Description of laboratory gas transport experimental set-ups and procedures

FORGE Report D5.3 – VER.0

	Name	Organisation	Signature	Date
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Euratom 7th Framework Programme Project: FORGE

EURATOM

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.

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1 Introduction

This report describes the experimental set-ups for the laboratory experiments related to gas transport in undisturbed clay within WP5. Forge participants, i.e. BGS and CIEMAT were asked to complete a detailed pro-forma the results of which are presented in the following pages of this document, fulfilling deliverable D5.3.

2 Completed pro-formas

Experimental Systems and Planned Activities

Organisation
BGS
Work package
WP 5.1.1 Baseline hydraulic and gas transport properties of the Callovo-Oxfordian argillite
Description of planned research/modelling work
The specific objective of the experimental programme is to examine the baseline hydraulic and gas transport parameters of the Callovo-Oxfordian argillite in order to investigate the validity of two-phase concepts applied to argillaceous formations. The study will form part of a wider programme of work focused on the Callovo-Oxfordian argillite and will compliment ongoing experimental activities currently underway at the BGS. The project will comprise: (i) a detailed programme of carefully controlled laboratory experiments under simulated in situ (isotropic) conditions of stress, pore pressure and chemical environment; (ii) full interpretation of data and the development of process understanding; (iii) numerical modelling using a conventional two-phase flow code; (iv) dissemination and publication of results, including full participation within the EU FORGE project. Tests will be performed under simulated in situ (isotropic) conditions of stress, pore pressure and chemical environment. As well as defining the long-term gas transport parameters, emphasis will also be placed on examining (a) the sensitivity of gas flow to the evolution of effective stress, (b) the non-linearity of the gas flow law caused by cyclic gas loading of the formation (hysteresis) and (c) the use of nano-particles to try and examine/directly image the location and distribution of gas pathways within the clay. All of which will provide additional data to guide process understanding.
Experimental set-up/code description
The basic permeameter (Figure 1) consists of five main components: (1) a specimen assembly, (2) a 70 MPa rated pressure vessel and associated confining pressure system, (3) a fluid injection system, (4) a backpressure system, and (5) a PC-based data acquisition system. The specimen is subject to an isotropic confining stress, with injection platen

mounted on the base of the specimen. A novel feature of the apparatus is the use of porous annular guard-ring filters around the inflow and outflow filters. The pressures in these two guard-rings can be independently monitored. The advantages of the guard-ring approach are: (a) pore pressure evolution can be studied, (b) hydraulic anisotropy can be quantified in a single test, (c) a check can be made of flow symmetry in the specimen, (d) excess gas pressure at gas entry can be determined, and (e) uncertainties associated with possible sheath leakage can be eliminated from data interpretation. Injection through the base.

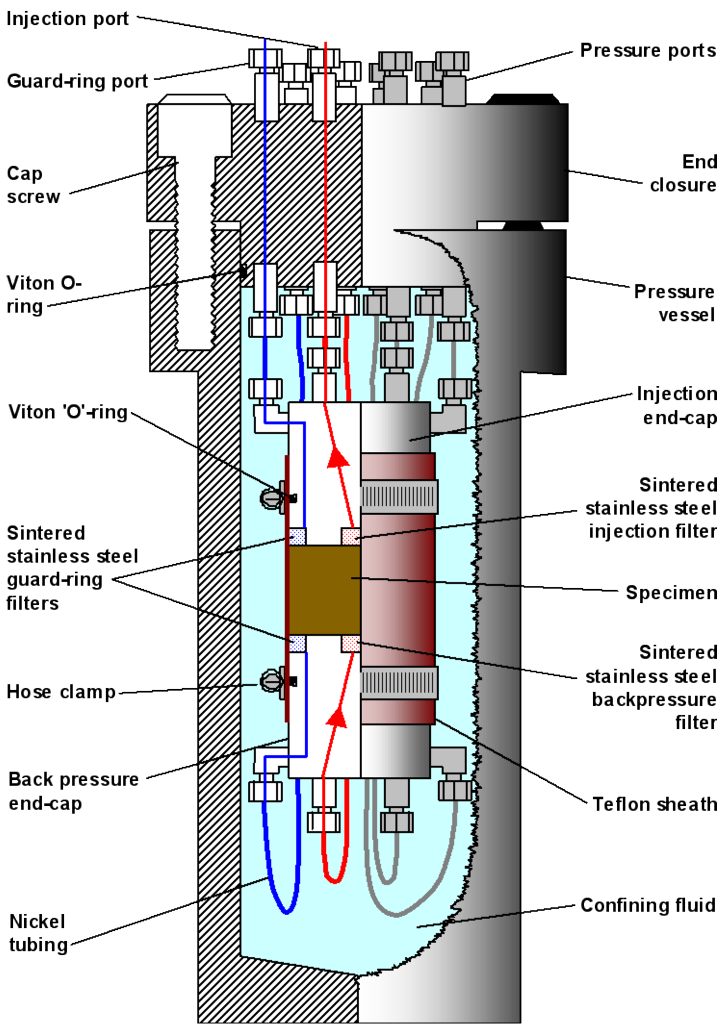

Figure 1. Schematic diagram of the pressure vessel and sample assembly for the BGS guard-ring permeameter. The pressure vessel is a custom-built stainless steel vessel rated to 70 MPa. The cylindrical clay specimen is sandwiched between two stainless steel end-caps, each with two filters recessed into the load-bearing surface, and jacketed in heat shrink Teflon to exclude the confining fluid.

The test specimen is sandwiched between two stainless steel end-caps and jacketed in heat-shrink Teflon to exclude confining fluid. Tapered locking rings compress the Teflon against two Viton "O"-rings in each end-cap to provide a leak-tight seal. The inlet and outlet zones for water or gas flow through the specimen are provided by porous filter discs 20 mm in diameter which are recessed into the bearing surface of the end-caps (Figure 1). These act as either source or sink for the injection of test permeants. Annular guard-ring filters with an internal diameter of 48.4 mm and an external diameter of 54.4 mm are recessed into the end-caps so that they completely encircle the inlet and outlet filters. A seal between the guard-ring and source/sink filters is achieved through the application of the confining stress, compressing the carefully machined surface of each platen against the clay. During hydraulic measurements, all the filters are saturated with an aqueous porewater solution.

Volumetric flow rates are controlled or monitored using a pair of ISCO-260, Series D, syringe pumps operated from a single digital control unit. The position of each pump piston is determined by an optically encoded disc graduated in segments equivalent to a change in volume of 16.6 nL. Movement of the pump piston is controlled by a micro-processor which continuously monitors and adjusts the rate of rotation of the encoded disc using a DC-motor connected to the piston assembly via a geared worm drive. This allows each pump to operate in either constant pressure or constant flow modes. A programme written in LabVIEWTM elicits data from the pump at pre-set time intervals. Testing is performed in an air-conditioned laboratory at a nominal temperature of 20 ºC.

Boundary conditions

In order to limit osmotic swelling of the specimen, a synthetic porewater solution was prepared for use as the backpressuring fluid and permeant during hydraulic test stages. Details of the hydrochemistry of the interstitial fluid were provided by Andra. A stock solution comprised of the following components was used as the aqueous test fluid in all hydraulic and consolidation test stages: Ca2+ (227 mg l-1); Mg2+ (125 mg l-1); Na+ (1012 mg l-1); K+ (35.7 mg l-1); SO42- (1266 mg l-1); Si (4.59 mg l-1); SiO2 (9.83 mg l-1); Sr (13.5 mg l-1); total S (423 mg l-1); total Fe (0.941 mg l-1). ICP-AES analysis of the stock solution was undertaken to verify the fluid composition. This fluid was saturated with fluorescein prior to testing. Post-test analysis of the core will be undertaken to try and identify the location and extent of the fluorescein within the sample. Helium gas (selected as a safe substitute for hydrogen) will be used to measure the gas transport properties of the argillite. *In situ* (isotropic) confining stress data has been provided by Andra with the initial confining stress nominally set to 12.5 MPa with a backpressure of 4.0 MPa.

Measured parameters/output from code

Measured parameters are:

(a) the hydraulic conductivity and intrinsic (absolute) permeability;

(b) the (threshold) capillary displacement pressure;

(c) the effective gas permeability and relative permeability to gas for a range of conditions;

(d) the post-test gas saturation.

Test/modelling programme

Tests will be undertaken on intact samples of Callovo Oxfordian Clay. The first test planned within Forge will examine the response of CoX to destressing and is likely to comprise the following steps:

1. Specimens will be fully resaturated with a synthetic groundwater (S_w>0.99) while subject to the original conditions of effective (isotropic) stress.
2. The intrinsic permeability of the specimens will then be measured by a combination of controlled flow rate and/or constant head methodologies using the synthetic groundwater solution.
3. After a period of pore pressure equilibration, gas testing (using H_e as the test permeant) will commence using a combination of controlled flow rate and/or constant pressure techniques in order to determine the initial gas transport parameters.

4. Effective stress will then be decreased in a number of steps while continually monitoring changes in flux in order to determine the change in gas permeability for each test stage as a function of stress state. This will provide information to help understanding the processes in relation to stress unloading of the argillite.
5. This will be followed by a second stage of hydraulic measurements to examine the self sealing characteristics of the argillite at the lower stress state and the effect of residual gas on the hydraulic parameters specific storage and permeability.
6. If time permits a repeat gas test (again using H_e as the test permeant) may then be undertaken in order to confirm the self sealing characteristics of the argillite and to determine the gas breakthrough pressure for the argillite at this particular stress state.
7. Basic geotechnical properties will be re-measured at the end of the test.
8. Test stages may be added or deleted depending on previous results.
9. Full interpretation and modelling of the data (where appropriate) will be undertaken.

Provisional time line

D4.1: Description of the experimental set-up: March 2010
Preliminary timeline:
Test 1 March 2010 to October 2011.
Test 2 November 2011 to December 2012.
Additional tests will be performed if time permits.

Organisation

CIEMAT

Work package

WP 5.1.2 Determination of two-phase flow parameters and analysis of fracturing by gas overpressure in Opalinus Clay

Description of planned research/modelling work

5.1.2.1 Determination of two-phase flow parameters: Tests will be performed in suction controlled oedometers in which the conditions in the repository with respect to stresses will be reproduced as well as possible. Different suctions will be applied to similar samples until stabilisation of the vertical strain. The retention curve under constant stress conditions and the capillary pressure will be thus determined.

5.1.2.2 Analysis of fracturing induced by gas overpressure: Study of fracture induced by local concentration of gas pressure as a function of the direction of measurement with respect to the bedding planes. The tests will be performed in core samples submitted to confining conditions similar to the *in situ* ones. When possible, the permeability of the argillite will also be determined.

Experimental set-up/code description

5.1.2.1 Suction controlled oedometer equipments
These equipments allow the study of the unsaturated behaviour of clays by imposing suction instead of measuring it, that is to say, by subjecting the sample to a given and

known suction that conditions its water content, while the other variables (stresses, strains) are modified or measured. Two different techniques will be used to impose suction: axis translation (matric suction, in membrane cells) and the imposition of relative humidity (total suction, in cells with deposit for solution).

The principle of axis translation consists in modifying suction by increasing the pressure of the gaseous phase. The sample is placed in a cell in contact with water at atmospheric pressure through a membrane permeable to water but not to gas. These regenerated cellulose membranes are amorphous and gel type in nature and have a pore diameter of 2.4 nm, as a result of which they are flexible and suitable for filtration and osmosis work. The pressure in the cell is increased by injecting gas at the desired pressure, this increasing the air pressure in the pores of the sample. This new situation forces the sample to exchange water through the membrane until equilibrium is reached once again. Given that the changes in capillary suction are caused by the difference between the pressure of the air in the pores (u_a) and the pressure of the water (u_w), when air pressure is applied to the sample an increase in u_a is induced, while u_w remains the same as atmospheric pressure. In this way, capillary suction varies by the same amount as gas pressure. The membrane allows ions to pass through, as a result of which osmotic suction is not controlled by this method.

The membrane cell is manufactured in stainless steel and consists of a base, cover and central body (Figure 1). The oedometer ring is housed in the body of the cell. The upper part of the cell has a central orifice for passage of the loading ram. This rests on the load distributing piston, which has a porous stone attached to it at its lower end, which remains directly in contact with the sample. The cover of the cell also has a gas inlet with a manometer for values of up to 16 MPa. Externally, a strain dial gauge with a level of accuracy of thousands of a millimetre, coupled to the loading ram, rests on the cell cover so that to measure the vertical deformation of the sample. The base of the cell has an embedded porous stone, below which there are two inlet and outlet orifices, connected to a deposit with water at atmospheric pressure. The semipermeable, regenerated cellulose membrane is placed over this stone, with the sample resting directly on it. A peristaltic pump, installed between the deposit and the cell inlets, facilitates the removal of the gas that could diffuse through the membrane. Given the mechanical limitations of the cell, it is possible only to apply matric suctions of less than 14 MPa. Industrial nitrogen is used as the gas.

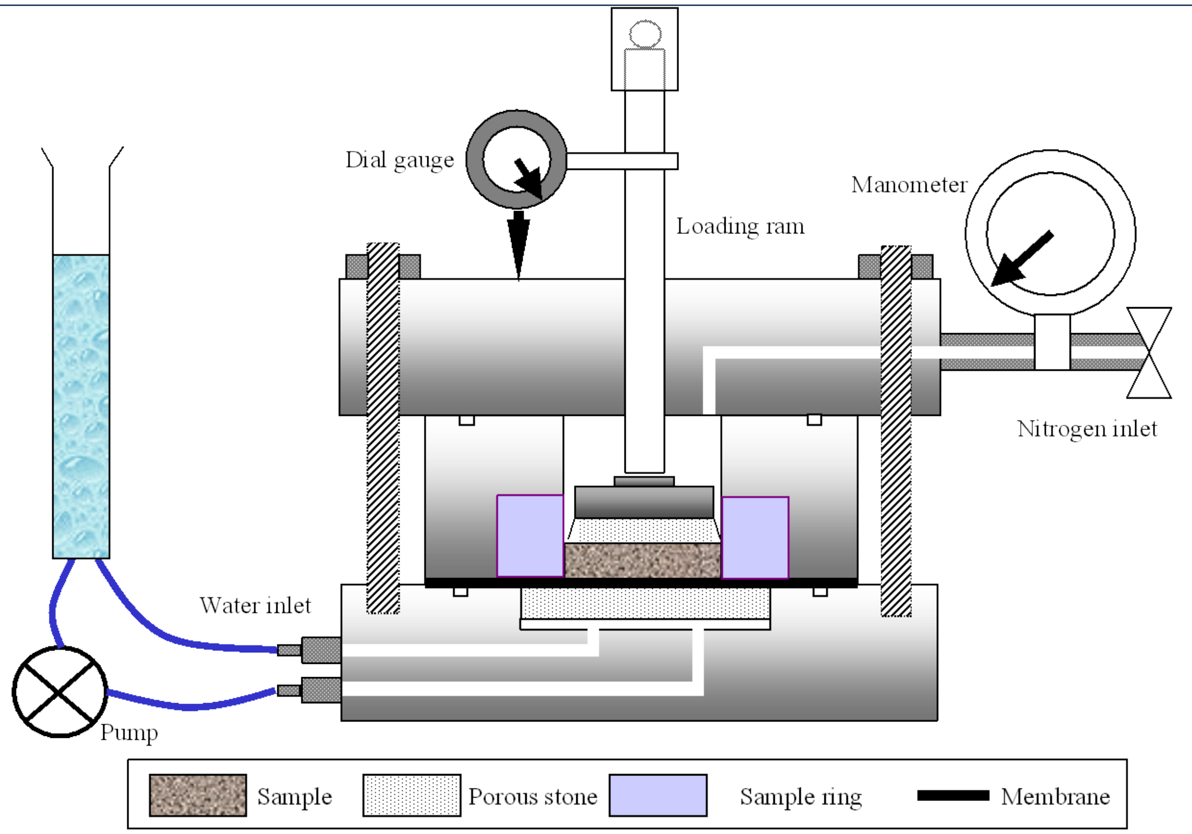

Figure 1: Schematic cross section of an oedometric membrane cell

The method of imposing relative humidity (*RH*) is based on the fact that this conditions the pressure of the water and gas in the pores (u_w and u_a). This humidity may be imposed by means of solutions of sulphuric acid (although any other solution of known water activity may be used). The sample exchanges water with the atmosphere until thermodynamic equilibrium is reached with the vapour pressure of the solution, as a result of which total suction is modified. The suction in the pores of the sample (*s*, in MPa) is related to the activity of the water in the solution (a_w=*RH*/100) by means of Kelvin's law:

$$s = -10^{-6}\,\frac{R \times T}{V_w}\ln\left(\frac{HR}{100}\right)$$

where *R* is the universal constant of the gases (8.3143 J/mol·K), *T* is absolute temperature and V_w is the molar volume of the water ($1.80 \cdot 10^{-5}$ m^3/mol).

The relation between the activity of the solution and the percentage in weight of sulphuric acid used to prepare it is reflected in experimental tables. The transfer of water between the clay and the atmosphere may cause the density of the solution to vary, as a result of which this should be checked prior to and following stabilisation, this being accomplished by means of pycnometers. There is an experimental relation between the specific gravity of the solution and the percentage in weight of the sulphuric acid in the solution (which in turn depends on activity), which is temperature-dependent.

Total suctions of between 3 and 500 MPa may be obtained using this method. In view of the influence of temperature on the activity of the solutions, this should be kept constant and known throughout the entire test.

The oedometric equipment used includes modified oedometric cells in which suction may be applied. The cell with a deposit for solutions consists of a base and cover of high corrosion-resistant stainless steel (AISI 316L) and a cylindrical body of transparent material with an internal border on which rests a ring-shaped glass deposit (Figure 2). A porous stone rests on the base of the cell, over which is the oedometer ring with the sample and finally, the upper porous stone attached to the piston. This assembly is attached to the base of the cell by means of a steel flange. The upper cover has an orifice for the loading ram, a perforation for insertion of the sulphuric acid and an inlet for the creation of the vacuum. Externally, a dial gauge accurate to thousands of a millimetre rests on the cell cover, coupled to the loading ram, to measure the vertical deformation of the sample.

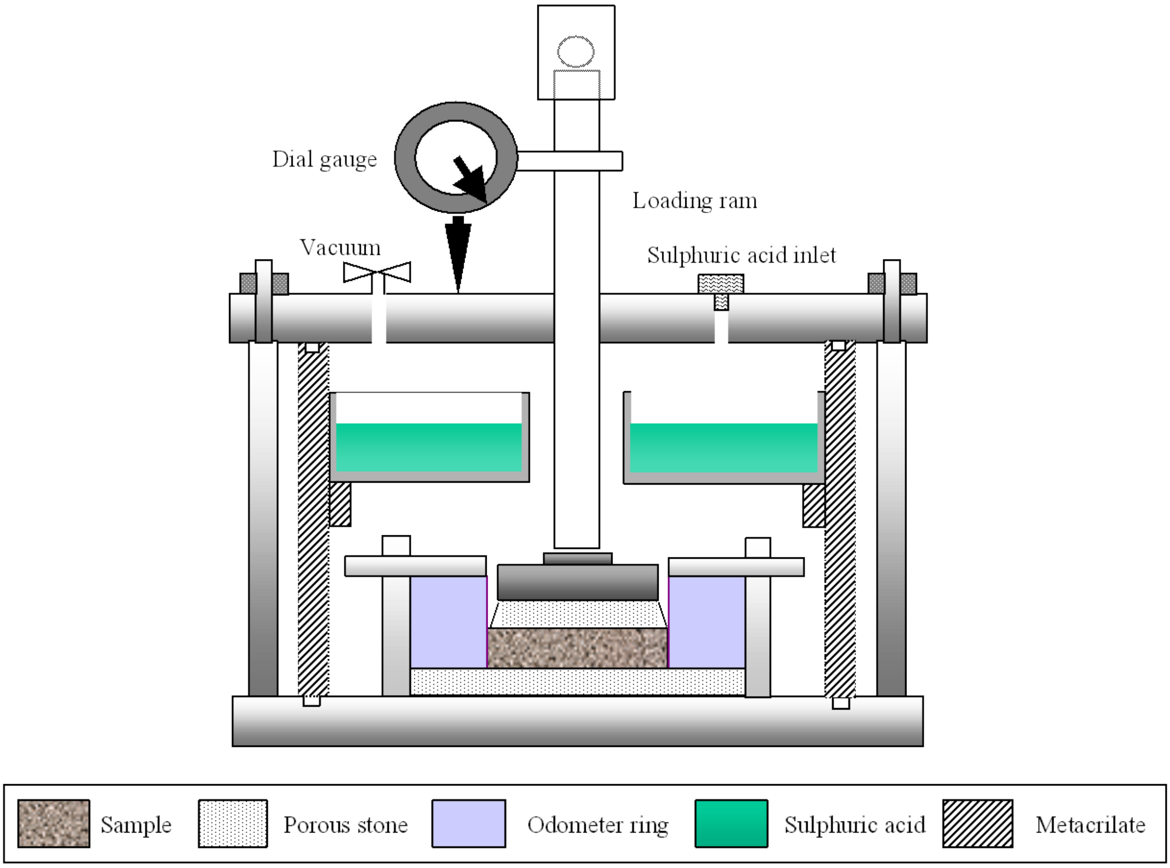

Figure 2: Schematic cross section of an oedometric cell with deposit for solutions

5.1.2.2 Analysis of fracturing induced by gas overpressure

The setup has been designed to perform steady gas permeability measurements under different gas pressures (Figure 3). The cylindrical sample is confined in a triaxial cell. The injection and downstream pressures can be independently varied and kept constant during the period of time necessary to get steady flow. Different range flowmeters measure the inward and outward flows. For the confining conditions two possibilities exist:

- The sample is wrapped in a latex membrane and the water in the triaxial cell is pressurised to the desired confining pressure.
- The sample has a rigid jacket that hinders its deformation and assures a perfect contact with the sample. In this way the confining pressure is given by the sample swelling characteristics.

The equipment works like a constant head permeameter, with the possibility to change the head value and measure the gas flow value. The system applies the pressures to the sample and registers flow and pressures from the measurement devices. In and outflow gas rates, up and downstream pressure, and the confining pressure are monitored.

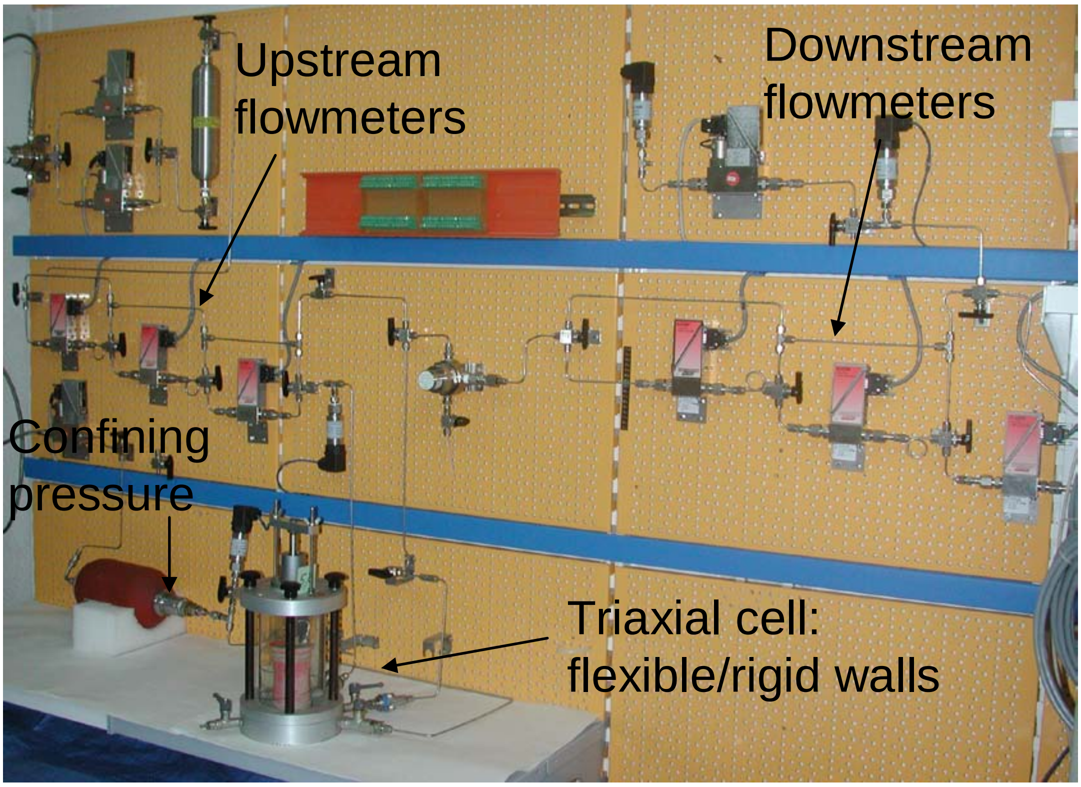

Figure 3: Appearance of the setup for measurement of gas permeability and breakthrough pressure

A more detailled drescription of the component of the experimental setup includes:

- *Test cell*. It is a modification of a commercial triaxial cell. The cell walls are made out of transparent plastic and are capable of withstanding pressures to 2700 kPa. Each cell has four inlets drilled in the base, one for sample top drainage/back pressure, two for sample bottom drainage/pore pressure, and one for confining pressure. Four no volume change valves are connected to the ports and an anvil fits the cell head. The tests will not be performed under real triaxial conditions.
- *Tubing, fitting and valves*. All the SWAGELOK fitting materials and valves are made out of stainless steel, SS316. The SANVICK tubing material is SS316 1/8”. The maximum leakage rate that manufacturer assures at each valve packing is around 0.1 cm^3/min at 68 bar g.
- *Water/nitrogen separator*. An OLAER’s pressure accumulator (to 330 bar) with an elastic membrane acts as separator between the nitrogen and the water phases of the confining pressure system.
- *Gas buffer*. A WHITEY gas sample SS-316 cylinder (300 cm^3), placed upstream, removes the fluctuations introduced by the pressure controller in the flow measurement. Also, it permits to keep constant the expected flow even in case of pneumatic fracturing.
- *Gas mass flowmeters*. The injected fluid flow rates are measured using three pairs of HITECH Gas mass flowmeters with different measurement flow range: 0.2-10, 2-100 and 20-1000 STP cm^3/min. It means that the minimum and maximum values measured without uncertainty in the system are around 0.003 and 16.7 STP cm^3/s, respectively. HI-TEC flowmeters operate on a principle of heat transfer by sensing the temperature

increment along a heated section of a capillary tube. They are calibrated to the consigned conditions: gas type N, pressure 70 bar a, and temperature 20ºC. The output signal is 0-5 VDC. Application software, such as FLUIDAT, enables to calculate accurate conversion factors from the calibration data, not only at 20ºC/1 atm but also at any temperature/pressure combination. This software will be used used to calculate the conversion factors to be applied.

- *Pressure controllers* (Figure 4). HI-TEC Gas forward pressure controllers are calibrated to the consigned conditions: gas type N and temperature 20ºC, at different pressures and maximum flow capacities. The output and control signals are 0-5 VDC. The maximum differential pressure is 30 bar. The PIC1 and PIC4 controllers give the target gas injection pressure. The PIC2 controller regulates the internal pressure within the nitrogen/water separator, the water acting as confining medium in the triaxial cell. The PIC3 controller gives the target gas outlet pressure.
- *Pressure transmitters* (Figure 4). Associated with the pressure controllers, DRUCK pressure transmitters, PTX1400 series, have been placed at several points: the inlet port of the triaxial cell (injection pressure), the outlet port of the water/nitrogen separator (confining pressure), and the outlet of the system (oulet pressure or atmospheric pressure). The transmitters range is 100 bar a (0.25% BSL). The output signal is 4-20 mA.

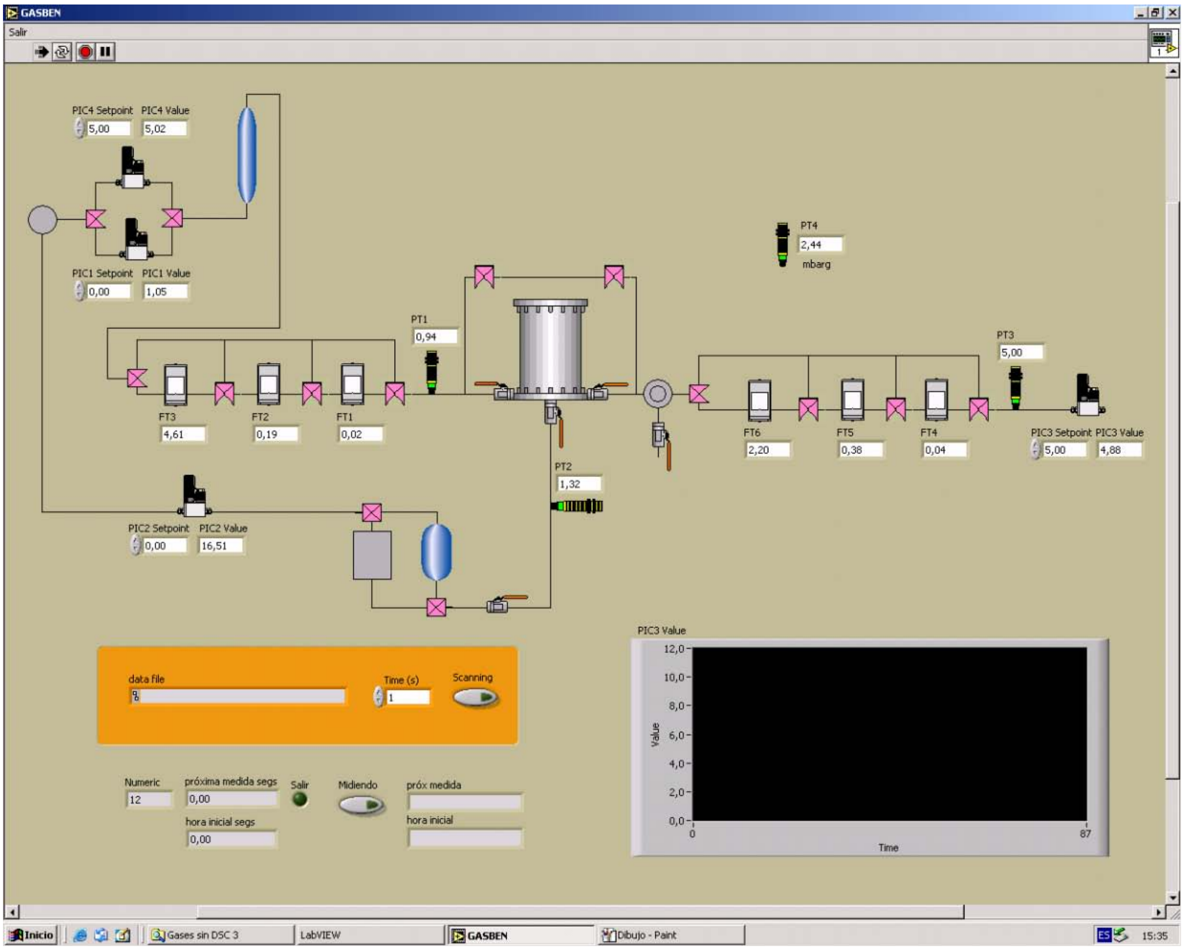

Figure 4: Schematic diagram of the setup for the gas permeability tests

Boundary conditions

5.1.2.1 Determination of two-phase flow parameters in suction controlled oedometer equipments

- The test will be performed under constant vertical stress.
- Gas pressure will be varied between 0.1 and 14 MPa and pore water pressure will be atmospheric in the membrane cell tests.
- Total suction will be applied by controlling the relative humidity in the cells with deposit.
- Temperature will be 20°C.

5.1.2.2 Analysis of fracturing induced by gas overpressure

- The confining pressure will be similar to the in situ one.
- The downstream pressure will be kept constant.
- Gas injection pressure will be varied by steps.
- Zero vertical displacement.
- Effective tension on the sample: variable.
- The tests will be performed at room temperature.

Measured parameters/output from code

5.1.2.1 Determination of two-phase flow parameters in suction controlled oedometer equipments

- Vertical strain
- Final water content (or equilibrium water content) and dry density
- The continuous measurement of water exchange in the membrane cells will be attempted, although the preliminary results are not supportive.
- Suction (imposed)
- Vertical stress (imposed)

5.1.2.2 Analysis of fracturing induced by gas overpressure

- Confining pressure.
- Gas flow in and out the cores.
- Gas pressure upstream and downstream.
- Estimation of the apparent permeability under gas stable flow conditions, corresponding to the pressure step in which the gas breakthrough is produced and subsequent steps. Also, the deduction of the possible generation of preferential paths.
- Final density and water content of the clay
- Confining pressure (can be changed or kept equal to the in situ one)
- Gas pressure upstream and downstream, in order to know the minimum pressure at

which the gas breakthrough is produced.

Test/modelling programme

5.1.2.1 Determination of two-phase flow parameters in suction controlled oedometer equipments
The clay will be used in undisturbed conditions with respect to water content, suction and dry density (as much as possible). In oedometers with suction controlled by solutions, the tests will begin at a suction similar to that existing in situ according to the results obtained in previous tests, this subsequently being increased stepwise. In the tests performed using oedometers with suction control by nitrogen pressure, the maximum applicable suction, 14 MPa, is close to the in situ value, for which reason this will be the initial value, and subsequently, wetting paths will follow. Alternatively, the sample can be initially saturated under suction 0 MPa and submitted to a drying path up to 14 MPa.

5.1.2.2 Analysis of fracturing induced by gas overpressure
The samples will be trimmed from intact core samples, thus the clay will be used in undisturbed conditions with respect to water content and dry density (as much as possible). Two types of experimental procedures are foreseen as a function of the expected gas permeability:

- Medium-low permeability samples: This procedure is related to samples with an open porosity and low water content that allow gas flow and consequently computation of matrix permeability. The injection and confining pressures will be progressively increased and permeability will be computed from the outflow once it is steady.
- Fracture or breakthrough in low permeability samples: The injection pressure will be increased stepwise (0.2 MPa per day), until reaching the breakthrough value. Accordingly, confining pressure must be also increased in the same rate. After breakthrough is obtained, the gas pressure paths will be closed for 24 hours, and afterwards the procedure described above will be repeated, in order to determine the possible development of preferential paths which could reduce the value of the gas breakthrough pressure.

Provisional time line

5.1.2.1 Determination of two-phase flow parameters in suction controlled oedometer equipments
Tests will start in February 2010 and go on for at least 1 year.

5.1.2.2 Analysis of fracturing induced by gas overpressure
Preliminary tests will start in February 2010. Systematic measurements will start along 2010, the exact date depending on the preliminary results.