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Fate Of Repository Gases

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Synthesis Report: Updated Treatment of Gas Generation and Migration in the Safety Case

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	Name	Organisation	Signature	Date
Compiled	S. Norris	NDA-RWMD		30th September 2013
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Synthesis Report: Updated Treatment of Gas Generation and Migration in the Safety Case[1]

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Work Package leader: Simon Norris

Authored by:

Belgium	Frank Lemy & Charles-Aymeric del Honeux (FANC-AFCN), Geert Volckaert, Eef Weetjens, Katinka Wouters (SCK-CEN),
France	Jacques Wendling (ANDRA), Magdalena Dymitrowska & Delphine Pellegrini (IRSN)
Sweden	Patrik Sellin (SKB)
Switzerland	Lawrence Johnson (Nagra), Manuel Sentis (ENSI)
United Kingdom	Jon Harrington (BGS), Simon Norris (NDA RWMD)

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EC Project Officer: Date:

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Fate **O**f **R**epository **G**as**E**s (FORGE)

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. Key issues to be further examined relating to an enhanced understanding of gas-related processes include: dilational versus visco-capillary flow mechanisms; long-term integrity of seals, in particular gas flow along contacts; role of the Engineered Disturbed Zone (EDZ) as a conduit for preferential flow; and laboratory to field up-scaling. Such issues are the focus of the integrated, multi-disciplinary European Commission FORGE project.

The FORGE project links international radioactive waste management organisations, regulators and academia, and is specifically designed to tackle the key research issues associated with the generation and movement of repository gases associated with waste disposed in a geological repository. 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.

Contact details:

EC FORGE Work Package 1 Leader:

Dr Simon Norris

Nuclear Decommissioning Authority – Radioactive Waste Management Directorate

Tel +44 (0) 1925 802891 / +44 (0) 7887 775309

Fax +44 (0) 1925 802948

Email simon.norris@nda.gov.uk

Web address: www.nda.gov.uk

Address NDA Harwell
Building 587
Curie Avenue
Harwell Science and Innovation Campus
Didcot
Oxon OX11 0RH
United Kingdom

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FOREWORD

The multiple barrier concept is the cornerstone of all proposed schemes for underground disposal of radioactive wastes. The concept invokes a series of complementary barriers, both engineered and natural, between the waste and the surface. Implementing this concept, from site appraisal and characterisation to repository design and construction, is the primary objective of all disposal programmes.

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-scale to field-scale 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.

FORGE was targeted to address these issues through a series of laboratory-scale and field-scale experiments, including the development of new methods for upscaling allowing the optimisation of concepts through detailed scenario analysis.

This report summarises overall understanding on the implications of gas generation and migration for the safety functions of the Engineered Barrier System (EBS) and the host rock as informed by the output of the FORGE project.

Achievements and recommendations from the FORGE project are noted, emphasising that FORGE has provided confidence in the basic physical understanding and modelling approaches relevant to repository-derived gas that can be applied for different concepts – such understanding, and the derived component and system models, can be used in the safety case to address regulatory guidance.

EXECUTIVE SUMMARY

The European Commission FORGE (**F**ate **O**f **R**epository **G**as**E**s) project is a pan-European project with links to international radioactive waste management organisations, regulators and academia, specifically designed to tackle the key research issues associated with the generation and movement of repository gases.

This report, a key milestone deliverable of the FORGE project, links new understanding derived throughout the duration of the project with its consideration in the safety case for the geological disposal of radioactive waste. Considering the output from the EC FORGE project, the overall key message in relation to the updated treatment of gas generation and migration in the safety case are noted below. Progressing beyond FORGE, these key messages and the outcomes from the EC FORGE project now need consideration in safety case studies undertaken by national implementing organisations, to ensure the derived understanding is embedded and to ensure repository-derived gas is managed such that the safety case is compliant with national regulatory requirements.

Key messages:

- 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).

- 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.

- Based on studies undertaken in the EC FORGE project, and on input from complementary studies, we have enhanced our understanding of repository-derived gas in relation to a range of concepts for the geological disposal of radioactive waste. Such understanding provides a justification for increased confidence in analyses of the gas issue as undertaken within the safety case.

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1 INTRODUCTION

The European Commission FORGE (**F**ate **O**f **R**epository **G**as**E**s) project is a pan-European project with links to international radioactive waste management organisations, regulators and academia, specifically designed to tackle the key research issues associated with the generation and movement of repository gases.

This report, a key milestone deliverable of the FORGE project, links new understanding derived throughout the duration of the project with its consideration in the safety case for the geological disposal of radioactive waste. Work undertaken in the FORGE project will benefit a range of customers, e.g. implementers, regulators, industry and academia, via the provision of new information and understanding into gas-relevant features, events and processes (FEPs), including mechanisms governing gas generation and migration, for consideration in the safety case.

1.1 Multiple Barrier Concept for Geological Disposal

A safety case for a repository is a set of claims concerning the environmental safety of the geological disposal of radioactive waste in a repository, substantiated by a structured collection of arguments and evidence. Such a safety case needs to address environmental safety at the time of disposal and in the long-term, after wastes have been emplaced and the facility has been closed. Materials emplaced underground will slowly degrade and even the most stable geological environments will eventually change with the passage of geological time; the hazard potential of the wastes also decreases by radioactive decay. A safety case looks at the balance of these processes so that we can evaluate the environmental safety of a repository far into the future, as well as at the time of disposal.

An intrinsic part of developing a safety case for a geological repository is the consideration of the various FEPs that could affect the long-term environmental safety of the repository. This can be done by identifying FEPs that are relevant to the performance of a particular disposal concept, or using existing international FEP lists, with intention being to analyse systematically all potentially relevant FEPs, thereby ensuring that the safety case is comprehensive in its coverage. Development of process models, component models and a total system model are subsequent aspects of a safety case.

The multiple barrier concept is the cornerstone of all proposed schemes for the underground disposal of radioactive wastes. Based on the principle that uncertainties in performance can be minimised by conservatism in design, the concept invokes a series of complementary barriers, both engineered and natural, between the waste and the surface environment. Each successive barrier represents an additional impediment to the movement of radionuclides.

For low and intermediate level waste (L-ILW), the waste may be incorporated in a relatively stable and inert matrix such as cement, bitumen, lead-alloy, polymer resin (the choice varying depending on the waste management organisation); glass may be used in the case of certain high level (HLW) reprocessing wastes. Due to the very low leach-rate of glass in groundwater, vitrification is widely accepted to be one of the best methods of immobilising the aqueous products from the reprocessing of spent fuel (SF) [1]. Many waste containers will provide some form of physical barrier to groundwater. However, because of the relatively small volumes of waste involved, spent fuel, vitrified waste and other highly active wastes will be totally encapsulated

in corrosion-resistant metal canisters which are designed to prevent groundwater entry for very extended time periods in excess of 100,000 years [2].

Depending on the disposal concept, engineered barriers may comprise the buffer/backfill medium enclosing the waste containers, the tunnel/borehole liner, and the backfill and high integrity seals placed in the repository access ways or emplacement boreholes. The buffer/backfill medium enclosing the waste will often also provide both a physical and a chemical barrier to radionuclide migration. The functions of the engineered/chemical barriers are:

- To reduce the rate of corrosion of the waste containers and thus extend their life;
- To limit the rate of hydraulic transport;
- To limit the release of radionuclides from the waste-form to the far-field (geosphere) after container failure;
- To limit the migration of radionuclides along the pathway provided by the access tunnels and shafts of a repository or the boreholes in the case of a deep borehole emplacement.

For L-ILW disposal in vaults, the backfill may be a porous, cementitious grout, which is intended to pH-buffer the pore water for an extended time period. Typically the buffer/backfill for HLW/SF might comprise compacted bentonite or other clay-based material, providing a low permeability, alkaline pH-buffered pore water to limit solubility and mobility of certain radionuclides (e.g. actinides), plus good retention/retardation properties including high sorption and a capacity to filter colloids.

The geological barrier is the final impediment to radionuclide migration. Depending on details of the local geology, this may be considered to constitute the host formation itself, extending above, below and laterally away from the repository. Alternatively, the entire sequence of low permeability rocks, which may separate the repository from the surface and/or more permeable, water-bearing, strata may be included. The practical realisation of the multiple barrier concept is the primary objective of all stages of a disposal programme, from site appraisal and characterisation through to design and construction. The general performance of the repository as a whole (waste, buffer, engineering disturbed zone, host rock), in particular its gas transport properties, is being intensively studied in many national programs. Issues relating to basic process understanding related to gas transport such as dilational versus displacement flow mechanisms, the long-term gas transport characteristics of seals, gas flow along interfacial contacts and up-scaling from laboratory to field conditions are a particular focus of study.

1.2 Gas in the Context of Geological Disposal of Radioactive Waste

Within a repository and assuming the availability of groundwater, corrosion of various metals and alloys, in particular ferrous materials, under anoxic conditions will lead to the formation of hydrogen. Radioactive decay of the waste and the radiolysis of water particularly will produce additional gas. If present, biodegradable wastes will produce carbon dioxide and methane through microbial action and other (minor) gaseous species may also be generated. If groundwater is not available, the gas generation rate will be very low to insignificant, although the potential gas source term itself (e.g. ferrous materials), will persist. If the gas production rate exceeds the rate of diffusion of gas molecules in the pores of the engineered barrier or host rock, the solubility limit of the gas will eventually be exceeded, leading to the formation of a discrete gas phase. Gas would continue to accumulate until its pressure becomes sufficient for it to enter the engineered barrier or host rock. Understanding gas generation and

migration is thus one of the key issues in the assessment of repository performance [3] and is the focus of the FORGE project.

The generation of gases in the repository environment is of concern for a number of reasons [4]:

- Pressurisation of waste containers;
- Perturbation of any groundwater flux;
- Effect on repository backfill and seals;
- Effect on Excavation Damage Zone (EDZ) and self-sealing properties;
- Effect on host-rock mass transport properties;
- Effect on heat dissipation;
- Release of active gases;
- Displacement of contaminated groundwater.

The impact of gas generation on repository infrastructure and evolution has been the source of several pan-European projects/initiatives during the last decade including GASNET. Of particular importance was the fundamental review undertaken by Rodwell et al. (1999) [4] which assessed the current state-of-the-art knowledge in relation to gas generation and migration in repository systems and the GASNET (2003) [5] project which dealt with the treatment of gas in repository performance assessment.

These projects identified a number of uncertainties linked to the quantitative treatment of gas generation, migration and subsequent evolution of repository systems. Uncertainties indentified in GASNET (2003) [5] of particular importance to FORGE are:

1. The definition of long-term corrosion rates of ferrous metals under repository conditions (considered in FORGE Work Package 2)
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)
3. The effect of elevated gas pressures on the movement of groundwater and aqueous borne contaminants (considered in FORGE Work Package 4)
4. The role of gas on the evolution of the near field and the EDZ (considered in FORGE Work Package 4)
5. The possible coupling of effects to compromise repository performance (considered in FORGE Work Package 1)

To address these fundamental issues, the FORGE project has been 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 helps to 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.

In the ideal situation, gas would be in solution in the host-rock groundwater and would be transported away from the repository by advection / diffusion processes. Furthermore, attenuation mechanisms (e.g. chemical reactions) in the host-rock could result in depletion of the flux of gas in solution so that, at some distance from the repository, the radiological significance of the flux could be minimal. However it is ironic that the sought-after characteristic of the host-medium of very low permeability

becomes somewhat problematic in the context of gas migration. Calculations of the probable maximum diffusive flux of gas (in solution) in a clay suggest that Fickian diffusion in the host-medium may be too slow a mechanism to accommodate the quantity (and rate) of gas produced in a repository [6]. This is, of course, highly dependent on specific details of the repository design and the waste inventory. Nevertheless, there is a strong possibility that a gaseous phase will accumulate in the available void-space within a repository resulting in a rise in gas pressure.

It seems highly unlikely that the high integrity seals constructed at key points in the repository could be engineered to withstand such a substantial differential pressure for an extended time-period. As gas pressure rises, it is inevitable that the gas entry pressure will be reached for the rock and /or repository sealing system. This may have consequences for the repository design and safety assessment, thus this aspect needs to be carefully examined. In low-permeability formations, gas production and migration is likely to be an important issue, since these rocks cannot easily accommodate the gas flux. If gas production is likely to be significant, then its possible repercussions must be examined as an integral part of the host-rock/site selection process.

To fully understand the impact of gas generation and transport on current engineered barrier concepts, its effect on self-sealing within the EDZ and its long-term effect on the hydrogeological characteristics of the far-field, requires an integrated, multi-disciplinary project to address these key research areas.

The Work Packages in the FORGE project 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.

Figure 1 summarises the linkages between these work packages. The experimental emphasis of the proposed work programme focuses on process understanding and the measurement of key generation, transport and hydromechanical properties governing the movement of gas from the canister surface through the Engineered Barrier System (EBS) and in to the far field. Laboratory and field-scale measurements will be performed in eleven European countries in support of this activity.

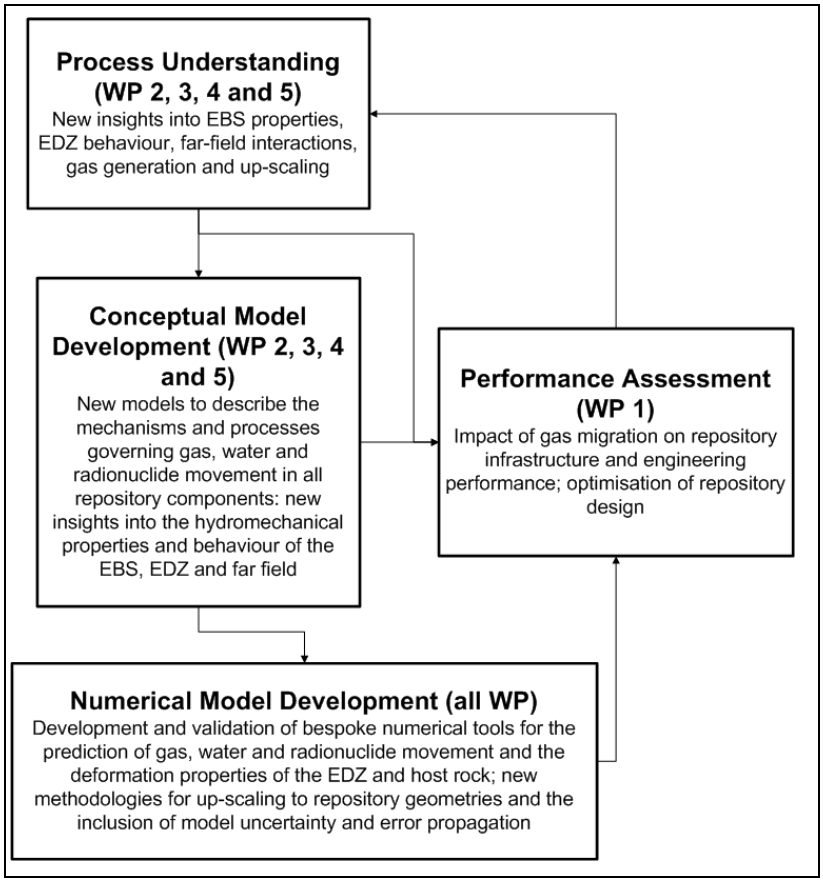


Figure 1 FORGE project: basic scientific linkages and knowledge development beyond the current state-of-the-art.

1.3 Structure of this Report

Following this **Section 1** Introduction, **Section 2** of this report provides information on the Work Packages of the FORGE project, noting how they are inter-related and designed to provide an information flow throughout the project. This relationship between the Work Packages is intended to ensure, for example, that the development of understanding of gas processes at a research level is reflected appropriately in the treatment of gas in performance assessment, and that the research studies themselves are focussed on key performance assessment issues.

Section 3 presents a summary of gas-related issues in the context of geological disposal, considering various geological disposal concepts, the related safety functions and the treatment of gas issues linking to repository design and optimisation.

Section 4 reports on developments in understanding gas generation and migration across FORGE Work Packages over the duration of the project, in the context of key safety case-relevant issues where input has been derived from Work Package participants via a questionnaire developed by Work Package 1. Relevant recent work from outside of FORGE is also reported, for completeness.

Section 5 summarises overall understanding on the implications of gas generation and migration for the safety functions of the EBS and the host rock. Linking to experimental observations in Work Packages 2-5, and modelling work from WP1.2

and 1.3, this section draws conclusions on how gas issues are to be treated in the safety case, including related uncertainties.

Section 6 presents the achievements and recommendations from the FORGE project, emphasising that FORGE has provided confidence in the basic physical understanding and modelling approaches relevant to repository-derived gas that can be applied for different concepts – such understanding, and the derived component and system models, can be used in the safety case to address regulatory guidance.

This report was produced over the period November 2012 to September 2013, according to the revised schedule of the EC FORGE project. It meets FORGE Milestone 68, and supersedes FORGE Deliverable D1.2-R (which was submitted to the EC in July 2009) and FORGE Milestone 15 (which was submitted to the EC in January 2010) [7].

2 EC FORGE PROJECT: DESCRIPTION OF WORK

This section provides information, on a Work Package by Work Package basis, of the activities that collectively form the work undertaken in the FORGE project. Work under each Work Package is detailed under the headings 'Objectives' and 'Description of Work'. This text is derived from the original Description of Work submitted as part of the FORGE proposal to the EC.

2.1 Work Package 1 Treatment of Gas in Performance Assessment

2.1.1 WP1.1 State of the Art – Gas Generation and Gas Migration

At the outset of the project, the stated purpose of this Work Package was to:

- Describe the current treatment of gas issues in long-term safety assessments for intermediate-level waste, high-level waste and spent nuclear fuel. Note significant input to this task is already provided by the output of the EC GASNET project [5] – this task therefore sought primarily to present an update on progress subsequent to that project;
- Identify the merits and shortcomings of the current treatment of gas issues in long-term safety assessments;
- Analyse the limitation of the previous studies and the different types of uncertainties related to the gas transport in host rock and engineered barriers;
- Discuss the needs for additional studies of gas migration issues and how they can support future assessments. Identify relevance of work being progressed by FORGE WPs 2-5, and the expected benefit to be gained from this work.

Throughout the duration of the project, the stated purpose of this Work Package was to:

- Integrate the information produced by this and the other WPs throughout the duration of the project, considering the implications for the current state of the art;
- Examine the extent to which other WPs' findings provide satisfactory answers to the needs identified at the outset of the project;
- Define "scenarios" related to gas issues. Discuss the uncertainties related to the scenarios. Identify the need of models and data for the treatment. Link to work in the EC PAMINA project looking at uncertainties in the consequences of repository-derived gas;
- Make recommendations for, and propose an updated treatment of, gas issues in relation to long-term safety assessments, based on the integrated findings from the project. This will consider, for example, issues to be dealt in a safety case and future needs, and a set of recommendations related to the design of the repository in relation to gas issues.

The current report is the conclusion of FORGE Work Package 1.1, and presents the achievements of the work package. The above bullet points are considered, but the report is more wide ranging and reflects new understanding regarding gas generation and gas migration that has been developed as part of the FORGE project, and outside the FORGE project where available for comprehensiveness, that now can be considered by e.g. implementing organisations in their development of safety cases that appropriately contextualise the gas issue and provide a treatment of it that attains respective regulatory requirements.

2.1.2 Work Package 1.2 Repository-scale Numerical Simulations of Gas Migration

(a) Benchmark Studies

A series of benchmark studies on repository-scale numerical simulations of gas migration were undertaken. The test cases considered in this task were defined by WP 1 participants at the outset of the project. These studies aimed 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. The benchmark studies were not repository-specific.

(b) Repository Scale Gas Migration Calculations

The aim of this task was to undertake a suite of calculations to progress understanding in repository-scale gas migration, linking to the output of WPs 2-5, throughout the duration of the project, as appropriate. These calculations were 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. The work undertaken includes:

- Development of methodologies for dealing with heterogeneities;
- Development of high performance calculation methods to handle large mesh sizes and complex geometries; Investigation of upscaling methodologies to simulate gas migration at repository scale;
- Integration of ‘small scale’ information into km-scale model.

2.2 Work Package 2: Gas Generation

This work package examined the rate of hydrogen production in order to provide information in support of repository performance assessment. The key processes to be better understood are the impact of radiation on near field materials and the role of chemical processes, in particular the corrosion of metals. Experiments have measured hydrogen production rates from the corrosion of steel in contact with bentonite under different test conditions, improving our understanding of these issues. Understanding the effectiveness of processes, which may mitigate the volume of hydrogen close to the point of origin, are an important input for assessing the influence of the global evolution of hydrogen generation within the repository. Short and longer term irradiation experiments were undertaken in parallel to understand the influence of the boundary conditions of the tests and the temporal evolution of parameters.

The expected outcome of these studies is to provide a more accurate assessment of the kinetics of hydrogen production and its migration than is available at present. This knowledge is an important factor in the understanding of the likelihood of the development of discrete gas phase within a repository. These processes have been addressed by experimental and numerical modelling and specific issues studied include:

- Consumption of hydrogen by reaction with radiolytic products such as hydroxide ions;
- Inhibition of corrosion by the products of radiolysis;
- Effect of water chemistry e.g. the influence of carbonate reactions with hydroxide ions;
- Effect of clay/cement barriers on hydrogen production and its migration through samples under irradiation.

The sensitivity of these processes on hydrogen production is investigated.

2.3 Work Package 3: Engineered Barrier and Seals

Gas generation from either the waste form or the engineered barriers is an unavoidable but generally undesired effect in most European repository concepts for radioactive waste. Gas generation and migration can potentially alter the hydraulic and mechanical properties of the repository (possibly the thermal and chemical properties as well). The purpose of this work package was to investigate gas migration processes and the consequences of gas migration in the EBS of the repositories. The WP delivered results that can be used for:

- Direct qualitative and quantitative confirmation of the consequences of gas migration to be used in long-term safety assessments;
- Scientific knowledge about the gas migration processes to be used in the development of conceptual models;
- Quantitative data to be used in the development and testing of numerical models for the simulation and prediction of gas migration and its consequences.

The work in this work package will be divided into 4 areas:

- **Bentonite Underground Rock Laboratory (URL) Experiments**: Field-scale experiments with bentonite buffers and seals that can be used directly as a confirmation in safety assessment and also give important information on the effects of up-scaling and realistic boundary conditions.
- **Bentonite Laboratory Experiment:** The test will complement the URL in the sense that it is possible to investigate the importance of different parameters and processes (materials, boundary conditions, etc) and provide detailed and high quality data to both conceptual and numerical models.
- **Interface Laboratory Experiment:** Test that will be specifically designed to study the importance of interfaces between different materials or construction parts for the gas migration processes. These tests will supply data to modelling, but also aid in the interpretation of other laboratory and field scale tests.
- **Concrete Laboratory Experiments:** Tests that will be used to study the gas migration process in concrete structures and barriers in the repository. The aim is to study the effects of degree of saturation, gas pressures and alteration of the cement as well as effects of gases on the cementitious materials themselves and provide detailed and high quality data to both conceptual and numerical models.

This Work Package consisted of the following sub-tasks, listed here to indicate the breadth and depth of work undertaken within the FORGE project.

WP 3.1 Bentonite URL Experiments

WP 3.1.1 Large Scale Gas Injection Test (LASGIT);
WP 3.1.2 In situ experiment at Bure site URL – Competition between gas production and seal resaturation, gas migration around the seal;
WP 3.1.3 In situ experiment at the Bure site URL – Gas migration at full scale HLW disposal cell (response of a bentonite plug to gas injection);
WP 3.1.4 Gas migration at a host rock (Boom clay) / bentonite interface;
WP 3.1.5 Gas migration modelling of *in situ* experiments.

WP 3.2 Bentonite Laboratory Experiments

WP 3.2.1 Gas migration in bentonite – fundamental issues;
WP 3.2.2 Gas migration in bentonite - stress effects;
WP 3.2.3 Bentonite laboratory experiments;
WP 3.2.4 ` Modelling of laboratory experiments;

WP 3.2.5 Detailed investigation of gas migration through sand/bentonite mixtures;
WP 3.2.6 Small scale experiments and modelling;
WP 3.2.7 Gas migration at various degrees of saturation;
WP 3.2.8 Effect of gas pressure on saturation and swelling behaviour of bentonite.

WP 3.3 Interface Laboratory Experiment
WP 3.3.1 Transport through interfaces among blocks of sealing materials;
WP 3.3.2 Influence of joints between blocks;
WP 3.3.3 Granite/bentonite interface;
WP 3.3.4 Gas migration through argillite-bentonite interface;
WP 3.3.5 Hydrochemical interaction of sand / bentonite mixtures with the cementitious tunnel backfill and its potential impact on gas migration.

WP 3.4 Concrete Laboratory Experiments
WP 3.4.1 Carbonation reactions of buffer/backfill cements and their impact upon gas and radionuclide migration;
WP 3.4.2 Gas migration in concrete;
WP 3.4.3 Concrete laboratory experiments.

2.4 Work Package 4: Disturbed Host Rock Formations

Construction of any underground opening results in a re-distribution of the local stress field. At depth it is possible to remove mass from the system (e.g. tunnelling), but it is not possible to remove the stress. Therefore the rock surrounding the opening has to accommodate the load that was originally borne by the removed rock, leading to a localised stress concentration. In most geological settings, rocks at depth are at a point of limiting equilibrium, i.e. they are at a stress state just short of failure. Therefore, any stress re-distribution is likely to result in failure of the host rock. Failure is usually observed in the form of a complex fracture network, which is heterogeneous in distribution around a circular tunnel opening because of the heterogeneous stress distribution. The orientation of stress with respect to the fracture network is known to be important. The complex heterogeneous stress trajectory and heterogeneous fracture network results in a broad range of stresses and stress directions acting on the open fracture network. During the open stage of the repository, stress will slowly alter as shear movements occur along the fractures, as well as other time-dependent phenomena. As the repository is backfilled, the stress field is further altered as the backfill e.g. settles and changes volume due to resaturation (dependent on choice of backfill). Therefore, a complex and wide ranging stress regime and stress history will result. As such, there is a need to understand the roles of the stress tensor, the stress path and associated mechanical deformation in determining permeability changes affecting the sealing efficiency of the host rock.

The work package was split into three main areas: laboratory, field and numerical simulation. The objectives of this integrated work package were to:

- Perform laboratory scale experiments to: (a) provide data to test, develop and validate theoretical frameworks and predictive tools to analyse the effects of the stress tensor, the stress path and associated mechanical deformation in determining permeability changes affecting the water and gas sealing efficiency of the host-rock following repository closure, (b) examine the role of the stress tensor orientation with fracture orientation and examine the conditions under which fractures become conductive, (c) examine possible radionuclide movement in an artificially damaged plastic clay formation (Boom Clay) supported by X-ray tomographic techniques (a technique successfully applied in the SELFRAC Project), (d) examine the

permeability evolution of the seal plug/host rock interface supported by X-ray tomographic imaging of test cores

- Perform field-scale experiments to: (a) provide a comprehensive insight into the hydro-mechanical behaviour of a fractured EDZ in an indurated mudrock formation (Opalinus Clay) in transporting gas along the backfilled tunnels and seals; (b) examine EDZ-sealing, radionuclide (Radionuclide) migration and gas movement in a plastic clay formation (Boom Clay) simulating the expected sequence of phenomena in a medium-level waste (MLW) repository that could lead to gas-driven radionuclide transport; (c) investigate the hydro-mechanical behaviour of the EDZ in a disturbed crystalline rock formation (granite) and its role in the movement of repository gases; (d) examine issues of up-scaling from laboratory to field-scale experiments
- Undertake detailed numerical modelling of laboratory and field scale experiments with particular emphasis placed on the assessment and application of constitutive models to describe hydraulic and gas flow properties in a clay-based EDZ. This will be facilitated through the development of strong interactive links between modeling and experimental teams.

The numerical data generated in WP 4 is used in the development and validation of process models aimed at repository performance assessment. As such, WP4 has direct links with WPs 1, 2, 3 and 5.

This Work Package consists of the following sub-tasks.

WP 4.1 Laboratory-scale experiments in argillaceous and crystalline formations

WP 4.1.1 Effect of stress field and mechanical deformation on permeability and fracture self-sealing;
WP 4.1.2 Validation of critical stress theory applied to repository concepts;
WP 4.1.3 Gas-driven radionuclide transport through closed fissures of EDZ and seal/host rock interface – Boom Clay.

WP 4.2 Field-scale experiments in argillaceous and crystalline rock

WP 4.2.1 Gas transport in EDZ at Mont Terri Test Site;
WP 4.2.2 Gas transport in the EDZ: In situ experiment in Boom Clay;
WP 4.2.3 In situ study of gas transport in disturbed crystalline rock.

WP 4.3 Modelling of laboratory and field-scale experiments

WP 4.3.1 Modelling of gas migration through disturbed argillaceous media
WP 4.3.2 Modelling gas migration processes in the Callovo-Oxfordian (COx) argillite
WP 4.3.3 Modelling the coupling between geomechanics and fluid (gas and liquid) flow in disturbed host rock formations.

2.5 Work Package 5: Undisturbed Host Rock Formations

Gas generation and migration may have an impact on the hydraulic and the mechanical properties of the host rock. Consequently, these processes could affect the safety function of the host rock to retard and spread in time the release of radionuclides.

Earlier studies have shown that the ratio between the gas generation rate and the diffusive gas flux through the undisturbed host rock determines the development of a separate gas phase as well as the rate of increase of gas pressure. The two-phase flow properties of the host rock will determine the gas pressure at which gas flow will

start as well as the quantity of water that will be displaced. The latter is particularly important in case of MLW disposal where gas generation and radionuclide release in the near-field can occur at the same time. There is now a general consensus that in the case of plastic clay-rich clays and in particular bentonite, classic concepts of porous medium two-phase flow are inappropriate and continuum approaches to modelling gas flow may be questionable depending on the scale of the processes and resolution of the numerical model. The mechanisms controlling gas entry, flow and pathway sealing in general clay-rich media are not yet fully understood. The "memory" of dilatant pathways within a clay could impair barrier performance.

WP5 has direct links with all the other work packages. Results directly relevant to PA are introduced into WP1. Gas migration in the undisturbed host rock sets the outer boundary condition to the transport processes in the EDZ and the engineered barriers (WP3 & WP4). Many processes may be common for bentonite buffer (WP3) and a clay host rock. The experimental results will be used for models applied (WP1).

The main objectives of this WP were to:

- Establish the conditions under which the different gas migration processes are dominant;
- Identify how those processes can be modelled and to determine the values of the main parameters;
- Measure the parameters that may have an impact on the long-term safety as a consequence of enhanced radionuclide transport through the host rock.

This Work Package consisted of the following sub-tasks.

WP 5.1 Gas transport laboratory experiments

WP 5.1.1 Baseline hydraulic and gas transport properties of the Callovo-Oxfordian argillite;

WP 5.1.2 Determination of two-phase flow parameters and analysis of fracturing by gas overpressure in Opalinus clay;

WP 5.1.3 Gas driven radionuclide transport in Boom Clay.

WP 5.2 Gas transport *in situ* experiments

WP 5.2.1 PGZ1: Gas migration in undisturbed indurated clay formation, Callovo-Oxfordian Clay, at the URL site in Bure;

WP 5.2.2 The HG-C gas injection experiment in intact Opalinus clay at the Mont Terri URL.

WP 5.3 Gas transport in undisturbed host rock modelling

WP 5.3.1 Modelling of gas transport in indurated clay;

WP 5.3.2 Modelling of the PGZ1 in situ test in the Callovo-Oxfordian clay in Bure;

WP 5.3.3 Modelling of the PGZ1 in situ test in the Callovo-Oxfordian clay in Bure and the HG-C in situ test in the Opalinus clay in Mont Terri;

WP 5.3.4 Interpretation of experimental results achieved in WP 5.1 and 5.2 by hydro-mechanically coupled 2-phase-flow models, and model validation;

WP 5.3.5 Modelling for understanding of flow physics and evaluation of uncertainty of gas transport in a wide range of sedimentary rocks and geological settings.

2.6 FORGE Exclusions

Certain gas-related issues were not studied in FORGE. This is 'by project design', as the subject areas that were studied are viewed as priority topics by a range of waste management organisations. The following areas were not studied in FORGE:

- Gas flammability (primarily an operational safety issue and manageable by repository operating procedure, ventilation etc).
- Gas in the biosphere (although some insight from outside of FORGE is provided in subsection 3.4.2.7).
- Gas in the context of the variant human intrusion scenario.
- Explicit consideration is not given to any minor gases / chemotoxic gases, although the assumption herein is that the behaviour of such in a repository environment is bounded by the consideration of major bulk and radioactive gases.
- No gas-related radiological risks are calculated or presented; derivation of such is highly site-specific (gas migration cannot be de-coupled from site specificity), and the approach to calculating radiological risk at the level of the waste management organisation needs to consider e.g. national regulations).

3 GEOLOGICAL DISPOSAL CONCEPTS AND ASSOCIATED GAS-RELATED ISSUES

3.1 Introduction

Within a geological repository, the formation of gas is unavoidable. Wastes and their containers, as well as steel used in the construction of the repository, are the principal sources of gas generation. The primary objectives of this section are to identify potentially safety-relevant issues associated with gas production and to put these issues in the context of the various repository concepts considered in FORGE. It also provides an overview of the key gas generation and migration processes and of the associated uncertainties. The approaches that are typically followed to treat these processes and uncertainties in a safety assessment are briefly discussed. The section provides links between identified gas-related issues and FORGE work packages (WP), but does not incorporate project results - these are discussed in subsequent sections of the report.

This section focuses on issues relevant to nuclear safety and radiation protection after the closure of geological repositories constructed in clay and crystalline host rocks. It is mainly based on the information available in the state-of-the-art report produced in the framework of the FORGE Milestone M15. Reference is also made to work performed outside FORGE when relevant.

3.2 Description of geological disposal concepts

3.2.1 General considerations

The two primary safety objectives of geological disposal of radioactive waste are to isolate the waste from the accessible biosphere (i.e. to keep the waste and its associated hazard apart from the biosphere) and to contain the radionuclides associated with the wastes within the containment barriers of the disposal system (i.e. to avoid or minimize the release of radionuclides). A variety of disposal concepts have been developed in the different countries that are carrying out a geological disposal project. These concepts provide different degrees of containment and isolation capability appropriate to the radioactive waste that they will receive.

The application of the defence in depth principle to radioactive waste disposal implies that containment and isolation of the waste shall be provided by means of a number of physical barriers of the disposal system [8]. The performance of these barriers shall be achieved by means of diverse physical and chemical processes such that overall performance of the disposal system is not unduly dependent on a single safety function[2]. A schematic representation of multiple barrier systems is provided in **Figure 2**.

[2] A **safety function** is a physical or chemical property or process that contributes to safety, by isolating or containing the disposed waste. The barriers that make up the geological disposal system may contribute to satisfying a number of different safety functions and these contributions may be independent of one another.

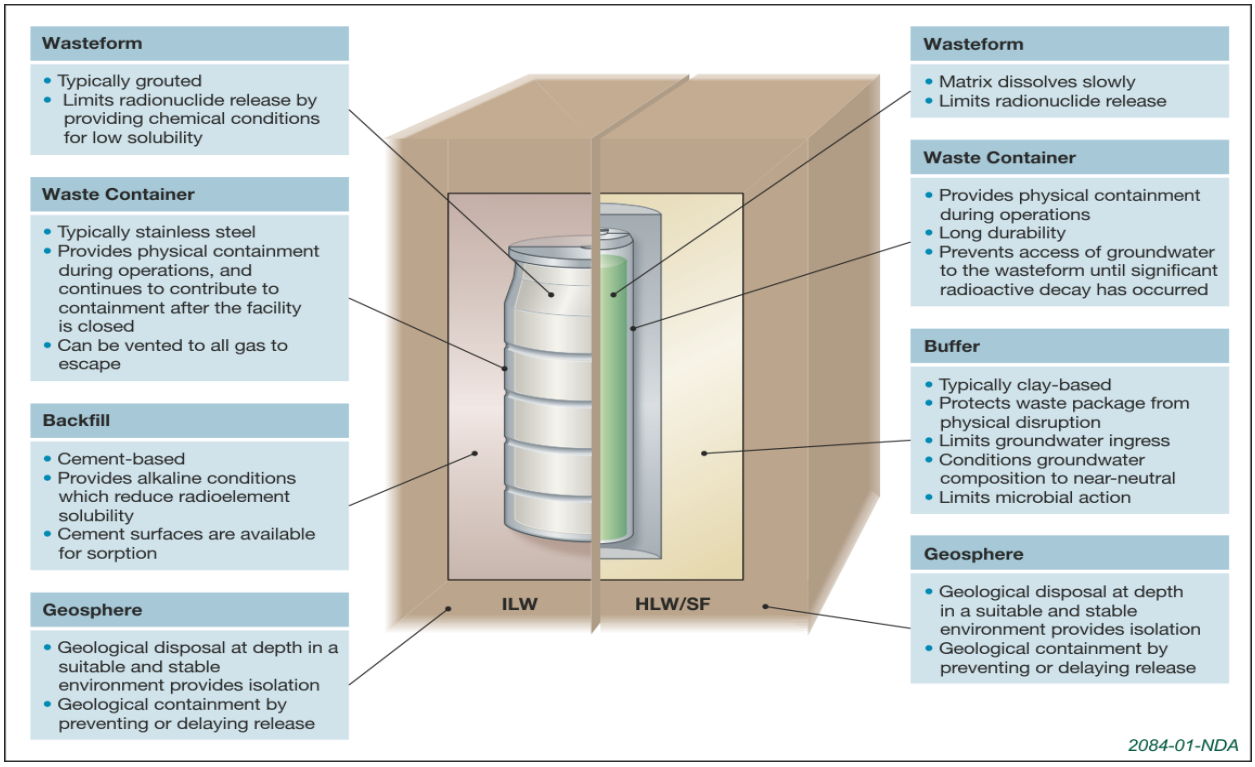


Figure 2 Schematic representation of a multiple barrier system, both for ILW and for HLW/SF (modified from NDA, 2010 [15])

The allocation of safety functions to the engineered and natural components of a repository and the required degree of complementarity and redundancy depend on national regulations, on the evolution of the hazard associated to the waste, as well as on technical aspects, such as the host formation considered.

3.2.2 Clay host rock concepts

3.2.2.1 Belgian concept (plastic clay)

In the Belgian concept, the vitrified high-level waste canisters and the spent fuel assemblies are placed in an engineered waste package entity called "supercontainer". The main barriers hindering the leaching of radionuclides in this concept are the waste matrix, the carbon steel overpack, the concrete buffer, and the clay formation (Boom Clay). Concrete containing Portland cement has been chosen for the buffer because this would provide a high alkaline chemical environment for a long period of time in order to inhibit corrosion of the overpack. The gas source term comes primarily from the corrosion of the C-steel overpack and to a smaller extent from the stainless steel envelope. There is also a potential gas source owing to radiolysis of components of the buffer.

Low and intermediate level waste are placed in concrete monoliths. Corrosion of the galvanised steel drums and of the reinforcement bars in the concrete monoliths are considered as a major potential gas source term.

Waste disposal cells are sub-horizontal concrete lined tunnels approximately 3m in diameter.

3.2.2.2 French concept (indurated clay)

In the French repository concept in Callovo-Oxfordian (COx) clay, intermediate-level and long-lived (MAVL), and high-level and long-lived (HA) wastes, will be emplaced at around 600 m depth in separated wastes zones linked by a network of drifts to the shaft zone, also including sub-horizontal ramps.

MAVL and HA waste primary packages are conditioned in disposal packages:

- MAVL waste primary packages are grouped in concrete containers.
- HA waste primary packages are placed in carbon steel over-packs.

HA disposal cells are sub-horizontal dead-end tunnels approximately 100 m long and 0.7 m in diameter. Each tunnel has an inner lining consisting of a thick, low or non-alloy steel.

The MAVL disposal cells are sub-horizontal concrete tunnels with an excavated diameter of approximately 10 m and a useful length up to 450 m. A supplementary volume of concrete is added to the coating so as to constitute a disposal chamber with rectangular section dimensions suited for the shape of the stack of packages.

It is expected that gas would be mainly generated by anoxic corrosion of metallic materials in the repository or radiolysis of organic matter contained in the waste packages.

3.2.2.3 Swiss concept (indurated clay)

The Swiss repository for SF/HLW in Opalinus Clay envisages an array of long (~800 m) parallel tunnels at a depth of 600 to 900 m where spent fuel and high-level waste (HLW) canisters would be emplaced, with the region around the canisters filled with bentonite. Canisters are assumed to be fabricated from carbon steel with a wall thickness of 13-15 cm (an alternative of a copper canister with iron insert is also considered). Depending on excavation stability considerations, a liner may or may not be used. Anaerobic corrosion of steel is the most significant source of gas production.

Low and intermediate-level wastes (L/ILW) would be disposed of in caverns. The waste drums would be placed into pre-fabricated reinforced concrete containers and the void spaces within and between the containers are filled with viscous cementitious mortar. The corrosion of metals and the degradation of organics are the most significant sources of gas production. The majority of the L/ILW would be disposed of in the L/ILW repository, with only the much smaller quantity of long-lived intermediate level waste disposed of in separate caverns associated with the repository for SF/HLW in Opalinus Clay.

3.2.2.4 Gas-related issues associated with clay host rock concepts

Considering that gas migration should occur along the path of least resistance, several pathways are possible in clay host rock concepts. Before saturation of the engineered clay-rich barrier sealing the disposal cells (around one hundred years), gas may migrate along the interface between the seal and the host rock through the interstices in the seal or the EDZ. After saturation of the seals, due to the relatively low permeability to gas of clay rocks, gas is usually expected to migrate axially along the EDZ of the disposal tunnels and/or along the interfaces between the host rock and the engineered components of the repository.

Seals and backfills can also be designed to keep the gas pressure below the magnitude at which preferential pathways within the host rock would start to develop. The objective is to increase the gas transport capacity of the engineered barriers and thus to prevent high pressure being reached without compromising their safety functions.

3.2.3 Crystalline host rock concepts

3.2.3.1 KBS-3 concept (Sweden, Finland and Lithuania)

The KBS-3 concept aims at long-term isolation and containment of spent fuel assemblies within durable copper-iron canisters. The disposal system consists of the spent nuclear fuel, the canisters (composed of an outer protection of copper and a cast iron insert with channels for the fuel assemblies), the bentonite buffer and the geosphere in the vicinity of the repository.

Two variants of the KBS-3 concept are being considered: KBS-3V, in which the disposal canisters are emplaced vertically in individual deposition holes, and KBS-3H, in which the canisters are emplaced horizontally in long deposition drifts.

Gas in deposition holes may originate from the air trapped in the buffer during saturation, radiolysis of water, microbial activity or from the corrosion of the cast iron insert in a damaged or breeched canister. This last source is considered as the most important.

Crystalline host rocks are usually assumed to have a high capacity for gas migration due to the presence of pre-existing geological discontinuities. Therefore, the main gas issues associated with the KBS-3 concept are related to the migration of gas through the bentonite buffer. The bentonite buffer is an important barrier in the KBS-3 system serving as a diffusional barrier between the canister and the groundwater in the rock.

3.2.3.2 UK concept for ILW/LLW disposal

Gas generation in association with UK ILW/LLW is sourced from:

- Corrosion of metals (Magnox, uranium, aluminium, steels and Zircaloy);
- Degradation of organic materials;
- Radiolysis of water and polymers;
- Radioactive decay.
- Additionally, evolution of graphite in a repository environment will result in some carbon-14 bearing gas being formed.

Bulk gas formation will be dominated by H_2, CH_4 and CO_2; minor gases may also be generated.

Radioactive gas formation will be dominated by $^{14}CH_4$, $^{14}CO_2$, ^{3}HH and Rn-222.

Bulk gas can act as a ‘carrier’ for the volumetrically small amounts of radioactive gas generated. Carbon-14 bearing gases are potentially a significant issue in the post-closure period.

Studies currently being undertaken by NDA RWMD are not UK geology-specific. A range of generic geological disposal concepts is available internationally; these concepts have the potential to provide safe and secure geological disposal options for any suitable UK geological environment.

Work carried out by NDA RWMD in 2008 [9, 10] reviewed the range of possible concepts for geological disposal of ILW, HLW and SF in the UK. The work drew on

previous work in the UK, and disposal programmes in other countries, to identify disposal options for generic geological environments (rock formations and their surrounding geological setting).

Illustrative geological disposal concept examples for higher strength rock, lower strength sedimentary rock and evaporite rock were selected by:

- Consideration of the concepts identified in the Geological Disposal Options studies for ILW/LLW and HLW/SF [9, 10].
- Basing the illustrative examples on geological disposal concepts that have previously been developed in the UK or are being developed in national programmes in other countries.
- Using the criteria developed previously to select KBS-3V as an example reference HLW/SF Concept, that is, the concept must be well-developed and supported by extensive R&D and have been subject to detailed safety assessment; regulatory scrutiny and international review (see Reference [11]).

The illustrative concepts are used to:

- Support the scoping of the impacts of a GDF.
- Develop the disposal system specification, engineering design and safety case methodology.
- Support prioritisation of the R&D programme.

One such concept currently under consideration in the UK programme is the UK ILW/LLW disposal concept, for higher strength rock. This draws heavily on knowledge gained from previous Nirex studies.

The consideration of gas generation and gas migration in the context of these three illustrative geologies is part of NDA RWMD's ongoing research and safety case studies.

3.2.4 Canadian concept

Ontario Power Generation (OPG) is proposing to build a Deep Geologic Repository (DGR) for Low and Intermediate Level Waste (L&ILW) near the existing Western Waste Management Facility (WWMF) at the Bruce nuclear site in the Municipality of Kincardine, Ontario [12].

The DGR is planned to be located in competent and low permeability Ordovician argillaceous limestone with Ordovician shales and Silurian shales, dolostones and evaporites and Devonian dolostones above. The porewater in the Silurian and Ordovician sediments is highly saline brine (with total dissolved solids of 150 to 350 g/L) and reducing, with pH buffered by carbonate minerals. The repository is planned at a depth of about 680 m below ground surface. The repository is not planned to be backfilled.

During the years following closure, it is expected that there will be corrosion of the carbon steel containers and degradation of organic materials in the wastes. The atmosphere in the repository will become anaerobic as oxygen is consumed by corrosion, and subsequent slow anaerobic degradation of the wastes and packaging materials (i.e. containers and overpacks) in the DGR will generate various decomposition products, in particular gases (predominantly CO_2 and CH_4 from the microbial decomposition of organics, and H_2 from the corrosion of metals).

The low permeability of the shaft seals and the host rock, plus the gas pressure in the repository and the water consumption by corrosion reactions, all limit the

resaturation of the repository. The repository might take many hundreds of thousands or even millions of years to resaturate completely.

Contaminants are released from the waste by dissolution into repository water and, especially for H-3 and C-14, the formation of radio-labelled gases. Once released into the water or gas in the repository, the migration of contaminants from the repository is limited by the low permeability shaft seals and very low permeability host rock. The excavation of the repository results in a damaged zone developing around the shaft, emplacement rooms and tunnels, with higher porosity and permeability. This is also a potential pathway for contaminant transport. Most radionuclides decay within the repository and the surrounding rock. However, slow migration of some dissolved or gaseous contaminants will occur into the geosphere surrounding the repository and into the repository shafts.

3.3 Relevance of gas-related issues for safety in the context of geological disposal

The generation of both radioactive and non-radioactive gases in a geological repository may have several implications on long-term safety. Under certain conditions, they have the potential to affect, either negatively or positively, safety functions fulfilled by one or several repository components. Volatile radionuclides originating from the waste can also be released out of the repository system and lead to radiological impacts on man and the environment.

The main gas-related issues that should typically be considered in a safety case are identified below. The importance of a particular issue will vary with the repository concept, the site characteristics and the waste inventory.

3.3.1 Potential impacts on safety functions

It is generally expected that non-radioactive gases will be generated in amounts sufficient for influencing some of the processes and conditions prevailing in a geological repository and upon which safety functions allocated to components of the repository may be relying. This section describes briefly potential gas-induced perturbations of the most common safety functions that may have to be considered in a safety case. Advantageous effects of gas generation on safety functions, and hence on the overall performance of the disposal system, are also identified. A detailed discussion of the possible implications of gas generation and migration on safety functions is provided in section 5.

3.3.1.1 Containment

Spent fuel and HLW waste packages typically fulfil a containment function aimed at preventing any form of release until radiotoxicity of the waste has sufficiently decreased. LLW/ILW waste packages are usually not assigned a containment function and are sometimes vented to mitigate gas generation during packaging and before repository closure.

Gas generation may have both negative and positive impacts on containment. Pressure rise inside or outside metal waste packages due to corrosion-induced gas production can potentially damage containers. The generation of gaseous hydrogen and carbon can also alter the chemical conditions in the repository which may in turn favour or hinder metal corrosion (e.g. Libert et al., 2011 [13]). Moreover, desaturation due to accumulation of gases in repository components located in the vicinity of the waste may also result in a reduction of corrosion rates.

Possible effects of gas generation on containment have been considered in WP3.

3.3.1.2 Flow limitation

If the generated gas cannot be totally dissolved in the pore water of the near field, the formation and movement of a free gas phase have the potential to displace contaminated water through the repository or to provoke the development of preferential pathways increasing the migration rate of the radionuclides present in the liquid or in the gas phase. Gas generation may also contribute to limiting groundwater flow due to the desaturation induced by the accumulation of gases in repository components.

Gas-induced water displacement and effects on radionuclide migration in the host rock have been studied in WP5.1.3. The development of gas-induced pathways in clay engineered barriers and interfaces between repository components have been investigated in WP3.

(a) Gas-driven displacement of water containing radionuclides

Whether or not gas-induced groundwater movement occurs is dependent on e.g. site-specific properties, gas generation rate. Several processes are known at least to have the potential to produce water advection as a result of gas pressure and flow:

- Gas-pressure driven advection: Water flow may be induced by elevated gas pressure in regions of a repository (e.g. hydraulic gradients induced by heterogeneous saturation) or by the expulsion of water due to the generation of a free gas phase in a defective canister or in the pores of repository components.
- Gas-flow driven advection (GASNET, 2003) [5]:
  - Colloidal clay particles may be transported by gas bubbles that may lead to enhanced transport of radionuclides sorbed onto the clay;
  - Groundwater movement may be caused by the collapsing and reforming of gas-induced preferential pathways and the entrainment of groundwater in streams of bubbles, effects which seem more likely to be associated with flow in fractures than in the porosity of rock.

(b) Permeability increase due to gas pressure build-up

Gas pressure rise within the repository, were it to occur, could eventually lead to an over-pressurisation resulting in the development of temporary or permanent preferential pathways for contaminated water or volatile radionuclides due to mechanical damage or pore dilation. The potential for over-pressurisation is largely determined by the gas generation rate (itself generally dependent on the availability of water) and the gas transport capacity of the host rock, engineered barriers and interfaces between repository components.

Gas generated at rates expected in repositories is likely to be able to escape through most fractured crystalline rocks without creating a gas pressure build up [5]. However, pressure build up is a potential issue for low-permeability host rocks such as salt and clay-rich host rocks. In these cases, the quantification of the water displacement and Darcy velocities due to gas pressure build-up and the impact of gas pressure and release on the transport characteristics of the rock are considered as key questions in several disposal programmes.

Preferential pathways are also commonly expected to develop in low permeability buffers or at the interface between repository components. The relevance for safety

of these pathways depends on their longevity (i.e. temporary vs. permanent) and their geometry (e.g. length, location).

(c) Gas-induced desaturation

Gas generation has a significant influence on the saturation degree of different repository components after closure. This influence may have beneficial or detrimental effects on components contributing to flow limitation. The desaturation of a clay host rock would tend to reduce its permeability. On the other hand, desaturation of a clay-based seal would reduce its swelling pressure and hence its performance.

3.3.1.3 Ensuring stable conditions

The potential impacts of gas migration on the ability to maintain stable conditions within the repository is considered as a relevant issue in several disposal programmes where such a safety function is being relied upon. For instance, cementitious materials are sometimes used to establish homogeneous near-field alkaline conditions ensuring favourable chemical conditions for corrosion or for the sorption of carbon dioxide gas generated from evolving waste.

Under some circumstances, the reactivity of gases with the engineered barriers or the host rock may result in modifications of the initial physico-chemical conditions (e.g. concrete carbonation, reducing conditions created by the generation of hydrogen, gas-induced microbial activity). These modifications may have negative or positive implications for safety. For instance, the production of hydrogen in the near-field is known to protect UO_2 against oxidative dissolution [14].

3.3.1.4 Retention of radionuclides

Possible modifications of chemical conditions associated with the reactivity of gases with engineered and geological barriers may alter the ability of these barriers to chemically retard radionuclides. For instance, gaseous hydrogen produces reducing conditions that may increase the ability of the host rock or of the engineered barriers to retard redox-sensitive radionuclides.

3.3.2 Transport of volatile radionuclides

Depending on the waste types to be disposed of, several volatile radionuclides originating from the waste may migrate through the engineered and geological barriers and be eventually released to the biosphere. Although the volumes of these gases may be small relative to non-radioactive gases [15], their safety significance should be considered because of their radioactivity.

Gases containing the following radionuclides may have to be considered in a safety assessment:

- Waste inventories may contain large quantities of ^{14}C in a variety of materials. Volatile ^{14}C may originate from the instant release fraction (IRF) of spent fuel, irradiated graphite, and irradiated metals (steels, Zircaloy, uranium, Magnox) as they corrode. Hence, ^{14}C-containing gases (principally $^{14}CH_4$ and $^{14}CO_2$) are commonly expected to be the radiotoxic gases causing the most significant radiological hazard. As a consequence, the behaviour these gases in cementitious barriers, in clayey host rocks and in the EDZ associated to these rocks is considered as a relevant issue in several disposal programmes. .

- ^{129}I is a long-lived fission product that originates principally from the IRF of spent fuel.
- ^{222}Rn, ^{3}H and ^{85}Kr (which is the only radioactive noble gas remaining in irradiated fuel elements after a cooling period of one year [16]) are short-lived radionuclides with respect to the time-frames associated to deep geological disposal but they may have to be considered to assess safety during the operational phase. The long-term radiological impact associated with ingrown ^{222}Rn may also be significant were ^{226}Ra and higher members of the ^{238}U decay series originating from disposed wastes to migrate from the repository (in a groundwater pathway, if present) to reach the near-surface or biosphere. ^{222}Rn would also need consideration in the context of an assessment of the variant human intrusion pathway scenario.
- Under reducing conditions, long-lived fission products such as ^{79}Se may be biologically transformed in volatile organic species (e.g. dimethylselenide, dimethyl-diselenide) if they are metabolised by micro-organisms [17].

The radiological impact of any potential gaseous releases has to be assessed in order to demonstrate that any impact will remain acceptable for different scenarios considered in the safety assessment, such as those considering gas movement through the geosphere or the release of radioactive gases caused by an intrusion into the repository. The identification and quantification of the potential amount and generation rates of these gases is therefore of importance for the safety assessment.

3.4 Gas issues in the safety case

3.4.1 Gas generation

In all cases, the assessments of the impacts of gas generation starts with an assessment of the amounts and rates of production of the range of safety-relevant gases that can be generated in a repository [5].

There is general consensus about the mechanisms that may generate significant quantities of gases in repositories and that need to be considered in the safety case, although the relevance and priority of contributing gas generation mechanisms varies from waste management organisation to waste management organisation (reflecting e.g. inventory differences). These mechanisms are:

- Anaerobic corrosion of steels and other metals, including the reactive metals e.g. aluminium and Magnox, if present;
- Microbial degradation of organic materials;
- Water radiolysis; and
- Denitrification processes of nitrate-bearing waste.

The dominance of these mechanisms will mean that the bulk gas produced within a geological repository under saturated and anoxic conditions would be mainly hydrogen and methane [18]. In the case of nitrate-bearing wastes, their decomposition (nitrate reduction) may also lead to the production of molecular nitrogen. Some gas generation from radioactive decay will occur but has always been assessed to be insignificant as far as the volumes of gas are concerned [5].

3.4.1.1 General considerations on the treatment of gas generation in the safety case

The relative amounts of gas generated are mainly dependent on the waste materials to be disposed of and on the materials used in the EBS. The rates at which most of the gases will be generated are sensitive to environmental factors, which might change with space and time, such as the presence of oxygen, water, hydrogen or

chloride ions and temperature. In a safety assessment, it would also be appropriate to consider what interactions there might be between these processes, e.g. the ways in which the gas generation processes can affect conditions within the repository, particularly the redox conditions [5].

The gas generation source terms used for gas migration calculations are typically supplied as constant or time varying representations of the results of separate gas generation calculations, often with a measure of conservatism built in. For instance, it is sometimes assumed that water is instantaneously available for use in gas generation processes at the onset of the post-closure period and water desaturation resulting from gas generation is often ignored. Other assumptions on the starting time of gas generation may be considered dependent on site-specific geology. However, it is commonly recognized that the gas generation source terms for LLW and ILW cannot be quantified as effectively as for HLW and SF because of the rather heterogeneous nature and the intrinsic humidity (relevant in those rocks in which resaturation may be very slow) of many of the wastes [5].

3.4.1.2 Corrosion

Generalised corrosion has to be considered for hydrogen gas generation. Localised forms of corrosion, although they may lead to a loss of container integrity, are unlikely to contribute significantly to total metal loss or gas generation [15]. In the case of ILW / LLW, carbon steel and stainless steel can be present in the wastes and also as waste containers. Zircaloy, Magnox, aluminium, zinc and uranium can be present as wastes. Spent fuel can include stainless steel and Zircaloy; stainless steel can also be associated with HLW in the form of the containers [18]. Possible container metals include carbon steel, stainless steel, copper, titanium and nickel alloys [19].

Generally, a rather simple approach is followed in the safety case for the estimation of the rate of hydrogen production from the corrosion of metals [5]. The rate of H_2 gas production is proportional to the rate at which the metal corrodes. This is considered to occur uniformly and is calculated as the product of the corrosion rate constant [$m \cdot a^{-1}$] and the metal surface area (which may change with time, depending on the shape of the metal items in the waste). However, uncertainties on the surface area of the different metal types found in LLW/ILW inventories can be significant. In order to calculate rates of H_2 gas generation, corrosion rates may have to be provided for a number of combinations of metals and environmental conditions.

A source of ^{14}C that has been considered in some assessments is that of irradiated metals, which may release $^{14}CH_4$ as they corrode [5]. These metals include irradiated uranium, Magnox, Zircaloy and steels. While the first two seem likely to release at least some of their ^{14}C in gaseous form as they corrode, the behaviour of Zircaloy and steels is subject to more uncertainty. Nevertheless, in a number of programmes, a conservative allowance for such possible release is being made to ensure robustness to the safety analysis [5].

3.4.1.3 Microbial degradation of LLW and ILW

Microbial populations are ubiquitous and thus a geological repository is expected to contain microbes, both in the waste packages and introduced during construction and operations (e.g. on soil and dust particles) ([13], [20], [21], [22], [23] and [24].) However, gas generation from microbial degradation is only an issue for certain types of LLW and ILW such as organic and nitrate-bearing wastes. These biodegradable wastes can comprise a variety of materials, for example, cellulose, nitrate-bearing

bitumen, ion-exchange resins, plastics, rubbers, and oils. A variety of gases can be produced by microbial degradation of organic materials and nitrate-bearing wastes under anaerobic conditions, mainly CO_2, CH_4, H_2, N_2, and H_2S. In the context of gas generation from organic materials in a geological repository, the most important microbial process involves the degradation of cellulose [18].

In the long term, the main constraints on microbial activity are the availability of nutrients, space and water restrictions and environmental conditions such as high pH [18]. However, adaptation and selection may result in microbial populations that can tolerate extreme environments. Gas generation from microbial degradation of cellulose at high pH is considered as a relevant issue for the disposal of LLW/ILW making use of cementitious components.

Gas production from microbial action within the geological repository after closure would be very heterogeneous with broad ranges of possible generation rates and time dependencies due to the range of possible microbes and the large diversity of wastes. The conventional approach has been to use bounding upper estimates [18]. A pair of basic parameters that can be used to simply characterise microbially mediated gas generation are the total yield of organic materials under repository conditions and the gas production rate per unit mass of undegraded wastes [5].

3.4.1.4 Radiolysis

Hydrogen can be produced by the radiolysis process. The main process is decomposition of water resulting in the production of hydrogen and oxidizing species whose recombination rate in closed system is very slow ([15]; [25]; [26]). Usually, radiolysis will be subordinate to gas generation by anaerobic corrosion of metals, which will be dominant in terms of volume, or degradation of organic matter [18].

3.4.1.5 Radioactive decay

The decay of several radionuclides originating from the waste can result in the production of radioactive gases, principally ^{222}Rn.

The decay of alpha-emitting radionuclides will also release one atom of helium per alpha particle emitted. This process has been assessed not to have any significance for safety in spent fuel wastes [5] as the volume of helium produced is small relative to the void volume of the canister [27].

3.4.1.6 Natural gas sources

The EC FORGE Project was focussed on the effects of gas generated from waste constituents or materials used in repository engineering. It is however worth mentioning that, in the safety case, it may also be necessary to consider the effects on repository performance of gas from natural sources.

Gas may be present naturally in the geological formation and may also be formed when the geological environment is changed by mining operations [5] or following waste emplacement. Naturally occurring methane has been observed in rock formations studied as potential repository sites, specifically the Valangian Marls of Switzerland and some granitic rocks in Finland [5].

Under a low thermal stress, the immature natural organic matter intimately associated to clay minerals may quickly decompose to produce a non-negligible amount of CO_2 as observed by Deniau et al. (2005) [28] in Boom Clay. The kinetic of kerogen decomposition leading to the early generation of CO_2 in Boom Clay has

been studied in detail by Lorant et al. (2008) [29] and its impact on pore water pH evolution modelled by Pitsch et al. (2007) [30]. The alteration of geochemical conditions induced by this process may affect the migration properties of radionuclides complexed by carbonates such as uranium.

3.4.2 Gas migration

This section identifies gas migration processes that may have to be considered in the safety assessment of a geological repository. Possible treatments or representations of these processes in safety assessment and associated key uncertainties are also discussed. The links with FORGE WPs and subsequent sections of the report discussing the outcomes of the project are also identified when relevant. Indeed, the lack of data and existing uncertainties on gas migration processes in various types of engineered barriers and host rocks constitute two important issues addressed in the FORGE project.

Safety-relevant gas migration processes include processes:

- Governing or influencing the accumulation and flow of safety-relevant gases in the repository;
- Potentially affecting safety functions fulfilled by one or several components of the repository through:
  - an alteration of the mechanical, hydraulic or chemical conditions that need to be met in order to reach the expected level of performance, or
  - a disruption of the physical integrity of one or several components;
- Governing or influencing the radiological impact associated to volatile radionuclides.

Transport mechanisms prevailing in a particular system are greatly dependent on its geometry, the characteristics of the different materials composing the system, its boundary conditions (stress state etc) and the gas generation rate.

3.4.2.1 Gas diffusion

(a) Process description

As soon as a gas being produced becomes dissolved in the pore water surrounding the waste packages, the gas concentration gradient initiates diffusive transport of dissolved gas from gas source towards the host rock and the EBS. Gas diffusion is commonly considered as the prevailing gas transport mechanism when gas generation remains limited and most of the generated gas is dissolved in the liquid phase of the porous materials. However, in many cases, diffusion is not sufficient to allow generated gas to escape from the repository.

(b) Uncertainties relating to and treatment in the safety case of gas diffusion

Reliable confirmation from experimental data of the large-scale aqueous phase intrinsic diffusion coefficient for the principal gases generated in a repository is required to sustain an argument that diffusion will disperse the gases generated [5]. The gas flux through the repository is also greatly dependent on the actual value of the diffusion coefficient.

However, diffusion coefficients for hydrogen and methane for clay host rocks may, in some cases, widely scatter due to the limited test results and the complexity of performing such tests due to the small pore size of the material. However uncertainties on this parameter could be also due to the chemical reactions of

hydrogen during diffusion experiments and to limitations of the experimental set-up used [31].

Moreover, with the decrease of water saturation, the tortuosity for diffusion increases and hence the diffusion coefficient decreases. The quantitative relationship between diffusion coefficients and saturation degree of pores has not yet been appropriately quantified for materials such as intact and disturbed clay host rocks or concrete.

Parameter sensitivity analyses or the use of conservative assumptions are typically used in the safety case to cope with these uncertainties.

3.4.2.2 Free gas phase formation

(a) Process description

If the generated gas cannot dissolve, a free gas phase will form within the repository. Gradual desaturation of the EBS and possibly of the host rock is expected when gas pressure starts to build up in the near field. A small fraction of the interstitial water may be expelled. Under some circumstances there may be an overlap between the re-saturation process of bentonite buffers and gas generation, with the latter starting before the former has been completed.

(b) Uncertainties relating to and treatment in the safety case of free gas phase formation

Gas dissolution is generally quantified in the safety case using Henry's Law. Uncertainties relating to gas dissolution in the pore-water of engineered or geological barriers are dependent on various factors such as groundwater salinity and are sometimes considered as a relevant issue. However, in many cases, the amount of gas that can dissolve into pore water is expected to be negligible with respect to the amount of generated gas or it is difficult to rule out the formation of a gas phase. In these cases, uncertainties on gas solubility can be handled making use of pessimistic assumptions.

As explained in section 3.3.1.2, desaturation may have beneficial or detrimental effects on components contributing to flow limitation. Additionally, gas-induced de-saturation of backfill materials and its influence on their mechanical properties, investigated in WP3.1 and 3.2, are issues of interest for several programmes. For instance, the validation of available unsaturated soil models when applied to backfilled materials is an area needing further investigation. It may also be assumed that water is instantaneously available for use in gas generation processes at the onset of the post-closure period; any alternative postulate being considered via variant scenario analysis. Gas-induced groundwater movement, if any, also needs to be considered in the safety case.

3.4.2.3 Two-phase flow

(a) Process description

Gas generation may keep going on until isolated gas bubbles ultimately form a continuous gas phase. As of this stage, two-phase flow begins in engineered barriers with a relatively high permeability and possibly in the surrounding rock. Gas is evacuated mainly through advective flow, and water is being further expelled.

(b) Uncertainties relating to and safety case treatment of two-phase flow

Reliable predictions of multiphase flow depend on the understanding of the physical processes that take place at the pore scale, and on the ability of accurately representing their effects at the field scale by means of appropriate conceptual models. The general model based on Darcy-type two-phase flow is generally considered as capable of describing the evolution of gas/water pressure/degree of saturation before gas preferential pathways are formed. A variety of well-established computer models are available to carry out such conventional two-phase flow calculations.

The unsaturated soil model used to describe two-phase flow needs a large number of parameters. However, few data are available regarding gas transport properties of concrete material, backfill materials and seal plugs as well as of interfaces between different materials or construction parts. The following parameters are of particular importance:

- The gas entry pressure is an important parameter when describing the transition from saturated to unsaturated condition. It represents the difference between gas pressure and water pressure needed to displace the porewater from the initially fully saturated medium. Uncertainties on the gas entry pressure of clay host rocks, investigated in WP5, and of bentonite buffers are considered as a key issue as it determines the maximum pressure that can be created in the near-field of the repository. The variations in the values obtained using different measuring techniques and the scale dependence of transport properties were identified as important areas of study for clay rocks. Gas permeability of crystalline host rocks is generally considerably greater than that of bentonite buffer so that gas entry rate in these buffers is much smaller than host rock's ability to transport gases. In the area of gas transport for a L/ILW repository, hydro-chemical interactions at the interface between cementitious backfill and sand/bentonite seals may also play an important role in influencing gas entry pressures of these materials. The extent to which these occur under saturated conditions is not well understood and may influence gas release.

- The relative permeability of gas and liquid in the unsaturated material greatly influences the timescale and magnitude of the excess gas pressure built up in the repository. However, there are few reliable experimental results available for clay host rocks. This issue has been considered in WP 3.2, 4 & 5.

Anisotropy, geological structures (e.g. faults, bedding planes, foliation) and heterogeneities are known to influence significantly the mechanical and hydraulic properties and behaviours of rock masses and, hence, gas migration in host rocks. Their possible influences on gas migration in clay host rocks are considered as relevant issues and need to be taken into account when upscaling gas transport properties. This topic has been investigated in WP5. The scale dependence of transport properties and the extension of existing data bases on gas-related parameters of EBS materials are also important areas of study. For instance, the influence of heterogeneities in cementitious materials, including effects of cracking, shrinkage and curing-related morphological change (in comparison with assumption of properties of homogeneous medium) on gas migration is an area needing further development.

The case of water ingress into a defective canister and the possible effect of gas generation following water ingress in causing expulsion of contaminated water

involves complex coupled processes: water flow (or vapour diffusion) through small defects, gas generation with water consumption by corrosion, mechanical stresses due to corrosion products, chemical evolution and its effects on contaminant release from spent fuel; water expulsion; gas and water transport through the bentonite buffer; etc. [5]. Assessment cases or scoping calculations with simplifying assumptions are therefore used to assess the potential consequences of gas-induced displacement of contaminated water from a defective canister. The release of volatile radionuclides from a defective canister may also be examined through the development of a specific assessment case.

3.4.2.4 Pathway dilation & gas-induced fracturing

(a) Process description

If the gas generation rate remains significant, gas flow may not be sufficient to evacuate all the gas generated in the repository due to the low permeability of the surrounding material. Hence, for a clay-rich host formation there is a major concern over whether gas will be able to escape through the rock without the gas pressure rising to a level at which fractures will occur and create pathways for groundwater transport to bypass the geological barrier. There is similar concern in connection with low-permeability buffers.

Two types of processes may prevail when gas pressure locally exceeds the gas breakthrough pressure of a natural or engineered clay-rich material or of an interface between two repository components:

- Current evidence suggests that gas is able, in several cases, to create preferential pathways through these media at pressures below those at which macroscopic fracturing would occur. There is e.g. strong evidence that gas flows in bentonite through a network of pressure-induced pathways. One hypothesis is dilatancy-controlled gas flow as described by NAGRA (2008) [32] where it is assumed that micro-fractures form as a result of overpressure. The process of gas-driven micro-fracturing leads to an increase of the pore space, which is accompanied by an increase in intrinsic permeability.

- Once the gas pressure locally exceeds the stress required for macroscopic fracturing (usually equated with the sum of the minimum principal stress and the tensile strength of the rock), a preferential pathway can be also created due to tensile failure in a plane perpendicular to that stress. This is regarded as an important issue because of the potential for macroscopic fractures to provide a route by which groundwater transport of radionuclides could bypass confinement provided by the host formation.

The nature of the preferential pathway that will develop in a clayey host rock and the pressure at which it will be induced is strongly dependent on the gas entry pressure and on the stress distribution around the excavations and hence on the depth of the disposal units. The opening and/or propagation of preferential pathways come to a halt when the gas pressure becomes less than the value of the minimum principal stress (shut-in stress).

A study undertaken by Mallants and Jacques (2004) [33] proposed a small water flux would occur during the generation of a preferential gas flow path. The radionuclide transport due to this flux would be much faster than that due to a diffusive flux. Gas pathways may also transport radionuclides that are present as volatile species.

(b) Uncertainties relating to and treatment in the safety case of pathway dilation & gas-induced fracturing

Uncertainties on the stress-strain behaviour of clayey host rocks and engineered barriers in response to gas pressure build-up (dilation, micro- and macro-fracturing, cyclic opening and closing of a preferential flow path, post-breakthrough behaviour) as well as on flow processes in gas pressure-induced fractures are considered as particularly relevant issues for all waste types. Indeed, under these circumstances, it is not certain that conventional two-phase flow-models are properly applicable to the gas transport mechanisms operating in these materials [5].

Because pathway dilation does not create a discontinuous pore network, gas flow may be still represented by a continuous two-phase flow model applicable to a porous medium. The main difference with respect to the conventional two-phase flow is that the transport properties of the porous media are deformation dependent and can no longer be viewed as invariants. Moreover, a conventional porous medium type approach is frequently used in PA, even if there is doubt about the accuracy with which this represents the processes occurring, as models of this type are the only ones currently available for application at the field scale (GASNET, 2003). Parameters are chosen on the basis of available information and varied to ensure that predictions bound the range of possible behaviour.

However, continuous fluid flow models are no longer be applicable to discrete flow paths (i.e. macro-fractures) where non-Darcian flow is expected. Where such models are considered inappropriate, other sorts of simple scoping or bounding calculations of the way gas would migrate from a repository can be undertaken. These would try to represent the controlling physical processes in a simplified way (see e.g. Volckaert and Mallants, 1999 [34]). The significance of the increase in the calculated radionuclide flux is greatly dependent on the hypotheses made regarding the extent and geometry of the pathway, the availability of dissolved radionuclides at the time of pathway development or the amount of contaminated water flowing through the pathway.

Given the uncertainties associated with flow modelling after the onset of pathway dilation or gas-induced fracturing, uncertainties on the break-through and closure pressures of clay barriers are of particular importance. Additionally, these may determine the maximum pressure that could occur in the repository. Uncertainties on the role of anisotropy and geological structures in accommodating excess gas in clay host rocks are also considered as an important issue. In particular, the possible influence of anisotropy of plastic clay on the development of fractures after gas breakthrough has been identified as an issue deserving further attention.

The potential consequences of gas-induced irreversible damage to the clay host rock and to clay-based seals are sometimes assessed through the use of an alternative evolution scenario. It may also be covered by the expected evolution scenario when e.g. the healing of the fractures created by excavation is not envisaged. Different test cases may also be used to assess the likelihood of exceeding a critical pressure value.

Gas breakthrough in response to gas pressure build-up in clay-rich engineered barriers and host rocks was investigated respectively in WP3 and WP5.

3.4.2.5 Gas migration through interfaces

Regardless of the host rock and waste types being considered, the way and the extent to which gases migrate within a geological repository are greatly influenced by

the gas transport properties of the interfaces between its engineered and natural components. Hence, the lack of data and uncertainties on gas transport processes and on the presence of voids at the interface between components (due e.g. to ground support corrosion, alkaline disturbance) have been identified as important issues.

The EDZ around excavations in clay host rocks is one of the interfaces of interest as it could act as a preferential gas migration path and reduce gas pressure in the repository. Indeed, the increased permeability and reduced strength of the EDZ is expected to lead to reduced gas entry pressure and an increase in pore space for gas storage. In particular, the possible impacts of the near field evolution on porosity and pore structure, anisotropic gas migration properties, resealing capacity and gas breakthrough pressure of EDZ are considered as issues needing further attention. Although EDZ in crystalline rock masses might also locally alter gas migration, it is commonly expected that gas flow will remain mainly governed by pre-existing fractures.

Uncertainties on the migration processes as well as on the break-through and closure pressures along the interfaces between bentonite and other materials have also been identified as safety-relevant issues.

Gas migration through interfaces has been considered in WP3.1, 3.3 and 4. Sensitivity analyses considering the existence of different interfaces have also been performed in the benchmark study carried out in WP1.2.

3.4.2.6 Gas reactivity

Gases generated in a repository may interact in different ways with the media through which they migrate. Beside more physical processes such as sorption and dissolution, gases may undergo chemical (abiotic) and microbially mediated reactions in the subsurface environment. The potential implications for safety of these interactions are manifold as they may alter the physical and chemical characteristics of the migrating gas as well as of different phases of the media where they take place.

(a) Gas uptake

Gas uptake can occur in different media through chemical reactions (e.g. cement carbonation), precipitation or sorption onto solid phases possessing a high porosity and specific surface (e.g. clay minerals, zeolite, coal). Didier et al. (2011 [35], 2012 [36]) have e.g. studied the sorption of hydrogen gas at high temperature on dry clay in relation to redox processes. Due to their chemical affinity, methane and other alkane gases are also known to sorb onto natural organic matter intimately associated with clay minerals.

Uncertainties on gas sorption properties of different host rocks and of surrounding formations into which a gas phase could migrate are considered as an issue in several programmes in particular for the disposal of LLW/ILW for which larger amounts of generated gases are expected. However, gas sorption processes are sometimes neglected to cope with associated uncertainties. This issue has been considered in WP5.3.

Uncertainties on carbon dioxide sequestration by carbonation of cementitious components have also been identified as a relevant issue for several disposal

concepts relying of such components. WP3.4.1 intended to study the effect of carbonation on gas migration in cementitious barriers.

(b) Gas-induced redox reactions

Hydrogen is a potent reducing agent, which could contribute to maintain or to restore strongly reducing conditions generally considered to decrease the mobility of radionuclides. However, the reaction of hydrogen with clay components and iron-containing materials could also affect the stability of clay minerals and the corrosion of the metallic barriers.

In the absence of catalyst and reactive surface, hydrogen is relatively inert and therefore abiotic reactions may be slow at room temperature. At high temperature hydrogen can be more rapidly consumed by various inorganic reduction reactions such as Fe(III), sulphate and pyrite reduction (for instance, H_2 can react with the magnetite (Fe_3O_4) layer passivating steel overpacks). The reduction of the structural Fe^{3+} present in Fe_3O_4 can lead to the transformation of magnetite into more soluble ferrous hydroxide. In the same way, hydrogen may also be consumed by the dissolution of iron oxy-hydroxide produced by pyrite oxidation in the EDZ.

H_2 and other reducing agents generated as intermediate products (e.g. sulphide, thiosulphate) can also react with Fe(III)-bearing clay minerals as observed by Stucki et al. (1996 [37], 2011 [38]), by Lear and Stucki (1985, 1989) and by Manceau et al. (2000). These studies have shown that the structural Fe^{3+} present in the dioctahedral layer of iron-rich clay minerals can be reduced into Fe^{2+} by hydrogen or other powerful reducing agents. The isomorphic substitution of Fe^{3+} by Fe^{2+} in the octahedral layer may induce the collapse of smectite, possibly impacting safety functions of clay barriers. Truche et al. (2010) also studied the reaction of H_2 with pyrite. Pyrite is thermodynamically unstable in the presence of H_2 and is reduced into pyrrhotite (FeS) and H_2S. Pyrite reduction already significantly occurs at 90 °C and could represent a potential sink for H_2 in the immediate vicinity of heat emitting waste. The release of H_2S may also increase the corrosion of metallic canisters.

(c) Gas-affected microbial activity

H_2 is also one of the most energetic substrates for microbial life in deep subsurface environment and can fuel bacterial activity. Micro-organisms are able to use hydrogen as electron donor in many redox processes on which they rely for their energy source. Hydrogen can support the development of many chemolithotrophic bacteria such as nitrate reducing bacteria (NRB), iron reducing bacteria (IRB), sulphate reducing bacteria (SRB), methanogens, and homoacetogen bacteria.

Iron reducing bacteria (IRB) can perform the bio-reduction of structural Fe(III) in ferric smectite (nontronite) in the presence of hydrogen ([13]; [38]; [39]; [40]; [41]; [42]). In a similar way, they can also transfer electrons to the Fe(III) present in passive magnetite (Fe_3O_4) layer coating the steel overpacks. In both cases the production of Fe^{2+} leads to the destabilisation of the crystal lattice structure of the iron-bearing minerals. Hence, IRB have also the potential to provoke the collapse of the smectite and to promote the bio-corrosion of steel canisters in the presence of hydrogen under anoxic conditions ([13]; [41]).

Gas conversion by biochemical reactions has been identified as a relevant process for LLW/ILW. The bacterial development may indeed contribute to gas conversion or consumption. Various chemical transformations induced by microbial activity can also contribute to a net decrease of the number of gaseous molecules and hence of total

gas pressure in the disposal system. Methane containing ^{14}C may also be converted to $^{14}CO_2$ or $H^{14}CO_3^-$ during its migration to or within the biosphere which may lead to retention as carbonates. This process is of importance as $^{14}CO_2$ represent a greater radiological hazard than $^{14}CH_4$ [5].

Many more reactions are possible as illustrated by Meleshyn (2011 [43]) in a recent literature review showing how the bacterial development may alter the porewater chemistry and geochemical conditions. However, space and water restrictions (e.g. use of high density bentonite), high pH (e.g. use of cementitious barriers) and nutrients depletion (controlled by diffusion) are important limiting factors of the microbial activity after the closure of a deep repository. Hence, it is of prime importance to limit by design the voids and the open spaces left in a repository after its closure.

3.4.2.7 Gas migration in the biosphere

Gas in the biosphere did not form an area of study in the FORGE project. The remainder of the section provides a background to this subject only.

The radiological impact associated to radioactive gases is dependent on the manner of their release to the biosphere, including e.g. the area over which it is released and the biological uptake path that is followed [5]. The manner of release is likely to be highly concept-, site- and scenario-specific (e.g. gradual release vs. human intrusion scenarios). Hence, the pathways that need to be considered in a safety assessment have to be identified for each particular site and concept.

Once the gas has left the host rock, its migration to the biosphere would be determined by the physical nature of the geosphere (e.g. the presence of cap rocks), the contact with deeper groundwater and dissolution and dilution in near-surface groundwater and the composition of those waters. In some cases the combination of such site-specific features may remove free gas by dissolution in groundwater and prevent the release of free gas to the biosphere [15].

^{14}C may be released as methane ($^{14}CH_4$) or carbon dioxide ($^{14}CO_2$). It is sometimes assumed that if ^{14}C is released as methane from the repository, it will be oxidised to carbon dioxide by soil organisms. Exposure pathways which may have to be assumed in the assessment of the radiological impact of ^{14}C include [5]:

- Release of methane directly to the atmosphere, followed by inhalation;
- Conversion in the near-surface soils to carbon dioxide, which is taken up by foodstuff plants and then ingested;
- Dissolution in near-surface aquifers from which drinking water is drawn.

The radiological impact associated with radon (^{222}Rn) may be significant in case of human intrusion or when ^{226}Ra (initially present or as a result of ingrowth) migrating from a repository accumulates near the surface. In that case, the radiological impact is site-specific and can be assessed employing conservative assumptions or by means of a model of radon stripping from geological formations. Radon is a noble gas and does not undergo chemical transformations. The radiotoxicity of radon and its radioactive daughter products is often assessed assuming an indoor inhalation exposure pathway. When the release from the repository and subsequent geosphere transport occurs via groundwater, this impact can also be assessed as part of the groundwater release pathway [5].

3.4.3 Management of gas issues: possible strategies

Safety-relevant gas-related issues need to be considered from the outset in developing a repository concept and in formulating an approach to a safety case, so that a consistent approach is adopted across all issues [5]. The generation, accumulation and migration of gas in a geological repository will vary with the waste types to be disposed of and the repository concept. Applying adequate strategies to control these processes will limit their potential adverse impacts on the performance of the various barriers of the repository and, ultimately, on its long-term safety.

3.4.3.1 Design strategies

Different design strategies aiming to reach one or several of the following objectives may be adopted:

1. Ensure that the release of gases to the biosphere does not result in regulatory limits being exceeded;
2. Prevent the degradation of the performance of repository barriers;
3. Reduce uncertainties on factors controlling the generation and migration of gases;
4. Limit adverse consequences of gas release in case of human intrusion.

The strategies to be followed are directly dependent on the waste inventory as well as on the boundary conditions associated with the host rock and its environment. These boundary conditions include the availability of water and chemical conditions (controlling the corrosion processes and the free gas phase), gas entry pressure (controlling the gas accumulation and the subsequent pressurisation) and sealing capacities (controlling the resilience of the host-rock) [15].

3.4.3.2 Approaches for treating gas-related issues in the safety case

The possible impacts on safety of repository gases that cannot be avoided by design need to be addressed in the safety case. The consideration of gas generation and migration in the safety case may have several purposes:

- To identify safety-significant uncertainties;
- To assess the radiological impact associated with gas migration through the repository as a consequence of gas-induced displacement of contaminated water or of migration of volatile radionuclides;
- To assess the possible implications of gas generation and migration on safety functions;
- To substantiate that the design of the repository has been optimized with respect to these possible implications.

Recognition and appropriate treatment of uncertainty is an important aspect of any safety case for the geological disposal of radioactive waste, and applies to gas issues. 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.

The treatment of gas issues in the safety case starts generally with the identification of relevant processes and uncertainties e.g. through the use of simple scoping calculations. Processes that can be shown to be insignificant for safety can be ignored in subsequent steps of the assessment. When a process needs to be treated explicitly in the safety assessment, this is typically done considering specific modelling assumptions or parameter values in dedicated assessment cases or scenarios. Conservative model assumptions are often used to deal with uncertainties on gas-related processes. The calculations may also take the form of "what-if" studies rather than treatments in detail of possible evolutions of the system. Regardless of the approach used, the description of a particular process must be sufficiently detailed to justify, by arguments founded on scientific understanding, its treatment in the safety assessment and in particular the way uncertainties are taken care of.

The way in which the construction of gas-related scenarios is carried out varies quite widely from waste management organisation to waste management organisation, primarily because of differences in repository concept. Whether it is appropriate to consider the effects of gas in alternative evolution scenarios rather than in the expected evolution scenario depends on the quantities of gas expected to be produced and the confidence that exists that this is not likely to affect barrier performance [5]. The radiological impact associated with volatile radionuclides may also have to be examined through the use of specific scenarios such a human intrusion scenario.

The justification and validation of the models used for representing gas-related processes constitute important aspects of the quantitative treatment of gases in the safety assessment. In particular, the up-scaling of gas transport properties and processes from the pore scale to the repository scale is regarded as a key issue in all disposal programmes. For example, two-phase flow models are commonly used to simulate gas transport at the scale of the disposal system although not necessarily representative of the phenomenology observed at smaller scales. Materials are also typically assumed homogeneous in PA models. Therefore, uncertainties associated with the presence and spatial variability of heterogeneities in the different repository components (e.g. geological structures, EDZ features, cracks in cementitious components) have to be considered in comparison with the assumption of properties of homogeneous components.

Whether the use of simplified models is appropriate greatly depends on the purposes of the modelling (e.g. process understanding, scoping calculations, design optimisation). The substantiation of their suitability has to rely on a thorough conceptual understanding of the processes taking place at the pore scale and of their significance at the scales relevant for the safety assessment. Therefore, the availability of experimental data for model justification and validation is a major issue in relation to the treatment of gas in the safety assessment.

3.5 Conclusions

A number of processes associated with the production of gases may have to be considered and taken into account when assessing the long term safety of a geological repository. The relevance of a particular gas-related process varies according to the characteristics of the wastes to be disposed of, the characteristics and layout of the engineered barrier system and the host environment. In the context of a safety case, the relevance and treatment of a specific process will also be greatly determined by the safety functions upon which the safety of the repository relies and the possible implications of gases on these functions.

The potential impacts of gas generation and migration on safety functions commonly considered in current geological disposal concepts in clay and crystalline rocks have been identified in section 3.3. Latter sections provide a more detailed discussion of these possible implications and of the issues to be dealt with in a safety case.

When a gas-related process is deemed to be of relevance to safety, an appropriate approach to its treatment in the safety case has to be chosen. The way the various types of uncertainties associated with a process are handled in the safety case is particularly important. Main sources of uncertainties include the lack of data or experimental evidence, insufficient process understanding, upscaling from laboratory to repository scale as well as shortcomings of modelling tools.

Justifying the suitability of the adopted approach requires sound arguments founded on scientific understanding. The most recent developments in the understanding of key gas generation and migration processes identified in the current section are presented in section 4; this section also addresses specific issues of particular relevance to the modelling of gas migration processes such as upscaling and the role of interfaces between different repository components.

Recommendations for the treatment of gas in the safety case and for developing strategies to control, limit and minimize potential adverse impacts on long-term safety, as discussed in section 3.4.3, are provided in section 6.

4 RECENT DEVELOPMENTS IN UNDERSTANDING REGARDING GAS-RELATED ISSUES IN THE CONTEXT OF GEOLOGICAL DISPOSAL

4.1 Introduction

This section discusses recent developments in understanding of gas generation and gas migration in the context of geological disposal. Output from the FORGE project is discussed, and to ensure comprehensiveness, output from relevant work outside of FORGE is also discussed.

The output provided from FORGE Work Packages 1.2, 1.3, and 2 to 5 are authored by respective work package leaders, and summarise learning derived from these tasks. The output was provided in response to a series of structured questions derived by Work Package 1.1 participants, with the aim being to allow an overall synthesis of the consideration of gas in the safety case for a geological disposal facility to be produced.

Summaries are provided in this section from FORGE WP2, 3, 4, 5, 1.2 and 1.3 in that order. Hence gas generation (WP2) is discussed initially, followed by gas issues in the context of the EBS (bentonite, cement), gas issues in the context of disturbed host rock and gas issues in the context of undisturbed host rock. Consideration of the treatment of gas issues in modelling of the type that supports the safety case is then provided.

4.2 Gas generation relevant to geological repository conditions: Work from FORGE and from outside of FORGE

4.2.1 Introduction

Section 3 presented information on gas generation in the context of geological repository conditions. Building on this, the current section presents the main outcomes of the work carried out within FORGE WP2 on the gas generation rate issue, as well as results and key references of various relevant studies performed outside the FORGE project.

4.2.2 Gas production through corrosion of metals in cementitious L/ILW disposal systems

Accurate measurement of corrosion rates of metals and alloys under alkaline conditions in which passivation occurs (e.g. carbon steel, stainless steels, nickel alloys and Zircaloy) was of limited interest prior to studies of disposal of L/ILW in the 1980s because rates of corrosion of much less than 1 µm/a were low enough that it was of little technological interest to determine the rates more accurately. In the case of repositories in clay rocks, however, in which gas pressure build-up may be an issue, reliable determination of the actual associated gas production rates may be important.

Several methods of measuring the corrosion rate of carbon steel in high pH anoxic environments have been employed during the past decades. These methods include gas evolution, weight loss, and electrochemical techniques. However, these methods usually provide different values for the corrosion rate for a number of reasons. The high sensitivity of gas evolution measurements requires only a short exposure duration (a few days) to obtain a measurable amount of H_2 gas. Thus gas

measurements permit an almost continuous determination of corrosion rates, and the (near) instantaneous rate can be determined after long times. In contrast, weight loss methods inherently overestimate the corrosion rate since a small amount of non-oxidized metal always dissolves in the solution used to remove the surface oxides prior to weighing. Additionally, weight loss measurements require long exposures (months or years) in order to obtain the relatively large amount of corrosion products necessary to minimize the effects of measurement errors. As a result, the measurements represent an average value of the total exposure period including the initially high rate that occurs prior to stabilization of the passive film and can provide little accurate information on the evolution of the corrosion rate with time. In addition, electrochemical measurements have also been found to give higher corrosion rates than those derived from gas production ([44], [45]) and correct interpretation of the measurements requires a detailed understanding of all surface reaction mechanisms taking place, something which is not always straightforward especially in complex geological systems. Consequently, the results discussed here are largely based on measurements using gas generation methods.

It should be noted that the early stages of repository evolution may differ significantly for different repository concepts and that this may affect the timing of the onset of gas evolution. Furthermore, if the depth of corrosion penetration is of interest because of the need to evaluate containment lifetime, other methods for assessing corrosion mechanisms and measuring corrosion rates are also relevant, but this is not further discussed here.

4.2.2.1 Corrosion of carbon steel

Overview – Under the high pH conditions of Portland cement-dominated systems, carbon steel and cast iron either present in waste packages or used as container materials for such packages corrode passively. For low salinity systems (salinity approximately below that of seawater) and for conditions in which the pH remains high (>10.5), passive films form on these materials and the corrosion rate (gas generation rate) gradually decrease towards a near steady state value, which is probably not attained in many of the studies. For the first ~100 hours, corrosion rates may be high (~10 µm/a), however the rate declines rapidly to values below 0.1µm/a. Passive film formation and dissolution at a constant rate does not represent an equilibrium condition, thus it is important when considering the use of such gas generation rates in safety assessment to consider factors that might affect the film properties. These include changes in flow conditions, pH and salinity, as well as stress and physical blocking effects that might influence evolution of porosity [46]. Additional factors, such as the presence of organic materials in wastes, the degradation of which might produce organic acids, may also influence the rates. It can be stated that provided a high pH is maintained, chloride concentrations do not exceed that in seawater and organic wastes do not influence corrosion chemistry, the long-term passive corrosion rate will remain the same or decline gradually almost indefinitely if there is little porewater exchange.

Results of a number of studies that determined the corrosion rate of carbon steel using gas generation methods are summarized in **Table 1**. The results represent instantaneous measurements of the rate after very long periods, although it should be noted that rates were continuing to decline in most cases. The results clearly show that the long-term corrosion rate of carbon steel in dilute and slightly saline groundwaters (up to 35000 ppm Cl) are typically in the range of 10-40 nm/a at ~25°C. Two measurements indicate somewhat higher values. In one case, the reasons appear to be related to the relatively short duration of the experiments (i.e. the rate measured after 375 days by Kaneko et al. (2004 [47]). In the case of Smart

et al. (2004 [48]), the reason for one experiment giving a higher rate of about 100 nm/a is not clear.

Rate (nm/a)	Time (days)	pH	Temperature (°C)	Other parameters	Reference
20	900	12.4	25	5000 ppm Cl	Mihara et al. 2002
4	375	12.6	15	0.4 ppm Cl	Fujisawa et al. 1997
20	375	12.6	30	0.4 ppm Cl	Fujisawa et al. 1997
~20	375	12.6	45	0.4 ppm Cl	Fujisawa et al. 1997
5-30	~450	12.5-12.9	21	~0 Cl	Kreis 1991
<100	375	12.5	20	20000 ppm Cl; short-term experiment	Kaneko et al. 2004
30	540	12.5	20	5000 ppm Cl	Kaneko et al. 2004
10-30	450-540	10.5 to 12.5	20	0 Cl	Kaneko et al. 2004
10-30	450-500	12.5	20	5000 ppm Cl; mortar-covered samples	Kaneko et al. 2004
30	800-900	12.5	35	0 Cl	Kaneko et al. 2004
20	800-900	12.5	20	5000 ppm Cl	Kaneko et al. 2004
40	460	13	50	20000 ppm Cl, OPC backfill-covered samples	Naish 2001
2-10	25-30	12.5.-13	50	mortar-covered samples; 100% RH	Newman and Wang 2010
2-16	25-30	12.5-13	50	Samples dipped in NaCl solution, then mortar covered, 100% RH	Newman and Wang 2010
2-16	25-30	12.5-13	50	mortar-covered samples; Opalinus Clay pore water	Newman and Wang 2010
20-30[3]	>1000	12.5	30, 50, 80	Various low Cl solutions	Smart et al. 2004
~30	3750	13	30, 50, 80	3.5% NaCl	Smart et al. 2004

Table 1 Long-term corrosion rate of carbon steel in alkaline porewaters determined using gas generation methods

Effect of salinity – The corrosion rate increase above about 30000 ppm Cl. There is little data at Cl^- concentration exceeding that of seawater.

[3] One of a large number of long-term measurements reported in Smart et al (2004, [48]) gives a value of about 100 nm/a.

Effect of coverage by cementitious backfill or grout – In many cases, carbon steel in a repository may be covered by cementitious backfill or wastes within containers may include cementitious mortar. There is some evidence that this can reduce the corrosion rate of steel [46], which may be due to Ca uptake by the passive film.

Effect of unsaturated conditions – The subject of corrosion under unsaturated conditions has been little studied, but is of some interest because the inflow rate into L/ILW caverns is expected to be very low in some clay rocks [32]. In such conditions, the corrosion rate as measured by gas production can be even lower, possibly in the range of 5-10 nm/a [49]. The reasons for this are not clear, but may involve blocking of pores thus limiting access of water or Ca uptake on in the corrosion product leading to a reduction in dissolution rate of the passive film.

Effect of temperature – The results of Smart et al. (2004 [48]) show no indication of a temperature dependency in the range of 30-80 °C. Nonetheless, the initial rate can be significantly accelerated at 45 vs. 15 °C, as noted by Fujiwara et al. (1997[50]). Careful inspection of his data nonetheless shows that the rate at 45 °C after very long times is in the range of 20 nm/a.

Kursten (2012 [46]) provides a more comprehensive discussion of results from many studies of corrosion in alkaline systems. In addition, Smart and Hoch (2010 [51]) have published a comprehensive review of corrosion data, in which they conclude, based on fitting of the data generated in the UK programme, that the long-term anaerobic corrosion rate of carbon steel in alkaline systems is about 5 nm/a.

Effect of pH– The decomposition of organics such as cellulose in various L/ILW packages can lead to the production of organic acids and CO_2 that may result in reduced pH values that can accelerate corrosion of metals. This will depend on various factors, including the organic/cement ratio within the waste package and fluid mass transfer considerations. In near neutral or slightly acidic conditions, corrosion rates may increase to several µm/a, thus hydrogen gas production can be greatly increased. However, in such conditions hydrogen may also be utilized as an electron source by microbes, thus the net hydrogen production may be dramatically reduced [52].

4.2.2.2 Corrosion of stainless steel

The corrosion rate of stainless steel in alkaline systems has been studied by Kaneko et al. (2004) [47], Smart et al. (2004) [48], Mihara et al. (2002) [53] and Wada et al. (1999) [54] using gas production methods. The long-term corrosion rates are typically in the range of 0.1 to 10 nm/a.

4.2.2.3 Corrosion of other metals

The corrosion rate of aluminium in cement grout is reported to be less than about 10µm/a (Fujisawa at al. 1997 [50]); however, galvanic corrosion through contact with steel can significantly increase the corrosion rate. Some metals evolve hydrogen under both aerated and oxygen-free conditions, notably magnesium and aluminium.

4.2.2.4 Corrosion of Zircaloy

The corrosion rate of Zircaloy at high pH was measured by Wada et al. (1999 [54]) in sea water-based groundwater with a pH of 10 to 12.5 over the temperature range of 30-50°C. The range of measured rates after 325 days was 0.05 to 0.3 nm/a. Kurashige et al. (1999 [55]) measured rates at 30°C and 45 °C in sea water-based solutions at pH values of 10.5 and 12.8. The reported long-term rates were 1-2 nm/a. Mihara et al. (2002 [53]) measured a corrosion rate of about 5 nm/a, as discussed in

Kaneko et al. (2004 [47]). The numbers of experimental studies for these conditions are limited, in comparison to the case for steels.

4.2.3 Gas production as a result of corrosion of metals in SF/HLW disposal systems

The principal materials under study for encapsulation of HLW include carbon steel and copper with cast iron insert. Depending on the repository design concept, carbon steel may be in contact with compacted bentonite, claystone or cementitious material. In contrast to L/ILW repositories, the canister surface temperature in temperature may be in the range of 90-100°C for many decades.

4.2.3.1 Carbon steel corrosion in bentonite or claystone

In the case of corrosion in bentonite, several studies of the corrosion rate using gas generation methods have been performed, as summarized by King (2008 [56]).The long-term corrosion rate in anoxic compacted bentonite at 25-80°C is in the range of 1-2 µm/a in dilute and moderately saline (seawater) groundwaters. It is notable from the studies referenced by King that the achievement of steady state rates can take years for corrosion in contact with compacted bentonite, in contrast to weeks to months in water without clay, which may be related to strong sorption of Fe(II) on clay that destabilizes the protective film. The corrosion rate of carbon steel in claystone may be somewhat higher (Crusset et al. in press). It should also be noted that except for archaeological analogue studies, there are few in situ corrosion data for steel, which is a limitation since potential microbial impacts on corrosion need to be assessed. The effect of gamma radiation on hydrogen gas production has received limited attention, but has been studied within the FORGE project (see below).

The objective of WP2 studies was to further assess the effects of parameters representative of repository conditions (T, Eh, reactive surface, solid and/or liquid phases in contact with steel, irradiation etc) on corrosion rates of carbon steel and consequently on hydrogen production rates. The experiments carried out by UJV (Ustav Jaderneho Vyzkumu; *Czech Republic Nuclear Research Institute*) involved measuring continuously the volume of hydrogen generated from iron powder or carbon steel samples of various reactive surfaces in synthetic bentonite pore water, within a closed system at various temperatures (40 to 80°C) and anaerobic conditions. In addition, a series of experiments was performed in presence of compacted bentonite. The effect of irradiation was assessed by IRSN (Institut de Radioprotection et de Sûreté Nucléaire) using iron powder in de-aerated pure water, in an “open” (flushed) system with continuous monitoring of hydrogen production by gas chromatography, for two dose rates (50 and 100 Gy/h).

A rather large range of hydrogen production rates was measured in the UJV studies, with initial rates up to 10 mol $H_2.m^{-2}.yr^{-1}$ and rates after formation of a corrosion layer from 0.02 to 6 mol $H_2.yr^{-1}.m^{-2}$ (corresponding to corrosion rates in the range 0.1 to 30 µm/a approximately) depending on conditions of corrosion. Though most of the experiments were carried out on short durations (30 days), a 1-year experiment based on hydrogen continuous measurements as well as on weight loss measurements after dismantling, leads to values within this range, with a steady state value of 1 $mol.m^{-2}.yr^{-1}$ corresponding to about 5 µm/a) (**Figure 3**).

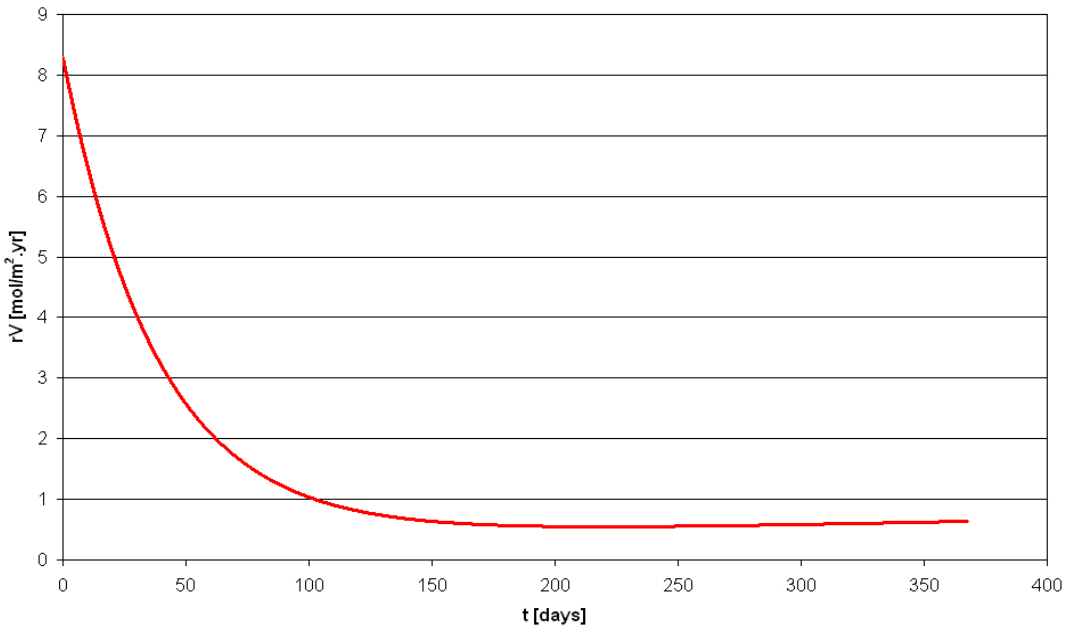


Figure 3 **Hydrogen generation rates measured continuously during a 1 year experiment (ratio of carbon steel surface to synthetic bentonite water 141 cm^2/300 ml, temperature 70 °C)**

Overall, WP2 experiments show that reactive surface area acts on hydrogen generation rates through its effect on the evolution of redox conditions as a result of corrosion, which conditions the stability of the corrosion layer. As illustrated in **Figure 4** for a series of experiments, the Eh is initially slightly above 0 mV but decreases to low values immediately after immersion of the sample in the solution. This indicates that the remaining oxygen quickly reacts with the carbon steel surface to form corrosion products, such as ferrous hydroxide or magnetite. Then Eh increases due to oxidation and the formation of corrosion products with ferric ions, such as maghemite or hematite. The observed trends of increasing corrosion rates at the end of the experiment with 1 plate suggests that the reaction of iron with oxygen was not sufficient (Eh of -70 to -250 mV) to form a stable protective corrosion product layer in comparison with the experiment with 10 plates (Eh of -200 to -350 mV). In another series of experiments with powders, a stronger decrease in Eh (around -400 to -500 mV) produced a less protective corrosion (higher hydrogen generation rates per m^2) than that observed for a lower Eh decrease (around -200 mV). An important parameter is thus the ratio of surface of metals to the volume of water, as it primarily affects the oxidation potential of water in contact with metals: while the (slight) presence of oxygen impacts hydrogen generation rates at the beginning of corrosion until a sufficiently protective layer is formed, strongly reducing environment can also favour hydrogen generation.

The oxygen content in the surface layers of corroded samples was found to be higher than the stochiometric ratio of Fe/O in magnetite or maghemite, suggesting that the composition of passive corrosion products layers is not only formed by an inner layer of magnetite and outer layer of maghemite as proposed in previous papers (Kruger, 1989 [57], Lu and Macdonald, 2008 [58]).

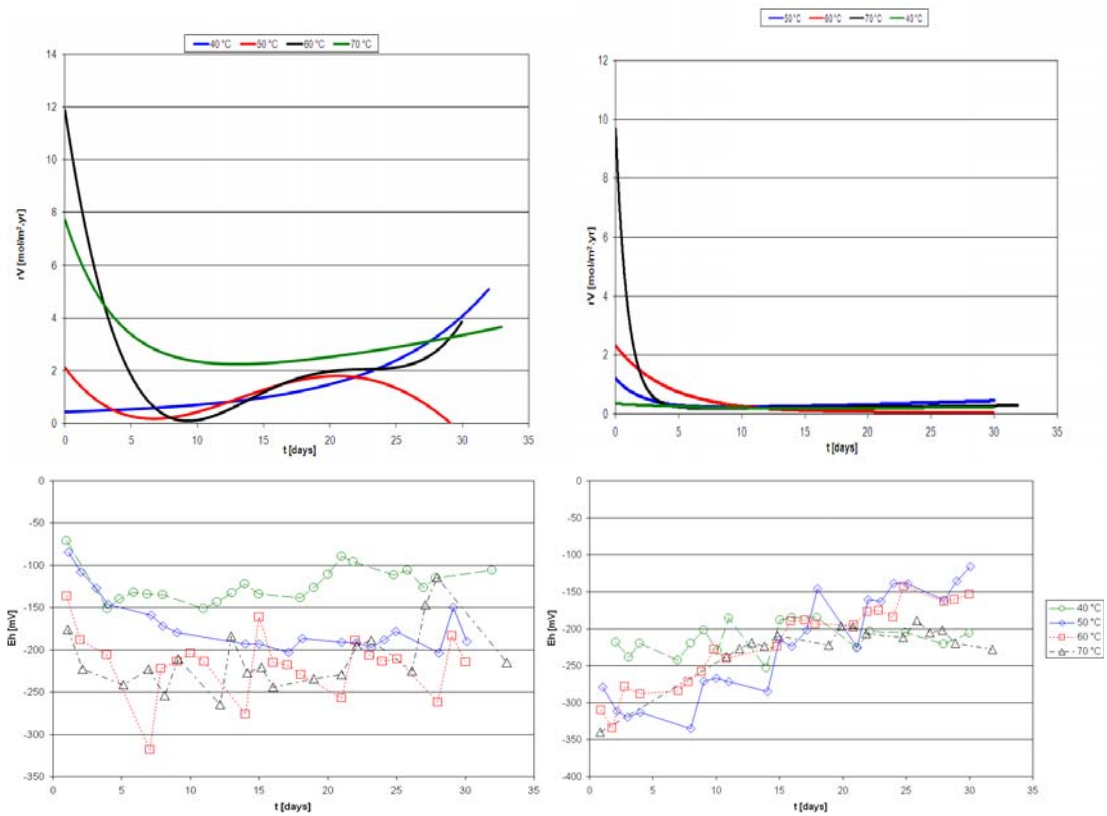


Figure 4 **Hydrogen generation rates and Eh evolution for experiments with one sample (left) and ten samples (right) of carbon steel in 2 litres of bentonite water at various temperatures in anaerobic box (O_2<0.1 ppm)**

In the experiments illustrated in **Figure 4**, a strong enhancement effect of temperature (40 to 70°C) on hydrogen generation rate was shown at the very beginning of corrosion: the higher the temperature is, the higher the initial hydrogen generation rate. The temperature effect depends on the nature and stability of the corrosion layer. Globally, WP2 experiments tend to indicate that hydrogen generation rate is not proportionally related to temperature (in the tested range), except at the very beginning of the corrosion process.

Bentonite in the solid form emplaced in a permeable bag added to the solution leads to a slight increase in the hydrogen generation rate form iron powder. On the contrary, the average corrosion rate of a carbon steel plate in contact with compacted bentonite after 30 day experiments was one order of magnitude higher compared to that measured in solution without bentonite. Such an increase due to a direct contact between carbon steel and compacted clay, consistent with previous studies [56], may be due to sorption of iron species on bentonite which thus do not contribute to the formation of an adherent corrosion layer.

In the IRSN gamma irradiation studies, similar behaviour was observed for the two experiments carried out at 50 and 100 Gy/h: an increase of hydrogen production because of water radiolysis, followed by a plateau and again a continuous increase in hydrogen production (**Figure 5**). 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 2 to 6 10^{-2} mol.m^{-2}.y^{-1}(0.14 to 0.4 µm/a), i.e. in the range of values reported in literature. After irradiation the corrosion rate is

increased by a factor higher than 10 while no stationary state was obtained at the end of the experiments. These experiments indicate that corrosion processes are modified under ionizing radiations, the result being a significantly enhanced production of hydrogen. However, no clear explanation is found at this stage of investigation regarding the apparently continuous acceleration of hydrogen production rate.

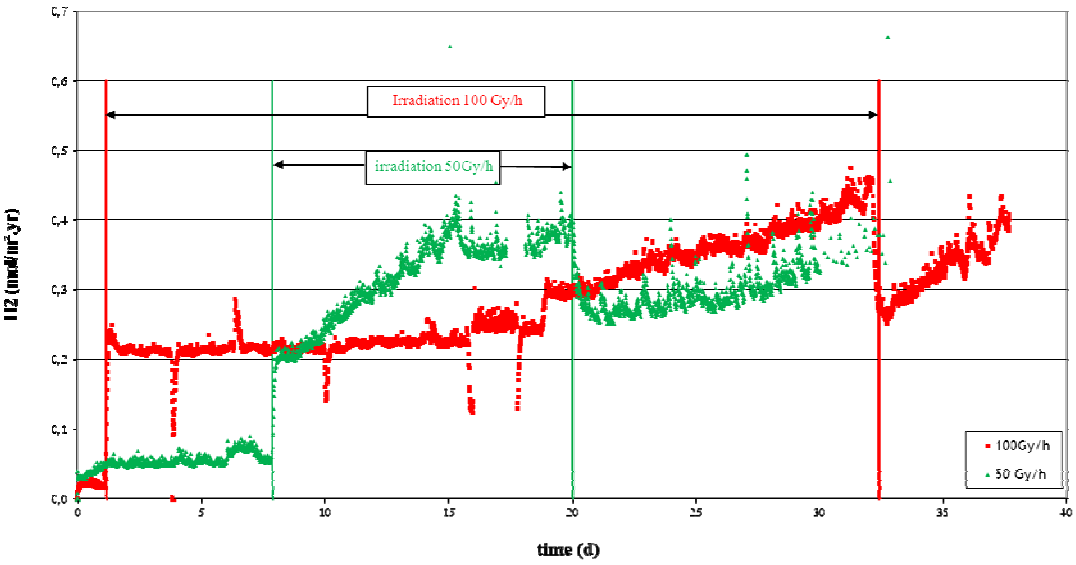


Figure 5 **Hydrogen production rate (mol.m².yr^{-1}) versus time (day) for 50 and 100 Gy/h experiments - mass of iron ~10 g; mH_2O ~100 g**

4.2.3.2 Carbon steel corrosion cementitious porewater

In the Belgian supercontainer concept for SF and HLW disposal, carbon steel may be in direct contact with cementitious material [46]. The overall corrosion behaviour is as discussed in Section 4.2.1, except of higher temperatures and a more significant gamma radiation field. In cementitious porewaters at temperatures of ~80-90 °C, the long-term corrosion rate is in the range of 0.1-0.2 µm/a in the absence of radiation. This rate is expected to decline to <0.1 µm/a [46]. No difference has been measured between long-term corrosion rates in the absence and presence (25 Gy/a) of gamma radiation.

4.2.3.3 Copper corrosion in bentonite

Some studies have reported to have measured the production of hydrogen from the corrosion of copper under anoxic conditions in pure water [59]. These studies have proven difficult to reproduce [60]. Nonetheless, even if they are correct the equilibrium hydrogen pressure for anoxic corrosion of copper would appear to be in the range of 1mbar. Such a low equilibrium pressure would lead to corrosion by this mechanism stopping because the hydrogen would accumulate in the bentonite buffer [61]. From a gas production perspective the process can be considered irrelevant.

4.2.3.4 Zircaloy fuel cladding corrosion

In the case of spent fuel disposal, the gas production as a result of corrosion of Zircaloy is also relevant. Shoesmith and Zagidulin (2010 [62]) in their detailed review of the electrochemistry and corrosion of Zircaloy suggested that the corrosion rate of Zircaloy would be about 5 nm/a at near neutral pH.

4.2.4 Gas production as a result of degradation of organics

An overview of microbial degradation of organics in L/ILW is given by Rodwell et al. (2003 [5]). The microbial degradation rates of organic wastes such as cellulose, plastics, ion exchange resins and bitumen and the associated gas production are difficult to predict and are likely to depend on the viability of microbes at pH values above 12.

4.2.5 Microbially induced gas consumption and production

Various types of microbes can influence gas production and consumption in repository environments. It has been established, e.g. that some SRB can utilize hydrogen (Pedersen [63]). In addition, other bacteria can utilize organic matter and nitrate present in bituminous wastes to produce nitrogenous gases, methane and CO_2 [64]. In a cementitious near field, the high pH is expected to suppress microbiological activity, although it may not stop it altogether.

The activity of microbes can also be significantly diminished in material with very small pore size, such as highly compacted bentonite ([63], Stroes-Gascoyne [65]) and claystones. In contrast, Boom Clay, with its larger pore size, exhibits substantially higher microbial activity, which could consume hydrogen produced by corrosion, thus increasing sulphide formation.

4.2.6 Modelling of gas generation under repository conditions

The modelling of gas generation under disposal conditions typically involves a set of simplifying assumptions, including:

1) Hydrogen generation due to anaerobic corrosion is assumed to begin upon waste container emplacement. The initial oxidizing period of some years to decades is thus ignored from a gas production perspective, although for many metals the oxygen must be consumed before hydrogen evolution can begin.
2) The high initial corrosion rate of metals that form passive or corrosion-resistant films (steels, nickel alloys and Zircaloy)are neglected as the period of high rates (months to a few years) represents a very small fraction of the total corrosion duration. Although this is not a conservative assumption it is a reasonable approximation.
3) Gas production rates are assumed to remain constant until the metal is consumed; this implicitly assumes that corrosion conditions (pH, salinity) remain constant or at least within the bandwidth of values derived from the gas generation experiments that have been used for the assessment calculations.
4) Consumption of gases (e.g. utilization of hydrogen by SRB) does not occur.

4.2.7 Synthesis and summary – FORGE Work Package 2

Studies in WP2 dealt with 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 pore water, a process related to early evolution in a repository. These represent areas among a much larger range of issues that had already been studied prior to FORGE related to gas generation in repositories. The discussion below includes all these aspects.

The principal sources of gas generation in repositories are anaerobic corrosion of metals and decomposition of organic matter, the latter being essentially only a significant issue for disposal of L/ILW. The gas production rates from corrosion are

strongly influenced by the specific conditions in the repository, thus the corrosion evolutionary path, which describes processes such as oxygen consumption, radiolysis, salinity changes, pH changes and temperature evolution, must be clearly defined as a basis for establishing the hydrogen gas production rates to be used in safety assessments.

Considerable progress was made prior to the FORGE project in the area of determining hydrogen gas production associated with anaerobic corrosion of metals. In the case of cementitious environments, very low anaerobic corrosion rates (typically much less than 100 nm/a) have been measured for steels. There are nonetheless some conditions for which further study may be required, as differences in repository design concepts can influence the gas evolution rates. If the pH can gradually decline, for example, as a result of groundwater flow through the near field, corrosion rates and associated hydrogen production may increase as a result of loss of passivity. Furthermore, in the case of the presence of organic material in wastes, the influence of their degradation on corrosion-induced gas production rates, e.g. through production of organic acids, may need to be considered.

In the case of carbon steels in neutral pH disposal environments, WP2 studies of the corrosion rate in compacted bentonite show that the initial corrosion rate is greatly accelerated (up to tens of µm/a) in the first month compared to the rate in bentonite porewater. The increased rates are thought to be due to enhanced dissolution of the protective film on the steel as a result of sorption of Fe(II) onto the bentonite. The long-term corrosion rate, i.e. the steady-state rate after several years, has been determined in several prior studies to be a few µm/a, which still represents a higher rate than is measured in pore water without compacted bentonite. Other WP2 findings show that the initial corrosion rate of carbon steel is significantly higher at elevated temperature (70°C) than at lower temperatures, but the rate decreases rapidly and there is no significant temperature dependence after approximately one month. As a result, the higher initial hydrogen production rate is not likely to be important from a long-term performance assessment perspective.

Additional studies in WP2 found that gamma radiation at dose rates in the range of 50-100 Gy/h enhances both hydrogen production and the corrosion rate of carbon steel. Furthermore, the enhancement of the rate was sustained even after irradiation ceased, the reasons for which are not yet fully understood. The studies may be relevant to some disposal concepts, but cannot be extrapolated to other conditions (e.g. lower dose rates or the presence of clay).

A process that may be relevant to corrosion and hydrogen generation for both steel and copper is microbial corrosion. This was not studied in FORGE, but received some attention in prior laboratory studies. Further in situ studies may be warranted.

The approach to modelling gas generation for safety assessments includes some simplifications that need to be checked depending on the application of the modelling. For example, higher short-term gas production rates are often not considered in long-term calculations of gas build-up, as the amount of gas produced is very small relative to the total produced over tens of thousands of years. However, in some cases, for example determining if there are any effects from short-term gas transients, this may not be appropriate and must be checked for the specific EBS design and hydraulic boundary conditions. A process that is normally conservatively ignored in assessing gas pressure build-up is the utilisation of hydrogen as an electron donor by microbes (e.g. SRB), which can reduce gas pressure.

4.3 Gas Transport Mechanisms

4.3.1 Bentonite-based EBS

4.3.1.1 Gas diffusion

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

The diffusion coefficient for air has been determined from measurements of steady-state volumetric flow through cylindrical samples of pure montmorillonite and natural bentonite (MX-80) exposed to certain air pressure gradients in a 1D geometry, see **Table 2**.

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

Table 2 Determined diffusion coefficient for air in given clay samples

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

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

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

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

In experiments with continuous increase of pressure from the reaction of iron with water, it was found that before the breakthrough a plateau with a relatively constant pressure was formed (**Figure 6**). The diffusion coefficients estimated from the region of this plateau was 3.7×10^{-10} m^2/s. In the similar experiment with sodium bentonite (Volclay KWK-20-80), the estimated value of the diffusion coefficient was 2.9×10^{-11} m^2/s.

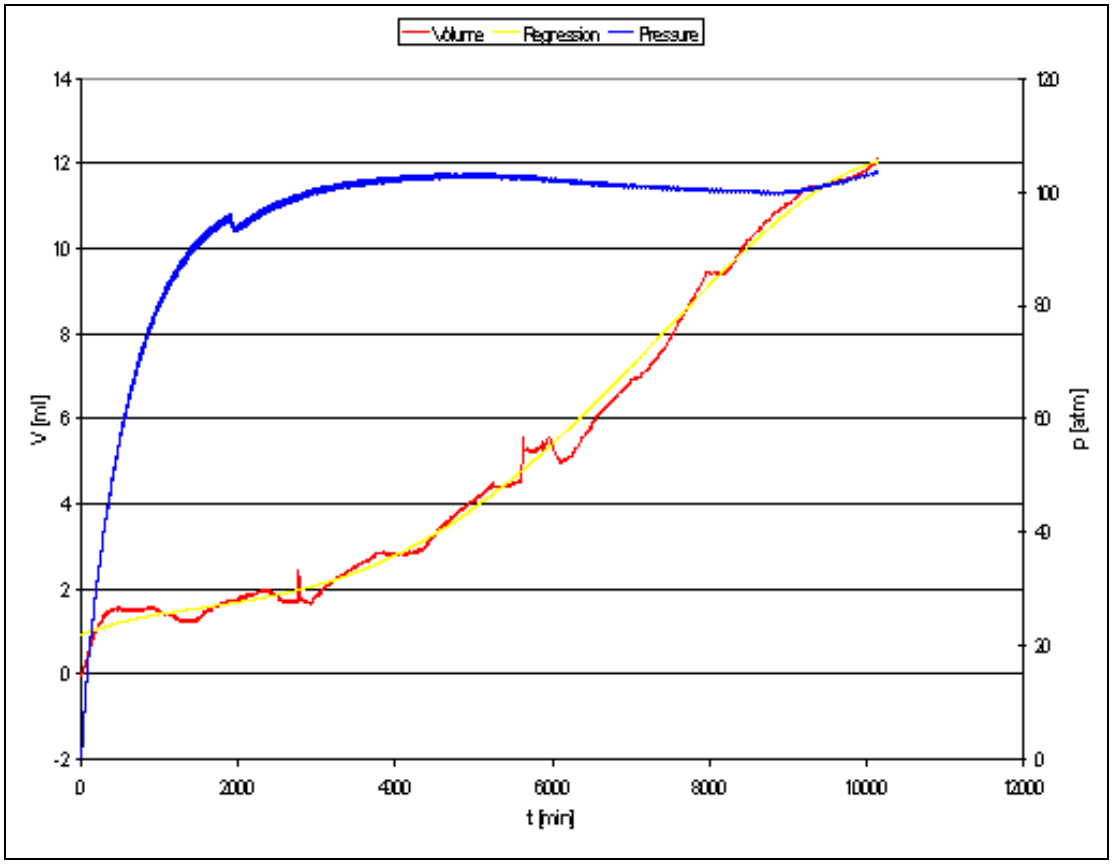


Figure 6 **Pressure plateau formed before the breakthrough of hydrogen through bentonite of density 1600 kg/m3 and related flow of hydrogen**

There is a remaining uncertainty regarding the true value of Henry's constant in the clay environment:

- The measurements done within FORGE are too few to give a comprehensive picture of the diffusivity of dissolved gases.
- However, within the density range reported, it seems like the diffusion coefficient decreases with density, molecular size and sodium content in the bentonite.

4.3.1.2 Free gas phase formation

In the case where more gas is generated than what can escape with diffusion a free gas phase will form. Initially, the gas phase will consolidate the clay phase. Experiments by Clay Technology have demonstrated, not only that a gas phase does not interact mechanically with water-saturated bentonite when its pressure is below the pressure of the bentonite, but also that mechanical interaction inevitably takes place when the gas pressure exceeds the initial pressure of the clay. This is clearly illustrated in **Figure 7**, which shows the pressure response of an MX-80 bentonite sample at differently applied gas pressures gradients.

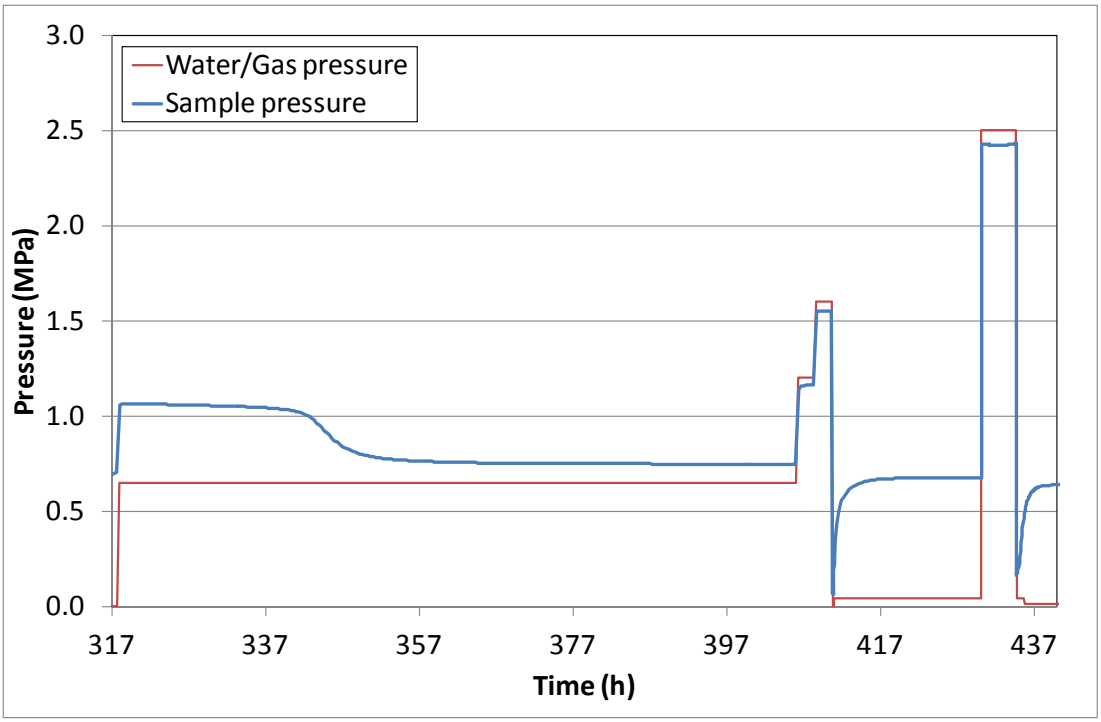


Figure 7 Response of an MX-80 sample which is being pressurized with gas above and below initial swelling pressure. The pressure of the sample is independent of gas pressure when the gas pressure is below the initial swelling pressure (~0.75 MPa), while it is basically equal to the gas pressure at higher injection pressures. Note that water is the pressurizing fluid at the beginning of the displayed pressure evolution (317 – 340 h).

An interesting feature of the test output recorded in **Figure 7** is that although it clearly demonstrates mechanical influence of the gas on the clay, it does not indicate any additional transport mechanism apart from the ever-present gas diffusion. In contrast, in other tests, gas breakthrough events have been demonstrated to occur for gas pressures at or above the pressure of the clay sample.

4.3.1.3 Two-phase flow

Two-phase flow conditions are encountered when gas invades a porous medium as a separate phase. This process is associated with the drainage of interstitial fluid as the non-wetting phase (gas) displaces the wetting phase (pore water). Flow is within the original inter-granular porosity resulting in an iso-volumetric boundary condition. Considerable care has to be taken on terminology – what is meant with regard to bentonite is visco-capillary flow where the properties of the clay capillaries are playing a control on gas displacing water.

Based on experimental data from CIEMAT work, two-phase flow seems to take place for degrees of saturation lower than about 93% in compacted bentonite and concrete [66].

The experimental work within FORGE clearly demonstrates that no two-phase flow occurs in saturated bentonite [67, 68].

4.3.1.4 Pathway dilation and gas-induced fracturing

Experimental evidence from LASGIT[4] shows that dilation is the predominant advective flow mechanism [68]. At the point of gas breakthrough there is a co-incident pressure and stress response seen at the deposition hole wall. Qualitatively similar results have been seen in laboratory tests and in gas injection tests 1 & 2 [68]. For gas test 3 (see **Figure 8**) gas break-through has been accompanied by a secondary rise in gas pressure, followed by a second break-through event with a more gentle form. These results strengthen the dilatancy path propagation hypothesis and cannot be explained by classical visco-capillary (two-phase) flow concepts.

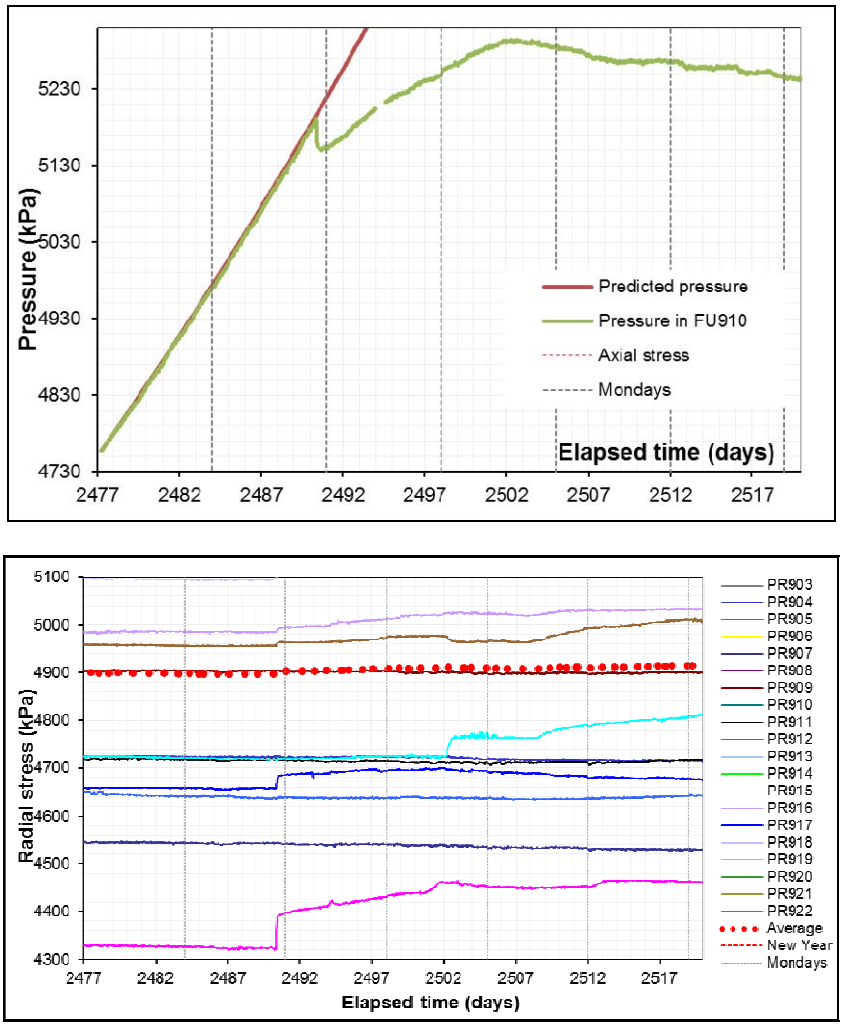


Figure 8 **Gas break-through (top) and radial stress response (below) at the deposition hole wall.**

[4] LArge Scale Gas Injection Test - a full-scale field experiment located 420 m below ground level at the Äspö laboratory. The laboratory design involves placement of copper/steel canisters containing spent nuclear fuel in large-diameter disposal boreholes drilled into the floor of the repository tunnels. Spaces around each canister are filled with pre-compacted bentonite blocks which over time, swell with water and reduce any construction gaps. Once hydrated, the bentonite will act as a low-permeability barrier to fluid flow from the waste canisters. In the LASGIT experiment, a full-scale waste canister has been modified to provide points for gas injection to simulate canister defects. The borehole, bentonite and canister are fully instrumented to monitor stress, porewater pressure and relative humidity. Additional monitoring includes temperature, relative displacement of the borehole lid and anchor restraining forces. The state-of-the-art experimental monitoring and control systems for LASGIT are housed in the 'gas laboratory', a self-contained, temperature-controlled unit.

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

The laboratory studies by BGS (British Geological Survey) [69, 70] show that under all tested conditions during the project, observations indicate that the ***primary*** mode of gas transport is by dilatant pathway formation. These observations include:

- Strong coupling between σ, Π and Pp[5] (**Figure 9**)
- Localised changes in σ, Π and Pp (**Figure 9**)
- Unstable flow, exhibiting spatio- temporal evolution (**Figure 9**)
- Localised outflows during gas breakthrough
- No measurable sample desaturation in any test samples, indicating that gas has not passed through the bulk of the bentonite

These findings are consistent with other recent studies involving argillaceous materials (Ortiz et al., 2002 [71]; Angeli, et al. 2010 [72]; Cuss et al., 2010 [73]; Skurtveit et al. 2010 [74]; Harrington et al., 2009 [75]), as well as earlier studies in bentonite (Pusch et al., 1985 [76]; Harrington and Horseman, 2003 [77]; Horseman et al., 1999 [78]).

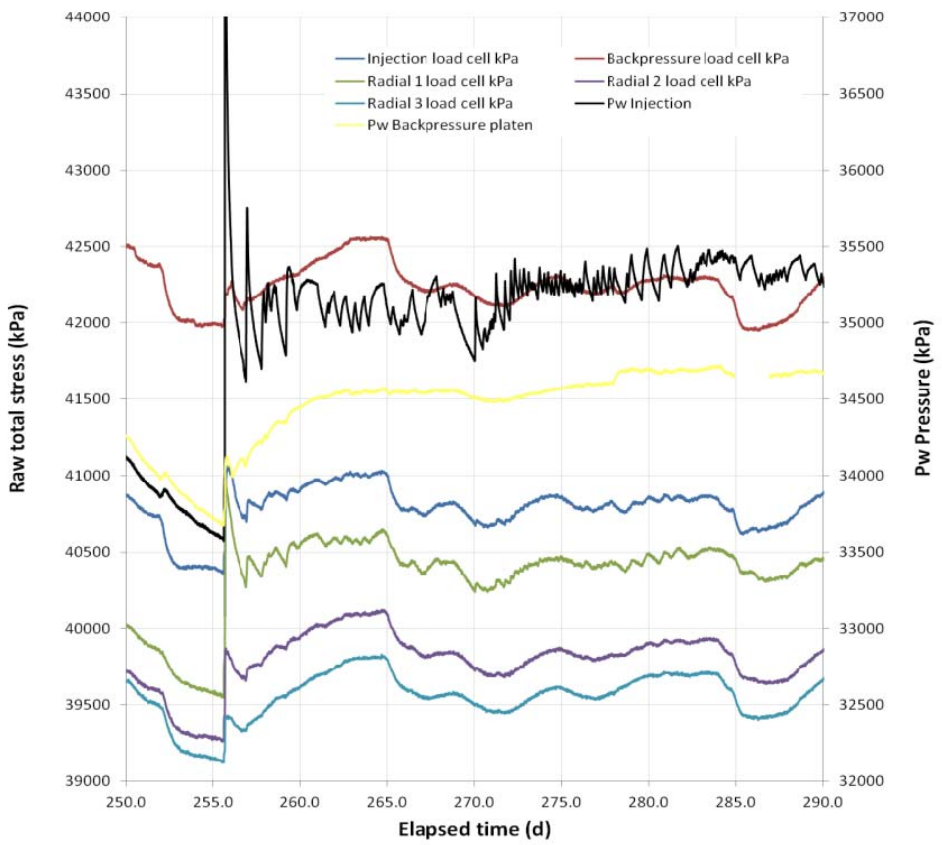


Figure 9 **Output of laboratory studies by BGS, showing that under all tested conditions during the project, observations indicate that the primary mode of gas transport is by dilatant pathway formation.**

[5] Where σ = Total stress, Π = Swelling pressure and Pp = pore pressure (indeterminate of phase)

Clay Technology has tested the gas breakthrough behaviour by lowering the total pressure of the bentonite system by flushing a strong NaCl solution on the outlet side while a constant gas pressure, below the initial pressure of the bentonite, was maintained on the inlet side. The bentonite pressure immediately started to fall after the flushing, and when it became comparable in size to the injection gas pressure a gas breakthrough event occurred (**Figure 10**). This behaviour was demonstrated in the same sample for different gas injection pressures, giving a clear-cut evidence of the osmotic nature of the system.

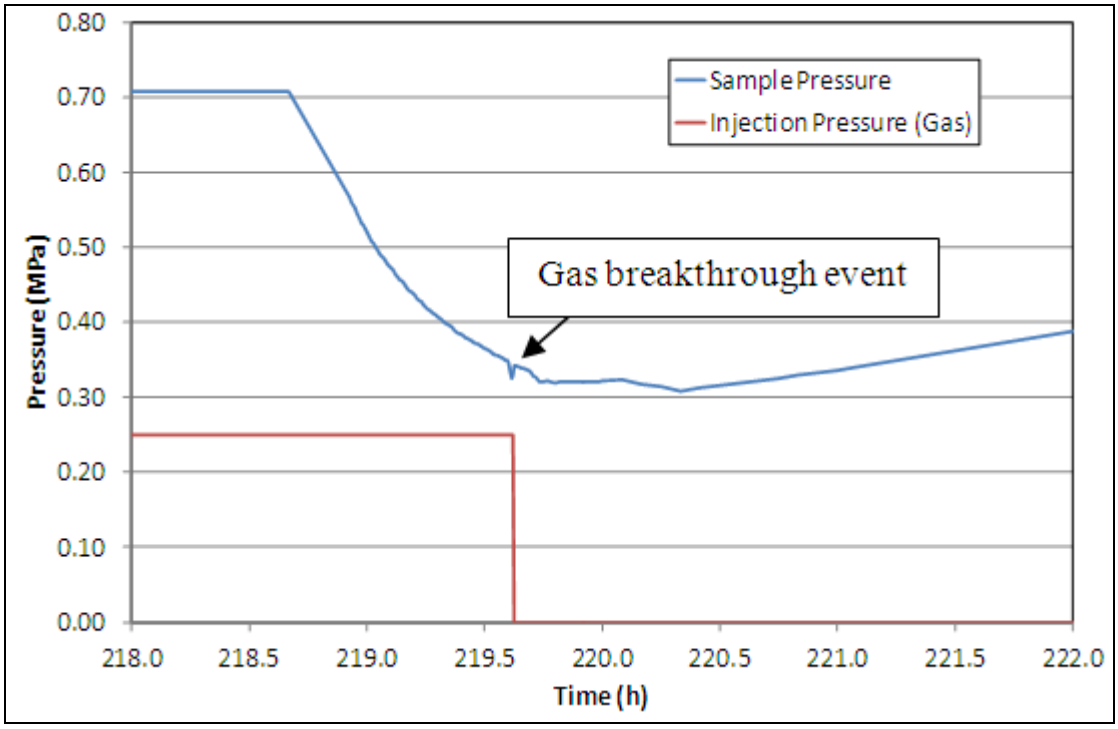


Figure 10 **Response of an MX-80 sample which is being flushed by a 3 M NaCl solution at the top filter (at ca 218.75 h). The bottom filter is pressurized with air at 0.25 MPa. A breakthrough event occurs when the sample pressure becomes similar to the gas injection pressure.**

4.3.2 Cement-based EBS

4.3.2.1 Gas diffusion

This subsection reports work led by BGS, see [79] for further details. For the experiments with transport of CO_2 in cement it should be noted that the CO_2 is not an inert migrating phase. It is highly reactive towards the cement, and so its overall migration is controlled by both reaction and transport. The net effect of this will be to slow the rate of CO_2 migration relative to a purely non-reactive diffusional case.

However, some of the experiments did reveal the positions of migrating carbonation fronts as a function of time (**Figure 11**). Diffusional transport of CO_2 played a role in the migration of the fronts (though so did reaction of the CO_2 with the cement). It should be noted that the fronts do not record the position of the leading edge of CO_2 ingress, as minor amounts of CO_2 reacted with cement in advance of the fronts. Instead, the carbonation fronts record the position where sufficient CO_2 had permeated the core to enable sufficient carbonation to occur to change the structure

of the cement (including *complete* carbonation of the reactive cement minerals). The fronts migrated by a few millimetres over several weeks.

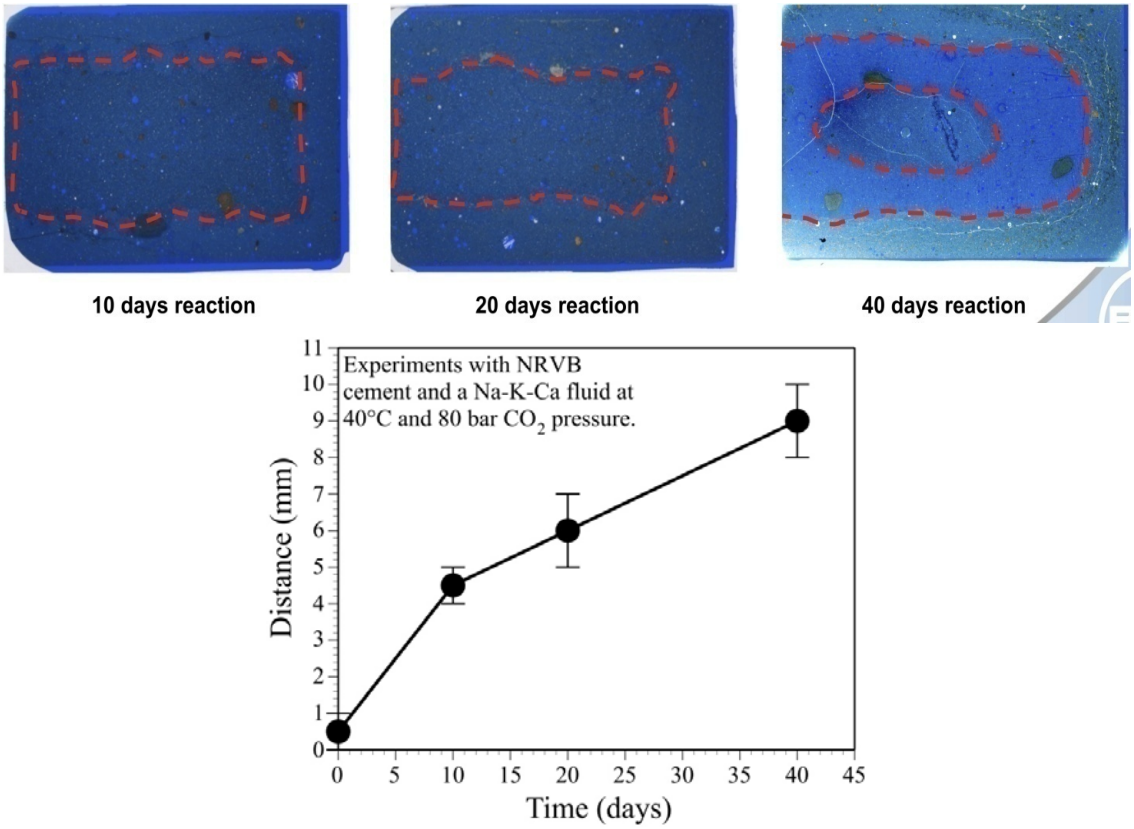


Figure 11 **Progressive migration of carbonation reaction fronts through 25mm diameter samples of NRVB cement.**

In WP3 gas reactivity has only been studied for systems with cement and CO_2. CO_2-cement chemical reaction caused two types of *localised* temporary shrinkage cracks to develop, and in particular in the unconfined samples. One set of shrinkage cracks were oriented parallel to the flow direction (i.e. at 90° to the reaction fronts) and the other set were perpendicular to it (associated with the most recent, as well as older, reaction fronts). Secondary precipitation eventually sealed these potential flow features (see **Figure 12**, a more detailed view of **Figure 11** at 40 days).

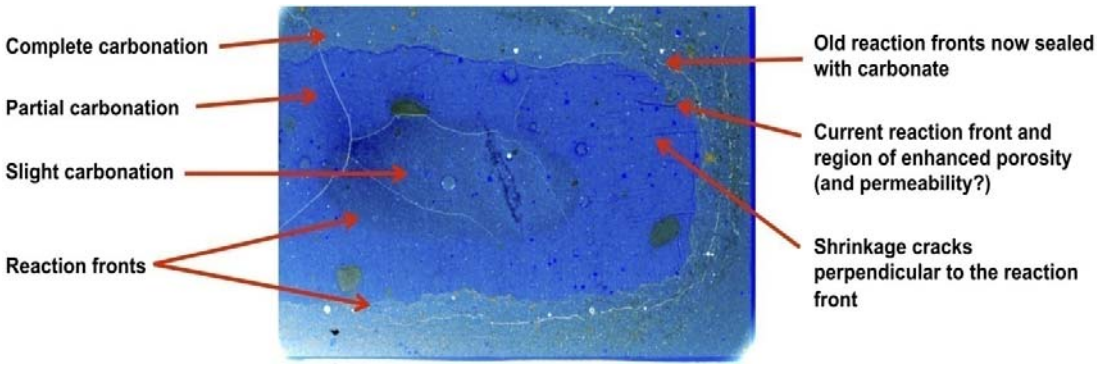


Figure 12 **Partially carbonated cement sample showing zones having different degrees of carbonation together with associated reaction fronts (Rochelle et al. BGS technical report [79]).**

4.3.3 Interfaces within the EBS

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

In general it seems like gas will select a path in the interface between a clay and a metal barrier if there is a possibility. This is confirmed in a number of experiments in FORGE. However, it still seems like the properties of the clay alone will determine the gas transport process.

A test was undertaken by Clay Technology that specifically addressed the issue of the interface pathway. The nature of the formed breakthrough pathways was studied in test cells of a type schematically pictured in **Figure 13**. Gas was injected in a small central filter in the bottom of the cell, while filters for outflow were located both at the top of the cell as well as at the bottom circumference ("guard filter"). In this way it can be concluded that any gas ending up on the top side must have propagated through the clay. However, in all cases where this has been studied (with the exception of some very thin samples) the gas was detected in the "guard filter" although the dimensions were such that this path was typically twice as long as the shortest path through the clay.

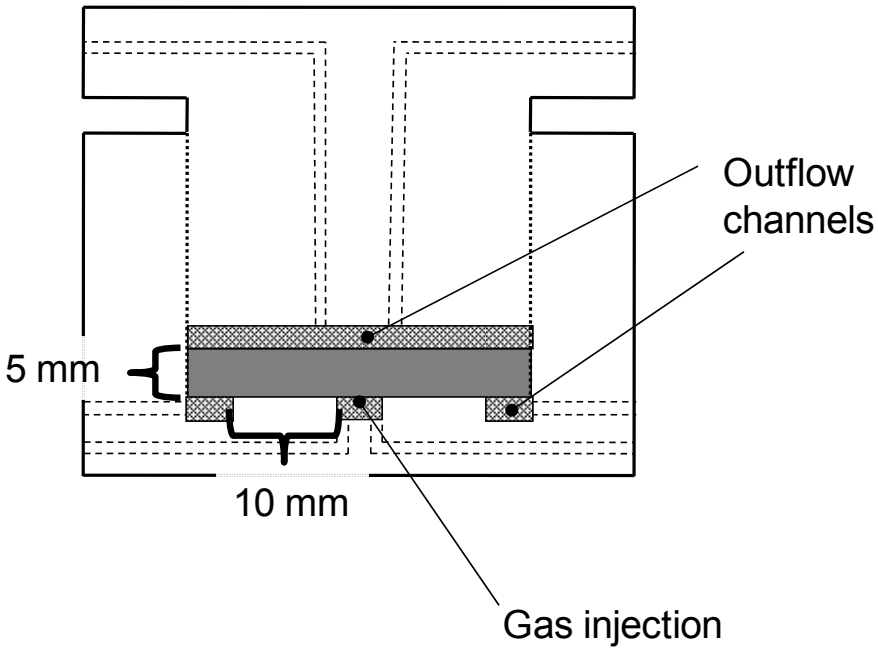


Figure 13 Schematics of the test cell used for detecting preferential path for gas between clay body and cell wall.

Consequently, these results strongly indicate that the cell/clay interface is a

preferential path for the gas in a breakthrough event.

Further relevant results from FORGE are:

- Tests at Institut für Gebirgsmechanik GmbH (IfG) with bentonite-bentonite interfaces showed that, at dry conditions, gas flow along interfaces is at least four orders higher than through the matrix.
- Increase of confinement on the contact zone significantly lowers the gas flow also in the dry buffer material but the effect is more pronounced for interfaces; crack sealing is postulated to occur.
- 'Flooding' of the EBS results in saturation of the interface (or contact zone), however our results document that regarding the contacts block/block (i.e. inside the bentonite plug composed of single bentonite blocks) the water consumption during swelling is not dependent on the injection geometry, and is only weakly dependent on the axial load. For 'flooding' of the bentonite block seal it can be concluded that between the blocks no preferred flow along the interface exists, i.e. matrix swelling dominates. In addition during the tests, one year of test time was not sufficient to reach full saturation.

4.3.4 Disturbed host rocks

4.3.4.1 Gas diffusion

The movement of gas by diffusion will occur in all porous media as soon as gas is produced. Dedicated experiments focussing on the evaluation of gas diffusion coefficients in disturbed host rock formations were beyond the scope of FORGE, which was originally designed to address key issues associated with advective gas flow. While WP4 experiments were not specifically configured to measure gas diffusion behaviour, certain observations within the test programme can be attributed to the diffusion of gas. However, such phenomena are only relevant in long-duration tests and are invariably concept and material specific. For example, the EDZ in a fractured crystalline host rock may have a stronger influence on the direction and magnitude of diffusive gas flow than in clay-based repository systems exhibiting temporal self-sealing.

Outside of FORGE, a large number of experiments defining gas diffusion coefficients have been under taken. Nagra has commissioned many laboratory experiments on Opalinus Clay samples with emphasis on the evaluation of gas diffusion processes (e.g. transient gas pressure decay experiments at RWTH Aachen, Mont Terri FM-C [80]; Mont Terri Hydrogen Transfer (HT) experiment; NEA-Claytrac Project [81]; OPA/Schlattingen). In addition, the gas diffusion properties of Boom Clay have also been extensively investigated. SCK•CEN have determined diffusion coefficients both perpendicular [82] and parallel to bedding, defining an anisotropy ratio of between 0.62 and 0.58, depending on the type of gas used.

However, all of these measurements have been performed on intact (undamaged) samples. In the absence of direct experimental data, the role of the disturbed zone could be modelled using diffusion coefficients for the intact material and 'free' water in conjunction with an appropriate description of the 'fractured' medium.

4.3.4.2 Two-phase flow

Two-phase flow conditions are encountered when gas invades a porous medium as a separate phase. This process is associated with the drainage of interstitial fluid as the non-wetting phase (gas) displaces the wetting phase (pore water). Flow is within the original inter-granular porosity resulting in an iso-volumetric boundary condition.

In reviewing the data from this section, it should be acknowledged that the majority of tests were performed on intact rock specimens, reflecting the difficulties in obtaining representative 'damaged' material and the large matrix of tests that would be required to cover all 'disturbed' scenarios.

In Opalinus Clay, gas permeability tests performed on isostatic/triaxially stressed specimens provide evidence for the displacement of water by a gas phase (NTB 02-03, [83], [84]). Indirect evidence from micro-structural analyses [85], suggesting that (sparsely) connected systems of macro- and meso-pores exist in Opalinus Clay, which can be invaded by a gas phase. These observations support the hypothesis of classical two-phase flow.

Interpretation of the data from the PGZ[6] field experiment performed by ANDRA at the Bure URL indicates gas flow is initially focussed within the excavation damaged zone, which acts as a preferential pathway for gas. Numerical interpretation of PGZ by EDF appears to confirm the hypothesis of classical two-phase flow in the undisturbed host rock. However, the question of gas flow in disturbed host rock (EDZ) is very complex due to the difficulties of characterising this zone. To adequately model such behaviour, parameterisation of the EDZ (permeability/suction/damage) would be necessary.

In contrast, experiments performed on undisturbed samples of Boom Clay by SCK•CEN have not yielded, as yet, evidence for the movement of gas by classical two-phase flow. SCK•CEN cite high gas entry pressures, no desaturation and the prevailing stress regime around a repository as arguments against classical two-phase flow.

While considerable effort has been placed on examining the fundamental controls governing the advective movement of gas, definitive interpretation of the data to identify the underlying physics controlling gas flow, currently remains elusive. In part, this may stem from differences in experimental procedures, types of material, control of boundary conditions and terminology.

4.3.4.3 Pathway dilation & gas-induced fracturing

Dilatant gas flow is taken to relate to the expansion/disruption of the inter-granular pores, where permeability is now a dependent function, related to the number of pathways in the plane normal to flow, together with the width and aperture distributions of these features. Evidence for the existence of dilatant flow stems from volume change of the sample at the onset of gas movement: this change in sample volume is in stark contrast to classical two-phase flow, which assumes an iso-volumetric boundary condition. The majority of laboratory tests have been performed on initially undisturbed samples.

Nagra has commissioned various laboratory tests aimed at exploring the regime of pathway dilation. However, as yet, clear evidence for dilatancy controlled gas flow

[6] The PGZ experiments are part of an experimental and demonstration test programme at the Meuse/Haute-Marne Underground Research Laboratory. One of the objectives of this experimental and demonstration test programme is to study the response of argillites to disturbances caused by a repository. In the PGZ experiments, the subject is the disturbances caused by gases generated by the radiolysis or anoxic corrosion of some of the waste packages and, where applicable, by the corrosion of the repository's steel components, e.g. high-level waste cells. This will particularly include gas transfer in undisturbed and disturbed argillites, as well as transfer through the sealing plugs and at interfaces.

has not been observed in these laboratory experiments [84]. Additional laboratory tests have been commissioned outside of FORGE to further investigate this phenomenon. In contrast, recent field experiments (Mont Terri HG-C/HG-D) have provided indications for pathway dilation and self-sealing during gas testing [83] Under these conditions, numerical simulations by Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH show that two-phase flow models, which need to displace water in order for gas to flow, are not able to capture important aspects of transport in the pathway dilation regime. As such, GRS have found it necessary to include dilation in existing codes in order to model experimental data.

Laboratory experiments performed on samples of COx clearly demonstrate localised mechanical dilatancy at the onset of gas flow. This behaviour cannot be explained by the mechanical response of the sample to changes in effective stress alone. The clear correlation between dilation and outflow, along with no measureable desaturation of the sample after testing, provides strong evidence that dilatancy is a key process in the movement of gas in COx. Experimental observations indicate that these pathways are temporal in nature, evolve over time, are unstable, and may heal, which can make re-initiation of flow difficult. A separate and independent study performed by LAEGO-ENSG (Laboratoire Environnement Géomécanique et Ouvrages - École Nationale Supérieure de Géologie, Nancy, France) [70] outside of FORGE observed similar behaviour with dilatancy occurring at the onset of gas flow.

Experiments undertaken by SCK•CEN on undisturbed and pre-fissured Boom Clay samples suggest dilatancy occurs during gas flow. Experiments injecting nanoparticles (Harrington et al., 2012 [86]) in a carrier gas (performed at BGS) provide direct evidence for the formation of dilatant pathways during gas flow: as with COx, dilatant pathways were very unstable after breakthrough [87]. While gas breakthrough occurred at a lower pressure in artificially fissured cores, there was no indication for gas flow along the plane of the pre-existing fracture. Experiments will continue beyond FORGE investigating these processes further, including the influence of micro-fracture orientation to bedding.

Analogue tests performed by BGS investigating the onset of gas flow in an EDZ also show the formation of localised dilatant pathways. The geometry of the features appears to vary between tests, suggesting the direction of gas flow in clay-filled (kaolinite) fractures is difficult to spatially predict. In loading and unloading cycles fracture transmissivity exhibits a high degree of hysteresis, which suggests the complex stress history observed in the EDZ during the full life cycle of a repository is of fundamental importance.

While there is clear evidence to support pathway dilation, a definitive description of flow processes has yet to be realised due to the paucity of available data.

Gas fracturing is associated with the rapid build-up of gas pressures significantly above the minimum principal stress. In reality, the likelihood of gas pressure build-up to such high values is unlikely as the gas moves by diffusion and advection. Other arguments can also be used here:

- the rates of pressure increase in gas tests are usually much higher than what is expected in situ as resulting from a very slow corrosion reaction;
- the potential presence of local heterogeneities may locally facilitate gas entry. The probability of encountering such heterogeneities is scale dependent.

Frack tests have been performed by Nagra (HG-B experiment) though the focus of these was on the determination of *in situ* stress fields.

4.3.5 EBS - host rock interfaces

Considerable care and attention is taken by all experimentalists, either in the laboratory or field, to minimise/negate experimental interfaces. This includes elements in the design of experiments, methodologies, sample preparation, interpretation of data etc. Unless stated to the contrary, tests are designed to reflect realistic in situ conditions, with separate detailed bespoke tests specifically designed to interrogate the interfaces in water and gas flow.

Laboratory and in-situ experiments in clay-based systems support the hypothesis that interfaces within a repository are regions of stress redistribution and fabric disruption. Interfaces are likely to represent favourable pathways for gas flow when there is either a continuous pathway/plane of weakness along the interface or when the contact pressure (created by swelling and convergence) is less than the minimal principal stress. The HG-A experiment at Mont Terri is a typical example for the inter-dependence of the leakage rate and radial stress distribution around a sealed tunnel. However, such interfaces are not stable with time as also demonstrated in laboratory experiments performed on Boom Clay. In analogue tests (BGS), gas pathways along clay-filled interfaces continue to evolve after initial breakthrough with decreasing peak pressure with subsequent breakthrough events. A multitude of mechanisms may affect the gas transport capacity of an interface including stress and pore pressure conditions, stress history, orientation of the interface to the stress field, strains and hydro-chemical porewater-rock interactions (e.g. swelling, precipitation, filtration, erosion). Dedicated experiments have been conducted in the past to address the issue of interface opening/closure (within [88]).

The importance of interfaces is also recognised in crystalline host rock disposal concepts with experiments performed by CTU (Centre of Experimental Geotechnics, Czech Republic) clearly demonstrating an increase in permeability of the EDZ in contrast to intact rock. The evolution of these features is likely to evolve temporally and spatially, though no direct observations were recorded as part of FORGE.

4.3.6 Compilation of information from outside of FORGE: Gas migration processes in porous media

A conceptual model of gas migration processes is developed based on experimental data; mathematical / numerical studies are undertaken based on such a conceptual model.

Only relevant work undertaken in parallel work to FORGE is considered here; references were taken into account only from 2009 to 2012. The research was made using exclusively the internet sites of BRGM (Bureau de Recherches Géologiques et Minières, France), Science Online (Scientific publications), One Petro (petroleum and CO_2 sequestration industries references) and ANDRA's International Department concerning the nuclear waste references. All the cited references are public domain ones. A total of 44 references are taken into account, from which 6 are from BRGM, 19 from Science Online, 16 from One Petro and 3 from ANDRA's International Department.

The major fields where works on gas migration processes are developed are numerical research studies undertaken by a range of universities (16 references), petroleum field (14 references), CO_2 sequestration (10 references) and to a lesser extent (excluding FORGE work) nuclear waste management (4 references). The porous media investigated are therefore mainly fractured reservoirs and clay rocks (cap-rocks). Some work was also done with highly saline brine; the most interesting geological layer for the gas/petroleum/CO_2 industry is several kilometres below the ground surface.

Globally, the processes developed/confirmed can be gathered into 5 major groups:

- Diffusion (mainly of dissolved gas) (1 reference);
- Two-phase flow, including Darcy and non-Darcy behaviour (non linearity, Knudsen effect etc). These topics include a large amount of numerical work trying to improve the numerical behaviour of the available codes (considering phase change numerical complexity, upscaling techniques) (21 references);
- Hydraulic coupling with other processes, especially mechanic (pathway dilation, fracturing) and/or chemistry (sorption of gas on solids, chemical interaction of gas with solids and or other solutes) (10 references);
- Pore scale processes (simulation of molecular of nanoscale processes not assuming any macro-processes and trying to define them) (10 references).
- Safety approach in an industrial point of view concerning gas related topics (2 references).

4.3.6.1 Gas diffusion

From all the processes actually considered for gas migration, the diffusion of the solutes is the most well understood in low permeability porous media and the work done on this topic is not very significant compared to the work done on other topics. Nevertheless, in the CO_2 sequestration industry this field is still active trying to reduce the uncertainty on the parameters controlling this process [89].

4.3.6.2 Two phase flow

(a) Darcy formulation

The work done concerning strictly the Darcy approximation is mainly linked to code numerical problems related to the phase change and new numerical methods to handle this issue [90, 91, 92, 93, 94]. These are mainly based on numerical formulations with continuous primary variables whatever the phase state, like “negative saturation”, use of mass fraction, use of variable changes, but some work is also done on new numerical discretization scheme [95].

Some complementary work is done on the hysteresis processes, especially in the CO_2 sequestration industry as a significant process to capture CO_2 [96, 97].

(b) Non Darcy formulations

In these papers the general framework of the Darcy flow is supposed to be globally consistent, but other processes are added (Klinkenberg and/or Knudsen effects) or a non linearity between flow and water head gradient is taken into account for low head gradients [98, 99, 100, 101].

(c) Multi-porosity model

A significant amount of work is aiming at different representations of multi porosity media. Most of these papers are referring to double porosity models, trying to include the effect of natural fractures in the flow [102, 103, 104, 105]. Some are referring to “Triple porosity” models, introducing in addition to the matrix and the fractures small features (micro-fractures) making the link between the intact rock and the inside of the macroscopically opened fractures [106, 107]. In the petroleum industry, specific problems related to the fact that organic matter is filling the pores and can contain gas, lead to introducing a porosity linked to this organic matter [108, 109].

(d) Upscaling

Some work on mathematical upscaling (homogenization of the main hydraulic parameters, including shape of the retention and relative permeability curves) is developed in the framework of the CO_2 sequestration industry [110]. A more complete description about upscaling, and linked bibliography, can be found in section 4.4.

4.3.6.3 Coupling effects

Most of the codes are already taking into account thermal coupling and code developments are mainly focused on mechanical and chemical coupling

(a) Mechanical coupling

Mechanical coupling is investigated by petroleum industry, CO_2 sequestration industry and nuclear waste disposal industry. The main work is about real coupling integrating complete mechanical behaviour into the hydraulic transient process. Some of the works are assuming continuous porous media and mechanical damage is determined through the use of “damage coefficients” [111, 112], but some papers are assuming predetermined fractures [113] and some others are using “explicit fracture initiating” formulations [114, 115].

In the context of nuclear waste disposal, mechanical coupling papers are mainly focused on bentonite-gas related behaviour [116, 117, 118].

(b) Chemical coupling

Chemical coupling is mainly considered in the CO_2 sequestration field and the developed chemical equations are complex and aside of the FORGE work [119, 120].

4.3.6.4 Pore scale simulations

This approach is mainly used to understand more thoroughly the behaviour or gas-water(-oil) at the pore scale and to analyze the deviation from generalized Darcy equations. One explanation provided by these “nanoscale” simulations is that slippage effects due to fluid-matrix interactions can be very significant [121, 122, 123]. Use of “power-law fluids” is also an explanation for oil and gas migration in low permeability petroleum fields [124]. Other papers are introducing a mechanical coupling at pore scale as an explanation to these deviations [125]. But some papers are concluding that a (linear) relationship exists between “macroscopic dissipation rate” and “pressure drop through the REV” [126]. Other tools used in pore scale simulation are dynamic time dependent interface-tracking trying to improve the simulation of the water-gas-(oil) interface at pore scale [127, 128]. The nanoscale simulation is also used to determine macroscale relative permeability curves based on microscale pore understanding [129, 130].

4.3.6.5 Safety approach

For regulatory reasons, the CO_2 industry tries to define safety procedures including topics linked explicitly to the gaseous form of the CO_2 [131, 132].

4.3.6.6 Summary of information from outside of FORGE: gas migration processes in porous media

The references found from the public domain published articles between 2009 and 2012 are showing that the main work on the gas migration processes in porous media are developed in the gas and petroleum industry as well as in the universities.

The CO_2 sequestration industry is also providing a large amount of work on this topic but mostly focused on specific chemical interaction between dissolved gas, other solutes and/or solid matrix, which is a topic aside of the FORGE main goal.

As a general conclusion, the work done outside FORGE has showed no important research topic which is not (at least partly) developed inside the project. However:

- Pore scale simulations could help a more thorough understanding of the gas/water/solid interactions at very small scale and could provide a better understanding of the macro-scale behaviour;
- Promoting relations with the gas and petroleum industry could help broaden the radioactive waste community view on the subject, and could help in finding practical ways to incorporate new processes in software codes, especially a simple representation of mechanical coupling.

For granite sites it could be interesting (if not already done) to test the impact of the "triple-porosity" model in the way to represent this naturally fractured media.

4.3.7 Compilation of information from outside of FORGE: Experimental studies on EDZ

Most of the papers on experimental results are concerned with the EDZ characterisation with respect to their HM properties [133, 134, 135]. In [133] and [134] the samples of the Callovo-Oxfordian argillite and the Opalinus clay were studied. The samples were artificially cracked by tensile strength tests. The cracked samples were put into a triaxial cell and the permeability was measured during stepwise increase of confining pressure (up to 5MPa and 15 MPa respectively). The corresponding gas (nitrogen) effective permeability was in the range $3.2\ 10^{-20}$ - $1.9\ 10^{-16}$ m^2 for Opalinus and 10^{-16} - $1.9\ 10^{-13}$ m^2 for COx. In both studies the gas effective permeability decreased during loading of about 2 and 4 orders of magnitude respectively, and was partly reversible with unloading. Under constant loading the gas permeability decreased over time (over several days) indicating a ''creep'' behaviour of clays resulting in a slight closure of cracks. The long term (up to 1.5 year) tests of sealing were done in [134] at moderate confining pressures (2.25-3 MPa). The measured absolute water permeability deceased rapidly (up to seven orders) and continued to decrease with time to levels corresponding to the intact rock.

Some other conclusions drawn from these works include relatively low impact of heating on water permeability. In [136] the experimental results were reinterpreted in terms of fractal theory to obtain a relationship between porosity and permeability. For these cracked Opalinus clay samples, the fractal dimension D=2.80 obtained was similar to previous determinations of D for fractured rocks.

In [135] the possibility of reactivating a pre-existing shear band by thermal pore water pressurisation is demonstrated in saturated Boom clay samples. However, it is shown that the presence of a shear band does not affect significantly the water permeability, except the effect it may have on porosity. A slight decrease of permeability is observed (from 3.10^{-19} m^2 to 2.10^{-19} m^2) after shearing test, which can be attributed to the sample compaction (decreasing of the porosity of 0.238%) during the drained triaxial shear test. The permeability test at 80 °C on the sheared sample shows a slight increase of the permeability (up to $2.8\ 10^{-19}$ m^2) which is reversible upon cooling.

Similar work was also conducted on other materials, like coal and sandstones [137] and concretes [138]. In [138] the concrete specimens were sequentially loaded

(Brazilian test) and tested in a gas permeameter after several unloading cycles. The water flow percolating through most cracked specimens gradually reduces with time, due to water–cement matrix interaction. If only the flow produced during the first part of the water permeability test is considered, then water and gas permeability measurements tend towards the same value. Results suggest that permeability increases proportionally to the cube of the fracture aperture, as shown theoretically for viscous flow in rough fractures. In damaged specimens, the cracks convey the larger part of flow, with no influence from the saturation degree for water saturations from 0.01 to 0.35.

In [137] fractured samples of sandstone and are tested and the relationship between their gas permeability and confining pressure is obtained. The results show that fracture permeability strongly depends on confining pressure for both types of rocks and it is found that for all specimens, the gas permeability increases as the confining pressure decreases (3-5 times for coal samples, 7-11 times for sandstone). In order to investigate the impact of a load cycle on permeability, one of the coal samples was reloaded 1–3MPa confining pressure after unloading of 3–1MPa. During reloading, gas permeability declines nearly in a straight line in contrast to the unloading stage where the relationship between confining pressure and permeability is a power law. The authors suggest that the loading–unloading–reloading course generates abundant and extensive distributed cracks in the samples, so that cracks can gradually get interconnected. Further analysis of the data obtained in this work is presented in [139]. It shows that an universal relationship can be proposed between the unloading rate (Pr=(P-P0)/P0) and the “rate of increase” of permeability (kr=(k-k0)/k0) which fits well the data presented [137] kr = A exp(b Pr).

4.3.7.1 Modelling of single fracture or interface

There are several papers proposing microscale models to interpret laboratory results on fractured samples. The paper [140] intends to investigate the influence of calcium leaching on the mechanical behaviour of rock–mortar interfaces by means of numerical simulations. The materials considered in these simulations are a mortar and granite. The leaching of the interface walls leads to a local increase of porosity in the bulk material and an augmentation of the interface transmissivity could logically be expected. On the contrary, in [141] it is observed that the leached interface would not conduct water upon shearing before physical breakage of the interface. The results presented show that passed a certain transitional leaching depth, the interface contracts upon shearing. As a result, the mechanical and hydraulic aperture also decrease and the overall transmissivity of the interface will decrease. The experimental results after [141] are thus corroborated by these mechanistic simulations.

Another paper [142] deals with tortuosity effects in coupled transport and mechanical properties of fractured geomaterials. Experimental results on confined fractured argillite samples [143] are presented in terms of progressive fracture reclosure and permeability evolution, both depending on confining pressure. The observed non-linear mechanical response (fracture reclosure law) is physically interpreted as the progressive reclosure of local pores. The weak correlation between mechanical and hydraulic measurements is attributed to tortuosity effects in the morphological evolution of the fracture interfacial domain which enhance the initial decrease of the permeability. The derived micromechanics analysis is able to propose estimates for the longitudinal permeability coefficient on the basis of fracture reclosure tests [144].

4.3.7.2 Modelling of the EDZ

The article [145] describes the development of a structured approach to abstract complex geoscientific models of two-phase flow through the EDZ to simplified models suitable for use within a Probabilistic Safety Assessment (PSA). The modelling presented aims at developing of a structured methodology for treatment of gas, solute, and water flow through the EDZ. The approach utilizes three different models: a discrete fracture network (DFN) model of the EDZ, an equivalent heterogeneous continuum porous medium (CPM) model and a simplified CPM model suitable for use within PSA. Equivalent properties of the elements of the heterogeneous CPM models are upscaled from DFN realizations. Results from gas injection simulations with the heterogeneous CPM models are then used to derive appropriate parameters for the simplified CPM model.

4.3.7.3 Modelling of in-situ experiments / set ups

The paper [146] presents an international, multiple code, simulation study of coupled thermal, hydrological, and mechanical (THM) processes and their effect on permeability and fluid flow in fractured rock around heated underground nuclear waste emplacement drifts. Simulations were conducted considering two types of repository settings (1) open emplacement drifts in relatively shallow unsaturated volcanic rock, and (2) backfilled emplacement drifts in deeper saturated crystalline rock. The study was part of the DECOVALEX-THMC project (2004–2007). The analysis conducted within this project shows that the main stress response in both repository settings is an increase in horizontal thermal stress that has two possible consequences on the permeability field. On one hand, an increased horizontal stress tends to reduce the aperture and permeability in steeply dipping fractures. On the other hand, an increased horizontal stress also leads to increase the shear stresses that could induce shear failure and dilation of shallowly dipping fractures. For the assumed properties and repository settings considered in this study, shear induced permeability changes were ruled out. This led to a system in which permeability changes during the heating period were controlled by a reduction in vertical permeability, caused by horizontal thermal stress. This reduction in permeability critically depends on (1) an accurate prediction of regional thermal–mechanical stress evolution, and (2) a correctly bounded estimate of the relationship between stress and permeability for fractured rock. However, even for the most sensitive relationships between stress and permeability, the impact of mechanically induced changes in permeability on the flow field was relatively small, with most changes occurring in the vicinity of the emplacement drifts.

The next paper [147] is concerned with the hydro-mechanical responses of unsaturated heterogeneous structures of fissured argillaceous formations to tunnelling. The study started from the case-history of a deep tunnel excavated in a highly tectonised clay formation (Raticosa tunnel, Italy). A wide range of conceptual models for the short-term response were compared, ranging from the conventional undrained behaviour to specifically formulated models for stiff fissured clays, for which the pore pressure reduction during unloading stress paths may be particularly strong as a consequence of shearing and opening of fissures. For the long-term situation, two idealised cases were considered, depending on the permeability of the plastic annulus around the tunnel, which can be assumed either equal to that of the undisturbed rock or comparatively infinite, again as a consequence of fissure opening. It was found that the different models predict significantly different tunnel wall convergences in the situation before lining installation, but have only a modest influence on the support loads. Even the simplest model of a fully dry-medium can provide a fair estimate of the short (and long-term) load on the lining.

There are also several modelling papers of in-situ experiments dedicated to sealing capabilities and/or gas flow in multi-material systems. In most cases [148, 149, 150] the material interfaces are not represented explicitly, however the differences between the measurements and model predictions indicate that the flow though interfaces is a key ingredient of the system [151].

4.4 Upscaling

4.4.1 Upscaling of experimental results and understanding

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

Rigorous upscaling approaches for immiscible flow have been proposed in various geoscientific disciplines ([152], [153], [154], [155], [156], [157]). All approaches are based on stochastic assessments of the spatial variability of an appropriate porosity/connectivity indicator. These approaches provide a solid theoretical framework, defining the model assumptions and the range of validity of the presented numerical/analytical solutions.

For a given gas transport problem, the following methodology may be followed to provide the basis for process and parameterisation upscaling: (i) analysis of the relevant structural elements of the rock fabric along the composite gas flow path (identification of the scales of heterogeneity); (ii) characterisation of the multi-phase flow properties of the relevant structural elements of the composite flow path; (iii) perform, if possible, gas transport experiments on the scale of the composite gas flow path (e.g. laboratory tests, mock up experiments, *in-situ* tests).

However, in practice this may not be practical and system components may need to be reduced to individual elements and a phenomenological approach adopted[7]. Only with a full knowledge of the stress and gas flow boundary conditions (including temporal and spatial evolution/influence) will it be possible to understand and predict the real behaviour of gas at a repository scale. Either way, validation of model predictions to repository scales can be achieved using the following steps: (i) blind predictions based on lab tests; (ii) calibrated predictions from field data; (iii) final predictions and evaluation of results (based on all appropriate data).

This approach, once complete, can be scaled to natural flow boundary conditions based on a detailed characterisation and knowledge of the initial and boundary conditions at the appropriate repository scale. However, the validity of any theoretical framework has to be demonstrated explicitly (e.g. assumption of weak disorder, separation of scales, etc.).

[7]Even for large-scale *in situ* experiments it is difficult to characterise, for example, local stress states due to on-going hydromechanical transients, which complicates interpretation of results.

4.4.2 Numerical upscaling

Experiments often show that “reality” is too complex to be fully captured by the available models. Upscaling in geological systems often requires ‘homogenisation’ which needs to be validated to have confidence in the output of the model. However, it is also important to recognise that models may not need to capture all observed behaviour, but only those directly relevant to the safety of a repository.

4.4.3 Time upscaling

Assuming the approach outlined above has been followed, simulations should be reliable for a wide range of gas source terms, with uncertainly analysis performed to provide additional confidence where required.

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

4.4.4 Compilation of information from outside FORGE

The concept of upscaling is based on the fact that experimental data are collected at a scale different from (usually smaller than) the one used to discretize the aquifer in the numerical model. By means of parameter upscaling a grid of parameter values defined at the scale of the measurements is transformed into a coarser grid of block of parameter values amenable for input to a numerical flow simulator. The need for upscaling stems from the disparity between the scales at which measurements are taken and the scale at which aquifers are discretized for the numerical solution of flow and transport. Various scales are used in different fields for example, the typical length scales considered in petroleum engineering (over hundreds of metres to kilometres) differ significantly from those in environmental engineering (from meters to tens of meters).Different forces are likely to dominate on different scales. While on smaller scales capillary effects are more pronounced, the gravity effects and the viscous effects become more important at larger scales. The dominating forces need to be determined prior to choosing a particular upscaling method. In this section approaches and statements about upscaling in porous media for homogeneous as well as for heterogeneous are presented based on a large bibliography.

In [4] unresolved issues for the modeling of two-phase flow in porous media are identified. For example for fractured rock the determination of what is the appropriate scale on which to represent processes (e.g. what might be the appropriate size of a representative elementary volume for a fracture network). It is indicated also the problem of how to deal with viscous and capillary instabilities what again is partly a scale dependent issue. In opinion of this author another unresolved issue is the problem of providing macroscopic conceptual models that will bridge the gap between microscopic understanding and the prediction of large scale behavior, in particular when it is necessary to represent, on a macroscopic scale, processes that are controlled by smaller scale features or that sample only a small part of the pore space. Another unresolved issue is the determination of suitable two-phase flow parameters, notably the saturation functions, on an appropriate scale.

In [5] it is affirmed that the continuum approach can become inadequate when the scale on which a process can be satisfactorily averaged (using the conventional

framework) is smaller than the scale on which a model is discretized in a numerical model (and there is always a practical lower limit to this scale for simulations on a domain of a given overall size). In opinion of the author this may occur in water infiltration into unsaturated rock or the development of gas pathways through saturated rock. It does not necessarily follow that a satisfactory macro-scale model cannot be developed to describe such processes, but this will be a different (possibly phenomenologically based) model from the conventional two-phase flow model. As an alternative to the conventional two-phase models, a number of discrete pathway models have been proposed in attempts to model gas migration through water saturated clay buffers or host rock materials. In these models the movement of gas and water through a porous medium is followed by tracking the movement of gas-water interfaces (conceptually similar models have been used to track gas invasion of a fracture network). The movement of interfaces may be tracked through stochastic or discrete representations of the pathways. At present these models should be regarded as tools for interpreting gas migration experiments and for elucidating gas migration mechanisms. The development of suitable macroscale models from such models, when they have been suitably tested, is an issue for future work.

In reference [5] is reported that confidence in the use of models for performance assessment needs, however, to be demonstrated on scales more typical of the repository system than can be generated in the laboratory. This means that steps need to be taken to obtain data on a larger scale from field experiments, either undertaken from the surface or from underground laboratories, as discussed for most rock types in the report. Unfortunately, the larger the scale of the system the more difficult it is to properly characterize it, so that when it comes to modeling the experiment there is substantial uncertainty in the parameter values and boundary conditions that should be applied. In many cases, the difficulty of gas migration experiments in the field also means that there are problems in obtaining good data.

In opinion of the author of [158] the upscaling of gas migration models from the typical repository scale to the site scale do not appear to have improved. Upscaling representations of gas migration remains a significant problem when modeling of gas migration. The insignificant advance in this area most probably reflects the inherent difficulty of addressing this issue. Nirex used two codes GWNet and FRAC specifically to model laboratory-scale experiments of flow in single fractures or simple fracture networks to gain a deeper understanding of the often complex interactions seen at this scale. They were relevant therefore to gas flow in the host rock. Other codes like techSIM and TOUGH2 used an equivalent continuum representation of two phase flow and are therefore more appropriate to gas migration in the overlying sedimentary rocks. They were also used to model gas migration in the fractured host rock in performance assessment calculations. This requires the derivation of relevant upscaled values for two phase flow.

For low permeability fracture rocks it is unclear how well upscaled properties represent the fine details of gas migration in fracture networks. Of particular concern is the extent to which upscaling accurately reproduces the actual contact area between the gaseous and aqueous phases. The larger this contact area, the greater will be the degree of gas dissolution. To explore the significance of this upscaling, the modellers changed the Henry's law constant which, as noted above, let to gas breakthrough at the surface. This demonstrated the sensitivity of the model to assumptions and the method of deriving upscaled continuum parameters for such fracture networks.

The reworking of the Nirex 97 gas calculations using a more sophisticated two phase flow model based on TOUGH2 [159] provided a starting point for Nirex to update its gas migration modelling capability. It confirmed the site specific nature of gas migration but also revealed the sensitivity of the results to assumptions about upscaling two phase flow in fracture networks to an equivalent continuum.

In [159] an assessment was carried out with TOUGH2 that confirmed that the detailed characteristics of the geology could exert a significant effect on gas migration. It was also demonstrated that whether or not gas reached the surface depended on assumptions used to upscale two-phase flow in fracture networks so as to parameterize equivalent porous medium models.

Reference [5] identifies uncertainties about gas migration through the geosphere, particularly regarding the appropriate scale at which to represent processes and models that upscale microscopic processes to predict large-scale behavior (in particular determining whether the models are appropriate)

The study of [159] and [160] confirmed the sensitivity of gas migration to fine details of the overlying geology and the difficulty of capturing the fine details of two phase flow in fracture networks in upscaled equivalent continuum models.

In [161] it is reported that while it was believed that the physics of two-phase flow in fractured rock is satisfactorily understood, modelling this at the field scale presents problems because of the difficulty of obtaining consistent hydrogeological parameters and constitutive relationships at the appropriate scale. The applicability of conventional continuum models on a scale much larger than the intrinsic scale of the flow phenomena occurring is also questionable. Work is needed to address these issues, in conjunction with the consideration of gas-water interactions. The “upscaling” required could be supported by field experiments designed to test the aggregated response of a rock mass or, in principle, by natural analogue data.

References that present the state-of-the-art in upscaling of two phase flow in fractured rock are [162] and [163] (note the respective dates when these studies were undertaken); the latter makes a review of recent progress on the upscaling of geocellular models for reservoir flow simulation. A report of the PAMINA project [164] describes also the state of the art on upscaling techniques. In this report different techniques are presented for the upscaling of flow parameters and of transport parameters under a Fickian and also a non-Fickian approach.

In [165] is presented a review of the state of the art of upscaling multiphase flow in porous media. Current description of macroscopic multiphase flow behaviour is based on an empirical extension of Darcy’s law supplemented with capillary pressure-saturation-relative permeability relationships. However, these empirical models are not always sufficient to account fully for the physics of the flow, especially at scales larger than laboratory and in heterogeneous porous media. An improved description of the physical processes and mathematical modelling of multiphase flow in porous media at various scales was the scope of a workshop held at the Delft University of Technology in 2003 some of which results are presented in [165].

The paper [166] presents the simplest form of a pore-scale model, namely a bundle of tubes model. Despite their extremely simple nature, these models are able to mimic the major features of a porous medium. In fact, due to their simple construction, it is possible to reveal subscale mechanisms that are often obscured in

more complex models. They use their model to demonstrate the pore-scale process that underlies dynamic capillary pressure effects.

The authors of the paper [167] employ static pore-scale network models to obtain hydraulic properties relevant to single, two- and three-phase flow for a variety of rocks. The pore space is represented by a topologically disordered lattice of pores connected by throats that have angular cross sections. They consider single-phase flow of non-Newtonian as well as Newtonian fluids. They show that it is possible to use easily acquired data to estimate difficult-to-measure properties and to predict trends in data for different rock types or displacement sequences.

The choice of the geometry of the pore space in a pore-scale network model is very critical to the outcome of the model. In the paper [168] a procedure is developed for the reconstruction of the porous structure and the study of transport properties of the porous medium. The disordered structure of porous media, such as random sphere packing, Vycor glass, and North Sea chalk, is represented by three-dimensional binary images. Transport properties such as Knudsen diffusivity, molecular diffusivity, and permeability are determined through computational experiments.

The pore-scale network model of [168] is employed in [169] to study drying processes in porous media. These include mass transfer by advection and diffusion in the gas phase, viscous flow in the liquid and gas phases, and capillary effects. Effects of films on the drying rates and phase distribution patterns are studied and it is shown that film flow is a major transport mechanism in the drying of porous materials.

Reference [170] also starts with a pore-scale description of two phase flow, based on Washburn equation for flow in a tube. Subsequently, through a conceptual upscaling of the pore-scale equation, they develop anew continuum description of two-phase. In this formulation, in addition to the two fluid phases, a third continuum, representing the meniscus and called the M-continuum, is introduced. The properties of the M-continuum and its governing equations are obtained from the pore-scale description. The new model is analyzed for the case of one-dimensional flow.

A procedure for upscaling dynamic two-phase flow in porous media is discussed in [171]. Starting with the Darcian description of two-phase flow in a (heterogeneous) porous medium, they perform fine-scale simulations and obtain macro-scale effective properties through averaging of numerical results. They focus on the study of an extended capillary pressure-saturation relationship that accounts for dynamic effects. They determine the value of the dynamic capillary pressure coefficient at various scales. They investigate the influence of averaging domain size, boundary conditions, and soil parameters on the dynamic coefficient.

The dynamic capillary pressure effect is also the focus of the paper [172]. They analyze a few alternative formulations of unsaturated flow that account for dynamic capillary pressure. Each of the alternative models is analyzed for flow characteristics under gravity-dominated conditions by using a travelling wave transformation for the model equations. It is shown that finger flow that has been observed during infiltration of water into a (partially) dry zone cannot be modelled by the classical Richard's equation. The introduction of dynamic effects, however, may result in unstable finger flow under certain conditions.

Non-equilibrium (dynamic) effects are also investigated in the paper [173].They study counter-current imbibition, where the flow of a strongly wetting phase causes

spontaneous flow of the non-wetting phase in the opposite direction. They employ an approximate analytical approach to derive an expression for a saturation profile for the case of non-negligible viscosity of the non-wetting phase. Their approach is particularly applicable to water-flooding of hydrocarbon reservoirs, or the displacement of non-aqueous phase liquid (NAPL) by water.

In the paper [174], a multistage upscaling approach is pursued. They recognize the fact that reservoirs are composed of a variety of rock types with heterogeneities at a number of distinct length scales. Thus, in order to upscale the effects of these heterogeneities, one may require a series of stages of upscaling, to go from small-scales (mm or cm) to field scale. They focus on the effects of steady-state upscaling for viscosity-dominated (water) flooding operations.

Reference [175] presents a dynamic pore-scale network model of two-phase flow, consisting of a three-dimensional network of tubes (pore throats) and spheres (pore bodies). The flow of two immiscible phases and displacement of fluid–fluid interface in the network is determined as a function of time using the Poiseuille flow equation. They employ their model to study dynamic effects in capillary pressure saturation relationships and determine the value of the dynamic capillary pressure coefficient. As expected, they find a value that is one to two orders of magnitude larger than the value determined by [166] for a much simpler network model.

In [176] it is remarked that in spite of increasing computational power, the simulation of multi-phase flow in porous media is still restricted to comparably coarse grids, prohibiting the resolution of small-scale features. In the simulation of multi-phase flow on larger scales, it is therefore necessary to parameterize the effects of small-scale heterogeneities on the large-scale flow behavior. A variety of techniques have been developed and applied to transfer the information from the process scale to the simulation scale. These techniques are commonly referred to as upscaling. The upscaling techniques are classified [176] into the following categories:

- A-posteriori methods (effective parameters are derived from the analysis of highly resolved computation or measurement) [e.g. [177]; [178]; [179]; [180]];
- Stochastic methods (determination of the effective parameters through assumptions of the statistical distribution of the heterogeneities and a stochastical averaging of the equations) e.g. [181], [182] and [183]; [184]; [185]; [186];
- Analytical methods (computation of the effective parameters for simple configurations, volume averaging) e.g.[187]; [188]; [189]; [190];
- Analogy methods (transformation of upscaling-approaches from other scopes of research to multi-phase flow) e.g. [191]; [192];[193];
- Equilibrium methods (simplification of the systems by assuming an equilibrium of forces) e.g. [194]; [195]; [196]; [197]; [198]; [199]; [200]; [201]. The focus here lies on the reduction of variables by assuming an equilibrium of (capillary) forces. This assumption allows for the application of a percolation model which, together with an appropriate averaging method, results in effective constitutive relationships for the macroscale.

In [176] a system based on equilibrium is presented an approach to handle the upscaling of a two-phase flow model in an heterogeneous porous medium. It is assumed that capillary effects dominate the processes on the small scale and the changes of variables on the large scale are very slow compared to changes of

variables on the small scale. And that means that from a large-scale point of view the small-scale reaction on a change of large-scale variables is quasi instantaneously. The fluid distribution on the small-scale model is given from the local P_c-S relations that are presented by Brooks-Corey type functions. A percolation model is used to obtain the macroscopic P_c-S relationship and of a single-phase flow-approach to determine the effective permeabilities as a function of mean saturation. A renormalization method is used to obtain the first upscaling of the relative permeabilities. Based on the local saturation distribution known from the percolation model for a given capillary pressure and on the known local kr-S relationship a local effective permeability is obtained. A single-phase flow-averaging method is then used to solve the pressure equation for a single phase imposing a unit pressure gradient. It is assumed that the motion of one fluid has no impact on that of the other fluid. From the pressure distribution a velocity field and finally the effective permeability of the phase considered is determined. This yields a single point on the upscaled relative permeability-saturation relationship. Repeating the analysis for different capillary pressures, and thus different saturations the entire relative permeability curve is constructed. This approach is used to compare measured and simulated NAPL distributions. The results are promising, since the overall spatial extent of the DNAPL plume could be approximated well.

[202] provides an extensive review and critique of modeling approaches for fractured and faulted rocks, concluding that the idealizations: equivalent continuum (ECM), single continuum (SCM), dual continuum (DCM), DCM with connected matrix (DCCM), DCCM with matrix gradients (DCDM) have serious shortcomings when applied to the full THMC problem.

4.5 Syntheses and Summaries

4.5.1 FORGE Work Package 3

The purpose of FORGE WP3 was to examine how unresolved issues related to gas migration could detrimentally alter the hydraulic and mechanical (and potentially the thermal and chemical) properties of the engineered barrier systems. A detailed series of laboratory and field scale experiments was undertaken to provide new fundamental insights into the processes and consequences of gas migration through the engineered barrier and seals of repositories.

The main focus of the work has been on the migration of hydrogen gas through barriers and seals consisting of bentonite or bentonite sand mixtures. This also includes the interfaces between bentonite and other system components. However, some work has also been done for gas migration in cementitious barriers.

4.5.1.1 Bentonite based barriers

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

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

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

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

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

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

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

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

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

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

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

4.5.1.2 Concrete barriers

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

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

4.5.2 FORGE Work Package 4

At the onset of FORGE the rationale of WP4 was 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).

After repository closure, 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 last, but not least, by the nature of the actual gas source term (gas generation rates, gas species).

While previous EU projects such as SELFRAC [203] and NF-PRO [204] improved basic understanding of EDZ creation and evolution along selected stress paths, a paucity of data defining key HM responses remained, in particular those associated with unsaturated and saturated conditions. WP4 (see [205] for the related summary report) identified a number of issues including the evolution of sealing and gas migration during complex stress paths, fracture self-sealing, tunnel convergence, radionuclide displacement and possible repository over-pressuring. As common with other WP's, a combination of focussed laboratory, field and numerical simulation techniques were employed to develop process models in support of repository performance assessment (WP1).

All experiments followed a similar approach, resaturating the sample/borehole before the injection of gas. It is important to acknowledge that obtaining representative 'damaged' material (both laboratory and field) is technically challenging and would require a large matrix of tests in order to cover all 'disturbed' scenarios. Within the timeframe of FORGE this ambition could never be fully realised, but our understanding of processes governing gas flow in the EDZ has significantly improved as a result of the project.

The movement of dissolved gas by diffusion in the liquid phase will occur in all porous media as soon as gas is produced. Gas diffusion occurs predominantly in the connected micro-, meso- and macropores of the intact host rock, whereas the contribution of the EDZ fractures to enhance diffusive gas transport capacity is limited. For this reason, dedicated experiments focussing on the evaluation of gas diffusion coefficients in disturbed host rock formations were not in the scope of FORGE, which was instead designed to address key issues associated with advective gas flow. Gas diffusion in the liquid phase is omnipresent in the WP4 experiments. Such phenomena are only relevant in small rock samples, because the Peclet number (ratio between advective and diffusive flow) is proportional to the travel distance through the specimen. Outside of FORGE, a large number of experiments defining gas diffusion coefficients in intact rock have been undertaken for a range of candidate repository materials.

Data from WP4 clearly demonstrates that gas flow is initially focussed within the EDZ, the network of discrete EDZ fractures acting as a preferential pathway for gas migration. Within the EDZ, flow is seen to be highly localised along the largest EDZ fractures, exhibiting a complex inter-dependence between flow rate and the distribution of radial stress around the tunnel. This zone is not stable over time, as demonstrated in laboratory experiments. Analogue tests with clay-filled interfaces (representing fractures) clearly demonstrate temporal evolution of the flow behaviour. A multitude of mechanisms may affect gas transport properties including 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).

However, interpretation of the data to elicit the fundamental processes governing gas flow in the EDZ is complex, due to the difficulties of characterising this zone. For laboratory experiments it has been difficult to reproduce EDZ damage and fully represent its behaviour (i.e. the anisotropic distribution of fractures around a tunnel). In the field, it has also been difficult to characterise the evolution in properties of the EDZ following closure (i.e. limited instrumentation in respect to localised phenomena; monitoring boreholes intersecting the EDZ will have their own corresponding damaged zone). Despite these challenges, significant progress has been made on understanding gas flow in the EDZ. However, incomplete descriptions of hydro-mechanical processes exist and can lead to non-unique interpretations of the data resulting in a limited predictive capability of the models.

In light of these observations, identification of the appropriate theoretical framework to describe gas flow in the EDZ is far from trivial. Numerical simulations based on two phase concepts are able to represent elements of the data by homogenisation and simplification of the system. However, the validity of such an approach to forward prediction of repository performance is restricted, because these concepts have a limited capability to mimic hydromechanical coupling evident in the data(e.g. pressure-dependence of porosity, permeability, entry pressure).Models have been developed that mimic dilatancy using a number of different approaches in order to better represent the data and to improve simulations. While current modelling approaches do not capture all the elements observed in the experimental data, they provide a powerful tool with which to test and validate conceptual models and theoretical frameworks used to describe these systems.

Upscaling of the EDZ related features and processes in time and space is another challenge. In practice, modelling the evolution of entire system may not be practical. Instead, it may be necessary to reduce the system to individual simplified elements and to adopt a phenomenological approach. Only with a full knowledge of the stress state and gas flow boundary conditions together with a detailed description of the initial EDZ fracture network in terms of geometry and hydraulic characteristics will it be possible to understand and predict the real behaviour of gas at the repository scale. Partial validation of model predictions to repository scales can be achieved using the following steps: (i) blind predictions based on lab tests; (ii) calibrated predictions from field data; (iii) final predictions and evaluation of results (based on all appropriate data).

While considerable progress has been made on examining the fundamental controls governing the advective movement of gas in the EDZ, definitive interpretation of the data to identify the underlying physics controlling gas flow, remains elusive.

4.5.3 FORGE Work Package 5

The aims of WP5 were to establish the conditions under which the different gas migration processes are dominant, to identify how those processes can be modeled and to determine the values of the main parameters and finally to establish whether an impact on the long-term safety as a consequence of enhanced radionuclide transport through the host rock could be expected.

When one wants to perform experiments on undisturbed clay host rock, one needs to take into account some major experimental challenges i.e.

- In laboratory experiments, it is quite difficult to obtain and prepare samples that are representative for undisturbed clay host rock and it is very difficult to obtain reliable two phase flow parameters at very low degree of desaturation which is the most representative of what is expected in situ.

- For in situ experiments, one needs to be aware that drilling boreholes causes an EDZ at the borehole wall. Although self-sealing will occur, this borehole EDZ will have a strong influence on the pressure evolution of test intervals. This effect might be stronger in stiffer clays. In in situ experiments it can also be quite difficult to have exact measurements of the initial gas filled volume in a gas injection interval.

Measuring the water retention curves of an indurated clay like the Opalinus clay remains a difficult task especially at high water saturation where the spread in results remain high. No difference was noted between applying matric or total suction and the expected hysteresis behavior was observed. Increasing mechanical stress seems to further increase the air entry value which is already in any case high (6 to 34 MPa)

It is quite 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 when a gas phase flow through an undisturbed clay that very little water is displaced. Very carefully performed laboratory experiments in which all mechanical and hydraulic boundary conditions are well controlled point to hydro-mechanical coupling and pathway dilatancy as gas transport mechanism.

It is still debatable as to whether or not hydro-mechanical coupled modeling is required to model the actual gas flow and pressure transients. In some cases one was successful to model experimental outcomes using standard two-phase flow codes however modifications for permeability and water retention were always required. In other cases only through hydro-mechanical coupling one was able to grasp the main features of the experimental results. From the modeling exercises it was also very clear that it is essential to correctly represent the details of the experimental set-up, its in situ installation and experimental history to have a correct representation of e.g. the gas injection boundary condition and its evolution. In general one needs to take into account a borehole EDZ what causes additional uncertainty in the modelling.

As very little water can be displaced by a gas phase through an undisturbed clay host rock, there is little risk for advective transport of contaminated water form a repository through the clay host rock. As the gas will take the easiest way the disturbed host rock around excavations (galleries or disposal cells) and the access ways to repository will most probably act as preferential pathways for the gas to escape. Large scale simulations for simulated geologies show that gas flow is very sensitive to local variations in gas transport properties with the gas taking always the way with the lowest resistance with also the inclination of layers (or pathways) having an impact due to buoyancy. As the gas generation rate and local variations in gas transport properties (thresholds for gas entry) determine the dynamics of the pressure buildup, the gas generation rate will have an impact on in which pathways the gas flows and thus how the gas spreads over the repository and geology.

4.6 Implications for system description of repository-scale gas behaviour

4.6.1 Introduction

Most experimental data are acquired to define or comfort phenomenological conceptual models and are therefore independent of national repository layouts and concepts. But when it comes to estimate gas and water behaviour at repository system scale, waste emplacement concepts and general architecture become more essential.

More than this, behaviour estimation at repository scale implies gas migration at kilometre scale during several thousands of years at minimum and thus spatial and time upscaling compared to experimental data acquisition (mostly centimetre to decimetre spatial scale and years as characteristic time scale).

Nowadays, only numerical simulation is able to approach these goals. The mathematical formulations used for phenomenological constitutive laws implemented in the codes are based as far as possible on results from experiments and the numerical mesh represents specific repository geometry as finely as computer power enables it. Thus, gas (and water) behaviour estimated from numerical models are deeply connected to conceptual phenomenological models and repository structure at all scales (from emplacement cells to shafts including a drift network).

Nevertheless, some generic behaviour can be derived from global repository scale numerical simulations. Only these types of behaviour are described below, being representative of some generic repository concepts (for clay or crystalline rocks) widely used in Europe and of phenomenological models implemented in most porous media two-phase flow numerical codes.

These behaviours are mostly derived from work done in FORGE (WP2 to WP5 for experimental data and WP1.2 for numerical results at repository scale) but results from outside FORGE were also taken into account to derive the synthesis below.

4.6.2 Conceptual phenomenological model at system scale

Based on experimental data, phenomenological conceptual models implemented in the porous media two-phase flow codes are sometimes adapted to be more representative of the space and/or time scale characteristics of the simulations, especially if upscaling or simplifications are needed mainly to fulfil the numerical constrains linked to computer power limitations.

4.6.2.1 Gas generation – simulation at repository scale

For simulation at repository scale, gas source terms are simplified compared to the chemical knowhow about this process mainly to limit the number of non linearities in the code, thus in most numerical modelling at repository scale corrosion is:

- Constant over time and representative of a mean value of the long term corrosion rate (some micron/year for non-alloyed metal in contact with clay, one order of magnitude lower for metal in contact with concrete);
- Constant for each type of material containing metal or in contact with metal (mainly clay and concrete);
- Constant whatever the water saturation (relative humidity). Experimental data are showing that corrosion rate is lowered if water saturation (or relative humidity) is under a defined level which is generally around 70%-80%. Justification of this assumption is made *a posteriori* as water saturation for all

material as over 60% at all times and mostly over 80% after several thousand years. These figures can of course vary with repository concepts and fine simulations at national level can adopt more complex formulation (at a numerical nonlinearity cost);

- Not implying consumption of water. As for water saturation, this simplification is justified afterwards once the results are known showing that water flow is generally largely sufficient compared to the flux needed for corrosion. Of course this assumption is conservative for water saturation and thus for gas pressure. More phenomenological representations are possible at national level if needed.

In most of the cases, repository gases are assumed to be hydrogen and all other gases are neglected. This is true as hydrogen generated from corrosion and radiolysis represents more than 90% to 95% of all the gases produced in a repository (depending on national waste treatment scheme before storage). This assumption also simplifies the numerical modelling restricting the number of gas components which migration has to be simulated.

4.6.2.2 Water and gas movement

Water and gas movement are linked to two main processes: convection, sometimes coupled with mechanical processes, and diffusion. Phase changes are also a dominant process in the over whole gas and water transient period at repository scale.

(a) Dissolution and phase changes

In most of the codes used for water and gas migration at repository scale, the dissolution limit is estimated using Henry's law:

> "*At a constant temperature, the amount of a given gas that dissolves in a given type and volume of liquid is directly proportional to the partial pressure of that gas in equilibrium with that liquid*."

An equivalent way of stating the law is that the solubility of a gas in a liquid is directly proportional to the partial pressure of the gas above the liquid. The coefficient relating solute concentration and partial gas pressure is varying with water pressure and temperature.

This simple model gives a good estimation of the phase change phenomena even if for very specific cases the estimation of solubility limit can be over or under estimated from 10% to 20%.

(b) Diffusion

At the beginning, when gas is still all dissolved, it can migrate following water convection or by molecular diffusion in the water contained in the porous media porosity. In an eventual gas phase, each gas components (at minimum hydrogen and water vapour) can migrate either by gas convection or by molecular diffusion.

In both phases the molecular diffusion is estimated by Fick's law. In non-saturated cases, both for water and gas, the influence of porosity and porosity saturation are taken into account. One of the main empirical models to do this is the Millington-Quirk model. The uncertainties on these model parameters are high as there are very few data especially for concrete, clay host rock and bentonite. Some other empirical models can be used, but this one is at least commonly used in vadose zone hydrology and is validated for soils.

(c) Convection: two phase flow or pathway dilation

Two phase flow model by Darcy law assumes that gas is penetrating a water-saturated porous media by displacing water in the pores once that a certain level of pressure is reached. This level is linked to the pore structure, the smaller the connected pores, the higher the gas entry pressure. For most of the natural porous media, convection can be approximated by extended Darcy law assuming a constant coefficient linking pressure gradient and flow, both for water and gas.

In a repository, porous media are far less permeable than in the surface or subsurface soils mainly investigated by this model and thus some deviations can appear. For backfill and conventional concrete these deviations are not significant. However, for clay (including bentonite) and for granite specific models have sometimes to be used.

For granite, assuming double porosity models can ease the problem and Darcy law can mostly still be applied in two distinct media representing the fractures and the matrix between the fractures.

For very dense plastic clays (including bentonite at high density as used in certain concepts plugs) the porosity structure can imply very small pores diameter and gas entry pressure can be higher than mechanical constrains, which in first approximation is more or less equal to the swelling pressure of the clay. In this case, the gas is entering the porous media when its pressure is reaching the mechanical stress and not the gas entry pressure: the process enabling gas migration in the porous media is not Darcy two-phase flow. Conceptually and schematically speaking, the gas is opening the smallest existing pores to create a pathway (a micro fracture) through the porous media. This process is called pathway dilation. This process is not displacing water, and the opened paths are not modifying the porosity significantly. Conceptually and schematically speaking, the gas phase is concentrated in a coarse network (if not in one) micro fracture and the surrounding porous media is fully water saturated. Once the gas pressure is falling lower than the mechanical stress, the opened pathways are closing, gas is no more able to flow through it and the porous media can immediately be considered as fully saturated.

Whilst data exist on this pathway dilation process, the conceptual model described in brief above has not been synthesized in a simple mathematical formulation that is (a) recognized and adopted by the European researchers' community and (b) able to be implemented in codes used for repository-scale water and gas migration simulations.

Furthermore, in consideration of resaturation, Darcy two-phase flow can be applied in most of the cases. This is because during re-imbibition water is entering the porous media desaturated by another process than pathway dilation (this process could be desaturation in an air atmosphere at low relative humidity).

Thus for most simulations at repository scale, Darcy two-phase flow is assumed for bentonite and host rock clays. Except for specific buffer concepts (see section 4.3.1), this assumption is somehow further justified by the fact that bentonite and host rock clays are the less permeable porous media on a path from gas generation zone toward shafts and are thus hardly used by gas to migrate at repository scale (gas is following the easiest path flowing through EDZ, interfaces or backfill).

4.6.3 Generic implications for description of system behaviour

As already stated before, implications at repository scale can only be obtained by numerical simulations and are at least partly linked to repository layout. Specific behaviour linked to waste package and/or overpack, emplacement cell design or global repository layout cannot be described in this general report.

However, from the different repository concepts studied in European countries, and from the knowledge gained from several numerical simulations, the gas behaviour at some specific scale can be related to disposal concepts.

- At waste emplacement scale, the difference in gas behaviour will be focused on the choice of EBS including waste and packaging.
- At larger scale, the type of host rock is of most importance and the sealing concept can also impact the general gas behaviour.

Gas behaviour in relation to a repository hosted in an indurated clay is considered in the remainder of section 4.6.3, by way of an illustration of system behaviour.

4.6.3.1 Behaviour at cell scale

At cell scale, where most of the gas is produced, the way waste packages are put in place has a significant role in the general gas migration at more global repository scale. One of the main issues affecting gas generation is whether or not the waste packages are fully embedded in a bentonite (or similar plastic clay) buffer or not.

(a) With a bentonite buffer

If embedded in such an impervious buffer, water can reach the packages only after flowing through the bentonite. As this material is put in place in the cells at high density and partial water saturation, no water can reach the waste packages before complete resaturation of the buffer. This can take several tens to several thousands of years depending on the buffer itself:

- Thickness of the bentonite walls;
- Density of the bentonite;
- Type of emplacement method to create the bentonite buffer : high compacted bentonite blocks or rings, pellets and powder;
- Initial water saturation
- Voids around the waste packages and between the buffer and the host rock.

It also depends on the host rock ability to conduct water. In very low permeability clay host rock, the water needed to resaturate the bentonite buffer will need more time to reach the emplacement cells than in fractured granite. However, whatever the buffer and the host rock, water will never be available in sufficient quantity to corrode significantly the waste packages until the buffer is completely resaturated (or at least very close to complete saturation).

It is assumed that the bentonite buffer will need to be saturated for gas (hydrogen) to be produced. As inside the buffer the technical voids are reduced to a minimum, hydrogen pressure will rapidly increase. Once reaching the bentonite swelling pressure, the gas will force its passage through the bentonite plug migrating by pathway dilation. As part of the gas escapes the buffer, the pressure drops down and the pathway closes. This periodic process will continue until corrosion of all the metallic part of the waste packages with average gas flow being representative of the evolutionary surface of metal offered to corrosion and the corrosion rate depending on the availability of water.

If the buffer is made of blocks, it could be that interfaces between blocks represent a weak path from the inside to the outside of the buffer and that gas will essentially flow through these.

If the buffer is more meant to generate a chemical interface (to be certain that water reaching the waste has some beneficial characteristic relating to e.g. its pH) than an hydraulically impervious cage (quite low density pellets and powder), than it could be that the gas entry pressure by smaller than the swelling pressure and Darcy two-phase flow can apply even during desaturation.

(b) Without a bentonite buffer

If the waste packages are not embedded in a bentonite buffer, or within any other low permeability material, then water could access any metallic parts soon after ingress from the host rock; in this instance no buffer resaturation would have to occur before groundwater could be used in e.g. corrosion reactions with the waste package.

Such emplacement cells concepts are mainly found in very low permeability clay host rock being able to compensate the lack of buffer; the water availability is low and gas will hardly desaturate the rock. Once a gas phase appears in the voids around the waste canisters, and once its pressure will be sufficient, it will flow through the weakest path. As the host rock is very impervious it will mainly use the interfaces along the cells walls. These interfaces can include an EDZ or be reduced to the contact surface between the rock and the cell sleeve. As the amount of generated gas is quite small, theses interfaces are enough to allow migration along the cells wall toward the gallery network and finally toward the shafts.

Depending on (i) repository depth, defining (at least partially) the mechanical constraints in the host rock, (ii) rock hydraulic characteristics and (iii) thermal load of the wastes, some gas could also migrate into the host rock radially to the cell walls.

If gas entry pressure in the rock is higher than the mechanical constraints, then gas could enter the host rock by pathway dilation. This process is generally enough to allow gas migration and no macro fracturing should occur. However, if mechanical constraints are significantly higher than host rock gas entry pressure, than gas can partially desaturate the clay rock and migrate by Darcy two phase flows. However, as host rock has very low permeability and high retention curve, desaturating the host rock is more energy demanding than passing through the interfaces along the cell walls. Thus clay host rock is hardly desaturating and only a small part of the free gas is migrating inside the rock itself.

In the case of heat-generating waste, the increase of temperature is (i) expanding the gas, (ii) reducing the dissolution potential and (iii) generating a water overpressure around the cell (implying an increase water flux toward the cell). These three processes all contribute to an increase of gas pressure compared to a non-exothermic cell. These overpressures can be significant but are not lasting as they are vanishing with the temperature decrease inside the cell once the exothermic potential of the waste is lowering.

4.6.3.2 Behaviour at module and repository scale

Once out of the emplacement cell the gas migrates into the repository and the host rock, depending on the general layout concept and the type of rock.

(a) In a crystalline rock

In a repository build in a crystalline host rock, gas can escape the repository by any of the secondary fractures intercepted by the gallery network and not plugged by a bentonite seal (or whatever type of plug used). In this context, it is difficult to define precisely the path of the gas, and potentially it could move relatively rapidly towards the geosphere either (i) by developing a continuous gas phase or (ii) by dissolving in the geological water present in the host rock fractures and migrating by convection and diffusion as a solute. Behaviour of gas in the geosphere is highly site-specific.

The second migration scheme is more likely to happen if the crystalline rock has a higher porosity and/or a higher permeability and/or if this rock is heavily fractured, all this enhanced properties allowing for more gas/water contact and more dissolution possibilities.

In a crystalline host rock the expansion and duration of the gas phase are very variable depending on the layout concept and the rock hydraulic properties. It may be that once escaping the bentonite buffer surrounding the waste canisters, the gas immediately dissolves in the host rock water. The gas can as well move quite far from the emplacement zone before complete dissolution. Inside the bentonite buffer, a free gas phase is likely to be maintained as long as metal is still to be corroded; the duration of the continuous gas phase is highly concept dependent.

(b) In a clay rock

In a repository built in a clay host rock, the gas is somehow trapped into the rock. The easiest path to the geosphere is not the rock itself (very impervious and with high retention curves) but the gallery network.

Depending on the type of rock (mainly its mechanical characteristics) and the depth of the repository (mechanical constraints), the repository concept will imply, or not, a backfilling of all galleries after the operational phase as being required.

In case of a concept that requires backfilling, the backfill material is mainly designed for its mechanical characteristics (to avoid the collapse of the galleries) and its hydraulic characteristics are not optimized. Therefore the backfill material is potentially quite important compared to the one of the clay rock and even to the EDZ with respect to gas, which can migrate into the backfill wherever it exists. The backfill can constitute a high volume of material into which gas can migrate, which may help reduce gas pressure as a whole in the repository (the role of the backfill as a 'storage volume' for gas could be important). In case of a concept not requiring any backfill, the expansion volume and the facility for the gas to migrate is even higher.

Most, if not all, repositories are sealed for long term safety reasons mainly to prevent and reduce solute radionuclides migrating with water toward the shafts. The design and number of seals vary with the national concepts especially regarding performance toward gas migration. However, whatever the seal design, it represents a barrier for gas migration compared to the backfill. Pressure upward of the seals is thus building up. The values of the maximum pressures are depending on the seal hydraulic characteristic (including the eventual EDZ and interfaces surrounding the bentonite plug of the seal) and on the gas flux:

- The higher the gas flux, the higher the pressure;
- The higher the gas entry pressure in the seal, the higher the gas pressure upward;
- The higher the seal permeability to gas, the lower the gas pressure.

The main gas pressure loss during its way toward the shafts is by passing through seals which are generally emplaced to separate storage zones dedicated to different type of wastes. Having redundant seals in the shafts and/or in the gallery network helps restricting the gas flux reaching the overlying geosphere.

Water movement due to gas pressurisation is a secondary process and even if it exists, the water is moved only during the gas phase expansion (afterwards gas is no more able to displace water) and the maximum length of the water displacement is negligible compared to the total distance from emplacement waste to the shafts.

During the whole travel path from emplacement cell (where gas are mainly generated) toward the shafts, the gaseous phase is in contact with the host rock water. This implies dissolution all along this path; the longest the path, the higher the dissolved percentage of the initial gas. If the path is long enough, it can arrive that all the gas dissolves before reaching the shafts and no free gas is arriving in the overlying geosphere.

Once dissolved, the gas migrates (i) by convection with the host rock water and (ii) by diffusion. As host rock permeability is very low, in most of the cases the migration of dissolved gas in the host rock is dominated by diffusion. These processes (dissolution/diffusion) are in many cases the main processes of gas migration at repository scale over longs periods of time.

Depending on the seal concept (especially the way the different storage zones for different types of waste are separated from each other) and on the emplacement cells concepts (type and amount of metal present, volume of technical voids), the resaturation time can be significantly different in different repository zones. As the shaft zone is somehow separated by a seal from the emplacements zones and is (in most concepts) not containing high gas generating products (except some metallic parts in the gallery walls for mechanical reinforcement), this zone is the first to resaturate. Time of resaturation of the emplacement zones is highly dependent on the concept and complete resaturation time can reach hundreds of thousands of years.

During resaturation, water saturation values can vary significantly in time but also in space depending on the type of material; the higher the retention curve, the higher the water saturation for a given gas pressure. In general, the host rock and the bentonite plugs are hardly desaturated by the gas pressure (some percent at the most), but backfill and in a lesser extend EDZ and concrete can desaturate more significantly. So even at cell scale final resaturation time covers a variety of behaviour and some material can be very close to water saturation at quite early time whereas other material can still be far from water saturation. On a general basis, if technical voids exist and are maintained open during the whole resaturation process, they will be the last part to resaturate. Not being porous media, they have no retention curve: relative humidity will reach 100% only when the material in contact with the voids will already be saturated and liquid water will (in first approximation) appear only once relative humidity has reached 100%.

On a general basis, whatever the repository layout and the precise hydraulic characteristic of the host rock, the key controls on migration of gas in clay host rock are:

- Dissolution and diffusion processes in the host rock;
- Hydraulic parameters of the host rock, and its EDZ, mainly intrinsic permeability for gas and capillary curve;

- Depth of the repository, relating to hydrostatic pressure and mechanical constraints.

4.6.4 Possible implication for scenario description

These implications are quite different depending on the type of host rock.

4.6.4.1 Crystalline host rock

For crystalline host rock, on a general basis, the implication can be separated between the cell scale and the host rock.

At cell scale, a gas phase can be maintained inside the buffer during all the gas generation period. This is highly concept-dependent as the gas generation rate and the potential gas escape rate are linked respectively to the waste package conception (type of metal, surface of metal, thickness of metal) and to the buffer conception (type of plastic clay, percentage of non-swelling material, density of the clay, designed swelling pressure, thickness of the buffer, buffer emplaced by blocs of pellets). If a gas phase is present during a long period inside the buffer it can potentially delay the release of some soluble radionuclides. On the other hand, a continuous gas phase (even if by intermittence) from inside the buffer to outside the buffer may increase the migration of gaseous radionuclides toward the geosphere.

Once out of the buffer, the path and type of migration for the (gaseous or soluble) radionuclides is very host-rock dependent (density of fracturing, porosity and permeability of the rock matrix). If a gas phase cannot be maintained in the rock over a long distance compared to the distance of the biosphere, no radionuclides can move under gaseous form toward the biosphere. If on the contrary such a continuous path exists, and if fracture permeability to gas is high and porosity is small, the time for gaseous radionuclides to reach the biosphere could be very quick (quicker than the half-life of key species such as carbon-14 as might be present in $^{14}CH_4$, although note that some free gas migrating in a fracture would diffuse / dissolve in the groundwater present).

Whatever the crystalline host rock, fractures gas permeabilities are high enough to prevent any additional fracturing; over-pressurisation will not occur.

4.6.4.2 Clay host rock

In clay host rock, the difference between cell scale and repository scale is less important regarding gas oriented scenario as once out of the cell the gas has to move along the gallery network before reaching the shaft, only way to eventually reach the biosphere under gaseous form (before dissolving).

Before describing the potential impact for gas scenarios, let's consider the potential impact on water and soluble radionuclides migration. During resaturation of the repository, no long distance organized water movement is possible as there is no continuous water gradient throughout the drift network from emplacement cells toward the shafts. During this period water gradient are mainly radially oriented to permit the resaturation of the repository elements by the host rock water. Assuming a continuous gas phase, some gas driven movement of water can exist along the drifts but (i) it will last only during the gas phase expansion and (ii) the movement of water will be small compared to the length of the path from cells to shafts (typically several tens of meters at most).

Concerning the gas phase – which will eventually be continuous from the waste emplacement cells to the shafts - several considerations can be taken into account concerning gas related scenarios:

- Whatever the concept, the generated volume of gaseous radionuclides cannot create a continuous gas phase by themselves. The only way for radionuclides gases to migrate is to follow the hydrogen generated gas phase;
- Gases in the hydrogen generated gas phase are flowing from cells toward the shafts only during the expansion of the gas phase and only during the time this phase is continuous from cells to shafts. As in general there is no (or small amount of) gas generated near the shafts, this zone is resaturating first in a repository;
- As in most concepts the emplacements zones are well separated from the shafts zone by long galleries, the time for gas to migrate from a cell to a shaft can be high compared to the half-life of most gaseous radionuclides.
- During the migration of gas from cells toward the shafts, dissolution is an important process acting all along the path and only very small amounts (if none) of the gas volume generated in the cells are reaching the shafts under gaseous form.
- Even if in some cases dilation pathway (creation of reversible micro-fractures) can be the main gas migration process there is no evidence that gas pressurization at repository scale could produce macro-fracturing of the host rock.

5 IMPLICATIONS OF GAS-RELATED ISSUES FOR THE SAFETY FUNCTIONS OF THE EBS AND THE HOST ROCK

5.1 Introduction

It is generally expected that non-radioactive gases will be generated in amounts sufficient to potentially influence some of the processes and conditions prevailing in a geological repository and upon which safety functions allocated to components of the repository may be relying. This section builds on section 3.3.1, and on the developments in understanding regarding gas generation and migration that derive from FORGE and from work outside of FORGE, to provide a detailed discussion of the possible implications of gas generation and migration on safety functions.

5.2 Containment

The safety function “containment” in a geological repository is generally fulfilled by a metal waste container. Containment is still provided to a degree in disposal concepts that have vented waste packages, as the package itself and the choice of vent material limit the rate at which groundwater can interact with in-package waste. The possible implications of gas generation and migration on containment are discussed in the present section. In other terms, the present section discusses how gas generation and migration can affect the favourable properties of waste containers in a geological repository. The discussion is based on the contributions from the different WPs.

Gas generation is closely related to the degradation of containment, as a large part of the gas in a repository is produced by anaerobic corrosion of metal waste containers (although this is noted to vary dependent on the national waste inventory under consideration). Several processes related to gas production and migration can affect containment but waste container corrosion is a prerequisite.

The following processes may influence the ability of a waste package to contain radionuclides:

- Processes altering geochemical conditions:
    - Microbial sulphate reduction by microbes that consume H_2 may lead to the production of sulphide and thus enhances steel corrosion. On the other hand H_2 oxidation would reduce H_2 concentration.
    - External corrosion of waste containers may be enhanced due to iron reduction induced by the generation of gaseous H_2 (e.g. [1]).
    - Gas generation may influence the geochemical conditions in the near field and/or the host formation. A potential positive effect is the even more reducing conditions imposed by the presence of hydrogen gas. A potential negative effect is the lower pH conditions imposed by the presence of CO_2 gas produced through microbial degradation of organics.
    - Carbonation can consume the pH buffering capacity of the cement, and hence increase the metal corrosion rate.
    - pCO_2 has a major control on the pore water pH most notably in bentonites and this can influence corrosion rates (to some extent, although most safety assessments tend to assume a linear corrosion rate

for high pH cement / near-neutral conditions for generally oxidising and reducing systems).

- Saturation degree:
  - The partially unsaturated conditions linked to gas generation in a clay host over several thousand years are favourable to waste containment. The gas pressure could force the water around the waste containers away. These conditions are retarding the water flow and increasing the period over which metal waste containers fulfil their containment function.
- Internal corrosion:
  - The presence of water in metal waste containers induces a pressure rise through internal corrosion. The relevance of this process varies according to the amount of water contained in the waste package and on the waste type. However, in most cases, internal corrosion is more likely to be a consequence of containment loss through external corrosion than its cause.
- Gas pressure build-up and hydrogen embrittlement:
  - Gas pressure rise in the near-field could also affect corroded waste containers. However, external pressurization of the containers is expected to be more or less isostatic and hence relatively high pressures would be required to damage these components.
  - Hydrogen production could initiate premature failure of steel containers due to its diffusion into the metal lattice (H_2 embrittlement).

In conclusion, the safety function “containment” can be either negatively or positively affected by gas generation and migration. However, corrosion remains the most detrimental process for containment.

5.3 Flow limitation

Bulk gas generation (which will in the main be hydrogen derived from e.g. corrosion processes affecting components of the near-field) in a repository and subsequent gas migration could affect the movement of any groundwater present and could also affect the movement of any radionuclides that are able to move either in a liquid phase or in a gas phase. This subsection discusses, primarily on the basis of work undertaken in FORGE, an understanding of how gas generated in a repository could interact with groundwater present in / derived from a clay host rock.

5.3.1 Variation of water and solutes movement

Independently of gas generation, at closure time, most of the materials present in a repository, or just around the excavation walls, are only partially saturated mainly for two reasons:

- Ventilation during the operational phase. This is typically the case for the EDZ (when it exists) and concrete lining in all galleries, as well as for waste containers (mainly in concrete) emplaced in ventilated cells/vaults;
- Emplacement of a clay-based component of the EBS at partial saturation, which is typically the case of all EBS including bentonite plugs and backfill.

The gas present in the unsaturated pores of these porous media has to migrate before hydraulic conditions near hydrostatic could occur. During this period, which can be of several thousands of years for bentonite plugs in high diameter galleries of

a repository and in a low permeability clay host rock, water flow cannot re-establish itself along the excavation axes over long distances as water fluxes are mainly radial.

If gas is generated in sufficient quantity it can prevent the complete resaturation of the porous media when its pressure increases over their gas entry pressure. When this will happen, for bentonite plugs the water saturation will be near 100%, but for backfill it can be much less (down to 60-70%, depending on its retention curve). This "near saturation" situation can last as long as the gas flow is maintained at a sufficient level, typically several tens of thousands of years in a low permeability clay host rock. During this period, as water saturation is not complete, water flow will not re-establish itself over long distances. This could prevent the migration of solutes along the drift network as long as resaturation is not complete.

However, in some circumstances, the expansion of a gas phase along the gallery network of a repository in a low permeability host rock can generate water movement in the same direction, if this expansion arrives at a moment when complete resaturation of the EBS is nearly complete. In this case, assuming Darcy two phase flow processes, gas needs to reopen some part of the pores and to do so will have to move some water aside. As the surrounding rock is very impervious, gas and water will mainly flow along the drifts. But over time part of the water can eventually migrate inside the host rock and thus even if the gas phase becomes continuous at repository scale the migration of water linked to this process is never extending over more than a few tens of metres, which is far from the distance between emplacement cells and shafts.

This description is more realistic for a clay host rock then for a crystalline host rock, as for this latter type of rock the transport of the repository-derived gas through natural fractures in the host rock may prevent any gas pressure build up inside the gallery network. For a repository concept in crystalline host rock that used a bentonite-based EBS, once gas is out of the bentonite buffer surrounding the waste canisters, it will flow quite freely inside the nearest fracture in the host rock and no high pressure should arise outside the emplacement cells. Nevertheless, gas can partially desaturate in some of the fractures around the repository and interfere with the natural water flow, but it will not flow preferentially along the drift network.

5.3.2 Variation of gas movement

Some waste may contain material that could under certain physico-chemical conditions evolve to produce gases. Dependent on the gas volume and gaseous species present, the amount of groundwater present and whether or not it already contains dissolved gas, all-to-some-to-none of the gas produced in a repository will exist as a free gas, with the remainder dissolving into the surrounding host rock water and migrating as solute. To migrate in free gas form, radioactive gases generated in the repository – which volumetrically are very small – need a bulk free gas to be presented to act as a supporting transport medium. This bulk free gas phase can be created by e.g. hydrogen generated by corrosion and/or radiolysis in the repository (bulk methane could also play such a role, if this were to be produced; this is an artefact of the disposal inventory under consideration).

In a clay host rock, if the hydrogen flux is high enough it can generate a gaseous phase that can expand gradually over the whole repository throughout the drift network toward the shafts. During this expansion phase, hydrogen gas pressure gradients are directed toward the shafts and other gaseous elements can use this path toward the repository access.

In most of the repository architecture, the waste emplacements zones are quite far away from the shafts and, even for a quite mobile gas as hydrogen (and also because hydrogen fluxes are quite small even at a repository level), it will take several thousands of years to travel this far. Radionuclides present in gaseous species in a repository that have a half-life significantly less than this period will have decayed to insignificant amounts during the journey. Furthermore, all along the path from emplacement cells toward the drift the gas is in contact with the host rock water as the material in which it flows are only partially desaturated - the available surface over which gas dissolution can take place is huge and an significant amount of gas is calculated to dissolve along this transit. When arriving at the shaft, most of the initial gas has been dissolved and is migrating as a solute in the host rock water.

Nevertheless, a gas flux will be maintained throughout the shaft seals as long as the hydrogen gas flux will be high enough to maintain a significant desaturation of the porous material (mainly EDZ and backfill) all along the path. This can last several tens of thousands of years.

Concerning crystalline host rock, once out of the bentonite buffer, the migration path of the hydrogen created gas phase will not follow the drift network but use a hydraulically-active fault/fracture if present (note this is a site specific issue, and the rate of gas migration in the geosphere overlying the repository could also be affected by the presence of e.g. low permeability sediments and structural traps). Gas may be able to spread over several fractures, dependent on the fracture density and connectivity; the contact surface area with host rock water can become important and a significant proportion of gas may dissolve and not migrate further as a free gas.

5.4 Ensuring stable conditions / retention of repository-derived gas

When assessing the fate of repository gases, a principal objective of the FORGE project, the potential reactivity of gases in the repository system should be evaluated as well. Although generally representing a sink term for gas pressure build-up, it may potentially have adverse consequences. Host formations and engineered barriers are selected to provide favorable conditions for the fulfillment of the safety functions of the disposal system. These conditions should be maintained as long as required in the safety concept and any phenomenon or process that may impact these conditions should be assessed in a safety case. The main gas-related phenomena identified with the potential to significantly alter the physico-chemical conditions in the near field and host formation are the following:

1) Carbonation of concrete barriers
2) H_2 chemical reactivity
3) Gas induced microbial activity

In the following sections, the relevant observations from the FORGE experiments are summarised, as well as important recent progress from research outside of FORGE.

5.4.1 Concrete carbonation

Carbon dioxide can react with the calcium hydroxide and CSH-phases in concrete to form calcium carbonate. This process is called carbonation, which has two effects: it increases the mechanical strength of concrete, but it also decreases alkalinity, which is essential for corrosion prevention of steels. Below a pH of 10, the steel's thin layer of surface passivation dissolves and corrosion is promoted.

Carbonation has been studied in construction (general) and low-level waste repository development where the impact on corrosion of the (iron) reinforcement is evaluated. The source of CO_2 is often only atmospheric and the porous media unsaturated. Some additional information is available from the field of carbon capture and storage (CCS). In the context of geological disposal, dissolved CO_2 (HCO_3^-) is present in the pore water and additional CO_2 is generated from biotic and abiotic (*e.g.* through radiolysis) organic waste degradation. Several direct and indirect consequences need to be assessed[8]. Carbonation processes can change the hydraulic properties (porosity/permeability) of concrete barriers and filling materials, thereby on the one hand reducing porewater influx and radionuclide out-flux, but on the other hand reducing the near field gas storage capacity, potentially leading to increased gas pressures or earlier structural damage in the concrete. Carbonation processes will also lower the pH buffering capacity of the cement, and hence potentially increase metal corrosion and locally affect radionuclide migration. The degree in which these effects take place is dependent on the amounts of CO_2 produced, the amounts of cement used, the transport characteristics of the EBS and host rock and the considered disposal depth, which makes evaluation of the consequences of carbonation very concept dependent.

Within FORGE, carbonation has specifically been addressed in WP3.4. BGS studied carbonation in a rather permeable and homogeneous cement system (Nirex reference vault backfill, NRVB) for both saturated systems containing dissolved CO_2 (diffusion) and systems imposing CO_2 in the gas phase (flow). The emphasis was on determining the progress of the carbonation front as a function of time using visualisation techniques and studying alterations in concrete (pore water) composition. It was observed that the fronts migrated by a few mm over several weeks (see **Figure 11**).

BGS [79] also found that carbonation of the NRVB concrete reduces its permeability by about a factor 2 (from 2×10^{-19} to 1×10^{-19} m^2), and it is acknowledged that slower carbonation reactions (*i.e.* at rates more realistic of a repository) might produce a narrower reaction front with more efficient infilling of porosity, and consequently larger reductions of permeability. Intermediate effects at the carbonation front seem to be rather complex, involving development of shrinkage cracks both parallel and perpendicular to the flow direction, which get sealed later on by secondary precipitation.

Information on interface effects of carbonation has been gathered by BGS mainly from the CCS field. It seems that in conditions allowing flow of acidic, CO_2-rich water at an interface, there is a potential for enhanced dissolution thus increasing interface permeability, on the condition that there is enough flow to remove dissolved components. In low or no flow conditions (such as in geological disposal systems), carbonation reactions rather result in a reduction of porosity/permeability, improving interface sealing [79].

The results from BGS in WP3.4 also suggest an enhanced localised uptake of Cl^- due to carbonation (see **Figure 14** and **Figure 15)** [79], [206]. The potential exists therefore, for possible retardation (or at least reduction in local mobility) of ^{36}Cl. This effect has been observed in tests from the FORGE project, and also in cement

[8] Regarding the Belgian programme, note that organic wastes (filters, bituminized waste) are generally classified as MLW and therefore generally do not consider complete containment provided by watertight metallic barriers in their safety concept. However, the pH lowering as a result of carbonation might have an effect on the corrosion rates to be applied while assessing the corrosion gas (hydrogen) source term in the considered disposal system.

carbonation tests/investigations as part of CCS studies [207]; [208], [209]. All experiments conducted as part of the FORGE project showed decreased concentrations of dissolved (non-active) chloride, which was more pronounced in the CO_2-pressurised experiments. It appears that Cl^- was being precipitated in a solid phase inside the cement. Such uptake in CO_2-free systems concurs with observations undertaken as part of previous radioactive waste-cement studies [210]. Cl^- uptake in CO_2-rich systems has previously been noted, with Rochelle *et al.* [208], [209], finding increased Cl^- uptake in CO_2-rich experiments compared to CO_2-free experiments - concurring with data from the FORGE project. Detailed mapping of element distribution in the region around carbonation fronts, showed Cl concentration on the cement-side of the reaction front. It would appear that the Cl-rich solid phase is only stable under higher pH conditions (*i.e.* where CSH phases were still present). Analysis of the fluid chemical data also suggests that: a) the Cl-rich phase is more stable at 20°C compared to 40°C, b) it is stabilised by the presence of some CO_2 (suggesting that it may exist as part of a solid-solution series with Cl- and CO_3-end-members.

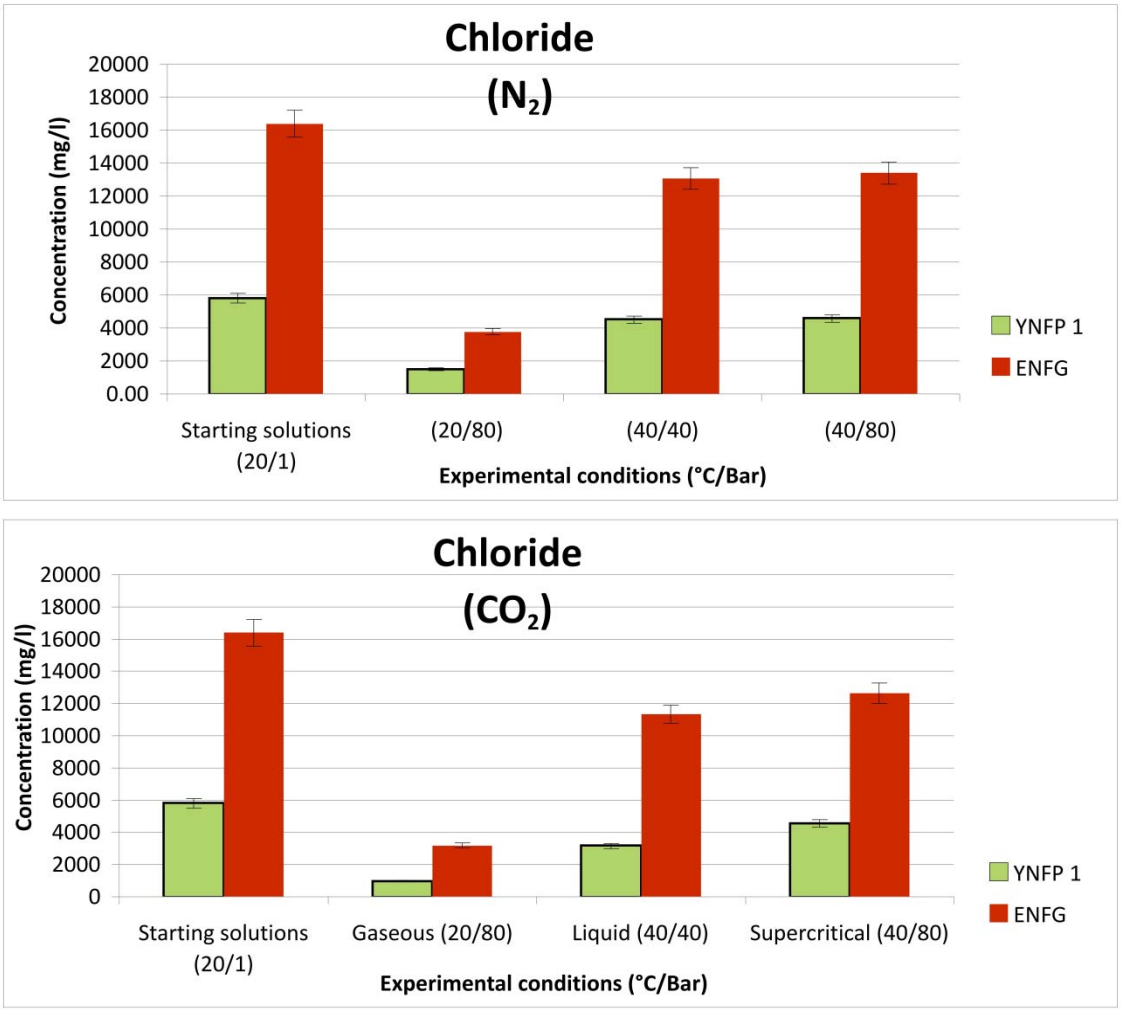
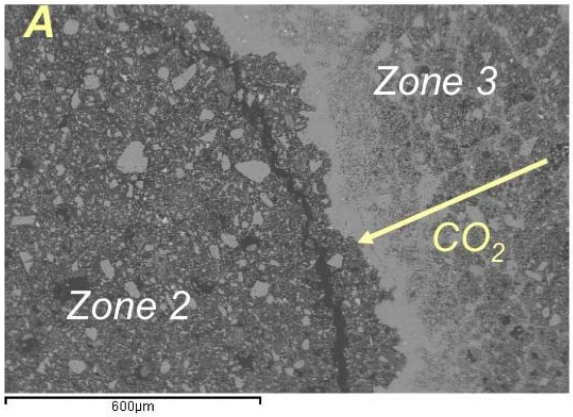


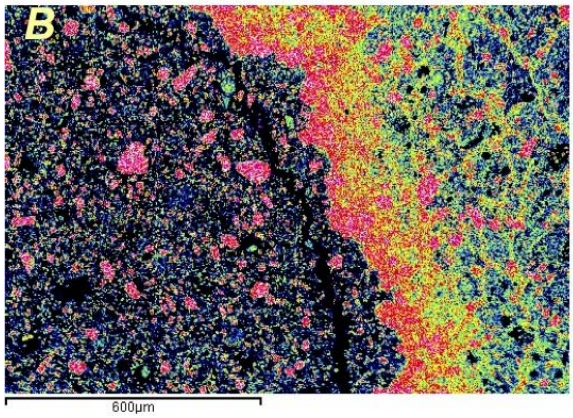
Figure 14 Chloride uptake in experiments conducted as part of the FORGE project. Note decreases relative to the starting solutions, especially at 20°C. YFNP1 = young (Na/K/Ca/OH) near-field cement porewater, ENFG = evolved (Ca/OH) near-field cement groundwater.



A) Backscattered SEM image of main carbonation reaction front.

B) Map of Ca abundance (high Ca = pink). Note the high abundance at the carbonation front, and also the limestone grains in cement.

C) Map of Cl abundance (High Cl = green). Note its abundance at the edge of Zone 2, just in advance of the main carbonation front.



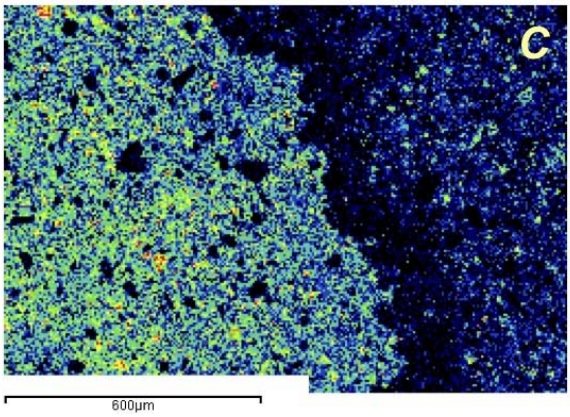


Figure 15 High resolution images of a cement carbonation front. Note the abundance of Cl on the partly-carbonated side of the reaction front, and its near absence in the fully-carbonated cement.

5.4.2 H_2 reactivity

The favourable properties of the components of the engineered barrier system and the host rock rely among others on their capacity to impose reducing conditions. Hydrogen, as a reducing agent, is therefore not directly expected to affect this favourable property. Moreover, its *in situ* reactivity could be limited because it requires high temperatures (mostly exceeding the range expected in disposal conditions, see *e.g.* research on abiotic nitrate and sulphate reduction by hydrogen at high temperatures by Truche et al., [211]-[212]), high radiation fields, or bacterial or surface catalysis.

Nonetheless, several phenomena involving hydrogen interactions with waste, engineered barrier components and host rocks are likely to occur and make that hydrogen will not act as conservative species. For instance, it is well known that hydrogen protects spent fuel against corrosion/dissolution by scavenging molecular radiolytic oxidants. To act as reductant, hydrogen is dissociated into reactive radicals, either radiolytically, on catalytic surfaces or by a combination of both. Notably, three of the ε-particles[9] (Rh, Pd and Ru) are exceptionally good catalysts for the $H_2/H^{\cdot}/H^+$ reaction [213].

[9] ε-particles are elements in spent fuel which are unstable as oxides and segregate to form noble metal particles (Mo, Ru, Pd, Rh and Tc).

Within FORGE, experiments focus mainly on gas migration through concrete or bentonite barriers/seals and through clay host formations. Since many experiments within FORGE are not performed with H_2, but with other gases (*e.g.* N_2, or inert gases He, Ar), the (new) experimental evidence for making statements on H_2 reactivity in relevant disposal conditions is not particularly large. However, some conclusions from modelling perspective can be drawn and there are some recent publications dealing with possible hydrogen interactions in disposal conditions. These are summarised below.

Particularly in argillaceous systems, hydrogen may react with structural Fe(III) in the clay fraction, pyrite or other clay minerals. Such structural and mineralogical redox changes could affect the migration of redox-sensitive radionuclides in the long term. Didier et al. [214], [215] used synthetic montmorillonites with increasing structural Fe(III) and COx samples in the form of powders to study adsorption of hydrogen and redox processes at 90°C and 120°C and at a hydrogen partial pressure of 0.45 bar. It is observed that the amount of adsorbed hydrogen does not depend on the structural Fe(III) content, and there was no significant reduction in the COx samples. Therefore, the adsorption is attributed to mainly weak surface interaction mechanisms as van der Waals, supported by the observation that at higher temperatures, favouring desorption through molecular thermal movement, adsorption is lower. It is concluded that more than 18 m^3 of $H_2(g)$ could be absorbed per m^3 of raw COx, which could considerably reduce the gas pressure in the repository near field. This value seems to be consistent with the work of Gil et al. [216], who studied H_2 adsorption on aluminium pillared montmorillonites. Importantly, the adsorption seems to occur without redox modification of the COx host formation (since no significant reduction was observed). Although Truche et al. [217], [212] have observed (abiotic) reduction of pyrite to pyrrhotite by hydrogen gas, this happened at experimental conditions of 90°C-180°C, hence in excess of relevant disposal conditions. In other words, the COx in situ mineralogical and geochemical conditions seem to remain stable whilst providing significant H_2 adsorption capacity.

A re-evaluation of the hydrogen diffusion experiments performed in the MEGAS project [218] assessing diffusive transport of molecular hydrogen in water-saturated Boom Clay from the HADES URF in Mol, Belgium, revealed that the apparent diffusion coefficients ($D_{app,H2}$) could be as low as 5×10^{-12} m^2/s, *i.e.* significantly lower than the values reported in the MEGAS report [219]. Interactions of H_2 with the Boom Clay are lumped into a retardation factor (R) and wide scatter is observed for the product ηR(with η: porosity): from 1 to 40. Recently, an innovative test design was developed to determine the H_2 diffusion coefficient more accurately (Jacops et al. [82]). In these diffusion experiments, precautions were taken to avoid microbial conversion of hydrogen by irradiation of the sample prior to the test. Results now suggest that D_p = 7.14 $x10^{-10}$ m²/s and R=1 (Jacops et al., paper in progress), hence no sorption of H_2 on Boom Clay is observed. The low diffusion values and high retardation values found in previous studies can therefore be considered biased due to microbial conversion, leakage of H_2 and outgassing.

It is acknowledged that hydrogen may also interact with iron-bearing phyllosilicates, thereby reducing the structural Fe(III) present in the dioctahedral layer [37] [38]. These redox mechanisms may result in the loss of swelling properties and possible changes in cation exchange and sorption properties of swelling clay minerals (notably smectites of the nontronite type). Therefore, Fe-poor montmorillonite is preferred for application as engineered barrier in geological disposal systems.

In order to assess H_2 migration behaviour at relevant scales, it can be noted that modelling advancements have been achieved within FORGE. As part of work contributing to WP3, IRSN implemented a new geochemical modelling approach including multi-species diffusion, where each aqueous species has its own diffusion coefficient and where the electroneutrality condition of the pore water has to be met, thereby imposing constraints on the migration of individual species (example results are shown in **Figure 16**). This could impact the overall hydrogen behaviour in the cement/clay-based system under consideration. First calculations (in a saturated 1D medium at 25°C, without kinetics nor surface complexation reactions, water equilibrated with 1bar of gaseous hydrogen, inflow of hydrogen on the left side) leads IRSN to suggest that implementation of the electroneutrality condition slows down the diffusion of dissolved hydrogen and thus limits the amount of migrating hydrogen. This is shown for the case of local consumption of protons by redox reactions in presence of H_2, whereby additional protons diffuse from the neighbouring clayey formation pore water and contribute to pH buffering. At the H_2 gas phase entry side of the domain, the consumption of protons is stronger so that a sharp pH front can temporarily take place and thus stimulate chemical reactions.

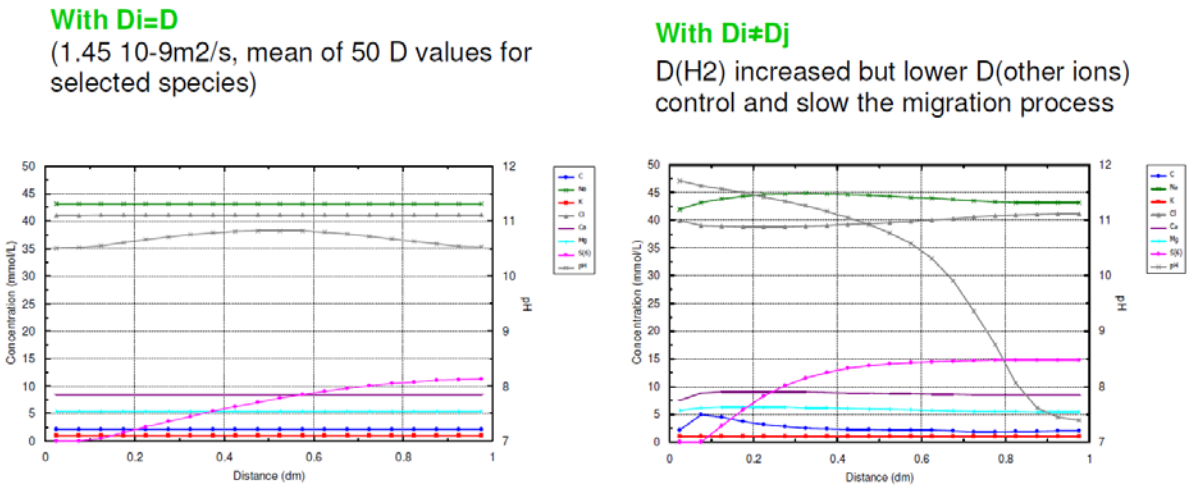


Figure 16 Concentration profiles in synthetic pore water in contact with a H_2 source after 2 years. Left: all the species have the same mean diffusion coefficient; right: diffusion is species specific.

5.4.3 Biotic H_2 reactivity

Growing awareness of the ubiquity and versatility of microbial life, expanding towards the most extreme environments on Earth, has led to an increased concern of interactions of microbial activity within and around the EBS when safety of nuclear waste disposal is addressed. Indeed, microbial activities are expected to have an impact on, among others, corrosion, pore water chemistry, clay mineral integrity, radionuclide speciation and the net balance of gasses ([40]; [220]; [43]). Considering the latter, a complex network of microbial metabolic pathways may contribute to consumption, production or conversion of gasses in and around the EBS, producing unexpected metabolites when looking from an abiotic point of view, and altering the total gas volume balance.

In order to sustain active microbial life, the presence of an electron donor and electron acceptor are essential as means to provide energy and a source of carbon is required for biomass production. The most straightforward microbial metabolism would be fuelled by oxygen as an electron acceptor and any kind of organic carbon compound as both electron donor and carbon source. In the deep subsurface

environments selected for nuclear waste disposal however, typically little or no OM (organic matter) and oxygen are available. In this case, hydrogen gas (H_2) is considered one of the most energetic substrates for microbial life, replacing OM as electron donor [13]. H_2 is expected to be readily available in nuclear waste repositories due to radiolysis and/or corrosion. Electron acceptors for microbial activity in this case would be NO_3^- (and downstream NO_2^- and NO), SO_4^{2-}, Fe(III) or CO_2, with varying optima regarding redox potential, as shown in **Table 3**. As an alternative to H_2, CH_4 is also known to be coupled to the microbial reduction of NO_3^-, SO_4^{2-}, Fe(III) [43]. The effect of these H_2- or CH_4- mediated microbial reactions is two-sided. On one hand, all of the reactions shown in **Table 3** will result into a net decrease of the gas volume, even when for example H_2 gas is consumed to produce N_2, due to the reaction stoichiometry [13]. Such evolution would be considered beneficial in terms of safety assessment. On the other hand, some metabolites produced through reduction should be considered in closer detail.

Respiration type	e-donor	e- acceptor	Product	E0 (V)
Aerobic metabolism	OM, H_2	O_2	H_2O	+8.16
Nitrate reduction	OM, H_2, CH_4	NO_3^-	N_2	+0.71
Iron reduction	OM, H_2, CH_4	Fe(III)	Fe(II)	+0.14
Sulphate reduction	OM, H_2, CH_4	SO_4^{2-}	HS^-	-0.22
Methanogenesis	OM, H_2	CO_2	CH_4	-0.26
Acetogenesis	OM, H_2	CO_2	acetate	-0.30

Table 3 Overview of possible microbial respiration types catalysed by a gas compound (H2 or CH4) as electron donor, as an alternative to organic matter (OM). Electron acceptors, reaction products and redox potentials (E0) of the redox reactions are provided.

Microbial reduction of Fe(III) to Fe(II) fuelled by H_2 is considered a major driving force in extreme environments like the deep subsurface. The process has two main consequences that interfere with abiotic geochemical modelling. The first possible consequence as indicated by [13] and [38] could be enhanced illitisation of iron(III) rich smectite clay, thereby altering the structural integrity of the clay. Note however that the clays considered as host or buffer in geological disposal applications are of the low-Fe montmorillonite type. Furthermore, smectite illitisation is known to be kinetically controlled by several physico-chemical parameters such as temperature and potassium-availability ([221] and references therein). The second possible consequence is the dissolution of the passive protective layer of oxidized Fe(III) as suggested by Esnault et al. (2011 [41]) and Libert et al. (2011) [13]), which would cause the exposure of more iron to oxidation and hence increase corrosion rates. However it can be expected that, if this phenomenon would occur at all in the very restrictive high radiation conditions, it would be transitory and an equilibrium condition will probably be established.

Sulphate reduction is considered a major pathway in microbial induced corrosion (MIC) either direct or indirect, for example by H_2S attack of a metal surface. MIC driven by sulphate reduction is typically characterized by the presence of newly-formed sulphide minerals on the corroding metal surfaces, or by dissolved H_2S in the environment [43]. Microbial reduction of sulphates can have implications on the mobility of radionuclides as well. Causes are i) the impact on the governing redox potential, however more reducing conditions in general lead to decrease in mobility,

ii) complexation effects of metabolites or extracellular enzymes, iii) precipitation effects *e.g.* of metallic radionuclides with sulphides and iv) radionuclide sorption on bacteria themselves which, in case of clay host rocks, will presumably lead to radionuclide immobilisation through filtration [40].

The microbial production of CH_4 or acetate by a specialist group of micro-organisms from CO_2 and H_2 can be considered a fuel reaction, since the metabolites can be used as energetically favourable compounds by other micro-organisms [13]. Acetate and CH_4 can be used as electron donor in all reactions shown in **Table 3**, thereby broadening the part of the microbial community that might be active in repository conditions. The possible microbial pathways involving H_2, CO_2, CH_4 and acetate can cause a complex network of interactions, as shown in **Figure 17**.

When considered as a source in these schemes, acetate can be replaced by other organic compounds as well, both simple and complex. The pathways displayed in **Figure 17** (a), (b), (c) and (d) can be performed at the same time, by different groups of micro-organisms. However, depending on the dominance of a specific community of micro-organisms, different end products and a shift in the total net gas volume are expected. A shift in community composition towards fermentation or direct methanogenesis of organic matter would increase the net volume of gasses around the EBS, while a dominance of homoacetogens and autotrophic methanogens would result in a net decrease of the gas volume.

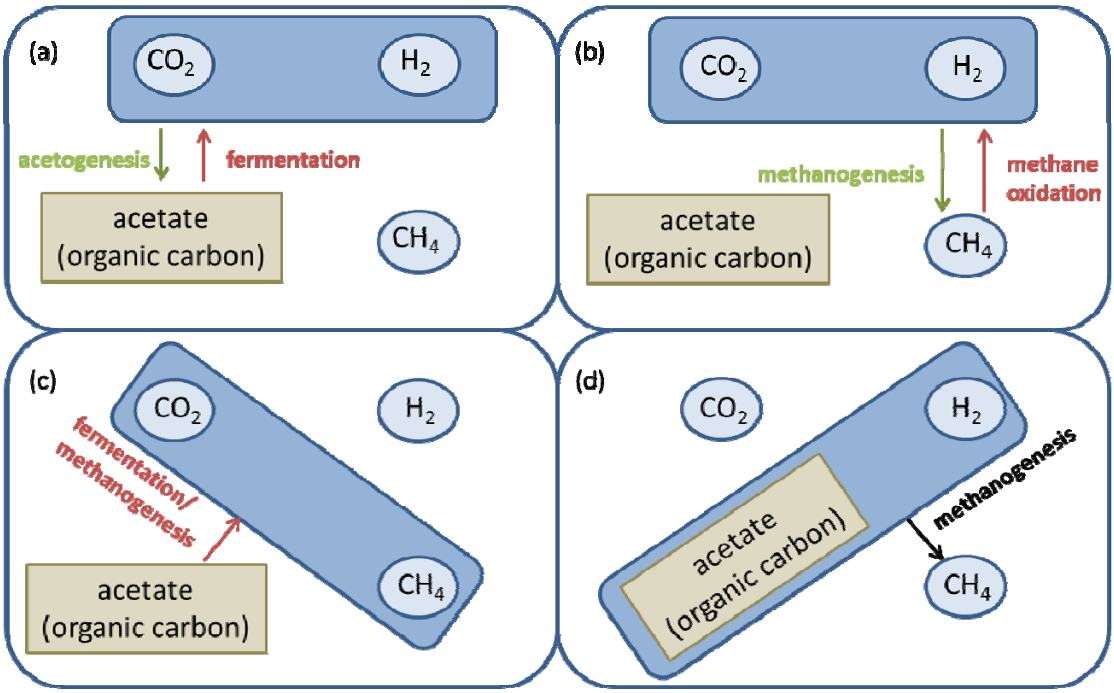


Figure 17 **Possible microbial pathways involving H_2, CO_2, CH_4 and acetate/organic matter. Gases are indicated in circles while others are indicated in a square. Reactions in green, red and black result respectively in net gas consumption, production or conversion.**

The proliferation of a specific sub-community of, for example, methanogens, is dependent on (i) the composition of the original subsurface microbial community, (ii) the introduction of allochtonous micro-organisms, (iii) the evolution of their geochemical and physical environment. Within and around the EBS, radiation, temporary high temperature (>65 °C), temporary high pH (>11) and spatial

confinement are considered the most challenging restrictions for microbial life, although they should not be considered irreversible [41]. In fact, high temperatures and pH might even be optimal conditions for specialised groups of organisms (extremophiles) ([222]; [43]). As an example, methanogenesis is a reaction mostly performed by thermophilic micro-organisms thriving at temperatures beyond 45 °C or even beyond 80 °C (hyperthermophilic) [222]. Reactions that have been documented at such elevated temperatures are NO_3^- reduction, (autotrophic) SO_4^{2-} reduction, (autotrophic) acetogenesis, (autotrophic) methanogenesis and (autotrophic) Fe(III) reduction, thereby nearly covering all the above mentioned reactions [222].

An assessment of the estimated impact of microbial activity on the global gas balance and gas reactivity in deep subsurface nuclear waste repositories is at this stage a subject of continuing study. The short and long term impact of expected environmental stresses like radiation, heat, high pH and the transition of oxidising to reducing conditions on microbial community dynamics, on the expression of gas generation pathways and their impact on EBS, host rock and waste stability should be addressed. This information would not only prove highly valuable in terms of the assessment of in situ gas balance and reactivity, but will also help evaluate and interpret laboratory experimental results which are prone to bias by microbial activity.

5.5 Completeness

FORGE has provided confidence in the basic physical understanding and modelling approaches relevant to repository-derived gas that can be applied for different concepts – such understanding, and the derived component and system models, can be used in the safety case to address regulatory guidance. 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 but they were not identified as 'show-stoppers' in terms of adequate and appropriate consideration of gas in the safety case. Further targeted work (e.g. EC CAST[10] project; various studies investigating gas migration in bentonite; on-going national and international laboratory and URL studies) is being performed to consider such gas-related uncertainties. Deployment of an approach to managing uncertainty is a required component of any safety case, and is applicable to gas issues.

5.5.1 Output from the EC PAMINA project

The EC PAMINA project considered performance assessment methodologies in application to guide the development of the safety case. An area of study within PAMINA was repository-derived gas. The PAMINA project summary report [223] notes the following regarding gas; this text is reproduced here as – for points of commonality - it aligns with understanding derived from the FORGE project:

- *Generation of gas from the waste, generally hydrogen, by corrosion of steel containers and overpacks and by other means is accepted as an important issue that must be assessed in the safety case. Gas generation may affect repository performance by increasing pressure, which could affect*

[10] CArbon-14 Source Terms, scheduled to commence in FY13/14.

radionuclide transport in the near field and immediate geosphere. Pressure increase could also affect the behaviour of buffer and backfill materials and possibly impact on dissipation of heat from some wastes. Not least, the potential radiological impacts of radionuclide release in gaseous form need to be considered in a safety case. It is therefore essential to understand the balance between gas generation and gas migration.

- *Modelling of gas generation carried out within PAMINA suggests that in the disposal concepts currently proposed in participating countries, repository overpressurisation is not expected to be a cause for concern, owing to the likelihood that any gas pressure build up would dilate existing pathways in the near field and fracture systems in the immediate geosphere, allowing gas to migrate away from the repository. Radionuclide migration through the buffer and near-field is expected to be dominated by two-phase flow, for which suitable analysis tools exist. One such tool, TOUGH2, was used to investigate the potential impacts of hydrogen gas on the disposal concept proposed in Belgium, where large amounts of steel are to be used as waste containers. Again, overpressurisation was shown not to be a significant issue in either the buffer material or the near field. However, further effort is required in terms of model qualification and validation in order to reduce uncertainty.*
- *The details of gas migration were shown in PAMINA to be extremely site (and, therefore, geology) specific. Perhaps the most important of the assumptions affecting the migration behaviour of gas is the extent to which any free gas moving away from the repository will contact the groundwater within the rock volume represented by a grid block. In addition, the presence of low-permeability units in the geosphere may have a significant effect on gas migration. It is therefore important that the potential for gas transport through the host rock and overlying geological units is explicitly recognised in the development of a site-specific safety case, and that research, assessment and site characterisation studies develop an understanding of the gas transport properties of such strata in order to better understand how gas migration could occur, particularly in instances where the uncertainties could be significant to the safety case (e.g. where unfractured argillaceous rocks form part of the geological "barrier").*

6 FORGE PROJECT ACHIEVEMENTS AND RECOMMENDATIONS

This section presents key messages from the EC FORGE project. These document the achievements of the project over its duration, and are information guides / learning points that should be drawn on as recommendations to inform and direct any future gas study (national or international) in the context of the safety case for the geological disposal of radioactive waste.

Work undertaken in the FORGE project will benefit a range of customers, e.g. implementers, regulators, industry and academia, via the provision of new information and understanding into gas-relevant FEPs, including mechanisms governing gas generation and migration, for consideration in the safety case. 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.

6.1 EC FORGE project key messages

(a) WP2 Gas generation

- Experiments on carbon steel showed:
  - Initial corrosion rate in compacted bentonite in neutral pH disposal environments is greatly accelerated (up to tens of µm/a) 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.

(b) WP3 Engineered barrier systems

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 (e.g. when the gas pressure reaches the sample pressure, as seen in LASGIT).
- 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 CO_2 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 there 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.

(c) WP4 Disturbed host rock formations

- 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 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 small-scale variability of certain rock properties (strength, permeability) enabled models to better simulate gas flow localisation.

(d) WP5 Undisturbed host rock formations

- 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.

(e) WP1.2 Benchmark studies

- 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.

(f) WP1.3 Repository Scale Gas Migration Calculations

- 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 agreement between the modeling teams for the gas pressures variations but less accurate agreement for the 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.

(g) Two phase flow / pathway dilation 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, with permeability depending only on water saturation).
- For small scale experiments, 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, two-phase flow models can be implied to give a 'good' approximation of the experimental results. This issue requires further consideration, and related conclusions drawn to date could be due to e.g. less accurate measurements in situ, but also to homogenization effect at metre scale.

6.2 Application of EC FORGE project output: repository design

Safety-relevant gas-related issues need to be considered from the outset in developing a repository concept and in formulating an approach to a site-specific safety case, so that a consistent approach is adopted across all issues [5]. The generation, accumulation and migration of gas in a geological repository will vary with waste inventory and with repository concept. Applying adequate strategies to manage gas processes will limit their potential adverse impacts on the performance of the various barriers of the repository and, ultimately, on its long-term safety.

Different design strategies aiming to reach one or several of the following objectives may be adopted:

- Ensure that the release of any gases to the biosphere does not result in regulatory limits being exceeded;
- Prevent the degradation of the performance of repository barriers;
- Reduce uncertainties on factors controlling the generation and migration of gases;
- Limit adverse consequences of gas release in case of the variant human intrusion scenario.

The strategies to be followed are directly dependent on the waste inventory and related disposal concept, as well as on the boundary conditions associated with the host rock and its environment. These boundary conditions include the availability of water and the chemical conditions (controlling the corrosion processes and the free gas phase), gas entry pressure (controlling the gas accumulation and the subsequent pressurisation) and sealing capacities (controlling the resilience of the host-rock) [15].

With respect to repository-derived gas, the EC FORGE project has considered gas generation and gas migration, with migration concerning both clay-based and cement-based EBS materials, and disturbed and undisturbed host rock. Significant new numerical modelling has also been undertaken, at the cell, module and repository scale, to better understand how gas could interact with repository infrastructure (e.g. plugs and seals, interfaces) and how migration of waste-derived gas could occur over the post-closure period. Interaction with groundwater has been considered (both in the desaturation period occurring whilst the repository is open, and in the resaturation period after the repository is closed).

On the basis of understanding derived from FORGE output, the basis for the consideration of waste-derived gas in the repository has been enhanced, such that, going forward, repository design, operation, sealing and closure can more thoroughly account for the gas issue than has been the case in the past. This allows optimisation of the disposal inventory (bulk material and radioactive waste) and optimisation of repository management to be considered holistically from the perspective of repository-derived gas management, and allows for the treatment of gas such that compliance with relevant regulatory criteria can be demonstrated in the safety case.

6.3 Overall Key Messages from FORGE

Considering the output from the EC FORGE project as detailed in this report and in related references, the overall key message in relation to the updated treatment of gas generation and migration in the safety case are noted below. Progressing beyond FORGE, these key messages and the outcomes from the EC FORGE project now need consideration in safety case studies undertaken by national implementing organisations, to ensure the derived understanding is embedded and to ensure repository-derived gas is managed such that the safety case is compliant with national regulatory requirements.

- 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 managed prior to emplacement in the repository etc) and the safety functions required to be provided by complementary barriers (e.g. EBS, geology).

- 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.

- Based on studies undertaken in the EC FORGE project, and on input from complementary studies, we have enhanced our understanding of repository-derived gas in relation to a range of concepts for the geological disposal of radioactive waste. Such understanding provides a justification for increased confidence in analyses of the gas issue as undertaken within the safety case.

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