European Joint Programme on Radioactive Waste Management

Deliverable 6.6.

Achievements of EURAD-GAS

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 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 designs 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[1] (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 effective 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 the case that 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.

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 engineered and geological clayey materials, their couplings with the mechanical behavior and their impact on the properties of these materials.
- To evaluate the gas transport mechanisms that can be active at the scale of a geological disposal system and their potential impact on barrier integrity and repository performance.

The program of work aimed (i) to provide results that are applicable to a wide range of national programs and (ii) to transfer knowledge gained from laboratory and in situ experiments to configurations that are commonly found in current repository designs, to address key questions from the end-users:

- How could gas be transported throughout the disposal system and which water-soluble and gaseous radionuclide transport could be associated with it?
- How and to what extent could the hydro-mechanical perturbations induced by gas affect barrier integrity and long-term repository performance?

[1] The EBS comprises all man-made components, including the lining of galleries. In the French concept, most of the hydrogen production is expected to come from the corrosion of the metallic reinforcement of the concrete lining around the access galleries and large ILW cells.

For five years, twenty-nine partners from ten countries collaborated to achieve the objectives. The results obtained are significant, although some experiments are still ongoing at the time of writing, due to delays mainly caused by the COVID-19 crisis. Nine deliverables have been produced over the course of the project.

This document, Deliverable D6.6, briefly presents the main achievements of EURAD-GAS. It provides an overview of the main results, per task and per partner, and conclusions of the different tasks. Additionally, it presents the current understanding of gas transport in clayey materials, the impact at the repository scale and the fate of radionuclides. In conclusion, it lists the impacts of EURAD-GAS, in terms of implementation needs for radioactive waste management, safety, and increased scientific and technical knowledge. The addendum contains a summary of all articles/conference papers/PhD theses published under EURAD-GAS.

Deliverable D6.6 is as an entry point to other deliverables of EURAD-GAS:

- Three thematic technical reports with detailed scientific achievements: *Gas transport mechanisms: diffusion, retention and advection processes* (Deliverable 6.7, Task 2) (Jacops and Kolditz, 2024), B*arrier integrity: gas-induced impacts and model-based interpretation* (Deliverable 6.8, Task 3) (Marschall *et al.*, 2024) and *Modelling of a generic geological disposal: evaluation of various approaches to model gas transport through geological disposal systems* (Deliverable 6.9, Task 4) (Wendling *et al.*, 2024). These three comprehensive reports detail all the work carried out by each partner during EURAD-GAS. These documents are addressed to scientists with a background in geological disposal of radioactive waste and/or transport of gas in clayey materials.

- One document with the full integration of the shared understanding of the EURAD-GAS partners on gas transport processes and their controls in clayey materials with guidance on what could be the controls of gas and its impact at the repository scale: *State of the art on gas transport in clayey materials – Update 2023* (Deliverable 6.2) (Levasseur *et al.*, 2024). D6.2 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. This document is primarily intended for end-users (RD&D managers, implementers and safety authorities).

EURAD-GAS has increased confidence in the overall understanding of gas behavior in clayey materials, building on the FORGE EC project and beyond. This has in turn improved its integration into the conceptualization process for the different components of a disposal system, supporting and justifying the use of robust evaluation approaches. Overall, the discussion among all members of EURAD-GAS, including research entities, technical support organizations, and waste management organizations, has helped to strengthen the expert judgment at the end of FORGE: gas is not a showstopper for geological disposal, but rather requires effective management of uncertainties.

The impacts of EURAD-GAS can be categorized in terms of implementation needs for geological disposal, safety of geological disposal systems, and increased of scientific and technical knowledge:

- *In terms of implementation needs for geological disposal*: EURAD-GAS has produced documents for implementers that can inspire design measures to further reduce the gas impacts, or uncertainties associated with gas transport through geological disposal systems. Two state-of-the-art reports have been written. The first one presents some fundamentals on gas transport in clayey materials (Levasseur *et al.*, 2021) while the second focuses on key messages for end-users such as program managers (Levasseur *et al.*, 2024).

- *In terms of safety*: EURAD-GAS has provided experimental evidence on the processes involved in gas transport throughout a disposal system and on the effects of these processes on barrier materials (Jacops and Kolditz, 2024; Marschall *et al.*, 2024). This evidence can be referred to by national programs in the arguments supporting claims about long-term safety of a geological system. EURAD-GAS had also collected elements that make it possible to identify the inherent strengths and limitations of various approaches for the treatment of gas in safety cases and to assess their suitability in different contexts, recognizing that this may depend on the disposal system that is being evaluated (host rock/design) or even the advancement of the (national) program (Wendling *et al.*, 2024). Similarities of approaches between national programs were identified and the rationale behind differences explained (Levasseur *et al.*, 2021).

- *In terms of increased scientific and technical knowledge*: EURAD-GAS has succeeded in bridging the gap between experimentalists and modelers. Building on the lessons learned from the FORGE EC project, modelers were embedded with the experimentalists to encourage dialogue in the design of experiments and the development of shared conceptualizations of the observed behavior (Jacops and Kolditz, 2024; Marschall *et al.*, 2024). By doing so, EURAD-GAS has built confidence and extended the scientific bases on the fundamentals of gas transport in clayey materials. It has confirmed that the fundamental gas transport mechanisms that can develop in different clays are similar. Because a wide enough, but realistic, range of conditions were explored for representative clayey materials, EURAD-GAS has provided data which are of relevance for all disposal systems that include clayey barriers. Testing over a range of conditions spanning low (diffusion) to high (advection) gas generation rates, a better understanding of processes has been acquired which then has broader end-users' appeal. This understanding is described in the final technical reports (Jacops and Kolditz, 2024; Marschall *et al.*, 2024; Wendling *et al.*, 2024).

Based on the FORGE EC project, EURAD-GAS has made a major step forward on the mechanistic understanding of the transport of gas in clayey materials. However, further research is needed to address remaining questions.

Regarding the scientific basis, it is recommended to perform the following actions:

- Expand experimental databases (mainly gas transport properties and, when relevant, relationship between properties), by conducting experiments under conditions that are representative of repository conditions.

- Develop and share best practices in terms of sample handling and experimental protocols, in line with the efforts initiated by Nagra during EURAD-GAS with the support of CIMNE/EPFL/BGS (including the use of visualization techniques) (Marschall *et al.*, 2024).

- Strengthen dialogue between modelers and experimenters in the design of experiments and the development of shared conceptualizations of observed behavior.

- Further develop coupled microscopic models to refine small-scale process understanding.

- Develop a portfolio of models at different scales (microscopic, centimeter, component, repository) (stepwise abstraction process) and assess their applicability. Having a range of models available (each with their advantages and limitations, but always based on a physical basis) allows for the selection of the most appropriate one(s) for a particular disposal system.

Regarding end-users' needs, organizations involved in radioactive waste management should continue to exchange information. This includes:

- Sharing and further developing best practices for dealing with gas transport processes and impacts. Efforts are needed to apply the knowledge gained from small-scale experiments appropriately at the repository scale. Gas requirements evaluated through modeling for the whole disposal system need to be broken down into requirements at lower levels of design.
- Developing common strategies to ensure that gas transport requirements are compatible with all other requirements of the geological disposal system and are well integrated in the design and optimization processes.

Contents

Acronyms

Organizations – The organizations involved in EURAD-GAS are in bold.	
Aalto University	Aalto University (*Aalto-yliopisto / Aalto-universitetet*) (Finland)
Andra	National Agency for Radioactive Waste Management (*Agence Nationale pour la Gestion des Déchets Radioactifs*) (France)
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)
CIEMAT	Centre for Energy, Environment and Technology (*Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas*) (Spain)
CIMNE	International Centre for Numerical Methods in Engineering (Spain)
CNRS	*Centre national de la recherche scientifique* (France)
CNRS-U Lorraine	GeoRessources, depending on the University of Lorraine and CNRS (France)
CNRS-U Poitiers	Institute of Chemistry of Materials and Media *(Institut de chimie des milieux et matériaux de Poitiers – IC2MP),* depending on the University of Poitiers, IMT Atlantique and CNRS (France)
CNRS-U Grenoble	Institute of Earth Sciences (*Institut des sciences de la Terre – ISTerre*), depending on the University of Grenoble and CNRS(France)
COVRA	Central Organisation for Radioactive Waste (*Centrale Organisatie Voor Radioactief Afval*) (the Netherlands)
CTU	Czech Technical University (Czech Republic)
EC	European Commission
EDF	*Electricité de France* (France)
EPFL	*Ecole Polytechnique Fédérale de Lausanne* (Switzerland)
GRS	*Gesellschaft für Anlagen- und Reaktorsicherheit* (Germany)
IRSN	Institute for radiation protection and nuclear safety (*Institut de radioprotection et de sûreté nucléaire*) (France)
LEI	Lithuanian Energy Institute (Lithuania)
Nagra	National Cooperative for the Disposal of Radioactive Waste (*Nationale Genossenschaft für die Lagerung radioaktiver Abfälle*) (Switzerland)
NWS	Nuclear Waste Services (UK)
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)
PSI	Paul Scherrer Institute (Switzerland)
SCK CEN	Belgian Nuclear Research Centre (*Studiecentrum voor Kernenergie / Centre d'Étude de l'énergie Nucléaire*) (Belgium)
SKB	Swedish Nuclear Fuel and Waste Management Company (*Svensk Kärnbränslehantering*) (Sweden)

Organizations – The organizations involved in EURAD-GAS are in bold.	
SÚRAO	Radioactive Waste Repositories Authority (*Správa Úložišť Radioaktivních Odpadů*) (Czech Republic)
TU Delft	Delft University of Technology (*Technische Universiteit Delft*) (the Netherlands)
UKRI-BGS	UK Research and Innovation - British Geological Survey (United Kingdom)
UFZ	Helmholtz Centre for Environmental Research (*Helmholtz Zentrum für Umweltforschung*) (Germany)
ÚJV Řež	Nuclear Physics Institute (Czech Republic)
U Liège	University of Liège (Belgium)
UPC	Technical University of Catalonia (*Universitat Politècnica de Catalunya*) (Spain)
ZHAW	Zurich University of Applied Sciences (Switzerland)

Projects/facilities	
Äspö HRL	Äspö hard rock laboratory in Sweden
BEACON EC project	Bentonite mechanical evolution – *EC project (2017–2021)*
CAST	Carbon-14 source term and fate – *EC project (2013–2018)*
EURAD	European joint programme on radioactive waste management – *EC funded (2019–2024)*
EURAD-DONUT	RD&D work package of EURAD devoted to the development and improvement of numerical methods and tools for modeling coupled processes
EURAD-HITEC	RD&D work package of EURAD devoted to the study of the influence of temperature on clay-based material behaviour
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)*
HADES URL	High-activity disposal experimental site – underground research laboratory in Mol (Belgium)
LASGIT	Large-scale gas injection test (full-scale demonstration project performed at the Äspö hard rock laboratory in Sweden)
MEGAS	Modeling and experiments on gas migration in repository host rocks (under the umbrella of the PEGASUS EC project) – *EC project (1992–1997)*
PEGASUS	Project on the effects of gas in underground storage facilities for radioactive waste – *EC umbrella project (1991–1998)*

Other acronyms	
BC	Boom Clay formation (Belgium and the Netherlands) (a poorly indurated clay rock)
BCV	Czech bentonite
COx	Callovo-Oxfordian claystone (France) (an indurated clay rock)
EBS	Engineered barrier system
EDZ	Excavation damaged zone
FEBEX bentonite	Spanish bentonite
FIB-SEM	Focused ion beam scanning electron microscope
GDF	Geological disposal facility
HLW	High-level waste
ILW	Long-lived intermediate-level waste
L/ILW	Low- and intermediate-level waste
µ-CT	Micro-computed tomography
MX-80	Wyoming bentonite (USA)
OPA	Opalinus clay (Switzerland) (an indurated clay rock)
SEM	Scanning electron microscope
SOTA	State-of-the-art report
TH(M)	Thermo-hydro(-mechanical)
THM(C)	Thermo-hydro-mechanical(-chemical)
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 designs under development.

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- How and to what extent could the hydro-mechanical perturbations induced by gas affect barrier integrity and long-term repository performance?

EURAD-GAS work program included three scientific tasks. Task 2 mainly focused on the characterization of gas transport mechanisms in clays while Task 3 analyzed the consequences of gas transport in terms of material perturbations. Based on the results of Tasks 2 and 3, Task 4 provided phenomenological descriptions of gas transport and likely consequences for radionuclide transport at the repository scale. These three scientific tasks have strongly interacted with each other during the project, under the supervision of Task 1, responsible for the coordination of the project and the dissemination of results and knowledge gained (Figure 1). The clayey materials studied were of two types: geological (Opalinus clay formation (OPA), Callovo-Oxfordian claystone (COx) and Boom Clay formation (BC)) and engineered (three types of bentonites and their mixture with sand (Wyoming MX-80, Spanish FEBEX and Czech BCV)). EURAD-GAS was closely linked to EURAD-HITEC which studied the influence of temperature on the behaviour of clayey materials behaviour. These two work packages had a common objective, namely to provide building blocks to support the evaluation of the robustness of the repository concepts (Levasseur *et al.*, 2022).

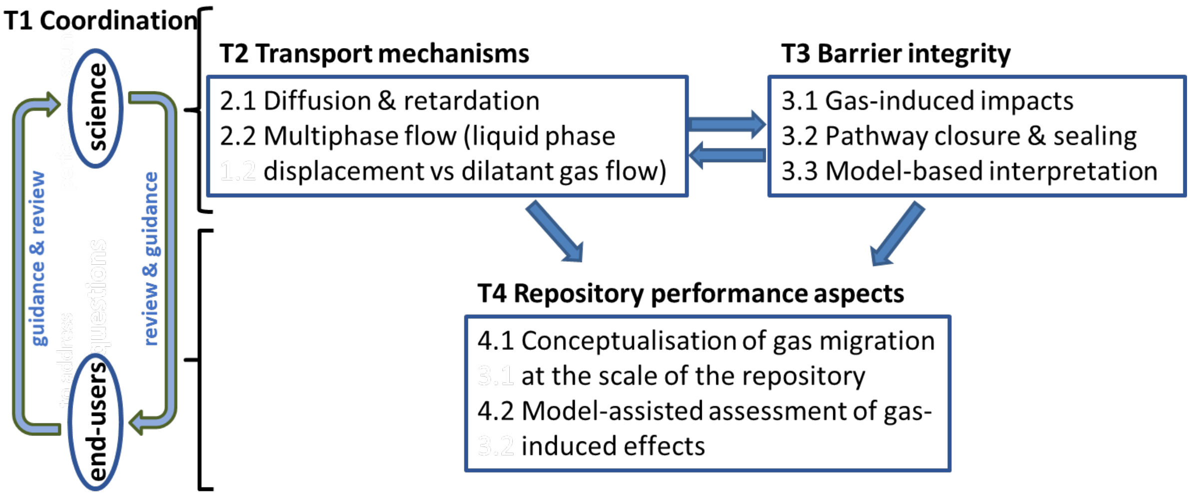

Figure 1 – Structure and organization of EURAD-GAS, as stated in the beginning of the project.

This document, Deliverable D6.6, is as an entry point to other deliverables of EURAD-GAS:

- Three thematic technical reports with detailed scientific achievements: *Gas transport mechanisms: diffusion, retention and advection processes* (Deliverable 6.7, Task 2) (Jacops and Kolditz, 2024), B*arrier integrity: gas-induced impacts and model-based interpretation* (Deliverable 6.8, Task 3) (Marschall *et al.*, 2024) and *Modelling of a generic geological disposal: evaluation of various approaches to model gas transport through geological disposal systems* (Deliverable 6.9, Task 4) (Wendling *et al.*, 2024). These three comprehensive reports detail all the work carried out by each partner during EURAD-GAS. These documents are addressed to scientists with a background in geological disposal of radioactive waste and/or transport of gas in clayey materials.

- One document with the full integration of the shared understanding of the EURAD-GAS partners on gas transport processes and their controls in clayey materials with guidance on what could be the controls of gas and its impact at the repository scale: *State of the art on gas transport in clayey materials – Update 2023* (Deliverable 6.2) (Levasseur *et al.*, 2024). D6.2

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. This document is primarily intended for end-users (RD&D managers, implementers and safety authorities).

This document contains six chapters. After this introduction, Chapters 2, 3 and 4 provide an overview of the main results and conclusions of Tasks 2, 3 and 4 respectively, in relation to their own objectives as defined at the beginning of the project. These chapters are based on the technical reports delivered by each task at the end of the project (Jacops and Kolditz, 2024; Marschall *et al.*, 2024; Wendling *et al.*, 2024). Chapter 5 presents the current understanding of gas transport in clayey materials, impact at repository scale and fate of radionuclides. In conclusion, Chapter 6 lists the impacts of EURAD-GAS, in terms of implementation needs for radioactive waste management, safety, and increasing scientific and technical knowledge, and ends with the future perspectives.

The appendix contains a summary of most articles written by EURAD-GAS members or in collaboration with members of DONUT and PhD theses published in the frame of EURAD-GAS (as of 20 May 2024), listed per task. They are all quoted in the main text. The document ends with a bibliography. Additional references are listed in the two state-of-the-art reports on gas transport in clayey materials (Levasseur *et al.* (2021; 2024)).

2. Task 2 – Transport mechanisms

Chapter 2 presents the objectives of Task 2 as defined at the beginning of the project, followed by a short overview of the main results (Section 2.2) and achievements (Section 2.3). Detailed scientific achievements are available in the thematic technical report: *Gas transport mechanisms: Diffusion, retention and advection processes* (Deliverable 6.7, Task 2) (Jacops and Kolditz, 2024). The full integration of the results is presented in Deliverable 6.2 (*State of the art on gas transport in clayey materials – Update 2023* (Levasseur *et al.*, 2024)).

2.1 Task 2 key objectives

Task 2 (Transport mechanisms) focused on the main gas transport mechanisms which may take place in a disposal system in the post-closure phase, in clayey materials. Different mechanisms are generally expected to be of importance in a geological disposal system:

- 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 adsorption on the solid phase. Improving the mechanistic understanding of diffusive transport of dissolved gas and retention mechanisms was the main objective of Subtask 2.1 (Gas diffusion and retardation processes at high level of water saturation).
- If the gas production rate exceeds the rate at which gas can be dissolved and evacuated by diffusion, a free gas phase will develop. Gas pressure gradients will give rise to the flow of the two immiscible fluids, gas and water. Two primary mechanisms for gas transport as a separate phase are possible: (i) visco-capillary two-phase flow and (ii) the formation and propagation of dilatancy-controlled gas pathways. Exploring which transport mechanism prevails under which range of conditions and understanding how the coupling between pressure in the liquid and gas phases and the stresses in the solid phase control gas transport was the main objective of Subtask 2.2.

2.1.1 Subtask 2.1 objectives

In order to compare the rates of gas generation and gas evacuation by dissolution and diffusion, knowledge of the diffusion parameters of dissolved gas through the used materials is essential. Up to now, diffusion parameters are available essentially in water-saturated systems. As partial desaturation might occur at some point during the repository evolution, EURAD-GAS extended the available experimental data for gas diffusion to partially desaturated conditions. In line with the general objective to improve mechanistic understanding, interpretation of the experimental results was supported by pore network modeling. The experimental and modeling work program aimed in particular to investigate how petrophysical parameters (e.g., mineralogy and density), material anisotropy and the state of stress influence the diffusion parameters. One objective of Subtask 2.1 was thus to determine gas diffusion parameters on different clayey materials at different degrees of water saturation and support experimental data interpretation by pore network modeling.

While diffusing through the pore water, dissolved gas might also interact with the barrier materials. Before EURAD-GAS, interactions (mainly gas sorption) were only studied under dry conditions. In particular, hydrogen adsorption due to sorption processes has only been studied under dry conditions. One objective of this subtask was to determine whether gas sorption could be a relevant retardation mechanism for diffusive transport under repository conditions, in clays that are partially or fully saturated with pore water

2.1.2 Subtask 2.2 objectives

From previous work (including work done in the FORGE EC project), two primary mechanisms for advective gas transport through natural mudrocks were proposed; (i) visco-capillary two-phase flow, where capillary forces must be overridden in order to allow displacement of the wetting phase, by migrating gas and (ii) the formation of dilatancy-controlled gas pathways. The potential for one specific mechanism to prevail is dependent on a range of factors, including the saturation state of the clay and the ratio of the clay to sand fraction. For fully saturated pure bentonite materials, such as those used in engineered barriers, experimental evidence indicates that gas flow by dilatancy-controlled gas pathways is the preferred mechanism of transport. In such cases, significant hydro-mechanical coupling is observed, which cannot be satisfactorily explained by visco-capillary two-phase flow. At the start of the EURAD-GAS project, it was considered necessary to refine this hydro-mechanical coupling between the immiscible phase, the interstitial fluid, and the total stress for all types of clayey materials. In that sense, understanding these relationships, how they vary from one studied clay to another, and what impact, if any, they have on the integrity of the EBS and the host rock, was a research issue of EURAD-GAS.

Through new experimental studies, Subtask 2.2 aimed to:

1. Provide reference data for various engineered and geological clayey materials under a sufficiently broad range of conditions to make it possible to investigate in a more systematic way how petrophysical parameters, like mineralogy or density, and conditions such as the stress state influence transport of a free gas phase.
2. Improve understanding of the observed gas transport mechanisms and identifying their main controls, through interpretation of the experimental results by models in which the representation of dilatancy-controlled gas pathways and crack propagation is implemented at process level.
3. Compare conceptualizations of the transport mechanisms limited to the macroscale (continuum representation + discrete conducting features) such as those developed, for instance, in the FORGE EC project to conceptualizations in which the microscale is also represented, to better improve understanding of how microlevel heterogeneities and deformations affect macroproperties.

The experimental and modeling programs of Subtask 2.2 aimed at characterizing and understanding gas transport in clayey materials close to saturation. For bentonite, it benefited from the results of the BEACON EC project that characterized the hydro-mechanical coupling of swelling clay materials during resaturation phase. For bentonite barriers, BEACON EC project, EURAD-GAS and EURAD-HITEC were complementary as these together support the global description of hydro-mechanical processes from the installation of materials to their evolution over the long term (including the thermal phase in the case of heat-emitting waste) and the consequences of gas flow. The integration of knowledge was facilitated by involvement in this subtask of EURAD-GAS of several BEACON and EURAD-HITEC members.

2.2 Task 2 main results

This subsection summarizes the final results of Subtasks 2.1 and 2.2 as achieved by each partner of the Task 2 and reported in Jacops and Kolditz (2024). The gas of interest was chiefly hydrogen (H_2), but other apolar gases such as methane (CH_4, by-product of bacterial activity), helium (He), argon (Ar) (naturally occurring noble gases) and neon (Ne, H_2 proxy) were also investigated in order to i) avoid artifacts related to bacterial activity triggered by H_2, ii) overcome safety problems especially in underground research laboratories, and iii) probe the effect of polarizability and kinetic diameter on apolar gases transport in porous networks.

Results of Subtask 2.1 per partner are succinctly presented hereafter:

- **Quantification of hydrogen adsorption by clays by CNRS-U Grenoble (ISTerre)**. The assessed hydrogen adsorption by dry COx at 25°C and 100 bar is 0.02 wt%. However, dry conditions are not representative of disposal systems in clayey host rocks. The evaluation of hydrogen adsorption by COx and Boom Clay in partially and fully saturated conditions is still ongoing at the end of EURAD-GAS. This adsorption is expected to be very low.

- **Determination of the diffusion coefficient in synthetic and natural clayey materials (UKRI-BGS)**. UKRI-BGS has successfully measured the permeability and diffusion coefficients for Boom Clay, a sample of the Eigenbilzen sands under two different stress regimes, and a range of synthetic samples manufactured with different mineralogical compositions, all adding to the knowledge pool for these materials. Diffusion of helium was also shown to occur preferentially along bedding planes with approximately 60% of the diffusional capacity of Boom Clay moving parallel to bedding. However, of greater importance was the observed coupling between diffusion and intrinsic permeability which was very clear in the synthetic sample data. Examining the data as a whole suggests a fundamental relationship exists between permeability and diffusivity, and when plotted in logarithmic scale the Boom Clay and Eigenbilzen sand fall on a common projection. If correct, such relationships could be used to predict diffusivity across a range of material and permeability scales.

- **Diffusion of gases in unsaturated clay (SCK CEN)**. SCK CEN has developed a setup to measure diffusion of gases (He and Ar) in slightly unsaturated synthetic clay samples mimicking host rocks. So far, diffusion has been measured in two sample types, containing different amounts of clay, silt and sand, at saturation degrees between 73% and 100%. Results showed that gas diffusivity, as a combination of diffusion of dissolved gas and gaseous gas, increased only slightly, about 20% when desaturating the sample toward 73% (Gowrishankar *et al.*, 2023). As a general conclusion and based on the data collected so far, one can state that the rate of gas transport is marginally increased in partially saturated clay-based materials as opposed to rates of solute ionic tracers, which decrease with desaturation[3]. It is recommended that lower saturation degree and samples with variable clay content be studied to improve process understanding and to be representative of engineered clayey materials.

- **In situ diffusion experiments with neon (SCK CEN).** The NEMESIS experiment (neon diffusion in MEGAS in situ), which measures diffusion of neon on a meter scale in Boom Clay at the level of the HADES underground research laboratory (HADES URL), was started in September 2023 and will run for at least five years. The aim of this in situ experiment is to obtain values of the diffusion coefficients of dissolved neon in different directions on the scale of several meters. During the preparation phase, a validation in-diffusion experiment was performed with helium in 2 filters already present in HADES URL (these filters are not involved in the final NEMESIS experiment). Based on the helium pressure loss due to in-diffusion, and by using a newly developed model, the fitted diffusion coefficient was estimated at 80% of the value measured in laboratory experiments – which is very satisfactory[3].

- **Characterization of the pore networks of clayey materials (IRSN).** IRSN has studied the water-air distribution during onset of desaturation in clay-containing rocks, in order to confirm whether small-scale samples can be considered homogeneously desaturated, assumption which is often made for further evaluation of unsaturated transport properties (suction, diffusion, ...). For this purpose, x-ray synchrotron imaging and laboratory-based computed tomography facilities

[3] Estimating the diffusion coefficient is not an easy task. It is done by back-analysis of laboratory experiments. The values can be subjected to large uncertainties. A variation of 20% can be viewed as small compared to uncertainties.

were used to study several clay-containing rocks (Toarcian, Eigenbilzen sands) at a submicron scale (samples were cylindrical with a very small diameter (6–10 millimeters)). Porosity profile observations indicate that even if desaturation takes place in the largest pores first, desaturation front may be viewed as homogeneous within subcentimeter size samples, even when close to the full water saturation.

- **Modeling gas diffusion in saturated and unsaturated conditions (PSI).** PSI has investigated the transport mechanisms of gas in porous clay materials and explored the relationships between the structural properties of the clay minerals and the mobility of small gaseous molecules. Specifically, it focused on smectite-rich clay rocks. Molecular dynamics simulations have been employed to observe fluid behavior at the nanoscale, and a methodology has been developed to upscale the results to a pore scale using pore-scale simulations. Findings revealed that the diffusion of gases is significantly influenced by three key factors: the mineralogy of the clay, the size of the pores, and the hydrodynamic radius of the gas molecule (which depends on the gas type). Simulation results for unsaturated clay suggest that the gas-diffusive behavior in this system exhibits a transition regime characterized by a combination of molecular diffusion and Knudsen diffusion. In other terms, gas diffusion is influenced by the thickness of the water films, the mean free path of the gas molecules, and the average available pore width. As a result, Fick's law, which is commonly used to describe diffusion processes, should be seen as an approximation for diffusive process of gas at this nanoscopic scale; at this nanoscale, diffusion processes may be described by a combination of the Fick's law and of the Knudsen diffusion (Owusu *et al.*, 2022; Owusu *et al.*, 2023; Owusu, 2023).

- **Modeling gas diffusion in saturated conditions (BGR and Aalto University).** The diffusion processes of gas dissolved in a pore liquid are characterized by multiphase effects. In the framework of EURAD-GAS, these effects are captured by BGR by using the TH2M model OpenGeoSys 6 (OGS-6). The TH2M model was validated against the double through-diffusion experiment performed by SCK CEN. The model assumed full saturation of the sample (modeling single-phase flow), no deformation, isothermal conditions and no liquid pressure gradient (advection negligible). The diffusion processes of the components were captured by multiple superimposed TH2M runs (validated with a 4-component formulation implemented in Python). Overall, the experimental and numerical results matched very well, thus increasing confidence in the numerical model (Pitz *et al.*, 2024). In addition, Aalto University (Gupta, Jacops *et al.*, 2023) has developed a numerical framework to model diffusion tests by incorporating gas mixture flow for two incompressible and inert 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 the BGR approach, the approach developed by Aalto University successfully captures the results of the SCK CEN experiment in terms of the diffusive gas flux through the sample and hence the gas concentration increase in the two vessels. These two approaches are useful for the characterization of the diffusion process on a large scale.

Results of Subtask 2.2 per partner are succinctly presented hereafter:

- **Gas injection under controlled loading (SCK CEN).** SCK CEN, in collaboration with ONDRAF/NIRAS, performed a gas injection experiment with one Boom Clay sample under isostatic conditions. In this setup, gas pressure is slightly inferior to the confinement pressure, which has been set to be representative of the level of the HADES URL. In a first stage, diffusion experiment is performed with dissolved helium and then pure gaseous helium and results are compared to diffusion coefficients estimated from constant volume cell experiments. The test enabled to show that (1) only minor differences are observed when different loading conditions (constant volume versus isotropic loading) are considered and (2) tests performed with pure gaseous gas (without any liquid) or dissolved gas in pore water provide same results. Then, gas injection is slightly and stepwise increased to test the limits of diffusive transport of dissolved gas and a possible transition to free gas flow during a long gas injection time. At time of reporting, no clear conclusions can be drawn yet on this possible transition.

- **Triaxial tests (UKRI-BGS).** UKRI-BGS performed four triaxial gas injection experiments on Boom Clay and Callovo-Oxfordian claystone at two different confining pressures in order to determine whether stress controls the transition from visco-capillary two-phase flow to dilatancy-controlled gas pathways formation. The apparatus used allowed axial and radial strains to be measured during each test. All tests showed that (1) visco-capillary two-phase transport was not observed during gas injection with dilatancy-controlled gas transport the only mechanism observed; (2) the control of the gas-entry pressure was the minimum principal stress and (3) the dilatancy-controlled gas pathways are not classical fracture related. Significant differences between tests of different geometry (small injection filters versus full-face filters) were observed and thus, the direct comparison between tests performed in different laboratories might be controversial as a result of the filter geometry selection. No conclusive conclusions can be drawn yet regarding (1) the role of stress as a control of the physics of gas transport and whether (2) there is a transition from visco-capillary two-phase flow to dilatancy-controlled gas flow.

- **LASGIT (UKRI-BGS).** LASGIT was a full-scale demonstration experiment operated by SKB at the Äspö Hard Rock Laboratory (Sweden) at a depth of 420 meters, with the two main objectives of providing quantitative data to improve process understanding and validating modeling approaches used in performance assessment. All observations from LASGIT support the idea that gas is mainly transported through dilatancy-controlled gas pathways (and diffusion), which is consistent with observations made within the laboratory. Full-scale observations showed that (1) the movement of gas occurred at a pressure very close to the local total stress; (2) no signs of localized consolidation of the bentonite were observed; (3) the measured peak gas pressures should not lead to any mechanical damage to the buffer or to other barrier components in the repository; (4) peak gas pressure is linked to the hydraulic permeability of the buffer and the ease at which gas can exit a disposal hole; (5) gas is transported through a small number of time-dependent dilatancy-controlled gas pathways; (6) no desaturation of the bentonite buffer as a result of gas transport was observed; (7) over the timescale of the project, pathway closure was only partially successful in the absence of injected water; (8) the dilatancy-controlled gas pathways are expected to slowly close at a finite "shut-in" pressure; (9) gas migration through a bentonite is highly unlikely to alter the favorable hydro-mechanical properties of the barrier and (10) the impact of emplaced and long-lived persistent heterogeneities within the bentonite on gas pressure remains unclear (Cuss *et al.*, 2022).

- **Gas flow analysis using a fracture visualization approach (UKRI-BGS).** UKRI-BGS has performed a series of twenty-five experiments using the fracture visualization rig to answer fundamental questions about gas flow in clayey rocks. These experiments have confirmed that (1) multiple pathways form at the same time, and these vary in size and distribution within the sample; (2) the velocity of pathway growth varies between pathways, with some pathways migrating quicker and others at a slow rate; (3) the distribution of pathways is stochastic; (4) the walls of a dilatancy-controlled gas pathway elastically compress to accommodate the pathway and (5) self-sealing is observed.

- **Laboratory-scale testing of synthetic materials (UKRI-BGS).** UKRI-BGS has carried out six different tests to examine the role of clay content (in relation to other constituents) on the dominant gas transport mechanism and the resulting distribution of transported gas within the flow regimes. A sample compaction procedure has been developed to create synthetic compacted clay samples with controlled mineralogy. These samples have then been subjected to gas injection experiments in a constant volume cell after an initial rehydration and swelling phase. A heavily instrumented and novel test cell has been designed and constructed and hence, the mechanical response of the clay has been monitored in detail to provide meaningful insight into the mechanisms and distribution of gas flow during these experiments. The analysis of these tests leads to the following points: (1) as expected, swelling pressure is correlated with clay content in engineered clays and anti-correlated with sand + silt fraction in synthetic mixtures; (2) at fully saturated conditions, dilatancy-controlled gas flow is expected to occur when clay content exceeds 40%; and (3) clay content appears to play a significant role regarding the stress field behavior and to directly relate to gas-entry pressure. Two different metrics have been defined to provide some information regarding the increase and decrease in clay permeability as pathways open and close. This should allow a substantial improvement in the capacity to simulate this complex process.

- **Characterization of advective gas transport in FEBEX bentonite (CIEMAT).** CIEMAT has undertaken an experimental program aimed at discerning the mechanisms controlling advective gas flow in the Spanish reference barrier material (FEBEX). The tests have been performed on initially fully saturated FEBEX bentonite and in them, two gas transport mechanisms have been identified, depending on the gas pressure level. Below a value referred to as gas-entry pressure, the predominant transport mechanism is the diffusion in pore water. Once this gas-entry pressure is exceeded, gas flows through existing porosity or via self-created and stress-induced pathways. Gas flow is then localized, and little water is displaced. At the tested dry densities (1 500 kg/m^3, 1 600 kg/m^3 and 1 700 kg/m^3), gas flow seems to occur by the formation and propagation of dilatancy-controlled gas pathways. Therefore, this knowledge needs to be included into the thermo-hydro-mechanical (THM) modeling (as done for instance by Gupta *et al.* (2021; 2023a)) and thus, discrete pathways approaches could probably help to reproduce the experimentally observed breakthrough phenomena.

- **Gas entry and flow through a bentonite barrier (CTU, SÚRAO).** An experimental program has been undertaken by the Czech consortium with the objective of investigating the gas breakthrough behavior of samples of Czech BCV bentonite with varying dry densities. Although this material was already known to behave similarly to bentonites tested in other countries such as Wyoming bentonite MX-80 in terms of gas permeability, tests in this experimental design have shown that (1) the formation of dilatancy-controlled gas pathways via gas-exerted pressure occurs following the exceeding of the measured axial total stress, which is related to the swelling pressure of the material; and (2) the type of gas injected (air or hydrogen) exerts no influence on the breakthrough pressure value.

- **Gas breakthrough experiments on different types of bentonites (ÚJV Řež, SÚRAO).** The aim of the ÚJV Řež experimental research was to conduct gas injection tests and subsequently evaluate the gas pressure evolutions of two types of bentonites (BCV and MX-80) in order to compare BCV with MX-80, the bentonite that is most commonly used in geological disposal programs. Multiple gas injection tests using a relatively simple apparatus were performed on samples of different dry densities (1 400 kg/m^3 and 1 600 kg/m^3). The experiments confirmed the existing knowledge of gas flow through the formation of dilatancy-controlled gas pathways: in general, the behavior of BCV bentonite does not differ from the reference material (MX-80) in terms of gas transport mechanisms and gas breakthrough occurs at pressure levels lying within the theoretical swelling pressure range. Further knowledge acquired within the experimental program concerns (1) the acquisition of data from the direct comparison of the behavior of hydrogen and air, from which it was determined that the type of gas has no effect on the gas breakthrough pressure values; and (2) the demonstration of the self-sealing capacity of BCV bentonite.

- **Visualization of multiphase flow in nanopores network (CNRS-U Poitiers (IC2MP)).** IC2MP has developed a gas injection test on a small sample inside a miniaturized and optimized triaxial cell to investigate the microstructural changes inside the claystone (COx) caused by gas entry and propagation in the porous network. This setup has been combined with a visualization technique in a laboratory computed tomography facility to monitor the formation of gas pathways in real time. At the time of writing the EURAD-GAS conclusions, the test was still in progress. No experimental results are available yet.

- **A coupled pneumo-hydro-mechanical (PHM) finite element model within LAGAMINE (TU Delft).** (The term *pneumo* refers to zero-thickness coupled interface finite element.) TU Delft has developed a numerical model capable of simulating gas migration in water-saturated clay. This model is able to simultaneously reproduce, within a unified framework, the diffusion and advection of dissolved gas and the displacement of the liquid phase filling the clay pores by the invading gas, together with the localized gas flow along macroscopic cracks induced and propagated by the gas pressure (Liaudat *et al.*, 2023). To do so, this model extends a pre-existing two-phase–two-species hydro-mechanical model for bulk porous media by means of zero-thickness interface elements. For this purpose, a new PHM triple-node zero-thickness interface element has been developed. Main progresses comprise: (1) the explicit representation of the PHM behavior of sample–device interfaces; (2) the initiation and propagation of macroscopic gas cracks inside the water-saturated bulk material; (3) the modification of the flow patterns inside the bulk material as a consequence of the localized flows along the cracks; (4) the sharp drop of the gas injection pressure when the gas crack reaches the back-pressure filter, followed by some fluctuations before reaching a steady state; and (5) the closure (self-sealing) of the gas cracks when the gas injection ceases. The performance of this approach has been validated with synthetic benchmark examples. However, at the time of writing the EURAD-GAS conclusions, quantitative validation of the model is still ongoing as well as its extension to the dilatancy-controlled gas flow, the introduction of variability/heterogeneity of the material properties, and extension to a three-dimensional setting.

- **Conceptualization of the gas transport processes taking place in the post-closure phase within LAGAMINE (U Liège).** A multiscale modeling approach has been adopted by the University of Liège to simulate the gas transport mechanisms in low permeable sound clay rocks as well as their accompanied hydro-mechanical processes. This model takes into account the role played by the rock structure at a microlevel in the initiation and propagation of such a macroscale gas transport mechanism. Based on experimental data, a suitable representative element volume (REV) has been defined to idealize the material microstructure with different families of discontinuities, and an assembly of tubes substituting the flow behavior of the porous matrix blocks. The transition between the two scales is achieved by means of homogenization and localization techniques. The approach has been implemented in the finite element code LAGAMINE and first applied to a laboratory-scale case study (Corman *et al.*, 2024). Then, it has been extended to a more general and random configuration, which has been upscaled in order to evaluate the possibility to extrapolate the laboratory observations of gas-induced damage processes on a larger scale. Simulations highlight two essential aspects in the development of preferential pathways: (1) the more continuous the connectivity between the disturbed planes is, the faster the gas flow through this discrete zone; and (2) these links between the planes of weakness (also called bridging planes) need to be numerous to ensure a rapid gas propagation at a larger scale. Even if such approach is very promising for process understanding, predictive and upscaling capabilities are limited as some model parameters are tuned in order to mimic the experimental data and since the microstructural heterogeneity of some clay properties (stiffness and strength) and the connectivity of the macropores and microfractures cannot be easily defined at a large scale.

- **Benchmarking of a new non-isothermal two-phase flow in deformable porous media approach within the open-source simulator OpenGeoSys (BGE, UFZ and BGR).** BGE, in collaboration with UFZ and BGR, has further developed an approach to simulate non-isothermal two-phase–two-component flow in deformable porous media (TH2M). Different gas permeability models have been tested and implemented: (1) a gas pressure-dependent permeability model, which assumes that permeability changes increase significantly when a specific gas pressure threshold is exceeded with microcracks being formed through the development of a percolation network; (2) a strain dependent permeability model, which assumes that gas permeability depends on both the total volumetric elastic strain and an equivalent plastic strain, and describes an increase of permeability as soon as a plastic failure occurs; (3) a failure index permeability model, which describes intrinsic permeability as a function of a Mohr-Coulomb failure index; and (4) an embedded fracture permeability model, where the clay is supposed to be an idealized setting with fractures, whose parameters are employed to define the intrinsic permeability. This OGS approach has been validated via a benchmark test, originally proposed by Andra as part of the MoMaS project (*Mathematical Modeling and Simulations for Radioactive Waste Management*) and successfully captures the transition from a single-phase to a two-phase gas transport (Grunwald *et al.*, 2023).

- **Direct Numerical Simulations (DNS) of multiphase fluid transport through deformable nanoporous materials (IRSN and CNRS-U Lorraine (Georessources)).** IRSN and CNRS-U Lorraine have conducted numerical simulations of multiphase flow through deformable materials using DNS modeling approaches, which require the description of both (1) the physical phenomena at the pore scale and (2) the pore space (Amrofel *et al.*, 2024). On the one hand, a DNS of a dilatancy-controlled gas flow has been performed using an extended Smoothed Particle Hydrodynamics (SPH) code. This method can be successfully employed to simulate (1) the explicit interaction of a pressurized fluid with a deformable elastic media and (2) fracturing if damage is assumed. On the other hand, a DNS of flow and transport phenomena using the mesoscopic lattice Boltzmann method (LBM) has been developed. These two models (SPH and LBM) have been used to assess different processes regarding gas flow, including evaporation (considering Kelvin effect), visco-capillary two-phase flow, dilatancy-controlled gas flow and fracturing. Both have proved to be relevant in terms of numerical stability for adequate physical parameters. It has also been observed that FIB-SEM (Focused ion beam scanning electron microscope) images can be used with DNS to compute properties at various scales. However, there are still limitations in computational capacity to describe (1) domain sizes smaller than the representative elementary volume (REV) or (2) multiscale heterogeneities. It has also been proved that the Kelvin effect is a significant phenomenon highly impacting the time of breakthrough and saturation that needs to be accounted for.

- **Hydro-mechanical simulations of breakthrough tests on FEBEX bentonite using CODE_BRIGHT (UPC).** A standard two-phase flow model has been used to simulate gas breakthrough tests on FEBEX bentonite. This has been enhanced with a complex hydro-mechanical model, based on a rate dependent permeability (assuming a cubic law) and a calibrated Barcelona Basic Model (*BBM*) for the mechanical part. The model has proved successful to describe gas breakthrough tests (including saturation level, gas injection rates, draining of gas, resaturation and second gas injection) under a three-dimensional full geometry. The model considers arbitrarily random distributions of permeability zones. Adjustments to the experimental setup (better instrumentation, refined sensor data, use of imaging techniques and transparent walls) are needed to improve the model (Toprak and Olivella, 2023).

2.3 Task 2 achievements

During EURAD-GAS, numerous experiments and modeling activities were conducted, resulting in significant progress in the characterization and the understanding of gas transport processes. This progress is presented hereafter in the form of answers to some fundamental questions about gas transport identified at the start of the project.

How does clay permeability and its anisotropy influence the diffusion parameters?

The analysis by UKRI-BGS of the results from gas diffusion tests obtained on Boom Clay with regards to its anisotropy suggests that helium diffuses preferentially along bedding planes with approximately 60% of the diffusional capacity of Boom Clay moving parallel to bedding. Examining these data as a whole in comparison with the clay intrinsic permeability suggests that a fundamental relationship exists between permeability and diffusivity. Such relationships may be useful to predict diffusivity across a range of material and permeability scales.

Up to now, diffusion of gases was measured mainly on samples of a few centimeters in diameter and not at larger scale. The NEMESIS experiment, which measures diffusion of neon on a meter scale in Boom Clay, was launched in the frame of EURAD-GAS with the aim to upscale diffusion of dissolved gas in both directions (parallel and perpendicular to bedding) (Jacops *et al.*, 2023). This 3D in situ through-diffusion experiment will run for at least five years to capture the anisotropy of the diffusion coefficient of neon. Preliminary results obtained during the preparation phase on in-diffusion tests[4] with helium suggest a in situ diffusion coefficient of about 80% of the value measured perpendicularly to the bedding planes in laboratory experiments.

How do petrophysical parameters, like mineralogy or density, and conditions such as the stress state, influence free gas transport?

Experiments carried out by UKRI-BGS on Boom Clay and Callovo-Oxfordian claystone showed that (1) transport through dilatancy-controlled gas pathways was the only mechanism observed at the tested gas injection rates (which are higher than those expected in a geological system); (2) the control of the gas-entry pressure was the minimum principal stress and (3) the dilatancy-controlled gas pathways are not classical fracture related. So far, no firm conclusions can yet be drawn regarding the role of gas injection rates and stresses as controls of the physics of gas flow and whether there is a transition from visco-capillary two-phase flow to dilatancy-controlled gas flow.

Gas injection experiments in synthetic samples mimicking bentonites by UKRI-BGS leads to the following key learning points: (1) swelling pressure is correlated with clay content in engineered clays and anti-correlated with sand + silt fraction in synthetic mixtures; (2) at fully saturated conditions, dilatancy-controlled gas flow can occur even at low clay contents; and (3) clay content appears to play a significant role regarding the stress field behavior and to directly relate to gas-entry pressure.

Gas entry and flow through Czech Ca-Mg BCV bentonite at different dry densities by CTU showed (1) the formation of dilatancy-controlled gas pathways via gas-exerted pressure occurs following the exceeding of the measured axial total stress, which is related to the swelling pressure of the material; and (2) the type of gas injected exerts no influence on the breakthrough pressure value. Gas injection experiments in BCV and MX-80 bentonites at two different densities by ÚJV Řež showed no difference with respect to the tested gas (air versus hydrogen). The experiments confirmed the self-sealing

[4] Unlike a through-diffusion test, an in-diffusion test does not capture the anisotropy of diffusion coefficient. The estimated value is an average value deduced from the pressure loss in the injection filter to diffusion.

capacity of BCV. In addition, all bentonite materials behaved in a similar way regarding gas flow mechanisms.

How is gas diffusivity changing in partially saturated clay?

Depending on the host rock and disposal design, slightly unsaturated conditions may be encountered. Therefore, the impact of desaturation on gas diffusivity was studied in EURAD-GAS on synthetic samples, containing different amounts of clay, silt and sand, at saturation degrees between 73% and 100%. Results showed that gas diffusivity, as a combination of diffusion of dissolved gas and gaseous gas, increased only slightly when desaturation of the sample is limited. Indeed, the diffusion of dissolved gas should decrease with desaturation whereas the diffusion of gaseous gas should increase. At high saturation degree, these effects are limited and cannot be clearly seen in the balance as expected from Archie's law (Savoye *et al.*, 2010). The analysis of porosity profile with imaging techniques performed by IRSN shows that even if desaturation takes place in the largest pores first, desaturation front can be viewed as homogeneous within subcentimeter size samples, even when close to the full water saturation.

Modeling of gas diffusion in saturated and unsaturated conditions using molecular dynamic simulations (Owusu *et al.*, 2022; Owusu *et al.*, 2023; Owusu, 2023) revealed that the diffusion of gases is significantly influenced by three key factors: the mineralogy (specifically, the type of clay), the size of the pores, and the hydrodynamic radius of the gas molecule (which depends on the gas type). In unsaturated clay, gas diffusion is influenced by multiple factors, including the thickness of the water films, the mean free path of the gas molecules, and the average available pore width.

Could gas adsorption be a relevant retardation mechanism for transport under repository conditions?

Adsorption of hydrogen on clay has been considered in the past to be a potentially relevant process in a geological disposal system as it would lead to reduced gas pressures. Complementary work performed during EURAD-GAS has shown that these results could have been biased by experimental artifacts. With respect to gas adsorption, the assessed hydrogen adsorption by dry COx at 25°C and 100 bar is 0.02 wt%. However, dry conditions are not representative of disposal systems in clayey host rocks. Hydrogen adsorption by COx and Boom Clay in partially and fully saturated conditions should be very limited, however its quantitative evaluation is still ongoing at the end of EURAD-GAS. One of the conclusions of EURAD-GAS is therefore not to consider sorption as a relevant retardation mechanism of gas transport under disposal conditions (conservative assumption).

Do we understand the observed gas transport mechanisms, and which are the main controls? Can experimental results be described by models in which the representation of crack propagation and of dilatancy-controlled gas pathways is implemented at a process level?

Experiments on FEBEX bentonite (CIEMAT) confirm that below a value referred to as gas-entry pressure, the predominant transport mechanism is the diffusion in pore water (Gutiérrez-Rodrigo *et al.*, 2021; Villar *et al.*, 2021). Once this gas-entry pressure is exceeded, gas flows locally through either the existing porosity or through self-created and stress-induced pathways (the distinction between the two mechanisms of transport cannot be clearly set out in this experimental setup). Little water is displaced by the gas flow.

According to the experiments in a fracture visualization rig of UKRI-BGS, multiple dilatancy-controlled gas pathways form at the same time, with variable size, with variable velocity and the distribution of pathways is stochastic. The walls of these pathways elastically compress to accommodate the pathway and self-sealing is observed. With respect to upscaling, all observations from LASGIT point in the same direction, namely that gas is mainly transported through dilatancy-controlled gas pathways.

From these observations, standard hydraulic models should be enhanced with mechanisms to account for discrete pathways. Following this direction, different models implemented within different software have been developed in EURAD-GAS. Major progress has been made since several models have proved successful in describing key experimental features such as (1) the initiation and propagation of gas pathways inside deformable materials, (2) the sharp drop of the gas injection pressure when the gas pathway reaches the back-pressure filter, and (3) the closure of the gas pathway when the gas injection ceases (Toprak and Olivella, 2023; Corman *et al.*, 2024).

How does conceptualization of transport mechanisms on macroscale (with discrete conducting fractures) compare to microscale (with heterogeneities and deformations)?

Over the duration of EURAD-GAS, different multiscale modeling approaches have been developed, where the rock structure at a microscale level is included into the description of the macroscale gas transport mechanisms. Special emphasis is placed on the transition between the two scales, which is currently achieved by means of homogenization and localization techniques. The University of Liège has highlighted the importance to consider the connectivity between the planes of weakness to model gas breakthrough tests (Corman *et al.*, 2024). UPC proposed to use random distributions of permeability zones (Toprak and Olivella, 2023). IRSN and CNRS-U Lorraine noted that Direct Numerical Simulations approach could be combined with FIB-SEM images to compute upscaled properties (Amrofel *et al.*, 2024).

While these different multiscale modeling approaches are able to represent key phenomena observed in laboratory-scale results, predictive capabilities and upscaling to the in situ-scale behavior are still challenging, since the microstructural heterogeneity of some clay properties (e.g., stiffness, strength, porosity or permeability distributions) and the connectivity of the macropores and microfractures created cannot be easily defined a priori and at a large scale. Multiscale approaches also require large computational capacity to describe domain sizes smaller than the representative elementary volume (REV). These computational demands for upscaling heterogeneities and geometries are currently seen as limitations to the use of multiscale approaches at large-scale.

3. Task 3 – Barrier integrity

Chapter 3 presents the objectives of Task 3 as defined at the beginning of the project (Section 3.1), followed by an short overview of the main results (Section 3.2), modeling developments (Section 3.3), and achievements (Section 3.4). Section 3.5 provides Task 3 inputs for the assessment of gas transport at the repository scale. Detailed scientific achievements are available in the thematic technical report: B*arrier integrity: gas-induced impacts and model-based interpretation* (Deliverable 6.8, Task 3) (Marschall *et al.*, 2024). The full integration of the results is presented in Deliverable 6.2 (S*tate of the art on gas transport in clayey materials – Update 2023* (Levasseur *et al.*, 2024)).

3.1 Task 3 key objectives and work program

Accumulation of gas in the backfilled structures of a deep geological repository associated with the build-up of excessive gas pressures may impair the long-term safety functions of the repository's multibarrier system, namely the radionuclide retention and attenuation capacity of the engineered barrier system (EBS) and the geological barriers. Clayey rocks and clayey EBS are particularly prone to failure due to their low mechanical strength. On the other hand, it is well known that clays exhibit the favorable feature to self-seal after sustaining mechanical failure. Task 3 was aimed at gaining a mechanistic understanding of the hydro-mechanical phenomena and processes, associated with:

- the gas-induced failure of clay barriers, i.e., within the engineered barrier system (EBS), within the excavation damaged zone (EDZ) and within the host rock (Subtask 3.1);
- the effectiveness of self-sealing processes along dilatancy-controlled gas pathways in the clay barriers of a geological repository (Subtask 3.2).

The evaluation of achievements was accomplished by model-supported data analyses, predictive modeling, and the application of the newly developed modeling tools on in situ experiments (Subtask 3.3).

As mentioned in Task 2, after the FORGE EC project, the hydro-mechanical couplings between the gas phase, the pore water, and the total stress, were still a priority research issue. For bentonite barriers, the task benefited from the results of the BEACON EC project that characterized the hydro-mechanical coupling of swelling clay materials during the resaturation phase. This knowledge transfer was facilitated by the involvement of several BEACON members in this task.

3.1.1 Subtask 3.1 work program

Laboratory experiments that were designed and executed, aimed at investigating the evolution of damage in clayey materials when subjected to excessive water/gas pressures. Different test setups and different test procedures were required for the investigation of argillaceous formation and EBS material, respectively.

Potential clayey host rocks

Fracture initiation and fracture propagation processes in the intact potential host rock, fracture reactivation in the excavation damaged zone (EDZ), and the reactivation of tectonic features depend on the rock fabric and on the prevailing in situ conditions (pore pressure and stress). Fracture opening and fracture sliding mechanisms were investigated along typical stress paths, which are representative for real repository conditions:

- Hydro-mechanical laboratory experiments were performed on potential clay host rock samples. The test setups ensured the precise control of water/gas flow, pore pressure/total suction, stress state and strains during all test stages (isostatic/triaxial cells). The impact of texture and tectonic overprint on gas-induced damage evolution was addressed by testing the samples parallel, perpendicular and/or at oblique angles to the principal orientations of microfabric, respectively.
- Further phenomenological experiments were conducted by using a direct-shear-rig for water/gas injection experiments. Shear fractures were created within the apparatus while tensile fractures were created externally. Quantitative textural information of all fracture surfaces was determined prior, and after, detailed gas testing. The evolution of damage and self-sealing processes in the sample during gas injection and after shut-in was followed by using a high-resolution x-ray computed tomography and/or by microstructural analyses.

Engineered clayey materials

Studied engineered clayey materials included compacted granular bentonite, bentonite blocks and sand/bentonite mixtures used as buffer materials (mainly backfill and seals).

Long-term water/gas injection tests in oedometric/isostatic cells were conducted to investigate dilatancy-controlled gas flow. The laboratory experiments were performed to reveal the impact of the as-compacted state (e.g., dry density, grain size distribution, initial water content) and the hydration process (e.g., imbibition process, water chemistry) on the gas breakthrough pressure of the hydrated material.

Microstructural analyses of the investigated geomaterials (damaged or intact potential host rocks and EBS materials, respectively) before and after gas injection was conducted to get insight in the prevailing failure mechanisms. This included the evaluation of apertures, extensions and surface roughness of the induced fractures and statistical descriptions of the pore networks of the intact and of the damaged material.

3.1.2 Subtask 3.2 work program

From a mechanistic point of view, the self-sealing capacity of clayey materials is associated with various (thermo-)hydro-mechanical and chemical processes and controlled by the prevailing state conditions. Mechanical closure of fractures (e.g., crack closure, fracture sliding), hydro-chemical interactions of the pore water with the clayey 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 clayey materials. In this context, it is important to consider also phenomena and processes that could prevent fracture closure, such as mineral transformations and hydro-mechanical reorganizations of the fabric caused by gas seepage through the clayey barriers over long time periods of several 10 000 years.

The self-sealing capacity of the clayey materials was investigated with the same test configurations as used for the gas injection experiments. After termination of the gas injection experiments, the test specimen was subjected to different pressure recovery conditions including resaturation with different water compositions and different total stress scenarios. Subsequently, final water permeability testing was conducted to qualify the loss of hydraulic barrier function of the test specimen. Different potential host rocks and different fracture modes were addressed. Visualization techniques such as micro-computed tomography (μ-CT) or scanning electron microscope (SEM) allowed revealing the microstructural changes of the investigated test samples at the end of the self-sealing stage. The phenomenological gas injection experiments were also extended beyond the shut-in phase to visualize at pore scale the complex hydro-mechanical-chemical (HM-C) interactions, which are associated with the self-sealing process.

The self-sealing studies of this subtask were closely connected to the ones performed in Subtask 2.1 of EURAD-HITEC ("Clay host rock" task – "Experiment near field with EDZ" subtask). The aim of these studies were to characterize (T)HM(-C)(+gas) processes associated to self-sealing mechanisms in clayey materials.

3.1.3 Subtask 3.3 work program

Subtask 3.3 was dedicated to the interpretation of the experimental results and the synthesis of the scientific achievements (development of process models for dilatancy-controlled gas flow and self-sealing). The modeling teams contributed to the experimental design of the laboratory experiments and the model-based interpretation of the acquired experimental results. Back-analysis of the experiments fed in the development of conceptual process models of gas-induced damage evolution and self-sealing processes for potential damaged or intact host rocks and EBS materials, respectively. Validation of these models took place in a series of prediction-evaluation exercises, covering different loading paths in different geometric configurations (potential host rocks: isostatic/triaxial; EBS material: oedometric/isostatic).

3.2 Subtasks 3.1 and 3.2 research activities per partner

This subsection presents the research activities per partner in Subtasks 3.1 and 3.2, divided in such a way as to address the general objective of Task 3. Details of the results per partner are reported in Marschall *et al.* (2024). Main collective achievements are summarized in Section 3.4.

3.2.1 Research activities related to the engineered barriers

The favorable barrier properties of clayey materials (low permeability, high swelling capacity, contaminant retention capacity, mineralogical stability in the conditions of geological disposal systems) suggest their use as sealing elements in EBS. Most geological disposal designs worldwide use bentonite as sealing material, a naturally occurring absorbent swelling clayey material with a low permeability (Levasseur *et al.*, 2024). Depending on the intended safety functions and the corresponding design specifications of the engineered barrier system, bentonite is processed in different forms and compacted to various densities.

Two experimental activities were conducted in Task 3 with focus on gas-induced failure and self-sealing capacity of bentonite:

- **CTU – Cyclical water and gas injection experiments with Czech BCV bentonite.** Phenomenological studies were carried out with constant volume cells, comprising cyclic hydration/gas invasion tests on BCV bentonite samples of different dry densities. Tests were conducted both on homogeneous samples and on samples with an artificial joint. The experimental program complements previous studies on gas transport in bentonite, which were carried out with MX-80. The objective was to evaluate whether different kinds of bentonite have a similar behavior[5]. This aspect complements experimental work carried out in Task 2 by the same team.

- **IRSN – Gas migration processes in initially heterogeneous bentonite mixtures.** The evolution of microfabric of a mixture of MX-80 pellets with bentonite powder was investigated in response to hydration and gas invasion processes. The hydration/gas invasion processes of the pellet/powder mixture in a constant volume cell were monitored by x-ray computed tomography. Data analyses included the evolution of porosity, gas/water permeability and local displacements of the solid skeleton, highlighting the paramount relevance of microfabric for a mechanistic understanding of gas transport processes in bentonite.

3.2.2 Research activities related to the geological barriers

When subjected to excessive gas pressures, clayey rocks are particularly prone to failure due to their low mechanical strength. The impacts of gas accumulation, gas pressure build-up and gas transport processes on the performance of the clayey barriers depend not only on the hydro-mechanical properties of the barrier materials but also on the environmental conditions at repository level, on the repository design and on various other aspects such as waste-related source terms. Consequently, an in-depth understanding of the gas-related failure mechanisms is required for a balanced assessment of the safety-related impacts on barrier performance (see also (Levasseur *et al.*, 2021)).

The mechanical characteristics of geological clayey barriers cover a wide property range in terms of strength and stiffness, representing the full spectrum of deformation behavior in the transition between indurated and poorly indurated clays (see also (Gens, 2013)). Three potential host rocks were investigated in Task 3, namely Boom Clay (BC), a poorly indurated clay, Callovo-Oxfordian (COX) and Opalinus clay, two indurated clays.

Five laboratory experiments were conducted in Task 3 with focus on gas-induced failure and self-sealing capacity of clayey host rocks:

- **GRS – Gas transport along fractures in clayey rocks and impact of self-sealing.** The experimental program consisted of long-term self-sealing and gas injection tests in triaxial cells on artificially fractured claystone (COx, OPA). Exceptional databases were acquired by monitoring the evolution of hydraulic conductivity, axial and radial strains over a period of up to 700 days. Gas injection tests on the self-sealed material were aimed at exploring the gas pressure needed to invade the self-sealed material under a given confining stress. Self-sealing tests after completion of gas injection sequences and postmortem investigations of the dismantled test samples complemented the experimental program (Zhang and Talandier, 2023).

- **CNRS-U Lorraine – Visualization of gas transport in fractures and impact on their self-sealing capacity.** Water and gas permeability tests with flow direction parallel and perpendicular to bedding were conducted in triaxial cells on intact and artificially fractured claystone (COX). The innovative aspect in the experimental program was the use of a µ-CT-equipment for the visualization of gas transport processes during gas invasion phase and, subsequently, the self-

[5] An important issue considered in EURAD-GAS is the type and the quality of the bentonite. A broad range of different qualities with respect to exchangeable ion, accessory minerals and production techniques are used by the waste management organizations, depending on the intended application. Comprehensive geotechnical databases exist for MX-80, encompassing not only the basic geotechnical characteristics of the material but also dedicated investigations on gas-induced failure and its self-sealing capacity. Extending this database to other types of bentonites is of a high interest for a valuable transfer of knowledge for all radioactive waste management organizations.

sealing processes in response to rehydration of the sample. Evolution of permeability, volumetric strain and fracture volume were monitored during the gas invasion phase and during rehydration (Agboli *et al.*, 2023).

- **UKRI-BGS – Effects of gas transport on fracture transmissivity and self-sealing.** A novel test concept for fracture transmissivity measurements under well-defined shearing conditions was pursued using a highly instrumented (normal stress, shear stress, shear displacement, normal displacement, pore water pressure/flow, etc.) bespoke direct shear apparatus. The experiments were conducted with indurated claystone (COx, OPA). The test results encompassed not only the transients of gas/water flow, stresses and shear strains but also scans of the fracture surfaces before and after testing. Such complementary spatial information about the evolution of the fracture surface in response to gas injections and re-hydrations provides invaluable insights in the gas transport and self-sealing mechanisms.

- **EPFL – Gas transport in intact and remolded/recompacted claystone.** Combined water/gas injection experiments were conducted on indurated clayey samples (Opalinus clay, OPA) using a high-pressure oedometric cell, specifically designed for testing at high confining stresses. Special focus was on accurate measurement of the volumetric behavior of the material during the entire test sequence, comprising the initial sample equilibration phase, water permeability tests, gas injection tests, rehydration tests and a final pressure recovery phase. The experiments were carried out on intact material and on material, which had been remolded and recompacted for mimicking the microfabric of fault gouge material. The impact of gas transport on the microfabric of the test samples was studied as part of a postmortem investigation program.

- **CIMNE – Hydro-mechanical response of claystone on gas injections.** Combined water/gas injection experiments were conducted with a poorly indurated clay (Boom Clay). For this, a new oedometric cell was designed and tested, allowing for highly accurate measurements of the volumetric behavior of the material and for monitoring of the lateral stress acting on the test sample. Outstanding experimental results were achieved with the new cell, indicating a distinct dependence of the gas transport capacity of the Boom Clay samples on the gas injection rate. Postmortem studies indicated a measurable impact of the gas injections on the microfabric of the material (Gonzalez-Blanco *et al.*, 2023).

3.2.3 Two-phase flow properties derived from pore-scale imaging

Methods of digital rock physics (DRP) such as µ-CT or SEM imaging are emerging as a complementary part of the geotechnical characterization of clayey materials. The digital outputs can be used to create a physically based model of the material under investigation. IRSN and CNRS-U Lorraine used such techniques successfully to visualize the pore space evolution of bentonite mixtures and of fractured COx in response to hydration, gas injections and subsequent rehydration phases. UKRI-BGS has developed a method which allows to scan fracture surfaces and to reconstruct fracture aperture in three dimensions (3D). Imaging and reconstruction of the pore networks of intact claystone with dominant pore sizes in the order of 10 nm is a topic of ongoing research, even though pioneering work has been carried out in this field during the last decade (e.g., (Keller *et al.*, 2013)).

A dedicated research activity (ZHAW) in Task 3 was aimed at developing new workflows for the analysis of digital pore models (Keller, 2021; Keller, 2022; Keller, 2023):

- Development of workflows for statistical analysis of 3D images of pore size distribution as input for water retention behavior and relative permeability – saturation relationships;

- Development of workflows for statistical analysis of fracture roughness as input for mechanistic models for damage evolution and self-sealing (e.g., shear-box experiments).

3.3 Subtask 3.3 main modeling developments

This subsection summarizes the final results of Subtask 3.3 achieved by each partner of the Task 3 and reported in Marschall *et al*. (2024).

Subtask 3.3 activities mainly focused on the development of conceptual process models of gas-induced damage evolution and self-sealing processes for damaged or intact host rocks and EBS materials. These models were then validated in a series of configurations of relevance for geological disposal in clayey host rocks. The experimentalists of Subtasks 3.1 and 3.2 closely interacted with the modelers of Subtask 3.3 during the EURAD-GAS. This facilitated a traceable and transparent handover of data to the modelers, which was used as input for the development of gas-related process models, for model benchmarking and for back calculation of experimental data.

Important model developments were performed during EURAD-GAS concerning the process chain and code implementations, e.g.:

- **In CODE_ASTER (EDF):** Coupling of two-phase flow model with elasto-viscoplastic mechanical law with second gradient (i.e., H2M model). Development of couplings between permeability and plastic deformation (Mhamdi Alaoui *et al.*, 2023; Faivre *et al.*, 2023).
- **In CODE_BRIGHT (UPC):** 3D heterogeneous, coupled hydromechanics and transport of gas (HM-G), BBM + cubic law for permeability, comprehensive protocol of hydration intervals and gas injection intervals (Toprak and Olivella, 2023).
- **In LAGAMINE (U Liège):** Extension of the second-gradient method to two-phase flow hydro-mechanically coupled conditions including strong couplings between transport properties and the deformations (Corman *et al.*, 2022). Development of a hydro-mechanical interface constitutive model to reproduce the self-sealing process in an artificially fractured sample (Quacquarelli *et al.*, 2024).
- **In LAGAMINE (TU Delft):** Development of a pneumo-hydro-mechanical (PHM) framework to model gas-induced crack initiation and propagation in clays (Liaudat *et al.*, 2023).
- **In OpenGeoSys (OGS-6) (UFZ, BGR, BGE):** monolithic TH2M model (Pitz, Grunwald *et al.*, 2023; Radeisen *et al.*, 2023; Pitz, Kaiser *et al.*, 2023; Grunwald *et al.*, 2023; Pitz *et al.*, 2024) and fracture mechanics based on phase-field method (Mollaali *et al.*, 2023).

Compared with the FORGE EC project, EURAD-GAS has made huge progress in the development of (T-)HM process models, including model calibration and back calculation of experimental data. The model portfolio obtained in EURAD-GAS provides new tools for systematic model abstraction (geometric, process abstraction, scaling/homogenization) in performance assessment as the ones detailed in the following subsections.

3.3.1 Models representing dilatancy-controlled gas flow

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 clayey materials as observed at laboratory scale. Dilatancy-controlled gas 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 Task 3 as in Task 2 of EURAD-GAS, the modeling of dilatancy-controlled gas flow was investigated by introducing permeability models that affect the gas transport in clayey materials. Different permeability models describing permeability by pore gas pressure and by deformation were developed and implemented in the open-source finite element code OpenGeoSys 6 (OGS-6) (Bilke *et al.*, 2019; Lehmann *et al.*, 2024) and in the finite element code Thebes.

To validate these models, the modeling of a gas injection test in Opalinus clay performed by Popp *et al.* (2007) from Institut für Gebirgsmechanik (IfG) has been carried out by BGE and Aalto University (Gupta, Abed *et al.*, 2023b). In this test, a reversible dilatant behavior of the clayey sample upon gas injection was provoked. 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 when the permeability is coupled with the mechanical strains. Based on the obtained results, it can be concluded that the dilatancy-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 clayey material.

Further numerical investigations were conducted in order to verify the occurrence of visco-capillary two-phase flow in low permeable clays. For this purpose, BGE modeled an experiment conducted at EPFL by Minardi (2018). This experiment explored the hydro-mechanical behavior of water-saturated samples during gas injection at varying gas pressures. The results of this experiment evidence that the sample showed a reversible behavior during the experiment. Minardi (2018) interpreted this finding as evidence of visco-capillary two-phase flow as the predominant gas transport mechanism in this experiment.

The experiment proceeded in two stages. First, the sample underwent resaturation. Subsequent to achieving water saturation, a water pressure gradient was applied across the sample. Upon reaching a steady state, hydraulic conductivity was calculated following Darcy's law. After the first stage, the upstream water pressure was halted, and gas (air) was injected. Concurrently, the downstream water pressure was reduced and then held steady. Both radial and axial stresses remained constant during this gas injection period.

BGE simulation outcomes, using the numerical process model in OpenGeoSys developed by Grunwald *et al.* (2022) that accounts for the visco-capillary two-phase flow, align well with the experimental data. The model indicates that only water contributes to the outflow volume, as no gas outflow is discernible at the model's downstream end. Further numerical examinations of the saturation evolution within the sample have shown that the sample remained saturated during the experiment. It results from these observations that the gas did not penetrate into the pores of the clayey sample. From the modeling perspective, BGE concluded that the visco-capillary two-phase flow was not the dominant gas transport mechanism in this experiment. This confirms the conclusions of the FORGE EC project stating that no experimental evidence of two-phase flow could be identified in experiments on low permeability porous media such as clays and bentonite close to saturation with water (Shaw, 2013). Thus, the dilatancy-controlled gas flow can be seen as the predominant gas transport mechanism in clays at gas pressures below the gas fracturing level. More experimental investigations are necessary to examine the conditions of under which visco-capillary two-phase flow occurs in low permeable clayey materials.

The activation and prevalence of the different gas transport mechanisms depend on several factors, including the hydraulic and mechanical properties of the clay, the gas pressure at the injection locus and the hydro-mechanical state of the material. A new pneumo-hydro-mechanical (PHM) modeling framework for crack initiation and propagation has been developed and implemented in LAGAMINE (Liaudat *et al.*, 2023). The modeling approach uses continuum elements to represent the mechanical and flow processes in the bulk clayey material and zero-thickness interface elements to represent existing or induced discontinuities (cracks). Thereby, gas flow through the bulk material (through diffusion and advection of dissolved gas, and two-phase flow) as well as existing or induced discontinuities can be modeled within a single modeling framework. The developed framework enables the investigation of the factors controlling the onset of each gas transport mechanism and the interaction between these mechanisms.

The H2M model with a viscoplastic law (LKR) and second gradient has also been applied to triaxial tests with gas injection on COx clays (EDF, CNRS-U Lorraine). A straightforward relationship between permeability and deformation allows the fitting of permeability evolution due to deviatoric stress. Main tendencies observed in such a test with gas injection are well reproduced depending on the initial state of the sample.

The second gradient H2M model for gas transport in EDZ and surrounding intact rock has been successfully applied to MEGAS and MAVL experiments in Boom Clay and COx, respectively (Corman *et al.*, 2022; Corman, 2024).

3.3.2 Models taking into account heterogeneity

UPC developed an elasto-plastic model derived from the Barcelona Basic Model (BBM) to simulate irreversible strains induced by gas injection under heterogeneous conditions. Two types of heterogeneities are observed during gas injection tests. The first type of heterogeneity is associated with the variation of dry density (porosity) during hydration. Gas breakthrough pressures are associated with the dry density after hydration (while swelling pressure varies according to dry density) prior to gas injection rather than the initial dry density. Consequently, the hydration step is integrated in the modeling in order to improve the model predictions. The second type of heterogeneity is related to gas effective permeability. In intact clay (without advection of free gas), gas permeability and water retention curves are porosity dependent. In damaged clay (that includes dilatancy-controlled gas pathways), in contrast, gas permeability and water retention curves are strain dependent. The magnitude of the apertures in dilatancy-controlled gas pathways is variable. Hence, UPC has shown that considering at least two different dilatancy-controlled gas pathways with different aperture characteristics in their model allows not only to reproduce the development of swelling pressures during the hydration part but also to simulate irreversible strains induced by gas injection under the heterogeneous model configuration (Toprak and Olivella, 2023).

3.3.3 Models including self-sealing

During excavation, an excavation damaged zone (EDZ) is created, where the hydro-mechanical properties of the host rock are modified. The EDZ is characterized by a network of shear and tension fractures, where the permeability increases. However, after circulation of water within the fracture, it is observed both at the laboratory scale and in situ that the initial permeability is progressively recovered. The underlying process is the self-sealing of the fractured zones. A hydro-mechanical model for an interface element was developed during EURAD-GAS by U Liège (Quacquarelli *et al.*, 2024). It encompasses two low-density and fairly compressible zones around the fracture. They are integrated into the interface elements to avoid their re-meshing, with a considerable simplification of the problem. Thanks to the clay transmissivity, the water injected can permeate the clay, first involving the EDZ and then the rest of the sample. This model is able to reproduce the evolution of crack opening during wetting and drying tests on artificially fractured COx samples as well as reproducing the influence of the confining pressure on the self-sealing process. The upscaling of such an approach from the laboratory scale to the gallery one remains an open question.

3.3.4 Coupled hydro-mechanical models for EDZ

An important aspect that goes along with the creation of an EDZ in clayey host rocks is the modification of their hydro-mechanical properties, which has a non-negligible impact on gas transport processes. The numerical modeling should therefore not only reproduce fracture development inherent to the EDZ but also integrate strong interactions coupling the flow and transport properties (e.g., intrinsic permeability, retention curve) to the mechanical behavior. In this way, the models developed thanks to EURAD-GAS demonstrates an ability to replicate the transition between gas transport mechanisms with increasing gas pressures in line with the current state of knowledge in the EDZ (as for instance done by U Liège (Corman *et al.*, 2022)). First, a slow background process of gas transport by diffusion is certain to occur but with a limited capacity of gas transport. For larger gas production sequences, the activation of a visco-capillary two-phase flow mechanism in the discontinuities leads to a faster propagation of gas through the entire EDZ in the form of a gaseous front. In the model, the transition from purely diffusive gas transport to gas transport that combines diffusion and visco-capillary two-phase flow is more pronounced when the EDZ is active (that means that fractures in the EDZ are considered open and/or sealed fractures re-open because of gas). Yet, some uncertainties remain in the ranges of variation of the key flow parameters that govern the two-phase flow model, which in this scenario is the dominant transport mechanism.

3.4 Task 3 achievements

The achievements of Task 3 are divided in two parts, from the perspective of end-users (Subsection 3.4.1) and from perspective of the scientists (Subsection 3.4.2).

3.4.1 Achievements from the end-users' perspective

According to Levasseur *et al.* (2021) *"… the results of previous efforts on the identification and characterization of the possible gas transport processes suggest that the mechanisms at play in different clays are generally similar"*. In other words, in the recent years a fundamental level of trust has been developed regarding the effective gas transport mechanisms and the gas-related failure mechanisms, which is complemented by a mutual understanding of the efficiency of self-sealing mechanisms. On the other hand, the report revealed honestly a deficiency of quantitative approaches and modeling tools, which are needed to implement such basic knowledge in quantitative performance assessment workflows for a sufficiently wide range of relevant repository settings.

Significant further achievements have been made in the course of the EURAD-GAS project, both on the experimental and the modeling side. Their evaluation is carried out in the subsequent sections with reference to the questionnaires, which were compiled in the SOTA 1 and SOTA 2 reports to formulate the end-users' needs according to the general mission of the project (Levasseur *et al.*, 2021; Levasseur *et al.*, 2024).

How could gas be transported within the repository and which water-soluble and gaseous radionuclide transport could be associated with it?

Evidence for dilatancy-controlled gas transport in clayey host rocks has been reported by CNRS-U Lorraine, UKRI-BGS, EPFL and CIMNE, which builds confidence in the existing conceptual frameworks as described in the SOTA 1 report. Beyond that, new insights were gained about the volumetric behavior of poorly indurated and indurated clays in response to water/gas injections. CIMNE conducted water/gas injection tests on Boom Clay samples and EPFL did similar tests on Opalinus clay in a high-pressure oedometric cell, allowing for accurate measurements of the volumetric behavior of the tested material. Both teams could demonstrate independently with their experimental setups that clayey media exhibit a distinct drained/undrained volumetric response depending on the applied gas pressure build-up rates (respectively slow/high). The experiments of EPFL and CIMNE show that the slower the gas injection rate (i.e., the gas invasion occurs in drained conditions), the less the clay expands, and thus the weaker the effect of gas-induced microfracturing (Gonzalez-Blanco *et al.*, 2022; Gonzalez-Blanco and Romero, 2022). It can be assumed that gas transport in the clayey host rock of a real repository will occur under drained conditions due to the slow pressure build-up in the backfilled repository structures. These observations on the volumetric behavior of the Boom Clay and the Opalinus Clay is of great significance for the assessment of repository performance in these two clays, because gas pressure build-up in a real repository will take place slowly, at time scales in the order of 10^3 to 10^4 years.

Innovative visualization techniques at pore scale provide new insights in gas/liquid flow and transport of dissolved/gaseous radionuclides in clayey barriers supporting the evidence for dilatancy-controlled gas pathways. In Task 3, visualization of gas transport pathways has been the subject of several laboratory experiments. IRSN's experimental program was dedicated to the visualization of hydration and gas invasion processes in initially heterogeneous bentonite mixtures. Hydration processes and gas injections were imaged in an x-ray transparent constant volume cell with a 3D µ-CT scanner. Hydration of the initially heterogeneous bentonite mixture ended in a rather quick homogenization of the material with little residual variability in bentonite density. During the subsequent gas invasion phase, clear evidence was found for preferential gas flow along grain boundaries and interfacial flow along the wall of the constant volume cell. This indicates that the gas transport pathways in saturated bentonite are still affected by the initial distribution of bentonite density ("memory effect"), even though the hydration process reduces the density variations significantly. Future modeling strategies for seal design can make use of such insights to optimize the gas transport capacity of bentonite seals by adapting the grain size distribution of the bentonite mixtures.

CNRS-U Lorraine performed visualization experiments on gas transport and self-sealing in artificially fractured COx in a triaxial cell. Emphasis was on the fracture mechanisms, gas permeability and fracture closure for a wide range of stress conditions parallel and perpendicular to bedding. For different flow configurations, a systematical determination of the threshold for gas breakthrough and the formation of dilatancy-controlled gas pathways was carried out, representing the turning point from which the opening induces a significant increase in the gas permeability. This increase was greater when the main principal stress was parallel to the bedding planes due to the development of bedding parallel microcracks. Visualization of fracture closure during the self-sealing tests was not restricted to the qualitative description of the associated self-sealing mechanisms but included a quantitative estimation of the evolution of the crack volume with time (Agboli *et al.*, 2023). This is a new category of information that can be used in the context of digital rock physics for the development of pore-scale models and the validation of process models associated with the gas transport in fractured rock and self-sealing of gas-induced fractures.

Broad consensus has been gained among the experimentalists, confirming that gas transport through clayey materials is barely associated with any displacement of pore water from the clay matrix. Corresponding evidence is not only based on the gas injection experiments by EPFL and CIMNE, but can be deduced indirectly from the huge existing databases, which have been compiled in recent years to characterize the water retention behavior of clayey barrier materials (e.g., (Levasseur *et al.*, 2021)). Mercury intrusion porosimetry and water retention measurements on all sorts of clays indicate that a high mass fraction of typically 80 to 90% of the pore water occupies the meso- and micropores of the saturated clay matrix. This pore water can be hardly displaced by an invading gas phase; this is of high relevance for the gas-induced transport of dissolved radionuclides in clayey barriers. It can be concluded that the expulsion of dissolved radionuclides by gas is not a relevant transport mechanism.

How and to what extent could the hydro-mechanical perturbations induced by gas affect barrier integrity and long-term repository performance?

The gas-induced failure mechanisms of potential relevance are well established (Levasseur *et al.*, 2021) and have been largely confirmed within the EURAD-GAS project. Dilatancy-controlled gas flow is a plausible failure mechanism of special importance for clayey media with low tensile strength. Due to the expected microscale variability of the microfabric of the clayey material, it is conceivable that gas-induced microfractures will form before yielding (localized deformation due to the dilatant "opening" of pathways and also the localized compaction of the surrounding pores to accommodate this). The process of gas-driven microfracturing leads to the generation of new porosity and/or coalescence of pores, which is accompanied by a detectable increase in intrinsic permeability and a change in the capillary pressure-saturation relationship.

Several tests performed on bentonite by CTU showed no significant change in hydraulic conductivity and swelling pressure after one year of cyclic loading with high build-up rates. The integrity of the EBS appears to be preserved despite extreme stresses. CNRS-U Lorraine and GRS developed experimental setup showing that the gas transport through the fractured clay does not limit the self-sealing ability of the material. This ability was observed on large fractures that could be associated with the initial EDZ (Zhang and Talandier, 2023; Agboli *et al.*, 2023).

How do the gas transport mechanisms in the clayey barrier materials of a disposal system depend on the conditions to which these materials are subjected, primarily mechanical stresses and fluid pressures?

Multiple experimental evidence has been provided in Task 3 to point out the impact of the in situ state conditions (in particular pore pressure and stress) on the gas transport mechanisms in clayey host rock. GRS developed empirical relationships between the gas breakthrough pressure, water permeability and the effective confining stress (Zhang and Talandier, 2023). Further relationships were aimed at establishing new fracture closure laws. The correlation between clay mineral content and self-sealing capacity was demonstrated for COx and Opalinus clay.

CNRS-U Lorraine carried out systematic determinations of the threshold for gas breakthrough and dilatancy-controlled gas pathways formation for different flow configurations and different loading paths. A significant increase in the gas permeability was observed close to failure during triaxial test and deviatoric loading (Agboli *et al.*, 2023). The experiments can serve as input for model-based assessments, which allow to extrapolate the findings to a wider range of environmental conditions.

The shear-box experiments by UKRI-BGS were aimed at analyzing the impact of the stress state and the prevailing tectonic regime on water/gas transport in fractured Boom Clay, COx and OPA. Considerable variation was noted in initial gas flow and in the self-sealing capacity for all three rock types tested. The origin of these differences is not yet understood; further model-supported analyses are required to interpret the experimental results in the context of the prevailing hydro-mechanical state conditions. It is worth mentioning that the acquired experimental database will serve as indispensable input for model development.

Gas transport in clayey barrier materials is strongly controlled by heterogeneities at various scales, ranging from microfabric to large-scale heterogeneities such as sedimentological or tectonic features. EPFL carried out gas injection tests on remolded and recompacted OPA, aimed at mimicking the gas transport characteristics of fault gouge material. The experiments indicate a low gas-entry pressure and a high gas permeability of the remolded material, whereas the water permeability remained low. This demonstrates that natural and excavation-induced fractures in clayey rocks act as distinct preferential gas transport pathways, whereas the hydraulic barrier function of the fractured rock is maintained due to its self-sealing capacity.

What are the relevant material and fluid properties controlling these mechanisms?

Gas transport in engineered and geological clayey barriers is largely controlled by the mineralogical composition and the microstructure of the materials. Clay mineral content is typically above 40% with significant amount of swelling clay minerals such as smectites or illite-smectite mixed layers. The pore space is formed by a network of micro/meso- and macropores with typical pore sizes in the range of 1–100 nanometers. Water permeability of clays is generally low, the water retention curves display high gas-entry pressures. Geomechanically, clays are characterized by low strength, low stiffness and distinct swelling pressures. Comprehensive geotechnical databases were compiled in the SOTA 1 report (Levasseur *et al.*, 2021), which have been written at the beginning of the EURAD-GAS project. All experimental teams of Task 3 contributed to the existing databases with new geotechnical characterization of the tested material as done for instance by Zeng *et al.* (2023b) for granular bentonite (Marschall *et al.*, 2024).

How to characterize the material properties, accounting for the fact that some of these might well be affected by the passage of gas?

Reliable test protocols have been developed to characterize the basic geotechnical properties of clayey barriers. The consistency of the collected data from different geotechnical laboratories builds confidence in the quality of the existing gas-related databases for a wide range of clayey materials.

Water/gas permeability testing under well-constrained hydro-mechanical conditions may represent the most challenging type of geotechnical laboratory tests on clayey materials. In the past, the comparison of experimental results from different geotechnical laboratories has shown discrepancies. Often the origin of such discrepancies could not be explained unequivocally. Further efforts are required in this context to benchmark the test protocols and test equipment. Comparisons of experimental procedures for various loading and/or water/gas injection conditions, as initiated by UKRI-BGS, EPFL and CIMNE in the context of Task 3 may serve as a basis for launching new international benchmark exercises and ultimately defining reference lab test protocols.

Another additional source of potential bias is associated with the choice of the test material. It is common practice to choose “intact” rock samples for gas testing, which means that test material is selected with low variability of the fabric at the sample scale. Such a sample bias may lead to an overestimation of the effective gas-entry pressure and the water retention curves at the in situ scale. At the decameter to hectometer scale of gas accumulation in the backfilled repository structures, lithostratigraphic variability of the host rock and tectonic features form a more heterogeneous host rock. In essence, increasing heterogeneity of the rock mass corresponds to lower gas-entry pressure and less distinct water retention behavior, which is mainly controlled by the “flaws” in the rock fabric. Geostatistical analyses of the microfabric of OPA samples from different lithostratigraphic units provide clear evidence for the scale dependency of pore size distributions and the corresponding water retention behavior as done by ZHAW (Keller, 2021; Keller, 2022; Keller, 2023). From a safety assessment perspective, visco-capillary two-phase flow parameters derived from “intact” rock samples may represent an upper limit of gas-entry pressures and water retention curves. The influence of the natural variability of clay microstructure on gas properties needs to be verified on different potential clayey host rocks such as Boom Clay or COx.

Which gas-related processes could impair repository performance with respect to the intended safety functions radionuclide retention and waste confinement?

The SOTA 2 report (Levasseur *et al.*, 2024) summarizes the potential detrimental effects of gas produced within a geological repository after closure that could impair long-term repository performance:

- The potential for over pressurization and damage if gas would be produced at a faster rate than it can diffuse out of the multibarrier system.
- The potential for release of gaseous radionuclides to the biosphere (see Section 4.3.3).
- 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 (the latter was not addressed by EURAD-GAS).

In the light of the new experimental results obtained in Task 3 of the EURAD-GAS project, the potential for accelerated gas-driven expulsion of dissolved radionuclides in particular can be downgraded in terms of its safety relevance. The recent gas injection experiments by CIMNE, CNRS-U Lorraine, UKRI-BGS and EPFL on Boom Clay, COx and OPA provide clear evidence for negligible pore water expulsion in response to gas invasion, which is expressed in the two-phase flow formulation of gas transport processes by a marginal phase interference (distinct separation of the relative permeability curves for gas/water).

The feature of drained/undrained behavior of clayey rocks observed by CIMNE (Boom Clay) and EPFL (OPA) may be considered as new evidence to consider in the evaluation of the potential for gas-induced over pressurization and damage. It can be assumed that gas transport in the clayey host rock of a real repository will occur under drained conditions due to the slow pressure build-up in the backfilled repository structures. The recent experiments have shown that the observed strains in response to slow gas invasion (i.e., drained conditions) are less pronounced than in the case of fast pressure build-up (i.e., undrained conditions).

What are the safety-related consequences on the barrier properties during and after the passage of gas to be considered?

During gas passage, the transport of dissolved radionuclides by advection and diffusion is limited, because the gas phase preferentially occupies the macropores of the clayey matrix and blocks the most transmissive transport pathways for dissolved radionuclides. Diffusion and advection of dissolved radionuclides is restricted to the connected pore space constituted by the meso- and micropores. The observed low phase interference is a clear indication of the insignificant role of pore water expulsion during gas passage. Even in the case of dilatancy-controlled gas transport, there is no evidence for major desaturation of the clayey matrix.

The investigation of self-sealing of gas-induced (micro-)fractures has been the subject of all experimental teams involved in Task 3 (CIMNE, CNRS-U Lorraine, CTU, EPFL, GRS, IRSN, UKRI-BGS). All teams report an efficient self-sealing of the tested clayey materials (engineered barriers, geological barriers). Visualization of gas-induced fractures during the rehydration process provides convincing evidence for the efficient self-sealing of clayey materials.

3.4.2 Further achievements of general scientific interest

During Task 3, a wealth of new experimental results has been collected, which are of general scientific interest from the viewpoint of the radioactive waste management organizations. The questionnaire in previous subsection addresses only a little fraction of relevant achievements in the field of gas-induced impacts on barrier integrity.

In this context the confirmation and extension of the comprehensive gas-related databases for a wide range of clayey barriers are of invaluable relevance for building a mechanistic understanding of gas transport in engineered and geological clayey barriers. From a performance assessment perspective, it is not only the clarification of scientific issues of gas-induced impacts that build confidence in the performance of the disposal system, but also the confirmation of a sound understanding of the basic phenomena and processes. A rigorous and honest evaluation of phenomena and processes of potential safety relevance is an indispensable step to build trust in performance assessment workflows.

CTU's comprehensive laboratory program consisting of cyclical water and gas injection experiments with Czech bentonite BVC is a typical example for the extension of existing databases which is of value for all radioactive waste management organizations. The experimental program complements previous studies in bentonite barriers, which were mostly carried out with MX-80. A long-lasting laboratory program was executed by CTU using oedometric cells and consisting of a series of cyclic gas injection and resaturation phases. Water and gas injections were conducted with homogeneous bentonite samples of different dry density and with samples that included a discontinuity. In good agreement with other types of bentonites, the experiments did not reveal significant changes in the self-sealing performance of the material (hydraulic conductivity and swelling pressure) after one year of cyclic loading. An interesting detail of the results with the heterogeneous test samples was the formation of distinct preferential gas pathways even after long resaturation times. A similar behavior has been observed in IRSN's experiments, where gas transport pathways in saturated bentonite were still affected by the initial distribution of bentonite density, even though the hydration process had reduced the density variations significantly. Imaging of the hydration processes and gas injections with a 3D µ-CT scanner may provide further mechanistic insights in the saturation processes during hydration and flow localization during the gas injection phases. The experimental results of CTU demonstrate that the BVC bentonite may represent a reliable alternative to the expensive MX-80 which is used in other geological disposal programs.

From the end-users' side, the remarkable experimental databases for model development may be seen as important achievements. The close interactions of the experimentalist with the modeling teams of Task 3.3 during the EURAD-GAS progress meetings facilitated a traceable and transparent documentation and handover of data to the modelers. Innovative new visualization techniques allowed to test state-of-the-art methods of digital rock physics. The use of 3D visualization data to reconstruct digital pore structures and fracture surfaces is now applicable for multiphase flow and transport simulations and, in a longer perspective, may help to build digital models of the geological and engineered barrier systems at spatial scales from micro to macro. The use of the experimental databases of Tasks 3.1 and 3.2 by the modeling teams of Task 3.3 is essentially documented in Marschall *et al.* (2024).

3.5 Input from Task 3 for the assessment of gas transport at the repository scale

The dominance of dilatancy-controlled gas flow above a certain pressure threshold has been demonstrated experimentally at laboratory scales in clay media with low tensile strength (such as Boom Clay, COx and OPA) (Marschall et al., 2024). Such media cannot withstand long-term gas pressures with a magnitude greater than the minimum principal stress acting on the material. Due to the expected microscale variability of the geomechanical material properties, it is plausible that microfractures will form before yielding (ductile fracturing). The process of gas-driven microfracturing leads to an increase of porosity and/or coalescence of pores, which is accompanied by a detectable increase in intrinsic permeability and a change in the capillary pressure-saturation relationship. Porosity, permeability, and capillary strength are linked to rock deformation through empirical relationships.

In dilatancy-controlled gas flow, the gas flow is still controlled by visco-capillary forces. However, the main difference compared with the conventional visco-capillary two-phase flow is that the transport properties of the solid phase can no longer be viewed as invariants, since they depend on the state of deformation of the material. An expected consequence of this conceptual framework is a smooth transition between visco-capillary two-phase flow and dilatancy-controlled gas flow, when the geomechanical behaviour of the porous medium changes from elastic toward plastic.

At present, dilatancy-controlled gas flow through clayey materials cannot be characterized on a large scale and therefore cannot be modeled at the repository scale. The latter would require a very fine description of the mechanisms on a large scale and an explicit representation of all couplings between each individual pathway and its mechanical behavior. Nevertheless, dilatancy-controlled gas flow on a large scale can be approximated by visco-capillary two-phase flow approaches, in which the values of the parameters of the transport properties consider the state of deformation in the various components of the disposal system. In this two-phase flow approach, the mechanical behavior of clay is implicitly represented. Clay is assumed to behave elastically (the state of deformation corresponds to elastic deformation only, without any plastic deformation). This approach, used in Task 4, is conservative and overestimates the gas pressures in the host rock[6].

[6] For a given stress level, the deformation of the media is less in the case of an elastic response than in the case of an elasto-plastic response. As a result, the gas pressure will be higher in the former case.

4. Task 4 – Repository performance aspects

Chapter 4 presents the objectives of Task 4 as defined at the beginning of the project (Section 4.1), followed by an overview of the objectives and main outcomes of Subtask 4.1 (Section 4.2) and Subtask 4.2 (Section 4.3). Detailed scientific achievements are available in the thematic technical report: *Modelling of a generic geological disposal: evaluation of various approaches to model gas transport through geological disposal systems* (Deliverable 6.9, Task 4) (Wendling *et al.*, 2024). The full integration of the results is presented in Deliverable 6.2 (S*tate of the art on gas transport in clayey materials – Update 2023* (Levasseur *et al.*, 2024)).

4.1 Task objectives

Task 4 aimed at fulfilling the second high-level objective of the EURAD-GAS, which is to evaluate the gas transport mechanisms that can be active at the scale of a geological disposal system and their potential impact on repository performance. It was dedicated in particular to answer end-users' questions concerning:

- the effects of gas on the transport of soluble and gaseous radionuclides;
- the consequences of gas-induced hydro-mechanical perturbations on barrier integrity and long-term performance.

Built on the basis of the FORGE EC project, Task 4 was driven from an end-users' perspective of gas-induced effects in order to improve repository scale modeling by:

- including hydro-mechanical couplings in the analysis;
- including the transport of soluble and gaseous radionuclides in the analysis;
- promoting the use of multiple assessment approaches, supported by different numerical modeling tools.

Task 4 benefited from recent advances in phenomenological understanding from the CAST (CArbon-14 source term and fate) and BEACON (bentonite mechanical evolution) EC projects allowing respectively (i) a better understanding of potential release mechanisms of carbon-14 (in the form of methane for instance) from radioactive waste materials under conditions relevant to geological disposal systems and (ii) a better characterization of hydro-mechanical coupling in swelling clayey materials (from the installation of bentonite materials in the repository to their evolution over the long term).

Task 4 was broken down into two complementary subtasks.

4.2 Subtask 4.1 objective and main outcomes

The objective of Subtask 4.1 was to assemble phenomenological descriptions of gas transport at repository scale and of its consequences on the mechanical integrity of the host rock as well as on the transport of radionuclides, soluble or gaseous.

At the very beginning of the project, the radioactive waste management organizations of Belgium (ONDRAF/NIRAS), France (Andra), the Netherlands (COVRA), Switzerland (Nagra) and UK (NWS), as well as the French regulatory technical safety organization (IRSN) have described the current conceptualization of gas transport through repositories in their respective countries (Levasseur *et al.*, 2021). This was complemented by a brief overview of the main elements of modeling strategies for gas-related issues as developed for the needs of safety/performance assessment (SA/PA) analyses by ONDRAF/NIRAS, COVRA, Nagra, Andra and IRSN.

From these conceptualizations, a generic storyboard at repository scale was derived (Subsection 4.2.1), together with a generic repository model (Subsection 4.2.2), for use in Subtask 4.2.

All over the duration of EURAD-GAS, the radioactive waste management organizations and technical support organizations involved in Subtask 4.1 challenged and stimulated the scientific program from an end-users' perspective. This was achieved through active participation of Subtask 4.1 participants to all meetings of Tasks 2 and 3 and involvement in the reviewing of milestone documents and of the deliverables of these tasks.

4.2.1 Generic storyboard at repository scale

The generic storyboard developed in EURAD-GAS is biased toward GDFs in clayey host rocks because only two members of EURAD-GAS have chosen a disposal site so far, and in both cases the host rock is clay.

The main drivers and controls of the gas transient phase in a radioactive waste repository in a clayey rock could be as follows (Figure 2):

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[7], there is little or no flow of water into the rest of the repository from that path at this stage.
2. A slow and radial saturation of the EBS by the pore water 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. Radiolysis and decomposition of materials found in low and intermediate waste can also produce 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.

[7] 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.

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 percent 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 of the gas phase through the EBS can however be slowed down and/or hindered as it encounters seals.
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 further desaturation of the EBS (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 a maximum acceptable 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 Task 4.2 indicate that the onset of these releases can span from several tens to several hundred thousand years.

From the above, the most important elements in terms of repository performance can be identified. If a gas phase develops through the repository and even though it will consist mainly of inactive gas (hydrogen), the presence of this phase could affect the transport of water-soluble and/or gaseous radionuclides through the system:

- Dissolved radionuclide transport is not expected to be enhanced because the water displacement associated to the evacuation of gas is limited and would occur only over short distances. In fact, total system performance assessment models in most national programs considering clayey barriers and/or clayey host rocks do not currently represent directly the effects of gases on dissolved radionuclide transport. Indeed, it is assumed that the transport rate of solutes, among which soluble radionuclides, is higher in a system that would remain saturated with water, because a gas phase is an obstacle for solute diffusion. It is thus deemed conservative in estimating the transport of radionuclides in water-saturated conditions.

- Gaseous radionuclides that would not completely dissolve into the pore water upon release from the waste form can be carried toward the shafts and/or ramps along with the inactive gas (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, design dependent) and only radionuclides with half-lives around this duration or higher will present a significant concentration in the gas phase when arriving in the upper formations. In addition, all along the gas pathways, part of the gaseous radionuclides will also dissolve (as inactive gas also does) into the pore water present in the surrounding materials.

When the gas source term reduces to zero, typically after several tens to hundreds or thousands of years after closure (depending on the design), the repository will be/become fully saturated and the gas pathways (if any) will close thanks to the self-sealing capacity of the clay. This will ensure that the clay barriers maintain their very low hydraulic conductivity.

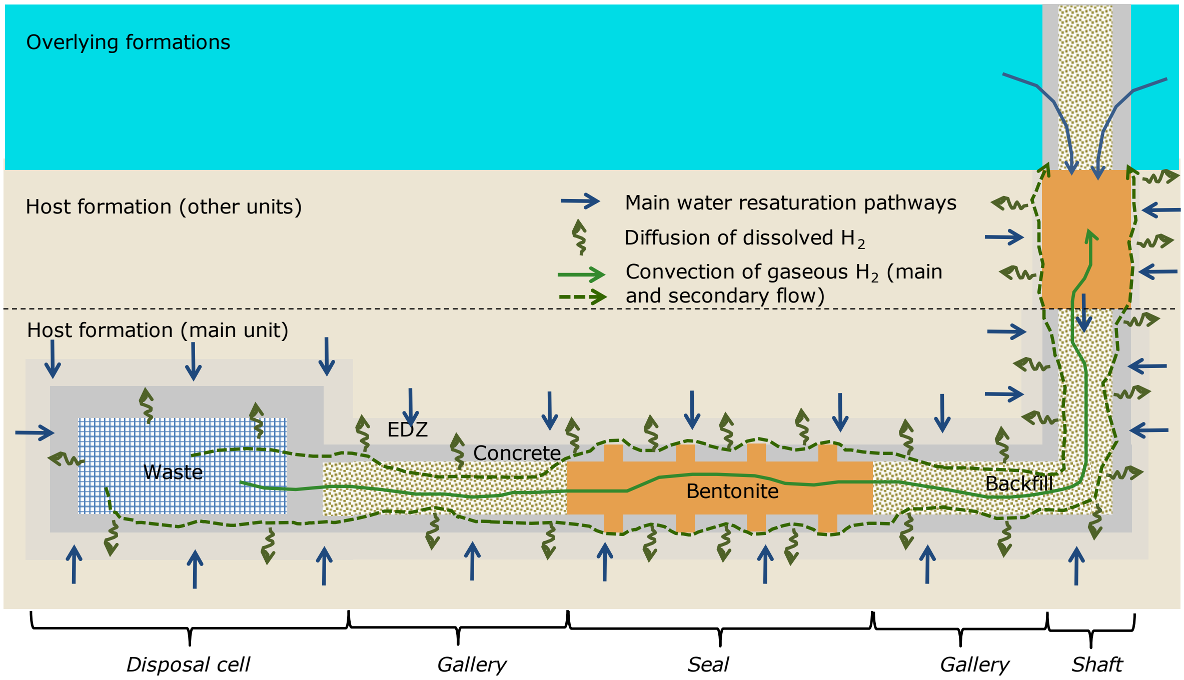

Figure 2 – Schematic representation of the main elements driving the storyboard at repository scale.

4.2.2 Generic repository model

A generic repository model is proposed, to provide a practical support for a comparison of modeling approaches at repository scale and of the use of different numerical simulation tools. The model proposed in Figure 3 incorporates features from different repository configurations considered in national programs taking part to EURAD-GAS. Salient characteristics and simplifications of this generic repository include:

- disposal on a single level,
- separation of zone for high-level, heat-emitting wastes (Zones B and C) from a zone for long-lived intermediate-level waste,
- backfilling of access galleries at once at zone closure,
- positioning of intermediate seals in the accesses to the disposal area in addition to the sealing of the (single) access shaft.

As for the storyboard, this generic model is biased toward disposal in clays but it can support the testing of approaches and tools that may be useful in other host rocks too (e.g., treatment of seals).

An example of the modeling results obtained for this generic case can be found in Narkuniene *et al.* (2023). This generic repository modeling exercise has been shared with the EURAD-DONUT, community. They have used it to develop numerical tools (see for example (Amri *et al.*, 2022; Amri *et al.*, 2023; Saâdi, 2024)).

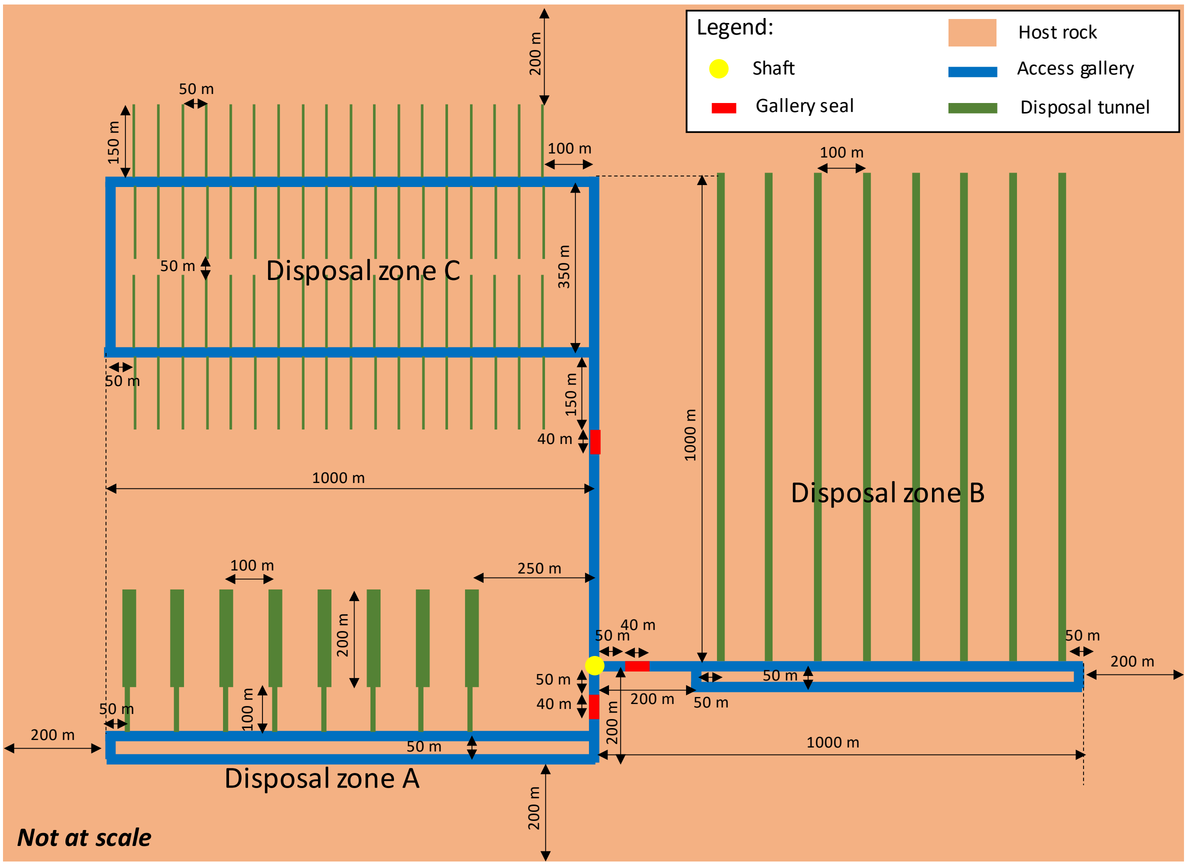

Figure 3 – Generic repository: schematic representation at main repository depth.

4.3 Subtask 4.2 objective and main outcomes

Subtask 4.2 aimed to compare modeling approaches at the repository scale and to assess the capabilities of numerical simulation tools, based on the generic repository model developed in Subtask 4.1. An additional purpose of this was to consolidate the phenomenological knowledge described in the first state-of-the-art report of EURAD-GAS (Levasseur *et al.*, 2021) and to assess the need to integrate (or not) new results and data acquired by Tasks 2 and 3 in repository-scale models, based on the results of calculations and parameter sensitivity analyses carried out. Numerical aspects were also discussed. For example, should the evaluation be based on 2D models, which are faster and allow for more sensitivity, or on 3D models, which are more representative of the disposal systems but allow for less sensitivity analyses?

The outcomes of Subtask 4.2, presented hereafter and detailed in Wendling *et al.* (2024) are generic in nature, being based on the generic storyboard and repository model proposed by Subtask 4.1 and cover:

- Uncertainties associated to the complexity of the problem and inherent limitations of modeling tools (Subsection 4.3.1);
- Assessment of the mechanical integrity of the host rock (Subsection 4.3.2);
- Assessment of the impact of gas on the transport of radionuclides (Subsection 4.3.3);
- Recommendations for a treatment of gas at the repository design stage (Subsection 4.3.4).

4.3.1 Uncertainties and inherent limitations of modeling tools

With respect to modeling of gas transport at repository scale, gas dissolution and diffusion is well understood, process-level models show good predictive capacity and upscaling to repository scale in a continuum framework is simple. For the other transport mechanisms, the current knowledge allows to estimate the threshold gas pressure for the activation of each. The capacities for gas evacuation through each of these transport mechanisms are sufficiently known to compute gas pressures and fluxes throughout a given disposal system.

The modeling approaches and tools compared in EURAD-GAS confirm the conclusions of the FORGE EC project. Currently, it is not possible to model gas transport at the repository scale while explicitly representing all couplings with the mechanical behavior of the barriers and the development of individual pathways through clayey materials.

Carrying out a complex hydro-mechanical numerical simulation on the scale of a repository was beyond the reach of all Task 4.2 teams, in the current state of computing power. The conceptual complexity of the problem implies that the modelers carrying out simulations at repository scale have to introduce simplified representations of processes, such as for example:

- Visco-capillary two-phase flow (without representation of dilatancy-controlled gas flow);
- Linear elastic mechanical behavior (without representation of the elasto-viscoplastic behavior of clays).

Carrying out two-phase flow simulations on the scale of a whole repository is possible, but this requires specific numerical approaches such as the use of "macroelements", non-conforming meshes and homogenization of materials. For example, in Task 4.2, simulations integrate:

- A mechanical coupling through the use of a single, bulk compressibility coefficient considering the compressibility of the grains of the host rock, the compressibility of water and porosity;
- Simplifications of the geometry of the components/repository taken into account component-scale/repository-scale models;
- Homogenization of the different materials present in and around the repository, for example the use of a single material on the extrados of the backfill in the galleries homogenizing a concrete lining and the EDZ;
- Simplification of initial and boundary conditions and/or their temporal evolution, for example by not considering the ventilated exploitation phase of the repository.

Task 4.2 teams (Aalto University, Andra, BGR, EDF, IRSN, LEI, SCK CEN, and U Liège) performed hydro-mechanical numerical simulations on the scale of a 2D section of a gallery or at best of a complete 3D gallery. Limited-scale models can also be built for the different components of the system that are expected to be passed through (e.g., seals, backfilled galleries, EDZ). These component models can explicitly describe the hydro-mechanical couplings. Gas pressure-flux relationships estimated from these smaller scales models under specific pressure conditions can then be integrated into repository scale model as complement to the more conceptual storyboard approaches. In turn, repository-scale model results can be compared to the assumed pressure conditions in the component-scale models, which can be adjusted if necessary.

It has to be noted, however, that except for what concerns diffusion of dissolved gas, the predictive capabilities of repository-scale models is expected to remain limited in the foreseeable future due to inherent uncertainties in the development of gas pathways that are unstable by nature and the uncertainties associated to the simplifications above, that cannot be avoided. Moreover, differences in the sets of parameters in the broad sense (mesh, homogenization, physical process, temporal management, boundary/initial conditions, etc.) produced by different teams to represent the same physical evolution on the same real object was found to lead to differences in terms of maximum gas pressure of a few MPa. Such uncertainty in the maximum gas pressure has to be taken into account in the treatment of gas at the repository design stage.

These differences integrate all the results independently of the model extension (2D, 3D at cell/zone scale, with and without exit to the shaft, total repository) and the numerical implementation of the process models (especially formulations on the way how compressibility is taken into account) in the used codes. A complementary work initiated in the framework of Task 4.2 has shown that for similar models, having the same size, the same representation of gas route toward the exit and the same set of parameters, uncertainty can be significantly reduced. However, this conclusion has to be confirmed (i) as the number of independent simulations on which this work was performed during EURAD-GAS is very limited (3, by Andra, BGR and SCK CEN) and (ii) as it is derived from a generic model and has to be verified on specific national repository design.

4.3.2 Assessment of the mechanical integrity of the host rock

In the evaluations carried out under Task 4.2, the mechanical integrity of the host rock is assessed by evaluating the maximum gas pressure in the repository. This single indicator allows, in turn, to estimate the change in stress in the different components of the repository. However, to achieve this, it is essential to consider the full disposal system. If this is not the case:

- The assessment may overestimate the maximum gas pressure when the simulated area does not include the access galleries and the accesses which represent an escape path for gases.
- The assessment may underestimate the maximum gas pressure when the simulated zone does not include the part of the repository in which the gas source term is maximum (difficult to quantify a priori).

The tools available today (computer codes, in particular the solvers, and the hardware) make it possible to carry out two-phase simulations on the scale of a whole repository. These simulations integrate the mechanical couplings through a (constant) storage coefficient (assuming linear elasticity). It is essential to match the value of this coefficient to the available data. This coefficient can be estimated directly from hydraulic experiments in transient conditions. It can also be evaluated based on its mathematical formulation, which can be different from one code to another, from the porosity, the compressibility of water and the compressibility of the porous matrix (that is related to the Young's modulus), potentially anisotropic.

This type of simulation gives a representative estimate of the maximum gas pressure in the repository if this value remains in the linear elastic part of the mechanical behavior of materials important for long-term safety. These materials are typically the seals and the host rock, but other materials may be included in this list depending on repository designs and national requirements. It is therefore important to experimentally determine the mechanical behavior of each of these materials, considering all the associated uncertainties, in order to know the stress limit beyond which linear elastic behavior is no longer representative and to be able to compare this value to the stresses resulting from the maximum gas pressure estimated in the repository, taking also into account all the uncertainties inherent in its estimation.

4.3.3 Assessment of the impact of gas on the transport of radionuclides

It is essential to represent the disposal system and its environment as a whole to correctly assess the transport of radionuclides. The gas potential exit pathways toward the EBS, including the accesses galleries, shaft(s) and/or ramp(s), the host rock, and its environment, must therefore be integrated in the simulation.

For a good estimate of the transport of gaseous radionuclides, considering the hydraulic-gas transient phase is essential as these radionuclides can migrate quickly through a gas phase essentially composed of (inactive) hydrogen and be carried along with it, if this gas phase is expanding through the system. Without taking this transient phase into account, gaseous radionuclides are supposed to dissolve and therefore migrate by diffusion in the pore water, which is a very slow process. In terms of the gaseous radionuclides fluxes through the top of the host rock and out from the shafts for the generic repository depicted in Figure 3**Error! Reference source not found.**, these two types of evaluations (with and without considering a gas phase) give estimates which can differ by several orders of magnitude, with the evaluation taking into account the presence of hydrogen as a carrier gas resulting in the largest flux.

The presence of gas means that the near field is not completely saturated. The pores filled with gas are not available for the transport of soluble, non-gaseous, radionuclides. The calculations conducted in Task 4.2 show that fluxes of soluble radionuclides through the host rock are slightly lower than those estimated under the assumption of a fully saturated environment, but still within the same order of magnitude. If an expanding gas phase displaces pore water, radionuclides dissolved in that water are displaced with it. However, results from the calculations performed in Task 4.2 for a generic repository show that water is principally displaced radially, over a short distance, around the galleries and drifts, not axially along the galleries. In general, this will depend on hydraulic properties and the state of saturation of the EBS materials (in particular on the composition of the backfill and seals but also of the structural elements) and is therefore design dependent.

Overall, regarding the impact of gas on radionuclide transport, the results in Task 4.2 are consistent with the generic storyboard at repository scale (Section 4.2.1).

4.3.4 Recommendations for the treatment of gas at the repository design stage

The results of Task 4 and the input of Task 3 for the assessment of gas transport at repository scale (Section 3.5) make it possible to propose some elements that should be integrated by end-users early in the repository design phase to manage the impact of gas on the barriers:

- Estimate the gas source term and carry out simulations of gas transport by dissolution and diffusion and by visco-capillary two-phase flow at the scale of the repository. These simulations require at least the provision of the following elements:
    - A repository architecture which in addition to the geometry of the excavations (galleries, cells, ramps, wells, etc.) integrates the location of the metals present in the different zones of the repository after closure (metallic reinforcement of concrete liners, disposal packages, primary waste packages, waste, etc.) in terms of mass and surface area, because the corrosion of these metals can be a significant or even the main part of the gas source term.
    - An estimate of the other components of the gas source term (e.g., radiolysis of organics).
    - An estimate of the gas generation rate per metal part as a function of its environment (i.e., corrosion rate under relevant conditions).
    - A characterization of diffusion and visco-capillary two-phase flow (water and gas permeability, relative permeability curves, retention curve, gas-entry pressure, porosity, storage coefficient, diffusion coefficient of hydrogen in water and variation depending on saturation, etc.) for all materials present in the disposal system, including the host rock and the EDZ. The diffusion and visco-capillary two-phase flow properties for each material should be derived from laboratory and in situ experiments through a dialogue between modelers and experimentalists.
    - A characterization of the mechanical behavior of the materials present in the disposal system (engineered barriers and the host rock), which are design dependent, and in particular the stress range corresponding to linear elastic behavior.
- Evaluate the maximum gas pressure in the repository by numerical simulation, considering all the associated uncertainties
    - If the maximum gas pressure is lower than the maximal elastic stress of the materials important for long-term safety, considering all the associated uncertainties, the numerical evaluations can be considered representative. Visco-capillary two-phase flow numerical simulations of radionuclide transport can be carried out, both on gaseous radionuclides and on soluble radionuclides.
    - Otherwise, at least one of the materials important for long-term safety could incur mechanical damage at least during the hydraulic-gas transient phase.

The loss of the mechanical integrity of a component that has no self-sealing capacity, even if it is temporary, being unacceptable, an evolution of the repository design is necessary to reduce the maximum gas pressure if any. Such a reduction may be possible by focusing design evolutions on one or another of the following elements:

- Reduce the source term, by reducing the mass and external surface area of metal components in the repository after closure. For example, consider replacing metallic reinforcements in concrete structural elements with non-metallic alternatives.

- Modify the composition and/or the design of the repository closure system, in particular the backfill(s) and/or the seal(s), to facilitate the transport of gas. For example, by the addition of sand to a swelling clay for the mixture constituting a seal or by increasing the porosity of backfill to let gas go through.

In any case, a design approach that considers gas from the start of the design process is required. The general knowledge about gas transport mechanisms through a repository and its components, as presented in the generic storyboard (Section 4.2.1) is readily available for that purpose. However, uncertainty on the values of key properties that control gas transport can be large at the start of a geological disposal 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.

5. Understanding of gas transport in clayey materials, and its impacts

This chapter first presents the understanding of gas transport in clayey materials at the end of EURAD-GAS. As a support, the drawing of Marschall *et al.* (2005) is revisited with the perspective of gas flowing from a disposal tunnel into a clayey host rock, per transport mechanism (Figure 4 to Figure 7) (Section 5.1). Sections 5.2 and 5.3 then respectively explain the impact of gas transport on barrier integrity (at repository scale) and the fate of radionuclides.

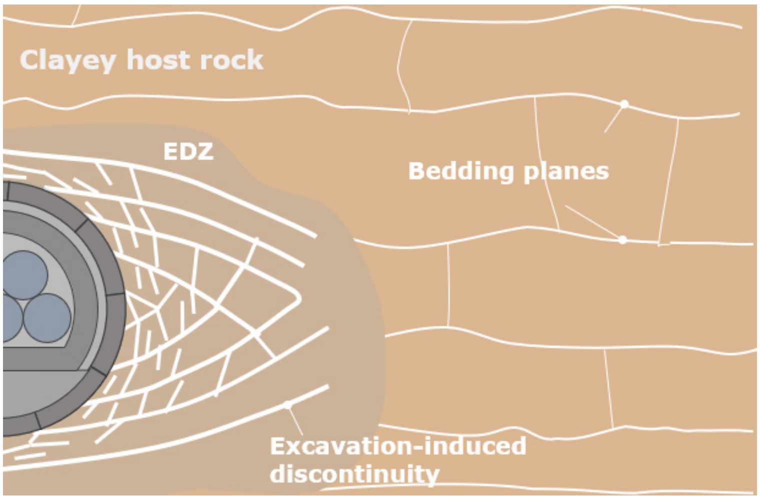

Figure 4 – Schematic representation of a gallery, excavation damaged zone (EDZ), with its discontinuities, and clayey host rock, with bedding planes. The gas flux originates from the gallery. The anisotropy of the material and/or the anisotropy of in situ stresses determine the shape of the damage pattern. 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) (Jacops and Kolditz, 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. 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.

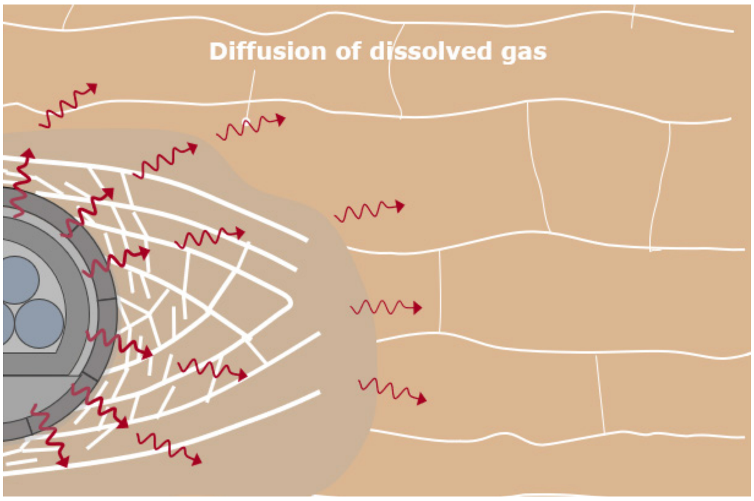

Figure 5– Diffusion is driven by concentration gradient. Depending on the host rock, the diffusion coefficient might be anisotropic.

5.1.2 Gas adsorption on clay minerals

In repository conditions, experimental evidence suggests that adsorption[8] of hydrogen on clayey materials should be neglected. In addition, neglecting adsorption is a conservative assumption as it would lead to reduced pressures.

5.1.3 Advective transport

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 6). 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 visco-capillary two-phase flow process. For what concerns geological disposal systems, a distinction is made between clayey 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[9], 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[10] of the materials that affect the

[8] 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.

[9] 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.

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

gas-entry pressure (Jacops and Kolditz, 2024) (Villar *et al.*, 2012; Sellin and Leupin, 2013). It is thus very design and material specific.

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 and Collin, 2023; Corman, 2024). 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).

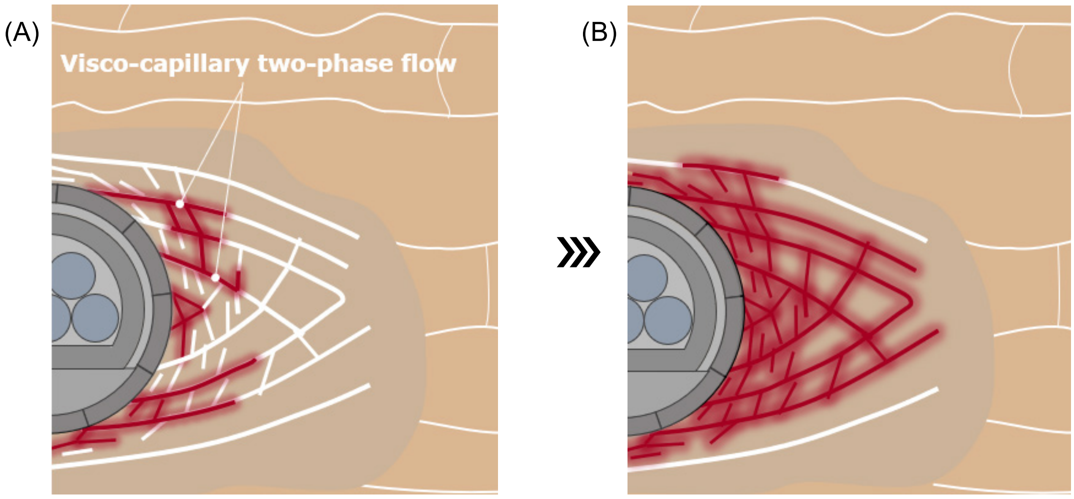

Figure 6– At higher gas pressure, visco-capillary two-phase flow occurs: gas invades the EDZ, starting with discontinuities (from (A) to (B)), in addition to diffusion (not shown). The chevron pattern shows increasing gas flux.

Flow through dilatancy-controlled gas pathways

There is a consensus that the creation of dilatancy-controlled gas pathways in clayey barrier materials is possible and controlled by the interplay between water retention, stress-strain behavior, and hydro-mechanical coupling. If pressure build-up is too high, continuous dilatancy-controlled gas flow would happen. Depending on the disposal design and/or on the host rock, dilatancy-controlled gas pathways may form in the EDZ and/or the intact host rock (Figure 7). The initiation and propagation of these pathways mainly depend on the deformation behavior of the clayey barrier and on the evolution of gas pressure build-up[11]. In the particular case of saturated engineered clayey barriers, such as bentonite with moderate emplacement density, dilatancy-controlled gas flow 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) (Marschall *et al.*, 2024).

The formation of dilatancy-controlled gas 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) (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 dilatancy-controlled gas 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).

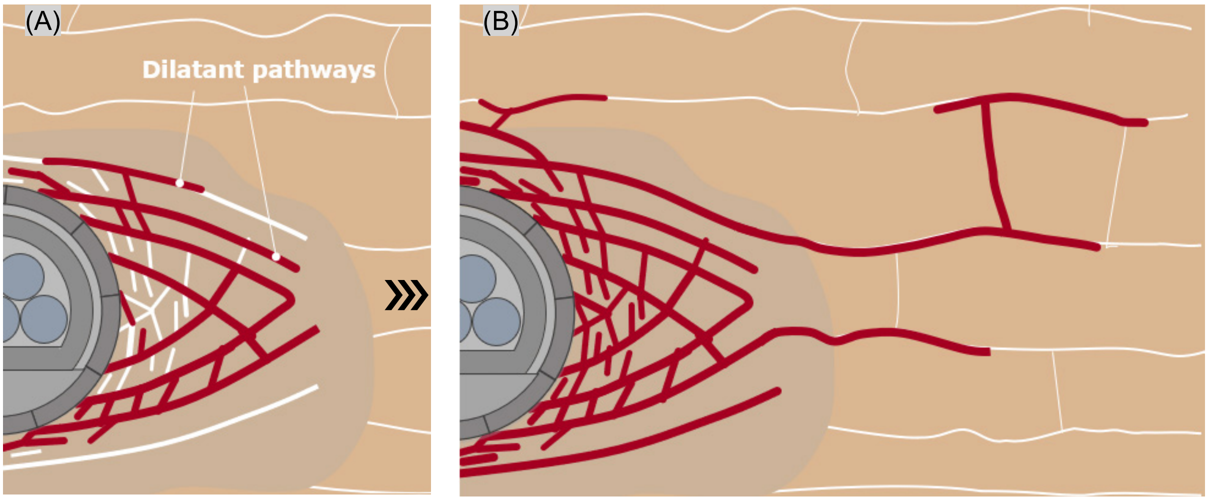

Figure 7 – (A) If the pressure continues to increase, dilatancy-controlled gas transport occurs in the EDZ, in addition to diffusion and possibly two-phase flow (not shown). (B) At even higher gas pressure, dilatancy-controlled gas pathways can even develop into intact host rock. The chevron pattern shows increasing gas flux.

5.2 Impact of gas transport on barrier integrity

Of the possible gas transport processes described above, the first mechanism, diffusion of dissolved gas is always certain to occur. It is widely accepted that this process has no impact on clayey barrier integrity.

The second mechanism, 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, by definition. Hence, no alteration of the properties of clayey barriers materials are expected for that transport mechanism either, although displacement of water and consequent desaturation of the barrier is

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

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, dilatancy-controlled gas flow, 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) (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). 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, 2023; 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 (Graham and Harrington, 2024). No evidence for the permanent accumulation of damage has been observed (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 ease this quantification in future.

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 clayey 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 *et al.*, 2017). The evidence base relating to the behavior of dilatancy-controlled gas pathways that could develop along interfaces between clayey 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[12]. The chosen strategy would be strongly design 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.

5.3 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 gaseous 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

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

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-gaseous. 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 clayey 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 (involved in EURAD-GAS) may thus neglect the effects of gas phases on dissolved radionuclide transport.

- Gaseous radionuclides that would not completely dissolve into the pore water upon release from the waste disposal packages 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, design dependent), and only radionuclides with half-lives around or longer than this duration (C-14 mainly) 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.

6. Conclusions: impacts of EURAD-GAS and perspectives

EURAD-GAS has increased confidence in the overall understanding of gas behavior in clayey materials, building on the FORGE EC project and beyond. This has in turn improved its integration into the conceptualization process for the different components of a disposal system, supporting and justifying the use of robust evaluation approaches. Overall, the discussion among all members of EURAD-GAS, including research entities, technical support organizations, and waste management organizations, has helped to strengthen the expert judgment at the end of FORGE: gas is not a showstopper for geological disposal, but rather requires effective management of uncertainties.

The impacts of EURAD-GAS are detailed in the next sections, in terms of implementation needs (Section 6.1), safety (Section 6.2), and scientific and technical knowledge (Section 6.3), followed by future perspectives (Section 6.4).

6.1 Regarding implementation needs for geological disposal

EURAD-GAS has produced documents for implementers that may inspire design measures to further reduce the gas impact, or the uncertainties associated with gas transport through geological disposal systems. Two state-of-the-art reports have been written. The first one presents some fundamentals on gas transport in clayey materials (Levasseur *et al.*, 2021) while the second focuses on key messages for end-users such as program managers (Levasseur *et al.*, 2024).

6.2 Regarding safety of geological disposal systems

EURAD-GAS has provided experimental evidence on the processes involved in gas transport throughout a disposal system and on the effects of these processes on barrier materials (Jacops and Kolditz, 2024; Marschall *et al.*, 2024). This evidence can be referred to by national programs in the arguments supporting claims about long-term safety of a geological system. EURAD-GAS had also collected elements that make it possible to identify the inherent strengths and limitations of various approaches for the treatment of gas in safety cases and to assess their suitability in different contexts, recognizing that this may depend on the disposal system that is being evaluated (host rock/design) or even the advancement of the (national) program (Wendling *et al.*, 2024). Similarities of approaches between national programs were identified and the rationale behind differences explained (Levasseur *et al.*, 2021).

6.3 Regarding increased scientific and technical knowledge

EURAD-GAS has succeeded in bridging the gap between experimentalists and modelers. Building on the lessons learned from the FORGE EC project, modelers were embedded with the experimentalists to encourage dialogue in the design of experiments and the development of shared conceptualizations of the observed behavior (Jacops and Kolditz, 2024; Marschall *et al.*, 2024). By doing so, EURAD-GAS has built confidence and extended the scientific bases on the fundamentals of gas transport in clayey materials. It has confirmed that the fundamental gas transport mechanisms that can develop in different clays are similar. Because a wide enough, but realistic, range of conditions were explored for representative clayey materials, EURAD-GAS has provided data which are of relevance for all disposal systems that include clayey barriers. Testing over a range of conditions spanning low (diffusion) to high (advection) gas generation rates, a better understanding of processes has been acquired which then has broader end-users' appeal. This understanding is described in the final technical reports (Jacops and Kolditz, 2024; Marschall *et al.*, 2024; Wendling *et al.*, 2024) and is summarized in the next subsections.

6.3.1 Diffusion and adsorption

Adsorption

- Issues on gas adsorption on clayey materials were cleared out. New gas adsorption tests have been performed under well-controlled conditions. These have led to the conclusion that the capacity of clayey materials for gas sorption is very low in the repository conditions tested during EURAD-GAS and has been overestimated in the past due to experimental artifacts.

Diffusion

- Diffusion coefficients of several dissolved gases (that can be used as proxy for hydrogen) are available for a wide range of water-saturated clayey materials (Jacops *et al.*, 2017). Various types of laboratory setups exist for determining the diffusion coefficient over a wide range of water-saturated clayey materials and under various mechanical conditions. The reproducibility of these tests is high.
- Diffusion of dissolved gas is now well understood in water-saturated clayey materials. A clear correlation between permeability and diffusion coefficient exists. Relationships between diffusion coefficient and permeability are established for different clayey saturated materials over a wide range of mechanical conditions. In all studied clays, it is observed that variations of permeability over several orders of magnitude result in variations of dissolved gas diffusion coefficients of less than one order of magnitude. In practice, it means that knowledge of permeability can give a first estimate of diffusion coefficient. However, this relationship is material dependent.
- At relatively high levels of water saturation (75-100%), laboratory tests performed within the experimental program suggest that the evolution of dissolved gas diffusivity with such high level of saturation is limited. The experimental database is, however, not as comprehensive as for water-saturated conditions. If necessary (host rock/concept dependent), it could be extended to more clayey materials and a wider range of saturation degrees.

6.3.2 Advective transport

The experimental program has improved the experimental setups previously developed (e.g., in FORGE EC project). Notable progress was done in visualization techniques (postmortem but also near-real time during experiments).

- For the materials and conditions examined within the laboratory program of the EURAD-GAS, tests evidence a dilatant behavior of the clayey materials associated to gas passage above a certain threshold. For these tests, dilatancy-controlled gas pathways tend to be multiple, taking advantage of local defects or planes of weakness within the materials. Nevertheless, these pathways do not affect permanently the water permeability which stays in the same range before and after gas breakthrough events thanks to self-sealing capacity of clayey materials. Interestingly, experiments that make it possible to measure the amount of water expulsed from the material being tested showed very little amount of water compared to the volumes of gas that passed through the material. For all samples that were initially saturated or close to saturation, high saturation degrees were preserved after testing.

- The coupling between gas transport and the mechanical behavior of the clayey material is confirmed. It is however recognized that breakthrough pressure (the pressure at which gas passage is detected at the outlet of an experimental setup) depends at least as much on the experimental initial and boundary conditions as on material properties. Hence, good understanding of – and transparency about – the specific conditions imposed by each setup is essential for correct interpretation of the experiments and use of their results (idem for postmortem interpretations).

Significant progress was made on the process-level modeling front.

- A variety of extensions of the classical visco-capillary process models, enhanced with mechanical features, have been developed and successfully tested with new experimental data sets.
- Process-level models that combine pathways activation and development mechanisms at the microscopic scale and the evaluation of field parameters and conditions at larger scale are progressively gaining more traction. These complement the more traditional approaches such as the use of coupled multiphase flow and mechanical continuum models with transport parameters that empirically depend on deformations.
- However, while most of the process-level models can generally be used to reproduce (fit) experimental results such as those obtained in EURAD-GAS and before with appropriate choice of parameters, predictive capabilities are still perceived as limited. These models are thus currently used mostly as a support to interpretation and for testing hypotheses on gas transport mechanisms.

6.3.3 Enhanced capabilities for assessment at repository scale

Next to the experimental evidence and process-level model developments gained from EURAD-GAS, attention was also given to the stepwise integration process of this scientific knowledge. This supports the conceptualization at the scale of a disposal system and can be used to justify the use of simpler evaluation approaches at that scale. For instance, if calculations performed at the scale of components of a disposal system with models that can resolve couplings between gas transport and the mechanical behavior of the barriers yield gas pressures that would not affect these barriers much mechanically, a two-phase flow approach can be used to obtain realistic orders of magnitude for gas pressures and transport rates through the system as a whole. This can be, in turn, used to evaluate the transport and release of gaseous radionuclides which are carried with the inactive gas.

More broadly, for the different concepts considered within EURAD-GAS in the studied clayey host rocks, the main findings of EURAD-GAS with respect to scientific basis and capabilities for assessment at repository scale can be summarized as follows:

- Evacuation of gas from the disposal system should take place
  - as dissolved gas or in gas form, through the EBS (backfill, seals and linings) and the EDZ, and their interfaces,
  - as dissolved gas only throughout the host rock, by diffusion.

If these conditions are respected:

- In water-saturated conditions, mobilization of water-soluble radionuclides by gas is expected to be very low because little or no water displacement is associated to the evacuation of gas.
- Gaseous radionuclides that would not completely dissolve into the pore water upon release from the waste form can be carried toward the shafts and/or ramps along with the inactive gas (generated in much larger quantities). However:
    - The duration of the transport from the gas source to the shafts and/or ramps may take several hundreds to several thousand years (order of magnitude, design dependent) and only radionuclides with half-lives around this duration or higher may present a significant concentration in the gas phase when arriving in the upper formations.
    - All along the gas pathways, part of the gaseous radionuclides will also dissolve (as inactive gas also does) into the pore water present in the surrounding materials.

6.4 Future perspectives

Based on the FORGE EC project, EURAD-GAS has made a major step forward on the mechanistic understanding of the transport of gas in clayey materials. However, further research is needed to address remaining questions.

Regarding the scientific basis, it is recommended to perform the following actions:

- Expand experimental databases (mainly gas transport properties and, when relevant, relationship between properties), by conducting experiments under conditions that are representative of repository conditions.
- Develop and share best practices in terms of sample handling and experimental protocols, in line with the efforts initiated by Nagra during EURAD-GAS with the support of CIMNE/EPFL/BGS (including the use of visualization techniques) (Marschall *et al.*, 2024).
- Strengthen dialogue between modelers and experimenters in the design of experiments and the development of shared conceptualizations of observed behavior.
- Further develop coupled microscopic models to refine small-scale process understanding.
- Develop a portfolio of models at different scales (microscopic, centimeter, component, repository) (stepwise abstraction process) and assess their applicability. Having a range of models available (each with their advantages and limitations, but always based on a physical basis) allows for the selection of the most appropriate one(s) for a particular disposal system.

Regarding end-users' needs, organizations involved in radioactive waste management should continue to exchange information. This includes:

- Sharing and further developing best practices for dealing with gas transport processes and impacts. Efforts are needed to apply the knowledge gained from small-scale experiments appropriately at the repository scale. Gas requirements evaluated through modeling for the whole disposal system need to be broken down into requirements at lower levels of design.
- Developing common strategies to ensure that gas transport requirements are compatible with all other requirements of the geological disposal system and are well integrated in the design and optimization processes.

Appendix: EURAD-GAS publications

This appendix contains a summary of most articles written by EURAD-GAS members or in collaboration with members of DONUT and PhD theses published in the frame of EURAD-GAS (as of 20 May 2024), listed per task. They are all quoted in the main text.

Articles written by EURAD-GAS members

Linked to Task 1

EURADWASTE'22 Paper – Host rocks and THMC processes in DGR EURAD GAS and HITEC: mechanistic understanding of gas and heat transport in clay-based materials for radioactive waste geological disposal (Levasseur *et al.*, 2022)

Deep geological disposal aims to contain and isolate radioactive waste from the biosphere. Repository systems are made of multiple barriers working together, typically comprising the natural geological barrier provided by the repository host rock and its surroundings and an engineered barrier system. Due to their excellent properties for the confinement of contaminants, including low permeability, high sorption capacity, and swelling/self-sealing capacity, clayey materials are considered as engineered and/or natural barriers in most repository designs under development in Europe. During the lifetime of the repository, clay barriers will be exposed to perturbations, among which those are resulting from gas and heat production within the system. It is important to verify that these perturbations will not be detrimental to the good functioning of these barriers. In this paper, it is shown how the two EURAD R&D work packages, GAS and HITEC use a combination of experimental and modelling approaches to increase the understanding and predictability of the impact on clay barriers of the fundamental processes and their couplings related to gas and heat transport respectively, providing building blocks to support the evaluation of the robustness of the repository concepts.

Linked to Task 2

Drying in nanoporous media with Kelvin effect: Capillary imbibition against evaporation by smoothed particle hydrodynamics method (Amrofel *et al.*, 2024)

Understanding drying processes in nanoporous media is of great importance in many technological and industrial situations. To better understand how gas moves through clayey rocks, of interest for underground disposal of radioactive wastes, we propose using pore-scale direct numerical simulations. In this study, we use the Smoothed Particle Hydrodynamics method, which has proved to be an effective approach for simulating complex fluid dynamics within porous media at the nanoscale. Our simulations consider capillary-dominated twophase flow with evaporation and condensation at liquid–gas interfaces, coupled to the diffusion of water vapor in the gas phase, as well as the Kelvin effect, which is a specific feature of nanopores. Our evaporation-condensation model is validated against analytical solutions. The size of the compact support of kernel function and the particle density required to obtain accurate and stable results of capillary pressure are investigated. Drying regimes, capillary-driven and evaporated-driven, are explored. A specific effort is made to highlight the influence of the Kelvin effect on desaturation and the creation of preferential paths for gas flow as well as its impact on drying rate. The role of condensation due to local vapor concentration conditions is also emphasized.

Hydro-mechanical modelling of gas transport processes in clay materials using a multi-scale approach (Corman *et al.*, 2024)

Deep geological disposal is the preferred solution in many countries to manage radioactive wastes, such as in Belgium where the Boom Clay is one of the potential candidate host formation. Over the long term, corrosion mechanisms are expected to release large amounts of gas that will rise in pressure and activate different gas transport processes in the system and the surrounding geological formation. Assessing which transfer mode prevails under which range of pressure conditions in the sound rock layers remains a major issue. This paper presents a multi-scale hydro-mechanical model capturing the influence of the microstructure features on the macroscopic gas flow, and especially the emergence of preferential gas-filled pathways. A detailed constitutive model for partially saturated clay materials is developed from experimental data to perform the modelling of a Representative Element Volume, and integrated into a multi-scale scheme using homogenisation and localisation techniques for the transitions to the macroscopic scale. Using this tool, numerical modelling of a gas injection tests in the Boom Clay are performed with the aim of improving the mechanistic understanding of gas transport processes in natural clay barriers.

An experimental methodology to assess the impact of desaturation on gas diffusion in clay based materials (Gowrishankar *et al.*, 2023)

The transport of gas in clay-based materials is dominated by diffusion. SCK CEN has studied and recorded an extensive data set of diffusion coefficients of gases in various saturated clays (Boom Clay, Opalinus Clay, Callovo-Oxfordian claystone and bentonites) used in Europe as host formations or in engineered barrier systems in the context of deep geological disposal of high and intermediate level nuclear wastes. However, partially saturated conditions may exist during the life cycle of a radioactive waste repository. The current study aims at establishing an experimental method to measure the diffusion coefficients of various gases in partially saturated clay-based materials by means of double through diffusion tests (cross diffusion of gases across the porous sample, driven by concentration gradients) and to assess the impact of desaturation on gas diffusivity. Historically, gas diffusion measurements have been performed under dissolved conditions with continuous water and dissolved gas flow around the clay sample. However, since the saturation degree of the sample has to be constant, a novel setup has been designed and optimized to operate under a pure gas phase under controlled relative humidity (RH). The experimental concept consists of cross diffusion of gas mixtures from two reservoirs at a fixed relative humidity on either side of the sample cell, namely, the upstream and downstream reservoirs. The RH is controlled using the same saturated salt solutions in both reservoirs. During the experiment, gases will diffuse from the upstream to the downstream reservoir. By measuring the change in gas composition in the downstream reservoir with a gas analyzer equipped with a mass spectrometer (which expresses the gas composition in terms of partial pressure), the gas flux and hence the diffusion coefficient is estimated by applying steady state Fick's first law. The experimental methodology has been validated by performing an experiment on a sandy-clay sample of known petrophysical properties, at full saturation.

Analysis of key thermal coupled factors in modelling of bentonite barriers (Gupta *et al.*, 2021)

Bentonite is a material considered to be used as a component of a barrier in deep geological repositories for nuclear waste. Its behaviour is affected by temperature, humidity and chemical composition of water saturating its pores. Reproduction of bentonite behaviour in such thermo-hydro-mechanical (THM) conditions involves extensive use of empirical and physical coupled relationships. This paper investigates parameters, which influence the bentonite behaviour in THM experiments relevant to the conditions in the repositories. For the study, a numerical investigation is performed based on test simulation computed with the finite element code Thebes (Abed and Sołowski 2017). A numerical simulation by Abed and Sołowski (2017) of a non-isothermal infiltration experiment (Villar and Gomez-

Espina 2009) has been taken as a basis for the investigation. The results of this simulation were compared with a series of 7 other simulations that are set up by inactivating the selected thermally coupled variables, one at a time. Presented results identify the key parameters the simulation is sensitive to and provide insights on the relevance of the underlying coupled processes.

Finite element modelling of multi-gas flow in expansive clay (Gupta, Jacops *et al.*, 2023)

In a nuclear waste repository, the interaction of groundwater with barrier materials such as canisters, steel liners or waste packages can generate multiple gases. The accumulation of such gases can adversely affect the integrity of an Engineered Barrier System (EBS) and host rock. Therefore, efficient repository modelling requires having a mechanistic understanding and predictive capabilities of these multi-gas interactions and transport processes. This study presents a numerical framework to model gas mixture flow for two incondensable and inert-type gases in expansive clays under hydraulic conditions. The framework is an extension of the finite element code Thebes, incorporating new functions for water retention and gaseous flow behaviour. The current work is limited to only gas flows in soil. For model verification, the study utilises two simplified benchmark tests and one experiment. Each test focuses on specific flow mechanisms i.e., advection, gas phase diffusion or dissolved diffusion. The benchmark test employs COMSOL and/or semi-analytical approaches for verification. The experimental replication uses the diffusion experiment of dissolved gas in Boom clay at constant volume (CH_4 and He), performed by SCK CEN. The presented results show a good match against the experimental data while giving critical insight into gas flow mechanisms in expansive clays.

Effect of interfaces on gas breakthrough pressure in compacted bentonite used as engineered barrier for radioactive waste disposal (Gutiérrez-Rodrigo *et al.*, 2021)

In a deep geological nuclear waste repository gas can be generated by different processes. Understanding the gas transport mechanisms across the engineered and natural barriers in a repository is relevant for its security assessment, both in terms of mechanical stability and of radionuclide transport. The engineered barrier may be composed of compacted blocks of bentonite and the interfaces between these blocks might evolve into preferential fluid pathways, in particular for the gas generated around the waste canisters. Small-scale laboratory tests were performed in sound samples and in samples crossed by an interface to determine gas breakthrough pressure values after saturation and the effect on them of the interface. The FEBEX bentonite, a Spanish bentonite composed mainly of montmorillonite, was used in the tests. The gas breakthrough pressure of the saturated compacted samples increased with dry density and was higher than the swelling pressure of the bentonite. Gas breakthrough could take place either in an instantaneous or in a gradual way, the difference between both modes being the flow rate, much higher in the first case. The gas transport mechanism would be microscopic pathway dilation, with microfracturing in the case of the instantaneous episodes. A sealed interface along the bentonite did not seem to affect the breakthrough pressure or gas permeability values, since the behaviour patterns were similar in both kinds of samples, depending mostly on the bentonite dry density.

Gas transport in Boom Clay: the role of the HADES URL in process understanding (Jacops *et al.*, 2023)

Since the 1990s, SCK CEN, EIG EURIDICE and ONDRAF/NIRAS have been investigating the impact of gas generation on the Boom Clay and the engineered barriers. Several experiments have been performed to study gas transport in Boom Clay at laboratory scale and in the HADES URL. This paper gives an overview of these experiments. The transition from the laboratory to the in-situ scale is still a challenging task. It is our ambition to address these issues for Boom Clay, starting with the diffusive transport of dissolved gas. A large set of gas diffusion coefficients in Boom Clay from small-scale lab *experiments (centimetre scale) is already available, and in order to validate these for use on a larger (metre) scale, an in-situ diffusion experiment with dissolved gas will be performed in the HADES URL, using the existing boreholes. In this new experiment, called NEMESIS, dissolved neon gas will be injected in one filter, and its diffusion will be monitored by three other filters. By re-using existing boreholes dating from the 1990s, the NEMESIS experiment will continue to provide new diffusion data for the next five years.*

Mobility of dissolved gases in smectites under saturated conditions: effects of pore size, gas types, temperature, and surface interaction (Owusu *et al.*, 2022)

In a nuclear waste repository, the corrosion of metals and the degradation of the organic material in the waste matrix can generate significant amounts of gases. These gases should be able to migrate through the multibarrier system to prevent a potential pressure build-up that could lead to a loss of barrier integrity. Smectite mineral particles form a tortuous pore network consisting of larger interparticle pores and narrow interlayer pores between the platelets of the smectite minerals. These pores are normally saturated with water, so one of the most important mechanisms for the transport of gases is diffusion. The diffusion of gases through the interparticle porosity depends on the distribution of gas molecules in the water-rich phase, their self-diffusion coefficients, and the tortuosity of the pore space. Classical molecular dynamics simulations were applied to study the mobility of gases (CO_2, H_2, CH_4, He, and Ar) in Na-montmorillonite (Na-MMT) under saturated conditions. The simulations were used to estimate the gas diffusion coefficient (D) in saturated Na-MMT as a function of nanopore size and temperature. The temperature dependence of the diffusion coefficient was expressed by the Arrhenius equation for the activation energy (Ea). The predicted D values of gases were found to be sensitive to the pore size as the D values gradually increase with increasing pore size and asymptotically converge to the gas diffusion coefficient in bulk water. This behavior is also observed in the self-diffusion coefficients of water in Na-MMT. In general, H_2 and He exhibit higher D values than Ar, CO_2, and CH_4. The predicted Ea values indicate that the confinement affects the activation energy. This effect is due to the structuring of the water molecules near the clay surface, which is more pronounced in the first two layers of water near the surface and decreases thereafter. Atomic density profiles and radial distribution functions obtained from the simulations show that the interaction of the gas with the liquid and the clay surface influences mobility. The obtained diffusion coefficient for different gases and slit pore size were parameterized with a single empirical relationship, which can be applied to macroscopic simulations of gas transport.

Diffusion and gas flow dynamics in partially saturated smectites (Owusu *et al.*, 2023)

Clays and clay rocks are considered good natural and engineered barriers for deep geological disposal of nuclear waste worldwide. Metal corrosion and organic waste degradation in underground repositories generate significant amounts of gas that should be able to migrate through the multibarrier system to avoid potential pressure buildup, which could be compromising the integrity of the barriers and host rocks. The gas is expected to accumulate in larger pores and eventually form an interconnected network. Under such conditions, the migration of gas molecules takes place both in pore water films and gas-filled macropores. Therefore, mass fluxes depend on the distribution of gas molecules between the water-rich and gas-rich phases and their mobility in both compartments.

Classical molecular dynamics (MD) simulations were employed to investigate the mobilities of He, H_2, CO_2, Ar, and CH_4 in a Na-montmorillonite mesopore as a function of the degree of saturation, as well as evaluate the hydrodynamic behavior of the pore fluid in partially saturated clays. The diffusivity of the gas molecules was determined by observing the asymptotic behavior of the mean square displacement in the gas-rich phase and at the gas−water interface. The partition coefficient and Gibbs free energy were analyzed to investigate the transfer of gas molecules between the gas-rich and water-rich phases by observing the molecular trajectories as they cross the vapor−liquid interface. The results *revealed that the diffusion coefficient in the gas phase increased with increasing gas-filled pore width and converged asymptotically toward the diffusion coefficient in the bulk state. It could be shown that the diffusion coefficient of gas molecules dissolved in the water films remained constant as long as the interacting water surface was in the bulk-liquid-like phase. This behavior changes in very thin water films. It was observed that the partitioning coefficient of gas molecules at the solid−liquid interface is nearly the same as that in the bulk-liquid-like phase. Partitioning is observed to be strongly dependent on the temperature and gas molecular weights. In the second part of the study, nonequilibrium molecular dynamics (NEMD) simulations were performed to investigate the mobility of gases in pressure-driven decoupled gas-phase dynamics (DGPD) and coupled gas and water phase dynamics (CGWPD) in a partially saturated Na-montmorillonite slit mesopore. The dynamic viscosity of the gas phase was calculated from NEMD simulations and indicated that the viscosity of the gas phase was almost the same in both methods (DGPD and CGWPD). The average slip length for gas molecules at the gas−water interface was also calculated, revealing that the slip-free boundary condition assumed in continuum models is generally invalid for microfluidics and that a slip boundary condition exists at the microscale for specific surface interactions. Finally, a Bosanquet-type equation was developed to predict the diffusion coefficient and dynamic viscosity of gas as a function of the average pore width, gas mean-free path, geometric factor, and thickness of the adsorbed water film.*

On multi-component gas migration in single-phase systems (Pitz *et al.*, 2024)

The present work deals with diffusion of gases in fully saturated porous media. We test and validate the gas transport mechanism of dissolution and diffusion, implemented in the TH2M process class in the open-source finite-element software OpenGeoSys. We discuss the importance of gas diffusion for the integrity of the multi-barrier system. Furthermore, we present a multi-component mass balance equation implementation in Python, which serves as a reference for the two-component TH2M implementation and allows for a discussion of multi-component gas diffusion in liquids. We verify and validate the numerical implementations as follows: First, we come up with a set of numerical benchmarks in which solutions obtained by the two-component TH2M and multi-component implementations are compared. Thus, we show under which conditions predictions made by the TH2M model can be used for multi-component gas systems. Finally, the work is validated using a through diffusion experiment performed at Belgium's Nuclear Research Centre SCK CEN and a sensitivity analysis is conducted based on the featured experiment. The results of this work illustrate that predictions by both the two- and four-component models match the laboratory findings very well. Therefore, we conclude that also the two-component implementation can reflect the multi-component processes well under the given constraints such as full saturation.

Modelling of gas injection tests in clay barriers, laboratory to field-scale (Toprak and Olivella, 2023)

The performance of different type of buffer or backfill materials (FEBEX and COx) in terms of gas transport capacity have been investigated by means of modelling of laboratory and/or full scale experiments. Modelling of laboratory and/or field-scale gas injection tests have been performed under different geometrical approaches that includes full 3D models and reduced 3D models. A set of sensitivity analyses (geological conditions, numerical simulation options and design options) has been proposed to provide a satisfactory reproduction of the experimental data. An initial heterogeneity has been incorporated to model geometries in a random way to simulate zones that have variable permeabilities. These modelling activities have been partly reported in EURAD-Gas Projects under Task 2 and Task 3.

The role of interfaces in the bentonite barrier of a nuclear waste repository on gas transport (Villar *et al.*, 2021)

The FEBEX in situ test provided bentonite samples that had been submitted to the conditions of the engineered barrier of a nuclear waste repository for 18 years. These samples can be considered quite evolved from the microstructural point of view (aged, matured) when compared with samples prepared in the laboratory under shorter and more usual time scales. The barrier, composed of bentonite blocks, was hydrated with granitic groundwater under natural conditions while it was submitted to the thermal gradient generated by a heater mimicking the waste canister. Some of the samples were drilled between two bentonite blocks, therefore they were crossed along by an interface. The gas permeability of samples with and without interface was tested in the laboratory under different triaxial boundary conditions. Samples with an interface drilled in the inner part of the barrier (i.e. closer to the heater and consequently drier) had higher gas permeability than samples of similar accessible void ratio (related to dry density and water content) with no interface, and it was necessary to apply higher confining pressures to reduce or suppress gas flow in them. Both observations point to the interface as a preferential pathway for gas flow in this kind of samples. In contrast, wetter samples drilled along interfaces of the external part of the barrier (which had very low accessible void ratio, because of the high saturation), had permeabilities similar to those corresponding to the same accessible void ratio in the reference, untreated bentonite. This would prove the healing of the interfaces between blocks as a result of full saturation. The importance of the testing boundary conditions, particularly with respect to confinement, on gas transport processes was also highlighted.

Linked to Task 3

Study under x-ray tomography of the impact of self-sealing process on the permeability of the Callovo-Oxfordian claystone (Agboli *et al.*, 2023)

To analyze the self-sealing process in the Callovo-Oxfordian claystone, self-sealing tests were performed on initially fractured samples under different temperatures with water and gas injection. Cylindrical samples oriented in parallel and perpendicularly to the bedding plane with an artificial initial fracture were used in a triaxial compression cell transparent to X-rays. Water and gas permeability were measured and the evolution of cracks volume was analyzed from X-ray tomography 3D images to characterize the self-sealing process. All tests performed at 20°C with water injection showed a rapid drop in permeability at the beginning followed by a progressive decrease and a stabilization after one month. The permeability of fractured samples decreases significantly after self-sealing but is still higher (by 2 orders of magnitude) than the permeability of healthy claystone. Otherwise, the less calcite the sample contains (i.e., the more clayey it is), the faster the crack self-seals. The smaller the opening of the initial crack is, the faster the water permeability decreases and the crack closes. No significant influence of the sample orientation on the self-sealing kinetic was identified at this stage. It seems that high temperature has a slight retarding effect on the self-sealing process. For the test with water and gas injection, the injection of gas delays the decrease of water permeability and the self-sealing process, which is probably due to the crack desaturation induced by gas injection.

Numerical investigation of the couplings between strain localisation processes and gas migrations in clay materials (Corman *et al.*, 2022)

Deep geological repository is the preferred solution in many countries to manage radioactive wastes, such as in France where the Callovo-Oxfordian (COx) claystone is the candidate host rock. In such clay rock formation, the drilling of storage gallery creates an Excavation Damaged Zone (EDZ) with altered flow properties in the short term, while corrosion processes release large amounts of gas in the long term. Assessing the evolution of gas pressures in the near-field and predicting the effect of the EDZ on *gas transport remains a major issue. This paper presents a second gradient two-phase flow hydro-mechanical (H2M) model tackling the multi-physics couplings related to gas transfers and fractures development. The EDZ is reproduced by shear strain localisation bands using a microstructure enriched model with a second gradient approach. The gas migration is captured by a biphasic fluid transfer model. The impact of fracturing on the flow properties is addressed by relating the permeability and the water retention curve to mechanical strains. Using this tool, numerical modelling of a drift in the COx claystone is performed with the aim of emphasising the influence of the HM couplings on gas migrations at nuclear waste disposal scale.*

On the use of a mass lumping technique compatible with a fully-coupled hydromechanical model for the case of strong discontinuities with the extended finite element method (Faivre *et al.*, 2023)

In the present article, we introduce a new approach that uses a standard lumping technique, also known as the row-sum technique, in the extended finite element method (XFEM) in order to stabilize a fully-coupled hydromechanical model in the presence of a strong discontinuity. Typically, numerical oscillations can be observed where high gradients appear (pressure shocks) due to the violation of the maximum principle. In order to remediate this abnormal behavior, it is of common use to lump the mass matrix associated with the discretized form of the diffusion equation, a parabolic partial differential equation. Several techniques are available to diagonalize the mass matrix in XFEM in the context of dynamic crack growth. However, these techniques were designed for a wave-like partial differential equation, a hyperbolic partial differential equation. The direct use of such techniques with a fully-coupled hydromechanical model does not conserve the mass and leads to the violation of the maximum principle. The approach proposed in this work fulfills the features of a parabolic partial differential equation. It is also adapted to the nonlinear case for which the mass density is given by a constitutive law with a distribution that depends on the position inside the domain but which can be different on each side of the discontinuity. In order to test the robustness of our XFEM formulation, only discontinuous enrichments are considered in this article and the discontinuity is supposed impermeable in order to recover two distinct materials on each side of the boundary-controlled interface for which we can exhibit independent analytical solutions

A multi-scale insight into gas transport in a deep Cenozoic clay (Gonzalez-Blanco and Romero, 2022)

The migration of gases is crucial to ensure the long-term feasibility of argillaceous formations for the deep disposal of radioactive waste. This paper presents an experimental investigation with a multi-scale perspective on the response to gas transport of initially saturated Boom Clay (Belgium). Gas injection tests have been performed under oedometer conditions at different controlled-volume rates, constant total vertical stress and different sample orientations (flow orthogonal or parallel to bedding planes). The results confirm soil expansion and consequent degradation during injection that has a significant impact on the aperture of localised gas pathways (fissures) and increases intrinsic permeability during the gas pressure dissipation stage. The analyses with complementary techniques (mercury intrusion porosimetry, field-emission scanning electron microscopy and X-ray micro-tomography) confirm the opening of fissures with different apertures and separations at the microstructural scale. Large-aperture fissures develop along the weaker bedding planes. These techniques allow the volume of fissures to be quantified, which does not significantly depend on gas flow direction, as also measured in the isotropic response of the gas effective permeability. A scalar damage variable derived from the fissured fraction has been used to assess the gas-entry pressure reduction and the intrinsic permeability increase after the gas tests in both directions.

Hydro-mechanical response to gas transfer of deep argillaceous host rocks for radioactive waste disposal (Gonzalez-Blanco *et al.*, 2022)

During recent decades, argillaceous sedimentary formations have been studied as potential host formations for the geological disposal of long-living and heat-emitting radioactive waste—Boom Clay in Belgium and Opalinus Clay and Brown Dogger in Switzerland. A significant issue in the long-term performance of these potential host rocks concerns the generation and transport of gases. The pressure resulting from the generation of gas in an almost impermeable geological medium in the near field of a repository will increase. Under high gas pressures, the mechanical and hydraulic properties of the host rock are expected to change significantly. Preferential gas pathways may develop which exploit material heterogeneity, anisotropy (bedding planes), rock discontinuities, or interfaces between the different components of the repository, and may eventually lead to the release of the produced gases. Gas flow through these clayey rocks is investigated on the basis of laboratory work. Priority has been given to studying the volume change response of these initially water-saturated materials through relatively fast and controlled volume-rate gas injections. The effect of the gas injection rate, the confining pressure and the bedding orientation on the gas transport properties have been studied with particular attention paid to the coupling with strain behaviour. The results have shown features common to the three formations concerning the gas transfer process through preferential pathways, despite their initially differential properties.

Self-sealing of Boom Clay after gas transport (Gonzalez-Blanco *et al.*, 2023)

In the geological disposal of high-level radioactive waste in argillaceous rocks, studying the barrier integrity after gas transport and the pathway closure thanks to self-sealing capacity is a crucial aspect for the safety assessment. This paper presents experimental research in Boom Clay (a potential host rock in Belgium) to evaluate the effectiveness of self-sealing and possible fissure reactivation during a second gas invasion event. Initial water permeability under oedometer conditions was first measured on samples at two bedding orientations, being higher the sample with bedding planes parallel to flow, highlighting marked anisotropy. Then, gas injection tests at a constant volume rate were performed. Results indicated that Boom Clay underwent expansion and degradation during gas injection due to the development of fissures that were quantified using microstructural techniques. The computed effective gas permeability was not significantly dependent on bedding orientation and was slightly larger than the initial intrinsic water permeability. The resaturation of the samples led to a recovery of the initial water permeability for both orientations, replicating the original anisotropy. The microstructural analyses confirmed the gas pathways' closure, indicating good self-sealing and the regaining of the hydraulic barrier function. However, a small volume of large unconnected pores was detected on undrained unloading before the microstructural study. An additional gas injection after the self-sealing resulted in a higher effective gas permeability and a larger increase in pore volume, suggesting the reopening of fissures generated during the first injection. Finally, the experimental data were compiled within a multi-scale phenomenological model to relate the microstructural information to macroscopic flow transport properties capturing the intrinsic permeability increase on gas invasion and its recovery during self-sealing.

Stress field disruption allows gas-driven microdeformation in bentonite to be quantified (Graham and Harrington, 2024)

Geological disposal of radioactive waste is being planned by many countries. Bentonite clay is often included in facility design, providing a barrier to radionuclide migration. Gas, generated by the waste or corrosion of waste canisters, may disrupt the properties of the bentonite. Robust prediction of this interaction is, therefore, necessary to demonstrate safe facility evolution. In some cases, gas may deform the clay, resulting in localised flow; however, the nature of this deformation has been widely debated. Accurate numerical representation of this behaviour has been limited by a shortage of *information on the degree/distribution of deformation. Using experimental data from gas injection tests in bentonite, we show that first order fluctuations in the stress field can provide this information. We show that hundreds of microdeformation events can be detected, with similar characteristics to established fracturing phenomena, including earthquakes and acoustic emissions. We also demonstrate that stress field disruption (i) is spatially localised and (ii) has characteristics consistent with gas pathway 'opening' and 'closure' as gas enters and exits the clay, respectively. This new methodology offers fundamental insight and a new opportunity to parameterise and constrain gas advection models in clays and shales, substantially improving our capacity for safe facility design.*

Identification of key thermal couplings affecting the bentonite behaviour in a deep geological nuclear waste repository (Gupta, Abed *et al.*, 2023a)

Deep geological nuclear waste repositories use the multi-layer Engineered Barrier System (EBS) to isolate nuclear waste from the environment. The key component of the barrier is densely compacted bentonite, closely resembling claystone. Therefore, to ensure safety, we need a numerical model for the bentonite and the barrier that predicts EBS behaviour during transient thermal, hydraulic, mechanical and chemical conditions. The paper identifies key mechanisms and processes affecting the bentonite in the barrier due to temperature changes (thermal couplings) based on advanced fully-coupled Finite Element Method simulations. The paper investigates 1) non-isothermal infiltration experiment on FEBEX bentonite (Villar and Gomez-Espina, 2009) and, 2) Centro de Investigaciones Energeticas Medioambientales y Tecnologicas (Ciemat) test (Martin et al., 2006), presenting 10 simulation configurations that are set up by inactivating one thermal coupling/variable at a time. The difference between these simulations and the baseline model results, examined in terms of the net mean stress (swelling pressure), suction and fluid flow, give insights into the significance of investigated coupling. Results suggest that thermal couplings related to vapour density, viscosity, water retention curve, and molecular diffusivity are among the most influential. The study additionally highlights the importance of water transport as liquid and gas, and water evaporation and condensation.

Implementation and validation of pressure-dependent gas permeability model for bentonite in FEM code Thebes (Gupta, Abed *et al.*, 2023b)

In an Engineered Barrier System of a nuclear waste repository, gas migrates through: a) diffusion/advection of dissolved gases, b) two-phase continuum flow, c) dilatant pathway flow and d) single-phase gas flow through macro-fractures in the soil. The gas production rate and the corresponding gas pressure accumulation affect the clay material behaviour and its properties such as air entry value. For the safe design of the EBS system, computational models need to account for the identified transport mechanisms. This study presents an enhancement in the finite element code Thebes that replicates the observed increase in permeability at higher gas pressures, e.g. due to pore dilatancy and gas fracture as proposed by Xu et al. The formulation links permeability to gas pressure and threshold/critical pressure. For model validation, the study utilizes a gas injection experiment carried out in IfG (Institute for Rock Mechanics, Germany) on Opalinus Clay. The results show a good fit against the measurements while giving insight into gas flow through clays.

3D pore microstructures and computer simulation: Effective permeabilities and capillary pressure during drainage in Opalinus Clay (Keller, 2021)

The 3D reconstruction of the pore space in Opalinus Clay is faced with the difficulty that high-resolution imaging methods reach their limits at the nanometer-sized pores in this material. Until now it has not been possible to image the whole pore space with pore sizes that span two orders of magnitude. Therefore, it has not been possible to predict the transport properties of this material with the help computer simulations that require 3D pore structures as input. Following the concept of self-similarity, *a digital pore microstructure was constructed from a real but incomplete pore microstructure. The constructed pore structure has the same pore size spectrum as measured in the laboratory. Computer simulations were used to predict capillary pressure curves during drainage, which also agree with laboratory data. It is predicted, that two-phase transport properties such as the evolution of effective permeability as well as capillary pressures during drainage depend both on transport directions, which should be considered for Opalinus Clay when assessing its suitability as host rock for nuclear waste. This directional dependence is controlled on the pore scale by a geometric anisotropy in the pore space.*

The hydro-mechanical behavior of Opalinus clay fractures: Combining roughness measurements with computer simulations (Keller, 2022)

The role of surface roughness of fractures in Opalinus Clay and in rocks in general is relevant in understanding the hydromechanical behavior of fractures. Two different fracture surfaces of shear fractures in the Opalinus Clay were investigated. The fracture surfaces were characterized based on their roughness power spectrum. It was found that slickensides fracture surfaces are near fractal-like up to the longest scale with a fractal dimension Df ~ 2.1 and in the absence of a roll-off region at long wavelengths. In contrast, the glassy fracture surfaces show a roll-off region, which is characteristic of a flat surface with rather small and local topographic height variations. The glassy fracture surface is near fractal like with Df ~ 2.0. The measured roughness power spectra were used to create fracture models to study the behavior of different fracture closure mechanism: 1) increasing congruence (matedness), 2) closure by compression and 3) closure by swelling. It turned out that the relationship between permeability and mean aperture depends on the fracture closure mechanism. Concerning closure by compression, the root mean square (rms) value of the aperture (aper) distribution aperrms influences the contact formation behavior, which in turn controls the hydromechanical properties. The lower aperrms is, the lower the fracture compliance. Apart from aperrms, the simulations show that in clay rocks, plastic deformation plays an important role in the closure of fractures by compression. In agreement with the experiments, the simulations predict that the permeability falls below 10% of the initial value at a compressive stress of 5 MPa. The simulations predict that fracture closure by swelling is rather ineffective for confining pressures exceeding ~1 MPa.

XCT analysis of drill cores of Opalinus clay and determination of sample size for effective properties evaluation (Keller, 2023)

In Switzerland, the Opalinus Clay unit was chosen as host rock for a repository for nuclear waste and has recently been investigated in a deep drilling campaign at possible repository construction sites. X-ray images of drill cores were compiled into virtual rock columns and were statistically analyzed with respect to layered compositional variations. This provides insight into scale-dependent homogenization and improves sampling strategy. To predict the repository behavior, using continuum-based models of Opalinus Clay, requires the knowledge of effective properties related to a minimum volume at which Opalinus Clay behaves homogeneously. It turned out that with respect to rock composition, such a volume does not exist in the sense that a single sample of manageable size provides a reliable mean composition. This is because the variation of the cm to dm thick layers, which differ in composition, does not sufficiently homogenize even at the 10-m scale. Thus, effective properties must be obtained by averaging several handleable samples. Regarding the composition of Opalinus Clay at a particular location, about 30 samples, distributed over the whole thickness, with a length of about 30 cm should be measured so that the relative error of the mean value is not higher than 5%–10%. For the statistical analyses computed tomography (CT) values of X-ray data were calibrated with respect to rock composition based on laboratory measurements. The CT values are largely controlled by the respective volume fraction of calcite, quartz, and porous clay matrix. These three components form >80 vol.% of the sedimentary rocks studied (also above and below Opalinus Clay). The relationship between CT value and component contents depends on the rock type. The use of data from different rock types to calibrate CT values with respect to composition can lead to erroneous results.

Modelling gas fracturing in saturated clay samples using triple-node zero-thickness interface elements (Liaudat *et al.*, 2023)

Geological Disposal Facilities (GDF) for radioactive waste will generally rely on clay-rich materials as a host geological formation and/or engineered barrier. Gas will be produced within the GDF, which can build up significant gas pressure and will activate the migration of gas through the clay materials via different transport mechanisms. These transport mechanisms are usually investigated in laboratory tests on small clay samples of a few centimetres. In this paper, a new Pneumo-Hydro-Mechanical (PHM) Finite Element model to simulate gas migration in saturated clay samples of this scale is presented. In the proposed modelling approach, continuum elements are used to represent the mechanical and flow processes in the bulk clay material, while zero-thickness interface elements are used to represent existing or induced discontinuities (cracks). A new triple-node PHM interface element is presented to achieve this. The performance of model is illustrated with synthetic benchmark examples which show the ability of the model to reproduce observed PHM mechanisms leading to propagation of cracks due to the gas pressure (gas fracturing).

Development and numerical implementation of a multiscale constitutive law for double-porosity swelling clayey rocks (Mhamdi Alaoui *et al.*, 2023)

The swelling of clayey rocks originates at different scales of the material. Clay mineral swelling is governed by two different phenomena: crystalline and osmotic swellings. In this paper, we adapt a multiscale numerical model that considers both crystalline swelling and interlayer osmotic swelling (four water layers and more) to efficiently implement it in a finite element method code. To this end, we itemize the different interactions arising in the interlayer space with respect to the disjoining pressure, and in the interparticular space considering the capillary effects. The overall model is based on the upscaling of the constitutive law from the nanoscopic scale to the macroscopic scale by adding all the different interactions that appear while upscaling. Additionally, the general model adapted in this paper can be considered as a new definition of the effective stress governing the chemo-hydromechanical behavior of clayey rocks. The numerical model is then implemented in Code_Aster software. The validation of our model is conducted by reproducing constant-volume swelling test. Simulation results for different pore size distributions demonstrate fair agreement between the experimental results and the numerical modeling in comparison with linear elasticity that overestimate the final swelling pressure compared to the model developed herein.

Benchmarking a new TH2M implementation in OGS-6 with regard to processes relevant for nuclear waste disposal (Pitz, Grunwald *et al.*, 2023)

In this paper, thermo-hydro-mechanically (THM) coupled processes triggered during the construction, operation and closure of a deep geological repository for heat generating, high level radioactive waste are discussed based on a generic disposal concept. For this purpose, we are using the numerical non-isothermal two-phase–two-component flow in deformable porous media (TH^{2}M) implementation (Grunwald et al. in Geomech Geophys Geo-energy Geo-resour, 2022) in the open-source software OpenGeoSys (Bilke et al. in Transport Porous Media 130(1):337–361, 2019, https:// doi. org/ 10. 1007/ s11242- 019- 01310-1). THM coupled effects covered in this work focus on single and two-phase-flow phenomena, gas and heat generation as well as poro-elastic medium deformation. A suitable set of benchmarks covering aforementioned THM-effects, devised in the scope of the BenVaSim benchmarking project (Lux et al. in Synthesis report. BenVaSim—International Benchmarking for Verification and Validation of TH2M Simulators with Special Consideration of Fluid Dynamical Processes in Radioactive Waste Repository Systems. Tech. rep., 2021, https:// doi. org/ 10. 13140/ RG.2. 2. 28998. 34887) is chosen and one additional benchmark is presented, allowing for the demonstration and comparison of the OGS-6 TH2M implementation against results obtained by other well-established codes used in the field. Apart from the code comparison, the benchmarks also serve *as means to analyze THM coupled processes in a repository based on very simplified geometries. Therefore, they can help to improve the process understanding, but any quantitative results should not be interpreted as predictions of the behaviour of a real repository. The results obtained in this work agree well with the results presented by the project partners in BenVaSim—both in single phasic, fully liquid saturated cases and in partially saturated two phase regions. Hence, the suitability of the OGS-6 TH2M implementation for the application in the field of radioactive waste management, supporting the safety case and analyzing the integrity of the geological and geotechnical barrier systems is demonstrated. Finally, a detailed discussion of observed phenomena in the benchmarks increases our understanding and confidence in the prediction of the behaviour of TH2M coupled systems in the context of deep geological radioactive waste disposal.*

Non-isothermal consolidation: A systematic evaluation of two implementations based on multiphase and Richards equations (Pitz, Kaiser et al., 2023)

In this paper, the governing equations of non-isothermal two-phase flow in unsaturated, deformable porous media are presented based on different representations of the gaseous phase along with their implementations in the open-source FEM code OpenGeoSys 6. As one implementation utilises the Richards equation to represent the unsaturated system whereas the second implementation relies on a two-component two-phase flow formulation, the impact of a free gaseous phase on model predictions is illustrated by a series of tests of increasing complexity, motivated by similar investigations in Task C of the DECOVALEX 2023 project. We demonstrate that the governing equations of both implementations collapse and become identical in fully saturated regions and that the numerical implementations remain stable in the special cases. Furthermore, the implicit assumptions made in the Richards equation are explicitly applied to the physically more complex and comprehensive TH^2M model. Thus, the impact of a constant residual gas pressure on the model predictions is investigated. Finally, the examples chosen for this work feature thermal consolidation and pore fluid pressurisation effects in the Opalinus Clay due to heat release from a nuclear waste canister as well as dry-out with subsequent resaturation of a bentonite buffer around the heat source. All test cases are evaluated on the level of the governing equations of each model as well as on the level of results obtained by each model for a discussion of conceptual model uncertainty.

Numerical modeling of self-sealing in fractured clayey materials (Quacquarelli *et al.*, 2024)

The fractures network generated during the excavation of underground research facilities can induce stress redistribution and alteration of flow and transport properties, becoming preferential paths for releasing radionuclides into the host rock. Nevertheless, in the long term, the fracture can be sealed through the resaturation of water coming from the rock as a function of its self-sealing potential. Despite the large number of experimental studies that have proven the self-sealing capacity of clay rocks, very few attempts have been made to describe and predict the phenomenon numerically. This may be due to the difficulty of measuring the initial hydro-mechanical conditions. Besides, samples artificially fractured in the laboratory can be disturbed by the preparation process itself, which can alter the hydro-mechanical state. This paper addresses that issue by bridging the gap between experiments and numerical modeling. Representative experimental tests performed on Callovo–Oxfordian Claystone (COx) are used to offer a hydro-mechanical fracture law taking into account the self-sealing capacity of the material. Implementing such a model in a finite element code allows its validation through comparison with laboratory tests. Furthermore, the role of the initial fracture size and the evolution of water permeability during the wetting/drying process is investigated. Due to its transmissivity, injected water can penetrate the rock, initially reaching the damaged zone around the fracture before spreading through the entire sample. This progression is accounted in the constitutive equation and represented numerically. Nevertheless, a larger initial crack leads to reduced recovery rates. These results match the experiments, offering a valuable perspective in the modeling of self-sealing in in situ conditions.

Derivation of heterogeneous material distributions and their sensitivity to HM-coupled two-phase flow models exemplified with the LASGIT experiment (Radeisen *et al.*, 2023)

Advective gas transport in bentonite, a possible buffer material in repositories for radioactive materials, is difficult to simulate in numerical continuum models, partly due to the complicated microstructure of bentonite. To generate reliable models of repositories nevertheless, spatially distributed heterogeneous material properties can be used to allow localization of gas flow. In this study, a pore-size-dependent stochastic approach of the gas entry pressure is derived from Mercury Intrusion Porosimetry, which is used to replicate measurements from the LASGIT experiment. In addition, three benchmark tests are simulated to investigate the dependence of heterogeneous distributions of material properties on the mesh discretization, the temporal dependence, and the coupling between the processes influenced by the heterogeneous parameters. The numerical modeling results of the LASGIT experiment show that the onset of gas flow into the system and the subsequent increase in pressure and stress can be well reproduced using heterogeneous distributions. Compared to a model with homogeneous material properties, heterogeneous distributions may allow the generation of dilatancy-controlled microfractures—an important feature with regard to the advective gas flow in bentonites. However, it can be observed that the heterogeneous distributions in LASGIT are less significant, as technical gaps or differences in material types could have a greater impact.

Microstructure and hydro-mechanical behaviour of compacted granular bentonite (Zeng *et al.*, 2023a)

In recent years, granular bentonite GB has become a reference material since it facilitates the backfilling operation due to high-density compacted granules with extended grain size distribution (maximum sizes around 10 mm). This investigation explores the initial microstructure of MX80-type GB and the hydro-mechanical HM response observed at the phenomenological scale. Samples were statically compacted at a fixed dry density (1.55 Mg/m³), encompassing dry and wet sides of the optimum. The microstructural study was performed using a combination of X-ray micro computed tomography and mercury intrusion porosimetry. The results indicate that the pore size density function of compacted GB can be simplified as a double-porosity network with micropores (intra-granular/aggregate pores) and macropores (inter-granular/aggregate pores). Compacting at the dry side increases macropores, making the sample more compressible on loading under as-compacted states despite the higher matric suction. The time evolution of the swelling pressure displays a double-peak pattern attributed to the particular microstructure of the samples. Moreover, the initial microstructure does not significantly influence the water retention properties without a marked hysteresis in the high-suction range under confined wetting/drying cycles. This work's outcomes highlight the microstructure's significance in the HM behaviour of compacted GB and provide further insight into the geotechnical properties of this material.

The importance of the microstructure on hydro-mechanical behaviour of compacted granular bentonite (Zeng *et al.*, 2023a)

Granular bentonite (GB) with extended grain size distribution, a candidate material to construct engineered barriers, has low water permeability, high swelling potential and self-sealing capacity and is desirable in portability and workability. However, few systematic data combining microstructural descriptions and hydromechanical (HM) behavioural features are available. The present work investigates the HM performance of Wyoming (MX80)-type GB samples compacted at different initial water contents and at a fixed dry density of 1 550 kg/m³. The samples' microstructure and its evolution along stress paths were studied using X-ray microcomputed tomography and mercury intrusion porosimetry. The results highlighted that the compacted GB presented local density heterogeneity and a multi-porosity network. Compaction at low initial water content increased the proportion of macropores and contributed to the enhancement of the compressibility and collapse on wetting under high stresses.

During isochoric saturation, two stages of swelling development were identified. First, the flooding of macropores and matric suction reduction was related to the initial increase in swelling pressure. The secondary swelling was due to the water absorption of clay sheets in micropores and the expansion of micropores within granules and aggregates, during which the density of pore water increased. A decrease in the compaction water content can result in a higher pore water density after isochoric saturation and a lower water permeability. Finally, the water retention behaviour of the compacted sample within drying/wetting cycles was less sensitive to the initial pore size distribution. The current outcomes underline the critical role of the microstructure on the HM behaviour of compacted GB.

Self-sealing of fractures in indurated claystones measured by water and gas flow (Zhang and Talandier, 2023)

Self-sealing of fractures in the indurated Callovo-Oxfordian (COX) and Opalinus (OPA) claystones, which are considered as host rocks for disposal of radioactive waste, was investigated on artificially fractured samples. The samples were extracted from four lithological facies relatively rich in clay mineral, carbonate and quartz, respectively. The self-sealing of fractures was measured by fracture closure, water permeability variation, gas penetration, and recovery of gas-induced pathways. Most of the fractured samples exhibited a dramatic reduction in water permeability to low levels that is close to that of intact rock, depending on their mineralogical composition, fracture intensity, confining stress, and load duration. The self-sealing capacity of the clay-rich samples is higher than that of the carbonate-rich and sandy ones. Significant effects of sample size and fracture intensity were identified. The sealed fractures become gastight for certain injection pressures. However, the measured gas breakthrough pressures are still lower than the confining stresses. The gas-induced pathways can recover when contacting water. These important findings imply that fractures in such indurated claystones can effectively recover to hinder water transport but allow gas release without compromising the rock integrity.

Linked to Task 4

Modelling of the corrosion-induced gas impact on hydraulic and radionuclide transport properties of geological repository barriers (Narkuniene *et al.*, 2023)

The geological disposal of high-level radioactive waste is the final step in the nuclear fuel cycle. It is realized via isolating the high-level radioactive waste in the geological environment with an appropriate system of engineered barriers. Radionuclides-containing materials must be isolated from the biosphere until the radioactivity contained in them has diminished to a safe level. In the case of high-level radioactive waste, it could take hundreds of thousands of years. Within such a long timescale, a number of physical and chemical processes will take part in the geological repository. For the assessment of radionuclide migration from a geological repository, it is necessary to predict the repository's behavior once placed in the host rock as well as the host-rock response to disturbances due to construction. In this study, the analysis of repository barriers (backfill, concrete, inner excavation disturbed zone (EDZ), outer EDZ, host rock) thermo–hydraulic–mechanical (THM) evolution was performed, and the scope of gas-induced desaturation was analyzed with COMSOL Multiphysics. The analysis was based on modelling of a two-phase flow of miscible fluid (water and H_2) considering important phenomena such as gas dissolution and diffusion, advective–diffusive transport in the gaseous phase, and mechanical deformations due to thermal expansion of water and porous media. The importance of proper consideration of temperature-dependent thermodynamic properties of water and THM couplings in the analysis of near-field processes was also discussed. The modelling demonstrated that such activities as 50 years' ventilation of the waste disposal tunnel in initially saturated porous media, and such processes as gas generation due to corrosion of waste package or heat load from the waste, also led *to desaturation of barriers. H_2 gas generation led to the desaturation in engineered barriers and in a part of the EDZ close to the gas generation place vanishing soon after finish of gas generation, while the host rock remained saturated during the gas generation phase (50–100,000 years). Radionuclide transport properties in porous media such as effective diffusivity are highly dependent on the water content in the barriers determined by their porosity and saturation.*

Articles conjointly written with EURAD-DONUT members

Parametric sensitivity to capillary entry pressure in two-phase water–gas flow models: Deep geologic disposal of radioactive waste (Amri *et al.*, 2022)

In a deep geological repository for the long-term containment of radioactive waste, the engineered barriers and host clay rock inhibit the migration of gases, due to their low permeability and high gas entry pressure. Some experiments in the literature have focused on the measurement of gas entry pressure ($P_{g,e}$), but there is a lack of 2-phase flow (water–gas) modeling studies that include entry pressure effects in such porous media. In the present work, the modified Van Genuchten–Mualem model (Vogel et al., 2000) is extended to two-phase flow, incorporating the capillary entry pressure parameter ($P_{c,e}$), and a new data analysis approach is developed in order to characterize the water–gas constitutive relations (saturation curve, water permeability curve, gas permeability curve). This constitutive model is then implemented in the iTOUGH2 code (Wainwright and Finsterle, 2016) in Global sensitivity and data-worth analyses in iTOUGH2: User's guide) with a change of primary variables to be described below (capillary pressure is set as primary state variable instead of gas saturation). After regression tests for verifying the change of primary variables in iTOUGH2, two problems were modeled: first, numerical flow experiments were performed in a clay soil (code-to-code benchmark tests, and comparisons focused on entry pressure effects); secondly, water–gas migration was modeled based on an in situ gas injection experiment (PGZ1) performed in the French URL (Underground Research Laboratory) of Bure. Sensitivity analyses show that gas entry pressure is an important controlling factor which should not be neglected in simulations of gas migration in clayey materials.

Modeling two-phase flow with hysteresis: Comparative study of hysteresis models and application (Amri *et al.*, 2023)

Drainage (drying) and imbibition (wetting) cycles are expected in many applications involving subsurface flows, such as underground CO storage, hydrocarbon extraction and production, and deep geological disposal of radioactive waste (particularly in the potential production of hydrogen due to corrosion and radiolysis). In the presence of imbibition/drainage cycles, the two-phase constitutive relations between water saturation, capillary pressure and relative permeability to liquid and gas, cannot be represented by simple one-to-one functions: they depend on the history of wetting/drying processes. Recent numerical and experimental studies highlight the importance of hysteresis and its effect on two-phase flows in porous media. In this study, an extensive critical review of conceptual hysteretic models is first presented. The performance of these models is evaluated and tested from a theoretical point of view based on their assumptions, and by confronting them with measured data for various porous materials extracted from the literature (cement, sands, Bentonite MX80). The advantages and disadvantages of the different hysteretic constitutive models are discussed. Finally, we implement the five hysteresis models in the iTOUGH2 code, and we develop a numerical simulation of a two-phase flow experiment with immiscible fluids at the laboratory scale (meters), to compare the flow results obtained with or without hysteresis, and also, with or without capillary entrapment. The results highlight the need to take into account hysteresis in the modeling of multiphase porous media flows, especially when they are subjected to drying–wetting cycles. It is suggested that the model of Beriozkin and

Mualem (Adv Water Resour 115:253–263, 2018) should be preferred both from a theoretical standpoint and for its computational advantages.

Gas-entry pressure impact on the evaluation of hydrogen migration at different scales of a deep geological disposal of radioactive waste (Saâdi, 2024)

Although the importance of gas-entry pressure in simulating two-phase liquid–gas flow in porous media has been studied at the column and borehole scales, its impact on the simulation of transient hydraulic-gas at different scales of a deep geological repository of radioactive waste (DGR) in low permeability clay rock during the post-closure phase has not yet been studied. The purpose of this work is to show that neglecting this phenomenon can lead to underestimation of the maximum gas pressure and water–gas fluxes simulated within the host rock and backfilled drift network. This could impact the performance of the engineered barrier system of a DGR. Simulations performed for a high-level waste disposal cell and for a simplified repository composed of hundreds of disposal cells situated in a clay host rock, show that gas preferentially migrates through the DGR components with low capillary entry pressures, such as the excavation damaged zone (Refers to the zone where fractures develop due to failure of the rock mass around galleries after tunneling) (EDZ), the engineered barriers materials (backfill, bentonite-plug…) and interfaces between the EDZ and these materials. Such a result could have significant consequences on the performance of a repository, due to the accumulation of gas in the drift network and high increase of gas pressure, which could lead to the host rock hydraulic fracturing.

PhD theses (defended during the course of the project)

Hydro- mechanical modelling of gas transport processes in clay host rocks in the context of a nuclear waste repository (Corman, 2024)

To date, the concept of a deep geological disposal is recognised as one of the most viable options for safely and durably storing high and intermediate level radioactive waste away from the biosphere. This mode of repository relies on a multi-barrier confinement system, which comprises a series of engineered and natural layers, aimed at delaying the radionuclide migration on a timescale consistent with the radioactive decay period. Due to their excellent insulation properties, clay materials are being considered in numerous repository designs not just as parts of the artificial barriers but also as a potential host formation, such as the Boom Clay in Belgium and the Callovo-Oxfordian claystone in France. Among the various multi-physics processes that could affect the long-term safety function of the geological barrier, the release of gases induced by the corrosion of the metal components within the system is a crucial issue. Given the impermeable nature of the host rock, an undesirable accumulation of gas in the system could trigger a succession of gas transport processes as a function of the gradual pressure build-up, jeopardising the long-term safety function of the repository.

In this context, the present work is dedicated to the modelling of the complex coupled hydromechanical processes governing gas flows in low-permeable clay materials, using the finite element code LAGAMINE. The aim is to gain a more comprehensive understanding of which transport regime prevails under which conditions in various zones of the geological barrier.

- *The first model is developed to reproduce the gas transport processes in the excavation damaged zone, which are assumed to be controlled by the hydraulic properties modifications caused by fracturing. This requires to simultaneously capture the multi-physics interactions related to gas transfers and the development of fractures. Particular attention is paid to the specific hydro-mechanical couplings between the transfer properties and the damage. The model is applied to simulate fieldscale gas injection experiments in the Boom Clay on the one* *hand, and a large-scale storage gallery set up in the Callovo-Oxfordian claystone on the other hand.*

- *The second model is developed to reproduce the gas transport processes in the undisturbed rock layers, which are assumed to be controlled by the rock structure at a microlevel. This requires to define a representative element volume that provides a detailed representation of the material microstructure with an explicit description of each constituent on their respective length scales. A comprehensive hydro-mechanical constitutive model can then be formulated at the microscale, which can be integrated into a multi-scale framework. This way, it is possible to capture the microstructure-induced phenomena that affect macroscopic gas flows. The model is applied to simulate laboratory-scale gas injection tests in the Boom Clay, as well as upscaled configurations.*

Through the development of these innovative numerical models, the aim of the research is to enhance the conceptualisation of the gas transport mechanisms in clay materials, to gain insight into the observed transport modes and their primary controls, and to provide a modest contribution to the mechanistic understanding of the hydro-mechanical phenomena associated with gas-induced failure.

Numerical simulations of gas transport in argillaceous rocks: A molecular dynamics and pore-scale simulation study (Owusu, 2023)

This dissertation investigates the gas transport and clay behavior within the context of deep geological disposal of nuclear waste. The repository for spent fuel and high-level waste can generate substantial amounts of gas through processes such as anaerobic corrosion of carbon steel, radiolysis of water, and radioactive decay in the waste. Likewise, gas production can occur in low and intermediate-level waste repositories due to chemical degradation of organic waste materials and corrosion of metals. If these gases cannot sufficiently escape from the vicinity of the repository, a localized build-up of gas pressure could compromise the integrity of the barriers and the safety design of the repository. Therefore, a thorough understanding of gas transport mechanisms and processes is crucial for assessing the repository's performance. Diffusion is the primary mechanism governing solute and fluid transport in these clays due to their low permeability. While experiments can provide valuable transport parameters for designing the barrier materials, they may not fully capture the long-term evolution of transport processes and specific subsurface conditions. Consequently, numerical and computer simulations become indispensable for determining the transport mechanisms and exploring the behavior of the system beyond the limits of experimental detection. These simulations offer the opportunity to explain experimental results, probe scales, and processes that are below the detection limit of experiments, and enhance our understanding of the transport mechanisms involved. Gas diffusion simulation in fully saturated Na-montmorillonite (Na-MMT) was performed and the effects of pore size, gas species, and temperature were investigated. Classical molecular dynamics simulations were utilized to study the diffusion coefficients of various gases (CO_2, H_2, CH_4, He, Ar). The findings indicate that the diffusion coefficients are influenced by the pore size, with H_2 and He demonstrating higher mobility compared to Ar, CO_2, and CH_4. The behavior of gases is affected by the confinement and the structuring of water molecules near the clay surface, as evidenced by density profiles and radial distribution functions. The obtained diffusion coefficients for different gases and slit pore sizes were parameterized using a single empirical relationship, enabling their application in macroscopic simulations of gas transport. Considering the long-term desaturation and resaturation process, the study extends to simulate gas diffusion in partially saturated Na-MMT and investigates the partitioning of gas molecules between the gas-rich and water-rich phases. Classical molecular dynamics simulations were employed to explore the impact of gas-filled pore widths, temperature, gas mean free path, gas viii size, and gas molecular weights on diffusion coefficients and partitioning coefficients. The results demonstrate that the diffusion coefficient in the gas phase increases with larger gas-filled pore widths and eventually converges asymptotically towards the diffusion coefficient in the bulk state. Partitioning coefficients were found to be strongly dependent on temperature and gas molecular weights. Furthermore, non-equilibrium *molecular dynamics simulations were conducted to investigate the mobility of gases in a pressure-driven flow within a partially saturated Na-MMT mesopore. The results reveal the presence of slip boundary conditions at the microscale, which challenges the assumptions made in continuum models. To predict the diffusion coefficient and dynamic viscosity of the gas, a Bosanquet-type equation was developed as a function of the average pore width, gas mean free path, geometric factor, and thickness of the adsorbed water film. Na-montmorillonite, being a swelling clay, undergoes changes in its swelling behavior when exposed to different chemical species like gas due to variations in chemical potential. These alterations can subsequently impact the hydraulic properties and transport mechanism of the clay. Consequently, we investigated the influence of gas presence on the swelling pressure of Na-MMT. To achieve this, classical molecular dynamics simulations were employed as a methodology to examine the effect of gas on swelling pressure. The findings indicate that gas molecules cause an increase in the swelling pressure of Na-montmorillonite, with an approximate rise of 3 MPa. The specific behavior observed is influenced by factors such as the dry density and the characteristics of the gas species. Additionally, the analysis includes a com prehensive exploration of structural transformations occurring within the clay interlayer, pro viding insights into the discrepancies observed between experimental and simulated curves, particularly at high levels of compaction. The thesis delves into pore-scale modeling to determine diffusion coefficients of water in compacted porous smectite clay structures. This exploration is motivated by the limitations inherent in conventional approaches used to obtain transport parameters, which tend to oversimplify the intricate porous nature of clay media by treating them as a continuum. This oversimplification neglects the behaviors occurring at smaller scales. To overcome this limitation, the thesis employs various techniques such as random walk simulations, lattice Boltzmann modeling, and large-scale molecular dynamics simulations to investigate transport mechanisms. These advanced modeling techniques take into account local diffusivities within the representative elementary volume, allowing for a more accurate understanding of transport phenomena. By considering local diffusivities, particularly near chemically reactive clay surfaces, this approach sheds light on the significance of accurately comprehending transport phenomena in porous materials. By overcoming the limitations of conventional approaches, the thesis provides valuable insights into the diffusion coefficients of water within compacted porous smectite clay structures. This thesis offers a comprehensive exploration of gas transport and clay behavior, focusing on their relevance to deep geological disposal of nuclear waste and energy storage. By establishing connections between simulations conducted under fully saturated and partially saturated conditions, examining the influence of gases on swelling pressure, and incorporating pore-scale ix modeling, this research provides valuable insights into diffusion, swelling, and pore-scale processes. These findings contribute to the development of effective barrier materials and enhance our understanding of waste management strategies in complex geological environments. The knowledge gained from this study has practical implications for improving the safety and efficiency of deep geological disposal systems and advancing energy storage technologies.*

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