Experimental results using aluminium-PlexiglassTM tubes (bentonite/argillite interface)

18 Months report

FORGE Report D3.07

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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 proposal for an integrated, multi-disciplinary project. The FORGE proposal is for 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.

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WP3 FORGE PROJECT – WP3.2.8 and WP3.3.4

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1. Introduction

Our research team, at LML Laboratory, is involved in two WP3 tasks: WP3.2.8 entitled "effect of gas pressure on saturation and swelling behaviour of bentonite" and WP3.3.4 entitled "gas migration through an argillite-bentonite interface". The WP3.2.8 program is a part of the PGZ experiment, which has been conducted by ANDRA at Bure for almost one year now. This *in situ* experiment is aimed at estimating the influence of gas pressure upon the swelling process of bentonite, when it is being water (re-)saturated. To help modelling such phenomenon, laboratory experiments are conducted under boundary conditions, which are better known and controlled than *in situ*. Experimental data are also to be obtained under homogeneous conditions carried out at a smaller scale than the *in situ* one. Our experiments have required the design of new set-ups: triaxial confinement cell, gas pressure control panel, etc… Even if performed on small samples, these experiments are time consuming. However, it has been possible to obtain several important results, which are presented further in this report. Complementary tests are currently conducted in our lab in order to complete and validate data already acquired.

The second part of our work in the FORGE project, as WP3.3.4, is related to gas migration through the argillite-bentonite interface. The main question about gas transfer in a storage structure, in which many kinds of interfaces (bentonite-argillite, bentonite-concrete, argillite-concrete, etc) are present, can be summed up as: is any kind of interface a privileged candidate for gas flow or gas entry? This is a crucial issue in terms of waste storage safety. Moreover, answering this question will help understand how gas pressure, mainly H_2 produced by corrosion, could dissipate through the whole storage structure and the host rock, without gas hydraulic fracturing (of a part of the structure). In this WP3.3.4, our experimental study mainly focuses on the bentonite-argillite interface and its "gas flow" properties. It is clear that these interface properties will depend on the swelling pressure (or contact pressure here) of bentonite, which value is controlled, firstly, by the bentonite dry mass density and its water saturation level, argillite acting mainly as the host rock. However, if a gas pressure is applied to bentonite, coupling effects will occur (i.e. variation in pore pressure), leading to changes in contact pressure upon argillite. Such a phenomenon has to be taken into account and will be the purpose of an important part of our study. To evaluate whether gas will be flowing through the interface, or through bulk argillite or bentonite, the gas entry pressures of argillite and bentonite alone have to be measured under realistic state of stresses: in our case under a mean *in situ* hydrostatic stress of 12MPa is chosen for argillite. For bentonite, the stress state will be induced by swelling, which will therefore be the *in situ* swelling pressure (as this is related to bentonite compaction process and water saturation, both conducted similarly as what is done *in situ*). The measurements will be performed on bentonite which has swelled into a calibrated Plexiglass™-aluminium tube. These tubes have been especially designed for the FORGE project. Only a few results concerning this second part are currently available, but most of the design and calibration operations are now completed.

2. The Plexiglass™-aluminium tubes and their calibration

We have chosen to let bentonite swell inside a Plexiglass™-aluminium tube for several reasons, which are detailed below. The first one is that it enables to control the bentonite sample preparation process used to target a swelling pressure, after *in* *situ* water saturation, and as expected in the storage structure. This pressure is around 7MPa for a mixture of silicate sand and bentonite. The mass sand proportion is 30% and the mixture is supplied to our lab by the CEA agency. Bentonite is a classical MX80 type. The required swelling pressure should be obtained for a mixture compacted to get a dry density of 1.77 with a water content of 15.2%. To obtain the adequate water content, the bentonite-sand mixture is placed in an atmosphere at controlled relative humidity (of 85%) and then compacted at about 12MPa axial pressure in a steel tube. The resulting sample is 25mm height and 42.5mm diameter (picture 1).

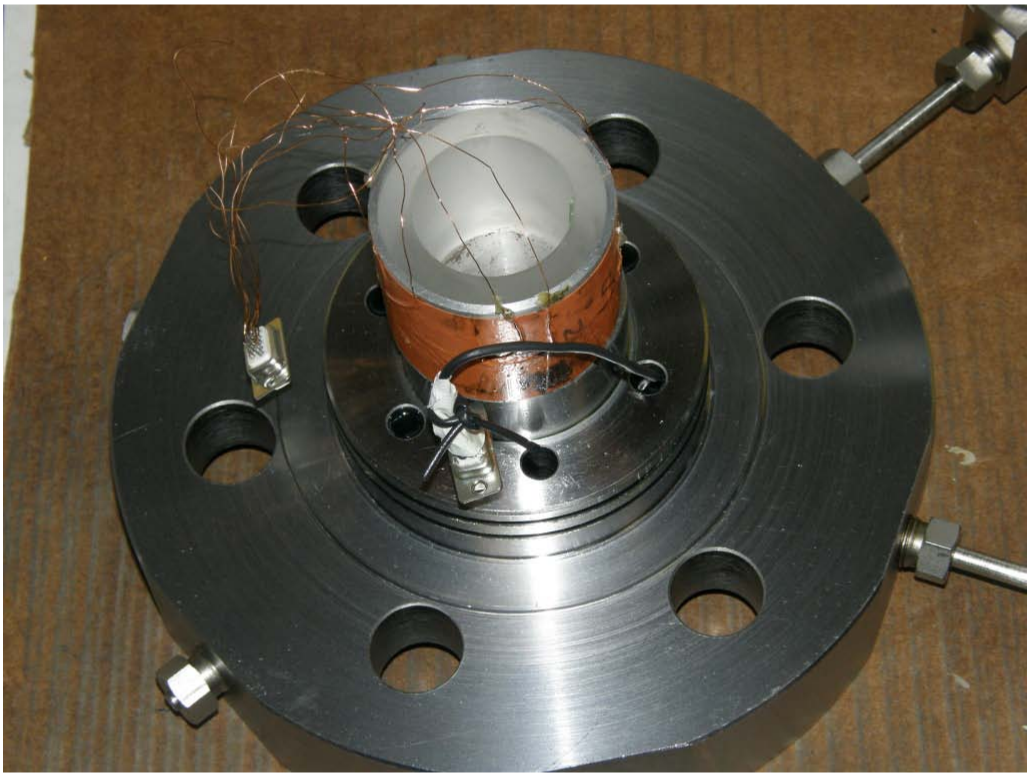
Picture 1: Bentonite plugs obtained after the compaction process

Dry density and water content are simply measured after compaction, by weighing one sample after complete drying in an oven at 65°C. In order to check that the target swelling pressure is obtained after complete water saturation and under oedometric conditions, the bentonite plug is placed inside a Plexiglass™-aluminium tube instrumented on its outer surface with strain gauges (picture 2).

Picture 2: Calibrated Plexiglass™-aluminium tubes. They are used to verify the swelling pressure

Each Plexiglass™-aluminium tube has been calibrated in a triaxial cell especially designed and manufactured for the FORGE project (in total, two cells have been manufactured for this project). The tube is placed in the triaxial cell and wrapped inside a Viton™ jacket. Confining oil is used to impose an hydrostatic stress to the tube of up to 12MPa. Therefore, gas is injected in the tube at different pressures P_i which simulate sample swelling. The 4 gauges record the lateral deformation of the tube in order to relate strains to gas pressure, and therefore, to the forthcoming swelling pressure due to bentonite (pictures 3-4 and figures 1 and 2). Very good linearity and reversibility were obtained for each tube.

Two types of Plexiglass™-aluminium tube have been manufactured, of either 50mm or 25mm height, with both an internal diameter of 42,5 mm. The smaller ones are used for the PGZ experiment (see next section). The higher ones will be used for poro-mechanical tests and some flow experiments through Plexiglass™-bentonite interfaces.



Picture 3: Plexiglass™-aluminium tube in a triaxial cell – first mounting step

Picture 4: The tube is wrapped inside the Viton™ membrane

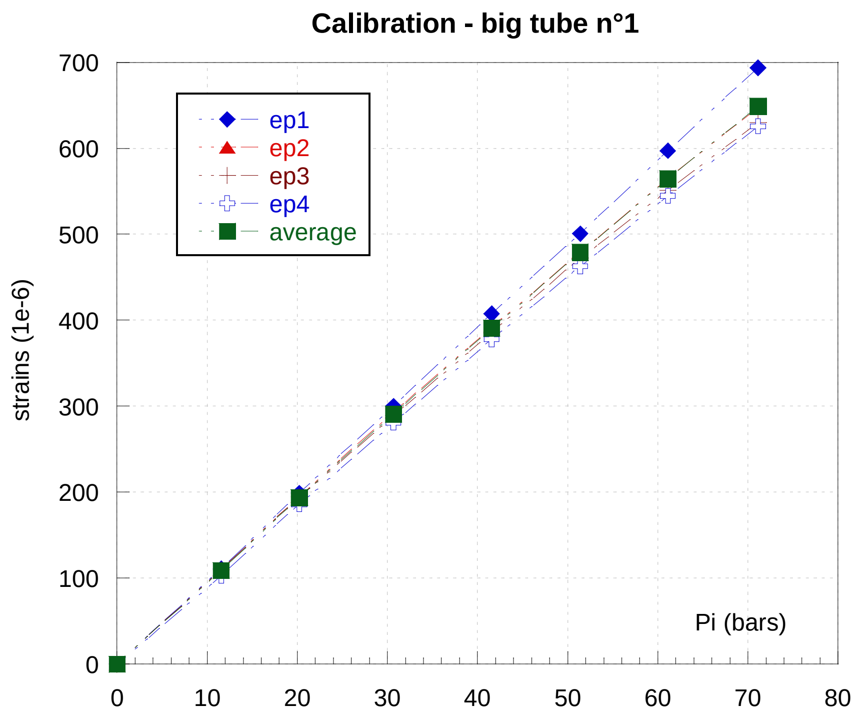


Figure 1: Example of tube calibration. The average value is used to relate the tube lateral strains to its internal pressure

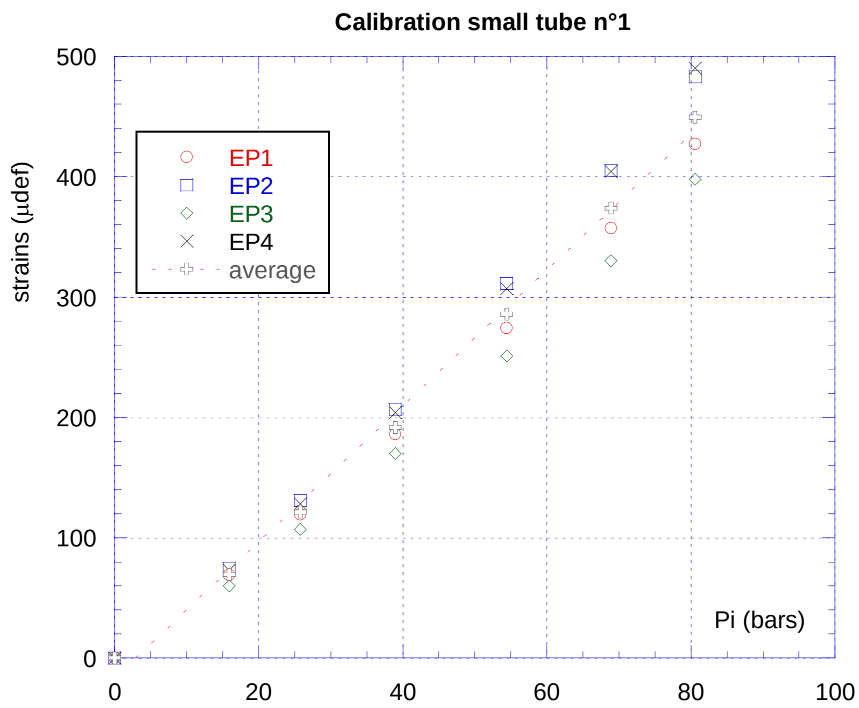


Figure 2: Example of small tube calibration. The average value is used to relate the strains to the tube internal pressure

Correction of thermal perturbations (thermal effect)

It must be mentioned here that every test performed for this study lasts quite a long time (at least several weeks) and, even if it is carried out in an air-conditioned room, small temperature variations in temperature may occur. This is sufficient to slightly deviate the strain results. As a consequence, a reference tube is used, which is not placed inside the triaxial cell but in the room beside it, in order to correct for thermal strains. Figure 3, below, shows an example of such a correction, which is now done for all tests.

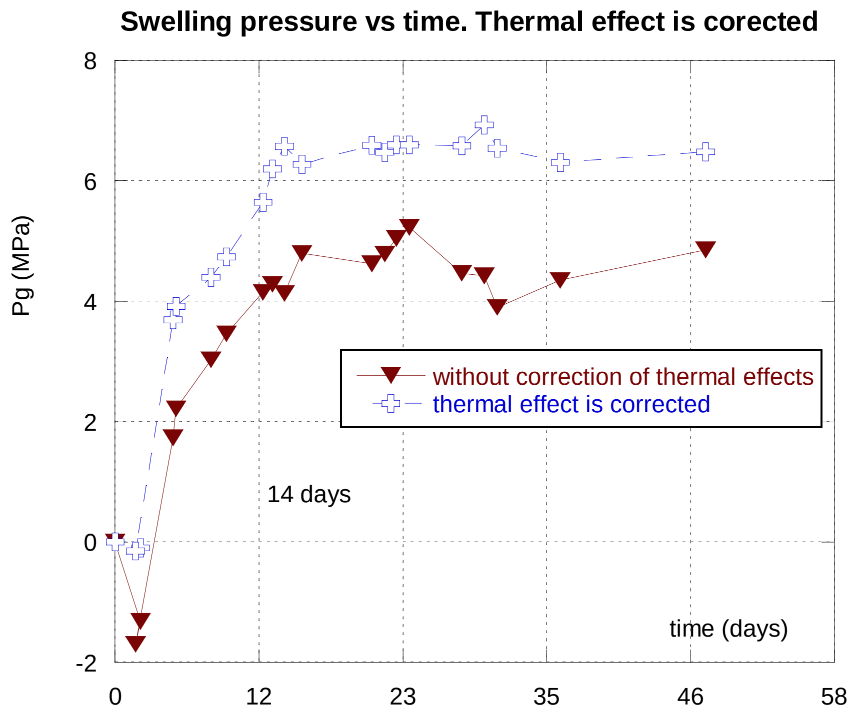


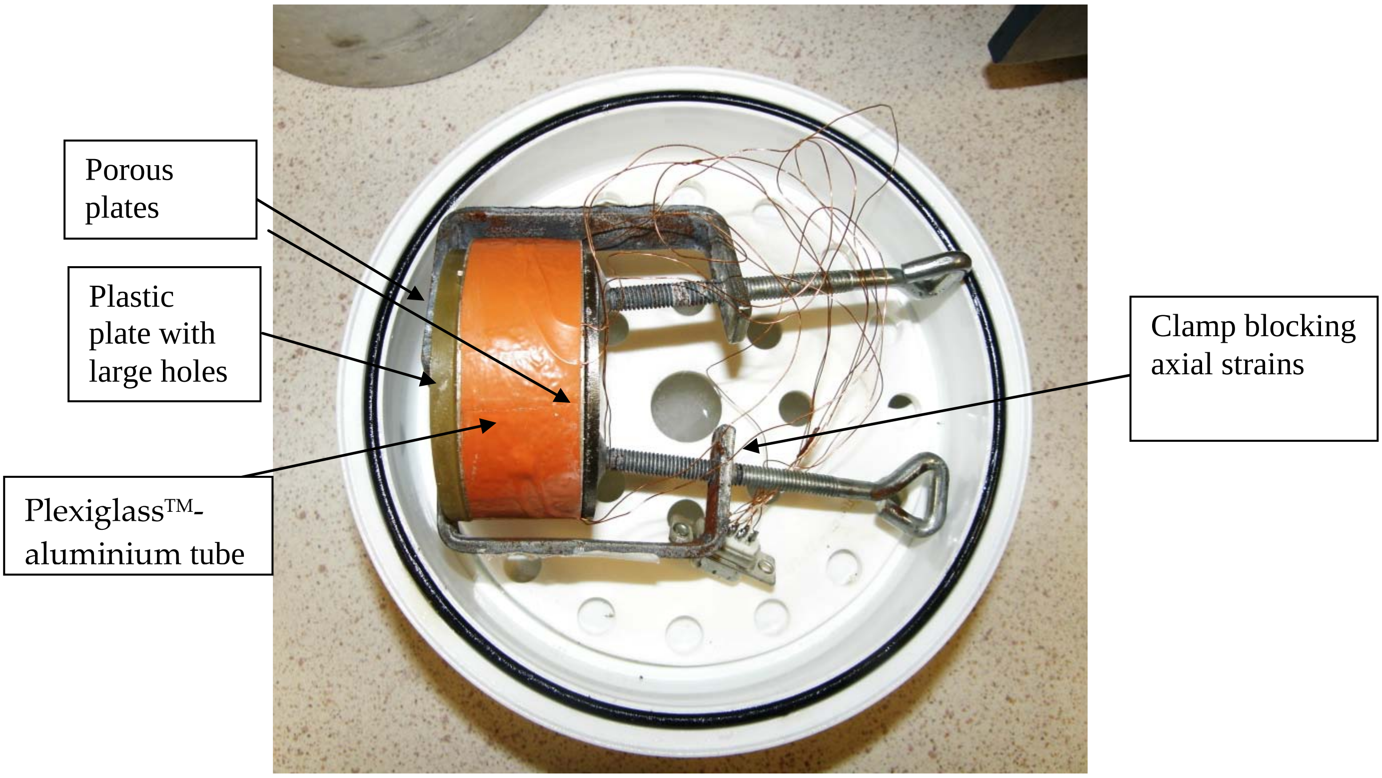
Figure 3 : Example of measurement corrections made in order to take into account temperature variations during the triaxial test

3. Swelling of compacted bentonite under oedometric conditions and variable relative humidity

This kind of test (and its results) are needed for the numerical simulation involved in the PGZ program, as follows. In the *in situ* experiments, bentonite swelling occurs in a fixed volume (i.e. the gallery volume), while axial strains are blocked. As a consequence, bentonite swelling is assumed to occur under oedometric conditions. Hence, in order to model the *in situ* material swelling, it is necessary to identify the retention curve (i.e. the relation between capillary pressure P_c and water saturation S_w) for a fixed volume of compacted (sand+bentonite) plug. This is the purpose of the experiments presented below.

Testing the (sand+bentonite) compacted mix under given relative humidity

We chose to let the (sand+bentonite) plug swell in a Plexiglass™-aluminium tube (as presented in previous section) while the axial strains are blocked by the use of two porous plates. The porous plates are made of stainless steel (picture 5). In fact, two tests have been performed already: two steel plates were used for the first one but the test lasted a very long time. Instead, it was decided to use a plastic porous plate with large holes to accelerate the saturation kinetics for the second test.



Picture 5: Experimental device used to make the (sand+bentonite) plug swell with (almost) no volumetric change

In a first attempt, the Plexiglass™-aluminium tube was instrumented with strain gauges, as we intended to measure the swelling pressure during bentonite saturation, but this was unsuccessful (erratic measurements). We have also tried to measure the sample gas permeability at various saturation steps, but gas flows through the tube-bentonite interface, giving a very high permeability with no physical meaning.

For both tests, the experimental methodology was as follows:

- Sand+bentonite sample compaction is performed twice, as described in previous section
- Weighing of both samples and of the parts of the experimental set-up (tube, plates, etc…)
- First equilibrium at 70% rh. This value was chosen because, at lower rh values, mass loss occurs (instead of mass invariance or increase)
- As the mass increase is not comparable for the two samples placed at rh=70%, further results are compared using the equilibrium at 70% as the initial state, and not the sample state after compaction
- Equilibrium at rh=75%, then 85, 92 and 98 %, followed by full water saturation using water injection

Nota: both sample behaviours are clearly not the same at rh=70%; 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.

Results

Figures 4 and 5 show the complete results for samples 1 and 2.

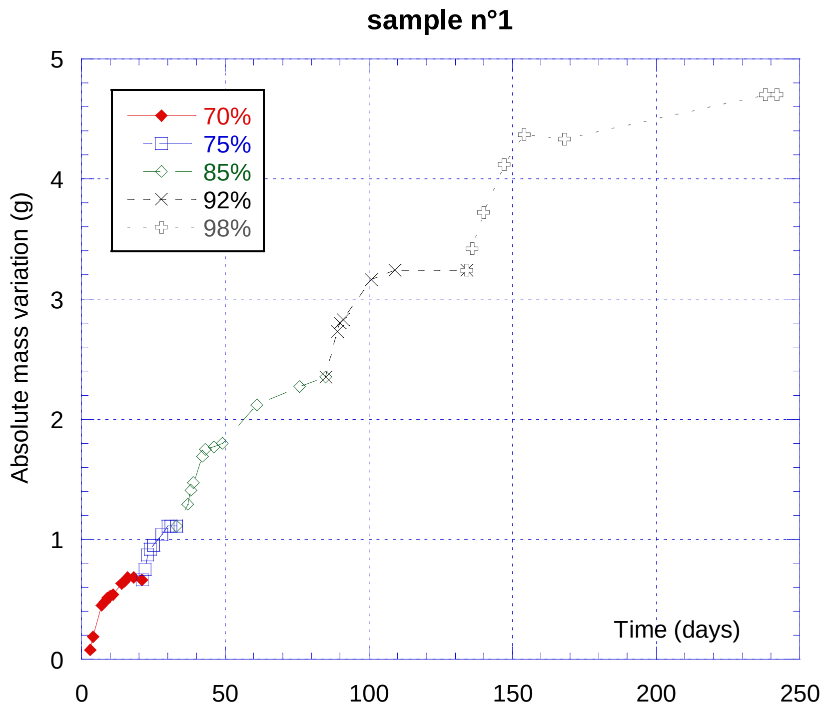


Figure 4: Mass variation of sample 1

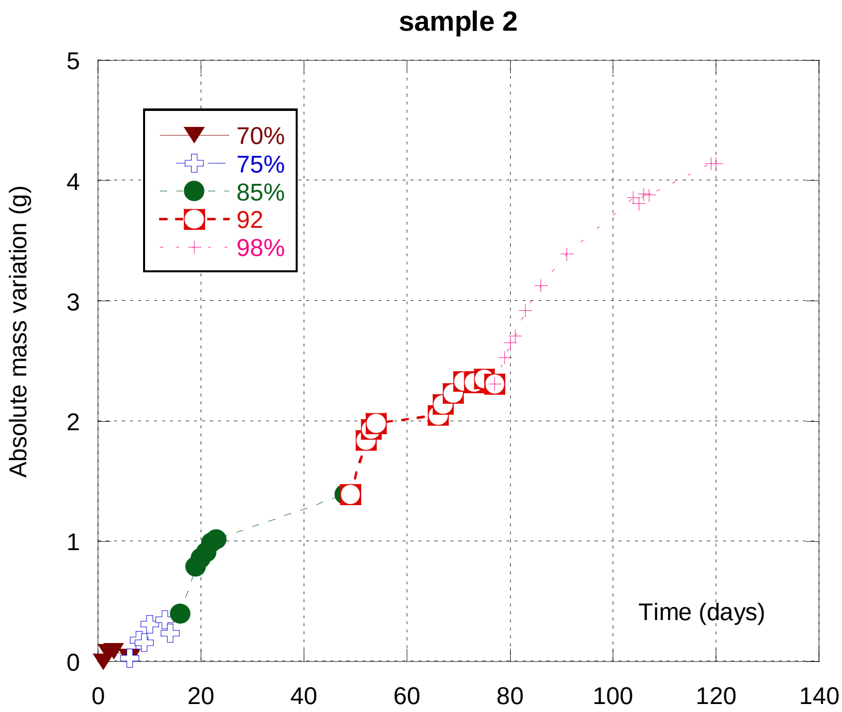


Figure 5: Mass variation of sample 2

We can see in figure 5 that sample 2 mass is not currently stabilized. This means that this test is not completed. Nevertheless, we compared results for both samples, see figures 6, 7 and 8.

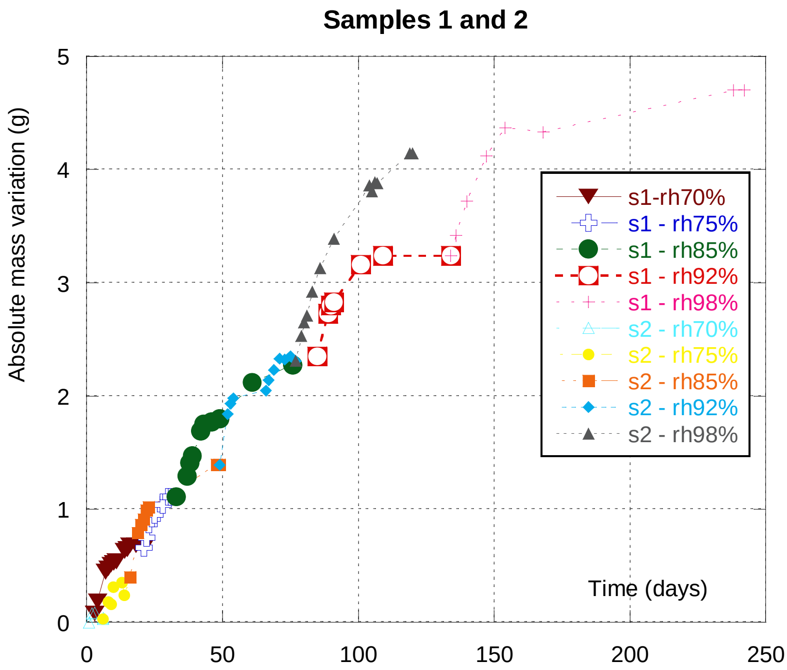


Figure 6: Comparison of absolute mass variation for sample 1 (s1) and sample 2 (s2)

Figure 7 and 8 are certainly the most useful to compare samples as they are drawn using relative mass variations. As mentioned before, samples behave differently at the first relative humidity step of 70%, with virtually no significant mass variation for sample 2 while sample 1 has gained more than 0.5% mass. This single aspect makes comparison more difficult. Hence, it was chosen to compare results from the stabilization step at 70% rh, i.e. figure 8.

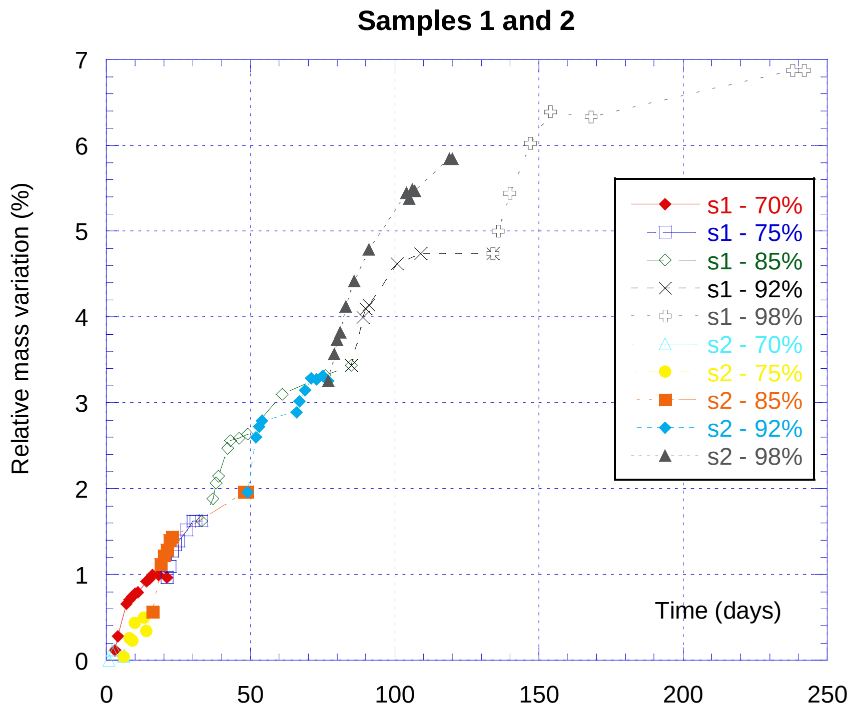


Figure 7: Relative mass variations – Samples 1 and 2

Figure 8 shows that for both samples, the mass increases at each humidity step are different. This is interpreted as the fact that the porous structures of both materials are not identical even if they have been submitted to the same preparation technique. In fact, slight changes in the waiting time before compaction (when bentonite powder matures at 85%rh) or in the compaction process itself can lead to small changes in the pore radii distributions. On another hand, the total mass variation is close to 6% for both plugs. Yet, observation of figure 8 allows us to assess that there are more large radius pores in sample 2 than in sample 1, because the increase in mass at rh98% is roughly 3% for s2 and only 2% for s1. The saturation kinetics are also very contrasted but this is supposed to be solely due to the plastic plate (for sample 2) which has replaced the steel plates (for sample 1). This allows faster humidity transfer towards the sample.

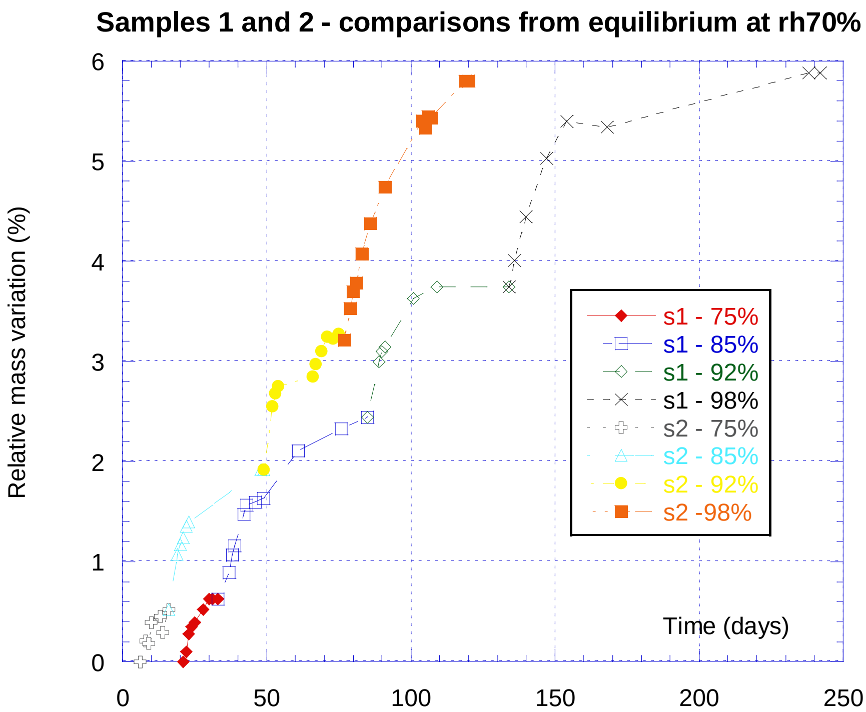


Figure 8: Relative mass variations plotted by starting from the equilibrium at 70%rh - Samples 1 and 2

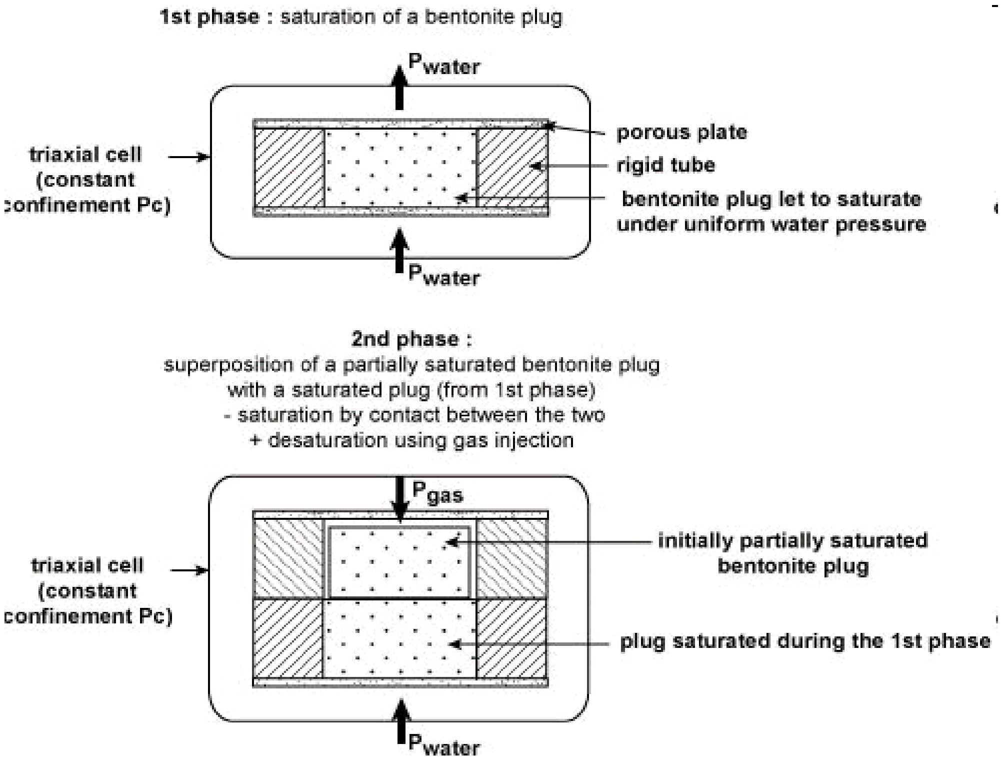
4. Swelling of compacted (sand+bentonite) under free volumetric changes and variable relative humidity

We are currently performing tests corresponding to next section. The comparison of results under free volumetric changes, with previous tests under oedometric conditions, will be done in the next report.

5. Swelling of compacted bentonite (into a Plexiglass™-aluminium tube) submitted to gas pressure – laboratory test design

One of the main *in situ* issue is to understand the swelling process of bentonite when it is submitted to gas pressure. This pressure would be due to hydrogen, mainly supplied by corrosion and water radiolysis. The analysis of this particular problem is the main purpose of the PGZ program, which is both experimental and numerical.

The numerical part needs many material properties which are not, by far, all available (relative gas and water permeability, suction curves, etc…). The actual *in situ* boundary conditions are not accurately known either. It is therefore useful to conduct laboratory experiments for which the boundary conditions are known accurately, in order to calibrate the numerical modelling or to use inverse methods so as to provide the properties mentioned above. In such context, we have designed an original and dedicated laboratory test with two objectives: 1) enabling numerical calculations and 2) investigating at the macroscopic scale (i.e. the REV scale) whether gas pressure influences the swelling pressure of a sand+bentonite plug, and changes (or not) its resulting characteristics after full water saturation, using a controlled gas pressure. Figure 9, below, presents a schematic diagram of the test we have designed for this part of the FORGE experimental study.



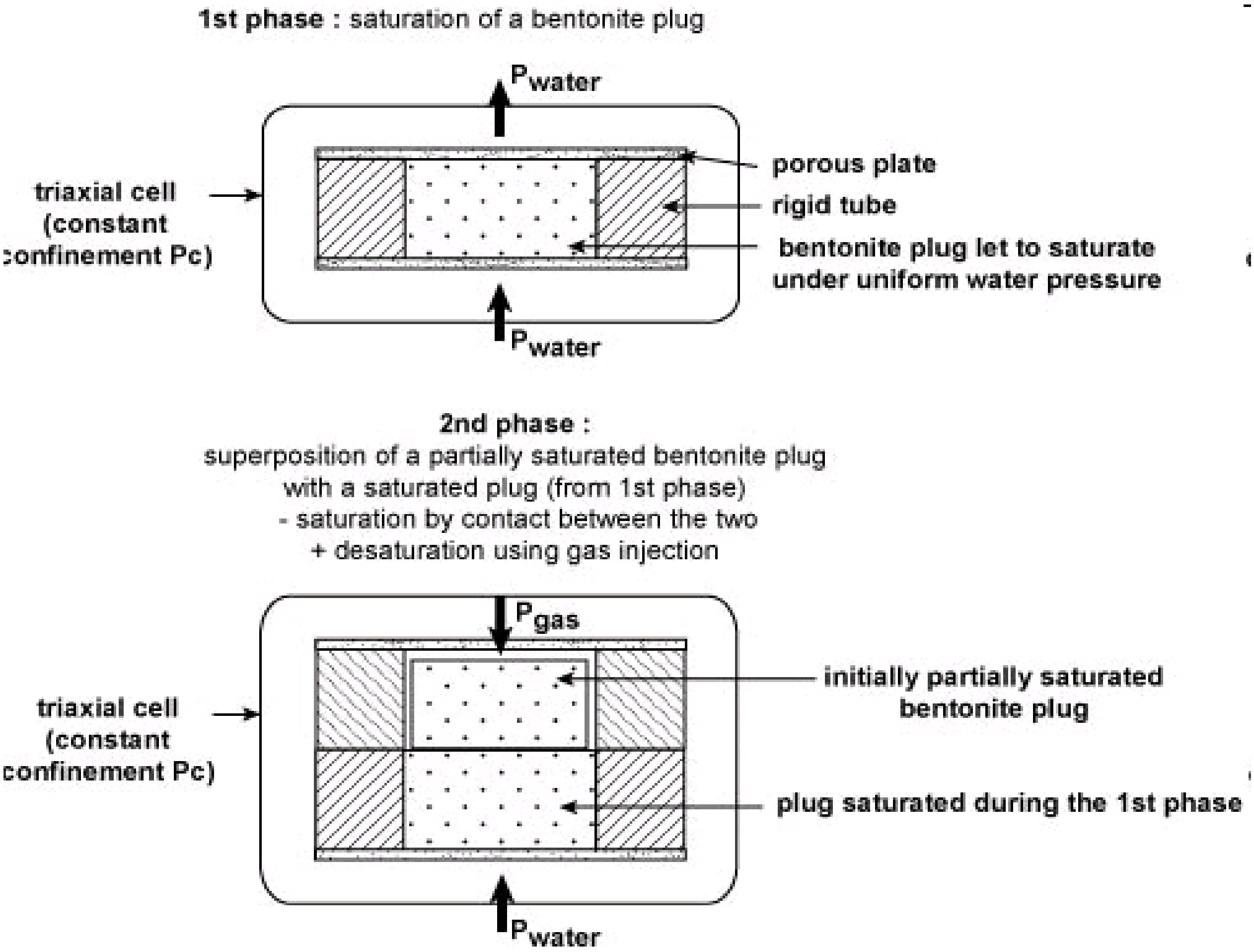


Figure 9: Schematic diagram of the "PGZ" laboratory experimental device

It can be seen on figure 8 that two different small Plexiglass™-aluminium tubes are used during the 2nd test phase (see also picture 6).

Picture 6 : Small tube used for the swelling experiment under gas pressure

The first tube is for the 1st phase of the test: it consists in water-saturating a sand+bentonite plug with *in situ*-like water. The second phase begins when this plug is fully saturated, and a second tube is placed just over the first one. Inside this tube is laid a (sand+bentonite) plug, which is in its initial partially-saturated state (i.e. just after compaction); this second plug is supplied with water by the first one placed below it. This is intended to be realistic toward a possible in-situ case. As presented in figure 9, gas pressure is applied at the top of the assembly. We have selected three possible cases: P_{gas}=0 (reference case), P_{gas}=4 MPa and P_{gas}=8MPa (which is the maximum value studied). For all tests, water pressure is the *in situ* average value i.e. 4MPa. The tube placed at the top, and submitted to gas pressure, is instrumented with strain gauges in order to follow the plug swelling pressure with time and to evaluate the change in the swelling process kinetics.

6. Swelling of compacted bentonite (into a Plexiglass™-aluminium tube) submitted to gas pressure – results

a. First series of tests

This first series can also be considered as feasibility tests as it was the first time that such an experiment was conducted in our laboratory. Figure 10 below shows the comparison of swelling pressure evolutions with 4MPa gas pressure or without it.

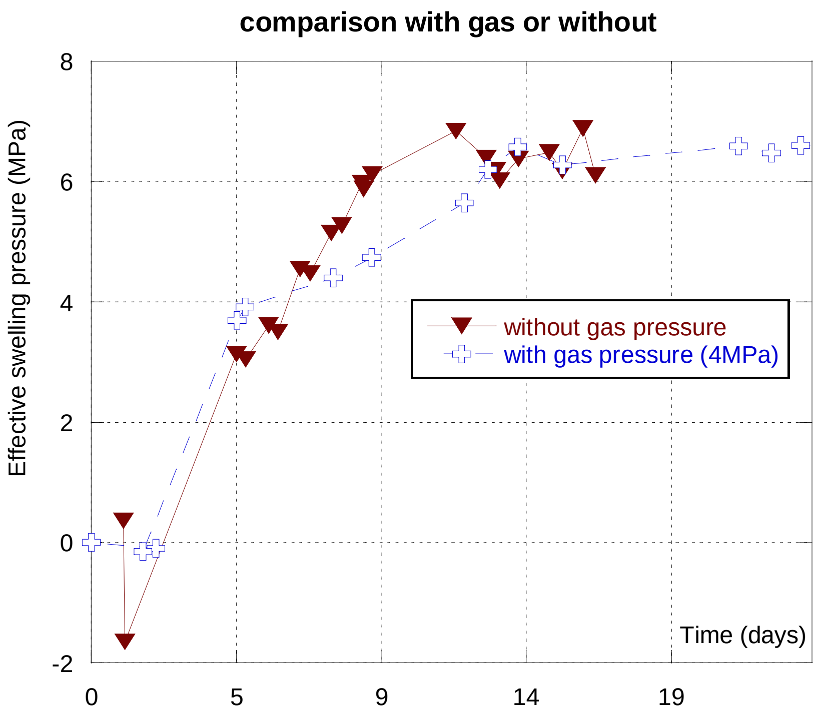


Figure 10 : Effective swelling pressures. Comparison with or without gas pressure. Gas pressure is 4MPa when applied.

The first and main observation which can be drawn from these results is that there is virtually no significant difference when gas is present or not (at least at this 4MPa pressure). The swelling kinetics seem to be almost identical at the beginning of the process but, when gas pressure is applied, it slows down from 4MPa pressure. This particular phenomenon needs to be confirmed as we are not sure that it is not an experimental artefact. Without gas pressure, the maximum swelling pressure is reached after 10 days injection, then it stabilizes. On the opposite, with gas pressure, 4 to 5 more days are necessary to reach the same effective value of swelling pressure. Gas entry pressure (GEP) experiments and permeability tests were carried out on both samples. For the sample swelled without gas, GEP was found to be between 6 and 7 MPa, and for the second one (swelled under gas pressure), GEP was detected from 4.4 MPa. For both samples, gas permeability was almost zero. One concludes that, at a pressure level of 4MPa, gas does not seem to strongly modify the swelling process, except that it induces a lower imbibition rate and a slightly lower gas entry pressure. The adverb "slightly" is used here, because, due to the very limited number of tests performed at present, we do not strongly rely on these first results, which have to be confirmed.

A third plug was then tested with a gas pressure of 8MPa. We were 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. Indeed, the injection water pressure is fixed at 4MPa but, when gas pressure is applied, the pore fluid pressure, being either gas (top plug) or liquid (bottom plug), is also increased, as water pressure measurement shows. This means that there is a coupling effect due to gas pressure. At this step, a choice had to be made, which was either letting gas pressure control the water injection pressure or controlling the water pressure by draining the bottom sample in order to maintain its pore water fluid at 4MPa. The first option was chosen because we supposed that the same kind of phenomenon would have taken place *in situ*. As a result and for the whole test, the actual water injection pressure was 7.2MPa, which is a stable value. Figure 11 gives the evolution of swelling pressure with time. As compared to previous results, one observes a huge decrease in swelling kinetics: under 8MPa gas pressure, 25 days are necessary to achieve full saturation (i.e. swelling stabilization). Moreover, the effective swelling pressure is now 4 MPa. This means that the top bentonite sample is not fully saturated because, otherwise, swelling pressure would have reached the target (between 6 and 7 MPa). At this stage, either we dismount the sample or we test it again with another gas pressure. This second option was chosen with a 6MPa gas pressure. The idea was to observe if swelling was going to increase and what would be the actual swelling pressure. Figure 12 shows our results. It is observed that an immediate new swelling process takes place and stabilizes within 6-7 days. The resulting effective pressure is now 7.6MPa which is higher than the target value.

Nota: effective swelling pressure is consistently obtained after water pressure is off, i.e. when pore pressure is zero.

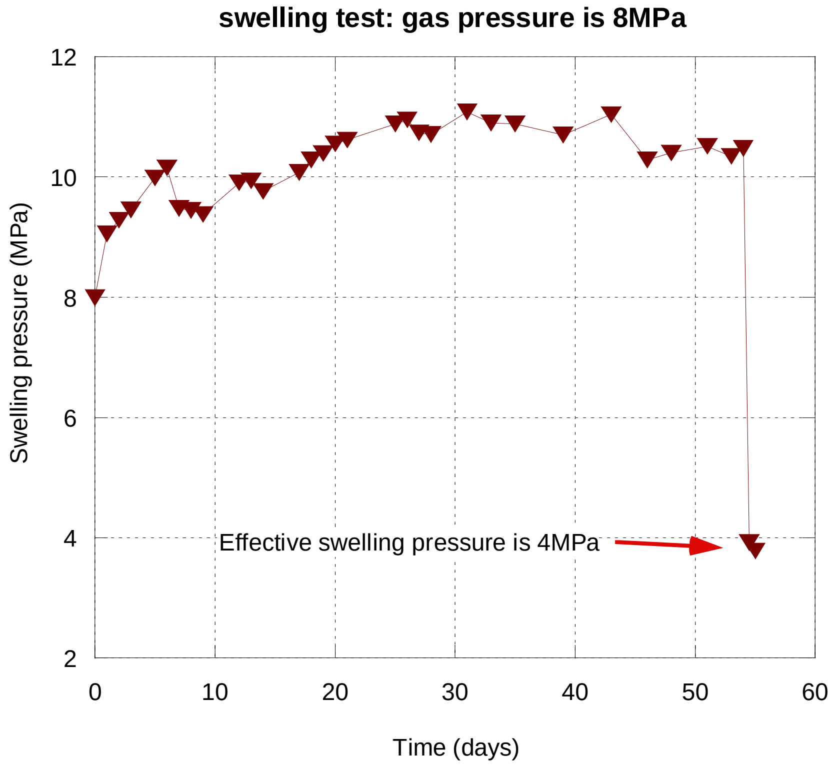


Figure 11 : Swelling pressure evolution with 8MPa gas pressure

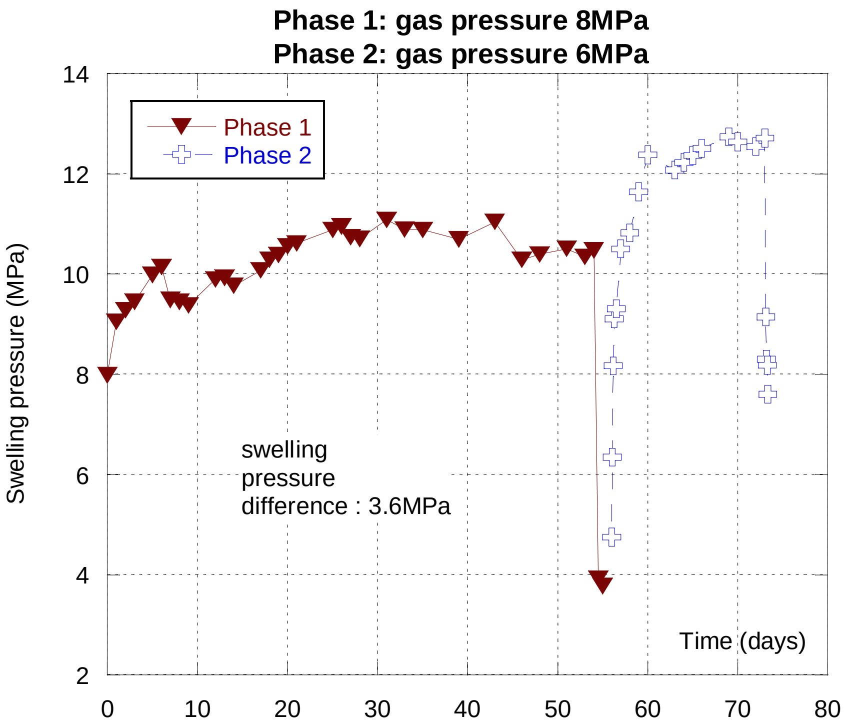


Figure 12 : Swelling pressure with two successive injection gas phases

After the swelling step at 6MPa gas pressure, the top sample was dismounted and submitted to gas entry pressure (GEP) experiment. It has been quite difficult to measure it accurately (as the expected range for GEP is unknown for a first test), but we have assessed that gas has gone through the sample at a pressure lower or equal to 2.5MPa. Compared to GEP values measured after a swelling with no gas pressure (6-7MPa) or with 4MPa gas pressure (GEP=4.4MPa), this new lower value of 2.5MPa is evidence that the porous structure is different, in a sense that there must now be bigger pore radii in the porous structure (which let gas pass more easily), although the effective swelling pressure is slightly higher than the one measured in previous experiments. We have also measured here that the sample gas permeability remains almost zero, even if some gas has passed through the material during the GEP experiment. All these observations need to be confirmed by the second series of tests which are currently carried out.

b. Second series of tests

This new series is being performed at present. It will allow to confirm with more accuracy the trends which were observed in the first phase. The first test of this second series consists in (sand+bentonite) swelling with no gas pressure. This test has just ended and the raw results are presented in figure 13 below.

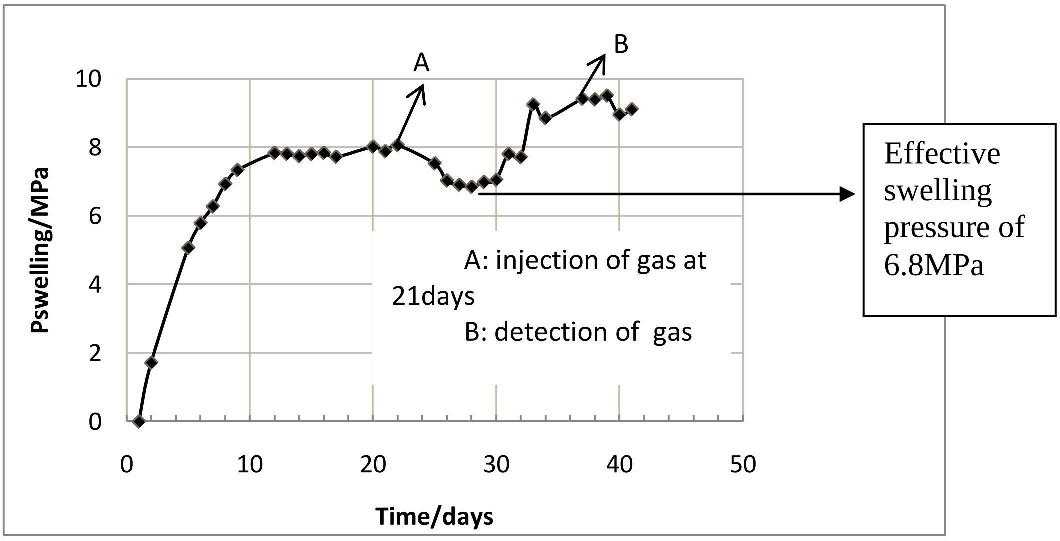


Figure 13: Swelling pressure with no gas pressure on top. PGZ experiment.

All these new tests (from this 2nd series) will be analysed and deeply commented in the next report

7. Swelling pressure of compacted bentonite (into highest Plexiglass™-aluminium tubes) – coupling phenomena

To be completed: several tests have already been performed without a sufficient amount of results to be presented

8. Gas entry pressure through argillite and bentonite

To be completed: several tests have already been performed without a sufficient amount of results to be presented

9. Partial conclusion

In this first part of our experimental work for both WP3.2.8 and WP3.3.4, it has been necessary to design and to build new experimental devices in order to manage our whole experimental program for FORGE. Two main issues had to be addressed: 1) gas influence upon the swelling process of a (sand+bentonite) mix and the behaviour of the sand+bentonite/argillite interface in relation with gas transfer. Materials and all experimental tools are now available and some important preliminary results have already been obtained. They are presented in part 3 to 6. To sum these up:

- Swelling under oedometric conditions has been studied and needs to be completed with swelling under free volumetric strain conditions. This last point is being tested.
- A complete "PGZ" experiment had been completed. We have shown that gas pressure has little influence on the (sand+bentonite) mix swelling pressure, up to a threshold between 4 and 8MPa, but this is still to be determined more accurately. It is now obvious that an 8MPa gas pressure deeply slows down the swelling process and decreases the swelling pressure. On another hand, if gas pressure is decreased from 8 to 6MPa, swelling starts again with a quite high rate (with 6-7 days only to get stabilization); swelling pressure finally reaches its target value, but the corresponding gas entry pressure of the swelled mix is lower than that obtained when the plug is saturated without gas pressure. In a first approach, this means that the porous structure has changed due to gas pressure effect.

A number of additional results will be obtained later in order to confirm these first partial conclusions and to complete the study, especially as regards coupling phenomena and the hydraulic behaviour of interfaces between bentonite and Plexiglass™-aluminium or argillite tubes.