FORGE

Fate Of Repository Gases

European Commission FP7

Results and interpretation of gas-driven radionuclide transport in undisturbed Boom Clay

FORGE Report D5.8 - SCK-CEN ER-158 – VER.0

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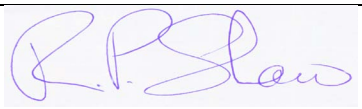
Approved	RP Shaw	BGS		18th July 2011

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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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Foreword

This report describes the results of the experiments performed by SCK•CEN in the framework of WP5.1: Undisturbed Host Rock Formations – Gas transport laboratory experiments. The goal of this research was to answer the question: "To what extent can a gas pressure build-up enhance the transport of radionuclides?".

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During gas breakthrough, some 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.

The work described in this report 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, gas breakthrough tests were performed on undisturbed Boom Clay cores. A permeameter cell was filled with 2 clay cores, one of them was infiltrated with porewater that is enriched with an anionic tracer (iodide). Gas pressure was applied at the side of the tracer infiltrated clay core until gas breakthrough occurred. The water expelled during breakthrough was tested on it's tracer content.

As iodide is an un-retarded tracer, its diffusion cannot be neglected when experiments last longer than 4 days because from that moment on the concentration of diffused I would be higher than the iodide background concentration in natural pore water.

Based on the amount of Iodide measured in the breakthrough sample, we can state that maximum 1% of the contaminated solution can be transported when gas breakthrough occurs. This is a conservative value, as we did not take into account the diffusion of iodide through the clay core

In general, breakthrough pressures were rather high and they were reached after only 4 – 5 days. By this time, the I has diffused through the clay and strongly influences the iodide concentration in the breakthrough sample. In order to have more solid proof of gas induced tracer transport more tests are needed. For that purpose, it might be more interesting to focus on tests on disturbed (fissured) Boom Clay, where we expect gas breakthrough at lower BT pressures and thus faster than the diffusive tracer breakthrough. Experiments of disturbed BC will be performed within WP4.

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 can 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 capacity for diffusive transport of dissolved gas, 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 is 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, some 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 performed experiments with permeameter cells using undisturbed Boom Clay. After infiltration of a clay core with porewater that contains an anionic tracer (I^-), a gas breakthrough test was performed to investigate gas induced transport of this tracer.

This report presents the result of the breakthrough tests on undisturbed Boom Clay.

2. Materials & methods

2.1 THE CONCEPT

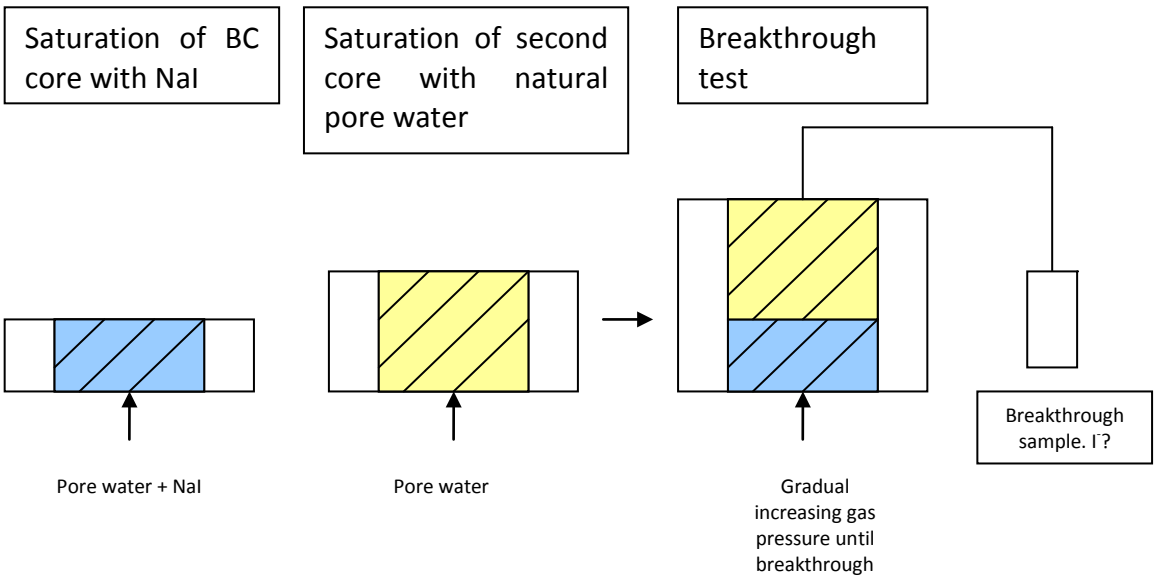

Figure 1: Basic concept

First, a small Boom Clay (BC) core is saturated with an anionic tracer (iodide [I^-], 0.01 mol/l NaI in Boom Clay natural pore water) and a second, larger core is saturated with natural pore water. Natural pore water (Real Boom Clay water – RBCW) is mainly 0.014 mol/l $NaHCO_3$ type of water.

As we don't want to disturb the electrical double layers from the clay, we used a NaI concentration of similar ionic strength as the natural pore water.

After the cores are completely saturated, the core resaturated with natural pore water is put on top of the NaI-conditioned core in a polycarbonate permeameter cell and confined (constant volume) between 2 porous stainless steel filters. The upper filter, and connected tubing, is filled with natural clay water which has a low background concentration of iodide ~5 10^{-6} mol/l (De Craen et al, 2004).

Gas pressure (helium) is imposed at the bottom of the cell, and stepwise increased until gas breakthrough occurs. When gas breakthrough occurs, water can be expelled. Therefore, the water at the outflow, expelled after gas breakthrough, is analysed (with ICP-MS) for iodine enrichment.

2.2 MATERIALS

2.2.1 The permeameter cell

A preliminary test was performed with a permeameter cell developed for the FP6 TIMODAZ project (WP 3.1: THM Characterization). Based on the lessons learned from this preliminary test, the design of the cell was optimised and new cells were constructed. The new cells are higher to allow the use of thicker undisturbed Boom Clay cores, the diameter of the gas inlet-filter was decreased to avoid breakthrough along the wall and a metal supporting ring is foreseen to avoid deformation of the polycarbonate permeameter cell (see figure 2 and figure 3) (Jacops et al., 2009).

The entire set-up was described in detail in FORGE report D5.1 (Jacops et al., 2009)

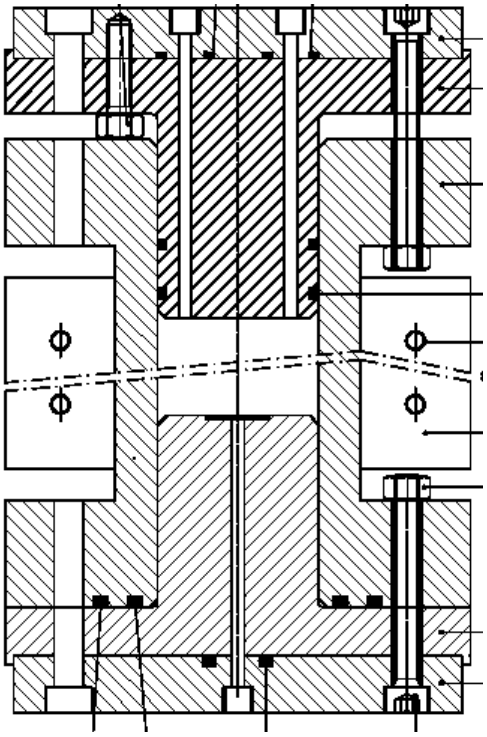

Figure 2: Drawing of the improved FORGE permeameter cell

Figure 3: Image of the new permeameter cell used for gas breakthrough experiments in the FORGE project

The Boom Clay 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 a cutting edge. 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.

2.2.2 The pressure system

A constant gas pressure is imposed at the inlet of the cell with a "pressure controlling volume measurement system" (see figure 4). This system was developed in the framework of the MEGAS EC project and is still operational (Volckaert et al., 1994).

The inlet of the system is connected with a gas bottle which pressurizes a mercury filled vessel. This vessel is connected to a calibrated cylindrical vessel. The lower part of this calibrated cylindrical vessel is also filled with mercury, the upper part is filled with gas. The calibrated cylindrical vessel is equipped with a displacement transducer which measures the mercury level. This cylindrical vessel is calibrated in such a way that by measurement of the mercury level, the change in the gas volume can be measured precisely and thus can be converted to the flow rate. The upper gas phase is connected to the outlet of the system which can be connected to the experimental set-up (Jacops et al., 2009).

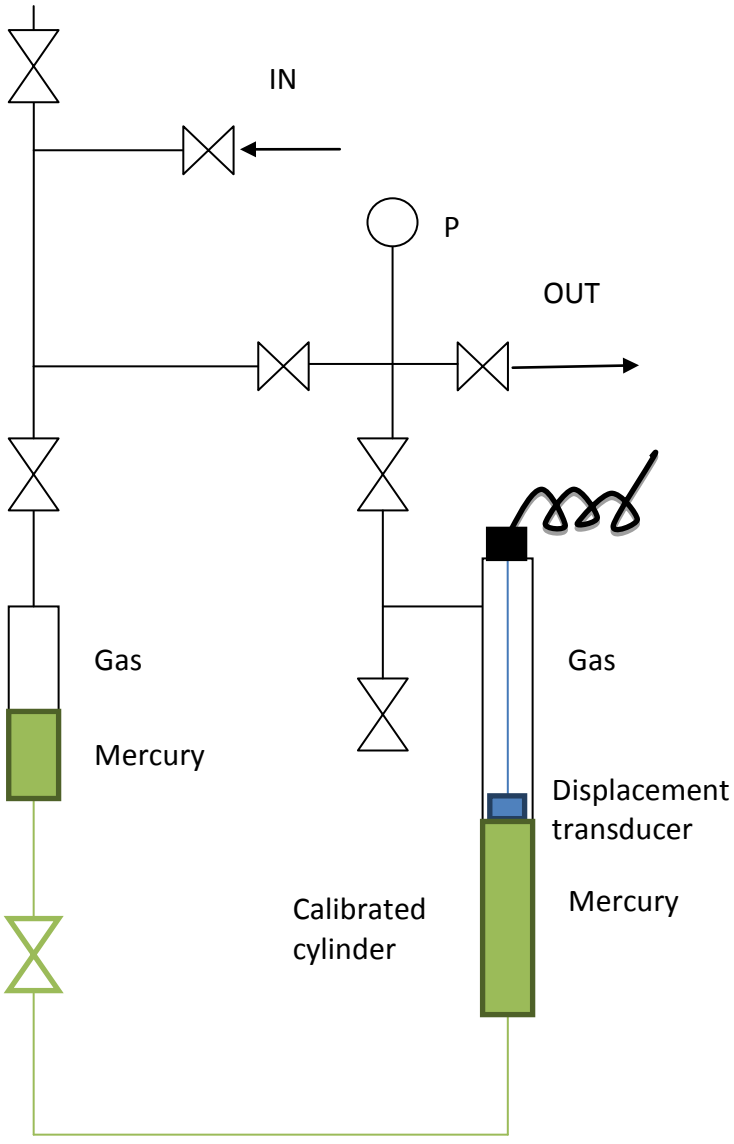

Figure 4: Gas injection system

2.3 METHODS

2.3.1 Saturation of the cores and hydraulic conductivity measurement

Prior to their use in a gas breakthrough test, cores are saturated and their hydraulic conductivity is measured.

To demonstrate the effect of a gas breakthrough on the transport of radionuclides, an anionic tracer is 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.

A small Boom Clay core of 10 mm high is saturated with a solution of 0.01 mol/l NaI in natural pore water. This iodine concentration is more than 1000x higher than the natural iodine concentration in pore water (~5 10^{-6} mol/l). (De Craen et al, 2004)

The second clay core of 37mm high is resaturated 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 a clay core of known dimensions in a certain period of time and at a known pressure gradient, the hydraulic conductivity can be calculated according to Darcy's law:

$$Q = K*(dh/dl)*A \quad (1)$$

With:

Q: flow rate (m^3/s)

K: hydraulic conductivity (m/s)

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

A: surface (m^2)

For Boom Clay, hydraulic conductivity has been measured during many years on different samples, as well in the lab as in-situ. The hydraulic conductivity vertical to the bedding plane is typically in the range 1 10^{-12} till 5-10^{-12} m/s (Yu et al., 2011).

2.3.2 Starting the breakthrough test

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

First the breakthrough pressure was estimated, using following empirical equation based on the hydraulic conductivity (Volckaert et al., 1994):

$$Pc = 4.71*10^{-4} *K^{-0.312} \quad (2)$$

with

P_c: breakthrough pressure (MPa)

K: hydraulic conductivity (m/s)

Both cores are combined in one experimental cell ("Forge Cell"). The clay core saturated with natural pore water (RBCW) is placed on top of the NaI saturated clay core and both cores are loaded into the Forge Cell. During the transfer of the cores, it is important that the walls of the Forge Cell do not become contaminated with I^-.

After the cell is filled, the upper part of the cell (filter and connected tubing) is filled with natural clay water, which will be expelled once gas breakthrough occurs. An enrichment in Iodine concentration after breakthrough can be seen as an indicator for gas driven tracer transport.

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

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

3. Results

3.1 TEST 2010/1

Table 1: Parameters for test 2010/1

	NaI core	RBCW core
Length (mm)	9.7	36.5
K (m/s)	$1.28\ 10^{-12}$	$1.20\ 10^{-12}$
Pc (MPa)	2.42	2.47

Table 2 Pressure evolution during test 2010/1

Days since start of experiment	Applied gas pressure MPa
0.0	1.0
0.8	1.4
1.7	1.8
2.7	2.0
3.7	2.2
3.8	2.4
3.9	2.6

Breakthrough occurred at 2.6 MPa (see table 2), a pressure only slightly higher than the calculated breakthrough pressure Pc (see table 1). The expelled water was sampled and sent for analyses (table 3). For this experiment, the upper part of the cell was filled with synthetic clay water (SBCW) instead of natural clay water (so no natural Iodine was present).

During dismantling we observed that it was hard to remove the stainless steel ring from the polycarbonate cell. Probably the polycarbonate deformed due to the high pressure. After removing the stainless steel ring, we observed gas at the interface clay-polycarbonate. This could be caused by a breakthrough along the wall, or by relaxation of the polycarbonate after removal of the steel ring.

Table 3: Measured concentration I (mol/l) in the breakthrough sample of test 2010/1

	Concentration I (mol/l)
Breakthrough sample (3.9 days)	$3.59 \quad 10^{-6}$

3.2 TEST 2010/2

Table 4: Parameters for test 2010/2

	NaI core	RBCW core
Length (mm)	12	37
K (m/s)	$1.42\ 10^{-12}$	$1.32\ 10^{-12}$
Pc (MPa)	2.34	2.40

Table 5: Pressure evolution during test 2010/2

Days since start pressure build-up	Pressure (MPa)
0.00	0.9
0.77	1.4
1.00	1.8
1.78	2.0
2.83	2.2
3.76	2.4
3.87	2.5
4.01	2.6
4.08	2.7
4.10	2.8
4.11	2.9

4.13	3.0
4.13	3.2
4.13	3.4
4.14	3.8
7.72	3.9
7.86	4.0
7.98	4.2
8.06	4.2
BT at 16.3 days	

Breakthrough occurred after 16.3 days at 4.2 MPa (see table 5); a pressure much higher than the calculated breakthrough pressure Pc (see table 4). The expelled water was sampled and sent for I-analyses (see table 6).

During dismantling, we observed gas at the interface clay-polycarbonate cell, but there was no clear preferential path along the wall.

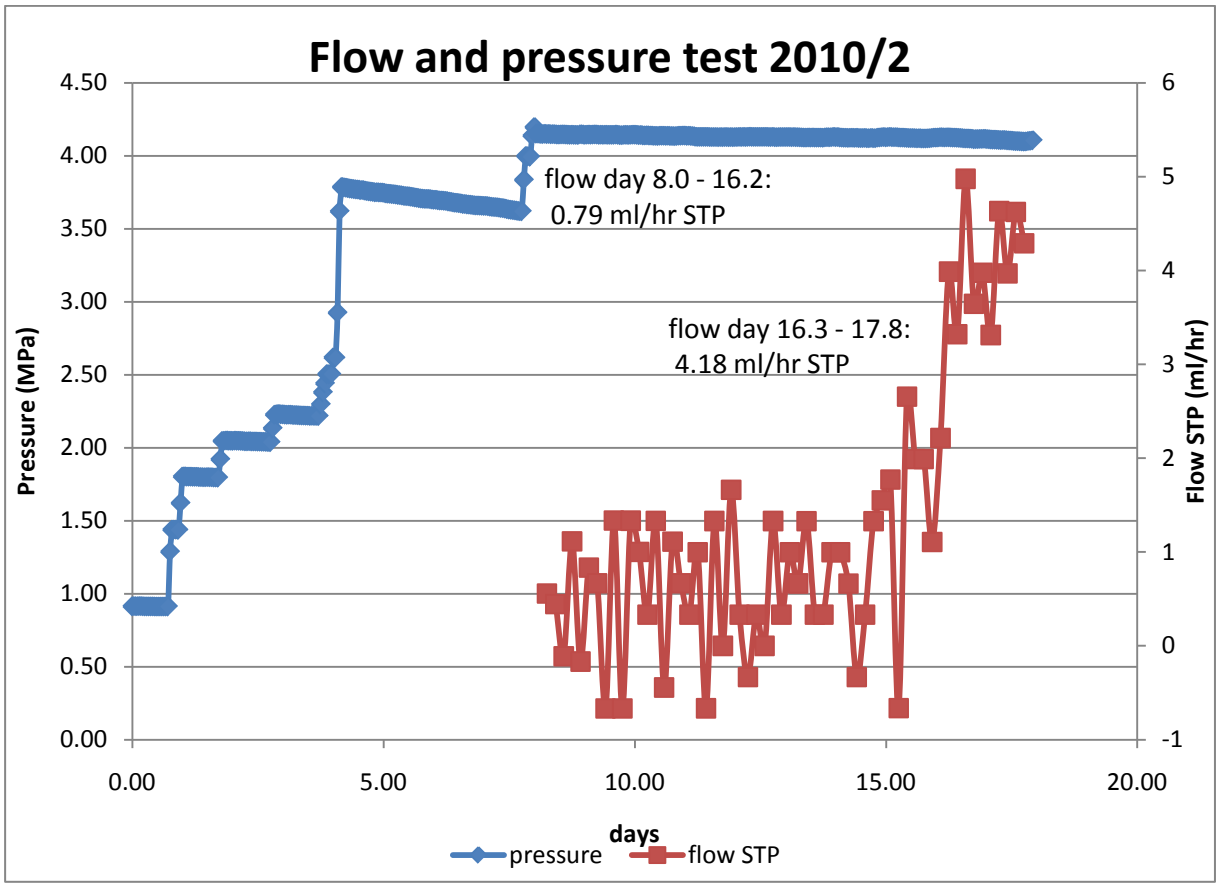

Figure 5: Flow and pressure during test 2010/2

Before breakthrough, a flow rate of 0.79 ml/hr STP was measured. After breakthrough, the flow rate was significantly higher: 4.18 ml/hr STP (see figure 5).

Table 6: Measured concentration I (mol/l) in the breakthrough sample of test 2010/2

	Concentration I (mol/l)
Breakthrough sample (16.3 days)	$225\ 10^{-6}$

3.3 TEST 2010/3

Table 7: Parameters for test 2010/3

	NaI core	RBCW core
Length (mm)	13.5	39
K (m/s)	$1.61\ 10^{-12}$	$1.31\ 10^{-12}$
Pc (MPa)	2.25	2.40

Table 8: Pressure evolution during test 2010/3

Days since start	Pressure (MPa)
0.00	1.10
0.81	1.44
1.11	1.83
2.02	2.21
3.01	2.41
3.88	2.64
3.96	2.83
4.06	3.03
4.10	3.20
4.13	3.58
4.15	3.83
4.16	4.01
4.18	4.21
42.92	4.11
42.92	4.43
43.07	4.58
43.11	4.81

After 4 days and a pressure of 4.2 MPa, no breakthrough happened. We decided to leave the experiment under pressure and wait for breakthrough. However, after 43 days we decided to increase pressure again and breakthrough occurred at a pressure of 4.81 MPa after 43.11 days (see table8).

The breakthrough pressure was much higher than the calculated breakthrough pressure Pc (see table 7).

After breakthrough the set-up was left under pressure for 1.5 day and the gas flow was recorded. On day 45 pressure was increased again to 4.81 MPa and again gas flow was recorded. Finally on day 46 we increased pressure till 5.0 MPa to evaluate the flow.

To measure the shut-in pressure, the valve of the pressure system was closed and we followed the pressure decrease.

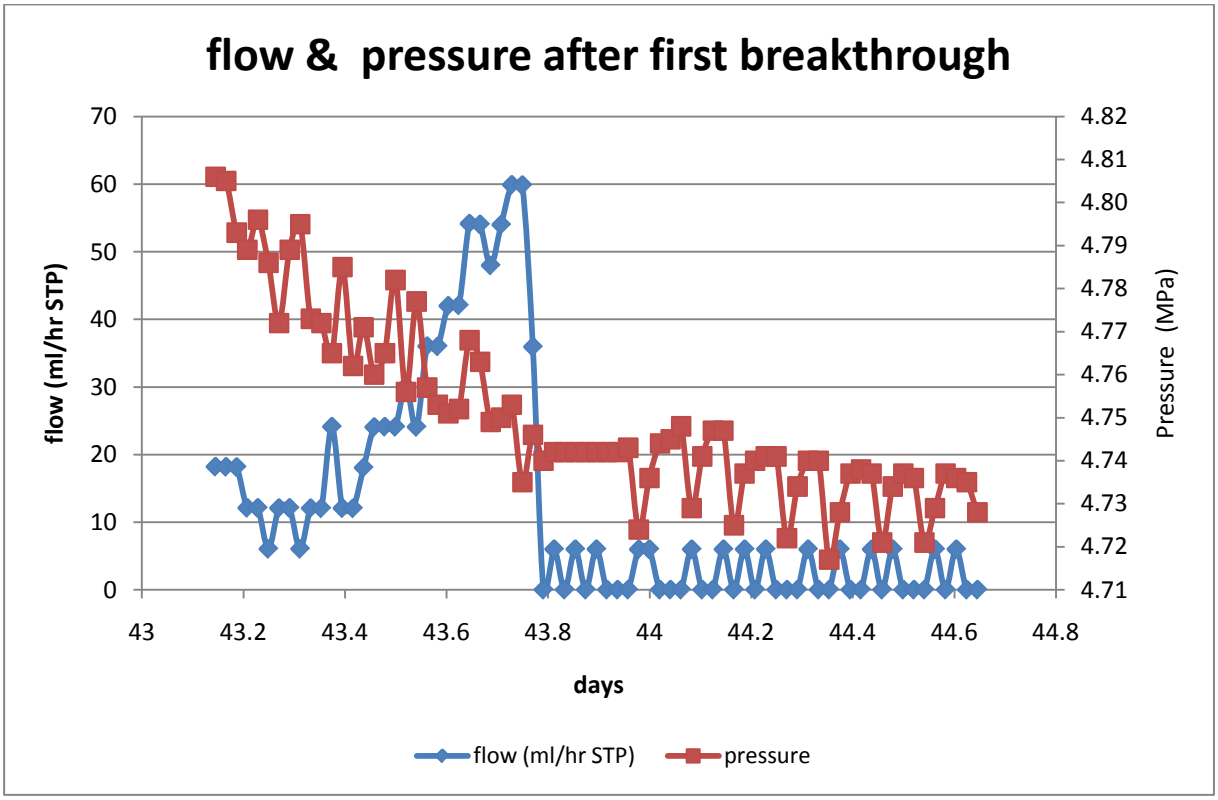

Figure 6: Flow and pressure after the first breakthrough of test 2010/3

After the first breakthrough, flow gradually increased until a peak flow of 60 ml/hr STP was reached, and then flow suddenly dropped to 0 (see figure 6). After the second and third breakthrough, gasflow was rather constant with values of respectively 3.37 ml/hr STP and 2.68 ml/hr STP (see table 9).

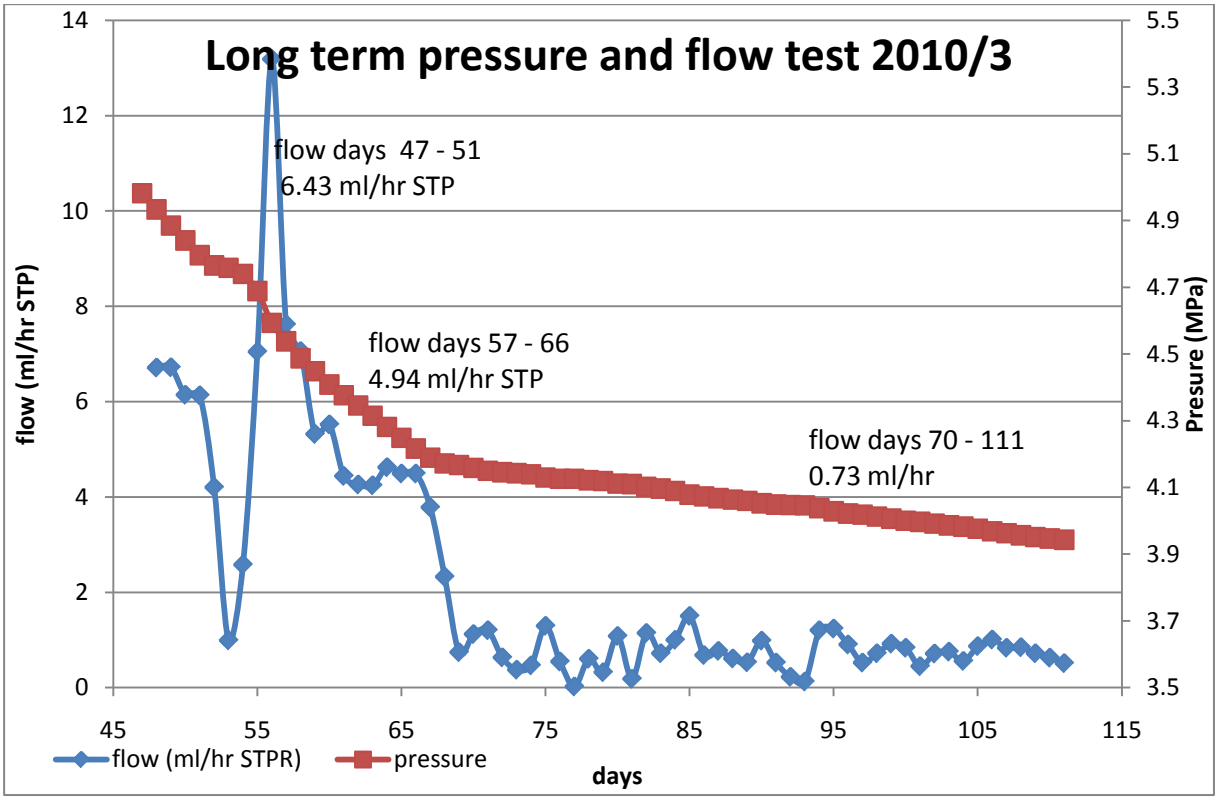

Figure 7: Long term pressure and flow evolution of test 2010/3

Table 9: Overview of flow during different stages of test 2010/3

Period	Flow (ml/hr STP)	Remarks
Day 5- 28	0.40	
Day 33 - 42	0.45	
1st breakthrough	60	Peak value
2nd breakthrough	3.37	
3rd breakthrough	2.68	
Day 47 – 51	6.43	
Day 56	13.2	Peak value
Day 57 - 66	4.94	
Day 70 - 111	0.73	

From day 46 on, pressure was followed to determine the shut-in pressure. On day 51, the gas flow suddenly stopped and restarted on day 54. A peak flow was reached on day 56, and from day 57 on the flow stabilised again. However, this flow is lower than the flow in the period 47-51 days.

From day 70 on, flow decreased to a steady-state value of 0.73 ml/hr STP. Probably the shut-in pressure was reached (4.16 MPa).

The experiment was stopped on day 112. (see figure 7 and table 9)

Table 10: Measured concentration I (mol/l) in the outlet sample of test 2010/3

	Concentration I (mol/l)
Sample after 8 days	$13.2\ 10^{-6}$

The water at the outlet of the cell was sampled after 8 days and sent for analyses. Analysis of this sample shows an iodine concentration $1.32\ 10^{-5}$ mol/l (see table 10) which is higher than the iodine concentration in natural clay water ($5\ 10^{-6}$ mol/l) (De Craen et al., 2004).

The water after breakthrough was sampled but not analysed as breakthrough occurred on day 60 and the Iodine in the water is mainly due to diffusion.

Because the samples were under quite high gas pressure for a considerable long time, desaturation might occur at the inlet. Therefore, immediately after stopping the experiment we dismantled the set-up and the clay was cut into slices of 1 mm. The slices were dried at 65°C for 2.5 days and the water content was calculated (see figure 8).

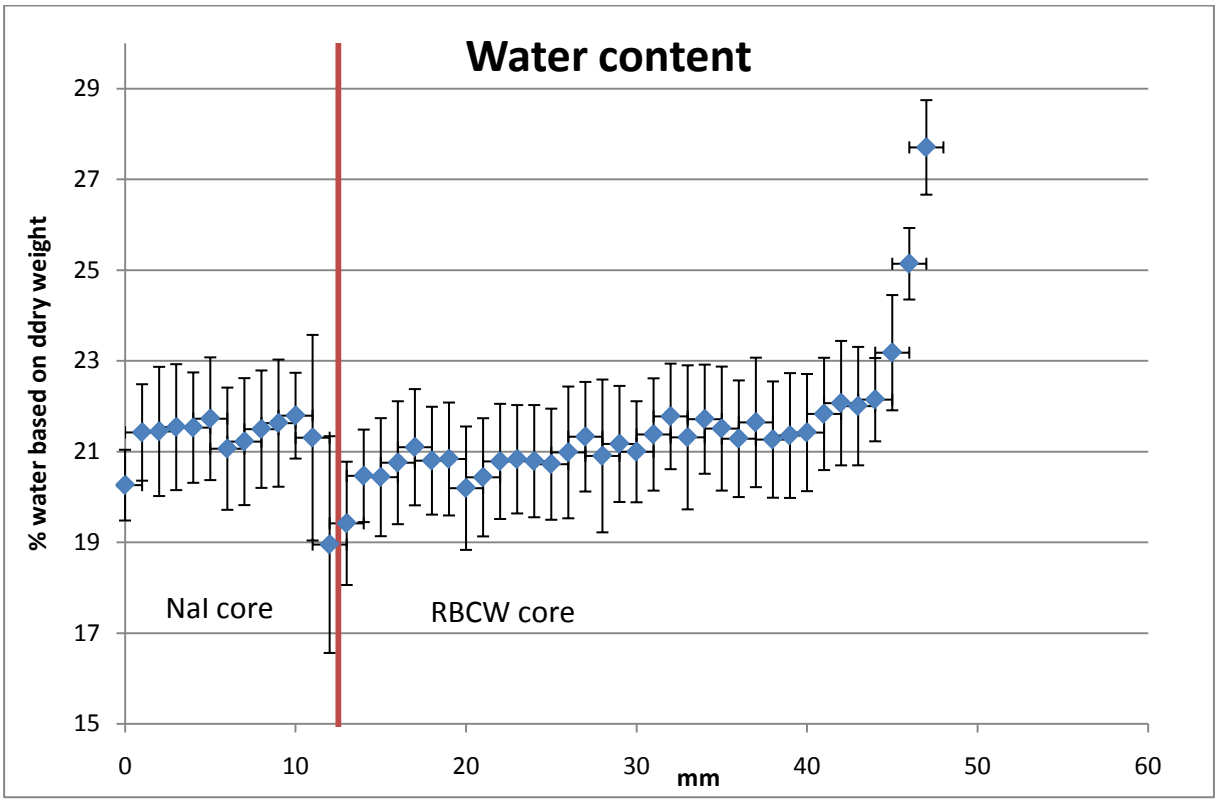

Figure 8: Water content of the slices

For the NaI-saturated core, the average water content is 21.4%. For the first slice at the interface gas inlet/filter/NaI Clay core, a lower water content is observed. However, also the last slice has a significantly lower water content: 19%. An equal situation is observed for the RBCW-core: the average water content is also 21.4%, but the first slice has a lower water content: 19.4% (see figure 8).

When dismantling the set-up, we observed an accumulation of gas at the interface of both cores (red line in the graph). Maybe, the accumulated gas did slightly dry out the clay at the interface. At the upper side of the clay core at the interface clay core/water saturated filter, the water content increased because of the contact with a water phase.

During dismantling, we also observed that both cores were not "sealed". In previous tests, both cores had been in close contact due to the exposure to a high pressure and after dismantling it was not possible to separate them. As this phenomenon can be compared to the sealing of fractured clay, we say that both cores are "sealed".

Because the cores of this experiment were not sealed, probably gas started accumulating at the interface between both cores already early in the test, avoiding good contact between both cores and causing the drying-out.

4. Discussion

The unretarded diffusion of iodide cannot be neglected when experiments last longer than 4 days. Transport calculations have shown that already after 3.5 days the concentration of diffused iodide is at the same level as the natural iodide concentration in Boom Clay water (5 10^{-6} mol/l – red line in graph) (Figure 9). So breakthrough should occur within 4 days to allow a determination of gas induced water displacement. In this case, only a water displacement 0.002 ml is sufficient to double the I concentration in the pore water.

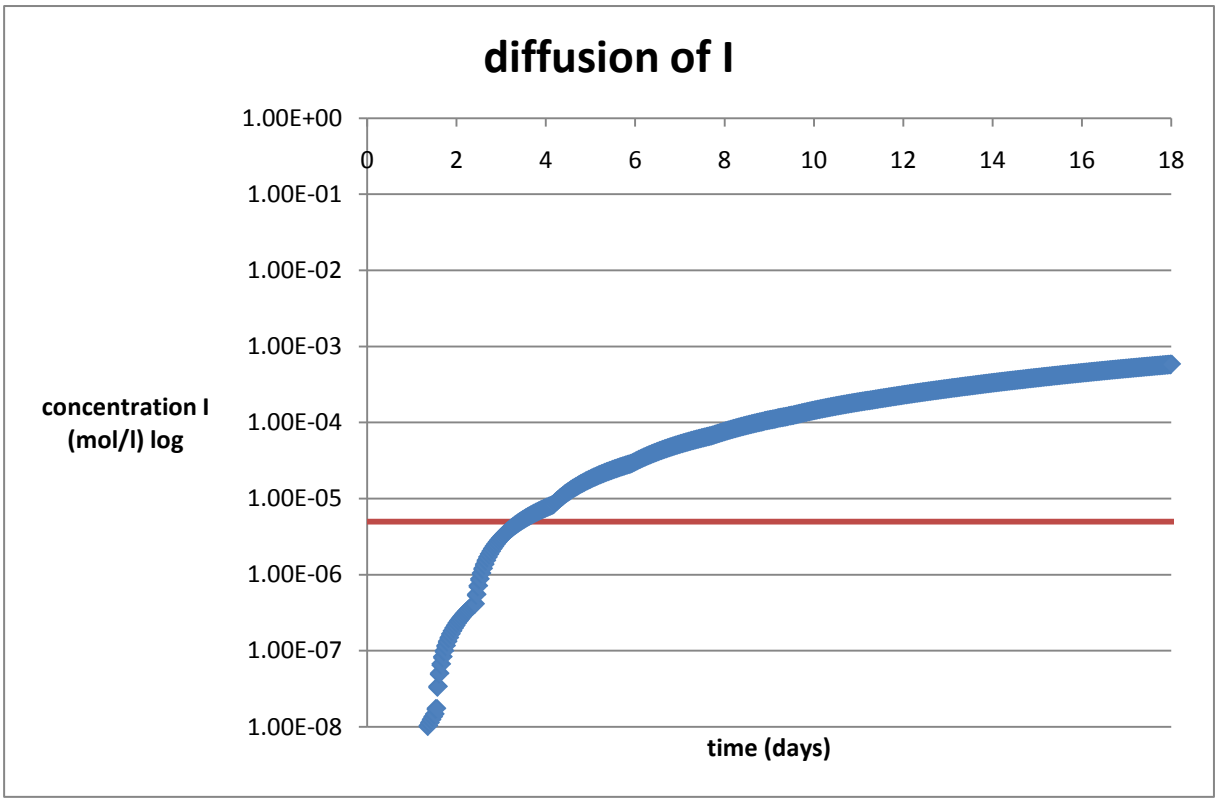

Figure 9: Calculated diffusion of iodine through a RBCW-saturated clay core of 37 mm high

In the first test (2010/1) synthectic pore water, which does not contain iodide, was used on top of the cell. After breakthrough, a concentration close to the natural I concentration of the pore water was obtained. However, since the Boom Clay core contains a natural background in iodide, this concentration will equilibrate with the synthetic clay water as soon as the test is started. If we neglect this equilibration and consider that all measured iodide is due to transport of I-enriched pore water from the bottom core, only 1.6 µl NaI solution was transported (see table 11).

In Test 2010/2, real Boom Clay water was used instead of synthetic clay water and breakthrough occurred after 18 days, resulting in an I concentration of 2.25 10^{-4} mol/l. This concentration is probably due to the diffusion of I only (calculated conc. ~5.8 10^{-4} mol/l). However if we reason in the same way as for experiment 2010/1 and consider that all measured iodine in the breakthrough sample is related to transport at the moment of gas breakthrough, 46.7 µl NaI solution was transported (see table 11). This provides an estimate of the volume "contaminated water" transported and the % of water displaced (or a measure of desaturation), knowing that in reality it will be lower.

In Test 2010/3, breakthrough only occurred after more than 40 days and therefore, no Iodide measurements were done at that time since it would be completely blurred by the Iodide diffusion. However, the water was sampled after 8 days (without BT) and an iodine concentration of 1.32 10^{-5} mol/l was obtained (see table 10). Diffusion of I through the clay core during 8 days would lead to a calculated concentration I equal to 7.61 10^{-5} mol/l.

Table 11: Overview of results

	Measured concentration I (mol/l)	Day of breakthrough	Concentration diffused iodide (calculated)	Calculated Volume NaI transported (ml)*	Volume of water in NaI core (ml)	% of water displaced
Test 2010/1	5.59 10^{-6}	3.9	7.2 10^{-6}	1.6 10^{-3}	4.4	0.036
Test 2010/2	225 10^{-6}	16.3**	579 10^{-6}	46.7 10^{-3}	5.4	0.86

*volume calculated neglecting the contribution of diffused iodide
** sample was taken after 18 days.

In previous breakthrough tests, a mean decrease in saturation less than 2% was reported (MEGAS project, Volckaert et al., 1994). The new tests provide indications that the decrease in saturation is less than 1%, which is even a conservative value, as we did not take into account the diffusion of iodide through the clay core. Probably the real value is even much lower than 1% desaturation.

During the MEGAS experiments, pressure was increased slowly and after each stepwise increase, the system had sufficient time to adapt to the new pressure state. Despite the faster pressure increase in our recent tests, the amount of water that is displaced during

breakthrough is even less. So a faster increase in pressure does not lead to more water displacement.

As observed, the breakthrough pressure for undisturbed Boom Clay is quite high, so it is not easy to reach it within 4 days, hampering the interpretation wrt. gas induced tracer transport. So, it might be more interesting to focus on tests on disturbed (fissured) Boom Clay, where we expect lower breakthrough pressures which can be reached easily within a few days.

At the moment of breakthrough for the 3 tests, quite different observations were made:

- in test 2010/2 after breakthrough, gas started to flow at moderate speed (4.18 ml/hr) (see figure 5).
- But in test 2010/3 flow increased during the first 15h after breakthrough till 60 ml/hr, and then suddenly decreased to 0 (see figure 6). Maybe at this point, the shut-in pressure was reached and pathways closed again? After the flow stopped, pressure was increased again until the first breakthrough pressure was reached and gas started to flow again, but at moderate speed (3.37 ml/hr). The same situation was observed after a third breakthrough (2.68 ml/hr). After the third breakthrough, gas kept flowing at moderate speed (6.43 and 4.94 ml/hr) until the shut-in pressure was reached (4.16 MPa). After reaching this shut-in pressure, gas kept flowing but at very low speed (0.72 ml/hr) (see figure 7 and table 9).

Apparently the created pathways are very unstable and successive breakthroughs on the same sample lead to the formation of different pathways. However, if the unstable flow is declared by the formation of unstable pathways, this indicates that breakthrough occurs through the clay, and not along the wall. Also the presence of a shut-in pressure indicates rather breakthrough through the clay than along the wall.

Besides differences during the gas breakthrough, we also observe some similarities:

When we compare the residual gas flow after reaching the shut-in pressure and compare it to the gas inflow rate before breakthrough for both tests 2010/2 (0.79 ml/hr) (see figure 5) and 2010/3 (0.40 and 0.45 ml/hr) (see table 9), we see that all values are in the same range.

We first considered that this could be due to the diffusion of helium through the clay. But when we calculate the diffusive inflow of He (D_{app} = 11.4 10^{-10} m²/s) (Jacops et al., 2011) into an infinite, porous medium (porosity 40%) with a diameter of 38mm – taking into account that the concentration He at the inlet is 0.0152 mol/l - the inflow of helium into our sample due to diffusion should be ~0.014 ml/hr STP. This value is an order of magnitude lower than the measured value, so not all inflow is related to in-diffusion of He. Secondly we considered possible leakage of the system. But, the measured leak rate of the pressure systems is less than 0.1 ml/hr. So the measured helium inflow is still considerable higher than the diffusion-related inflow plus leakage. This points to another phenomena that is governing this gas flow.

When looking at previous breakthrough experiments during the MEGAS project (Volckaert et al., 1994), the average flow rates obtained during these experiments (see table 12) are in the same order of magnitude as the flow rates obtained during tests 2010/2 and 2010/3 (see figure 5 and table 9).

Table 12: Average flow rates, calculated from MEGAS project (Volckaert et al., 1994)

Reference clay core	Average flow after breakthrough (ml/hr STP)
23B5.5K2	15.2
23B4.5	2.45
27B7.5K1	2.91
27B7.5K2	5.31

5. Conclusion

For all experiments, breakthrough occurred at a pressure higher than the calculated breakthrough pressure. This is probably due to the fast pressure increase regime used: because we want to measure I transport due to a gas breakthrough, this breakthrough should happen within 4 days after the start of the experiment as the unretarded diffusion of iodine will lead to iodine diffusive breakthrough and cause interference.

For this reason, pressure is increased quickly and the system does not get enough time to adapt to the new pressure state resulting in high and variable breakthrough pressures.

The tests were not entirely satisfactory wrt. evaluating gas induced tracer transport on undisturbed Boom Clay as the time to reach the relative high breakthrough pressures was equal or longer than the time needed for diffusive breakthrough. However, in one test, we can reasonably assume that iodine was transported due to gas breakthrough. The amount of displaced tracer saturated water is very small. This corresponds to low desaturation (<1%) of the tracer saturated clay core.

When looking at different flow rates and patterns that were measured during the different tests, we can state that the created pathways are very unstable: during breakthrough some pathways grow, other get closed and new pathways are formed. This can declare the variations in flow rate within one sample, but also between different samples.

However, most flow rates after breakthrough are in the same order of magnitude and correspond to flow rates measured during the MEGAS project. Also the low flow rates before breakthrough and after shut-in pressure are similar. These flow rates are an order of magnitude higher than the calculated helium inflow due to diffusion - pointing to an extra gas transport phenomena.

The goal of this research was to answer the following question: “to what extent can a gas pressure build-up enhance the radionuclide and contaminants transport in a clay host rock?” Based on the obtained results, we can state that the transport of radionuclides and contaminants due to a gas breakthrough is indeed possible but seems minimal. In order to have more solid proof of gas induced tracer transport more tests are needed. For that purpose, it might be more interesting to focus on tests on disturbed (fissured) Boom Clay, where we expect gas breakthrough at lower BT pressures and thus faster than the diffusive tracer breakthrough.

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