Description of experimental setups and procedures for CIEMAT gas transport tests of WP5

FORGE Report D5.7

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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 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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FORGE

Description of experimental setups and procedures for CIEMAT gas transport tests of WP5

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WP 5.1.2 DETERMINATION OF TWO-PHASE FLOW PARAMETERS AND ANALYSIS OF FRACTURING BY GAS OVERPRESSURE IN OPALINUS CLAY (CIEMAT)

CIEMAT plans to perform two kind of tests in indurated clay. Although in the proposal the material to be used was not specified, by taking into account the work to be performed by the other participants and our own experience, we have chosen the Opalinus clay.

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.

1.1 Description of setup: 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).

1.1.1 Membrane cells

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.

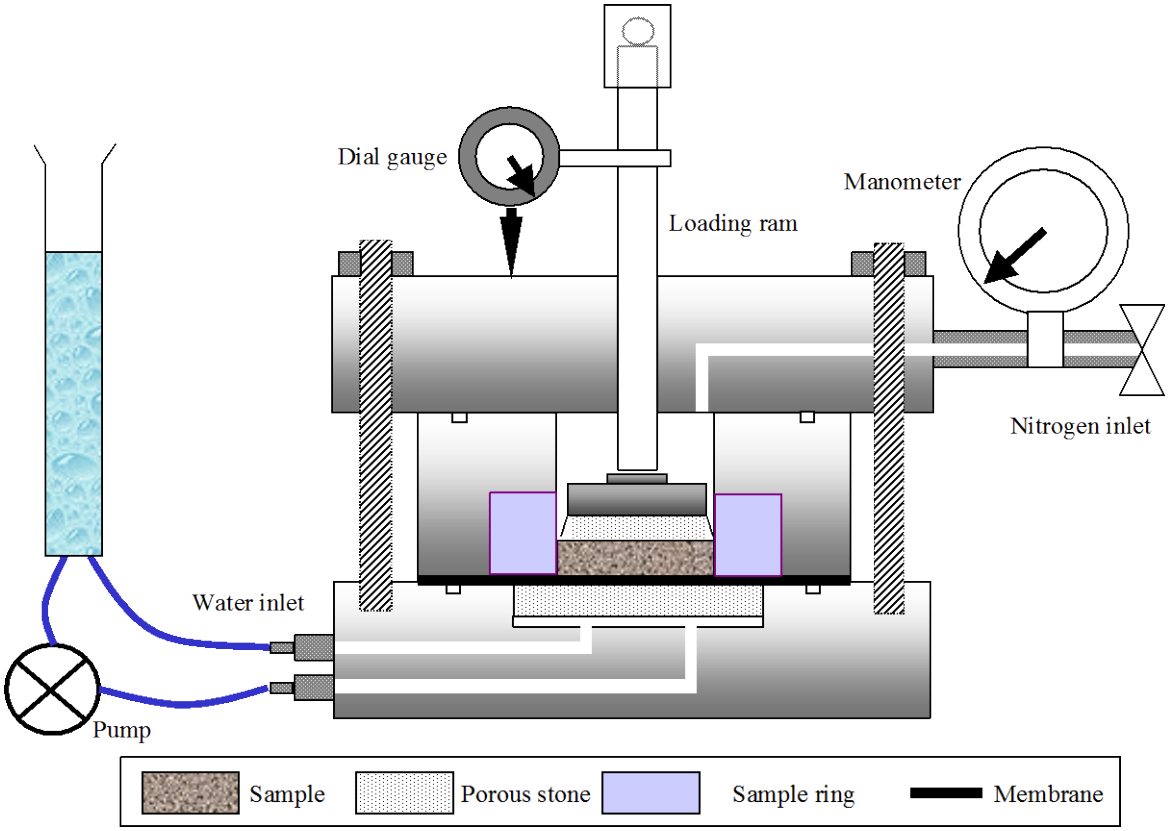

Figure 1: Schematic cross section of an oedometric membrane cell

1.1.2 Cells with deposit for solution

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.

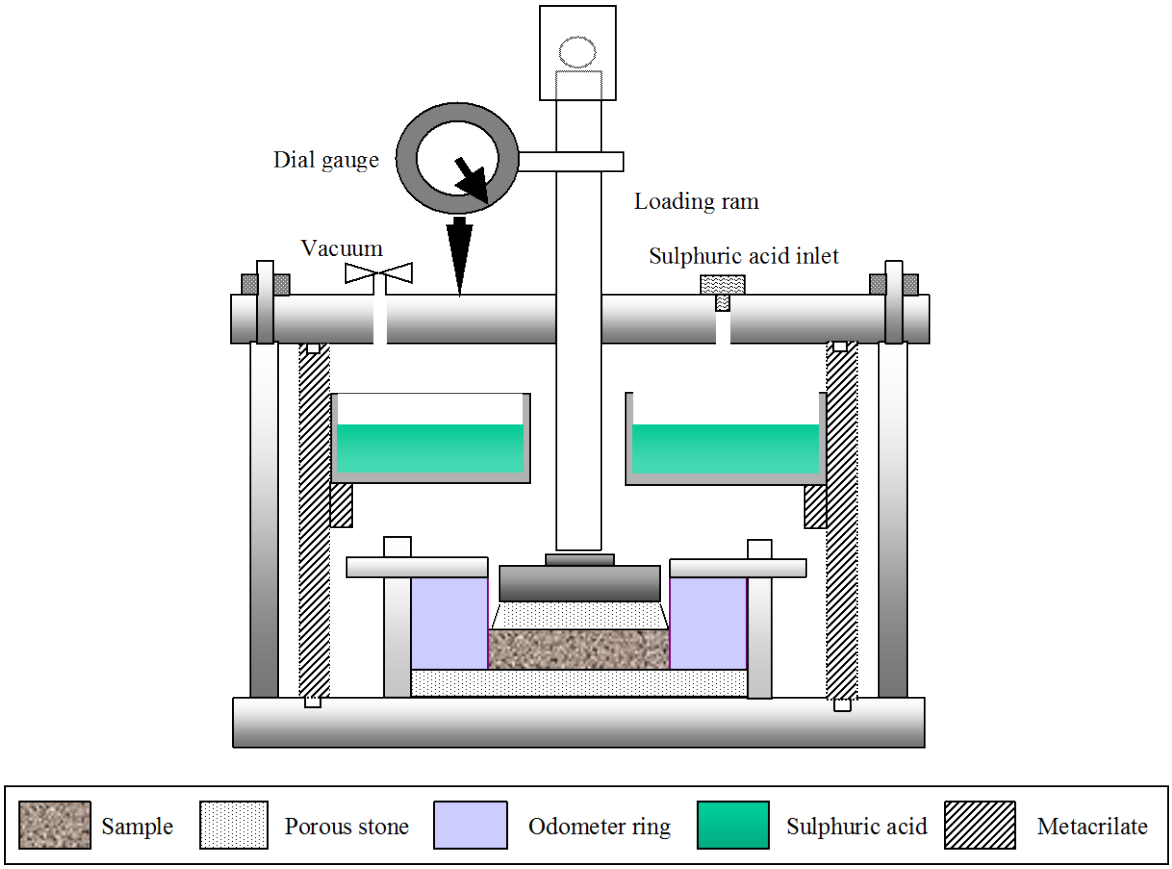

Figure 2: Schematic cross section of an oedometric cell with deposit for solutions

1.2 Description of experimental procedure

The as-received clay will be trimmed in cylindrical specimens to fit the oedometer ring, *i.e.* 1.2-2.0 cm in length and 11.40 or 19.24 cm^2 in cross section.

Once the oedometer ring with the sample is placed in the oedometer, the desired suction and vertical stress are applied and the strain recorded by the dial gauge is measured. Once equilibrium has been reached for a given suction and stress, that is to say, when there is no longer any strain, the cell is released, the ring and the sample are weighed (since the sample cannot be removed from the ring without undergoing disturbance) and the specimen is indirectly measured. Afterwards, the oedometer ring is immediately assembled in the cell and vertical stress is applied, along with the suction existing prior to disassemblage, waiting for 24 hours until equilibrium is reached again before going on to the next suction step. The duration of each step has been established at a minimum 20 days, even if the strain for a given load has stabilised earlier.

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.

The possibility of using a synthetic water reproducing the *in situ* salinity of the argillite will be investigated for the tests in membrane cells. Also, the scale effect can be checked by testing different height samples. Although the preliminary efforts to get a continuous measurement of water exchange in the membrane cells have failed, some modifications of the procedure will be tested to achieve this goal (using a less soluble gas than nitrogen (He, Ar), using lower air pressures, etc.).

1.3 Conditions and parameters

Initial conditions

The clay will be used in undisturbed conditions with respect to water content, suction and dry density (as much as possible).

Boundary conditions:

- 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.

Measured parameters:

- 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.

Varied parameters:

- Suction
- Vertical stress

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.

2.1 Description of setup: steady state gas permeability equipment

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.

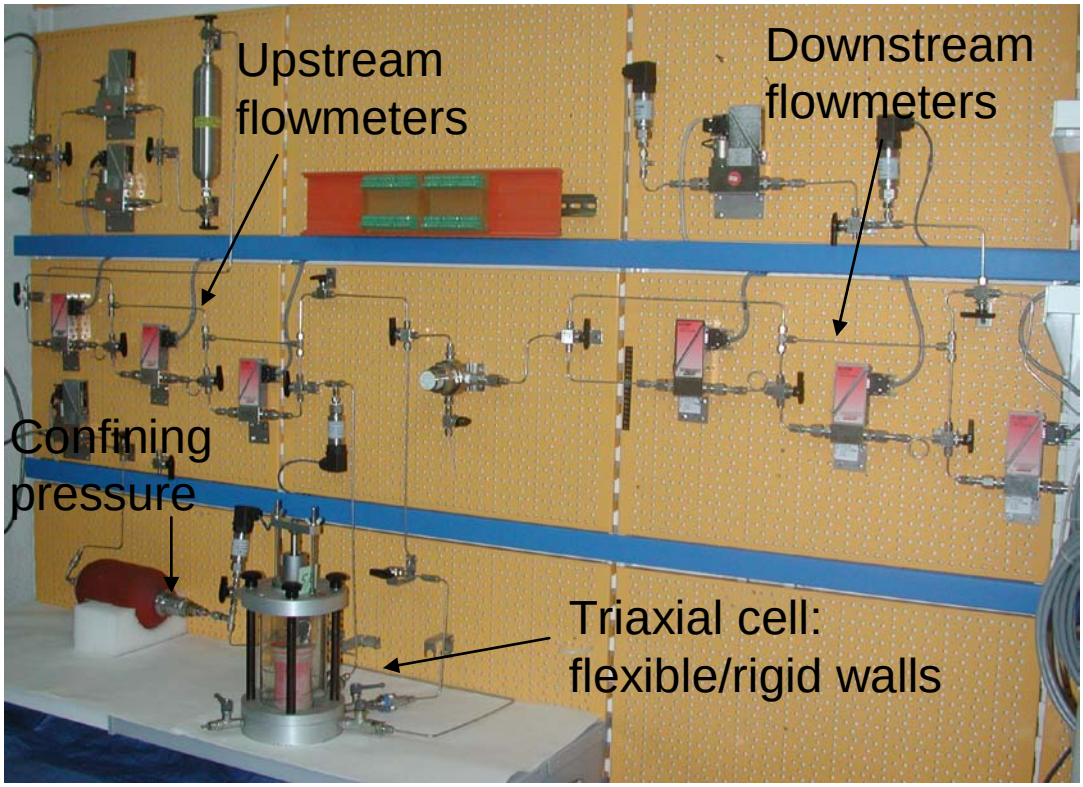

Figure 3: Appearance of the setup for measurement of gas permeability and breakthrough pressure

A more detailed description 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 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 (outlet pressure or atmospheric pressure). The transmitters range is 100 bar a (0.25% BSL). The output signal is 4-20 mA.

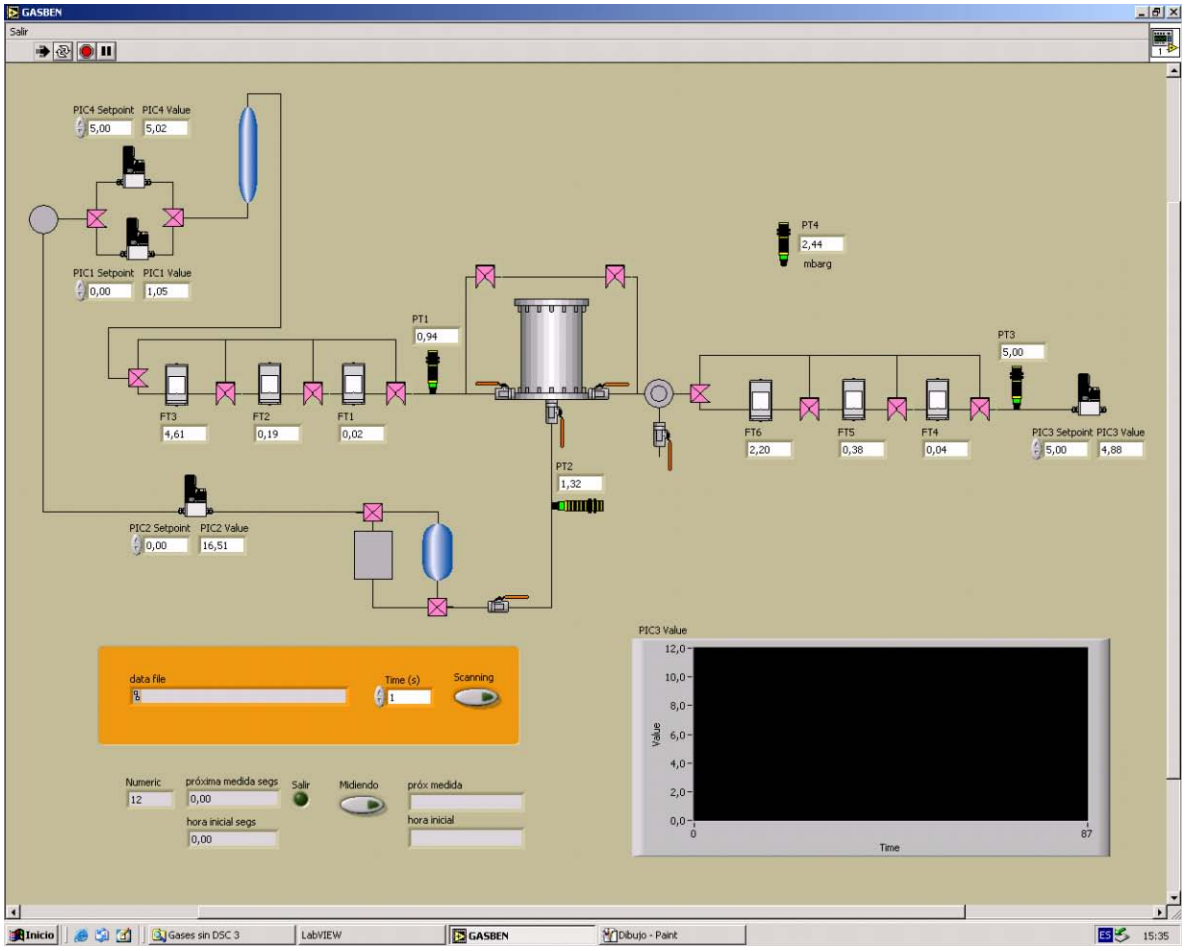

Figure 4: Schematic diagram of the setup for the gas permeability tests

2.2 Description of experimental procedure

The samples will be trimmed from intact core samples. The resulting specimens will be 6-8 cm in height and 4 cm in diameter. The gas used will be nitrogen.

In the selected test procedure the injection pressure is much lower than the confining pressure and will be applied in constant-pressure short multi-steps, with small pressure increments. The confining pressure will be as close as possible to the *in situ* conditions of the intact core samples and the maximum gas pressure is estimated to be in the range of 2.5 MPa. All pressure values are absolute.

Two types of experimental procedures are foreseen as a function of the expected gas permeability, but the following precautions must be taken in both: (1) the confining pressure will be enough to ensure that gas flows through the sample with no peripheral pathways (at least 0.6 MPa, differential pressure in relation to the injection pressure); (2) the first steps will be conducted under small pressure gradient to prevent the overload of the flowmeters.

2.2.1 Medium-low permeability samples

This procedure is related to samples with an open porosity and low water content that allow the gas flow and consequently the computation of matrix permeability. Each step has three phases:

1) Pressures are applied in the following sequence: confining pressure to stabilisation, downstream pressure is fixed and upstream pressure is applied to the sample.

2) Flowmeters are connected in this sequence: 1000, 100 and 10 mL/min. In this way, we obtain redundant flow values (two significant flowmeters if flow is lower than 100 mL/min).

3) If no gas flow is detected at the inlet after about 1 hour, the injection pressure is increased by 0.5 bar.

4) Once a steady value of flow is obtained (*i.e*. no fluctuations in flowmeters during a reasonable time) both at the inlet and the outlet, the next step of the experiment is applied (phases 1 and 2).

2.2.2 Fracture or breakthrough in low permeability samples

The previous considerations must also be taken into account, but in this case if no gas breakthrough is obtained for the applied pressure value after 24 hours, 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. All the possible modifications of the system will be recorded at short intervals.

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.

Overall, seven working days are required to increase the pressure up to the value of 20 bar absolute, which is the maximum pressure to be used in the experiment.

2.3 Conditions and parameters

Initial conditions

The clay will be used in undisturbed conditions with respect to water content and dry density (as much as possible). Thus, initially the samples will be quasi-saturated.

Boundary conditions (depend on the type of test)

- 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

- 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

Varied parameters

- 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.