakthrough pressure is not reproducible from one test to another. For sample E1, we don't obtain the continuous breakthrough pressure even through 10 MPa gas pressure is performed (limit of the device). However, at $P_g = 10$MPa, after 6 days and additional 20h, a continuous flow of gas is detected for sample E2, see Figure VI.7 and Table VI.2. This phenomenon can be attributed to the gradual decrease of water saturation of the sample. The swelling pressure was not measured, because the tube was not instrumented with strain gauges, but we can assume that it is identical to that obtained for other samples prepared under the same conditions.

As gas doesn't pass through the sample-tube under 10.5MPa, when recalling previous results of continuous gas breakthrough pressures of sample A1–A3 ($P_{con} = 7$–8MPa), see Figure VI.7 and Table VI.2, we can assume that, in the previous tests, gas transfer via the interface and not through the material matrix in the presence of a smooth tube. In the presence of a grooved tube, gas can migrate via the interface, but with more difficulty, or through the swollen
material, resulting in a significantly higher breakthrough pressure (beyond $P_g = 10$ MPa for continuous passage).

Similar phenomenon are also found by other researchers, which indicate that the interfaces between the clay and another material (argillite, granite, steel) are the preferential pathways for gas migration instead of the clay materials in a saturated system (Popp et al., 2013; Davy et al., 2009; Arnedo et al., 2011). This can be explained that it is very difficult for the gas to pass through the clay due to very fine pores, i.e., a very high capillary pressure (Push et Forsberg, 1983). In contrast, the bentonite/bentonite interfaces will not be preferential pathways for gas migration due to the cohesion between contact planes, while the system is completely saturated (Popp et al., 2013). On the other hand, we measured the intermittent gas flows from a lower pressure than the confining pressure: This indicates that preferential paths were created, but with an unstable flow. It is due to the effects of snap-off or capillary (Rossen, 2000) or progressive micro-cracking of the clay material (Horseman, 1999).

Anyway, continuous breakthrough pressure obtained for the argillite – that it is cracked like the EDZ (Excavation Damaged Zone) or intact (Skoczylas and Davy, 2011; M'Jahad, 2012, Davy et al., 2012), are always lower than 7 MPa. It means that the continuous gas breakthrough pressure of argillite is smaller than the $P_{con}$ of the plug-tube interface (measured on the bentonite-sand plug that they have swollen in the presence of a smooth or grooved tube). It is therefore unlikely that gas passes through saturated "bentonite-sand" material: instead it circulates at the interfaces or argillite itself.

For samples E1 (grooved tube) and A2 (smooth tube), Figure VI.8 present the rate of increase of the downstream pressure $Q_g$ (estimated from the data of the downstream manometer when the downstream chamber is closed), depending on the applied upstream gas pressure. It shows clearly that gas breakthrough pressure has changed considerably since the only significant change of the experimental conditions is the ease of passage through the interface: for $P_g \leq 10.5$ MPa, there is no free passage as measured by the $Q_g$ in the presence of a grooved interface, unlike the smooth interface, for which the passage takes place at $P_g = 7.1$ MPa, see Figure VI.8. This means that it is the tube-plug interface which governs the passage of gas through the assembly and not the material.
Table VI.2 Summary of the gas breakthrough tests of the series A and E.

<table>
<thead>
<tr>
<th>Test NO</th>
<th>( P_{\text{eff}} ) (MPa)</th>
<th>( P_{\text{dis}} ) (MPa)</th>
<th>( P_{\text{con}} ) (MPa)</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>7.42</td>
<td>4.1</td>
<td>N/A</td>
<td>the inner surface of the tube is smooth</td>
</tr>
<tr>
<td>A2</td>
<td>7.16</td>
<td>3.6</td>
<td>7.1</td>
<td>the inner surface of the tube is smooth</td>
</tr>
<tr>
<td>A3</td>
<td>7.39</td>
<td>4.6</td>
<td>8.1</td>
<td>the inner surface of the tube is smooth</td>
</tr>
<tr>
<td>E1</td>
<td>N/A</td>
<td>5~6</td>
<td>higher than 10</td>
<td>the inner surface of the tube is smooth</td>
</tr>
<tr>
<td>E2</td>
<td>N/A</td>
<td>10</td>
<td>higher than 10</td>
<td>the inner surface of the tube is grooved; ( P_{\text{con}} ) is measured at ( P_g=10\text{MPa} ) after 6 days and 20h.</td>
</tr>
</tbody>
</table>

Figure VI.7 Results of gas breakthrough test: samples E1 and E2; for sample E2, continuous gas flow is detected at \( P_g=10\text{MPa} \) after 6 days and 20h.
VI.3 Swelling and GBP of bentonite/sand plug + smooth argillite tube

VI.3.1 Water injection tests of plugs F1-1, F1-2 and F1-3

Figure VI.9 depicts water permeability and volume of water injection versus time for plugs F1-1, F1-2 and F1-3. The first and main observation was that more water was absorbed by the bentonite/sand-argillite assembly and the swelling kinetic was a little slower when comparing with plugs E1 and E2 (bentonite/sand plug swollen inside a grooved aluminium tube). This is logical, because the aluminium tube is impermeable to water while the argillite tube is permeable to water. Water permeability was measured after the occurrence of hydraulic cut-off, which was about $2.78 \sim 4.52 \times 10^{-19} \text{ m}^2$. This value was similar to plug E1 after five hour’s water injection ($3.09 \times 10^{-19} \text{ m}^2$). After 200 hours water injection, water permeabilities of plugs F1-1, F1-2 and F1-3 were stable at $0.68 \times 10^{-20} \text{ m}^2$, $0.94 \times 10^{-20} \text{ m}^2$ and $1.54 \times 10^{-20} \text{ m}^2$ respectively, while the corresponding values were about $0.75 (1.35) \times 10^{-20} \text{ m}^2$ for plugs F1-2 (F2-2) and $1.78 (2.22) \times 10^{-20} \text{ m}^2$ for plugs D1-1 (D1-2). This means that the water permeability of the completely saturated argillite tube $K_{\text{sat-w}}$ is similar to or lower than the bentonite-sand mixture. Other researchers in our laboratory (M’Jahad, 2012) have measured the $K_{\text{sat-w}}$ of argillite: it was similar to the bentonite-sand plug and bentonite/sand-argillite assembly, with the value of about $1.13 \times 10^{-20} \text{ m}^2$ (varying between $0.32 \sim 1.89 \times 10^{-20} \text{ m}^2$).
This means that the fully saturated water permeability (absolute permeability) of argillite, bentonite/sand plug and bentonite/sand-argillite assembly are of the same order of magnitude ($10^{-20}$-$10^{-21}$ m$^2$). From water permeability alone, it is then expected that tightness to water can be obtained for bentonite/sand plug, argillite tube and bentonite/sand-argillite assembly when they become completely saturated.

![Figure VI.9 Evolution of water permeability and water injection volume with time: plugs F1-1 F1-2 and F1-3.](image)

**VI. 3.2 Gas breakthrough tests of plugs F1-1, F1-2 and F1-3**

*Figure VI.10* shows the gas breakthrough pressures of mixed plugs F1-1, F1-2 and F1-3 (both discontinuous $P_{\text{dis}}$ and continuous $P_{\text{con}}$). It could be found that the discontinuous breakthrough pressures were about 1.54 MPa (plug F1-1), 3.60 MPa (plug F1-2) and 4.5 MPa (plug F1-3), and the continuous breakthrough pressures are 6.94 MPa (test F1-2), 7.41 MPa (test F1-2) and 6.03 MPa (F1-3). These values were close to or smaller than those of bentonite/sand plugs swollen into a smooth Plexiglas-Aluminium™ tube (e.g. plugs A1-2 and A2-1). As proved by previous tests, gas would pass through the interface between the bentonite/sand plug and the smooth Plexiglas-Aluminium™ tube, when gas pressure approached or exceeded the effective swelling pressure (between 7~8MPa). For the gas migration pathway through the bentonite/sand –argillite assembly, it means that there exist two possibilities: either through the plug-tube interface or through the argillite, which will be checked by experiments with a grooved argillite tube, namely plugs F2-1 and F2-2. Besides, for plug F1-2, it was re-saturated.
again after 1st time of GBT, and then GBT was performed again at the end of resaturation. It was found that both discontinuous breakthrough pressure and continuous breakthrough pressure were smaller than the corresponding values of the first time. In this respect, it is different with the situation that bentonite/sand plug swells inside a smooth Plexiglas-Aluminium™ tube, for which sealing ability ($P_{\text{con}}$ and $P_{\text{dis}}$) can be recovered after resaturation. This difference can be attributed to the different materials of tube, i.e. Plexiglas-Aluminium™ tube and argillite tube. This is also an indirect proof that the passage of gas through the bentonite/sand-argillite assembly is controlled by the argillite tube instead of bentonite/sand plug or sample-tube interface. Besides, as can been seen from Figure VI.11, the rate of increase of downstream gas pressure ($Q_g$) increases a lot when continuous gas breakthrough occurs.

![Figure VI.10 Gas breakthrough pressures of plugs F1-1, F1-2 and F1-3 (resaturation).](image)

Figure VI.10 Gas breakthrough pressures of plugs F1-1, F1-2 and F1-3 (resaturation).
Figure VI.11 Relationship between the rate of increase of the downstream gas pressure $Q_g$ and the applied upstream gas pressure $P_{\text{upstream}}$: plugs F1-1, F1-2 and F1-3.

## VI.4 Swelling and GBP of bentonite/sand plug+grooved argillite tube

### VI.4.1 Water injection test of plugs F2-1, F2-2

Plugs F2-1 and F2-2 were used to determine gas migration pathways, i.e. through the argillite or bentonite/sand-argillite interface. As presented in Figure VI.12, similar to previous tests, water permeability decreased rapidly at first then began to stabilize. When comparing with tests F1-1 and F1-2, it was noted that less time was needed for the $K_w$ to become stable, e.g. 56 h-F2-1(138 h-F2-2) vs. 150 h-F1-1(186 h-F1-2). In addition, the stable water permeability $K_{w,\text{stable}}$ were a little higher than the correspond values of plugs F1-1 and F1-2, e.g. $2.88 \times 10^{-20}$ m$^2$-F2-1 (3.88 $\times 10^{-20}$ m$^2$-F2-2) vs. $0.68 \times 10^{-20}$ m$^2$-F1-1(0.94 $\times 10^{-20}$ m$^2$-F2-2). Theoretically, the stable water permeability of plugs F2-1 (F2-2) should be similar with plugs F1-1 (F1-2), because the difference between them is just the inner surface of the tube (smooth or grooved). In fact, after the occurrence of hydraulic cut-off, the seepage path is bentonite/sand plug or argillite tube. While the bentonite/sand plug are always the same for the different series, the only reason for these difference is due to the property of the argillite tube. In order to machine the grooves in the inner surface, the tube would be placed on the machine tool to cut. Perhaps some micro-fissures were produced during this process, and these micro-fissures would lead to higher water permeability.
VI.4.2 Gas breakthrough tests of plugs F2-1, F2-2

Gas breakthrough tests of plugs F2-1 and F2-2 were initiated after swelling tests to compare with the results of previous tests (with smooth inner surface). It was quite interesting to note that the discontinuous breakthrough pressures were comparable to plugs F1-1 and F1-2, see Figure VI.13, e.g. 1.98 MPa-F2-1 (3.01 MPa-F2-2) vs. 1.54 MPa-F1-1(3.06 MPa-F1-2), while the continuous breakthrough pressures were similar (8.00 MPa-F2-2) or smaller (5.01 MPa-F2-1) than the corresponding values of plugs F1-1 and F1-2. Besides, for argillite alone, continuous gas breakthrough pressure was measured at value ranging from 0.2 MPa and up to several MPa (depending on argillite orientation); it has been recorded up to 6MPa for undisturbed argillite (Davy et al., 2012, Skoczylas and Davy, 2011). It means that the $P_{\text{con}}$ of argillite is similar or smaller than the $P_{\text{con}}$ of the plug-tube interface, while their values are much smaller than the $P_{\text{con}}$ of bentonite/sand plug, see Figure VI.14. Therefore when all materials become fully saturated, the most likely gas migration pathway is through the argillite (host rock), then is through the interface and the last possibility is through the bentonite/sand plug.
After resaturation, gas breakthrough tests were implemented again. As shown in Figure VI.13, both discontinuous breakthrough pressure and continuous breakthrough pressures were lower the corresponding values of the first time. The same phenomena, already found for previous test (F1-2), were confirmed: the sealing ability ($P_{\text{con}}$ and $P_{\text{dis}}$) of bentonite/sand-argillite assembly couldn’t be recovered after resaturation, while the sealing ability of bentonite/sand plug could be obtained again after resaturation.
For all tests (except for plug F2-1-resaturation), it can be discovered that the values of $Q_g$ are close to or higher than 0.001 MPa/h when continuous gas breakthrough occurs, see Figure VI.15. Therefore it is possible for us to use this value to help us to judge the phenomenon of gas passage, e.g. discontinuous or continuous. Besides, gas permeability was measured after continuous gas breakthrough, see Figure VI.16. It was observed that for all plugs, the order of magnitude of permeability was about $10^{-20} \sim 10^{-21}$ m$^2$. These values are extremely low, which mean it is very difficult for gas to pass through the bentonite/sand–argillite assembly. However, we are not sure that the measured values are the $K_g$ of argillite or other materials, because both argillite and the plug-interface are two possible gas migration pathways. Another phenomena could be found that the value of $K_g$ (after resaturation) were higher than the corresponding values of the 1$^{st}$ cycle. It was consistent with the results of gas breakthrough tests, i.e. the $P_{\text{con}}$ of the 2$^{nd}$ (after resaturation) was higher than the corresponding values of the 1$^{st}$ GBT. Therefore, this phenomenon can also regarded as an indicated proof that gas migration pathway is through the argillite not the interface and the measured gas permeability is the $k_g$ of argillite. It was checked that the Klinkenberg effect was not obvious for this material in the range of pressures applied.

![Figure VI.15 Summary of the rate of increase of downstream gas pressure ($Q_g$).](image-url)
V. 5 Conclusion

In this part, we presented the results of swelling tests and gas breakthrough tests under different boundary conditions from those of the Chapter V (only smooth Plexiglas-aluminum tube). It was then found that when the bentonite/sand plug is saturated with water in a smooth tube, the breakthrough pressure through the tube-plug assembly is at least equal to the effective swelling pressure of the material.

To attempt to remove the effects of interface, we firstly put a sample in the Viton™ jacket directly and then apply confining pressure on the jacket. Confining pressure will therefore play a role on the swelling pressure for the saturated material. It has never been possible to obtain the breakthrough pressure at a value lower than the imposed confining pressure. We can therefore conclude that the swelling pressure will always be a lower bound for the breakthrough pressure.

In a second case, the plug swells with a grooved tube to increase the roughness of contact surface and interface, and therefore it is difficult for the gas to create a pathway. The continuous breakthrough pressure was not obtained even at 10MPa gas injection. The only variation compared to other tests is the nature of the interface; therefore we can logically deduce that the gas passes through the interface at the moment of breakthrough in the presence of the smooth tube.
Finally, we perform some tests with an argillite-bentonite/sand plug assembly which is more close to in situ situation. Our results showed that the fully saturated water permeabilities of compacted bentonite/sand and argillite were similar: in the order of magnitude of $10^{-20}$-$10^{-21}$ m$^2$. Gas breakthrough tests revealed that continuous gas breakthrough pressure of bentonite/sand plugs was higher than 10.0~10.9 MPa. Due to the limits of our experimental device, we didn’t obtain continuous gas breakthrough for bentonite/sand plug when swollen inside a grooved tube. For argillite, this value was lower, with values consistently smaller than 5.0~8.0 MPa. For plug-tube interface (smooth inner surface), continuous gas flow was detected when gas pressure was similar to or higher than the effective swelling pressure (between 7-8MPa). This means that the interface and the argillite (host rock) are two preferential pathways for gas migration, and the possibility of gas passage through argillite is more likely when all clayey materials become fully water saturated.
General Conclusion

This research contributes to a better understanding of 1) the sealing ability of partially saturated bentonite-sand plug under variable confinement, 2) the kinetics and swelling pressure of plugs in the presence of gas pressure (at least 4 MPa) and in contact with water (contact direct or via a fully saturated plug) as well as their gas breakthrough pressure, and 3) the sealing efficiency of argillite-bentonite interface, obtained by accurately measuring swelling pressure of bentonite/sand mixture and the discontinuous/continuous gas breakthrough pressure, when this interface is subjected to a non-negligible gas pressure.

Previously, we conducted water retention tests, with different boundary conditions: free swelling conditions and constant volume conditions. Our work shows that the swelling speeds of the sample under free swelling conditions are faster than for the samples which swell under constant volume conditions. In addition, more water is absorbed under free swelling conditions (compared to constant volume conditions). We also found that at RH = 98%, the swelling is higher with an increase of 22.5% of the initial volume of the plug.

To evaluate the sealing ability of partially saturated bentonite/sand plugs with, we measured their gas permeability under variable confining pressure (up to 12MPa). We find that the porous structure accessible to gas and transport of gas are very sensitive to successive drying / imbibitions cycles, coupled with cycles of confinement / de-confinement. Besides, tightness to gas (supposedly obtained when the gas permeability is less than $10^{-20}\text{m}^2$) is obtained under 9MPa confinement and for the samples initially saturated at 86-91% only. After stabilization of the mass at a given RH under conditions of free swelling and given confinement, the results of gas permeability highlight two antagonistic effects. Firstly, the swelling is accompanied by an increase of the total volume and the pore volume accessible to the gas, so as to contribute to increased gas permeability. Moreover, at given sample volume, the increase in water saturation partially fills the volume of pores, so that the gas permeability decreases. These antagonistic effects already described in the literature, are observed simultaneously in the domain of intermediate saturations (50-60%): they allow to interpret satisfactorily our experiments of gas permeability in partially water-saturated conditions, for which an increase of the water saturation may lead to an augmentation of the gas permeability, so the effect of the swelling (and therefore an increase in the pore volume available to gas) is predominate.
In a third part, an experimental campaign is performed to determine the effect of gas pressure (4, 6 or 8 MPa) on the swelling capacity of bentonite-sand plug, as well as its gas breakthrough pressure (discontinuous and continuous). In parallel with the presence of water (favorable for swelling), the presence of 4 MPa gas pressure slightly limits the effective swelling pressure, but significantly affects the gas breakthrough pressure. For a gas pressure of 8 MPa, equal to the double of water pressure, a very significant decrease of the effective swelling pressure of is measured, and the gas passage occurs systematically, regardless of the pressure employed.

At last, a tube with a grooved inner surface is used to determine whether gas passes through the interface or through the water-saturated plug: our tests show that gas transfers preferentially via the interface. When the tube is smooth, gas breakthrough pressure is similar or slightly higher than the actual swelling pressure for the tests of A1 ~ A3, otherwise (rough interface), gas passes at a much higher pressure (beyond 10MPa). Our tests also showed that the absence of scale effect on the swelling pressure and breakthrough pressure, using a sample A4 twice longer than the others. For argillite, this value was lower, with values consistently smaller than 5.0~8.0 MPa. For plug-tube interface (smooth inner surface), continuous gas flow was detected when gas pressure was similar to or higher than the effective swelling pressure (between 7-8MPa). This means that the interface and the argillite (host rock) are two preferential pathways for gas migration, and the possibility of gas passage through argillite is more likely when all clayey materials become fully water saturated.
Reference:


Annex 1

(a) Volume changes of bentonite/sand plugs for test series S2

Figure A.1 volume change of samples S2-3, S2-4, S2-5 and S2-8 of test series 2, showing the limited changes due to the initial gas permeability testing phase (up to 5MPa confinement).
Annex 2

(b) Volume changes of bentonite/sand plugs for test series S3

[Graphs showing volume changes for test series S3]
Figure A.2  volume change of samples S3-9, S3-10, S3-11 , S3-12 and S3-13 of test series 3.