Summary of the work performed within WP3 in FORGE

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Euratom 7th Framework Programme Project: FORGE

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

In FORGE the objective of WP 3 was to examine unresolved issues related to gas migration which can detrimentally alter the hydraulic and mechanical (and potentially the thermal and chemical) properties of the engineered barrier systems. A detailed series of laboratory and field scale experiments was undertaken to provide new fundamental insights into the processes and consequences of gas migration through the engineered barrier and seals of repositories.

The current report summarizes the main findings from the WP3. The report was originally produced as the WP3 input to the work package 1 synthesis report.

Contents

Summary

The European Commission FORGE (Fate Of Repository GasEs) 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 gases.

This report contains a brief summary of the main findings from the work on the Engineered Barriers Systems (EBS) with the FORGE project. With respect to this, both clay and concrete based barriers have been studied. The purpose of WP3 was to examine how unresolved issues related to gas migration could detrimentally alter the hydraulic and mechanical (and potentially the thermal and chemical) properties of the engineered barrier systems. A detailed series of laboratory and field scale experiments was undertaken to provide new fundamental insights into the processes and consequences of gas migration through the engineered barrier and seals of repositories. The focus of this report is to describe how the findings can be used to represent issues around gas transport mechanisms, the role of interfaces and upscaling.

Bentonite based barriers

In an unsaturated or partially saturated bentonite there is a linear dependence between gas flow rate and pressure gradient, which indicates that two-phase flow is the dominating transport mechanism. This may also be the case for saturated sand-bentonite mixtures if the sand content is sufficiently high.

At a degree of saturation of ~80-90% or higher the behaviour changes entirely. No flow of gas will take place in the bentonite unless the applied pressure is equal to or higher than the total stress. The only transport mechanism is the omnipresent diffusion of dissolved gas. Diffusion has not been a key issue in Forge, but evaluated diffusivities are well in line with what has been presented elsewhere.

If the gas pressure reaches a higher value than that the pressure in the bentonite a mechanical interaction will occur. This will lead to either:

1. Consolidation of the bentonite, and/or
2. Formation of dilatant pathways

Consolidation means that a gas volume will be formed within the clay that and that the clay is compressed. This increases the clay density closest to the gas volume and the local swelling pressure is increased to balance the gas pressure. There is however a limit to the extent of consolidation.

At some critical pressure, pathways will be formed and the gas will become mobile. The pathways are characterized by a strong coupling between σ, Π and Pp, localised changes in σ, Π and Pp, unstable flow, exhibiting spatio- temporal evolution, localised outflows during gas breakthrough and no measurable desaturation in any test samples.

It is still unclear when consolidation ends and pathway formation starts. In some tests, pathways form when the gas pressure reaches the sample pressure. An example of this is the full scale Lasgit test. Other tests show pathway formation at an overpressure at about 20-30%, while there also are tests where breakthrough occurs at pressures 2-3 times higher than the sample pressure. The effect is clearly geometry dependent, but other factors may be involved as well.

However, it is clear that classical two-phase flow models cannot correctly represent gas migration in a compacted saturated bentonite.

In Forge, substantial effort has been devoted to the study of gas migration in interfaces. A simple summary of the findings is:

1. Interfaces will, not surprisingly, be the preferred pathway in an unsaturated system
2. If given the opportunity, gas will generally move along the interface between the clay and another material in a saturated system as well. This does not however seem to affect the transport mechanisms (previous paragraph).
3. In most cases bentonite/bentonite interfaces will seal and will not be preferential pathways for gas.
4. It is possible to design experiments where the gas is "forced" to move through the matrix

In Forge WP experiments have been performed in a multitude of different setups, boundary conditions, geometries (small and full scale) and materials. Overall, the results from the tests provide a consistent story. This indicates that the knowledge about the processes involved could be upscaled to repository conditions, both in time and in space.

Concrete barriers

The studies of gas migration in concrete within Forge have been limited in comparison with the studies of bentonite.

The key achievement have been an improved database for gas permeability in concrete under different conditions as well as understanding on how carbonation, from CO_2 gas, will affect the permeability of concrete.

Introduction

This report contains a summary of the responses from the participants in WP3 to a questionnaire that was prepared by WP1. The purpose was to cover key safety case-relevant issues. The focus of this report is to describe how the findings can be used to represent issues around gas transport mechanisms, the role of interfaces and upscaling.

This section gives phenomenological descriptions of transport mechanisms, which are diffusion, 2-phase flow, pathway dilation, gas fracing, that are frequently used in describing the domains of behaviour of clay-based materials - host rock, and clay-based engineered barrier - including its form (e.g. bentonite blocks versus bentonite pellets).

1.1 GAS DIFFUSION

Bentonite-based EBS

No experiments within FORGE were designed to specifically measure diffusion of dissolved gas in bentonite. However, since diffusion is an active process in all experiments it has been possible to evaluate the diffusion coefficient for certain systems.

The diffusion coefficient for <u>air</u> has been determined from measurements of steady-state volumetric flow through cylindrical samples of pure montmorillonite and natural bentonite (MX-80) exposed to certain air pressure gradients in a 1D geometry.

Sample	Density (post-analysis) (kg/m^3)	D_e (m^2/s)
Na-montmorillonite	616	$1.3 \cdot 10^{-10}$
MX-80	1075	$1.25 \cdot 10^{-10}$

Although the method primary measures volumetric flow (rather than making a more explicit analysis of transferred gas), it is reliable because the pressure response of the clay has been measured simultaneously. As the response due to pressurization with gas is very different as compared to the response due to pressurization with water, it can be fully assured that air is the pressurizing fluid at the time of measurement.

Diffusion coefficients for dissolved hydrogen were estimated under the assumptions that:

1. Before the break through of hydrogen through bentonite, the driving process for hydrogen transport is diffusion.
2. Concentration of hydrogen entering bentonite in aqueous phase corresponds to the pressure measure according to Henry's law.
3. The effect of transport of hydrogen by advective flow under pressure of hydrogen is negligible in a comparison with diffusion transport.

It was found that the values of diffusion coefficients depend on the density of bentonite, a higher density would yield diffusion coefficients. In experiments with the density of Ca, Mg bentonite (Czech bentonite from Rokle deposit) 1400 kg/m3 the diffusion coefficient was 3 x 10^{-10} m^2/s and for 1600 kg/m^3 7.6 x 10^{-11} m^2/s. For density of 1800 kg/m^3, diffusion of dissolved gas was not measureable. The effect of pressure is however noticeable suggesting that the effect of transport of hydrogen by advective flow initiated by pressure of hydrogen cannot be neglected.

In experiments with continuous increase of pressure from the reaction of iron with water, it was found that before the breakthrough a plateau with a relatively constant pressure was formed (Figure 1). The diffusion coefficients estimated from the region of this plateau was 3.7 x 10^{-10} m^2/s. In the similar experiment with Na bentonite (Volclay KWK-20-80), the estimated value of diffusion coefficient was 2.9 x 10^{-11} m^2/s.

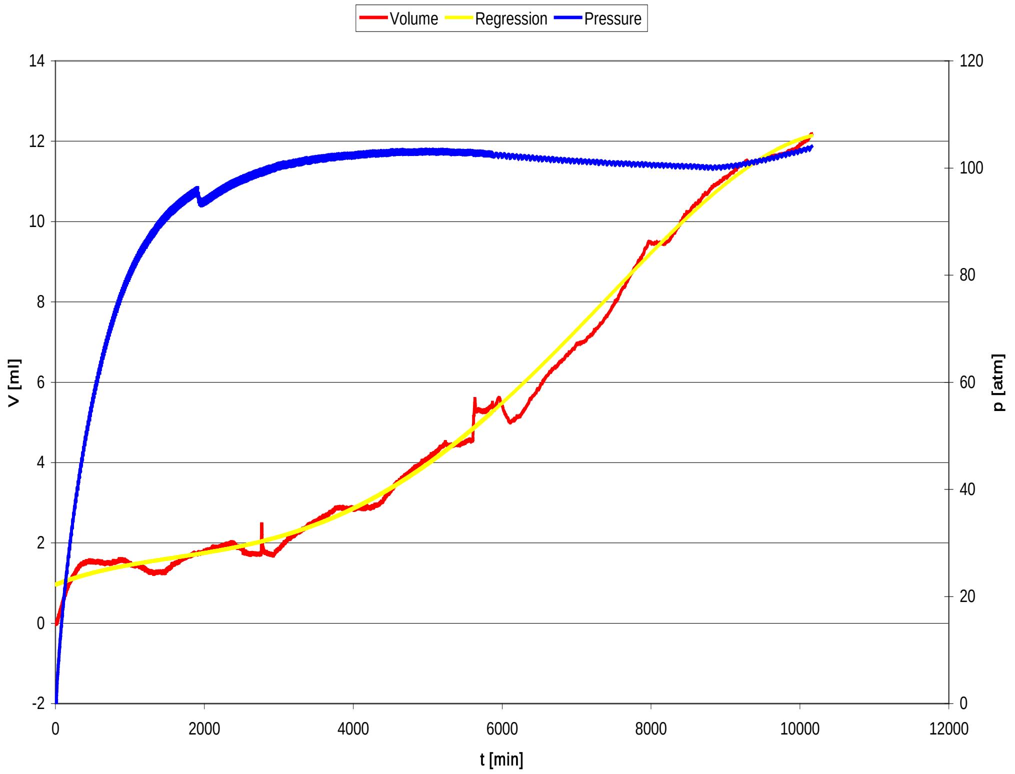

Figure 1 Pressure plateau formed before the breakthrough of hydrogen through bentonite of density 1600 kg/m3 and related flow of hydrogen

There is a remaining uncertainty regarding the true value of Henry's constant in the clay environment. The measurements done within FORGE are too few to give a comprehensive picture of the diffusivity of dissolved gases. Within the density range reported, it seems like the diffusion coefficient decreases with density (not surprising), molecular size and sodium content in the bentonite. However, there is a remaining uncertainty regarding the true value of Henry's constant in the clay environment.

Cementitious EBS

For the experiments with transport of CO_2 in cement it should be noted that the CO_2 is not an inert migrating phase. It is highly reactive towards the cement, and so its overall migration is controlled by both reaction and transport. The net effect of this will be to slow the rate of CO_2 migration relative to a purely non-reactive diffusional case.

However, some of our experiments did reveal the positions of migrating carbonation fronts as a function of time. Diffusional transport of CO_2 played a role in the migration of the fronts (though so did reaction of the CO_2 with the cement). It should be noted that he fronts do not record the position of the leading edge of CO_2 ingress, as minor amounts of CO_2 reacted with cement in advance of the fronts. Instead, the carbonation fronts record the position where sufficient CO_2 had permeated the core to enable sufficient carbonation to occur to change the structure of the cement (including *complete* carbonation of the reactive cement minerals). The fronts migrated by a few mm over several weeks.

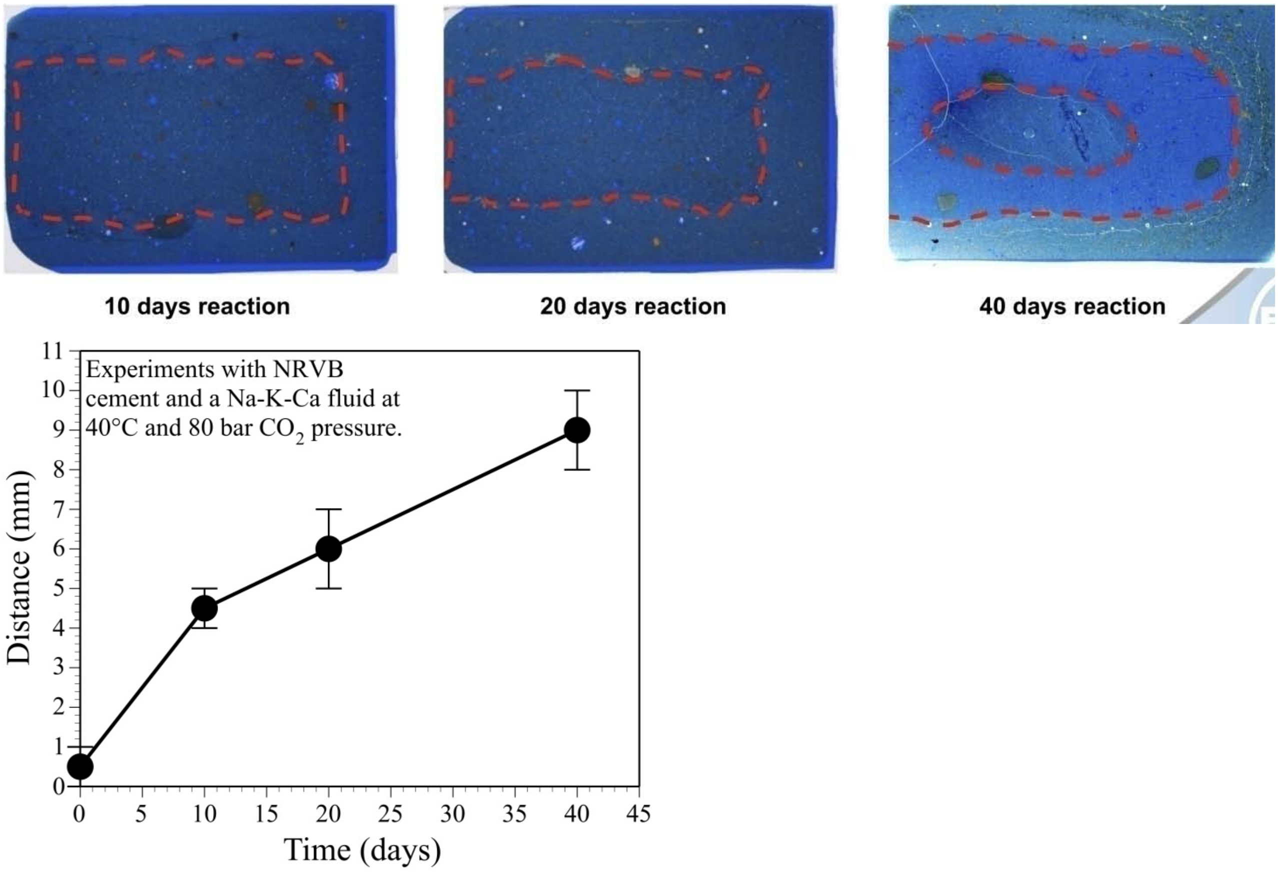

Figure 2 progressive migration of carbonation reaction fronts through 25mm diameter samples of Nirex reference vault backfill (NRVB) cement.

1.2 FREE GAS PHASE FORMATION

Bentonite-based EBS

In the case where more gas is generated than what can escape with diffusion a free gas phase will form.

Initially, the gas phase will consolidate the clay phase. Experiments by Clay Technology have demonstrated, not only that a gas phase does not interact mechanically with water-saturated bentonite when its pressure is below the pressure of the bentonite, but also that mechanical interaction inevitably takes place when the gas pressure exceeds the initial pressure of the clay. This is clearly illustrated in Figure 3, which shows the pressure response of an MX-80 bentonite sample at differently applied gas pressures gradients.

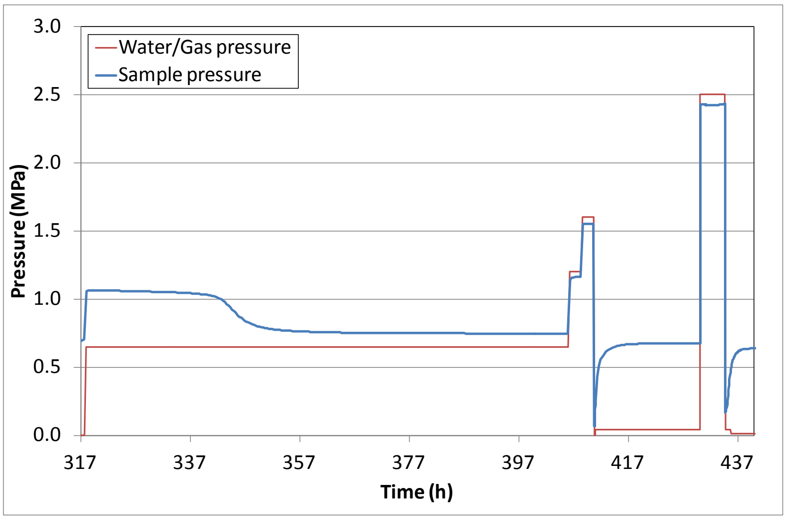

Figure 3 Response of an MX-80 sample which is being pressurized with gas above and below initial swelling pressure. The pressure of the sample is independent of gas pressure when the gas pressure is below the initial swelling pressure (~0.75 MPa), while it is basically equal to the gas pressure at higher injection pressures. Note that water is the pressurizing fluid at the beginning of the displayed pressure evolution (317 – 340 h).

An interesting feature of the test of Figure 3 is that although it clearly demonstrates mechanical influence of the gas on the clay, it does not indicate any additional transport mechanism apart from the ever-present gas diffusion. In contrast, in other tests gas breakthrough events have been demonstrated to occur when gas pressures at or above the pressure of the clay sample.

1.3 TWO-PHASE FLOW

Considerable care has to be taken on terminology. In the question it is stated “2-phase flow” – what is meant here is visco-capillary flow where the properties of the clay capillaries are playing a control on gas displacing water.

Based on experimental data from Ciemat data, two-phase flow seems to take place for degrees of saturation lower than about 93% in compacted bentonite and concrete.

The experimental work within Forge clearly demonstrates that no two-phase flow occurs in saturated bentonite.

1.4 PATHWAY DILATION & GAS-INDUCED FRACTURING

Experimental evidence from Lasigt shows that dilation is the predominant advective flow mechanism. At the point of gas breakthrough there is a co-incident pressure and stress response seen at the deposition hole wall. Qualitatively similar results have been seen in laboratory tests and in gas injection tests 1 & 2. For gas test 3 (see Figure 4) gas break-through has been accompanied by a secondary rise in gas pressure, followed by a second break-through event with a more gentle form. These results strengthen the dilatancy path propagation hypothesis and cannot be explained by classical visco-capillary (two-phase) flow concepts.

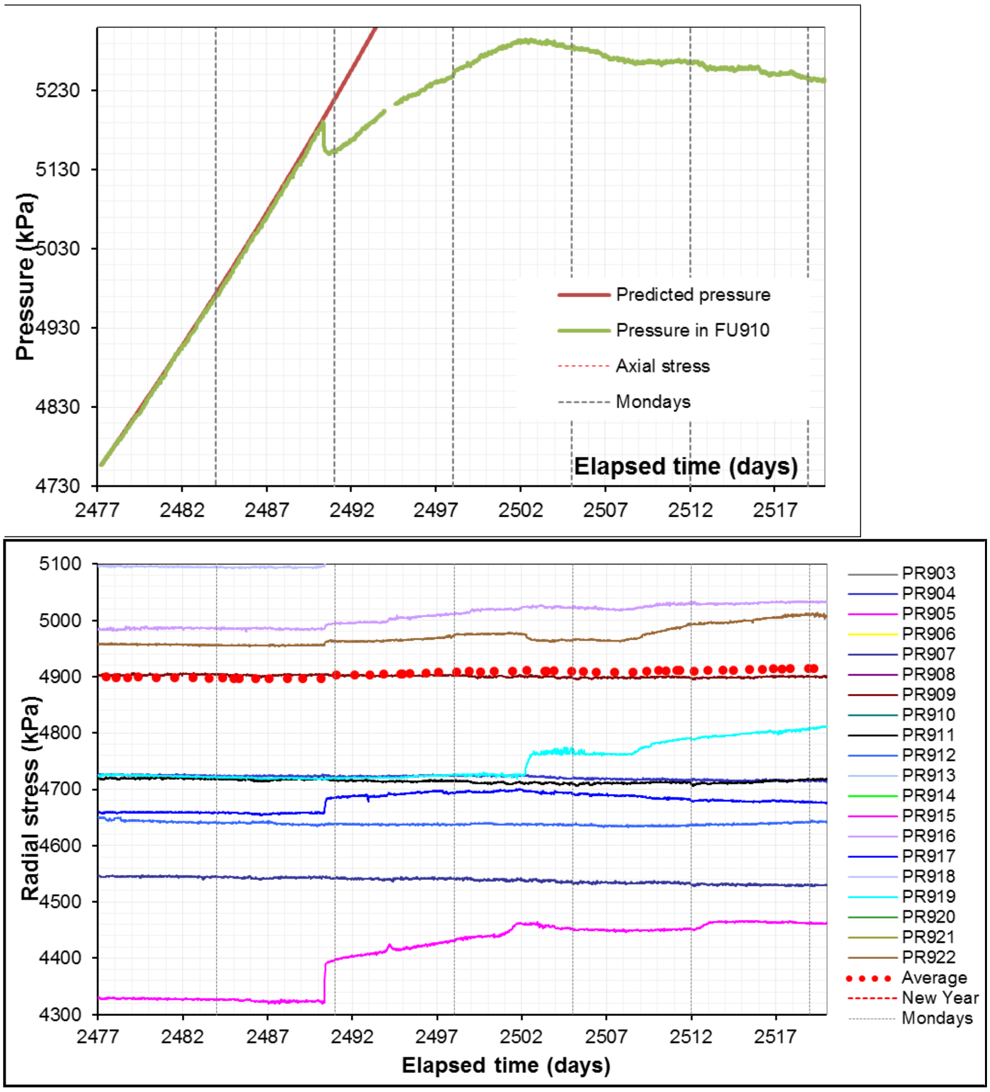

Figure 4 Gas break-through (top) and radial stress response (below) at the deposition hole wall.

There is no evidence of 2-phase flow by visco-capiliary flow.

The laboratory studies by BGS show that under all tested conditions during the project, observations indicate that the ***primary*** mode of gas transport is by dilatant pathway formation. These observations include:

- Strong coupling between σ, Π and Pp (Figure 5)
- Localised changes in σ, Π and Pp (Figure 5)
- Unstable flow, exhibiting spatio- temporal evolution (Figure 5)
- Localised outflows during gas breakthrough
- No measurable sample desaturation in any test samples, indicating that gas has not passed through the bulk of the bentonite

These findings are consistent with other recent studies involving argillaceous materials (Ortiz et al., 2002; Angeli, et al. 2010; Cuss et al., 2010; Skurtveit et al. 2011; Harrington et al., 2009), as well as earlier studies in bentonite (Pusch et al., 1985; Harrington and Horseman, 2003; Horseman et al., 1999).

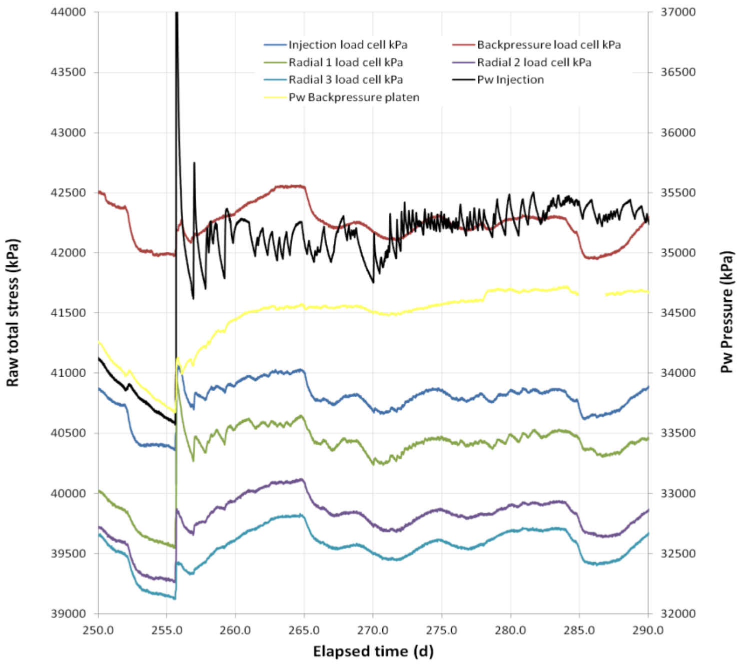

Figure 5 Output of laboratory studies by BGS, showing that under all tested conditions during the project, observations indicate that the primary mode of gas transport is by dilatant pathway formation.

Clay Technology has tested the gas breakthrough behavior by lowering the the total pressure of the bentonite system by flushing a strong NaCl solution on the outlet side while a constant gas pressure, below the initial pressure of the bentonite, was maintained on the inlet side. The bentonite pressure immediately started to fall after the flushing, and when it became comparable in size to the injection gas pressure a gas breakthrough event occurred (Figure 6). This behavior was demonstrated in the same sample for different gas injection pressures, giving a clear-cut evidence of the osmotic nature of the system.

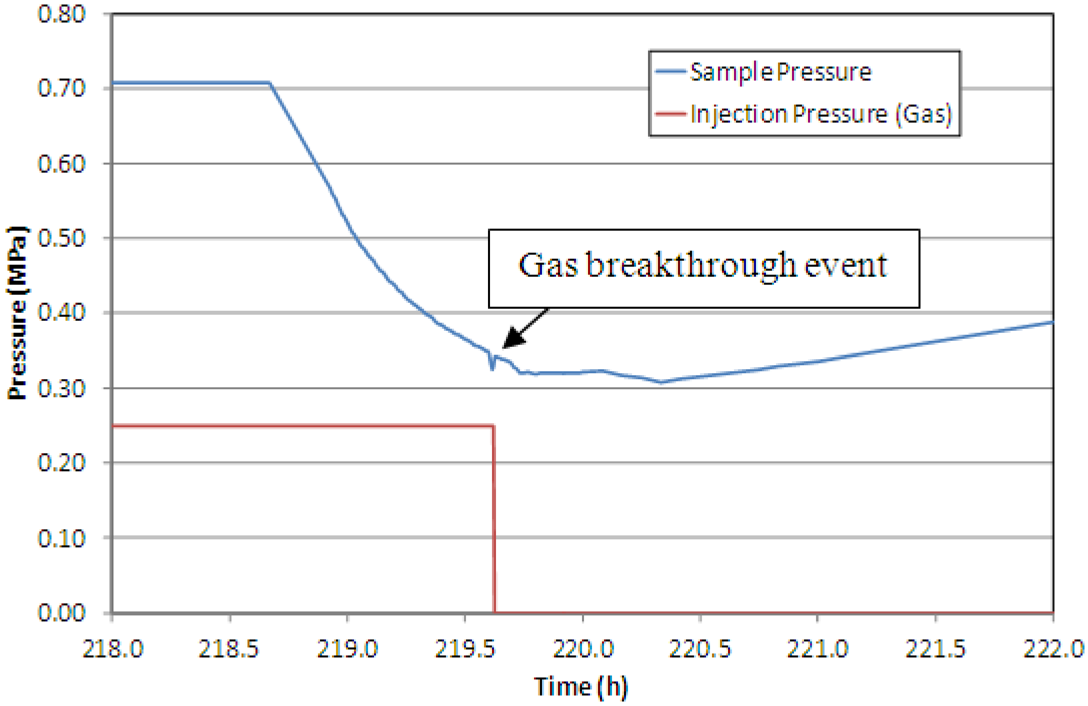

Figure 6 Response of an MX-80 sample which is being flushed by a 3 M NaCl solution at the top filter (at ca 218.75 h). The bottom filter is pressurized with air at 0.25 MPa. A breakthrough event occurs when the sample pressure becomes similar to the gas injection pressure.

1.5 GAS REACTIVITY

Cementitious EBS

In WP3 gas reactivity has only been studied for systems with cement and CO_2.

CO_2-cement chemical reaction caused two types of *localised* temporary shrinkage cracks to develop, and in particular in the unconfined samples. One set of shrinkage cracks were oriented parallel to the flow direction (i.e. at 90° to the reaction fronts) and the other set were perpendicular to it (associated with the most recent, as well as older, reaction fronts). Secondary precipitation eventually sealed these potential flow features.

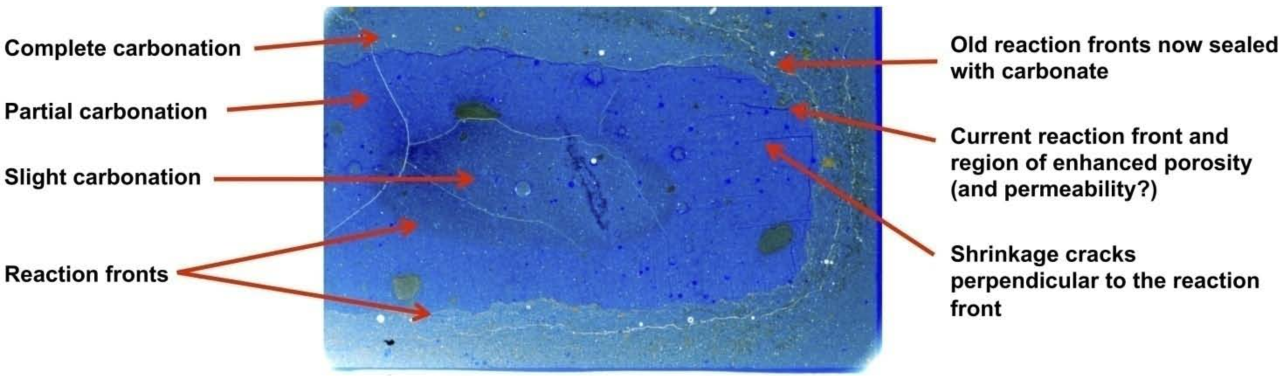

Figure 7 Partially carbonated cement sample showing zones having different degrees of carbonation together with associated reaction fronts (Rochelle et al. BGS technical report that is currently in draft).

2.1 INTERFACES WITHIN THE EBS

In general it seems like gas will select a path in the interface between a clay and a metal barrier if there is a possibility. This is confirmed in a number of experiments in Forge. However it still seems like the properties of the clay alone will determine the gas transport process. One example a test by Clay Technology that specifically addresses the issue of the interface pathway

The nature of the formed breakthrough pathways was studied in test cells of a type schematically pictured in Figure 8. Gas was injected in a small central filter in the bottom of the cell, while filters for outflow were located both at the top of the cell as well as at the bottom circumference (“guard filter”). In this way it can be concluded that any gas ending up on the top side must have propagated through the clay. However, in all cases where this has been studied (with the exception of some very thin samples) the gas was detected in the “guard filter” although the dimensions were such that this path was typically twice as long as the shortest path through the clay.

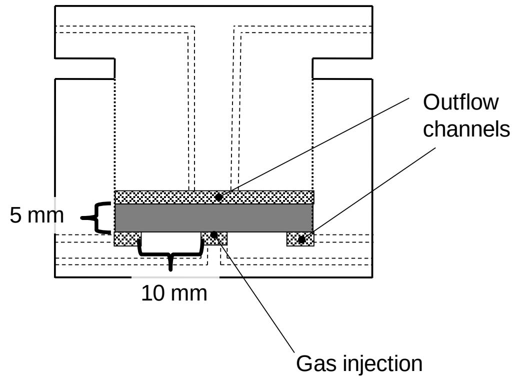

Figure 8: Schematics of the test cell used for detecting preferential path for gas between clay body and cell wall.

Consequently, these results strongly indicates that the cell/clay interface compose a preferential path for the gas in a breakthrough event.

Tests at IfG with bentonite-bentonite interfaces showed that, at dry conditions gas flow along interfaces is at least 4 orders higher than through the matrix.

Increase of confinement resp. normal stress on the contact zone significantly lowers the gas flow also in the dry buffer material, but the effect is more pronounced for interfaces → crack sealing.

Flooding of the EBS results in saturation of the interface (or contact zone), however our results document that regarding the contacts block/block (i.e. inside the bentonite plug composed of single bentonite blocks) the water consumption during swelling depends

a) obviously not on the injection geometry

b) and only weakly on the axial load

For flooding of the bentonite block seal it can be concluded that between the blocks no preferred flow along the interface exists, i.e. matrix swelling dominates. In addition during the tests, 1 year of test time was not sufficient to reach full saturation.

2.2 TREATMENT OF INTERFACES IN EXPERIMENTS

It is well known that interfaces in experiments may act as preferential paths. All experiments in Forge has been designed with that fact in mind. As an example from Ciemat, two different approaches are used depending on the tested material: If the material develops swelling pressure (as saturated bentonite), the material itself acting against a rigid cell forms the seal. This would be confirmed by the fact that the breakthrough pressures measured so far are higher than the expected swelling pressure. If the material does not develop enough swelling pressure (concrete, low-saturated bentonite, low-expansive clays), the samples are wrapped with elastic materials (rubber or neoprene) and confining pressures higher than the gas injection pressure were applied. Both approaches are used for homogeneous samples and interface tests. For tests within a rigid cell, the non-swelling material is glued with epoxy to the inner surface of the cell.

3.1 SPATIAL UPSCALING

Lab tests allow us to understand processes on a small scale, under carefully controlled conditions and suitably defined boundary conditions. Such understanding underpins experimental work at the URL scale (for example using laboratory testing to inform interpretation of observations at Lasgit, where similar behaviour is observed on both scales). Findings from lab and URL scale can then be used to inform the selection of the correct model and aid in its calibration/validation. This may be an iterative process, but provides a full and accurate understanding of the likely processes involved. Assuming the bentonite is homogeneous then laboratory observations must be directly scalable to field conditions, for regions of bentonite that are at similar levels of maturity.

3.2 UPSCALING OF EXPERIMENTAL RESULTS AND UNDERSTANDING

One approach could be:

1.) Develop a full understanding of each individual component on a range of scales
2.) Identify the dominant components/processes involved under expected conditions
3.) Identify linkages between the dominant components/processes involved under expected conditions
4.) A forecast of the expected system behaviour can then be made
5.) Further tests may be required to reduce the remaining uncertainties

3.3 TIME UPSCALING

Tests are run as slowly as possible, so as to solicit the underlying physics. The behaviour observed in many of the WP3 tests, and in the literature, indicates a distinct threshold for gas entry (related to the sum of Π and Pp). The evidence clearly suggests that this threshold is ***independent of rate*** and should scale well to longer time-scales. However, the potential for other effects to become significant at longer timescales may also need to be considered (for example, the coupled impacts of glacial loading and residual pore-pressures).

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