Deliverable 6.2:

State of the Art on Gas Transport in Clayey Materials – Update 2023

Work Package GAS

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Executive summary

Many countries have chosen to dispose of all or part of their radioactive waste in facilities constructed in deep stable geological formations. Geological disposal as a safe solution for the long-term management of radioactive waste is consistent with international recommendations and best practices. Owing to their excellent properties for the confinement of contaminants, clays are considered as potential host rocks for geological disposal in several countries in Europe. Clay-based materials are also expected to be used in engineered barriers in most geological repository concepts under development.

Considerable amounts of gas can be generated in a geological repository for radioactive waste. The largest fraction of the gas is expected to be hydrogen produced through the anaerobic corrosion of steel and reactive metals present in the waste, in their packaging and in the engineered barrier system (EBS). Radiolysis and the degradation of organics also produce gas. Even though the gas production processes are generally slow, it is important to verify that these will not be detrimental to the good functioning of the disposal system. The low permeability of clays, which is favorable to the containment function of a repository, also limits the evacuation of the generated gas. It is possible that gas could be produced at a faster rate than it can be removed through the engineered barrier components, resulting in the development of a pressurized gas phase within the repository. If the pressure of accumulated gas in the repository is too high, gas could then escape from the repository to the host rock by creating gas-specific pathways through the EBS and/or the host rock. In case the gas contains volatile radionuclides, its potential release to the biosphere is an area that needs specific considerations by the organizations responsible for developing a geological repository and therefore requires robust underpinning knowledge to be able to adequately consider it within a facility safety case.

To properly evaluate the impact of gas on the functioning of a deep geological repository, increasing the understanding of gas transport through low-permeability porous materials such as clayey materials was considered as a high priority in the EURAD Roadmap, within the geoscience theme. Consequently, a work package devoted to the mechanistic understanding of gas transport in clayey materials has been included in the first European Joint Programme on Radioactive Waste Management (EURAD) (2019-2024). The main objectives of the work package, called EURAD-GAS, were:

- To improve the mechanistic understanding of gas transport processes in natural and engineered clay materials, their couplings with the mechanical behavior and their impact on the properties of these materials.
- To evaluate the gas transport regimes that can be active at the scale of a geological disposal system and their potential impact on barrier integrity and repository performance.

Next to thematic technical and concluding reports, the list of deliverables of EURAD-GAS included two state-of-the-art reports on gas transport through clayey materials. The first one, called SOTA 1, written in the early stage of the project, provides an extended general overview of the state of knowledge with respect to the main RD&D (research, development and demonstration) topics of EURAD-GAS (Levasseur *et al.*, 2021).

This document is the second state-of-the-art report of EURAD-GAS. Building on SOTA 1, it provides a shared view of the EURAD-GAS partners on the current knowledge of gas transport through clayey materials, enriched by the knowledge gained within EURAD-GAS and beyond. It also shows how this knowledge is currently being used in the context of the development of geological disposal systems in several European countries. Experimental data and insights from modeling are provided mainly for three clayey host rocks (the Boom Clay, the Callovo-Oxfordian claystone and the Opalinus clay), and for three bentonites (the Wyoming bentonite MX-80, the FEBEX bentonite and the Czech bentonite BCV).

This document is primarily intended for end-users (RD&D managers, implementers and safety authorities). Starting with an overview of their main concerns regarding the generation and the transport of gas in clayey materials in the context of geological disposal of radioactive waste, it presents the shared understanding of the EURAD-GAS partners on gas transport processes and their controls in clayey materials. It then provides guidance on what could be the controls of gas and its impact at the repository scale. The document also identifies and formulates the remaining uncertainties, in their context and with transparency, in addition to this common view of gas aspects based on the knowledge gathered so far. Throughout the preparation of this report, the aim has been to enable end-users to easily assess the relevance of the collected knowledge and uncertainties about gas in their own programs. This will empower them to develop optimal strategies for making the best use of the available science base and for setting priorities for their own research and development efforts.

Overall, this state-of-the-art document reinforces the FORGE EC project's insight that gas is not a showstopper for the safety case, but a matter of managing uncertainties. It provides input to implementers that may inspire design measures to reduce the impact of gas on the disposal system and/or the uncertainties associated with gas flow through geological disposal systems. In particular, the authors suggest adopting a comprehensive design approach that considers the presence and effects of gas early on in any RD&D program for geological disposal of radioactive waste. Getting a first estimate of the expected gas volumes and rate of production should be a priority. Then, a good basis for the initial stages of development could be the available general knowledge of the mechanisms of gas transport in clay materials and the generic storyboard for its evacuation through a disposal system presented in this document and its references. In addition, it is recommended that an iterative design approach is put in place to address gas transport issues from the start, recognizing that adaptations can occur along the way. Indeed, uncertainty on the values of key properties that control gas transport can be large at the start of an RD&D program. Also, the passage from parameters derived from the characterization of gas transport through selected materials in laboratory or limited-scale in situ experiments to parameters that can be used for robust repository-scale evaluations requires a dialogue between experimentalists, modelers and repository designers. Indeed, gas transport strongly depends on the selection of EBS materials, and the conditions expected at the time of repository closure through the whole system. It is essential that gas transport models and parameters are consistent with these specific materials and conditions.

Contents

List of boxes

Eight text boxes are distributed throughout the document. Most of them illustrate results obtained in the framework of EURAD-GAS while others complement statements made in the text.

Acronyms

BC	Boom Clay formation (Belgium and the Netherlands) (a poorly indurated clay rock)
BCV	Czech bentonite
Aalto University	Aalto University (*Aalto-yliopisto / Aalto-universitetet*) (Finland)
ACED	RD&D work package of EURAD devoted to the assessment of chemical evolution of ILW and HLW disposal cells
Andra	National Agency for Radioactive Waste Management (*Agence Nationale pour la Gestion des Déchets Radioactifs*) (France)
Äspö HRL	Äspö hard rock laboratory in Sweden
ATLAS	Admissible thermal loading for argillaceous storage – *EC project (1992-2007)*
BGE	Federal Company for Radioactive Waste Disposal mbH (*Bundesgesellschaft für Endlagerung*) (Germany)
BGR	Federal Institute for Geosciences and Natural Resources (*Bundesanstalt für Geowissenschaften und Rohstoffe*) (Germany)
BGS	British Geological Survey (UKRI) (United Kingdom)
CEA	Alternative energies and atomic energy commission (*Commissariat à l'énergie atomique et aux énergies alternatives*) (France)
CIEMAT	Centre for Energy, Environment and Technology (*Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas*) (Spain)
CIMNE	International Centre for Numerical Methods in Engineering (Spain)
COVRA	Central Organisation for Radioactive Waste (*Centrale Organisatie Voor Radioactief Afval*) (the Netherlands)
COx	Callovo-Oxfordian claystone (France) (an indurated clay rock)
CTU	Czech Technical University (Czech Republic)
DECOVALEX	Development of coupled models and their validation against experiments – *international research and model comparison collaboration, initiated in 1992*
DONUT	RD&D work package of EURAD devoted to the development and improvement of numerical methods and tools for modeling coupled processes
EBS	Engineered barrier system
EC	European Commission
EDZ	Excavation damaged zone
ENRESA	National Radioactive Waste Company (*Empresa Nacional de Residuos Radiactivos*) (Spain)
EPFL	*Ecole Polytechnique Fédérale de Lausanne* (Switzerland)
EURAD	European joint programme on radioactive waste management – EC funded (2019–2024)
EURIDICE	European Underground Research Infrastructure for Disposal of nuclear waste in a Clay Environment (Belgium)
FEBEX	Full-scale engineered barriers experiment in crystalline host rock – EC project (1996-2004)
FEBEX bentonite	Spanish bentonite
FoCa7 bentonite	French bentonite (Fourges-Cahaignes clay)

FORGE EC project	Fate of repository gases. Investigation of process of gas generation and transport and their potential impact on a disposal system – *EC project (2009–2013)*
GDF	Geological disposal facility
GRIMSEL Test Site (GTS)	International underground research laboratory located in the Swiss Alps
GRS	*Gesellschaft für Anlagen- und Reaktorsicherheit* (Germany)
HADES URL	High-activity disposal experimental site – underground research laboratory in Mol (Belgium)
HITEC	RD&D work package of EURAD devoted to the study of the influence of temperature on clay-based material behavior
HLW	High-level waste
HTO	Tritiated water
ILW	Long-lived intermediate-level waste
IRSN	Institute for radiation protection and nuclear safety (*Institut de radioprotection et de sûreté nucléaire*) (France)
IUPAC	International Union of Pure and Applied Chemistry
L/ILW	Low- and intermediate-level waste
Lasgit	Large-scale gas injection test (full-scale demonstration project performed at the Äspö hard rock laboratory in Sweden)
LEI	Lithuanian Energy Institute (Lithuania)
MEGAS	Modeling and experiments on gas migration in repository host rocks (under the umbrella of the PEGASUS EC project) – *EC project (1992–1997)*
MHM URL	Meuse/Haute-Marne underground research laboratory in France
Mont Terri URL	Underground research laboratory in Switzerland
MX-80	Wyoming bentonite (USA)
Nagra	National Cooperative for the Disposal of Radioactive Waste (*Nationale Genossenschaft für die Lagerung radioaktiver Abfälle*) (Switzerland)
ONDRAF/NIRAS	Belgian Agency for Radioactive Waste and Enriched Fissile Materials (*Organisme national des déchets radioactifs et des matières fissiles enrichies / Nationale instelling voor radioactief afval en verrijkte Splijtstoffen)* (Belgium)
OPA	Opalinus clay formation (Switzerland) (an indurated clay rock)
PEGASUS	Project on the effects of gas in underground storage facilities for radioactive waste – *EC umbrella project (1991–1998)*
PSI	Paul Scherrer Institut (Switzerland)
RD&D	Research, development and demonstration
SCK CEN	Belgian Nuclear Research Centre (*Studiecentrum voor Kernenergie / Centre d'Étude de l'énergie Nucléaire*) (Belgium)
SF	Spent fuel
SOTA	State-of-the-art report
THM(C)	Thermo-hydro-mechanical(-chemical)
TH^{2}M	Two-phase hydro-mechanical model
TU Delft	Delft University of Technology (*Technische Universiteit Delft*) (the Netherlands)

UFZ	Helmholtz Centre for Environmental Research (*Helmholtz Zentrum für Umweltforschung*) (Germany)
ÚJV Řež	Nuclear Physics Institute (Czech Republic)
ULiège	University of Liège (Belgium)
UPC	Technical University of Catalonia (*Universitat Politècnica de Catalunya*) (Spain)
URL	Underground research laboratory
WMO	Waste management organization

1. Introduction

Many countries have chosen to dispose of all or part of their radioactive waste in facilities constructed in stable, deep geological formations. Geological disposal as a safe solution for the long-term management of radioactive waste is consistent with international recommendations and best practices. Owing to their excellent properties for the confinement of contaminants, clays are considered as potential host rocks for geological disposal in several countries in Europe. Clay-based materials are also expected to be used in engineered barriers in many geological repository concepts under development.

Considerable amounts of gas can be generated in a geological repository for radioactive waste. The largest fraction of the gas is expected to be hydrogen produced through the anaerobic corrosion of steel and reactive metals present in the waste, in their packaging and in the engineered barrier system (EBS). Radiolysis and the degradation of organics also produce gas. Even though the gas production processes are generally slow, it is important to verify that these will not be detrimental to the good functioning of the disposal system. The low permeability of clays, which is favorable to the containment function of a repository, also limits the evacuation of the generated gas. It is possible that gas could be produced at a faster rate than it can be removed through the engineered barrier components, resulting in the development of a pressurized gas phase within the repository. If the pressure of accumulated gas in the repository is too high, gas could then escape from the repository to the host rock by creating gas-specific pathways through the EBS and/or the host rock. In case the gas contains volatile radionuclides, its potential release to the biosphere is an area that needs specific considerations by the organizations in charge of developing a geological repository and therefore requires robust underpinning knowledge.

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This document is the second state-of-the-art report of the project EURAD-GAS. Building on SOTA 1, enriched by the knowledge gained within EURAD-GAS and beyond since its publication, this new SOTA provides a concise and integrated overview of the current state of knowledge on gas transport processes. It is mainly addressed to stakeholders such as RD&D managers. It presents the common understanding of the current state of the art of gas transport in clayey materials, shared by the organizations involved in EURAD-GAS. It provides the keys for transferring knowledge gained from laboratory and in situ experiments to configurations that are commonly found in current repository designs. It highlights the potential consequences of gas transport on barrier integrity and repository performance and lists the remaining uncertainties and knowledge gaps, with some recommendations for early-stage programs.

This SOTA 2 contains six chapters. After this introduction, Chapter 2 sets the scene with an overview of geological disposal and worldwide implementation. It then depicts the end-users' interests and needs regarding gas in this context. Chapter 3 first describes how clayey materials are used in geological disposal systems, either as potential host rocks or as engineered barriers. It then presents the clayey materials considered in this document and their properties. It concludes with the common features of repository concepts for disposal in clay host rocks. Chapter 4 reviews the scientific basis for understanding and analyzing the various gas transport mechanisms that may prevail in clayey host rocks and engineered barriers. For each gas transport mechanism, a concise state of knowledge is proposed, together with a common view of the associated uncertainties and knowledge gaps. Finally, Chapter 5 presents the current understanding of the impacts of gas on a disposal system at different scales. It begins with an integrated review of the individual transport mechanisms. The impact of gas transport on long-term barrier integrity is then discussed. Next, a generic conceptualization of gas transport at repository scale is proposed. It concludes with a repository-scale assessment and expert judgment. Chapter 6 ends this state-of-the-art report by presenting concluding remarks, with some recommendations to stakeholders. Eight text boxes, which are distributed throughout the text, complete the document. Most of them illustrate results obtained in the framework of EURAD-GAS while others complement statements made in the text.

2. Interests and needs of end-users regarding gas in the context of geological disposal

Chapter 2 sets the scene with an overview of geological disposal and worldwide implementation (Section 2.1). It then depicts the end-users' interests and needs regarding gas in this context (Section 2.2).

2.1 Overview of geological disposal and worldwide implementation

Geological disposal involves placing disposal radioactive waste packages deep underground in stable geological formations, to protect people and the environment from harmful levels of radioactivity. Geological disposal as a safe solution for the long-term management of radioactive waste is in line with international recommendations and practices.

The disposal system consists of packaged radioactive waste, surrounded by the multiple barriers of the disposal facility, constructed in a suitable host rock. The multi-barrier approach, which includes both engineered and natural barriers, ensures that the disposal system is not dependent on any single barrier. The disposal system isolates the radioactive waste far away from people and the surface environment and contains it there while it naturally decays back to less harmful levels (Figure 2-1).

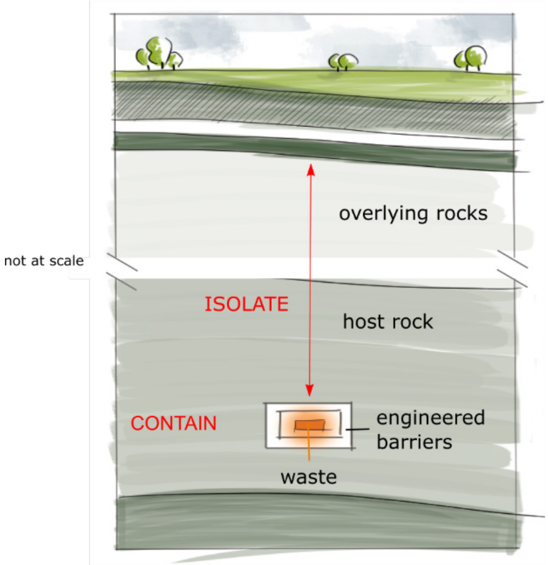

Figure 2-1 – Illustrative diagram of geological disposal. It provides protection of people and the environment without human interventions being necessary once the facility is closed (adapted from ONDRAF/NIRAS).

Geological disposal systems are designed to be passively safe once closed, which means their safety does not depend on human maintenance.

The safety of a disposal system is assessed and demonstrated by iterative safety cases of increasing detail as a GDF programme progresses (e.g., through early siting to site selection, construction license application, operational license application and eventually closure of the facility and delicensing of the surface site). The NEA (2013) defines a safety case as "*a formal compilation of evidence, analyses and arguments that quantify and substantiate a claim that the repository will be safe.*" IAEA (2022) precises that *"a safety case may relate to a given stage of development. In such cases, the safety case should* *acknowledge the existence of any unresolved issues and should provide guidance for work to resolve these issues in future development stages."*

Many countries have chosen to dispose of all or part of their radioactive waste in a GDF. Worldwide, GDFs are at different stages of development, as reported hereafter (Table 2-1). As of 2022, at least two GDFs are operational: in the United States (for military transuranic waste) since 1999 and in Hungary (for low- and intermediate-level waste) since 2005. Two GDFs are in construction: one in Finland for spent fuel and one in Germany for non-heat-emitting waste. In Sweden, following the Government's decision to allow the construction of a GDF for spent fuel, the licensing process continues. France, Switzerland, China and Russia have chosen the site of their future GDFs. The site selection is well advanced in Canada (for spent fuel). Siting is underway in Germany and the United Kingdom, without prejudging the type of host rock (both countries perform RD&D on several types of host rocks).

Table 2-1 – Status of geological disposal projects in the most advanced countries and EURAD-GAS members.

Country	Waste type *	Site and selection year	Host rock and depth (m)	Authorization of construction (year)	Start of the operational period
GDF operational					
Hungary	L/ILW	Bátaapáti (2005)	Crystalline rock (200 – 250)	2008	2012
United States	ILW (military)	Carlsbad, New Mexico (1979)	Salt rock (650)	1980	1999
GDF in construction					
Germany	Non-heat generating waste	Konrad (1976)	Clay under crystalline rock (800 – 1300)	2002 and confirmed in 2007	Planned for 2028
Finland	HLW	Eurajoki (2001)	Crystalline rock (400 – 450)	2016	Planned for 2024
Site chosen					
China	HLW	Xinchang (Beishan) (2016)	Crystalline rock (560)		Planned for 2050
France	ILW and HLW	Around MHM URL (2009)	Clay (500)	Submitted in 2023	Planned for 2027 (pilot phase)
Russia	ILW and HLW	Yeniseiskiy (2008)	Crystalline rock (450 – 525)		Planned around 2035
Sweden	HLW	Forsmark (2009)	Crystalline rock (500)	2022	Planned for 2030
Switzerland	All	Nördlich Lägern (2022)	Clay (900)	Expected around 2030	Planned around 2050 (L/ILW) and 2060 (HLW)
Siting in progress					
Canada	HLW	Ongoing selection (planned for 2024)		Expected in 2032	Planned between 2040 and 2045
Czech Republic	ILW and HLW	Ongoing selection (planned for 2030)	Crystalline rock	Expected around 2050	Planned for 2065
Germany	Heat generating waste	Ongoing selection (planned for 2031)			Planned for 2050
Hungary	ILW and HLW	Ongoing selection (planned for 2030)	Clay		Planned for 2064

Country	Waste type *	Site and selection year	Host rock and depth (m)	Authorization of construction (year)	Start of the operational period
Lithuania	ILW and HLW	Ongoing selection (planned for 2031)			Planned for 2068
United Kingdom	ILW and HLW	Ongoing selection	** (200 – 1000 m)	Planned for around 2040s	Planned in the 2050s'
Decision in favor of geological disposal					
Belgium	ILW and HLW			Not before 2050	
Netherlands	All		Salt rock or clay		Planned after 2100

* L/ILW: low- and intermediate-level waste, ILW long-lived intermediate-level waste, HLW: high-level waste.
** Open to all host rock types, but currently working with communities that have clays and mudstones (interbedded with evaporite).

2.2 Interests and needs of end-users regarding gas

Gas generation and transport in GDFs is an important issue for geological disposal for all waste management organizations (WMO). The issues to be addressed in a safety case depend on the repository concepts developed by each WMO in the specific context of its national program. While these contexts may significantly differ (in wastes to be disposed of, regulatory context, program maturity or stage, geological setting, disposal concept, etc.), several concerns and needs are shared by many end-users. These are summarized in the following sections.

2.2.1 Interests of end-users regarding gas in the context of geological disposal

Considerable amounts of gas can be produced in geological disposal facilities through a number of processes such as the corrosion of metals and the radiolytic, chemical and/or microbial degradation of the organics present in the repository (Norris, 2013). Gas produced within a geological repository after closure would be mainly hydrogen and methane. Although the production of gas is expected to be slow, spanning over centuries, millennia or more, the total quantities of gas involved are such that four concerns often arise:

- The potential for over pressurization and damage if gas would be produced at a faster rate than it can diffuse out of the multi-barrier system.
- The potential for release of volatile radionuclides to the biosphere.
- The potential for accelerated gas-driven expulsion of water containing dissolved radionuclides to the biosphere.
- The risk associated to the accumulation and/or sudden release of flammable gas.

There is therefore a risk that gas could undermine the safety of the repository if not dealt with appropriately. Addressing these concerns requires in the first instance adequate understanding of gas production and evacuation mechanisms. The focus of EURAD-GAS and of this document is on the mechanisms controlling the evacuation of the gas through clay barriers and the consequences of these mechanisms on the performance of these barriers. Gas generation processes and gas consuming reactions are out of scope of EURAD-GAS and so of this document. For more information on gas generation, reader can refer to Norris (2013), Nagra (2016), Neeft (2018) or Small (2019).

2.2.2 Shared needs of end-users regarding gas evacuation

To evaluate the capacity of a system for gas evacuation and any associated impacts to system performance, the transport mechanisms of the engineered and geological components of the multi-barrier system need to be identified and understood. Hence, all end-users who consider clayey materials for their GDF share the same need of knowledge of gas transport mechanisms in clayey materials and in particular,

- How these mechanisms depend on the conditions to which these materials are subjected, primarily mechanical stresses and fluid pressures.
- What are the relevant material and fluid properties controlling these mechanisms.
- How to characterize the material properties, accounting for the fact that some of these might well be affected by the passage of gas.
- In addition, possible physicochemical gas retention and (microbial) conversion processes are also considered by some end-users[1].

The understanding of gas transport mechanisms is based on laboratory experiments, supported by the development of process-level models. Laboratory experiments may focus on a particular transport mechanism in well-controlled conditions or on the transition between different mechanisms. Different process-level models may represent phenomena from the microscopic (pore size or below) to the macroscopic scale (homogenization). These models allow to test assumptions made on the actual mechanisms that govern gas transport and thereby refine their understanding.

Chapter 4 gives an overview of the scientific bases for understanding and analyzing the different gas transport mechanisms that may prevail in clayey host rocks and EBS components. For each gas transport mechanism, a concise state of knowledge is proposed, together with a shared view of EURAD-GAS partners about the associated uncertainties and knowledge gaps. In complement, the reader is also referred to SOTA 1 for the finer details of each gas transport mechanism and its controls (Levasseur *et al.*, 2021, Chapters 2 and 3).

2.2.3 Shared needs of end-users regarding gas impacts on barrier performance

From a safety case perspective, the effects of gas belong to the category of the “Repository-Induced Effects.” The two main causes of concern to the end-users are the potential for a loss of mechanical integrity of the engineered and geological barriers and a loss of performance with respect to the confinement of radionuclides. Hence, the processes associated to gas transport need to be examined again, this time to evaluate under which conditions gas accumulation, pressure build-up, and gas transport via all possible mechanisms may perturb the clayey materials and to which extent. If perturbations are expected, the consequences on the barrier properties during and after the passage of gas need to be characterized.

The involved processes depend not only on the type of barrier material and its fabric (granular/compacted bentonite, poorly indurated clay, indurated clay), but also on the repository concept, the site-specific and the design-specific conditions controlling the evolution of the repository

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[1] Gas conversion processes are out of scope of EURAD-GAS and this report.

near field[2]. For example, in repository concepts that are based on a bentonite buffer, gas-induced impacts in the bentonite buffer could hypothetically manifest in the mobilization of bentonite colloids, leading to a loss of swelling pressure, increase in buffer water permeability and, potentially, increased groundwater flow rates around the disposal waste packages. Experimental evidence for gas-induced mobilization of bentonite colloids has indeed been gained in laboratory experiments (Manca, 2016) and in situ experiments (GAST Experiment Grimsel), but unfavorable effects on swelling pressure / hydraulic conductivity were however not observed. In the near field of repositories in clay formations, re-activation of existing fractures or development of microfractures may be expected when gas invades the host rock, potentially facilitating transport of dissolved and volatile radionuclides along the newly created pathways. Next to processes associated to the possible creation of damage in clay barriers, self-sealing capacity of clays is also considered. The self-sealing capacity characterizes the ability of clays to restore hydraulic properties. There is a need to evaluate to which extent this capacity is preserved after the passage of gas.

For building the line of arguments toward a robust safety case, multiple lines of convincing evidence are required, which demonstrate the resilience of clay barriers and the efficiency of self-sealing for the conditions that could arise in a repository. This involves performing experiments at different scales in the laboratory and in situ in underground research laboratories, looking at natural analog examples and also developing and validating conceptual models that might then be used to extrapolate to the time and spatial scales of relevance to the post-closure performance of a repository.

2.2.4 The challenge of upscaling

Numerical modeling plays an important role in safety cases, e.g., in identifying key parameters based on experiments, in testing hypotheses about the involved processes and, because the spatial scale and especially the time scale associated with the post-closure functioning of a repository are not always accessible to experimentalists, in assessing the future evolution of the disposal system. Modeling is thus not only essential for fundamental process understanding but also for the transferability and upscaling of the scientific bases to the analysis of the system as a whole.

While certain processes are easily transferable from one scale to another (e.g., solute diffusion or heat conduction), the upscaling of gas transport is less straightforward. Indeed, some gas transport mechanisms (e.g., through the creation of discontinuities) are local by nature and the interpretation of experiments may in consequence be linked to the resolution of the observations. In any case, an appropriate upscaling remains a need for end-users.

The use of repository-scale models, which would explicitly take into account all couplings with the mechanical behavior of the barriers and the development of local gas pathways through clayey materials, is not currently being considered by any WMO due to its complexity and the fact that this capability (while desirable) is not necessary in order to underpin a robust design and safety case. Therefore, end-users are exploring alternative approaches to repository-scale modeling. They rely on gas transport storyboards to estimate the maximum gas pressures within the repository and to assess the transport pathways and the amount of radionuclides released. The most widely adopted approaches involve using modular representations of the various components of the disposal system, along with ad hoc gas transport models for each component. Results from modeling one component are used as boundary conditions for modeling the next. The simplified representations of the components, which may be based on the results of component-scale coupled models to improve understanding of the finer

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[2] "Near field" defined by IAEA as "the excavated area of a repository near or in contact with the waste packages, including filling or sealing materials, and those parts of the host medium/rock whose characteristics have been or could be altered by the repository or its content" (IAEA, 2022).

details of the hydro-mechanical behavior of the system, are assembled to derive an analysis of the functioning of the overall system.

In any case, predictions of gas transport at the scale of a disposal system could be subject to significant uncertainties, depending on the reproducibility of observations from laboratory and in situ experiments. However, rather than accurate predictions, an envelope of the possible effects of gas may be sufficient to meet the needs of the end-users for the purpose of designing a GDF and evaluating its performance and safety. This implies the definition of design requirements and safety targets associated with the transport of gas. In the end, however, performing assessments in support of a safety case is the responsibility of the end-users, with the support of experts, as this will depend to a large extent on the waste inventory, the repository concepts and site(s), and the national regulations.

3. Clayey materials and repository concepts

Chapter 3 first describes how clayey materials are used in geological disposal systems, either as potential host rocks or as engineered barriers (Section 3.1). It then presents the clayey materials considered in this document and their properties. It concludes with the common features of repository concepts for disposal in clay host rocks (Section 3.2).

3.1 Clayey materials in geological disposal systems

The geological disposal systems in operation or under development in many countries rely on clayey materials, either as host rock (Section 3.1.1) and/or as parts of the EBS (Section 3.1.2). Whatever their use, clayey materials present various characteristics that make them excellent barriers against the migration of radionuclides and chemical contaminants toward the surface environment. The deep clayey layers selected or considered as potential host rocks are in addition sufficiently thick and hydro-geologically, geochemically and mechanically stable over geological timescales, i.e., millions of years. While Sections 3.1.1 and 3.1.2 are generic, Sections 3.1.3 and 3.1.4 respectively present the clayey materials studied in the frame of EURAD-GAS and their main properties.

3.1.1 Clayey materials as potential host rocks

The research and development studies performed over several decades have highlighted the favorable features and properties of carefully selected clays studied as potential host rocks:

- Low permeability: thanks to their small pore size, there is almost no water movement in clays. Radionuclide and contaminant transport through clays is delayed relative to more permeable formations. In addition, because of this limited water movement, transport in clays is typically diffusive; species migrate primarily under the influence of their concentration gradient, and very little under the influence of pore water movement.

- Retention capacity: through various (ad)sorption processes, clays have a strong retention capacity for many radionuclides and chemical contaminants. Their migration through clays is thus considerably delayed.

- Buffer capacity: clays display a significant buffer effect regarding chemical perturbations. The thickness of the clay that is chemically perturbed by the disposal facility is therefore very limited.

- Self-sealing capacity[3]: thanks to their swelling and creep capacities, clays show a high capacity for self-sealing of discontinuities. Any fractures that occur, in particular those created by excavation activities, are typically observed to close over relatively short timescales (Figure 3-1). This process is known to be faster for poorly indurated clays than for indurated clays.

- Stability: carefully selected clay host rocks –and therefore their favorable properties– have remained unchanged over millions of years. The migration of natural chemical species through these clay host rocks has remained diffusive during at least the last million years.

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[3] Self-sealing should not be confused with self-healing. Where there has been a fracture, changes in the structure of the clay may persist and lead to permanent changes in the local hydro-mechanical properties at or near the “former” fracture.

- Vertical homogeneity: radionuclide and chemical contaminant transport properties are often very homogeneous almost throughout the entire thickness of the selected clay host rocks.
- Lateral continuity: clays are often present within simple geological structures, with a significant lateral continuity, which facilitates their large-scale characterization.

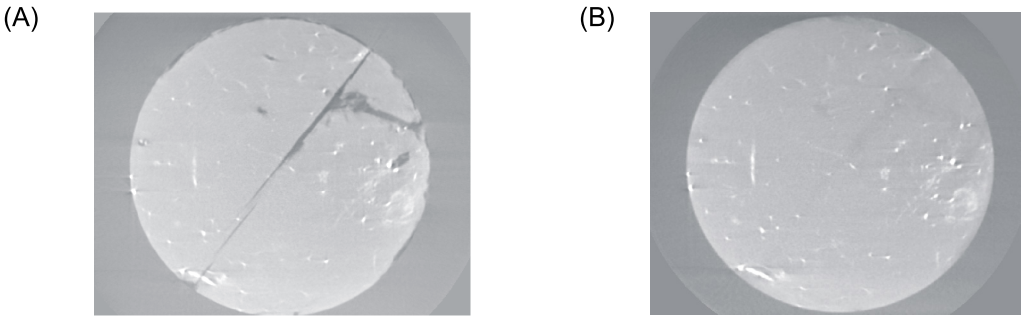

Figure 3-1 – Illustration of the self-sealing capacity of Boom Clay. (A) Clay sample in which a fracture has been induced. (B) The same sample four hours after hydraulic saturation in permeameter cell: the fracture has been sealed (Bastiaens et al., 2007; Van Geet et al., 2008).

3.1.2 Clayey materials as engineered barriers

The favorable properties of clay (such as its low permeability and swelling capacity, contaminant retention capacity and mineralogical stability in the conditions of geological disposal systems) make it a material of choice for engineered barriers. The engineered barrier systems of many geological disposal concepts (Table 3-1) contain bentonite, a naturally occurring absorbent swelling clayey material with a low permeability. Depending on the specifications required for use in a GDF, bentonite is processed in different forms and compacted to various densities (Figures 3-2 and 3-3).

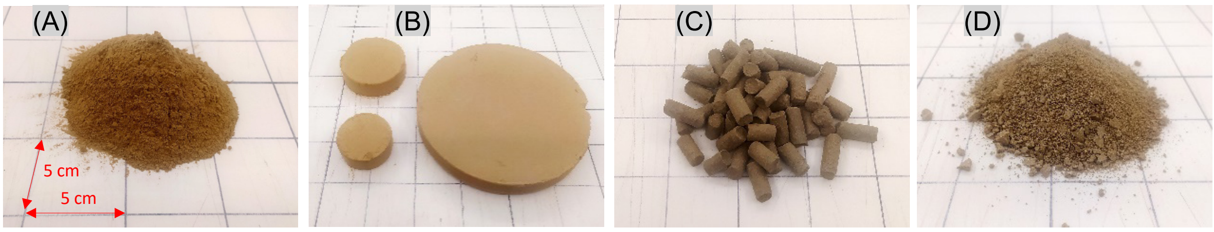

Figure 3-2 – Different shapes of Czech bentonite BCV prepared for testing. (A) Powder. (B) Compacted blocks. (C) Pellets. (D) Crushed pellets.

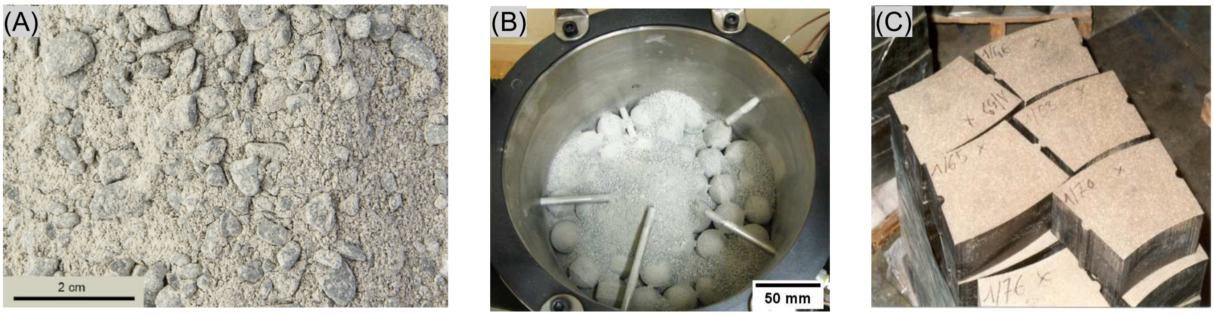

Figure 3-3 – Different shapes of bentonites tested for use in GDF. (A) Highly compacted granulated bentonite mix produced for the "Full-scale emplacement experiment (FE)" (Müller et al., 2017). (B) Mix of 32 mm pellets and crushed pellets (MX-80) (Bernachy-Barbe et al., 2020). (C) Blocks (FoCa7 bentonite) (Gatabin et al., 2016).

Clay is –or is planned to be– mainly used as:

- buffer material: the empty space between the disposal package and the host rock is filled with clay (Figure 3-4 A).
- backfill material: clay is used to fill excavated spaces (placement rooms, access ways), sometimes in combination with other materials (Figure 3-4 B).
- sealing material: clay, sometimes in combination with other materials, is used to isolate parts of the disposal facility. Seals are works of limited dimensions with specific purpose placed at key locations of the disposal facility, e.g., to limit water flow within the underground facilities (Figure 3-5).

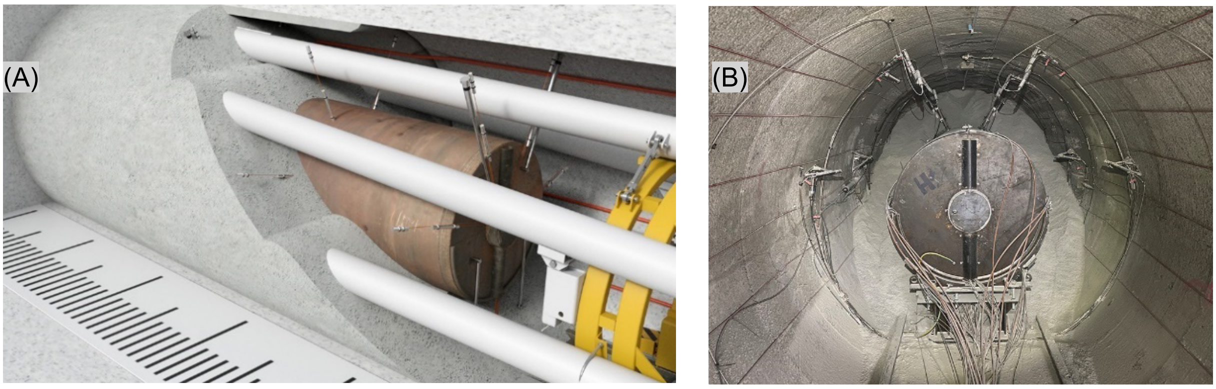

Figure 3-4 – Examples of uses of bentonite in the HotBENT experiment in Grimsel (granite host rock), which aims to assess high temperature effects on bentonite buffers. (A) Schematic representation. (B) HotBENT heater instrumented and partially backfilled with granular bentonite material[4].

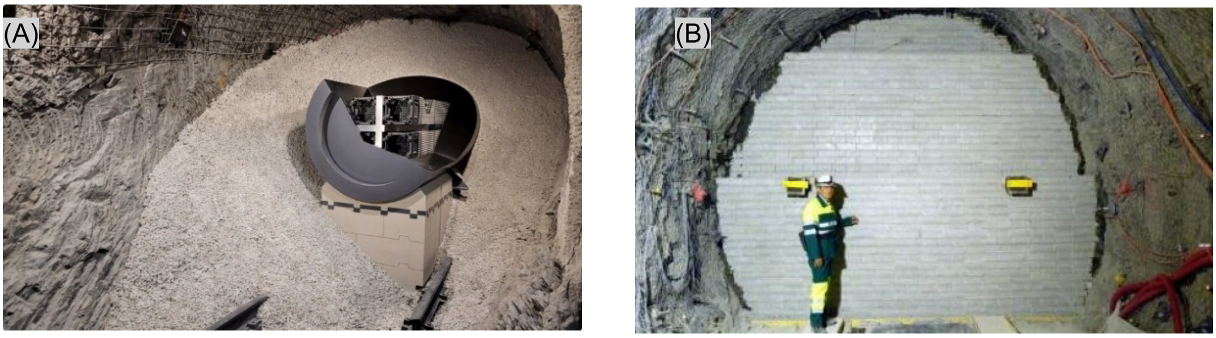

Figure 3-5 – Examples of uses of bentonite in GDF concepts under development (A) Mock-up Nagra high-level waste disposal in a clay formation (buffer with bentonite blocks and granules) (courtesy of Nagra, copyright Comet GmbH Weisslingen, CH). (B) Sealing wall made with bentonite blocks in a clay formation (GES gallery in MHM URL) (de la Vaissière, Conil et al., 2014)[5].

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[4] https://www.grimsel.com/gts-projects/hotbent-high-temperature-effects-on-bentonite-buffers/hotbent-site-preparation-work. Last accessed 2023-10-05.

[5] de la Vaissière, Conil *et al.* (2014) describes the test. Picture from web site *https://www.andra.fr/experimentation-les-premieres-briques-du-scellement-de-galerie*. Last accessed: 2024-02-12.

3.1.3 Clayey materials considered in this document

This document covers gas transport through three clay formations, defined as sedimentary rocks with a fine grain size, a low to moderate mechanical strength, a high clay content, and a low permeability:

- **Opalinus clay formation (OPA).** Switzerland has chosen OPA as a host rock to dispose of all its radioactive waste. The OPA formation has been extensively studied at the Mont Terri underground research laboratory (URL), Switzerland (Figure 3-6 A).
- **Callovo-Oxfordian claystone (COx).** France has selected COx as a host rock to dispose of its high-level and long-lived intermediate-level radioactive waste. The COx formation has been extensively studied at the Meuse/Haute-Marne URL, Bure, France.
- **Boom Clay formation (BC)** studied by Belgium as potential host rock for the disposal of its high-level and long-lived intermediate-level radioactive waste, together with Ypresian clays (both clays are poorly indurated rocks), and by the Netherlands, for the disposal of all types of radioactive waste. The BC formation has been extensively studied at the HADES URL, Mol, Belgium (Figure 3-6 B).

Figure 3-6 – Different types of clayey rocks. (A) Opalinus clay, survey during excavation of a gallery/drift for the Full-Scale Emplacement Experiment (Mont Terri URL, Switzerland) (courtesy of Nagra, @ Comet GmbH Weisslingen, CH). (B) Boom Clay (Belgium) (source: EURIDICE).

The engineered barrier systems of most disposal concepts developed worldwide contain clayey materials, and more specifically bentonite (Table 3-1). This document focuses on gas transport through three types of bentonites and their mixture with sand:

- **Wyoming bentonite (MX-80)**, extracted from Wyoming (USA). It has been selected in many disposal concepts as buffer and sealing material.
- **FEBEX bentonite**, extracted from the Cortijo de Archidona deposit in Spain. Its name comes from a 900-tons batch of bentonite processed in 1996 for the Full-scale Engineered Barrier Experiment (FEBEX) project (Huertas *et al.*, 2000). The FEBEX bentonite is considered as buffer in the Spanish concept for a GDF in crystalline rock (ENRESA, 2006).
- **Czech bentonite BCV**, extracted from the Černý vrch deposit in Czech Republic. The Czech reference geological concept includes use of this bentonite as buffer and backfill.
- **Sand/bentonite mixtures (S/B)** studied in the frame of EURAD-GAS contain quartz sand and MX-80 bentonite in proportion 80/20 and 60/40 in dry mass. Compared to pure bentonite, sand/bentonite mixtures allow to control/limit the swelling pressure, present a higher thermal

conductivity (at equivalent dry densities) and a better mechanical stability, and also facilitate gas transport while still limiting water transport (Box 1, page 14).

Table 3-1 summarizes the functions of clayey materials in the geological disposal concepts developed by members of EURAD-GAS and detailed in SOTA 1 (Levasseur *et al.*, 2021, Section 4.1).

Table 3-1 – Functions of clayey materials (in bold) currently foreseen in geological disposal concepts detailed in SOTA 1 (Section 4.1), as a function of the stage of development of the GDF.

Country	**Host rock (chosen or studied)**	**Waste group**	**Parts of the engineered barrier system**		
			Buffer/local backfill	**Mass backfill**	**Seals**
Site chosen					
France	**COx**	HLW	–	**Crushed COx + bentonite** and sand	**Bentonite; Sand/bentonite**
	COx	ILW	CM *	**Crushed COx + bentonite** and sand	**Bentonite; Sand/bentonite**
Switzerland	**OPA**	SF/HLW/ILW	**Bentonite**	Sand/**bentonite or crushed OPA**	**Bentonite; Sand/bentonite**
	OPA	L/ILW	High porosity mortar	Sand/**bentonite or crushed OPA**	**Bentonite; Sand/bentonite**
Other countries (host rock not selected yet)					
Belgium	**Poorly indurated clay**	ILW	CM	CM	**Bentonite**
	Poorly indurated clay	HLW/SF	CM	CM	**Bentonite**
Netherlands	**Poorly indurated clay**	HLW and LILW	CM	**Host rock *****	**Host rock *****
United Kingdom	Crystalline rock	HLW	**Bentonite**	**Bentonite** or crushed rock	SC **; **Bentonite**
		ILW	CM	Crushed rock	SC; **Bentonite**
	Clay	HLW	**Bentonite**	**Crushed host rock**	SC; **Bentonite; Sand/bentonite**
		ILW	CM	**Crushed host rock**	SC; **Bentonite; Sand/bentonite**

* CM: cementitious materials
** SC: structural concrete
*** Reconstituted compacted excavated host rock, mixed with some cement if necessary

Box 1 – Clayey materials used for the EBS can be tailored to requirements.

Gas production associated with metal corrosion and degradation of organic material may give rise to significant gas overpressures in the backfilled and sealed underground structures of a deep geological repository. Gas-related design options have been developed, allowing for a controlled gas accumulation and gas transfer along the underground structures at moderate gas overpressures (e.g., Nagra (2008)). Gas permeable gallery seals are known as a key element in gas-related repository optimization, exhibiting the favorable feature of low permeability to water, whereas the gas permeability is enhanced. High compressive strength of the sealing materials is required to ensure the mechanical integrity of the seals over long time periods.

Clayey backfill and sealing materials can be tailored to the design requirements by remolding and mixing with other geomaterials. Thus, design-specific adjustments of hydraulic conductivity, capillary pressure-saturation relationship, permeability-saturation relationship and thermal conductivity are obtained by appropriate mixing ratios of the materials and appropriate emplacement densities. Favorable chemical conditions are achieved by adequate mineralogical composition of the mixtures.

Comprehensive fundamental databases for the geotechnical characterization of sand/bentonite mixtures have been obtained in the past in various national programs e.g., (Graham *et al.*, 1997; Tashiro *et al.*, 1998; JAEA, 1999; Nagra, 2008; Manca, 2016). The characteristic relationship between the geotechnical properties and the bentonite content can be explained by the micro-fabric of the sand/bentonite mixture. For sand/bentonite mixtures with low bentonite content, the structural framework of the medium is determined by the sand because of the direct contact of the sand grains. A distinct microstructural variability is observed, which is expressed by the multi-modal pore size distribution of the material. The very fine bentonite particles (< 2 µm) are distributed in the void space between the sand grains; when water invades, they swell at these locations and the hydraulic conductivity of the sand/bentonite mixture is drastically reduced. As a result, sand/bentonite mixtures with low bentonite content ("grain supported") are characterized by a high compressive strength, a limited swelling capacity and a high sensitivity of the hydraulic conductivity to the bentonite/sand ratio (JAEA, 1999). With increasing bentonite content ("matrix supported") the geomechanical properties of the sand/bentonite mixture change significantly, because the bentonite not only occupies the pore space between the grains, but increasingly interrupts the contact between the sand grains. Thus, the swelling capacity of the material increases and the stiffness decreases. The hydraulic properties of the sand/bentonite mixture approach those of pure bentonite at same (bentonite) dry density. The mixing ratio between sand and bentonite determines not only the average hydraulic conductivity of the sand/bentonite mixture, but also the homogeneity and consequently the possibility of developing preferential flow paths.

Figure 3-7 shows the results of a series of permeameter experiments for three different sand/bentonite ratios (10%, 20%, 30% bentonite content) with different dry densities varying between 1.65 and 2.0 g/cm^3. It is shown that the permeabilities for bentonite contents of 10% vary strongly over a range of approximately 2 orders of magnitude and even for high dry density values between 1×10^{-10} m/s and 1×10^{-11} m/s are observed. For values of bentonite content of 20%, the range of variability corresponds to approximately one order of magnitude, whereas even smaller variations are observed for a bentonite content of 30%. For the entire spectrum of dry densities from 1.65 g/cm^3 to 2.0 g/cm^3, the hydraulic conductivity varies between 2×10^{-12} and 7×10^{-13} m/s, less than half an order of magnitude. Furthermore, mixtures with higher bentonite content allow higher densities to be reached, because through appropriate compaction measures, the fine particles of the bentonite part can be better distributed within the sand part; additionally, at such a high bentonite content the exact mixing and compaction procedures are less critical.

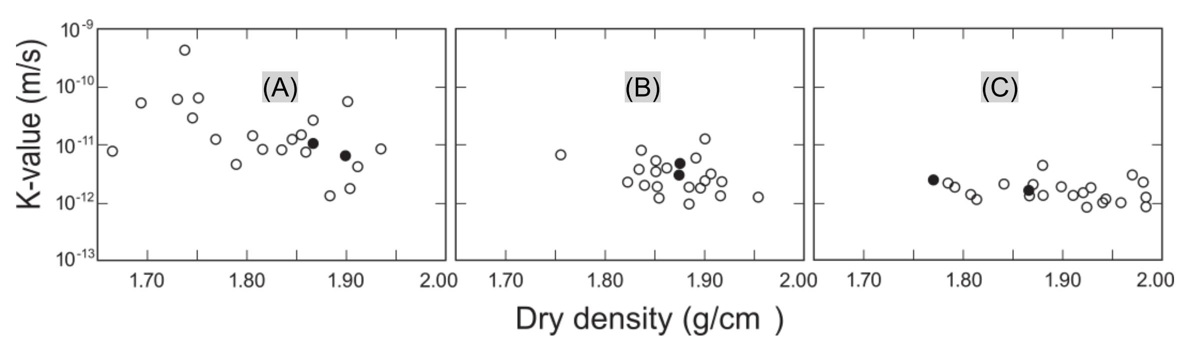

Figure 3-7 – Dependence of the hydraulic conductivity on the dry density of sand/bentonite mixtures for bentonite contents of 10% (A), 20% (B) and 30% (C) and different compaction methods ((Nagra, 2008); after (Tashiro et al., 1998)). Open symbols represent compaction by "combined roller" and full symbols represent compaction by vibratory plate compactor.

3.1.4 Properties of the studied clayey materials

Gas transport through clayey materials is a complex process that depends strongly on the coupling between the hydraulic and mechanical responses of these materials. Table 3-2 lists, for the three host rocks studied, some of the parameters that may influence the gas transport behavior in these materials (additional parameters are available in NEA (2022)). A more general overview of the geology, mineralogy and pore water composition of each of these formations is given in SOTA 1 (Levasseur *et al.*, 2021, Section 1.5).

Table 3-2 – Summary of basic parameters of the three host rocks studied in EURAD-GAS (References are from NEA (2022) unless otherwise noted).

Parameter [unit]	**Host rock – Best estimate value or range of values**		
	Boom Clay at HADES URL (Boerentang Member)	**COx at MHM URL (UA layer)**	**Opalinus clay at Mont Terri URL**
Type of clay	Poorly indurated	Indurated	Indurated
Formation			
Age [Ma]	32–28	163–158	172
Present burial depth (top) [m]	190	490	220
Over-consolidation ratio [–]	2.4	*n.d.*	4.0
Thickness [m]	102	140–160 ***	90–160
Mineralogical parameters			
Total clay minerals [wt%]	51.5	49	59
Quartz content [wt%]	34.3	18	15
Organic carbon [%]	1.0	1.0	0.7

Parameter [unit]	**Host rock – Best estimate value or range of values**		
	Boom Clay at HADES URL (Boerentang Member)	**COx at MHM URL (UA layer)**	**Opalinus clay at Mont Terri URL**
Petrophysical parameters			
Cation exchange capacity [meq/kg rock]	182	160	128
Bulk dry density [kg/m^3]	1 700	2 290 ± 40 ***	2 330
Grain density [kg/m^3]	2 695	2 710 ± 10 ***	2 710
Water content [wt%]	19.2	7.0	6.4
Physical porosity (based on water content) [%]	34–38 *****	15.9	13.7
Flow and solute transport parameters			
Hydraulic conductivity in situ [m/s]	4.7×10^{-12}	1.1×10^{-13} (⊥) *** 3.3×10^{-13} (//) ***	3.1×10^{-13}
Geochemical parameters			
Uniaxial compressive strength laboratory tests [MPa]	2.0 (⊥) – (//)	26 ± 6 (⊥) 21.6 ± 6 (//) ***	7.0–16.0 (⊥) 10.0–18.0 (//)
Young’s modulus static laboratory tests [MPa]	200 (⊥) 400 (//)	5 945 ± 2 500 (⊥) *** 11 428 ± 4 600 (//) ***	3 300 (⊥) 6 600 (//)
Young’s modulus static in situ tests [MPa]	700 (⊥) 1 400 (//) ****	*n.d.*	2 500 (⊥) 6 800 (//)
Swelling pressure laboratory tests [MPa]	1.0 *	*n.d.*	0.5 (⊥) 0.2 (//)
Cohesion laboratory tests [MPa]	0.3 (⊥) 0.3 (//)	*n.d.*	2–10 (⊥) 2–10 (//)
Internal friction angle laboratory tests [degrees]	18 **	*n.d.*	20 (⊥) 29 (//)
Dilation angle [degrees]	0–10 *	*n.d.*	*n.d.*

Note: *n.d.* no data available in (NEA, 2022).

* (Cui *et al.*, 2013)

** (Bernier *et al.* 2007)

*** (Yven *et al.*, 2007; Conil *et al.*, 2018; Andra, 2022)

**** from back-analysis of ATLAS III in situ heater test (Chen *et al.*, 2011)

***** (De Craen M., 2004)

Table 3-3 lists a few basic parameters of the three studied bentonites. Properties such as porosity depend on the degree of compaction. A more general description of these materials is given in SOTA 1 (Levasseur *et al.*, 2021, Section 1.5).

Table 3-3 – A few basic properties and mineralogical composition of the three bentonites studied in EURAD-GAS: MX-80 (Bradbury and Baeyens, 2002; Leupin et al., 2015; Seiphoori, 2016; Jenni et al., 2019); FEBEX (ENRESA, 2006); BCV (Červinka and Vašíček, 2018; Laufek et al., 2021; Šachlová et al., 2022; Svoboda et al., 2023).

Parameter / Composition [unit]	**Value (or range of values)**		
	MX-80 bentonite	**Czech bentonite BCV**	**FEBEX bentonite**
Mineralogical parameters			
Clay minerals [wt%]	80–85 montmorillonite 0.1–0.8 illite	65.7 ± 6.9 smectite 2.3 ± 2.2 kaolinite 2.3 ± 2.2 illite	92 ± 3 smectite (montmorillonite-illite mixed layer, with 10-15 wt% of illite layers)
Major Exchangeable cations	Na^+	Mg^{2+}/Ca^{2+}	$Ca^{2+}/Mg^{2+}/Na^{2+}$
Quartz content [wt%]	2.5–4.0	11.4	2 ± 1
Petrophysical parameters			
Cation exchange capacity [meq/100g]	76–88	63.7 ± 2.2	98 ± 2 (smectite)
Dry density [kg/m^3]	1 450 (granular) 1 800 (blocks)		1 600
Grain density [kg/m^3]	2 740–2 800	2 758 ± 17	2 700 ± 40
Water content [wt%]	20–30	10	13.7 ± 1.3
Porosity [%]	45–50 (granular) 35 (blocks)		
Flow and solute transport parameters			
Hydraulic conductivity [m/s]	1×10^{-13}–2×10^{-12} *	~1×10^{-13} **	~5×10^{-14} **
Geochemical parameters			
Plastic limit [%]	65	65	53 ± 3
Liquid limit [%]	420	140 ± 2%	102 ± 4

* depending on dry density, compaction, hydration, etc.

** for a dry density of 1.6 g/cm^3, saturated hydraulic conductivity at room temperature with deionized water as the percolating fluid

3.2 Common features of disposal concepts in a clay host rock

The concepts for a geological disposal facility built in a clay host rock for the disposal of radioactive waste developed in France, Switzerland, the United Kingdom, Belgium and the Netherlands have common features (as illustrated by the French concept in Figure 3-8):

- The spent fuel assemblies (SF) and the containers containing the vitrified high-level waste (VHLW) are packed into disposal waste packages in a surface facility and then transported to the disposal zone of the repository. The current reference SF and VHLW disposal waste package concept foresees the use of thick-walled carbon steel canisters (called overpacks) (as illustrated by the Belgian concept in Figure 3-9).
- The primary packages of long-lived intermediate-level waste are usually drums of diverse geometries and lengths. Disposal packages, manufactured in a surface facility, typically encapsulate several primary waste packages (as illustrated by the Swiss concept in Figure 3-10).
- GDF is built at a depth of several hundred meters. Disposal takes place in a single horizontal plane. Disposal gallery excavation techniques are selected to minimize formation disturbance and thus to limit the extent of the excavation damaged zone (EDZ).
- The walls of the excavated volumes are stabilized using different techniques to a degree depending on the host rock. In some clayey formations, the walls must be lined with concrete blocks to limit spontaneous convergence. In other host rocks, stabilization is not required everywhere and where it is necessary, it is provided by anchor bolts and welded mesh, with or without shotcrete.
- Access to the repository is provided, during construction and operation, by an access way (gallery/drift/ramp) and/or by shafts. The repository also contains ventilation and construction shafts, ventilation and operation galleries, and eventually a pilot facility and/or a test area.
- The waste is grouped by zone according to its main characteristics (radiological, thermal, etc.). The installation therefore often includes two distinct disposal zones, one for long-lived intermediate-level waste and the other for high-level waste, each consisting of disposal galleries parallel to each other. The two zones are several hundred meters apart, to avoid any potentially deleterious interaction (thermal, hydraulic, mechanical, chemical or gaseous) that could affect the system after its closure.
- The long-lived intermediate-level waste disposal packages can be stacked on top of and behind each other in large cross-sectional vaults (when the type of host rock permits) or placed one behind the other in smaller cylindrical galleries as they do not generate significant heat.
- The disposal area for high-level waste is designed to dissipate the heat they generate, to limit the temperature rise of the engineered and natural barriers to values that will not affect the confinement and isolation capacity of the disposal system and to limit the temperature rise in the aquifers to regulatory values. This constraint can be met in particular by adapting the duration of temporary storage, and therefore the duration of cooling at the surface, the thermal loading of high-level waste disposal packages, the spacing between disposal packages in the same disposal galleries and the spacing between disposal galleries.
- Once all the waste has been placed in the underground facility, the facility and its access ways are completely closed, i.e., backfilled and sealed, immediately or after a monitoring period, at once or in stages, but delayed closure must not jeopardize safety and security.
- The buffer/local backfill and mass backfill are either clayey and/or cementitious materials, of different porosities depending on the required properties.
- Engineered seals, made of clayey materials or of a sand/bentonite mixture, are installed during closure of disposal areas, access ways and shafts. These seals, depending on the disposal

concept, may need to allow gas to be released from closed disposal areas to prevent excess pressurization, but restrict the flow of water.

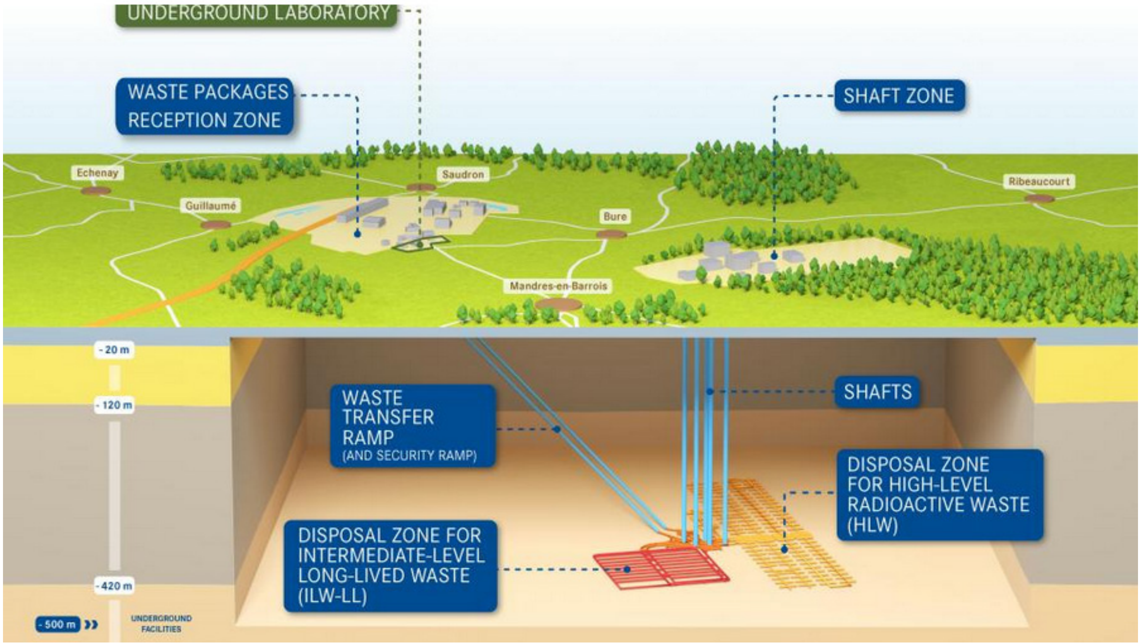

Figure 3-8 – Schematic representation of Cigeo, the French GDF concept[6].

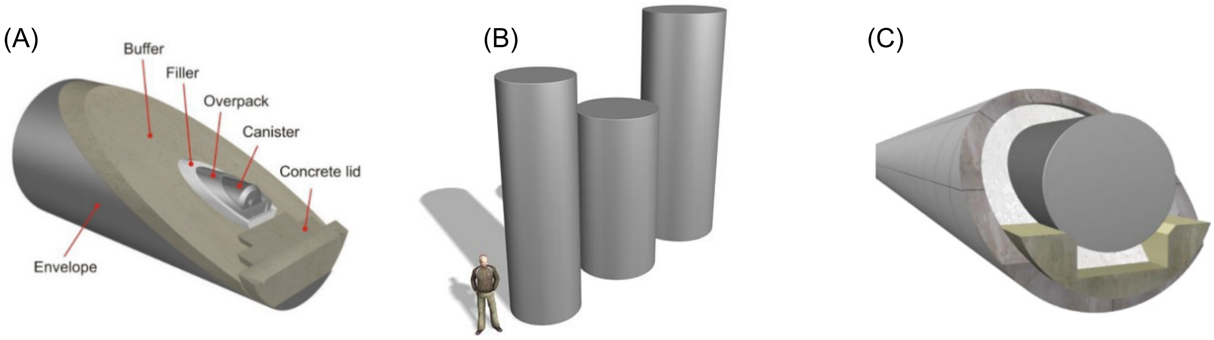

Figure 3-9 – (A) Disposal package for HLW in the Belgian concept (also called "supercontainer"). (B) The length of the supercontainers varies from 4 up to 6.2 meters to accommodate the lengths of the different types of HLW. (C) Disposal package disposed of in an underground gallery (ONDRAF/NIRAS, 2013).

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[6] https://international.andra.fr/projects/cigeo/cigeos-facilities-and-operation/project-siting-and-facilities-overview. Last accessed 2024-02-29.

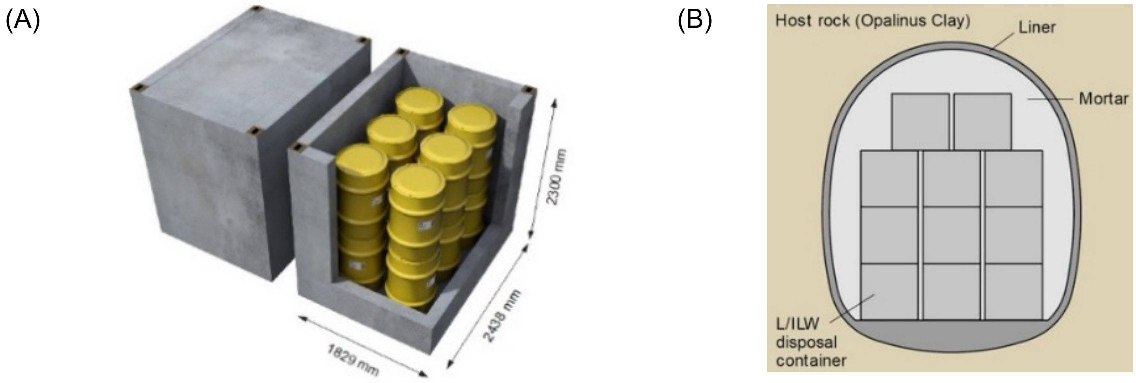

Figure 3-10 – Swiss concept. (A) Example of the packing of 200-liter drums of L/ILW (yellow) into one standard disposal package (gray). (B) Schematic layout of a L/ILW disposal cavern (Nagra, 2016).

4. Understanding of processes related to gas and remaining uncertainties and knowledge gaps

Different gas transport processes are expected in the post-closure phase of a geological disposal system once resaturated:

- Gas produced within the system can dissolve in the pore water and can then be transported by diffusion. The diffusion of dissolved gas may be retarded by sorption on the solid phase.
- If the gas production rate exceeds the rate at which gas can be dissolved and evacuated by diffusion, a free gas phase will develop. Three primary modes for gas transport as a separate phase are possible: (i) two-phase flow, in which gas displaces pore water to flow through the pre-existing porosity, (ii) the formation (and propagation) of dilatancy-controlled gas pathways –or dilatant pathways– and (iii) gas fracturing.

Gaining insight in which transport mechanisms prevail under which range of conditions and understanding (when relevant) how the coupling between pressure in the liquid and gas phases and the stresses in the solid phase control gas transport is fundamental to the analysis of the functioning of a geological disposal system with respect to the release of gas. A concise overview of the characteristics of each of these gas transport processes is provided in this chapter. For each, particular attention is given to the shared understanding and knowledge gaps. A more detailed analysis of each of these mechanisms is available in the literature, as documented in SOTA 1 (Levasseur *et al.*, 2021, Chapters 2 and 3).

4.1 Processes relevant to gas generation and consumption

There are several processes that can generate gas from waste packages and other materials in a GDF. There is 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 national program to national program (reflecting e.g., waste inventory differences). As detailed in (Norris, 2013), these mechanisms are:

- Anaerobic corrosion of steels and other metals, including the reactive metals e.g., aluminum, if present;
- Radiolysis of water and organic materials;
- Microbial degradation of organic materials and
- Denitrification processes of nitrate-bearing waste (e.g., bituminized waste).

As already stated in FORGE EC project (Norris, 2013), the dominance of these mechanisms will mean that the bulk gas produced within a geological repository under anoxic conditions would be mainly hydrogen and methane. 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.

The type, amount and rate of gas generated can affect its subsequent transport behavior. Gas generation will depend on the materials being degraded and on conditions that can evolve (e.g., pH, temperature, presence of sulfur ions that may enhance corrosion). Next to gas generation, some irreversible reactions, such as carbonation of cementitious materials and conversion of hydrogen gas to methane by microbes, can reduce the net quantity of gaseous molecules. Gas generation processes and gas consuming reactions are out of scope of EURAD-GAS and so of this document. It should nevertheless be mentioned that gas consumption reactions can have an effect on the timing and quantity of gas to be transported. For example, the conversion of hydrogen to methane in Boom Clay will produce four times fewer moles of a gas that dissolves better in water but that diffuses 3 to 4 times more slowly than hydrogen. For more information, reader can refer to Norris (2013), Nagra (2016), Neeft (2018) or Small (2019).

4.2 Processes relevant to gas transport

Gas initially present and gas generated within a geological repository will dissolve in the pore water. This dissolved gas will migrate by diffusion through the water and be displaced along with water in case of water movement. It can also react with or sorb onto some engineered materials or mineral phases of natural materials. If the gas generation rate exceeds the rate of evacuation of dissolved gas, a distinct gas phase may form. Gas can migrate as such through the disposal system by different mechanisms. The processes relevant to gas transport within a disposal system are outlined below, illustrated in Figure 4-1 and discussed in more detail later in this chapter (see also SOTA 1, Chapters 2 and 3 (Levasseur *et al.*, 2021) and Alkan and Müller (2006)):

- **Dissolution of gaseous molecules**. Where water is present, gaseous species will dissolve in the water until the concentration reaches the solubility limit under the prevailing temperature and pressure conditions.
- **Gas transport in solution**. Gas transport in solution involves two major gas transport mechanisms, diffusion and advection in the liquid phase (pore water) of the dissolved gas. Diffusion is driven by a concentration gradient in the pore water, whereas advective transport refers to the dissolved gas moving along with pore water, at the same velocity. In principle, the latter is limited in clayey materials as water usually hardly moves because of the very low hydraulic conductivity and low hydraulic gradient. In unsaturated environments, diffusion and advection of the gaseous species in the gas phase is another transport process to consider.
- **Gas sorption** may occur on engineered barrier materials, on their degradation products and on specific mineral phases within the clay host rock.
- Advective gas flow
    - **Visco-capillary two-phase flow.** If the production rate of gas is higher than the diffusional capacity of the clay, then a free gas phase will persist, or form, and its pressure will increase. In water-saturated clayey material, two-phase flow conditions are encountered when the gas pressure is sufficiently high driving gas to invade this material as a separate phase. To enter the porosity, gas has to displace water, overcoming viscous and capillary forces. The deformation behavior of the clayey material in response to gas invasion processes is considered to be reversible in the visco-capillary two-phase flow regime.
    - **Dilatancy-controlled gas pathways.** If the gas generation rate exceeds the capacity for pore water displacement by visco-capillary flow, or if the gas pressures required for the initiation of visco-capillary two-phase flow are too high, the gas will progressively deform the clay fabric to create additional, discrete, dilatancy-controlled gas pathways.
    - **Gas fracturing.** Gas fracturing may happen if the total accumulated gas volume in a given pore space is so high that transport by dilatancy-controlled gas pathways is not able to keep up with gas build-up and the gas pressure increase rate. The conditions necessary for gas fracturing to occur are dependent on the stress field which also controls the direction of fracturing.

Once the gas pressure is released, dilatancy-controlled gas pathways and gas fractures in clayey materials can close and seal. The efficiency of this process depends on the self-sealing capacity of the material and its hydro-mechanical properties and in situ conditions.

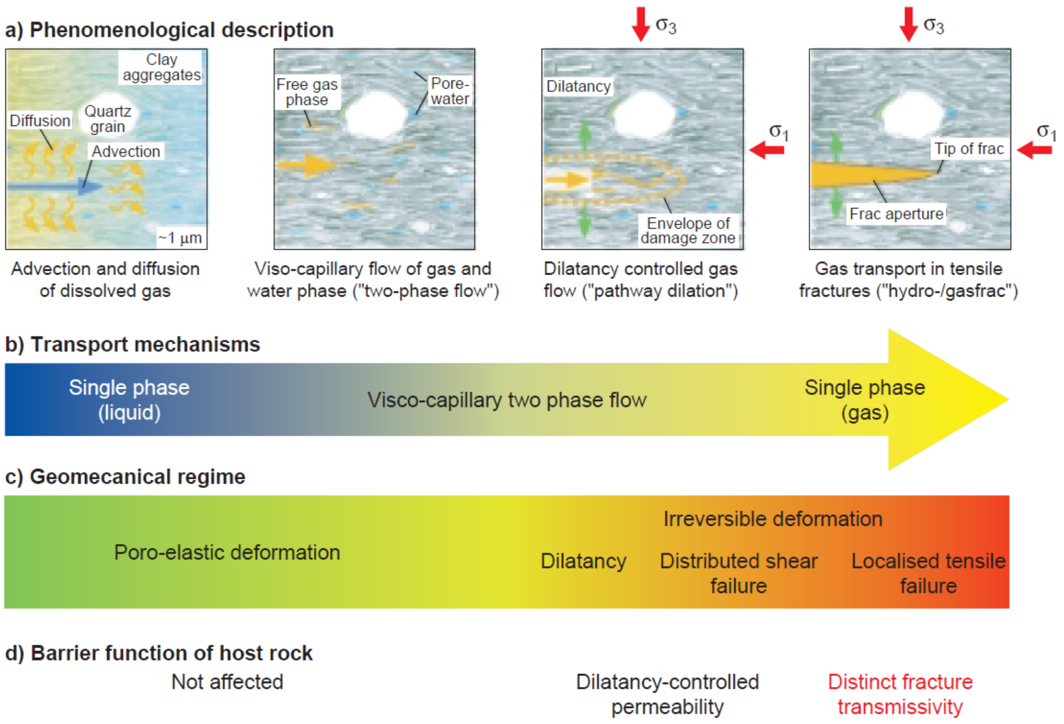

Figure 4-1 – Classification and analysis of gas transport processes in all clayey materials (Marschall et al., 2005). (a) phenomenological description based on the microstructural model concept (σ_1: horizontal total stress, σ_3: vertical total stress); (b) basic transport mechanisms; (c) geomechanical regime; (d) effect of gas transport on the barrier function of the host rock.

4.2.1 Diffusion of dissolved gas through clayey materials

4.2.1.1 Process overview / State of knowledge

The transport of gas dissolved in pore water is characterized by three fundamental laws: Henry's law describes the solubility of gas in pore water, Darcy's law governs the advective pore water flow and Fick's law represents the diffusion of dissolved gas due to concentration gradients in the pore water (Helmig, 1997). The transport of dissolved gas in water-saturated media occurs even at low gas pressures (Marschall *et al.*, 2005).

Where water and gas are in contact, gaseous species dissolve in the water according to Henry's law (Henry, 1803). The solubility of gas essentially depends on the gas species, the composition of the pore water (other species in solution), the temperature, and the gas pressure. Among these, gas pressure is the key parameter in the context of geological disposal. In water-saturated conditions, Henry's law states that solubility is linearly proportional to the partial pressure of gas. The proportionality factor between dissolved gas concentration and this pressure is called Henry's constant. In unsaturated conditions, as water and gas are not at the same pressure, it is necessary to take into account a correction factor, whose value evolves exponentially with pressure difference as shown in FORGE EC project (Dymitrowska *et al.*, 2015).

Dissolution and diffusion of gaseous molecules through the pore water is the background process of gas removal from a repository (Marschall *et al.*, 2005). It must always be accounted for in gas transport assessments. It reduces the amount of free gas in the disposal system to be dealt with by other processes. In fact, if gas generation is sufficiently slow (concept dependent) and transport of the dissolved gas is sufficiently rapid (clay-dependent), all the generated gas could potentially dissolve, and no free gas phase will form.

The diffusion of dissolved gas is driven by its concentration gradient in the pore water, following Fick's law. The proportionality factor between the rate of diffusion (also named flux) and the gradient of dissolved gas concentration is called the diffusion coefficient. In water-saturated porous media, the effective diffusion coefficient of dissolved gas, i.e., the coefficient of diffusion through the water-saturated material, is related to the morphology of the pore network (porosity, tortuosity, constrictivity) and is anisotropic (as shown for instance by Jacops (2018) for Boom Clay). In the frame of EURAD-GAS, BGS has measured the permeability and diffusion coefficients of helium for Boom Clay and Eigenbilzen sand samples under two different stress regimes (Jacops *et al.*, 2024). For the first time, tests on Boom Clay have been performed in conditions equivalent to a depth of burial of 400 meters thus providing new data which, in time, can be used to assess the impact of burial history on the diffusion properties of Boom Clay. As new data are still scarce, additional tests must be performed to allow comparison with data in Table 4-1. Nevertheless, data collected by the BGS confirm that, at greater depth, gas will also preferentially diffuse parallel to, rather than perpendicular to, the bedding planes. Like hydraulic conductivity, diffusion process is anisotropic as the pore space is better interconnected parallel to bedding planes than orthogonal to bedding planes.

During the last ten years, the availability of reliable diffusion coefficients for different dissolved gases, measured in a broad range of saturated clayey materials, has increased, as detailed in SOTA 1 (Levasseur *et al.*, 2021). Currently, comprehensive data sets of diffusion coefficients are available for several gases in the three studied host rocks (Table 4-1) (Jacops *et al.*, 2016; Jacops, Aertsens, Maes, Bruggeman, Swennen *et al.*, 2017; Jacops, Aertsens, Maes, Bruggeman, Krooss *et al.*, 2017). For Volclay KWK[7], a bentonite similar to the MX-80 bentonite, at dry density 1.4 and 1.6 g/cm³, data for He, Ne, CH_4 and C_2H_6 are available.

Diffusion coefficients for hydrogen, which is the most relevant gas in a geological repository, are however still lacking for almost all clayey materials. Measuring diffusion coefficients of hydrogen in clay-based materials is experimentally challenging because of microbial conversion of H_2 into CH_4 (Mijnendonckx *et al.*, 2019). Only Jacops *et al.* (2015) were able to measure the diffusion coefficient of hydrogen in one sample of Boom Clay by using a sophisticated protocol, including heat sterilization, gamma irradiation, gas filtration and using a microbial inhibitor. Measurement of the hydrogen diffusion coefficient in COx by using the same protocol is in progress at SCK CEN at the time of writing.

Nevertheless, the confidence in the measured diffusion coefficients is high as an exponential relationship, describing the decrease in diffusivity with an increase in the size of the diffusing molecule can be established for the three studied clays (Jacops, Aertsens, Maes, Bruggeman, Swennen *et al.*, 2017).

Similarly, a relation between diffusion coefficient and permeability has been established for Boom Clay by BGS as illustrated in the textbox hereafter (Box 2, page 26). This relationship is not surprising as both parameters depend on the porosity of the clay which is fairly constant for Boom Clay. It is therefore concluded that this relation should also be a function of some other clay property (e.g., mineralogy,

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[7] The "Volclay KWK" bentonite is a brand name of a bentonite similar to the "MX-80" bentonite. It is a fine-grained sodium bentonite with montmorillonite as the main component (Horseman et al., 1999). In contrast to "MX-80", "KWK" has been used in only a few research studies related to the disposal of radioactive waste.

preconsolidation state, etc.), not yet clearly identified at the end of EURAD-GAS. The results of Jacops (2018) showed that diffusion is indeed influenced by the mineralogical composition and density. In the framework of EURAD-GAS, BGS has developed a specific set of experiments to understand the variations of the diffusion coefficient as a function of mineralogy. At the time of writing, this work is still ongoing. It should provide relevant information for generic disposal concepts and improve the use of existing data by various end-users.

Table 4-1 – Diffusion coefficients for dissolved gases and HTO in different clayey materials (samples close to full saturation).

	D_{eff} [× 10^{-11} m²/s]						
	BC		**COX**		**OPA**	**Bentonite [g/cm³]**	
	⊥	**//**	**//**	**⊥**	**⊥**	**1.4**	**1.6**
He	46.8 ± 1.7 45.0 ± 1.0 50.0 ± 0.6 51.0 ± 2.0	74.7 ± 2.0	10 ± 1	8.1 ± 0.2	6.8 ± 0.3	17.7 ± 1.1	27.0 ± 0.9
HTO	18.7 ± 0.6 16.0 ± 0.5 20.6 ± 0.6 17.6 ± 0.9	27.8 ± 0.9	2.2 ± 0.78	–	1.2 ± 0.04	15.6 ± 1.2	8.07 ± 0.5
Ne	17.5 ± 0.3	22.9 ± 1.0	–	2.1 ± 0.1	0.6 ± 0.03	10.3 ± 0.4	8.7 ± 0.7
H_2	–	51.2 ± 1.1	–	–	–	–	–
Ar	6.9 ± 0.2	14.5 ± 0.2	1.3 ± 0.1	0.7 ± 0.02	0.4 ± 0.1	–	–
CH_4	9.7 ± 0.3 8.8 ± 0.3 11.0 ± 0.1 8.4 ± 0.2	15.5 ± 0.5	–	–	–	2.8 ± 0.1	0.9 ± 0.1
Xe	6.1 ± 0.2	6.6 ± 0.9	–	–	–	–	–
C_2H_6	4.6 ± 0.2	5.9 ± 0.1	–	0.2 ± 0.01	–	1.1 ± 0.02	0.3 ± 0.04

The geometric factor, which relates the diffusion coefficient in a porous medium to the diffusion coefficient in water, is not an intrinsic property of the studied material (Jacops, Aertsens, Maes, Bruggeman, Swennen *et al.*, 2017). Indeed, the observed relationship depends on the material. For the Boom Clay, the geometric factor increases only slightly when the diffusing molecule becomes larger: the difference in geometric factors between the smallest (He) and largest (C_2H_6) molecules is not more than a factor of two. For the other materials (COx, OPA and Volclay KWK (density 1.4 and 1.6 g/cm³)), the geometric factor clearly increases with an increasing molecule size. The ratio in geometric factors between the smallest (He) and largest (C_2H_6) molecule is about 16 for the Volclay KWK sample with density 1.6 g/cm³. This implies that the geometric factor is not an intrinsic property of the studied material but depends on both the material pore network and of the size of the studied diffusing molecule.

Box 2 – Gas diffusion: synthetic samples can act as early demonstrators for the establishment of relationship between key transport parameters.

In the frame of EURAD-GAS, BGS has measured the permeability and diffusion coefficients for a series of synthetic samples manufactured with mineralogical compositions that allow to mimic transport characteristics of the Boom Clay. The aim was to evaluate the relevance of synthetic samples for defining important correlations between hard to measure parameters (such as the diffusion coefficient) and easier to measure parameters (such as the intrinsic permeability). The advantage of synthetic samples is that they are "pure systems" (without the complexities introduced by the heterogeneities of natural materials).

A coupling between intrinsic permeability and diffusion coefficient was clearly put in evidence from the synthetic sample data, suggesting mineralogy, diffusion coefficient and permeability are fundamentally linked (Figure 4-2) (Jacops *et al.*, 2024). The data collected from literature by BGS and completed by their own tests for the Eigenbilzen sand and Boom Clay appear to follow a similar trendline. Given this commonality, it seems reasonable to extend this trendline in order to predict likely diffusion coefficient values for higher/lower permeability samples. However, all of these relationships remain hypothetical. Further experimental verification is needed.

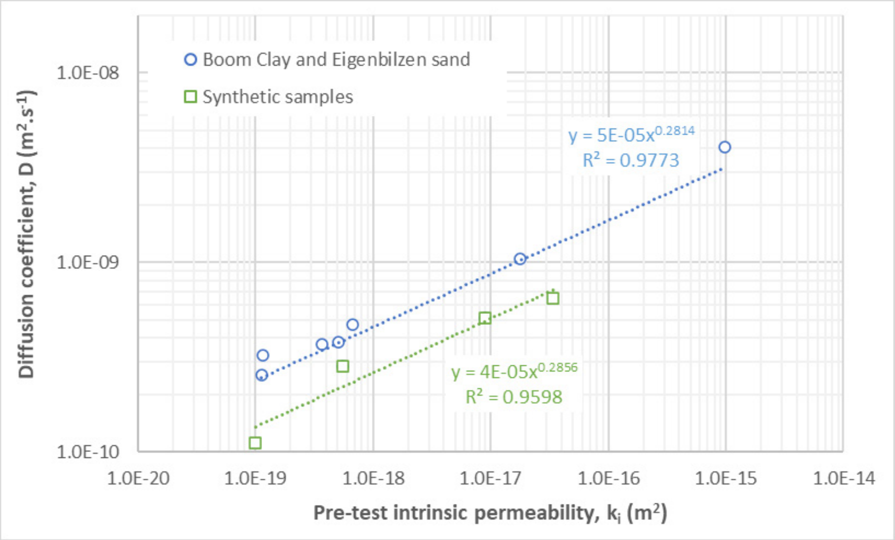


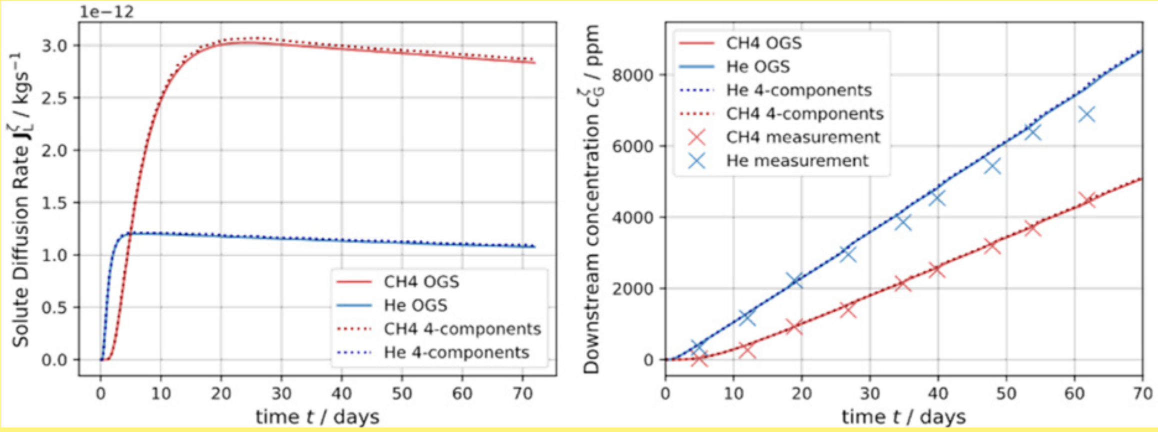
Figure 4-2 – Compilation of data for Boom Clay, Eigenbilzen sand and synthetic samples (Jacops et al., 2024).

The process of diffusion of dissolved gas in water-saturated clay is well understood. Indeed, the diffusion process has been successfully modeled at different scales with several approaches during EURAD-GAS (Jacops *et al.*, 2024) (molecular pore network modeling or macroscopic simulations by finite element approaches) and validated through comparisons with the experimental results of the dissolved gas through-diffusion experiments of SCK CEN (Jacops *et al.*, 2015) (Box 3, page 27).

Box 3 – The diffusion process of dissolved gas through clayey materials is well understood, both at molecular and macroscopic scales.

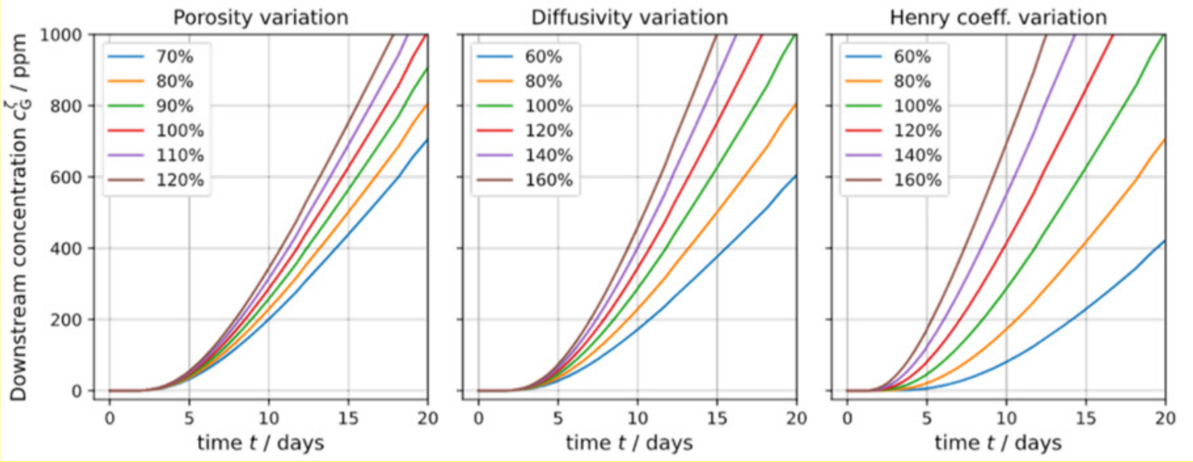
In the framework of EURAD-GAS, the double through-diffusion experiment performed by SCK CEN (Jacops *et al.,* 2015*)* was used to test the validity of a series of gas transport models considering diffusion processes at various scales.

BGR extended the two-phase hydro-mechanical model (TH^{2}M model) of the open-source finite element code OpenGeoSys 6 to the diffusion processes of dissolved gas (Pitz *et al.*, under review - 1). The comparison of their modeling with experimental data has allowed them to evaluate the contribution of each individual process (diffusion, advection in liquid, advection in gas) and associated parameters as shown in Figure 4-3. A sensitivity analysis, performed as a complement, suggested a higher impact of the Henry's law constant on the diffusion process compared to the impact of porosity and diffusion coefficient (Figure 4-4).



(A) (B)

Figure 4-3 – Comparison of experimental and numerical results for the double diffusion experiment (Pitz et al., under review - 1).

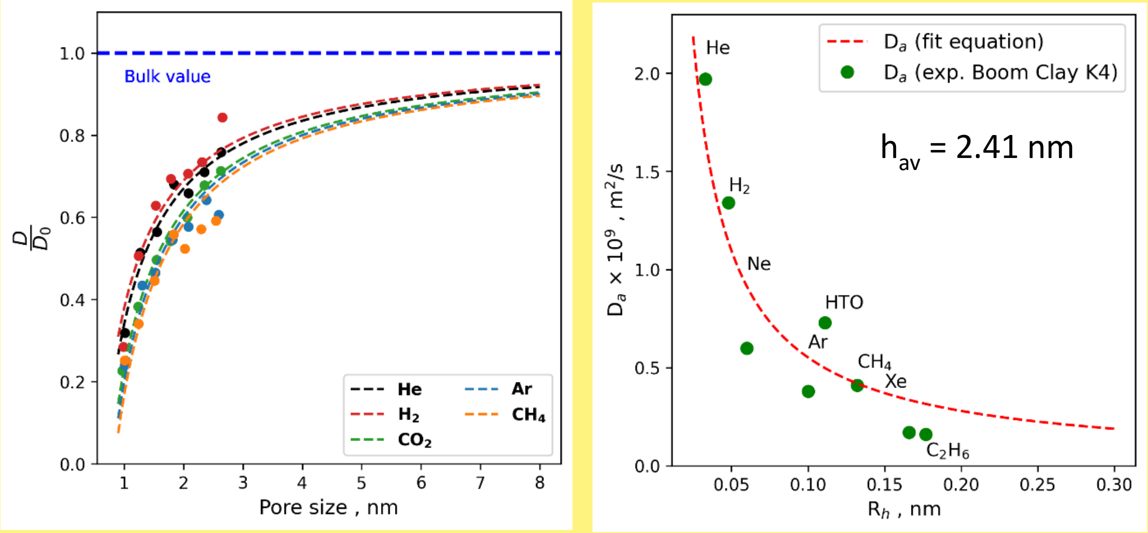


(A) (B) (C)

Figure 4-4 – Impact of selected parameters (A: porosity, B: diffusivity, C: Henry's law constant) on diffusion as modeled for the double diffusion experiment (Pitz et al., under review - 1).

To consider the effects of various gases simultaneously, Aalto University (Gupta *et al.*, 2023) developed a numerical framework to model gas mixture flow for two incompressible and inert-type gases in clays under hydraulic conditions. The framework is an extension of the finite element code Thebes, incorporating new functions for water retention and gaseous flow behavior. As with BGR approach, the approach developed by Aalto University successfully captures the results of the SCK CEN experiment in term of diffusive gas flux through the sample and hence the gas concentration increase in the two vessels. These two approaches are useful for characterizing diffusion process at large scale.

To understand more in details the mechanisms associated to the diffusion processes, PSI developed a diffusion model at molecular scale (Owusu *et al.*, 2022; Owusu *et al.*, 2023). By analyzing the molecular scale diffusion trajectories of various gases (Ar, CH_4, CO_2, H_2 and He), they established a semi-empirical equation connecting effective diffusivity of dissolved gas molecules in clay with their molecular mobility in bulk water (available experimentally), interaction with clay mineral surface (obtained by molecular simulation as function of inter particle distance), and "geometry factor" (experimentally available from HTO measurements of SCK CEN in Boom Clay) (Figure 4-5). This equation was found to be consistent with the results of SCK CEN experiments with gases in three different types of Boom Clay, with different porosities and pore size distributions (Jacops, Aertsens, Maes, Bruggeman, Krooss *et al.*, 2017). This empirical equation can now be used to assess gas molecule's mobility in Boom Clay, if no direct experimental data are available, as well as for interpolation of experimental data in THMC modeling.



(A) (B)

Figure 4-5 – Molecular scale and effective transport parameters of Ar, He H_2, CO_2 and CH_4 obtained by molecular dynamics simulations (Owusu et al., 2022; Owusu et al., 2023). (A) Relative diffusion coefficient of gases (D/D_0) as a function of pore size. (B) Apparent diffusion coefficients as a function of gas effective radius.

Clayey EBS materials and the EDZ of some clayey host rocks may not be fully saturated at the time of repository closure. Significant uncertainties remain regarding the duration of the saturation phase, which depends on the interplay between water inflow from the host rock and evacuation of gas, initially present or produced after closure. Therefore, diffusion through partially saturated clayey materials has also been investigated in the frame of EURAD-GAS.

In a non-saturated medium, gases do not only diffuse as dissolved in the liquid phase but also as gas through the gas phase. The efficiency of diffusion processes in both gas and liquid phase (water) depends on the fraction of the porosity filled with gas, which depends on the degree of saturation with water, and its continuity and connectivity as detailed in SOTA 1 (Levasseur *et al.*, 2021). In the frame of geological disposal, diffusion of gas in the gas phase is particularly of interest for volatile radionuclide transport.

SCK CEN (Gowrishankar *et al.*, 2023) developed a new setup to measure the diffusion of dissolved gas in unsaturated clays that is inspired from the double through-diffusion experiment of Jacops *et al.* (2015). Their approach was tested on synthetic clayey materials to limit the complexity brought by natural materials. The results obtained from these synthetic samples show that the effective diffusion coefficient increases slightly when the saturation decreases from 100 to 73%. These results were expected because gas diffuses more easily in the gas phase than in the liquid phase. At this range of saturation degrees, the connectivity of the gas phase is still limited, hence the moderate increase in the diffusion coefficient.

The diffusion coefficients of solutes (HTO (tritiated water), Cs-134, Na-22 and I-125) on desaturated COx samples have been measured (Savoye *et al.*, 2010; Savoye *et al.*, 2012; Savoye *et al.*, 2017). The results show that, for a decrease in saturation from 100 to 80%, the effective diffusion coefficient decreases by a factor of 60, 50 and 7 for Cs-134, I-125 and HTO, respectively. This decrease in diffusivity was expected since solutes diffuse only in the liquid phase.

Complementary to this work, the relationship between diffusive transport and the pore network of the studied materials was investigated through pore network models that allow to theoretically calculate diffusion coefficients under different conditions such as variable density and variable degree of saturation The values of the theoretical parameters identified by PSI (Owusu *et al.*, 2023) will soon be compared to the experimental results of the dissolved gas through-diffusion experiments in unsaturated conditions of SCK CEN (Gowrishankar *et al.*, 2023).

4.2.1.2 Uncertainties and knowledge gaps

The experimental protocol to be carried out to determine diffusion parameters for dissolved gas is well developed and mastered and can now easily be extent to a broad range of clayey materials under various mechanical conditions and, in the case of clayey host rocks, along different directions relative to the bedding. To extend this database, experiments studying the **potential effects of sample size and mechanical conditions on the diffusion coefficients** have been started during EURAD-GAS (e.g., Jacops *et al.* (2023)). The NEMESIS experiment (Neon Diffusion in MEGAS In Situ) which studies the upscaling of diffusion of neon from laboratory scale to in situ scale in the HADES URL (Jacops *et al.*, 2022) is currently ongoing. After a very long, well considered preparation phase, it was started in September 2023 and will run for 3 to 5 years. As such experiments are very time consuming, results are not yet available at the time of writing this document and the estimation of diffusion coefficient in in situ conditions is still considered a knowledge gap.

The work of BGS indicates a possible **relationship between the effective diffusion coefficient and intrinsic permeability.** The development of synthetic clay samples (compacted clay/silt/sand mixes) has allowed the exploration of various relationships (e.g., bulk density, clay content and permeability).

However, further expansion of the data set is required to fully define these relationships, including the role of in situ stress/compaction state of the clay, and their potential to predict the diffusion coefficient from more easily measured parameters. In this sense, the use of synthetic samples would be considered by a less advanced program as a good alternative for the characterization of clays prior the selection of a host rock.

Although the technique to measure diffusion of dissolved gases in clay-based samples is well established, **diffusion coefficients for hydrogen remain scarce**. So far, only one reliable value (for Boom Clay) has been published by Jacops *et al.* (2015). Measuring diffusion of hydrogen is very complicated because hydrogen itself easily leaks through joints and seals and is sensitive to microbial conversion into methane. Only by applying a complex sterilization protocol as described by Jacops *et al.* (2015), microbial conversion is avoided and reliable diffusion coefficients can be obtained. Given the very limited availability of diffusion coefficients for hydrogen for different host rocks and EBS materials, this is considered to be a knowledge gap too.

The experimental setups yield reliable parameter values for water-saturated materials. In clayey EBS materials or in EDZ which may be not initially saturated at the time of repository closure, significant uncertainties remain on the duration of the saturation phase. Hence, diffusion through partially saturated EBS materials should be further investigated in such cases. Research on the **effect of desaturation on the diffusion coefficients** of dissolved gases has been initiated during EURAD-GAS by SCK CEN on synthetic clay samples. First results on desaturated synthetic clay samples have shown that for samples with a saturation degree between 73 and 100%, the impact of desaturation on the diffusion coefficient is minor. At the time of writing, experiments are still ongoing, and more detailed data interpretation and sample characterization is required. Moreover, for deeper fundamental understanding, other sample compositions and lower saturation degrees have to be studied. This issue is therefore still considered as a knowledge gap.

In addition, **lack of knowledge on the actual water-gas distribution in partially desaturated clay samples of centimeter-decimeter size still remains**. In some cases, clayey samples prepared by drying from a water-saturated state may exhibit front desaturation near their surfaces rather than homogeneous desaturation throughout the volume. For the time being, imaging techniques do not always allow to clearly distinguish liquid-filled from gas-filled pores in clayey materials.

4.2.2 Gas sorption

4.2.2.1 Process overview / State of knowledge

While diffusing through the pore water, dissolved gas might interact with the clayey barrier materials (either parts of the EBS and/or the host rock) and gas sorption on the clay minerals could occur (Didier, 2012; Didier *et al.*, 2012; Truche *et al.*, 2018). On the one hand, this could lead to a retardation of gas transport. On the other hand, sorption processes would remove free and dissolved gas thus potentially reducing the pressure build-up in the host rock.

Hydrogen adsorption (physisorption) on various materials has been primarily studied in the context of solid-state hydrogen storage, mostly on synthetic materials. Some materials, such as Metal Organic Frameworks (MOFs), zeolites, carbon-based nanomaterials, have been thoroughly investigated for their potential to adsorb hydrogen: their $kg_{H2}/kg_{material}$ can be as high as 9 wt% H_2 at 77 K and 50 bar (Langmi *et al.*, 2003; Jordá-Beneyto *et al.*, 2007; Ramirez-Cuesta and Mitchell, 2007; Kaye *et al.*, 2007; Han *et al.*, 2011). However, hydrogen physisorption on microporous materials suffers from a dramatic loss of adsorption capacity as temperature increases above 77 K due to the low enthalpy of H_2 adsorption on these materials, i.e., 5 – 8 kJ/mol (Bhatia and Myers, 2006). The adsorption capacity of carbon-based nanomaterials typically decreases by one order of magnitude from 77 K to 298 K. For example, hydrogen uptake of single-wall carbon nanotubes is about 2 wt% H_2 at 77 K and 40 bar, but decreases down to about 0.2 wt% H_2 at 298 K and 200 bar (Jordá-Beneyto *et al.*, 2007).

There are few quality data on hydrogen adsorption on natural samples, such as smectites and other clay minerals, because sorption experiments with H_2 on such materials are difficult to perform. Setups are prone to leakage and microbial processes can interfere (transforming CO_2 to CH_4 or nitrate to N_2) (Vinsot *et al.*, 2014; Jacops *et al.*, 2015; Bagnoud *et al.*, 2016). Table 4-2 reports the maximum sorption capacities of natural clayey samples from literature. (Ziemiański and Derkowski, 2022) concluded that hydrogen intercalation within smectite interlayers is strongly dependent on hydration state of the interlayer space, itself controlled by the nature of the exchangeable cation.

Table 4-2 – Maximum sorption capacity of clayey compounds taken from literature.

Clayey compound	**Specific surface area [m^2/g]**	**Maximum sorption capacity [wt% H_2]**	**Temperature [°C]; pressure [bar]**	**Reference**
Al-pillared montmorillonite		0.2	-196; ~ 1	(Gil *et al.*, 2009)
K-bentonite		0.2	-196; ~ 1	(Erdoğan Alver, 2017)
Sepiolite		0.2	-196; ~ 1	(Erdoğan Alver, 2018)
Laponite®	350	0.63	-196; ~ 1	(Edge *et al.*, 2014)
Saponite	45	0.05	25; 100	(Masci *et al.*, 2023)
Laponite®	350	~ 0.1	25; 140	(Edge *et al.*, 2014)
Montmorillonite and illite		Lower than 0.04 wt% with one exception (0.1 wt%) *	Several temperatures (from 25°C to 70°C) and pressures (up to 145 bar)	(Ziemiański and Derkowski, 2022)
COx	20–40 **	0.1 ***	25; 60	(Bardelli *et al.*, 2014)
COx	20–40 **	0.12 ***	90; 60	(Bardelli *et al.*, 2014)

* The specific case when montmorillonite exchanged with tetramethylammonium cation. In this case, the hydrogen uptake was 0.1 wt% at 25°C and 145 bar.
** (Yven *et al.*, 2007)
*** These values must be taken with care, see text hereafter.

In the framework of EURAD-GAS, experiments with a well-defined protocol under dry conditions have been carried out with the following results:

- H_2 uptake is very low. At 100 bar, the hydrogen uptake by dry COx is 0.04 wt% at 0°C, 0.02 wt% at 22°C, and is almost zero at 50°C. At such temperatures, the measurements are close to the quantification limit of the technique used. To put these values into perspective, they can be compared with saponite, laponite® and MOF-177 (a metal organic framework): the H_2 uptake recorded for these three materials at 100 bar and 25°C is 0.05, 0.1 and 0.5 wt% respectively. Obviously, it is the specific surface area of these materials that is the primary driver of their H_2 uptake. The specific surface area as measured by the BET technique using N_2 as the sorbent gas (S_{BET}) is: 20 m^2/g – 40 m^2/g for COx (Yven *et al.*, 2007), 45 m^2/g for saponite, 350 m^2/g for laponite® and 3 800 m^2/g for MOF-177.
- The H_2 uptake by COx decreases with increasing temperature as thermodynamically expected.
- These results slightly differ with the data previously reported by Bardelli *et al.* (2014). 1) the authors report a higher H_2 uptake at 90°C than at 28°C (0.12 – 0.1 ± 0.01 wt% respectively), which seems physically not consistent, 2) the H_2 uptake at 28°C and 60 bar H_2 pressure is about

0.1 ± 0.01 wt%. This value is high considering the specific surfaces of laponite and MOF-177, that are 2 to 3 orders of magnitude higher than the one of COx. (Even with such a high surface area, MOF-177 can only adsorb 0.6 wt% of H_2 at room temperature and 100 bar (Li and Yang, 2007).)

Adsorption of hydrogen by the COx decreases drastically with the water saturation (Didier, 2012). Under repository conditions, COx will be mainly fully saturated (a few meters away from the tunnels) or close to full saturation in the vicinity of the tunnels and H_2 adsorption should be neglected. Furthermore, in disposal conditions, in clays that are partially or fully saturated with pore water, potential sorption sites might not be available for hydrogen and other volatile gas adsorption.

Based on EURAD-GAS results and on data from literature, gas sorption of hydrogen is not considered to be a relevant retardation mechanism for diffusive transport under disposal conditions.

4.2.2.2 Uncertainties and knowledge gaps

It may be of interest to conduct research to answer the following questions, although gas sorption of hydrogen is not considered a relevant retardation mechanism for diffusive transport under disposal conditions:

- What is the role of water saturation on adsorption of different gases in clays?
- Does the release of natural gases from the host rock due to heating or pressure changes are an additional source of gas? Is it significant enough to be considered?
- One can also question the adsorption mechanism itself. What is the nature of the gas sorption sites? Where is the gas adsorbed? What is the relative contribution of the different mineral phases, including organic matter? Neutron spectroscopy can be extremely valuable in addressing these questions in detail.

It could also be interesting to determine sorption isotherms for all relevant gases in all relevant materials. The Full-scale Emplacement – Gas (FE–G) experiment is currently investigating the potential for reversible sorption of oxygen to bentonite as a function of hydration and temperature. There is a play-off between temperature of the waste impacting saturation in the immediate vicinity of the disposal package, and a potential return to aerobic conditions once the temperature has decreased sufficiently to allow resaturation.

4.2.3 Advective gas flow through clayey materials

4.2.3.1 Processes overview

If the gas production rate exceeds the rate at which gas is dissolved, a free gas forms. Three primary mechanisms for gas transport through clayey materials exist (i) the visco-capillary two-phase flow, where capillary forces must be overridden to allow gas to enter the pores and displace the liquid phase, (ii) the formation of dilatancy-controlled gas pathways or (iii) the formation of gas fractures. The prevalence of one mechanism or another is largely controlled by the fabric of the material, influencing the capacity of gas to enter porous media (gas entry pressure), the mobility of gas (permeability), and by the susceptibility to deformation[8] resulting from the passage of gas (Marschall *et al.*, 2005).

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[8] Susceptibility to deformation depends on mechanical properties (strength, stiffness) and stresses.

4.2.3.1.1 Visco-capillary two-phase flow

Visco-capillary two-phase flow occurs when gas invades as a separate phase into a porous medium. This process is called invasion or drainage because, in a water-saturated medium, the non-wetting fluid (gas) displaces a wetting fluid (pore water). The propagation of the gas front is controlled by the complex interaction of viscous forces, capillary forces and gravity. Depending on the balance between these forces in the considered material, different types of desaturation fronts might develop: stable displacement, viscous fingering or capillary fingering as illustrated in Figure 4-6 after Lenormand *et al.* (1988). When a gas front invades an initially saturated matrix of a low-permeability clayey material, capillary fingering can be assumed as the prevailing regime. Gas release through a reactivated fault or along the EDZ and the backfilled underground structures may be associated with viscous fingering, a process which creates a distinct precursor flux (early gas breakthrough). This phenomenon is of potential relevance for the transport of volatile radionuclides.

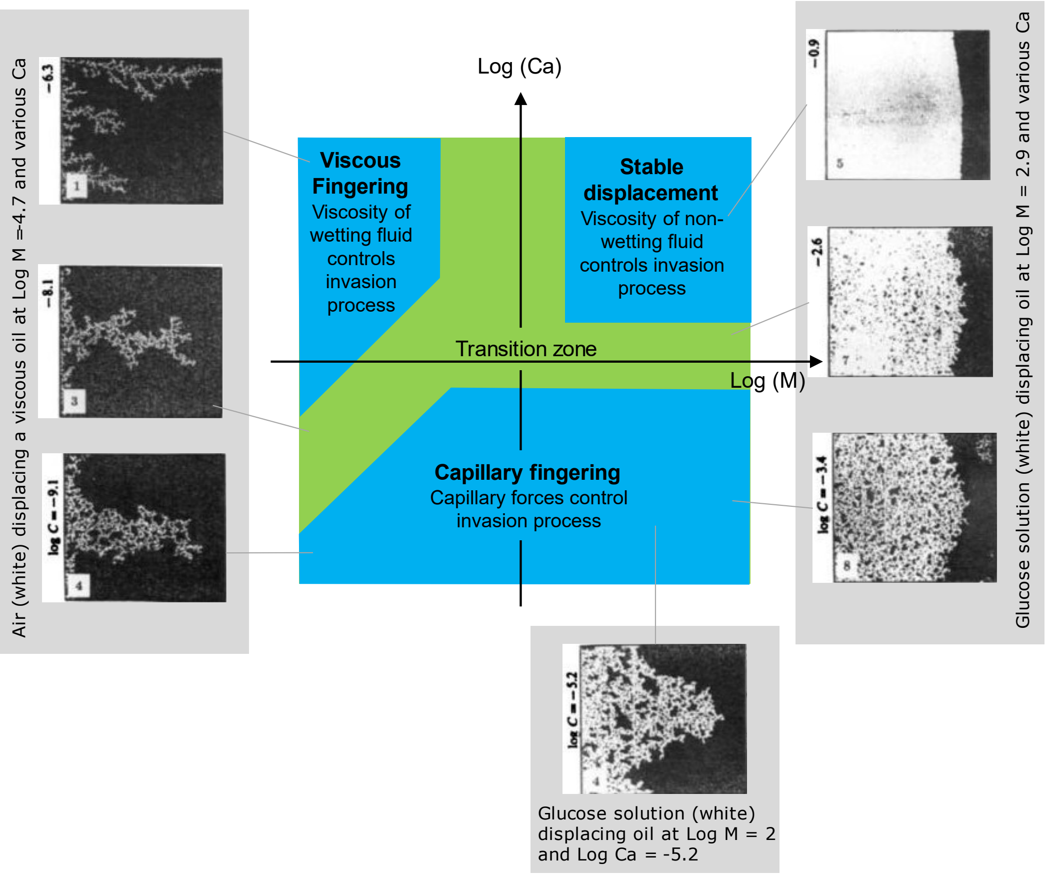

Figure 4-6 – Schematic representation of the three major flow regimes for a wide range of capillary number Ca and viscosity ratio M[9] after Lenormand (1988): viscous fingering, capillary fingering and stable displacement (Levasseur et al., 2021).

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[9] Capillary numbers *Ca* and viscosity ratios *M* being respectively defined as:

$$Ca = \frac{viscous \rightleftarrows forces}{capillary \rightleftarrows forces} = \frac{\mu_{wf} \cdot v_f}{\gamma_{wf,\rightleftarrows nwf}} \qquad M = \frac{\mu_{defending}}{\mu_{invading}}$$

where μ_{wf} and μ_{nwf} represent the viscosities of the wetting and non-wetting fluid [Pa.s], v_f is the characteristic (Darcy) velocity in the medium [m/s], $\gamma_{wf,\rightleftarrows nwf}$ is the surface tension between the two fluids [N/m], $\mu_{defending}$ is the viscosity of the defending fluid and $\mu_{invading}$ is the viscosity of the invading fluid [Pa.s].

The potential for visco-capillary two-phase flow depends on the fabric of the porous medium, in particular the characteristic pore throat radius and its distribution. The process of gas invasion can lead to a redistribution of fluid pressures and stress in the material resulting in deformations of the porous medium (Keller, 2021). The deformation remains, however, reversible in the two-phase flow regime.

Visco-capillary two-phase flow in a porous material can be defined in a similar way to single-phase flow, but taking account of the saturations of (i.e., the fractions of the porosity occupied by) each phase in the material. The permeability of the material that defines single-phase flow is modified by a relative permeability for each phase that depends on the saturation (e.g., van Genuchten-Mualem model (Mualem, 1976; Helmig, 1997)). In addition, there is a difference in pressure between the phases, the capillary pressure, which a non-wetting phase (gas) must exceed for it to displace a wetting phase (pore water). Capillary forces are inversely proportional to pore size. Hence, the largest pores are invaded first by gas (gas can displace water from the largest pores at a lower capillary pressure than for the smaller pores). Thus, invasion of gas only starts once gas pressure exceeds the capillary pressures of the largest pores. This threshold pressure is called the gas entry pressure. To increase gas saturation and thus invade smaller pores, higher capillary pressures have to be overcome and thus gas pressure must increase (Figure 4-7). SOTA 1 explained and illustrated these concepts (Levasseur *et al.*, 2021). Box 4 (page 35) illustrates how to explore the transition between single-phase and two-phase flow conditions in clayey rocks has been explored through an example of a numerical approach developed in EURAD-GAS.

Clayey materials can have a wide range of permeabilities and gas entry pressures. On the one hand, gas transport by visco-capillary two-phase flow will occur readily in saturated porous geomaterials with relatively high permeabilities and low gas entry pressures or in partially saturated materials, even with very high gas entry pressures, as bentonite. On the other hand, for saturated materials with very low permeabilities and high gas entry pressures (like saturated clayey host rocks and compacted bentonite), gas transport will occur by visco-capillary two-phase flow only after substantial pressure build-up, and then with limited ability for gas to flow. As stated by Sellin and Leupin (2013), in the particular case of bentonite currently considered for radioactive waste disposal, when unsaturated or partially saturated, a linear dependence develops between the gas flow rate and the pressure gradient, which indicates that visco-capillary two-phase flow is the dominant mechanism (Villar *et al.*, 2012). This may also be the case for saturated sand/bentonite mixtures if the sand content is sufficiently high. However, at a degree of saturation of 80-90% or higher, the bentonite behavior changes: in almost saturated bentonite—as in clayey host rocks—a distinct change of micro-fabric is observed, with the disappearance of connected macropores. Gas entry pressures are sufficiently high that the pressure of the invading gas phase would lead to volumetric deformation (i.e., dilatancy) of the material prior to displacing water from pores. This deformation results in dilatant gas-specific pathways (Horseman *et al.*, 1999; Villar *et al.*, 2012; Graham *et al.*, 2012; Sellin, 2014; Cuss *et al.*, 2022).

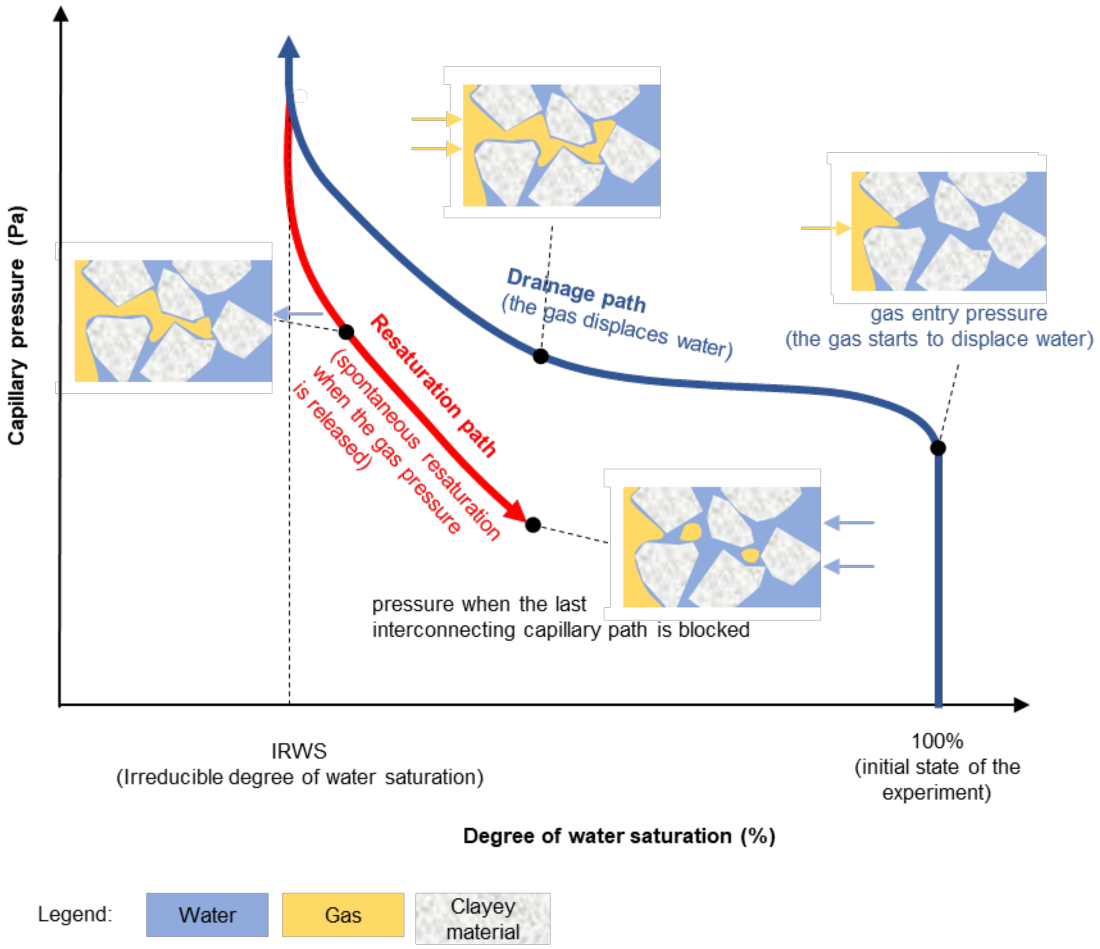

Figure 4-7 – Conceptual sketch of capillary processes in clayey materials. The graph shows the evolution of the capillary pressure as a function of water saturation in a stepwise desaturation and resaturation experiment starting with a fully saturated sample. The blue line marks the drainage path, where the gas displaces the water (increasing capillary pressures). The red line describes the resaturation path (decreasing capillary pressures) (Levasseur et al., 2021).

Box 4 – Transition from diffusive to advective gas transport in clayey materials has been modeled in the frame of EURAD-GAS.

A comprehensive numerical study, based on continuous two-phase (visco-capillary) flow models, has been conducted in EURAD-GAS jointly with the MoMaS[10] working group of Andra to explore the behavior of gas flows in clayey rock, specifically targeting the transition from single-phase (i.e., diffusion of dissolved gas) to two-phase flow conditions, an often-considered scenario in radioactive waste disposal (Figure 4-8). The modeling of phase transition was developed in a theoretical framework for low-permeability porous media through a benchmark initiated by Andra (Grunwald *et al.*, 2023). This approach has now to be validated on experimental data such as the long-term gas injection tests which are ongoing at SCK CEN (Jacops *et al.*, 2024) and that aim to capture the

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[10] Groupe de Recherche “Mathematical Modeling and Numerical Simulation for Nuclear Waste Management Problems”

transition from diffusive to advective gas transport. The success of such an approach can provide valuable insights into the behavior of gas in clayey materials, allowing to improve the understanding and confidence in models used for large-scale repository simulations.

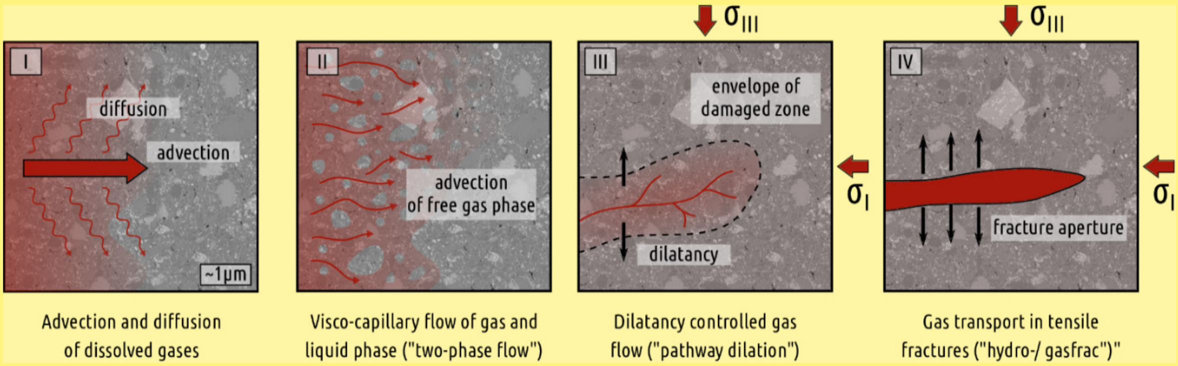


Figure 4-8 – Modified version of an overview of gas transport regimes in low-permeable clayey rock after Marschall et al. (2005, Figure 2) and Cuss et al. (2014).

4.2.3.1.2 Dilatancy-controlled gas transport

Dilatancy-controlled gas transport is a transport mechanism of special importance for clayey materials with very fine porosity and low tensile strength. In such materials, visco-capillary two-phase flow is difficult, and no transport of gas phase can occur until a significant gas pressure has built up. When gas pressure exceeds the sum of the local stress and tensile strength of the material, gas invasion will occur through the localized creation of new porosity. Significant debate as to the nature of that deformation, e.g., mode 1 (tensile) or mode 2 (shear) or a combination of both, however, the term pathway dilatancy is often used to reflect this change in porosity (Harrington and Horseman, 1999; Villar *et al.*, 2012; Graham *et al.*, 2012; Cuss *et al.*, 2014; Levasseur *et al.*, 2021; Gonzalez-Blanco *et al.*, 2022).

In the majority of experiments, no measurable desaturation or expulsion of water is observed (e.g., (Harrington and Horseman, 2003; Jacops *et al.*, 2014; Graham *et al.*, 2019; Levasseur *et al.*, 2021)). Confidence in these measurements is high enough to assert that water is not being displaced from existing pore space by gas, indicating that water displacement is not a pre-requisite for gas flow.

Should pathway formation be sufficient to relieve gas pressure, then closure of the pathways can occur (Horseman *et al.*, 1999; Graham *et al.*, 2012; Gonzalez-Blanco *et al.*, 2023). The consequences of this opening-closing process on gas pathway sealing and its impact on material properties are not yet fully understood.

In the terminology of multiphase flow concepts when applied to numerical models, dilatancy-controlled gas flow is still represented by visco-capillary forces; the main difference with respect to conventional two-phase flow is that the transport properties of the solid skeleton (permeability, relative permeability, capillary pressure relationship) can no longer be viewed as invariants. Instead, they are considered dependent on the state of deformation of the material (Marschall *et al.*, 2005), which in turn depends on the mechanical properties of the material.

Characterization of dilatant pathways in clays is challenging because of the small scale, local and discrete nature of the couplings between hydraulic and mechanical properties (Harrington, Graham *et al.*, 2017; Harrington *et al.*, 2019). In EURAD-GAS, experimental work examining changes in the stress field during gas injection indicates substantial information relating to pathway dilatancy can be obtained (Graham and Harrington, 2024). This includes quantification of the degree of deformation, an initial understanding of the size distribution of pathways, a measure of the degree of pathway opening/closure along with an insight into the spatial evolution of deformation. These new approaches demonstrate the potential to better inform the development of numerical models as a result of work undertaken in EURAD-GAS (Kolditz *et al.*, 2024; Marschall *et al.*, 2024). Nevertheless, spatial distribution of the pathways is not so easily observable even using imaging techniques. Its characterization also suffers from the weak repeatability of tests, highlighting the importance of identifying deterministic and stochastic processes which occur during dilatancy-controlled gas flow. So far, it is generally still not sufficiently detailed for predictive modeling of gas transport at repository scale (Corman *et al.*, submitted). Therefore, in materials where gas transport is expected to occur by dilatant pathway, the transport behavior is often approximated by modeling it using enhanced visco-capillary two-phase flow approaches with suitably chosen parameters (see for instance Corman *et al.* (2022)) in the frame of EURAD-GAS and as detailed in SOTA 1 (Levasseur *et al.*, 2021). Such approaches also have been applied by Tamayo-Mas *et al.* (2021; 2024).

4.2.3.1.3 Gas fracturing

The transition between dilatancy-controlled gas flow and macro scale gas fracturing is strongly dependent on the gas pressurization rate. Macroscopic tensile fractures develop when the gas pressure is larger than the sum of the minimum principal stress and the tensile strength of the material (e.g., (Valkó and Economides, 1995)). In classical linear elastic fracture mechanics, macroscopic fractures are initiated quasi-instantaneously and propagate at about the velocity of a shear wave. Gas flow in such macroscopic tensile fractures can be seen as single-phase flow processes. The propagation comes to a halt when the gas pressure in the fractures becomes less than the value of the minimum principal stress (shut-in pressure).

Sudden fracture initiation and propagation is typically observed in case of high gas pressure build-up rates. From a fracture mechanics perspective, this phenomenon refers to the process of stable crack growth, i.e., the development of fractures as gas pressure continues to increase. Gas fracturing may happen in low-strength materials if the transport by dilatancy-controlled gas pathways is not able to keep up with gas generation and pressure build-up rates. If this scenario were to occur within a repository, this could result in a large reservoir of pressurized gas from which a fracture(s) could propagate a significant distance.

Given the likely implications for GDF performance of potentially large changes in material properties associated with fracturing, this scenario should be avoided, or its likelihood minimized through appropriate provisions in the GDF design process. Therefore, it is important to recognize the circumstances under which fracturing could occur. Besides gas generation and pressure build-up rates, key controls, or parameters are the minimum principal stress on the material and its tensile strength. However, the coupling between these may not be trivial. In theory, a macroscopic fracture develops in a low tensile strength material only when the gas pressure build-up is rapid, i.e., when the combined effect of pore water displacement and formation of small-scale dilatant pathways no longer counterbalances the gas production rate. In experimental conditions combining high gas injection rates associated with rapid gas pressure build-up, gas fracturing was observed in in situ conditions in COx below the principal minimum stress by de la Vaissière *et al.* (2019; 2020). This is expected to be due to the presence of a damaged zone, i.e., the presence of initial fractures due to excavation and a modified stress field around the cavity, favoring gas fracturing. It should be noted, however, that such high pressurization rates are not expected in normal repository scenarios; gas fracturing is therefore considered unlikely.

4.2.3.2 Shared understanding

Among visco-capillary two-phase flow, creation of dilatant pathways and gas fracturing, the transport mechanism that will prevail in a fully saturated clayey host rock is largely controlled by the fabric and mineralogy of the material, as well as by in situ conditions, such as current stress state and stress history (Cuss *et al.*, 2017; Harrington, Cuss *et al.*, 2017; Gonzalez-Blanco *et al.*, 2022). This material can either be undisturbed or locally disturbed in the form of discontinuities.

A balanced assessment of gas transport processes requires, among other aspects, careful consideration of the structure of the solid skeleton and, even more important, of the connectedness of the pore network. The displacement of pore water by a gas phase is mainly restricted to the network of macropores (equivalent pore radius R_{eq} > 25 nm according to IUPAC nomenclature), as smaller pores are hardly invaded by the gas phase due to their high gas entry pressure (Rouquerol *et al.*, 1994).

High gas pressures are needed to invade a fully water-saturated clayey material. Gas invasion is not associated with major desaturation of the material. Consequently, characterization of the water retention behavior (Figure 4-7) requires advanced laboratory methods to explore the suction range close to saturation and thus close to the gas entry pressure (as detailed in SOTA 1, Levasseur *et al.*, 2021). In addition, water retention behavior is hysteretic when the material is subjected to successive drying/wetting paths notably because once entrapped in small pores gas cannot easily escape (Busch and Amann-Hildenbrand, 2013).

In gas injection experiments at laboratory scale, gas flow rates within clay are generally low, if at all, during the initial water displacement by visco-capillary two-phase flow. The gas transport capacity of the clay sample increases as dilatancy-controlled gas pathways progressively develop until gas breakthrough is reached. The invaded material may undergo deformation in response to gas pressure build-up, depending on the mechanical confinement and the tensile and shear strength of the test sample. Similar to Cuss *et al.* (2012), Gonzalez-Blanco *et al.* (2022) show that the typical deformation behavior of the material in response to gas invasion exhibits first a continuous volume expansion as long as the gas propagates through the test specimen, followed by contraction when gas breakthrough at the downstream end of the sample is reached (see Box 5, page 39). At the end of the gas invasion phase, when pore pressure recovers to its initial state, a minor component of irreversible strain may be observed, corresponding to moderate changes in pore structure. Indeed, postmortem evidence for distinct localization of gas flow can be often found in these experiments, as in Harrington *et al.* (2012) and Gonzalez-Blanco *et al.* (2022). Hence, the concept of intrinsic permeability has to be used with caution in this context as measurements of gas and water permeability may yield different values.

Furthermore, the experiments in Figure 4-10 (Box 5) suggest that the effects of gas transport may differ not only with the magnitude of gas pressure but also with the gas pressure build-up rate as shown by Gonzalez-Blanco (2017) for Boom Clay and confirmed by EPFL (Marschall *et al.*, 2024) for Opalinus clay). As long as the gas pressure build-up at the locus of gas generation is slow, visco-capillary two-phase flow and dilatancy-controlled gas pathways represent efficient transport mechanisms to accommodate gas flow in clayey materials (Marschall *et al.*, 2005). In this context, “slow” means that gas generation induces little or no generation of excess pore water pressure during the transient phase (drained geomechanical conditions). The determining factor for distinguishing between “slow” and “rapid” gas pressure build-up is the coefficient of (vertical) consolidation of the clay.

Box 5 – The rate of gas pressure build-up affects the deformation response of the clay.

In the frame of EURAD-GAS, UPC and EPFL investigated the hydro-mechanical response of saturated Boom Clay and Opalinus clay on gas transport, performing dedicated gas injection experiments in oedometer cells (Gonzalez-Blanco *et al.*, 2022; Gonzalez-Blanco and Romero, 2022) (Marschall *et al.*, 2024). The experiments revealed a marked dependence of the volumetric behavior of the test samples on the gas pressure build-up rate.

For both clays, experiments showed that the increase in injection pressure was accompanied by expansion (negative axial strains), followed by compression strains throughout the dissipation stage coupled with the increase in outflow volume, which is considered the gas breakthrough. The observations are explained by examining the gas pressure-front propagation into the sample, which induces a local increase in the pore pressure and, consequently, a local decrease in the effective stress. For the faster injection rate considered in these studies, the deformation response is delayed, since the pore pressure increase was restricted to the lower boundary of the sample. In contrast, for the lower injection rate considered in these studies, samples expanded during the injection stage, reaching their peak expansion at the maximum gas pressure (the slow pore pressure increase propagated into the sample reaching minimum local effective stress). Furthermore, a distinct effect of bedding orientation is observed for Boom Clay: BC⊥ systematically showed higher expansions regardless of the injection rate, since the material is less constrained from expanding due to the oedometer conditions compared to the samples with bedding planes parallel to the flow and the BC anisotropy (Figures 4-9 and 4-10).

Figure 4-9 – Boom Clay samples with gas flow (A) perpendicular and (B) parallel to bedding as tested in oedometer cells by Gonzalez-Blanco et al. (Gonzalez-Blanco et al., 2022; Gonzalez-Blanco and Romero, 2022).

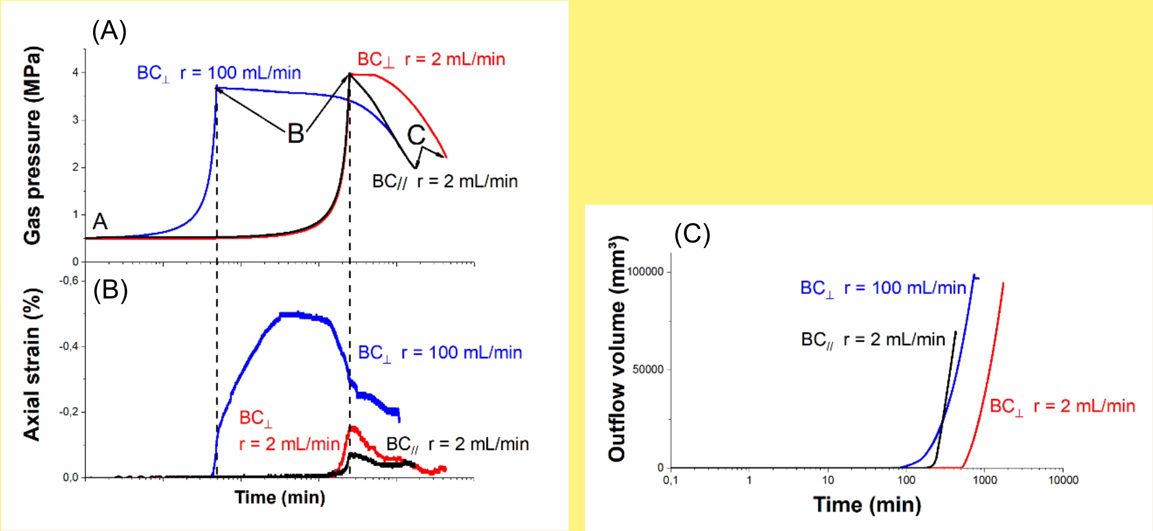


Figure 4-10 – Gas injection experiments on Boom Clay samples in an oedometer cell with gas flow respectively parallel and perpendicular to bedding, using controlled volume rates of respectively 2 and 100 mL/min (Gonzalez-Blanco et al., 2022). Time evolution of (A) gas pressure at the injection point (MPa). (B) Average axial strain (%). (C) Outflow volume (mm^3).

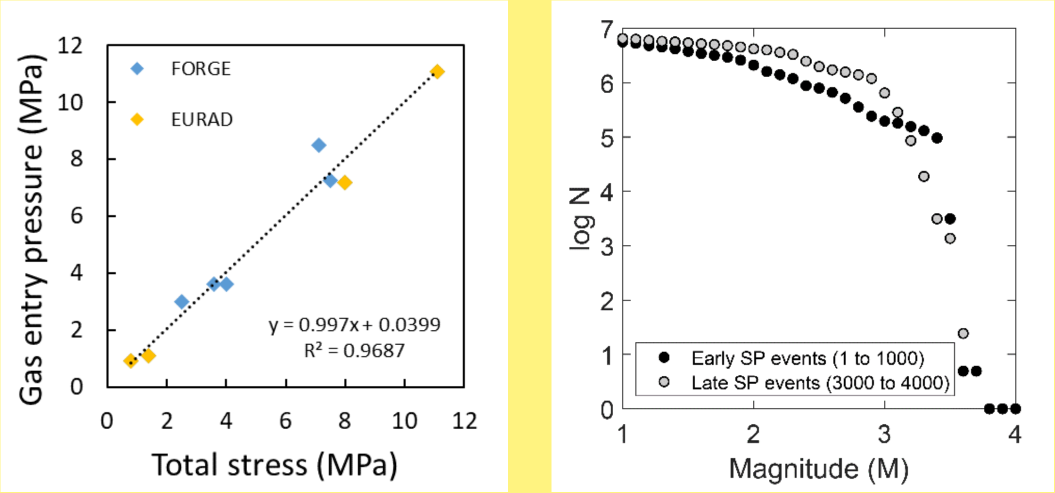
In the particular case of compacted bentonite, ÚJV, CTU and CIEMAT confirmed during EURAD-GAS, through different experimental campaigns on BCV, MX-80 and FEBEX bentonites, that breakthrough events, and so the formation of gas pathways, occurred at pressure levels that lie in the range of the swelling pressure for the considered dry density of the bentonite, or even higher, and always exceeded the value of the total axial stress in the test cell. These observations are in line with previous studies on MX-80 bentonite such as (Horseman *et al.*, 1999; Harrington and Horseman, 2003; Graham *et al.*, 2012). By analyzing the perturbation of the stress field occurring during a breakthrough event in MX-80 bentonite, BGS makes it possible to quantify the development of gas-induced deformation in term of magnitude, scale and mode of deformation during gas pathway dilatancy, as well as the degree of self-sealing (Box 6, page 40). This approach developed by BGS during EURAD-GAS could be of particular interest for improving predictive simulations in the future.

Visco-capillary two-phase flow **models**, in their simplest forms, are generally not able to reproduce the results of gas injection experiments. When the gas injection pressure increases and the material is subjected to deformations, the introduction of hydro-mechanical coupling linking the permeability and the retention curve to deformation is often required to capture (a posteriori) the development of preferential pathways. This can be achieved through an extension of classical visco-capillary two-phase flow models, either with stress and pressure-dependent global transport parameters or through the explicit representation of discrete dilatancy-controlled gas pathways (Box 7, page 42) (see also for instance Pitz *et al.*, under review - 2; Gerard *et al.*, 2014; Radeisen *et al.*, 2022; Corman *et al.*, 2022; Liaudat *et al.*, 2023). However, the predictive capabilities of these numerical models are inherently limited because the actual development of dilatancy-controlled gas pathways fundamentally depends on the microscale heterogeneity of the material.

Box 6 – Stress field disruption allows development of gas-driven deformation in compacted clay to be quantified.

Multiple experimental programs document the occurrence of gas flow in clays above a critical threshold (Pusch *et al.*, 1985; Horseman *et al.*, 1997; Tanai *et al.*, 1997; Gallé and Tanai, 1998; Horseman *et al.*, 1999; Sellin, 2014; Graham *et al.*, 2016; Shimura *et al.*, 2017; Levasseur *et al.*, 2021), which has been shown to relate to the internal stress state of the clay, σ_{ij}, (Horseman *et al.*, 1997; Gallé *et al.*, 2008; Graham *et al.*, 2016). This association was demonstrated for bentonite constant volume gas injection experiments in FORGE EC project and has now been expanded across a wider range of gas entry pressures for bentonite and clay/sand/silt mixtures in EURAD-GAS (Figure 4-11 A). Tests were conducted across a range of very low pressurization rates (in EURAD-GAS: generated from an initial gas volume close to 300 mL and an injection rate of 0.003 mL/min). Despite the observation of mechanical coupling in pathway dilatancy, limited information about the evolution of the microscale changes that occur during gas entry has been available in the past. This is partly due to limitations in resolution for conventional imaging methodologies on a representative scale (Busch *et al.*, 2017; Godinho *et al.*, 2019) and the self-sealing or closure of gas pathways after testing. However, work conducted in EURAD-GAS using a heavily-instrumented monitoring system during these gas injection experiments shows that first order fluctuations in the stress field can provide this information (Graham and Harrington, 2024).

Using an automated algorithm to detect stress perturbation (SP) events, the accumulation of microscale deformation can be assessed. This approach has the advantage that it can be conducted in real-time, under pressurized conditions, before pathway closure or sample damage due to depressurization can occur. These stress field disturbances show many similar features to those resulting from microfracturing in rock compression tests, including a rapid increase in occurrence before gas outflow and a power-law frequency-magnitude distribution (Figure 4-11 B). This latter observation indicates gas migration through a network of many pathways (100’s-1 000’s in a single test), with a wide range of sizes. The slope of this distribution changes with time, demonstrating evolution of the gas pathway network during progression to major gas outflow.



(A) (B)

Figure 4-11 – (A) Gas entry pressure versus total stress for constant volume gas injection tests on compacted clays in FORGE EC project and EURAD-GAS. (B) Frequency-magnitude distribution of first order perturbations in the stress field during gas injection into a compacted clay (Graham and Harrington, 2024).

Furthermore, source analysis of SP events also provides evidence of the micromechanisms involved in pathway dilatancy (Figure 4-12). A strongly dilatational behavior is apparent as gas enters the clay (polarity = positive), followed by compressional changes once gas is able to escape (polarity = negative). This directly demonstrates the capacity for gas pathways to open during gas pressurization and close on depressurization. This approach, therefore, provides new quantitative data on the magnitude, scale and mode of deformation during gas pathway dilatancy, as well as the degree of self-sealing, which can be used to inform predictive simulations in future.

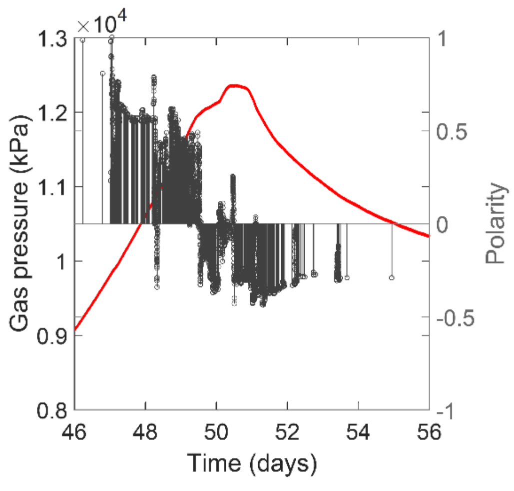


Figure 4-12 – Gas pressure evolution (red line) during gas entry and breakthrough in bentonite testing. An additional metric (polarity) relates to the degree of dilatation (positive polarity) or compression (negative polarity) measured in individual stress perturbation events detected over this period (Graham and Harrington, 2024).

Box 7 – In the frame of EURAD-GAS, the modeling of dilatancy-controlled gas pathways has made great progress.

One of the outcomes of the FORGE EC project was that the visco-capillary two-phase flow alone was not sufficient to explain the gas transport in clay materials. Dilatancy-controlled gas pathways flow has been identified as a major transport mechanism in clay (Shaw, 2013). The mechanism of dilatancy-controlled gas flow arises when gas pressure triggers localized consolidation or creates microfractures. This process effectively amplifies the local porosity, resulting in a marked increase in permeability and a decrease in the gas entry pressure value (Cuss *et al.*, 2014). In the frame of EURAD-GAS, the modeling of dilatancy-controlled path flows was investigated in various ways.

In a first approach, the simulation of gas transport in clayey materials was done using three types of permeability models describing permeability evolutions with pore gas pressure and deformations: (i) In a failure-index permeability model, a stress-based criterion is defined. Using this criterion, a gas flow transport in the continuum model is enhanced, when a threshold value is exceeded. (ii) In a strain-dependent permeability model, it is assumed that microfractures due to dilatancy path flow can be related to the mechanical strains. (iii) In a gas pressure-dependent permeability model, dilatancy-controlled gas flow takes place above a threshold gas pressure. To validate these models, a gas injection test in Opalinus clay performed by Popp *et al.* (2007) was modeled by BGE and BGR. In this test, a reversible dilatant pathway was formed in the clay sample upon gas injection. The modeling of this test (in OpenGeoSys) under single- and two-phase-flow conditions using the developed permeability models shows a good agreement with experimental results with best results obtained for the strain-dependent permeability model (Figure 4-13). The two others were not able to reproduce the reversible behavior of the sample upon gas pressure. Based on the obtained results, it can be concluded that **the dilatant-controlled gas transport can be captured numerically through the introduction of permeability models that depend on the mechanical strains induced by the gas pressure in the clay material**.

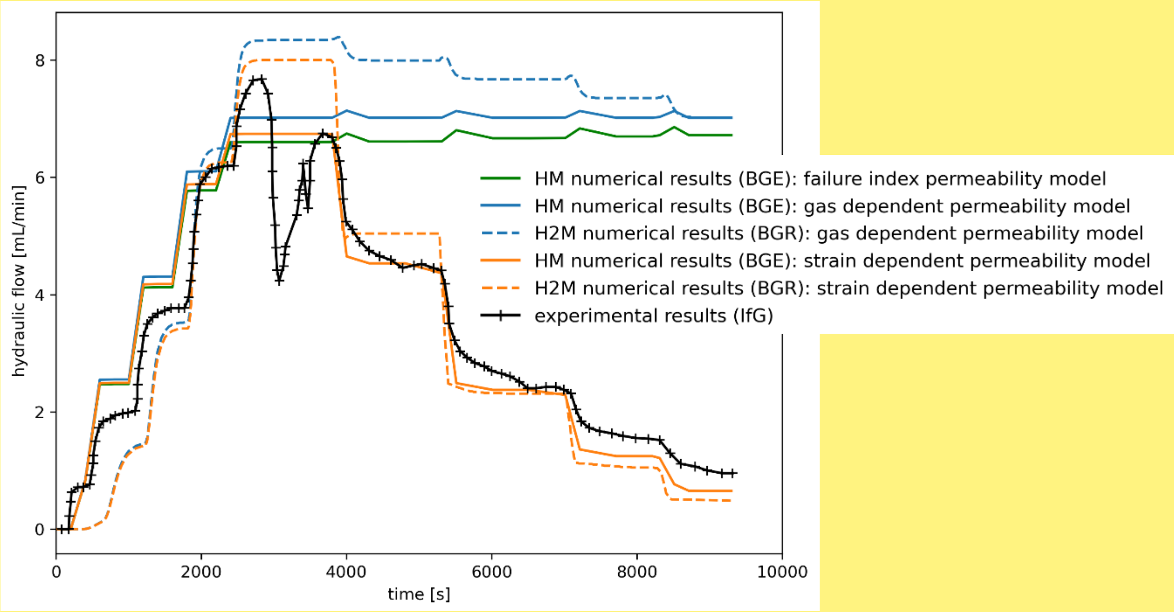
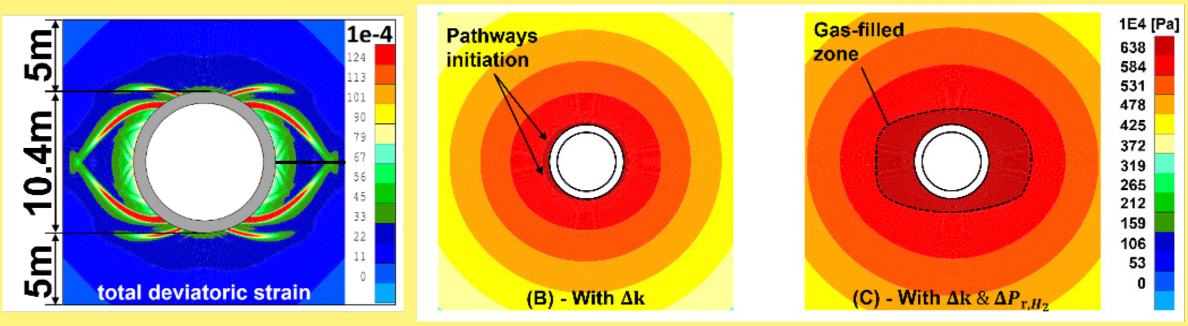


Figure 4-13 – Modeling results and comparison with experimental data of two gas injection tests with different permeability models implemented in OpenGeoSys.

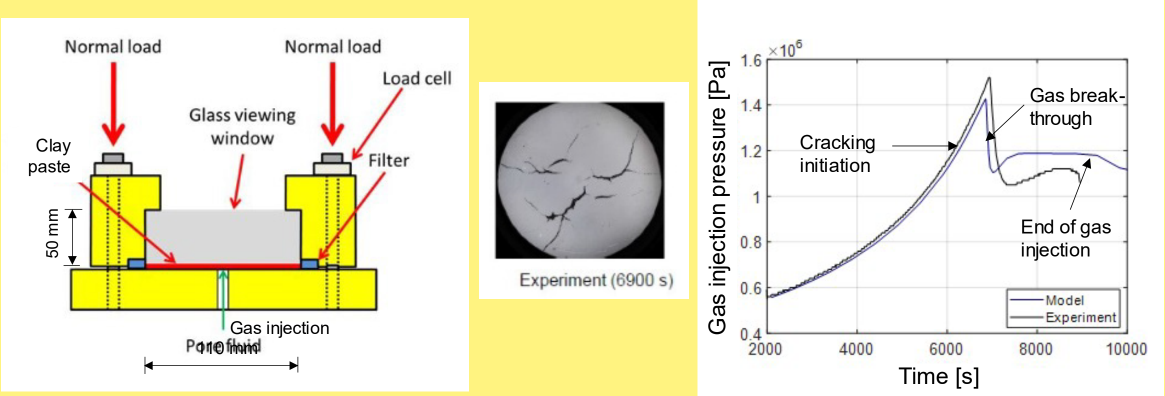
In a second approach, the gas transport in clay materials was done by ULiège (Corman *et al.*, 2022) with a specific focus on the impact of the EDZ on the gas transport kinetics. This numerical tool includes a regularization technique of second gradient type, in order to properly reproduce the fracturing process characterizing the EDZ, with strain localization in shear band mode. It also incorporates the features of a two-phase flow transport. Given the important transport properties modifications induced by the damage, specific coupled effects linking the intrinsic permeability and the gas entry pressure to strain development, are additionally taken into account. These tools are implemented in the finite element code LAGAMINE. This model has been applied to the long-term modeling of a disposal gallery with a diameter of 10.4 meters excavated in the Callovo-Oxfordian claystone, following the French concept. A complete constitutive HM model encompassing viscoplastic effects is employed to reproduce the claystone behavior. Figure 4-14 (A) presents results of a full characterization of the development and shape of the EDZ induced by the claystone deconfinement imposed by the excavation. This process ends up with a clear and well-constructed shear band pattern which initiates in the zone under plastic loading around the gallery and is associated with the anisotropy of the material and of the initial stress state. Considering first a reference simulation without these advanced HM interactions in the EDZ, only a slight desaturation of the claystone mass over a small radial distance occurs as soon as dissolved hydrogen in the water phase is not sufficient enough to ensure transport of gas under the largest amount of released gas, leading to the creation of a distinct gas phase. If the evolution of permeability with strains is taken into account, then hydrogen propagates more efficiently in the EDZ, as displayed in Figure 4-14 (B). More specifically, preferential flow paths corresponding to the localized shear bands seem to initiate around the gallery/drift due to the high increase in permeability within these discontinuities where the deformation is concentrated. Finally, accounting for the reduction of gas entry pressure with strains in Figure 4-14 (C) facilitates even more the penetration of gas into the claystone. It follows that desaturation is amplified by the cracking process in the EDZ. This way, hydrogen is no longer dissolved in water but is almost only transferred in the gaseous state. Under such a desaturation sequence, it is especially possible to model the propagation of gas in the form of a uniform front across the whole zone affected by the hydraulic properties' modifications, which is more in line with experimental observations. Incorporating these advanced HM couplings in the modeling therefore leads to a more realistic and accurate reproduction of gas transport through the EDZ.



(A) (B) (C)

Figure 4-14 – Development of the EDZ (A). Gas pressure around the gallery after 100 000 years with variation of permeability with strains (B) and with variation of permeability and of gas entry pressure with strains (C).

In a third approach, TU Delft developed a model for gas transport in clays with explicit representation of gas-driven fractures (Liaudat *et al.*, 2023). The model uses a fully coupled Pneumo-Hydro-Mechanical (PHM)—also called two-phase hydro-mechanical (TH^{2}M) model—formulation in 2D and is implemented in the finite element code LAGAMINE. Two different types of finite elements are used: continuum and zero-thickness interface elements. Continuum elements are used to represent the mechanical and flow processes in the bulk clay material and interface elements are used to represent cracks. Interface elements are introduced a priori in between the continuum elements to provide potential cracking paths. The model parameters are such that, as long as the interface elements remain closed, they do not have any significant effect on the response of the model. However, when a certain mechanical threshold is reached (e.g., the tensile strength) and the interface element starts to open, localized mechanical and flow processes are triggered. The model has been used to simulate gas injection experiments performed by BGS on Boom Clay samples using a novel testing setup (Kolditz *et al.*, 2024; Marschall *et al.*, 2024). This setup makes it possible to induce and observe the formation of "2D" cracks in clay-rich low-permeability materials by the injection of gas or water (Wiseall *et al.*, 2015; Wiseall *et al.*, 2019). In this setup, a thin layer (~1 mm) of clay paste is compressed between a 110 mm diameter sight glass and a steel plate, while it is held laterally in place by a ring filter at atmospheric pressure (Figure 4-15 A). Then, gas is injected at the center of the lower platen at a controlled volumetric rate while the cracks induced are registered by a camera through the sight glass (Figure 4-15 B). Additionally, the evolution of the average vertical stress is monitored by means of donut load cells placed in series with the bolts used to compress the sample. In order to estimate the initial state variable (stress, porosity, etc.) of the clay sample at the beginning of the gas injection, preliminary simulations of the clay sample consolidation upon the vertical loading were performed. These simulations indicate that the sample develops non-uniform radial profiles of state variables upon loading and that this non-uniformity increases with loading rate. The subsequent simulation of the gas injection test show that the model can reproduce the initiation and propagation of gas cracks, as well as the gas injection pressure and average vertical stress variations with time (Figure 4-15 C). Unfortunately, qualitative aspects of the cracking pattern are not well reproduced. This could be attributed to the high uncertainty on the initial state variables as well as to the intrinsic randomness of a cracking when the minimum and the intermediate principal stress are equal.



(A) (B) (C)

Figure 4-15 – (A) BGS Experimental gas injection setup and (B) experimental crack pattern (Marschall et al., 2024). (C) Experimental and numerical evolution of the gas injection pressure with time (as modeled by TU Delft) (Kolditz et al., 2024).

4.2.3.3 Uncertainties and knowledge gaps

The compilation of the outcomes from several decades of investigations of gas transport through clayey materials reveals a great diversity of experiments and results in terms of observed phenomena in a wide range of thermo-hydro-mechanical (THM) state conditions. Comparison of experimental results is not trivial particularly due to the lack of experiment reproducibility and/or replication. One of the challenges is to dissociate the deterministic and stochastic parts of processes and features of gas transport. Some commonalities in gas transport responses have been explored in the frame of EURAD-GAS and DECOVALEX 2019 and 2023 [11] and should be further developed. In addition, beyond the passage of gas, the actual transport mechanisms are most often only observed indirectly, through its effects in terms of e.g., measured changes in stress and strain, permeability of the materials or postmortem characterization of the porosity by tagging gas with nanoparticulates. By consequence, a clear mechanistic understanding of the gas flow processes is still desirable to properly bound the effects of these processes using simple and robust descriptions. Significant progress has been made in the context of EURAD-GAS by comparing the impact of experimental procedures and experimental setups on the test results (Box 8, page 46). Work is still underway to properly compare, evaluate and improve advanced test procedures and setups for clayey materials.

Reducing uncertainties in terms of experimental characterization and understanding of gas transport processes has to be coupled with numerical model developments. However, the modeling of gas transport in saturated clayey materials is still a challenging task, for the following main reasons:

- Gas transport in a water-saturated clay sample is controlled by the microstructure, the connectivity of the macropores and the heterogeneities. Clear progress has been made by introducing these aspects in enhanced hydro-mechanical models. However, pore scale modeling generally suffers from the lack of complete characterization of the pore structure. In the future, it could be interesting to further develop pore scale simulations (DNS)[12]. Methods such as Lattice Boltzmann (LBM) or Smoothed Particle Hydrodynamics (SPH) allow easy treatment of multi-physical coupling while accounting for detailed pore space geometries. They can form the first level of multistage upscaling procedures based on both experimental data and novel numerical approaches such as neural networks, reduced models, etc.
- Lack of model predictability. Enhanced hydro-mechanical models are able to reproduce gas transport processes from laboratory tests. However, parameters of such models are generally tuned in order to mimic the experimental response. To avoid this and improve the predictive capacity of models, the concepts of visco-capillary two-phase flow and dilatancy-controlled gas pathways still require further refinement to better capture the development of gas pathways, its coupling to the stress field and its consequences in term of permeability evolution. Additional experimental evidence at the laboratory- and field-scales are also required to limit the uncertainties of the existing process and parameter at various scales and conditions.
- Upscaling: while dilatancy-controlled gas pathways models are very useful to reproduce the experimental data at the laboratory scale (centimeter scale), the visco-capillary two-phase flow approaches are generally preferred for in situ tests (meter scale) and repository-scale modeling as the effects of pore structures are less pronounced at that scale and the response is less constrained by experimental boundary conditions. The transition between the laboratory scale and in situ test and their modeling approaches is not straightforward. In this respect, the coarseness of model meshes decreases from laboratory to field scales, which means that further improvement in the identification key processes that span both scales is important. This

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[11] https://decovalex.org/D-2023/task-b.html. Last accessed: 2023-12-07.

[12] Today, these approaches are computationally demanding. This currently limits the size of the simulation domains that can be processed and makes it difficult to scale up without simplifying/losing process details.

echoes the importance of identifying the deterministic and stochastic features associated with gas transport and how best to estimate and incorporate them into models.

Box 8 – Work is underway to evaluate and improve advanced test methods for clayey materials.

A systematic comparison of gas invasion test methods in three experienced laboratories (BGS, CIMNE and EPFL) was initiated during EURAD-GAS. The motivation for this work is the apparent discrepancies in the results obtained by the three teams, which may lead to slightly different conceptualizations of gas transport in clayey materials. The aim of this comparison is to evaluate and improve advanced test methods for clayey materials (both clay host rocks and bentonites). Most experimental facets are being scrutinized:

- Gas testing equipment (type and sample size, sensors and auxiliary devices).
- Testing protocols and procedures (pre-conditioning, hydro-mechanical paths prior to gas injection, water permeability, water/gas exchange, gas injection features, gas injection protocol, resaturation protocol, second gas injection, unloading protocol).
- Sample preparation and characterization protocols (supply of core material, sample preparation, interim storage of sample, size prior to testing, pore fluid, analysis of off-cuts, post testing analyses).

Specific experiments are also compared.

At the time of writing, it is too early to present the results of this comparative work that is ongoing. Subsequent phases of this work could include the development of a common evaluation concept based on the feedback from the participating experts, and a "round robin" type laboratory program (beyond EURAD-GAS). The benchmark should be extended to other laboratories.

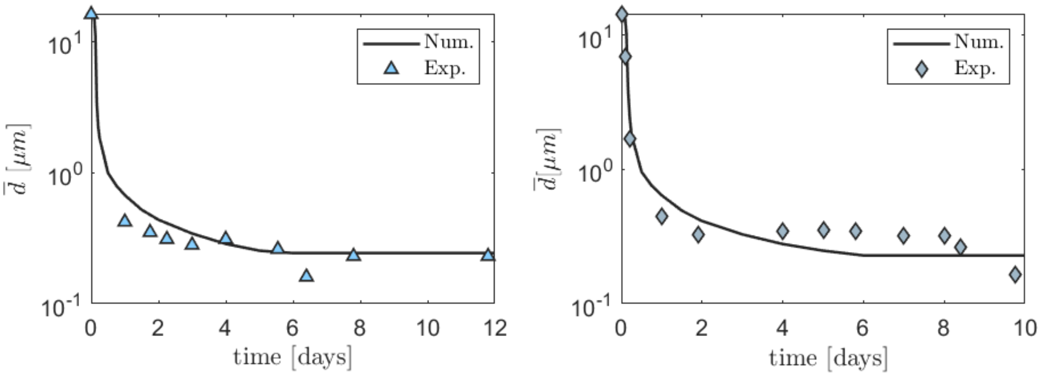
4.2.4 Self-sealing after gas transport

The principles that underpin self-sealing mechanisms are well understood for the three studied host rocks (BC, OPA and COx) and bentonite-based materials used for engineered barriers. Self-sealing depends on (thermo-)hydro-mechanical and chemical processes, its swelling capacity (resulting from the mineral composition of the material, the initial void ratio, and composition of the pore fluid) all of which is coupled to the prevailing stress conditions. Mechanical closure of fractures (e.g., crack closure, fracture sliding), hydro-chemical interactions of the pore water with the clay-bearing solid phase of the geomaterial (e.g., swelling, disaggregation) and colloidal transport processes (e.g., sedimentation, clogging) have been identified as typical self-sealing mechanisms in clay-rich materials. A summary of these mechanisms is given in SOTA 1 (Levasseur *et al.*, 2021).

Pathways that have been created for gas transport are expected to close and seal once the gas pressure is released. Indeed, gas tests in clayey materials have shown that the hydraulic conductivity after the passage of gas is not significantly different to initial values prior to gas flow (Harrington, Cuss *et al.*, 2017; Zhang and Talandier, 2023). This indicates that after the creation of gas pathways, the rock retains a capacity for self-sealing and that the conditions for this sealing (stress relief and water availability) are still met when gas pressure decreases (Harrington *et al.*, 2019; Gonzalez-Blanco *et al.*, 2023; Graham and Harrington, 2024).

The recovery of the hydraulic conductivity does not mean that some gas transport properties are not permanently modified by the passage of gas. In particular gas entry pressures may still be lower in the sealed zone than in the intact clay (Gonzalez-Blanco *et al.*, 2023). In that sense, self-sealing capacity of clayey materials is preserved for what concerns hydraulic conductivity but not necessarily with respect to gas entry. Should gas pressure rise again, it is possible that pathways will be reactivated at a lower threshold pressure (Horseman and Harrington, 1994; Harrington and Horseman, 2003). In the particular case of bentonite materials, CTU has shown however that the longer the resaturation time after gas passage is, the better the recovery of bentonite favorable properties (Kolditz *et al.*, 2024; Marschall *et al.*, 2024). At the time of writing, it is still unclear what would be the effects on gas entry pressure of repeated gas entry over long timescales.

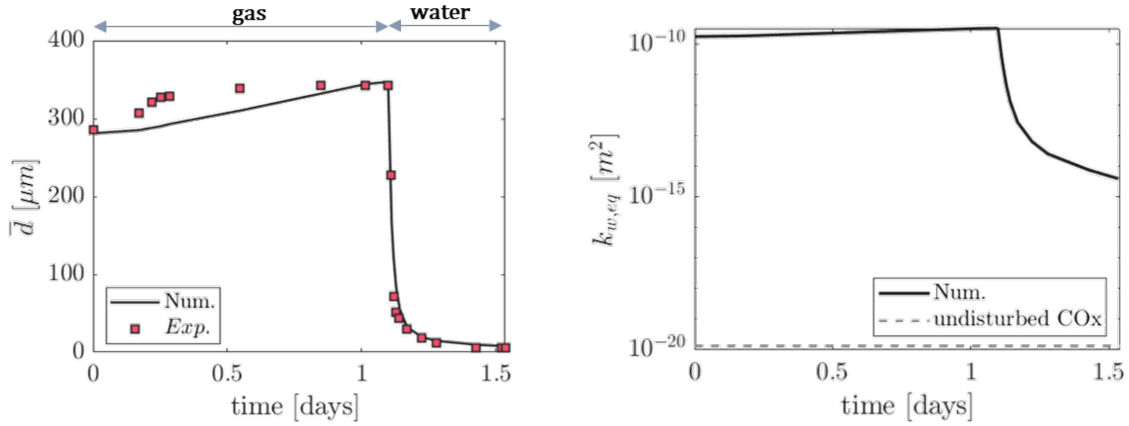
(Quacquarelli *et al.*, under review) proposed to model self-sealing capacity of clays by introducing a new constitutive model for an interface element reproducing the sealing effects observed in a fracture. This model takes into account the existence of damaged zones along the interface surface, where the swelling phenomenon will concentrate quickly during hydration. The hydro-mechanical parameters of the damaged zones have been calibrated against sealing experimental results obtained by Wang *et al.* (2022) on Callovo-Oxfordian claystone (Figure 4-16). The model is able to capture the rapid closure of the fracture during wetting, which stabilizes after a few days.



(A) (B)

Figure 4-16 – Temporal evolution of the average fracture opening during the wetting test, comparison between numerical and experimental results obtained by Wang et al. (2022) on COx: (A) Test UA3-C1; (B) Test UA3-C2.

This model has also been used to model more complex experiment on COx comprising gas injection followed by a water injection step (Figure 4-17) (Di Donna *et al.*, 2022). During gas injection, the facture aperture is actually increasing due to the desaturation of the damaged zones. On the contrary, when water injection starts, a rapid closure of the fracture is observed both numerically and experimentally.



(A) (B)

Figure 4-17 – Temporal evolutions of (A) the average fault opening $\bar{d}$ and (B) the water permeability within fault during gas injection followed by water injection phase, test 8182 (Di Donna et al., 2022).

4.3 Expert judgments / Critical overview

The concepts and mechanisms of gas transport through clayey materials are now widely accepted and supported by a significant body of experimental evidence. Nevertheless, the phenomena and features associated with gas-induced damage evolution in clayey materials remain difficult to predict. The limited predictability of localization phenomena in geomechanics concerns both the locus of damage initiation and the propagation of damage in space and time. There are two main reasons for this limitation:

- Microstructural information (e.g., small-scale variability of stiffness and strength or undetected microfractures) that is incomplete by nature.
- Irreducible uncertainties in the prevailing initial and boundary conditions (e.g., on actual stresses and pressures at the local scale, beyond average field values).

It is therefore still a challenge to upscale laboratory scale results to behavior at in situ scale (Daneshy *et al.*, 2004; Marschall *et al.*, 2005; Marschall *et al.*, 2013; de la Vaissière, Gerard *et al.*, 2014; de la Vaissière *et al.*, 2019; de la Vaissière *et al.*, 2020). While comprehensive theoretical frameworks for modeling gas-induced damage propagation in clayey materials have been developed, effects of the mineralogical variability and/or microstructural heterogeneity of the clays cannot be easily anticipated at large scale. Further work can be envisaged to better capture the effects of inherent parameter uncertainty of the gas transport mechanisms. For instance, more probabilistic approaches to these models could be an option given the inherent uncertainty in parameterization. Statistical approaches (e.g., variograms) may also be used to try to represent heterogeneities. Furthermore, large-scale in situ experiments (repository-scale or half-scale tests) are valuable tools that should be carried out, as they can provide additional data for the proper transposition of key gas transport concepts into repository simulations (as proposed in Chapter 5). While sensitivity analyses and statistic approaches may form a useful part of future model development, it is also essential to continue to gather evidence, through experiments supported by mechanistic models, of the fundamentals of pathways creation and propagation. That is, that pathway creation and propagation are mainly tied to local deformations and that water displacement, if any, is predominantly lateral, not in the direction of propagation.

5. Understanding of the gas transport and impacts at repository scale

Having explored in Chapter 4 which transport mechanisms prevail under which range of conditions and understanding how the coupling between liquid and gas phases pressure and the solid phase stresses controls gas transport, this chapter focuses on the impact of gas on the functioning of a geological disposal system. To support this analysis of gas impacts at the repository scale, the drawing of Marschall *et al.* (2005) (Figure 4-1) is revisited with the perspective of gas flowing from a disposal gallery into a clayey host rock (Figures 5-1 to 5-4). In this context, each transport mechanism is first reviewed (Section 5.1). Then, potential consequences in terms of barrier integrity (Section 5.2), conceptualization of gas transport at the repository scale (Section 5.3), and long-term repository performance and radionuclide transport (Section 5.4) are discussed with respect to the state of knowledge on gas transport at the end of EURAD-GAS.

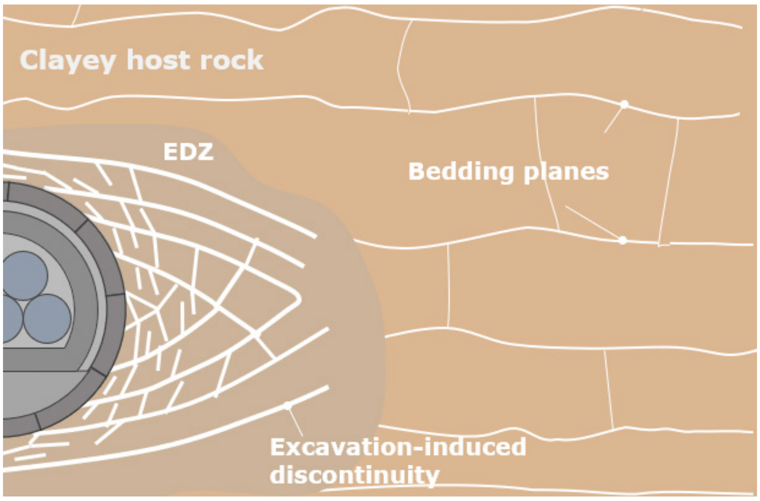


Figure 5-1 – Schematic representation of a gallery, excavation damaged zone (EDZ), with its discontinuities, and clayey host rock, with bedding planes. The gas flux originates from the gallery. The anisotropy of the material and/or the anisotropy of in situ stresses determine the shape of the damage pattern. Excavation-induced discontinuities, even if sealed, and bedding planes can be preferential pathways for gas transport.

5.1 How gas moves through the system

5.1.1 Diffusion of dissolved gas in water-saturated clay

The diffusion of dissolved gas in water-saturated clay is well understood ((Figure 5-2) and (Jacops *et al.*, 2024)). This transport mechanism is always certain to occur, but the solubility of hydrogen being low, the capacity of gas transport by diffusion of dissolved gas in water-saturated clay is limited. The determination of diffusion parameters for dissolved gas through a broad range of clayey materials under various mechanical conditions and, in the case of clayey host rocks, along different directions relative to the bedding, is well developed and mastered. The experimental setups yield reliable parameter values for water-saturated materials (Section 4.2.1). In particular, uncertainties are limited in saturated (or close to saturation) host rocks. In clayey EBS materials, which are not initially saturated at the time of repository closure, significant uncertainties remain on the duration of the saturation phase, which depends on the interplay between water inflow from the host rock and evacuation of gas, initially present or produced after closure. This is thus system specific. Hence, diffusion through partially saturated EBS materials should be further investigated in such cases.

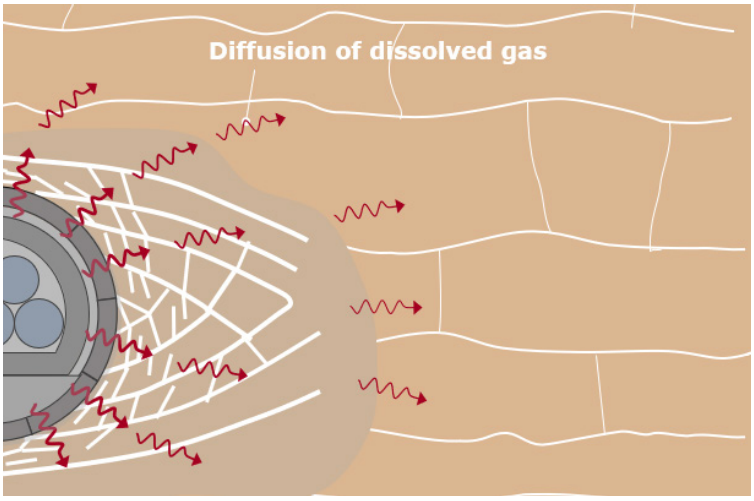


Figure 5-2 – Diffusion is driven by concentration gradient. Depending on the host rock, the diffusion coefficient might be anisotropic.

5.1.2 Gas sorption on clay minerals

In repository conditions, adsorption[13] of hydrogen on clayey materials should be neglected.

5.1.3 Visco-capillary two-phase flow

There is a consensus that visco-capillary two-phase flow of gas and water is a priori possible through the porous media and/or through discontinuities (Figure 5-3). For what concerns geological disposal systems, a distinction is made between clay host rocks and EBS materials:

- For the host rocks studied in EURAD-GAS, it is expected that, if visco-capillary two-phase flow develops, it will be mainly localized within the EDZ and in particular within the discontinuities of this EDZ (Levasseur *et al.*, 2021). There is no clear evidence of significant visco-capillary two-phase flow through mechanically undisturbed host rock.
- In the clayey components of the EBS such as buffers, plugs and seals, which are either partially saturated or have a high sand content, visco-capillary two-phase flow may develop[14], or not, depending on the degree of saturation, the composition of the buffer (i.e., the percentage of sand), the availability of water, and the long-term homogeneity[15] of the materials that affect the gas entry pressure (Jacops *et al.*, 2024) (Villar *et al.*, 2012; Sellin and Leupin, 2013). It is thus very concept and material specific.

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[13] In addition, some irreversible reactions, such as carbonation of cementitious materials and conversion of hydrogen gas to methane by microbes, are expected to reduce the net quantity of gaseous molecules. Such gas consuming reactions are however out of scope of this report.

[14] It cannot be excluded that in some systems, the EBS with limited water content may initially offer continuous gas pathways that will remain so until gas production becomes sufficiently low for resaturation to proceed.

[15] Or homogenization, e.g., during the saturation of a bentonite barrier.

The theoretical framework for visco-capillary two-phase flow is well established and broadly implemented in many modeling tools. These have shown to be capable of reproducing experimental results, at least qualitatively a priori, or quantitively through a posteriori calibration (Tamayo-Mas *et al.*, 2021; Corman, 2023; Corman and Collin, 2023). All experiments typically involved gas injection rates that were higher than those that would be expected from the slow gas production processes in a repository. This induces significant uncertainty. Indeed, to capture the dependence of experimental results on injection rates as previously demonstrated in several European programs (Rodwell, 2000; Gonzalez-Blanco, 2017), new calibration of model parameters may be needed. To elucidate this matter, very slow injection tests have been set up in the framework of EURAD-GAS, but the results of these long-term experiments are not yet available at the time of writing. This highlights the difficulty in assessing the long-term behavior of gas in disposal systems. Another source of uncertainty is the role of material heterogeneities in the initiation and development of visco-capillary two-phase flow. A scale effect has been evidenced, with laboratory tests on small samples being more sensitive to the presence of heterogeneities, which are not explicitly represented in a two-phase flow model. On large-scale tests, e.g., in situ tests, gas transport within the EDZ is inevitably affected by the presence of discontinuities, which appear to play a similar role to the heterogeneities in the laboratory tests. In modeling at the scale of a disposal system, this is usually treated by homogenization through the use of adapted parameters values (e.g., reduced gas entry pressure) in the visco-capillary two-phase flow model. The uncertainties are thus reflected in the ranges adopted for the values of these parameters (Wendling *et al.*, 2024).

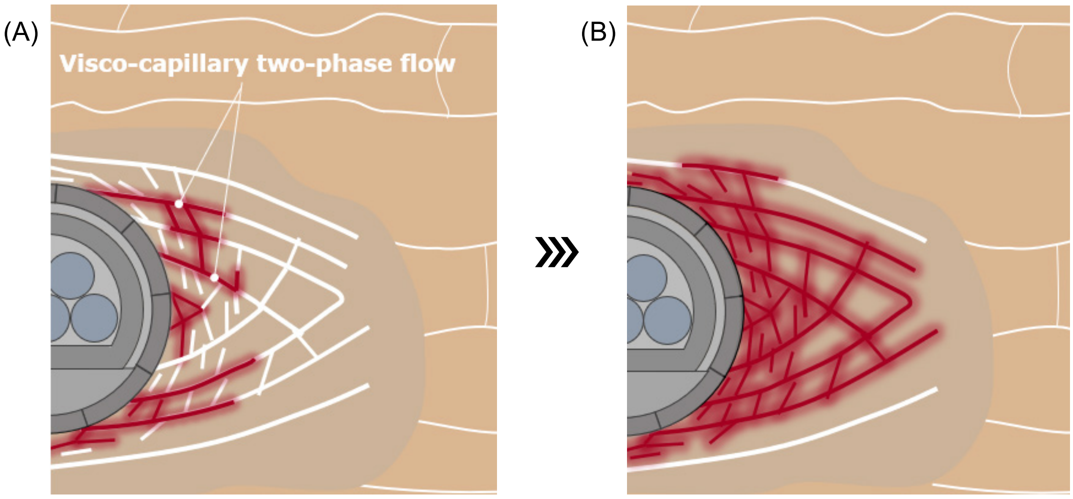

Figure 5-3 – At higher gas pressure, visco-capillary two-phase flow occurs: gas invades the EDZ, starting with discontinuities (from (A) to (B)), in addition to diffusion (not shown). The chevron pattern shows increasing gas flux.

5.1.4 Formation of dilatancy-controlled gas pathways

There is a consensus that the creation of dilatancy-controlled gas pathways in clay-rich barrier materials is possible and controlled by the interplay between water retention, stress-strain behavior and hydro-mechanical coupling. When gas pressure is high enough to overcome the gas entry pressure of the intact rock matrix, but still below the local minimum stress along a pre-existing fracture, gas invasion under drained conditions may happen as a conventional two-phase flow process. If pressure build-up is still too high, continuous pathway dilation would happen. Depending on the disposal concept and/or on the host rock, pathways dilation may form in the EDZ and/or the intact host rock (Figure 5-4). The initiation and propagation of dilatancy-controlled gas pathways mainly depend on the deformation behavior of the clayey barrier and on the evolution of gas pressure build-up (Section 4.2.3)[16]. In the particular case of saturated engineered clay barriers, such as bentonite with moderate emplacement density, dilatancy-controlled gas pathway propagation is expected when the applied gas pressure exceeds the sum of the local swelling and pore water pressure (which is generally close to the local minor principal stress) (Section 4.2.3) (Kolditz *et al.*, 2024; Marschall *et al.*, 2024).

The formation of pathways is understood as the generation of new porosity and/or coalescence of pores as a result of local, gas-induced, stress redistribution (Horseman *et al.*, 1999; Harrington *et al.*, 2003; Graham *et al.*, 2012; Busch and Amann-Hildenbrand, 2013) (Kolditz *et al.*, 2024; Marschall *et al.*, 2024). Depending on the speed of pressure build-up, gas production rate could be balanced steadily by a variation of pressure in the pore network and by the newly created pore volume at the pathway tips. If so, the pathways will be controlled by dilatancy and may propagate within the EDZ. In some cases, it can even extend toward natural flaws in the rock matrix such as bedding planes and tectonic features. These processes increase the exchange surfaces with the rock matrix, giving rise to significant enhancement of diffusive transport. Moreover, very little of the interstitial water present in the pores prior to the formation of dilatant pathways is expected to be displaced along the pathway (Horseman and Harrington, 1994; Rodwell, 2000; Harrington and Horseman, 2003; Graham *et al.*, 2012; Jacops *et al.*, 2014). In experiments where high gas injection rates are associated with rapid gas pressure build-up, gas fracturing might be expected to occur (Section 4.2.3.1) with propagation velocities up to the shear velocity of the material (Valkó and Economides, 1995). However, such high pressurization rates are not expected in normal repository scenarios.

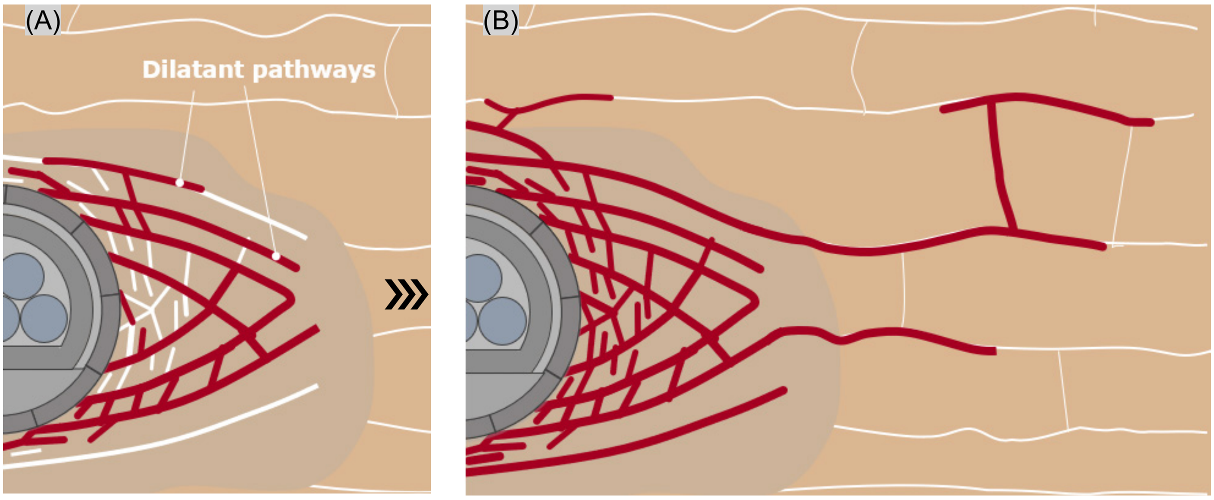

Figure 5-4 – (A) If the pressure continues to increase, dilatancy-controlled gas transport occurs in the EDZ, in addition to diffusion and possibly two-phase flow (not shown). (B) At even higher gas pressure, dilatant pathways can even develop into intact host rock. The chevron pattern shows increasing gas flux.

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[16] As suggested by experimental evidence derived from the materials and conditions studied within the EURAD-GAS and earlier laboratory programs.

5.2 Impact of gas transport on barrier integrity

Of the possible gas transport processes described above, diffusion of dissolved gas is always certain to occur. It is widely accepted that this process has no impact on clay barrier integrity. By definition, visco-capillary two-phase flow in a porous media involves the displacement of the liquid phase (water) by the gas phase without irreversible deformation of the media. Hence, no alteration of the properties of clay barriers materials are expected for that transport mechanism either, although displacement of water and consequent desaturation of the barrier is expected as a result. If, however, it can be shown that no detrimental effects result from this, integrity of the barriers should be preserved for visco-capillary two-phase flow, as well as for diffusion.

The third mechanism, the formation of dilatancy-controlled gas pathways is always associated with minimal displacement of water, but leads to changes in the microstructure of the material, through localized deformation (due to the dilatant 'opening' of pathways and also the localized compaction of the surrounding pores to accommodate this) (Harrington and Horseman, 2003; Graham *et al.*, 2012; Graham *et al.*, 2016; Cuss *et al.*, 2022) (Jacops *et al.*, 2024; Kolditz *et al.*, 2024; Marschall *et al.*, 2024). Nevertheless, it is well recognized that clayey materials exhibit the favorable feature of self-sealing after sustaining mechanical failure (Bernier *et al.*, 2007; Van Geet *et al.*, 2009) and this ability is evidenced in experiments showing episodic gas flow, correlated with increase and decline in gas pressure (Graham *et al.*, 2016) (Box 6, page 40). Also, in the experiments performed and conditions tested within EURAD-GAS, it has been observed that gas pathways close after gas flow ends and water permeability is generally restored. This process has been observed to repeat itself upon subsequent gas injection (Zhang and Talandier, 2022; Gonzalez-Blanco *et al.*, 2023). While it is possible that some of the previous gas pathways are reopened during a subsequent pressurization, experimental work performed in EURAD-GAS also shows their capacity to close again after depressurization (Box 6) (Graham and Harrington, 2024). No evidence for the permanent accumulation of damage has been observed (Kolditz *et al.*, 2024; Marschall *et al.*, 2024), although there is currently limited quantifiable data demonstrating the rate and degree to which self-sealing occurs. Experimental work conducted in EURAD-GAS has, however, led to the further development of methodologies that will facilitate this quantification in future (e.g., Box 6).

Consequently, because of the local nature of the perturbations induced by dilatancy-controlled gas pathways and because of the observed self-sealing, it is expected that the integrity of clay barriers will not be significantly impacted by the passage of gas through this transport mechanism, even in the case that gas would be expelled in a cyclic fashion and locally perturb the stress field (Graham *et al.*, 2016; Harrington, Graham *et al.*, 2017). The evidence base relating to the behavior of dilatancy-controlled gas pathways that could develop along interfaces between clay barriers and other components is less substantial, though this behavior has been observed both at the laboratory and field scales (Sellin, 2014; Cuss *et al.*, 2022). This should be addressed either by reducing uncertainties in the behavior of interfaces or by designing the repository in such a way that this behavior is avoided, or organized[17]. The chosen strategy would be strongly concept dependent but in both cases the objective would be to make the functioning of the system more predictable. The knowledge gained by EURAD-GAS at the end of the project provides a solid knowledge base upon which such efforts can be pursued.

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[17] Interfaces may be one way of managing gas pressure, depending on where they lead and what parts of the disposal systems they negate.

5.3 Conceptualization of gas transport

Today, the knowledge base is sufficient to derive a concept of gas transport in and from a fully saturated GDF built in a clayey host rock after closure. The communication of the conceptualizations of gas transport is based on “gas storyboards”, a sequence of successive steps, as done by ONDRAF/NIRAS, COVRA, Nagra, Andra and IRSN in SOTA 1 (Levasseur *et al.*, 2022). These storyboards highlight the main drivers for the development of a gas transient phase in a GDF, as illustrated for instance in Figure 5-5 for the French concept developed by Andra. The storyboard generalized to a generic radioactive waste repository in a clayey host rock presents the following steps:

1. Rapid saturation (a few decades) of the access shafts and ramps up to the access seals by water from overlying aquifers. Because of these seals, there is little or no water flow from this path to the rest of the repository at this stage.

2. A slow and radial saturation of the engineered barrier system (EBS) in the repository galleries by the waters of the host rock, limited by its low permeability. Inflow through the now saturated access seals is also very limited.

3. During the saturation of the EBS, no significant desaturation of the host rock is expected beyond the EDZ because of the high gas entry pressure prevailing in the intact rock. Air initially present in the porosity of the EBS materials and the technological voids is expected to dissolve in pore water (in the case of oxygen, part of it will be consumed by oxidation processes).

4. Gas, mainly hydrogen, is continuously produced within the repository. A first important source is the anaerobic corrosion of metals. Metals are present in waste packages, but can also be present in other EBS components, depending on the repository design (e.g., metal lining of disposal cells, reinforcement of concrete gallery lining) and in some waste forms (e.g., claddings). It is to be noted that full saturation is not necessary for corrosion to take place, the presence of humidity in a gas phase is sufficient. Hence gas production is already active during the EBS saturation phase[18]. Radiolysis and decomposition of materials found in low and intermediate waste can also produce large amounts of gas, again mainly hydrogen but also methane in the case of microbial activity (Norris, 2013; Small, 2019).

5. Gas dissolves into the pore water and diffuses away, through the EBS and the host rock. If the gas production exceeds the system’s capacity for dissolution and diffusive removal, it can prevent the engineered barriers (and possibly the EDZ) attaining full saturation for a very long time or can induce and maintain partial desaturation if full saturation was reached earlier.

6. The intact host rock remains almost saturated with water due to its very high gas entry pressure. Hence, if a gas phase persists or reappears in the system due to gas production, it will be confined to the near field and only dissolved gas will migrate into the far field. Desaturation, if it is considered to happen, would be restricted to the immediate vicinity of the galleries (meter scale), and saturation degree would not decrease by more than a few per cent from full saturation in that zone.

7. The fraction of the produced gas that does not dissolve in the surrounding pore water can move through the galleries toward the repository access structures, also increasing the contact surface between the gas and liquid phase, favoring further dissolution. An expanding gas phase is not expected to displace large quantities of water along the galleries as water is more easily pushed into the immediate surroundings. Because a limited desaturation of the EBS/EDZ is sufficient to obtain high enough transmissivities for gas, the gas phase can extend over long distances within the system. The progress

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[18] In particular, the UK inventory includes waste packages containing reactive metals and water, so gas generation will occur from emplacement to the disposal gallery. The packages are therefore vented, which means that radioactive gases and large amounts of hydrogen can migrate from the waste packages early on, even before resaturation. Generation rates are expected to be highest shortly after the introduction of a cementitious backfill, because of the effect of the curing exotherm on gas generation rates and the rapid transition to anoxic conditions.

of the gas phase through the underground infrastructures can however be slowed down and/or hindered as it encounters seals. The role played by seals varies depending on their specific location in the system. Seals closing disposal galleries are made of a controlled mix of sand and bentonite to allow the passage of gas above a certain threshold while retaining a low hydraulic conductivity. Seals closing shafts and ramps are made of pure bentonite to prevent the passage of both gas and water.

8. Once the gas phase encounters a seal, its pressure will increase further. This will at first result in the expulsion of water from the gallery to the surroundings and so a more desaturation of the underground infrastructures (in particular, the backfill of galleries). At some point the pressure can attain a value at which gas pathways can develop either through the seal or through the EDZ around it and the progression of the gas phase will resume. Gas pathways through the EDZ would take advantage of planes of weakness created during the excavation phase. Depending on the repository concept and the expected quantities of gas to be managed, design requirements could be defined (e.g., properties of the seal such as its gas entry pressure or swelling pressure) to limit the pressurization of the gas phase.

9. Whether the pathways are through the seal or through the EDZ around the seal, they will close once gas pressure decreases due to the self-sealing capacity of clayey materials.

10. Depending on the disposal concept and on the gas production, a significant fraction of the gas, distributed over a long period of time, may eventually reach the geological layers above the host rock through the access shafts and ramps and/or along their EDZ, depending on the performance of the seals. Simulations performed in FORGE EC project and confirmed in EURAD-GAS indicate that the onset of these releases can span from several tens to several hundred thousand years. Additionally, the simulations highlight that the duration of these releases exhibits a similar temporal range.

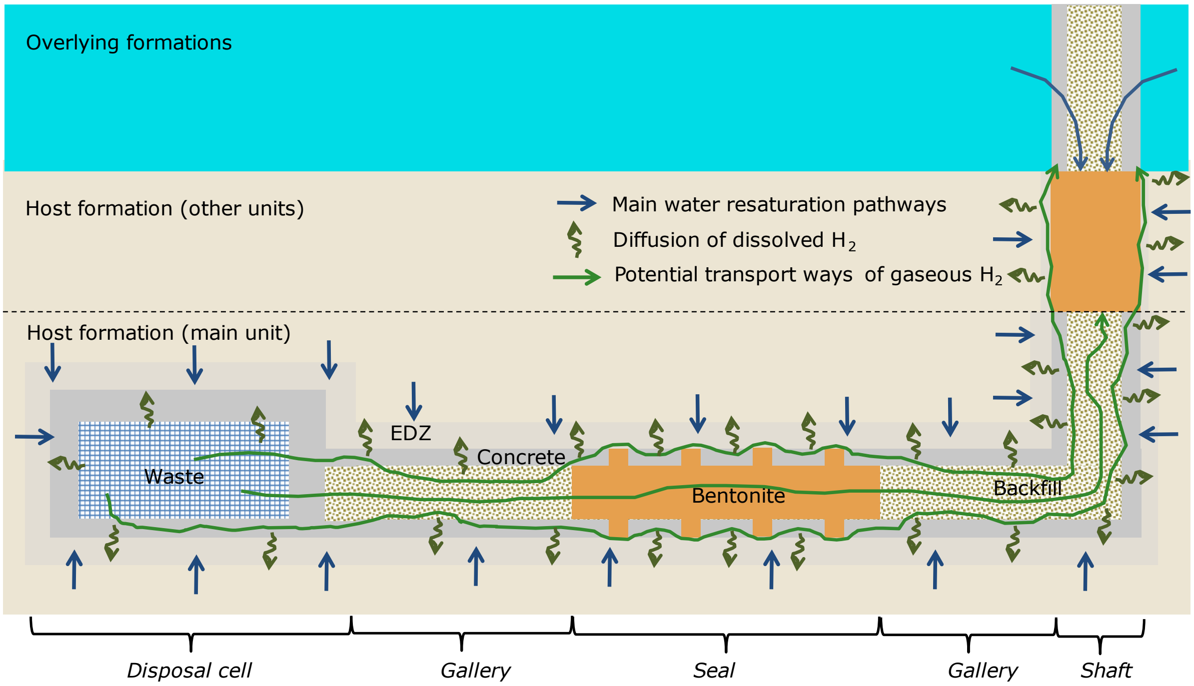

Figure 5-5 – Schematic representation of the Andra's repository concept and of the main phenomena structuring the hydraulic-gas transient (adapted from SOTA 1 (Levasseur et al., 2021)).

5.4 Assessment at repository scale and expert judgment

From the end-users' perspective, the following points should be evaluated at repository scale: gas pressures and gas fluxes throughout the disposal system, gas-mediated radionuclide transport and releases and the long-term integrity of barriers as a consequence of gas pressurization and passage. These points can only be assessed by modeling as space and time scales are too important to be assessed by experimentation. The development of repository-scale models and simulation tools should integrate the available results gained from laboratory experiments to in situ tests, both carried out in representative conditions and scales.

5.4.1 Gas transport modeling at repository scale

The use of repository-scale models, which would explicitly take into account all couplings with the mechanical behavior of the barriers and the development of local gas pathways through clayey materials, is not currently being considered by any WMO due to its complexity and the fact that this capability (while desirable) is not necessary in order to underpin a robust design and safety case.

From current knowledge, the conditions for the activation of different gas transport mechanisms and the capacity for gas evacuation through each of them are sufficiently known to compute gas pressures and fluxes throughout a given disposal system. In line with the conclusions of the FORGE EC project, modeling approaches and tools that have been compared in EURAD-GAS confirm that repository-scale modeling of gas transport explicitly representing all couplings with the mechanical behavior of the barriers and for the development of individual pathways through clayey materials is currently out of reach (Wendling *et al.*, 2024). It will probably remain so in the foreseeable future, both for reasons of computational cost and because of the local nature of gas pathways initiation and propagation (which cannot easily be represented by continuous numerical approaches generally used at repository scale).

Modeling at the scale of the repository is nevertheless possible today using simplified, modular approaches guided by the conceptual gas transport storyboards and based on dissolution, diffusion and two-phase flow phenomena. Smaller scale models can also be built for the different components of the system through which gas can pass through. These component models can explicitly describe the mechanical couplings and provide a better hydro-mechanical understanding of the whole system. Gas pressure evolution and fluxes estimated from these smaller scale models can then be integrated into repository-scale models as complement to the more conceptual storyboard approaches (Wendling *et al.*, 2024).

However, gas pressure deduced from component models must be taken with caution; predictive capability of full-scale coupled process modeling is currently limited due to inherent uncertainties in the development of gas pathways and associated to the simplifications above. Indeed, as gas tends to flow over the whole disposal system toward the shafts and/or ramps, the pressure level assessed by component-scale models may not be representative of the whole repository; it could be either overestimated (limited volume for gas expansion and limited surface for gas dissolution) or underestimated (in the case the modeled component is not the one in which the higher pressure is developed at repository scale) by a few MPa (Wendling *et al.*, 2024).

Modeling on the scale of a repository should therefore be seen more as a tool for bounding the evolution of gas pressures, which is already sufficient to support a robust design and safety cases.

5.4.2 Long-term barrier integrity

From the current state of knowledge, no significant impact on the long-term integrity of clay barriers is expected after the passage of gas provided that the repository is designed in such a way that it favors the progressive release of gas. Thus, not all national programs will require gas-induced damage to be fully predictable, just that there are sufficient safety arguments to demonstrate that gas-induced damage is not detrimental to repository performance, so long as gas pressures can be managed with design.

For that purpose, it is important to assess if gas pressure will induce mechanical damage to the clayey engineered and natural barriers and if such damage would be transient only, thanks to self-sealing capacity of clay, or would have a lasting effect, and how this would affect (or not) the global functioning of the repository. From such an assessment, design measures could be set up to avoid as much as possible gas pressure induced damage in the EBS and the EDZ. Possible ways to mitigate with (i.e., preserve the host rock integrity) to high gas pressures (i.e., over fracturing pressure) could be:

- Design backfills and/or seals to increase their potential gas transport capacity. One way to achieve this is to use specific seals and/or backfill concepts enhancing two-phase flow gas transport (Box 1, page 14);
- Limit as much as possible the quantity of metal present in the repository at closure (for example use non-metallic rebars for concrete reinforcement).

Gas will be generated in the GDF during several tens of thousands up to hundreds of thousands of years, depending on the concept of disposal. Many factors significantly influence the rate of gas generation, and by consequence may play a role on gas transport and its effect at repository scale. As an example, in the Belgian concept, the HLW is surrounded by a carbon steel overpack embedded in a high pH environment. Such an environment protects this overpack, leading to a relatively low corrosion rate and thus to a low gas generation rate during an extended period.

5.4.3 Fate of radionuclides

Although most of the gas flowing through gas-induced pathways would be inactive, it could at some point displace dissolved and/or volatile radionuclides, but the amounts involved are expected to be very limited, because:

- In water-saturated conditions, the transport of dissolved radionuclides (i.e., dissolved in pore water) is expected to be very low and driven by diffusion only because no water displacement is associated to the evacuation of gas in such conditions.
- If a gas phase appears and gas pathways develop through clay-based components, it has been shown that very little water, if any, is expelled along these pathways because most of it is more easily pushed laterally into the immediate surroundings. Hence, radioactive solutes in that water will also be displaced accordingly over a short distance to the sides of the pathway, not over a large distance along it.
- In unsaturated conditions, the diffusion of dissolved radionuclides is not expected to be enhanced as a gas phase is an obstacle for solute diffusion for species that are non-volatile. The diffusion rate of solutes, among which soluble radionuclides, is higher in a system that would remain saturated with water. Because degrees of saturation in clay barriers are expected to remain high at all times, it is expected that the reduction of diffusion of solutes will be limited. By consequence, it is deemed conservative to estimate the transport of dissolved radionuclides in water-saturated conditions. Total system performance assessment models considering

clayey host rocks and/or engineered barriers[19] may thus neglect the effects of gas phases on dissolved radionuclide transport.

- Volatile radionuclides that would not completely dissolve into the pore water upon release from the waste form may be transported to the shafts and/or ramps together with the inactive gas (which is generated in much larger quantities). However, the duration of the transport from the gas source to the shafts and/or ramps may take several hundred to several thousand years (order of magnitude, concept dependent), and only radionuclides with half-lives around or longer than this duration may present a significant activity in the gas phase when they reach the upper formations. In addition, all along the gas pathways, a fraction of the gaseous radionuclides will also dissolve (as also does inactive gas) into the pore water present in the surrounding materials.

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[19] And involved in EURAD-GAS

6. Concluding remarks and recommendations

This state-of-the-art document summarizes the current knowledge of gas transport through clayey materials and how this knowledge is currently being used in the context of the development of geological disposal systems in several European countries. Experimental data and insights from modeling are provided mainly for three clayey host rocks: the Boom Clay, the Callovo-Oxfordian claystone and the Opalinus clay, and for three bentonites: the Wyoming bentonite (MX-80), the FEBEX bentonite and the Czech bentonite BCV.

This document is primarily intended for end-users (RD&D managers, implementers and safety authorities). Starting with an overview of their main concerns regarding the generation and the transport of gas in clayey materials in the context of geological disposal of radioactive waste, it presents the shared understanding of the EURAD-GAS partners on gas transport processes and their controls in clayey materials. It then provides guidance on what could be the controls of gas and its impact at the repository scale. The document also identifies and formulates the remaining uncertainties, in their context and with transparency, in addition to this common view of gas aspects based on the knowledge gathered so far. Throughout the preparation of this report, the aim has been to enable end-users to easily assess the relevance of the collected knowledge and uncertainties about gas in their own programs. This will empower them to develop optimal strategies for making the best use of the available science base and for setting priorities for their own research and development efforts.

it first focuses on the shared[20] understanding about the gas transport process and their controls. Then, it gives indications of what could be the impacts of gas and their controls at repository scale. In that sense, the document identifies and formulates the existing knowledge as well as the remaining uncertainties of all types, with sufficient context and with full transparency. The aim is to allow end-users to eventually assess the relevance of this knowledge and these uncertainties within their national program and, subsequently develop optimal strategies to deal with the transport of gas and, if necessary, to effectively address the identified uncertainties where appropriate.

Overall, this state-of-the-art document reinforces the FORGE EC project's insight that gas is not a showstopper for the safety case, but a matter of managing uncertainties. It provides input to implementers that may inspire design measures to reduce the impact of gas on the disposal system and/or the uncertainties associated with gas flow through geological disposal systems. In particular, the authors suggest adopting a **comprehensive design approach** that considers the presence and effects of gas early on in any RD&D program for geological disposal of radioactive waste. Getting a first estimate of the expected gas volumes and rate of production should be a priority. Then, a good basis for the initial stages of development could be the available general knowledge of the mechanisms of gas transport in clay materials and the generic storyboard for its evacuation through a disposal system presented in this document and its references. In addition, it is recommended that an **iterative design approach** is put in place to address gas transport issues from the start, recognizing that adaptations can occur along the way. Indeed, uncertainty on the values of key properties that control gas transport can be large at the start of an RD&D program. Also, the passage from parameters derived from the characterization of gas transport through selected materials in laboratory or limited-scale in situ experiments to parameters that can be used for robust repository-scale evaluations requires a dialogue between experimentalists, modelers and repository designers. Indeed, gas transport strongly depends on the selection of EBS materials, and the conditions expected at repository closure throughout the whole system. It is essential that gas transport models and parameters are consistent with these specific materials and conditions.

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[20] i.e., the vision of EURAD-GAS partners

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