EURAD-GAS: OVERVIEW OF KNOWLEDGE GAINED ON GAS TRANSPORT IN CLAYEY MATERIALS AND ADDED-VALUES FOR THE BELGIAN PROGRAM

October 25, 2024 • S. Levasseur (WP Leader, ONDRAF/NIRAS), E. Jacops (Task 2 leader, SCK CEN) and colleagues from ONDRAF/NIRAS, SCK CEN, UKRI-BGS, CIMNE, ULiège, TU Delft, BGR, PSI, LEI, EDF, Aalto University, COVRA, IRSN, Nuclear Waste Services, Andra and NAGRA who contributed to provide relevant input for the Belgian programme

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MECHANICAL UNDERSTANDING OF GAS TRANSPORT IN CLAYS (EURAD-GAS) – CONTEXT

1. Considerable amounts of gas can be produced in a Geological Disposal Facility (GDF)

- Mostly hydrogen, produced through anaerobic corrosion of metallic barriers & waste components
- 2nd potentially important source is degradation of organic wastes (again, mostly hydrogen)

→ Significant total gas volume even though its production will be spread over a long period of time

2. Clays as potential host rock and material for engineered barrier components

- Owing to their excellent properties for the confinement of contaminants
- Also, a barrier for gas: low solubility of hydrogen, low diffusion coefficient for dissolved gas, high gas entry pressure, low permeability

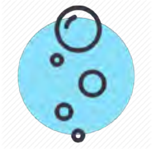
→ not easy for dissolved gas to diffuse away, not easy for free gas to displace pore water

Combining 1. and 2. above, even though gas production is slow, it is possible in some cases that it would be produced faster that it could be removed without significant gas pressures

- What consequences?

EURAD-GAS – "RAISON D'ETRE"

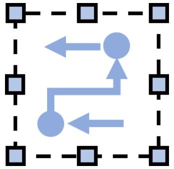
Address key end-users' questions



- **How is gas transported?**
  - Possible transport mechanisms throughout repository system, with focus on clay(ey) components
  - How much can gas displace/carry away soluble and volatile radionuclides?

- **Effects of gas pressure on barrier integrity?**
  - Gas-induced (mechanical) damage?
  - Any lasting effect on performance?
  - (Input for design options)

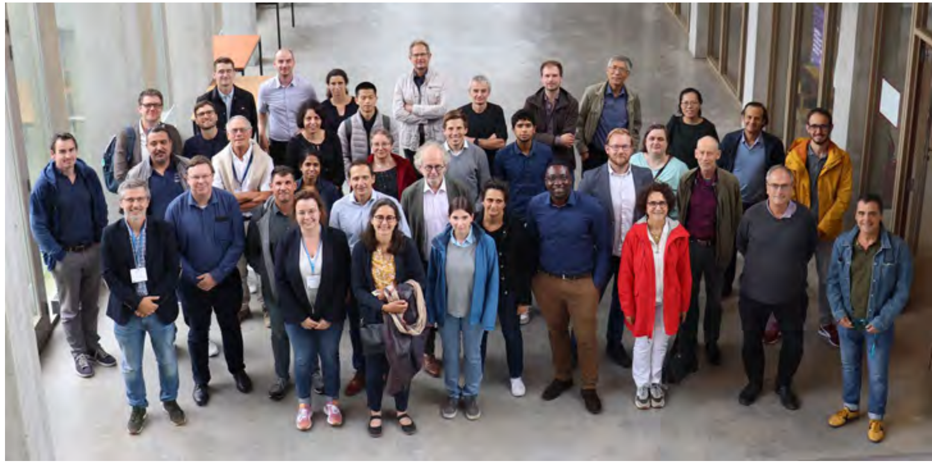
Support the safety case for gas aspects



- **Show how gas impact can be bound**
  - Simple/robust conceptualizations built on properly integrated scientific bases: storyboards
  - Comfort expert judgement from FORGE: gas is not a showstopper, but a question of managing uncertainties

- **Develop a shared vision on gas management in *varietate concordia***
  - Clear statements on scientific consensus
  - Recognizing differences between national programmes as contexts can be different

GAS WP – ORGANISATION



Process-level modelling embedded with tasks 2 & 3!

T1 Coordination

science

guidance & review

review & guidance

end-users

T2 Transport mechanisms

2.1 Diffusion & retardation
2.2 Advection (liquid phase displacement vs dilatant gas flow)

T3 Barrier integrity

3.1 Gas-induced impacts
3.2 **Pathway closure & sealing**
3.3 Model-based interpretation

Joint HITEC-GAS subtask

T4 Repository performance aspects

4.1 Conceptualisation of gas migration at the scale of the repository
4.2 Model-assisted assessment of gas-induced effects

A number of end-users are embedded in T4

+ Liaise with KM SoC, guidance & training

EURAD-GAS – OVERVIEW OF KNOWLEDGE GAINED

Mechanistic understanding of gas transport in clay(ey) materials

- **What did we learn** from an experimental & modelling programme on gas transport?
- **What do we expect** about the impact of gas passage on clay materials and barrier integrity?

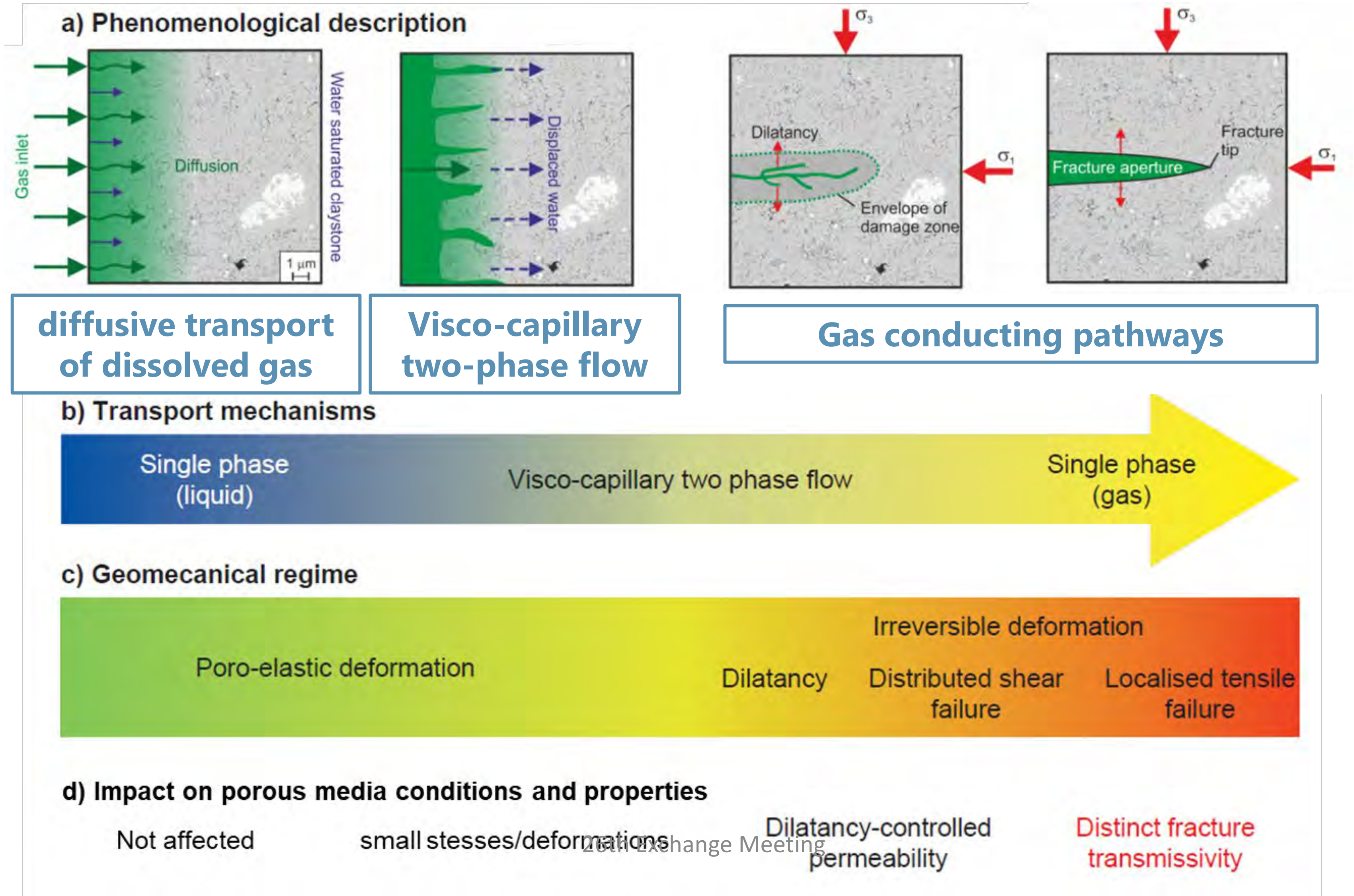
→ **focus on the science**

Integration in conceptualizations of the functioning of a repository system

- **Expert judgment and Storyboards** for typical repository configurations
- **Testing ≠ approaches** to represent gas effects at the component & system level

→ **focus on the applications and impacts for end-users**

GAS TRANSPORT MODES IN CLAYS



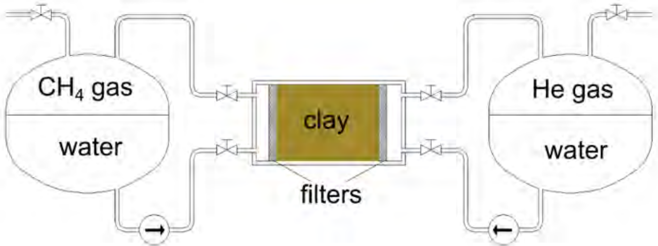
(adapted from Marschall et al. 2005 and Cuss et al. 2014)

DIFFUSION OF DISSOLVED GAS IN SATURATED CONDITIONS

- **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**

  ➔ Repeatability of such tests is good, large database

  ➔ Gas appears to **preferentially diffuse parallel to bedding** with nearly 60% of the diffusional capacity of the Boom Clay moving within the bedding planes

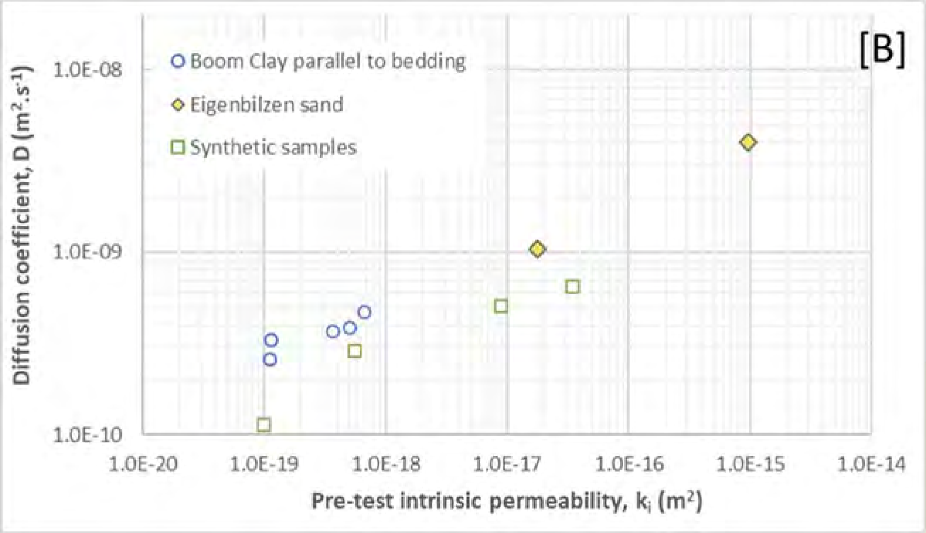


Dissolved gas diffusivity exp. In constant volume cell (SCK CEN)

- **A clear correlation between permeability and diffusion coefficient exists**

  ➔ **Variations of permeability over 3 orders of magnitude result on variations of dissolved gas diffusion coefficient of less than one order of magnitude**

  ➔ **Material-dependent.** But quantitative relation to mineralogical composition & compaction remains elusive

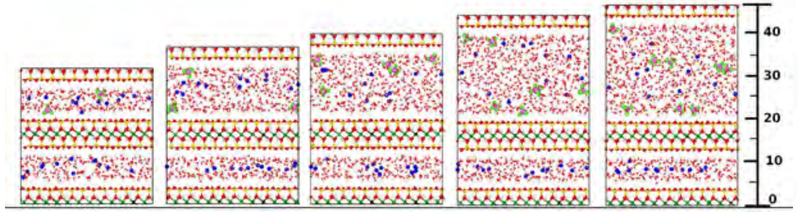


Compilation by BGS

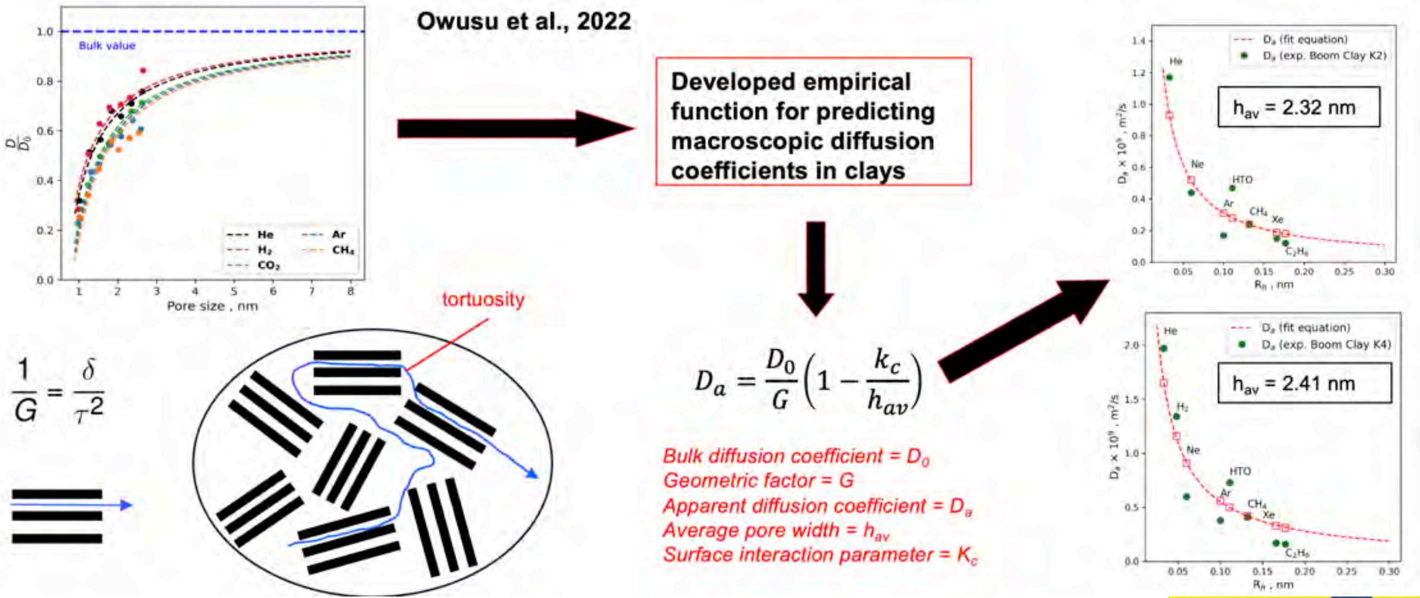
DIFFUSION OF DISSOLVED GAS IN SATURATED CONDITIONS

- **Modelling of diffusion processes at various scales**
  - **Macroscopic scale** modelling by BGR, Aalto University and LEI to consider the effect of various gases simultaneously
  - **Molecular scale modelling** by PSI to understand in detail the mechanisms associated to diffusion processes through the analysis of the molecular diffusion trajectories of various gases

→ allow to capture the results of double through diffusion experiment performed by SCK CEN and well estimate macroscopic diffusion coefficient in Boom Clay

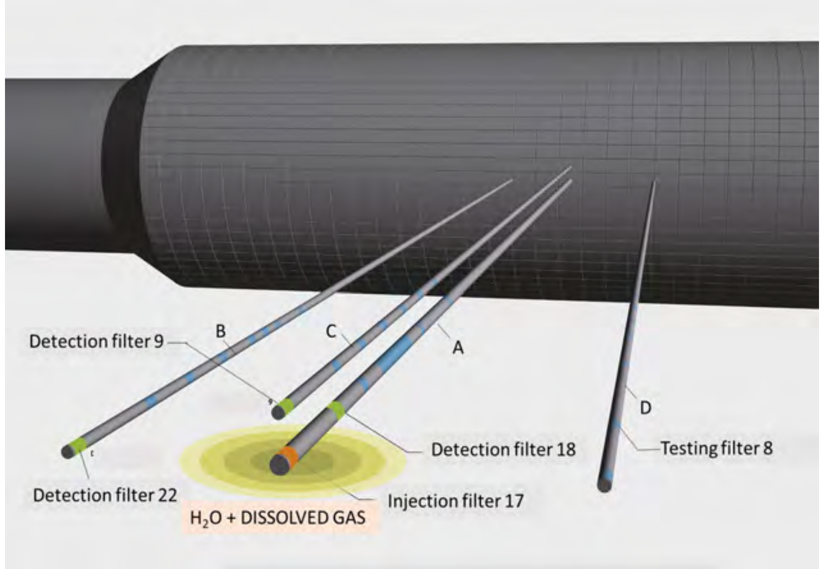


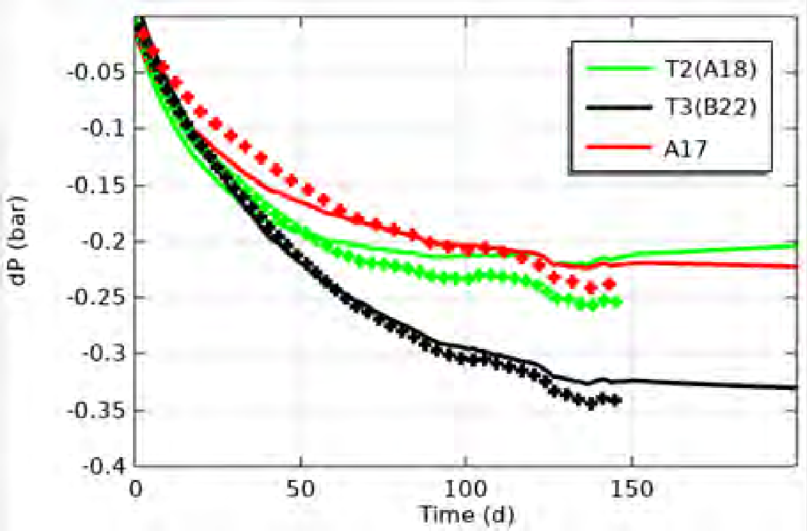
Molecular dynamics simulation by PSI of the SCK CEN dissolved gas diffusivity experiment



DIFFUSION OF DISSOLVED GAS IN SATURATED CONDITIONS – UPSCALING

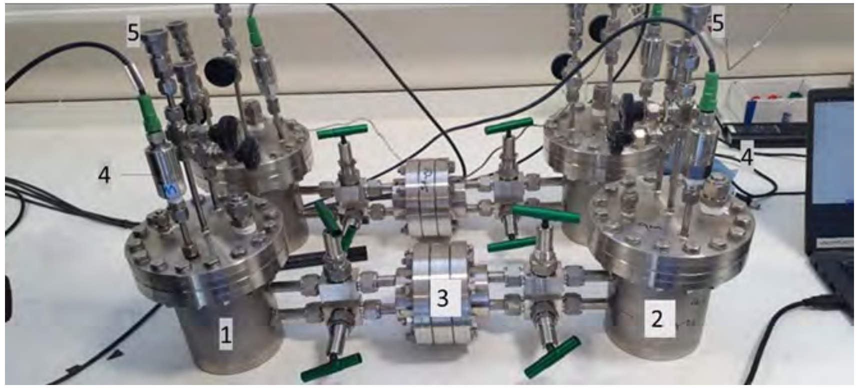
- **NEMESIS Experiment: Neon diffusion in MEgaS In Situ experiment** (in situ through-diffusion exp. In HADES URL)
  - First results from Helium in-diffusion experiment → **very promising results, confirming at large scale our knowledge on diffusion coefficient**
  - Still has to be validated by a through-diffusion test that should be more accurate over time and able to capture anisotropy





DIFFUSION OF DISSOLVED GAS IN UNSATURATED CONDITIONS

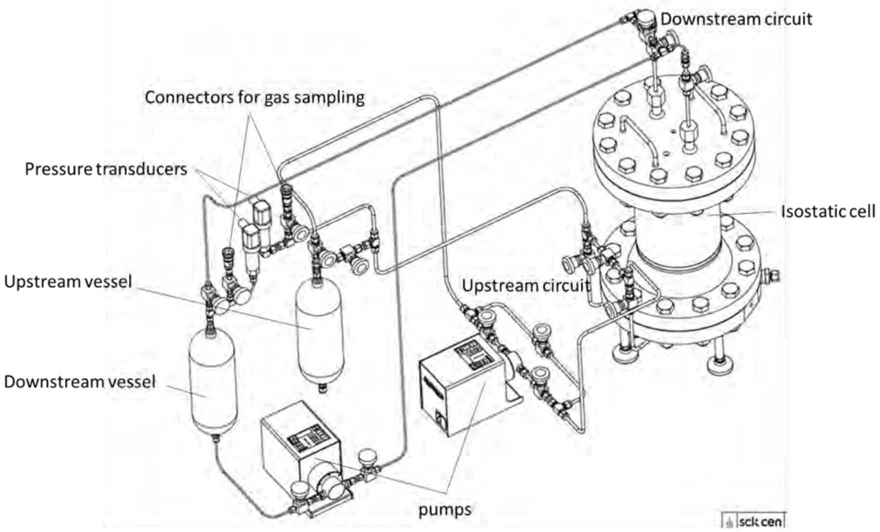
- **At relatively high levels of water saturation, between 75-100%, effect of the presence of gas-filled pores appears limited**
  - Connectivity of gas-filled pore probably limited; net gas transport remains controlled by diffusion in dissolved form
  - The experimental database is however not as comprehensive as for water saturated conditions and limited so far to synthetic clays

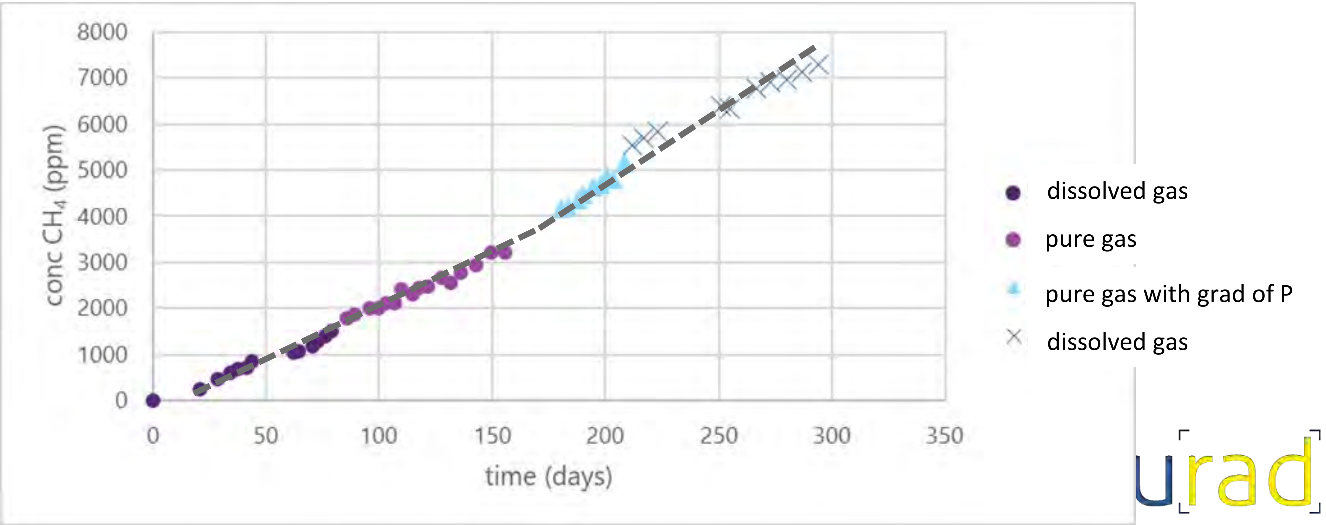


Double-through diffusion experiment setup
for unsaturated clay (SCK CEN)

LIMITS OF DIFFUSIVE TRANSPORT OF DISSOLVED GAS AND TRANSITION TO FREE GAS FLOW

- **The transition between diffusive transport of dissolved gas and free gas flow is known to be gas pressure dependent. However, the passage from one transport mode to another is not so simple in clays ...**
- **Gas injection test under controlled loading (in isostatic cell)**
  - **1st results are comparable with diffusion coefficients estimated from constant volume cell experiments**
  - Tests performed with pure gaseous gas (without any liquid) or dissolved gas in pore water provide same results

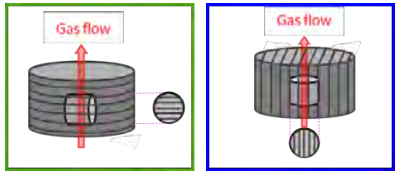


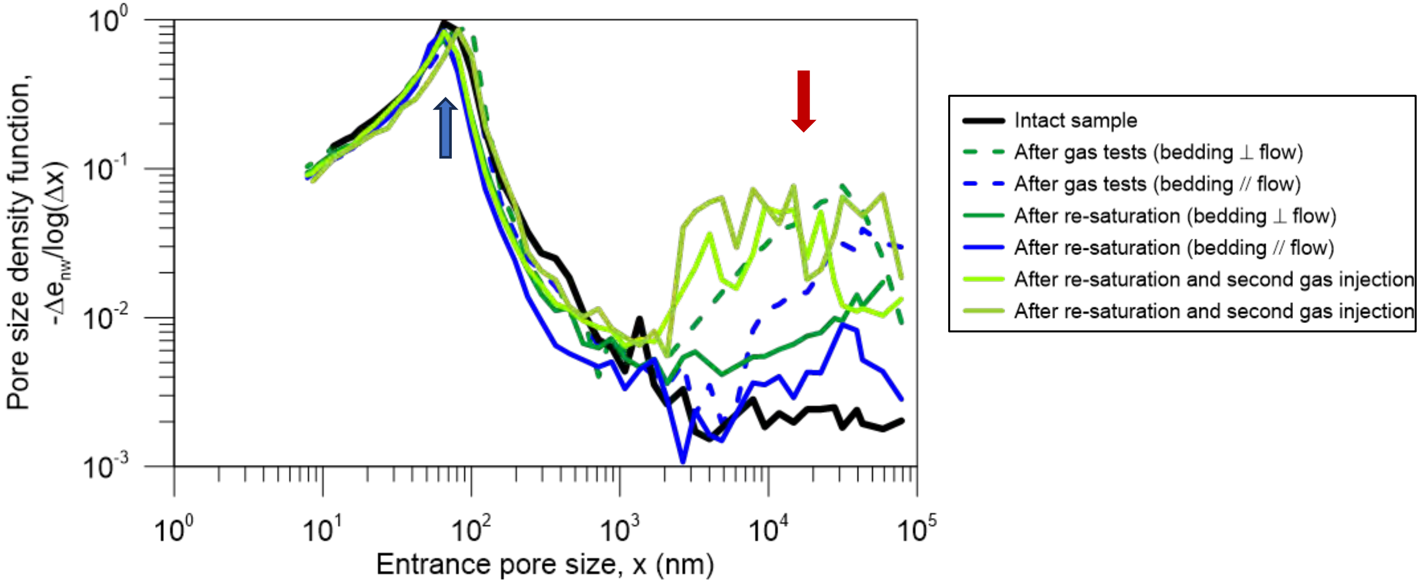


GAS PATHWAYS AND SELF-SEALING IN BOOM CLAY

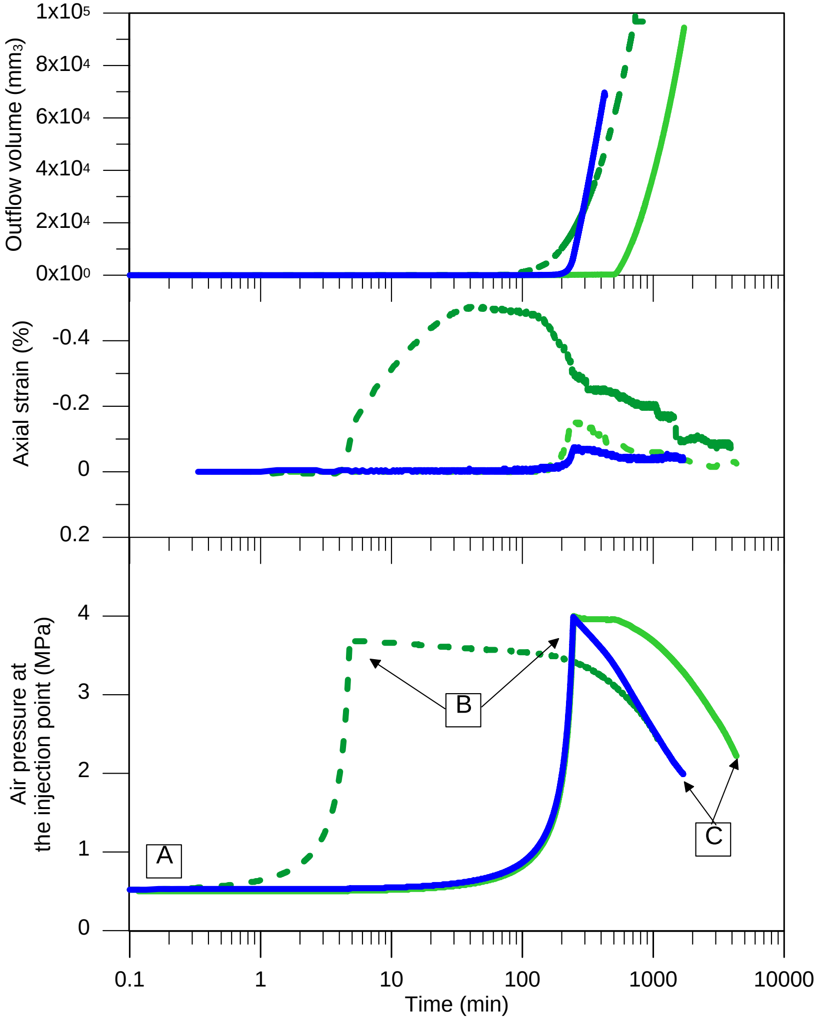
Gas injection tests under oedometer conditions:

- Different bedding orientation (parallel and normal to the flow)
- Different volumetric gas injection rates (slow and fast)
- Several gas injections + re-saturation in between





Bi-modal pore size distribution after gas tests: natural pores (matrix) and fissures (damage/degradation)



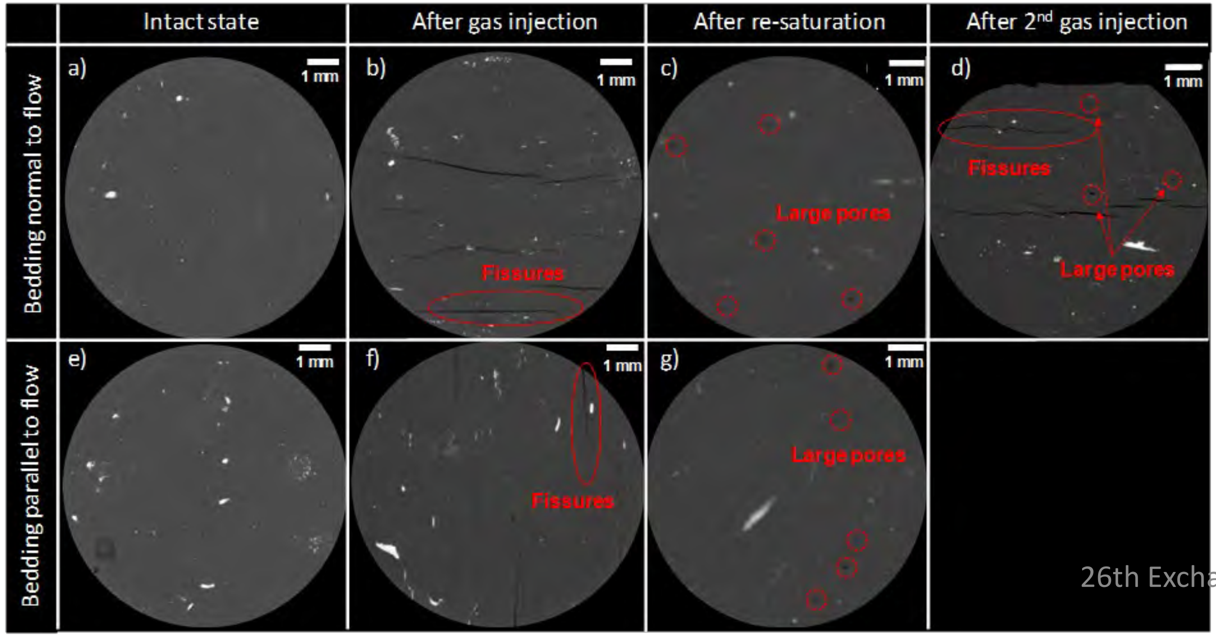
Small deformations during gas injection in both orientations, larger for higher injection rates

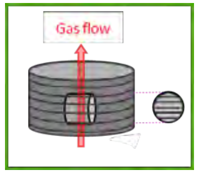
GAS PATHWAYS AND SELF-SEALING IN BOOM CLAY

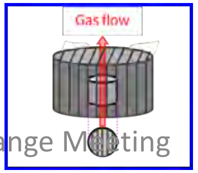
- **Evidence a dilatant behaviour of the Boom Clay associated to gas passage**
  - Gas mainly flowed through multiple dilatant preferential pathways (fissures) **taking advantage of local defects or planes of weakness**, confirmed by microstructural tests
  - After re-saturation, initial values of water permeability were restored despite the detection of some pores due to gas entrapment

    → **effective self-sealing**

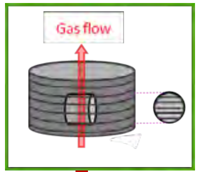
MICRO-CT *Resolution 20 μm*

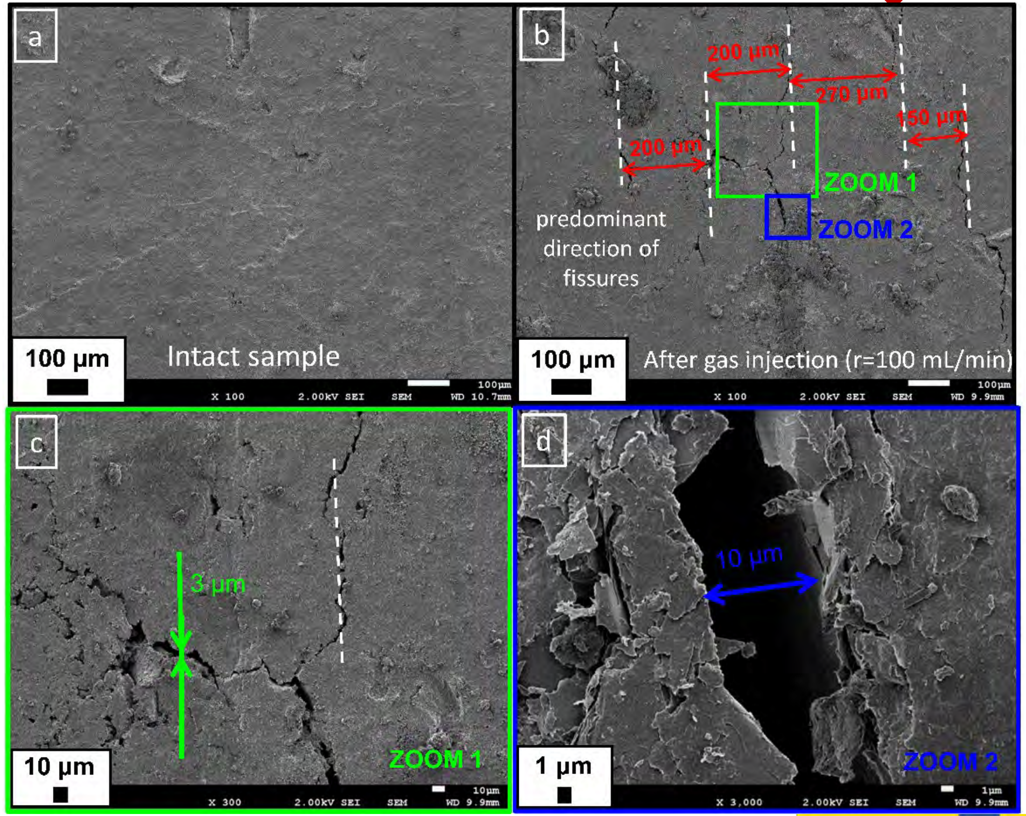






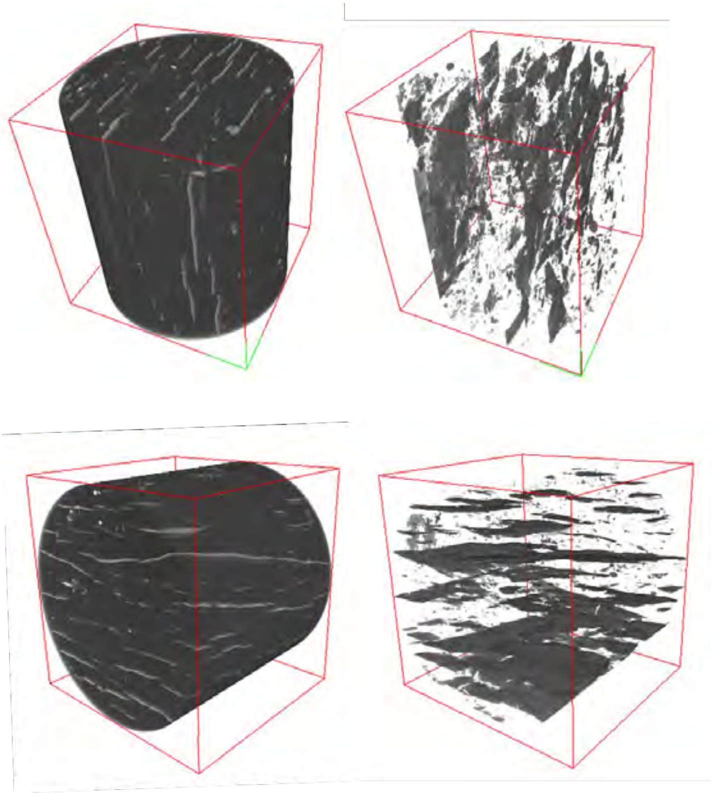
FESEM

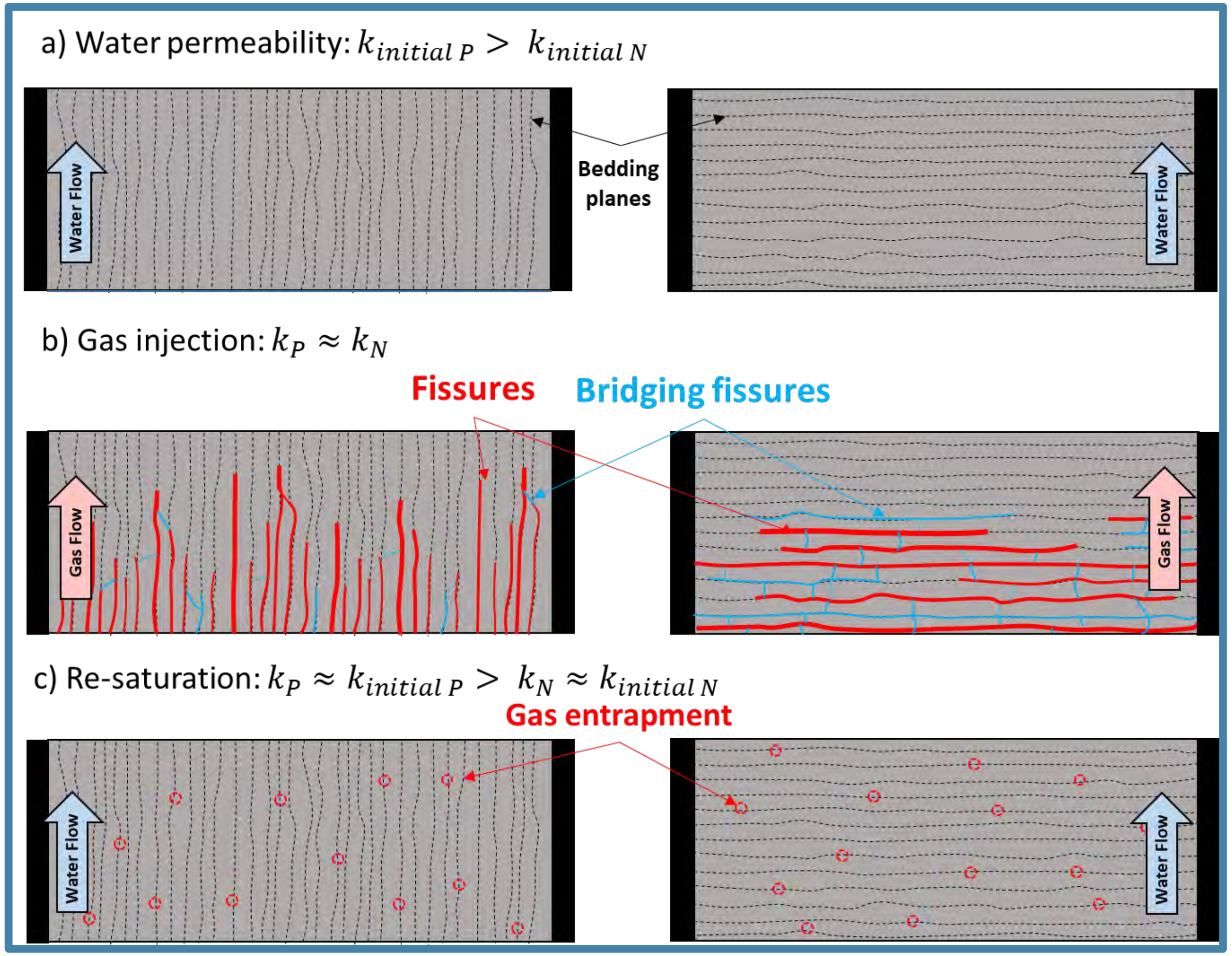




GAS PATHWAYS AND SELF-SEALING IN BOOM CLAY

- **Evidence a dilatant behaviour of the Boom Clay associated to gas passage → Conceptualization**

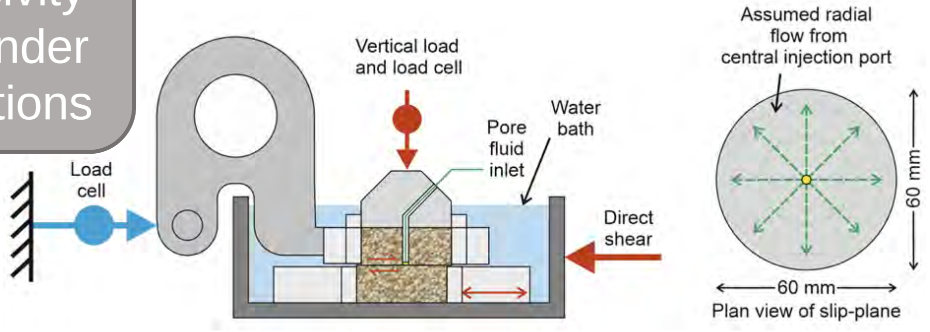




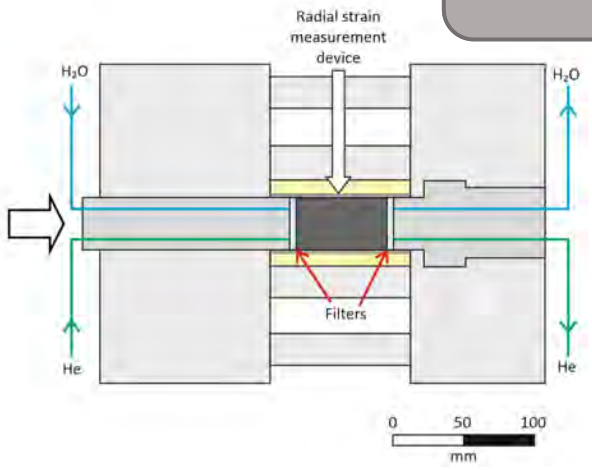
GAS PATHWAYS AND SELF-SEALING IN BOOM CLAY

- **Evidence a dilatant behaviour of the Boom Clay associated to gas passage and efficient self-sealing confirmed in various mechanical load conditions**

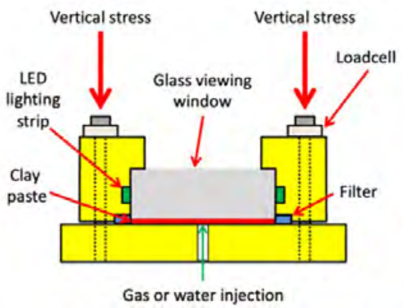
Gas transport on fracture transmissivity and self-sealing under direct shear conditions



Gas injection tests under triaxial conditions



Fracture visualization and self-sealing



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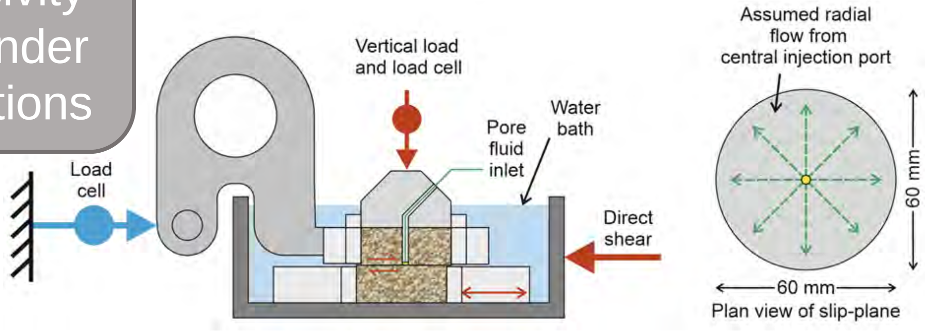

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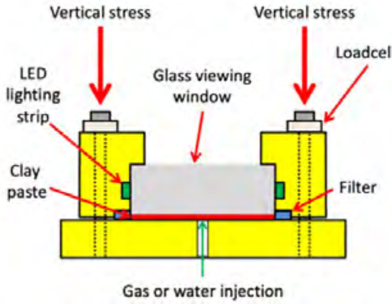

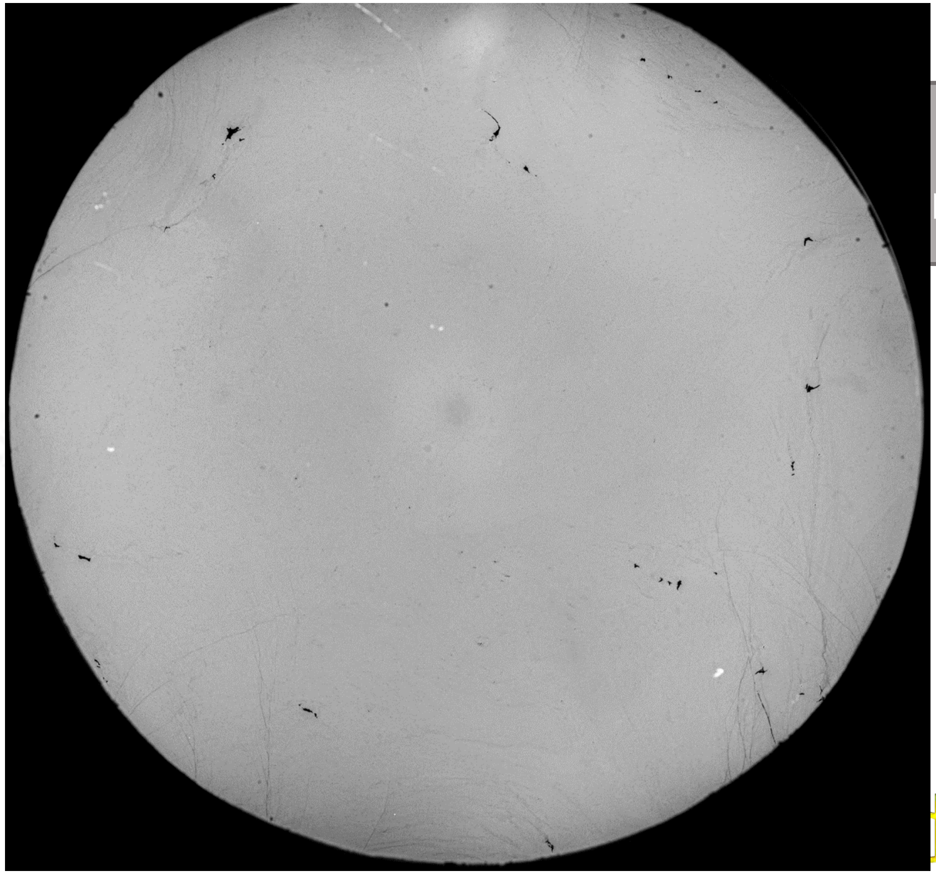

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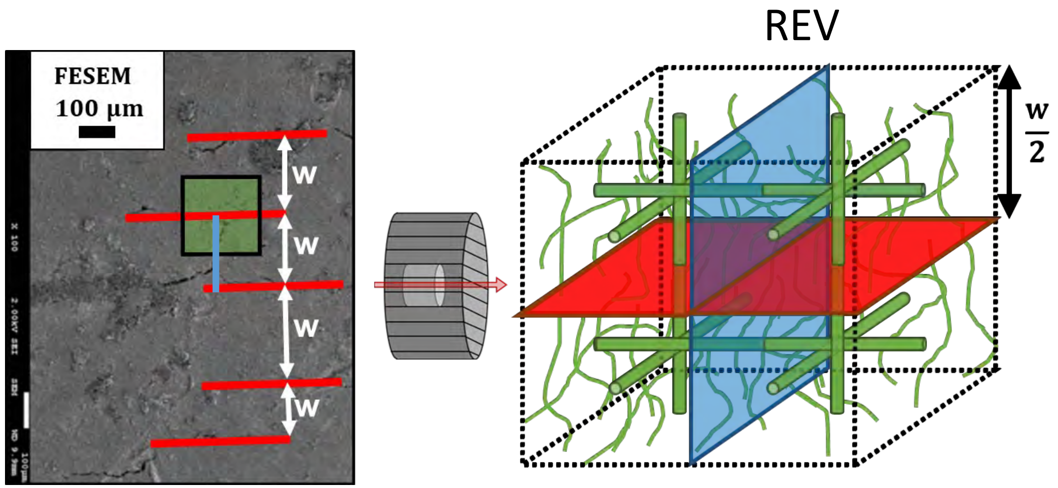
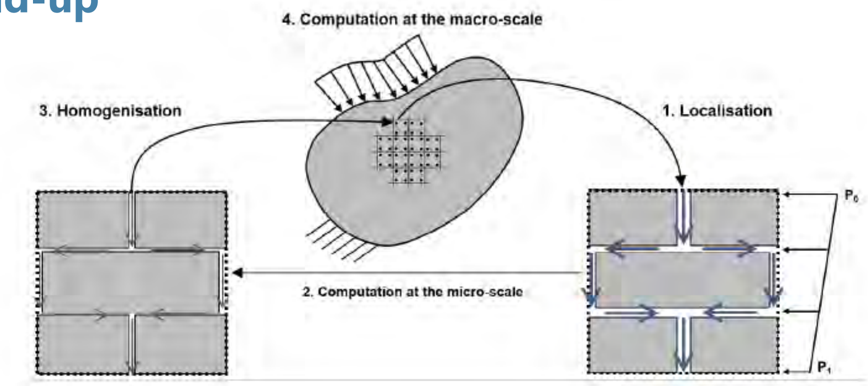
GAS PATHWAYS AND SELF-SEALING IN BOOM CLAY

Multi-scale hydro-mechanical modelling of gas transport

- Constitutive model at the micro-scale (REV) integrated into a multi-scale scheme using homogenisation and localisation techniques for the transitions to the macro-scale.
- Advection-diffusion model for multiphase flow along fractures and tubes embedded in a REV
- Implicit Hydro-mechanically coupling: dependency of the permeability on the fracture and tube apertures (flow channel model), which are stress-dependent

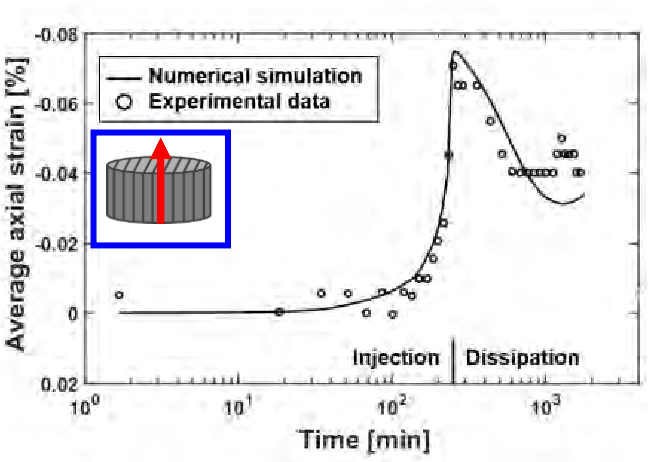
→ diffusion mechanism of dissolved gas at low pressure + development of gas-filled pathways through the fractures within the microstructure of the material when pressure build-up

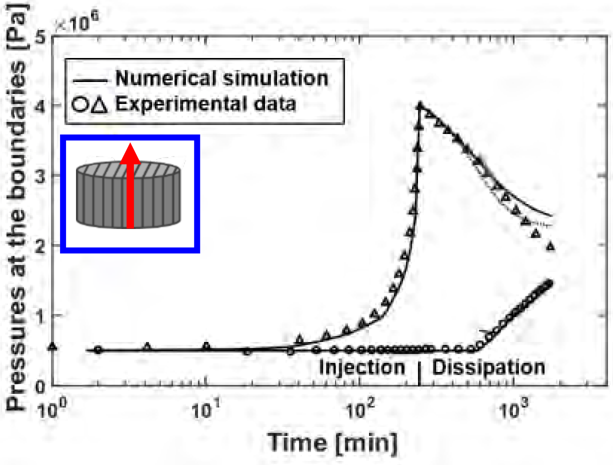


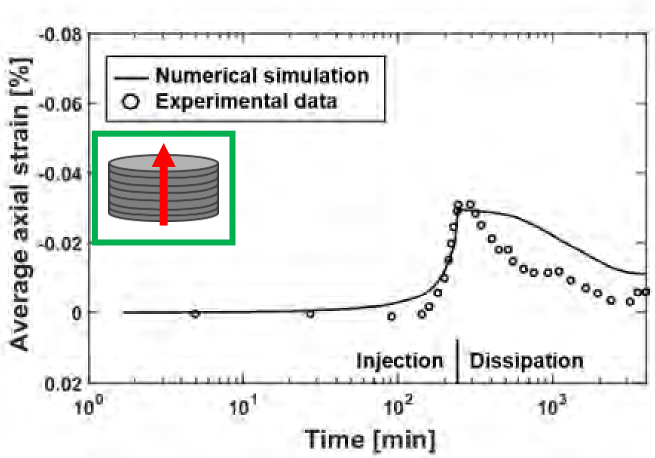


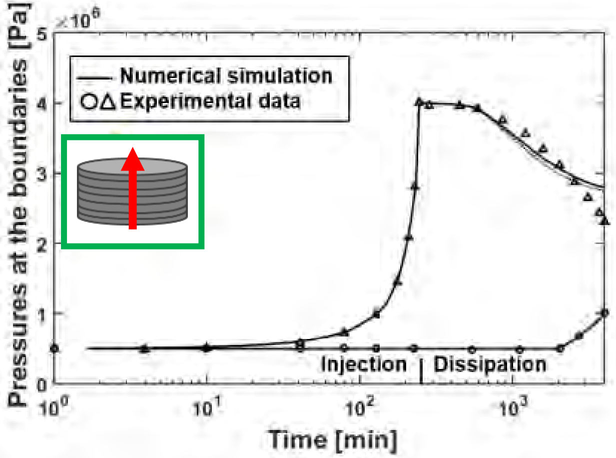
- ► 1 fracture = bedding plane + a network of connected porosity
- ► The pore network is substituted with an assembly of tubes
- ► 1 thinner fracture = bridging plane

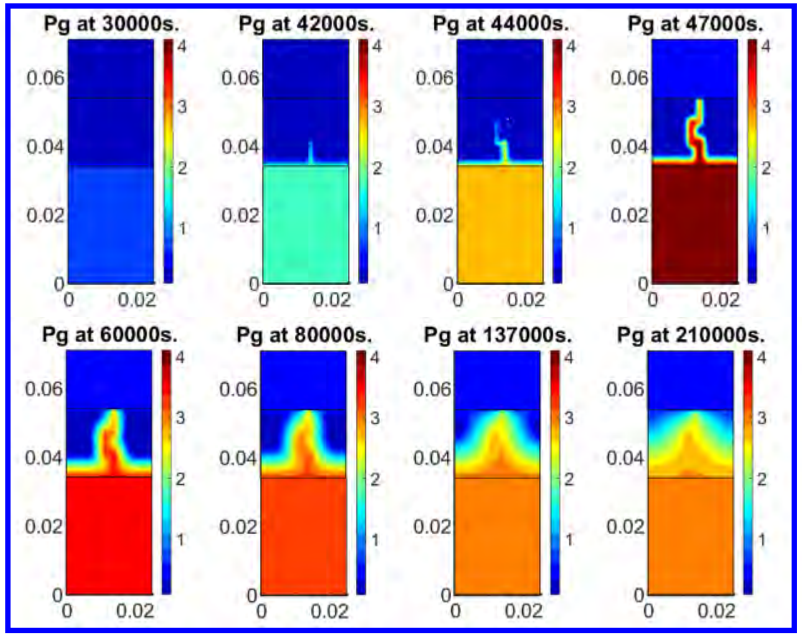
GAS PATHWAYS AND SELF-SEALING IN BOOM CLAY

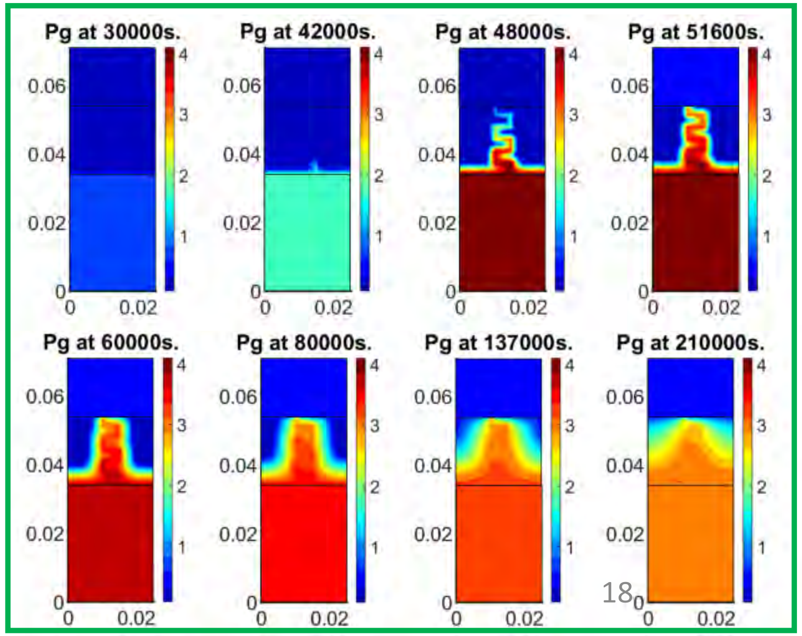










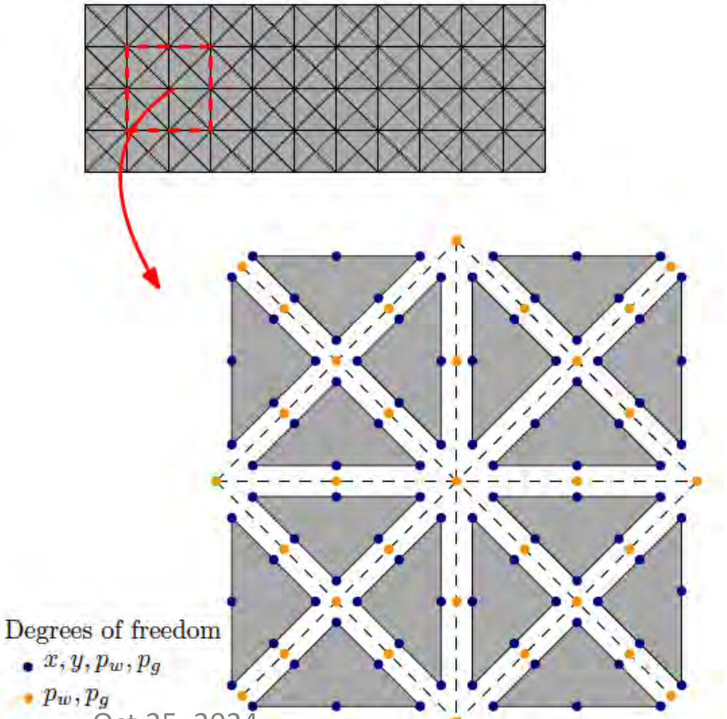


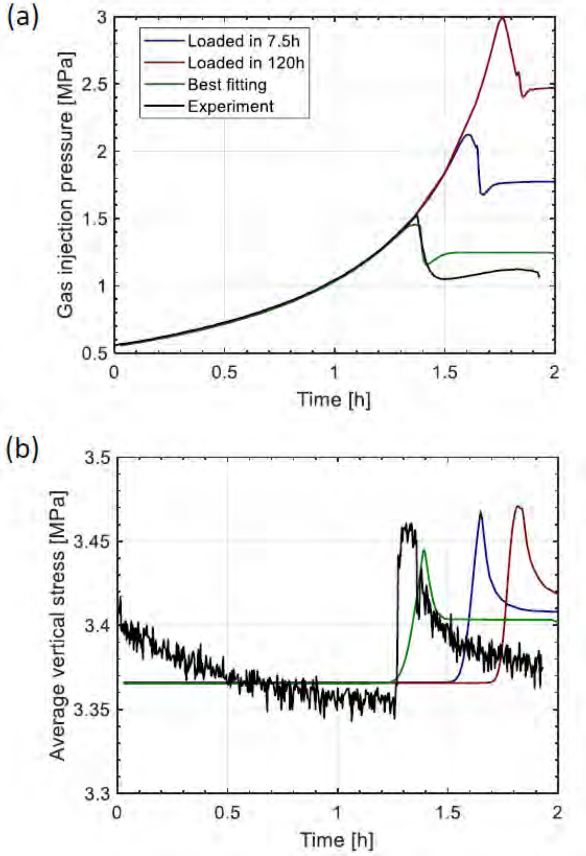
Model simulations well capture experimental results in terms of gas pressure evolution, volume change and outflow volume computation → **very useful support to interpretation and for testing of hypotheses on gas transport mechanisms**

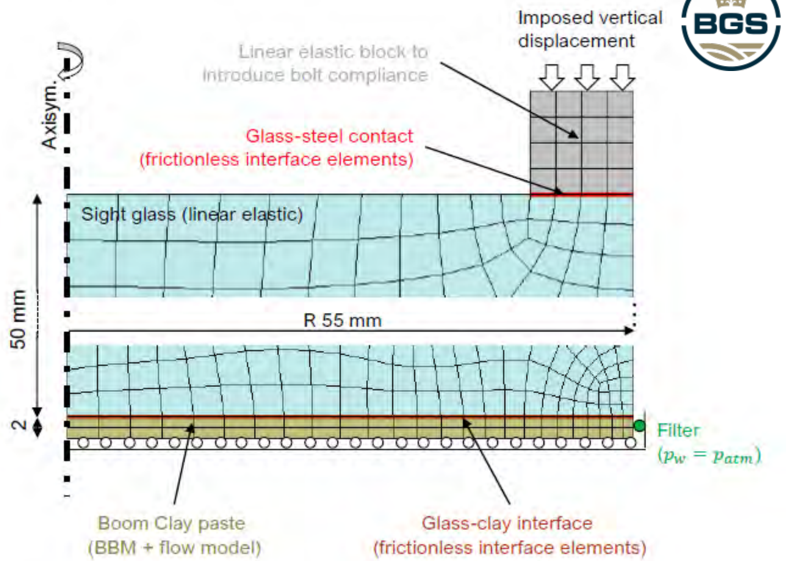
GAS TRANSPORT IN FRACTURES

Pneumo-hydro-mechanical (PHM) model for gas cracking

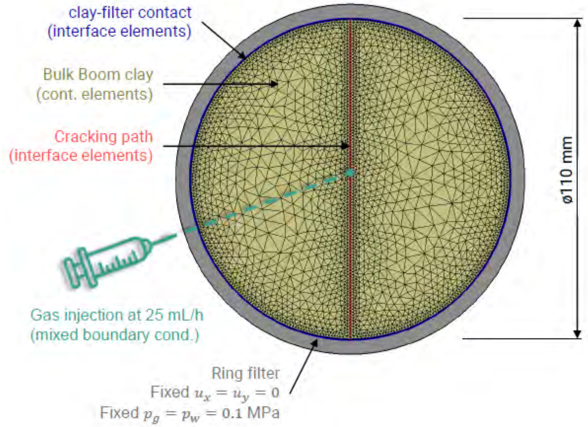
- Explicit representation of gas cracking via zero-
elements equipped with cohesive fracture cons
introduced a priori in between continuum elem
cracking paths







Gas injection



With single straight cracking path, the model can qualitatively replicate the gas injection pressure and average vertical stress

EURAD-GAS – KNOWLEDGE GAINED

- The experimental programme has taken the setups developed in FORGE and before to a new level

→ **Notable progress was done in visualization techniques** (post-mortem but also near-real time during experiments)

- **The numerical programme has made significant progress in support to interpretation and for testing of hypotheses on gas transport mechanisms**

→ development of process-level modelling that combines pathways activation and development mechanisms at the microscopic scale and the evaluation of field parameters and conditions at larger scale **→ predictive capabilities are still very limited**

Mechanistic understanding of gas transport in clay(ey) materials

- **Diffusion of dissolved gas is pretty well understood and characterized**
- **Advective transport of gas is more likely to take place via specific pathways, without significant displacement of water from the matrix**
- **Role of coupling between gas transport and mechanical behaviour is confirmed**
- **Good understanding of the specific hydromechanical conditions imposed by each setup is essential for correct interpretation of the experiments and use of their results**

EURAD-GAS – OVERVIEW OF KNOWLEDGE GAINED

Mechanistic understanding of gas transport in clay(ey) materials

- **What did we learn** from an experimental & modelling programme on gas transport?
- **What do we expect** about the impact of gas passage on clay materials and barrier integrity?

→ **focus on the science**

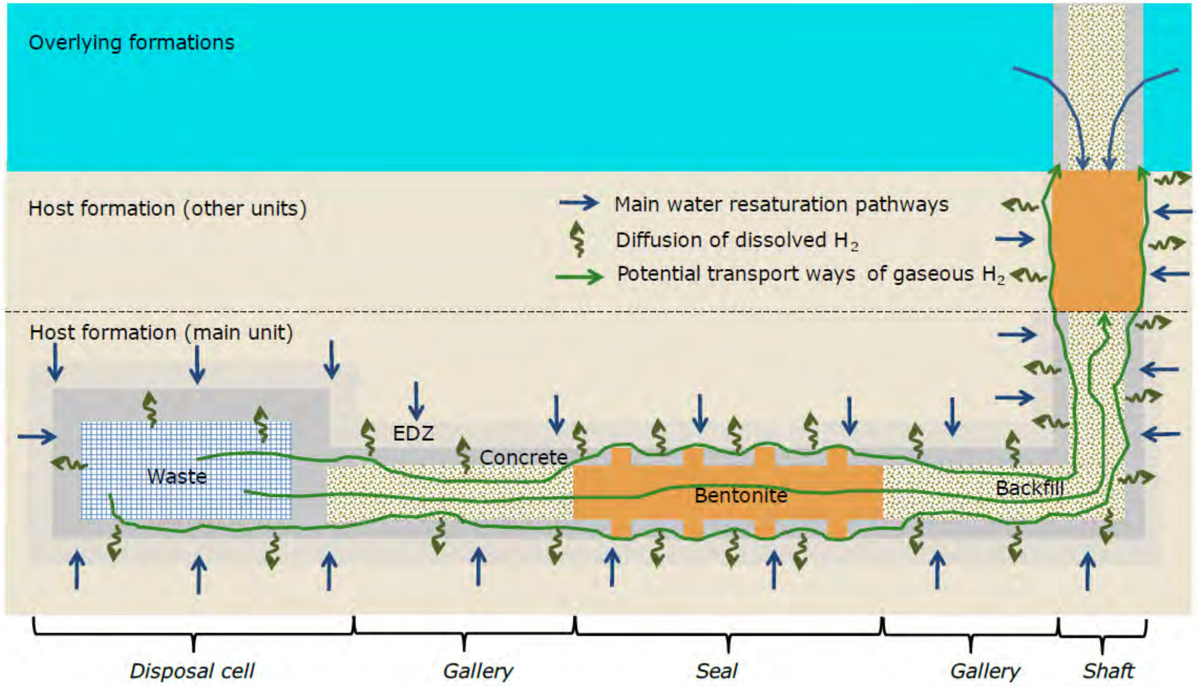
Integration in conceptualizations of the functioning of a repository system

- **Expert judgment and Storyboards** for typical repository configurations
- **Testing ≠ approaches** to represent gas effects at the component & system level

→ **focus on the applications and impacts for end-users**

CONCEPTUALISATION OF GAS TRANSPORT AT THE SCALE OF DISPOSAL SYSTEM

- **Integration of scientific knowledge in storyboards to evaluate**
  - gas pressures and gas fluxes throughout the system,
  - long-term integrity of barriers after the passage of gas
  - gas-mediated radionuclide releases



Evacuation from the disposal system is expected

- in **gas form**, through the EBS (gas-specific pathways through buffer, backfill and seal materials + lining & interfaces) & the EDZ
- only as **dissolved gas** throughout the undisturbed host rock, by diffusion

Should pathway formation be sufficient to relieve gas pressure, then closure of the pathways can occur

→ **self-sealing capacity of clays is preserved** for what concerns hydraulic conductivity but not necessarily with respect to gas entry (pathway may re-open at lower gas pressure)

CONCEPTUALISATION OF GAS TRANSPORT AT THE SCALE OF DISPOSAL SYSTEM

- **Mobilisation of water-soluble radionuclides by gas → expected to be very low because little or no water displacement is associated to the evacuation of gas**
- **Volatile radionuclides can be carried from the gas source to the shafts and/or ramps along with the inactive gas. However:**
    - **Part will continue to dissolve** (as inactive gas also does) into the porewater present in the surrounding materials
    - **Transport may take several hundreds to several thousand years** (order of magnitude, concept dependent)
    - Only radionuclides with ½ lives around this duration or higher will present a significant concentration in the gas phase when arriving in the upper formations

In-depth, quantitative evaluation is system-specific, but should be based on such a prior conceptualisation of the functioning of a disposal system

ONDRAF nagra ANDRA IRSN COVRA Radioactive Waste Management

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GAS IMPACT ASSESSMENT BY REPOSITORY SCALE MODELLING

- **Repository-scale modelling of gas transport accounting for <u>all</u> couplings**

→ **Out of reach and will remain so in the foreseeable future**

→ **However, modelling at the scale of the repository is possible today using simplified, modular approaches guided by the gas transport storyboards and based on dissolution, diffusion and two-phase flow phenomena**

  - Gas is expected to flow over the network of excavation of the repository toward the shafts and/or ramps. Quantification of such process can be done by:

    → Simplified models at repository scale to get an estimate of maximum gas pressures in the repository

    → Component scale coupled models for finer, hydromechanical understanding of the system

RECOMMENDATIONS TO AVOID HIGH GAS PRESSURES

- **Hand-in-hand approaches for the development of the design**
  - Dialogue between Science, Engineering and Safety
  - Example of mitigation measures against too high gas pressures (i.e. preserve the host rock integrity, avoid fracturing pressure):
    - limit as far as possible the surface of metal present in the repository at closure (e.g., limit quantities, use non-metallic rebars for concrete reinforcement,…)
    - design repository layout, backfill and/or seals to favor diffusion of dissolved gas

→ Good practices to avoid painful adaptations at a later phase require a management of gas requirements together with all other requirements early in the design stage!

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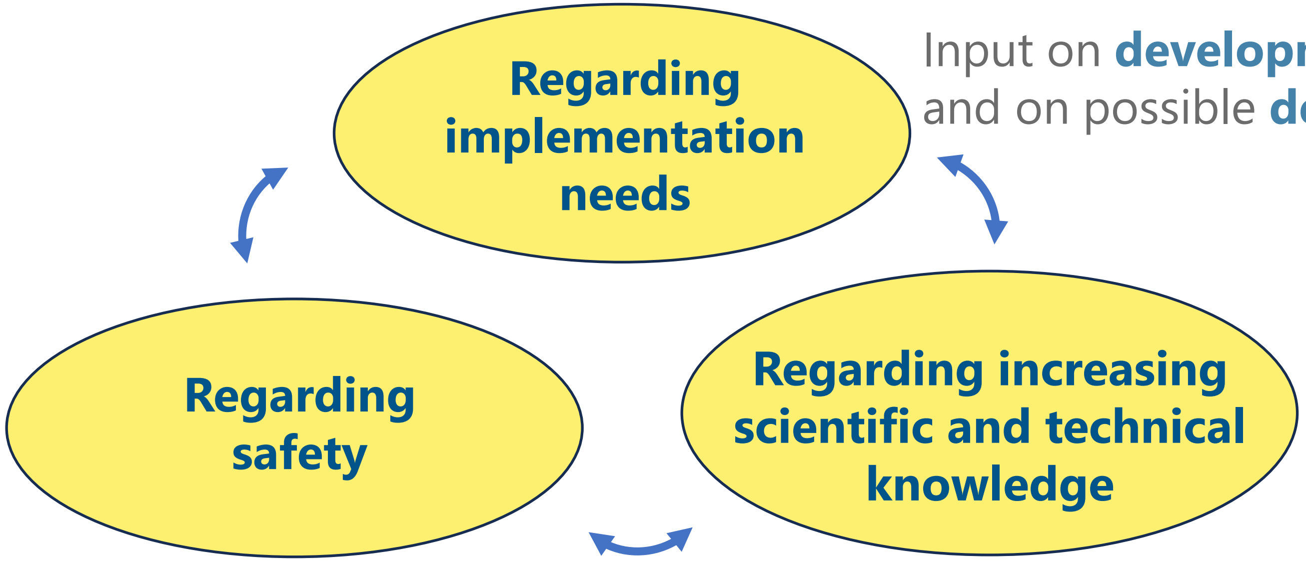
IMPACTS OF EURAD-GAS FOR END-USERS



Input on **development of gas requirements** and on possible **design measures**

Experimental evidences that can be referred to by national programmes in the **arguments supporting claims about long-term safety**

Build confidence in and extending the scientific bases
Data of relevance for a large variety of disposal concepts
Good understanding of processes which has broad end-user appeal

Identification of **strengths and limitations of various approaches** to assess their **suitability in different contexts**

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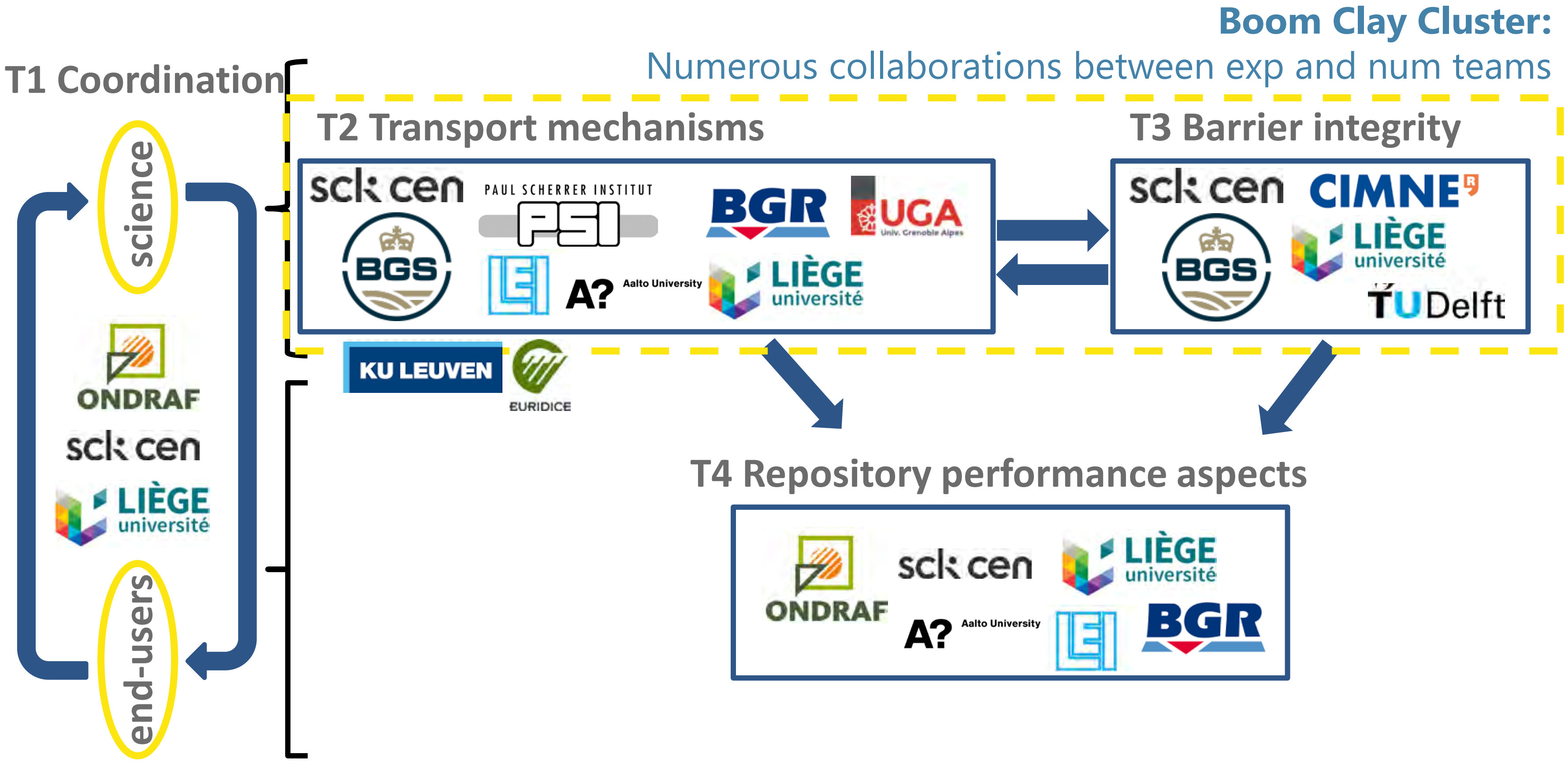
EURAD-GAS AS A MAJOR STEP FORWARD …

- **Consolidation of knowledge:** extension of the scientific bases, both for EBS and clayey rocks
- **Shared understanding and confirmation of the FORGE insights about gas-related processes**
- **Stepwise integration of the scientific knowledge under the form of a generic description of gas transport at repository scale and of its impact in terms of barrier integrity and fate of radionuclides**

- **EURAD-GAS results and conclusions are**
  - Summarized in **EURAD deliverables**: state-of-the-art report (D6.1), main conclusion report (D6.6) and Technical reports detailing the results of each scientific tasks (D6.7 – Characterization of gas transport modes, D6.8 – Effects of gas on barrier integrity, D6.9 – Repository scale modelling) - https://www.ejp-eurad.eu/publications
  - Put in perspective as a **state-of-knowledge** on gas transport in clay barriers in the 2nd state-of-the-art (D6.2)

+ the **publication of a special issue of *Environmental Geotechnics*** (to be published beg. 2025)

… WHICH IS BENEFICIAL TO THE BELGIAN PROGRAM



WP has benefited from the mobility and training program sck cen, as well as significant support from nagra. ANDRA

13 mobility grants obtained for EURAD-GAS

2 training courses organized by and at LIÈGE université *, one jointly with ALERT Geomaterials network*

EURAD-GAS IS A SUCCESS FOR THE BELGIAN PROGRAM

- **SCK CEN** → European leader for experiments on diffusion of dissolved gas
- **CIMNE and ULiège** → key contributions to characterizing gas transport, an example of collaboration
- **ONDRAF/NIRAS** → Conceptualisation of gas transport on the scale of a repository acclaimed by peers

EURAD-GAS is a success for the Belgian Programme, an example of fruitful and beneficial collaboration between ONDRAF/NIRAS and SCK CEN, which began right from the development of the initial project proposal

- 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) – **ONDRAF/NIRAS – NAGRA – Andra**
- Gas transport in Boom Clay: the role of the HADES URL in process understanding (Jacops et al., 2023) – **SCK CEN – ONDRAF/NIRAS – EURIDICE**
- Finite element modelling of multi-gas flow in expansive clay (Gupta, Jacops et al., 2023) – **SCK CEN – UAalto**
- On multi-component gas migration in single-phase systems (Pitz et al., 2024) – **SCK CEN – BGR**
- An experimental methodology to assess the impact of desaturation on gas diffusion in clay-based materials (Gowrishankar et al., 2023) – **SCK CEN – KULeuven**
- Mobility of dissolved gases in smectites under saturated conditions: effects of pore size, gas types, temperature, and surface interaction (Owusu et al., 2022) – **PSI**
- Diffusion and gas flow dynamics in partially saturated smectites (Owusu et al., 2023) – **PSI**
- A multi-scale insight into gas transport in a deep Cenozoic clay (Gonzalez-Blanco and Romero, 2022) **– CIMNE**
- Hydro-mechanical response to gas transfer of deep argillaceous host rocks for radioactive waste disposal (Gonzalez-Blanco et al., 2022) – **CIMNE – NAGRA – ONDRAF/NIRAS**
- Self-sealing of Boom Clay after gas transport (Gonzalez-Blanco et al., 2023) – **CIMNE – ONDRAF/NIRAS**
- Hydro-mechanical modelling of gas transport processes in clay materials using a multi-scale approach (Corman et al., 2024) – **ULiège – CIMNE – ONDRAF/NIRAS**
- Numerical investigation of the couplings between strain localisation processes and gas migrations in clay materials (Corman et al., 2022) – **ULiège – Andra**
- Modelling gas fracturing in saturated clay samples using triple-node zero-thickness interface elements (Liaudat et al., 2023) – **TUDelft**

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