Gas Generation and Migration

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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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FORGE

Fate of Repository Gases Project

Gas Generation and Migration

International Symposium and Workshop

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FORGE (Fate of Repository Gases) Project

Various gases will be generated in a repository including hydrogen (from metal corrosion) and methane and carbon dioxide (both from decomposition of organic materials contained in some wastes). Understanding where and how these gases form and how they move through the repository and surrounding rocks is the focus of the FORGE project. By using small scale laboratory experiments, large scale field tests (performed at a number of underground research laboratories throughout Europe), data and numerical modelling the results from FORGE is providing information to help guide repository design and predict future radionuclide migration.

The understanding and prediction of the evolution of repository systems over geological time scales requires a detailed knowledge of a series of highly-complex coupled processes. There remains significant uncertainty regarding the mechanisms and processes governing gas generation and migration in natural and engineered barrier systems. It is important to understand a system to an adequate level of detail to allow confidence in the assessment of site performance, recognising that a robust treatment of uncertainty is desirable. Of particular importance to the European radioactive waste management programmes are the long-term engineering performance of bentonite buffers, plastic clays, indurated mudrocks and crystalline formations. To reduce uncertainty, further experimental data are required to address:

- Corrosion and gas generation rates in repository environments;
- Key issues relating to the migration and fate of repository gases;
- Validation of numerical codes;
- Derivation of new methodologies for up-scaling from laboratory to field to
- repository scales;
- Optimisation of repository concepts through detailed scenario analysis.

FORGE is now nearing completion and these proceedings arise from a three day meeting held in Luxembourg, in February 2013, to disseminate the outcomes of the project.

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Gas Generation and Migration

International Symposium and Workshop
5th to 7th February 2013
Luxembourg
Programme

Day 1 Tuesday 5th February 2013

9.00 Welcome Richard Shaw (BGS - Co-ordinator FORGE) and Christophe Davies (EC Project Officer)

9.15 Keynote Presentations

Why is gas important for radioactive waste disposal safety case? - Simon Norris (NDA-RWMD)

Is gas an issue? - Ulrich Noseck (GRS on behalf of OECD-NEA)

10-15 to 10.45 Break

10.45 Lessons learned from FORGE – summaries from each FORGE Work Package.

10.45 to 11.05	WP1.2	Jacques Wendling (Andra)
11.05 to 11.25	WP2	Delphine Pellegrini (ISRN)
11.25 to 11.45	WP3	Patrik Sellin (SKB)
11.45 to 12.05	WP4	Jon Harrington (BGS)
12.05 to 12.25	WP5	Geert Volckaert (SCK.CEN)

12.30 Lunch

14.00 Parallel Sessions 1

	A. Waste form and engineered barriers (WP1,2 and 3)	B. Disturbed and undisturbed host rocks (WP4 and 5)
14.00 to 14.20	1. Gas Intrusion in Bentonite – Results of Small Scale Experiments M. Birgesson and O. Karnland (Clay Technology, Sweden)	1. Gas flow in anisotropic claystone. Modelling triaxial experiments D Arnedo et al (UPC, Spain)
14.20 to 14.40	2. Role of Interfaces in Bentonite-Block Assemblies as Favoured Pathways for Gas Transport T Popp et al (IfG, Germany)	2. Thermodynamical Modelling of Hydrogen Migration in Argillite for a deep Geological Repository – ISRN contribution to FORGE M Dymitrowska et al (ISRN, France)
14.40 to 15.00	3. 3D Gas Transport Model of LASGIT Gas Injection Tests N Calder et al (Geofirm Engineering, Canada)	3. Characterisation of Gas Migration in Claystone through the Modelling of a Field-Scale Gas Injection Test P Gerard et al (University of Liege, Belgium)
15.00 to 15.20	4. Gas Flow in Compact Bentonite RJ Cuss et al (British Geological Survey, UK)	4. Determination of Diffusion Coefficients for dissolved He and CH_4 in Opalinus Clay A Grade et al (SCK.CEN, Belgium)
15.20 to 15.40	5. Laboratory Investigation of hydrogen generation from Carbon steel corrosion under deep geological repository conditions D Dobrev and A Vokal (UJV Res, Czech Republic)	5. Numerical Modelling and Interpretation of Field-Scale Gas Injection Experiment PGZ1 S Granit and R de La Vaissiere (EDF and Andra, France)
15.40 to 16.00	6. Microbial Transformation of Radioactive Wastes: Implications on Gas Generation and Radionuclide Speciation AJ Francis (Pohang University, Korea)	6. Dilatancy Driven Gas Flow in Callovo-Oxfordian Claystone (COx) JF Harrington et al (BGS, UK)

16.00 to 16.30 Break

16.30 to 18.00 Brief introductions poster

18.00 to 20.00 Poster Session based on two themes above with buffet reception

Day 2 Wednesday 6th February 2013

9.00 Continuation of parallel sessions

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9.20 to 9.40	2. Gas Generation in real L/ILW container drums and near	2. The Role of the Excavated Damaged Zone in HG-A Field-Scale Experiment Modelling

	surface vaults M Molnar et al (INR, Hungary)	S Levasseur et al (University of Liege, Belgium)
9.40 to 10.00	3. Carbon Steel Corrosion and Hydrogen Gas Generation in Cementitious Grout RC Newman et al (University of Toronto, Canada)	3. Gas Generation and Migration through Salt Formations T Popp et al (IfG, Germany)
10.00 to 10.20	4. Carbonation of Repository Cement: Impact of CO_2 on Cement Mineralogy and Porewater Chemistry G Purser et al (BGS, UK)	4. Numerical Interpretation of Gas Injection Tests at Different Scales H Shao et al (BGR, Germany)
10.20 to 10.40	5. FORGE Mock-up Experiment to Simulate Controlled Gas Release from a L/ILW Repository J Rueedi et al (Nagra, Switzerland)	5. The Experimental in-situ Study of Gas Migration in Crystalline Rock with a Focus on the EDZ J Svoboda and J Smutek (CTU, Czech Republic)

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11.50 to 12.10	2. Gas and Water Permeability of Concrete M-V Villar et al (CIEMAT, Spain)	2. 3D Water and Gas Transport Model of the HG-A Experiment R Walsh et al (Geofirma Engineering, Canada)
12.10 to 12.30	3. Mechanical Stability of Engineered Barriers in Sub-surface Disposal Facility during Gas Migration Based on Coupled Hydro-Mechanical Modelling S Yamamoto et al (Obayashi Corporation, Japan)	3. Investigations of Gas Migration through Undisturbed and Resealed Clay Rocks C-L Zhang et al (GRS, Germany)

12.30 to 14.00 Lunch

14.00 to 14.15 Introduction to working groups

Working Groups with topics/questions

	Working Group 1	**Working Group 2**	**Working Group 3**	**Working Group 4**	**Working Group 5**
14.15 to 15.45 and 16.15 to 17.45	vi) i) ii) iv) iii) v)	iv) iii) vi) ii) i) v)	i) v) iii) iv) ii) vi)	ii) i) iii) iv) v) vi)	iii) i) ii) iv) v) vi)
15.45 to 16.15	Break				

Working Group Questions

i) **Hydraulic properties:** In your opinion, can gas, under any circumstances, affect the favourable hydraulic properties (e.g. low permeability) of components of the engineered barrier system and the host? If yes, specify how, indicate significance and clarify the circumstances.

ii) **Mechanical properties:** In your opinion, can gas, under any circumstances, mechanically disrupt components of the engineered barrier system and the host rock? If yes, specify how, indicate significance and clarify the circumstances.

iii) **Solute migration:** In your opinion, can gas, under any circumstances, affect water and solute (typically radionuclides) displacement within the engineered barrier system and the host rock? If yes, specify how, indicate significance and clarify the circumstances.

iv) **Chemical properties:** In your opinion, can gas, under any circumstances, chemically affect the favourable properties of components of the engineered barrier system and the host rock? If yes, specify how, indicate significance and clarify the circumstances.

v) **Containment:** In your opinion, can gas, under any circumstances, affect the favourable properties of canisters/overpacks in the repository? If yes, specify how, indicate significance and clarify the circumstances.

vi) **Completeness:** In your opinion, are there other gas related phenomena that could affect the safety functions that are not considered in the other questions? If yes, specify how, indicate significance and clarify the circumstances.

Day 3 Thursday 7th February 2013

9.00 to 10.30 Reports from Working Groups (ca 15 minutes each)

10.30 to 11.00 Break

11.00 to 12.00 Panel discussion

12.00 to 13.00 General raporteurs feedback

13.00 to 13.15 Closure

13.15 Lunch

Posters

A. Waste form and engineered barriers (WP1,2 and 3)	**B. Disturbed and undisturbed host rocks (WP4 and 5)**
1. FORGE WP1.2: Numerical Benchmark on Gas Migration at Repository Scale: Involved Teams and Codes, Conceptual Basis and Main Results E Ahusborde et al (CNRS, France)	1. Dissolved Gases in Crystalline Rock, Observations from Outokumpu Deep Drill Hole L Ahonen et al (Geological Survey of Finland)
2. 3D Numerical Simulation of Gas Migration Through Engineered and Geological Barriers for a Deep Repository for Radioactive Waste: FORGE WP1.2 Benchmark E Ahusborde et al (CNRS, France)	2. Simplified 1D Modelling of the HG-A Test J Alcoverro et al (UPC, Spain)
3. ‘Second Order’ Exploratory Data Analysis of the Large Scale Gas Injection Test (Lasgit) Dataset, Focused Around Known Gas Migration Events Bennett et al (Cardiff University, UK)	3. FORGE WP4-5: Gas Migration Through Callovo-Oxfordian Claystone: Mechanisms Description and Remaining Issues J Talandier and R de La Vaissiere (Andra, France)
4. FORGE Repository Scale Benchmark Modelling Using T2GGM N Calder et al (Geofirma Engineering, Canada)	4. Validity of Critical Stress Theory Applied to the Movement of Water and Gas Along a Fracture Plane S Sathar et al (BGS, UK)
5. Hydrogen Production by Iron Corrosion under Gamma-Irradiation D Stammose et al (ISRN, France)	5. Determination of Gas Diffusion Coefficients in Boom Clay: Effect of Molecular Size and Anisotropy E Jacops et al (SCK.CEN, Belgium)
6. Modelling of Localised Gas Pathways in Long-Term Gas Injection Test P Gerard et al (University of Liege, Belgium)	6. Noble Gas Mobility in Brines: Case Study on the Salting Out Effect S Stadler et al (BGR, Germany)
7. The Impact of Elevated Pore-Pressure on Gas Flow in the Buffer; Experimental Observations in Pre-Compacted Mx80 Bentonite CC Graham et al (BGS, UK)	
8. Gas Injection Tests on a Sand-Bentonite Mixture: Investigation of the Effect of Pore Water Chemistry D Manca et al(Ecole Polytechnique Federale de Laussanne, Switzerland)	
9. Laboratory Gas Injection Tests and Modelling on Compacted Bentonite Buffer for TRU Waste Disposal in Japan K Namiki et al (RWMC, Japan)	
10. Simulating the HG-C and HG-D Experiment M Navarro (GRS, Germany)	
11. Modelling Lasgit Experiment S Olivella et al (UPC, Spain)	
12. Numerical Simulation of Gas Migration Test of Compacted Bentonite Using Model of Two-Phase Flow Through Deformable Porous Media Y Tanaka and M Hironaga (CRIEPI, Japan)	
13. Hydraulic Gas Behaviour of all Relevant Time and Space Scales of a Generic Repository. Contribution of Andra’s Study in the Framework of FORGE WP1.2 E Treille and J Wendling (Andra, France)	
14. Gas Permeability and Breakthrough Pressures of FEBEX Bentonite M-V Villar et al (CIEMAT, Spain)	

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FORGE: General outcomes: A view from the general rapporteurs

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[2]Ondraf/Niras, Belgium

1. Introduction

In the post closure phase of a deep geological repository for radioactive waste, significant quantities of gases may be generated, which could impact the long-term safety of the repository in different ways. Mitigating the various processes presents the designer with a possible conflict of goals. While radioactive gases are subject to the strategy of the system providing isolation and containment, non-radioactive gases may need to be dissipated to minimize the pressure build-up in the repository system. Consequently, a sound strategy for addressing these issues, and a body of robust arguments, are needed to support a post-closure safety case in order to give an adequate level of confidence that gas generation is not an issue likely to compromise the safety of a deep disposal system.

This paper is compiled by the general rapporteurs of this symposium. It aims to present salient results and key issues raised at the symposium and to place them in the perspective of the safety case. Account is also taken in this document of key aspects discussed in a topical meeting held earlier (Thirteenth Meeting of the Integration Group for the Safety Case (IGSC), NEA topical session on the relevance of gas for the post-closure safety case of DGR for HLW and spent fuel, Paris, 19 October 2011**)** by the member organisations of IGSC and that were synthetized in an introductory presentation at the symposium opening.

One general and often repeated statement from the FORGE project is that gas behaviour is highly concept- and conditions-specific. This concerns for example the type of engineered barrier system (EBS) used, the saturation state, the type of host rock, or the layout of the repository. As a consequence, future research might be more focused on specific needs from national programmes, e.g. the detailed composition of the backfill material.

2. Gas sources in repository system

In general, the results presented at the FORGE symposium are in line with the existing knowledge regarding gas sources. Gas generating processes in a repository include anaerobic metal corrosion, (bio-)chemical degradation of organics and radiolysis of water and waste. In addition, some radionuclides released by the waste as it degrades might be in gas form.

Corrosion processes have been extensively investigated early on in national programmes for geological disposal to verify that containment requirements could be met. Long-term experiments in conditions representative of a disposal system show carbon steel corrosion rates of the order of tens of nanometres per year in cement-based systems and tens of microns per year in bentonite-based systems (Newman et al., 2013 and Pellegrini et al. 2013). The asymptotic decrease of carbon steel corrosion rates because of the progressive build-up of a protection layer suggests rates of an order of magnitude lower at the time scales relevant for geological disposal systems.

Once confidence in meeting the basic containment requirements is obtained, R&D tends to shift towards the study of the initial transient and of the effect of possible perturbations (e.g. ingress of aggressive species). Indeed, unsaturated transient conditions might prevail for a significant time, depending on the type of repository system. For instance, unsaturated conditions are inherently part of the expected evolution of a repository in dry host formation, like rock salt (Beuth et al., 2012). Such conditions might also exist for decades in the near field of some clay-based concepts. For such conditions where water vapour corrosion would occur, few data are available. However, recent results for steel corrosion in cementitious material at 100% humidity show corrosion rates similar to those observed in liquid water (Newman et al, 2013).

A variety of gases can be produced from microbial degradation of cellulose, resins, bitumen and plastics present in intermediate-level, long-lived wastes (ILLW)(GASNET, European Commission, 2003). The rates are difficult to predict because of the limited experimental data and the great variability. It is believed that geological conditions are sufficiently hostile to limit the microbial activity. However microbes show also remarkable adaptation behaviour. In addition to inactive gas, the possibility of formation of volatile radioactive compounds (including I and Se) from microbial activity, e.g. methylation has been discussed by Francis (2013). It has also been pointed out that microbial activity is not necessarily detrimental and can also be a sink of gas (e.g. the microbial conversion of hydrogen) but the issues of variability and long-term viability of microbes in repository conditions also apply here.

3. Gas transport

In system components saturated with groundwater or close to saturation, gas transport will start with molecular diffusion of dissolved gas. No particular issues are expected for systems in which the gas production rate is low enough for all gas to be continuously evacuated through this well-characterised process (Grade et al., 2013). Difficulties – and differences between disposal systems – arise if the capacity for diffusive removal of dissolved gas is exceeded: characterisation of free gas transport through low-permeability porous materials is a challenging endeavour, especially when those materials are close to saturation with water.

Teams involved in the identification and characterisation of free gas transport modes on small samples in the lab stressed the high sensitivity of the results to the experimental conditions and the initial state of the samples. Some of these conditions can be constrained by adequate control of the stress or strain boundary conditions and the geometry of the experimental setup. Nevertheless, for natural materials, sample variability may still lead to very different results for otherwise similar tests: gas always takes advantage of heterogeneities to flow through the path of least resistance. For engineered materials, large variability in gas migration properties might also show up as a result of discrepancies in formulation and curing conditions used to produce the material (Rueedi et al., 2013). The difficult characterization of concrete with respect to its gas transport properties was described by Villar et al. (2013). In situ characterisation of gas transport in the host rock around underground laboratories presents additional challenges such as the presence, around the experimental setup, of a disturbed zone. Modelling studies reveal that the outcomes of such experiments can be quite sensitive to the extent and the properties of this disturbed zone (Levasseur et al., 2013; Gerard et al., 2013a; Granet and de La Vaissiére, 2013). A correct interpretation of in situ tests would thus ideally require an adequate characterisation of this zone. In absence of this, some properties might be deduced from the injected gas volumes and observed pressures but this calls for a certain level of faith in the process of back-analysis and parameter identification.

From the large amount of laboratory and in-situ studies performed in FORGE on gas transport in clay systems (e.g. Birgersson and Karnland, 2013; Cuss et al., 2013; Harrington et al., 2013; Zhang et al., 2013; Graham et al., 2013) a consensus emerged that the transport of free gas in such low permeability porous media saturated with water or close to saturation occurs by the creation of specific gas pathways, which translate into sample dilatancy or the creation / reactivation of discontinuities in the material that is being tested. Dilation is clearly observed in laboratory experiments on clays (e.g. Harrington et al. 2013). Interestingly, measurements of water saturation indicate no loss of water even after many hundreds of days of gas testing at elevated pressures. Moreover, hydraulic conductivity tests performed before and after gas flow through these samples did not exhibit notable differences.

Around a repository, experimental evidences from laboratory and in situ testing (e.g. Zhang et al., 2013; Svoboda and Smutek, 2013 among others) confirmed that discontinuities in the disturbed zone can act as preferential gas transport pathways that become active above a threshold gas pressure and shut down once the pressure drops. Besides the disturbed zone, the possibility of localised gas transport along interfaces between repository components has also been mentioned by several participants to the symposium (see for example Popp et al., 2013). Ample evidence has been collected in previous EC projects and again in FORGE of spontaneous sealing of discontinuities in confined clay systems (e.g. Zhang et al., 2013). A priori, discontinuities activated by gas transport should thus not act afterwards as preferential groundwater and solute transport pathways. Given that gas release may spread over quite long periods, it should be checked, however, that other perturbations will not in the meantime negatively affect this "self-sealing" capacity.

While no experimental evidences of two-phase flow have been collected in FORGE experiments on low permeability porous media – typically clays and bentonite – close to saturation with water, this gas transport mode can be relevant for materials with a lower gas entry pressure, such as sand-bentonite mixtures and high porosity concrete. In that case, it can be possible for a pressurized gas phase to displace the porewater, without the creation of new, gas-specific pathways. In unsaturated conditions, Darcian gas flow is also possible even for low-permeability materials provided that continuous, water-free pathways are maintained. See Manca et al. (2013) for an example of controlled gas release test in a bentonite-sand mixture and Villar, et al. (2013) for the determination of water and gas permeabilities in unsaturated cementitious materials. An additional challenge presented by cement-based systems is the complex, dynamic physico-chemical evolution of the materials. This evolution can have different effects on porosity and gas permeability as these can be affected, for instance, by crack formations but also pore clogging due to formation of new phases (Purser et al., 2013).

4. Process-level modelling

Many presentations at the FORGE symposium project dealt with interpretative modelling of laboratory and in situ gas transport experiments. In addition, a system-level modelling benchmark exercise was performed (Ahusborde et al. 2013).

The models used are in most cases based on the concept of two-phase flow through continuous porous media or extensions of this concept. Pressure-dependent porosity and permeability are often used as a way to better reproduce a rapid increase of gas flow above a threshold injection pressure. Other participants suggested to explicitly couple two-phase flow transport models with poro-mechanics models (e.g. Arnedo et al., 2013; Gerard et al., 2013b; Olivella et al., 2013; Shao et al., 2013 and Yamamoto et al., 2013) to better take into account the role of the evolving stress field and, possibly, to better reproduce the effect of pathway dilation on gas transport at a system-relevant scale. To take into account material anisotropy and the possible presence of preferential gas transport pathways along a given orientation, Arnedo et al. (2013) and Gerard et al. (2013b) used embedded fracture permeability models, adding joint elements to a continuum model.

In many cases, experimental results – principally observed gas pressure evolutions – could be reproduced reasonably well by the models used in FORGE. However, experimenters and most modellers were cautious with respect to predictive capabilities at this point. Convincing, scientifically based, process-level models for dilatant gas flow might still be missing at this point.

5. Radiological impact

Gas can possibly affect radiological performance of a repository in two ways. The first one corresponds to the direct release of radioactive gas itself. The second is of an indirect nature: if large amounts of inactive gas are produced in the repository, these might enhance radionuclide transport within the disposal system by displacing contaminated water, acting as carrier gas for volatile nuclides or inducing damage to the multi-barrier system as a result of excessive pressures.

Radioactive gas migration through, and release from a repository was not systematically investigated in FORGE. Nevertheless, some considerations could be formulated on the occasion of this symposium. Due to a usually large initial inventory and a longer half-life, ^{14}C can be of safety relevance depending on its degree of conversion to methane and on the period of confinement and transport of this gas in the geological system before it reaches the biosphere. The behavior of ^{14}C is a matter of further research and will be addressed in the CAST project currently in negotiation as part of the EC Seventh Framework Programme. Depending on the concept, scenarios in which a fraction of volatile iodine can be released from the waste could be considered. However, the contribution of radioactive gas to the total dose rate in the biosphere is usually considered minor, except for dry systems in which radioactive gas releases can make up most of the radiological impact.

The influence of non-radioactive gas on radionuclide transport depends on the gas production rate. If the gas production rate is low enough to be dissipated by dissolution in groundwater and diffusion then the radionuclide transport is not affected. If conditions in the repository are such that two-phase flow can develop, gas might expel amounts of contaminated water but these effects can be bounded. In case of gas transport by pathway dilatancy, experimental evidence suggests that little water is moving with gas in clay materials (Jacops et al. 2013) resulting in a low impact. Should gas breakthrough occur, the well-documented self-sealing properties of a bentonite-based barriers and/or clay host rock should prevent the persistence of a preferential open pathway.

6. Treatment in performance assessment

The dominant gas generation process for geological disposal of vitrified high-level waste and spent fuel is anaerobic corrosion. In Performance assessment (PA), the rate of gas generation by metal corrosion is generally determined as the product of the surface area and the corrosion rate. A large body of knowledge with respect to metal corrosion rates in saturated systems is available. Estimations of metal surface areas are not straightforward for certain ILW and LLW waste types which may lead to over-conservative hypotheses in some cases. Corrosion rates in unsaturated systems like in salt formations can be limited by the availability of water.

Gas production by a possible bio-chemical degradation of organic wastes could be represented in PA using conservative estimates based on limiting factors or simple assumptions, assuming for instance full conversion into ultimate degradation products.

Diffusive transport of dissolved gas through groundwater is a well understood and easily modelled mechanism. Models are also available to represent two-phase flow when it applies. Extension of two-phase flow models coupled to mechanical poro-elastic models have been proposed as closer representation of pathway dilatancy. However, as stated in GASNET [European Commission, 2003], physically consistent process-level models are required to better support the use of conventional continuum models in PA.

7. Strategy

In order to address the uncertainties in the safety case, different strategies exist. With respect to the source term uncertainties can be reduced by the estimation of more realistic corrosion, dissolution rates and specific surfaces. A second option is to optimize the design of the repository, by (i) minimizing the amount of gas producing materials, and/or (ii) minimizing gas generation rates by optimization of geochemical conditions and/or (iii) limiting the availability of water within the engineered barrier system.

Comparable strategies can be followed with respect to gas transport. A reduction of the uncertainty associated with complex gas transport modes might be reached by the choice of EBS materials through which gas transport can be more easily characterized. The design for gas transport might be optimized by maximizing the exchange surface for transport of dissolved gas, *i.e.* adapting the repository geometry and/or organizing storage & transport capacities to cope with gas production rates, e.g. by the choice of porous, non-compacting backfilling material, or by explicitly providing long-term stable gas evacuation pathways.

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Why Gas is an Important Consideration in a Radioactive Waste Disposal Safety Case - Key Messages from EC FORGE project

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Introduction and Background

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.

Considering the performance of the repository as a whole (waste, buffer, engineering disturbed zone, host rock), and in particular its gas transport properties, key issues for further study are: dilational versus visco-capillary flow mechanisms; long-term integrity of seals, in particular gas flow along contacts; role of the Engineered Disturbed Zone as a conduit for preferential flow; laboratory to field upscaling. Of particular importance is 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.

Understanding gas generation and migration is thus vital in the quantitative assessment of repositories and is the focus of the research in the integrated, multi-disciplinary, European Commission "**F**ate **O**f **R**epository **G**as**E**s" project (FORGE) - 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.

The EC FORGE project builds on EC GASNET project (2003) and work thereafter; driven to meet aims of EC waste management organisations. Uncertainties identified in GASNET relevant to FORGE include:

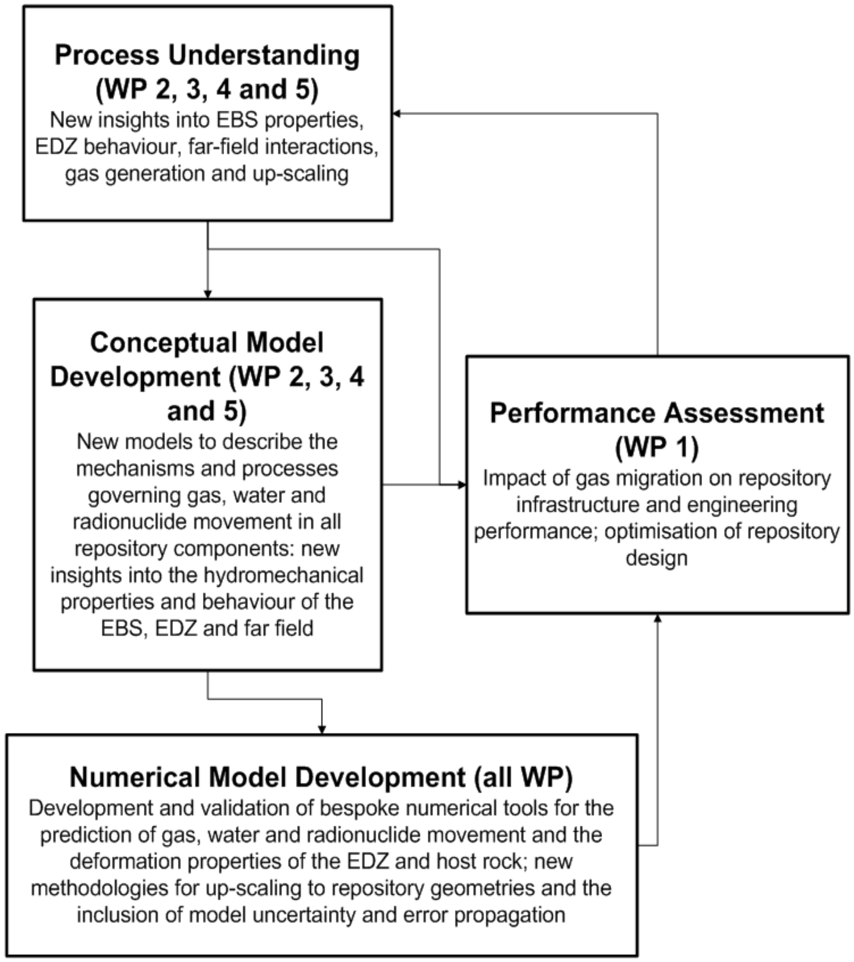
- The definition of long-term corrosion rates of ferrous metals under repository conditions (considered in FORGE Work Package 2);
- A better understanding of the processes and mechanisms governing gas migration in clay-based engineered barriers and host rocks (considered in FORGE Work Packages 3, 4 and 5);
- The effect of elevated gas pressures on the movement of groundwater and aqueous borne contaminants (considered in FORGE Work Package 4);
- The role of gas on the evolution of the near field and the EDZ (considered in FORGE Work Package 4);
- The possible coupling of effects to compromise repository performance (considered in FORGE Work Package 1).

To address these fundamental issues, the FORGE project was structured in such a way as to provide new insights into the processes and mechanisms governing gas generation (WP2) and migration (WP3-5) through the acquisition of new experimental data, aimed at repository performance assessment (WP1). FORGE also aimed to help address the paucity of high-quality data currently available for future activities such as benchmarking and validation of numerical codes for the quantitative prediction of gas flow, the development of HM (Hydrogeological – Mechanical) models for the prediction of EDZ and near-field processes and to assist in the assessment of the long-term evolution of the potential geological barriers.

The FORGE project consists of five Work Packages (WP), which are:

- WP 1 Treatment of gas in performance assessment;
- WP 2 Gas generation;
- WP 3 Engineered barrier systems;
- WP 4 Disturbed host rock formations;
- WP 5 Undisturbed host rock formations.

The structure of the EC FORGE project is shown below:



The aims of each Work Package, and key messages derived from work undertaken in FORGE, are outlined in the following sections.

WP2 Gas Generation

Aim: *To study the effect of redox, temperature, presence of bentonite on hydrogen generation and associated corrosion of steel, as well as the effects of gamma radiation, on the generation of hydrogen during the corrosion of steel in clay porewater*

Key Messages from work undertaken in FORGE WP2:

- Experiments on carbon steel showed:
  - Corrosion rate in compacted bentonite in neutral pH disposal environments is greatly accelerated (up to tens of µm/a) at least in the first month compared to the rate in bentonite porewater (the long-term corrosion rate has been determined in several prior studies to be a few µm/a).
  - Initial corrosion rate is significantly higher at elevated temperature (70°C) than at lower temperatures. The rate decreases rapidly, no significant temperature dependence after approximately one month. This should not be a significant issue for post-closure assessment.
  - Gamma radiation at dose rates in the range of 50-100 Gy/h enhances both hydrogen production and the corrosion rate – need to extrapolate to 'general' repository conditions of lower dose rates or the presence of clay.

- In the case of cementitious environments, further study may be required for some conditions, e.g. changes to corrosion rates and associated gas generation rates influenced by:
  - pH changes and loss of carbon steel passivity;
  - Effect of organic degradation products.
- Microbial issues where further (in situ) studies may be warranted:
  - Microbial corrosion of steel and copper: not studied in FORGE, but considered in prior laboratory studies.
  - Utilisation of hydrogen as an electron donor by microbes (e.g. sulphate-reducing bacteria) - this process is normally conservatively ignored in assessing gas pressure build-up, but can reduce gas pressure.

- Overall, gas generation processes are generally well-understood for different types of metals, including how generation rates are globally affected by changes in experimental conditions / conditions in repository.
- Effects of higher short-term gas production rates need to be checked for the specific EBS design and hydraulic boundary conditions.

WP3 Engineered Barrier Systems

Aim: *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*

Key Messages from work undertaken in FORGE WP3:

For Bentonite-based barriers:

- Two-phase flow is the dominating transport mechanism in unsaturated or partially saturated bentonite (also for saturated sand-bentonite mixtures if the sand content is sufficiently high).
- Classical two-phase flow models cannot correctly represent gas migration in a compacted saturated bentonite.
- High gas pressure may significantly delay the saturation of the bentonite.
- If the gas pressure reaches a higher value than the pressure in the bentonite a mechanical interaction will occur, leading to either:
  - Consolidation of the bentonite; and/or
  - Formation of dilatant pathways (allowing gas mobility).
- Dilatant pathways exhibit spatio-temporal evolution - localised outflows during gas breakthrough and no measurable desaturation in any test samples.
- A detailed stress analysis is required to capture the transition from consolidation to dilatant pathway formation; effect is clearly geometry dependent, but other factors may be involved:
  - When the gas pressure reaches the sample pressure (e.g. as seen in LASGIT);
  - At an overpressure at about 20-30%;

    - At pressures 2-3 times higher than the sample pressure.
- Self-sealing of bentonite always occurs after a gas migration event

For concrete barriers:

- For other materials studied in FORGE, the deformation of the solid phase is less important and therefore two-phase flow can be considered as the main mechanism for gas flow even near water saturation. Improved database and process understanding has been gained for:
    - Gas permeability in concrete under different conditions;
    - How carbonation, from CO2 gas, will affect the permeability of concrete.

Gas migration in interfaces

- Gas will generally move along the interface between the clay and another material in a saturated system (because gas entry pressure in the interface is generally lower then in the surrounding materials).
- Interfacial flow depends on surface roughness, geomechanical properties, wettability, etc of the materials
- In a saturated system bentonite/bentonite interfaces will seal (or heal - as demonstrated by the development of cohesion) and will not be preferential pathways for gas. Gas pressure induced re-opening of healed interfaces is not observed.
- Shear displacement in the contact zone due to pressurisation of a plug will not result in mechanically-induced pathways because the saturated bentonite behaves plastically.

WP4 Disturbed Host Rock Formations

Aim: *To examine the evolution of the EDZ around the backfilled underground structures of a disposal/storage facility as a potential escape route for gases (and dissolved radionuclides)*

Key Messages from work undertaken in FORGE WP4:

- Our understanding of fundamental processes governing gas flow in the EDZ has significantly improved as a result of FORGE. Relevant mechanisms affecting gas transport properties include stress and pore pressure conditions, stress history, orientation of EDZ fractures to the stress field, strains and hydro-chemical porewater-rock interactions (e.g. swelling, precipitation, filtration, erosion).
- Gas flow is initially focussed within the repository excavation damaged zone (EDZ), the network of discrete EDZ fractures acting as a preferential pathway for gas migration. Flow in EDZ highly localised along the largest EDZ fractures, exhibiting a complex inter-dependence between fracture transmissivity and the distribution of radial stress around tunnel.
- Tests show temporal evolution of flow behaviour.
- The evolution of the EDZ as a gas transport path is controlled by a variety of features, events and processes, such as the connectedness of EDZ fracture network, the resaturation of the repository near field, pore pressure recovery, build-up of swelling pressures in the clay-bearing EBS, rock creep in response to the local stress field and by the nature of the actual gas source term (gas generation rates, gas species).
- Linking to WP3, models have been developed that consider pathway dilatancy using a number of different approaches in order to better represent the data and to improve simulations.
- Generic modelling studies emphasised the relevance of the spatial variability of rock properties in gas transport simulations. The inclusion of subscale variability of certain rock properties (strength, permeability) enabled models to simulate gas flow localisation.

WP5 Undisturbed Host Rock Formations

Aim: *To establish the conditions under which the different gas migration processes are dominant; to identify how those processes can be modelled; to determine the values of the main parameters; and to establish whether an impact on the long-term safety as a consequence of enhanced radionuclide transport through the host rock could be expected.*

Key Messages from work undertaken in FORGE WP5:

- Major experimental challenges for undisturbed clay host rock have been identified and considered in improved test protocols.
- Difficult to create a gas flow into an intact clay host rock as the gas entry pressure and water retention is very high.
- Both laboratory and in situ experiments show that very little water is displaced by gas phase flow through undisturbed clay.
- Evidence for hydro-mechanical coupling and pathway dilatancy found as gas transport mechanism in very carefully-performed laboratory experiments in which all mechanical and hydraulic boundary conditions are well controlled and sample is near water saturation (or saturated).
- As its gas entry pressure is generally lower, free gas will preferentially flow through the EDZ rather than in intact rock in a clay host rock.
- Implicit and explicit formulations have been tested to introduce hydro-mechanical coupling in gas transport simulations (also WP4-relevant):
    - Implicit formulations are based on an extension of classical two phase flow codes to cope with fluid flow and gas transport processes in deformable media. Successful applications are reported for gas permeability tests, associated with low and moderate volumetric strains in response to the gas pressure build-up.;
    - Explicit formulations (fully coupled HM modeling) were needed in other cases to reproduce the main features of the experimental results.
- Measuring the water retention curves of an indurated clay (e.g. Opalinus clay) reveals new issues to be addressed in a future research programme:
    - No significant difference was noted between applying matrix or total suction, suggesting that osmotic suction (i.e. porewater chemistry) has only a minor impact on gas transport. Dedicated investigations need to assess impact of porewater chemistry on the water retention behavior of indurated clays.
    - Increasing mechanical stress seems to further increase the capillary strength value which in any case is already high (6 to 34 MPa). Dedicated studies of stress dependence of the two-phase flow parameters needed.

WP1.2 and 1.3 Modelling

6.1 Benchmark Studies (based on ANDRA concept):

Aim: *To test the capabilities of software tools WP 1 participants are using, and to investigate how decisions made by modellers when using these tools affect model output and its interpretation.*

Key Messages from work undertaken in FORGE WP1.2:

- Simulations of gas movement at repository scale show gas flow is very sensitive to local variations in gas transport properties: gas takes pathway with lowest resistance.
- Disturbed host rock around excavations (galleries or disposal cells) and the access ways to repository will therefore most probably act as preferential pathways for gas migration.

- During migration, free gas is always in contact with water (present in the partially desaturated pores) - dissolution can take place.
- Only a very small part of the total generated gas volume (if ever) may reach access ways as free gas.
- Overall, most of the gas is migrating by diffusion in dissolved form towards surrounding geology.

6.2 Repository Scale Gas Migration Calculations

Aim: *To undertake a suite of calculations to progress understanding in repository-scale gas migration, linking to the output of WPs 2-5. Such calculations are typically specific to gas issues in individual national programmes, although it is important that lessons learnt from these studies are communicated to other project participants in a timely manner.*

Key Messages from work undertaken in FORGE WP1.3:

- With respect to gas migration, it is possible to model a whole repository, taking into account both large and very small scale features.
- To achieve such a numerical simulation some simplifications have to be considered (e.g. no complex mechanical coupling), and upscaling techniques have to be addressed
- Overall, these simulations give good estimations of gas pressures variations but less accurate estimations of gas fluxes.
- Some attempts were made to introduce a simple 'proof of principle' mechanical coupling (in order to roughly take into account 'pathway dilation' processes), however, there is currently insufficient information to properly parameterize such a model.

6.3 Overall messages from FORGE regarding two phase flow / mechanical coupling models

- Localization of gas pathways for low permeability porous media such as indurated rock is difficult to handle with classical two phase flow models (based on generalized Darcy law for each phase, permeability depending only on water saturation).
- For small scale experiment, using two phase flow models without any coupling with mechanical effects or any evolution of rock properties due to gas pressure and/or deformations leads to low accuracy results, especially on laboratory-scale experiments in which localized pathways can be monitored.
- For large scale experiments when gas is injected in undisturbed host rock, such models seem to give a good approximation of the experimental results. This could be due to less accurate measurements in situ, but also to homogenization effect at metre scale.
    - Two phase flow models seem to give a good representation of gas migration on large scale experiments in undisturbed indurated rock.
    - At small scale, "classical" two phase flow model is not well adapted to reproduce all the details of the test.

Overall Key Messages from FORGE

Work undertaken in the FORGE project will benefit a range of customers via the provision of new information and understanding into the processes and mechanisms governing gas generation and migration. FORGE has also provided high-quality data that could be used for future activities such as benchmarking and validation of numerical codes for the quantitative prediction of gas flow, the development of HM (Hydrogeological – Mechanical) models for the prediction of EDZ and near-field processes and to assist in the assessment of the long-term evolution of the potential geological barriers.

The project has enhanced, in relation to the consideration of gas in safety case studies:

- Information and understanding;
- Methods, models and computer codes;
- Qualitative safety arguments;
- Quantitative assessments;
- Development and maintenance of expertise, collaboration.

Some gas-related uncertainties still remain – which is to be expected - and are already areas for further targeted work (e.g. EC CAST[1] proposal). Recognition and appropriate treatment of uncertainty is an important aspect of any safety case for the geological disposal of radioactive waste. Areas in which uncertainty may influence a safety case include:

- Uncertainty over future states of the system;
- Data uncertainty;
- Model uncertainty;
- Uncertainty about human behaviour.

Strategies have been developed to manage such uncertainties, ensuring that a safety case can be developed even given the presence of uncertainties. Such strategies consider:

- Demonstrating that the uncertainty is irrelevant:
  - e.g. safety controlled by other processes;
- Addressing the uncertainty explicitly:
  - representing by PDFs in a probabilistic calculation;
  - scoping effect of range of uncertainty by deterministic sensitivity calculations;
- Bounding the uncertainty:
  - making conservative assumptions;
- Ruling out the uncertainty on the basis of low probability;
- Explicitly ignoring uncertainty or agreeing a stylised approach for handling an irreducible uncertainty.

Gas-related uncertainties are not 'show-stoppers' in terms of adequate and appropriate consideration of gas in the safety case now; deployment of an approach to "Managing uncertainty" is a required component of any safety case, and is applicable to gas issues.

Overall Key Messages from FORGE are:

- Features, Events and Processes (FEPs) relevant to the consideration of gas in the safety case (the 'gas issue') are well-known, although therein there are uncertainties that need to be managed as a standard aspect of developing a safety case.

- Understanding the 'gas issue' provides coupled mitigation opportunities that can be considered on repository-specific basis, e.g. inventory optimisation, choice of materials for the Engineered Barrier System (EBS), repository design and repository operation (including repository sealing and closure).

- The relative importance of the 'gas issue' in the safety case is a function of the disposal concept under consideration, which is itself a function of the disposal inventory (including the gas source term, the approach to waste treatment and packaging, and how the packaged waste is management prior to emplacement in the repository etc) and the safety functions required to be provided by complementary barriers (e.g. EBS, geology).

[1] CArbon-14 Source Terms

- Repository-derived gas needs to be considered at an appropriate level in all repository safety cases. This can be done on the basis of existing knowledge.

- **We have a sufficient understanding of the 'gas issue' now to be confident, on the basis of work undertaken in the EC FORGE project and complementary studies, that repository-derived gas presents no 'show-stoppers' to implementing geological disposal in a wide range of disposal systems.**

Gas Intrusion in Bentonite – Results of Small Scale Experiments

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Summary

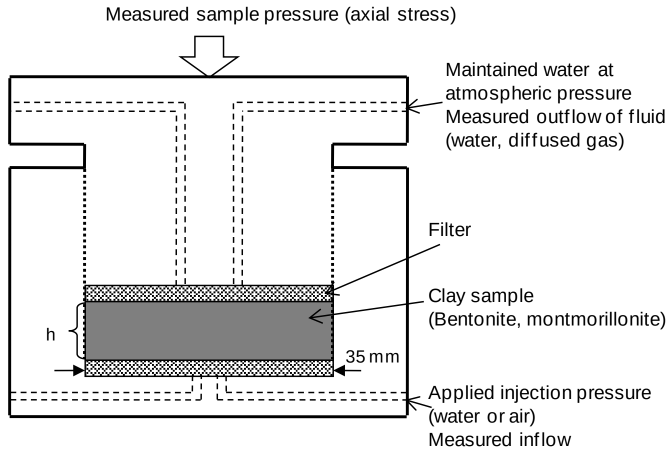
A large set of gas (air) pressurization tests have been performed on small scale (cm) bentonite samples. The results clearly shows that mechanical interaction between the gas phase and the clay body only takes place when the gas pressure exceeds the initial pressure of the bentonite sample. It was further demonstrated that the migration of gas after intrusion depends strongly on test details such as injection geometry and sample density. In many cases gas breakthrough events was induced, while in some cases the gas phase consolidated the clay body in a controlled fashion, with only diffusive gas transport through the clay. In the case of breakthrough, the gas usually followed a preferential path formed at the interface between clay and the wall of the test cell. The results are all in accordance with a montmorillonite interlayer-only interpretation (osmotic approach) and incompatible with a "conventional" two-phase flow view of the bentonite system.

1. Introduction

An investigation on the flow and pressure response in compacted water saturated bentonite/montmorillonite systems due to external gas pressurization has been conducted. The strategy has been to use small scale samples (mm- to cm-scale) in very simple geometries in order to have full control of the boundary conditions and to be able to work relatively quickly. The saturation time for a bentonite sample basically increases quadratic with sample size and may be as large as several months even for samples on the dm-scale. In contrast, the major part of the samples used in this investigation has a characteristic length scale of 5 mm and becomes saturated in just a few days. Due to this enormous time gain, a substantial number of samples have been tested and statistical knowledge has been gathered on the gas intrusion process – something which to a large extent has been lacking in the past.

2. Methodology

Gas intrusion experiments in water saturated bentonite (MX-80) and purified montmorillonite (calcium and sodium type) have been performed in constant volume test cells. Cylindrical samples of diameter 35 mm and height in the range 2 – 20 mm where pressurized with air via a filter in the bottom of the test cell. Gas injection was performed both in cells with a tiny centred point-like inlet filter and in cells where the inlet filter covered the entire bottom area. At the top side the samples was contacted with non-pressurised (deionised) water via a filter covering the whole top area. A schematic picture as well as a photograph of a test cell is shown in Fig. 1. Some of the tests with point-like injection had an extra ring-like outlet filter on the circumference of the bottom area. With such a "guard filter", the path taken by the transported gas could to a certain degree be determined. The different inlet types are shown in Fig 2.



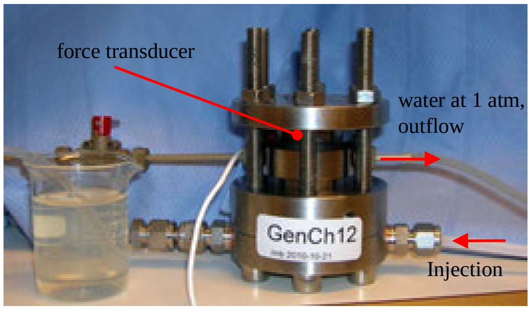


Figure 1:Test cell schematics

The axial force exerted by the clay as well as the volumetric flow of fluid out of the system was continuously measured during the course of the tests in all of the tests. The density of the samples was chosen in order to achieve a clay sample pressure in the absence of external pressurization (P_s^0) in the range 0.5 – 1.5 MPa.

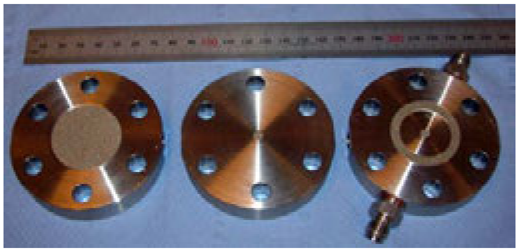


Figure 2: Filter in full bottom area (left), point-like injection filter (middle) and point-like injection filter + circumferential outflow filter (right). The circumferential outflow channel is isolated from the top outflow channel.

In total was eight different samples has been tested for gas intrusion: three MX-80 samples (two with point-injection, two with full area injection), three Na-montmorillonite samples (two with point-injection, one with full area-injection), and one Ca-montmorillonite sample (with full area-injection).

3. Results

All performed tests clearly demonstrate that there is no response in the equilibrium sample pressure for gas injection pressures below P_s^0. Consequently, the gas phase does not interact mechanically with the clay under these circumstances. In contrast, for injection pressures above Ps0 the gas phase is inevitably interacting mechanically with the clay as seen from the observation that the sample pressure always response in this case. These results show that the fundamental criterion for gas intrusion – i.e. the incorporation of a separate gas phase within the volume of the test cells – is that the injection pressure exceeds the initial pressure of the sample (P_s^0 + possible external water pressure).

The detailed system response when gas intrusion occur is quite complex and depends on many variables. Two main types of behaviour have been identified: gas breakthrough and clay consolidation. The gas breakthrough is characterized by a sudden event where the clay system "breaks" and the gas reservoir flows through the sample and is emptied within minutes. As compared to typical diffusive flux of gas, the flow in the breakthrough events is enormously larger (a factor of 10^8 or more). Figure 3 shows the pressure response in two samples in which breakthrough events were observed.

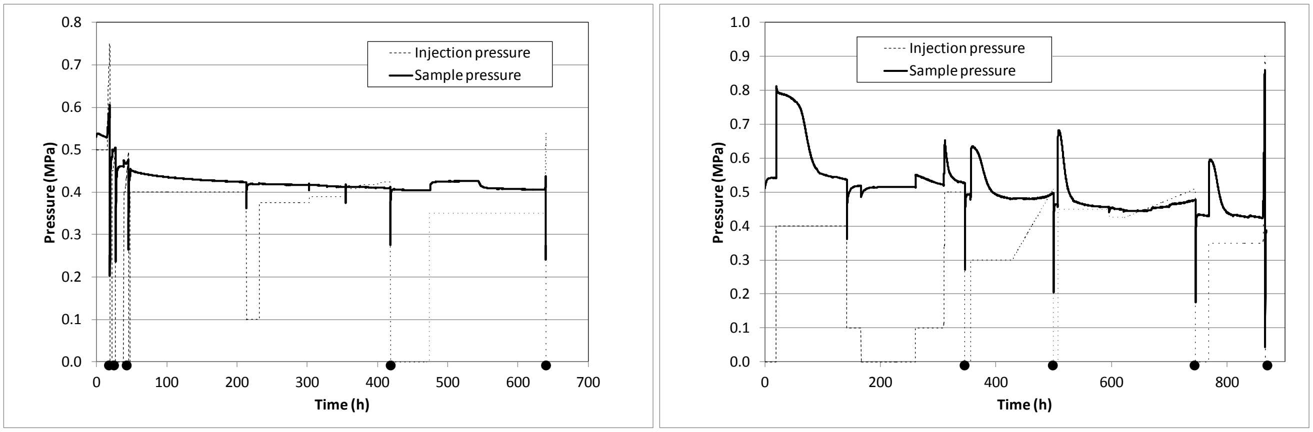


Figure 3: Sample pressure response in two samples of Na-montmorillonite in which gas breakthrough events where induced. The diagram on the left shows the pressure response in a 5 mm sample with point-like injection (see Fig. 2) and the diagram on the right shows the response in as 5 mm sample with full area-injection (Fig. 2). The hunch-like behaviour of the sample pressure response, most clearly seen in the right diagram, are due to residual water in the inlet which initially is the pressurising fluid when an injection pressure is applied (The response due to water is very different from the response due to gas[1]). The dots on the time axis show where gas breakthrough occurred. In these samples, such events were induced as soon as the injection pressure reached the value of the sample pressure. The diagrams also show that the sample pressure is independent of the injection pressure when this is lower than P_s^0.

The clay consolidation process is instead characterized by that the sample pressure equals the injection pressure while only diffusive flow of gas occurs through the sample. The gas is in this case basically functioning as a piston pushing on the clay body. That the clay body really is consolidated (i.e. decreased in volume) is evident from the pressure response as the injection pressure is released in this state (Fig. 4). The sample pressure is then falling to zero, indicating that the clay body temporarily is not filling up the test cell volume (the pressure then increases again as water is being taken up from the reservoir).

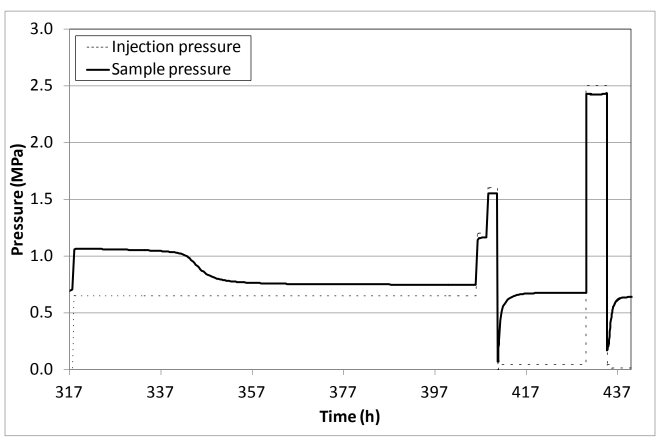


Figure 4: Consolidation by air in an MX-80 sample (5 mm, full area-injection). The initial sample pressure response (317 h – 350 h) is due to residual water at the inlet[1]. As this water is expelled and replaced by air as pressurising fluid, the sample pressure falls back to P_s^0 *(0.75 MPa) as long as the injection pressure is below this value (ca 350 h – 406 h). At 406 h the injection pressure is increased above* P_s^0 *in two steps to 1.6 MPa without inducing a breakthrough event. In this state the clay body is consolidated which is evident from the sample pressure response as the injection pressure is lowered to 10 kPa (411 h). The sample pressure basically reaches zero temporarily, demonstrating that the clay body is not filling up the test cell volume.*

In cases where diffusion of gas was the only transport mechanism, the flow mainly went through the clay and out through the top filter. In contrast, the favoured path taken by the gas during a breakthrough event was along the interface between test cell and clay body. Furthermore, it was noted in several of the samples that processes induced in the system due to intrusion of gas may occur on quite long time scales, although the sample size was rather small.

4. Discussion and conclusions

The most robust conclusion to be drawn from this study is that gas and clay inevitably interacts mechanically when the gas pressure equals (or exceeds) the pressure of the clay sample, while no such interaction takes place for gas pressure below the sample pressure. This observation is in full agreement with an osmotic interpretation of saturated bentonite [1][2] and is incompatible with a two-phase flow interpretation – the results clearly demonstrates that the interaction between a gas phase and the clay is such that the gas has to consolidate the clay to a certain degree in order to create its "own" volume within the test cell and cannot be interpreted as the gas expelling and replacing water in a pre-existing pore structure.

In situations where gas intrusion occurs (i.e. when the gas pressure exceeds the initial sample pressure) the fate of the gas phase within the volume of the test cell depends on several of the test details. Breakthrough events have been induced in many of the samples, but also states of clay consolidation and only diffusive flux have been achieved, specifically in cells with full area-injection and in samples with relatively high density. Thus, the detailed behaviour of the gas migration after intrusion is at least dependent on injection geometry, sample density and time (quick increase of injection pressure may lead to higher transient sample pressure before breakthrough). It may also be speculated that the gas migration mechanisms are further dependent on e.g. clay type and sample geometry.

Furthermore, in the specific case of gas breakthrough events, it was observed that the flow path of the gas usually followed the interface between the clay body and the test cell. Thus, this interface constitutes a preferential path for gas.

5. Acknowledgements

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Role of Interfaces in Bentonite-Block Assemblies as favoured Pathways for Gas Transport

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Summary

Gas will be created in a radioactive waste repository performance assessment, which requires quantification of the relevancy of various potential pathways. Our lab investigations give hints that in sealing plugs consisting of (dry or saturated) bentonite bricks gas will preferentially move along the interface between the clay and another material (e.g. the host rock with the EDZ). This is, because bentonite/bentonite interfaces will heal during saturation - not only seal - as demonstrated by the development of interfacial cohesion. Coevally the gas entry pressure in the contact zone to the host rock is lower than the pressure acting on the interface. With respect to overall plug integrity it can be stated that during pressurisation, shear displacement in the contact zone will not result in mechanical induced pathways because the saturated bentonite behaves plastic.

1. Introduction

As radioactive waste remains hazardous for up to one million years, the safest long term solution is to dispose of it in deep underground repositories which generally rely on a multi-barrier system to isolate the waste from the biosphere. The multi-barrier system typically comprises the natural geological barrier provided by the repository host rock and its surroundings and an engineered barrier system (EBS), i.e. the backfilling and sealing of shafts and galleries to block any preferential path for radioactive contaminants. Because gas will be created in a radioactive waste repository performance assessment requires quantification of the relevancy of various potential pathways.
Referring to the sealing plugs it is expected that in addition to the matrix properties of the sealing material conductive discrete interfaces inside the sealing elements itself and to the host rock may act not only as mechanical weakness planes but also as preferential gas pathways. For instance despite the assumed self-sealing capacity of bentonite inherent existing interfaces may be reopened during gas injection. Our lab investigations are aiming on a comprehensive hydro-mechanical characterization of interfaces in bentonite buffers, i.e. (1) between the prefabricated bentonite blocks themselves and (2) on mechanical contacts of bentonite blocks and concrete to various host rocks, i.e. granite.

2. Methodology

We used as reference material pre-compacted bentonite blocks (250 mm x 125 mm x 62.5 mm; Table 1) consisting of a sand clay-bentonite mixture. The investigations consist of:

- long-term water injection tests in a new designed oedometer cell (duration of several month up to two years) with different sample constellations (block/block resp. block/host rock) under well controlled stress and swelling conditions to provide data about

  - time dependent interface "permeability" changes (i.e. sealing) during long-term compaction and fluid injection
  - gas entry pressures and relative gas permeability changes during pressure dependent gas injection (under constant volume conditions);
- shear tests (in addition to triaxial tests) to quantify mechanical interface properties of the dry and the saturated bentonite blocks under well controlled shear forces or displacements.

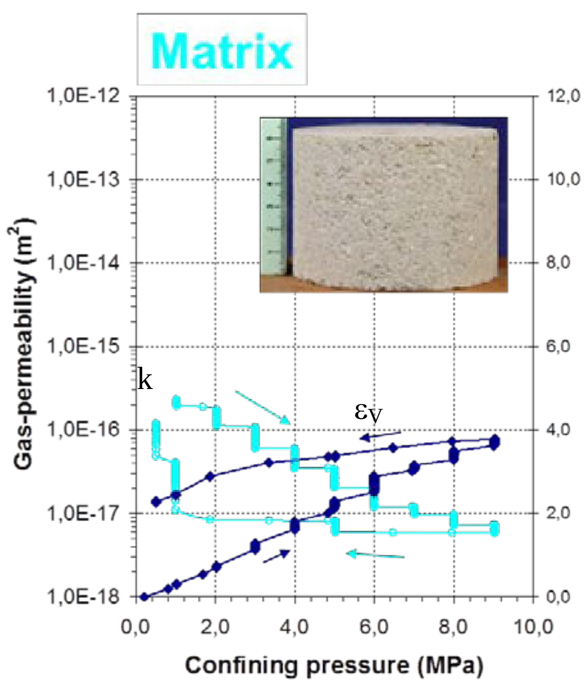
Table 1: Basic properties of the bentonite bricks (taken from Sitz, 2003.

Wetro FS40	
Compression Force (MPa)	40 – 50
Composition	Ca. 60% bentonite (calcigel) + 40% sand
Dry density (g/cm^3)	1.89±0.02
Initial saturation (-)	0.77±0.04

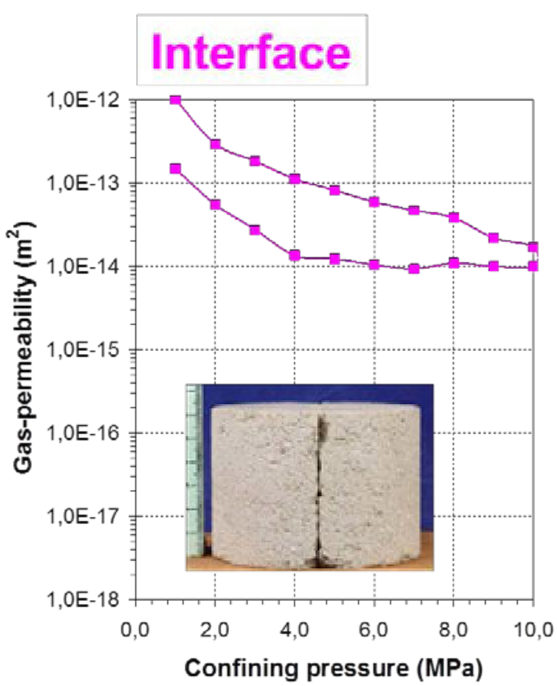
3. Results

3.1 Initial permeability of the dry material

As a prerequisite for evaluation of the later saturation tests the short-term gas permeability of the bentonite bricks was determined in a triaxial cell. As depicted in Figure 1 two different sample arrangements were used to realize axial gas-injection tests, both on the intact matrix and also on an artificial interface between two bentonite bricks. For permeability determination the injection gas pressure and the gas outflow rate were measured at various triaxial confinements.
At low confining pressure (σ_3 = 5 bar) the interface permeability amounts to ca. $1 \cdot 10^{-12}$ m², which is 4 orders of magnitude higher than the matrix permeability. By increasing of the confining pressure the permeability decreases by more than 2 orders, whereas the effect for the matrix is lower. A crack sealing of the dry material was observed during the unloading of the sample.



a)



b)

Figure 1. Permeability testing at hydrostatic loading: Gas permeability (k) and volumetric strain (ε_V) vs. confining pressure (σ_3). a) Bentonite brick matrix permeability. b) Bentonite brick with artificial interface (saw cut).

3.2 Saturation tests

For pre-saturation of the samples respectively for the later gas injection tests a new oedometer-like pressure cell was constructed facilitating both (1) controlled saturation at defined loading conditions and (2) gas flow measurements under defined axial sample consolidation respectively swelling pressures depending with different injection geometries. The cell diameter is 90 mm and the sample height is ca. 120 mm. During the saturation tests the influence of different flooding geometries and stress fields of bentonite/bentonite and also of the bentonite/granite interfaces was studied. For the bentonite/bentonite interface samples the water consumption during swelling depends not on the injection geometry and only weakly on the stress field. The flooding of the interface in the bentonite/granite sample depends on the mechanical properties and the swelling characteristics of the bentonite. In both cases after more than one year of water injection the material didn't reach a full saturation state.

3.3 Gas-injection tests

After saturation several gas-injection tests were performed on the bentonite/bentonite interfaces as well on the bentonite/granite interfaces applying different loadings. During stepwise gas pressure increase the gas breakthrough is indicated by quasi-constant gas in-flow at the upstream side or gas outflow, which could see as bubbles in a water cylinder at the downstream side.
For sample assemblies consisting only of bentonite bricks the gas breakthrough occurred only at significant overpressures, i.e. between 1 and 3 MPa higher than the minimal stress σ_{min} (Fig. 2a). The saturated aggregate behaves like a homogenous matrix that means that there is no interface characteristic. In contrast, the injection tests demonstrate that for bentonite/granite aggregates also in the saturation state the gas breakthrough pressure is significantly lower than σ_{min}. A spontaneous breakthrough (Fig. 2b) was reached after a timeframe of 16 hours, where the gas pressure was 20 percent lower than the minimal stress.

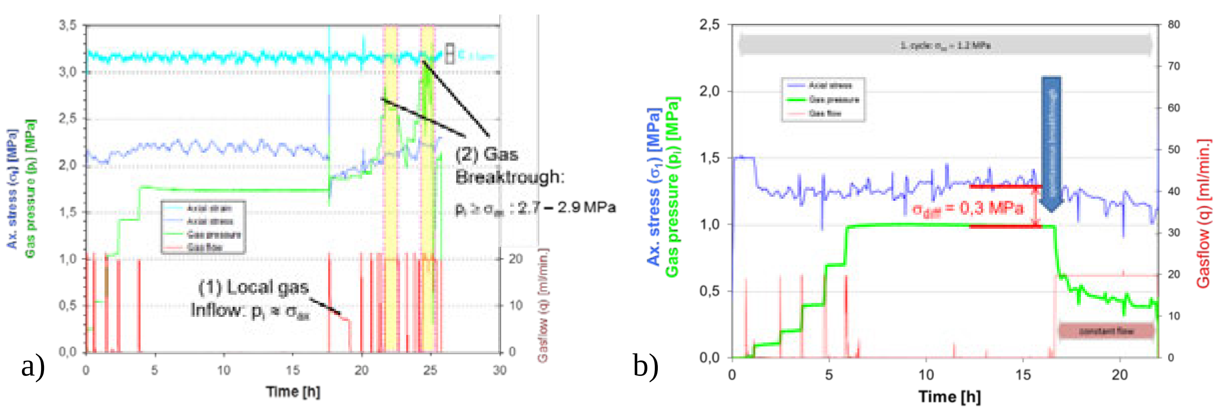


Figure 2. Results from gas breakthrough tests (measured parmeters vs. time): a) bentonite/bentonite interface; $\sigma_{ax} \approx 2.3$ MPa. b) bentonite/granite interface; $\sigma_{ax} \approx 1.2$ MPa.

3.4 Shear tests

Several shear tests with different sample combinations (bentonite/bentonite interfaces, bentonite/granite interfaces and compact bentonite) in the initial dry and in the saturated stadium have been performed. Most important, the shear test results of the bentonite/bentonite interfaces shows cohesion, so real healing is confirmed.

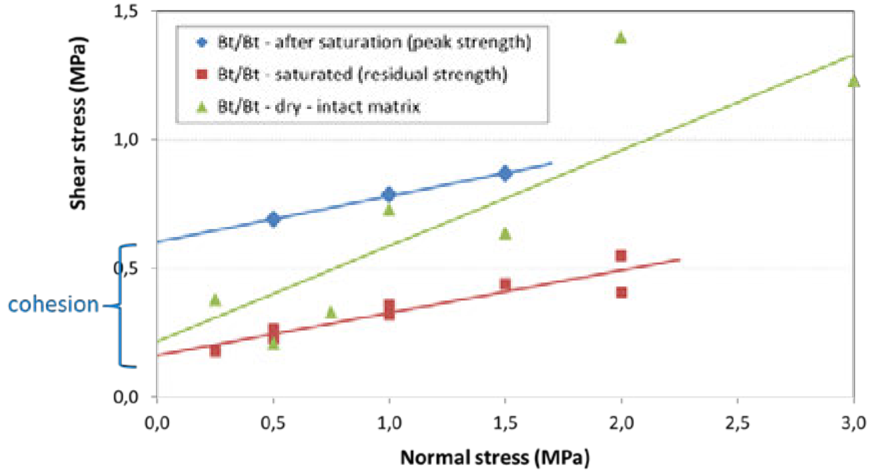


Figure 3. Mechanical properties of the dry and saturated compact bentonite and also for the dry and saturated bentonite/bentonite interfaces

4. Conclusions

The performed lab investigations cover a wide field of hydro-mechanical properties of bentonite bricks, which represent a favorable option for constructing sealing plugs in different host rock environments. In summary the following conclusions can be drawn:

- At dry conditions gas flow along interfaces is at least 4 orders higher than through the matrix, i.e. interfaces will be the preferred pathway in an unsaturated system. Increase of confinement (unsaturated case) significantly lowers the gas flow but the effect is more pronounced for interfaces $\Rightarrow$ at least partial crack sealing is demonstrated
- Saturation of bentonite blocks is not affected by the interfaces and only weakly by the acting confining pressure. However, in a saturated system bentonite/bentonite interfaces will heal – not only seal - as demonstrated by the development of cohesion. Thus saturated bentonite blocks will not act as preferential pathways for gas. In addition, gas pressure induced re-opening of healed interfaces was not observed.
- Despite saturation of the buffer gas will generally move along the interface between the clay and another material (e.g. the host rock with the EDZ). Because the gas entry pressure in the contact zone is lower than the pressure acting on the interface the saturation does not seem to affect significantly the transport mechanisms.
- The gas break through results in stationary flow but no significant effect on the total stress is measured, probably due to the localized gas flow.
- For the saturated bentonite matrix the measured gas pressures (at gas threshold) under constant volume conditions significantly exceed the sum of the swelling pressure and externally-applied

porewater pressure. The assembly of swelled bentonite blocks behaves like a homogenous matrix, i.e. no interface characteristics.

- Shear displacement in the contact zone due to pressurization of the plug will not result in mechanical induced pathways because the saturated bentonite behaves plastic

5. Acknowledgement

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3D Gas Transport Model of LASGIT Gas Injection Tests

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Geofirma Engineering, Canada

Summary

The LASGIT (Large Scale Gas Injection Test) project is a full-scale test of gas transport in bentonite at the Äspö Hard Rock Laboratory in Sweden. The experimental set-up consists of a full size copper container surrounded by bentonite buffer within an excavated deposition hole in crystalline rock. The purpose of the LASGIT experiment is to improve the understanding of full-scale gas migration through the bentonite buffer under repository conditions and to validate different modelling approaches which may be used in future performance assessments. There is significant evidence that gas flow in compacted bentonite often occurs along discrete pathways, rather than as conventional 2-phase flow. The current gas-transport modelling approach selected for the LASGIT experiment uses T2GGM, a modified version of TOUGH2 with optional gas generation model, with modifications to the code to approximate dilational gas transport mechanisms.

A 3D model representing the full bentonite ring was developed in attempt to qualitatively represent experimental results of the gas injection test conducted in 2009. The wall sensor where gas breakthrough is most readily apparent is on the opposite side of the deposition hole from the injection port, and gas breakthough at this sensor occurred approximately 1.5 days after pressure at the injection port starts to drop. Due to the complexity of the gas propagation behaviour, the goal of this model was to simulate the pressure drop at the injection port, and a pressure-rise at the rock-wall, preferably not immediately adjacent to the injection port, within a similar time frame to experimental results.

The model successfully simulated the pressure drop at the injection port; however, the gas transport path was too diffuse and propagated too slowly, with gas pressure rising at the rock wall immediately adjacent to the injection port in a minimum of 5 days. A heterogeneous permeability field was required in order to obtain the relatively short travel times between the injection port and the rock-wall. Several modifications to the model are recommended to improve the model's ability to simulate a distinct gas pathway at a location not immediately adjacent to the injection port.

1. Introduction

The LASGIT experiment examines full-scale gas transport in the bentonite buffer surrounding a copper container. The container and bentonite buffer is located within an excavated deposition hole in granite rock within the Äspö Hard Rock Laboratory in Sweden. Neon gas is injected into the bentonite through ports installed in the surface of the copper container. Modelling the gas injection tests of the

LASGIT experiment is intended to improve the understanding of full-scale gas migration through the bentonite buffer under repository conditions and to validate different modelling approaches which may be used in future performance assessments.

Gas flow in compacted bentonite by two-phase flow processes is limited, and there is considerable evidence that gas flow often occurs along discrete pathways, referred to as dilational flow or flow through micro-fractures (Senger and Marschall, 2008 [1] , Cuss et al. 2011 [2]). The modelling described here uses the two phase flow code T2GGM (Quintessa and Geofirma, 2011 [3]), a TOUGH2 code with optional gas generation capabilities, with modifications to abstract the processes of dilational flow through pressure-dependent permeability and capillary pressure. Permeability (k) is modified linearly based on input pressure thresholds (P1 and P2) and scaling factor (kfactor):

$$k = k_0(1 + (kfactor - 1)\left(\frac{P - P1}{P2 - P1}\right)) \quad where\, k_0 \le k \le k_0 \times kfactor$$

Capillary pressure (P_c) is modified based on the pressure-dependent permeability change,

$$P_c = P_{c0}\sqrt{\frac{k_0}{k}}$$

Alternate pressure-dependent permeability and capillary pressure functions are possible. mView, developed by Geofirma Engineering, was used for model pre- and post-processing.

The work described here focuses on the 2009 gas injection tests (Cuss et al., 2011[2]). Future modelling work this year will analyze and model the 2010/2011 gas injection. In the 2009 test, neon gas was injected at an injection port in the lower array (LFA3). 1.5 days after a pressure drop was observed at LFA3, a pressure response was observed at a pressure transducer on the rock wall farthest from the injection port (UR907). 3.5 days after the pressure drop at LFA3, a pressure response was seen at the injection port on the other side of the container, LFA1 (near the rock-wall where the first pressure response was observed). A pressure response was also observed at the bottom of the deposition hole 15.5 days after the pressure drop at LFA3. The extended time between the pressure drop at the injection sensor and the pressure responses at the rock-wall and other sensors suggests that the pressure drop at the injection sensor is due to an increase in the volume available for occupation by the neon gas, rather than the establishment of a permeable gas filled channel connected to the surrounding formation.

2. Methodology

A 3D model was developed representing the full circumference of the bentonite buffer surrounding the container, and limited to approximately 1 m above and below the injection port. The top, bottom and sides (including the rock boundary) were no flow boundaries as a modelling simplification, to allow the modelling work to focus on the goal of qualitatively modelling gas migration within the bentonite, simulating the pressure drop at the injection port, and a pressure increase at the rock wall. The rock wall no-flow boundary condition would cause the modelled pressure increase at the rock wall to increase with time, rather than level out as in the experiment.

Previous modelling results suggested that initial water saturations in the bentonite can have significant effects on the model results. Since the initial water saturation conditions in the bentonite are not known, a simple 2D saturation model was developed in attempt to improve the estimate of initial saturations in the bentonite for the 3D model. The model was executed from time zero of the experiment, considering different initial water saturation conditions in the bentonite, and different permeable interfaces between the container and the bentonite buffer. Each scenario was evaluated based on a hydraulic shut-in test at 1300 days. The drop in pressure at the injection ports during this hydraulic shut-in test provides an indication of the degree of saturation of the bentonite, due to the suction pressure observed during the test. While several scenarios were possible, a scenario using a variable permeability at container interface (starting at a permeability of 10^{-18} m^2 and ending at 10^{-20} m^2 at 1000 days) and variable initial saturations (initial saturation varying per bentonite block) was selected for use as initial conditions for the 3D modelling.

3. Results and Discussion

Using a homogeneous bentonite permeability field, a pressure increase at the rock wall was not observed until approximately 78 days after the pressure drop at LFA3. As might be expected, the gas travels in a diffuse pathway towards the rock-wall immediately opposite the injection pathway. Of particular interest in this slow homogeneous model is the impact of initial saturations. Once the gas travels through the saturated bentonite near the injection port, the gas pressure increases around the full extents of the bentonite ring where initial gas saturations are present, before any gas reaches the rock wall. In the model, gas reaches the far side of the bentonite ring from the injection port (middle of the ring, not the rock wall) 18 days, after the gas pressure begins to drop, longer than the observed 1.5 days to the rock wall at UR907. See Figure 1 for gas pressure and gas saturation profile after 30 days.

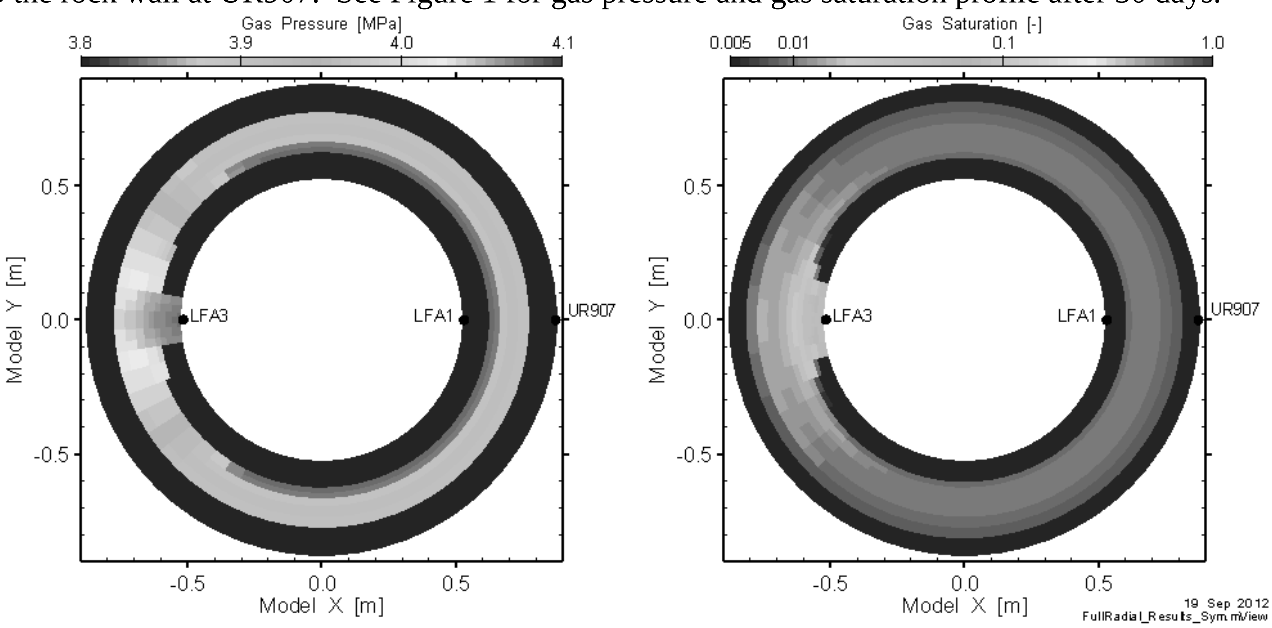

Figure 1: Gas pressures and saturations in bentonite 30 days after drop in pressure at injection port. Note that UR907 is located 30 cm above the injection ports.

Three heterogeneous permeability fields were generated, each a different realization of the same normal permeability distribution with the majority of elements having permeabilities between 10^{-20} and 10^{-22} m^2. The intent of the heterogeneous permeability fields is to provide a more discrete pathway through the bentonite for the gas, which would also reduce the time to reach the rock-wall. While the heterogeneous permeability field was successful in reducing the travel time for gas to reach the rock-wall (closest to the injection port) to 5 days (see Figure 2), the pathway was still diffuse rather than discrete.

The presence of a container-bentonite interface is a possible explanation for gas breakthrough at the rock-wall at locations far from the injection port (UR907); however, it is not considered a robust explanation given that: (1) gas breakthrough occurs at UR907 before LFA1, and (2) gas breakthrough does not occur at LFA2 or LFA4, the injection ports immediately between LFA3 and LFA1. Initial condition modelling also found that interfaces with permeabilities more than one order of magnitude higher than the bentonite buffer resulted in rapid saturation of the bentonite, which is inconsistent with the hydraulic shut-in test. A test case using an interface with a permeability enhancement of one order of magnitude (in addition to a heterogeneous permeability field) found a greater but more immediate drop in pressure at the injection port; a longer time of 10 days before a pressure response at the rock wall was predicted (still immediately adjacent to the injection port), and pressure responses at LFA2 and LFA4 within approximately 100 days.

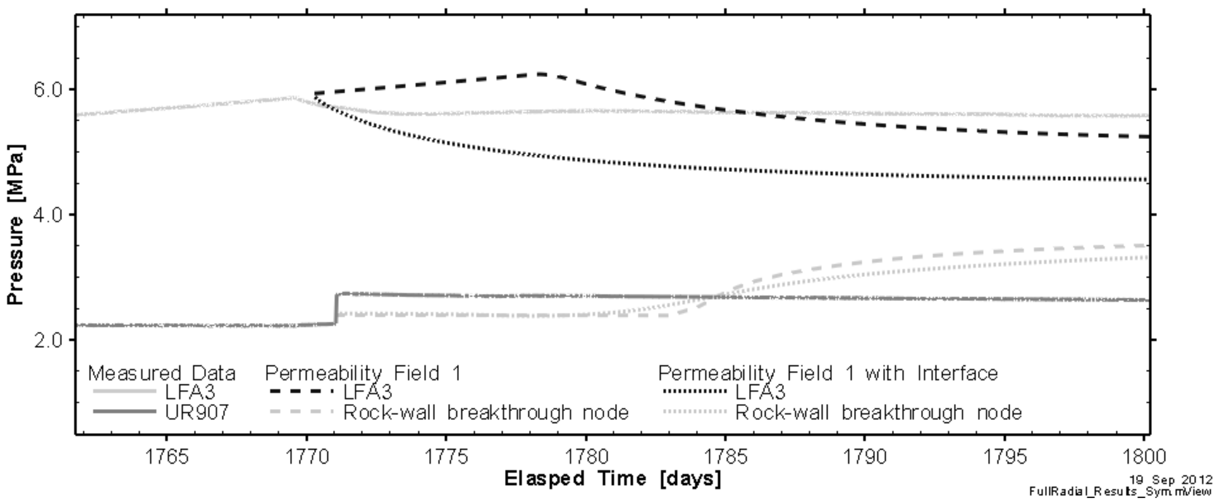

Figure 2: Gas pressures at the injection port (LFA3) and rock wall, comparing measured data (solid lines) to simulation data (dashed lines) for a heterogeneous bentonite permeability field.

4. Conclusions

Further refinement of the model to represent dilational flow is required in order to qualitatively represent the LASGIT experimental results. The modelling to date is promising in its ability to model gas breakthrough into the bentonite at the injection port; however, the gas pathway is too diffuse and propagates too slowly. There are many possible refinements to improve the model, including changing the pressure-dependent permeability and capillary pressure functions or including a stress-dependent permeability/capillary pressure function.

5. Acknowledgements

This modelling work was supported by the Nuclear Waste Management Organization (NWMO), Canada.

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Gas flow in compact bentonite

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Summary

Laboratory experiments conducted on compacted bentonite have shown strong coupling between gas flow and the local stress field with a clear hydromechanical response observed at the onset of gas movement, attributed to dilatant pathway formation. The Large scale gas injection test (Lasgit) was initiated to investigate and validate the up-scaling of the laboratory observations. In the 8-year history of Lasgit a total of three gas injection tests have been conducted. System behaviour is seen to be similar between the laboratory and field scales, although the stress states seen at field scale are lower than those experienced in the laboratory.

1. Introduction

In recent years, significant effort has been placed on defining the phenomenological controls governing the movement of gas in compact engineered bentonite. These activities have focussed on defining the flow of gas at different scales, combining results from on-going laboratory experiments with those derived from large scale field test.

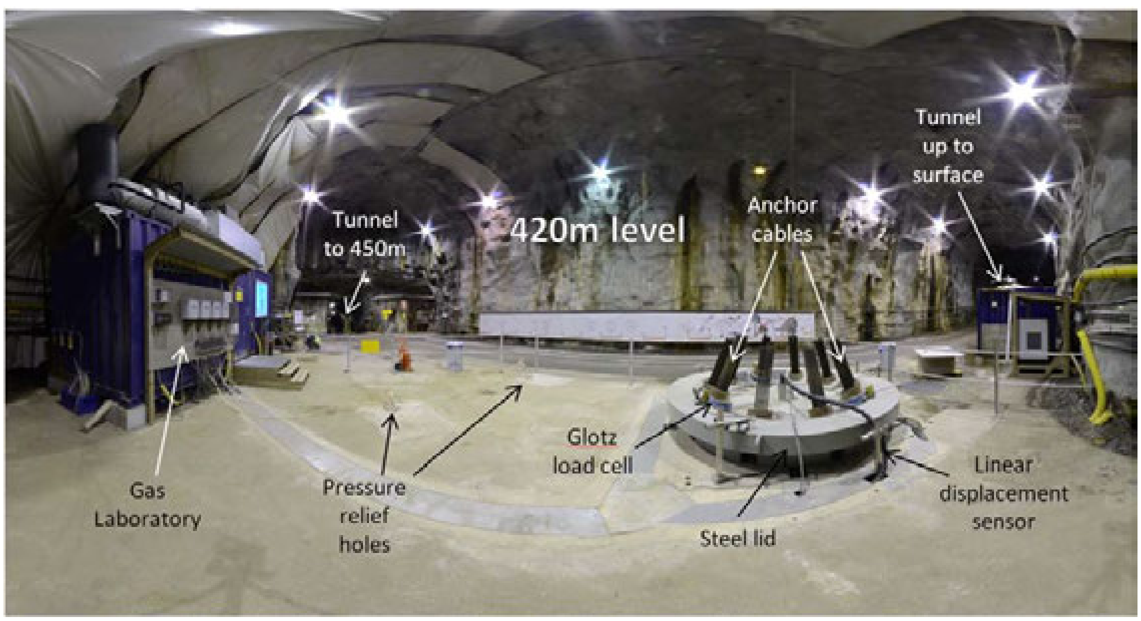

Figure 1: A panoramic view of the Large scale gas injection test (Lasgit) test site located 420m below ground at the Äspö Hard Rock Laboratory in Sweden. The photo shows the position of the deposition hole, gas laboratory, pressure relief holes and some of the instrumentation attached to the steel lid.

2. Methodology

An important tool in the development and validation of conceptual models for gas flow in bentonite is the Lasgit experiment (Cuss *et al.* 2011)[1]. Lasgit, which stands for large scale gas injection test, is a full scale demonstration test located at 420m depth within SKB's Äspö Hard Rock Laboratory (HRL) in Sweden. The primary objectives of Lasgit are to provide quantitative data to improve process understanding and test/validate modelling approaches used in performance assessment. This experiment has been in continuous operation since February 2005 and while the first two years focused on artificial hydration of the buffer, subsequent activities have examined the evolution in flow behaviour during successive gas and water injection events.

3. Results

To date, three gas injection 'campaigns' have been undertaken, as shown in Figure 2. In each of these, baseline hydraulic properties were determined before and after gas injection. In tests one and two, large diameter filters located on the lower canister array (Sellin & Harrington, 2006)[2] were selected as points of injection. In each gas experiment, a small precursor flow was noted prior to major gas entry. However, this flow practically ceased when gas pressure was held constant and was only reinitiated once pressure again began to rise. At present the exact nature of this flow remains unclear, but may stem from the displacement of 'lower' density clay which has expanded into the initial void around the canister, or, from hydraulic disequilibrium related to the non-uniform development of stress and porewater pressure within the bulk of the clay.

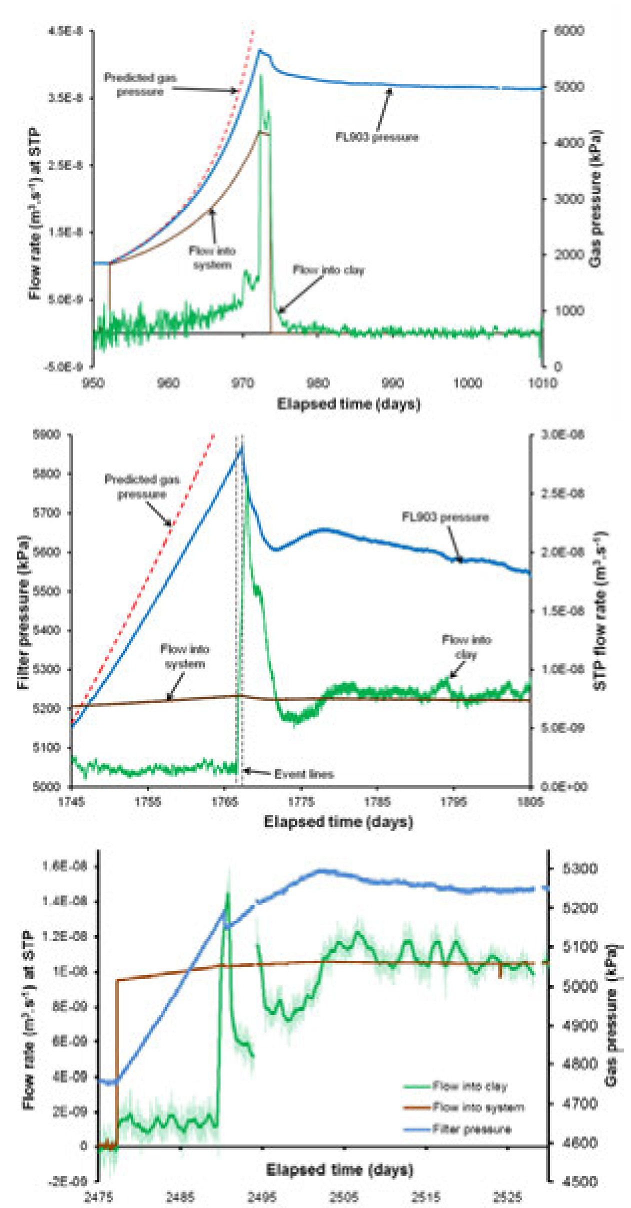

Figure 2: The gas entry events for the three gas injection tests conducted to date.

Major gas penetration and subsequent flow was accompanied by local dilation of the buffer clay, which strongly affected porewater pressure and total stress acting within the clay. Pathways were seen to evolve both temporally and spatially within the bentonite, exhibiting strong evidence for spontaneous self-sealing. Gas flow was observed to migrate generally down-hole in a somewhat tortuous path, possibly aligned to the prevailing stress gradient. Neon gas was also detected outside of the deposition hole, clearly indicating that gas had migrated through the bentonite and into the surrounding rock mass. Nevertheless, the exact mechanism governing this migration (i.e. advection and or diffusion) remains unclear. However, observations of strong coupling between gas flow and the hydromechanical response of the clay are highly consistent with results from small-scale laboratory experiments (Harrington & Horseman, 2003)[3], in which gas migration is attributed to dilatant pathway formation.

In the third gas test, a smaller diameter filter in the upper array (nearer the top of the canister) was selected, so as to provide data on the potential impact of the pressure gradient during gas injection. As in previous tests, a series of time dependent responses were observed in a number of radial stress sensors as major gas entry occurred. As before, examination of the data clearly demonstrates the temporal evolution of localised gas pathways. However, unlike the previous tests, gas pressure continued to increase following the initial inflow event and exhibited a second peak at a higher gas pressure significantly in excess of the local radial stress. Close examination of the data found no evidence for the escape of gas from the deposition hole, but instead, indicates gas periodically migrating downwards (Figure 3), following the prevailing stress gradient. Inspection of the pressure data from the adjacent filters on the test array suggests that, towards the end of the test stage, a network of open conduits exists with minimal gas pressure decay along the features. However, when gas injection stops and pressures begin to decay, this network fragments, leaving residual gas trapped within the clay at a pressure close to the local stress state. As in previous tests, the post-test injection of water indicates the gas has minimal impact on the hydraulic permeability of the clay, but causes a slight increase in specific storage.

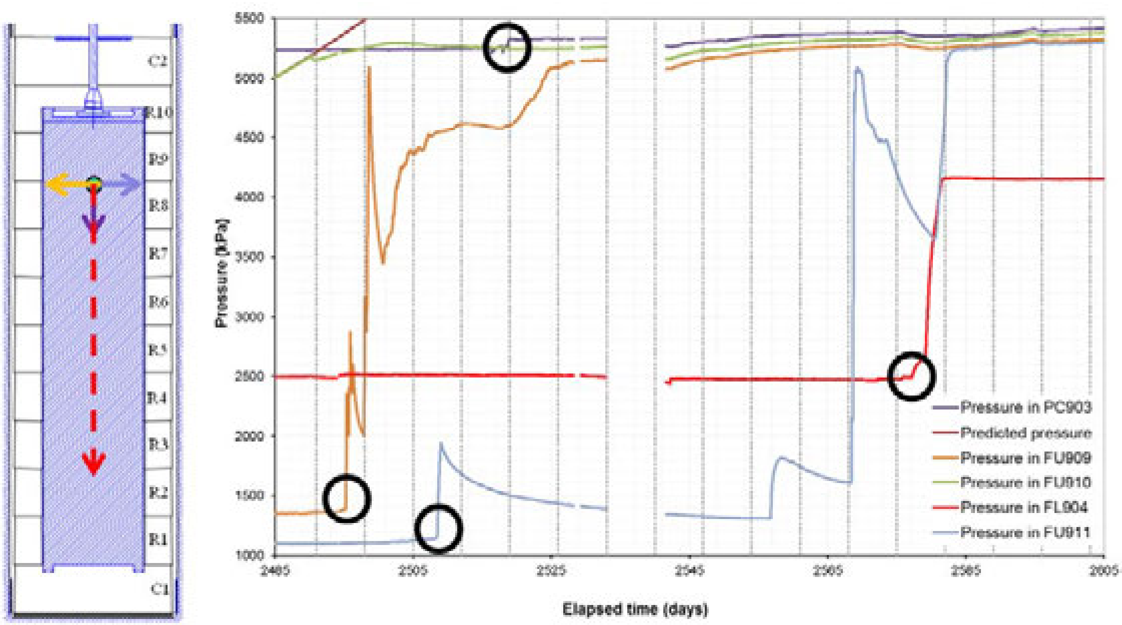

Figure 3: Response of selected sensors during prolonged gas injection. In order of first change, gas reached sensors FU909, FU911, PC903 and FL904. The evolution of pressure shows that several gas pathways must have formed and that these continued to evolve spatially and temporally.

Many features of the gas response are consistent between the tests at laboratory and full scale. All three gas tests have shown a link between local stress and gas break-through pressure, which is confirmed in Figure 4 where the dotted line represents the condition when applied gas pressure is equal to local stress. As seen, the gas breakthrough pressures plot close to this condition. Comparing Lasgit data with laboratory data from tests Mx80-10, Mx80-13 and Mx80-14 clearly shows that gas movement is strongly controlled by the local stress state. However, there are also a number of clear differences, the most notable of which are the observed precursor flow prior to major gas entry and the lack of very high observed gas pressures. While the exact cause of the precursor flow remains unclear, data from laboratory tests clearly shows that high gas pressures stem from the gas's ability, or lack of, to locate a suitable sink. Initial data from Lasgit would seem to suggest that the large volume of clay and the possible development of lower density zones as observed around the periphery of the bentonite in previous field tests may help to moderate the maximum gas pressure that can be generated. Nevertheless, this observation is obviously deposition hole specific and requires further examination as the buffer continues to mature.

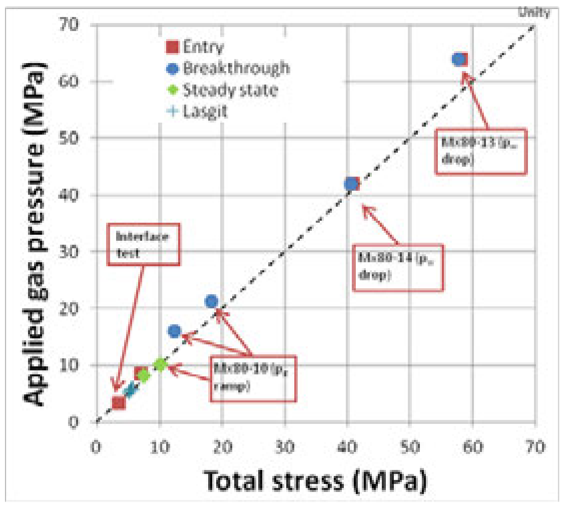

Figure 4: Close relationship between local stress and gas breakthrough pressure for laboratory and Lasgit-scale experiments.

One key observation from these full-scale tests is the general coupling between gas, stress and porewater pressure. This is extremely important from a phenomenological perspective and can be explained through pathway dilatancy concepts. These observations are similar to those reported from the laboratory tests, suggesting that scale has

4. Acknowledgements

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Laboratory investigation of hydrogen generation from carbon steel corrosion under deep geological repository conditions

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Extended abstract

There si concern that gases generated in a deep geological repository could adversely affect repository barriers if the pressure was allowed to increase above a certain limit. The problem can be solved either by reducing the rate of gases generation in a repository or by using barriers permeable enough not to allow accumulation of gas leading to the increase of pressure above barrier strength limits. The largest source of gases in repositories comes from corrosion of metals, particularly steel based canisters. Despite a huge number of publications devoted to the study of steels corrosion, there are still a lot of uncertainties concerning hydrogen generation rates under repository conditions. The problem is that corrosion and consequently hydrogen generation rate is affected by a large variety of factors such as: pH, temperature, composition of water, radiation and primarily amount of oxygen present in the water contacting metals. Reported values of hydrogen generation rates therefore range from less than one tenth of moles per square meter and year to the values exceeding several tens of moles of generated hydrogen per square meter and year. The main aim of our work conducted in the framework of 7th FM EC project FORGE was to decrease the uncertainties concerning the possible values of hydrogen generated from corrosion of carbon steels under anaerobic repository conditions.

The experiments were conducted with carbon steel samples of standard EN 10204/3.1 with content of carbon less than 0.1 %. The layers of carbon steel samples affected by atmospheric oxidation were removed before the experiment. Most of the experiments were conducted in the synthetic bentonite pore water corresponding to the pore water of bentonite Volclay KWK 20 – 80 of density 1600 kg/m^3. The bentonite water was at least 14 days in the anaerobic box before conducting anaerobic experiments. The hydrogen generation rate was determined by measuring hydrogen evolved from corroding iron or carbon steel using a device developed in ÚJV Řež, a.s. The device enables to measure hydrogen evolution continuously at different temperatures and at a constant pressure. The source of pressure is heavy piston, moving in mutually interconnected pressurized cylindrical vessels with a rolling membrane. The measurement of volume is based on the detection of the piston position, which depends on the medium volume. Most of the experiments were conducted in the anaerobic box filled with argon with concentration of oxygen below 0.1 ppm. A special corrosion cell was developed to conduct long-term (one year) experiments in the anaerobic box. The short term experiments were carried out at various temperatures controlled by a TDC 2 Temperature Controller (Gamry Instruments Inc, USA) with a Series 988 PID control unit (Watlow Controls, USA). Corrosion products on the carbon steel plates after the experiments were measured by Raman spectrometer LabRam (Horiba - Jobin Yvon, France) with two lasers (He – Ne and Ar), by X-ray diffraction Philips-Xpert PRO (PANalytical, Netherlands) and by ESCA Probe P (Omikron Nanotechnology, Germany) in CAE mode (Constant Analyser Energy).

The typical picture of hydrogen generation from short-term experiments (30 days) for one carbon steel plate of surface 100 cm^2 in 2 l of water conducted under various temperatures in the anaerobic box is shown in Figure 1. It can be seen that after fast hydrogen generation rate at the beginning of corrosion, the hydrogen generation rate slows down over time. This decrease is presumably connected with the formation of passivating corrosion product layers formed on the surface of carbon steel samples.

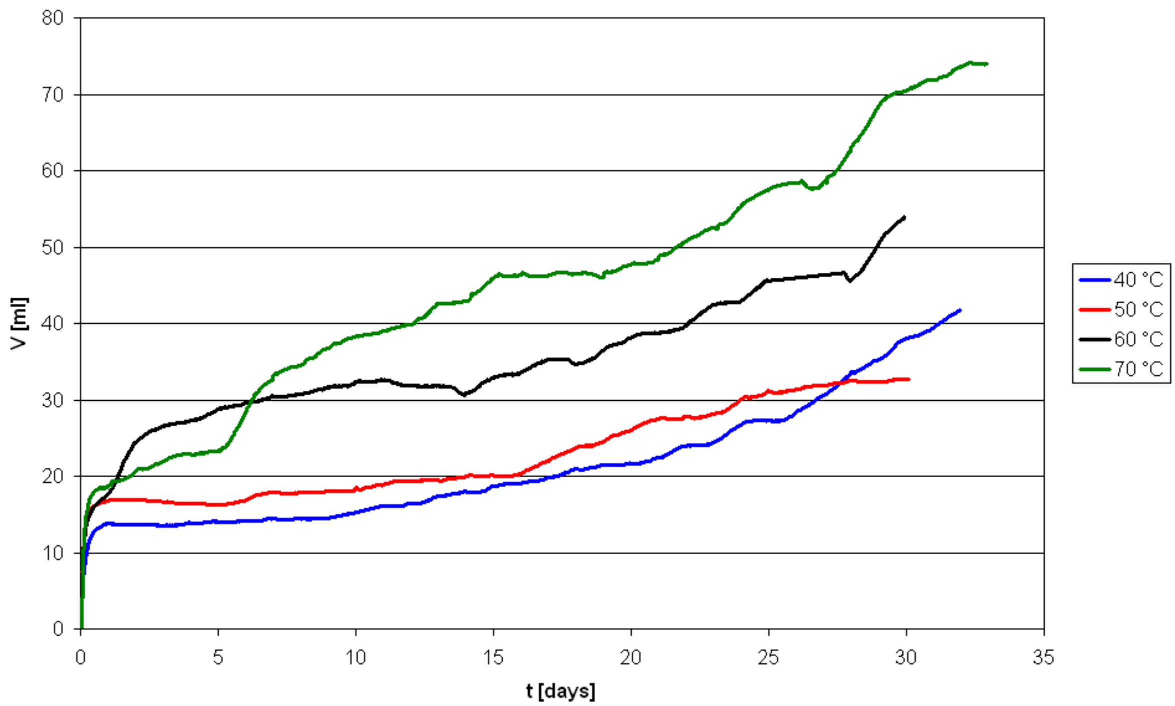

Figure 1. Hydrogen evolution rate under anaerobic conditions (one carbon steel plate (aprox. 100 cm^2) in 2 l of bentonite water)

The effect of temperature is strong, particularly at the beginning of corrosion, but then its effect depends on the nature and stability of the corrosion layer formed and hydrogen generation rates could be even greater at low temperatures. An important factor affecting the hydrogen generation rate is the ratio of surface of metals to the volume of water. This ratio affects primarily the oxidation potential of water contacting steels. At the beginning of corrosion oxygen can contribute to the greater hydrogen generation rates until a sufficient stable corrosion layer is formed, but in the strongly reducing environment the low oxidation potential of water can lead to the greater hydrogen generation rates, because the oxidation potential of water is not sufficient to reproduce stable corrosion product layers protecting carbon steels against corrosion.

The strong effect of repository conditions on hydrogen generation rates was also confirmed by experiments with pure iron powder of high surface (0.205 m^2/g) that was used for acceleration of experiments. The experiments with iron powder were also conducted in contact with solid bentonite. The example of the results obtained with corrosion of iron powder is given in Fig. 2. It is clearly shown that hydrogen generation is affected not only by the ratio of iron to water, but also by the presence of solid bentonite that accelerates hydrogen generation rates. It can be seen that hydrogen generation rates recalculated per 1 m^2 are greater after adding 30 g than after adding 5 g of iron. It was found that after adding 30 g of iron powder to the solution, Eh decreases more significantly that in the experiment with 5 g of iron. Presumably no protective layer inhibiting significantly corrosion can be reproduced under very low Eh values. The oxidation potential of water after adding 5 g of iron is in this case sufficient to form a layer of corrosion products on the iron powder inhibiting corrosion more than in the experiment with 30 g of iron.

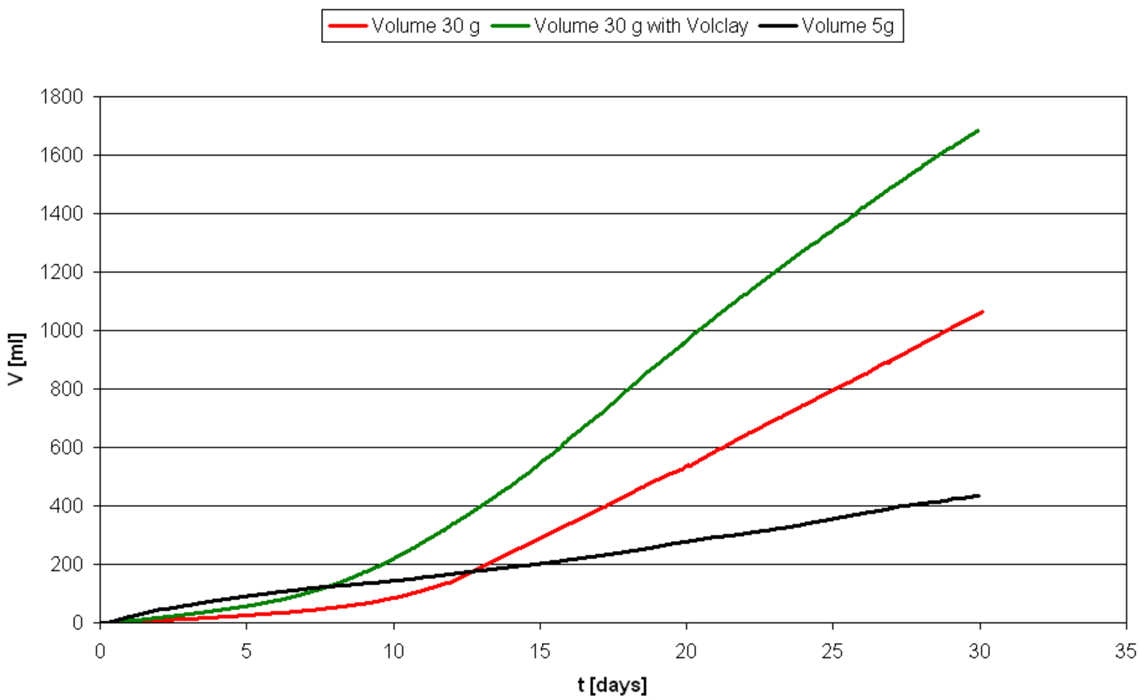

Fig. 2 Hydrogen generation form iron corrosion in bentonite with with and without presence of solid bentonite

The long term experiments were conducted in the corrosion cell in which a sample of carbon steel of cylindrical form (149 cm^2) were immersed in 300 ml of bentonite water at 80 °C. A corrosion product layer was removed from the sample by lathe, so that the real surface can be little larger than the surface estimated from geometrical dimensions. The result of the first experiment after derivation of the results is given in Fig. 3. The second experiment is still under way, but in principle the results of this second long-term experiment confirm the results obtained in the first one. It can be seen that, particularly at the beginning of corrosion, the corrosion and hydrogen generation rate (1 mol/m^2yr ~ 5.55 μm/yr) is high (> 8 mol/m^2yr). This is probably connected with the formation of a new corrosion product layer on the surface of the sample with the removed original corrosion layer formed under atmospheric conditions. Hydrogen generation rate was higher than 1 mol/m^2yr for the first 100 days. But even under steady state (after 100 days) the hydrogen generation rate is only slightly less than 1 mol/m^2yr. This long-term hydrogen generation rate is by order of magnitude greater than the values obtained before both in our laboratory in experiments conducted under not so strictly maintained strongly reducing conditions or in other laboratories (Smart et al, 2001, Hunter et al, 2007).

After dismantling the experiment, It was found that the carbon steel sample was covered by homogeneous, black layer of corrosion products, suggesting that magnetite is the main product of the corrosion layer formed. The corrosion rate obtained by the weight loss measurement of the carbon steel sample after the experiment was 5.58 μm/yr. This value corresponds almost exactly to the value obtained from measuring hydrogen generation rates.

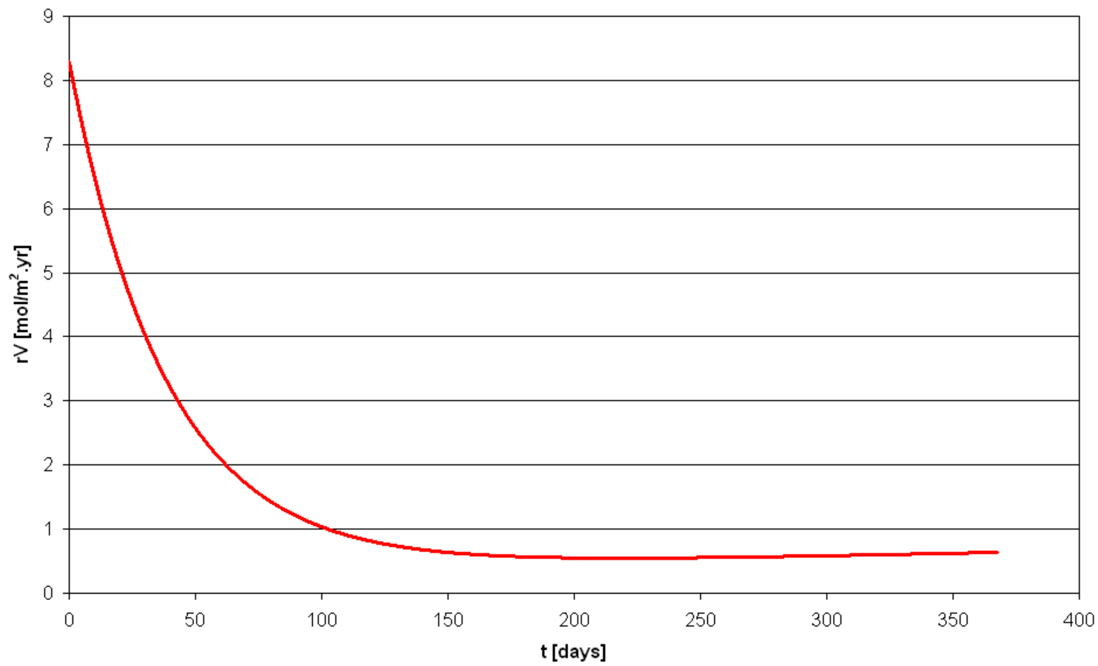

Fig. 3 Hydrogen generation rates from carbon steel in the long-term experiment in the anaerobic box.

The results of experimental investigation of hydrogen generation conducted in this work suggest that hydrogen generation rates cannot be predicted with any certainty if exact corrosion conditions are not known. It seems that the nature and stability of corrosion products formed on the surface of carbon steels, which is in turn affected by changes of geochemical conditions, particularly by the oxidation potential of water in various stages of repository evolution. At the beginning of corrosion oxygen present in the water can contribute to the greater hydrogen generation rates until a sufficient stable corrosion layer is formed, but in the strongly reducing environment the low oxidation potential of water can lead to the greater hydrogen generation rates, because the oxidation potential of water is not sufficient to reproduce stable corrosion product layers inhibiting corrosion.

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Acknowledgment

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Microbial Transformations of Radioactive Wastes: Implications on Gas Generation and Radionuclide Speciation

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Summary. Gas generation from the subterranean disposal of radioactive wastes is a major concern. Microorganisms have been detected in low- and intermediate-level wastes (L/ILW), deep geological formations, backfill materials such as bentonite, and waste repository sites designated for disposal of high-level waste (HLW). In general the levels of radioactivity present in the L/ILW and TRU wastes are not toxic to microorganisms. Biodegradation of natural and synthetic organic compounds in L/ILW and transuranic (TRU) wastes under aerobic and anaerobic conditions results in the production of gases such as CO_2, N_2O, N_2, H_2, H_2S, and CH_4 as well as HT, $^{14}CO_2$, $^{14}CH_4$, CH_3T, $^{14}CH_3T$. Microbes reduce $^{14}CO_2$ to $^{14}CH_4$ in the presence of H_2 and conversely, they oxidize $^{14}CH_4$ to $^{14}CO_2$ under aerobic conditions. In addition, they catalyze the methylation of iodine and selenium and potentially generate volatile methylated radioactive compounds such as methyl iodide ($CH_3{}^{129}I$), dimethylselenide $(CH_3)_2{}^{79}Se$, and dimethyldiselenide $(CH_3)_2{}^{79}Se_2$. In addition to gas generation microorganisms affect the speciation and solubility of radionuclides. The organic ligands and microbial metabolic products form soluble complexes with reduced actinides and fission products and affect their mobility.

1. Introduction

Gas generation from HLW and spent fuel are primarily due to radiolysis and corrosion while microbial processes play a dominant in role in L/ILW and TRU wastes. The harsh environmental conditions and the lack of nutrients in HLW are not conducive for microbial growth. The L/ILW generated from nuclear power plants, industries, hospitals, and research institutions contain a variety of radionuclides and a large fraction of biodegradable organic materials such as contaminated paper, plastics, rubber gloves, shoes, cotton, spent resins, cartridge filters, etc. Biodegradation of organic materials and corrosion of steel drums including microbially induced corrosion (MIC) can generate gas which has the potential to compromise the integrity of waste containers and facilitate pathways for radionuclide migration into the environment. Radiolysis of plastics and rubber materials may not only result in the production of volatile toxic organic compounds but render the plastics more susceptible to biodegradation. Microbial activity in L/ILW is regulated by the availability of electron donors and acceptors, moisture, nutrients (nitrogen, phosphorus); temperature, pH, Eh, radiation levels, and the presence of toxic compounds. Microorganisms play a major role in the transformations of radionuclides, toxic metals and organic compounds in the waste. Dissolution or immobilization of radionuclides is brought about by direct enzymatic (oxidation-reduction reactions which affect the valence state) or indirect non-enzymatic actions of microorganisms [1]. The mechanisms of microbial generation of gases and the microbial mobilization and immobilization of radionuclides in L/ILW and TRU wastes are briefly summarized.

2. Microorganisms in radioactive wastes, natural analog sites, backfill materials and radioactive waste repository sites.

Microorganisms have been detected in low-level radioactive wastes, TRU wastes, Pu-contaminated soils, and in waste-repository sites considered for disposal of HLW [1].

Low-level and Transuranic wastes: Several aerobic- and anaerobic- bacteria (*Bacillus* sp., *Pseudomonas* sp., *Citrobacter* sp., and *Clostridium* sp.) were isolated from the leachate samples containing Pu-238, 239, 240 and other radionuclides, and a variety of organic compounds from the LLW sites West Valley, NY, and Maxey Flats, KY. The radionuclides and the organic-chemicals present in the leachate were not toxic to the bacteria, which metabolized them, producing tritiated- and carbon-14 methane. Metabolically active microbes were identified at the Los Alamos National Laboratory's (LANL) TRU waste burial site containing ^{239}Pu contaminated soil and flammable waste. Even extreme environments, such as the hypersaline groundwaters at the Waste Isolation Pilot Plant (WIPP) site, and the extremely low-nutrient granodiurite pore-waters in Switzerland, harbour microorganisms capable of interacting with actinides in the wastes.

Natural anologue sites. Microorganisms representing autotrophic and heterotrophic groups including both native species, and those introduced by mining operations were found in water- and subsoil-samples from deep mines designated for disposal of HLW in Europe.

Pocos de Caldos, Brazil. Sulphur-cycle bacteria were found in soil core and groundwater samples at the Osamu Utsumi open-pit uranium mine and Morro do Ferro Th/REE ore body, Pocos de Caldos, Brazil.

Oklo, Africa: Bacteria were found at the Oklo site in Gabon, Africa, the only known natural fission reactor that has plutonium suggesting that microbes are likely to thrive in nuclear-waste repositories.

Cigar Lake, Canada. Cigar Lake, a high-grade uranium ore deposit site, investigated as a natural analogue site for HLW contained microorganisms in the ore zone and groundwater. Denitrifying, iron-reducing, and sulphate-reducing bacteria are common in both groundwaters and rocks and seems able to withstand the radiation. Anaerobes were more abundant than the aerobes, in agreement with the prevailing reducing environment in the deposit, thus retarding the mobilization of redox-sensitive radionuclides.

*Yucca Mountain Site***.** The abandoned geologic repository site at Yucca Mountain (YM), Nevada is located approximately 300 meters below the surface and 300 meters above the groundwater table in an unsaturated region of welded volcanic tuff. Studies at the YM and Nevada Test Site (NTS) showed that bacteria are present in the vadose zones representing thiobacilli and sulfate-reducing bacteria, which can corrode metals.

The presence of active microbial population in deep subsurface environments suggests that under appropriate conditions they could play a significant role in transformation of radionuclides in the wastes.

3. Generation of Carbon-14, Tritiated and other radioactive gaseous compounds.

Radioactive gaseous compounds such as CH_3T, HTO, ^{3}H, and other tritiated hydrocarbons as well as ^{85}Kr, ^{222}Rn, $^{14}CH_4$ and ^{14}C hydrocarbons were released from LLW sites [2]. Tritiated methane is one of the most abundant compounds; an estimated 200-6000 mCi/yr of CH_3T is released to the environment [2].

Microorganisms generate radioactive gases directly through their metabolic activity, or they indirectly enhance the release of trapped gases. Methane bacteria in anaerobic environments convert CO_2 and H_2 to CH_4. Tritium and carbon-14 released into the environment as tritiated and ^{14}C methane from the waste also can be oxidized by microorganisms to $^{14}CO_2$ and HTO under aerobic conditions. In addition to production of various gases, volatile low molecular weight organic acids, alcohols, aldehydes, ketones, esters are produced by microorganisms from the degradation organic materials in soils.

Biomethylation of Selenium-79 and Iodine-129. Microorganisms play a major role in the transformation of Se and I [1].

Selenium. A wide variety of bacteria and fungi are involved in the (i) oxidation of elemental selenium to selenite and selenate, (ii) reduction of selenite and selenate to elemental selenium, and (iii) methylation of selenium to dimethylselenide $(CH_3)_2Se$ and dimethyldiselenide $(CH_3)_2Se_2$. Methylation of selenium is carried out by a variety of bacteria and fungi [1]. Dimethyl selenide is released from soil into the atmosphere and the volatilization of the methylated compound is pronounced when the microflora is provided with methyl donor (such as methionine, cysteine), readily available carbon and selenate selenite and seleno-amino acids.

Iodine. Microorganisms affect the chemical behavior of iodine through processes such as volatilization (CH_3I), oxidation of I^- to I_2, reduction of IO_3^- to I^-, and bioaccumulation by bacterial cells intracellularly and extracelluarly [3]. Microbial volatilization of organic iodine was observed in soil slurries and seawater samples by aerobic bacteria through methylation of iodide (I^-) to form methyl iodide (CH_3I). The volatilization of iodide was also found in iodide-rich natural brine water. In addition to the organic iodine compounds, a significant amount of molecular iodine (I_2) was produced.

4. Long-term Microbial Gas Generation studies from L/ILW and TRU Wastes.

Long-term studies conducted with L/ILW at Olkiluoto, Finland VLJ repository for a period of ~9 years showed gas generation due to biodegradation of organic constituents with the potential to overpressure the repository and promote transport of radionuclides [4].

WIPP Repository. TRU waste is disposed of at the Waste Isolation Pilot Plant (WIPP), located in a salt bed (2,200 ft below ground) in southern New Mexico. Microbes in the repository are dominated by halophilic- or halotolerant-bacteria. Significant portion of the TRU waste disposed of in the WIPP is cellulose; its biodegradation under the repository's hypersaline conditions can produce CO_2 and also affect the solubility of the actinides. Gas generation from the microbial degradation of the organic constituents of TRU waste (mixed cellulose and electron-beam irradiated plastics and rubbers) showed aerobic-, denitrifying-, fermentative-, sulfate-reducing-, and methanogenic-activity [5]. Carbon dioxide was produced in samples incubated at 70% relative humidity. The total gas and CO_2 production from simulated waste are shown in Figures 1 and 2. Cellulose degradation products detected in solution include lactic-, acetic-, propionic-, butyric-, succinic-, and valeric-acids [5].

Effect of Bentonite on gas generation. Bentonite a potential backfill material stimulated microbial

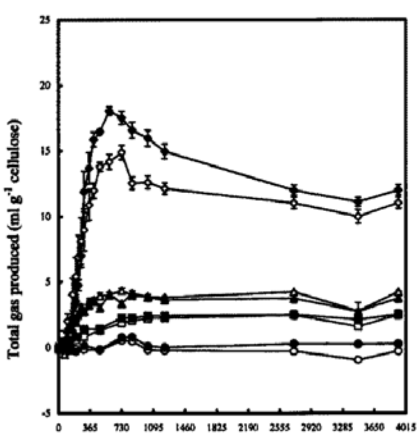

Figure 1. Total gas produced in anaerobic samples inundated with brine: unamended (○); unamended and inoculated (□); amended and inoculated (△); amended, inoculated, plus excess nitrate (◇). Closed symbols are samples with bentonite.

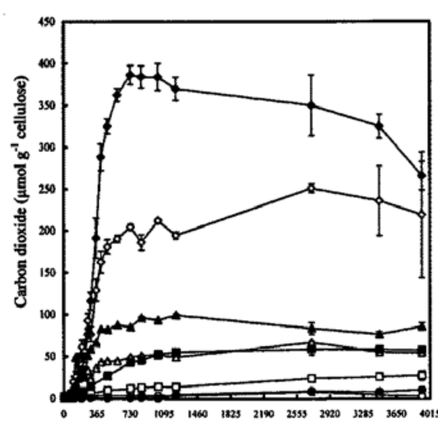

Figure 2. Carbon dioxide produced in anaerobic samples inundated with brine: unamended (○); unamended and inoculated (□); amended and inoculated (△); amended, inoculated, plus excess nitrate (◇). Closed symbols are samples with bentonite.

activity in WIPP waste and enhanced the total gas and CO_2 production and aqueous metabolites by several fold. The stimulatory effect of bentonite may be due to a combination of the use of ferric iron as an electron acceptor and/or buffering effect [5].

Degradation studies with actual and simulated TRU waste revealed that microorganisms produce far more gas than by physical and chemical means; including corrosion. Microbial activity is expected to be the dominant process responsible for degrading TRU waste and to have the greatest impact on performance assessment and compliance.

5. Microbial activities in wastes affect Radionuclide Speciation

An increase in microbial activity can alter the chemical speciation, solubility, and sorption properties and thus could increase or decrease the concentrations of radionuclides in solution and affect their bioavailability and mobility. Dissolution of radionuclides by microorganisms is due to changes in the Eh and pH of the local environment, by their production of organic acids, extracellular metabolites, dissolved carbonate species and production of chelating or sequestering agents such as oxalic acid, citric acid and siderophores. Immobilization or precipitation of radionuclides is due to changes in the Eh of the environment; enzymatic reductive precipitation (reduction from higher to lower oxidation state), biosorption, bioaccumulation, bioprecipitation reactions, biotransformation of radionuclide-organic and -inorganic complexes, and mobilization as biocolloids [1].

Naturally occurring as well as synthetic chelating agents, degradation products of plastics in the radioactive wastes form soluble complexes with radionuclides. Further, microbially generated fermentation products, dicarboxylic acids, ketogluconic acid, polyhydroxy acids and phenolic compounds form mono- and biligand complexes with actinides. Laboratory studies with uranium processing waste (sludge) from denitrification plant and contaminated sediment sample from the Oak Ridge Y-12 Plant produced gas, CO_2, H_2 and CH_4 when supplemented with a carbon and nitrogen source. Dissolution of a large fraction of metals was observed due to organic acid metabolic products as well lowering of the pH of the medium while U(VI) was reduced to U(IV) and precipitated from solution due to anaerobic microbial action. However, the microbially reduced U(IV) and Tc(IV) are not very stable and are readily reoxidized and thus can be remobilized. Also the presence of organic ligands such as citric acid, organic by-products, and macromolecules affect the precipitation of microbially reduced forms of U(IV), Pu(IV), Np(IV) and Tc(IV) by forming soluble complexes [1].

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Gas flow in anisotropic claystone. Modelling triaxial experiments

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Summary

Selected gas pulse tests on initially saturated claystone samples under isotropic confinement pressure were simulated using a 3D Thermo-Hydro-Mechanical code. The constitutive model considers the hydro-mechanical anisotropy of argillaceous rocks. A cross anisotropic linear elastic law is adopted for the mechanical behaviour. Elements for a proper modelling of gas flow along preferential paths include an embedded fracture permeability model. Rock permeability and its retention curve depend on strains through a fracture aperture. A constitutive internal dimension of the rock structure is required to simulate the aperture of joints. The hydraulic and mechanical behaviour have a common anisotropic structure. Small scale heterogeneity is also introduced in order to enhance the initiation of flow through preferential paths. Simulations are in agreement with recorded upstream and downstream pressures in the tests.

1. Introduction

In this paper, the modelling of two tests performed by Hildebrand et al (2002) [1] is presented. The constitutive model for the rock takes into account its natural cross anisotropy associated with the bedding planes of the rock. This anisotropy is considered both in its mechanical and hydraulic properties. The coupling between the mechanical and hydraulic behaviour of the rock and the possibility of bedding opening due to a decrease of the effective normal stress acting on the bedding planes are substantial improvements over conventional two phase flow models and allow the performance of more accurate analyses of gas flow phenomena in low permeability media such as argillaceous layered rocks.

2. Modelling approach

In The constitutive model for the rock is based on a cross anisotropic model for the mechanical and hydraulic behaviour of the claystone. Two angles describe the orientation of the rock bedding. Bedding has an important effect in the behaviour of this type of rocks since it controls their mechanical behaviour (e.g., highest compressibility in direction perpendicular to the bedding) and their hydraulic properties (e.g., higher fluid conductivity parallel to the bedding planes).

The permeability model used is described in detail in [2]. It simulates the effect of the presence of a planar discontinuity embedded in a given finite element on the value of its permeability. This discontinuity represents, in the cases analyzed in this paper, the bedding planes of the rock, which can be opened due to hydro-mechanical actions (e.g. increase of fluid pressure, effective unloading of the rock). The equivalent element permeability is computed assuming a laminar flow inside the planar fracture.

Figure 1 shows a scheme of the fracture geometry. The orientation of the fracture plane is given by two angles (dip direction angle, α, and dip angle, β). The permeability matrix in the reference cartesian coordinate system is obtained by means of a tensor "rotation", in terms of the intrinsic principal permeability values and the rotation matrices. The principal directions for k_1 and k_2 correspond to the fracture plane, while k_3 follows its normal direction. Changes of fracture aperture are computed through the normal strain to the fracture plane, *n* being the normal vector (see figure 1). The mechanical constitutive model is a transversal isotropic elastic model based on the model presented by Wittke [3], which considers 5 material parameters plus 2 anisotropy directions.

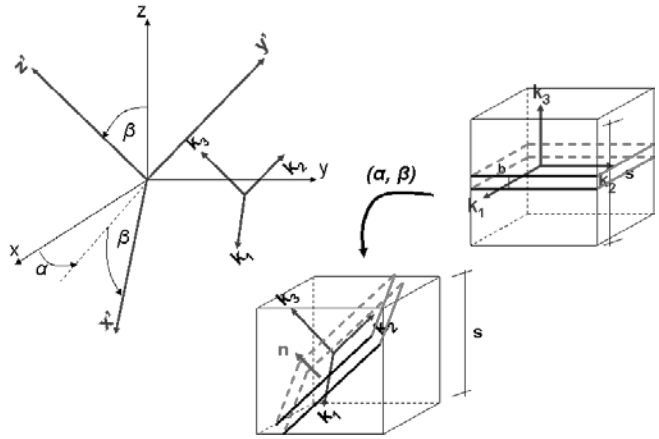

Figure 1. Scheme of the geometry of bedding planes and its relationship with the reference Cartesain coordinate system.

The gas injection on an initially saturated specimen leads to a desaturation of the existing or induced discontinuities. The rock matrix may become also partially saturated. Therefore, the mechanical model should handle the effect of variable suction. The effect of suction is properly accounted for by defining two stress fields: an isotropic one ($s\delta_{ij}$; δ_{ij}: Kronecker delta) and a so called "intergranular stress" which combines total stress and suction. In the model used, a Bishop-like expression defines the average skeleton stress *p'* in terms of the total mean stress, *p*, air and water pressures (u_a, u_w) and degree of saturation:

$$p' = p - u_a + S_r(u_a - u_w) = p - u_a + S_r s \qquad (1)$$

The constitutive model used is defined in detail in [4]. Only the elastic anisotropic component was activated in the simulations reported below. In addition, a small scale heterogeneity of the rock has been introduced in an effort to enhance the initiation of the gas flow through preferential paths following the bedding direction. Figure 2 shows a generated spatial distribution of the permeability for k_1 (permeability parallel to bedding direction). The distribution is defined in terms of the intrinsic permeability (log scale in the caption; units in m^2). The random field follows a normal distribution (mean value: $9\cdot10^{-21}m^2$; variance: $1.3\cdot10^{-40}m^4$). This randomness introduces a small variability of permeability.

3. Model validation

To properly simulate the tests a 3D representation of the sample and the upstream and downstream reservoirs (porous metallic stones) is necessary. The sample has been assumed to be cubical in order to facilitate the discretization by means of a structured mesh with cubical elements, which simplifies the hydraulic problem. A sample 30mm high was discretized into 150 quadrilateral linear elemets. The volumes of the two chambers have been adjusted in order to obtain a gas pressure equalisation in the two fluid reservoirs at the end of the simulations which is similar to the value recorded in the experiments.

Two tests were simulated. The first test corresponds to a sample with the bedding planes parallel to the imposed flow (bedding in vertical position), while the in the second test the bedding orientation is horizontal. The first sample was taken from a depth of 1580m, whereas the second sample was taken from a depth of 1515m. The duration of the test is highly dependent on the bedding orientation. The

maximum gas permeability recorded is one to two orders of magnitude higher in direction parallel to the bedding than perpendicular to it.
Figure 2 shows the evolution of upstream and downstream pressures for test 1 reported by Hildebrand et al [1] together with the pressures recorded in the test. The overall agreement is good, especially if one considers the evolution of the upstream pressure and the shape of both curves. The increase in the downstream pressure in the simulation of test 1 is delayed about one hour if compared with measurements. The model requires a time lapse for the gas to flow out of the sample into the downstream chamber, while in the actual test the increase in downstream pressure was recorded almost instantaneously.

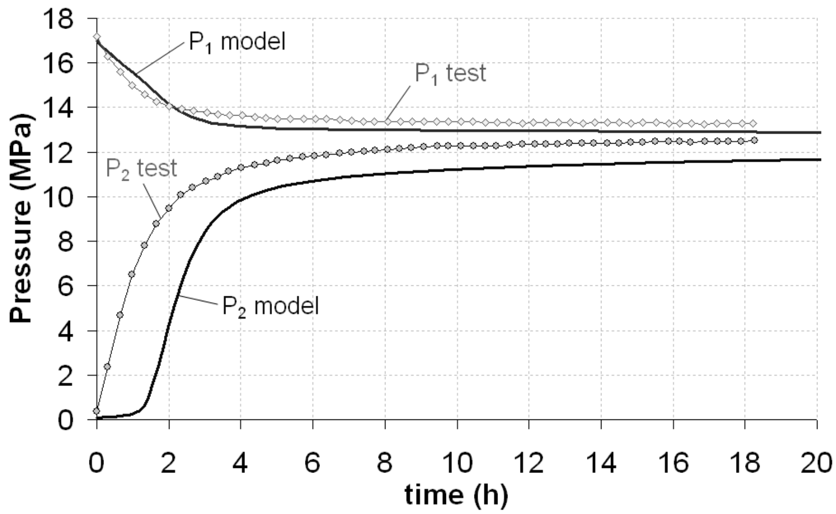

Figure 2. Comparison between simulated and measured pressure evolution at upstream and downstream reservoirs. Test 1.

Figure 3 shows the vectors of gas and water fluxes at a given time of the simulation of test 1. Initially, water flow clearly follows the direction of bedding. Gas and water vectors are at the same scale in figures 3. The effect of bedding opening was clearly visible in the water flux at the beginning of the test. Gas is pushing the water and forcing it to move towards the upper (downstream) reservoir. After the increase in permeability takes place in all the sample elements the vertical flow pattern becomes more homogeneous.
Once the gas pressure exceeds the new capillary air entry value of the material (the retention curve has changed due to deformation) it starts to desaturate the elements during the flow towards the downstream reservoir at the top of the sample.
Figure 4 shows the flow patterns (vectors of gas and water fluxes plotted in a common scale) obtained in the simulation of test 2. The effect of bedding direction is clearly visible and has a strong effect on the fluid flow towards the upper chamber. The orientation of the bedding is horizontal and this fact forces first the water, and afterwards the gas, to flow initially in sub-horizontal direction in each layer of elements.

4. Conclusions

Gas pulse tests on initially saturated claystone samples have been simulated considering the hydro-mechanical anisotropy of the rock. The presence of embedded fractures simulating the rock bedding is included in the permeability model. This is a significant improvement from conventional two phase flow models that allows performing more accurate analyses of gas flow in such type of low permeability layered argillaceous rocks. Two hydro-mechanical simulations of gas tests have been presented. Two bedding orientations, parallel and normal to the imposed flow in the test, have been modelled. Simulations show a good agreement between calculated and recorded pressure evolutions on the two tests.

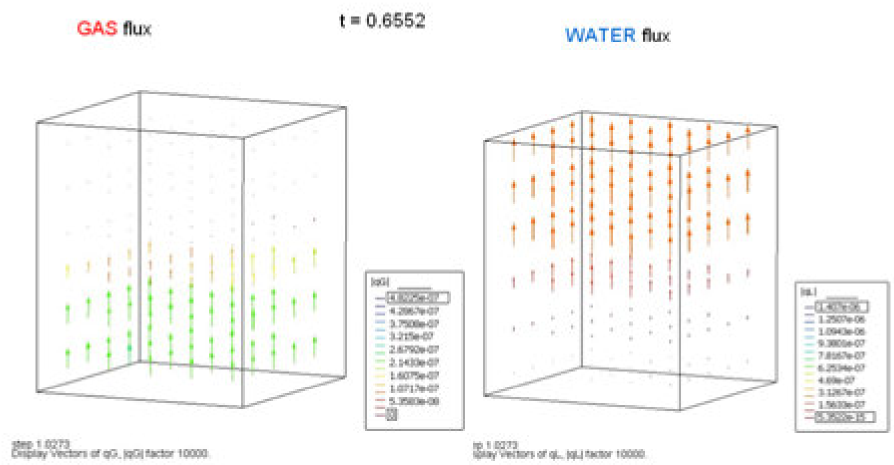

Figure 3. Gas (left) and water (right) advective flow vectors (m/s). Amplification factor 10^4. Test 1. t=0.6552h

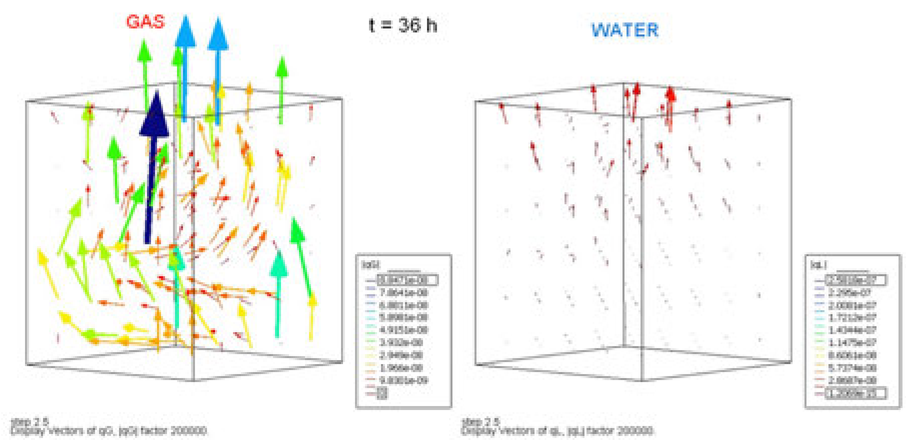

Figure 4. Gas (left) and water (right) advective flow vectors (m/s). Amplification factor $2{\cdot}10^5$. Test 2. t=36h

5. Acknowledgements

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Thermodynamical Modelling of Hydrogen Migration in Argillite for a Deep Geological Radioactive Waste Repository – IRSN contribution to FORGE.

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Summary

The aim of this work is to provide an improved mathematical and physical description of two phase flow in tight porous media in order to model hydrogen migration within a geological repository for radioactive waste as required to further asses the safety of such facilities. First, we introduce a general physical framework describing multi-component and two-phase (liquid and gas) flow in porous media with capillary pressure curve and mass exchange between phases. Assuming thermodynamical equilibrium, mass exchange between phases is governed by thermodynamical principles which we briefly expose. This physical modelling is able to describe single phase flow (only gas mixture or liquid solution) as well as two phase flow (both liquid and gas phases are present). On the basis of this treatment we obtain generalised formulation of Henry and Raoult-Kelvin laws. We show that for hydrogen-water mixture in repository conditions of pressure and succion these generalized relationships constitute an adequate mass equilibrium description. Second, we consider the mathematical formulation of the problem, which is mainly determined by the choice of primary variables. Contrary to existing approaches, we are interested in avoiding the need to switch primary variables during calculations (as it can be done in [1] with a less general physical framework). Indeed, such a switch is often proposed as a solution to treat phase appearance or disappearance (see e.g. [2] or [3]). We will show that, in our physical framework taking into account the capillary pressure and mass exchanges between phases, it is possible to work with universal primary variables to describe correctly gas phase appearance or disappearance. Simple numerical simulations are presented for validation as well as an application of this model to the Forge benchmark on the cell scale together with a sensitivity analysis guided by the physical model.

1. Physical framework

Considering multi-component and two-phase (liquid and gas) flow in a porous medium, the transport of each component is composed by 1) advection according to the flow rate of each phase and 2) diffusion in each phase according to spatial variation of phase composition. We consider two phases, gas and liquid, which fill the rock pores with gas (resp. liquid) saturation S_g (resp. S_l) and with gas (resp. liquid) pressure P_g (resp. P_l). The flow of each phase is governed by the generalized Darcy law and the difference between phase pressures is given by the capillary pressure curve:

$$P_g - P_l = P_c(S_g) \tag{1}$$

Furthermore, we consider that phases are not pure but composed by N +1 components: one solvent (typically water) and N solutes (or gas components, typically hydrogen, oxygen, etc). Their natural states at standard T,P are liquid for the solvent and gas for the gas components but due to vaporisation and gas dissolution, all components are present in each phase. Assuming that local thermodynamical equilibrium is satisfied, phase compositions are related by relations of the following general form:

$$P_g\, x^i_g = K^i\, x^i_l \qquad (2)$$

where i = 0 for the solvent and i = 1..N for the solutes ; x^i_α denotes the molar fraction of component i in phase $\alpha=\{g, l\}$. In the most general case the coefficient K^i depends on temperature and on pressure and composition of each phase but, in the case of ideal gas mixture and dilute liquid solution, this dependency is reduced to temperature and liquid pressure. Equation (2) recovers the classical relations like Henry's law, Raoult's law or Raoult-Kelvin's law. This correction takes into account the equilibrium displacement due to the capillary pressure and becomes more significant with higher capillary pressure. The correction $d_{r,i}$ obtained in repository conditions is of the order of 10% (Fig.1). An excellent agreement was found with the mass equilibrium predictions starting from the equation of state level [4].

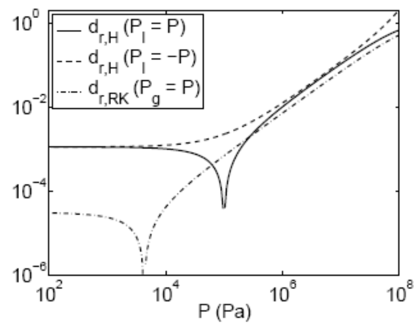

Figure 1: Relative corrections to the standard Henry constant $d_{r,H}$ and the Raoult-Kelvin relation $d_{r,RK}$ taking into account the existence of the capillary pressure. The curve where P_l=-P corresponds to the situation where the water pressure is negative as a result of strong capillary pressure. At moderate pressures (10MPa) the relative deviations are non negligible (10%)

2. Mathematical formulation

We propose to choose a unique set of primary variables to describe as well liquid and liquid-gas flow :

$$(P_l, (x_l^i)_{i=1..N}) \qquad (3)$$

These variables are physically well defined when a liquid phase is present and it is natural to use them to describe the liquid saturated flow. We will see that they can be extended in such a way to be able to determine whether a gas phase is present and, in this case, calculate the gas saturation, pressure and composition. Equation (1) and equation (2) provide two different expressions for gas pressure, π_g^m and π_g^t, given by :

$$\pi_g^m = P_l + P_c(S_g) \quad \text{and} \quad \pi_g^t = \Sigma_{i=0}^N K^i\, x^i_l$$

Local existence of gas phase is only possible when $\pi_g^m = \pi_g^t$. Thus, given pressure and composition of liquid phase, P_l, $(x_l^i)_{i=1..N}$, we look for a value of gas saturation S_g such as $P_c(S_g) = \pi_g^t - P_l$. The last equality is not always reachable, in particular because the capillary pressure curve is positive, $P_c \geq 0$. When $\pi_g^t - Pl < \min(P_c)$, the two expressions of gas pressure are not compatible and we conclude that gas phase can not exist (S_g=0). In a similar way we constrain the existence of the liquid phase and thus it is possible to describe in fully uniform and continuous way all thermodynamical situations. Now we are able to construct our mathematical formulation with primary variables given by (3), which leads to a system of N + 1 quasi-linear partial derivative equations.

3. Validation on MoMas test cases

In order to illustrate the ability of the formulation to treat gas appearance or disappearance, the model has been used on simple test cases supported by GNR MoMaS aiming at code testing and comparison [5]. Each of those test cases deals with only one physical/numerical problem at a time. We give here the results of one of these test cases (MoMaS test case 1a). It consists of a homogeneous domain with the material characteristics close to the Callovo-Oxfordian clay. At the beginning of the simulation the entire domain is partially saturated with water (gaseous hydrogen is present). Then a constant gas injection rate is applied on the left boundary. For most sets of initial and boundary conditions the gas would flow continuously from left to right with the maximal gas pressure obtained on the left border. However, it is possible to find such a set of particular conditions for which the continuity of the gas phase is at some time broken and a fully water saturated zone appears in the middle of the domain (Fig. 2).

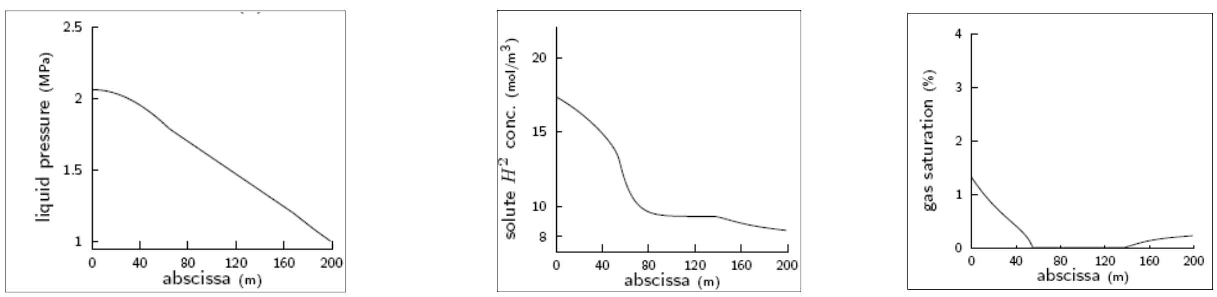

Figure 2 : Results at one intermediate time point of the MoMaS test case, showing that gas injection creates fully water saturated zone inside the domain.

4. Application to FORGE WP1 benchmark

In order to verify the capacity of our simulation code to deal with more complex applications we made calculations of a reduced version of the FORGE WP1 cell-scale test case [6]. The geometry of this test is presented in (Fig.3) together with measure points. The major deviations from the original FORGE benchmark consist in using a plane 2D geometry instead of the axisymmetrical one and replacing Van Genuchten liquid relative permeability in technological voids (interfaces) by $k_{r,l}=S_g^{4/3}$

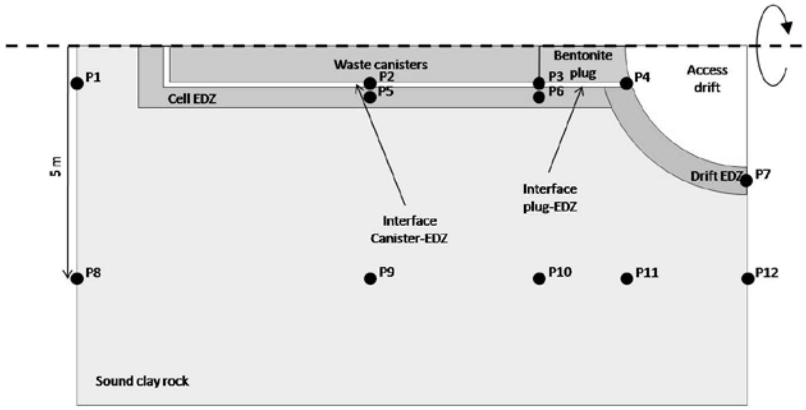

Figure 3: the general geometry of the FORGE call scale benchmark.

Figure 4 presents time evolution of pressures as measured in point P4 (interface) and in point P9 (inside undisturbed COX). We find a similar behaviour as the other benchmark participants, namely slow gas pressure increase during gas injection phase (till 10^4 years) accompanied by a decrease of water pressure in the interface (P4). In the case of point P9, where initially no hydrogen was present, the P_g variable "represents" the amount of dissolved hydrogen, and rises from zero to become equal to P_l which corresponds to the apparition of free gas phase. But as soon as P_g becomes lower than P_l, gas phase disappears. It can be noted that the value of gas saturation is small (of the order of 0.05%), and would usually not be seen by a code using a switch procedure. This can explain why we obtain some more pronounced minima of liquid pressure as compared to other benchmark participants. Further discussion of physically motivated differences will be included in the presentation.

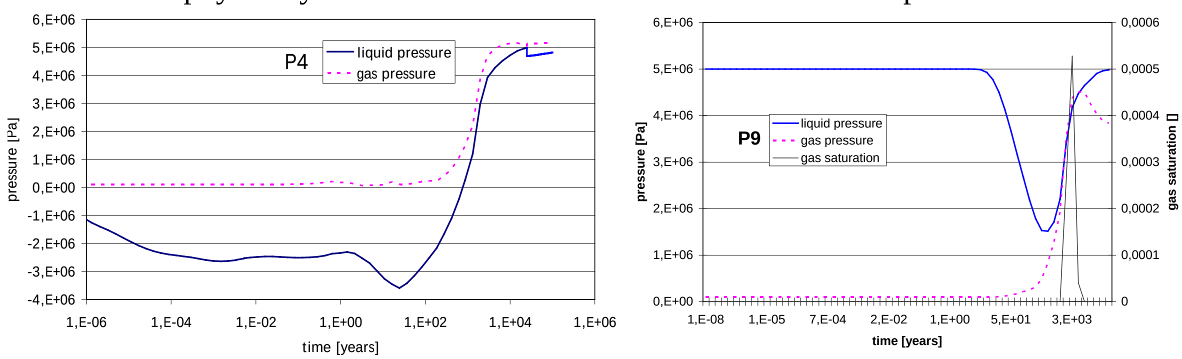

Figure 4: Time evolution of pressures in points P4 and P9. For P9 the saturation of gas is given.

5. Acknowledgements

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under Grant Agreement no230357, the FORGE project.

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Characterisation of Gas Migration in Claystone through the Modelling of a Field-Scale Gas Injection Test

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Summary

1D and 3D hydraulic modelling of a field-scale gas injection steps is performed with a two-phase flow model, in order to highlight how a predictive approach could reproduce the experimental observations. An iterative modelling process will show step-by-step how an accurate description of each component of the experiment system allows the reproduction and the understanding of the experimental data. For instance, the initial degree of saturation of the injection interval, as well as the presence of a disturbed zone around the boreholes, is necessary to obtain good agreement with the field data.

1. Introduction

In the field of radioactive waste confinement, the question of the gas transfers in argillaceous formations is a crucial issue. During long-term repository of radioactive waste in clayey rocks, a great amount of gases will be produced by the deterioration of the disposal components. This gas production could affect the safety function of the clay barriers. The understanding of their production and migration mechanisms in the host rock is of particular significance for the safety assessment of such solutions. A new large-scale gas injection test has been recently performed in the Meuse/Haute-Marne underground research laboratory (URL) in France. This experiment, called PGZ1, studies the migration of nitrogen in the host rock, i.e. Callovo-Oxfordian argillite. This contribution describes a finite element modelling of this large scale test and the interpretation we can obtain.

2. Field experiment description and observations

The experimental layout consists of two parallel boreholes that are equipped with a triple packer system to monitor pore pressures evolution with the gas injection conditions (see [1] for more details). An equilibrium phase (hydraulic phase) is first provided during 188 days after the drilling of the boreholes. The gas test is then composed by different periods of controlled nitrogen flow injection rate, interrupted by ‘shut-in’ phases when gas injection ceases. The injection is performed from the central packer of one of the boreholes. Figure 1 shows the pore pressures evolution observed in the three packers along the injection borehole. No interference is observed in the two external sensors.

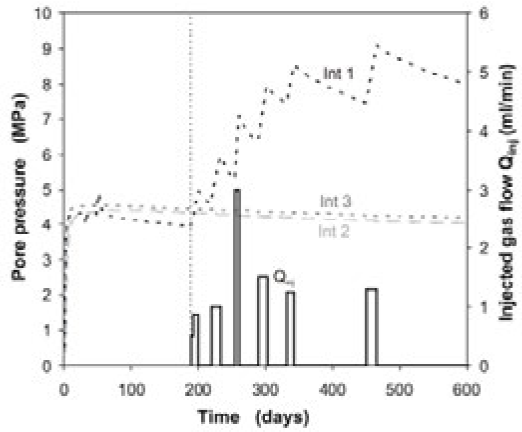

Figure 1: Time evolution of pore pressures in injection borehole before and during gas injection tests

3. Boundary value problem

A hydraulic modelling of the gas injection steps is performed with a two-phase flow model, in order to highlight how a predictive approach could reproduce the experimental observations. The geometry of the problem and the permeability anisotropy lead to perform 3D modelling, but for time consuming reasons 1D hydraulic modelling is first performed, which allows the understanding of the gas migration processes.

To reproduce water and helium transfers in partially saturated porous media, a two-phase flow model is used. This model is comprised of a liquid phase, composed of liquid water and dissolved helium and a gaseous phase, which is an ideal mixture of dry helium and water vapour. It takes into account the advection of each phase using the Darcy's law and the diffusion of the components within each phase (Fick's law). The retention curve and the water relative permeability curve are given by the van Genuchten's model (van Genuchten 1980). The gas relative permeability curve is a cubic function. The injection interval is modelled by considering an equivalent porous media, with high permeability, porosity equal to 1 and a low air entry pressure. No water and gas relative permeability curve is defined for this component.

Table 1 : Hydraulic parameters

K_w^{sat}	Water permeability in saturted conditions (m²)	$4\ 10^{-20}$
K_g^{dry}	Gas permeability in dried conditions (m²)	$4\ 10^{-18}$
ϕ	Porosity (-)	0.18
τ	Tortuosity (-)	0.25
P_r	van Genuchten air entry pressure (MPa)	15
n	van Genuchten parameter (-)	1.49
m	van Genuchten parameter (-)	0.55

4. Numerical results

An iterative modelling process shows step-by-step how an accurate description of each component of the experiment system allows the reproduction and the understanding of the experimental data. For instance, the initial degree of saturation of the injection interval, as well as the presence of a disturbed zone around the boreholes, is needed to obtain good agreement with the field data.

Indeed uncertainties remained on the volume of the residual water that was removed from the injection interval just before the nitrogen injection. After an accurate analysis of the experimental process, an initial degree of saturation of 0.221 in the interval can be deduced. It improves strongly the numerical results obtained during the first two gas peaks in comparison with a initially saturated interval (Fig.2 (a)).

Moreover the introduction of a damaged zone at the borehole wall improves also the agreement with the experimental observations, as shown in Fig. 2 (b). The extent of the EDZ is assumed close to the radius of the borehole (4 cm). In this disturbed zone, a constant permeability is considered, 500 times higher than the undisturbed claystone permeability. The retention properties are also modified by the excavation and the parameter P_r of the van Genuchten's retention curve is decreased due to the micro-fracturing of the rock mass ($P_{r,EDZ}$ = 3 MPa). The introduction of the damaged zone improves the reproduction of the experimental data during the first four injection peaks

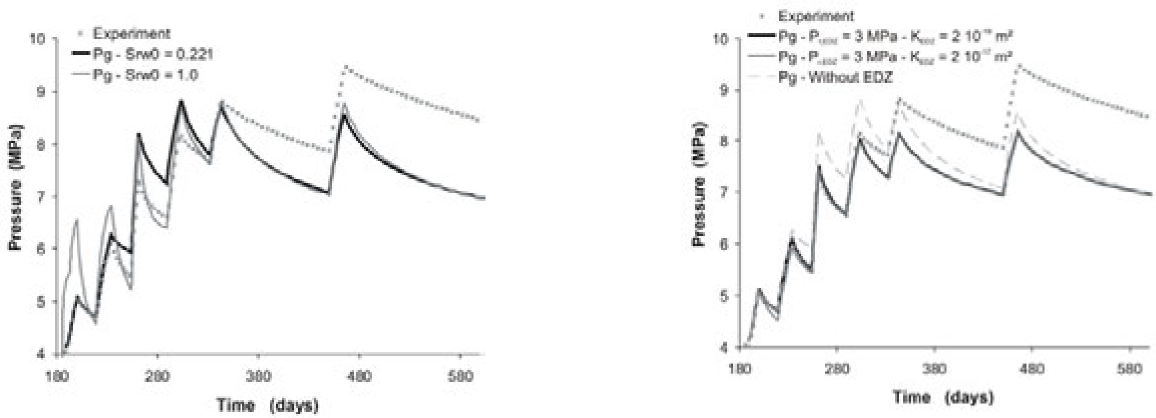

Figure 2: Influence of (a) the initial degree of saturation in the injection interval and (b) of the introduction of an excavated damage zone on the time evolution of pore pressures in the injection

Finally the last gas injection stages provide experimentally pore pressures higher than the numerical predictions. A modification of the gas relative permeability curve of the host rock allows a better reproduction of the experimental data, as shown in Fig.3 (a) when a Parker relationship is used instead of a cubic law.

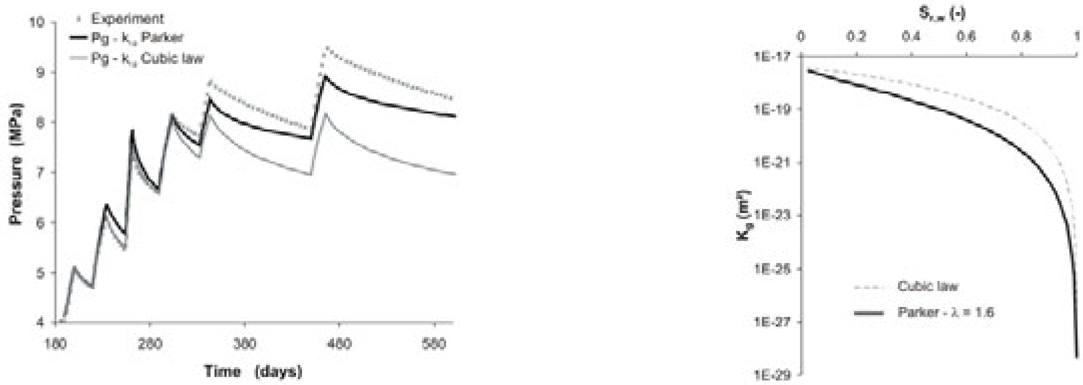

Figure 3: (a) Influence of the gas relative permeability of the undisturbed argillite on the time evolution of pore pressures in the injection interval – (b) Gas permeability vs. Degree of saturation

From this last satisfactory 1D modelling, we could extract a set of parameters characterizing the behaviour of the injection interval, the excavated damaged zone and the undisturbed rock. These parameters will be used in a 3D modelling of the problem, which will highlight the influence of the axial gas flows (see Gerard et al., 2012 for more details).

5. Discussion and conclusions

Even tough the experiment has been initially designed in order to study the gas transfers in a potential host rock for radioactive waste disposal, the modelling illustrates that the experimental response does not characterize the rock mass behaviour at the beginning of the nitrogen injection. The first injection phases test only the behaviour of the injection interval (water removal), whilst the response of the third and the fourth peaks are also influenced by the excavated damaged zone. It is only afterwards that nitrogen reaches the host rock and that pore pressures measurements allow characterizing the rock mass behaviour. The good agreement with the experimental data shows that a predictive model based on two-phase flow approach is able to reproduce experimental observations in large-scale system, as far as the injection flow rates and the gas pressures remain moderate and the development of preferential gas pathways is limited. More generally, the PGZ1 experiment has shown that gas would remain mainly confined in the borehole disturbed zone. Even though gas penetrates in the host rock, the quantities remain low and located near the injection interval with such gas injection conditions.

6. Acknowledgments

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under Grant Agreement no230357, the FORGE project.

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Determination of Diffusion Coefficients for dissolved He and CH_4 in Opalinus Clay

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Opalinus Clay is studied in Switzerland as a potential host formation for the geological disposal of radioactive waste. Good knowledge on the transport behaviour of radionuclides through the host formation is essential for safety assessment. Information pertinent to diffusive transport can be obtained from small scale lab experiments and on a larger scale by the modelling of natural tracer profiles. Helium is one such natural tracer that can aid in gaining a better understanding of transport processes in Opalinus Clay. It is produced by the decay of uranium and thorium, both of which are present in the formation. Furthermore, vertical fluxes and the interaction with embedding ground waters affect He concentrations in the pore water of the formation.

Modelling diffusive transport of He requires data on the spatial distribution of the concentration of dissolved He in the low-permeability sequence and in the embedding aquifers, and such data are available for 3 sites in northern Switzerland. On the other hand, diffusion coefficients have not been measured to date. For the transport models, estimations were made based on the known coefficients for water isotopes and anions. To obtain more accurate model calculations pertinent to the He diffusion profiles, information on the diffusion coefficient of helium in Opalinus Clay is required. Apart from helium, the diffusion coefficient of methane is also of interest.

Diffusion coefficients of dissolved gases are determined here in a cross through diffusion experiment. An Opalinus Clay core sampled perpendicular to the bedding plane, mounted in a stainless steel diffusion cell, is connected at both sides to a water vessel. Both sides are pressurised with a gas (He and CH_4) at equal pressure to avoid advective transport. Gas saturated water (Henry's law) is circulated along a filter in contact with the rock core. The dissolved gases will diffuse through the core due to a concentration gradient. The changes in gas composition at both sides are monitored by µGC, and so the through-diffusion fluxes of the gases can be quantified. The experiment is currently ongoing.

Numerical modelling and interpretation of field-Scale gas Injection experience PGZ1

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Summary

This work gives a numerical interpretation of PGZ1 experience leaded by ANDRA in experimental laboratory of Bure. Hence, the main goal of this field-scale gas injection test is to provide a better understanding and quantification of hydrogen migration in callovo-oxfordian claystone. We propose here a fully coupled hydromechanical simulation of this experience. An iterative process between the field analysis and modelling teams aims to finally lead to a better understanding of the experience behaviour.

Introduction

A great deal of attention is focused on radioactive waste management in nuclear industry and especially in the feasibility of deep geological underground disposal. In these deep repositories, the production of nitrogen involved by radiolysis of wastes or anoxic corrosion of steel components is an important phenomenom to take into consideration. In order to assess feasibility of the deep storage of radioactive waste, a large experimental program - leaded by Andra - has been engaged at the Meuse/Haute-Marne research laboratory. PGZ1 is one of these experiments. It focuses on perturbations induced by hydrogen migration in Callovo-Oxfordien claystone. This work proposes a numerical interpretation of this test, using a classical two-phase flow finite element model.

Methodology

PGZ1 experiment [2] consists of 3 boreholes: one borehole for gas injection and measurement and two boreholes for strain and gas pressure measurements (Figure 1). Each borehole is composed of 3 intervals (for measurement or injection) separated by packers. Six various gas injection steps, with constant fluxes, followed by shut-in phases have been done in intact claystone. This experiment began in 2009 and is still in progress. Pressure measurements are made during injection. Pressure remains lower than 10 MPa which is less than minimum principal stress. At this height, fracing threshold is not reached. Main goal of this in situ experiment is therefore a better understanding and quantification of gas transfer mechanisms in claystone.

An iterative process between the field analysis (ANDRA) and modelling teams (ULg and EDF) was required to improve numerical modelling in order to reproduce and understand the measurements [3]. Finite element software Code_Aster [1] based on a classical coupled two-phase flow model has been used. In a first step, hydromechanical computations took into account the digging of the borehole, which has been made in 3D and 2D. Those computations provided a good fitting of pressure for the 3 first steps of injection but were unable to reproduce the hydraulic comportment for the next steps. Then, a better understanding [2] of the comportment of injection interval allows us to provide better simulations. Indeed, a precise knowledge of initial gas volume in this chamber is required. This taken into consideration, , digging of the borehole (and mechanical comportment) is no more modeled and only gas injection is considered. Furthermore, influence of a “damage zone” around the borehole is under study. This area seems to play an important role but is hard to characterize. All these hypothesis lead us to grasp the main comportment of borehole and of the clay surrounding it.

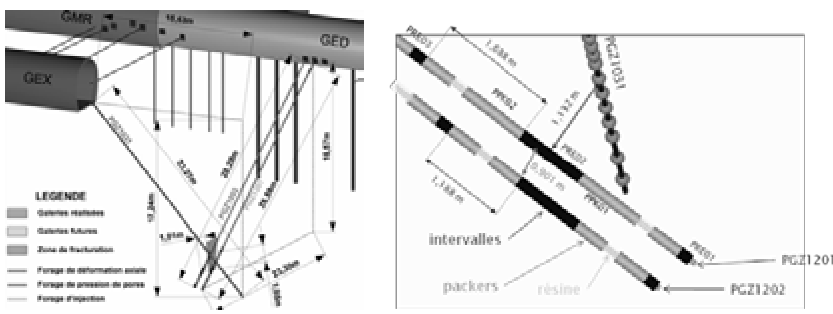

Figure 5. *PGZ1 experiment configuration and zoom on boreholes PGZ1201 and PGZ1202*

Results

We focus here on the second step of modelling (only gas injection is modeled and no more borehole excavation). Because anisotropy has no major effect in this test, axisymmetrical configuration is finally retained. Considering results from [2], precise initial gas saturation is taken into account in the injection chamber. This quantity has a major effect on pressure (Figure 2) and the first injection steps may essentially be composed of water displacement.

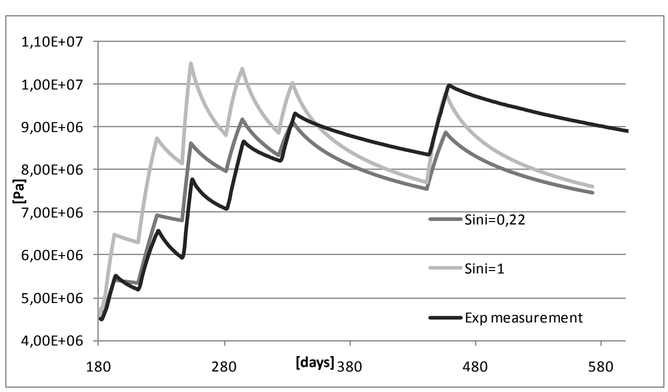

Figure 6. *Liquid pressure evolution in injection intervals: initial saturation goal*

Then, to improve those results, a transient zone (Borehole Damage zone BDZ) has been introduced. Experimental observations show that the influence of the gas injection in other borehole intervals is negligible. For this reason, we can imagine that the damaged zone has been closed around the swollen packers (Figure 3).

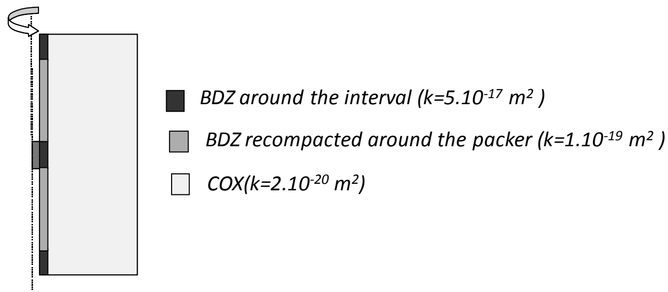

Figure 7. *Configuration and hypothesis of BDZ model*

Figure 4 shows that result with or without BDZ change very little till the middle of the second gas injection (≈220 days). From then on, we can make the assumption that injection interval behavior plays the main role and that water removal is the major phenomena. After 220 days, the results achieved with BDZ are much better than those obtained without it: this damage zone seems to play an important role. Results reflect experimental results till Day 230. After day 230, they differ. Nevertheless, differences between results with and without BDZ - seem to remain more or less constant. We can imagine that undisturbed claystone starts to play a role.

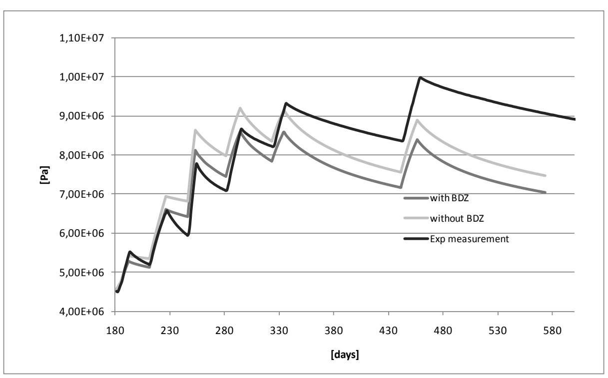

Figure 8. *Liquid pressure evolution in injection intervals: BDZ goal*

Finally, several parametrical studies have also been led. Specifically, the role of gas relative permeability has been studied by using a standard cubic law, improved by a Ck multiplied coefficient. This coefficient is not precisely known but proves to be decisive (Figure 5).

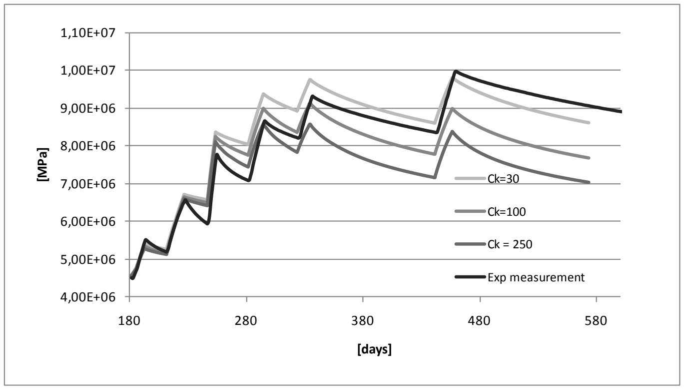

Figure 9. *Liquid pressure evolution in injection intervals: goal of relative permeability*

Conclusions

This contribution proposes a numerical modeling of a field scale test with a classical coupled two-phase flow model. The major difficulty in experience modelling is finally to understand and capture what happens in the instrumentation process and not only as far as the undisturbed clay is concerned. Indeed, it seems that different materials successively play a role: injection chamber and water displacement, borehole damage zone and at least undisturbed host rock. In both simulations and experimental results, we observe that the gas influence zone is very small and close to injection chamber. PGZ1 test is still in process.

Acknowledgements

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under Grant Agreement nº230357, the FORGE project. The authors would like to thank ANDRA (GL Transfert Gas) and ULg.

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Dilatancy driven gas flow in the Callovo-Oxfordian Claystone (COx)

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Summary

Observations from this study demonstrate the movement of gas in Callovo-Oxfordian Claystone is accompanied by bulk dilation of the fabric. Under these conditions, gas flow is along pressure-induced preferential pathways, where permeability is a dependent variable related to the number, width and aperture distributions of these features. These observations have led to the development of a new conceptual model for gas flow in COx, where the bulk movement of water is not prerequisite or required for advective gas flow.

1. Introduction

Movement of repository gases through argillaceous host rocks will occur by the combined processes of molecular diffusion (governed by Fick's Law) and bulk advection. In the case of a repository for radioactive waste, corrosion of ferrous materials under anoxic conditions will lead to the formation of hydrogen. Radioactive decay of the waste and the radiolysis of water will produce additional gas. If the gas production rate exceeds the rate of diffusion of gas molecules in the pores of the clay barrier, it is possible that a discrete gas phase could form (Ortiz et al. 2002) [1].

In a clay-based geological disposal facility (GDF), four primary phenomenological models describing gas flow can be defined (Marschall et al. 2005) [2]: (i) gas movement by diffusion and/or solution within interstitial fluids along prevailing hydraulic gradients; (ii) gas flow in the original porosity of the fabric, commonly referred to as visco-capillary (or 2-phase) flow; (iii) gas flow along localised dilatant pathways, which may or may not interact with the continuum stress field; (iv) gas fracturing of the rock similar to that performed during hydrocarbon stimulation exercises. Understanding the mechanisms controlling the advective movement of gas and its potential impact on the GDF host rock is crucial, both within performance assessment and the long-term prediction of the repository's evolution.

To investigate these issues the British Geological Survey (BGS) working in conjunction with the French radioactive waste management company Agence Nationale pour la Gestion des Déchets Radioactifs (Andra), undertook a series of laboratory-scale tests on preserved samples of the Callovo-Oxfordian argillite (COx), to define the key mechanisms governing gas migration in the COx.

2. Methodology

The basic permeameter consists of five main components: (1) a specimen assembly, (2) a 70 MPa rated pressure vessel and associated confining pressure system, (3) a fluid injection system, (4) a backpressure system, and (5) a PC-based data acquisition system (to minimise the chance of slug flow during gas testing). The specimen is subject to a confining stress, with the injection platen located on the base of the specimen. A novel feature of the apparatus is the use of porous annular guard-ring filters around the inflow and outflow filters (Figure 1). The pressures in these two guard-rings can be independently monitored. The advantages of the guard-ring approach are: (a) pore pressure evolution can be studied, (b) hydraulic anisotropy can be quantified in a single test, (c) a check can be made of flow symmetry in the specimen, (d) excess gas pressure at gas entry can be determined, and (e) uncertainties associated with possible sheath leakage can be eliminated from data interpretation.

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

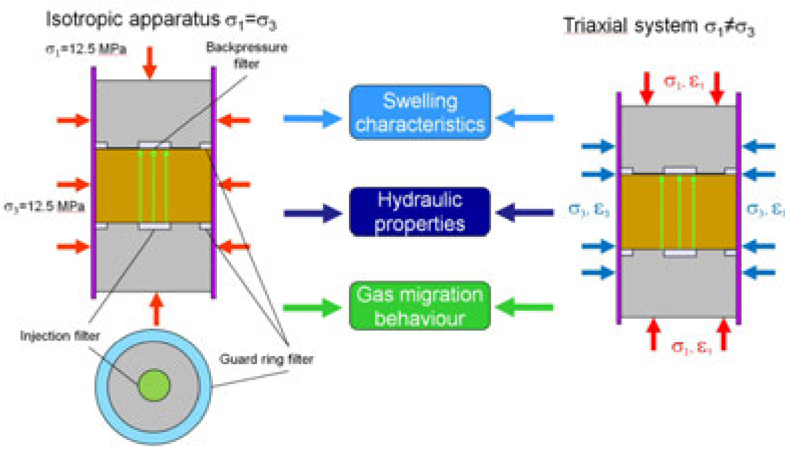

Figure 1 Schematic showing the sample assemblies for both isotropic and triaxial test systems.

Two variants of the apparatus were used. In the first, an isotropic confining stress was applied to the sample with data on volume change derived by careful measurement of the confining reservoir volume. In the second (tiaxial system), the volume change of the sample was directly measured using a combination of pressure balanced dash pots and LVDT's (Cuss et al., 2012) [3].

3. Results

To date, 5 long-term experimental tests have been performed on samples of the Callovo Oxfordian Claystone. In this way a detailed picture of the fundamental mechanisms controlling gas flow is beginning to emerge. The pressure at which gas begins to be mobile within the clay can be estimated from analysis of the injection and guard-ring filter data (Figure 2). This data suggests a significant degree of variability ranging from around 1 MPa to in excess of 5 MPa. This variability is thought to be linked to microstructural heterogeneity within the sample which may stem from engineered induced damage during excavation and sampling, local variations in

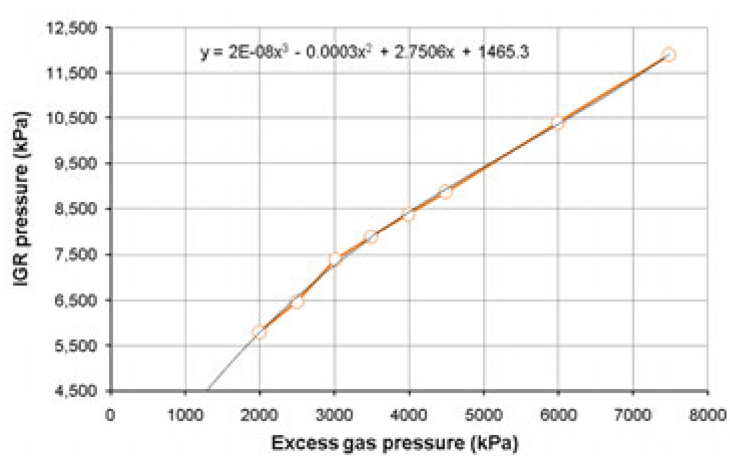

Figure 2 Example cross-plot of guard-ring versus excess injection gas pressure. Extrapolation of the line suggests a gas entry pressure of approximately 1.3 MPa

lithofacies (including perturbations due fossilised remains) and possibly natural occurring microfractures, a remnant artefact of the COx's burial history.

Gas breakthrough, signified by the emergence of gas from the downstream (opposite) end of the core, was also found to exhibit a significant degree of variability, though these pressures were in general, higher than those of breakthrough. This additional pressure component probably relates to scale and the associated increase in pathway lengths. Figure 3 shows a compilation of data from a number of tests performed under a range of test conditions. In general, breakthrough pressures appears to correlate to sample length, with low gas breakthrough pressures observed in short sample lengths and high gas breakthrough values seen in longer samples. The 'residual' values quoted by Aachen, support the hypothesis of localised gas flow, in effect bypassing the majority of the matrix, as their data clearly shows that the application of 3+ times the *in situ* confining stress, can yield similar gas breakthrough pressures to those normally associated with 'long-length' intact samples. However, the origin of the microfractures remains unclear and further work is required to understand this phenomenon.

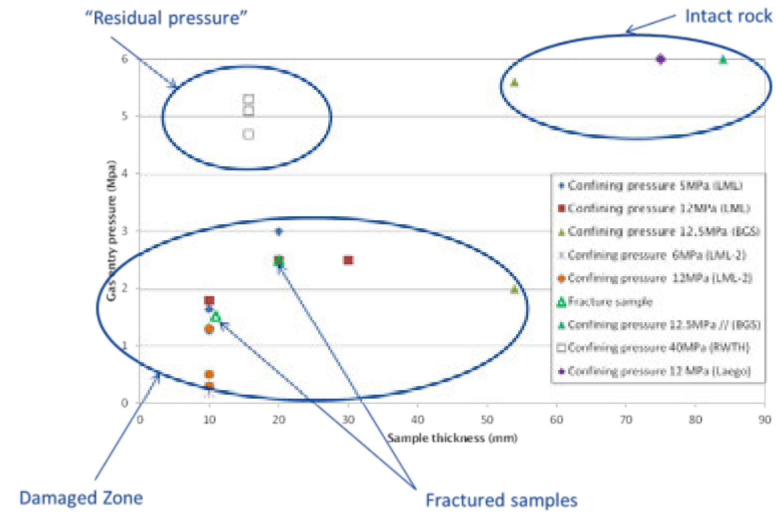

Figure 3 Cross plot of gas breakthrough pressure against sample length (image courtesy of Andra)

Inspection of the strain data during and after gas breakthrough clearly demonstrates that advective gas flow is accompanied by dilation of the samples (i.e. the formation of pressure induced micro-fissures) at gas pressures significantly below that of the minimum principal stress (Figure 4). As outflow evolves towards a steady-state, the sample undergoes an increase in volume, the size of which is beyond that that can be accounted for by the elastic compressibility of the sample.

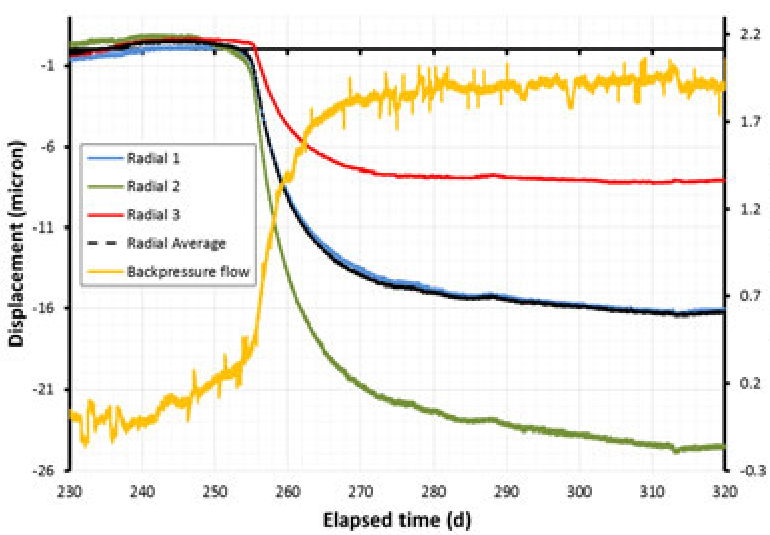

Figure 4 Data from a triaxial test showing a clear correlation bewteen sample volume and gas flow

Spontaneous changes in guard ring pressure and flow rate are indicative of dynamic pathway behaviour. These spontaneous changes in flux and pressure in and across the sample occur even when the system appears to be in some form of apparent steady-state.

Gas flow appears to occur through a local network of inherently unstable pathways, whose properties vary temporarily and spatially within the claystone signified by changes in flux and guard ring pressure (Figure 5). The coupling of variables results in the development of significant time-dependent effects, impacting many aspects of COx behaviour, from gas breakthrough time, to the control of deformation processes.

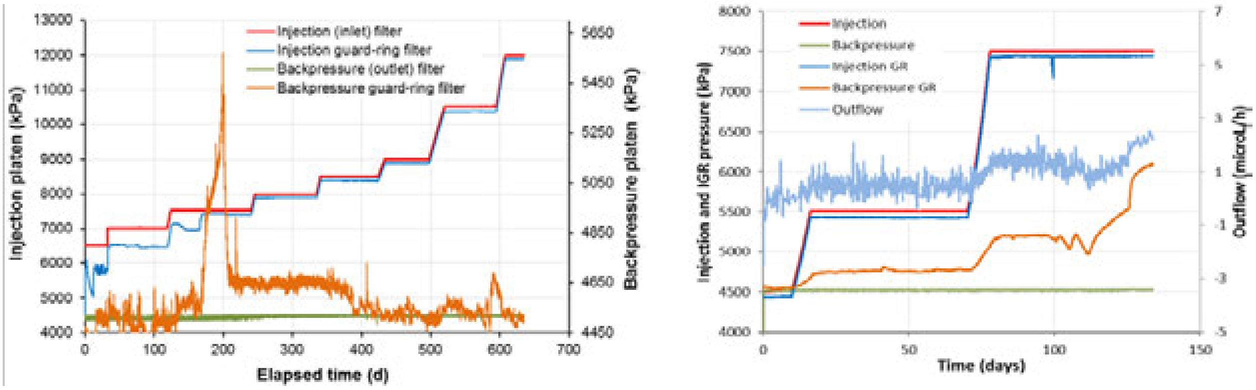

Figure 5 Data from isotropic testing showing spontaneous changes in flux and pressure symptomatic of an evolving network of dynamic and unstable pathways

Considerable evidence exists from analysis of pressure and flow data for the existence of multiple pathway networks which evolve independently in response to gas flow. Submersion and gentle heating of the samples in warm glycerol promotes degassing of the core (Figure 6). Localised streams of gas bubbles can be seen emanating from each face of the sample. While qualitative, the observation of localised gas flow may help to explain the apparent anisotropy in the radial strain data (Figure 4) and post-test measurements of water saturation which are always equal to unity. Visual observations of the degassing behaviour suggest a higher density of pathways on the downstream end of the core symptomatic of an expanding network of pathways which 'fan out' as they propagate through the core.

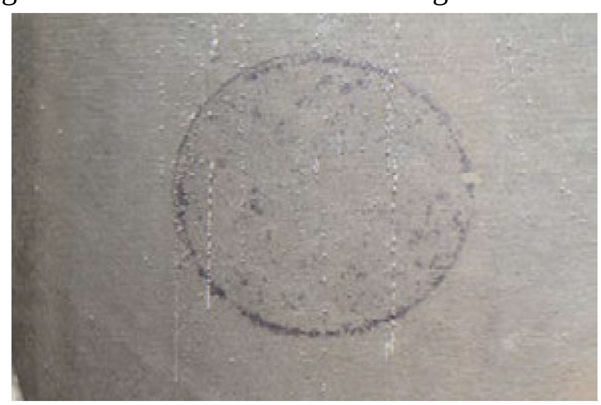

Figure 6 Post-test degassing of a sample. A control plug (not gas tested) yeilded little if any gas

In summary, a new conceptual model for gas flow in COx has been established, where the bulk movement of gas is accompanied by bulk dilation of the clay fabric, through a localised network of unstable, dynamic pathways, with minimal (if any) water displacement.

4. Acknowledgements

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under Grant Agreement no230357, the FORGE project.

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Sealing Efficiency of Bentonite/Sand Plugs: effect of gas pressure and location of gas pathways

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Summary

This study is an original contribution to understand the mechanism of gas transport in clay barriers and preferential gas pathways. Prior to gas breakthrough tests, bentonite samples are subjected to water pressure or both water and gas pressure for one or two months. To investigate the sealing efficiency of the interface between bentonite-sand plug and its surrounding (i.e. *in situ* claystone), three kinds of tubes are used, namely, a Plexiglass-aluminum^{TM} tube, an argillite tube and a dedicated aluminium tube with a grooved inner surface. Experimental results show that continuous gas breakthrough can be detected at 7MPa or more for a plug swelling without gas pressure, while gas can pass through the plug very easily for plugs swelling with water and a 4MPa gas pressure. No continuous gas breakthrough is found for a plug tested without surrounding tube. Gas permeability tests are performed on bentonite/sand plugs at different saturation levels under varying confinement: gas tightness can be obtained when saturation degree is more than 86% at an *in-situ* confinement, on the order of 7MPa.

1. Introduction

The design of long-term nuclear waste repositories includes the building of an engineered barrier around the waste containers, which aims to create a “low permeable zone” around them [1]. Bentonite-sand mixtures have been chosen by several industrialized countries as a buffer and backfill material to separate radioactive waste from the surrounding host rock. Indeed, bentonite-sand provides low permeability (when fully water-saturated), swelling and self-healing ability, together with high expandability and fair ductility. Nevertheless, several failure scenario need to be investigated to assess proper safety of this solution. Firstly, *in situ*, the interface between the buffer material and the disposal wall is the weak point whereby underground water may pass. Secondly, owing to water uptake from the host rock, sealing will be obtained gradually from bentonite-sand plug swelling [2]. Meanwhile, hydrogen gas generation is expected, due to humid corrosion, degradation of organic matter or water radiolysis, so that gas pressure may apply on the plug and hinder its swelling and sealing efficiency. Thirdly, into the massive disposal seal made of a number of bentonite-sand plugs (or bricks), a significant water saturation gradient is present between the core and the external surface, itself in contact with argillite and underground site water [3]. As a consequence, owing to a high water content, the external part of the massive barrier swells in contact with an extremely stiff host rock (e.g. argillite), so that it applies a confining pressure to the partially water-saturated core. The question here is to assess whether the central part of the bentonite/sand barrier is actually permeable to gas.

More precisely, this contribution summarizes the experimental investigation of 1) bentonite–sand plug swelling pressure and kinetics, and its gas breakthrough pressure, in presence of both water on one side, and gas (4MPa), on the other side, 2) gas permeability of samples under confinement (loading and unloading) and varying saturation levels, and 3) gas breakthrough test of a bentonite-sand plug swollen within a tube with a groove on its inner face (to show that gas passes at this interface in the absence of any groove), and also within a tube of argillite.

2. Methodology

2.1 Bentonite-sand plug and argillite tube preparation

The material used in this study is made of a single bentonite-sand mix intended to target a given swelling pressure after *in situ* water saturation, as expected in the storage structure. This pressure is around 7MPa for a mixture of silicate sand and bentonite, with a sand mass proportion of 30%. The mixture is supplied to our lab by the CEA Agency. Bentonite is a classical MX80 type. The swelling pressure is obtained for a mixture compacted at a dry density of 1.77, with a water content of 15.2%. To obtain the adequate water content, the bentonite-sand mixture is let in an atmosphere at controlled relative humidity (85%) until mass stabilization, and then compacted in a steel tube at 12MPa axial pressure. The resulting sample is of 25mm height and 42.5mm diameter. For the argillite tube, after coring, both the top and bottom surfaces are ground to ensure proper flatness.

2.2 Experimental method

In this study, four series of tests are presented, see Table 1.

Table 1: Definition of all test types performed: for samples A7 and B2, the inner tube surface is smooth, while for sample A5, the inner tube surface is grooved, i.e. rough, which makes potential gas passage more difficult; for samples S3-9~S3-13, each bentonite-sand plug is placed in the Viton™ membrane, without any surrounding tube.

Number	Boundary conditions		Note
	Pg(MPa)	Pw(MPa)	
A5	0	4	1) Direct contact with 4MPa water; 2) With a grooved inner surface of the Plexiglass™-aluminium tube
A7	0	4	1) Direct contact with 4MPa water; 2) Swelling within an argillite tube
B2	4	4	1) Direct contact with a fully saturated plug; 2) Swelling with a Plexiglas-aluminum™ tube
A5	0	4	1) Direct contact with 4MPa water; 2) With a grooved inner surface of the Plexiglass™-aluminium tube

Fig.1 displays the experimental setup. The triaxial cell provides hydrostatic loading to the tube by pressurizing oil around it. Prior to being placed inside the triaxial cell, the plug-tube is jacketed inside a protective Viton™ membrane. For the test on sample B2, two tubes are put over one another in the membrane, the bottom one is filled with a fully saturated bentonite plug, and the top one is filled with a second compacted plug. This second plug is supplied with water by the first one, which is intended to be realistic toward a possible *in situ* saturation case. Water is injected by a Gilson™–type pump from the upstream side at the *in situ* average pressure of 4MPa. Gas pressure is injected from the top side at a constant value, e.g. 4MPa. For the tests on samples A5 and A7, only one plug and tube are put inside the membrane, and no gas pressure is applied during the swelling test. For gas breakthrough tests, gas is injected from the upstream side and gas detection is conducted in a downstream chamber by both a manometer (+/-1-10mbar relative) and a dedicated argon gas detector (+/–10–4ml/sec). The gas pressure increases regularly (by 1MPa steps every one to three days) until discontinuous and continuous gas flows are detected on the other side.

3. Results and discussion

As shown in Fig. 2, three kinds of gas breakthrough pressures are measured. Firstly, for sample A7, bentonite swelling occurs inside a tube of argillite and without gas pressure: continuous gas breakthrough is measured slightly above Pg=7MPa, and this pressure is equal to the *effective swelling* *pressure* between the bentonite and the inner surface of the tube, see [4]. Secondly, for sample B2, bentonite has been swelling within a Plexiglas-aluminum™ tube and with 4MPa gas pressure: gas passes through the plug–tube at a much lower gas pressure, of about 2.5MPa. This means that the bentonite-sand plug is only partially saturated. Thirdly, for sample A5, bentonite has been swelling within a grooved tube: no continuous gas passage is measured until 10.5MPa gas pressure. This means that gas cannot migrate through the plug-tube so easily as when the interface is smooth, i.e. gas passage occurs at the bentonite plug/tube interface when the interface corresponds to a smooth tube.

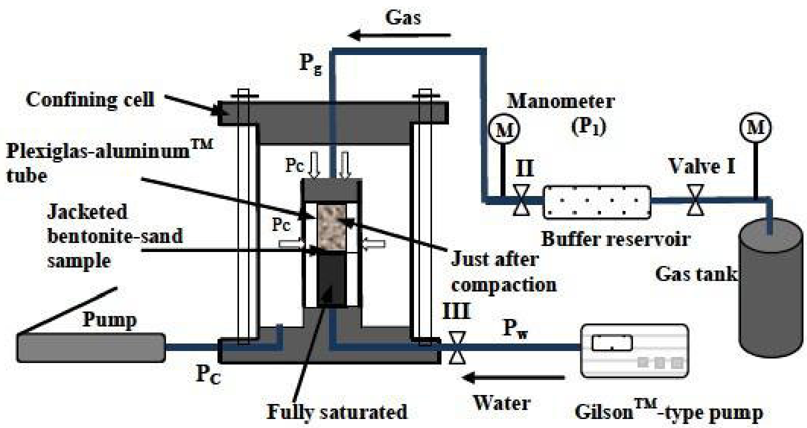
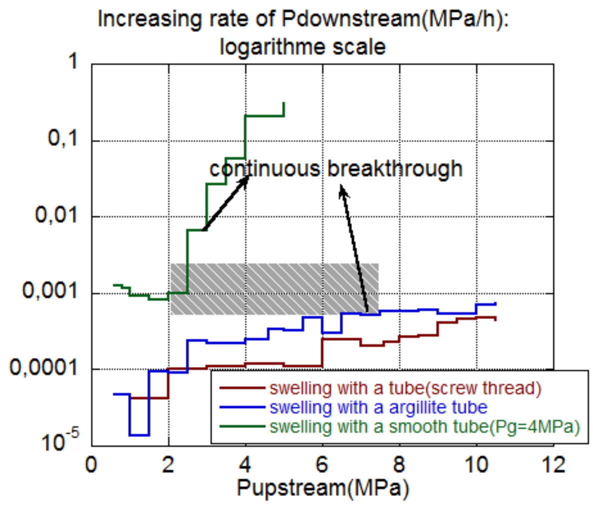


Fig. 1: Schematic diagram of the swelling test



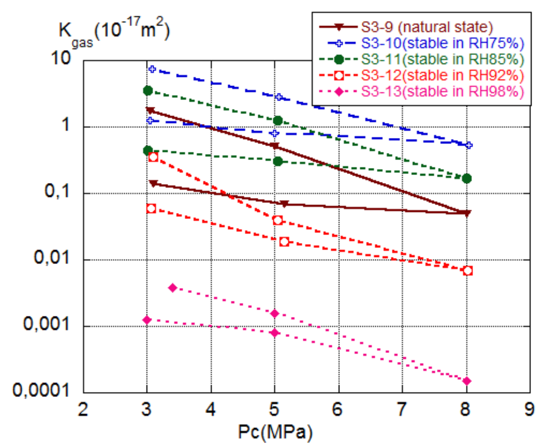


Fig. 2 (left): Gas breakthrough tests: relationship between the upstream gas pressure and the rate of increase of downstream gas pressure. Fig. 3 (right): Gas permeability of sample S3 series at different Pc and Sw.

Fig. 3, above, present the results of gas permeability tests at different saturation levels and confinement. It is observed that gas permeability becomes very low from 5 MPa confining pressure for sample S3-12 (RH=92%) and S3-13 (RH=98%), despite their incomplete saturation: Sw (S3-12, RH=92%)=72.8% and Sw(S3-13,RH=98%)=91.7%. When accounting for their initial difference in gas permeability, these samples follow parallel evolutions when Pc varies.

4. Conclusions

This contribution is a summary of our PhD results. It aimed at evaluating experimentally the sealing efficiency of an argillite-bentonite interface subjected to gas pressure. In order to identify actual gas pathway, e.g. through the tube/bentonite interface or within the bentonite bulk, a dedicated grooved tube is used. Our main results are as follows:

- When gas pressure (of at least 4MPa) is imposed together with water pressure, the bentonite-sand plug saturates only partially, and associated continuous gas breakthrough pressure is very low, at a value of only 2.5MPa.
- No continuous gas breakthrough is measured until 10.5MPa gas pressure, when the bentonite/sand plug-tube interface is made more tortuous by the inner surface grooving. This means that it is very difficult for gas to transfer through a fully saturated bentonite-sand plug, and that whenever a smooth interface with the tube is offered, gas passage is made easier.
- For bentonite swelling with an argillite tube (and yet no gas pressure at this stage), continuous gas breakthrough pressure is slightly greater than *effective swelling pressure*. When comparing with gas breakthrough pressure of sample A5, swollen in contact with a grooved inner tube surface, it is inferred that gas transfer occurs through the interface between bentonite/sand and argillite when continuous gas breakthrough occurs.
- Tightness to gas can be achieved under confinement for samples initially saturated up to 86-91% only.

Acknowledgments

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under Grant Agreement no. 230357, the FORGE project.

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Gas Generation in Real L/ILW Container Drums and Near Surface Vaults

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To obtain reliable estimates of the quantities and rates of the gas production in L/ILW a series of measurements was carried out since 2000 in Hungary using real L/ILW drums and real closed L/ILW vaults. The investigated L/ILW drums are filled with waste form the light water PWR type Paks Nuclear Power Plant of Hungary. The investigated L/ILW vaults are located in a near surface L/ILW disposal facility of Hungary. Gas samples from the drums and the vaults were taken using special tools and delivered to the laboratory for further analyses.

Qualitative and quantitative gas component analyses of headspace gases were executed by a quadrupole mass spectrometer (Omnistar 200 M2, Pfeiffer Vacuum). Tritum (H-3) samples separated form the gases are measured by a low background liquid scintillation counter (TRICARB 3170TR/SL, Packard). For C-14 activity measurements of the carbon content of the produced gas was measured by a low background gas proportional counter system (GPC, ATOMKI). ^{13}C and ^{15}N stable isotope ratio shift of carbon dioxide and nitrogen samples were measured by a DeltaXP^{plus} stable isotope mass spectrometer (Thermo Finnigan Co.). ^{3}He and helium content of the gas were measured by a VG5400 noble gas mass spectrometer (Fissons Instruments).

At the end of year 2003 ten drums (200 liter internal volume steel drums covered by plastic based protective painting) filled with representatively selected original L/ILW were placed into special hermetic containers equipped with sampling valves for repeated sampling. These hermetic containers were stored at the same site where the L/ILW is stored primarily in the light water PWR type Paks Nuclear Power Plant of Hungary (Paks NPP). The typical gas production rates were 0.05-0.2 STP litre gas/day for CO_2 and CH_4 generation, and less than 0.02 STP litre gas/day for H_2. No explosive gas mixture was indicated in the L/ILW drums during the investigated storage period.

Headspace gas analysis of real radioactive waste vaults closed between 1979 and 1995 in a near surface L/ILW disposal facility of Hungary was also carried out. Compositions of headspace gases in closed L/ILW vaults were in good agreement with gas generation processes observed in the L/ILW drums.

The stable carbon isotope measurements show that the main source of the CO_2 gas is the degradation of organic matter in the waste. The low ^{13}C content indicates microbial degradation processes as the main sources of CH_4 in the headspace gas. Typical tritium activity concentrations were between 0.1 and 10 Bq/liter gas in the drums and between 10 and 1000 Bq/liter gas in the vaults. Typical C-14 activity values of the headspace gases were between 0.1 and 2.0 Bq/liter gas in the drums and 10 and 1000 Bq/liter gas in the studied vaults.

Carbon Steel Corrosion and Hydrogen Gas Generation in Cementitious Grout

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Summary

Measurements of hydrogen gas production rates have been performed in cells containing carbon steel embedded in cementitious grout under anoxic unsaturated 100% relative humidity conditions. The experiments use a highly sensitive solid state electrochemical hydrogen sensor. Additional measurements of steel corrosion rates using the same sensor were performed in saturated grouts and in alkaline solutions without grout for comparison to previous work. The studies are aimed at providing data relevant to the extended unsaturated phase which is expected to occur after waste emplacement in the case of a repository in low permeability clay rocks.

1. Introduction

An important issue in safety assessment of low and intermediate level (L/ILW) repositories is the production of hydrogen gas from anaerobic corrosion of various metals used in waste drums and present in the wastes themselves. Even though the corrosion rates of many metals that passivate in the alkaline conditions of the repository are very low, the total surface area of metals in a L/ILW cavern may be large, thus accumulation of gas can gradually generate significant pressure within the waste caverns, depending on the gas permeability of the host rock and repository seal system. It is thus of some interest to have improved quantification of the gas production rates. In this study we present results for corrosion of carbon steel, the most common metal in the Swiss L/ILW inventory, under conditions closely approximating those in a repository. In particular the studies have focused on measuring hydrogen generation from steel samples embedded in humid unsaturated grout, for which few data are available. This is of particular interest as simulations of repository resaturation in low permeability clay rocks illustrate that unsaturated conditions can be maintained for many thousands of years after emplacement of the wastes (Nagra 2008). We note that the extent of non-uniform corrosion is not the present focus of work, because it is expected to occur only under aerobic conditions (Smart et al. 2004) and even if it does occur, it would have little impact on hydrogen gas generation.

2. Experimental

Carbon steel rods were embedded in cementitious grout (~25% porosity) that comprises only cement, sand and water and exposed to unsaturated conditions (100% relative humidity (RH)) in a deaerated sealed glass cell at 50°C, as shown in Figure 1. Separate measurements confirmed that ~100 % RH would be maintained during the corrosion experiments. Some experiments were also performed with fully saturated conditions (grout pores filled with synthetic Opalinus Clay pore water or with saturated $Ca(OH)_2$ solution). Control experiments (i.e. containing grout, but no steel) were also performed. In order to compare the results with those from prior studies, additional experiments were performed in saturated $Ca(OH)_2$ solution with no grout.

The hydrogen measurements were made using a solid state electrochemical hydrogen sensor, which permits equivalent corrosion rates down to about 0.1 nm/a to be determined under our experimental conditions. The probe responses are satisfactory for corrosion rate estimation to within a factor of 2 or better. Probe calibrations were performed with H_2 mixtures (typically 104 ppm H_2, balance N_2). Further details are given in Newman and Wang (2010).

The hydrogen production rates over time were measured periodically for up to several thousand hours, by which time the gradually decreasing rates typically appear to have stabilized.

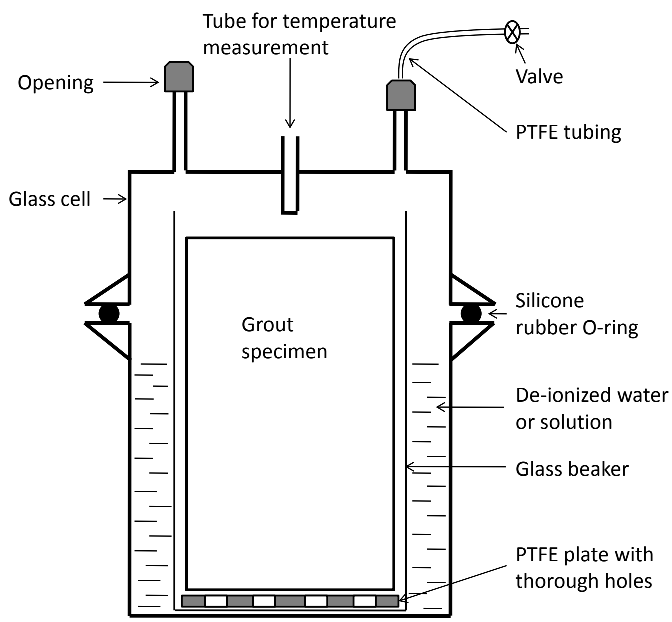

Fig. 1: Corrosion cell and grout specimen with embedded steel rods (Newman and Wang 2010)

3. Results and discussion

Corrosion rates were calculated assuming the reaction

$$Fe + 2H_2O \rightarrow Fe(OH)_2 + H_2\uparrow$$

Results of experiments performed with steel rods immersed in $Ca(OH)_2$ solution (final pH 12.6) with no grout were in excellent agreement with long-term values obtained at 25°C by Kreis (1991) and at 50°C by Smart et al. (2004). The corrosion rate was initially 30-40 nm/a, decreasing to 2-3 nm/a after 2000 hours, whereas Kreis and Smart et al. reported final rates of about 5-7 nm/a and 20-30 nm/a, respectively at somewhat longer times.

In the case of steel rods embedded in grout with pores filled with Opalinus Clay pore water (final pH 13), the corrosion rate after 1300 hours was 1-2 nm/a.

In the corrosion experiments with unsaturated grout, the derived corrosion rates also appeared to be about 1-3 nm/a, with some initial variations that may relate to different surface treatments of the steel prior to the experiments. Some typical results are shown in Figure 2. All experiments performed under unsaturated conditions showed a characteristic behaviour with initial rates of about 10-30 nm/a, decreasing to a few nm/a after 500 to 2000 hours. This differs somewhat from results of experiments performed in fully saturated conditions without embedment in grout (e.g. Smart et al. 2004 and Fujisawa et al. 1997, 1999), which typically showed much higher initial rates (ca. 100-200 nm/a, declining eventually to 5-30 nm/a). The reasons for the low initial rates are not clear, although it is possible that the presence of adsorbed water films in unsaturated grout may reduce mass transport of Fe(II) compared to the case of fully saturated systems. Alternatively, the blocking of the surface by cement phases may retard dissolution of Fe(II), leading to more rapid passivation.

Despite the relatively consistent results obtained in our studies, the results obtained with control cell measurements (i.e. grout only) throw the derived corrosion rates into question, as hydrogen production was detected with all grout samples tested at rates comparable to those determined for grouted steel rods. Separate experiments with either the cement or the quartz sand aggregate showed that the cement is largely responsible for the hydrogen production, which tends to decline significantly after 1000-2000 hours. The results suggest that our derived corrosion rates of a few nm/a may represent upper limit values. The reactions that produce hydrogen in grout are not known, but it is possible that it occurs as a result of corrosion of trace Fe(0) arising from the steel used to grind the cement clinker.

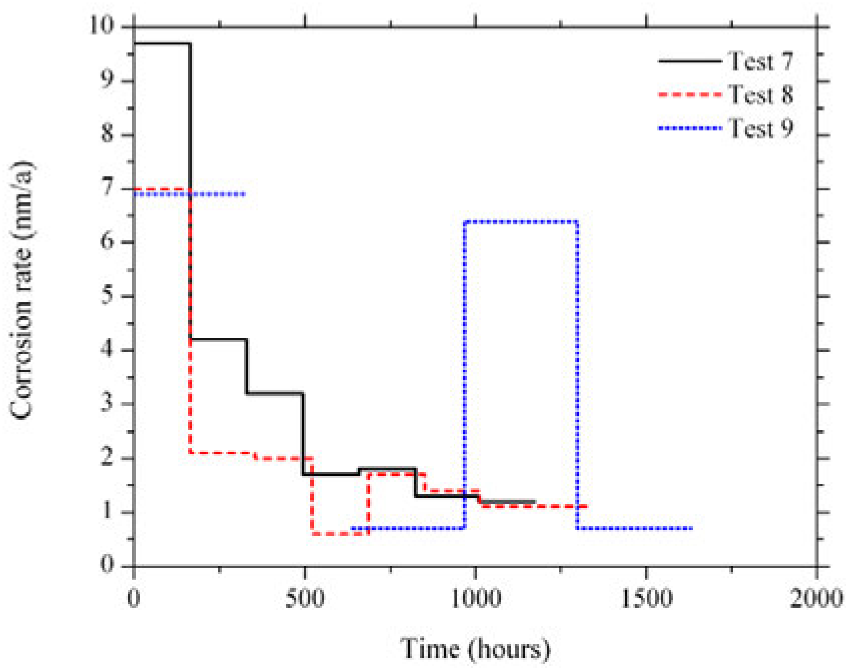

Figure 2: Derived corrosion rates based on H_2 gas generation for three samples of carbon steel embedded on cementitious grout with 100% RH: Test 7 – sample pickled prior to test in 10% HCl, Test 8 – sample sanded with emery paper, Test 9 – sample pre-corroded in 0.5 M NaCl (measurement missing for one time interval)

4. Conclusions

The long-term corrosion rate of carbon steel at 50°C under alkaline solutions can be summarized as follows:

1) Bare steel in saturated in $Ca(OH)_2$ solution: 2-3 nm/a
2) Steel embedded in cementitious grout with Opalinus Clay pore water: 1-2 nm/a
3) Steel embedded in unsaturated cementitious grout (100% RH): 1-2 nm/a.

The latter rate probably represents an upper limit, as some hydrogen is produced by unknown reactions in the grout.

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Carbonation of repository cement: Impact of CO_2 on cement mineralogy, water chemistry and permeability

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In the current UK reference concept for low/intermediate radioactive wastes, large quantities of cementitious materials will be used for repository construction and buffer/backfill. Degradation of organic material within the waste will produce CO_2, which will lead to cement carbonation. This will reduce the capacity of the buffer/backfill cement to maintain highly alkaline conditions, and as a consequence increase the likelihood of radionuclide migration. Conversely, some carbonation reactions might improve material properties, e.g. by slightly reducing cement permeability. The migration of CO_2 will be controlled by a complex interplay between transport processes and chemical reactions, and currently it is unclear whether the overall changes due to carbonation will be beneficial or deleterious to long-term radionuclide immobilisation.

As part of a pan-European Euratom 7th Framework initiative examining the fate of repository gases (FORGE project - Grant Agreement 230357), we have undertaken a laboratory study to examine the impact of carbonation on 'Nirex reference vault backfill' (NRVB) cement. Aims for the work were to quantify changes in cement mineralogy, structure, porosity/permeability, and the composition of coexisting aqueous fluids (Rochelle et al., 2013).

The cement composition consisted of approximately 26% Portland cement, 28.6% limestone powder, 9.8% lime, and 35.6% water. This cement is designed to be relatively permeable in order to prevent gas build up near canister vent holes, and hence dissipate excess gas pressure. Cement slurry was cast into cylindrical blocks of approximate size 12 cm diameter by 30 cm long, and was cured for at least 40 days prior to subsampling for use in the experiments. Subsamples consisted of small cores taken from these larger blocks.

The laboratory investigations involved two types of experiments to investigate processes happening under very low flow (diffusive) conditions, and at much higher flow rates:

- Thirty-two static 'batch experiments', where migration of CO_2 was controlled by diffusive processes. The small cylindrical samples of cement used were 25 mm in diameter and 50 mm long. The experiments had durations of 10-365 days, and were conducted under a range of potential *in-situ* conditions; 20°C or 40°C, 4 MPa or 8 MPa, with or without 'young' (Na/K/Ca-rich) or 'evolved' (Ca-rich) cement porewaters, with or without CO_2.
- Four dynamic experiments, where high-pressure pumps were used to create an advective flow through cement samples 49 mm in diameter and 49 mm long. Experiments involving flow of free phase CO_2 were conducted at 40°C and 4 MPa (two tests with gaseous CO_2) and at 40°C and 8 MPa (supercritical CO_2). An experiment involving flow of dissolved CO_2 was conducted at 40°C and 4 MPa.

All the cement samples showed rapid reaction with CO_2, manifested by a colour change from grey to light brown. The y remained intact, even after prolonged exposure to CO_2-rich fluids.

Careful measurements of sample dimensions from the static batch experiments showed that they did not shrink or swell during carbonation. However, carbonation was associated with an increase in weight of over 8% (Figure 1). Weight increase was favoured with increasing time, temperature, and CO_2 concentration. Free phase CO_2 gave slightly more reaction than dissolved CO_2, possibly because of its higher concentration and greater ability to penetrate the samples. CO_2-induced weight increases were far larger than those in equivalent nitrogen-pressurised experiments, where small weight increases were due to continued slow hydration. Weight losses occurred in the dry nitrogen experiments due to dehydration of the cement.

CO_2			Temperature °C							
			20°C				40°C			
			YNFP	YNFP+Cl	ENFG	DRY	YNFP	YNFP+Cl	ENFG	DRY
Pressure (bar)	40 Bar		Gaseous CO_2				Gaseous CO_2			
		40 Days	-	-	-	-	+4.84%	+2.56 %	+2.58 %	+8.20 %
		1 Year	-	-	-	-	-	-	-	-
	80 Bar		Liquid CO_2				Supercritical CO_2			
		10 Days	-	-	-	-	+3.06%		-	-
		20 Days	-	-	-	-	+4.49%		-	-
		40 Days	+4.23%	+5.25 %	+3.57 %	N/A	+5.59%	+7.68 %	+4.50 %	+7.63 %
		1 Year	-	-	-	-	-	+6.84 %	+4.79 %	+8.43 %
							Run 1351: N/A			

N_2			Temperature °C							
			20°C				40°C			
			YNFP	YNFP+Cl	ENFG	DRY	YNFP	YNFP+Cl	ENFG	DRY
Pressure (bar)	40 Bar		Gaseous N_2				Gaseous N_2			
		40 Days	-	-	-	-	+0.69%	-0.75 %	+0.58 %	-2.39 %
		1 Year	-	-	-	-	-	-	-	-
	80 Bar		Gaseous N_2				Gaseous N_2			
		40 Days	+0.65%	+0.97 %	+2.06 %	-3.03 %	+0.30%	+0.12 %	-0.04 %	-3.91 %
		1 Year	-	-	-	-	-	+3.37 %	+2.21 %	-8.83 %
							Run 1350: N/A			

Figure 1 Percentage weight changes of cement samples relative to their initial weights. YNFP was a young near-field porewater dominated by Na-K-Ca, YNFP+Cl was a Na-K-Ca-Cl porewater of more intermediate composition, and ENFG was an evolved near-field groundwater dominated by Ca and containing some Cl.

Weight increases during carbonation reflect the breakdown of portlandite ($Ca(OH)_2$) and calcium silicate hydrate (CSH) phases (the two main components of cement), and the formation of calcium carbonate phases and silica gel. The observed colour change was possibly caused by CO_2-enhanced reaction of small amounts of calcium ferrite minerals in the cement and oxidation of the iron to give a ‘rusty’ colour. Carbonation was associated with reaction zones/fronts that moved several mm through the cement over time (Figure 2):

Zone 1 (innermost zone) = minor carbonation with minimal apparent volume change

Zone 2 = significant carbonation and localised shrinkage

Zone 3 = complete conversion of portlandite and CSH

Zone 4 = dissolution of initially-formed carbonate minerals in the outer parts of the sample by the surrounding, slightly-acidic water.

The shrinkage in Zone 2 was expressed as small fractures (typically several mm long), though these do not appear to extend beyond this zone. Zone 3 contained a ‘3D chicken wire’ meshwork of interconnected, higher-density, carbonate-filled micro-fractures (typically on a 10s-100s μm scale) that separated silica-rich areas having lower-density and high porosity, and concentric ‘relic’ reaction fronts. The small fractures of Zone 2 appear to have filled with secondary precipitates in Zone 3. Carbonation was not always uniform however. Heterogeneities developed in some parts of the cement during casting, resulting in coarser layers dominated by portlandite, and finer layers dominated by CSH

(Figure 2). The portlandite was more reactive towards CO_2, and carbonation was enhanced along these layers.

Appreciable amounts of a secondary Cl-rich, calcium chloroaluminate phase were formed in both N_2- and CO_2-pressurised experiments. However, more of this formed in the CO_2 experiments, focussed in a region at the boundary of Zones 2 and 3. The formation of this phase appears to have been favoured by the alkaline conditions associated with portlandite and CSH phases. In the CO_2 experiments this resulted in the continued formation, dissolution and reformation of this phase in a region just in advance of the main carbonation front. The formation of Cl-rich phases within a repository could benefit its safety functions, as it might help to immobilise/retard ^{36}Cl if it were to leach from the waste.

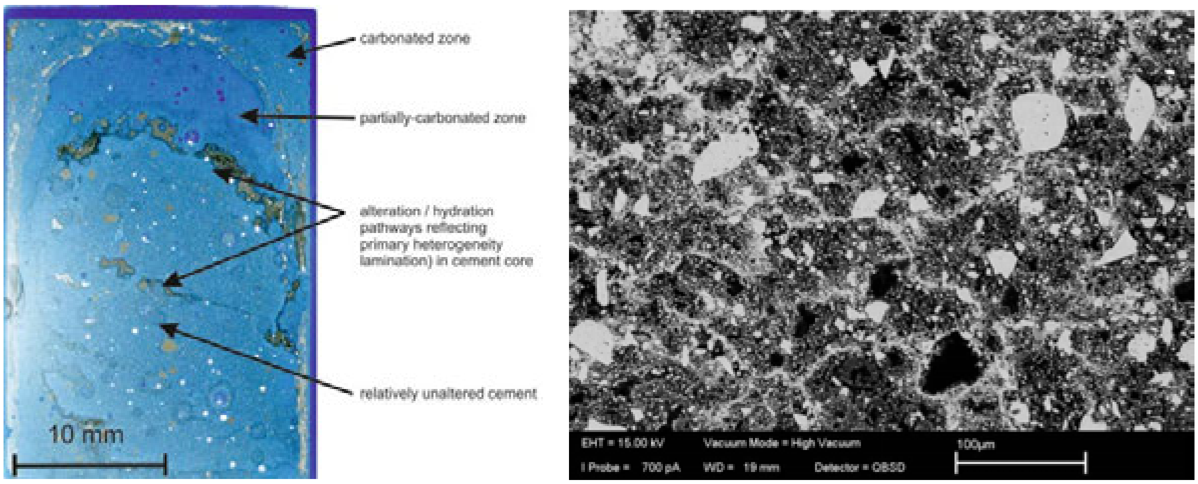

Figure 2 Left: Transmitted light image of a dyed polished thin section through core of NRVB reacted with 'YNFP+Cl' saturated with gaseous CO_2. Different degrees of carbonation are highlighted, together with pre-existing heterogeneity in the original cement. Right: Backscattered SEM image of typical 'chicken wire' fabric of higher density carbonate precipitation (light coloured lines), separating regions of lower density, mainly silica gel (dark).

Aqueous fluids in contact with the cement samples in the static batch experiments showed compositional changes that reflect the mineralogical changes. In the absence of CO_2, the pH of solutions in contact with the cement attained values of 12-13 expected for cement-equilibrated waters. On addition of a CO_2 the pH dropped to approximately 7 (as measured on depressurised samples at 25 °C) due to the consumption of hydroxyl ions and the formation of bicarbonate ions. This demonstrates the efficacy of CO_2 in reducing the high pH buffering capacity of buffer/backfill cement. Significant increases in concentration of dissolved Ca and SiO_2 were also found. These reflect the dissolution of the two most important phases in cement (portlandite and CSH phases) and equilibration with secondary phases such as calcite and amorphous silica.

Controlled flow rate carbonation experiments were conducted on isotropically-confined NRVB cement samples to investigate changes in flow properties in response to CO_2 ingress. Four tests were conducted to measure both hydraulic and gas transport properties (Figure 3):

1) Partial carbonation of a sample using CO_2 gas (NRVB-1)
2) Full carbonation of a sample using CO_2 gas (NRVB-2)
3) Full carbonation of a sample using supercritical CO_2 (NRVB-4)
4) Full carbonation of a sample using water saturated with gaseous CO_2 (NRVB-5)

These experiments revealed decreases in overall sample permeability as a consequence of porosity reduction due to conversion of portlandite and CSH to secondary carbonate minerals and silica gel. Injection of free-phase (gaseous and supercritical) CO_2 halved permeability, whereas use of dissolved CO_2 reduced hydraulic permeability by about 3 orders of magnitude. Detailed petrogaphic observations of partly-reacted cement samples show a series of reaction zones as per the static experiments. These observations, coupled with micropermeameter data, show the greatest reductions in porosity and permeability in a very narrow zone at the leading edge of the visible alteration front. Permeability reduction could be beneficial within a repository setting, as it could lower the potential for radionuclide migration. Permeability of the carbonated cement is not completely blocked however, so there is still potential for the cement to vent gas, and prevent build up of excess gas pressure in and around the waste canisters.

Test	Initial hydraulic permeability (m^2)	Hydraulic permeability after aqueous CO_2 injection (m^2)	N_2 gas permeability (m^2)	CO_2 permeability (m^2)
NRVB-1	4.3×10^{-17}	-	N/A	N/A
NRVB-2 before N_2	4.1×10^{-17}	-	-	-
NRVB-2 after N_2	4.2×10^{-17}	-	2.20×10^{-19}	-
NRVB-2 after CO_2	-	-	1.13×10^{-19}	1.00×10^{-19}
NRVB-4	4.0×10^{-17}	-	-	1.94×10^{-19}
NRVB-5	4.7×10^{-17}	4.28×10^{-20}	-	-

Figure 3 Summary of average permeability results from the NRVB cement carbonation tests.

Carbonation features and secondary phases observed in these experiments using a relatively porous/permeable cement, bear many similarities to those found in other systems. This includes experimental residues and recovered well samples of far lower porosity/permeability borehole cements used in CO_2-storage operations (Carey et al., 2007; Rochelle and Milodowski, in press). There are also similarities to samples of naturally-occurring CSH phases which have been naturally-carbonated over prolonged timescales (Rochelle and Milodowski, in press). A number of common carbonation processes may be operating in all these systems. Some uncertainties in the carbonation process remain however, and it would be useful to; quantify the longevity of the reaction zones and whether they evolve into a single reaction front over longer timescales, better define the role of micro-fracturing during carbonation, quantify how efficient secondary phases are at 'armouring' cement from further carbonation, and precisely identify the Cl-rich secondary phases and the role they might play in ^{36}Cl retardation.

Acknowledgements

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FORGE mock-up experiment to simulate controlled gas release from a L/ILW repository

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Summary

Gases (hydrogen, methane, carbon dioxide) may accumulate in the emplacement caverns of a geological repository for low/intermediate-level waste (L/ILW) due to the corrosion and degradation of the wastes. Nagra is evaluating the concept of an engineered gas transport system (EGTS), aimed at limiting the gas overpressures in the backfilled underground structures of a repository to an acceptable level without compromising the radionuclide retention capacity of the engineered barrier system (EBS). The main design elements of the EGTS are (i) specially designed backfill materials for the emplacement caverns with high porosity and high compressive strength and (ii) gas permeable tunnel seals, consisting of sand/bentonite (S/B) mixtures with a bentonite content of 20% to 30%.

Preliminary experimental studies on the laboratory scale confirmed the low water permeability and the enhanced gas transport capacity of S/B mixtures. These experiments showed that it is possible to design S/B mixtures with specific target permeabilities for water and gas flow. With respect to the impact of hyperalkaline solutions originating from concrete components (e.g. backfill mortar, waste fixation concrete, tunnel lining), modeling studies indicate that water transport will be reduced as a result of high-pH water interaction with the S/B material. However, there are uncertainties regarding the time scales of this process and whether and to what extent it influences the gas transport properties of S/B mixtures.

The FORGE Mock-up experiment consists of a 60 cm thick S/B layer (80/20) confined at both ends by a high-porosity (OPC) mortar (Fig. 1). The test aims at providing experimental evidence for water and gas transport through S/B under hyper-alkaline boundary conditions at an intermediate spatial scale (compared to repository scale). Water saturation with 20 bar pressure was started in June 2010 and water inflow, water saturation (by TDR), pore pressures and total pressures were observed continuously at various points. A synthetic Opalinus Clay pore water was used as injection water to mimic water ingress from the rock formations overlaying a future repository.

Saturation was completed in August 2012 and N_2 gas injection was started in October 2012 by exchanging the water at the gas side of the experiment and applying a pressure gradient between injection and extraction side of initially 0.5 bar. The pressure gradient was increased continuously to assess the gas entry pressure of the material and the gas transport capacity after breakthrough.

Introduction

Gases (hydrogen, methane, carbon dioxide) may accumulate in the emplacement caverns of a geological repository for low/intermediate-level waste (L/ILW) due to the corrosion and degradation of the wastes. Swiss low- and intermediate level waste (L/ILW) repository concept foresees cavern plugs and a repository seal to prevent excess gas pressures in the repository. The main aim of the plugs and the seal are to increase the gas transport capacity of the backfilled underground structures without compromising the radionuclide retention capacity of the engineered barrier system [Nagra, 2008]. The design option is called "engineered gas transport system (EGTS)" (Fig. 1). It involves specially designed backfill and sealing materials such as high porosity mortars as backfill materials for the emplacement caverns and sand/bentonite (S/B) mixtures with a bentonite content of 20 – 30 % for backfilling other underground structures and for the seals.

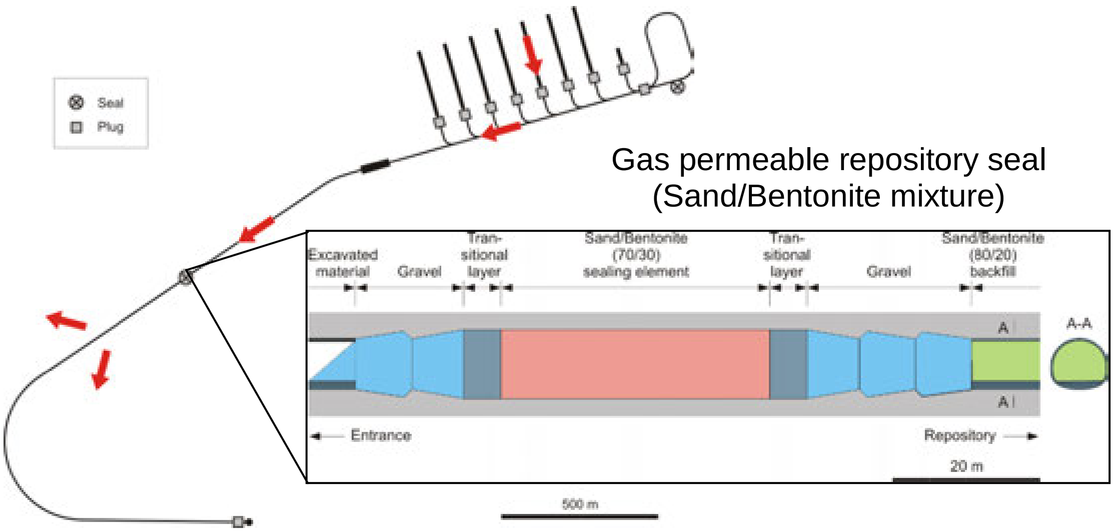

Figure 1: Schematic picture of repository layout and layout of repository sealing (bottom right). Red arrows indicate gas release pathways.

Preliminary experimental studies confirmed the high gas transport capacity of the S/B mixtures (Romero et al., 2003). These experiments have shown as well the ability to design S/B mixtures with specific target permeabilities for water and gas flow (e.g. Tashiro et al. 1998). First in-situ experiences were gained through the Gas Migration Test (GMT) at Grimsel Test Site (GTS), but with in-silo emplacement (Nagra, 2002).

At the contact zone between the different backfill materials (e.g. concrete - S/B) clogging effects may occur, which could affect the porewater flow and gas transport processes along the EBS. The influence of such a clogging layer on the water and gas transport properties across the material interfaces is investigated in detail through dedicated hydraulic and gas migration experiments. Final dismantling of the column will provide the necessary information to connect changes observed in material properties with observations from the column experiment.

The column to be used for this experiment is shown in Figure 2.

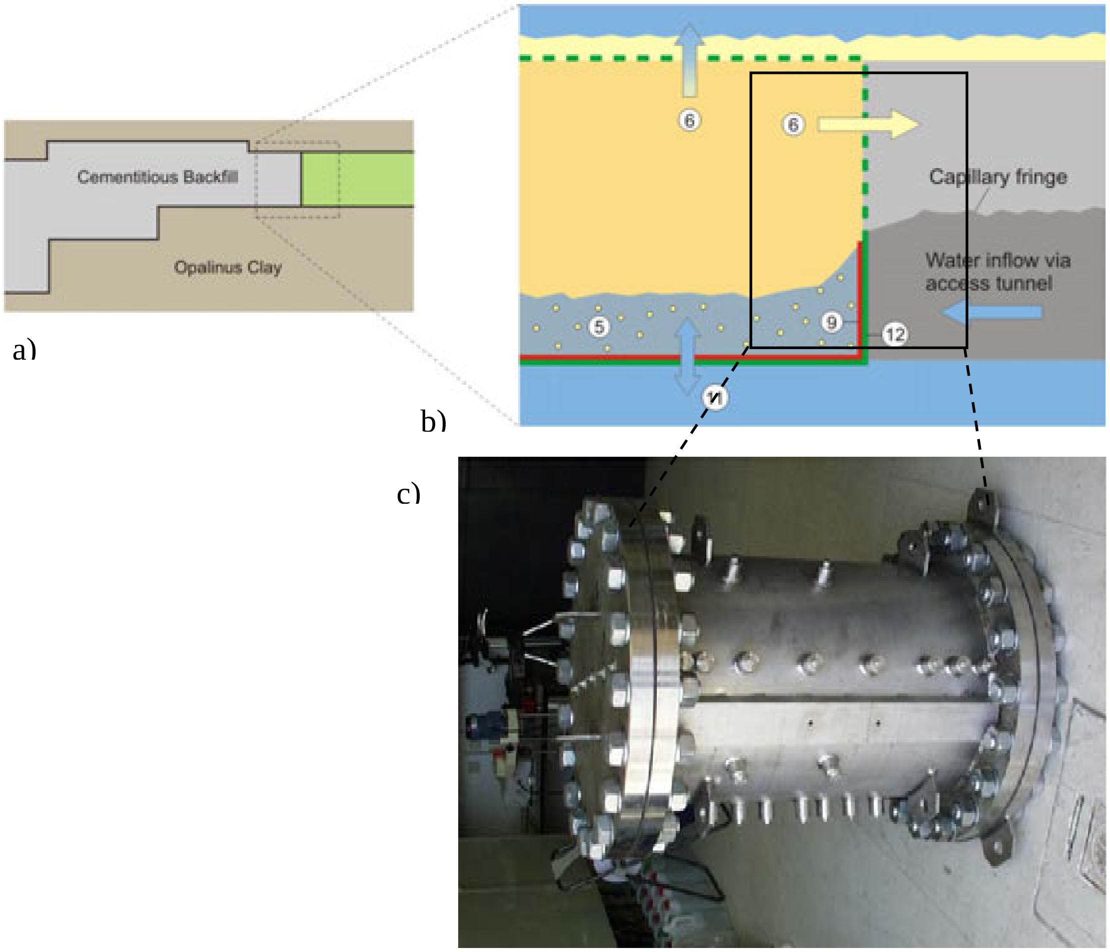

Figure 2: a) Schematic display of L/ILW repository, b) more detailed display of cavern plug (incl. expected water and gas flow paths), c) photograph of steel cylinder used to emplace the experiment.

The experiment aims at simulating the interface between porous mortar backfill and the S/B plug/seal (Fig. 2). Plugs and seals will be constructed with abutments on both sides to prevent horizontal displacement and subsequent deterioration of the water sealing properties.

Experiment Layout and construction

The instrumentation of the experiment was designed to provide continuous information on positive and negative pressures (water and total pressure), water saturation, and mass flows (gas and water) at the in- and outlets (Tab. 1).

Table 1: Specifications of selected sensors

Sensor Type	Code	Type	Range	Output signal	Accuracy
Water flowcontroller	WFC	Bronkhorst	0.5-10 g/h	4-20 mA	±0.1% Rd**
Back- Pressure Water and gas injection systems	PT	Keller-PAA-23SY	0 – 30 bar absolute	4-20 mA	0.5% FS*
Pressure Transmitters External	PE	Keller PAA-23SY	0 - 20 bar absolute	4-20 mA	0.5% FS*
Pressure Transmitters Tensiometer	TE	Keller PAA-23SY	0 – 1 bar absolute	4-20 mA	0.5% FS*
Total Presssure Sensor	TP	Keller PAA-25Y	0 - 50 bar absolute	4-20 mA	0.5% FS*
Differential Pressure Transmitter	DPT	Rosemount	0-1000 mm	4-20 mA	0.04% FS*
Ring-type Time Domain Reflectometry Sensors	RTDR	Solexperts	Humidity changes	Travel time	
3-rod Time Domain Reflectometry Sensors	TDR	Solexperts	Humidity changes	Travel time	

FS: Full scale** ****Rd: Reading

The geometry of the column is fixed to some extent but both in- and/or outflow could be filled with sand to reduce the total column length.

Materials used

The experiment setup consists mainly of two materials, namely the actual S/B seal with a target hydraulic permeability of 1E-11 m/s and the high porosity mortar with a target porosity of at least 30% (Fig. 2).

The sand used initially was sand from the bottom of Lake Geneva (Switzerland) because this is the reference material used by ETH Lausanne (LMS) in charge of the pre-tests. The major concern was that the mineralogy of this sand included Calcite, which could provoke chemical reactions while being in contact with some gases, whereas the main goal was to focus on the bentonite properties. Therefore, it has been decided to repeat these experiments using an inert sand with a different mineralogy, in order to avoid any chemical reaction.

The quartz sand, which was used for all following experiments, is grey quarry sand extracted in Germany, and which has already been sieved at 0.6 mm. Its specific weight is 2.65 g/cm^3.

The Wyoming bentonite reference MX-80 was selected because there is a considerable amount of research done with this bentonite. It is a highly plastic clay with a specific weight of 2.78 g/cm^3.

As the target property of the S/B mixture is its permeability, permeability tests had to be performed to assess the relation to emplacement density. Note that it is quite simple to assess the dry density of emplaced material in the field whereas permeability tests are only possible at the end of the saturation process.

As shown in Figure 3 the target hydraulic permeability of 1E-11 m/s can be reached with a dry density of about 1.8 g/cm^3.

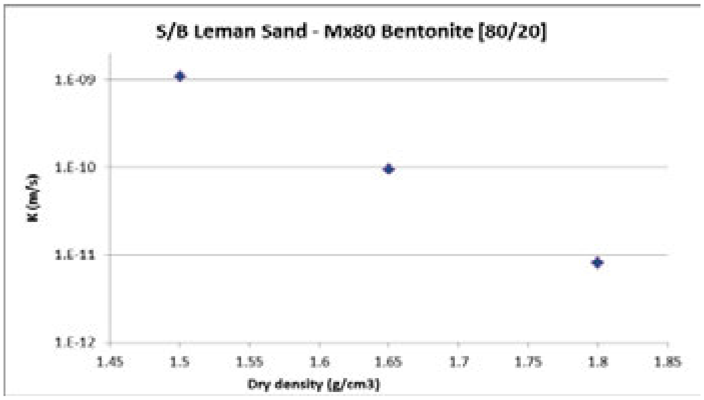

Figure 3: Evolution of saturated hydrualic permeability and dry density.

Four mortar types were tested to assess their stability and the compliance with the target of 30% porosity and high hydraulic permeability. In parallel, a support for the lifting of the cylindrical disks was developed and tested (Fig. 4). On 26.4.2010 both cylindrical disks were manufactured using the mortar "2-3 mm slightly compacted" with a hydraulic permeability of 1E-5 m/s, a water fillable porosity 29% and a bulk density of 1.66 g/cm^3. Until the transport of the cylindrical disks to the location of emplacement they were stored at 20 °C and > 90 % relative humidity, covered with plastic foil.

Figure 4: Photographs of mortar disks during emplacement

The finally emplaced experiment layout is shown in Figure 5 below. The figure shows the S/B body in the centre part with the pressure sensors and 3-rod TDRs emplaced, the ring of pure bentonite around the S/B seal to avoid preferential water and/or gas flow along the steel surface, and the two mortar discs at both sides. The right figure shows a schematic picture of steel column including the instrument distances found in the measurement labels. More details about the experiment setup and the materials used can be found in Nagra (2010).

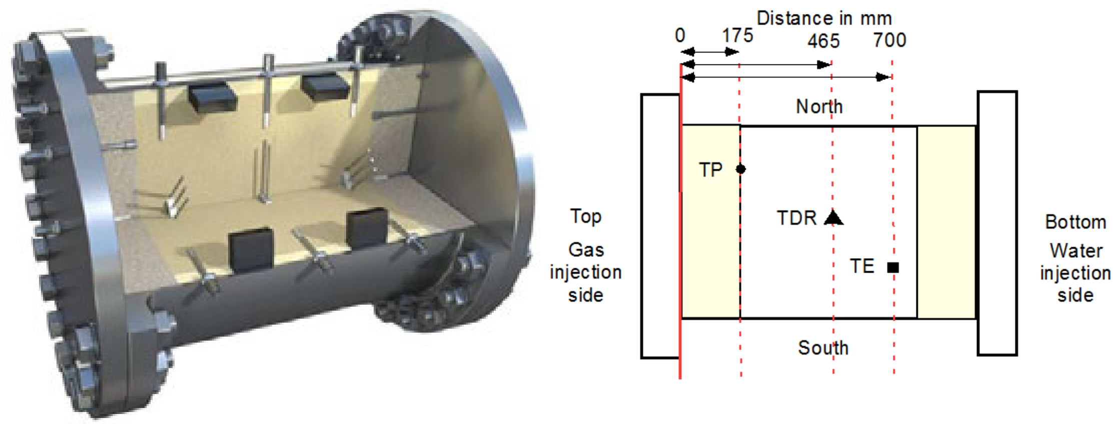

Figure 5: Schematic picture of experiment setup (left) and schematic picture of sensor positions in relation to water and gas injection side (right).

Experiment performance and results

Water injection into one of the mortar discs was started on 22.7.2010, still with the top valve open to avoid preferential flow along the column walls and potential breach of the bentonite sealing. On 13.10.2010 the backpressure was increased to 5 bar with a maximum flow set to 10 ml/h. As the saturation process was proceeding considerably slower than anticipated water injection from the other end of the column was started on 7.3.2012. Figure 6 shows the water pressures in the two mortar discs during the entire saturation phase. It can be seen that the injection pressure at the water side (PE-775-C) decreases steeply from time to time. This is because the flow controller located between the injection tank and the mortar disc clogged (zero inflow) frequently due to calcite precipitation in the steering valve.

The middle plot in Figure 6 shows the evolution of hydraulic pressures at the different sensor positions along the S/B seal. The four sensors 75 mm away from the water injection side were reached first (bluish lines in Figure 6) and all of them within a few days in October 2010. The sensors in the center of the S/B body (position 465 in greenish colours) showed saturation over a time starting from July 2011 until December 2011. The TDR measurements at the same position correspond well with these observations, showing an increase of saturation starting in July 2011 and reaching 100% in November 2011 (Fig. 6).

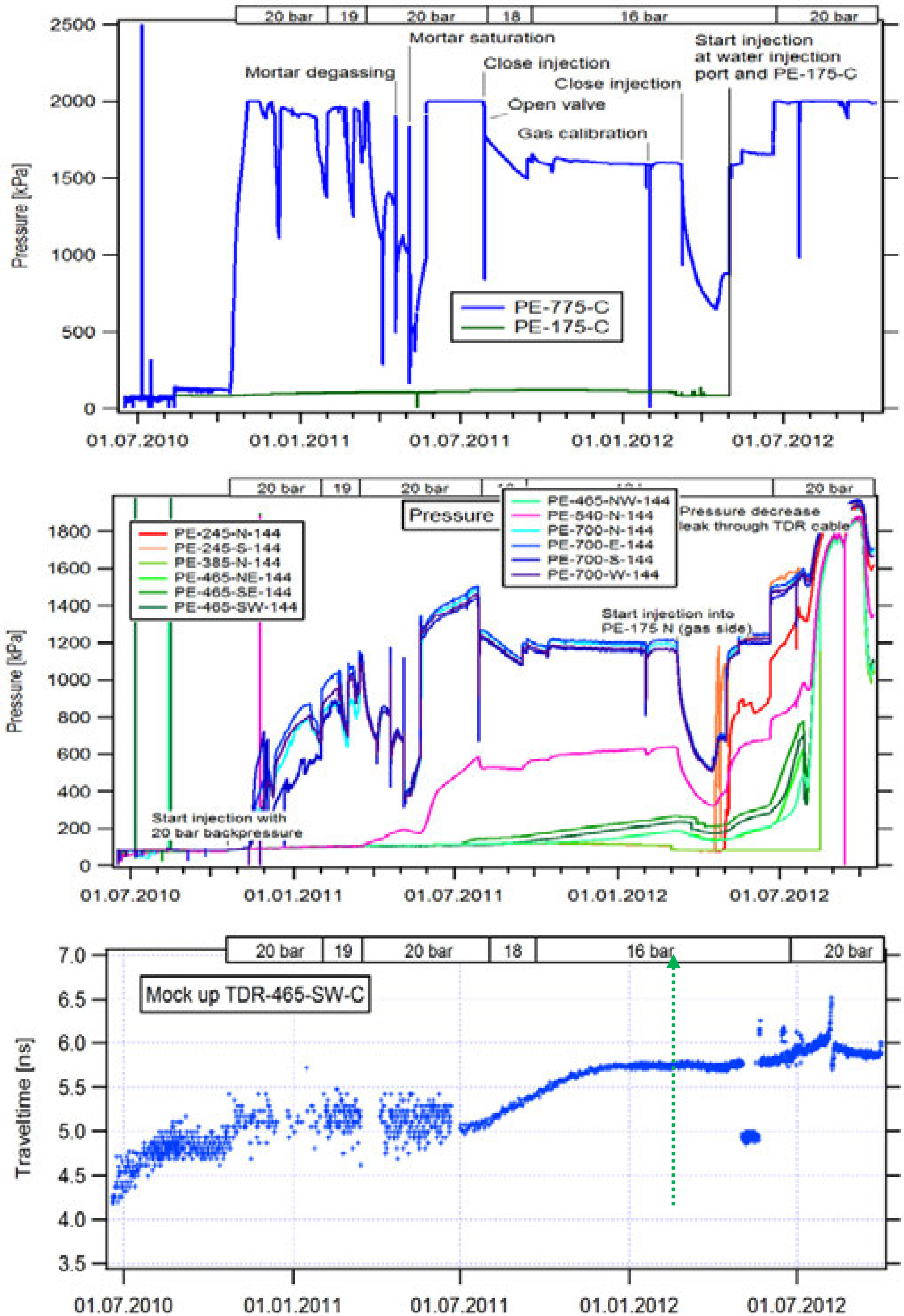

Figure 6: top plot: pressures at water and gas side; middle plot: pressures observed at the pressure sensors inside the S/B (positions according to Fig. 5); and bottom plot: TDR signal at the centre of the S/B seal.

To better understand the reasons for the longer saturation time the data were analysed in more detail. The main question was why the permeability of the S/B was considerably lower than expected based on the pre-tests in the laboratory. The analysis was done on two lines of evidence, namely an analysis of the gradients observed in the mock-up at different times, and the measurement of S/B permeabilities in the lab using the exact same materials as used for the mock-up experiment.

The pressure analysis was performed at four different times during the saturation phase during periods with minimal experimental disturbances. The hydraulic permeability was assessed using Darcy's law,

$$Q = \frac{-kA}{\mu} \cdot \frac{(p_x - p_i)}{\Delta x}$$

where Q is the injection rate, k is the intrinsic permeability, m is the viscosity (1.216 mPas at 13 °C), A is the cross section of the S/B body, p is the pressure at injection and observation point and x is the distance from injection.

The resulting permeabilities are between 2 and 2.8E-19 m^2, with an average over all sensors of 2.5E-19 m^2, thus about 5 times lower than designed.

The right plot in Figure 7 shows that there is no indication of a decrease in hydraulic permeability over the time scale of the expeirment performance despite the fact that water with a high pH of 12.6 to 12.7 was injected permanently into the S/B seal. So far, the only observable effect of applying realistic chemical boundary conditions was that the injection flow meter clogged regularly due to calcite precipitation inside the steering valve.

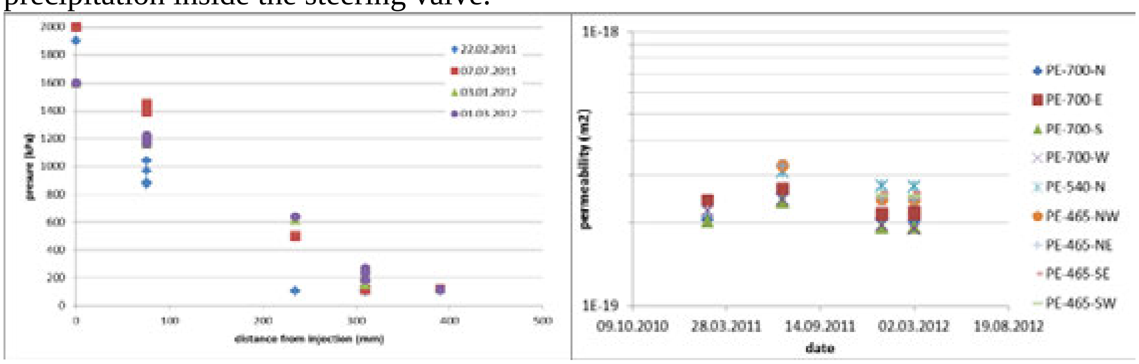

Figure 7: Hydraulic pressures measured at all pressure sensors at four dates during column saturation (left) and evolution of permeabilities over time.

The laboratory analyses of the hydraulic permeability at different dry densities of the identical material are shown in Figure 8. The figure shows clearly that the use of rounded sand (FORGE sand) as compared of the initially tested edged Leman sand has led to a considerable decrease of permeabilities. In fact the results coincide quite well with the observations of hydraulic gradients in the mock-up. The influence of the type of clay material on the permeabilities was found to have a minor impact (Fig. 8). The results provide further evidence that the observed lower hydraulic permeabilities are not a consequence of the high-pH water injected into the S/B but merely a consequence of the S/B properties as compacted.

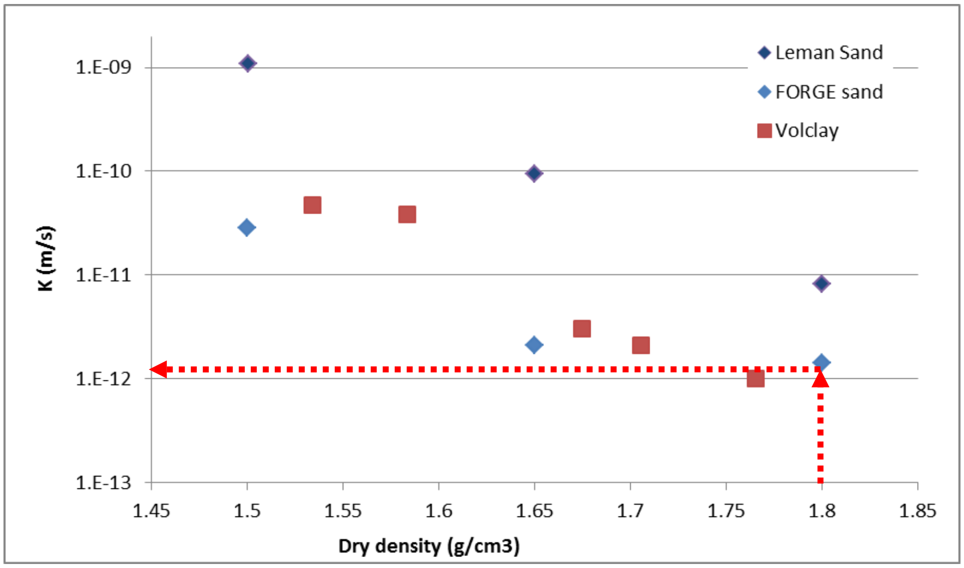

Figure 8: Relation of S/B (80/20) dry densities and resulting hydraulic permeabilities for two different sands and two different MX80 clay types.
Volclay is raw MX80 bentonite whereas the other two datasets were prepared with sieved granular bentonite.

Conclusions

The mock-up experiment constructed in the framework of the FORGE project was successfully setup to investigate and demonstrate the feasibility of gas-permeable seals at a larger scale. It was shown that constructing and instrumenting such an experiment has to be done carefully and that it is crucial to determine the density-permeability relation with exactly the same material as the one to be used for the emplacement. It was shown that particularly the use of different sands can lead to differences of as much as an order of magnitude in the resulting hydraulic permeability of the medium.

It was found that injection of high-pH water has no major impact on the hydraulic permeability of the S/B over the time scale of the experiment (i.e. about 2.5 years). Post mortem analyses will show whether or not there are changes of the pore space at the boundaries of the material interfaces.

The future analyses of the data will help to validate existing modelling codes and provide up-scaled model parameters for two-phase flow modeling that can be compared with those obtained in parallel on small scale laboratory experiments performed in the FORGE programme.

Acknowledgments

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GAS INDUCED RADIONUCLIDE TRANSPORT IN DISTURBED AND UNDISTURBED BOOM CLAY

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The Belgian agency for radioactive waste and enriched fissile materials ONDRAF/NIRAS presently considers Boom Clay as a potential host formation for the disposal of high-level and long-lived radioactive waste. In argillaceous formations like Boom Clay, radionuclide transport is dominated by solute diffusion. However, other phenomena like gas-induced transport may also occur, but are poorly described and quantified [2]. The present study focusses on the potential of gas-induced radionuclide transport after gas breakthrough in a clayey host rock (disturbed and undisturbed conditions) and at interfaces with engineered barriers. It was performed within the framework of the FP7 project FORGE (Fate of Repository Gasses).

In geological repositories for radioactive waste, gas can be generated by different mechanisms like anaerobic corrosion of metals, radiolysis of water and organic materials and microbial degradation of various organic wastes. The gas generated in the waste and engineered barriers will dissolve in the pore water and will 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 potential gas pressure build-up [1]. In case the gas pressure would exceed a local threshold value (e.g. tensile strength of concrete barriers or lithostatic stress in the Boom Clay host formation) this could lead to formation of discrete pathways. During a local gas breakthrough event in the clay, some water could be expelled by the gas phase. Depending on the timing of gas breakthrough, dissolved radionuclides and contaminants could be driven out of the clay faster than the normally expected diffusive transport.

To test the potential for gas-driven radionuclide transport, a column experiment was designed in which a water saturated clay core (h~4 cm, Ø= 3.8 cm) was put directly on top of a thin BC core (h~1cm) which had been previously saturated with a tracer solution. To mimic the presence of radionuclides in the pore water, a non-radioactive NaI solution of 0.01 mol/l was used as tracer solution. A He gas pressure (P~0.5-5 MPa) was applied at the bottom end of the iodide saturated plug while at the top end of the column, a known volume of natural pore water was put in contact with the plug. The gas pressure was stepwise increased until gas breakthrough occurred.

Upon gas breakthrough, the water on top of the column was expelled and analysed for its iodide content. The measured concentration of I was linked to the amount of NaI saturated pore water that was displaced, taking into account the natural background concentration of I and the transport of I due to diffusion (figure 2) [2]. Different types of experiments have been performed: experiments with undisturbed and disturbed (artificially fissured and let to self-seal for 1 night or 1 week) Boom Clay samples with different orientation wrt. bedding plane (parallel or perpendicular to the bedding plane). To investigate the role of interfaces, experiments on combined Boom Clay – bentonite samples were also conducted.

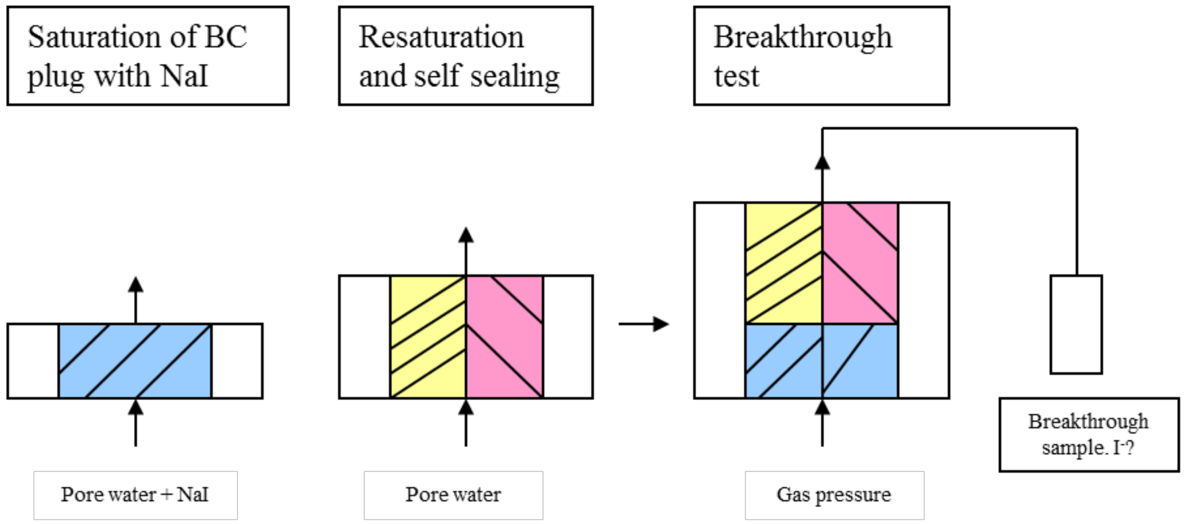

Figure 1: Basic concept of the tests to study gas driven tracer transport in the interface Boom Clay – Bentonite. Blue = NaI saturated Boom Clay core. Yellow = half Boom Clay core. Pink = half bentonite core.

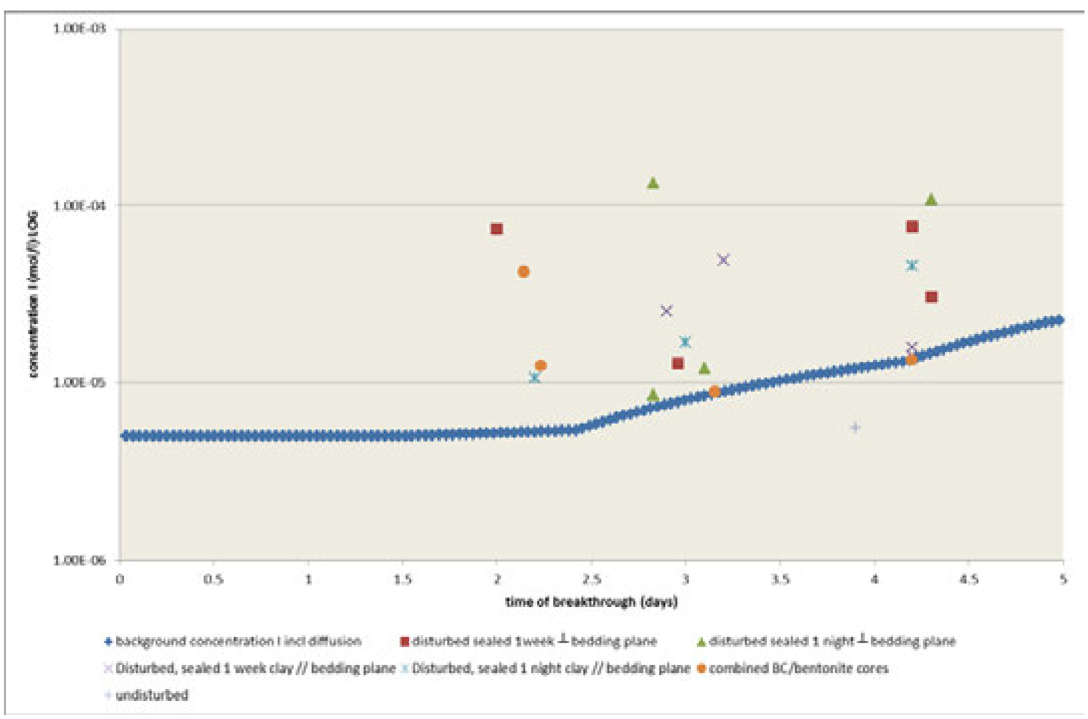

Figure 2: Iodide concentration in breakthrough samples (log scale) vs. time of breakthrough for all experiments. The dark blue line (diamonds) represent the expected background concentration of iodide, accounting for both the natural background concentratin in the pore water and the concentration increase due to iodide diffusion

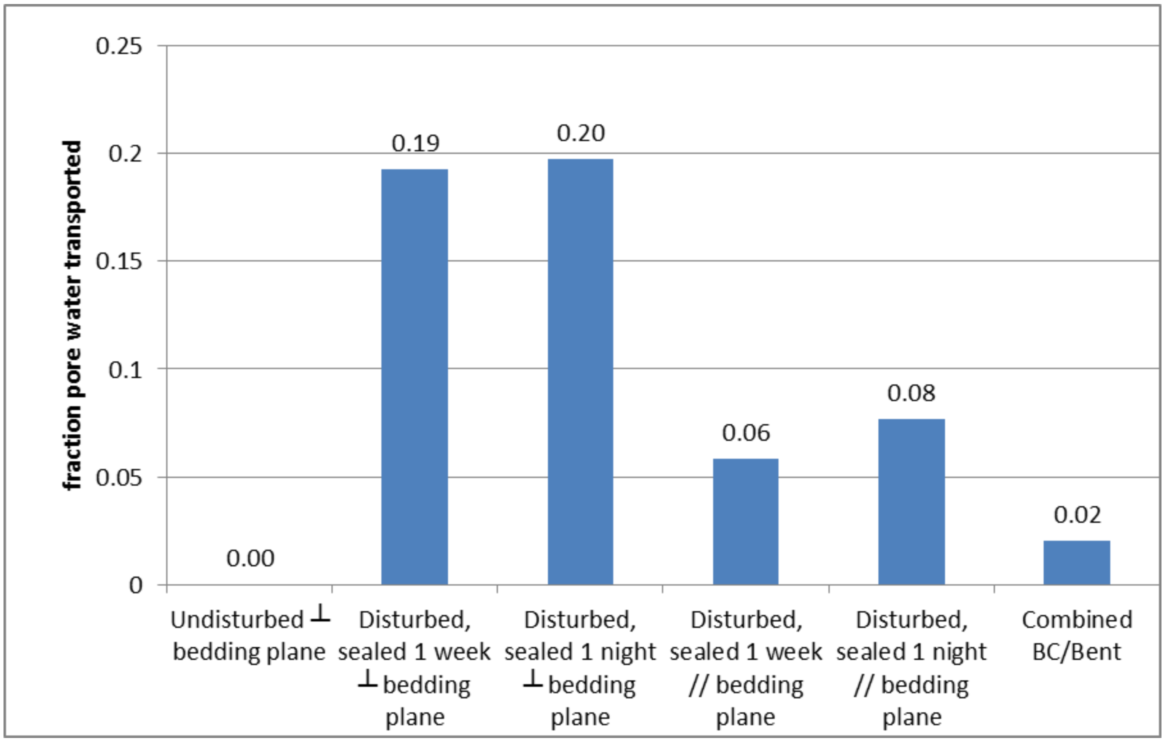

Figure 3: Average of the fraction of the volume pore water of the NaI saturated core transported during gas breakthrough for different sample types

Based on the obtained results, we can state that the transport of radionuclides and contaminants due to a gas breakthrough is indeed possible but remains very limited. Figure 3 shows that the amount of I that was transported and consequently the fraction of the volume pore water of the NaI saturated core transported during gas breakthrough was very low (< 0.5%). The effect of different parameters (orientation of the sample with respect to bedding plane, time of sealing of the fractures and porous medium type) on the fraction of the volume pore water of the NaI saturated core transported during gas breakthrough was investigated, but only orientation wrt. bedding plane played a significant role. This could be explained by the plate-structure of the clay: parallel oriented clay plates can more easily rejoin and sealing will be more efficient. Another significant parameter is the location of gas breakthrough: in 53% of the experiments, gas breakthrough pathways were observed at the interface clay - cell. In only 16% of the experiments gas breakthrough pathways were going through the clay core. So gas did find the weakest path which was in most cases the interface clay – cell. Transport through combined Boom Clay – bentonite samples is comparable to transport through disturbed Boom Clay.

Finally, it is important to note that during the experiments, observations of gas flow rather indicated another flow mechanism than classical visco-capillary 2-phase flow.

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The role of the Excavated Damaged Zone in HG-A Field-Scale Experiment Modelling

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Summary

HG-A field scale experiment has been developed in Mont-Terri Underground Research Laboratory to investigate the hydro-mechanical evolution of a backfilled and sealed tunnel section in Opalinus Clay. Based on this experiment, this study deals more particularly with the numerical modelling of the microtunnel excavation and ventilation. To take into account the anisotropic behaviour of Opalinus Clay, four sources of anisotropy are considered: in-situ stress, elastic modulus, failure criterion and water permeability. This modelling puts in evidence how the Excavated Damaged Zone, which developed around the microtunnel, plays a significant role in the hydro-mechanical behaviour observed in situ.

1. Introduction

The objective of the HG-A experiment is to investigate the hydro-mechanical evolution of a backfilled and sealed tunnel section [1]. In particular, the goals concern:

- understanding of the Excavated Damaged Zone (EDZ) generation and evolution in Opalinus Clay,
- upscaling of hydraulic conductivity determination from the lab test to the tunnel scale,
- investigation of self-sealing processes,
- estimation of gas leakage rates.

The geometry of the problem consists in a tunnel of 13m in length and 1.035m in diameter drilled in Opalinus Clay. More than 20 observation boreholes have been drilled parallel and oblique to the microtunnel axis and equipped with multipacker piezometer systems, inclinometer chains, chain deflectometers and stress cells to monitor the correspondent parameters in the host rock. After excavation, the micro-tunnel has also been instrumented with surface extensometers, strain gages, time domain reflectometers (TDRs), piezometers and geophones. The test plan consists in 6 stages:

- Phase 0: the drilling and instrumentation of the boreholes,
- Phase 1: the excavation of the microtunnel followed by backfilling and sealing,
- Phase 2: installation and inflation of the megapacker,
- Phase 3: hydraulic constant pressure and constant rate injection tests,
- Phase 4: gas injection tests,
- Phase 5: second hydraulic test series.

However, this particular study mainly concerns the numerical modelling of these two first stages to characterize the effects of anisotropic Opalinus Clay behaviour and the effects of damage on excavation and gallery ventilation processes.

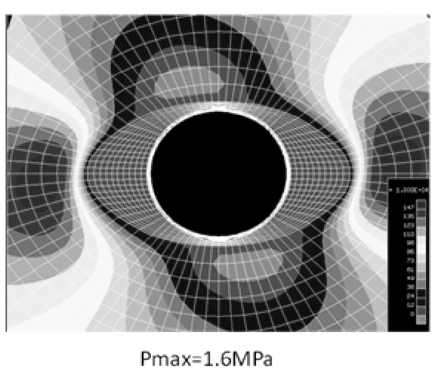
2. Methodology

The numerical modelling of HG-A experiment is performed in the finite element code Lagamine from Université de Liège. The hydro-mechanical behaviour of Opalinus Clay around the excavated gallery has been simulated through a 2D plane strain approach. To reproduce the dependency of the shear strength with the bedding orientation, an extended Drucker-Prager model with an anisotropic cohesion has been developed [2]. Then, four sources of anisotropy are considered to govern this behaviour: in-situ stress, elastic modulus, failure criterion and water permeability.

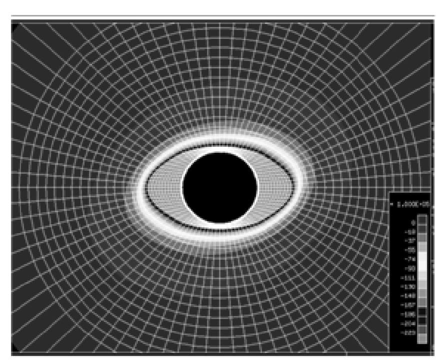
First, the ability of that model to reproduce the behaviour of Opalinus Clay has been proved by numerical simulations of triaxial tests performed with different orientations of loading with respect to bedding plane (see [2]). Second, numerical simulations of HG-A microtunnel are carried out. It consists in one week of excavation followed by one year of ventilation. Ventilation is putted in place with a relative humidity RH = 83% and a temperature T = 13°C in average. To reproduce gallery ventilation, pore water pressure is linearly decreased from 0 to a given suction in 7 days and then a constant pressure is maintained at -23.8 MPa.

3. Results

The modelling of HG-A microtunnel excavation and ventilation has shown that the hydro-mechanical response of the Opalinus Clay around excavation is fully governed by the four sources of anisotropy, keeping quite complex the global response of Opalinus Clay (see [2]). Nevertheless, good agreements with available in-situ measurements in term of displacement and water pressure evolutions can be obtained by also considering a damaged zone (EDZ) around HG-A microtunnel in numerical analysis. Based on plastic indicator PI field around the tunnel, an elliptic EDZ is expected (defined by PI > 0.5 or half-larger axis equals to 1m and half-smaller axis equals 0.6). This EDZ amplifies hydro-mechanical effects: tunnel convergence is higher and water pressure and suction fields increase after ventilation phase (Fig. 1). Model calibration on available in-situ measurements indicates that parameters have to be significantly modified in EDZ because of strong damage effects in Opalinus Clay: Young moduli have to be divided by 10, permeabilities have to be multiplied by 10^5 (Fig. 2). However, these modifications are not unrealistic for Opalinus Clay if we refer to what has been observed on SELFRAC experiment [3,4].

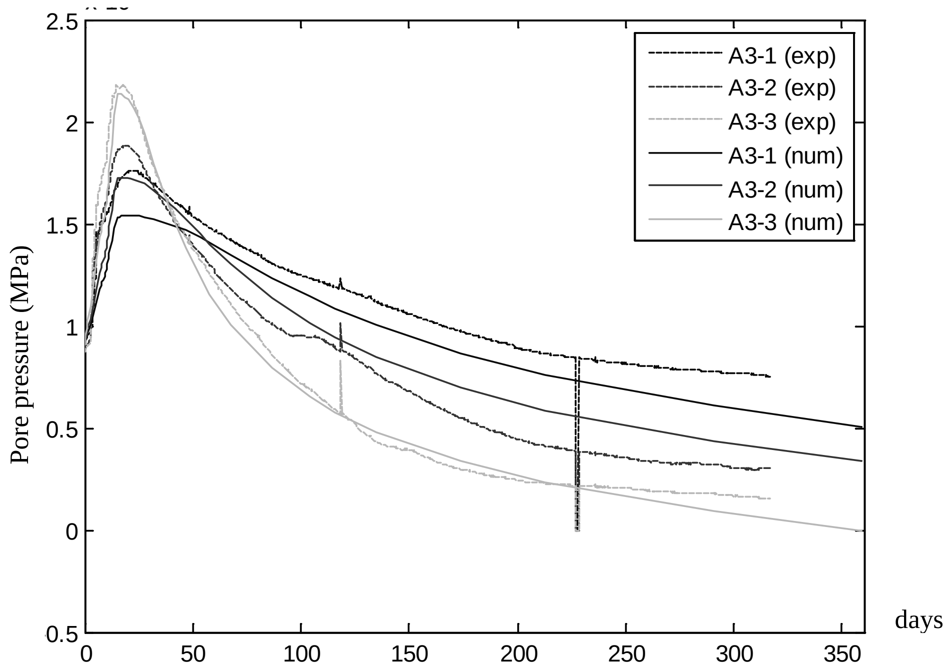


(a) water pressure

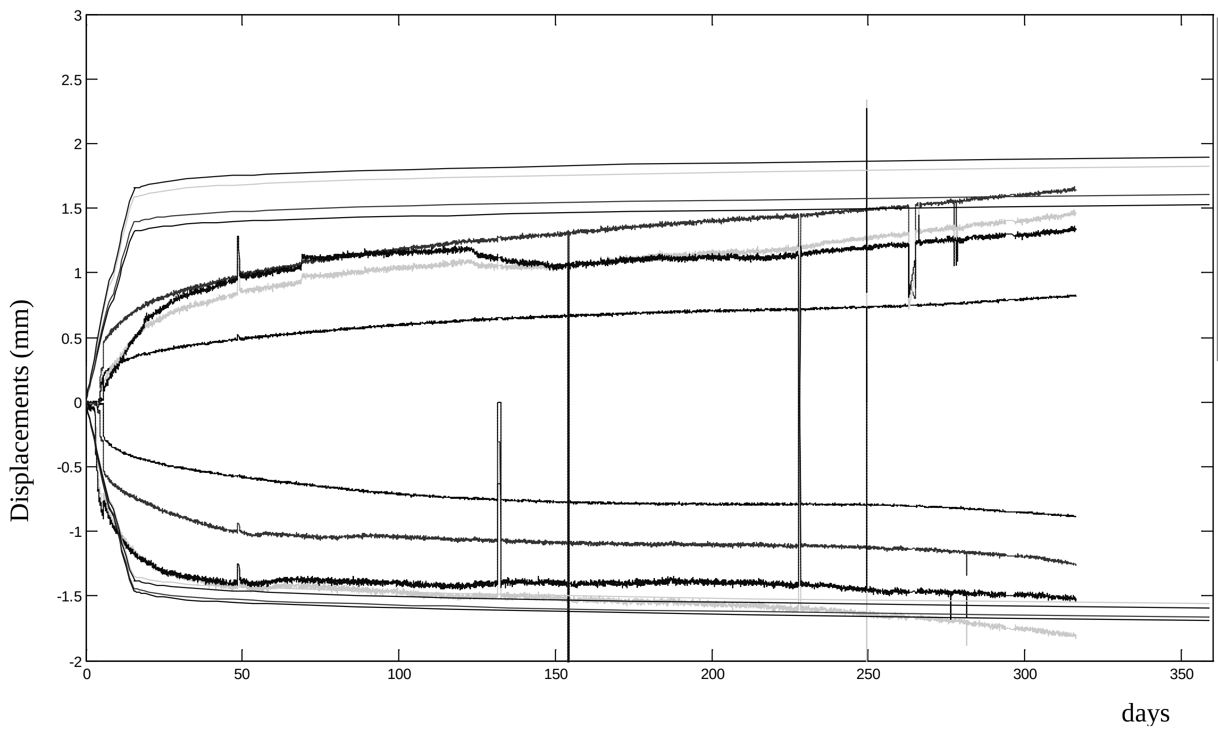


(b) suction

Figure 1: Water pressure and suction fields after ventilation phase



(a) water pressure evolution (A3 borehole)



(b) displacement evolution (A7 and A5 boreholes)

Figure 2: Water pressure and displacement evolutions with time

Furthermore, the presence of EDZ combined with clay anisotropies permits to justify the observed damaged (Fig. 3, [5]). Horizontally, damage (noted b) can be related to stress concentration providing plasticity and larger water pressures evolutions. At 45°, damage (noted a) results from hydraulic effects during ventilation when the gradient of saturation degree strongly decreases. Moreover, on the top of the microtunnel, the presence of a tectonic fault provides additional stress concentrations that amplify the damage, as we have shown numerically in [2].

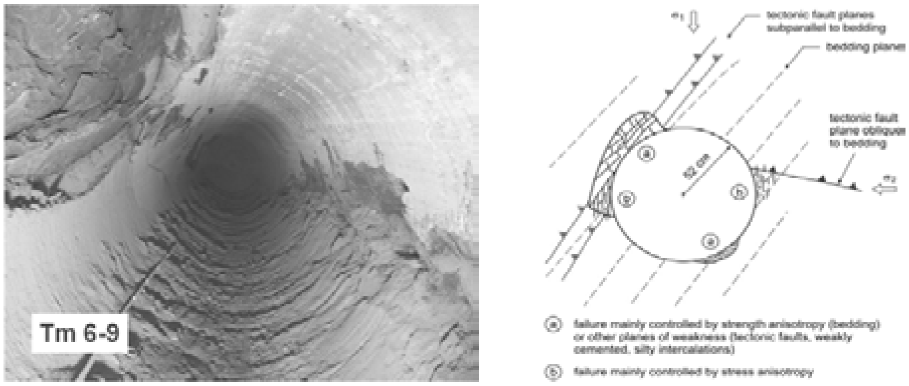


Figure 3: Damage observed in situ in HGA microtunnel (from [5])

4. Discussion

By the modelling of HG-A field-scale experiment, we have shown that hydro-mechanical response of the Opalinus Clay around excavation is strongly governed by 4 sources of anisotropy: in-situ stress, elastic modulus, failure criterion and water permeability. But these anisotropies are not sufficient to explain in situ observations. Displacements and water pressure fields are affected by a damaged zone that developed around HG-A borehole. An EDZ yields to larger overpressures at the interface between EDZ and undamaged zone and a realistic delay in water pressure evolution after excavation. Then, the proposed 2D modelling is very relevant on Opalinus Clay hydro-mechanical behaviour charaterization. However, it would be necessary in the future to improve the modelling calibration process with available in-situ measurements by establishing a more accurate relationship between EDZ parameter evolutions and plastic or damaged state of Opalinus Clay.

5. Acknowledgements

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Gas Generation and Migration through Salt Formations

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Summary

Salt formations ensure safe isolation of disposed waste due to their impermeability for gases and fluids. However, significant gas quantities may be generated in the long-term (e.g. due to anaerobic corrosion, if humidity is present) resulting in a time dependent pressure build-up. To assess the effect of increasing gas pressures on the integrity of rock salt, we present results from long-term gas injection tests in the laboratory and in sealed boreholes (up to 50 m^3 pressurized volume). Gas breakthrough occurs at pressures slightly above the minimum principal stress; an increase of permeability of three to four orders of magnitude is observed, independently from the investigation scale. Radial gas flow through a 2 m thick salt layer surrounding the large scale bore hole was identified as enhanced permeation zone by the performed AE-monitoring. Fortunately, with the subsequent pressure decay self-sealing is corroborated restoring the initial gastight behaviour. Our findings clearly disprove a single gas frac scenario.

1. Introduction

Tightness is a fundamental prerequisite for warranting the long-term safety of a radioactive waste repository in a salt formation, i.e. rock salt is attributed to be impermeable for gases and fluids, which means that the rock mass permeability is in the order of 10^{-20} - 10^{-22} m^2 or lower. The excavation of underground openings will perturb the rock sufficiently to cause fractures in the rock adjacent to the openings, i.e. the stresses will be in the dilatant domain, resulting in damage. However, because dilated salt is a visco-plastic material it will heal, and this will restore the original watertight and gastight performance [1].

As a consequence of tightness a time dependent pressure build up will occur due to various gas production processes (e.g. anaerobic corrosion and microbial degradation) and, in addition, due to excavation convergence, until the pressure inside the cavity equalizes the external stress in the salt bed. If the gas pressure exceeds the primary formation stress some scenario postulate that localized crack opening due to tensile stresses may take place in the overlying salt barrier, i.e. described as the so-called gas-frac scenario. However, to overcome the already existing uncertainties regarding such a scenario in salt in the last decade comprehensive lab and field investigations were performed by IfG delivering a valuable base for assessment of property changes in rock salt during gas-pressure build-up.

2. Methodology

The investigations consist of:

- extensive lab investigations with injection tests under well controlled lithological and stress conditions focusing on the gas break through process and its reversibility. In addition, a permeability-pressure relationship describing the effect of pressurisation of rock salt was obtained;
- three long-term in situ tests in bore holes (pressurized volume ca. 60 – 100 l), in the salt mines Bernburg and Merkers (D), respectively, whose results allow assessing of gas pressure effects in the undisturbed rock salt, in particularly during the gas breaktrough;
- a large scale injection test (50 m^3 gas volume) which was performed at the salt mine Merkers facilitates verification of the small-volume bore hole test results;
- a natural analogon of "gas frac" in salt exists in the salt mine Merkers (D) during a rock burst which facilitates the verification of long term salt healing with recovery of the barrier integrity after the gas-breakthrough.

Some of the results are already published in [1, 2].

3. Results

For determining gas permeability in the lab under well-defined loading conditions and at high gas pressures a conventional triaxial cell is used with a cylindrical salt sample equipped with a central sack hole, i.e. corresponding to the field borehole geometry. Measuring the radial gas outflow around the central injection borehole in the sample for a given pressure respectively measuring the equilibrium pressure for constant injection rates (i.e. stationary tests) facilitates the calculation of the gas permeability, using the well-known Darcy law for gases.

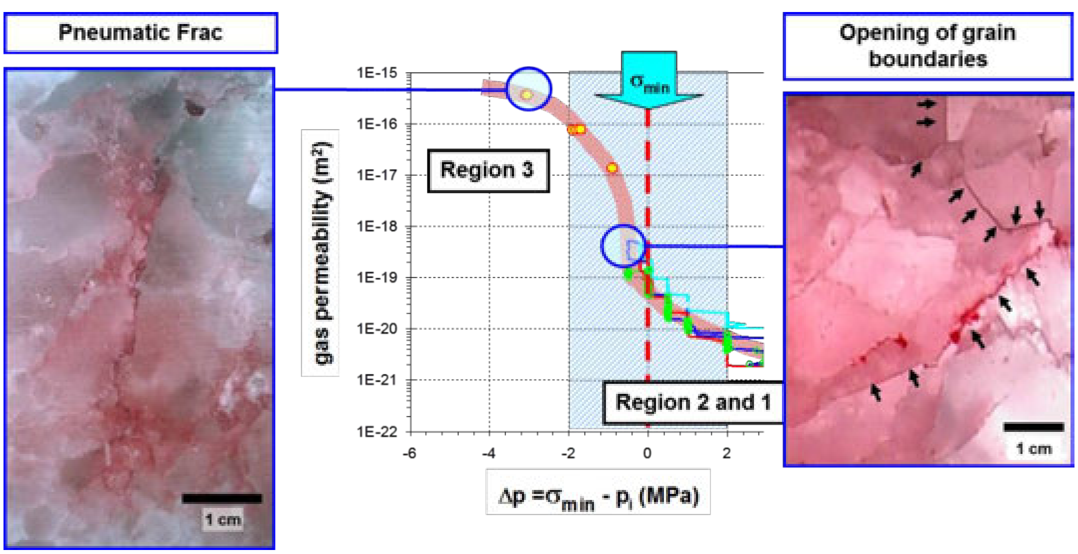

Figure 1. Lab test results of gas injection tests on cylindrical salt samples (∅ = 100 mm, l = 200 mm, central injection hole with radial gas flow) with microstructural observations after gas breakthrough. (Centre) Permeability vs. differential pressure. (Right) Dilated grain boundaries (indicated by arrows) in the pressure region of the gas breakthrough – Region 2 (p_i is in the order of σ_{min}). (Left) Singular fracture after gas injection at pressures with $\Delta p \leq -2$ – Region 3.

Generally, three pressure regions have to be discriminated:

- Region 1 - at low gas pressures ($p_i \ll \sigma_{min}$) the initially measured permeability of undisturbed rock salt is extremely low, i.e. $k < 10^{-20}$ m^2, but increases slightly during stepwise pressurization.
- Region 2 - when p_i approaches σ_{min} the gas breakthrough occurred resulting in a steep increase of permeability (up to 4 orders, whereby the lower the initial permeability the higher the rise) – only opening of grain boundaries forming a network of locally already existing pathways.
- Region 3 - at pressures $p_i \gg \sigma_{min}$ a plateau of permeability at $k > 10^{-16}$ m^2 is reached. But it has to be mentioned that in this region the database is only weak. Due to the drastically increased permeability, it was not easy to measure during high pressure shut-in tests the gas flow accurately – creation of transgranular fracturesdue to tensile stresses.

It is important to note that the permeability increase during progressive gas injection depends not on the absolute value of σ_{min} but on the difference between σ_{min} und $p_{injekt} \Rightarrow \Delta p$. In addition, in the regions 1 and 2 the Δp – permeability-relation is reversible.

Because in lab tests the pressurized gas volume is limited to some few tens of cubic centimetres additionally field tests with injection bore holes were performed increasing the scale of investigation by a factor of 1000 to some tens of cubic decimeters, respectively by a factor 10^6 with a large-scale field test in the salt mine Merkers with a pressurized volume of 50 m^3. Each test site consists of a complex borehole array with a central injection hole surrounded by various observation boreholes and monitored by a highly sensitive AE-network (operated by GMuG).

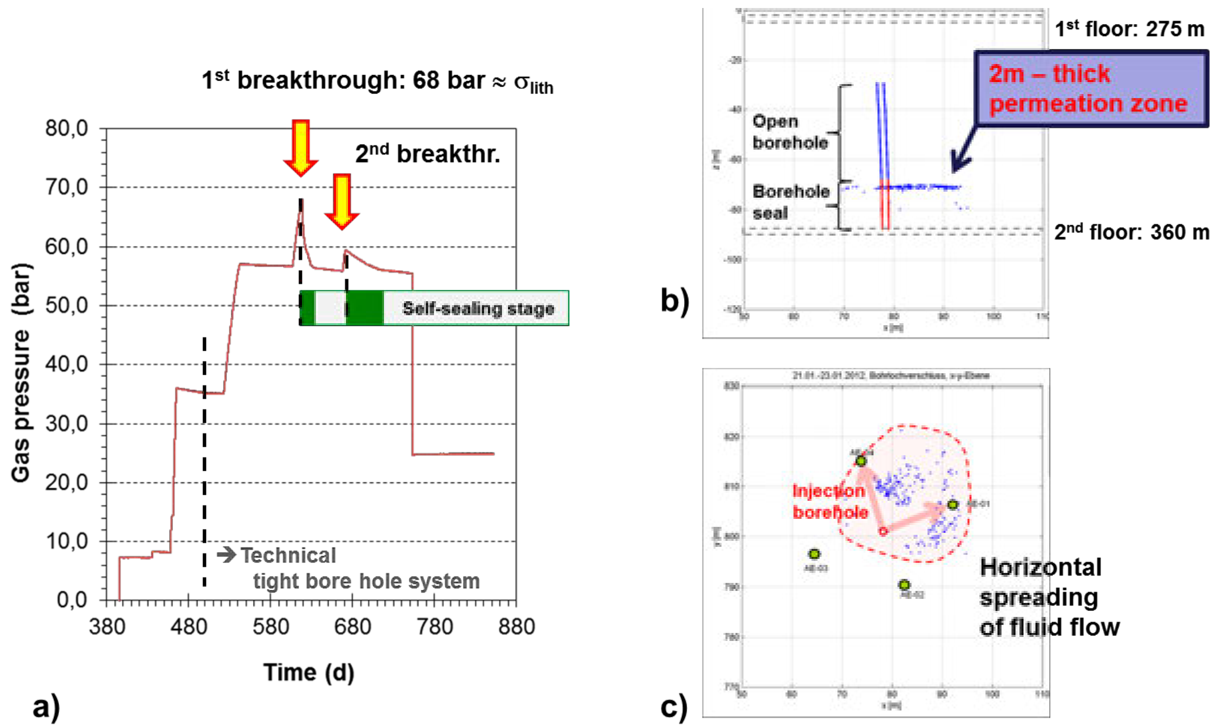

Figure 2. The large scale gas injection test in the salt mine Merkers (pressurized gas volume: 50 m^3). a) Multi-stage gas-pressure injection test, whereby at least two gas break-through events were observed as indicated by arrows. The colored section indicate phases where self sealing occurred. b) vertical section of the investigation site with the localized AE-events during the 1st break through period. c) horizontal section. Note the more or less radial fluid spreading through a 2 m thick salt layer.

The large-scale test site Merkers benefits from the unique mining situation in the bedded salt mass of the Werra salt formation (z1) where both potash seams were mined in a room-and-pillar system at 300 m (1st floor – Level 1) and 380 m depth (2nd floor – Level 2) respectively. From the second floor a nearly vertical 60 m high borehole was drilled with a diameter of 1.3 m. After equipping the borehole with pressure, stress and strain probes a gas tight seal plug of 20 m height is installed consisting of a special MgO concrete. In addition, two m^3 $MgCl_2$-brine were pumped in as capillary blocking medium. The gas injection borehole is surrounded by a highly sensitive AE-network consisting of twelve AE sensors installed in four observation boreholes drilled parallel to the main borehole at a distance of approximately 15 m to the center line of the borehole.
The pressurization took place in several steps, i.e. to about 8 bar (Step 1), to about 35 bar (Step 2) and to about 56 bar (Step 3). During the 4th step of pressure increase a widespread AE cluster was observed three days before the maximal pressure of about 68 bars was reached. At this maximal pressure a gas and brine breakthrough occurred which prevented a further increase of pressure followed by a pressure decrease to ca. 56 bars (comparable to the 3rd step). The AE events are located in a cluster 5 to 15 meters (forming a more or less radial fluid zone) eastward the large borehole and in another cluster close by 5 to 15 meters northeast the large borehole, but slightly below the sealing plug. Thus, firstly an outflow of brine followed by gas was recognised in two instrumentation boreholes. Video camera inspection demonstrated that a layer of 2 m thickness became permeable but the fluid outflow stopped when the gas pressure reached back 56 bars. Additional pressure tests confirmed that the initially rock tightness was restored after the pressure drop to 56 bar. Lowering the gas pressure to 25 bar documents no gas leak, i.e. rock tightness.

4. Conclusions

In extension of the already existing knowledge, our results from laboratory and in-situ tests give a valuable base for assessment of property changes in rock salt during gas-pressure build-up. The general characteristics of pressure driven gas transport in salt rocks has been convincingly enlightened, whereby in lab and field tests qualitatively the same phenomena are observed. The out-standing observation is that (independently from the investigation scale) if the gas pressure approaches σ_{min} the further pressure increase is diminished for a wide range of gas injection rates, due to the coeval permeability increase of 3-4 orders of magnitude (“secondary” permeability) at the gas breakthrough. However, the permeability increase is reversible, i.e. if the pressure decreases “self sealing” occurs spontaneously. Thus this process may act as a “safety valve” which clearly prevents the pneumatic gas-frac scenario at realistic pressurization rates.

5. Acknowledgements

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Numerical Interpretation of Gas Injection Tests at Different Scales

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Summary

To investigate gas migration processes in saturated Opalinus Clay, gas injection tests under different injection pressures have been carried out at the different scales: on the core samples at the laboratory scale, in the packed-off section of boreholes at the borehole scale (HG-B), and in the sealed micro-tunnel at the tunnel scale (HG-A) - an 1:2 scale experiment conducted in the Mont Terri Rock Laboratory. A fully coupled hydro-mechanical model has been developed taking account of elastic and plastic anisotropies, anisotropic two-phase flow based on the van Genuchten function, and strain dependent permeability to evaluate the experimental data. Special attention has been paid on dealing with a dilatancy-controlled gas flow regime in case of high gas injection pressure over confining or minimal principal stress. Based on the laboratory data two different flow regimes, two-phase-flow under low gas injection pressure and dilatancy-controlled gas flow under high gas injection pressure over minimal principal stress, have been understood properly. The developed model with calibrated parameter value from laboratory tests can be used very well to predict the in-situ borehole tests under good defined hydromechanical conditions. The gas flow regime in a large scale experiment, as the case of HG-A, was mainly predominated by experimental circumstances. Long-term and large scale (up-scaling) effects as well as local heterogeneity should be considered in the numerical model.

1. Introduction

The gas transport mechanisms depend strongly on the gas pressure and the hydro-mechanical state of the rock. In the saturated Opalinus Clay, advection and diffusion of dissolved gas is the dominant process at very low gas generation rates. A separate gas phase may form with increasing of gas generation rate and a visco-capillary two-phase flow may occur if the gas pressure is higher than the gas entry pressure. With continuous increase of gas pressure, gas may flow along micro-cracks network generated by high gas pressure because of relative low tensile strength of clay rock.

To investigate gas migration processes gas injection tests under different gas injection pressures have been conducted on the core samples in the laboratory [1], in the boreholes within the HG-B experiment [2] and in the sealed micro-tunnel within the HG-A experiment [3]. The core samples used for the laboratory tests were taken from the Mont Terri Rock Laboratory. Similar gas injection tests were done in situ in a sealed micro-tunnel within the HG-A experiment and in boreholes in the course of the HG-B experiment. The latter two experiments were performed in the Mont Terri Rock Laboratory in Switzerland.

2. Methods

The focus of this research work is the numerical analysis of the experimental data and the in-situ observations. A H²M coupled numerical model has been developed and implemented in FEM Code OGS. This model combines a hydraulic model (two-phase-flow model) and an elasto perfect plastic model (Mohr-Coulomb criterion with tension cut-off) in a monolithic manner. The effective stress is defined by using the saturation as weighting function for gas and water pressure. The relationship between capillary pressure and water saturation is described by the van Genuchten function based on laboratory data and the relative permeabilities to gas and water by the approach of Mualem.

Two permeability models have been used to describe the change of permeability during gas injection tests. The first approach is based on the assumption that permeability depends directly on the local gas pressure. A critical pressure value is introduced as a criterion distinguishing high and low gas pressures, which is determined to be the minimal principal stress acted on the system. The second approach assumes that permeability development is controlled by the deformation calculated by effective stress. The model uses the volumetric strain (ε_{vol}) and the equivalent plastic strain ($\bar{\varepsilon}^{p}$) and can be expressed by

$$\mathbf{k} = f\left(\Delta\varepsilon_{vol}\right) e^{b_1 \Delta \bar{\varepsilon}^{p}} \mathbf{k}_{\text{int}}^{ini} \tag{1}$$

in which

$$f\left(\Delta\varepsilon_{vol}\right) = \begin{cases} 10^{b_2 \Delta\varepsilon_{vol}} & , \quad \textit{compaction} \\ 10^{b_3 \Delta\varepsilon_{vol}} & , \quad \textit{extension} \end{cases} \tag{2}$$

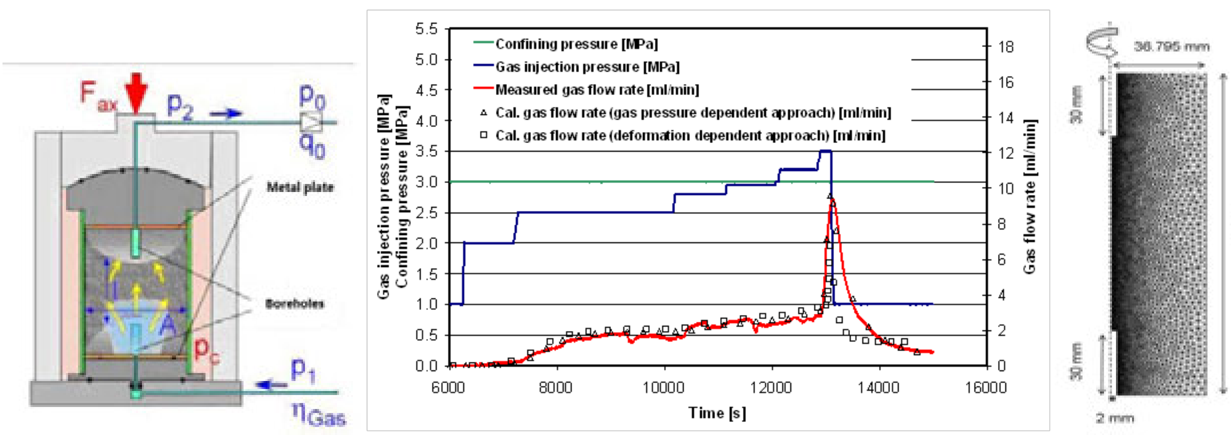

Figure 1: Laboratory experiment: test layout, results and meshed used

3. Experimental and numerical results

Laboratory test: Two mini-boreholes with a diameter of 4 mm were drilled in specimen, one borehole at the top and the other in the bottom of the specimen. Nitrogen gas was injected in the bottom hole for

5 hours and flows to the upper hole. The test was performed with a constant confining pressure and a stepwise increase of gas injection pressure. At the very low injection pressure, flow rate measured is so low that only dissolution of gas in water happened. Already under an injection pressure over 2.5 MPa, a flow rate of about 2 ml/min at STP was measured, which indicates a separate gas phase flow in the system. When the gas injection pressure reached to 3.5 MPa, an over-pressure of 0.5 MPa about the confining pressure, the gas outflow rate increased significantly. At this stage, a through-pathway for gas flow from local dilatant areas was created. Using the developed material models the measured pressure development can be very well simulated (Fig. 1).

HG-B test: For this test, four boreholes were drilled perpendicular to the bedding plane within the HG-B experiment. The central one is used for injection purpose and three surrounding holes for the seismic monitoring. Nitrogen gas was injected with a stepwise increased gas injection pressure under in-situ hydraulic and mechanical conditions. Gas flows parallel to the bedding plane which has relative high permeability due to hydraulic anisotropy. If the gas injection pressure is higher than 1.2 MPa, which is assumed to be the minimal principal stress, steep pressure decrease was observed both in the experimental data and in the calculated results (Figure 2). The analysis shows that the possible flow pathway induced by a high gas injection pressure is mainly influenced by the anisotropic material properties, the anisotropic initial stress distribution, and the in-situ test configuration. This result shows that in-situ measurements can also be interpreted in a realistic manner with this new material model.

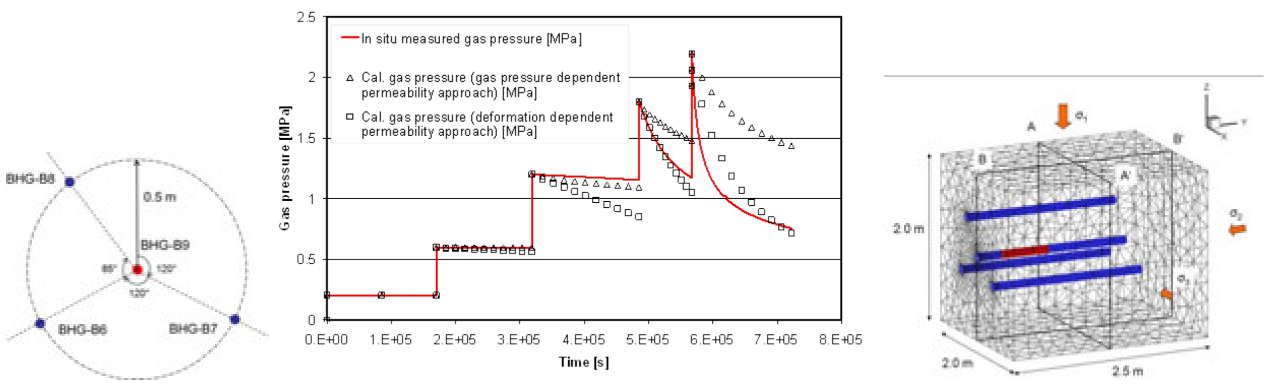

Figure 2: HG-B in-situ borehole experiment: test layout, results and mesh used

HG-A test: For the simulation of gas injection tests within the HG-A experiment, however, a multiple-step modelling concept was developed. This concept focused on the understanding of temporary changes of material properties, especially the generation of an excavation induced damage zone (EDZ) around the micro-tunnel. In the first step a coupled HM model was carried out to simulate the excavation induced processes. Location of observed breakouts can be interpreted by using an anisotropic elasto-plastic model taking into consideration shear failure and volumetric dilation. In the second step, local high permeability up to 10^{-15} m² in the EDZ with a thickness of about 20 cm was evaluated by simulating the water injection tests. Permeability variations have been done till a quite inhomogeneous model was built to simulate the gas injection tests. The long-term water injection test before gas injection tests leads to a reduction of permeability of one order of magnitude due to the sealing effect of Opalinus Clay. All three stages of gas injection tests for the duration of about 1.5 years have been simulated (Fig. 3). Satisfactory results have been obtained both in the injection area and sensors, which were installed around the mega-packer. The pressure decreases after two peaks may be explained by an opening of flow pathways somewhere in the system.

It can be concluded that if gas pressure exceeds the confining stress or the minimal principal stress, gas flow rate increases significantly in all cases.

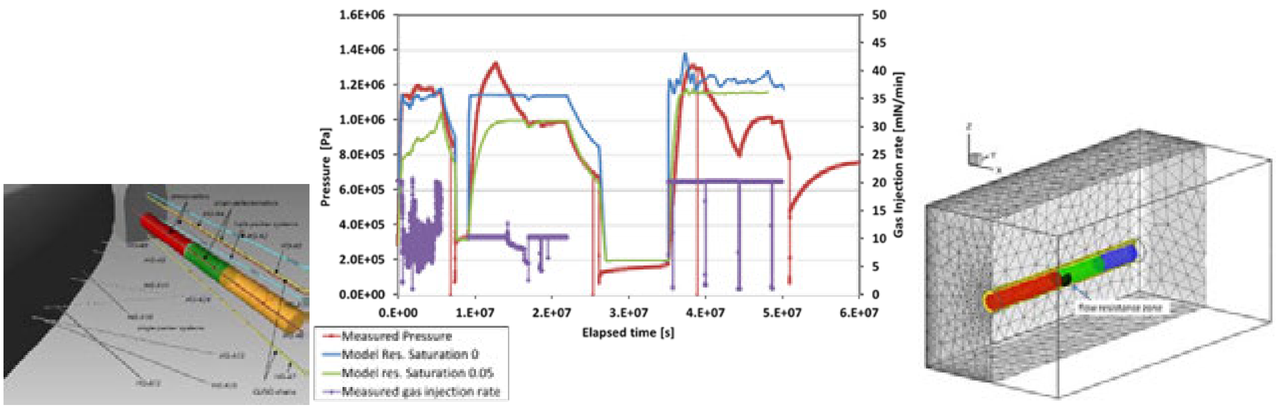

Figure 3: HG-A micro-tunnel experiment: test layout, results and meshed used

Table 1. Relationship between gas injection pressure and minimal stress.

Scale	Laboratory	In-situ borehole	In-situ micro-tunnel
Confining /minimal Stress [MPa]	3	1.2	1.2
Gas pressure [bar]	>30 ~ 32	>12 ~ 14	>12 ~ 14

4. Conclusions

Two different flow regimes, two-phase-flow under low gas injection pressure and dilatancy-controlled gas flow under high gas injection pressure over minimal principal stress have been investigated both experimentally and numerically. Three gas injection tests were performed at the different scales. At the laboratory scale, the determined processes can be well understood using a numerical method because of good defined experimental conditions. The applicability and predictability of the developed model with calibrated parameter from the laboratory tests is also proven to the in-situ tests. Local permeability may be increased up to 3 OOM due to dilatancy. Partial irreversible plastic strain may exist after gas injection. The gas flow regime in a large scale experiment, as the case of HG-A, was mainly predominated by experimental circumstances. Up-scaling effects (both long-term and large-scale) should be considered in the model.

5. Acknowledgements

This work was supported by BMWi (Bundesministerium für Wirtschaft und Technologie, Berlin).

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The Experimental In-Situ Study Of Gas Migration In Crystalline Rock With A Focus On The EDZ

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Summary

The Czech Technical University in Prague (CTU), Centre of Experimental Geotechnics (CEG) is currently participating in a FORGE project, part of WP4, which is focused on disturbed host rock formations. The Czech deep radioactive repository concept envisages crystalline rock as the host rock environment, therefore the CEG is concentrating on such formations. Large scale gas injection measurements are being carried out as part of the project the aim of which is to study the behaviour of both the excavation damaged/disturbed zone (EDZ) and the undisturbed rock environment.

Introduction

One of a number of deep RAW repository construction concepts considers crystalline structures as the host rock environment. The CEG's role in the FORGE-WP4 project is to study the behaviour of disturbed crystalline rock formations in terms of gas transport in connection with which in-situ gas injection measurements are being performed at the Josef Underground Laboratory, principally in the granodiorite section of the underground complex. The main objective is to study the various roles played by the EDZ as a provider of preferential routes for gas transport.

Methodology

The methodology of the experiments is based on the performance of large-scale in-situ gas injection measurements. The single and double packer systems are being used for gas injection purposes; compressed air is employed as the gaseous medium. A mobile measuring device was designed especially for this purpose. The measuring apparatus features gas pressure level control and the automatic registration of pressure, temperature and gas volume.

Several horizontal boreholes were used for pilot testing in the Josef Underground laboratory in various geological conditions. Following the evaluation of these tests research focused on a vertical borehole (MW-SP-67-3V) situated in the Mokrsko-west granodiorite section of the Josef Underground laboratory. The borehole was gradually deepened to 20, 30 and finally 40 metres. A series of air injection tests was performed using the single packer system each time the borehole was deepened. Two types of tests were used. The testing methodology of the first type of test (the Constant Head Injection Test - CHIT or Constant Flow Injection test - CFIT) is based on pressurizing the borehole to a prescribed pressure/flow level and then waiting until a steady state flow is attained. The second type of test, the pressure drop test (PDT), commences immediately following the CHIT and is based on ceasing injection once the air flow has been stabilized and subsequently observing the pressure drop curve. The tests were necessarily of a long-term nature since it takes a considerable period of time to stabilize injected air flow conditions and the team wished to study long-term behaviour including repeatability. Long-term testing in the granodiorite rock environment of the Mokrsko area lasted from several days to a number of weeks. Several water pressure tests were performed for the purpose of comparison with air pressure testing.

The flow rate of the injected air was measured by a Coriolis flow meter which formed part of the measuring apparatus. In the case of the extremely low permeability of undisturbed rock environments, where flow rate values were below the measurement limits of the flow meter, the flow rate was calculated from pressure drops/losses based on the known volume of the borehole injection section.

Results

More than 20 long-term test series (each test consisting of several subtests at distinct injection pressure levels) have been performed in the MW-SP-67-3V borehole. The levels of permeability of individual sections of the borehole were measured by placing the packer in different positions within the borehole (usually in 2.5m steps).

Multiple pressure levels and flow rates were used. The flow rate value was set within a range of between 0 and 40 Nl/min (normal litres per minute). In the undisturbed part of the rock environment pressure levels of between 10 and 50 bar were used (CHIT tests). Calculated flow rates ranged from 0.01 to 0.40 Nl/min. In the more permeable rock of the upper part of the borehole, a constant flow rate was used (approx 35 Nl/min) and borehole pressure was measured (CFIT testing). The maximum achieved pressure at a steady state in these sections was found to be lower than 25 bar which corresponds to the local primary stress level.

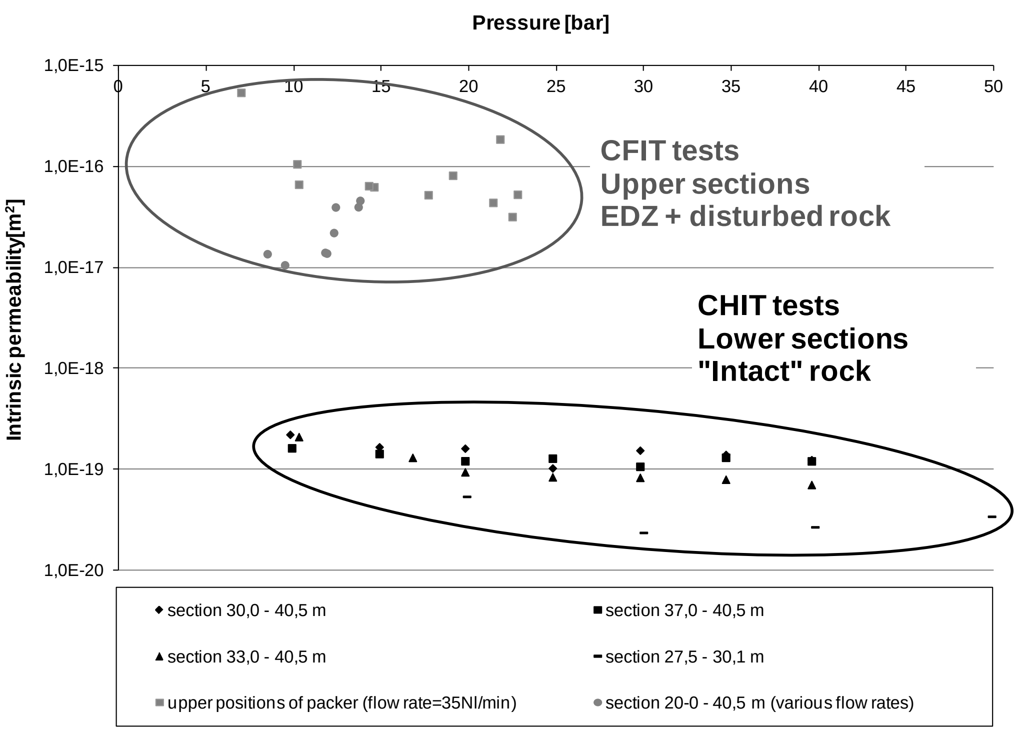

Figure 11: Dependence of intrinsic permeability on injection pressure

Intrinsic permeability and gas conductivity were determined using data obtained from the injection tests. A simplified analytical solution of flow was used for the calculation of permeability. Figure 1 shows the dependence of calculated intrinsic permeability on injection pressure for different borehole sections.

The permeability of each borehole section was back-calculated using the two adjacent measurements (positions of the packer); for this type of calculation average values from all the tests performed in the same section and at all pressure levels (flow rates) were used. The results are set out in Figure 2 which shows the dependence of permeability on distance from the start of the borehole (the gallery).
The results of the tests performed at the packer's upper positions (long injection zones) revealed that final permeability is in the order of between 10E-16 and 10E-17m^2 and that deeper in the borehole, in the undisturbed rock environment, values are around 10E-19m^2.
In some parts of the borehole, mainly between pressures of 15 and 25bar, the lower (shorter) injection section displayed a higher flow rate (or lower pressure) than the upper (longer) part. This was probably the result of injected air escaping back to the borehole through interconnected longitudinal joints; in such sections it was not possible to back-calculate permeability.

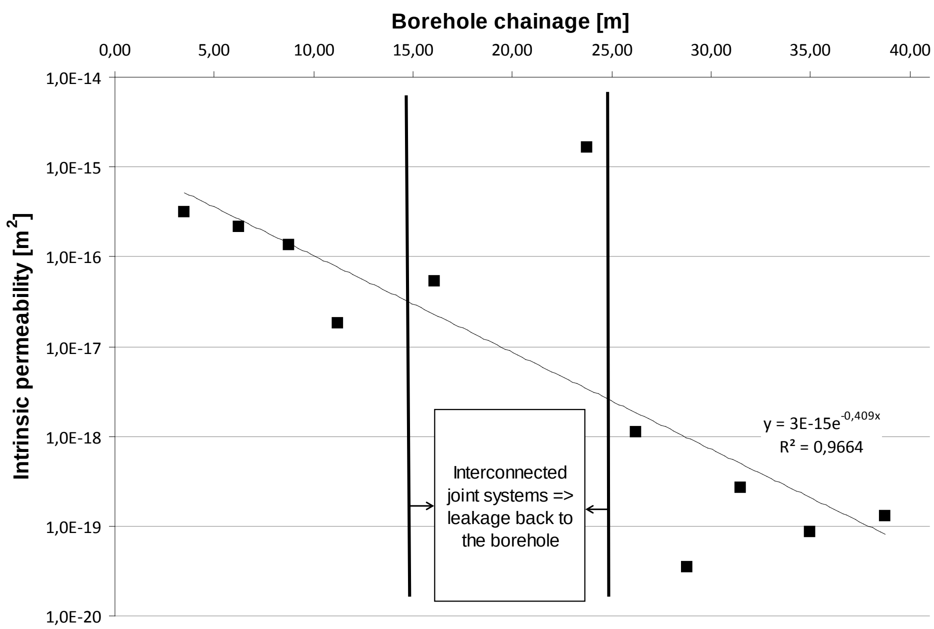

Figure 12: Dependence of intrinsic permeability on distance from the gallery

Discussion

The gallery was excavated using the drill and blast method and therefore featured a considerable EDZ. Gas permeability should therefore decrease with distance from the gallery. This decreasing trend has been confirmed to a surprisingly high degree. There is strong evidence of a large difference in this respect between the EDZ and the undisturbed rock; permeability differs by 3 orders of magnitude and decreases by one order of magnitude every ten metres.
The expectation that higher injection pressure levels will lead to the opening of preferential pathways for the escape of gas, which will in turn lead to an increase in gas permeability, was only slightly observed in certain tests with the packer in the upper position (section 20 – 40m) during CFIT testing. The opening of fractures had already been observed during water pressure testing [1]. However the trend for gas permeability to increase was not confirmed in the lower section of the borehole even with high injection pressures (up to 50 bar) exceeding the value of the primary stress level which was around 25 bar. The trend concerning gas testing in this case was even slightly the reverse.

Conclusions

A decrease in gas permeability with distance from the gallery has been confirmed. There was strong evidence of a difference between the disturbed (EDZ) and undisturbed rock formations.
A wide EDZ was present in the vicinity of the drifts which affected the gas permeability of the rock mass even up to distances of 20 metres.
No opening of the fractures (preferential pathways) was observed when the injection pressure exceeded the primary stress level in the undisturbed rock; however a slight increase in permeability was observed in the EDZ during CFIT testing.
According to the results, the EDZ plays a major role in terms of gas transport due to its significantly higher permeability; therefore gases could more easily open fractures within the EDZ due to lower stress levels and damage caused by excavation.
Conversely, sections further away survived excavation by blasting surprisingly well which suggests that it can be expected that the rock mass will seal the repository despite local damage.
However a combination of these two effects might cause problems for the repository in that the EDZ working as a “collector” within the undisturbed (“impermeable”) host rock could lead to a concentration of gases in certain locations (the equivalent of a stratigraphic trap). Effective sealing around the access shafts/tunnels is crucial therefore so as to prevent the EDZ working as a conduit to the surface.
Note: There is no guarantee that the use of bentonite-based backfill would help in this respect since the fractures used for gas transport are too small for bentonite to effectively penetrate and seal.

Acknowledgements

The research leading to the above results received funding from the European Atomic Energy Community's Seventh Framework Programme FP7/2007-2011 under grant agreement number 2300357.

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Gas Flow and Chemical Reactions in Unsaturated Bentonite Buffer.

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Summary
The generation and migration of gases within a potential nuclear waste repository has been the subject of considerable attention and interest for a number of years. The work presented here attempts to extend current knowledge to accommodate, in a systematic way, some "chemistry" related aspects, so that the impact of gas migration on the geochemical condition of a potential nuclear waste repository might be further explored. A reactive gas transport model has been developed within an established coupled thermo/hydro/chemical/mechanical (THCM) framework. The model comprises a module for multicomponent chemicals transport both in liquid and gas phases (COMPASS), together with an advanced geochemical reaction model (PHREEQC2). A series of numerical simulations have been carried out in which hydrogen gas has been injected into unsaturated bentonite buffer. The fate of hydrogen and pressure development due to the injection process have been investigated, when various geochemical interactions of gas, water and bentonite are included.

1. Introduction

The generation and migration of gases within a potential nuclear waste repository has been the subject of considerable attention and interest for a number of years. Research has been commissioned into a number of aspects, covering both the generation potential of the materials to be used in the repository and the migration of the generated gases through the surrounding engineered barriers and geosphere. Such attention has obviously been fully warranted in relation to the overall performance assessment requirements of a potential nuclear waste disposal facility. The design of potential underground repositories is based on the development of a multi-barrier system [1]. In such a system the metallic waste canisters are surrounded by compacted highly swelling clay buffer which is contained within the host rock. Various chemical reactions may take place between the generated gas and the clay/water system affecting the fate of the gas in the near field. Additionally these interactions may alter the behaviour of the clay buffer.

To the authors' knowledge, the previous body of research has largely focused on what might be described as the "physics" of the problem i.e. the impact of gas generation and migration, within performance assessment approaches, on the physical condition of a potential nuclear waste disposal repository. In fact the long term behaviours, considering gas transport and geochemical/gas-chemical interactions in the buffer have received less attention [2]. The work presented here attempts to extend current knowledge to accommodate, in a systematic way some "chemistry" related aspects, so that the impact of gas migration on the geochemical condition of a potential nuclear waste repository might be further explored. To address the aforementioned issues, a reactive gas transport model has been developed within an established coupled THCM framework. The newly developed model allows the reactive transport of gas to be modelled under variable physical, chemical and mechanical conditions. An application of the developed model to simulate hydrogen gas transport and fate through an unsaturated bentonite buffer and the influence of gas-chemical processes in predicting the long term buffer behaviour is presented.

The methodology adopted is based on the approaches proposed and developed by the senior author over many years [3,4]. Extensive studies of coupled transport processes in soils have been performed, examining so-called thermo/ hydro/chemical/mechanical (THCM) behaviour. Attention has been focused on a solution of the problem at various scales; ranging from soil element behaviour, to engineered barrier response, to the simulation of the whole system. A great deal of effort has been concentrated on the response of unsaturated soils to such conditions, with recent attention focused increasingly on physico/chemical response of highly swelling unsaturated clays [5].

The governing equations of multicomponent gas transport consider the major mechanisms of advective and diffusive flow. The developed in-house model comprises a transport module for multicomponent chemicals both in the liquid and gas phases (COMPASS), together with an advanced geochemical reaction model (PHREEQC2) [5,6,7]. Various geochemical features of gas-chemical interactions, such as ion exchange, redox processes, mineral dissolution/precipitation, have been coupled with the transport model. Numerical solutions of the flow and deformation equations have been achieved by employing the finite element method for spatial discretisation and the finite difference method for temporal discretisation. A sequential non-iterative approach has been adopted to couple the transport processes and geochemical interactions. The formulation has been developed for both isothermal and non-isothermal conditions.

2. Simulations

A series of numerical simulations have been carried out in which hydrogen gas has been injected into unsaturated bentonite buffer. It has been assumed that non-uniform re-saturation of the buffer will take place which permits the moisture to reach the canister-buffer interface at various discrete locations. As a result, corrosion and hydrogen generation has been initiated due to local anoxic conditions. The generated gas can then migrate through regions that remain unsaturated. Simulations have been performed under isothermal conditions. The gas influx (hydrogen) rate has been calculated considering a realistic corrosion rate for metallic canisters. According to various disposal concepts, a canister thickness for higher activity radioactive waste disposal of around 5 to 20cm has been suggested [8]. Gas generation for a 20 cm thick canister will last for about 10,000 years [8]. Therefore this duration will be considered in the simulations. Accordingly a constant hydrogen influx of 2.0×10^{-11} kg/m^2/sec has been considered in the simulations which fit within the range of realistic gas generation rates proposed [7]. At the injection face of the domain boundary, a constant influx of hydrogen has been considered while the opposite side has been kept impermeable to water and gas flow.

Modelling scenarios, using MX-80 bentonite, consider the effects of the geochemistry and the pore-water composition of the system. A buffer thickness or model domain of 0.35m has been considered following SKB's proposed KBS-3 concept [9]. The model domain has been discretised into 80 elements of 4-noded quadrilaterals. The initial pore water composition of compacted bentonite at various water contents have been calculated using geochemical modelling. Details of the material parameters and boundary condition are presented elsewhere [7].

3. Results and discussion

The results of hydrogen concentration development at the gas injection face of the buffer achieved from the simulation results are presented in Figure 1. Results have been plotted for three different water contents of 10%, 15% and 18% corresponding to degrees of saturation of approximately 40%, 60% and 70% respectively.

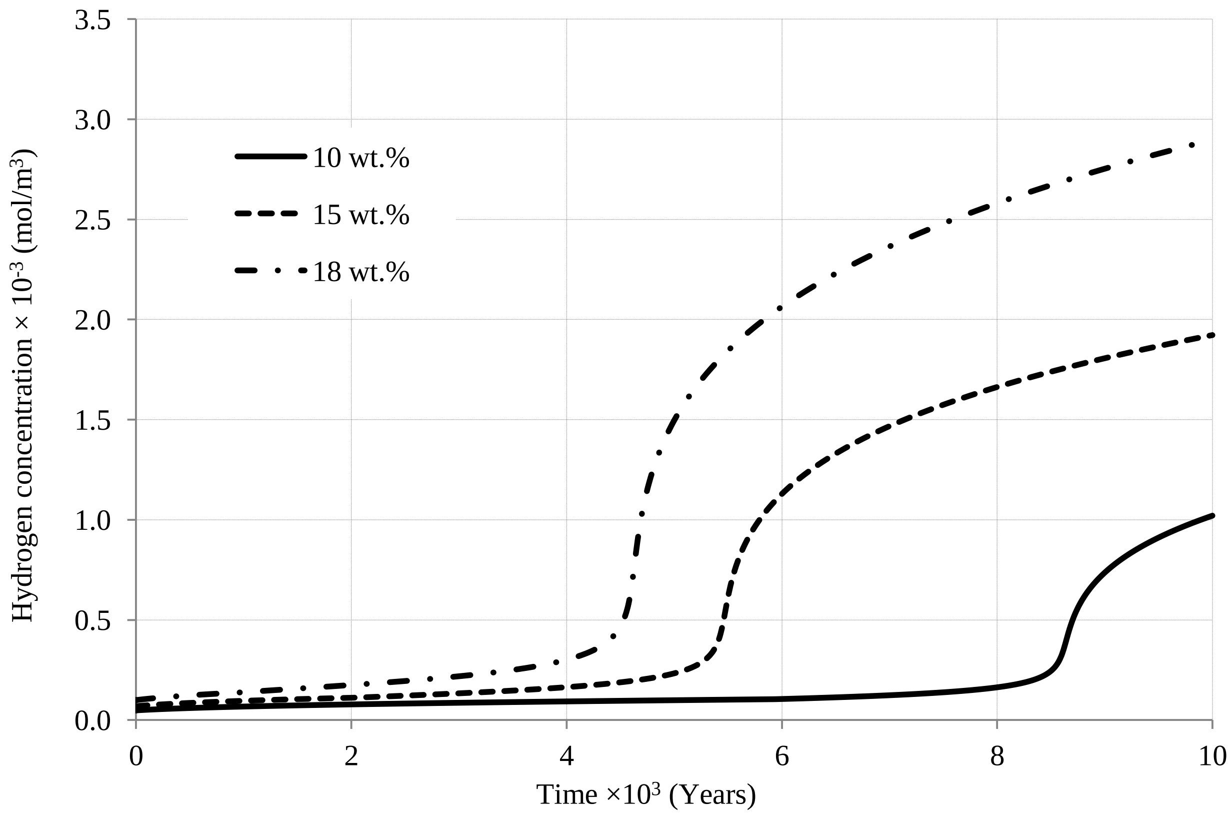

Figure 1: Evolution of hydrogen at various water contents at the injection boundary for a duration of 10,000 years.

The results of the numerical simulations also indicate that the gas pressure development is less in simulations where the pore water contains various chemical components and minerals, such as calcite, gypsum than simulations where pore water is considered as distilled water. This suggests that gas in the system will be buffered through gas-geochemical processes.

Dissolved gas in the system has also been found to be affected by the high buffering capacity of the bentonite-water system, resulting in a relatively stable pH environment. It has been observed that during simulation exercises, the pH of the clay-water system did not change significantly. Accelerated rates of dissolution of gypsum and precipitation of calcite have been recorded during the simulations. Based on the results obtained, it is postulated that the presence of various chemical components in the clay buffer may influence the transport and fate of the hydrogen gas. It is suggested that this investigation of gas flow/geochemical interactions have provided some new interesting insights.

4. Acknowledgement

Financial support from Arup, in the form of a PhD Fellowships to the third author, is gratefully acknowledged.

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Gas and Water Permeability of Concrete

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Summary

Gas permeability measurements were performed in concrete samples of different degrees of saturation and hydraulic history under low gas pressure. A steady-state setup for the measurement of gas permeability of concrete under different pressure conditions was also fine tuned. In addition, the saturated hydraulic conductivity of the samples was measured after saturating them with water. Intrinsic and relative gas permeabilities were deduced from these measurements.

1. Introduction

In most countries the final disposal of low and intermediate-level radioactive waste is performed on surface or near-surface facilities, in which concrete is frequently used as a barrier. The durability of concrete and its mechanical properties are intrinsically bound to moisture transport effects, especially when it is subjected to repeated wetting and drying regimes. This work is a contribution to the understanding of the behaviour of concrete barriers in surface disposal facilities and was carried out by CIEMAT in FORGE WP3.4.3 “Concrete laboratory experiments”.

2. Materials and methods

The concrete used was manufactured at the Spanish disposal facility El Cabril in the form of cylindrical blocks casted in PVC molds of different sizes, following the procedures used to manufacture the disposal cells. The OPC concrete had a characteristic strength of 350 kp/cm^2, with a water/cement ratio of 0.43 and a consistency of 14 cm. The percentage of pores of diameter smaller of 2 nm (micropores) was 39±7 and of those larger than 50 nm (macropores) was 49±5. The pore mode was around 92±13 nm, and the porosity about 10 percent.

The samples were cut in the laboratory to fit the cell dimensions (diameters of 35 or 50 mm and heights between 40 and 70 mm). These samples were allowed to air dry and their gas permeability was measured in several moments along the drying process, hence the gas permeability was obtained for different concrete water contents. The same samples were also water saturated to determine in them the hydraulic conductivity and then allowed to air dry again, measuring the gas permeability at different moments during this second drying process.

A steady-state setup for the measurement of gas permeability under different pressure conditions was fine tuned. The gas pressure of the samples was also measured in an unsteady-state equipment working under low injection pressures. The detailed description of both equipments is given in [1].

3. Results

It must be pointed out that no sample was completely dry (0% water content) during the determinations and therefore, the intrinsic permeability could not be directly obtained from the measurements performed, since to determine the intrinsic permeability with air flow (k_{ig}) the sample must be completely dry. In order to obtain completely dry samples it would have been necessary to dry them in the oven at 110°C and this would have caused changes in the microstructure of the concrete and consequently in its hydraulic properties. When there are two fluids present in the porous material (gas and water in this case), the permeabilities of each fluid depend upon the saturation of each fluid: these are called effective permeabilities. Hence, the value obtained in the determinations (apart from the gas permeability, k_g) is the intrinsic permeability measured with gas flow, k_{ig}, multiplied by the relative permeability to gas, k_{rg}.

Permeability decreased with water content, but it was also greatly affected by the hydraulic history of concrete, *i.e.* if it had been previously dried or wetted (Fig. 3.1). In particular, and for a given degree of saturation, the gas permeability of concrete previously saturated was lower than that of concrete that had been just air dried or saturated after air drying.

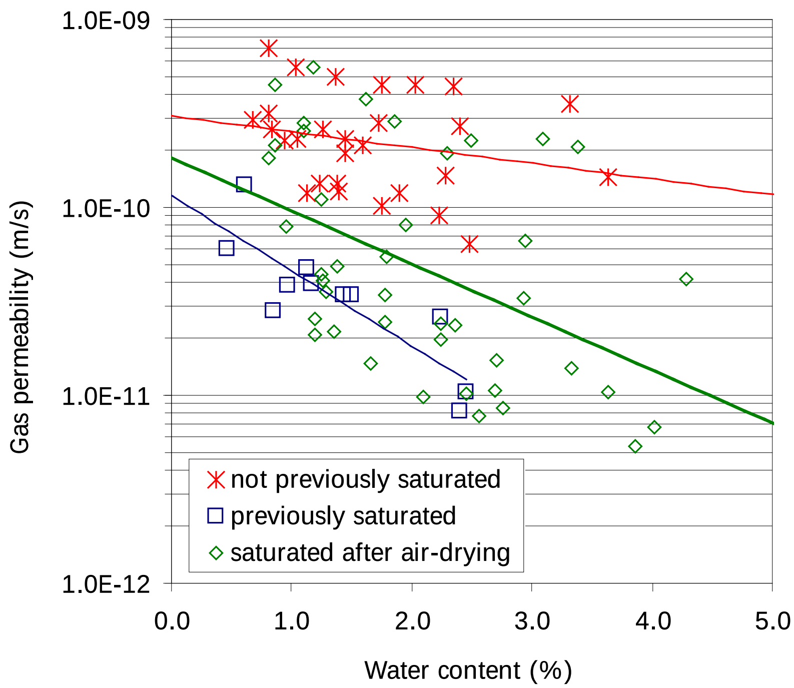

Figure 3.13: Change of gas permeability (k_g) with water content for samples with different hydraulic history

The hydraulic conductivity of these samples was measured after saturating them with deionised water. Normally, the longer the saturation time the lower the hydraulic conductivity. In some instances it took about 100 days to get a stable permeability value. When the hydraulic conductivity was determined again after air drying, the value obtained was much lower (up to two orders of magnitude), although the difference between both values tended to be lower over time. Thus, the average intrinsic permeability value for this concrete deduced from water permeability measurements would be $3.6 \cdot 10^{-18} \pm 5.1 \cdot 10^{-18}$ m^2.

From the relationships between gas permeability (k_g) and water content (w) shown in Fig. 3.1, the gas permeability for w=0% can be extrapolated, and from it the intrinsic permeability (k_{ig}) for each group of samples, since the relative gas permeability (k_{rg}) for a dry sample is 1. The intrinsic permeability obtained for the three groups of samples are of the same order of magnitude amongst them and two orders of magnitude higher than the value obtained in saturated samples with water flow (in the order of 10^{-16} m^2). Having these intrinsic permeability values, the relative gas permeability for each measurement performed can be computed through Equation 1, where μ_g is the dynamic viscosity of nitrogen ($1.79 \cdot 10^{-5}$ Pa·s) and ρ_g its density (1.12 kg/m^3). The change of relative gas permeability with degree of saturation (S_r) is more acute for samples previously saturated, whereas for samples not previously saturated the change of relative gas permeability with degree of saturation is not so significant and the relative gas permeability is distinctly higher for degrees of saturation higher than 20% (Fig. 3.2). Overall, the relative gas permeability of concrete increased sharply for water degrees of saturation smaller than 50%.

$$k_g = \frac{\rho_g \times g}{\mu_g} \times k_{ig} \times k_{rg} = 6.2 \cdot 10^5 \times k_{ig} \times k_{rg} \qquad (1)$$

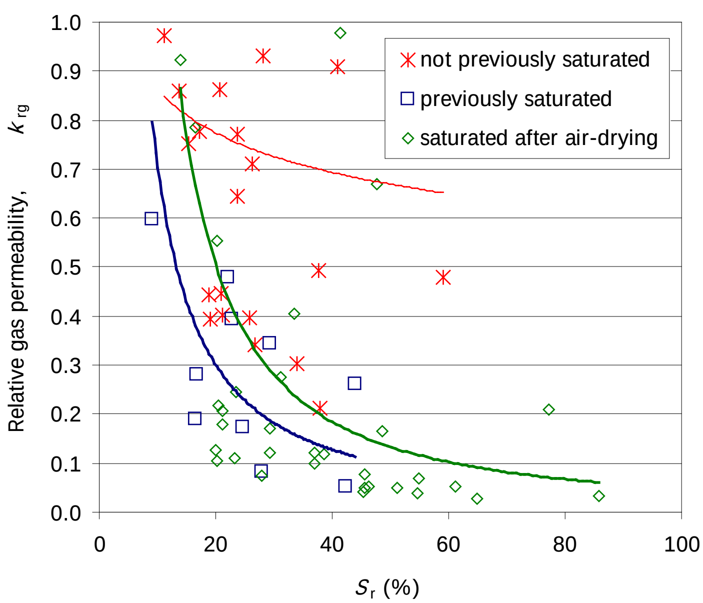

Figure 3.14: Relative gas permeability of concrete as a function of degree of saturation for different hydraulic histories

Also, the boundary conditions affected the gas permeability values, and thus, they seemed to be mostly conditioned by the backpressure and the confining pressure, increasing as the former increased and decreasing as the latter increased, *i.e*. decreasing as the effective pressure increased. Overall the increase of pressure head or injection pressure implied a decrease in gas permeability, although this effect was less significant when the pressure head was high. Consequently, all the values obtained with the unsteady-state equipment were higher than those obtained with the steady-state one, in which the confining and injection pressures applied were higher. Although microcracking during air-drying could not be ruled out as responsible for the decrease of permeability with confining pressure, the hydrostatic tests performed did not clearly confirm this hypothesis [1].

The effective permeability was calculated applying the Klinkenberg method to the results obtained with the steady-state equipment. The effective permeability thus obtained for a given effective pressure was only slightly smaller (by an average factor of 1.5) than the average of all the values measured for the same effective pressure range. For this reason it was considered that the Klinkenberg effect was not relevant in the range of pressures applied.

4. Conclusions

Gas permeability decreased with water content, but it was also greatly affected by the hydraulic history of concrete. The relative gas permeability of concrete increased sharply for water degrees of saturation smaller than 50%. Also, the boundary conditions affected the gas permeability values, and thus, they seemed to be mostly conditioned by the effective pressure.

The liquid water permeability of the concrete was about two orders of magnitude lower than the gas permeability (10^{-18} *vs*. 10^{-16} m^2), probably due to the chemical reactions taking place during saturation. Water reacts with the cement paste and can hydrate the anhydrated cement and change the chemical equilibria by dissolution, leaching and/or precipitation of calcite (if the latter is carbonated). This alteration is probably irreversible, since the gas permeability of previously saturated samples is lower than that of samples not previously saturated. Saturation after air drying implies a slight increase in gas permeability. The gas slippage or Klinkenberg effect is usually invoked as the reason for the difference between gas and water permeabilities, but this effect did not seem relevant in our tests.

The slowness of the processes occurring in concrete was illustrated by the time needed by the samples to get equilibrium water contents under room conditions and to reach a constant permeability value when they were saturated. The behaviour observed, in particular concerning hysteresis, highlighted the importance of microstructural analyses to understand the hydraulic properties of concrete.

5. Acknowledgements

The research leading to these results hasreceived funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under Grant Agreement n^{o}230357, the FORGE project. The laboratory work was performed by Ramón Campos and Juan Aroz, from CIEMAT.

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Mechanical Stability of Engineered Barriers in Sub-surface Disposal Facility during Gas Migration Based on Coupled Hydro-Mechanical Modelling

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[2] Japan Nuclear Fuel Limited (JNFL), Aomori, Japan

Summary

The "sub-surface disposal" concept has been proposed for relatively higher activity LLW in Japan. This concept includes a low permeability layer made of bentonite material (LPL) and a low diffusion layer made of dense cementitious material (LDL). The influence of gas generation and migration on mechanical stability of the engineered barrier system is one of the issues for long-term performance assessment of the disposal facility. In this study, coupled hydro-mechanical modelling and analyses are carried out in order to evaluate the mechanical stability of the system. Two gas generation rate cases are simulated here; 1) reference case, and 2) conservative case. It is found from the analyses that tensile stress developed in the cementitious components due to accumulated gas pressure is sufficiently less than the tensile strength of the materials and that stress developed in the LPL is kept compressive apart from the interface between the LPL and the LDL, which suggests opening could occur at the interface. These results indicate that the gas pressure would not mechanically damage the engineered barrier system of the sub-surface disposal even if a relatively high gas generation rate is assumed.

3. Introduction

In Japan, some of the low-level waste with a relatively higher concentration of radioactivity from nuclear facilities including power plants and fuel cycle facilities is to be disposed of in a facility located deeper than typical underground construction and exploration. This disposal concept, "sub-surface disposal" [1], is shown in Fig.1. It includes a low permeability layer (LPL) made of highly-compacted bentonite material and a low diffusion layer (LDL) made of a dense cementitious material. The LPL maintains an internal diffusive region, and the LDL prevents radionuclides from migrating outside the facility. Since the concept also includes steel containers, metallic waste [2] and concrete reinforcing bars, hydrogen gas can be generated by anaerobic corrosion of these metals after the closure of the facility.

If the generated gas can accumulate causing excessive gas pressure in the facility, the facility's barrier performance might be degraded by mechanical processes such as fracturing of surrounding rock and cementitious materials or large plastic deformation of the LPL. Kumagai et al. (2010) [3] performed mechanical analyses of the facility considering accumulated gas pressure inside the engineered barriers under typical loads including body force and bentonite swelling pressure, to evaluate the mechanical stability of the facility during gas generation and migration through the EBS. In that study a nonlinear finite element code, ABAQUS, was used for the mechanical analyses and the accumulated gas pressure applied to the EBS was estimated from two-phase flow analyses performed separately using a multi-phase flow analysis code, GETFLOWS [4].

In this study, coupled hydro(two-phase flow)-mechanical (HM) analyses are performed using

Code_Bright [5] to more realistically simulate hydro-mechanical behaviour during gas migration and consider the mechanical stability of the EBS. The results from the coupled HM analysis by Code_Bright are compared with the previous ABAQUS results together with the consistency of modelling assumptions.

4. Numerical modelling and simulation

2.1 Adopted constitutive model and material properties

The low permeability layer (LPL) made of bentonite (Kunigel-GX) is represented using the Barcelona Basic Model (BBM) [6] while all other components are treated as linear elastic materials. Most of the BBM material parameters are based on laboratory experiments and appropriate (conservative) assumptions are made for the rest (see Table 1). The linear elastic properties for waste, concrete pit, LDL, backfill and host rock are also based on laboratory experiments. Twophase flow parameters of the LPL and the host rock are determined by fitting experimental results [3], [7]. Those of other components are based on literature data for similar materials [3].

Table 1: BBM material properties and corresponding experimental data

Propety/ Material parameter	Recommended test (best)	Current status of test data for LPL (Kunigel-GX)	Parameter setting
Consolidation (saturated) λ_0, κ_0	• Standard consolidation test	Standard consolidation test data exist	Existing data are used
Consolidation (unsaturated) α, β, γ	• Controlled suction consolidation test • Uni-axial compression test	Uni-axial compression test data exist	Existing uni-axial compression test data are used for α with which swelling index is defined Independency of compression index of suction (β=γ=0) is assumed
Swelling κ_{s0}, α_{sp}	• Controlled suction loading/unloading test under constant confined stress, • Swelling pressure test under constant volume and swelling test under constant confined stress	Swelling test data exist	Existing data are used
Shear strength ϕ, c	• Controlled suction tri-axial compression test	Tri-axial compression tests data for saturated samples exist	Tri-axial compression tests data for saturated samples are used for ϕ and c, assuming conservatively that both internal friction angle and cohesion are independent of suction.

a. Finite element model

Figure 2 shows the finite element mesh and boundary conditions used for the coupled HM analyses by Code_Bright. Conservatively assuming that all porous media are saturated before gas generation, hydraulic and mechanical equilibrium under gravity and bentonite swelling pressure is initially reached. Following equilibrium, a flux of generated gas is applied uniformly to the waste layer area. Constant gas generation rates of 5 and 20 Nm3/y/Lot are assumed [3]. The former rate is a reference case; the latter is conservative. “Lot” is defined as the minimum unit cell in the concrete pit.

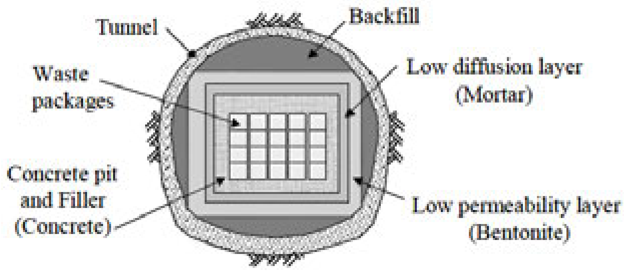


Figure 1. Basic concept of sub-surface disposal facility

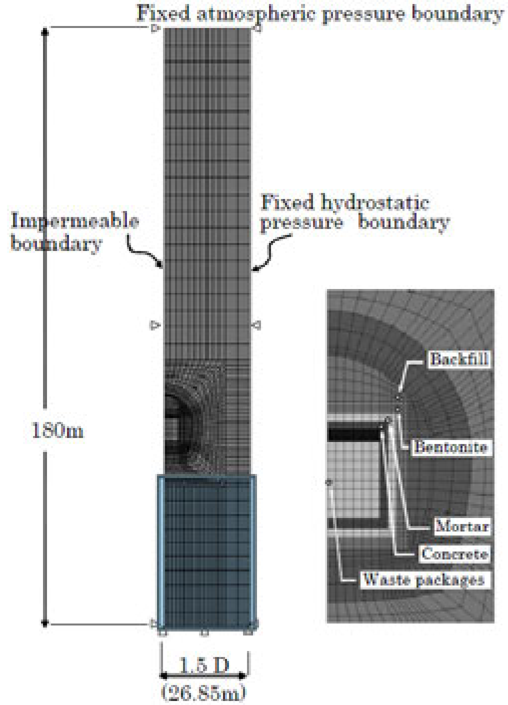


Figure 2: Code_Bright finite element mesh and boundary conditions

5. Simulation results and discussions

3.1 Accumulated gas pressure and gas flow path

Figure 3 shows the time evolution of gas pressure at the centre of the waste layer and upper right corners of the outer components. The gas pressure rises in the inner components and then develops toward the outer components. The gas pressure inside the LDL drops when the gas pressure in the backfill starts to increase, indicating that the gas reaches the more permeable backfill. The overall trends are quite similar in the both cases but the gas pressure magnitude is much higher and the phenomena (gas accumulation and migration) occur more quickly for the higher gas generation rate (20 Nm^3/y/Lot).

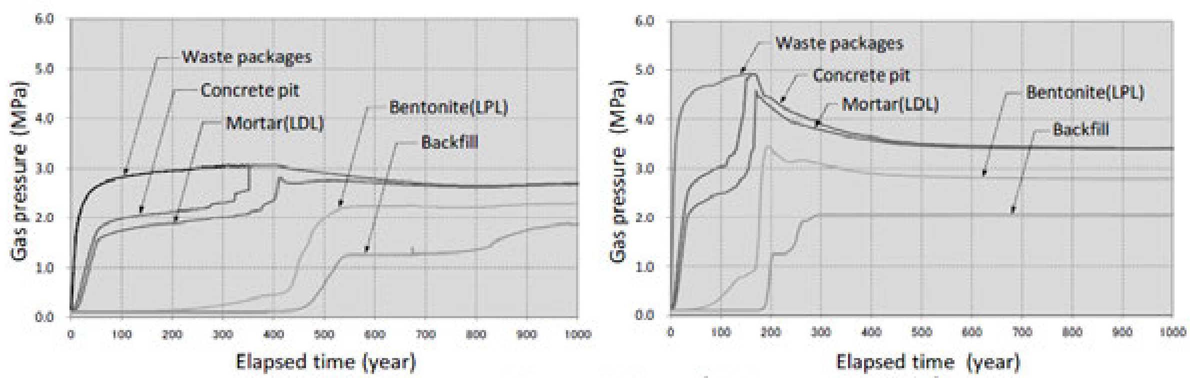
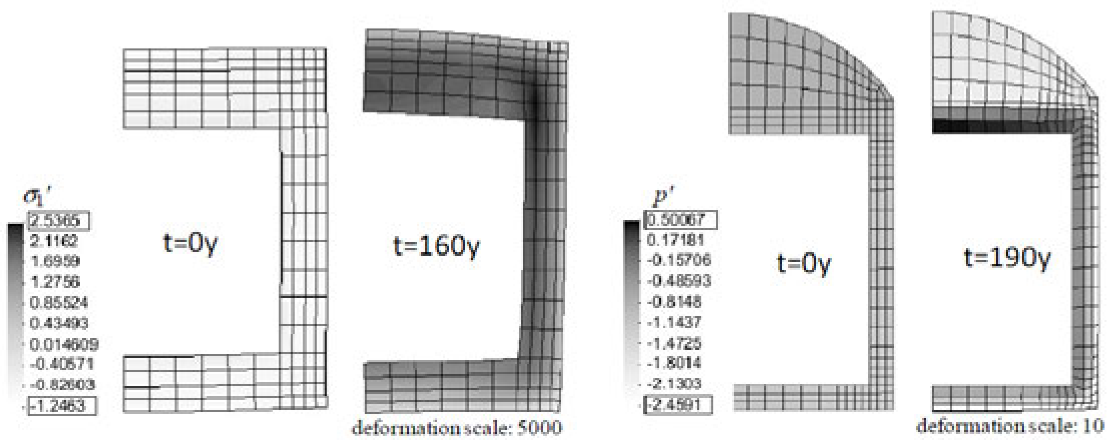


Figure 3: Gas pressure evolution: left $5Nm^3$/y/Lot, right $20Nm^3$/y/Lot

Mechanical stability of the EBS

Figure 4(a) shows the distribution of the maximum principal effective stress σ_1' in the LDL, the Concrete pit and the inner concrete filler for the 20 Nm^3/y/Lot gas generation rate case. The maximum value of σ_1' in all concrete components during the simulation is about 2.5 MPa. This is sufficiently lower than the tensile strength of the concrete (4.3 MPa), that cracking would not occur. Consequently the LDL and the concrete pit would be mechanically stable even if the higher gas generation rate (20 Nm^3/y/Lot) is assumed. The uncoupled analysis by ABAQUS showed cracking in some parts of concrete pit [3]. This suggests that the uncoupled analysis approach used by Kumagai et al. [3] would lead to a conservative evaluation of the mechanical stability of the facility. Figure 4(b) shows a representative distribution of mean effective stress p' in the LPL and the backfill for the 20 Nm^3/y/Lot generation rate. The stress developed in the LPL is kept compressive everywhere but the interface between the LPL and the LDL. This suggests that the LPL and the backfill could be kept mechanically stable, although interface opening might temporarily occur between the LPL and the LDL due to excess gas pressure for the case of higher gas generation rate. (p' is kept compressive over the entire area for the case of 5 Nm^3/y/Lot gas generation rate.)



(a) *(b)*

Figure 4 Distribution of (a) maximum principal effective stress σ_1' in the LDL, the concrete pit and the inner concrete filler and (b) mean effective stress p' in the LPL and the backfill

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Gas injection tests in the Meuse/Haute Marne underground research laboratory

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Summary

Several field scale experiments have been designed by the French national Agency for the management of radioactive waste (Andra) to understand: (i) the mechanisms controlling gas entry and gas migration in the Callovo-Oxfordian (COX) clay (PGZ1 experiment), the proposed host rock of the French deep geological repository, (ii) the interaction between gas and bentonite seals use to close the tunnels within the repository (PGZ2 experiments).

6. Layout

1.1 PGZ1

The experimental layout for PGZ1 consists of three boreholes. One dedicated to gas injection (PGZ1201) and the others to monitor pressure evolution associated with the hydraulic and gas injection test (PGZ1202), and potential deformation associated with the resulting stress changes (PGZ1031). The first gas injection phase (GAS1) was performed over a 14 months period between February 2010 and May 2011 into the middle interval of PGZ1201 borehole. It consisted of a series of six constant-rate gas injection tests at increasing peak pressure, each followed by a relatively long recovery period during which the gas pressure was allowed to decay. The maximum gas pressure imposed on the system was 9.1 MPa which is well below the minimum principal stress of around 12.3 MPa at the interval depth. Two hydraulics tests were performed before (HYDRO1) and after (HYDRO2) the long gas injection test GAS1 to see any change in term of water conductivity.

1.2 PGZ2

PGZ2 experiment is a set of a five horizontal boreholes drilled between two galleries to allow access from both sides and to avoid sensor equipment through the bentonite plugs. Two kind of materials have been tested: (i) pre-compacted bentonite plugs made with a mixture of MX80 and sand (70%/30% in mass), dry density about 1.6 g/cm3 (boreholes PGZ1011, PGZ1012 and PGZ1013) (ii) pellets/powder mixture made with MX80 directly built in place (borehole PGZ1021), dry density about 1,54 g/cm3. The re-saturation of the bentonite plugs was only done by natural water from the host rock without any kind of water injection. In two boreholes gas has been injected during the re-saturation phase of the plug to observe the perturbation induced by gas (in PGZ1011 during 70 days and PGZ1021 during 20 months). Other boreholes were used to see bentonite re-saturation without gas injection to be able to estimate gas effects. In each borehole multiple pore pressure and total pressure sensors have been installed to follow as best as possible swelling pressure evolution and water saturation. PGZ1001 is equipped with multi-intervals completion to monitor pore water pressure between the GEX and GMR drifts.

2. Observations & Results

2.1 PGZ1

An overall pressure decline is observed for the observation intervals in the injection borehole PGZ1201 and in all the intervals in the observation borehole PGZ1202, which was shown to most probably be due to the drainage effect through the backfilling cement in borehole PGZ1031 and towards the GEX drift at atmospheric pressure, as the hydraulic conductivity of the cement is several orders of magnitude larger than the value of the COX clay.

The pressure measurements during GAS1 clearly show a distinct change in the behavior between the first three steps and the last three steps.

Our interpretation of the field data is based on the fact that not all of the water in the gas injection interval could be removed during the water-gas exchange prior to the start of GAS1. The volume of the remaining water is not known mainly because the exact test interval volume is unknown. This is due to borehole breakouts and potential borehole creep during the six month period between equipment installation and water-gas exchange. The range of test interval volume was estimated based on different assumptions on the convergence of the clay rock around the test interval. In addition, the gas volume estimates were determined from the early linear pressure increase for each injection step based on the ideal gas law, assuming that no gas entered the formation.

The field analyses indicated that water was totally displaced from the injection interval by the start of the third injection step. Before that, the observed pressure responses in the injection interval represent displacement of water from the interval into the rock mass. The pressure responses after the third step represent gas migration into the rock. A maximum value of 2 MPa is obtained as the gas entry capillary pressure of the COX clay. This value is at the lower range of the values expected for this type of rock and might be a value representative of the borehole damaged zone of the COX clay. The analysis of the hydraulic tests shows the existence of a borehole damage zone of a few centimeters at the test interval of PGZ1201 with a hydraulic conductivity roughly two orders of magnitude higher than the sound clay rock.

This field analyses give key elements to reproduce well measurements by numerical simulation.

The hydraulic conductivities of the sound rock obtained from the hydraulic tests HYDRO1 and HYDRO2, preceding and following GAS1, are similar, which demonstrate that the 1.2 years long multi-step gas injection phase did not lead to any change in the water flow behaviour in the COX clay.

2.2 PGZ2

2.2.1. Bentonite-sand mixture plug

Natural hydration of the bentonite-sand mixture was monitored in PGZ1012 and PGZ1013 boreholes. Thanks to mock-up test, the hydration scenario is as follow:

- First, suction was applied during the steam phase via the moist air surrounding the plug. The duration of this phase was between a few hours and a few days.
- As soon as part of the plug came into contact with the claystone the plug would hydrate more quickly. On the whole, the amount of water reaching the plug was sufficient to maintain the swelling pressure.
    - the total pressure first increased significantly (for 20 to 40 days) because of the water hydrating the outer layers of the plug. Most of the forces were applied axially due to the rigidity at the centre of the plug, whose state of saturation was close to its initial state. Then the total pressure decreased (for 10 to 30 days) as a result of the water re-balancing from the outer layers of the plug to the centre, which became increasingly plastic. Finally, total pressure increased again, indicating the uniform saturation of the plug and, therefore, the uniform swelling pressure.

The total duration to reach full hydration was approximately 500-600 days for PGZ1012 and PGZ1013 boreholes and the swelling pressure was 6.5 and 4.5 MPa respectively. The swelling pressure was set at 7 MPa. Swelling pressure equals to total pressure minus pore pressure.

For PGZ1011, gas (nitrogen) was injected at different pressure during the natural hydration of the plug (October 2009 to February 2010). The only difference for PGZ1011 concerns the pore pressure build-up which is slightly slower than the other bentonite-sand plug. The swelling pressure of this plug is closed to 5 MPa.

The gas injection test showed that the gas did not penetrate the plug (based on the gas pressures and flow rates tested), even though it was injected directly in contact with the plug. This indicates that all of the bentonite / stainless steel, packer / claystone and resin / claystone interfaces play a major role in the transfer of gas in this type of configuration. Thus, some of the gas migrates along the borehole, and this migration is clearly facilitated by its geometry. The borehole's initial break-outs do not favour ideal contact between the rubber of the packers and the claystone, despite the convergence of the claystone around the packer.

In 2011, after full hydration, hydraulics tests were performed to get the water conductivity of the bentonites and plugs. All water conductivities were closed to 1 to 5 10-13 m/s. Those values are in agreement with the target value based on dry density of bentonite-sand mixture. The gas injection test performed in PGZ1011 during hydration had no impact on the value of the water permeability.

2.2.2. Pure bentonite powder and pellets plug

In the borehole PGZ1021, gas (nitrogen) was injected at constant flow rate immediately after installation, on one of the surfaces of the fritted disc. This gas injection test started in April 2011 and was stopped in December 2012 (during 20 months).

Despite the gas injection, measurements taken in PGZ1021 are, on the whole, fairly close to those taken in the other compacted bentonite plugs (PGZ1011 to PGZ1013). The main difference is in the hydration period, which would be considerably longer than in the other boreholes. Furthermore, the hydration process appears to be more complex due to the granular texture of the core.

A quick gas test performed in November 2012 has shown that both plug faces are still connected to gas. In details, this test indicates that the centre of the plug is quite largely saturated but the peripheral zone is largely de-saturated. Indeed, bentonite density is lower at the roof because of the mode of filling. It is likely that this area is the privileged pathway for gas flows.

3. Conclusion

Some observations made during those in situ tests gave elements to precise our analysis on gas migration into the underground repository:

- Role of the EDZ has been highlighted on PGZ1. A part of gas injected penetrated in the damaged zone around the borehole for low gas pressure compared to those expected into the repository;
- Few quantities of gas were able to migrate through the intact host rock. PGZ1 confirmed that gas entry pressure in Callovo-Oxfordian claystone seems to be at least over 5 MPa;
- Gas injection around a compacted bentonite plug in PGZ2 during re-saturation phase does not induce high perturbation. Main observation is a delay in reaching the full saturation state. Expected swelling pressure has been obtained in spite of gas injection;
- Gas injection around pellets/powder mixture seems to reveal the difficulties with that kind of material, to reach an homogeneous state of swelling and to fill the void at the roof.

3D Water and Gas Transport Model of the HG-A Experiment

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Summary

This study describes modelling of the Mont Terri HG-A field experiment, examining gas and water flow in the Excavation Damage Zone (EDZ) of a small, backfilled and sealed tunnel in Opalinus clay. A 3D radial model was developed using the code T2GGM, a modified version of TOUGH2 (with optional gas generation model). Reproduction of the experimental results during the hydraulic test phase required time-variable EDZ permeability. Models using constant EDZ permeability were inadequate. The permeability of the EDZ during the multi-rate hydraulic tests and gas injection tests of the HG-A experiment was likely affected by swelling of the damaged rock in the presence of water and hydromechanical coupling. At the end of the hydraulic test, permeability stabilised, and changes due to swelling or hydromechanical coupling were reduced. The model of the three gas tests used a heterogeneous EDZ, with relatively permeable channels located on the sides of the tunnel. Gas injection tests all followed a similar pattern. Injected gas initially dissolved in the water until it became gas saturated. Then gas pooled at the top of the test zone, remaining largely trapped. The pool of gas eventually reached a depth where it could begin entering the permeable EDZ channel which filled with gas, forming a continuous gas-filled pathway connected to the drift atmosphere. After the gas pathway formed there was a pressure breakdown as gas outflows exceeded inflows. There was evidence of unstable gas flow pathways.

1. Introduction

This extended abstract documents the work of Geofirma Engineering from 2011 and 2012, modelling the Mont Terri HG-A field experiment, focusing on gas and water flow in the Excavation Damage Zone (EDZ) of a tunnel in Opalinus clay. Of interest was the temporal evolution of permeability in the system due to swelling of clay, the effect of hydromechanical coupling (i.e., fluid pressure, confining stress) on the permeability of the EDZ, and the effect of EDZ heterogeneity on the escape of gas from the test zone. Some experimental data are shown in Figure 1 and Figure 2.

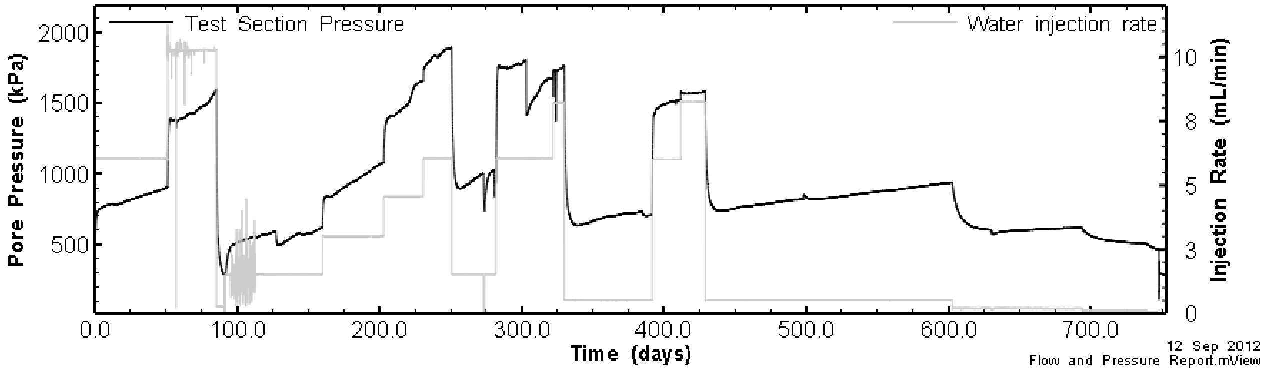

Figure 15: Fluid injection rates and test section pressures for the hydraulic test.

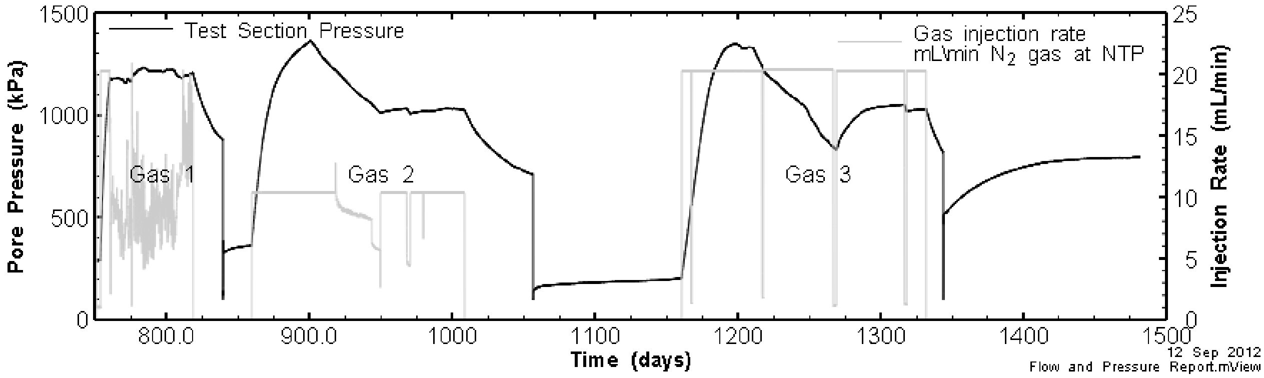

Figure 16: Fluid injection rates and test section pressures for the gas injection test.

2. Methodology

To model this experiment, we used the T2GGM code (version 3.1). T2GGM is a code that combines TOUGH2 with the Gas Generation Model, GGM (Quintessa and Geofirma 2011[1]). The TOUGH2 module EOS3 was used for calculations, with the properties of the gas component changed to reflect the fact that the gas is pure nitrogen rather than air. GGM components were not used in the modelling described here.

Initial model runs used constant values for EDZ permeability. However, this approach was found to be inadequate for matching the pressure history. A new subroutine allowing the specification of time-dependent permeability was implemented in T2GGM, allowing the user to modify the permeability in part of the model domain by specifying a time dependent permeability multiplier. To calculate the time dependent permeability in the HG-A test, the rate of water injection and the resulting change in pressure was used. The test section was idealised as a closed volume with one inlet (the injection port) and one outlet (the EDZ). It was further assumed that the response in any given time step could be modelled as steady-state, as supported by the very rapid equilibration of the system to changes in pumping rate (Figure 1). With these assumptions, it was possible to solve for the apparent permeability of the EDZ.

The HG-A experiment lends itself to modelling with a radial grid. Initial modelling was done with a 2D radial grid, assuming an axisymmetric problem and property distribution. The optimised properties from the 2D grid were applied to a 3D radial grid which incorporated the effect of gas buoyancy and the apparent heterogeneous permeability of the EDZ. All results presented here are based on this 3D radial grid.

The model was split into four stages: (1) the water-only hydraulic test from days 0 to 750, and three gas tests, which cover days 753 to 840 (Gas 1), 859 to 1057 (Gas 2), and 1160 to 1345 (Gas 3). The three gas tests were separated because the pressure and saturation in the test zone had to be reset before each test due to a gas/water exchange cycle.

3. Results and Discussion

The time-dependent permeability function (described in Section 2) was used to model the hydraulic test, and the results are presented in Figure 3. It is clear from the figure that using the time dependent permeability function and the measured injection rates resulted in a remarkable model fit to the observed test zone pressures. Taken together with the failure of constant permeability tests to produce a satisfactory result, this suggests that the only possible way to correctly model the HG-A experiment is with a non-constant permeability. It also suggests that the assumptions made when generating the permeability function (described in Section 2) provide an adequate conceptual model of what is actually happening in the HG-A test zone.

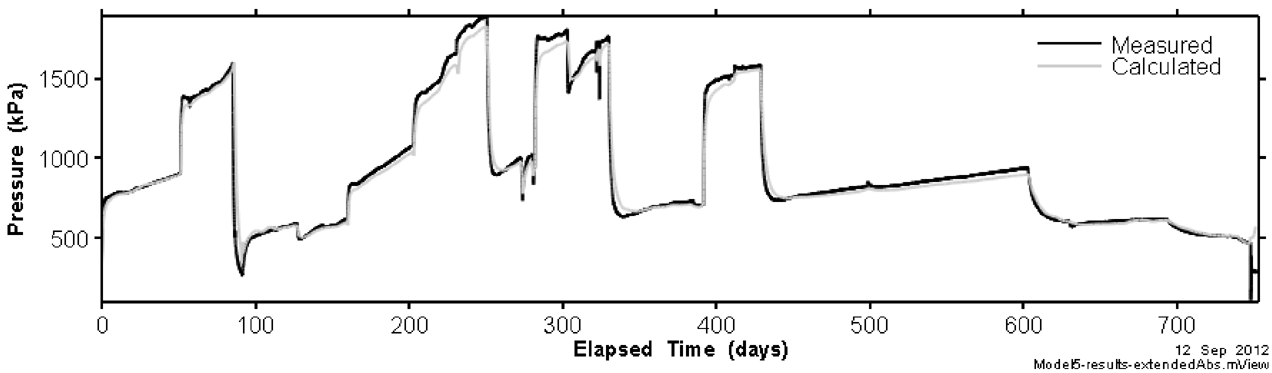

Figure 17: Measured and modelled test zone pressures during the multi-rate hydraulic test, variable EDZ permeability model.

A relatively small amount of gas escaped from the test zone during Gas test 1. Pressure was primarily dissipated by water escaping through the EDZ and channel. The model was able to match test zone pressure quite well. Results for Gas 1 are shown in Figure 4.

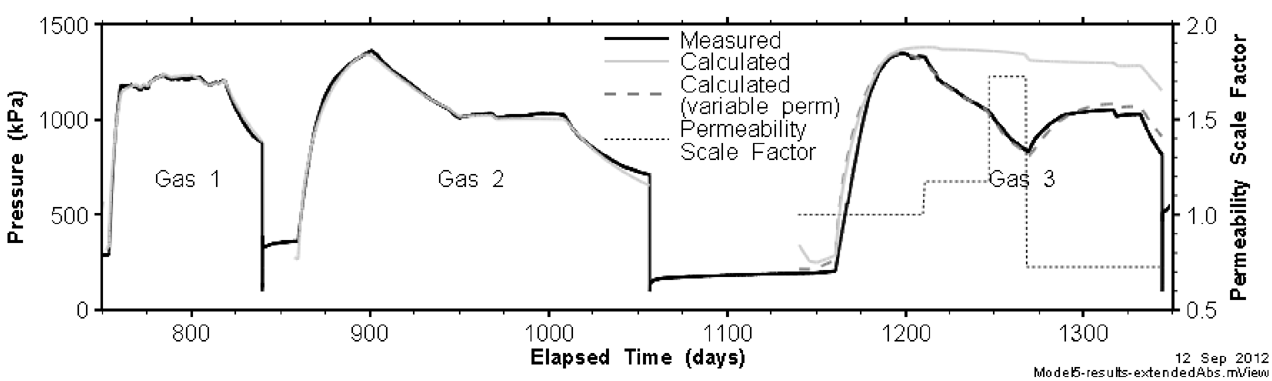

Figure 18: Measured and modelled test zone pressures during the three gas injection tests.

Results for Gas 2 are also shown in Figure 4. The pressure increased until approximately day 900, at which point, there was a sudden change in the trend and pressure began to decrease. The decline in test section pressure coincided with the formation of a continuous gas pathway along a channel in the EDZ, connecting the gas filled portion of the test zone to the atmosphere, allowing the gas to flow freely through the channel without the need to displace any water. At day 950, the pressure reached a steady state and pressures were maintained almost constant until gas shutoff at day 1009. Once again, the modelled pressures compare well with the measured pressures.

Results for Gas 3 are also shown in Figure 4. Gas tests 2 and 3 had approximately the same maximum pressure, and took approximately the same amount of time to reach that pressure, despite the fact that the injection rate for Gas 3 is double that of Gas 2 (Figure 2). In order to replicate this behaviour, it was necessary to raise the test zone compressibility by a factor of almost three, suggesting that something changed between the tests. Given the apparently higher test zone compressibility during Gas 3, the most likely explanation is trapped gas in the test zone.

A further complication in the Gas 3 test is that a time varying permeability was needed to match the pressure. Figure 4 shows three curves for Gas 3. The solid grey line (labelled “Calculated”) shows modelled pressure using the EDZ permeability value optimised for Gas 2. This model matches the initial pressure rise and gas breakthrough rather well, but is unable to reproduce pressures during the remainder of the test. The dashed grey line (labelled “Calculated (variable perm)”) shows a much improved model fit, which required minor adjustments to permeability (shown in the dashed black line, labelled “permeability scaling factor”). The test section pressure was on a downward trend when the gas injection was stopped between days 1267 and 1269. When gas injection restarted, pressure began to rise, and reached equilibrium at a pressure higher than the pre-stoppage pressure. We interpret this as an unstable gas pathway that expanded during the earlier part of the injection (before day 1267) and then closed during the two days that the gas injection was interrupted. This was modelled as a change in permeability at that time.

5. Conclusions

Modelling of the HG-A experiment, as part of FORGE WP4.3, aims to improve the understanding of gas and water flow in the EDZ of a tunnel in Opalinus clay. To date, the main conclusion of this modelling effort is that it is not possible to reproduce the experimental results (from January 2008 to April 2010) with a model using constant EDZ permeability. It seems clear that the permeability of the EDZ during the first 750 days of the HG-A experiment was affected by two distinct processes. Ongoing swelling of the damaged rock in the presence of water led to healing of fractures and a steady reduction in the permeability of the damaged zone. Additionally, hydromechanical coupling impacted permeability, as changes in pore pressure and confining stress (packer pressure) caused rapid changes in permeability. A simple analysis was able to quantify, to a reasonable degree of accuracy, the changes in permeability. Applying this variable permeability curve achieved good correspondence between modelled and measured pressures in the test zone.

The three gas injection tests followed a similar pattern of gas flow. At very early times the injected gas is dissolved in the water according to Henry's Law, until the water became saturated. At this point, the injected gas began to pool at the top of the test zone, remaining largely trapped. Eventually, the pool of gas reached a depth where it could begin entering a preferential flow pathway in the EDZ. The fractured pore space in the channel quickly filled with gas, and once the channel formed a continuous gas pathway to the atmosphere, there was a pressure breakdown as gas flowed out of the test zone faster than it is being injected, until a new equilibrium was established. In Gas test 3, there is evidence for the development and collapse of unstable gas flow pathways in the EDZ.

6. Acknowledgements

Modelling of the HG-A experiment is part of FORGE (Fate of Repository Gases) WP4.3. The modelling by Geofirma Engineering is supported by the Nuclear Waste Management Organisation (NWMO), Canada.

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Investigations of Gas Migration through Undisturbed and Resealed Clay Rocks

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Abstract

Gas migration in clay host rock of a nuclear waste repository is an important issue in safety considerations, because waste forms and other metallic components will produce corrosion gases throughout the whole time period regarded in performance assessment. During the last years, GRS performed various laboratory and in situ investigations addressing migration mechanisms, gas entry pressure and permeability, and resealing effects. The results imply that, rather than gas fracturing, the most relevant mechanism will be dilatancy controlled gas flow. Previously damaged clay rock, like the excavation damaged zone, will be resealed under load and resaturation, but still have the capacity for gas migration, with moderate gas entry pressures, thus inhibiting high pressure build up.

1. Introduction

An important issue in the safety assessment of a nuclear waste repository is the impact of gases produced mainly from corrosion of waste containers and other metallic components within the facility. Gas transport in consolidated clay formations may be hindered due to the very low permeabilities and full water saturation of both the host rock and the engineered barriers in the repository. Build up of high gas pressure which may cause fracturing in the multi-barrier system in case of exceeding the minimum principle stress is a potential issue that has to be investigated. However, the excavation damaged zone (EDZ) may act as a conduit for preferential gas flow, depending on the resealed state during the post-closure phase. Recently, GRS has investigated gas migration in undisturbed and resealed clay rocks in laboratory and in situ. The laboratory experiments were carried out mainly on resealed claystone samples from the Callovo-Oxfordian argillite (COX) and Opalinus clay (OPA), while the in-situ experiments were conducted in the Mont Terri Underground Rock Laboratory (URL) [1-4]. Main results will be presented in this paper.

2. Laboratory experiments

2.1 Methodology

Various kinds of laboratory gas migration tests have been designed and conducted on resealed COX and OPA samples of different sizes (diameter/length = 50/50 – 280/600 mm) and shapes (cylinder & hollow cylinder). Figure 1 shows schematically the principles of two types of the tests: (a) gas flow in axial direction nearly parallel to fracture planes remaining in the samples; and (b) gas flow in radial direction nearly parallel to bedding planes. The apparatus used in the first test type allows simultaneous testing of five samples confined in individual cells, while four samples can be simultaneously tested by means of another apparatus in the second type of test. In each test type, the samples are sealed in rubber jackets and loaded hydrostatically at identical levels. Gas injection is usually performed by stepwise increasing pressure at the upstream and keeping atmospheric pressure at the downstream. The downstream is connected to a burette for measuring gas outflow. In the test of the second type, gas is introduced into a thin central borehole filled with fine-grained quartz sand and allowed to flow into the annular gap between sample and jacket. Nitrogen gas was used in all the tests.

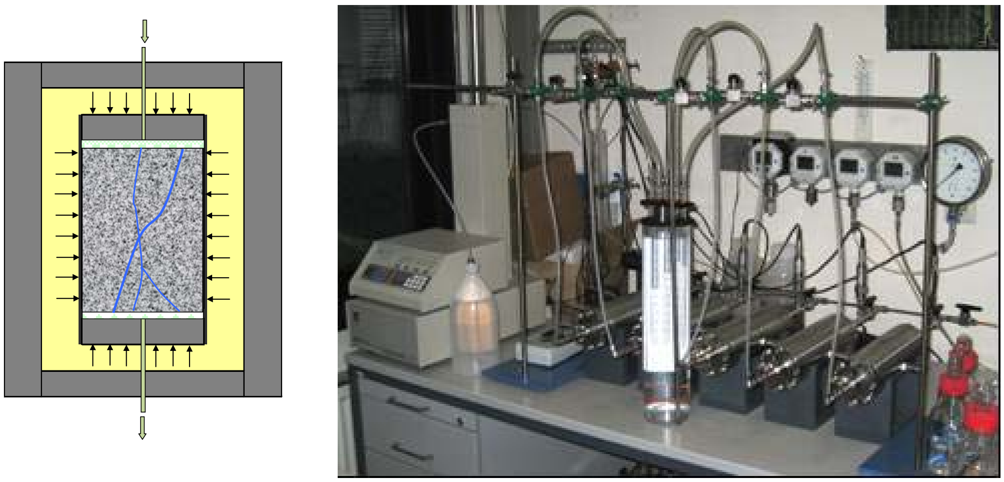

Fig. 1a: Axial gas flow through five samples

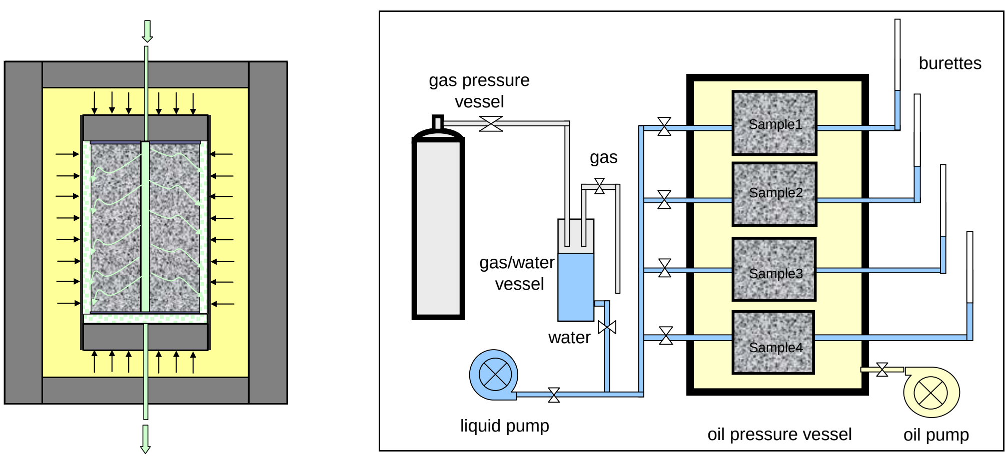

Fig. 1b: Radial gas flow through four samples

Figure 1: Layout and setup of gas migration tests with resealed claystone samples

2.2 Typical results

Five artificially-cracked COX and OPA samples of d=50mm and l=50mm were highly resealed during the previous sealing tests over more than 3 years [5-6]. Due to the fractures, their initial gas permeabilities before sealing were quite high, ranging at $k_g \approx 3\cdot10^{-12}$ m^2. Then, after flowing synthetic COX porewater through them under confining stresses of 2 – 3.5 MPa and temperatures between 20°C and 90°C, very low water permeabilities were reached, in a range from $3\cdot10^{-19}$ to $7\cdot10^{-21}$ m^2, close or even equal to that of the intact claystone. The pictures of the samples before sealing and the water permeabilities after sealing are summarized in Table 1. The strongly-resealed samples were used for the gas migration testing.

Table 1. Characters of the samples before and after sealing as well as key gas parameters determined during the gas migration test at confining stress of 2 MPa

cracked samples before sealing, gas permeability $k_g = 3\cdot10^{-12}$ m^2					
	OPA	COX1	COX2	COX3	COX4
water permeability k_w (m^2) after sealing	$3\cdot10^{-19}$	$2\cdot10^{-19}$	$2\cdot10^{-20}$	$7\cdot10^{-21}$	$2\cdot10^{-20}$
gas breakthrough pressure p_b (MPa)	0.20	0.40	0.60	0.60	1.00
gas entry pressure p_e (MPa)	0.02	0.27	0.31	0.6	0.19
gas permeability k_g (m^2) after breakthrough	$2\cdot10^{-18}$	$7\cdot10^{-20}$	$5\cdot10^{-20}$	$1\cdot10^{-21}$	$4\cdot10^{-20}$

The gas test began with a low injection pressure of 0.1 MPa at the upstream, followed by increasing the pressure step-by-step by 0.1 MPa with each step. Each step was kept for a sufficient time period of several days to a month. As gas outflow is observed, the corresponding gas pressure at the upstream is defined as *gas breakthrough pressure* (p_b). After breaking through, the *gas permeability* (k_g) can be measured. It is also possible to determine *gas entry pressure* (p_e) which was defined as the minimum constant shut-in pressure approached with time after switching off the inlet. This procedure was repeated for one sample to another with increasing gas injection pressure.

The preliminary results from all the five samples loaded at 2 MPa are plotted in Figure 2 in terms of gas upstream pressure and permeability after breaking versus the test duration of 4 months. The data show that:

a. the gas breakthrough took place sequentially from the weakly-resealed OPA sample at a pressure of p_b = 0.2 MPa to the strongly-resealed COX samples at increased pressures of p_b = 0.4, 0.6 and 1.0 MPa respectively, which are more than 50 % below the confining stress;

b. the gas permeability after breaking through is correlated with the initial water permeability, the higher water permeability yields a higher gas permeability;
c. the gas permeability increases with increasing gas breakthrough pressure and seems to be irreversible after decreasing the gas pressure (comparing the k_g-values of sample COX3 at p_g = 1.0 MPa before and after changing the gas pressure);
d. the shut-in pressure at which gas flow ceases decreases gradually with time to a constant value for each sample, and the stabilized gas entry pressure is significantly lower than the breakthrough pressure.

These observations suggest that the gas transport in resealed claystone is governed by opening and closing of the resealed fractures, namely dilatancy-controlled gas migration. This also implicates that the EDZ will act as a preferential conduit for gas flow before, during and after resealing. Possible impact of confining stress on the gas migration will be examined by continuing the tests at elevated loads up to 5 MPa, and in another test up to 15 MPa.

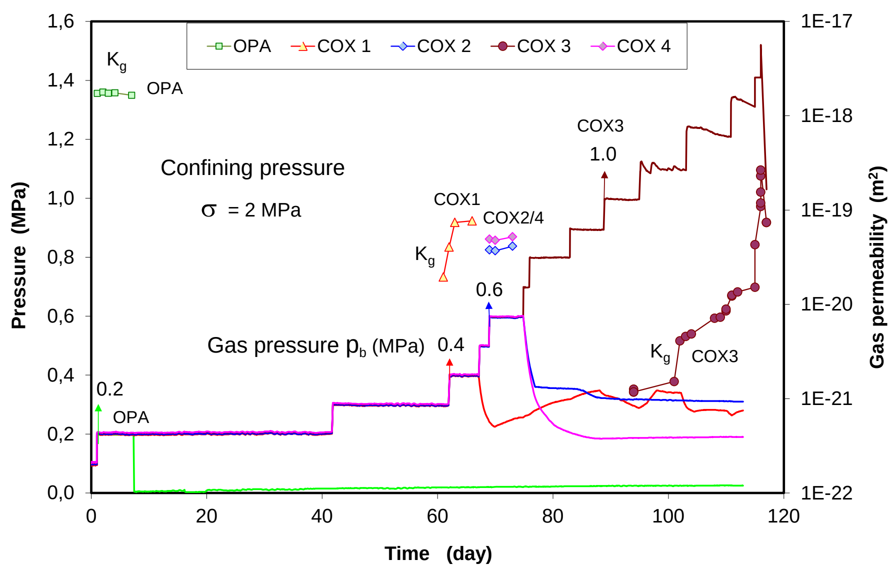

Figure 2: Evolution of upstream gas pressure and gas permeability after breaking through of resealed claystone samples under confining stress of 2 MPa

Up-scaling effect was investigated with large claystone samples [2, 4-5]. Figure 3 shows a test example on a large OPA sample (d=260mm/l=616mm). The sample was taken from the EDZ with a set of fractures along the bedding planes. Before resaturation, a gas permeability of ~10^{-16} m^2 was determined after loading/unloading cycles in a range of 3 to 18 MPa. The following water resaturation phase was conducted for about 2 months at injection pressure of 1.5 MPa and confining stress of 3 MPa, during which no water outflow was observed. This means that the sample might have been not yet fully resaturated. After that, gas was injected at a pressure of 0.5 MPa and a low permeability of $1 \cdot 10^{-19}$ m^2 was recorded. Increasing the gas pressure to 0.7 MPa accelerated the gas flow concurrent with an increase of permeability to $1 \cdot 10^{-18}$ m^2. These permeability values are 2 to 3 orders of magnitude lower than those observed before water injection. An additional stress increase from 3 to 6 MPa reduced the permeability significantly to less than 10^{-20} m^2, but the gas transport did not cease at the relatively low injecting pressure. A comparison with the results obtained on the small samples indicates no or little up-scaling effect. This conclusion was further confirmed for the rock mass by the in-situ measurements [1-3].

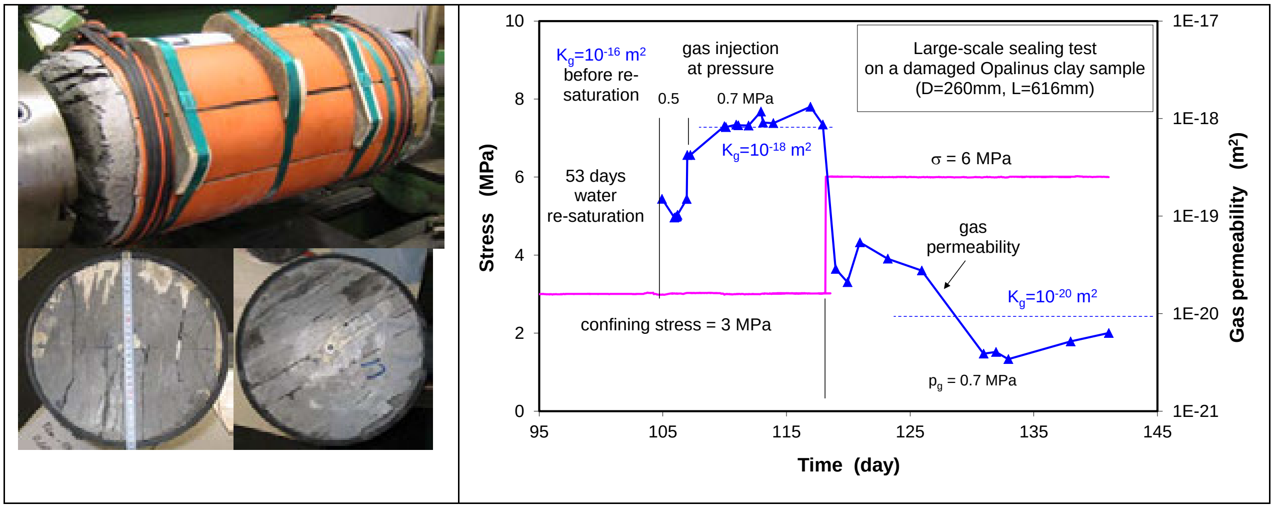

Figure 3: Results of the gas migration test on a big claystone sample

3. In-situ measurements

The objectives of the in-situ investigations were to gain more information on the gas migration mechanism at pressures close to minimum principal stress, to determine gas entry pressures and effective gas permeabilities, and to investigate resealing in situ.

3.1 Methodology

The in-situ measurements of gas migration were performed in the near-field around a tunnel in the Mont Terri URL. A total of six boreholes with diameter of 20 mm and lengths between 6 m and 10 m were drilled with an inclination of 45°, three of them parallel and the other three perpendicular to the bedding. The lowest 40 cm of each borehole were equipped with sinter steel mini-piezometers, the rest of the boreholes was filled with synthetic resin for sealing.

Figure 4: Drilling of boreholes; mini-piezometer before installation

After installation, the piezometer test intervals were filled with synthetic pore water. Then, water injection tests for determining water permeability were performed. Several cycles of gas injection tests were performed: After the first gas injection tests, the gas was left in the system. Half a year later, a second gas injection cycle was performed. After these tests, the gas in the piezometers was replaced by pore water, and several water injection cycles were performed, before, 1.5 years after the second gas injection cycle, a third gas injection cycle was performed for detection of resealing effects.

Water injection tests were performed as pulse injection tests at pressures of 0.5 - 1.9 MPa, the injected water mass was measured and the water permeability was determined from the pressure decay curves.

Gas injection tests were performed by first replacing the water in the test interval with nitrogen and then increasing the gas pressure in steps. The gas tank was put on high-accuracy scales in order to detect and measure the flow rate. Gas entry pressure is interpreted as the pressure at which a first flow rate is detected. Additional steps at higher pressure are added. From the flow rates and the corresponding pressures “effective” permeability values are derived.

3.2 Typical results

Before gas testing, water permeabilities between $2 \cdot 10^{-20}$ m^2 and $5 \cdot 10^{-19}$ m^2 were measured. The first gas injection tests in two of the piezometers resulted in values for the gas entry pressure of 1.8 MPa (BET 2) and 2.4 MPa (BET 5), respectively. These are assumed to be in the range of the minimum principal stress. Half a year later, the gas entry pressure in both intervals was by 0.6 MPa lower. This is interpreted by an inhibition of resealing by the gas that was left in the system after the first tests. After removal of the gas and water injection, permeability testing with water 16 months later seemed to imply that the original state was more or less reached again. When the gas injection tests were repeated afterwards, gas entry pressures close to the original ones were determined. Figure 5 shows the pressure and gas mass recording of the last gas injection test at BET 5 which yielded a gas entry pressure of 2.2 MPa.

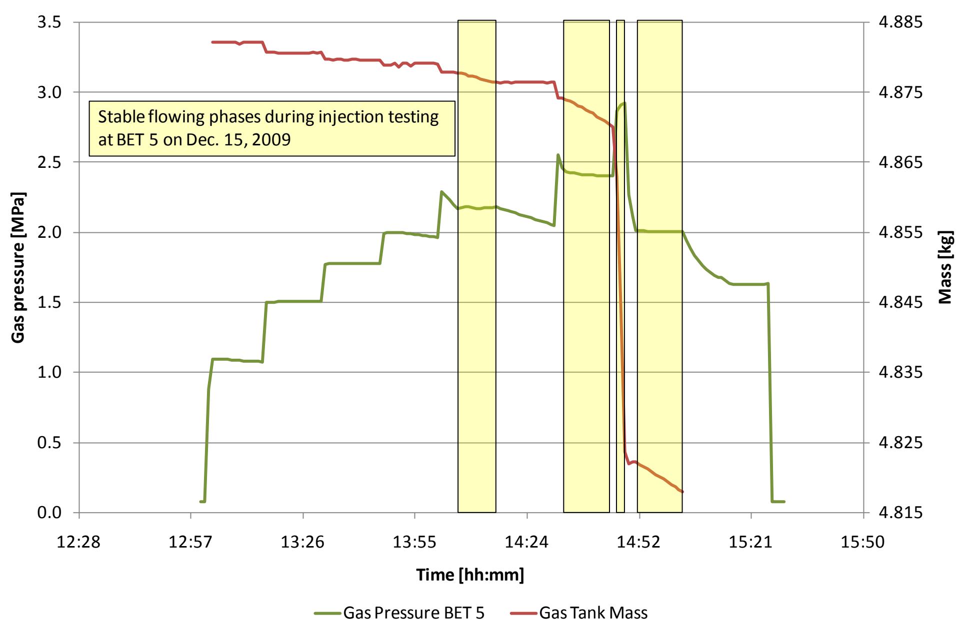

Figure 5: Last Gas injection BET 5: Gas injection pressure and gas tank mass

In Figure 5, the stable flow phases which are used for determining the effective gas permeability are marked in yellow. The measurements in all of the tests ranged between 10^{-19} m^2 and nearly 10^{-15} m^2, with most of the values in the order of 10^{-18} m^2.

4. Conclusions

Both the laboratory and in situ measurement results confirm the high resealing potential of the clay rock. At gas pressures close to the minimum principal stress, the relevant migration mechanism seems to be dilatancy-controlled flow. This is supported by the observation that after only 1.5 years of resealing the gas entry pressure was nearly restored. Previously disturbed rock, on the other hand, can provide preferential gas pathways again as soon as the entry pressure is reached.

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FORGE WP1.2: Numerical Benchmark on Gas Migration at Repository Scale : Involved Teams and codes, Conceptual basis and main results.

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Summary

As different countries, having all different clay based disposal concepts, are involved in WP1.2 activities, the reference exercises were defined as generic as possible (without targeting precisely on national concepts), with sensitivity analyses making it possible to cover national specificities. The aim here is more to understand the behaviour of the modelled system as understood by each participant, rather than to undertake an inter-comparison of codes. To give some added value to this exercise, a feature not yet well represented in usual gas simulations was introduced: the explicit representation of the interface between canisters and cell walls.

On the over whole, the results from the different teams are globally coherent and give the same general vision of the hydraulic-gas evolution at scales defined by the benchmark (cell scale, module scale, repository scale), showing that two-phase flow numerical simulations at repository scale is now available using some upscaling technics and that further reduction of some parameters uncertainties could help precise the expected hydraulic gas transient behavior.

1. Introduction

FORGE WP1.2 is a sub work package dealing with benchmark simulation concerning gas generation and migration at repository depth (many hundreds of metres below ground level) and repository scale and including teams from several countries, having all different clay based disposal concepts.

One of the main problems in dealing with different teams from different countries, was to find a common representation as a basis for the benchmark. This implies simplifications are necessary to real concepts, in order that they be represented at a basic level that has some degree of consistency to all participants. So at the start of WP1.2 activities, after discussion, there was a general agreement that the reference exercise would be as generic as possible (without targeting precisely on national concepts), with sensitivity analyses making it possible to cover national specificities.

The aim here is more to understand the behaviour of the modeled system as underastood by each participant, rather than to undertake an inter-comparison of codes. To give some added value to this first exercise, a feature not yet well represented in usual gas simulations was introduced: the explicit representation of the interface between canisters and cell walls.

2. Methodology

Although the final aim of the benchmark studies is to represent repository-scale simulations, the first exercise should be rather simple and at cell scale [1]. This first step was done during 2009 and first part of 2010. Results are showing a significant role of interfaces between plug and argillite and a transfer dominated at this scale by convection toward the drifts, radial diffusion being of secondary order.

Representing the transfer of gas at a larger scale including drifts and several tens of cells is then worthwhile. In this context, one of the major problems in representing gas transfers in a repository for radioactive waste is to model simultaneously all gas sources (generally located in the disposal cells) and the transfer pathways constituted by the network of interfaces, plugs and undergrounds drifts. The second benchmark at module scale (several tens of cells, Fig. 2.1) shows once more that interface plays an important role in the transient hydraulic-gas evolution as well as significant differences with the cell results such as the important role of dissolution/diffusion process along the pathways in the drifts. Although at this scale some teams had important numerical problems, especially concerning the establishment of the mesh, none of them used explicit upscaling technics.

The simulation of a complete generic repository including ten modules (Fig. 2.1) will force the teams to introduce a certain amount of upscaling in there simulations. More than this, having a complete repository will permit to validate the results at smaller scales (cell and module) and to give evaluation of the impact of gas at the shafts.

For this exercise we assume a repository having a simple architecture (Fig. 2.1). This architecture is more or less representative of some general concepts for HLW repository zones but without any national specificity. The main features are:

- Subdivision of the all repository into several modules linked by a main drift;
- All the modules are equal and contains an access drift serving 50 cells on each side;
- Two adjacent modules are not separated by a seal;
- In the main drift seals are placed between all modules connections.

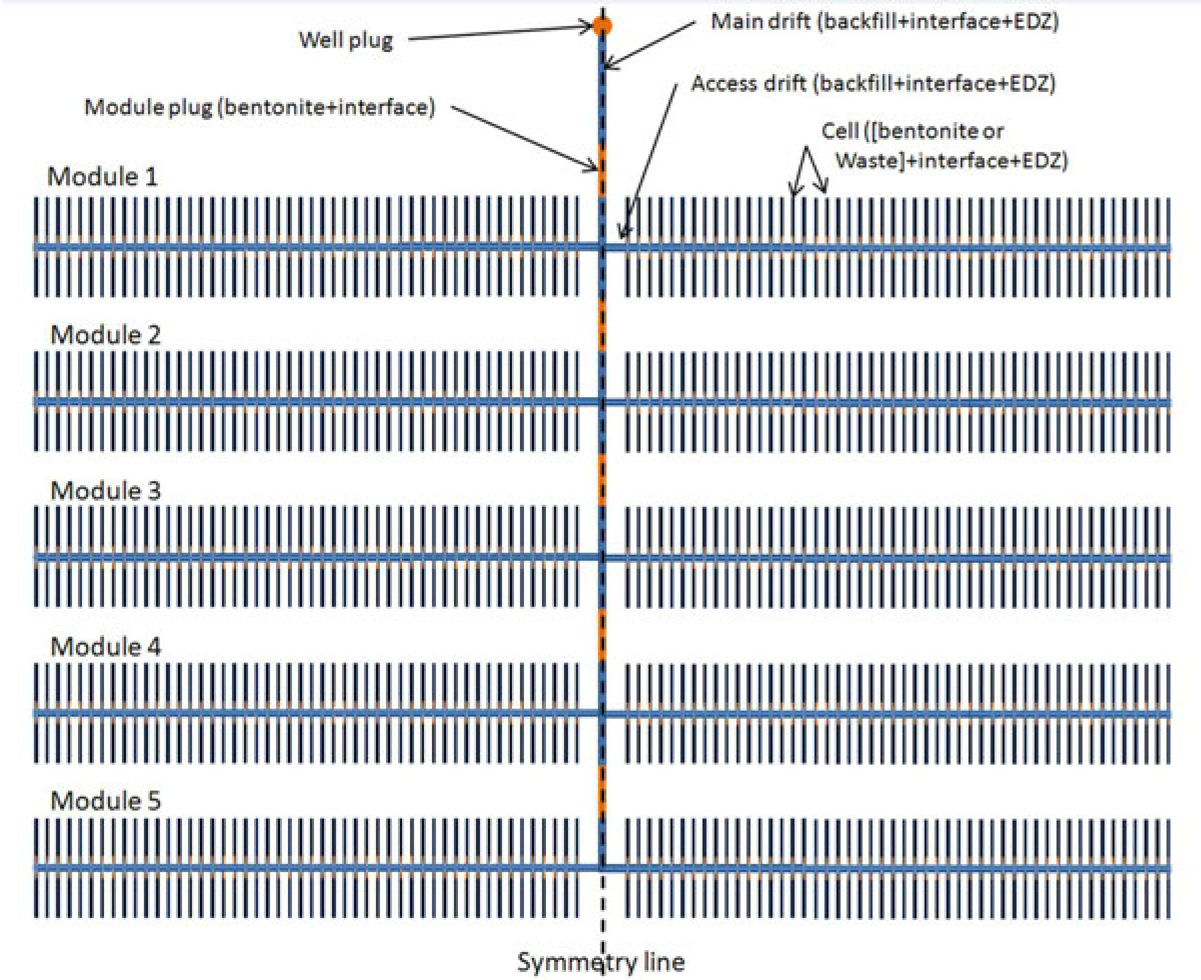

Figure 2.1: schematic representation of a repository for HLW

3. Results and conclusions

On the over whole, the results from the different teams are globally coherent and give the same general vision of the hydraulic-gas evolution at scales defined by the benchmark (cell scale, module scale, repository scale) (Fig. 2). However, significant discrepancies exists, and an important conclusion of this benchmark, consolidating similar ones in the past, is that numerical simulation is a powerful tool (if not the only one) to give a trend, a tendency, a general evolution, but cannot be used to determine precise values in time and space. Having this in mind, the main results can be summarized as follows:

- Using (numerical or mathematical) upscaling, nowadays computers allow complex two phase flow simulation of a complete repository with a precise representation at all scales (from cell to shaft including drift network and explicit representation of EDZ and host rock);
- As host clays are very impermeable and gas entry pressures are high, the desaturation is only of some percent and thus there is a need of better characterization of the “water saturated tail” of the relative permeability and retention curves for these materials;
- EDZ permeability (gas entry pressure) is much higher (lower) than in the clay rock, thus expressed gas is mainly flowing through this material : better experimental characterization of

the EDZ relative permeability and retention curves could also help increase the confidence on the simulation results.

- This last point is also true for the interfaces, adding intrinsic permeability values as well;
- Dissolution and diffusion are key combined processes in the comprehension of a repository hydraulic-gas transient and most of the produced hydrogen is finally migrating under dissolved form in the clay rock water. Decreasing the uncertainty on the host clay diffusion coefficient for H_2 could also help.

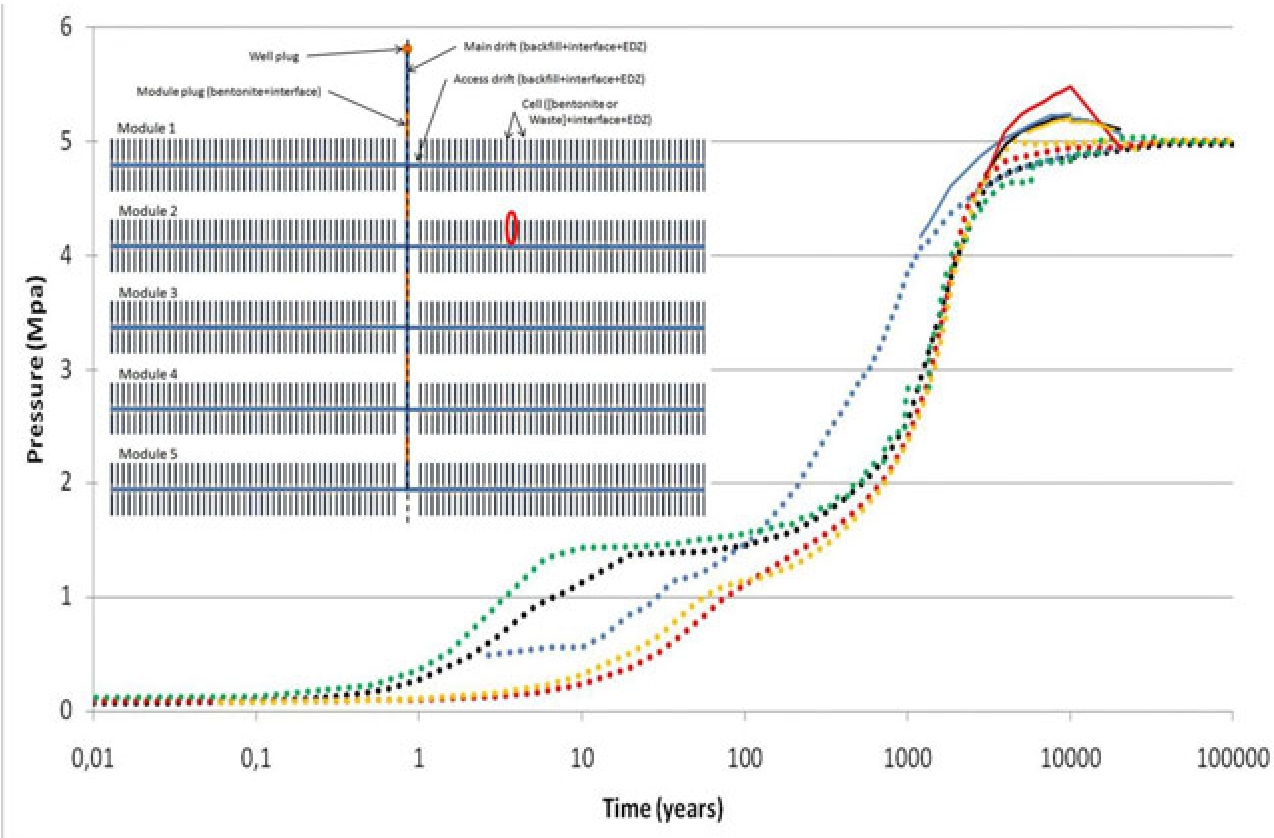

Figure 3.1: Water pressure (dashed lines) and gas pressure (continuous lines, only when gas phase exists) computed by different teams in the EDZ of a generic cell situated in the middle of a module (red ellipse on repository representation).

4. Aknowledgements

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme FTP/2007-2011.

5. References

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3D Numerical Simulation of Gas Migration Through Engineered and Geological Barriers for a Deep Repository for Radioactive Waste: FORGE WP1.2 Benchmark

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Summary

In the framework of the FORGE WP 1.2, three benchmark studies on repository-scale numerical simulations of gas migration were proposed. The first one was realized at cell scale and the second one is at a module scale (several tens on cells). This process can be formulated as a coupled system of partial differential equations modelling immiscible compressible two-phase flow in heterogeneous porous media. The purposes of this talk are to present a mathematical upscaling method combined to a vertex-centred finite volume discretization and numerical results for the two-dimensional benchmark at cell scale and the three-dimensional benchmark at module scale. A C++ homemade code and the DuMu^x code (http://www.dumux.uni-stuttgart.de/), a parallel multiphase flow simulator, have been adopted for this study.

1. Introduction

The type of cell considered in this benchmark is a somehow generic high level waste cell in a clay host rock. The aim of the first benchmark is to better understand the mechanisms of the gas migration at the cell scale and in particular to analyse the effect of the presence of different material and interfaces on such mechanisms. The main difficulties related to the second one are its size and the heterogeneities (see Figure 1). The main objective of the second benchmark is to understand how gas is moving from a cell toward a drift and finally toward a drift plugs (is convection still the main process at this scale, which part of the gas generated inside the cells is moving across the drift plugs, what is the characteristic time for this transfer, what pressure can be achieved, …). A secondary objective is to study different methods (homogenization, domain decomposition, high performance computing, …) used by the different teams to achieve this simulation taking into account all the physics described below and restraining the computation time and/or the mesh size to something manageable. The complete definition of the benchmark can be found in [3]. The mathematical model in the repository involves two-phase immiscible compressible, liquid and gas phases, transient flow processes in a porous medium under isothermal conditions. For gas flow, a mixture of gases is evolved, with hydrogen being the chief constituent. In this study, all gases produced by the corrosion phenomena of metallic components are treated as hydrogen. Hydrogen dissolution in the pore water follows Henry's law; gas and water flow are governed by Darcy-Muskat's law. In this work, we advance the applicability of the vertex-centred finite volume method in heterogeneous media on unstructured grids, where we show how to account for the discontinuity in saturation from different capillary pressure functions. An upscaling technique is developed to obtain an effective capillary pressure curve at the interface of two media. Our aim is to study a fully implicit finite volume scheme for the problem where a special discretization at the interfaces is developed and present numerical simulations for the 2D benchmark at cell scale and the 3D benchmark at module scale. We have developed our C++ homemade code based on the numerical scheme presented above for the 2D case. The DuMu^x code has been adopted for the 3D study.

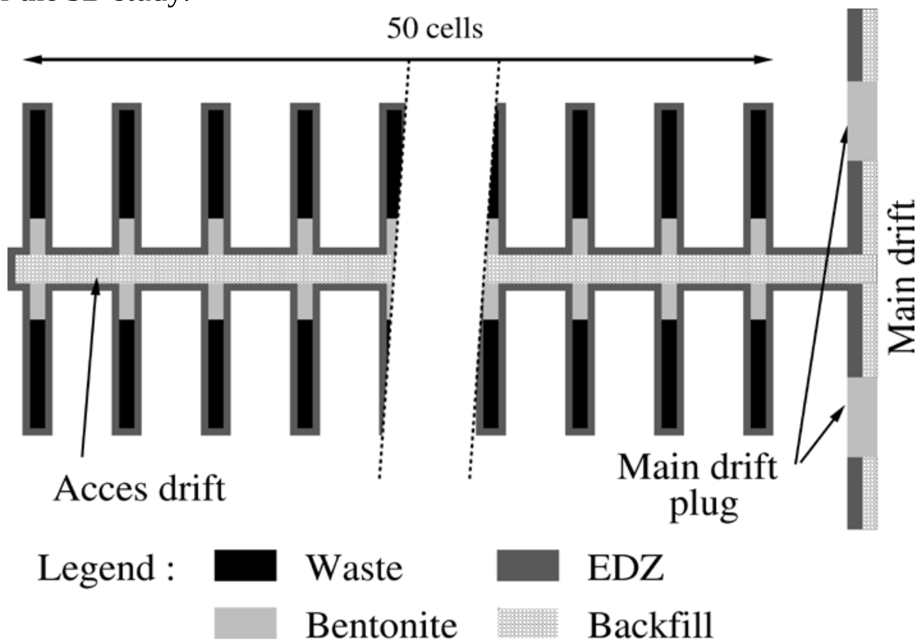

Figure 1: Schematic representation of the module to be simulated.

2. Methodology

2.1 Mathematical model used in simulations

We consider two-phase immiscible compressible flow, liquid and gas phases, in a porous medium under isothermal conditions. The index $\alpha \in \{w, g\}$ refers to the phase (water=liquid and gas=hydrogen), while the superscript $c \in \{H_2O, H_2\}$ refers to the component. K is the absolute permeability and ϕ is the porosity, $k_{r\alpha}, \mu_\alpha, \rho_\alpha, \mathbf{U}_\alpha, p_\alpha$ are respectively the relative permeability, the viscosity, the density, the velocity and the pressure of the α phase.

The Darcy-Muskat law is used as the equation for the conservation of momentum:
$\mathbf{U}_\alpha = \frac{-k_{r\alpha}(S_\alpha) K}{\mu_\alpha} (\nabla p_\alpha - \rho_\alpha \mathbf{g})$ where g is the gravity.

The mass conservation law for each component gives the following equations: $\frac{\partial}{\partial t}(\phi \sum_\alpha \rho_\alpha X_\alpha^c S_\alpha) + \sum_\alpha \nabla . (\rho_\alpha X_\alpha^c \mathbf{U}_\alpha + \mathbf{J}_\alpha^c) - \sum_\alpha Q_\alpha^c = 0$ where the sum goes over the water and the gas phases. S_α is the saturation of the α phase while X_α^c and Q_α^c are respectively the mass fraction and the source term of the component c in the α phase.

The diffusive fluxes are given by $J_\alpha^c = -\phi \rho_\alpha S_\alpha \frac{1}{\tau^2} D_\alpha^c \nabla X_\alpha^c$ where τ is the tortuosity and D_α^c the diffusion coefficient of the the component c in the α phase.

The phase pressures are connected by the capillary pressure law $P_g - P_w = P_c(S_w)$. The Van Genuchten-Mualem model is used to express the capillary pressure function and the relative permeabilities. Finally, the quantity of the dissolved hydrogen in the water is given by the Henry law. The model considers a system with two-phase initial and boundary conditions and time dependent gas sources from metal corrosion. In the repository, the hydrogen is partially dissolved in the water and partially exists as a free gas.

2.2 Numerical treatment

The second benchmark at Module scale is computationally very demanding due to the presence of the Interfaces and the EDZ, which leads to very fine grid to be used for the spatial discretization. In view of the whole repository, which contains ten modules, we have chosen to simplify the numerical representation of the module in order to reduce the size of the computational grid. For this, we use a mathematical upscaling method which replaces two or more heterogeneous materials with the so called effective material which is, in certain sense, equivalent to the heterogeneous material. This process is depicted on Figure 2, where we replace the Canister/Bentonite plug, the Interface, the EDZ and the surrounding Geological medium by a single effective material (left picture in Figure 2), and similarly we replace the materials in the Access Drift with an effective one (right picture in Figure 2). Three hierarchically upscaled models U1, U2, U3, each being more homogeneous than the preceding one, have been considered.

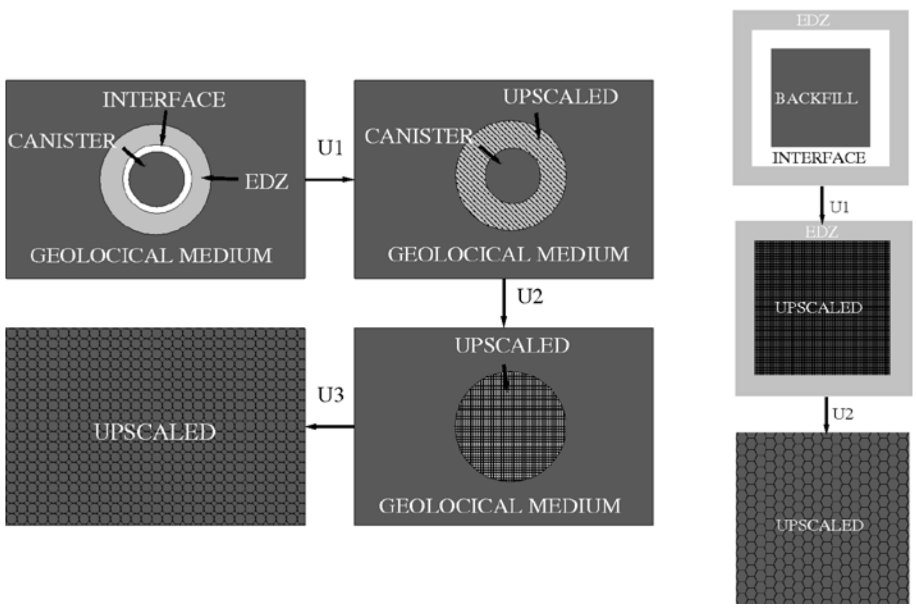

Figure 2: Different upscaling steps near canisters (left) and in the drifts (right).

In replacing the heterogeneous media by the effective, homogeneous, one we have to compute the effective properties of the homogeneous medium: porosity, permeability, capillary pressure, relative permeability functions and tortuosity. This is done by the mathematical upscaling technique which relies on steady state calculations by solving local problems in Representative Volume Elements (RVE). The heterogeneities present in the numerical model after this upscaling procedure are shown in Figure 3. Note that the Main Drift Plugs are present in the upscaled model with surrounding Interface, but the Plugs near the Canisters are completely removed by the upscaling procedure.

The simulations conducted in this study are computationally challenging because of the site-scale 3D grid and the complex nonlinear processes involved. A fully-implicit vertex-centered finite volume scheme is used for the spatial discretization, the implicit Euler scheme is applied for the temporal discretization. The simulations have been performed with DuMu^x [1]-[2]. DuMu^x is a free and open-source simulator for flow and transport processes in porous media, based on the Distributed and Unified Numerics Environment DUNE. DuMu^x has several mathematical flow models in different modules. Here we use the 2p2c model which implements a two-phase flow with two components.

3. Results

The simulation is performed on a grid with 27776 elements, see Figure 3. We will present all the requested results: Fluxes, solutions in points and solutions on lines.

Up to now, the numerical simulations carried out are for only the most homogeneous of the three models, i.e. the U3 model. Our simulations show that the maximum pressure in the module will be about 7 MPa, and that at the places where the fluxes were calculated, the convection will be the main way for the hydrogen transport. The transport of the hydrogen dissolved in water is about two orders of magnitude less significant than the transport of the gaseous hydrogen. As expected, the diffusion of dissolved gas in the water is responsible for reducing the maximum pressure in the repository for at least one MPa.

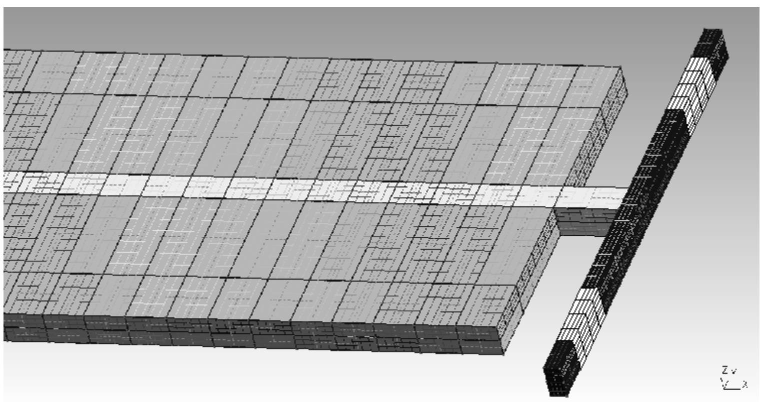

Figure 3: Grid used for U3 model.

4. Conclusions

We have presented results of numerical simulations of the module scale benchmark. Our approach for the numerical modelling includes the use of the mathematical homogenization to reduce heterogeneity of the model, thus reducing its complexity. We have build tools and procedures to build several upscaled models, with different level of small scale details; three such models are built. The simulation results have been compared to corresponding results from other teams involved in this work package and have shown reasonable match, with largest differences in calculated fluxes.

5. Acknowledgements
The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Program (FP7/2007-2011) under Grant Agreement no 230357, the FORGE project. This work was partially supported by the GnR MoMaS (PACEN/CNRS, ANDRA, BRGM,CEA, EDF, IRSN) France, their supports are gratefully acknowledged.

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'Second Order' Exploratory Data Analysis Of The Large Scale Gas Injection Test (Lasgit) Dataset, Focused Around Known Gas Migration Events

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Summary

Within large-scale experimental datasets a wealth of small scale information can typically be found. An example of such an experiment is the Large Scale Gas Injection Test (Lasgit). A toolkit has been developed to facilitate the investigation of the small scale or 'second order' detail contained within Lasgit's dataset. Results obtained through application include: quantifications of trends and seasonal effects; and second order gas transmission spurring from a primary macro scale transmission event.

1. Introduction

The Large Scale Gas Injection Test (Lasgit) is a field scale experiment run by the British Geological Survey (BGS) and is located approximately 420m underground at SKB's Äspö Hard Rock Laboratory (HRL) in Sweden. It has been designed to study the impact on safety of gas build up within a KBS-3V concept high level radioactive waste repository. Lasgit has been in almost continuous operation for approximately eight years and is still underway. Detailed information about the Lasgit experimental setup and procedures can be found in Harrington et al. (2008) [7] and Cuss et al. (2010) [5].

Lasgit is highly instrumented and frequently sampled. This has lead to a substantial dataset containing in excess of 14.7 million datum points. The data is anticipated to include a wealth of information, including many smaller scale or 'second order' features of interest that are not immediately apparent when considering the dataset as a whole. Given the magnitude of the dataset and the detail required in investigation, computational analysis is essential; however it is hampered by the non-uniform nature of the Lasgit dataset. To facilitate the second order analysis of such long-term, large-scale datasets a toolkit has been developed capable of accommodating non-uniform input [2].

2. Methodology

While specifically developed to be applied to the Lasgit dataset, the toolkit was designed to have generic applicability to long-term, large-scale time series based datasets, possibly containing non-uniformities. The resultant toolkit was applied to the Lasgit dataset both globally and locally in order to first identify and then investigate second order events.

The choice of capability incorporated into the toolkit and the development of methodologies used to achieve the chosen capability were guided by a requirement for minimisation of pre-requisite knowledge of the nature of information contained within a dataset before application. As such, the capability selection process favoured non-parametric techniques when information form is considered, and parameters related to the scales of interest to investigation (i.e. the desired second order scale) when they could not be avoided.

The toolkit's capability includes:

- Event detection sensitive to second order scale behaviour
- Detection of disparate datum points (spike detection) for QC purposes
- Frequency content analysis
- Trend detection / signal component derivation
- A range of smoothing and averaging capabilities

Modification (generalisation) of a number of established signal processing procedures (e.g. [1,3,4]) was required to accommodate non-uniform input in order to provide the capability listed above. Notably, windowing functions, typically used in smoothing functions and moving averages, were adapted from spanning a fixed number of datum points to spanning a fixed length in time and operating on the variable number of datum points hence encompassed. This allows the toolkit to be applied on a scale fixed relative to the processes of interest in the system studied, regardless of data sampling rate. Additionally a Discrete Fourier Transform (DFT) procedure was generalised to accommodate an arbitrary inspection range in the frequency domain along with the removal of reliance on a fixed input sample rate.

Event and spike detection made use of simple statistical indicators (such as local standard deviation) coupled with the time windowing approach described above. This approach removes the need for knowledge of the form of events. This approach however does only highlight fluctuations in the data, essentially highlighting 'candidates' for events of interest warranting further investigation.

Singular spectrum analysis (SSA) [6] was used for trend detection / signal component analysis due to its non-parametric estimation of form within a time series. Macro scale trend detection was primarily used to determine a long-term behaviour of the system that could subsequently be subtracted from the dataset allowing smaller scale features to appear more prominently, thus potentially exposing information otherwise obscured.

Application of the toolkit to the dataset was multi-stage, with a global application for reference and to assess the second order behaviour in the context of the macro scale behaviour, a trend removal stage, and subsequent reapplication of the second order assessment with macro obscurity removed. A number of second order event candidates were then chosen for a 'drill down', in which a more classical 'by hand', focused investigation was undertaken assisted by higher resolution output from the toolkit. The event candidates chosen for investigation were prioritised by relative prominence and proximity to known macro scale gas movement events.

3. Results

Exemplar results are presented below depicting both the general output generated by the application of the toolkit and specific discoveries made as a direct result of the information exposure achieved.

Figure 1 provides an example of typical toolkit outputs when applied to a pore-water pressure sensor installed down hole in Lasgit. Identified second order events (depicted by grey impulses in Subfigure a) are identified by the prominence of local noise in the system with time. Frequency content (Subfigure b) clearly depicts an annual (seasonal) cycle inherent in the magnitude of the sensor record of approximately ±10kPa. Subfigures c and d depicts the SSA derived trend and the residual signal after subtraction respectively.

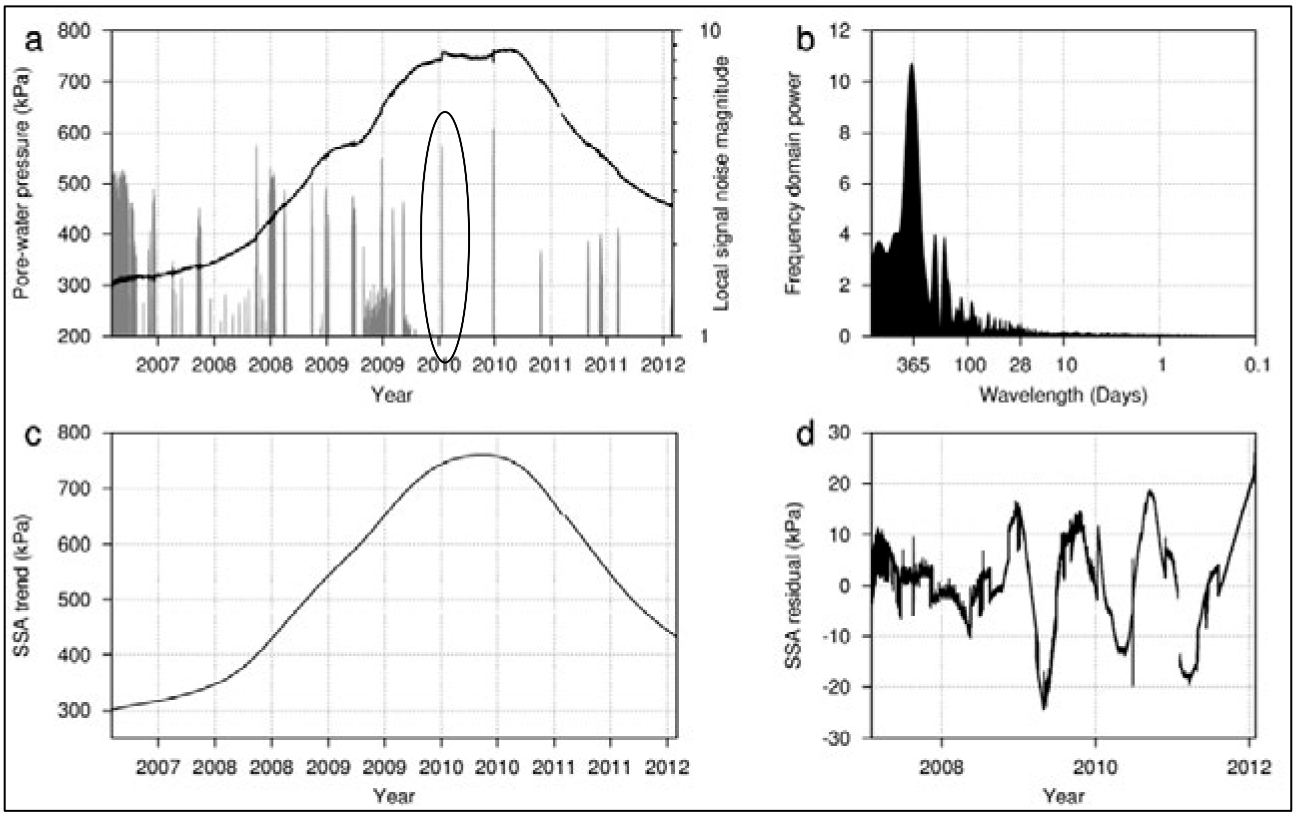

Figure 1. Example toolkit output when applied to a Lasgit pore-water pressure sensor. (a) The original time series with detected second order events depicted by impulses. (b) Frequency content of the de-trended time series showing annual (seasonal) variation. (c) and (d) the SSA derived trend and residual of the original signal respectively.

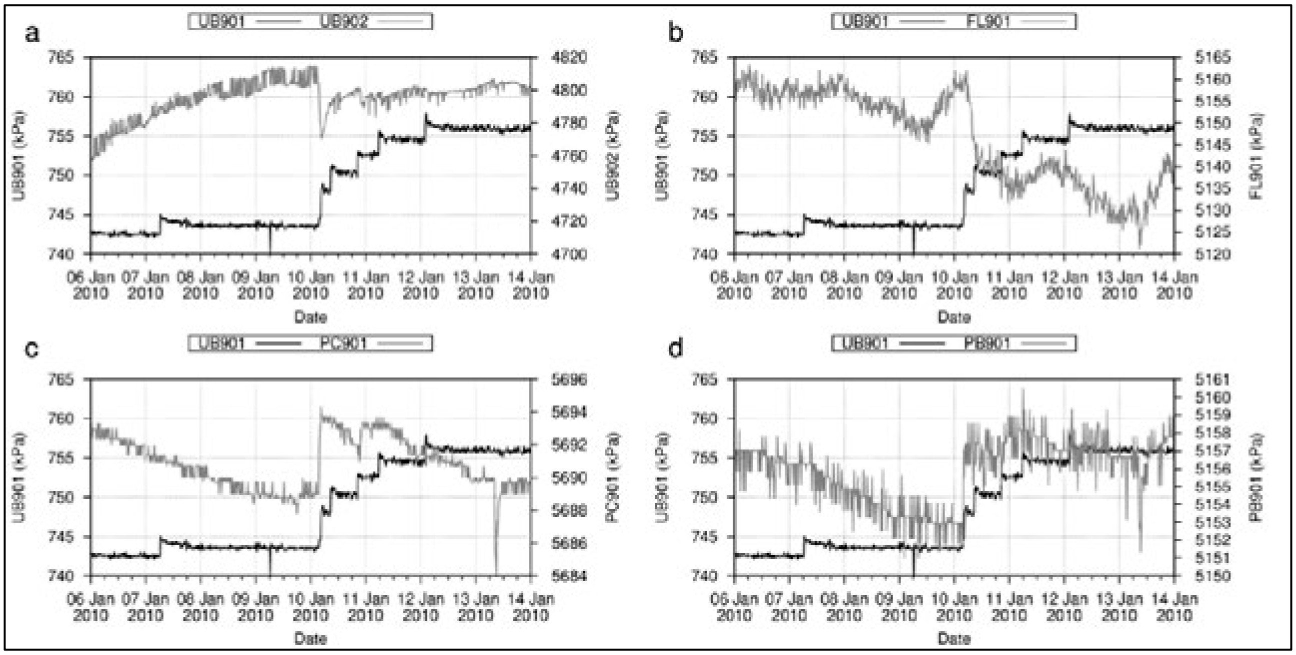

Figure 2. Sensors with second order event indicators in close proximity to the primary pore-water sensor being investigated. (a) a neighbouring pore-water pressure sensor that has experienced macro scale gas transmission. (b) The active gas injection filter. (c) and (d) nearby stress sensors.

Detailed scrutiny of the ten day period surrounding the impulse highlighted in Figure 1a is presented as an exemplar. Comparison of impulse results in the period identified four other sensors with second order events: the active gas injection filter; a neighbouring pore-water pressure sensor; and two nearby stress sensors. The records of these sensors are juxtaposed with the sensor record of the originally investigated pore-water sensor in Figure 2.

4. Discussion

The episodic pressure increase in the exemplar pore-water pressure sensor coupled with a decrease in pressure at the active gas injection filter and neighbouring pore-water pressure sensor that was previously the recipient of a macro scale gas transmission indicate a second order spurring flow has occurred from the original transmission path. The coinciding increases in stress locally indicate that dilatant flow was the mechanism for the spur. Other stress sensors in the region show no response to this event, suggesting it is a highly localised phenomenon.

5. Conclusions

The successful development of a toolkit capable of analysing a long-tern, large-scale, non-uniform dataset has been instrumental in facilitating the investigation of the Lasgit dataset. Small scale or ‘second order’ events have been identified as warranting detailed investigation with a high level of relevancy. Investigation of these events has lead to discovery of phenomenon that may otherwise be overlooked. An exemplar of such a discovery is spurring gas flow has been indentified within the Lasgit system.

6. Acknowledgements, References

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under Grant Agreement No. 230357, the FORGE project.

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FORGE Repository Scale Benchmark Modelling using T2GGM

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Summary

The FORGE benchmark modelling studies aim to model repository scale gas migration, with the objective of improving understanding of gas migration at the repository scale to support performance assessments. Modelling studies were divided into three phases: cell-scale, module-scale and repository-scale. T2GGM and T2GGM-MP were selected as the two-phase flow codes.

A 2D radial model was developed for the cell-scale, with each component of the cell explicitly represented, including a thin 1 cm interface. 3D models were developed for the module and repository scale models: the challenge in both of these models was developing a grid discretization that adequately represented the module and repository, while maintaining a tractable grid size. To obtain a working model, interfaces were upscaled, cells were converted from cylindrical to rectangular shape, and grids were nested and unstructured.

Both the cell-scale and module-scale models were strongly influenced by the boundary conditions in the drift. Within the cell-scale, the flow of gas was mainly advective along the cell EDZ and interface towards the access tunnel. Despite the presence of the more permeable interface, the EDZ transported the bulk of gas along the cell due to its greater cross-sectional area. The module-scale also found that free gas migrates along the cells toward the access drift, but only initially. Once pressures in the module begin to equilibrate with the host rock, water and gas flow directions were more complex throughout the module. When gas generation ceased at 10 000 years, a maximum gas pressure of 5.7 MPa at the cell scale, and 6.7 MPa at the module scale was observed. Module-scale results suggest that by 2000 years, bentonite seals are mostly water saturated, limiting the flow of gas through the main drift. At all three scales, model results compared well to those produced by other modelling groups (note that repository scale only compares preliminary results).

1. Introduction

As part of FORGE WP1.2, benchmark modelling studies at three scales (cell, module and repository) were conducted. The objective of the benchmark studies is to improve the understanding of gas migration at the repository scale to support performance assessments. Benchmark modelling has been conducted by several independent groups, using a range of codes.

2. Methodology

The benchmark repository (Figure 1) is located within an argillite host rock with a permeability of 10^{-20} m^2. 75 m above the repository is an aquifer with a permeability of 10^{-15} m^2. Fixed pressure (fully water saturated) boundary conditions exist 75 m below the repository (6 MPa), and at the Y = 0 and Y = 1437 m edges of the aquifer (4 MPa at Y = 0 and 4.5 MPa at Y = 1437 m). At the repository horizon, hydrostatic water pressure is approximately 5 MPa. Gas generation occurs over the first 10 000 years, at a constant rate of 100 mol of H_2/yr/cell. Within the repository, each cell is comprised of cylindrical water and gas impermeable waste surrounded by a thin 1 cm interface and 0.5 m of EDZ (Figure 1). A bentonite seal, surrounded by a thin interface and EDZ, separates the cell from the access drift. Access and main drifts are backfilled, and include interfaces and EDZ surrounding these drifts. 50 m bentonite seals (with interfaces but no EDZ) located in the main drift separate each module. A bentonite seal is also located in the shaft below the aquifer.

T2GGM (Quintessa and Geofirma, 2011 [1]), a modified version of TOUGH2 v2.0 with optional gas generation model, was selected as the two-phase flow modelling code. Standard T2GGM (without gas generation model) was used for cell-scale and module-scale modelling, while T2GGM-MP was used for repository-scale. Modifications were required to meet benchmark requirements, including addition of time-variable pressure and saturation boundary conditions, and addition of the Mualem gas relative permeability model. mView, developed by Geofirma Engineering, was used for pre- and post-processing.

A 2D radial model was developed for the cell-scale, and 3D models were developed for the module and repository scales. The greatest challenge in the module and repository scales was the inclusion of small features of the benchmark (e.g., the 1 cm interface) within a grid of tractable size. To obtain a working model, the interfaces were upscaled with adjacent elements, cells were converted from cylindrical to rectangular shapes of equivalent cross-sectional area, and grid discretization was nested and unstructured.

3. Results and Discussion

3.1 Cell-Scale

The boundary condition at the access tunnel is a strong driver in this model. It ensures flow along the cell and towards the access drift. Free gas flow occurs mainly in the cell EDZ, due to the greater cross-sectional area for flow compared to the more permeable 1 cm interface. The maximum gas pressure reached was 5.7 MPa in the bentonite plug adjacent to the canisters, at 10 000 years, the time at which gas injection stopped.

In the base case, advective flow along the cell EDZ dominates gas migration. A sensitivity case in which the diffusion coefficient was increased by one order of magnitude resulted in greater dissolution of gas, and halted migration of free phase gas towards the access drift.

3.2 Module-Scale

As in the cell-scale, the boundary condition in the main drift influenced results. Pressures and water saturations in the repository equilibrated more quickly than described in the boundary condition, and as a result, the boundary condition provided a strong gradient out of the module during gas generation. Despite this inconsistency of the boundary condition with the performance within the module, insights can be gained from this model. Also note that since the boundary conditions at both the cell and module scale are estimated for this exercise rather than based on a common repository scale model (i.e., these scales are modeled before the repository scale), these two model scales are not directly comparable.

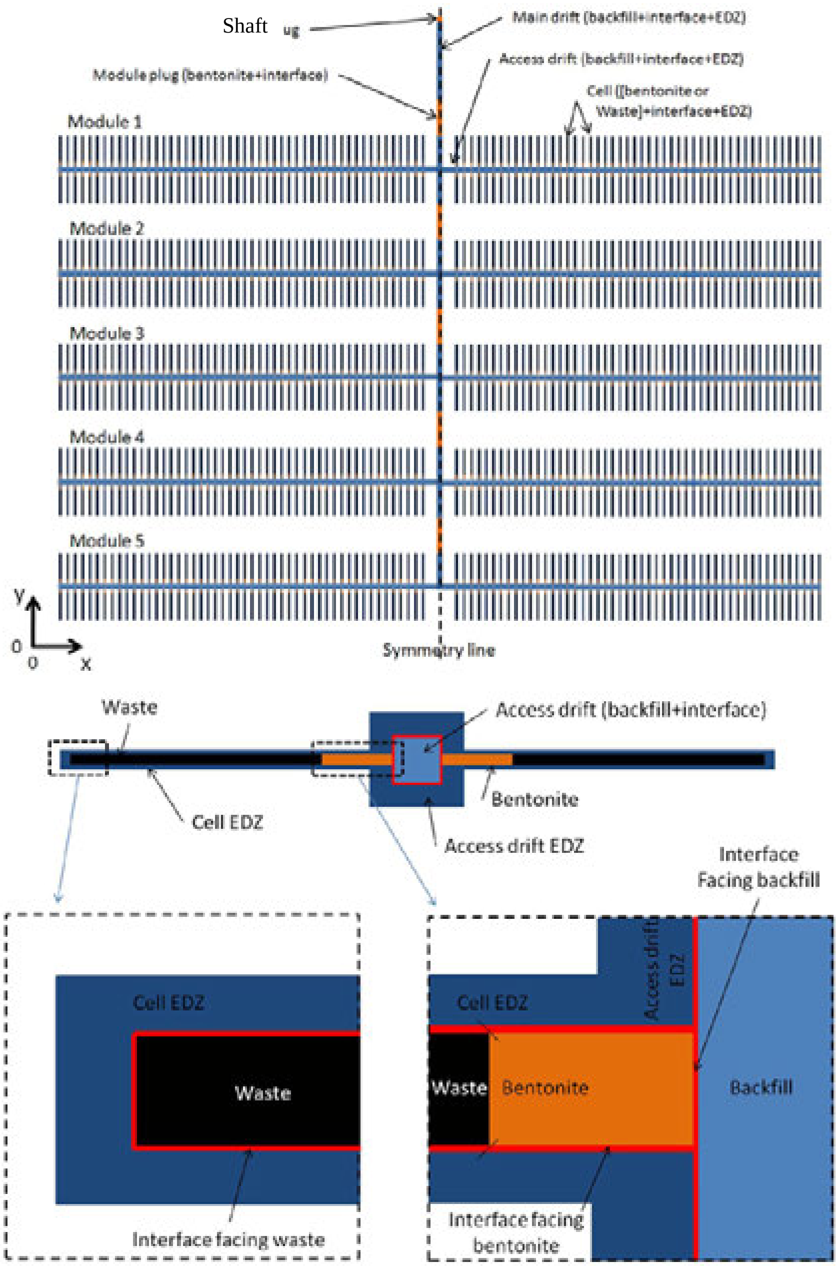

Figure 1: Schematic of benchmark repository (top) and a cell (bottom)

Bentonite seals in the main drift, which initially have high capillary pressures, are water saturated by approximately 2000 years (see Figure 2). As water saturation in the bentonite seals increases, gas pressures and saturations in the module begin to increase, until gas generation ceases at 10 000 years. The interface around the bentonite seals in the main drift plays a key role, representing three-quarters of all free gas flowing through the seals (note there is no EDZ surrounding the bentonite seals). Maximum gas flow through the bentonite seals is 3.5E-9 kg/s at 500 years.

Maximum gas pressures of 6.7 MPa are observed throughout the module cells and access drift at 10 000 years, the time at which gas generation stops. By 30 000 years, there is only a very small amount of H2 gas left in the system (0.001 kg), with effectively all of the generated gas present in a dissolved form (100 675 kg).

3.3 Repository-Scale

Two models were developed for the repository-scale: (1) a model representing the main drift, shaft and aquifer, using results from the module scale as gas and water input into the main drift; (2) a full model representing each cell and module in the repository. Work on these models is still in progress, and results are still preliminary. Results generally agree with those of the module-scale; bentonite seals in the main drift are mostly saturated by 2000 years, with no free gas migrating up through the shaft or host rock. Once bentonite seals saturate, gas pressures and saturations rise in the repository until gas generation ceases.

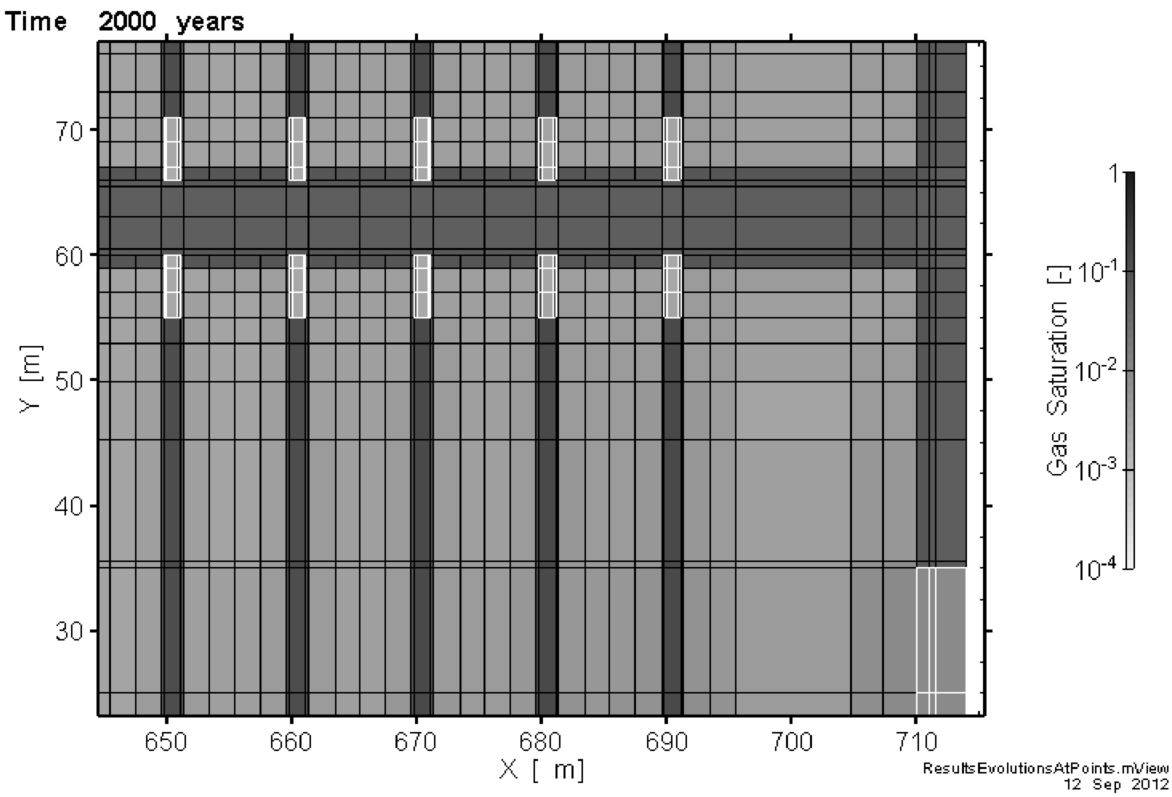

Figure 2: Gas saturations at 2000 years, near the junction of the access and main drifts, in plan view through the middle of the repository for the module-scale model. Shows grid discretization and almost fully water saturated bentonite plugs in the main drift and the end of each cell. Bentonite blocks are highlighted by white block outlines.

5. Conclusions

Each scale of the benchmark models provided insights as to the important two-phase flow processes of gas migration in a repository. The cell scale model highlights the need for well defined EDZ properties, as well as diffusion coefficients. The module and repository scales highlight the effectiveness of the repository design in minimizing gas flow out of the repository, even with interfaces surrounding bentonite seals. At all three scales, model results compared well to those produced by other modelling groups (note that repository scale compared preliminary results only).

6. Acknowledgements

This modelling work was supported by the Nuclear Waste Management Organization (NWMO), Canada.

Reference

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Hydrogen production by iron corrosion under gamma-irradiation

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Abstract

The influence of irradiation on iron corrosion was assessed through continuous monitoring of hydrogen production for 2 dose rates (50 and 100 Gy/h). Similar behaviour was observed for the two experiments: increase of hydrogen production because of water radiolysis, followed by a plateau and again a continuous increase in hydrogen production. When irradiation is stopped this acceleration of hydrogen production is maintained. The corrosion rates without irradiation deduced from these two experiments are in the range 0.1-0.4 $\mu m.y^{-1}$, i.e. consistent with values reported in literature. After irradiation the corrosion rate rises by a factor higher than 10 while no stationary state was obtained at the end of the experiments.

1 Introduction

The largest source of gas in a geological disposal facility for high level radioactive waste is the result of corrosion of iron materials (canisters, liners…). Despite the work already undertaken to study carbon steel corrosion mechanisms under various conditions, a number of uncertainties still remain concerning in particular, the effect of irradiation. Exposed to ionizing radiation, water decomposes to yield a range of chemically reactive species (oxidizing and reducing) which can strongly impact the aqueous redox conditions and influence metal corrosion kinetics. The purpose of this study was to evaluate the impact of radiation on corrosion processes of carbon steel and on the amount of gases released.

2 Methodology

Experimental setup is schematized in Fig.1. A mass close to 10 g of iron powder (particle diameter less than 210 µm) provided by Alfa-Aesar is added to a given volume (100 ml) of de-aerated water (Milli-Q quality) and introduced in a stainless steel cell. The degassing of the solution is ensured by Helium 4.5 at a fixed flow-rate (5ml/min). The gas mixture (H_2, O_2, N_2 in helium) was regularly (10 min) sampled (1 ml) and analyzed by Gas Chromatography (Varian GC 450).

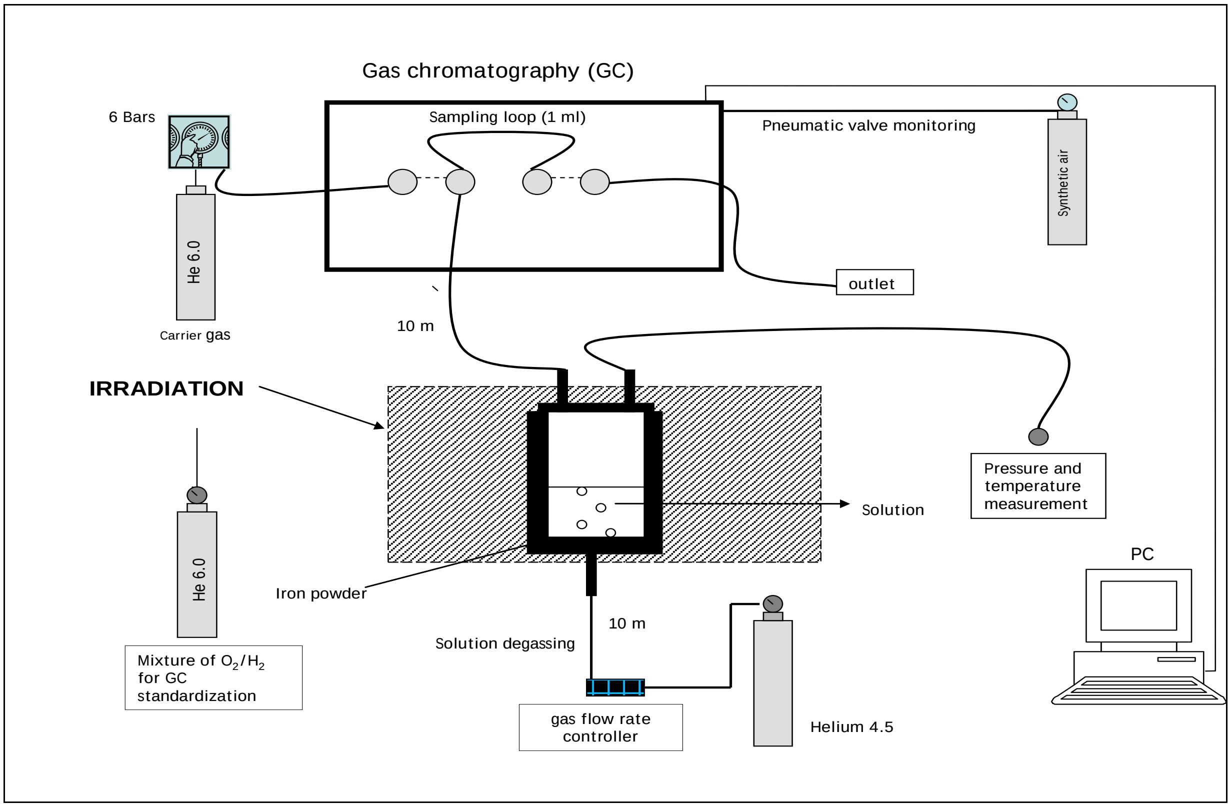

Figure 1: Experimental setup

The experiment is conducted as follows: a first step corresponds to corrosion without irradiation so as to obtain constant hydrogen production. Then irradiation is applied (step 2) for at least 3 weeks. In order to enhance the possible influence of radiation, two dose rates (50 and 100 Gy/h) were used, in two separate experiments. The optimal duration of step 2 corresponds to the time necessary to obtain a plateau (like in step 1). When irradiation is stopped, ideally hydrogen release is measured until to obtain also a plateau (step 3). Because of the availability of the irradiation facility, the experiment may have to be stopped earlier. It was the case for both experiments, which lasted at least 6 weeks, but with irradiation stopped before to reach a plateau. At the end of each experiment, the cell is dismantled for both solid and solution analyses.

3 Results

Hydrogen production expressed in mol/min versus time is plotted in Fig. 2 for the two experiments. The same shape is observed. A plateau is obtained during the corrosion phase (no radiation). As soon as irradiation is applied, hydrogen production increases sharply due to water radiolysis until to reach a plateau corresponding to the sum of hydrogen produced by corrosion and water radiolysis. For the higher dose rate, the plateau lasts 2 weeks, while for the lower dose rate, the plateau is only 2 days long. Then, an increase in hydrogen production rate is observed in both experiments; this increase continues when irradiation is stopped. No plateau was obtained before the experiments were stopped (2 weeks for the 50 Gy/h experiment and 1 week for the 100 Gy/h experiment).

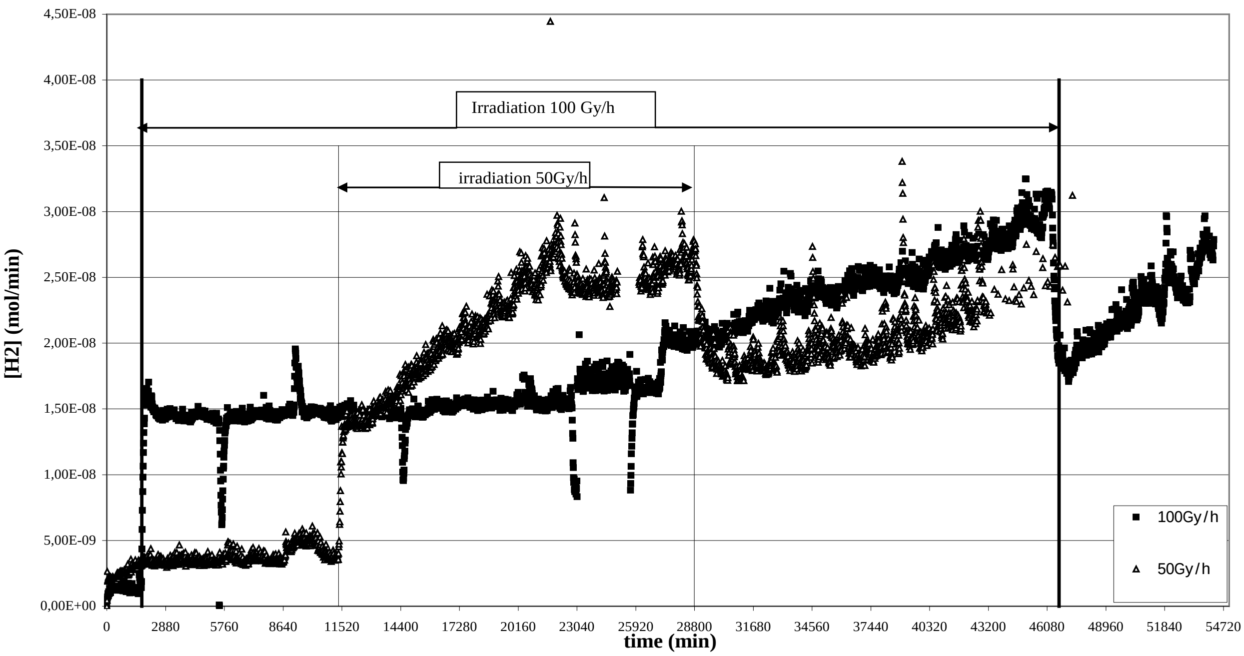

Figure 2: Hydrogen production rate (mol/min) versus time for 50 and 50 Gy/h experiments - mass of iron ~10 g; m_{H2O} ~100g

The hydrogen production rates deduced from these two experiments are gathered in Table 1 for the first step (corrosion without irradiation) and for water radiolysis.

Table 1: H_2 production rates at the different steps of the experiments

	Step 1 – Corrosion without irradiation		Step 2 - irradiation	Step 3 – irradiation stopped
Dose rate [Gy/h]	H_2 [mol/min]	Corrosion rate [µm/y]	H_2* [mol/min] (water radiolysis)	Corrosion rate [µm/y]
50	$3.81\ 10^{-9}$	0.4	$1.03\ 10^{-8}$	2.7
100	$1.37\ 10^{-9}$	0.14	$1.34\ 10^{-8}$	3.2

* determined on the plateau (corrosion under irradiation) mentioned in the text

The corrosion rates of iron powder deduced from these experiments are in agreement with those presented in literature (0.1-0.4µm.y^{-1}), but a small difference is observed between the experiments (not explained). Besides, hydrogen production rate by water radiolysis being proportional to the dose rate, a

factor 2 between the two experiments should be obtained, instead of a factor 1.3. In an aerated solution, the production of hydrogen is higher than in a de-aerated one. It is possible that some oxygen still remains in solution inducing a modification of recombination of radiolytic products (Allen chain). At the end of the two experiments, no stationary state was obtained and the corrosion rate has increased by a factor higher than 10 for the 2 experiments compared to corrosion rate determined before irradiation. At the end of experiments, pH of solutions is closed to 11 and Eh slightly reducing. No data was obtained by solid analysis (Raman spectroscopy), the duration of experiments being too small to allow the formation of a detectable corrosion layer.
The experimental data will be compared to the results of a kinetic model that incorporates reaction data set combining water radiolysis and corrosion reactions. This modelling work using ChemSimul software is still in progress and will be detailed in the conference presentation.

4 Conclusion

Considering these two experiments, it seems that corrosion processes are modified under the tested ionizing radiations, the result being a significantly enhanced production of hydrogen. This result has to be confirmed for lower dose rates more representative of disposal conditions (e.g. 10Gy/h, in progress), on the basis of longer experiments until to reach a constant hydrogen production rate (> 3 months at least). These additional experiments will also allow the characterisation of corrosion layers formed under irradiation and the comparison with those obtained without irradiation, which should help to understand the involved processes.

5 Acknowledgments

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Modelling of Localised Gas Pathways in Long-Term Gas Injection Test

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Summary

Long-term gas migration tests on Callovo-Oxfordian argillite show that gas migration is often accompanied by the development of preferential pathways, which propagate through the sample. Hydro-mechanical modelling of these tests is performed. It appears that a continuum approach to the movement of gas is not sufficient to reproduce the experimental observations. A hydro-mechanical coupling that takes into account the evolution of the permeability and the retention curve with the strains is proposed. The numerical results show that such couplings play an important role in a successful simulation of such gas flows.

7. Introduction

Over time, after closure of a deep radioactive waste repository in an argillaceous formation, steel containers will corrode, water and organic material will be irradiated. The processes lead to the generation of hydrogen gas. As such, understanding the migration of gas in the host formation and in the engineered barrier systems are important issues, which may affect the safety function of the clay barriers. Indeed some laboratory scale experiments of gas injection on clay samples exhibit complex hydro-mechanical behaviour especially for initially water saturated conditions. Laboratory tests highlight that gas entry and breakthrough are often accompanied by the development of preferential pathways, which propagate through the sample [3,5].

Such preferential pathways could not be reproduced with the classical models, based on a continuum two-phase flow approach. Initially soil heterogeneity can provide an explanation of such observations [1]. Another way to reproduce these flow instabilities numerically is to introduce an additional hydro-mechanical coupling between the permeability, the water retention curve and the strains [6]. This second option is introduced in the finite element code Lagamine to reproduce observations obtained from a new long term gas injection test performed by the British Geological Survey on Callovo-Oxfordian argillite. In this paper, a modelling of a long-term gas injection test is proposed, with a hydro-mechanical coupling that takes into account the evolution of the permeability and the retention curve with the strains.

8. Experiment description

Long-term gas migration tests have been performed by the British Geological Survey on the COx, the proposed host rock for the French repository. Helium injection occurs through the base of the sample by slowly increasing gas pressure in a series of steps (from 6.5 to 12 MPa). A backpressure of 4.5 MPa is continuously imposed at the top of the sample through the central filter and an isotropic confining stress of 12.5 MPa is maintained during the entire experiment. The argillite specimen has an initial diameter of 54.4 mm and a height of 53.9 mm. The orientation of the sample is perpendicular to the bedding plan.

Results show first a small emergent flux during the early stages of testing, before a spontaneous increase of discharge rate after 170 days, which is interpreted as evidences of major breakthrough (Fig. 1). The movement of gas is then probably through a localised network of pathways, whose properties vary temporarily and spatially within the claystone [4]. In their entirety, these observations are difficult to explain with standard porous medium flow models and suggest that a strong hydro-mechanical coupling is therefore needed to adequately explain the formation and subsequent behaviour of gas conductive pathways within the Callovo-Oxfordian argillite.

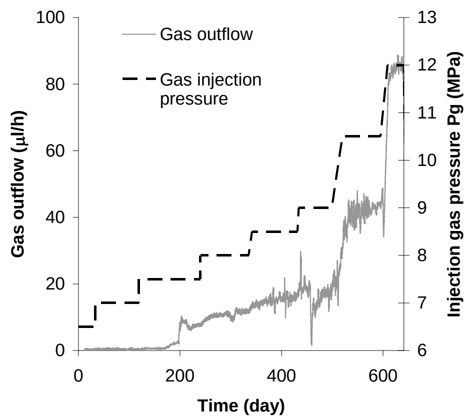


Figure 1. Experimental gas outflow and gas injection pressure evolution plotted against time.

9. Hydro-mechanical model

To reproduce water and helium transfers in partially saturated porous media, two-phase flow model is used. This model is comprised of a liquid phase, composed of liquid water and dissolved helium and a gaseous phase, which is an ideal mixture of dry helium and water vapour. It takes into account the advection of each phase using the Darcy's law and the diffusion of the components within each phase (Fick's law) (see [2] for more details). The retention curve and the water relative permeability curve are given by the van Genuchten's model (van Genuchten 1980). The gas relative permeability curve is a cubic function. The hydro-mechanical parameters for argillite are presented in Table 1. The initial anisotropy of argillite is deduced from hydraulic tests performed before the injection of gas.

Table 1. Hydraulic and mechanical parameters for argillite

k_{hor}	Horizontal intr. permeability (m²)	4×10^{-20}	E_0	Young's modulus (MPa)	3800
k_{vert}	Vertical intr. permeability (m²)	1.33×10^{-20}	ν_0	Poisson's ratio (-)	0.3
Φ	Porosity (-)	0.18	c	Cohesion (MPa)	3
m	Van Genuchten coefficient (-)	0.55	φ	Friction angle (°)	20
n	Van Genuchten coefficient (-)	1.49	b	Biot's coefficient (-)	0.6
P_r	Van Genuchten parameter (MPa)	15			

To reproduce the development of preferential gas pathways in COx, we consider that gas migration is associated with the development of preferential paths along existing or pressure-dependent discontinuities. A hydro-mechanical coupling between the pathways aperture, permeability and air entry pressure is proposed. A threshold strain corresponding to a maximal opening of the fracture is nevertheless added in comparison with the original model proposed by [6]. In the finite element code, this aperture is linked with the strains in the discontinuities.

$$K_{ij} = K_{matrix} + K_0 \left(1 + \lambda\left(\left\langle \varepsilon_n - \varepsilon_0 \right\rangle\right)\right)^3 \left(\delta_{ij} - n_j n_i\right) \tag{1}$$

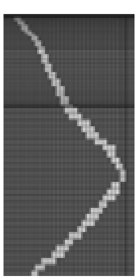
$$P_r = P_{r,0} \frac{1}{1 + \lambda\left(\left\langle \varepsilon_n - \varepsilon_0 \right\rangle\right)} \tag{2}$$

$$\text{with } \left\langle \varepsilon_n - \varepsilon_0 \right\rangle = \begin{cases} 0 & \text{si } \varepsilon_n < \varepsilon_0 \\ \varepsilon_n - \varepsilon_0 & \text{si } \varepsilon_0 \le \varepsilon_n \le \varepsilon_1 \\ \varepsilon_1 - \varepsilon_0 & \text{si } \varepsilon_n > \varepsilon_1 \end{cases} \tag{3}$$

and with ε_n the normal strain to the fracture, ε_0 the threshold strain required to initiate fracture opening, ε_0 the threshold strain corresponding to the maximal opening of the fracture, λ a parameter reflecting the density of fractures and the rugosity, K_{matrix} the permeability of the undisturbed argillite, K_0 the initial permeability along the fracture and $\underline{n}$ the normal to the fracture (considered parallel to the maximum principal strain ε_1). The mechanical model of argillite is a non-associated linear elastic-perfectly plastic model (with Van Eekelen yield surface). This model is written in terms of net stress and the mechanical parameters of argillite are presented in Table 1.

10. Numerical results

A 2D axisymetrical model of the gas injection test is performed with the finite element code Lagamine. The geometry, the initial conditions and the boundary conditions of the problem are defined in [2]. A pre-existing fracture is assumed in the sample (Fig. 2 (a)) and corresponds to the zone where the development of localised pathways is possible. This fracture is initially closed due to the high confining stress, but will be then be allowed to open as gas pressure in the sample increases.



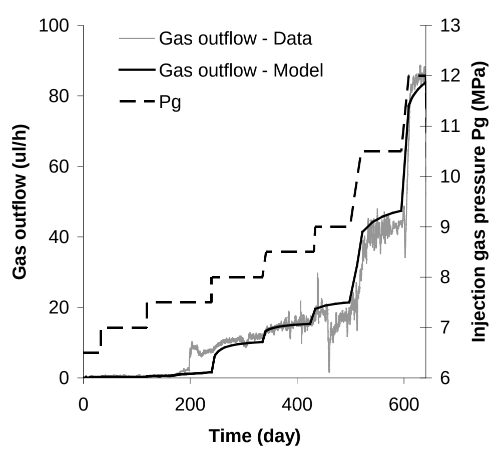


Figure 2: (a) Initial fracture in sample – (b) Gas outflow: comparison between experimental and numerical results with or without embedded fracture model.

The numerical results show that two-phase flow model allows the reproduction of the small gas outflow noted during the first stages of testing, but is not sufficient to model the strong raise of the measured outflow during the breakthrough (Fig.2 (b)). The spontaneous gas outflow observed after 170 days can be then reproduced with the proposed hydro-mechanical couplings (λ=7000; ε_0=-6.4 10^{-4}; ε_1=-4.9 10^{-4}). Pathway permeability increases and air entry pressure decreases with the extension induced by the gas injection, until the injection and backpressure platen are “gas connected”. When this “gas connection” is reached, the analysis of the numerical results shows it is necessary to consider that the aperture does not evolve any more despite the strain evolution in the discontinuities with the increase of the gas pressure. The “modified permeability” of the sample allows the reproduction of discharge rate observed during the last gas injection stages.

11. Conclusions

A 2D axisymetrical model of a gas injection test on Callovo-Oxfordian argillite has been performed. Evolution of permeability and air entry pressure is induced along a pre-existing fracture by changes in fracture aperture and its coupling with strain. The numerical results show the need for such hydro-mechanical models, in order to better reproduce the development of localised gas pathways. Moreover a maximum opening of the fracture must be introduced in the model to avoid overestimation of the discharge rate. The definition of the pre-existing fracture geometry is based on experimental evidences, but remains arbitrary. Modellings with other geometry of the pre-existing fracture have shown the ability of the model to reproduce localised pathways, but the set of parameters used must be adapted. Considering hydro-mechanical couplings must be considered as a way to interpret and explain gas migration mechanisms in claystone, but it does not consist in predictive modelling.

12. Acknowledgments

The research leading to these results has received funding from the European Atomic Energy Community’s Seventh Framework Programme (FP7/2007-2011) under Grant Agreement no230357, the FORGE project.

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The impact of elevated pore-pressures on gas flow in the buffer; experimental observations in pre-compacted Mx80 bentonite

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Summary

In the Swedish repository concept it has been suggested that generation of gas, by canister corrosion, may lead to the development of pressures sufficiently high enough to mobilise a discrete gas phase, by advection, into the bentonite buffer. We present laboratory results, examining the impact of pore-pressure cycling on gas flow behaviour in Mx80 bentonite. Such cycling is relevant in relation to likely episodes of pore-pressure deviation from expected hydrostatic conditions. The results indicate that gas entry follows conventional behaviours, in spite of previous pore-pressure cycling in Mx80 bentonite. The post-gas breakthrough observations highlight the potential for episodic and cyclic gas migration episodes in systems where there are limited sinks available and the system remains energised. Nevertheless, expansion of the test data set is required in order to further verify and better understand this phenomenon.

1. Introduction

Laboratory and field testing indicate that in order for gas entry into the buffer to occur, the applied gas pressure must exceed the total stress acting on the clay, which is a function of the pore-water pressure, Pw, and the swelling pressure, Π (Van Geet et al., 2007[1]; Harrington and Horseman, 2003[2]; Cuss et al., 2010[3]). However, laboratory studies have shown that the relationship between total stress, Pw and Π may be hysteric in nature, indicating that pore-pressure cycling has the potential to impact the bentonite in a permanent or semi-permanent fashion (Harrington and Birchall, 2007[4]). During the lifetime of a repository, a number of possible mechanisms may lead to noticeable deviations from expected hydrostatic conditions, which may be oscillatory in nature (e.g., glacial loading). As such, it is important to determine whether this phenomenon is liable to change the gas-transport characteristics of the buffer material and in what way. We present data from one of a series of laboratory tests investigating this scenario.

2. Methodology

Testing was carried out on a sample of pre-compacted Mx80 bentonite (Mx80-14), which was constrained under a constant volume boundary condition within a non-compliant stainless steel pressure vessel (Figure 1). The experimental apparatus was designed to allow the total stress exerted on the bentonite to be measured at two axial and three radial locations, whilst an adaptable filter-array geometry allowed pore-water pressure to be monitored in a number of axial and radial locations about the sample. Further detail on the configuration of the experimental apparatus is given by Graham et al., 2012[5]. The bentonite was subjected to two cycles of applied water pressure loading, before being returned to an elevated pore-water pressure condition. A gas overpressure was then applied through a central injection rod, directly into the middle of the clay sample, in order to ascertain the gas entry and flow behaviour of the clay after pore-pressure cycling.

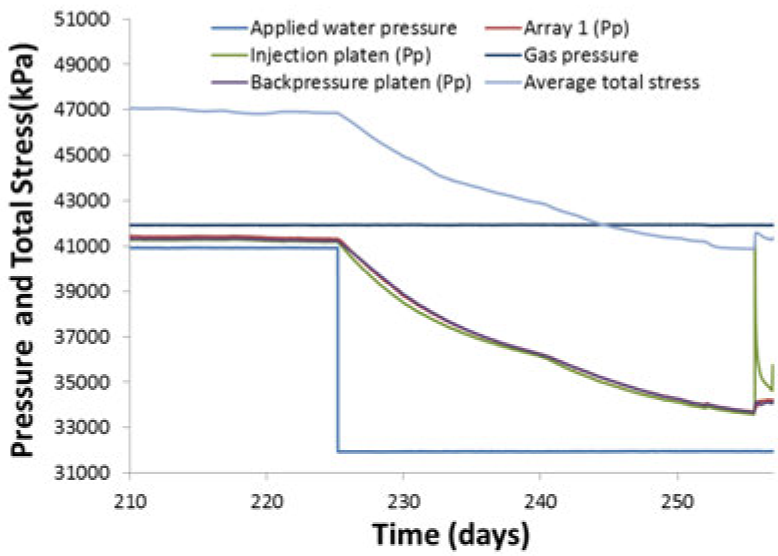

Figure 1: Gas entry was only detected after the monitored total stresses in the bentonite had fallen below the value of the applied gas pressure. Entry occurred at approximately day 260, at which time the observed pore pressure at the injection platen rose to that of the applied gas pressure.

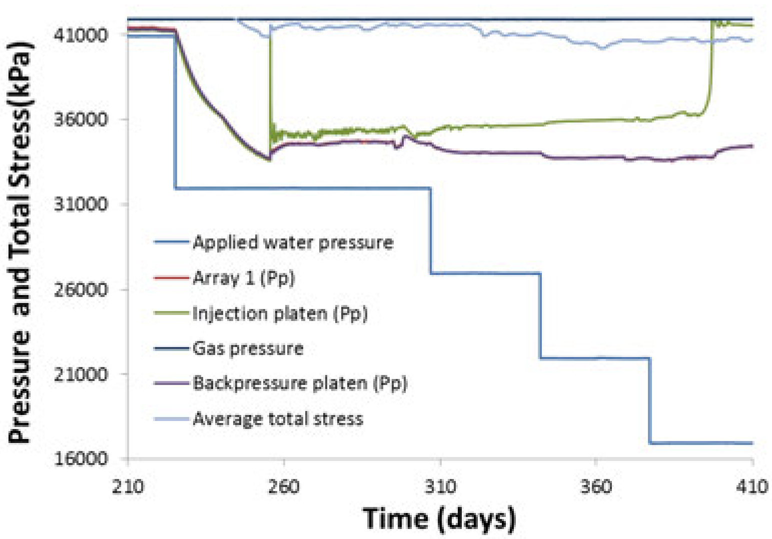

Figure 2: However, no major outflow was observed to occur over the subsequent 30 days, so the applied water pressure was successively reduced over the next 100 days. During this time a significant differential pressure was sustained by the bentonite without breakthrough occurring.

3. Results

Data from previous experiments in this test programme, gas entry has only been observed to occur once the applied gas pressure exceeds the value of the total stress within the clay (Graham et al., 2012)[5]. The results from this test indicate that this association also holds true for sample Mx80-14, in spite of previous pore-pressure cycling. This test represents an extreme situation, where elevated pore-pressures and gas flow occur with an unfavourable sink geometry, where gas cannot easily escape. In spite of this, observations indicate that gas entry still behaves following the conventional conceptual model, only occurring once total stresses have dropped below the applied gas pressure (Figure 1). Gas entry and migration through the bentonite, resulted in a series of localised perturbations in the monitored total stresses and pore-water pressures within the clay. These observations are highly indicative of gas flow through a network of pathways, which is consistent with previous findings for advective gas migration in argillaceous materials (Horseman et al., 1999[6]; Harrington and Horseman, 2003[2]; Angeli, et al. 2009[7]; Skurtveit et al. 2010[8]; Harrington et al., 2012[9]). However, the observed post-gas entry behaviour is unlike that generally reported in previous constant volume gas injection tests. The evolution of gas migration through the sample was observed for over 150 days, yet major gas breakthrough proved exceptionally difficult to instigate (Figure 2). So as to encourage major gas outflow, three steps in pore-water pressure were sequentially applied, without any notable evidence of gas breakthrough. During this time, the system was shown to sustain significant pressure gradients (up to 25MPa). Breakthrough finally occurred spontaneously (Figure 3), at approximately day 413.5, when major outflow was detected. The resulting period of gas flow was observed to be highly episodic and unstable in nature, switching from one state to another in a spontaneous fashion (Figure 3). The subsequent episodic events were often highly cyclic in nature, clearly pressure-controlled and displayed evidence of continued temporal evolution in spite of apparent self-regulation (Figure 3). These results imply that whilst the system remains energised, the gas will continue to seek a sink. As such, the path the gas takes to exit (and hence the time to breakthrough) appears highly unpredictable and potentially unique.

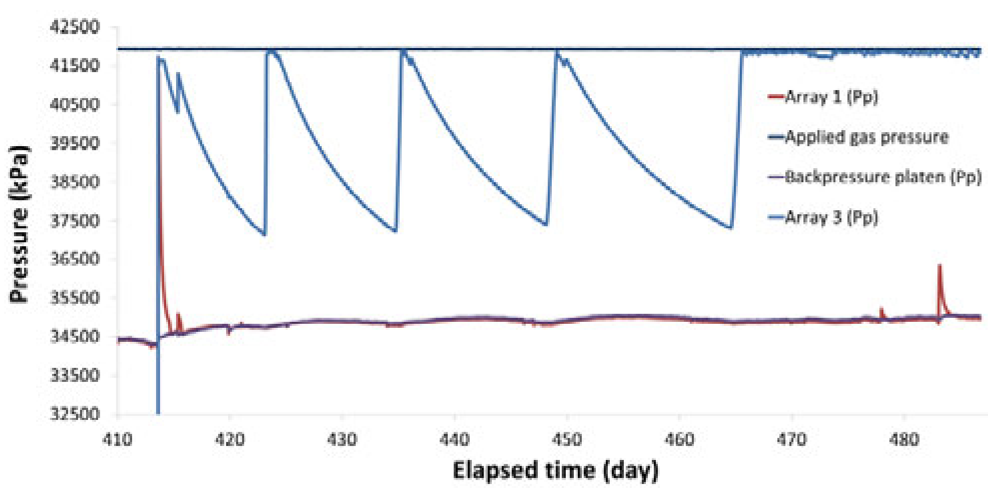

Figure 3: Gas breakthrough occurred at day 413.5, in a spontaneous fashion whilst the system was apparently unperturbed. Observed *gas migration during this phase was highly episodic, displaying extreme cyclicity combined with continued temporal evolution.*

4. Conclusions

Pore-pressure cycling was carried out on a sample of Mx80, before following this with a routine gas injection experiment. The results indicate that the gas entry behaviour of the material was unchanged by the previous episode of pore-pressure cycling, with the onset of gas entry appearing to still be relatively predictable under these conditions. However, observations of the gas flow behaviour post-breakthrough are much less typical, in terms of previously reported studies. It is possible this may be due to changes to the material during pore-pressure cycling. However, recent results from a similar study (presented elsewhere) indicate that the extreme sink geometry (with only four filters acting as potential routes for gas to escape) is more likely to have encouraged this behaviour. Evidence of time-dependent evolution during the observed cyclic episodes is suggestive of gas pressure acting to 'work' the clay before major outflow becomes possible. It is therefore possible that applying a sink geometry which is unfavourable for gas flow highlights system behaviours which are more likely in cases where gas is unable to escape the clay for significant periods of time. Nevertheless, this is a single test and requires verification and further investigation in order to fully understand system behaviour when such high gas pressure gradients are sustained over the reasonable long timescales.

5. Acknowledgements

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under Grant Agreement no230357, the FORGE project.

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Gas injection tests on a Sand Bentonite Mixture: Investigation on the effects of pore water chemistry

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Summary

This work presents the results of gas injection tests performed on fully saturated specimens of an 80/20 sand bentonite (80/20 S/B) mixture, mixed in dry condition. To study the influence of the water chemistry on the transport properties of the mixture two different types of water (demineralized water and synthetic water reproducing the in-situ composition expected for an underground nuclear waste repository) are used to saturate the specimens. The experimental results show that the breakthrough pressure is reduced and the time required for the breakthrough is shorter when the specimens were saturated with the synthetic water. This observation is attributed to the dependency of the swelling capacity of the bentonite on the water used as permeate fluid, as confirmed by complementary constrained swelling tests.

Introduction

Sand bentonite (S/B) mixtures are currently investigated as backfilling materials for cavern plugs and repository seal in the Swiss concept for nuclear waste storage of low and intermediate level (L/ILW). One important aspect regarding the performance assessment of these repositories is the consequence of the gas generated by microbial degradation of organic material and by anaerobic corrosion of the waste canister on the clay host rock. The main aim of the repository plugs and the repository seals is to increase the gas transport capacity of the backfilled underground structures without compromising the radionuclide retention capacity of the host rock as well as the engineered barrier system. Initially the generated gas is evacuated by diffusion through the pore water, then, once the gas concentration exceeds the gas solubility, the pressure increases and other transport mechanisms may appear and become predominant (two-phase flow and gas breakthrough). During the working life, the backfilling material is saturated with water coming from the surrounding host rock; therefore the chemical composition of the water may alter the transport properties of the mixture. To investigate the effect of the water chemistry on the transport properties of the selected S/B mixture gas injection tests were carried out on samples saturated with demineralized and a synthetic water which reproduces the composition of the water in situ.

Material description

The investigated material is a mixture of quartz sand and MX-80 bentonite mixed in proportion 80/20 (in dry weight).

The used bentonite is a Na bentonite composed by 84.9 % montmorillonite, 4.8 % muscovite, 3.7% quartz, 5.2 % feldspar and 1.3 % calcite. Thanks to the swelling capacity of the smectite minerals the bentonite in the mixture reduces the hydraulic conductivity and provides self-sealing capacity upon wetting. The particle density of the mixture is 2.68 g/cm^3 and it is obtained as weighted average of the particle density of the two components.
The specimens were statically compacted at a dry density of 1.5 g/cm^3 and at a water content of 11% (corresponding to the optimum water content, for a compaction energy of 3.4 J/cm^3) [1], applying a maximum compressive stress of approximately 1 MPa. Details on the chemical composition of the used synthetic water could be found in Table 2.1.

Table 2.1: Composition of the in synthetic water [3]

Compound	Mmol/kg$_{H2O}$	Mg/kg$_{H2O}$
Na	163.8	3765.7
K	2.551	99.75
Ca	11.91	477.4
Mg	9.166	222.8
Cl	169.0	5672.5
SO_4	24.00	2305.5
HCO_3	0.5431	33.14

Experimental setup and methodology

The experiments were performed under two different boundary conditions, triaxial and constant volume conditions. For the former case a standard triaxial cell was used and an isotropic stress of 300 kPa was maintained during the test. For the tests in constant volume condition, the cell consists of a ring placed between two metallic plates that are maintained together by four screws. The ring contains a cylindrical specimen which is 51 mm in diameter and 20 mm high. For both experimental setups, the bases of the specimen were in contact with two filter papers and two coarse porous stones that allowed a proper distribution of the water along the specimen surface. During the gas injection test the upstream was connected to the gas supplier and the downstream was connected to a water pressure/volume (PV) controller.
Before starting the test the specimens were saturated by injecting water at both sides by means of PV controllers. The water volume exchange was constantly monitored in this phase. Initially the water pressure was set equal to 20 kPa, for the specimen saturated with distilled water, and 5 kPa for the specimen saturated with the synthetic water. Once the specimen was saturated, the upstream was connected to the gas injection line and it was flushed with gas in order to evacuate the water from the porous stone. Then, the gas pressure was initially set equal to the water pressure and it was increased in steps while the water pressure was maintained constant. The gas pressure was controlled by means of a pressure regulator and recorded with a pressure probe whereas the mixed outflow was monitored by the PV controller connected to the downstream. The test was considered as finished once a steep increase of the outflow was observed.
Complementary tests to investigate the effects of the injected pore water composition on the swelling properties of the S/B mixture were carried out. Constrained and free swelling tests were performed on specimens compacted at different dry densities in the range of 1.3 to 1.9 g/cm^3.

Results

Examples of constrained swelling test results are depicted in Figure 4.1. The use of the synthetic pore water leaded to a reduction about 60% of the swelling pressure at the dry density of 1.5 g/cm^3.

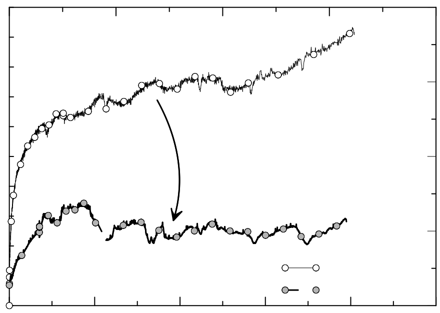

Figure 4.1: Constrained swelling tests on the S/B mixture compacted at 1.5 g/cm^3 of dry density using distilled and synthetic water

Examples of results for the gas injection tests are showed in Figure 4.2 and Figure 4.3 for distilled and synthetic water, respectively. In these figures the results are presented in three phases. Each phase corresponds to an increment in the upstream gas pressure. The outflow measured with the water PV controller showed for both tests a linear trend during phase 1. During this phase the mixed outflow was attributed to gas diffusion through a fully saturated specimen. In phase 2, the mixed outflow is much higher than in phase 1. The gas pressure in this phase was higher than the air entry of the mixture and the two phase-flow regime was established. Consequently the pore water was displaced by the gas phase and the material was progressively dried. Finally in phase 3 a steep increase in the outflow was observed. At this moment the sudden change in outflow volume was associated to an increase of the pressure in the water PV controller showing the fast passage of the gas through the specimen.

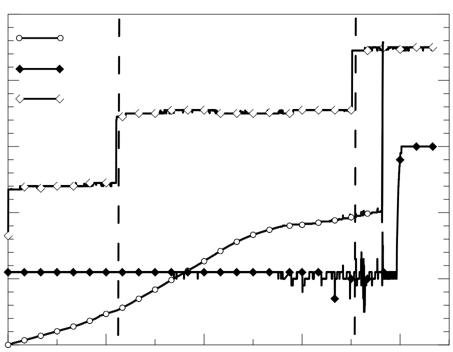

Figure 4.2 : Gas injection test in the triaxial cell at 300 kPa of confinement after saturation with demineralized water

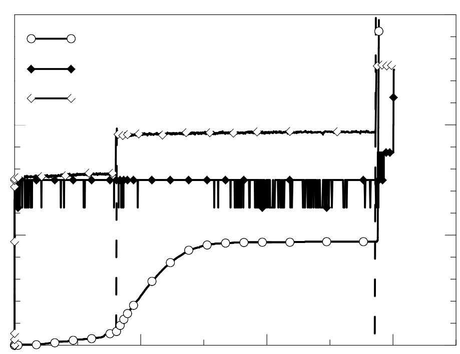

Figure 4.3 : Gas injection test in constant volume condition after saturation with synthetic water

Table 4.1: Data from gas injection test in triaxial cell at 300 kPa of confinement after saturation with in demineralized water

Phase n°	$P_{g,bot}$ (kPa)	$P_{w,top}$ (kPa)	Q (ml/h)
1	34	20	0.023
2	45	20	-
3	55	20 to 40	≈35.28

Table 4.2: Data from Gas injection test in constant volume condition after saturation with synthetic water

Phase n°	$P_{g,bot}$ (kPa)	$P_{w,top}$ (kPa)	Q (ml/h)
1	6	5-6	0.084
2	7.7	5-6	-
3	10.2	6 to 10	≈76.31

Comparing the two tests it was observed that the pressure required for the formation of the continuous gas pathway was considerably lower and that the outflow results much higher when synthetic water was used (see Table 4.1and Table 4.2). Results of the constrained swelling tests suggest that these results can be associated to the decrease of the swelling capacity of the bentonite when it was in contact with the synthetic water (Fig. 4.1).

Conclusions

The effects of the pore water chemistry on the breakthrough pressure of a S/B mixture has been investigated by means of gas injection tests carried out on specimen saturated using both distilled and synthetic water. The result shows a reduction of gas breakthrough pressure for the specimen saturated with the in-situ water. This result was explained by the reduction of the swelling capacity of the bentonite in contact with this type of water rather than the distilled water.

Acknowledgments

The authors wish to acknowledge the collaboration of Dr. Jörg Rüedi (NAGRA) and the funding from the European Atomic Energy Community’s Seventh Framework Programme (FP7/2007-2011) under Grant Agreement n° 230357, the FORGE project.

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Laboratory Gas Injection Tests and Modeling on Compacted Bentonite Buffer for TRU Waste Disposal in Japan

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Summary

Radioactive Waste Management Funding and Research Center (RWMC) is leading a research program to evaluate the gas transport mechanisms through a TRU waste disposal facility in Japan and acquire information on gas migration properties. In this paper, we carried out a series of laboratory gas injection tests together with a validation study using the test results with an advanced two-phase flow analysis code which can consider moisture dependent swelling-shrinkage properties of bentonite. In the validation study, significant hysteresis in the two-phase flow parameters was identified. Thanks to the outcome of this research project, advanced knowledge in the conceptualisation of TRU waste disposal is expected.

1. Introduction

Radioactive Waste Management Funding and Research Center (RWMC) is leading a research program devoted to study the gas transport mechanisms through a TRU* waste disposal facility in Japan. One of the crucial issues in this research is the acquisition of fundamental data on the bentonite buffer material (Kunigel V1) planned to be used in Japan.
In previous researches, conventional large-scale gas migration tests under realistic site conditions [1] were carried out for the purpose of understanding relevant gas migration phenomena. However, there are few examples of gas injection tests in the existing international literature.
In order to evaluate the mechanisms and acquire information on gas migration properties, we performed gas injection tests using small specimens of bentonite together with a validation study using these test results in a two-phase flow analysis code.
* TRU (TRans-Uranium) waste: LLW generated reprocessing and MOX fabrication. “TRU” comes from transuranium elements, i.e. elements with atomic numbers higher than that of uranium [2].

2. Methodology

Our gas migration experiments consider the situation where the buffer material is resaturated from the host rock, followed by gas generation from radioactive wastes and packaging after disposal in the TRU waste disposal facility. Figure 1 shows the scheme of a typical TRU waste disposal in Japan [3]. The experiment consisted of saturation of the sample followed by gas injection when the target saturation state was reached. Figure 2 shows the experimental setup including the injection points for water & gas and discharge points for the gas/water paths (outer and center) on top. The specifications of the specimen are shown in Table 1. As the saturation of the bentonite sample requires a significant time and only limited time was available for these experiments, we used a target saturation of 90% to shorten the saturation time. We then tried to reproduce the test results using an inverse analysis.

3. Results

Experiments were performed for two different sample heights: 25 and 50mm. A minimum of three tests were performed for each sample height. No significant differences were found between tests or sample heights and the results presented here all relate to a single test that shows a response that is typical of the observations from all the tests.
Results from the water saturation are shown in Figure 3. Figure 4 and 5 show the accumulated water volume and gas flux from the outlet in the gas injection test. The gas breakthrough was observed when the inlet gas pressure reached to 1.62MPa during the gas injection test. We observed that the main gas flow followed the lateral interface between the sample and the equipment. Unexpectedly gas was only expelled at the side of the top interface. An advanced two-phase flow code which can consider the moisture dependent swelling-shrinkage properties of bentonite was used in the modeling of this experiment. Through an inverse analysis, a significant hysteresis in the two phase flow parameters was identified (shown in Figures 6-8). An excellent match was attained between the measured and simulated responses to the breakthrough phenomenon as shown in Figures 3-5.

4. Conclusion

We conducted hydration and gas migration experiments on compacted Kunigel V1 bentonite and performed a modeling study using the test data in an advanced two-phase flow analysis. The test results have provided significant basic data to support understanding of the gas migration mechanisms through bentonite buffer material in a TRU waste disposal facility. The hysteresis in the two phase flow parameters was identified through the validation study.

Acknowledgements

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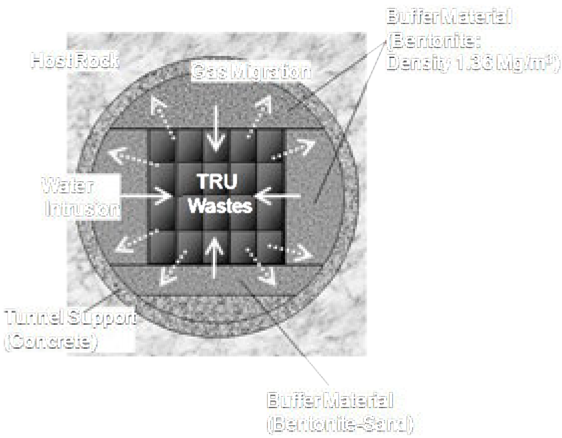

Figure 1: Considered gas migration in the concept of TRU waste disposal in Japan

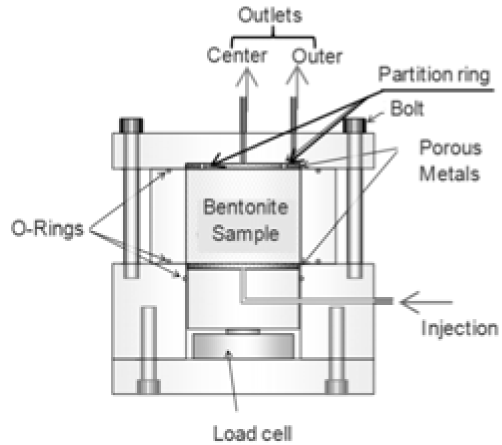

Figure 2: Outline of sample container

Table 1: Specifications of the specimen

Material	Kunigel V1
Dry density	1.36 Mg/m^3
Diameter	60 mm
Height	25 mm, 50 mm
Target saturation ratio	90 %

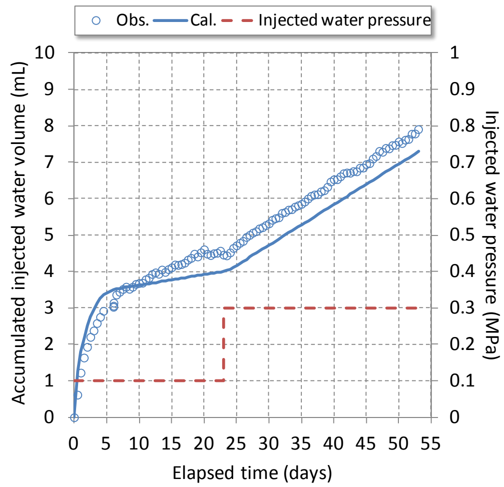

Figure 3: Comparison between measured and simulated injected water volume. Injected water pressure (hydration process) also shown

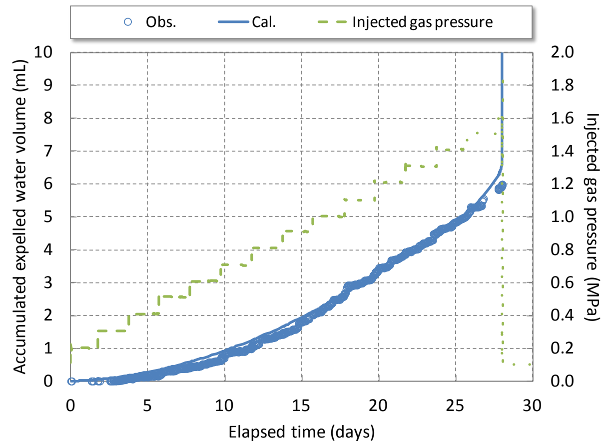

Figure 4: Comparison between measured and simulated expelled water volume during gas injection.Gas injection pressure also shown.

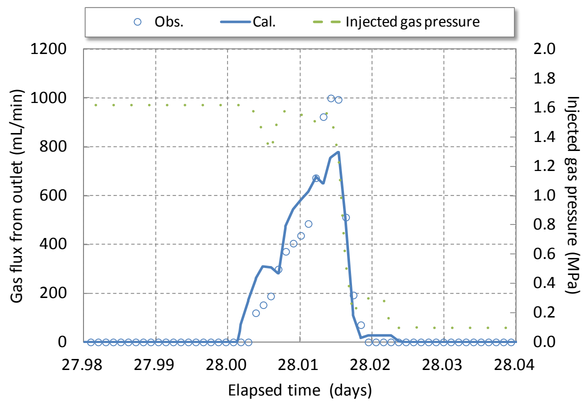

Figure 5: Comparison between measured and simulated gas flux around the breakthrough

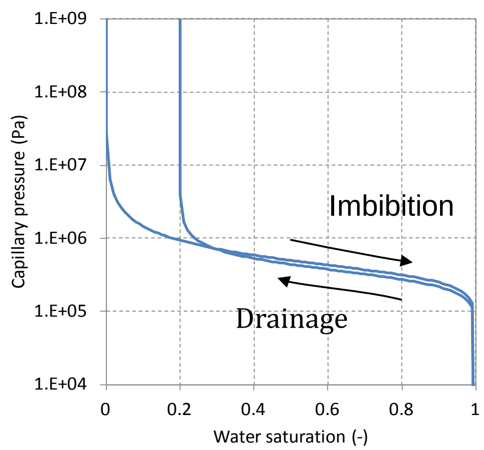

Figure 6: Identified hysteresis in capillary pressure

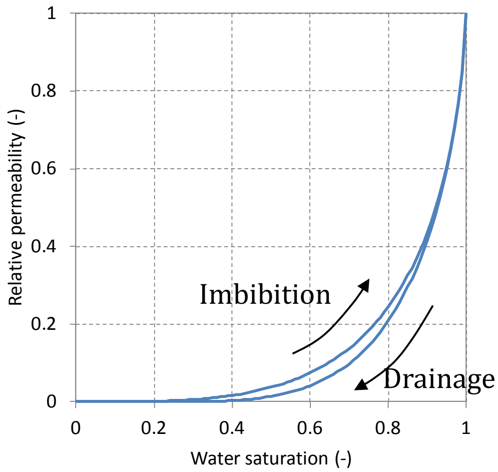

Figure 7: Identified hysteresis in water phase relative permeability

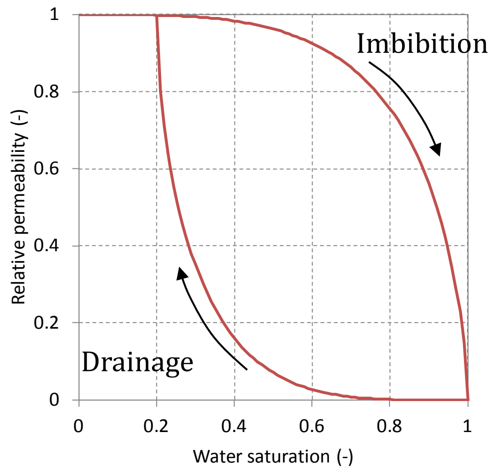

Figure 8: Identified hysteresis in gas phase relative permeability

Modelling Gas and Water Flow Through Dilating Pathways in Clay Stone: The HG-C and HG-D Experiments

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GRS, Germany

Summary

In the FORGE project, the two long-term water and gas injection tests HG-C and HG-D, which have been performed in the Opalinus Clay of the Mont Terri Rock Laboratory, Switzerland, were analysed by numerical modelling. For this purpose, continuum models based on the code TOUGH2 as well as so called “tube-chamber models” with analytic flow calculations were used. The present modelling study succeeded to reproduce the injection flows of the HG-C and HG-D experiments with high accuracy.

1. Introduction

In order to study the process of gas migration in Opalinus Clay, Nagra has conducted several long-term in-situ tests at the Mont Terri Rock Laboratory, Switzerland. From 1999 to 2012, four experiments have been carried out at the same site: The GP-A, GS, HG-C, and HG-D experiments, which have used an array of four to five boreholes for fluid injection and monitoring. In the GP-A experiment, a hydraulic fracture has been created. This fracture has been tested in the following GS experiment. From 2006 to 2009, the fracture has been re-tested in the HG-C experiment [1-3]. In order to investigate the behaviour of the undisturbed clay, a new borehole has been drilled for the HG-D experiment, approximately 1 metre below the expected location of the GP-A fracture. From 2009 to 2012, liquid and gas (nitrogen) injection tests have been performed using the new bore hole.

In order to simulate the HG-C and HG-D experiments by numerical modelling we used continuum models on the basis of TOUGH2 as well as new models (termed “tube-chamber models”) with analytic flow calculations. This paper will focus on the pathway dilation model and on the gas injection phase of the HG-C experiment. For the tube-chamber model analysis, the HG-D experiment and the water injection tests see [4].

2. Methodology and Results

Figure 1 (left) shows the injection pressure and injection flow during the gas injection phase of HG-C. According to the figure the main gas entry event started at day 147 with a precursory gas flow, which set on at day 100. This indicates that the initial threshold pressure for the main gas entry ranged between 2.34 MPa and 3 MPa at the site. Figure 1 (left) shows that there was still a positive injection flow at day 225 although the injection pressure had already dropped to 1.57 MPa, i.e. below the initial threshold for gas entry.

In the project, models with pressure dependent permeability, porosity and gas entry threshold were tested. Yet, none of these models was capable of creating and maintaining a low pressure regime in the rock, which would allow for the observed long lasting injection flow. This suggested the presence of an additional mechanism for pressure reduction inside the rock.

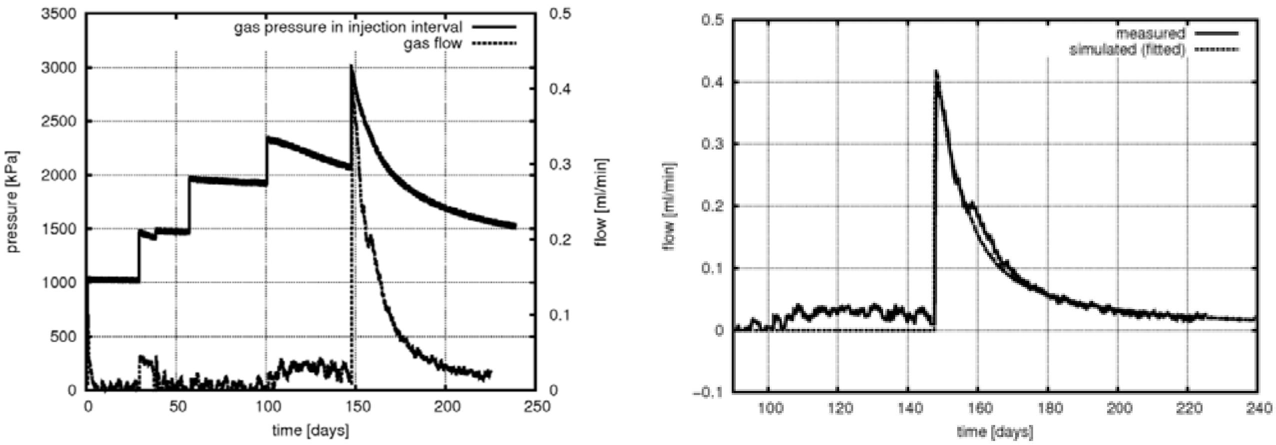

Figure 1: *Gas injection phase of the HG-C experiment. Left: Measured injection pressures and injection flows. Right: Measured and simulated injection flow.*

These considerations gave rise to the formulation of a pathway dilation model which introduces a pressure dependent rate of porosity change ("dilation rate"). With a positive dilation rate pressures are successively reduced in the rock due to an expansion of the contained gas phase. This allows the uptake of gas over long periods of time.

In the conceptual model it is assumed that micro cracks open inside the rock if the local pore pressure exceeds a certain pressure threshold. The aperture of the cracks depends on pore pressure. The evolving crack network is divided into main flow paths and dead end branches (see Fig. 2). The opening of dead end branches shall be a time-dependent process reflecting a time-dependent relaxation of the material. The mean porosity therefore increases with time. The macroscopic permeability shall be controlled by those parts of the crack network that constitute the main flow paths. Since the number of main flow paths shall not increase with time and crack apertures are assumed to be in equilibrium with pressure the macroscopic permeability is only pressure-dependent but not explicitly time-dependent. This means that permeability and porosity are independent.

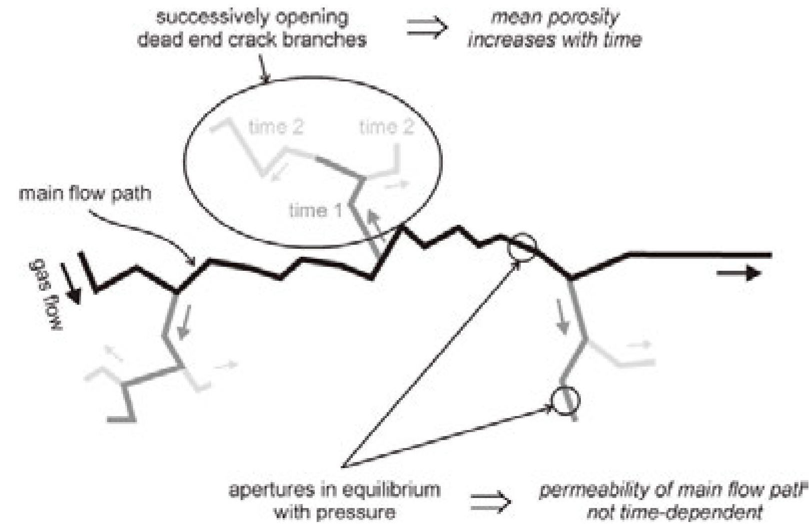

Figure 2: *Conceptual model of the dilation process. – Rock dilation is caused by a pressure induced opening of crack networks (black denote main flow paths, grey lines denote dead end crack branches).*

The model is characterised by the following features:

- There is a threshold pressure p_{thr} below which the main flow paths are closed. This implies that the crack network's permeability vanishes for pore pressures below the threshold pressure.
- For pore pressures $p > p_{\mathrm{thr}}$, the pressure threshold p_{thr} decreases from an initial value p_{thr}^{0} to a lower limit $p_{\mathrm{thr}}^{\mathrm{min}}$ with increasing pressure p (irreversible softening against microscopic tensile failure).
- Gas permeability is anisotropic and increases linearly with pressure difference $p - p_{\mathrm{thr}}$. There is no gas permeability if $p < p_{\mathrm{thr}}$.
- Porosity is variable and time-dependent. Porosity change is governed by a "dilation rate" $\frac{d\phi_{\mathrm{dil}}}{dt}$, which increases linearly with $p - p_{\mathrm{thr}}$ and becomes zero for pressures below the current threshold pressure. This implies that, except for $p \leq p_{\mathrm{thr}}$, porosity grows steadily. Yet, the system equilibrates because the gas phase inside the pores expands due to the dilation of the rock. This causes a decrease of pore pressures. This process continues until pore pressures reach the threshold pressure p_{thr} thereby stopping the dilation process.

A positive dilation rate reflects a time-dependent damage of the rock on the micro-scale. The model is only applicable for a low degree of damage and crack connectivity so that dead end crack branches can still exist. This should be reflected in porosities remaining well below 1.

Probably most of the clay's pore water is not in direct contact with the crack network. In order to avoid overestimation of phase interactions in the standard model, water was immobilized by setting the permeability of the original pore space to zero. A gas flow therefore only establishes by pathway dilation. To eliminate gas storage by processes other than pathway dilation the density of water was kept constant and the solubility of the gas component in the liquid phase was set to zero. Herewith neither water compression nor gas dissolution can take place. (The consequences of these processes were also investigated.) Thus, the system virtually is a one-phase flow system.

The code TOUGH2 of the Lawrence Berkeley National Laboratory, California, USA, was extended to implement the model. Figure 1 (right) shows that the model reproduced the injection flow very well.

3. Conclusions

The present modelling study succeeded to reproduce the injection flows of the HG-C and HG-D experiments with high accuracy. For this purpose, continuum models based on the code TOUGH2 as well as tube-chamber models with analytic flow calculations were used. The pathway dilation model implemented a time-dependent relaxation process. The fact that both, the pathway dilation model and the tube-chamber model, reproduced the data shows that there is no unique physical interpretation of the gas injection phase of the HG-C experiment.

The applied models showed that the mechanical processes in the experiments were simple enough to be approximated by time- and pressure-dependent relations for porosity and permeability. This shows that coupled hydro-mechanical modelling is not always mandatory for processes with hydro-mechanical interactions.

This study failed to fit the measured injection flow if phase interactions like water displacement, water compression or gas dissolution were taken into account. This indicates that the interface between gas and liquid phase was small in the experiment, which again suggests a high localisation of gas pathways. Initial thresholds for gas injection roughly agreed with the minimum principal stress for disturbed rock. This and the non-linear character of the flow response indicate the presence of dilating pathways. Shut-in thresholds were lower than the initial thresholds for gas injection indicating hysteretic processes.

In order to inject water into the saturated system the injection pressure has to be considerably higher than the minimum principal stress. This phenomenon cannot be explained by viscosity effects alone. Water seemed to be unable to activate fissures aligned normal to the axis of minimum principal stress. Possibly, the apertures of such fissures were too small so that water adsorption at mineral surfaces became significant and advective flow was impeded. Due to the specific pore structure of the clay it probably was easier to establish a water flow along the bedding planes than across the bedding planes. This may explain why water apparently preferred to open fractures parallel to the bedding planes (at pressures near to the normal stress on these planes). The difficulty to inject water might decrease if injection pressures are raised more slowly, as it would be the case in a repository. Although HG-C and HG-D are long term experiments, pressurisation has been performed quickly. Slow pressurisation could lead to a different water mobility of the clay.

The two-phase flow theory in its pure form does not seem to be appropriate to describe the observed experimental phenomena. Especially, the noticed lack of phase interactions conflicts with this theory. In the two-phase flow theory, gas and liquid share the same pore space which means that water has to be displaced in order to inject gas into a saturated rock. Also, phase interactions are more intense because gas flow is less localised. The concept of intrinsic permeability, which is part of the two-phase flow theory and which postulates phase independent filter characteristics of the rock, becomes meaningless if different pore spaces are used for liquid and gas flow.

4. Acknowledgements, References

This study was funded in the frameworks of the EU project FORGE and the project 3609R03210 of the German *Federal Ministry for the Environment, Nature Conservation and Nuclear Safety*. The author wants to thank *Nagra* for the provision of experimental data and internal reports. Special thanks to George William Lanyon and Stephan Hotzel for valuable comments and ample support.

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Modelling Lasgit Experiment

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Summary

Gas flow through barriers is a challenging problem because hydromechanical processes take place. This paper presents a modelling work carried out to understand the Lasgit in situ experiment which is intended at investigating the way that gas may escape from a corroded canister towards the rock migrating through a bentonite barrier.

1. Introduction

A large scale gas injection test (Lasgit) is carried out at the Äspö Hard Rock Laboratory in Sweden. Lasgit is the first demonstration project designed to study gas migration in bentonite under full-scale repository conditions. The objective of this experimental programme is to provide data to improve process understanding and test/validate modelling approaches, which might be used in performance assessment. Specific objectives are: (1) perform and interpret a large-scale gas injection test based on the KBS-3 repository design concept, (2) examine issues relating to up-scaling and its effect on gas movement and buffer performance, (3) provide additional information on the process of gas migration, and (4) provide high-quality test data to test/validate modelling approaches.

2. Methodology

Figure 1 shows a schematic representation of the test which among other elements contains a bentonite buffer material constructed using manufactured block rings. The test has a number of injection possibilities, some of them essentially intended for hydration (mat filters above and below canister) and other intended for gas injection (canister wall point sources).

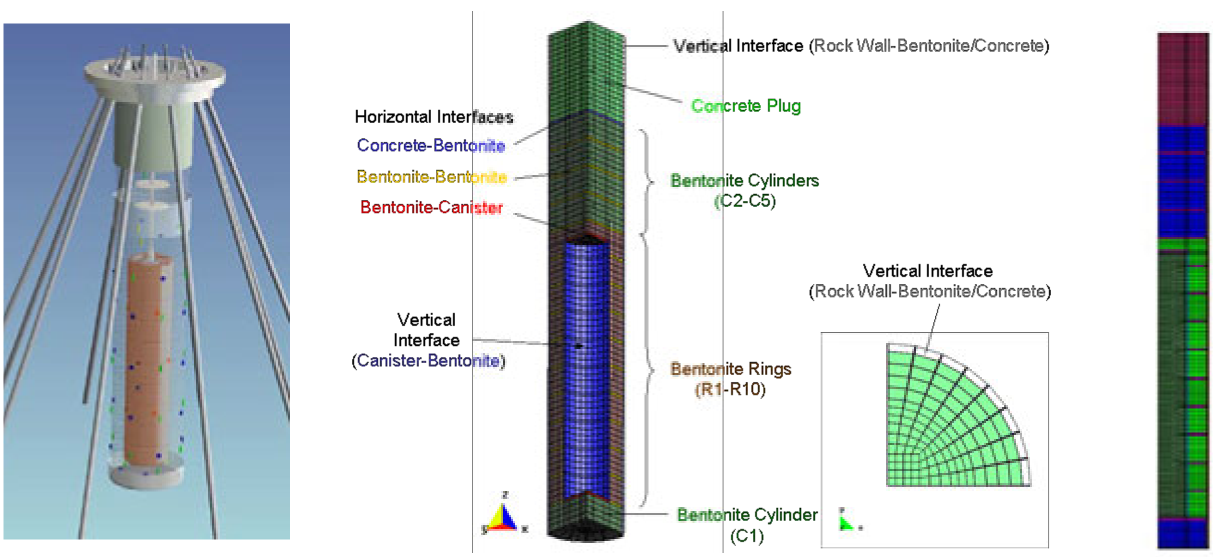

Figure 1. Schematic representation, 3D model and 2D model domains.

The modelling work is significantly complex as the water injection phases are extremely fast and thus produce high velocities of water through the interfaces. In a real repository, the interfaces are expected to close when the bentonite expands. However, in a fast test if water penetrates before the saturation of the rings, the opposite effect may be produced, i.e. interfaces opening by pore pressure development.

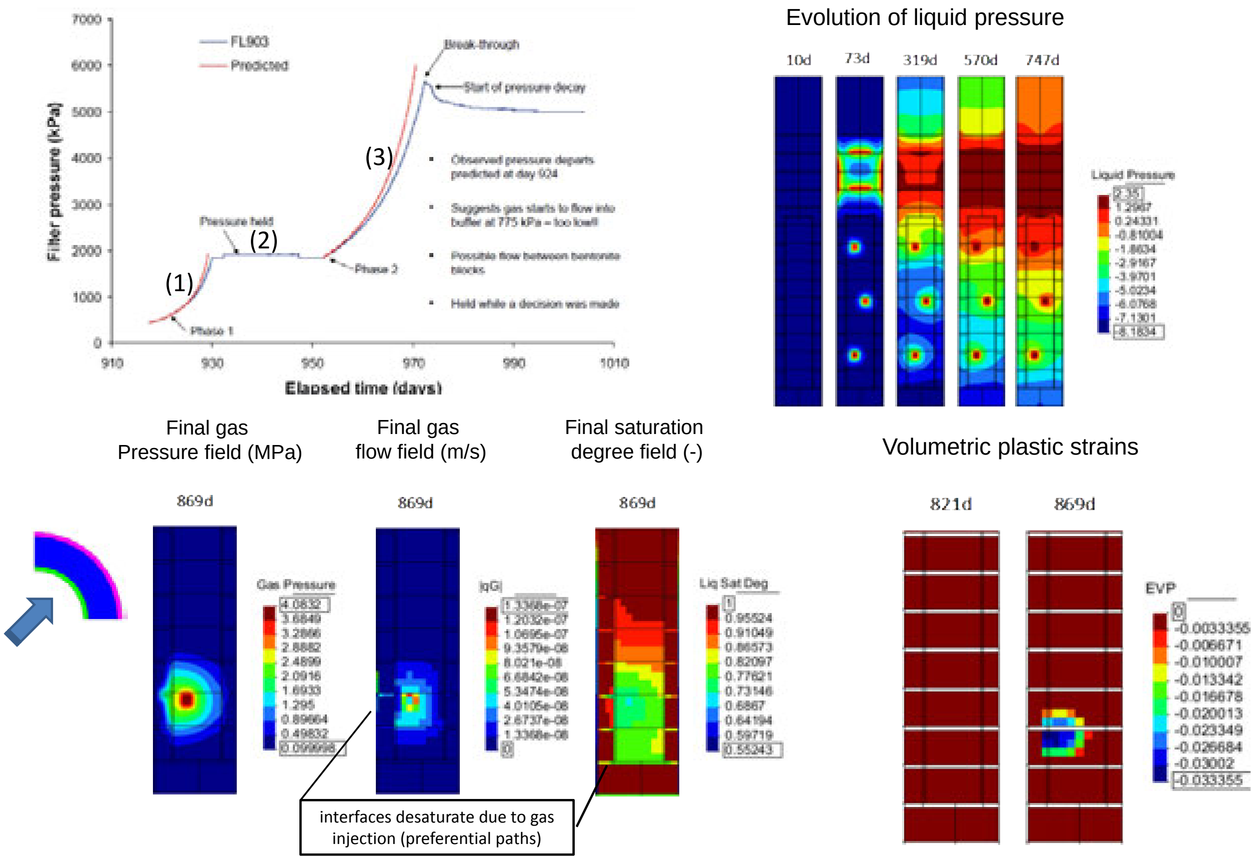

Figure 2. Injection protocol, water pressure for different times, final state after gas injection of different variables.

The modelling of this type of in situ experiments requires the incorporation of a constitutive model that is able to reproduce swelling induced by hydration of the buffer. The installation of the bentonite rings is done in such a way that the contact between the clay blocks and the canister remains as a discontinuity or interface. A similar situation happens on the contact surface between bentonite blocks. It has been observed that under natural hydration (i.e. experiments that do not use forced hydration), these interfaces tend to close as hydration and swelling of the bentonite progresses. Normally, the interface sealing would happen and therefore special treatment of interfaces would perhaps not be necessary. Later, when gas generation by corrosion was generated, these interfaces would play a role and hence they cannot be neglected in a model. The special case of modelling an experiment forces to include the interfaces. The necessity of including the interfaces explicitly appears both for the short term during water injection and for the long term as gas is generated. Even with forced hydration the interfaces manage to close, especially if the bentonite has a high swelling capacity. In case of mixtures of bentonite and sand, the interface closure during an experiment may be compromised as swelling capacity is lower. In any case, gas injection and migration is likely to be controlled by the presence of these interfaces. Therefore modelling of this kind of tests, especially if the water hydration is made very fast, requires the incorporation of interfaces and will improve the capabilities of the model.

The present approach considers interfaces as a zone of a given thickness which contains a so called "embedded" discontinuity model. This is necessary to model the penetration of water during the phase of hydration and later, the pressure responses induced by gas generation. The pressure response at sensors indicates that preferential paths through the discontinuities develop.

3. Results

Figure 2 shows the results obtained using the 3D model. Water distribution is first shown for different times. After hydration, the porous elements are not fully saturated in this calculation. The gas injection produces development of plastic strains around the zone of injection. This is due to effective stress in tensile conditions as the plastic strains are of dilatancy. This tendency to dilatancy, will produce permeability increments, and if concentrated in contacts, the permeability variations may be dramatic, thus permiting the gas to migrate towards the access drift and surrounding rock.

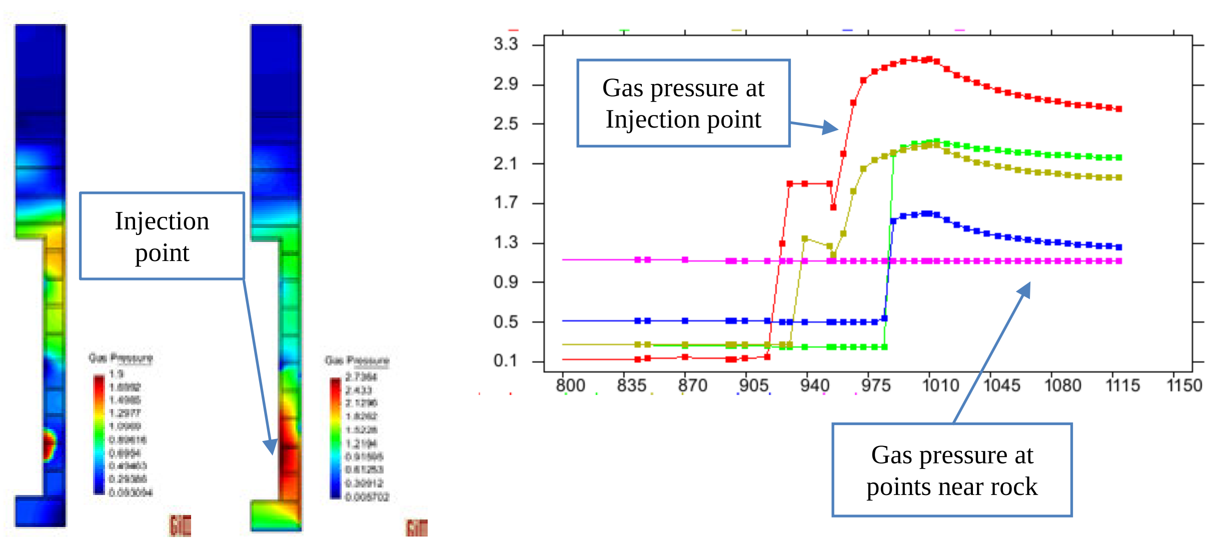

Figure 3. Gas pressure distribution during gas injection and gas pressure evolution at various points (axisymmetric model).

A possible simplification of the model consists in assuming axisymmetry. With this assumption, the model is simpler and permits to do faster calculations and undertake a sensitivity analysis in an easier way. As can be seen in Figure 3, gas injection at a point in the canister wall generates a gas propagation front. The gas fluxes are higher at the interfaces than in the bentonite body because the higher permeability of the interfaces. The interfaces play a major role as the migration of the gas takes place through preferential paths. It can be seen that gas pressure development at points situated at some distance increases very suddenly, and at some distance it remains constant. Gas outflow is permitted on the model boundary in a zone representing the fractures of the host rock (the rock is not modelled, it is substituted by appropriate boundary conditions representing inflow and outflow of fluids).

According to the model, permeability may increase up to two orders of magnitude when the peak of water or gas pressure occurs (Figure 4). Actually water injection at early times produces higher permeability increases. Hydration and the induced bentonite swelling counteract the opening effect and therefore, permeability decreases as the interfaces close, going back to low permeability values.

One of the main drawbacks of the axisymmetric model is that the in situ gas injection is performed in reality as point source but in the 2D axisymmetry the point source becomes a ring source. This reduces the gas pressure increments given the same properties. So developing a 3D model is a necessary but challenging task in view of the complex protocols that are considered in the test. As indicated above, figure 4 shows the evolution of permeabilities at different points within the axisymmetric model. The results indicate that the interfaces near the canister respond to water injection while the interfaces near the rock do not show permeability variations.

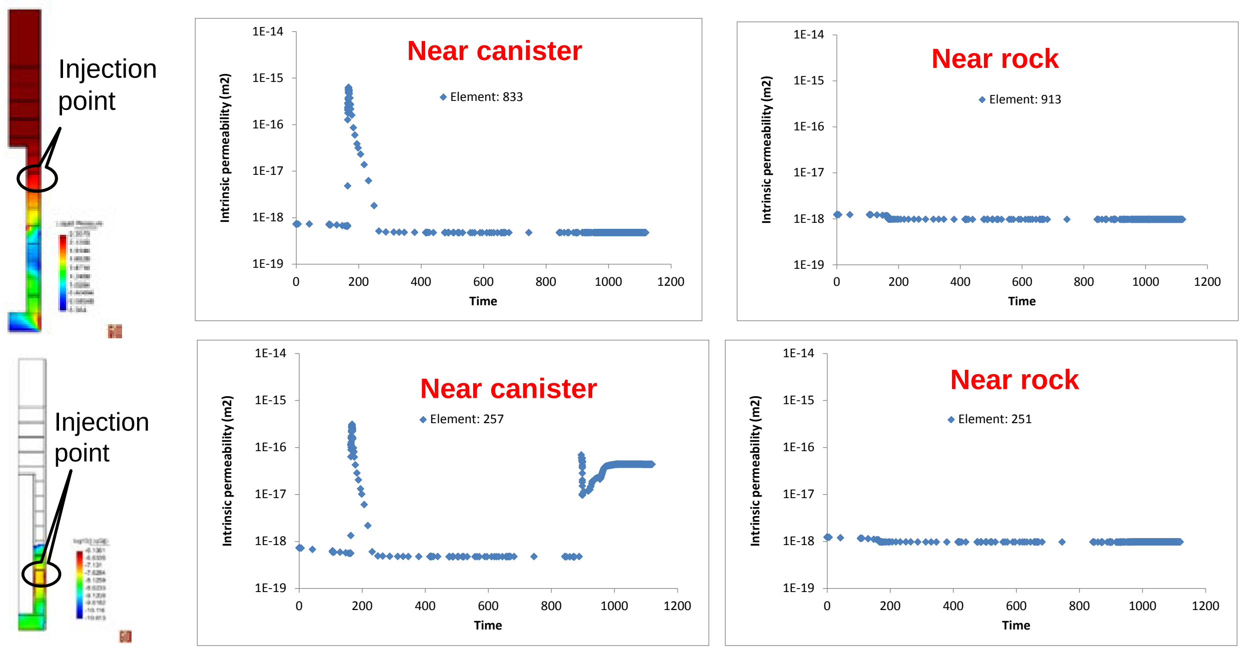

Figure 4. Evolution of permeabilities at interfaces in the model during water injection and gas injection (axisymmetric model).

5. Conclusions

Modelling the LASGIT in situ test is a complex task. In this paper, the modelling of hydration and gas injection based on two geometrical configurations is presented. Representation of the gas injection phase which in the experiment is carried out using a volumetric pump, is difficult due to the compressibility of the gas. The 3D model is necessary to represent point sources but, given the complexities of the physics (fast injection/nonlinear/coupled), may be non-competitive and thus a 2D axisymmetric model may be useful for preliminary understanding/ calibration and to perform sensitivity calculations with a controlled computational cost.

6. Acknowledgements

This work is funded by the EC through the FP7 research program (FORGE Project).

Numerical Simulation of Gas Migration Test of Compacted Bentonite using Model of Two-phase Flow through Deformable Porous Media

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Summary

CRIEPI already proposed an analytical method for simulating gas migration through the compacted bentonite using the model of two-phase flow through deformable porous media. It is already verified that the CRIEPI's method can simulate the behaviour of compacted bentonite during gas migration tests before large breakthrough, which defined as a sudden and sharp increase of amount of discharged gas. In this study, a failure criterion is introduced into the CRIEPI’s method to simulate behaviour of compacted bentonite throughout gas migration tests.

1. Introduction

In the current concept of repository for radioactive waste disposal, compacted bentonite will be used as an engineered barrier mainly for inhibiting migration of radioactive nuclides. Hydrogen gas can be generated inside of the engineered barrier by anaerobic corrosion of metals used for containers, etc. Therefore a method to evaluate the effect of gas pressure generation and gas migration on the engineered barrier is necessary. In this study, a failure criterion is introduced into the CRIEPI’s method, which is already proposed [1], to simulate behaviour of compacted bentonite throughout gas migration tests. Verification of the modification is also conducted in this paper by comparing calculated results with test results conducted under various test conditions.

Test No.	Bentonite	Dry density (Mg/m^3)	Initial axial stress (MPa)	Specimen size		Porous metal of gas inflow
				Height (mm)	Diam. (mm)	
K1	KunigelV1	1.22	0.69	20	60	Disk (60mm in diam.)
K3	KunigelV1	1.41	1.00			
K5	KunigelV1	1.59	2.03			
B1	KunigelV1	1.62	3.36	20	60	Disk (60mm in diam.)
B2	KunigelGX	1.62	1.77			
B3	Powdered KunigelGX	1.60	2.64			
S1	KunigelV1	1.20	0.59	20	60	Disk (60mm in diam.)
S2	KunigelV1	1.61	2.80			
S3	KunigelV1	1.61	2.79			Slit (60mm in length and 0.3mm wide)

Table 1 Test conditions of gas migration tests. Note : Back pressure is 0.3MPa for every test case listed above.

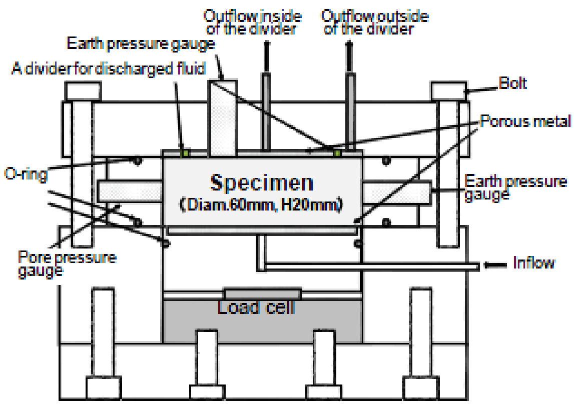
2. Gas migration test

2.1 Methodology

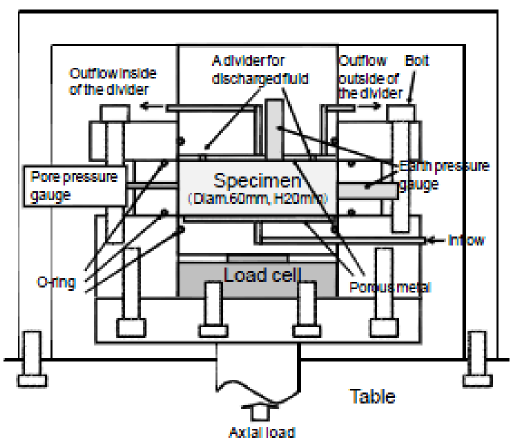
Table 1 shows test cases of the gas migration test conducted in this study. In the test cases of No.K1, No.K3 and No.K5, the effect of dry density of the specimen on the gas migration characteristics is investigated using the test apparatus shown in Fig. 1(a).

In the test cases of No.B1, No.B2 and No.B3, the effect of initial axis stress, which exceeds swelling pressure, on test results is investigated using the test apparatus shown in Fig. 1(b), while the effect of size and shape of inlet of applied gas on test results in the test cases of No.S1, No.S2 and No.S3 using the test apparatus shown in Fig. 1(c).

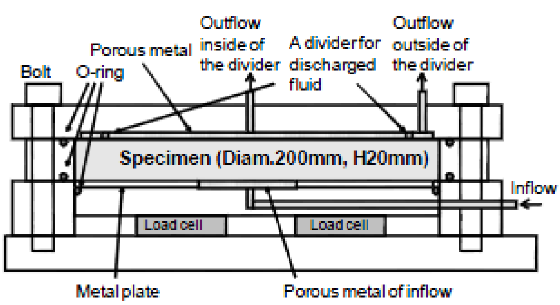
Further details of test conditions of the test cases are described in references [2] and [3].



(a) Test No.K1, K3 and K5



(b) Test No.B1, B2 and B3



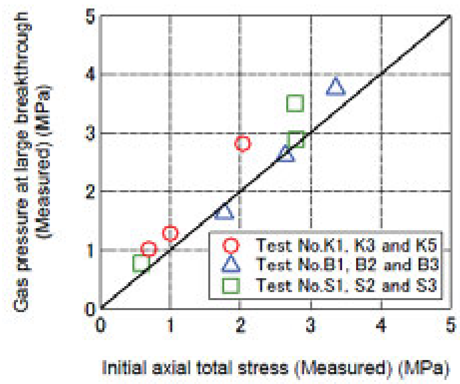


Fig.2

(above) Relationship between gas pressure at large breakthrough and initial axial stress.

Fig.1 (left) Cross sections of test cells used for the gas migration tests

2.2 Test results

Figure 2 shows the relationship between gas pressure at large breakthrough and initial axial total stress. All the test cases in Fig.2, large breakthrough occurred at boundary between the side face of the specimen and the internal face of the test cell. For Test No.B1, B2 and B3, gas pressure at large breakthrough approximates initial axial total stress. This means gas pressure at large breakthrough is not dominated by swelling pressure but by initial axial total stress. Contrary to Test No.B1, B2 and B3, pressure at large breakthrough is larger than initial axial total stress for Test No.K1, K3, K5.S1, S2 and S3.

Figure 3 shows an example of results of the test, showing time history of gas pressure, earth pressure, pore water pressure, volume of discharged water and effective gas permeability [1].

3. Numerical simulation

As previously mentioned in 2.2, large breakthrough occurred at between the side face of the specimen and the internal face of the test cell in all test cases in Table 1. As described by Tanaka et al. (2010) [2], when applied gas pressure exceeds the initial axial total stress, the specimen shrinks in axial direction with causing the clearance between the end of the specimen and the lower porous metal or metal plate as illustrated in Fig,4. Thus, a crack by shear appears along the side face of the specimen. In that case, if radial total stress is less than the gas pressure, the crack would develop and large breakthrough would occur as illustrated in Fig 4.

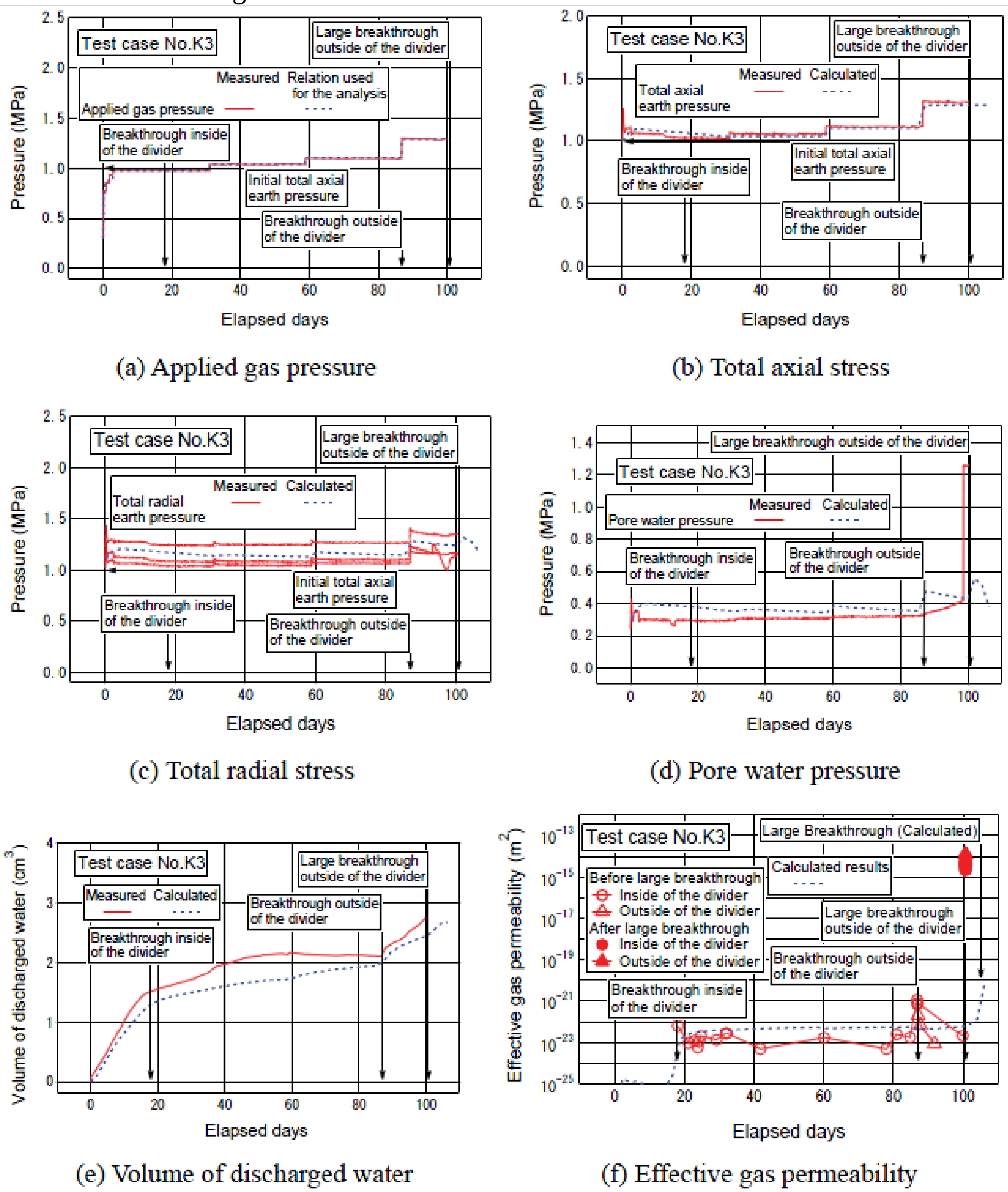
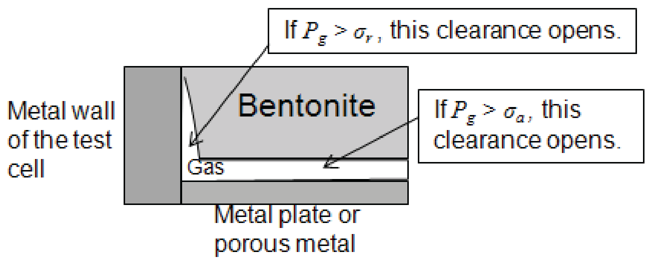


Fig.3 Measured results and calculated results with the passage of time (Test case No.K3)

Consequently, criterion of large breakthrough between the side face of the specimen and the internal face of the test cell is expressed as follows:

$P_g \geq \mathrm{Max}\ [\sigma_a , \sigma_r]$ (1)
where, P_g , σ_a , σ_r : gas pressure, axial total stress, radial total stress, respectively.

Introducing Eq. (1) into the CRIEPI's method, gas pressure at large breakthrough is calculated. Figure 5 shows the relationship between the calculated results and the measured results. Furthermore, calculated results with the passage of time are compared with measured results in Fig. 3. Figs. 3 and 5 show that the calculated results coincide well with the measured results.



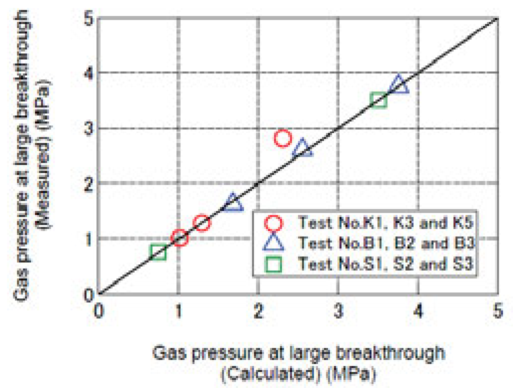


Fig.4 (left) Deformation of the bentonite specimen subjected to gas pressure. Fig.5 (right) relationship between measured results and calculated results of gas pressure at large breakthrough.

By summarizing described above, it is proved that behaviour of compacted bentonite can be simulated throughout gas migration tests by introducing Eq.(1) into the past CRIEPI's method.

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Hydraulic gas behavior of all relevant time and space scales of a generic repository. Contribution of Andra's study in the framework of FORGE WP1.2.

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Summary

The multiple barrier disposal concept is the cornerstone of all proposed schemes for geological disposal of radioactive wastes. The concept is based on a series of passive complementary barriers, both engineered and natural, that act to achieve the required level of safety for radioactive waste disposed in a geological repository.

Demonstrating an appropriate understanding of gas generation and migration is a key component in a safety case for a geological repository for radioactive waste. On the basis of work to date, the overall behaviour of waste-derived gas and its influences on repository system performance require improved understanding and numerical simulation is a suitable tool to do so.

Numerical simulations rely on TOUGH2-MP, a massively parallel general-purpose numerical simulation program for multi-phase fluid and heat flow in porous and fractured media. Tough2 was developed at the Earth Sciences Division of Lawrence Berkeley National Laboratory (LBNL).

Different simulations at various scales (cell, module and total repository scales) were performed by Andra in the framework of WP1.2 on a generic repository in a clay rock assuming no bentonite buffer surrounding the waste packages.

Some general conclusions, valid at all repository scales, can be drawn from the sensitivity process.

1. Introduction

FORGE WP1.2 is a sub work package dealing with benchmark simulation concerning gas generation and migration at repository depth (many hundreds of metres below ground level) and repository scale and including teams from several countries, having all different clay based disposal concepts.

The aim of this benchmark is more to understand the behaviour of the modeled system as underastood by each participant, rather than to undertake an inter-comparison of codes. To give some added value to this exercise, a feature not yet well represented in usual gas simulations was introduced: the explicit representation of the interface between canisters and cell walls.

2. Methodology

Although the final aim of the benchmark studies is to represent repository-scale simulations, the first exercise should be rather simple and at cell scale [1]. This first step was done during 2009 and first part of 2010. Results are showing a significant role of interfaces between plug and argillite and a transfer dominated at this scale by convection toward the drifts, radial diffusion being of secondary order.

Representing the transfer of gas at a larger scale including drifts and several tens of cells is then worthwhile. In this context, one of the major problems in representing gas transfers in a repository for radioactive waste is to model simultaneously all gas sources (generally located in the disposal cells) and the transfer pathways constituted by the network of interfaces, plugs and undergrounds drifts. The second benchmark at module scale (several tens of cells, Fig. 2.1) shows once more that interface plays an important role in the transient hydraulic-gas evolution as well as significant differences with the cell results such as the important role of dissolution/diffusion process along the pathways in the drifts.

The simulation of a complete generic repository including ten modules (Fig. 2.1) will force the teams to introduce a certain amount of upscaling in there simulations. More than this, having a complete repository will permit to validate the results at smaller scales (cell and module) and to give evaluation of the impact of gas at the shafts.

For this exercise we assume a repository having a simple architecture (Fig. 2.1). This architecture is more or less representative of some general concepts for HLW repository zones but without any national specificity. The main features are:

- Subdivision of the all repository into several modules linked by a main drift;
- All the modules are equal and contains an access drift serving 50 cells on each side;
- Two adjacent modules are not separated by a seal;
- In the main drift seals are placed between all modules connections.

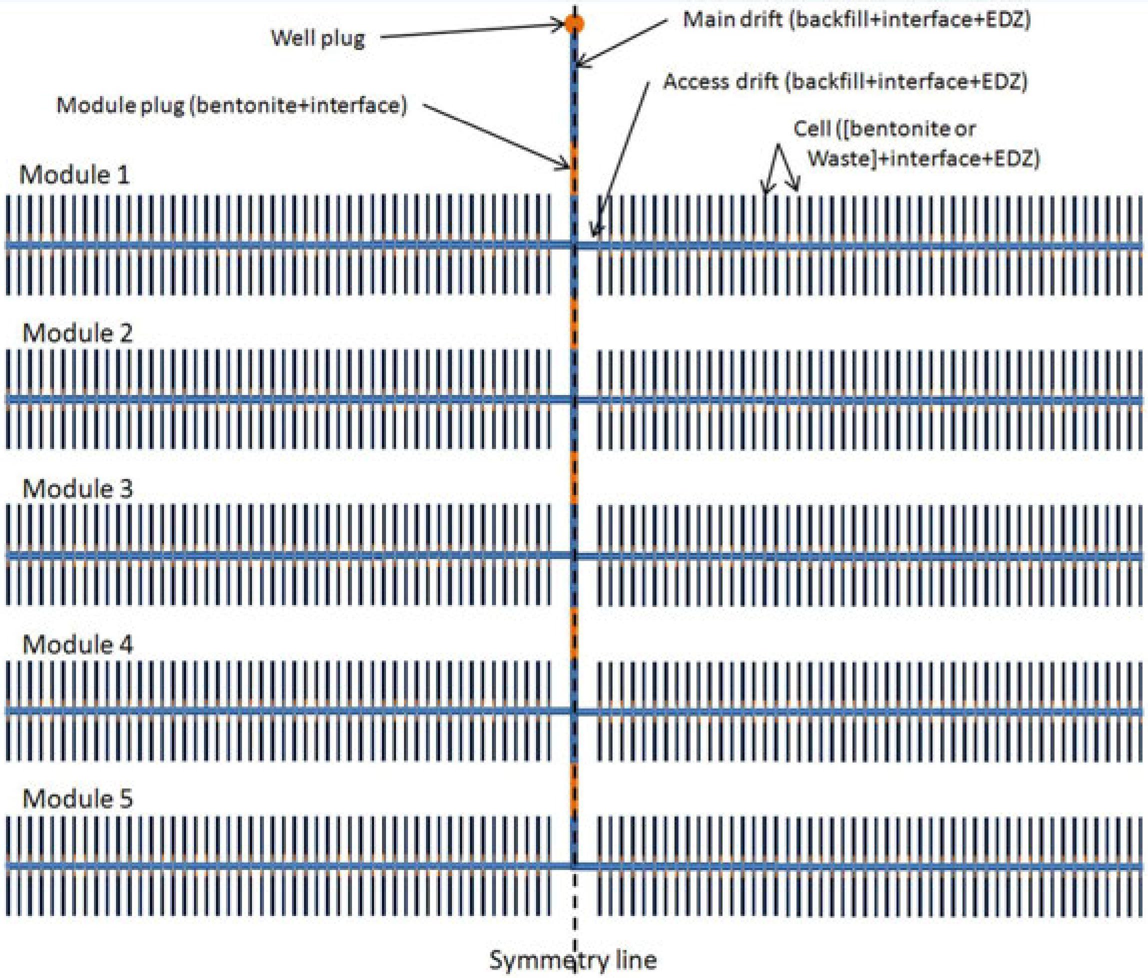

Figure 2.1: schematic representation of a generic repository for HLW

3. Results and conclusions

Different simulations have been made by Andra at the three different scales proposed by the work packages. The phenomenological results at all scales are coherent. The main general phenomenological results valid at all scales, and for all repository concept in a clay host rock, can be summarized as follows:

- Using (numerical or mathematical) upscaling, nowadays computers allow complex two phase flow simulation of a complete repository with a precise representation at all scales (from cell to shaft including drift network and explicit representation of EDZ and host rock);
- As host clays are very impermeable and gas entry pressures are high, the desaturation is only of some percent and thus there is a need of better characterization of the "water saturated tail" of the relative permeability and retention curves for these materials;
- EDZ permeability (gas entry pressure) is much higher (lower) than in the clay rock, thus expressed gas is mainly flowing through this material : better experimental characterization of the EDZ relative permeability and retention curves could also help increase the confidence on the simulation results.

- This last point is also true for the interfaces, adding intrinsic permeability values as well;
- Dissolution and diffusion are key combined processes in the comprehension of a repository hydraulic-gas transient and most of the produced hydrogen is finally migrating under dissolved form in the clay rock water. Decreasing the uncertainty on the host clay diffusion coefficient for H_2 could also help.

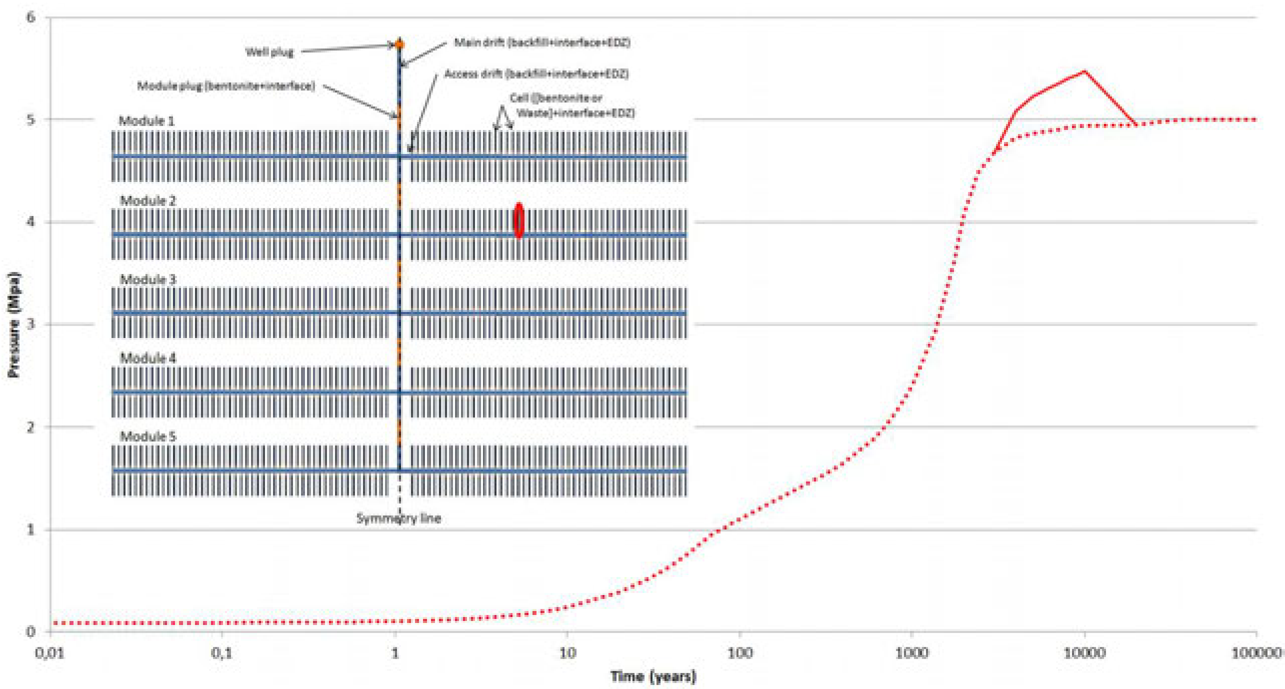

Figure 3.1: Water pressure (dashed lines) and gas pressure (continuous lines, only when gas phase exists) computed in the EDZ of a generic cell situated in the middle of a module (red ellipse on repository representation).

4. Aknowledgements

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme FTP/2007-2011.

5. References

[4] FORGE report D1.1 "Draft report on definition of benchmark studies on repository-scale numerical simulations of gas migration"

[5] FORGE report D1.3 "Progress report on benchmark studies on repository-scale numerical simulations of gas migration"

[6] FORGE report D1.4-R "Preliminary report on benchmark studies on repository-scale numerical simulations of gas migration"

Gas Permeability and Breakthrough Pressures of FEBEX Bentonite

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Summary

The gas permeability of the Spanish FEBEX bentonite compacted at dry densities of between 1.4 and 1.8 g/cm^3 with high water contents was measured for different confining, injection and backpressures in a steady-state equipment. The gas breakthrough pressure of saturated samples and along interfaces as a function of dry density was also determined.

1. Introduction

This investigation was carried out in the framework of Work Package 3 of the FORGE project, and its aim was the determination of the gas transport properties of a compacted bentonite to be used as engineered barrier in a radioactive waste disposal facility.

2. Materials and methods

The material used was the Spanish FEBEX bentonite, which is mainly composed of montmorillonite (90%). The saturated hydraulic conductivity and the swelling pressure of compacted samples are exponentially related to their dry density. For a dry density of 1.6 g/cm^3 the saturated permeability of the bentonite is about $5 \cdot 10^{-14}$ m/s and the swelling pressure has a value of about 6 MPa [1].

Gas permeability was measured in specimens of compacted bentonite previously mixed with different quantities of water. Cylindrical samples of 3.8 cm diameter and 7.8 cm height were obtained by uniaxial compaction. The setup to perform gas permeability measurements was designed to work as a constant head permeameter, allowing the change of the head value, the control of the confining pressure and the measurement of the gas inflow and outflow [2]. The injection and downstream pressures could be independently varied and kept constant by gas forward pressure controllers during the period of time necessary to get steady flow. Nitrogen gas was used as fluid.

A series of stainless steel cells were designed and manufactured to perform gas breakthrough tests in saturated bentonite [2]. The cells consisted of a body, pistons with o-rings at both ends of the samples and threaded caps. The samples, of 3.8 and 5.0 cm in diameter and 2.0 and 5.0 in height, were obtained by uniaxial compaction of the bentonite with its hygroscopic water content directly inside the cell body and were saturated with deionised water. Once saturated the cells were connected to a setup specially designed to measure breakthrough pressure consisting of two stainless steel deposits connected to the ends of the cell (Fig. 2.1). The pressure on both deposits was measured by means of pressure transmitters. Vacuum was applied to the downstream deposit and the other one was pressurised with nitrogen gas to 400 kPa. If no changes in pressure were recorded during 24 h, the injection pressure was increased by 200 kPa and kept constant for 24 h. The process was repeated until gas started to flow through the sample, causing a decrease of pressure in the upstream deposit and an increase in the downstream one.

Figure 2.4: Setup for measurement of breakthrough pressure in bentonite

3. Results

It was checked that gas permeability was greatly affected by dry density, decreasing about three orders of magnitude when it increased from 1.5 to 1.8 g/cm^3 for similar water content. The increase of water content caused also a decrease in gas permeability [2][3]. It was found that both gas permeability and the relative gas permeability were mainly related to the accessible porosity (Fig. 3.1). These relationships could be fitted to potential expressions with exponents between 3 and 4, as well as the relationship between intrinsic permeability and void ratio.

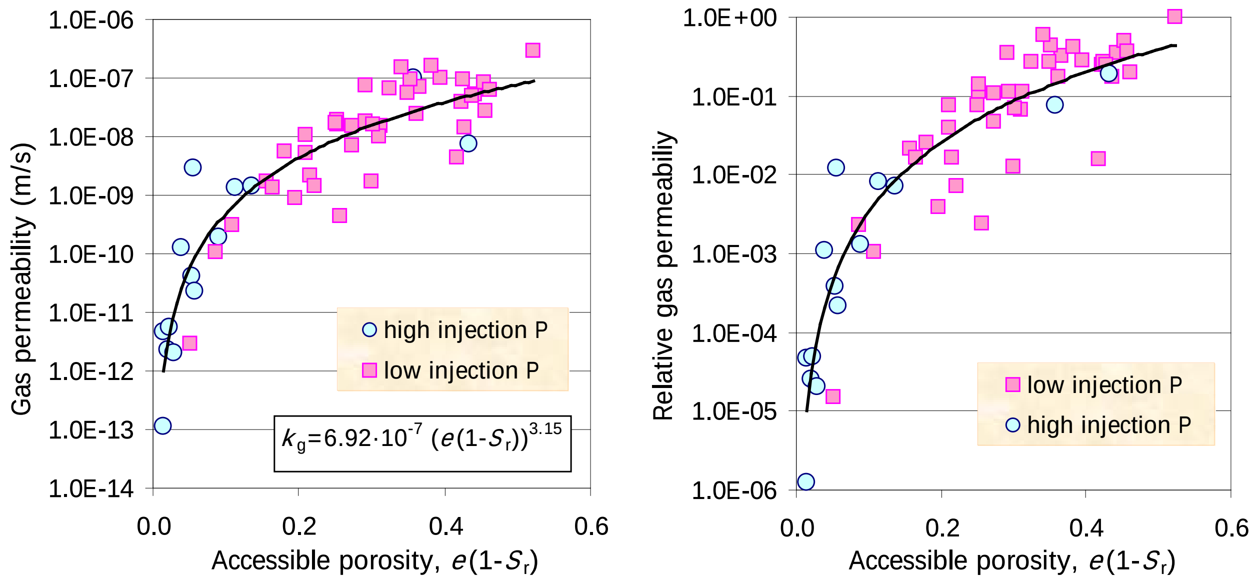


Figure 3.1: Gas permeability (left) and relative gas permeability (right) as a function of the accessible porosity for FEBEX

For gas pressures below 1200 kPa no effect of the injection or confining pressures on the value of permeability was detected, although when confining pressure increased from 1000 to 3000 kPa, the permeability decreased almost two orders of magnitude (Fig. 3.2, left). For a given confining pressure the permeability value decreased as the effective pressure increased (Fig. 3.2, right), especially if the increase in effective pressure was due to a decrease in gas backpressure. It was checked that the Klinkenberg effect was not significant for this material in the range of pressures applied in the tests.

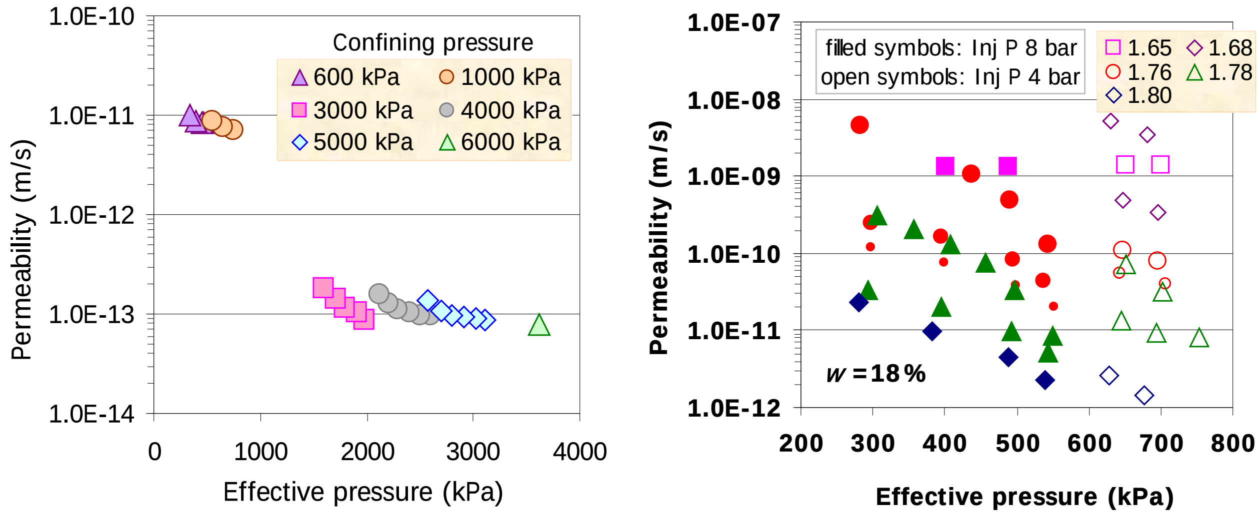
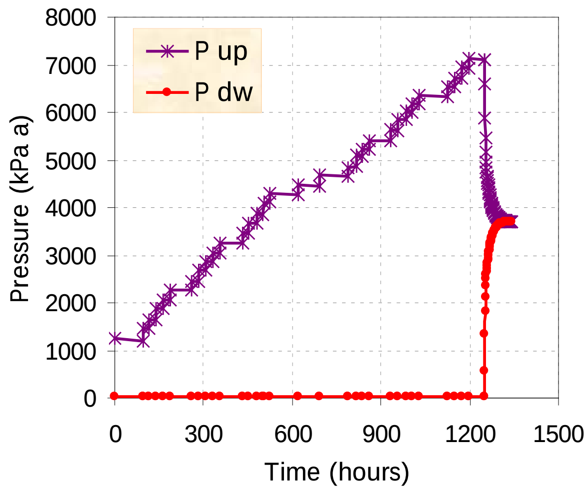


Figure3.2: Effect of effective pressure on gas permeability for samples of dry density 1.78 g/cm^3 (left, water content 19%, atmospheric backpressure) and average water content 18% (right, the dry density of the samples is indicated in the legend in g/cm^3)

The breakthrough pressure for the saturated bentonite increased with dry density, and the preliminary values obtained were in the order of 5 MPa for dry densities around 1.4 g/cm^3 and of 15 MPa for a dry density of 1.73 g/cm^3 (Fig. 3.3, left). These values were higher than the swelling pressure values measured for the same dry densities in saturated samples (Fig. 3.3, right). After breakthrough, the gas permeability values were much lower than those measured in samples of the same dry density and lower water contents not previously subjected to breakthrough. This highlighted again the influence of water content on gas permeability. Following breakthrough and resaturation of the samples, the same breakthrough pressure values were found again, what points to a perfect healing of the material upon saturation.

The effect of interfaces on breakthrough pressure values was also investigated. The breakthrough pressure along saturated bentonite interfaces tended to be lower than that for intact samples of the same dry density (Fig. 3.3, right). A saturated granite/bentonite interface (bentonite dry density 1.56 g/cm^3) allowed the passage of gas under a pressure of 0.7 MPa. After resaturation of this interface, the same breakthrough pressure was found.



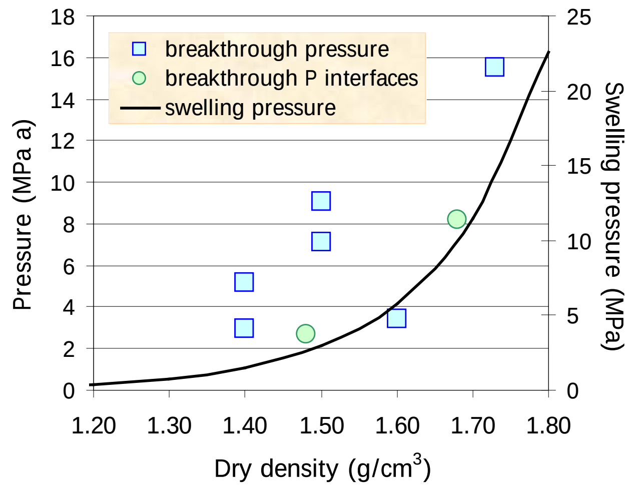


Figure 3.3: Pressure changes in the upstream (up) and downstream (dw) deposits during a breakthrough test in a saturated FEBEX sample of dry density 1.5 g/cm^3 (left). Gas breakthrough pressure and swelling pressure measured in saturated samples of different dry density, with and without interfaces (right)

4. Conclusions

The gas permeability of high water content bentonite samples decreased with dry density, whereas the breakthrough pressure of saturated bentonite samples increased with dry density, reaching values higher than the swelling pressure for the same dry density. After breakthrough and resaturation of the samples the same breakthrough pressures were recovered, which implied that healing was achieved during resaturation. The influence of the size of the samples and of the pressure increase rate on the breakthrough pressure values has to be assessed.

Preliminary results indicate that saturated interfaces between bentonite blocks are preferential paths for breakthrough, since their existence decreases the value of gas pressure needed for passage for a given average bentonite dry density.

5. Acknowledgements

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under Grant Agreement n^{o}230357, the FORGE project. The laboratory work was performed by Ramón Campos and Juan Aroz, from CIEMAT.

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Dissolved gases in crystalline rock, observations from Outokumpu deep drill hole

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Outokumpu deep drill hole (2500 m) in eastern Finland represents comparable lithologies and conditions met in the characterization of different sites for nuclear waste disposal in crystalline rock. Upper part of the bedrock pierced by the drill hole consists predominantly of mica schists, followed by an ophiolitic sequence (Outokumpu assemblage) at 1300 – 1500 m, whereas a coarse-grained granodioritic rock becomes predominant below 2000 m. Since the first observations during drilling, common existence of gases in formation fluids has been a major focus of interest. In addition to determination of the volume and composition of the evacuated gas phase, our current research aims at explaining the origin of the dissolved gases, processes involved and time scales required for their generation.

Saline, gas-bearing fluid was observed first time at the depth of about 1000 m during drilling (in 2004), when pressurized fluid samples were taken within a hydraulic-geophysical monitoring program. Later on, when the fresh drilling fluid was replaced by saline formation fluids, dissolved gases in drill hole water were sampled by taking a depth profile using a tube sampler. Gas compositions of samples from each depth section were analyzed, but quantitative volumes of gas could not be determined. To obtain more representative water samples from the bedrock, long-term pumping of fracture zones isolated by inflatable packers was conducted. Gas phase can thus be separated and analyzed on the ground, and the gas/water ratio can be estimated from the fluid flowing out. However, on-line monitoring of the gas composition during pumping revealed the complex behavior of gas flowing from high hydrostatic pressures of deep bedrock towards the atmospheric pressures: Both the gas flow rate and gas phase composition varied as a function of time. Composition-volume relationship indicates that the oscillation is at least partly attributed to the differences in the fugacities (solubilities) of the gaseous components.

Main components of the gas phase are methane (70-80%) and nitrogen (20-30%), followed by He (about 3-8%) and Ar (<1%). Repeated samplings also indicated presence of small but somehow inconsistently varying amounts of hydrogen in water samples delivered onwards in laboratories elsewhere. In a later sampling, analyses of the gas composition were performed on-site using mass spectrometry. The results show consistently low hydrogen concentrations (0.01 mM) for the uppermost 1.5 km of borehole, whereas deeper in the borehole hydrogen concentrations increase up to 40 % at the depth of 2400 m.

Total gas volumes were determined most reliably from fluid samples retrieved preserving their original hydrostatic pressures. Gas to water ratios were in the range 0.2-1 L/L at different depths, highest values were found at the depth of about 1000 m. Partial pressures (fugacities) of gases depend on the temperature and salinity of water, which are about 40 ^{o}C and 70 g/L at hole bottom. Highest calculated gas-phase pressure was about 50 bars at 1000 m.
This study is a part of the Finnish research programme on nuclear waste management KYT2014.

Simplified 1D Modelling of the HG-A Test

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Summary

The HG-A in-situ test located at the Mont Terri Underground Rock Laboratory (Switzerland) is part of the FORGE project. In this test, a horizontal micro-tunnel was excavated in the ultra-low permeable Opalinus clay, thereby creating an EDZ around it. The behaviour of the EDZ has been investigated by applying a varying pressure on the tunnel surface and forcing liquid or gas flux along the EDZ by applying varying water and gas injections rates. A simplified 1D model is obtained by adopting appropriate simplifying assumptions. A key feature of this model is a deformation-dependent intrinsic permeability law for the EDZ. The comparison between the measured evolution of upstream pressures at the EDZ and the results of the 1D simplified model shows that the main features of the measured evolution are well reproduced, but some other aspects could not be modelled.

1. Introduction

The HG-A test is located in the Mont Terri Underground Rock Laboratory, near Saint Ursanne (Jura, Switzerland) and is operated by NAGRA in the framework of the FORGE project. It consists of a horizontal tunnel of 1 m in diameter and 13 in length excavated in the ultra-low permeable Opalinus clay. During the tunnel drilling, the Opalinus clay near the tunnel wall was damaged, giving rise to an EDZ (Excavation Damaged Zone) around the tunnel. The tunnel was divided into three sections: (1) the test section at the last 4 m, close to the tunnel end, which was backfilled with gravel; (2) the megapacker section at the next 3 m, where an inflatable rubber packer of 1 m in diameter was installed; and (3) the liner section at the next 6 m, close to the tunnel mouth, where a steel liner was installed. After saturation with water of the test section and application of pressure to the megapacker, a series of liquid and gas injection tests were carried out at varying megapacker pressures, whereby liquid or gas was injected into the test section and, due to the very low permeability of the intact Opalinus clay, forced to flow back along the EDZ. One of the key aspects of this test is the variation of the intrinsic permeability of the EDZ due to the opening and closing of the existing cracks induced by the megapacker pressure and the injected liquid and gas flows. More information on the HG-A test may be found in Lanyon et al. (2008) [1].

2. Modelling of the HG-A Test

Modelling of the HG-A test was done within the framework of multiphase deformable porous media. Details of this formulation may be found in Olivella et al. (1994) [2].

Both the (undamaged) Opalinus clay and the EDZ were considered to be porous media, with an incompressible solid phase (with one species: clay), an incompressible liquid phase (with two species: water and air) and a gas phase (with two species: water and air). The properties of the liquid phase were assumed to be independent of the concentration of dissolved air and the gas phase was assumed to be a mixture of dry air and water vapour, both assumed to be ideal gases, such that Dalton's law holds. Exchanges of both species water and species air between the liquid phase and the gas phase were allowed, but it was assumed that they were always in equilibrium, defined by the psychrometric and Henry's laws.

It was assumed that motions are slow so that terms involving accelerations and products of velocities may be neglected. Motion of the liquid phase and of the gas phase were described by generalisations of Darcy's laws appropriate to unsaturated porous media and motion of the species water and the species air in the liquid phase and in the gas phase were described by Fick's laws. In the (undamaged) Opalinus clay the intrinsic permeability was assumed to be constant, but in the EDZ it was assumed to depend on the volumetric deformation of the solid skeleton via an aperture-based cubic law, as presented in Olivella et al. (2008) [3]

A generalised form of Terzaghi's effective stress principle was assumed to hold both in saturated and in unsaturated conditions, whereby the maximum of the liquid and gas pressure is considered to be the equivalent fluid pressure acting on the porous medium. The constitutive laws for generalised effective stress for both the (undamaged) Opalinus clay and the EDZ were assumed to be given by isotropic linear elasticity. A van Genuchten's water retention curve governed the degree of saturation of the porous medium and, in particular, the transition between saturated and unsaturated states. Finally, it was assumed that temperature remained constant everywhere.

By combining the balance equations, the equilibrium restrictions and the constitutive relations, we get the field equations (balance of momentum for the porous medium as a whole, balance of mass of the species water and balance of mass of the species air), that with suitable initial and boundary conditions (provided by the test protocol) allow to determine the unknown functions (displacement vector of the solid phase, the pressure of the liquid phase and the pressure of the gas phase) of the space position and time.

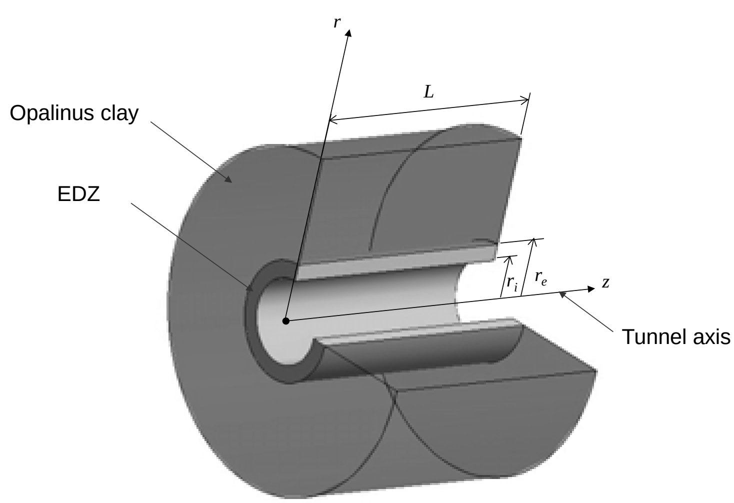

Figure 1. Geometry and coordinate system around the HG-A tunnel in front of the megapacker.

In the considered case of the HG-A test, we have considered simplifying assumptions that greatly reduce the complexity of the field equations. In order to present them, reference is made to Figure 1, which shows the geometry of the modelled region (the EDZ in front of the megapacker and the (undamaged) Opalinus clay behind it) and the coordinate system used. Mechanical assumptions: both on the EDZ and on the (undamaged) Opalinus clay (1) there is axisymmetry about the tunnel axis (z axis); (2) there are no volume forces (gravity is neglected); and (3) slices $z = const$ move independently and in plane strain. Hydraulic assumptions: both on the EDZ and on the (undamaged) Opalinus clay (1) there is axisymmetry about the tunnel axis (z axis); and (2) there are no volume forces (gravity is neglected). Furthermore, on the EDZ, it is assumed that $(3)^{EDZ}$ the flows of the liquid phase and of the gas phase are parallel to the tunnel axis (z axis); and, on the (undamaged) Opalinus clay, it is assumed that $(3)^{OPA}$ there is no gas phase (saturated state), there is no flow of the liquid phase (undrained conditions) and there is no air in the liquid phase.

With these simplifying assumptions, the problem reduced to a coupled system of two differential equations in two unknown functions (the pressure of the liquid phase in the EDZ and the pressure of the gas phase in the EDZ) of the cylindrical coordinate z on the 1D domain, representing the part of the EDZ in front of the megapacker, and of time t. The boundary conditions at the end of this 1D domain close to the test section were derived from the protocols (time evolutions) of the volume injection rate of the liquid phase and of the volume injection rate of the gas phase. The boundary conditions at the end of this domain close to the liner were generalized conditions involving the mass rates of the liquid phase and of the gas phase and the values of the external pressures of the liquid phase and of the gas phase. The protocol of the megapacker pressure was transformed into a body term on the 1D domain. The initial conditions for the unknown functions were derived from the assumed conditions of the test. This coupled system of equations was solved using the FEM, and has been implemented in an Excel worksheet. Once the two unknown functions are known, all the field variables both on the (undamaged) Opalinus clay and on the EDZ are determined in closed form using the two unknown functions. A comprehensive description of this simplified formulation and of its numerical implementation may be found in Alcoverro et al. (2012) [4]

3. Results

Figure 2 shows the evolution of the fluid pressure measured at the test section compared with the evolution of the effective fluid pressure (the maximum of liquid and gas pressures) at the contact between the test section and the megapacker (upstream of the EDZ) resulting from the 1D model. The considered time period covers both water injection tests (days 0 to 1250) and gas injection tests (days 1250 to 1750). It may be seen that the 1D model reproduces well the main features of the aforementioned evolution, but the increasing base trend could not be modelled.

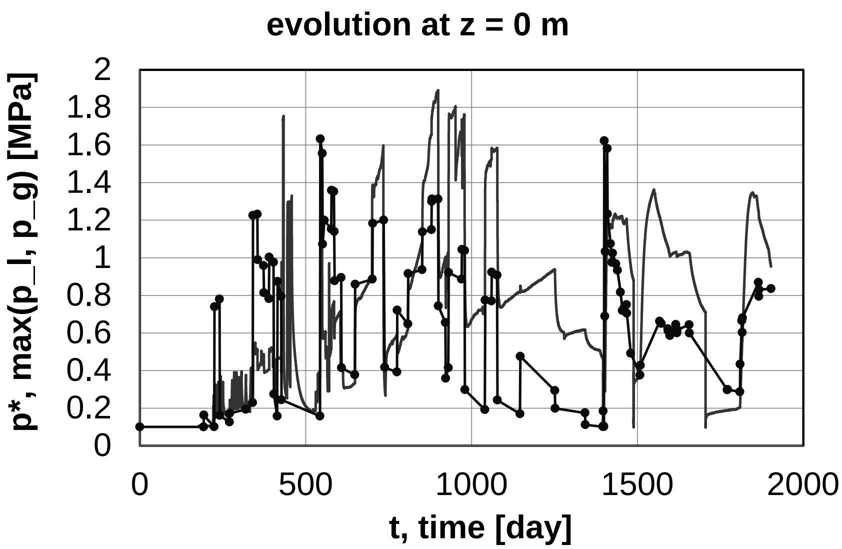

Figure 2. Evolution of the effective fluid pressure at the test section: measured (thin line) and 1D model (thick line with full circles).

Figure 3 shows the evolution of the intrinsic permeability at the contact between the test section and the megapacker (upstream of the EDZ). The large variations in the intrinsic permeability (note the logarithmic scale) result from the variations of crack apertures in the EDZ.

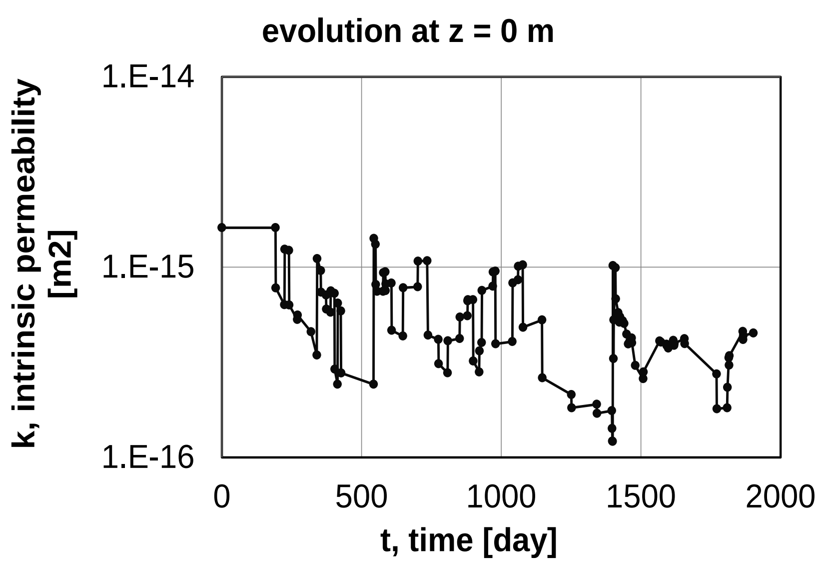

Figure 3. Evolution of the intrinsic permeability at the EDZ at the contact between the test section and the megapacker.

It should be mentioned that a 2D axysimmetric model for the HG-A test using CODE_BRIGHT (a FEM code developed at the UPC) showed that the simplifying assumptions leading to the 1D model were appropriate, except for small zones of the EDZ near the ends of the megapacker.

Since the simplified 1D model requires little computing time, it may be used for the parameter calibration of a 2D axisymmetric model. On the other hand, the validity of the simplifying assumptions allows gaining insight into the complex HG-A test.

4. Acknowledgements

This work is funded by the EC through the FP7 research program (FORGE project).

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FORGE WP4-5 : Gas migration through Callovo-Oxfordian claystone: Mechanisms description and remaining issues

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Andra, France

The French national Agency for the management of radioactive waste (Andra) plans to build a deep geological repository in the Callovo-Oxfordian (COX) clay layer To assess the impact of gas generated within the repository, a program has been set up to investigate gas properties of clay materials. This program consists in lab tests experiments on clay samples and large scale experiments in the underground Laboratory of Meuse/Haute-Marne. A part of those experiments have been done within the FORGE project. All the data obtained and the observations made from the experimental program have been used to build evolution models. Those models are integrated in the safety assessment tools to simulate gas migration at repository scale.

Understanding gas migration mechanisms in clay materials needs to develop investigation tools at several scales. To model gas flow through porous media; most of the time classical two phase flow based on generalized Darcy law for each phase are used. To be able to run that models, macroscopic properties have to be known with a sufficient precision. In that context, water and gas permeability function of the water saturation state; relationship between capillary pressure and water content have been measured. The dependency of those properties with the mineralogical composition of Cox has been shown and a discussion will be made about the variability of the measure due to the difficulty to obtain clay sample without any damaged. The extension of those properties for the EDZ based on lab test and in situ test can also be proposed.

One of the main elements investigated about gas transfer in clay is the evaluation of the gas entry pressure. Different kind of technics has been developed to estimate the pressure values at which the gas will penetrate the clay and the dependency of those values in regard of environment context has been explored: scale effect, several gas used (H_2, N_2, Ar, He), confining pressure … To complete gas migration observation on samples and field scale experiments, investigation on material microstructure have been done. Technics as FIB/SEM are used to extract a 3D pore network. The point is to confirm if there is enough connected pores with a size around 50nm which is the corresponding size of gas entry pressure measured in terms of Laplace law.

During the meeting, the different parameters needed to model gas migration at the underground repository scale will be discussed. Mechanisms identified through the experimental program and the remaining issues at the end of the FORGE project concerning Cox will be presented.

Validity of critical stress theory applied to the movement of water and gas along a fracture plane

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Summary

Fractures and faults can be conduits of flow around a geological disposal facility (GDF) and as such an understanding of their transmissivity is important. Critical stress theory states that fractures at certain orientations to the stress field are more conductive than others. This study examined the interplay of fracture orientation and fracture transmissivity using a bespoke shear apparatus. Other important aspects of fracture flow were also examined, showing that fracture flow is a complex problem. The study looked at both water and gas flow along idealised fractures made of kaolinite gouge.

1. Introduction

Critical stress theory states that the transmissivity of a fracture/fault is strongly dependent upon its orientation to the prevailing stress tensor. Studies of hydrocarbon reservoirs confirm this general approach when active shearing occurs along a fault network. However, the inter-play of fracture orientation and stress tensor direction is also of crucial importance in understanding the evolution of fluid migration in a geological disposal facility (GDF). If valid for the specific lithologies of interest to a GDF, critical stress theory could provide a framework for the description of fluid (gas and water) movement in both: i.) the engineered disturbed zone (EDZ), where a range of fracture orientations will be present in a highly-complex localised-stress field, and ii.) the far field, where either a pre-fractured host rock is located, or, joints, fractures or faults are encountered.

However, while critical stress theory suggests that fractures close to localised shear failure are critically stressed and therefore most conductive, analysis of the geometry and stress conditions of the various fracture networks within the Sellafield area, UK, have revealed that no such features exist, despite the fact that fluid flow was observed within the boreholes.

2. Methodology

In order to understand the underlying causes for the apparent conductivity of non-critically stressed fractures, a matrix of tests were performed by the British Geological Survey. A bespoke, Angled Shear

Rig (ASR, Figure 1) was built to study the relationship between water and gas flow along a clay-filled fracture surface, as a function of normal load and shear stress.

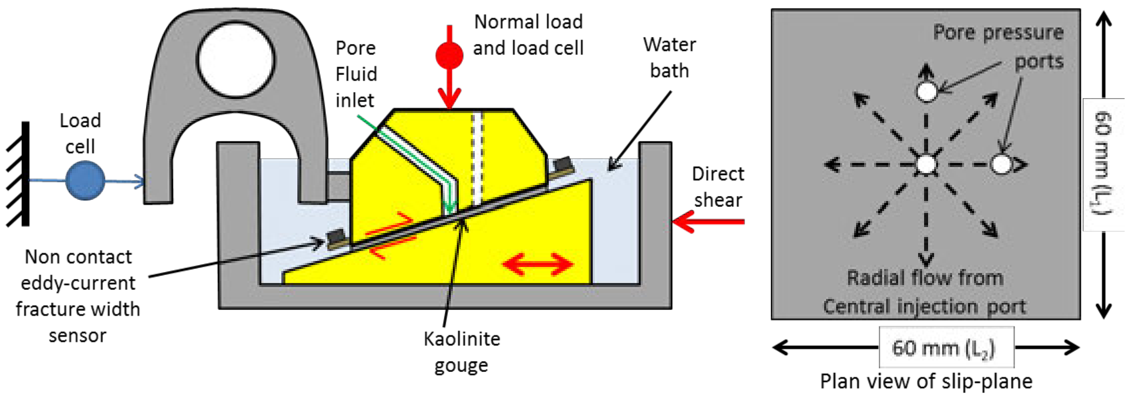

Figure 1: The bespoke Angled Shear Rig (ASR).

3. Results

The results clearly demonstrate that as normal load increases, fluid movement along the fracture plane significantly reduces (Figure 2). However, as the fracture is unloaded, significant hysteresis is observed, with the fracture exhibiting only a partial recovery in flow. This simple observation illustrates the importance of stress history in defining the conditions under which fluids are mobile within the fracture plane.

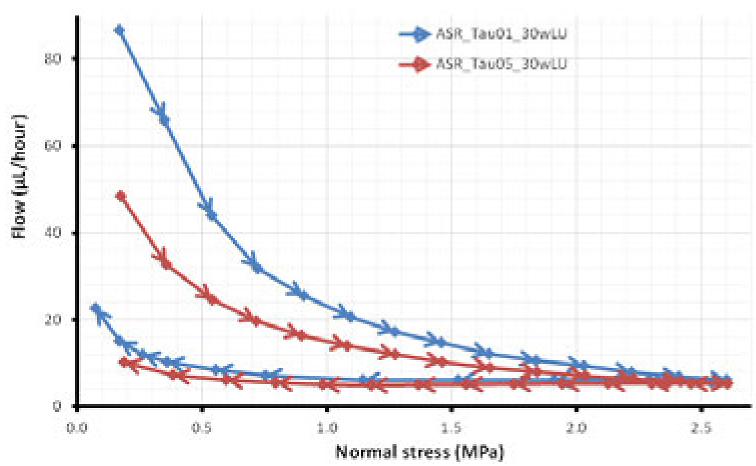

Figure 2: Results from two loading-unloading tests displaying considerable hysteresis during unloading.

Hysteresis in shear stress was also observed during the unloading stages of the experiment. Examination of the data suggests that the shear stress to normal stress ratio appears to control the movement of fluids during the unloading stage of the experiment. This observation is particularly important for fractured systems undergoing stress relaxation by exhumation/uplift or compaction through continued burial. In these systems, flow will be strongly linked to burial history and the resultant shear to normal stress ratio. It is interesting to note that the exhumation of the rocks in the Sellafield area during the Paleogene-Neogene uplift, would have resulted in considerable stress relaxation. This may account for the non-critically stressed nature of the fractures and explain the observed flow responses seen within the boreholes.

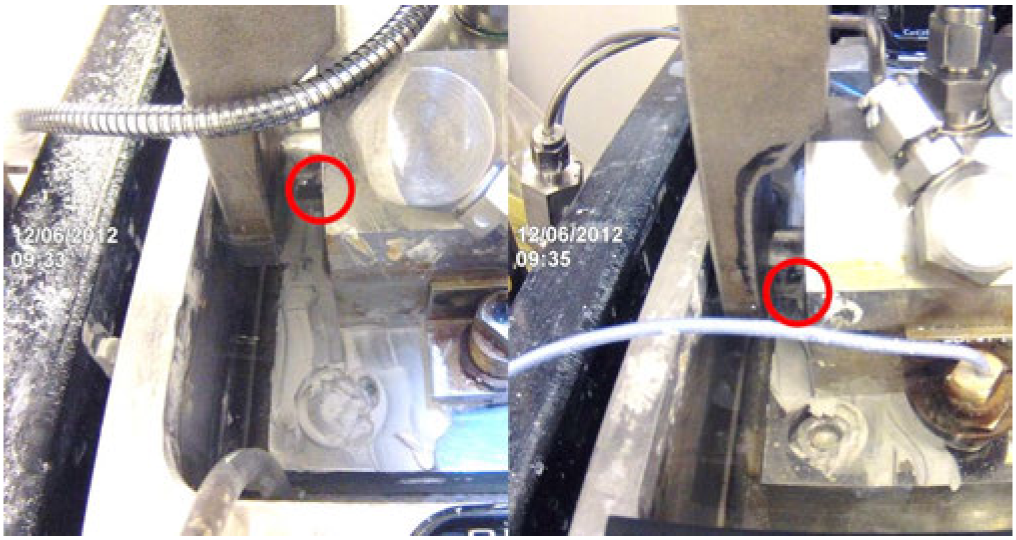

Figure 3: Three tests were conducted and recorded using time-lapse photography to observe the escape of gas from the slip-plane into the bath of the apparatus. All three of these experiments showed that a small, isolated stream of bubbles escaped from a single or multiple location/locations.

Analysis of the pressure distribution across the fracture plane, combined with post-test characterisation of the gouge, suggests water and gas flow is non-uniformly distributed within the fracture plane, with the latter occurring through localised preferential pathways (Figure 3). While repeat gas injection experiments show a consistent gas entry pressure, peak gas pressures were found to vary. It is suggested that this may reflect subtle changes in the number, distribution and geometry of the conductive pathways.

Fracture transmissivity was also found to be sensitive to a number of other factors including shearing (Figure 4), with flux decreasing by an order of magnitude, and swelling, again exhibiting a significant decrease in flow and fracture roughness.

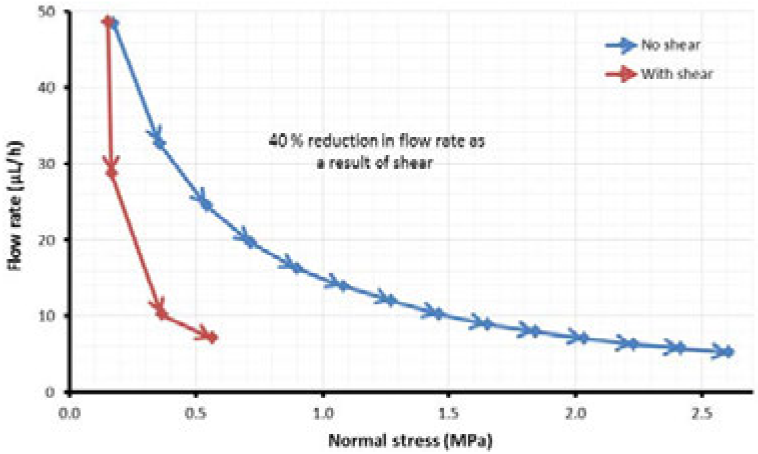

Figure 4: The reduction in fracture flow as a result of shearing, clearly showing that shear is an effective self-sealing mechanism.

The results from this study, combined with those from previous field observations, illustrate that understanding the stress-history (both uplift and burial) of the rock mass and associated fractures is essential, and an understanding the current stress regime is insufficient to estimate the flow behaviour of present-day fractures.

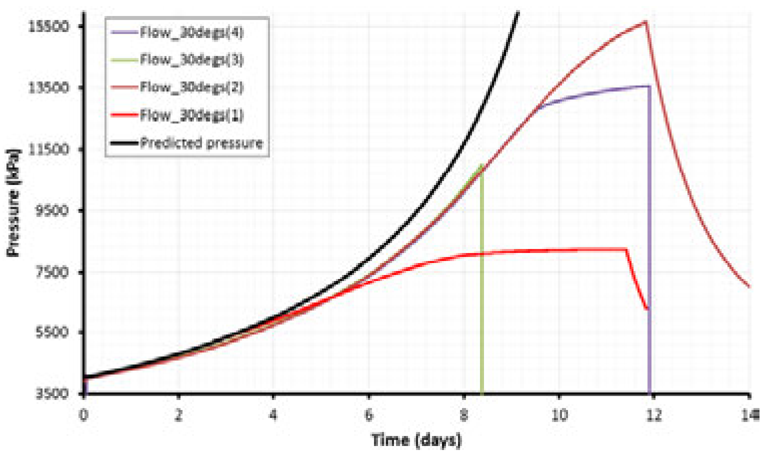

Figure 5: Repeatability of gas injection tests on a discontinuity oriented at 30°.

Repeat gas injection tests have shown that considerably different gas peak pressures are observed, as shown in Figure 5. This is consistent for fractures oriented at 0°, 15°, 30° and 45° to the shear direction. Careful analysis of the data can determine the gas entry pressure, and this appears to show relatively similar results. However, some tests do appear to reach entry pressure at lower pressures. As no consistency is observed in gas peak pressure, and therefore fracture transmissivity, during repeat testing at all angles, there is no consistent result with gas peak pressure and fracture orientation. However, the experimental study has clearly demonstrated a variation in gas entry pressure with discontinuity orientation (Figure 6). This demonstrates that the critical stress theory is applicable in the absence of stress relaxation.

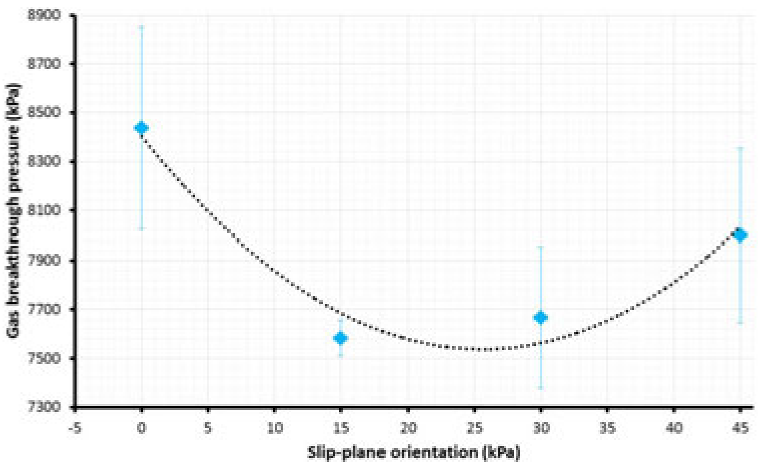

Figure 6: Gas injection pressure variation with discontinuity orientation.

4. Acknowledgements

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under Grant Agreement no230357, the FORGE project.

Determination Of Gas Diffusion Coefficients In Boom Clay: Effect Of Molecular Size and Anisotropy

Elke Jacops, Geert Volckaert, Norbert Maes, Joan Govaerts, Eef Weetjens, Arno Grade

SCK•CEN, Mol, Belgium

Summary

Within a geological repository, the production of gas is unavoidable. Gas generation rates in general are slow and the produced gas in the near field of a geological repository will dissolve in the pore water and is transported away from the repository by diffusion as dissolved species. Should the gas generation rate become larger than the diffusive flux, the porewater will become oversaturated and a free gas phase will form, possibly leading to tensile fractures within the rock fabric. This entire process may locally and at least temporarily alter the hydraulic and mechanical properties of the engineered barriers and the clay and, perhaps, their performance. Therefore it is important to assess whether or not gas production rates might exceed the potential removal rate by molecular diffusion of dissolved gas. These assessments depend largely on the applied parameter values. Due to the large uncertainty on the available diffusion coefficient for H_2, combined with the uncertainty on the gas source term, made it impossible to exclude the formation of a free gas phase. The described method allows to measure diffusion coefficients for various dissolved gasses with good accuray. Moreover by using the relationship between the effective diffusion coefficient D_{eff} and the kinetic diameter of the gasses, we are able to estimate D_{eff} for gasses like H_2, traditionally suffering from experimental difficulties.

1. Introduction

The Belgian agency for radioactive waste and enriched fissile materials Ondraf/Niras presently considers Boom Clay as a potential host formation for the disposal of high-level and long-lived radioactive waste. Long-term repository evolution studies have pointed out that the production of gas is unavoidable within a geological repository. Gas is produced by different mechanisms: anaerobic corrosion of metals in waste and packaging, radiolysis of water and organic materials in the waste and engineered barriers and possibly microbial degradation of various organic wastes. Corrosion and radiolysis yield mainly hydrogen while microbial degradation leads to methane and carbon dioxide [1]. Gas generation rates in general are slow and the produced gas in the near field of a geological repository will dissolve in the pore water and is transported away from the repository by diffusion as dissolved species. Should the gas generation rate become larger than the diffusive flux, the porewater will become oversaturated and a free gas phase will form. Initially, isolated gas bubbles will accumulate until a continuous gas phase is formed. As gas pressure continues to increase, discrete gas pathways may be formed by tensile fractures within the rock fabric. Consequently, this entire process may locally and at least temporarily alter the hydraulic and mechanical properties of the engineered

barriers and the clay and, perhaps, their performance [1]. Therefore it is important to assess whether or not gas production rates might exceed the potential removal rate by molecular diffusion of dissolved gas. The currently available gas diffusion parameters (D_{eff}: effective diffusion coefficient) for hydrogen in Boom Clay, obtained from the MEGAS project [2], and re-evaluated [3] [4] lead to an estimated D_{eff} between 1.9 10^{-12} m²/s and 1.5 10^{-10} m²/s. Sensitivity calculations [5] showed that the large uncertainty on the diffusion coefficient, combined with that on the gas source term, made it impossible to exclude the formation of a free gas phase.
To reduce the uncertainty, an experimental method was developed [6] to determine more precisely the gas diffusion coefficient for dissolved gases (especially dissolved hydrogen) in Boom Clay. The methodology was demonstrated for He and CH_4. As experiments with H_2 remained problematic due to recurrent bacterial activity, diffusion experiments with gasses of different size were started. If a relationship between size of the molecule and the diffusion coefficient can be found, the diffusion coefficient of H_2 and other gasses can be predicted.

2. Methodology

The method is based on the cross-diffusion of two dissolved gasses under (quasi-)constant concentration gradients, yielding two gas diffusion coefficients in a single test. In practice a clay core is sealed in a stainless steel cell and connected at both sides with water vessels that are pressurized with 2 different gasses at the same pressure. Gas dissolves into the water and because of the gas concentration gradient, these dissolved gasses diffuse through the clay. The dissolved gas that diffuses into the other water vessel will equilibrate with the gas atmosphere and the changes in the gas composition are determined by gas chromatography.

In the first experimental phase, we focussed on reproducibility. In the second phase, we focussed on the effect of the bedding planes in the clay sample to determine the anisotropy in the gas diffusion parameters. In the third phase, we investigated the relationship between the size of the diffusing gas molecule and D_{eff}. For this topic, we performed diffusion experiments with gasses of different size (C_2H_6, Ne, Ar, Xe). Based on observed relationships D_{eff} vs. the gas kinetic diameter we investigated the possibility to estimate D_{eff} for other gasses as H_2. In the last experimental phase we wanted to confirm the estimated D_{eff} value for H_2.
The tests are interpreted with a simple diffusive transport model. The model solves the diffusive transport equation in a 1D geometry and is based on the second law of Fick for diffusive transport in porous media. As output, fluxes at both faces are calculated with the first law of Fick, as well as concentration profiles at regular time intervals and concentration. All calculations are performed with COMSOL multiphysics 3.5a, Earth science module. The diffusion coefficients are obtained by using a least squares fitting procedure to the experimental data with the MATLAB Optimization Toolbox.

3. Results

3.1 Reproducibility

Diffusion experiments were performed on different Boom Clay cores, using He and CH_4. For both gasses, the results were reproducible and we obtained a good accuracy (rel. error < 10%) (table 1).

Table 1: D_{eff} and 95% confidence interval for He and CH_4 on different samples

	D_{eff} (m²/s)	95% confidence interval
He core 1 (⊥)	$4.51\ 10^{-10}$	$4.30\ 10^{-10} – 4.74\ 10^{-10}$
He core 2 (⊥)	$4.37\ 10^{-10}$	$4.26\ 10^{-10} – 4.44\ 10^{-10}$
He core 4 (//)	$7.18\ 10^{-10}$	$6.81\ 10^{-10} – 7.55\ 10^{-10}$
CH_4 core 1 (⊥)	$0.90\ 10^{-10}$	$0.87\ 10^{-10} – 0.92\ 10^{-10}$
CH_4 core 2 (⊥)	$0.83\ 10^{-10}$	$0.81\ 10^{-10} – 0.85\ 10^{-10}$
CH_4 core 4 (//)	$1.42\ 10^{-10}$	$1.40\ 10^{-10} – 1.45\ 10^{-10}$

3.2 Effect of anisotropy

The anisotropy ratio for diffusion was determined using He and CH_4 on a clay core with a stratification parallel to the bedding plane (table 1), and for He and CH_4 the same anisotropy ratio of 1.59 was obtained.

3.3 Effect of the molecular size of the gas

To investigate the effect of the molecular size (kinetic diameter) on D_{eff}, additional experiments were performed with Ne, Ar, Xe and C_2H_6 (table 2). Figure 1 shows the relationship between the kinetic diameter of the gas molecule and D_{eff}. To investigate the role of the kinetic diameter, the relationship between D_0 (diffusion coefficient in free water) vs kinetic diameter is plotted (fig. 1). As shown in (fig. 1) this relationship can be described as $D_0 = 5.96\ 10^{-8}\ e^{-0.899x}$ wih x the kinetic diameter of the gas molecule (expressed in Angstrom). When we do the same for D_{eff} we obtain $D_{eff} = 4.99\ 10^{-9}e^{-1.102x}$ but the D_{eff} for He strongly deviates and biasses the curve (not shown in graph). If we neglect He we obtain $D_{eff} = 1.55\ 10^{-9}\ e^{-0.805x}$ with a similar exponential factor as in the relationship D_0-x. Based on the kinetic diameter and by using this last equation, D_{eff} for H_2 was predicted.

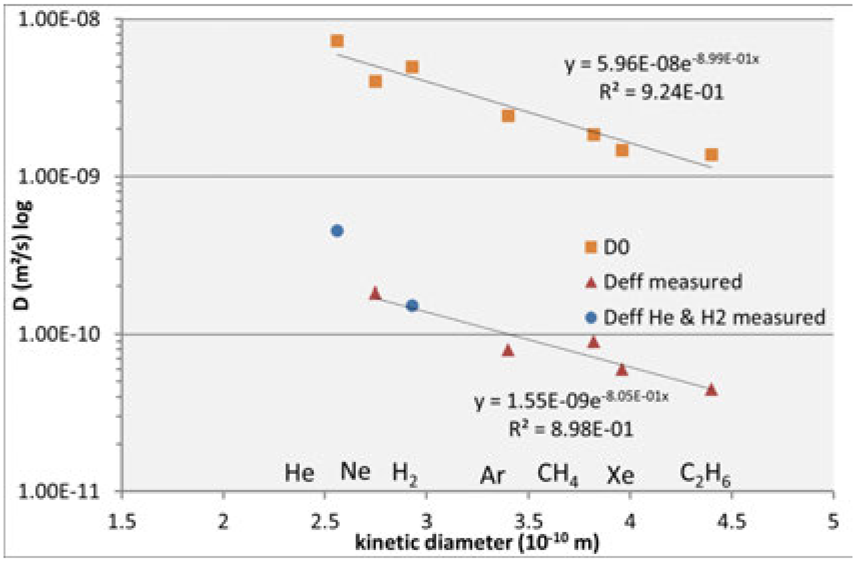

Figure 1: D_0 vs kinetic diameter (squares) and D_{eff} vs kinetic diameter (triangles) and their best fit. Measured D_{eff} values for He and H_2 are represented as circles.

Table 2:Experimental determined D_{eff} for the different gasses tested ⊥ to bedding plane

	Kinetic diameter (10^{-10}m) [7]	*D_0 (m²/s) [8]*	*Deff (m²/s)*	*core*
He	2.56	7.28 10^{-9}	4.51 10^{-10}	2
Ne	2.75	4.03 10^{-9}	1.82 10^{-10}	2
H_2	2.93	4.97 10^{-9}	1.52 10^{-10}	3
Ar	3.4	2.44 10^{-9}	0.80 10^{-10}	2
CH_4	3.82	1.84 10^{-9}	0.90 10^{-10}	2
Xe	3.96	1.47 10^{-9}	0.60 10^{-10}	2
C_2H_6	4.4	1.38 10^{-9}	0.43 10^{-10}	2

3.4 Diffusion experiment with hydrogen

To avoid bacterial activity (conversion of H_2 into CH_4), the entire set-up (except pressure transducers) was irradiated with Co-60 sources during 72 hours, leading to a total received dose of 43 kGy. Despite this sterilisation process, bacterial activity (methane production) was registered from day 76 on. The best fit for D_{eff}, using the 4 datapoints measured before day 76 is 1.52 10^{-10} m²/s (table 2) with a 95% confidence interval 0.56 10^{-10} – 4.14 10^{-10} m²/s. The predicted value for H_2, using equation $D_{eff} = 1.55\ 10^{-9}\ e^{-0.805x}$ is 1.47 10^{-10} m²/s and close to the measured one.

4. Discussion

When we compare the relationships D_0 vs kinetic diameter and D_{eff} vs kinetic diameter, both exponentials factors are similar. This indicates that the diffusion coefficient for a certain dissolved gas in Boom Clay mainly depends on its diffusion coefficient in free water. However, He is an exception for this statement: D_{eff} is much higher than one would expect based on the relationship D_{eff} – kinetic diameter. Further research in literature will be performed on this deviating behaviour. From our study it looks like He is not a good approximation to study H_2 diffusion, Ne is proposed as analogue. Due to bacterial activity the experimental derived D_{eff} for H_2 was only based on 4 datapoints leading to a large uncertainty. However the predicted value corresponds well to the measured value ($1.47\ 10^{-10}$ vs $1.52\ 10^{-10}$ m²/s).

5. Conclusions

The results of the diffusion experiments with dissolved gasses with different kinetic diameter allows to draw a relationship between D_{eff} and the kinetic diameter. By using the obtained equation, we can predict D_{eff} for gasses like H_2, traditionally suffering from experimental difficulties. The measured D_{eff} for H_2 was compared to the predicted value, and both values correspond well.

6. Acknowledgements

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. This work is performed with the technical support of T. Maes and L. van Ravestyn.

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Noble gas mobility in brines: Case study on the salting out effect

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We investigated highly saline groundwater that entered (drowned) salt mining structures in the city of Stassfurt, Germany, to identify the role of groundwater in ongoing subrosion processes in the investigated area (Stadler et al. 2012). In this context, we studied the behavior of the noble gases He and Ne and their isotopes. We found that in the study area He contents in the sampled water did not seem to further increase below a depth of about 100 m b.g.l. Neon concentrations were between half and 2.5-fold the value of solubility equilibrium of the site's pT conditions, which is a larger scatter than generally observed for excess air (Heaton and Vogel 1981, Aeschbach-Hertig, 2001). We found that i) in-situ production of ^{4}He from U and Th in rocks cannot account for the observed measured concentrations alone. In addition, ii) pure diffusion could not explain quantitatively the combination of ^{4}He and salt concentration gradients, but iii) pure (salt-induced) diffusion could explain qualitatively the combination of depth profiles of salt and Ne concentration. In our contribution, we show and discuss that our data on noble gases (especially Ne) give indications that salt-induced diffusive processes are non-negligible as compared to groundwater flow in the aquifers of Stassfurt.

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Working Groups

Part of the symposium was dedicated to five working groups which were each tasked to consider six questions in relation to gas generation and migration. These questions were:

i) **Hydraulic properties:** In your opinion, can gas, under any circumstances, affect the favourable hydraulic properties (e.g. low permeability) of components of the engineered barrier system and the host? If yes, specify how, indicate significance and clarify the circumstances.

ii) **Mechanical properties:** In your opinion, can gas, under any circumstances, mechanically disrupt components of the engineered barrier system and the host rock? If yes, specify how, indicate significance and clarify the circumstances.

iii) **Solute migration:** In your opinion, can gas, under any circumstances, affect water and solute (typically radionuclides) displacement within the engineered barrier system and the host rock? If yes, specify how, indicate significance and clarify the circumstances.

iv) **Chemical properties:** In your opinion, can gas, under any circumstances, chemically affect the favourable properties of components of the engineered barrier system and the host rock? If yes, specify how, indicate significance and clarify the circumstances.

v) **Containment:** In your opinion, can gas, under any circumstances, affect the favourable properties of canisters/overpacks in the repository? If yes, specify how, indicate significance and clarify the circumstances.

vi) **Completeness:** In your opinion, are there other gas related phenomena that could affect the safety functions that are not considered in the other questions? If yes, specify how, indicate significance and clarify the circumstances.

Summary powerpoint presentations of the discussions of the working proups are provided below.

Workgroup 1

Completeness : In your opinion are there other gas related phenomena that could affect the safety functions that are not considered in other questions?

Difficult to separate all the questions : in particular the two first one, Hydraulic and mechanic but also chemical for example with concrete

→ Mainly all processes are coupled

What is missing in FORGE salt, granite… radionuclides transport, few things on concretes

> → All different materiel have to be considered different behavior between for example: bentonite, concrete conclusion will not be the same

How Scientific results are transferred to PA?
Integration at the different scales from lab to large scale test and to repository
Time scale and processes evolution on long durations: ex EDZ and self-sealing

Generation of gas realistic term and times dependent evolution in some cases is still unknown: coupling with water availability

Definition of the main terms use in the project : two phase flow, gas breakthrough… (→ conclusions might be misleading)

Need a criteria to estimate if gas in an issue (relative to integrity or other….) minimum pressure but also saturation time? Impact of gas on materiel water saturation

Optimization of the repository is a solution: examples new packaging, EGTS in Nagra, large buffer volume for gas expansion…

How gas pressure could affect the rock properties, hydraulic, mechanical and solute migration coupling ?

Effective pressure concept which parameters should we use? Biot coefficient ?

Benefic contribution of gas pressure, ex → limit degradation by corrosion

Chemical:

Hydrogen embrittlement was not part of FORGE

Radionuclides transport in liquid and in gas phase should be more investigated : different pathways in clay and fracture media, salt ?

Chemical properties : clogging and carbonation of concrete permeability (increase or lower permeability)

Modification due to gas of the pH on solubility and mobility of radionuclides?

What is necessary for performance assessment ?

- How much do we need to know?
- Understanding the processes
- In some areas prediction is problematic but is that really a problem for PA?
- What do we have to predict?
- At the moment, models are able to reproduce experimental results

Models

Benchmark at different levels could be useful: physical model comparison, blind predictions,

Validation of the code and the model

WG2 Feedback

FORGE Symposium

Gas Generation and Migration

Luxembourg

6 Feb. 2013

nagra.

Hydraulic properties

- Lot of evidence from URL experiments and lab experiments that clay rocks have very good resealing characterisitics. After gas paths form and gas pressure is reduced
  - Self-sealing is observed
  - Hydraulic properties return to the original state
- Residual negative effects on hydraulic properties have not been observed for clay rocks or bentonite barriers
- For porous grouts in LILW, gas can displace water, thus lowering diffusive transport rates, but water could also be expelled
- Not an issue for crystalline rock
- In salt, trapped gas can slow or limit convergence, leaving salt more hydraulically permeable – impact would depend on scenario
- If gas phase forms and C-14 is present as e.g. methane, C-14 could be transported with a moving hydrogen/methane phase

Mechanical properties

- High gas pressures can create dilational pathways in rock – EDZ and bedding planes (but resealing is observed)
- Some bentonite gas studies evidence consolidation – transition from consolidation to pathway formation not well understood, but probably this is not ‘disrupting’

Solute migration

- Evidence from experiments that there is little water displacement by gas
  - Studies on bentonites – almost no water displaced
  - Studies on Boom Clay – iodide transport enhanced only very slightly
  - Studies on other clay rock cores – no significant displacement
- HG coupling scenarios possible
  - Displacement of water in canister by gas pressure rise
  - Displacement of water in porous grout
  - Gas keeping water out of a porous grout
- Negative or positive effects – dependent on materials, inventories, timing of phenomena

Chemical properties

- Carbonation of cement by CO_2 can use up buffering capacity, decreasing pH
  - Potential loss of passivation of some metals
  - Changing radionuclide K_d values and solubilities
- H_2 can theoretically reduce Fe(III) in bentonite to Fe(II), possibly changing swelling properties and hydraulic conductivity
- H_2 can be consumed by SRB, precipitating sulphide, which might affect other processes – not necessarily a negative effect
  - Sulphide could affect corrosion
  - Gas pressure could be reduced

Containment

- High gas pressure at a canister external surface likely could not affect structural integrity of a SF/HLW canister (LILW drums perhaps, if they have void space)
- Some metals take up H and HIC might occur – not so relevant for copper or carbon steel canisters

Completeness

- Are there as yet undiscovered processes?
- Water consumption by corrosion and high gas pressures leading to reduced RH and suppression of corrosion – not discussed in FORGE but looked at in clay repository programs
- Combinations of processes may be relevant e.g H_2 good for stabilizing spent fuel, but can other processes negate the positive effect (e.g. increased water flow in a fractured rock system)
- Conversion of hydrogen to methane may be good to lower total pressure but may produce $^{14}CH_4$
- Coupled HMCG processes are complex – for any system the system evolution and processes have to be looked at carefully

Working group 3

Hydraulic properties in our opinion

How gases can affect H-properties?

- **Cement** gets sealed by carbonation due to CO_2 flow.
- Large amounts of gases increase **clay**s' permeability
- The induced mechanical state in different **host rocks** (crystalline vs. claystone) may cause different and complicated alterations.

Significance?

- Gas needs to be released before it compromises the integrity of barrier system.
- We don't know the limits of acceptability

Directions?

- Compilation of full data base
- Descriptions/models for (host's) discontinuities (EDZ, etc.)
- significance of gas flows during partially saturated (= higher permeability) conditions
- Replacement of Darcy
- Vocabulary: should we continue relating pathway dilatation to an increase in permeability?

Containment in our opinion

Can gases can affect the properties of waste containers?

- Generally not considered to have an effect.
- However, we feel not being competent to evaluate this topic any further.

Solute migration in our opinion

How gases can affect water and solute migration?

- CO_2 changes retention properties
- What's Cl- 's migration velocity wrt gas velocity

Significance?

- Before dilatant macro fractures are opened, micro fractures have released pressure.

Directions?

- Tracers are transported in clays slowly, but upscaling to repository scale is not known.
- E.g. tortuosities are fairly carelessly applied for materials&solutes for which they are not measured for
- Description of migration in "dilatant paths"
- Migration of radioactive gases
- migration in non-dilatant components - dilatant displacements are not foreseen e.g. in sands and concrete
- Are existing fractures connected?

Chemical properties in our opinion

How gases can affect chemical properties?

- There is H_2 reactivity, but some H_2 induced reaction paths are affected by microbial processes.
- Porewater chemistry (solubilities, complexation, ...) is altered influencing RNT
- extensive literature available (from 1970's @ Germany)

Significance?

- Information the extents of generated gases would be useful.

Directions?

- Assessment of thermodynamic constraints for chemical changes.
- H_2 reactions

Mechanical properties in our opinion

How gases can affect mechanical properties?

- Unlikely to exceed tensile strength.
- Boom can self-seal but not self-heal

Significance?

- Mechanical model for clays missing, so that it's difficult to assess significance of saturation/drying
- "Where does gases go and why?" - if they dissolve, does it matter?
- Information the extents of generated gases would be useful.

Directions?

- C-changes induced M-changes in cement not known
- FORGE test results are not integrated into models: Models for interfaces and dilatant paths (in addition to Lasgit-code_bright)
- make test data publicly available
- is there enough data to make proper process model?

Completeness in our opinion

Are there any other gas related phenomena not considered?

- microbial activity
- gas migration HMC-effects in partially saturated conditions
- system optimisation to make EBS to perform certain gas related tasks (like gas collection)
- uncertainty management
  - more data/investigations
  - show irrelevancy / other ways to go around
- gas consuming processes

Advances

- self-sealing has been quite well been demonstrated?
- are radioactive gases generated in significant amounts?
- is overall buffering capacity affected?
- does small deficiencies mean that we can't use the current global models?
  - e.g. there are no pure 2-phase flows in EDZ?
  - are conservative assumptions sufficient to cover these kinds of deficiencies? (extend flow path instead of early dissolution)

FORGE

Fate Of Repository Gases

European Commission FP7

FORGE International Symposium and Workshop

Working Group 4 Report

J.D. Barnichon, M. Birgersson, N. Calder,
S. Granet, J. Harrington, K. Liberda,
S. Olivella, D. Pellegrini, C. Rochelle,
J. Rueedi, X. Sillen, D. Stammose

Luxembourg, February 7, 2013

FORGE
Fate Of Repository Gases
European Commission FP7

Working Group 4 report

- **Caveat: questions asked to WG are rather generic but WG notes that for gas, answers tend to be more specific, i.e. very much depend on**
  - material
  - conditions (saturation,…)
  - disposal concept
  - repository layout
  - …

- **WG answers are broadly structured distinguishing EBS components (bentonite, concrete) and host rock (disturbed / undisturbed)**

- **4 (out of 6) questions addressed: fate of mechanical and hydraulic properties, gas impact on solute migration and chemical properties**

Fate of mechanical properties ?

- **Bentonite, saturated**
  - High gas pressures and bentonite consolidation can be observed in some lab experiments. Due in part to geometry of test setup but consolidation cannot be ruled out in repository conditions.
  - Confident about pressure at which gas start to move. However, Peak gas pressure are less well defined.
  - Depending on material, injection test conditions (sequence), successive breakthrough events observed to occur at comparable or different pressures (for instance, repeated breakthrough that occur at a lower pressure than the first) but the latter case does not imply significant changes to mechanical properties.
  - Nature of (swelling!) material is such that it self-heals

- **Bentonite, unsaturated ➔ saturated**
  - Uncertainties remain about homogeneity in the long term of bentonite.

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Fate of mechanical properties ?

- **Cementitious EBS components**
  - Engineered material ➔ Mechanical disruption not expected if cement designed to be permeable enough to dissipate gas.
  - Mechanical properties change are expected if exposed to reactive gas (e.g. CO_2 ➔ carbonation ➔ CHM coupling)
  - Structural failure of concrete plug (at the end of a disposal drift…) ?
    - Design-dependent but host rock (clay) likely to act as relief valve before plug failure

Fate of mechanical properties ?

- **Host rock**
  - Remark: no classical mechanical tests on samples after gas flow experiments – could be indicative of perceived (lack of) significance of mechanic strength of host rock (post-closure, at time of gas transport)

  - Most mechanical damage done <u>during excavation</u>
    - No additional discontinuity creation expected in the excavation damaged zone (EDZ)
    - If significant self-sealing can occur before gas pressure build-up, formation of new pathways cannot be ruled out.

  - Gas transport through <u>path of less resistance</u>
    - Intrinsic host rock features: HG-C, dilation measurement suggest opening of bedding (as expected).
    - EDZ (unavoidable, not easily characterized) can play an important role (may depend on when gas is generated, "longevity" of DZ)

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Fate of hydraulic properties ?

- **EBS components**
  - Bentonite based
    - Saturated bentonite: no permeability increase, consolidation is possible (lowers permeability, storage)
    - Unsaturated bentonite: high mobility of gas is noted, could be an issue for radioactive gas
  - Cementitious materials
    - CO_2 through high pH concrete ➔ carbonation

      ⇒ Permeability decrease, except at small scale where reaction is actively happening
      - Transferability to low pH concrete?
      - Will the concrete end up being more brittle (thus more easily damaged by stress change later, leading to higher permeability)? Not done yet.

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Fate of hydraulic properties ?

- **Host rock**
  - Hydraulic conductivity: if water available, self-sealing of damaged zone around excavations is generally expected in clay host rocks.
    - well documented in the lab and in situ
    - rock and concept-dependent
  - Gas permeability?
  - Possible role of microbes in (open) fractures: availability of CH_4, H_2 may promote microbial activity ➔ formation of biofilms ➔ clogging

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Effect on solute migration ?

- **Gas-driven (contaminated) water displacement ?**
  - Concept, materials dependent
  - in FORGE (experiments), gas flow through low-permeability porous media induced only very little porewater flow.
  - Bentonite consolidation: water (which can be contaminated depending on timing) is being pushed out
  - In unsaturated conditions, less (or no) water available for solute transport but high mobility of inactive (carrier) gas and volatile radionuclides

- **Gas flows through <u>localised</u> pathways in clays**
  ➔ little effect of gas on <u>bulk</u> porosity and D_{eff}

- **Indirect, secondary effect if gas promotes microbial activity ➔ perturbation of porewater chemistry, can affect RN speciation.**

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Chemical properties ?

- **Cement**
  - CO_2 (from LILW) ➔ carbonation ➔ lower pH
    - loss of passivation
    - radionuclide speciation changes
    - Sorption characteristics (mineral phases) can change
    - Incorporation of some radionuclides (^{14}C, ^{36}Cl) within new phases
- **Bentonite, Host Rock**
  - CO_2 release in host rock can make water more acidic
  - Specific experiments, thermodynamics, suggest that suction (in unsaturated conditions) may affect thermodynamic constants ➔ precipitation of new mineral phases ?

Working Group 5

Gas Generation and Migration

International Symposium and Workshop

Luxembourg, 5 February 2013

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Nuclear Decommissioning Authority

EURATOM

Question 3 (in the context of deep disposal)

- <u>**Solute migration**</u>**:** In your opinion, can gas, under any circumstances, affect water and solute (typically radionuclides) displacement within the engineered barrier system and the host rock? If yes, specify how, indicate significance and clarify the circumstances.
- **Majority of gas generation processes require water – availability of water affects gas generation rate and duration of gas source term. Type of gas generated is a function of the inventory.**
- **Bulk and radioactive gas generation could occur early in post-closure period of a repository, when majority of non gaseous radionuclides are likely to be in-container still (in wasteform). Free gas vs dissolved gas.**
- **Need to know canister failure scenario – does time of failure coincide with e.g. resaturation of a repository to an evolved steady state? Gas generation affects rate of resaturation.**
- **Evolving Eh and pH conditions can affect radionuclide migration in a positive way (effect of reducing conditions on actinide migration) or negative way (loss of alkaline conditions). Carbonation.**

Question 3 (in the context of deep disposal)

- <u>**Solute migration**</u>**:** In your opinion, can gas, under any circumstances, affect water and solute (typically radionuclides) displacement within the engineered barrier system and the host rock? If yes, specify how, indicate significance and clarify the circumstances.
- **Repository-scale calculations (ANDRA et al) indicate some gas-driven groundwater movement along plug / seal interfaces on scale of several 10's m over 10k years+**
- **Gas will exploit interfaces ('weakest' zones, higher k), will gas migrating through such localised features influence solute migration on a repository scale?**
- **Overall - depends on inventory, disposal concept, geology etc – could have a degree of interaction between waste-derived gas, water and solutes. c.f. Q(iv) re Chemical Properties.**

NDA

EURATOM

Question 1 (in the context of deep disposal)

- **<u>Hydraulic properties</u>:** In your opinion, can gas, under any circumstances, affect the favourable hydraulic properties (e.g. low permeability) of components of the engineered barrier system and the host? If yes, specify how, indicate significance and clarify the circumstances.
- **Key issue is gas pressure.**
- **Cannot reasonably engineer a seal / plug such that pressurisation can arise to the extent that hydraulic properties of the host rock can be affected (gas fracking not likely).**
- **For volatile radionuclides, gas pressurisation maintaining open pathway via EDZ could significantly reduce transport time from source to geosphere.**
- **Gas pressurisation will delay the self-sealing of bentonite and EDZ Transient effect.**
- **?Acidic gas generation affecting carbonate content in a rock and therefore porosity?**
- **Carbonation affects rock properties (reduced poroperm) c.f. Q(iv) re Chemical Properties.**

NDA

EURATOM

Question 2 (in the context of deep disposal)

- **<u>Mechanical properties</u>:** In your opinion, can gas, under any circumstances, mechanically disrupt components of the engineered barrier system and the host rock? If yes, specify how, indicate significance and clarify the circumstances.
- **As per Question 1 re effects of gas pressurisation. Mechanical properties could be affected by e.g. carbonation.**
- **During desaturation period, there will be a stress redistribution**
- **EDZ and engineered barriers will have different properties to resaturated state.**
- **Cycles of gas pressurisation (higher than hydrostatic) and depressurisation might lead to stress redistribution.**

Question 4 (in the context of deep disposal)

- **<u>Chemical properties</u>:** In your opinion, can gas, under any circumstances, chemically affect the favourable properties of components of the engineered barrier system and the host rock? If yes, specify how, indicate significance and clarify the circumstances.
- **Overall - depends on inventory, disposal concept, geology etc**
- **Carbonation affects rock properties (reduced poroperm)?**
- **Acidic gas generation affecting carbonate content in a rock and therefore porosity?**
- **Evolving Eh and pH conditions can affect radionuclide migration in a positive way (effect of reducing conditions on actinide migration) or negative way (loss of alkaline conditions). Carbonation.**

Question 5 (in the context of deep disposal)

- **<u>Containment</u>:** In your opinion, can gas, under any circumstances, affect the favourable properties of canisters/overpacks in the repository? If yes, specify how, indicate significance and clarify the circumstances.
- **Some containers are vented, so are not designed to be gas-tight**
- **For gas-tight containers, need to consider over-pressurisation and possible breach if there is a reasonable and viable internal gas generation mechanism (e.g. corrosion of furniture in spent fuel canister mediated by carry-over water; He generation from radioactive decay)**
- **Explosion / flammable event in the pre-closure phase? (repository management is out of spec if this occurs)**
- **Formation of acidic niches? But this is not a gas issue**
- **In desaturated geology (no or limited water contacting waste), corrosion rate will be lower – containment will be prolonged**

Question 6

- **<u>Completeness</u>:** In your opinion, are there other gas related phenomena that could affect the safety functions that are not considered in the other questions? If yes, specify how, indicate significance and clarify the circumstances.
- **Have not responded re "Significance" on any of Q1 to Q5, as this is determined by a safety case and is not a black and white issue**
- **We have answered the 5 Qs of the basis of phenomenology, biased towards post closure period – gas management in the pre-closure period is a challenge, dependent on source term and concept.**
- **Gas issue is understood to an adequate level, and can largely be managed by choice of engineered barrier system in complement with role of geology to achieved desired function of isolation and containment, so long as you have a good handle on the inventory, geology**
- **Our responses are on the basis of a deep geological repository - response could be different if a surface or shallow disposal facility for low level or short-lived intermediate level waste were under consideration.**
- **Gas generation on stability of spent fuel not considered – has a positive effect.**

Question 6

- **Completeness:** In your opinion, are there other gas related phenomena that could affect the safety functions that are not considered in the other questions? If yes, specify how, indicate significance and clarify the circumstances.
- **Supercritical fluid formation – is this a reasonable FEP for a deep geological repository given the rate of gas generation, volume, pressure? Unlikely (very) to be an issue for hydrogen and methane, at least theoretically possible for carbon dioxide. "So what"? Sudden phase change? Effect on e.g. rate of carbonation**

Spare text

- Gas issue can be engineered out, so long as you have a good handle on the inventory, geology – can design and operate a repository such that gas in the post-closure phase is not a significant issue.