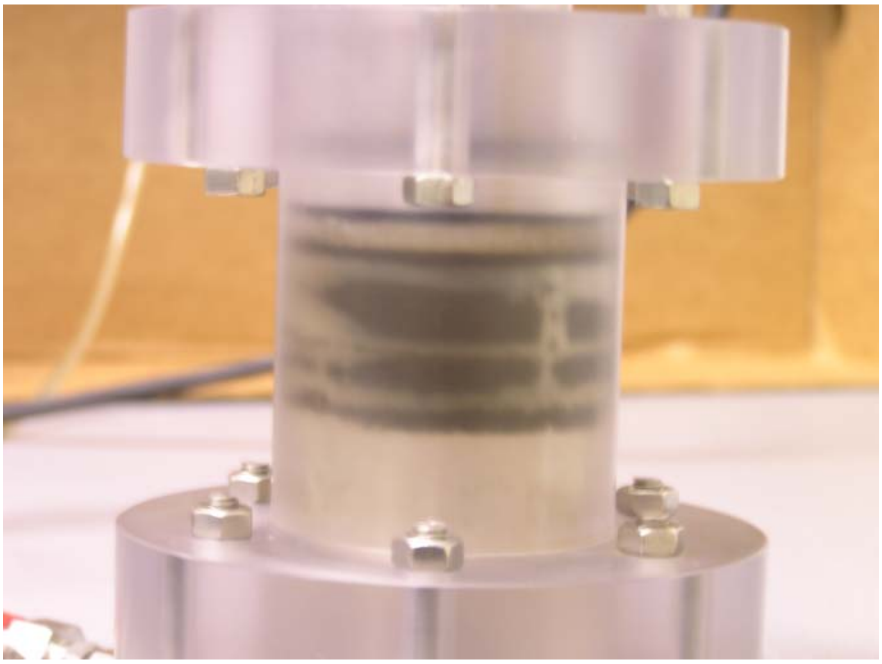
Detailed description of the experimental set-up and procedures for gas-driven RN-transport

FORGE Report D5.1 (SCK•CEN ER-105)



Detailed description of experimental set-up and procedures for gas-driven RN-transport

FORGE Report D5.1 (SCK•CEN-ER105)

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Compiled by: E. Jacops Date: 11/09/2009

Checked by: N. Maes Date: 06/10/2009

Approved by: R. Shaw Date: 12th October 2009

Keywords

Boom Clay; Gas breakthrough pressure.

Front cover

Clay core right after gas breakthrough.

Bibliographical reference

Jacops, E., Maes N., Volckaert G. (2009) Detailed description of experimental set-up and procedures for gas-driven RN-transport. - FORGE Report D5.1., SCK•CEN-ER105, Mol, Belgium.

Euratom 7th Framework project: FORGE 2009

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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Foreword

This report describes test procedures developed/used to investigate the gas-driven radionuclide transport in clays conducted in the framework of the EC FP7 project FORGE WP5.

Acknowledgements

The research was realised with the technical support of T. Maes, M. Van Gompel, J. Peeters and F. Slegers.

Next to the financial support of the European Commission in the framework of the FP7 FORGE project, this work is performed in close cooperation with, and with the financial support of ONDRAF/NIRAS, the Belgian Agency for Radioactive Waste and Fissile Materials, as part of the programme on geological disposal of high-level/long-lived radioactive waste that is carried out by ONDRAF/NIRAS.

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Summary

In deep geological repositories, gas can be generated by: anaerobic corrosion of metals in wastes and packaging; radiolysis of water and organic materials in the packages, and microbial degradation of various organic wastes. Corrosion and radiolysis yield mainly hydrogen while microbial degradation leads to methane and carbon dioxide.

The gas generated in the near field of a geological repository in clay will dissolve in the ground water and be transported away from the repository by diffusion as dissolved species. However if the gas generation rate is larger than the diffusive flux, the pore water will get oversaturated and a free gas phase will be formed. This will lead to a gas pressure build-up and finally to an advective gas flux. However with this gas flux, contaminated water could be expelled. Radionuclides and contaminants could be driven out of the undisturbed clay by a two-phase flow mechanism faster than the normally expected diffusive transport.

Work package 5 of the FORGE research programme should help us to answer the question: “to what extent can a gas pressure build-up enhance the radionuclide and contaminants transport in clayey materials?”

To answer this question, we will perform experiments with permeameter cells using undisturbed Boom Clay. After infiltration of a clay core with pore water that contains an anionic tracer, a gas breakthrough test will be performed to measure gas induced transport of this tracer.

To verify whether the basic concept will work or not, a preliminary test was executed with an existing cell. The results of this preliminary test are presented in this report, and based on this results the design of the experimental cells was improved.

1 Introduction

1.1 BACKGROUND

The main mechanisms by which gas will be generated in deep geological repositories are: anaerobic corrosion of metals in wastes and packaging; radiolysis of water and organic materials in the packages, and microbial degradation of various organic wastes. Corrosion and radiolysis yield mainly hydrogen while microbial degradation leads to methane and carbon dioxide.

The gas generated in the near field of a geological repository in clay willl dissolve in the ground water and be transported away from the repository by diffusion as dissolved species. However if the gas generation rate is larger than the diffusive flux, the pore water will get oversaturated and a free gas phase will be formed, leading to a gas pressure build-up. The gas production rates for various waste types and packages, especially ILLWs, although marred by large uncertainties, is expected to be significantly higher than the diffusive flux. Hence, one of our research objectives is to improve the understanding of gas transport modes through the EBS and clay when the capacity for transport of dissolved gasses is exceeded. Indeed, the processes by which gasses are transported in clays are still poorly understood: 2-phase flow, pathway dilatation (μ-fracturing), fracturing…?

Next to transport in the undisturbed host rock and EBS materials, the migration of gas via interfaces and the damaged zone of the host rock are important to assess: should these be considered as preferential flow paths for gas migration and could these result in gas driven RN transport?

These questions are addressed in the context of the EC project FORGE (Fate of repository gases), which aims at acquiring a deeper insight in the gas transport processes from a phenomenological point of view and studies the gas migration behaviour in different host rocks, EDZ and EBS.

1.2 OBJECTIVE OF WP5: UNDISTURBED HOST ROCK FORMATIONS

During gas breakthrough, contaminated water could be expelled by the gas phase. Radionuclides and contaminants could be driven out of the undisturbed clay by a two-phase flow mechanism faster than the normally expected diffusive transport.

This work package should help us to answer the question: “to what extent can a gas pressure build-up enhance the radionuclide and contaminants transport in clayey materials?”

To answer this question, we will perform experiments with permeameter cells using undisturbed Boom Clay. After infiltration of a clay core with pore water that contains an anionic tracer, a gas breakthrough test will be performed to investigate gas induced transport of this tracer.

2 Detailed description of the set-up

2.1 BASIC CONCEPT

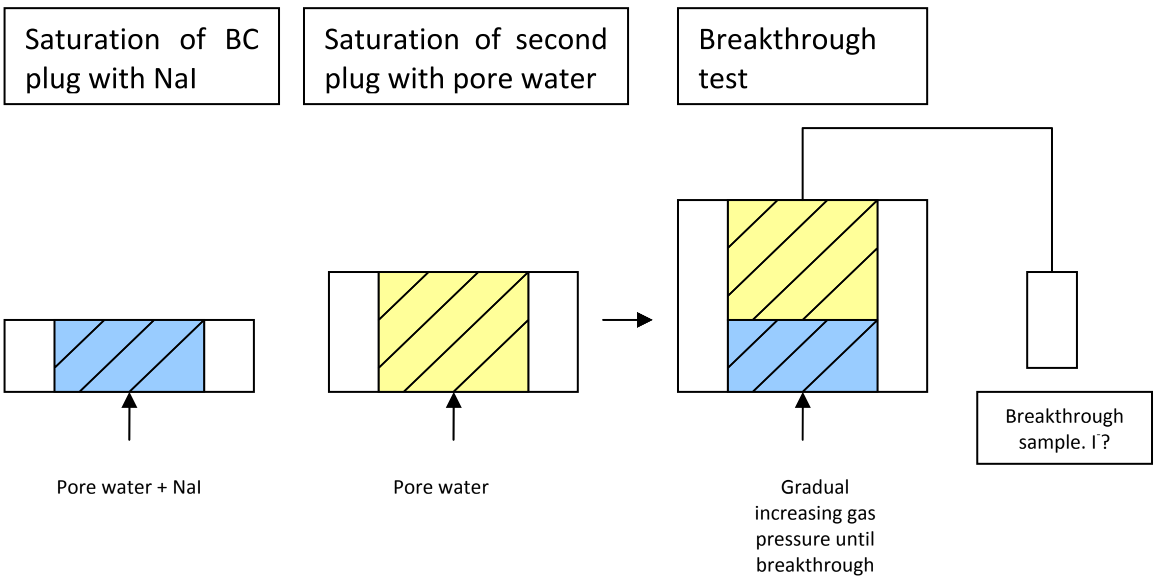


Figure 1: Basic concept

First, a small Boom Clay (BC) plug is saturated with an anionic tracer (iodide [I^-], 0.01 mol/l NaI) and a second, larger plug is saturated with natural pore water. For both, the hydraulic conductivity is determined.

After the plugs are completely saturated (constant hydraulic conductivity for at least 2 weeks), the plug re-saturated with natural pore water is put on top of the NaI-conditioned plug in a polycarbonate cell and confined (constant volume) between 2 porous stainless steel filter plates. The upper filter, and connected tubing, is filled with natural clay water (free of iodide).

Gas pressure is imposed at the bottom of the cell, and gradually increased until gas breakthrough occurs. When gas breakthrough occurs, water can be expelled. Therefore, the water at the outflow is analysed for the presence of iodine used as tracer.

After breakthrough, a µ-CT analysis will be performed to image the de-saturation front that might be generated as well as possible preferential gas pathways if they are large enough compared to the CT-resolution.

2.2 DESCRIPTION OF THE DIFFERENT COMPONENTS

2.2.1 The permeameter cell

For the preliminary test, an existing permeameter cell was used. This cell was already used in the FP6 TIMODAZ project (WP3.1: THM Characterisation) and was suitable to test the experimental procedure. After this first test, the design of this cell will be adapted to specific needs of this type of gas tests and new cells will be manufactured.

At the end of each test, µ-CT analyses will be conducted. As metal parts lead to artefacts in the µ-CT imaging process, metal components located close to the clay plug have to be avoided. For this reason, the cell is made from polycarbonate which is however less pressure resistant than, for example, stainless steel.

2.2.2 The pressure system

The system used to impose a constant gas pressure at the inlet of the cell is a “pressure controlling system buffered with mercury” (see figure 2). This system was developed in the frame work of the MEGAS EC project and is still operational (Volckaert et al., 1994).

The inlet of the system is connected with a pressure controller (DPI 520, Druck LTD) which imposes a constant pressure on an Hg filled vessel. This vessel is connected to a displacement transducer that has been calibrated so that displacement can be quantitatively converted in volume change and thus flow rate. The lower part of this displacement transducer is also filled with mercury; the upper part is filled with gas. This gas phase is connected to the outlet of the system which can be connected to the experiment.

The inlet and outlet of the system are at equal pressure. If the pressure at the outlet drops (for example due to a leak or gas breakthrough), the higher inlet pressure displaces the mercury and this displacement will be registered by the displacement transducer. As the pressure controller tries to keep the inlet at a constant level, gas is added to the system and keeps displacing the mercury until the displacement transducer is at its end. The use of this system avoids that in case of gas breakthrough gas keeps flowing until the gas bottle is completely consumed. Supplementary, the displacement transducer provides the evolution of the gas flow rate during the test.

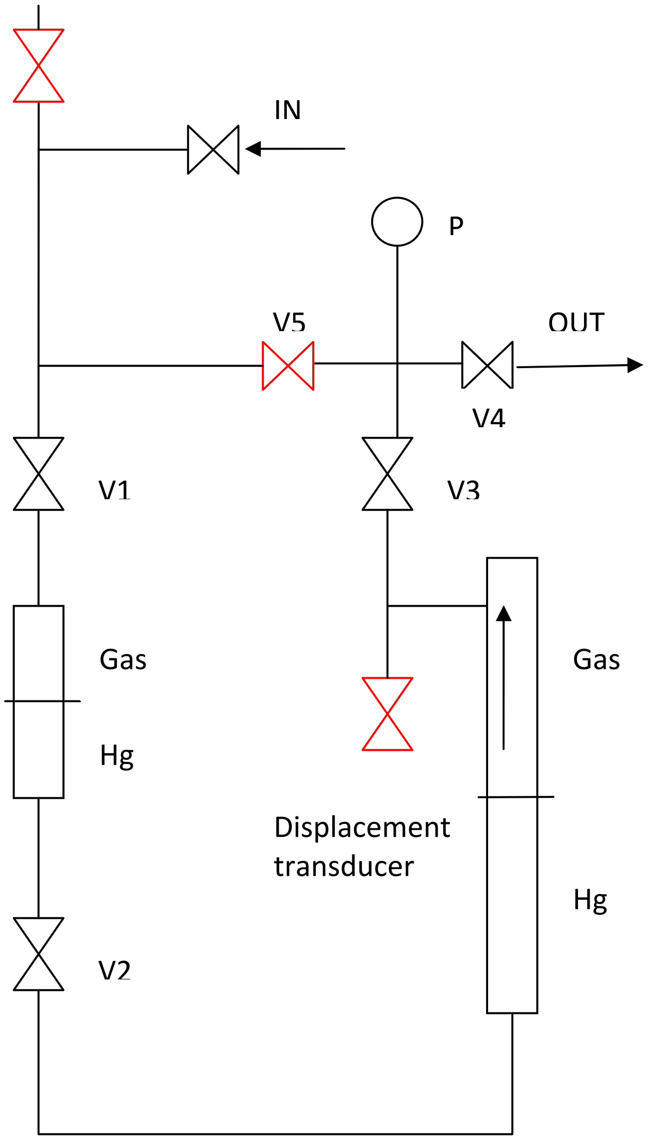


Figure 2: Pressure system

3 Procedure for experiments on gas-driven radionuclide transport

3.1 CONDITIONING OF THE CLAY CORES

3.1.1 Preparation of the clay cores and their installation in the cells

Two clay cores are prepared from a vertical stainless steel tube with a cutting edge.

The BC samples used for the experiments were cored in the HADES underground research facility (Mol, Belgium), perpendicular to the stratification, making use of stainless steel tubes with cutting edges. The cores were conserved in their original tubes that had been sealed and stored at 4°C to prevent dehydration and oxidation and to minimise microbial activity.

On the starting day of the experiment, the cores were extruded from their tubes with the aid of a hydraulic press.

The diameter of the clay core is larger than the internal diameter of the used cells (38mm). In order to prepare clay cores of the desired diameter that fit perfectly in the cells, a ring with a sharp edge (diameter 38mm) is mounted in front of the cell and with the aid of a press; a clay core is pushed onto this ring. The sharp edge will cut off the outer layer of the clay core. When the desired length is mounted into the cell, the core is cut by means of a knife.

Two cells are filled with clay cores respectively with a length of 10mm (for the saturation with an anionic tracer) and 14mm (for saturation with natural pore water)

3.1.2 Saturation of the cores and hydraulic conductivity measurement

To saturate the cores and measure their hydraulic conductivity, porous stainless steel filters are placed at the top and bottom of the plug and the cores are confined between the filter plates (i.e. constant volume), the lower part is connected with a vessel containing the saturating solution, the upper part is connected to a recipient.

To demonstrate the effect of a gas breakthrough on the transport of radionuclides an anionic tracer will be used.

Iodide, I^-, is chosen because its transport is not retarded under normal Boom Clay conditions: the sorption to clay minerals is negligible and it does not react with natural organic matter.

To obtain a very low detection limit, radioactive isotopes can be used, but this requires strict working conditions. Therefore our first test is performed with non-radioactive iodine in a concentration 1000x higher than the natural iodine concentration in pore water (5 10^{-6} mol/l) (De Craen et al, 2004)

A solution of 0.01 M NaI in natural pore water (Real Boom Clay water which is mainly a 0.01 M $NaHCO_3$ water type (De Craen et al, 2004) was prepared and transferred into a stainless steel vessel. A gas buffer of ca. 1 MPa argon was put on top of the solution, and the outlet of the vessel was connected to the clay core. The outlet of the cell is connected to a small bottle where the out-flow water is collected (see figure 3).

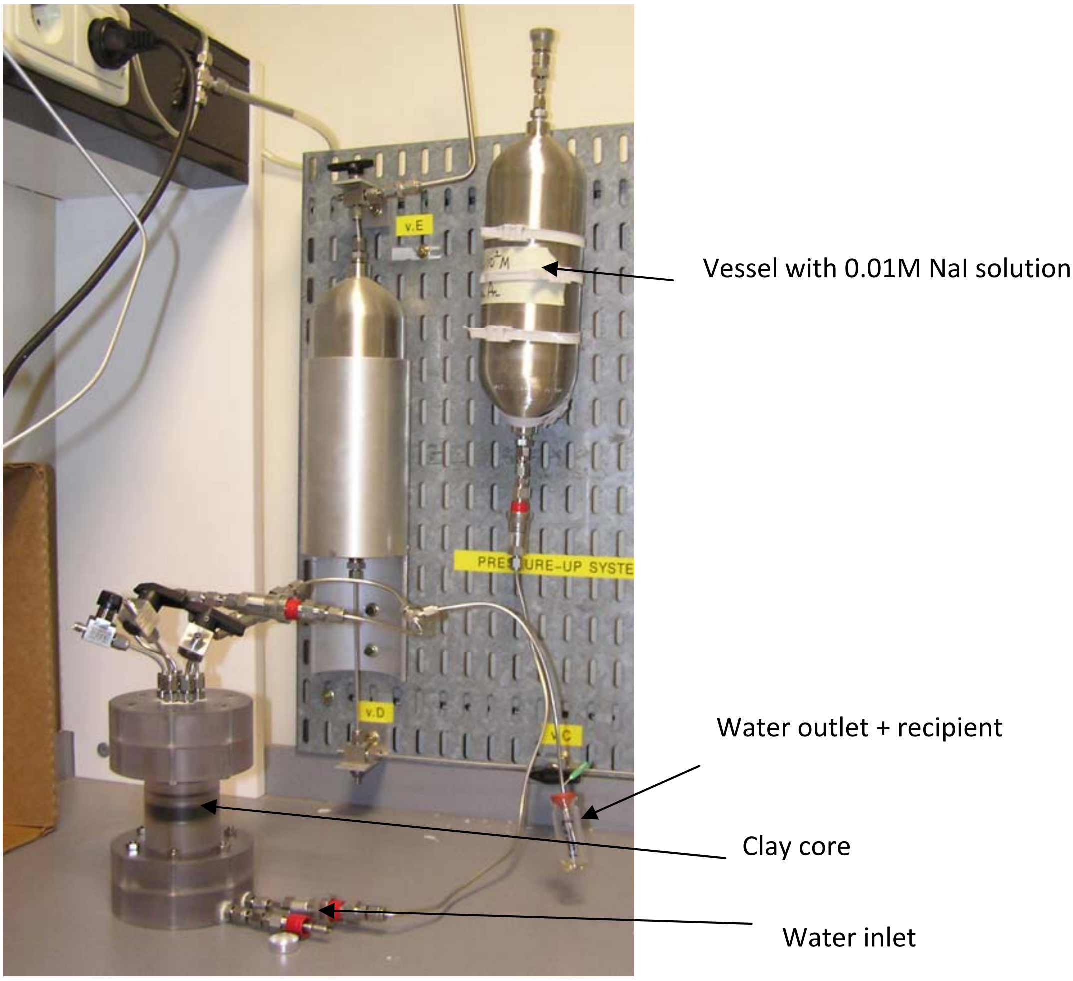


Figure 3: Saturation of the NaI clay core.

To follow-up the saturation process, water samples are taken on a regular base and for each sample the electric conductivity is measured. The electric conductivity of Real Boom Clay water (RBCW) is 1,7 mS/cm, those of the 0,01M NaI solution 2,77 mS/cm. The evolution of the electric conductivity of the flow-through water in time indicates the progress of the saturation process.

In addition to the electric conductivity measurement, samples of the flow-through water can be analysed for iodine by ICP-MS.

The second clay core is re-saturated similarly but with a solution of natural clay water.

For each clay core to be used, we determine the hydraulic conductivity K as “quality check”. Cores with deviating K values are not to be used for the experiments typical for undisturbed Boom Clay.

By registering the amount of water that passes through the clay core in a certain period of time, the hydraulic conductivity can be calculated according to Darcy’s law:

$Q = K*(dh/dl)*A$

With:

Q: flow rate (m^3/s)

K: hydraulic conductivity (m/s)

dh/dl: pressure gradient (in m water column/m)

A: surface (m²)

For Boom Clay, a hydraulic conductivity, orthogonal to bedding, between 1,3 and 2,4 10^{-12} m²/s is considered to be normal. *(De Craen et al, 2004)*

3.2 STARTING THE BREAKTHROUGH TEST

After the complete saturation of the clay cores and the determination of the hydraulic conductivity, the breakthrough test can be started.

The entire test is conducted in a temperature controlled room, at a constant temperature of 21°C (± 2°C).

Both cores are combined in one experimental cell ("Forge Cell"). The clay core saturated with RBCW is transferred on top of the NaI saturated cell. During the transfer of the cores, it is important that the walls of the Forge Cell are not yet contaminated with I^-.

After the cell is filled, the top part of the cell (filter and connected tubing) is filled with RBCW water, which will be expelled once gas breakthrough occurs. Analyses of this breakthrough water for iodine should indicate whether iodine was transported during gas breakthrough or not.

Finally, the "Forge" cell can be connected to the pressure system and the test is started.

To estimate the breakthrough pressure, following empirical equation based on K was used (Volckaert et al,, 1994):

$P_c = 4{,}71 * 10^{-4} * K^{-0{,}312}$ with

Pc: breakthrough pressure (MPa)

K: hydraulic conductivity (m/s)

(Volckaert et al., 1994)

The calculated breakthrough pressure for clay cores with normal K values ($1.3\ 10^{-12} < K < 2.4\ 10^{-12}$) is between 2.0 and 2.4 MPa.

When looking at the Megas results, the lowest breakthrough pressure for a vertical Boom Clay core was 1,1 MPa. As we do not expect to have lower breakthrough pressures (unless the calculations would indicate this), the experiment was started at 1,1 MPa.

To avoid that the results are disturbed by the diffusion of I^-, the test has to be finished within approximately 5 days. For this reason the following pressure schema was used:

Day 1: 1,1 MPa

Day 2: 1,5 MPa

Day 3: 1,9 MPa

Day 4: 2,1 MPa

Day 5: 2,3 MPa

4 Results

4.1 SATURATION AND HYDRAULIC CONDUCTIVITY

During the saturation process, water samples are taken on a regular base to calculate the flow rate and to follow-up the electric conductivity of the NaI core.

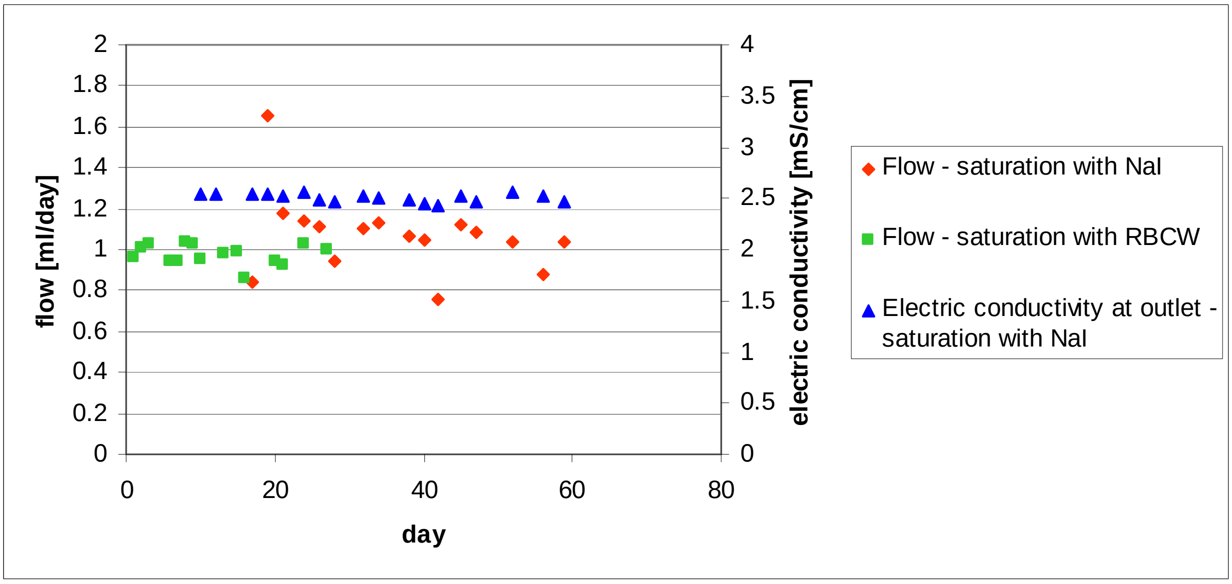


Figure 4: Saturation of the clay cores: follow-up of flow and electric conductivity

Table 1: Overview of measured and calculated parameters of the clay cores used

	Flow rate (m^3/s)	Δp (meter water column)	L (m)	K (m^2/s)	Pc (MPa)
NaI core	1,21E-12	82,3	0,010	1,29 10^{-12}	2,41
RBCW core	1,13E-12	69	0,014	2,02 10^{-12}	2,10

Compared to the reference hydraulic conductivity values for Boom Clay, both clay cores are considered to be “normal” and can be used for the experiment (see table 1).

Ten days after the start of the saturation process with NaI, the first flow-through water sample was taken and measured. The samples that were taken on day 17 and 59 were analysed for iodine with ICP-MS.

Table 2: electric conductivity and concentration [I] for different flow-through samples

Sample	Electric conductivity [mS/cm]	I [mol/l]
Sample day 10	2,549	-
Sample day 17	2,546	8,67 10^{-3}
Sample day 59	2,468	9,22 10^{-3}

NaI solution	2,770	$10\ 10^{-3}$
RBCW	1,7	-

Already after 10 days, the clay core was well saturated with NaI. This was confirmed with the iodine-analyses at day 17 and 59 (see table 2).

4.2 GAS BREAKTHROUGH

According to the calculated Pc, the proposed pressure-schema (see 3.2) should be feasible.

After the cell was filled with both plugs and prepared for the test, the set-up was left unpressurised to let the cores "equilibrate" in the cell.

Finally the test could be started with an initial He gas pressure of 1,1 MPa.

Table 3: Overview of events during the breakthrough experiment

Time	Pressure	Observation
Day 1	1,1 MPa	nothing special was observed
Day 2	1,5 MPa	After a few hours, a little amount of gas was accumulating at the bottom of the cell
Day 3	1,9 MPa	After a few minutes, more and more gas started to accumulate along the wall of the cell and also at the interface between the 2 clay cores. Only 30 minutes later, gas breakthrough occurred. Due to this gas breakthrough, the water present at the top of the cell was expelled. This water was collected and analysed for iodine by ICP-MS analyses.

Visual inspection of the cell showed 2 damaged zones. Probably, this was caused by a small piece of pyrite inclusion which damaged the clay core wall and/or the cell during transfer of the core in the cell. Due to this damage, a preferential path was created that allowed gas breakthrough along the clay-cell interface at a lower than expected pressure. (See figures 5 and 6)

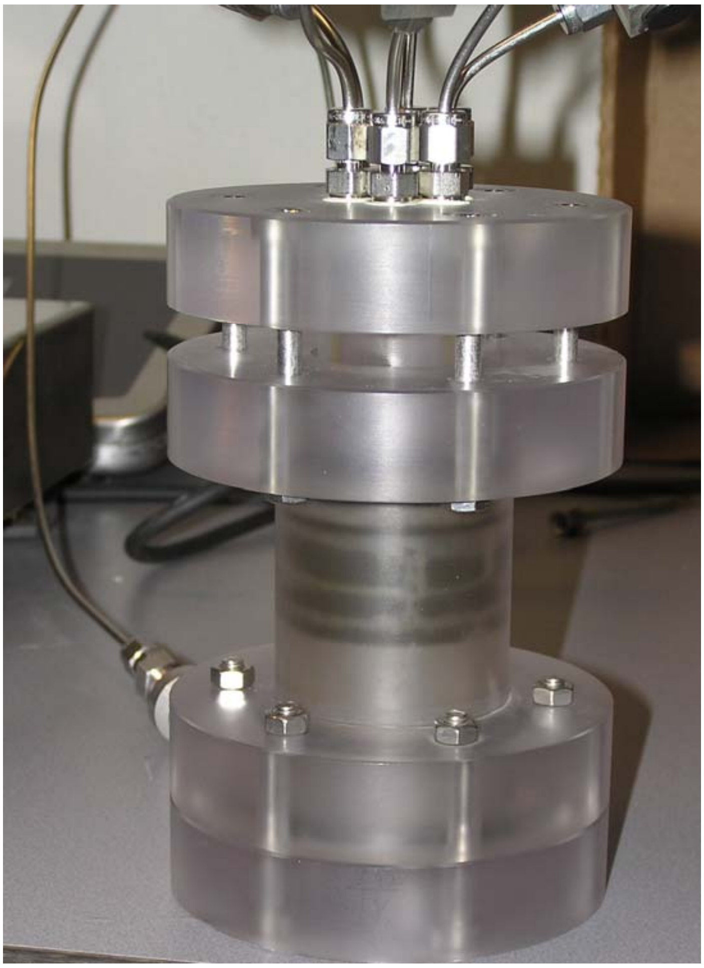
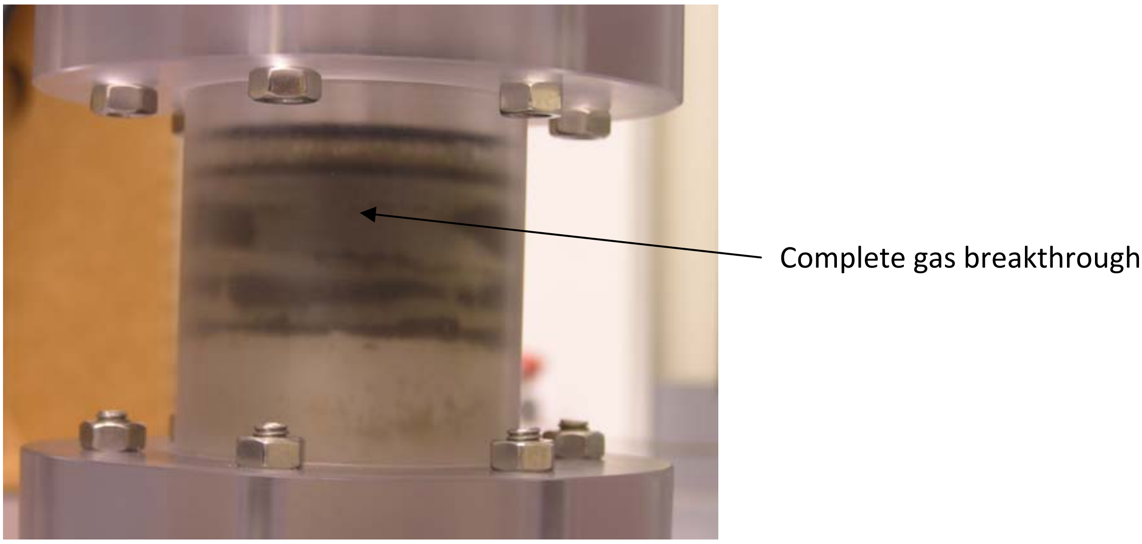


Figure 5: Set-up right after gas breakthrough



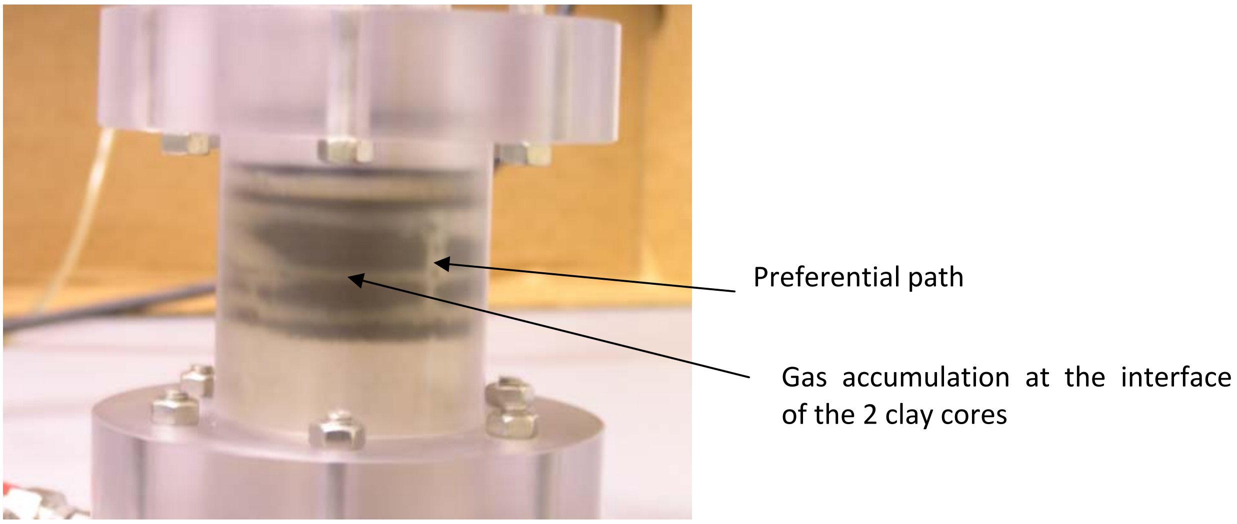


Figure 6: details of the cell after gas breakthrough

Two days after gas breakthrough, the clay cores were pushed out of the cell. Visual inspection revealed – as expected – a little piece of pyrite which damaged the clay core and the wall of the cell (see figure 7).

Figure 8 reveals another damaged zone where especially the upper plug is deformed. This deformation is certainly a preferential path.

The grey inclusions are probably carbonate-inclusions.

The horizontal cracks are a consequence of the drying of the clay during 2 days.

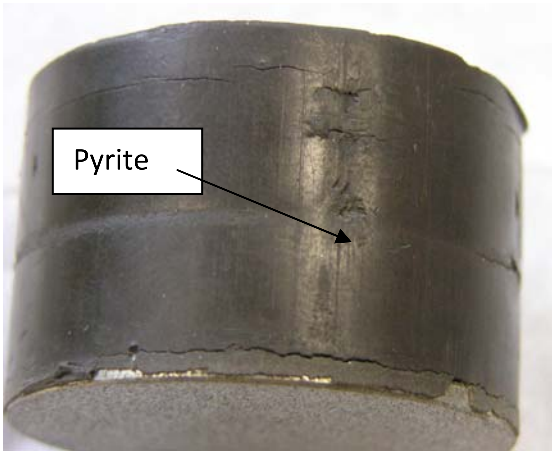


Figure 7: damaged clay core: pyrite inclusion

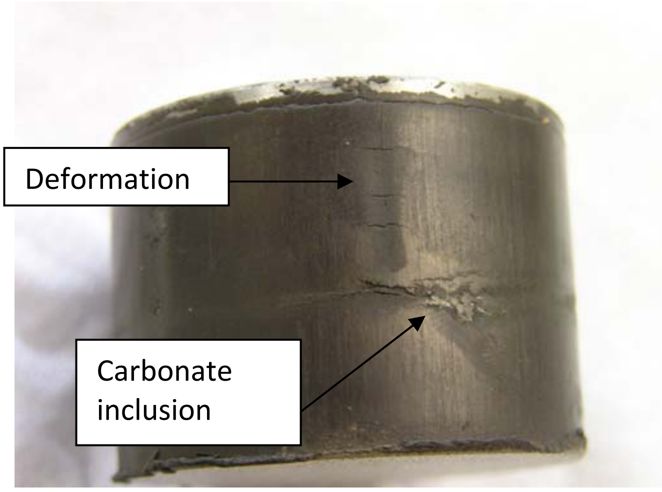


Figure 8, deformations and a carbonate inclusion.

Analyses of the different water sample gave following results:

Table 4: analyses of the different water samples

Sample	Electric conductivity [mS/cm]	I [mol/l]
Breakthrough sample	1,838	$8{,}04\ 10^{-5}$
RBCW core outlet sample	-	$6{,}22\ 10^{-6}$
RBCW core inlet sample	-	$6{,}46\ 10^{-6}$
NaI core outlet sample	2,468	$9{,}22\ 10^{-3}$
NaI core inlet sample	2,770	$10{,}0\ 10^{-3}$

The concentration iodine in the breakthrough sample is higher than the concentration in the RBCW water used to fill the upper part of the cell.

According to these data, a volume of 37 µl NaI water was transported along the wall of the vessel.

4.3 µ-CT ANALYSES

As the gas breakthrough occurred via the interface clay-vessel, no µ-CT analyses were performed.

5 Conclusion

The aim of this first experiment was to check whether the concept is feasible and if necessary to optimise the set-up.

Regarding the results of the breakthrough test, NaI containing water was transported during gas breakthrough and this could be detected by ICP-MS analyse of the breakthrough water. So for the time being, it is our evaluation that it is not necessary to use radioactive tracers in order to obtain a lower detection limit.

Unfortunately gas breakthrough occurred along the wall of the cell due to different causes:

- Damage of the plug/wall due to the presence of pyrite inclusions causing a preferential path at the interface
- Deformation of the polycarbonate cell at high pressure

To avoid these problems in the future experiments, following improvements will be made:

- The diameter of the inlet filter will be smaller than the core diameter so that gas is injected only at the centre of the clay core, and not at the wall.
- Longer RBCW-cores will be used. This can limit the effect of inclusions and other damages.
- To limit the deformation of the cell, a metal supporting ring will be foreseen.
- To increase the tightness between the clay core and the walls of the cell, these walls will be rubbed in with high viscosity grease.
- When closing the cell, it will be placed under a hydraulic press to remove eventually present air inclusions.

All these improvements led to an optimised cell design:

- The body of the cell is enlarged so that clay cores with a total maximum height of 55mm can be used. In future experiment, a clay plug of 10mm height will be used for saturation with NaI, and a plug of 30mm will be used for saturation with RBCW.
- During the EC-TIMODAZ tests, many cells showed shears where the metal screws were screwed into the polycarbonate. To avoid this shearing, the cells were adapted and metal plates are foreseen at the top and bottom. They are placed at such a distance from the core that they do not disturb CT-imaging. The use of these metal plates changes of course the whole closing system of the cell.
- The diameter of the inlet filter decreased from 38mm tot 15mm.
- A supporting metal ring is foreseen, to limit the deformation of the polycarbonate at high pressure. This ring can be removed when the cell is scanned with μ-CT.
- 1 inlet is foreseen at the bottom and at the top 2 outlets are foreseen.

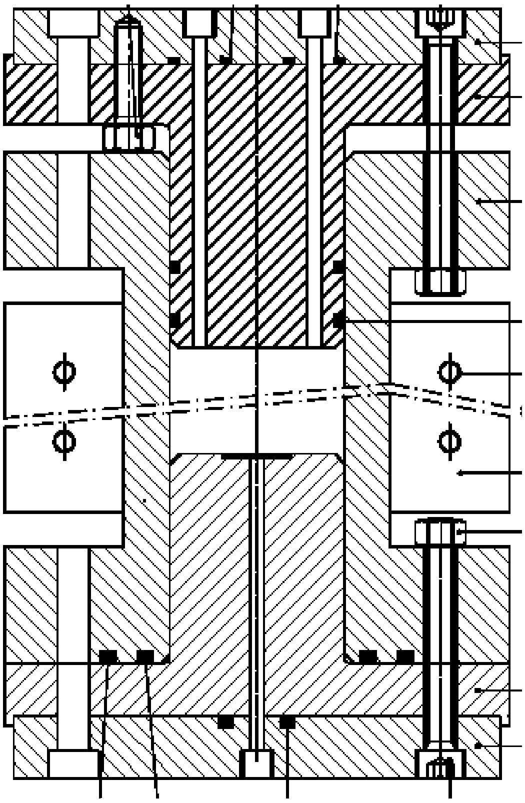


Figure 9: Final improved design of the Forge Cell

During the first WP5 kick-off meeting, it was suggested to do these tests in double: one with gas pressure and one with water pressure. In this way, it should be more clear which effects are gas-related and which are not.

References

De Craen M., Wang L., Van Geet M., Moors H. (2004) *"Geochemistry of Boom Clay pore water at the Mol site"* SCK•CEN Report (BLG 990)

Volckaert G., Ortiz L., De Cannière P., Put M., Horseman S.T., Harrington J.F., Fioravante V. Impey M. (1994) "*MEGAS Modelling and Experiments on Gas Migration in Repository Host Rocks. Final Report Phase 1*". EUR16235 EN, Luxembourg.