Gas transport in Boom Clay: the role of the HADES URL in process understanding

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Abstract: 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.

The most widely accepted solution for the long-term management of high and intermediate level radioactive waste is disposal in a deep geological repository (DGR). A system of natural and engineered barriers ensures that the waste is contained and isolated for a sufficiently long time. In Belgium, no formal decision has yet been taken, but for R&D purposes, the Belgian radioactive waste management organization ONDRAF/NIRAS considers poorly indurated clays as potential host rocks for a DGR.

In 1974, SCK CEN started investigating the possibility of geological disposal of radioactive waste in Boom Clay. To study the properties and behaviour of this poorly indurated clay at several hundred metres depth, it was decided at that time to build an underground research laboratory (URL) in the Boom Clay. The construction of the HADES URL began in 1980 and, during the first phase, the First Shaft and First Gallery were constructed. In 1987, a second gallery known as Test Drift was constructed, followed by the Second Shaft in 1997 and the Connecting Gallery in 2002. The last phase was the construction of the PRACLAY gallery in 2007, which is perpendicular to the Connecting Gallery (Fig. 1). The history of the construction of the HADES URL is described in Li *et al.* (2022).

Significant quantities of gas can be generated in a DGR containing radioactive waste. The largest fraction of the gas is expected to be hydrogen produced by the anaerobic corrosion of steel and reactive metals present in the waste and the engineering barrier system (EBS). Even though the gas production processes are generally slow, it is important to verify that they will not be detrimental to the proper functioning of the disposal system. The low permeability of clays is, on the one hand, favourable with respect to the containment function of a DGR, but, on the other hand, it limits the evacuation of the generated gas. In some cases, gas may be generated faster than it can be removed through the EBS and host rock. As a result, a pressurized gas phase may develop in the DGR, which in turn may create discrete, gas-specific pathways through the EBS and/or the host rock.

There are a number of processes associated with the transport of gas from a DGR, regardless of the type of host rock (Marschall *et al.* 2005; Levasseur *et al.* 2021). The rate and amount of gas generated within the DGR are important in determining the subsequent gas transport behaviour. Gas generation will depend on the gas-generating materials and the conditions prevailing in the DGR, including the presence of water, oxygen and some ions (e.g. chloride or sulfate that may promote metal corrosion), pH and temperature (Diomidis *et al.* 2016). In addition, some chemical or biochemical interactions can

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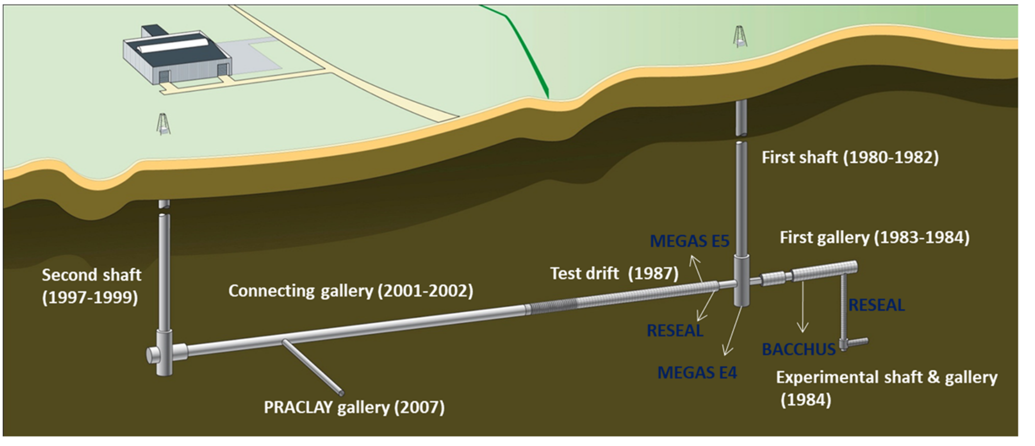

Fig. 1. Location of the different gas injection experiments in the HADES URL.

occur and reduce the volume of gas in the repository (e.g. gas sorption on specific mineral phases, carbonation of cementitious materials and/or conversion of hydrogen gas into methane by microbes) (Leupin *et al.* 2016).

As detailed in Figure 2, the gases produced in the DGR will first dissolve in pore water and be transported by diffusion and advection. Because the solubility of gas in the pore water is limited, a separate gas phase may form, depending on the production

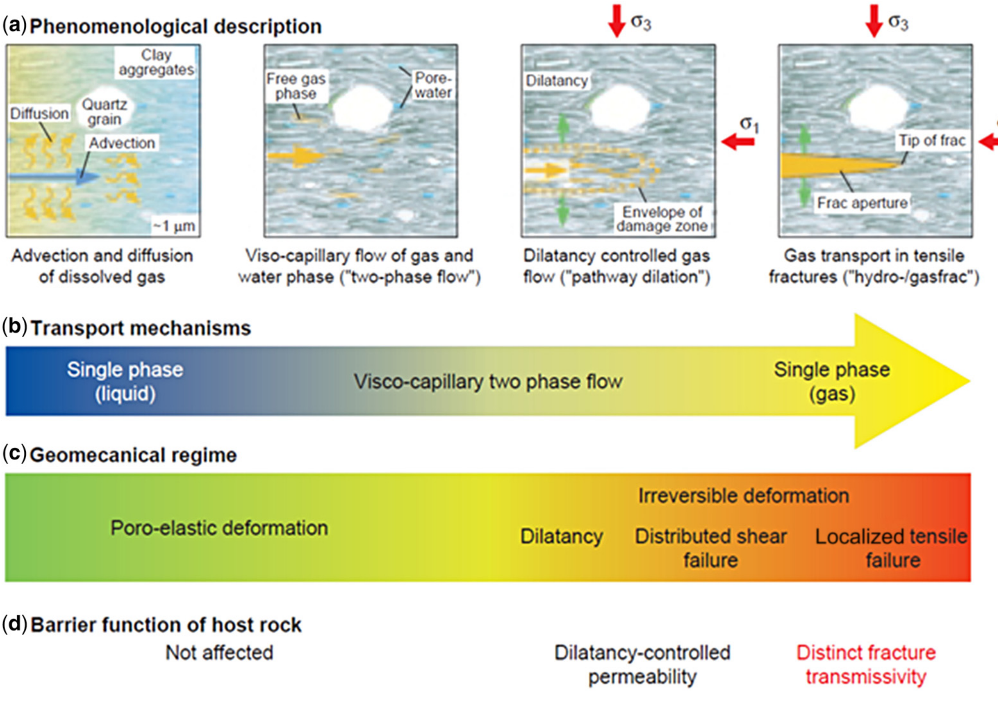

Fig. 2. Classification and analysis of gas transport processes in all clayey materials (Marschall *et al.* 2005). (**a**) phenomenological description based on the microstructural model concept (σ_1: horizontal total stress, σ_3: vertical total stress); (**b**) basic transport mechanisms; (**c**) geomechanical regime; (**d**) effect of gas transport on the barrier function of the host rock.

rate. If this happens, the gas may be transported by dilatancy-controlled gas flow ('pathway-dilation') or through tensile fractures created by the increased gas pressure. Whether and how a gas phase is transported through an engineered barrier system (EBS), host rock or overlying rock material depends on the properties of those materials.

To evaluate the impact of gas on the functioning of a DGR, adequate understanding of possible gas transport modes through clay barriers is essential. This paper aims to summarize the state-of-knowledge on gas transport in Boom Clay and to introduce the upcoming *in-situ* gas diffusion test, NEMESIS (Neon diffusion in MEGAS *in situ*), which will be carried out in the HADES URL to enhance current understanding.

State of knowledge on gas transport in Boom Clay

Small-scale gas injection experiments in the surface laboratory

Diffusive transport of dissolved gas. A free gas phase will form in the DGR when more gas is generated than can be dissolved in the pore water. The amount of gas that dissolves, in turn, depends on the rate at which the gas is transported away. In a low-permeable medium such as Boom Clay, the transport of dissolved gas will mainly take place via diffusion. Assessing whether a free gas phase will form therefore requires knowledge about the gas diffusion coefficients in the pore water.

A first attempt to obtain gas diffusion parameters for hydrogen in Boom Clay was made through the EC MEGAS (Modelling and Experiments on Gas Migration in Repository Host Rocks) project (Volckaert *et al.* 1995; Ortiz *et al.* 1997). It consisted of through-diffusion experiments and in-diffusion experiments with hydrogen (H_2 pure gas phase). In the through-diffusion experiments, clay samples were placed between two reservoirs: one with a high concentration of hydrogen and the other without hydrogen. By monitoring the evolution of the hydrogen in both reservoirs, the hydrogen diffusion coefficient of the clay was determined using a transport model based on Fick's law. The in-diffusion experiments used only one reservoir of water with a known initial concentration. The decrease in hydrogen concentration in this reservoir was then used to calculate the diffusion coefficient.

These experiments, however, did not allow an accurate measurement of the diffusion coefficient (Volckaert *et al.* 1995; Aertsens 2009). The through-diffusion experiments suffered from CO_2 outgassing from the clay samples and the in-diffusion experiments were disturbed by hydrogen leakage. In addition, the duration of the experiments was considered too short to give accurate parameter values. It was estimated that there was an uncertainty of up to two orders of magnitude. Therefore, only the apparent diffusion coefficient D_{app} (between 5×10^{-12} m^2 s^{-1} and 4×10^{-10} m^2 s^{-1}) could be estimated.

That is why SCK CEN, with the support of ONDRAF/NIRAS, has developed a new method to measure diffusion coefficients of dissolved gases in Boom Clay. The new test setup (Fig. 3) consists of two water reservoirs filled with approximately 500 ml synthetic pore water and 500 ml gas at 1 MPa. These are placed on either side of a saturated clay sample (diameter 80 mm, length 30 mm). The sample is placed into a constant volume cell and maintained at a constant temperature of 21 ± 2°C. According to Henry's law (Henry 1803), the free gas in the gas phase is in equilibrium with the dissolved gas in the water. The water on both sides is then circulated over the sample and each of the dissolved gases diffuses through the sample from the high concentration reservoir to the low concentration

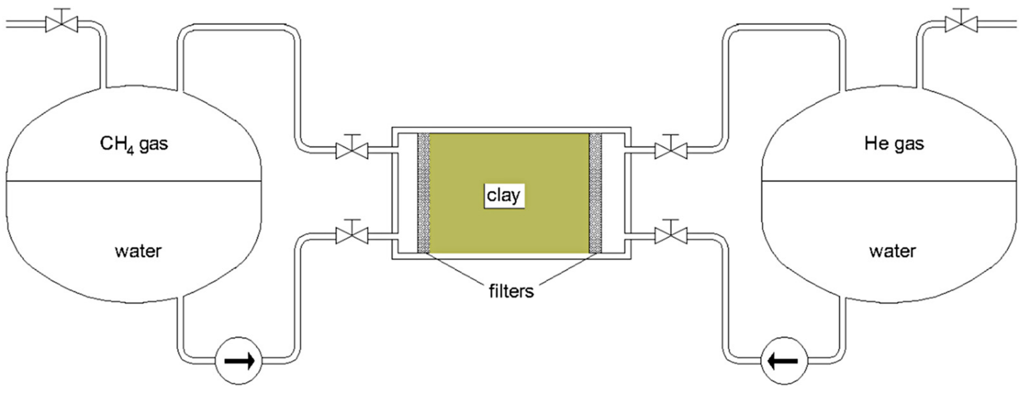

Fig. 3. Layout of the lab-scale gas diffusion setup. Source: Jacops *et al.* (2017*a*).

reservoir. Once a constant gas flow regime is reached, the gas phase is sampled on a regular basis until 10 data points are obtained and their composition is analysed by gas chromatography. To avoid leakage and microbial activity, Jacops *et al.* (2015*b*) developed a protocol that combines different sterilization techniques such as heat sterilization, gas filtration, gamma irradiation and the use of a specific inhibitor.

This new method was applied in a research programme that aimed to (i) estimate diffusion coefficients of Boom Clay for different gases, and (ii) assess how lithological variations in the Boom Clay (mainly variations in clay/silt/sand content) affect its transport properties. Therefore, samples from different layers were selected: samples from the 'clayey' middle part (Putte and Terhagen Member), from the 'silty' top part (Boeretang Member) and from the 'sandy' formation above the Boom Clay (Eigenbilzen Sand). These samples were then used to measure the diffusion coefficients of tritiated water (HTO) and a series of gases (He, Ne, H_2, Ar, CH_4, Xe, C_2H_6) (Jacops *et al.* 2020*a*). The measured gases have been selected because they are (1) relevant in a DGR (e.g. H_2 and CH_4), (2) a good proxy for the difficult to measure H_2 gas (Ne) or (3) part of the phenomenological studies that assess the impact of size of the gas molecule on diffusion (He, Ar, Xe, C_2H_6). The diffusion was determined both parallel and perpendicular to the (sub-horizontal) bedding plane. More information on the origin and lithology of the samples can be found in Jacops *et al.* (2017*a*). Diffusion coefficients have been obtained for samples confined in a constant volume cell, hence the stress state of these samples might be different from the *in-situ* conditions. In order to assess the impact of a variable stress state on the diffusion of dissolved gases, SCK CEN and ONDRAF/NIRAS recently launched a new experimental programme, which is currently on-going.

The resulting diffusion coefficients for HTO and a fixed series of gases, which have been measured for the majority of the samples (He, Ne, CH_4, C_2H_6), are given in Table 1, along with the corresponding anisotropy factors. The measured effective diffusion coefficients $D_{\mathrm{eff,h}}$ and $D_{\mathrm{eff,v}}$ are around 3.0×10^{-10} $m^2\ s^{-1}$ and $2.0\times10^{-10}\ m^2\ s^{-1}$, respectively. For neon, the following values for the diffusion coefficients in the Boom Clay were obtained: $D_{\mathrm{eff,h}} = 2.3\times10^{-10}\ m^2\ s^{-1}$ and $D_{\mathrm{eff,v}} = 1.8\times10^{-10}\ m^2\ s^{-1}$ (Jacops *et al.* 2017*b*). The diffusion in the horizontal direction is higher than in the vertical one (Aertsens *et al.* 2009; Jacops *et al.* 2017*a*). The anisotropy is mainly caused by the typical stratification of clay platelets (Vandenberghe *et al.* 2004). This also explains why the anisotropy factor is lower for the more silty or sandy samples from the Eigenbilzen Sand. It is expected that there is a threshold value for the clay content below which the anisotropy starts to decrease. This threshold value is currently unknown.

For all samples, an exponential relationship could be observed between the effective diffusion coefficient and the size of the gas molecules. This indicates that the microstructure of the sample can strongly influence the diffuse transport.

Subsequently, the petrophysical (e.g. mineralogy, grain size distribution, porosity, pore size distribution) and petrographical (CT, light microscopy) properties of the samples were analysed. This showed that the origin and composition of the samples strongly influence the pore size distribution and thus the microstructure of the sample (Jacops *et al.* 2020*a*, *b*). These differences in microstructure in turn affect both the diffusivity and hydraulic conductivity of the samples. The hydraulic conductivity in the Eigenbilzen Sand is two orders of magnitude greater than in Boom Clay, and the effective diffusivity (D_{eff}) is a factor 1.7 greater.

The mineralogical composition and grain size distribution of samples from the 'silty' Boeretang Member are comparable to those of samples from the 'clayey' Putte and Terhagen Member. Nevertheless, the measured hydraulic conductivity in the Boeretang Member turned out to be a factor 5–10 greater than in the Putte and Terhagen Member.

Table 1. *Determined diffusion coefficients for dissolved gases (He, Ne, CH_4 and C_2H_6) and for HTO, measured in Boom Clay and Eigenbilzen Sand samples and the corresponding anisotropy factors*

Determined diffusion coefficient [$m^2\ s^{-1}$] and calculated anisotropy factor	Boom Clay	Eigenbilzen Sand
$D_{\mathrm{eff,h}}$ (He, Ne, CH_4, C_2H_6)	3.0×10^{-10}	3.8×10^{-10}
$D_{\mathrm{eff,v}}$ (He, Ne, CH_4, C_2H_6)	2.0×10^{-10}	3.4×10^{-10}
calculated anisotropy factor	1.5	1.1
$D_{\mathrm{eff,h}}$ (HTO)	2.8×10^{-10}	3.8×10^{-10}
$D_{\mathrm{eff,v}}$ (HTO)	1.9×10^{-10}	3.0×10^{-10}
calculated anisotropy factor	1.5	1.3

This can be explained by a difference in the microstructure (Jacops *et al.* 2020*b*). The locally larger pore sizes can provide a preferential flow path for advective water flow. However, the diffuse transport of solutes, including dissolved gas, may be dominated by the micro- and mesopores in the clay phase.

Visco-capillary two-phase flow. As recalled in Levasseur *et al.* (2021), the classical formulation of flow of immiscible fluids in porous media (i.e. 'two-phase flow') describes the combined flow of a wetting fluid (water) and a non-wetting fluid (gas) in the connected pore system of a rigid/elastic porous medium on the action of viscous and capillary forces. Classical two-phase flow concepts are described by a capillary pressure–water saturation relationship (or water retention curve) and the relative permeability–saturation relationships of the liquid and the gas phases.

The retention curve of Boom Clay water was estimated by Gonzalez Blanco (2017) and Le *et al.* (2008). They used a dew-point psychrometer on samples that were first stepwise dried and then stepwise wetted until saturation. Mercury intrusion porosimetry (MIP) tests were also conducted to determine the pore size distribution, the relationship between matrix suction and degree of saturation, and the gas entry value corresponding to the dominant pore mode. Figure 4 shows the estimated water retention curve including curves based on MIP data and psychrometer measurements and the fitted van Genuchten equation (van Genuchten 1980), defined as a relation between the degree of saturation and the soil suction (van Genuchten 1980). Using Laplace's equation, the Boom Clay gas entry value, i.e. the suction for which desaturation becomes significant and corresponding to the dominant pore mode detected from MIP data, was found to be approximately 4.8 MPa (Gonzalez Blanco 2017).

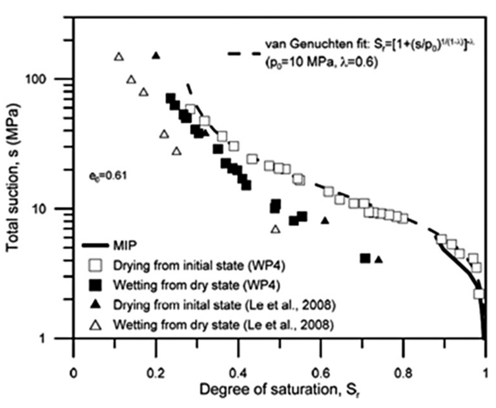

Fig. 4. Boom Clay water retention curve. Source: Gonzalez Blanco (2017).

Water and gas permeameter experiments were performed in the lab to determine the mobility of the liquid/gas phase in intact Boom Clay, expressed in terms of relative permeability relationships for both phases. The intrinsic permeability depends on the pore structure and the relative permeability controls the variation of permeability in the unsaturated regime. Empirical relationships are used to establish the dependency between the liquid and gaseous phases and the degree of saturation. According to Delahaye and Alonso (2002), the gas relative permeability of the Boom Clay can be expressed as a generalized power law fitted on experimental data from Volckaert *et al.* (1995):

$$k_{\mathrm{rg}} = A\,(1 - S_{\mathrm{r,w}})^{\beta}$$

where k_{rg} is the gas relative permeability [–], $S_{\mathrm{r,w}}$ is the degree of saturation and A and β are material parameters equal to 1 and 2.8, respectively (see Fig. 5).

In multiphase flow concepts, the gas flow is controlled by visco-capillary forces (phase interference between wetting and non-wetting liquid). In the case where the gas phase causes microfracturing, the increase in pore space leads to a detectable increase in clay permeability and a change in the capillary pressure–saturation relationship. The transport properties of the clay (permeability, capillary pressure) depend on the deformation state of the clay and can therefore no longer be considered invariants.

Gas specific pathways. Visco-capillary two-phase flow is unlikely in low strength materials, such as Boom Clay. This means that gas transport is unlikely

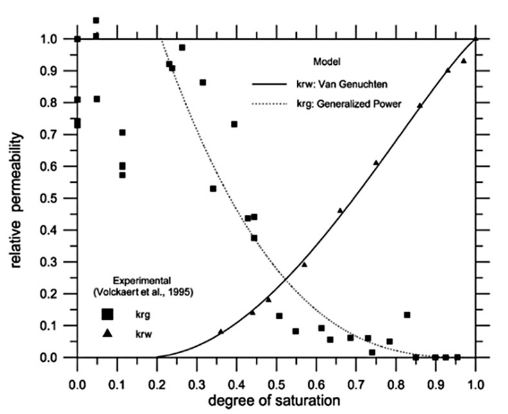

Fig. 5. Experimental data for relative permeability in Boom Clay (Volckaert *et al.* 1995) together with model fitting (Delahaye and Alonso 2002) (k_{rw}: relative permeability of water/liquid phase; k_{rg}: relative permeability of gas).

to occur until gas pressure build up is sufficiently high to cause the formation of specific pathways (microfractures or 'pathway dilation'). Once pathways are formed, gas flows along these. Once the gas pressure is released, they are expected to close and seal. The efficiency of the pathway sealing depends on the self-sealing capacity of the clay.

As part of the EC MEGAS project, SCK CEN and the British Geological Survey (BGS) conducted gas injection experiments on Boom Clay samples in oedometer and isotropic cells with different gas injection rates and stress states (Volckaert *et al.* 1995). These tests showed that gas breakthrough only occurred above a threshold pressure, which was followed by a negative transient leading to steady-state gas flow. A non-linear relationship between the flow rate after breakthrough and the pressure was observed. Despite the sometimes large volumes of gas flow, the degree of saturation did not change significantly. The authors concluded that these experimental observations are consistent with gas flow by dilatant pathway formation and that the measured gas permeabilities depend on the sample deformation and the stress applied on the samples. Further, according to authors, the gas flow pathways open and close as the gas pressure varies, which explains the observed non-linear behaviour of the flow rate. Some tests showed an intermittent or burst-type flow response which was attributed to pathway propagation/collapse.

The gas flow mechanically disturbs the clay. Gonzalez Blanco (2017) and Gonzalez-Blanco *et al.* (2022) evaluated how the gas flow changes the microstructure of the clay. They suggest that flow mainly occurs along natural discontinuities. Gonzalez-Blanco *et al.* (2022) revealed that the bedding plane plays a fundamental role in gas transport and demonstrated the high hydromechanical coupling during gas injection experiments. Under oedometric conditions, samples with bedding planes perpendicular to the gas flow expanded more during the gas equalization stage and compressed more during the gas dissipation stage than samples with bedding planes parallel to the of gas flow (Fig. 6). This shows that under these oedometric conditions gas pathways develop more easily along bedding planes.

This effect is even more pronounced when the gas injection rate is increased. Increasing the injection rate resulted in a higher expansion as the pressure front propagates (Fig. 6). At a low injection rate, the pressure front did not propagate after gas injection was stopped and the remaining gas volume in the inlet line was allowed to dissipate. Gonzalez Blanco (2017) showed that the pore pressure was close to equilibrium during slow gas injection and that the volume change was quasi-reversible during the dissipation stage. Consequently, gas pathways form more easily and their effect on the volumetric strain is more limited when the gas flow is slow.

After the sample was dismantled, the pathway was checked for desaturation. This confirmed that the clay matrix had remained saturated and so the degree of desaturation was low (Gonzalez Blanco 2017). The gas pathways resulted in new pore sizes without significantly changing the matrix porosity. This means that only very small additional classes of (large) pore radii (>2 µm) become accessible for gas transport and changes in the intrinsic permeability of Boom Clay are limited (Hildenbrand *et al.* 2002, 2004). Sample deformation is observed during gas breakthrough experiments, but as illustrated by Harrington *et al.* (2012), the gas flow only occurs locally via a gas-specific pathway and is correlated with the local increase in gas transport capacity.

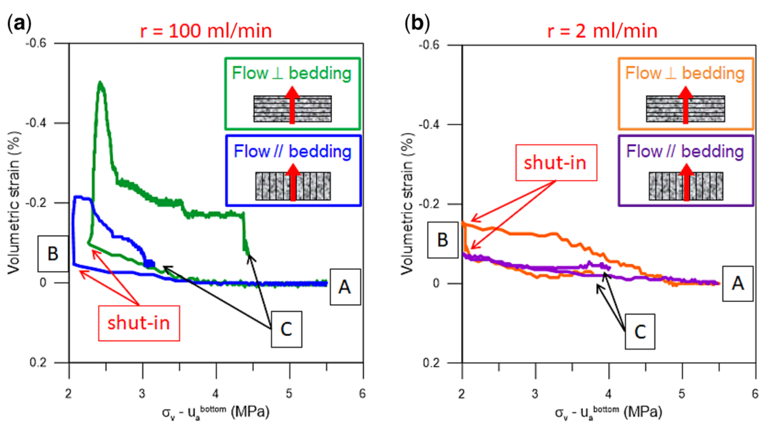

Fig. 6. Evolution of volumetric strains with vertical stresses during air injection/dissipation stages in (**a**) fast and (**b**) slow injection experiments. Source: Gonzalez Blanco (2017).

As part of the EC FORGE project, SCK CEN examined gas transport within discontinuities in Boom Clay samples (Jacops *et al.* 2015*a*). Undisturbed Boom Clay samples were used, together with samples that were made up by placing two samples against one another. In that way the gas transport along the interface between these two samples could be investigated. All samples were placed above a NaI-conditioned sample and confined at constant volume within a permeameter cell. The downstream filter was immersed in natural Boom Clay pore fluid. At the upstream end of the sample, an excess gas pressure was applied using helium. The gas pressure was increased stepwise until there was gas breakthrough and water was expelled from the downstream end of the sample. This water was then analysed for iodine enrichment to assess whether gas-driven transport of the tracer had occurred.

Assuming that the unretarded diffusion of iodine and background concentration of iodine in the natural Boom Clay pore water is slow, the volume of iodine that was transported by gas breakthrough was limited. Furthermore, the tests showed that there was no significant displacement of water before and during gas breakthrough and that the degree of sample desaturation was less than 0.5%. The downstream iodine concentrations were similar for recombined Boom Clay/Boom Clay samples which was, according to the authors, evidence of the rapid self-sealing of gas-induced fractures and artificial interfaces in Boom Clay. As described by Jacops *et al.* (2015*a*), the experiments were performed in constant volume cells in which the stress state of the samples was not controlled. Therefore, the results should be interpreted carefully and no extrapolation to, for example, the scale of a repository can be made.

Gas injection experiments in the HADES URL

The research discussed so far enabled us to gain mechanistic understanding and property measurements at lab scale. It was then tried to validate and refine this knowledge and understanding to larger scales using mock-up and long-term *in-situ* experiments. One of the specific objectives was to examine whether the strong coupling between the geomechanical and hydraulic properties of the Boom Clay, as observed in the lab, could be confirmed at larger scale. Field-scale gas injection tests using helium were conducted at the HADES URL as part of the EC MEGAS project (Volckaert *et al.* 1995). These experiments, MEGAS E4 and MEGAS E5, are described in detail by Ortiz *et al.* (1997) and Volckaert *et al.* (1995).

MEGAS E4. The MEGAS E4 gas injection experiment (1992–93) was performed using a vertical piezometer drilled at the bottom of the first shaft of the HADES URL. The piezometer has several injection filters installed in the Boom Clay formation (Figs 1 & 7). Once the pore water pressures were in equilibrium with the *in-situ* pressure, gas was injected into one filter and the pressures at filters above the injection filter were monitored. Two different tests were performed using two different injection filters.

These tests have led to some important observations. One such observations was that the gas breakthrough occurred at 0.6 to 0.65 MPa, which was smaller than the predicted value of 1.25 MPa (based on the total stress at the location of the filter). At least one preferential pathway was created along the length of the piezometer. Gas eventually entered the HADES URL, which led to the test being halted. The pressure at which gas started to enter the HADES URL was relatively low. It is believed this was due to the damage induced by borehole drilling. However, it is expected that the convergence of the clay will minimize the risk of gas flow below the predicted total stress value.

MEGAS E5. The MEGAS E5 gas injection experiment (1994) was performed from the Test Drift (Fig. 1). Four horizontally oriented piezometers were installed in the Boom Clay around the HADES URL: one central piezometer for gas injection (Piezo A) and three monitoring piezometers (Piezo B, C and D) (Fig. 8). The injection pressure and the pore water pressures were monitored in 29 filters.

First, baseline parameters, such as pore water pressure, total stress and water permeability were determined. This was followed by injecting helium whose pressure was increased by 0.1 MPa per week until breakthrough occurred (Fig. 9). Gas inflow was observed in a neighbouring filter on the same piezometer as that with the injection filter. This showed that a preferential pathway had formed between the injection filter and this neighbouring filter, i.e. along a path where stresses are the lowest. Subsequently, some, but not all, of the surrounding filters showed increasing pore water pressures, which points to the hydromechanical coupling. The gas fluxes measured after breakthrough were of a similar magnitude to those found in lab tests. Observations from *in-situ* gas testing are in line with lab test conclusions. The formation of gas-induced pathways in Boom Clay depends on the applied hydromechanical boundary conditions and on the Boom Clay behaviour depending on material heterogeneities (Volckaert *et al.* 1995).

Gas injection experiments combining gas transport and self-sealing of Boom Clay. In 1998, an additional gas injection experiment was conducted using the MEGAS E5 experimental setup. Its goal was to examine the self-sealing capacity of the Boom

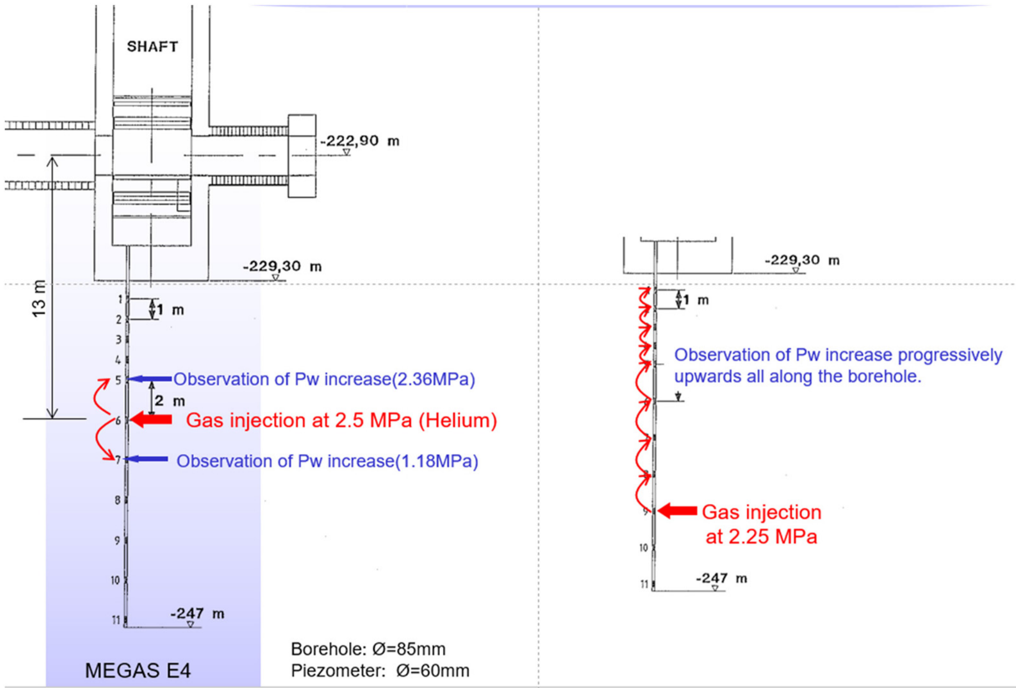

Fig. 7. Observations during the MEGAS E4 gas injection experiment. Source: modified from Volckaert *et al.* (1995).

Clay and to find out how the hydraulic and mechanical properties recovered after gas flow. Helium was injected between filters 13 and 14 at an overpressure of 0.3 MPa (relative to the pore water pressure). The gas flow was maintained for a year. Forty days after the end of the gas injection phase, a HTO migration test was performed by injecting HTO in filter 13. The measured activity concentrations were consistent with simulated predictions, indicating that the depressurized gas pathway does not represent a preferential flow path for gaseous radionuclides. Rodwell (2000) and Ortiz *et al.* (2002) suggested that this is due to the effective self-sealing capacity of the clay.

Other borehole and shaft sealing tests were performed at the HADES URL, as part of the EC RESEAL project (Van Geet *et al.* 2009). While the primary aim of this project was to demonstrate the sealing of boreholes and shafts in plastic clay using bentonite, gas transport through and around the seal was also tested.

The first such gas injection test (1999) used the setup which was installed during the BACCHUS 2 experiment. The details of this BACCHUS 2 setup

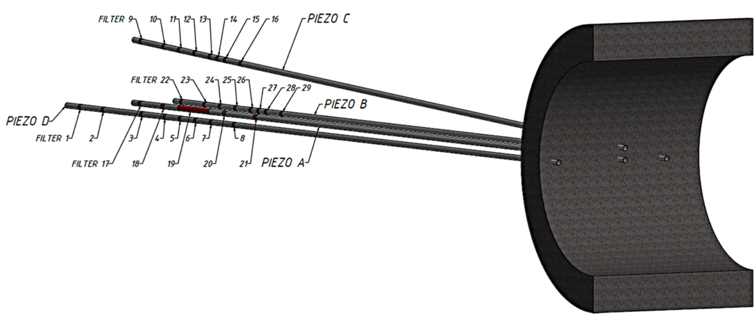

Fig. 8. Location of the different piezometers and their filters of the MEGAS E5 experiment.

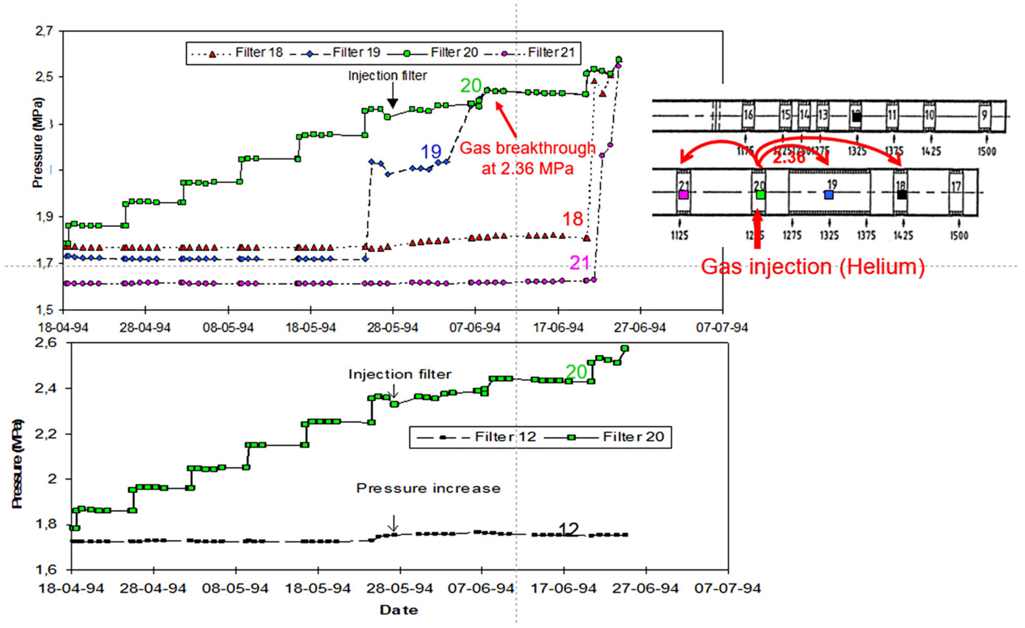

Fig. 9. Results of the MEGAS E5 gas injection experiment: pressure evolution in different filters. Source: modified from Volckaert *et al.* (1995).

have been described by Volckaert *et al.* (2000). The test setup was originally used to study the hydration and behaviour of a backfill consisting of a 50/50 pellet/powder mixture of Boom Clay. The test setup included a 50 cm wide borehole that was filled with the backfill mixture. The borehole also had a central filter tube (Fig. 10). This tube was used to perform the gas injection test in filter PW09C. Above and below this filter total stress sensors were installed.

After 30 days, gas breakthrough occurred at a pressure of 1.04 MPa. This was indicated by a sharp pressure drop in the injection system. Only the pore water pressure sensor PW06V (at a radial distance of 9.5 cm) showed a response to the gas breakthrough. The total stress sensors showed no significant response. This means that the gas flowed in a radial direction and not axially along the central tube. Had that been the case, the gas flow would have been detected by the neighbouring filters PW08C or PW10C. Hydraulic conductivity measurements were made in each filter in the backfill after the gas breakthrough. The authors stated that no significant changes in conductivity were observed, demonstrating the self-sealing capacity of the backfill.

The second gas injection test used a 2.6 m long piezometer that was installed in a 15 m deep borehole as part of a RESEAL borehole experiment (Fig. 11) (Van Geet *et al.* 2007). At the end of this piezometer were two 55 cm long sealing compartments. One consisted of compacted blocks of FoCa clay, the other of compacted blocks of Serrata clay. The total pressure could be measured by means of sensors at different locations in the piezometer. After the whole setup was saturated, gas was injected into filter PW1 and the injection pressure was increased stepwise.

Gas breakthrough occurred at 3.1 MPa. This corresponded to the total radial stress measured in the FoCa seal before gas injection. Upon breakthrough, the pressure suddenly decreased in filter PW1 and increased in filter PW5. Filters PW2, PW3 and PW4, all in the FoCa seal, showed a very weak response. This indicates that the gas did not flow through the FoCa seal, but along the interface between the seal and host rock or through the excavation-damaged zone of the host rock.

The third gas injection experiment was performed in a shaft seal that was installed as part of a RESEAL shaft seal experiment (Van Geet *et al.* 2009). The seal was installed in a small experimental shaft in the HADES URL (Fig. 12). Before the seal was placed in the shaft, the shaft lining was removed over a vertical length of 2.2 m. This section was then filled with a mixture of 50% of powder and 50% of highly compacted pellets of FoCa clay.

It took about 6 years to fully saturate the seal. After this saturation phase, gas was injected into filter PW-S-HLm-SW, a disc shaped filter close to the interface seal/host rock. The gas pressure was

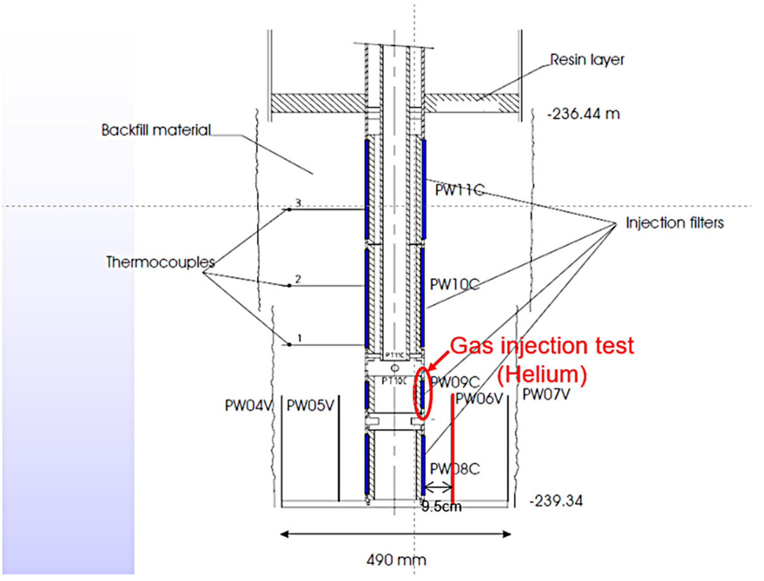

Fig. 10. Layout of the BACCHUS 2 experiment. Note that gas was injected at filter PW09C. Source: modified from Volckaert *et al.* (2000).

increased in steps of 0.1 MPa. Gas started to dissipate slightly at a pressure of 1.43 MPa and breakthrough occurred at 1.3 MPa. This shows that gas breakthrough does not only depend on the gas pressure, but also on time-related phenomena. The breakthrough could already have happened at 1.3 MPa if the system had been given sufficient time to respond.

Upon breakthrough, most filters showed a response, but the magnitude of this response was limited to 0.05 MPa. The total stress sensors showed a variation of 0.02 MPa, while the sensor on the western rod of the bottom instrumental level showed a 0.1 MPa decrease in total pressure. These observations showed that there was no direct connection between the injection filter and the other sensors. After breakthrough, the pressure decreased and stabilized at 0.83 MPa, which can be seen as the shut-in pressure.

One month after the first breakthrough, a second breakthrough test was conducted at the same location. This time gas started to dissipate at an injection pressure of 1.4 MPa. A few hours later breakthrough occurred at 1.28 MPa. In contrast to the first test, the pore water pressure and stress response in the other sensors were more pronounced. The pressure difference between the injection filter and the monitoring sensors remained, indicating that there was no direct connection between the two. Van Geet *et al.* (2009) concluded that gas breakthrough is likely to occur at the minimum total stress if the system is given sufficient time to respond.

Lessons learnt from the gas injection experiments. All these gas injection experiments – using the MEGAS E5, BACCHUS 2 and RESEAL test setups – provided valuable insight into gas transport

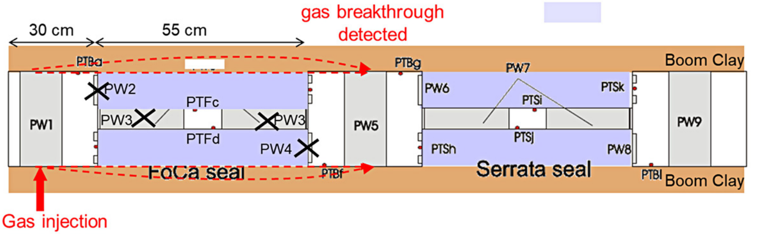

Fig. 11. Schematic view of the piezometer used in the RESEAL borehole experiment (PW, pore water pressure sensor; PTB, total pressure sensor in contact with Boom Clay; PTF, total pressure sensor in contact with FoCa clay; PTS, total pressure sensor in contact with Serrata clay). Source: modified from Van Geet *et al.* (2007).

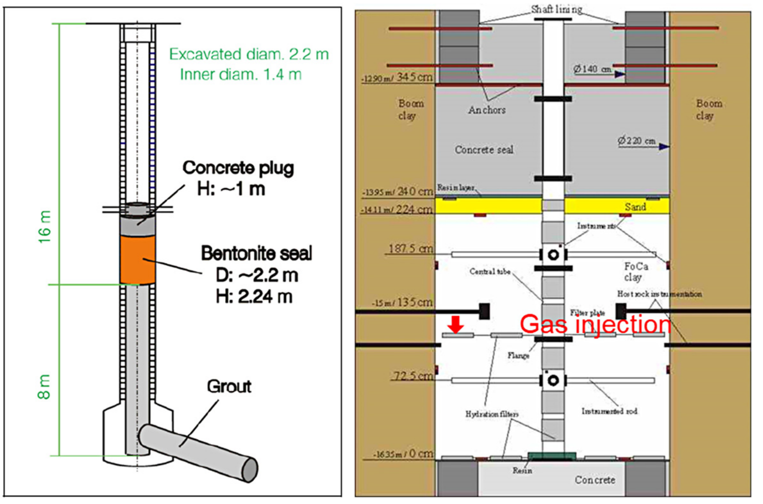

Fig. 12. Layout of the RESEAL II shaft sealing experiment. Source: modified from Van Geet *et al.* (2009).

through seals at a relevant scale. While in lab-scale experiments the gas flow flowed along interfaces (e.g. in Jacops *et al.* 2015*a*), this occurred only in the RESEAL borehole sealing experiment. With the test setups of the BACCHUS 2 and the RESEAL shaft sealing experiment, the gas flow was through the sealing material and no gas pathway was formed between the injection filter and any of the monitoring filters. In conclusion, gas will start to flow once the pressure reaches the local minimum effective stress. The direction of the flow strongly depends on the local stress situation.

The results of the gas injection experiment in the RESEAL shaft sealing test indicate that gas breakthrough also depends on time-related phenomena. This implies that, when gas pressure is increased too rapidly, the measured breakthrough pressure might be overestimated because the system is not given enough time to accommodate to the pressure and the measured breakthrough pressure is higher than it would be under repository conditions.

Main understanding on gas transport in Boom Clay

Over the past three decades, many lab and *in-situ* experiments have been conducted to study gas flow. The main outcomes have been discussed above. More details can be found in Levasseur *et al.* (2021).

The lab-scale experiments showed that for gas to flow into a saturated Boom Clay samples, a high gas entry pressure is required. As Boom Clay is a low strength material, it is prone to failure without desaturation when subject to high gas pressures (i.e. failure before reaching gas entry pressure). The experiments showed that the gas transport capacity depends more on the breakthrough pressure and the rate at which the pressure builds up than on the gas entry pressure. It was also observed that the samples deformed and increased in volume during the experiments. The volume increase is related to the locally increased gas transport capacity.

The *in-situ* experiments confirmed the coupling between the geomechanical and hydraulic properties of the Boom Clay. In all experiments, changes in the pressure with which gas is injected caused a hydraulic response in one or more monitoring filters. The MEGAS E4/E5 and the RESEAL borehole sealing experiment also confirmed the important role of interfaces in gas transport. In the MEGAS experiments, the gas flowed along the interface between the clay and the piezometer filter. In the RESEAL borehole sealing experiment, it was the interface between the clay and the FoCa seal that provided a pathway for gas transport.

In contrast, the BACCHUS 2 and RESEAL shaft sealing experiment showed no preferential gas flow along the interfaces. This may be due to the differences in the local minimum effective stress between the different experimental setups. For gas to flow, the injection pressure must be higher than the local minimum effective stress. This stress strongly depends on local conditions and therefore the pressure at which gas flows, will differ depending on the experimental setup.

It will be important to recognize the role the interfaces can play in gas transport when designing *in-situ* gas experiments in the future. To prevent gas flow along an interface, it is important that the *in-situ* stress field around filters is restored after it has been disturbed by borehole drilling and piezometer installation. This means that the gas injection must take place sufficiently long after the installation of the test setup to allow the host rock to recover.

Various processes and phenomena play a role in the transport of gas in Boom Clay. This can complicate comparing the outcomes of different experiments and sometimes different hypotheses can be formulated to explain the observations. This can make it difficult to reproduce the experiments, which in turn increases the experimental uncertainties. Furthermore, the results obtained in the lab need to be tested or validated under conditions that are representative of the conditions prevailing in a DGR, such as the *in-situ* stress fields in the host rock around a DGR. Scaling up the observations and knowledge gained from lab-scale tests to a scale that is representative of a DGR remains a challenge. Nevertheless, SCK CEN and ONDRAF/NIRAS are continuing their efforts to improve understanding of the diffusive transport of dissolved gas and investigate the impact of gas generation on the Boom Clay and the engineered barriers.

SCK CEN has extensively studied gas diffusion in saturated porous media in the lab resulting in a large database of gas diffusion coefficients in Boom Clay (Jacops *et al.* 2013, 2015*b*, 2016, 2017*a*, *b*, 2020*a*, *b*; Jacops 2018). These lab tests were performed at a small scale (centimetre scale). To validate the data obtained in the lab, ONDRAF/NIRAS and SCK CEN (with the support of EIG EURIDICE who manages and operates HADES URL) will conduct an *in-situ* diffusion experiment in the HADES URL: NEMESIS (Neon diffusion in MEGAS *In Situ*).

The NEMESIS experiment in the HADES URL

Objective and setup

The NEMESIS experiment aims to determine parameters associated with the diffusion of dissolved gas in Boom Clay. In addition to the laboratory programme, this *in-situ* experiment will allow current knowledge relating to diffusion of dissolved gases at a larger scale to be confirmed and/or improved.

Previous *in-situ* gas injection experiments showed the importance of limiting the disturbed stress field for transport experiments. Therefore, it has been decided to re-use the MEGAS E5 piezometers drilled in the Test Drift in 1992. This consists of four horizontally oriented piezometers in a 3D configuration (Figs 8 & 13). Each piezometer is 15 m deep and equipped with several filters. As detailed in Volckaert *et al.* (1995) and in Ortiz *et al.* (2002), this setup was used in 1994 to perform a gas breakthrough experiment by injecting gas (helium) in filter 20. Upon breakthrough, the gas reached filters 18, 19 and 21 (Fig. 8) (equal pressure in all filters), but only a small pressure response was observed in some filters of all three other piezometers. In 1998, a new gas breakthrough experiment was performed in filters 13 and 14 (Fig. 8), followed by a tracer diffusion experiment in which HTO (tritiated water) was injected in filter 13 and monitored in the surrounding filters. To avoid interaction with the previous experiments performed when using the MEGAS piezometers, another tracer gas, neon, will be used for the NEMESIS experiment. The neon will be dissolved in water and injected through

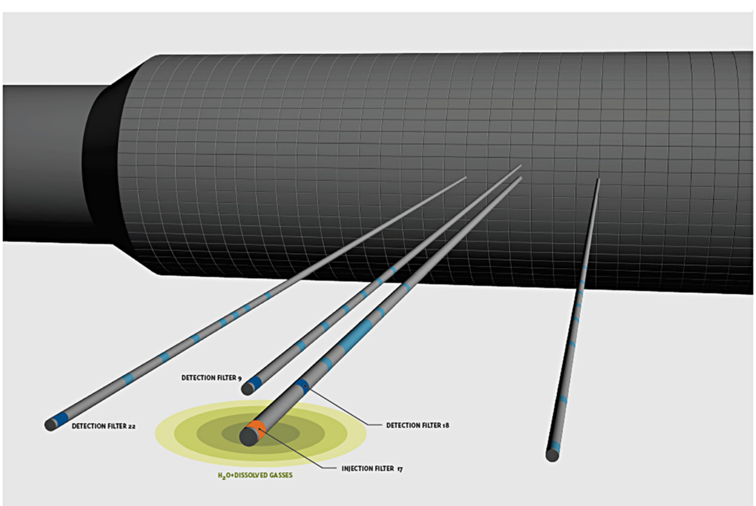

Fig. 13. 3D view of the neon diffusion in the NEMESIS experiment.

the Boom Clay from filter 17. Three neighbouring filters located in horizontal and vertical planes will serve as gas monitoring filters by measuring how the gas concentration evolves over time (filters 9, 18 and 22, Fig. 13). This configuration will allow the investigation of the diffusion of neon in three dimensions and determine the anisotropic diffusion coefficients of dissolved neon. Neon is selected because it is (1) considered as a good proxy for H_2 (the main gas generated in a GDR) (Jacops *et al.* 2017*b*) and (2) not naturally present in Boom Clay.

The NEMESIS setup to measure diffusion *in situ* will be a modification of the setup that has been used to measure diffusion of gases in the lab, which is described in detail in the previous section and in Jacops *et al.* (2013). This setup will consist of one injection circuit (source – connected to filter 17) and three monitoring circuits (target 1, 2 and 3 – connected to respectively filters 9, 18 and 22) (Fig. 14). The water of each filter will enter the vessel at the top and will be pumped back into the filter from the bottom of the vessel. Several sensors will monitor the gas pressure, water level, water flow and temperature of the system. The top part of each vessel will be connected to a CGC4 (Compact Gas Chromatograph 4, Interscience, The Netherlands) gas analyser. The gas analyser will be equipped with a multi-position valve allowing automated sampling from the different vessels.

Estimate of hydraulic conditions prior to the experiment

The NEMESIS piezometers have been impacted by the gas breakthrough tests performed during the MEGAS E5 experiment in 1994 (Ortiz *et al.* 1997) and 1998 (Ortiz *et al.* 2002) and by an HTO migration test that has been running in the MEGAS E5 setup since 1998 (Aertsens 2013). In order to check the impact on the hydraulic conductivity of the surrounding Boom Clay, and to know the initial hydraulic conditions of the NEMESIS experiment, *in-situ* permeability tests were performed on the source and target filters (i.e. respectively filters 17, 9, 22 and 18) and four of their nearby filters (i.e. filters 1, 8, 10 and 20, see Fig. 8). These nearby filters were selected taking into account that the Boom Clay around these filters will be the main influence zone for the gas diffusion test. The HTO contamination level of the pore water around these filters is low and does not affect the permeability tests. Although it is necessary to measure the permeability around the other nearby filter 23, a permeability test could

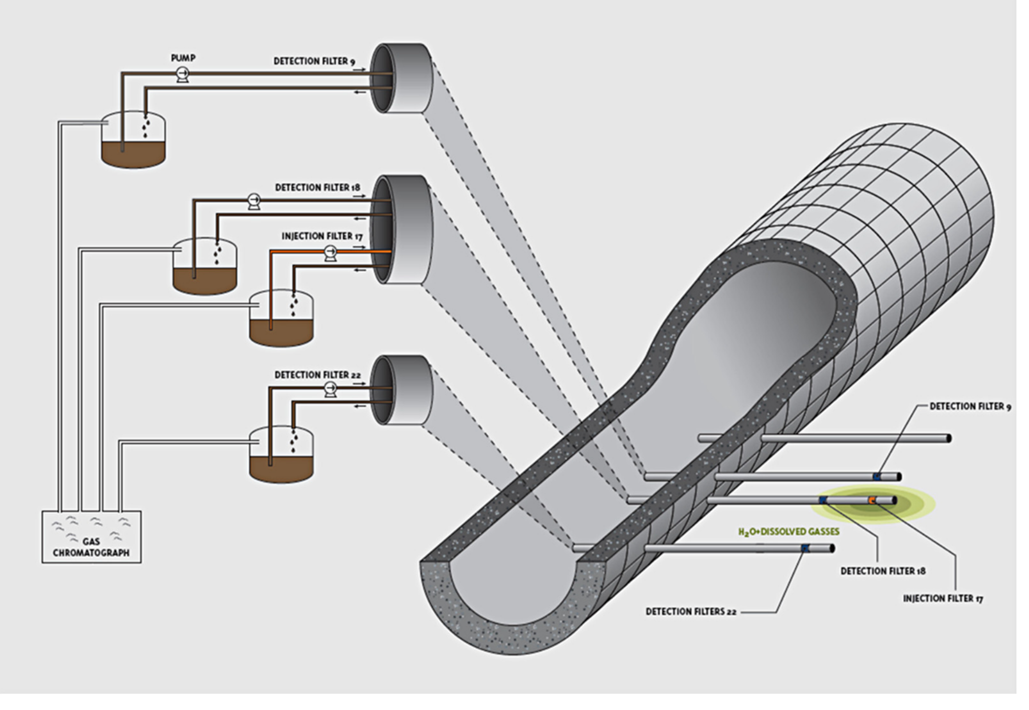

Fig. 14. Schematic view of the NEMESIS experimental setup.

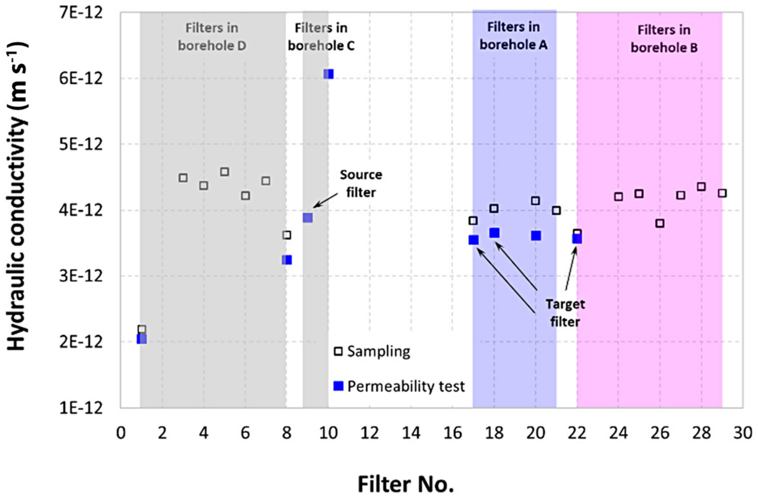

Fig. 15. Hydraulic conductivity estimated from HTO sampling campaign and permeability tests.

not be conducted because the tubes connected to this filter were blocked. In addition, an estimation of the Boom Clay hydraulic conductivity around each filter of the MEGAS setup was made by analysing the outflow rates measured during a HTO sampling campaign performed in 2019.

Estimates of the Boom Clay hydraulic conductivity around all these filters are presented in Figure 15. Except for filters 1 and 10 (Fig. 8), which could not yet be interpreted, the hydraulic conductivity measured at all other filters is within a narrow range of 3.2×10^{-12} to 4.6×10^{-12} m s^{-1}. The hydraulic conductivities estimated from the HTO sampling campaign are consistent with the results of permeability tests. The hydraulic conductivity at the four NEMESIS injection and monitoring filters is within an even narrower range, from 3.5×10^{-12} to 3.9×10^{-12} m s^{-1}. It demonstrates the homogeneity of the initial hydraulic conditions near the NEMESIS setup.

Scoping calculations

The NEMESIS experiment complements the long-term tracer diffusion test carried out since 1998 from filters of the MEGAS E5 piezometers (Fig. 8) (Ortiz *et al.* 2002; Aertsens 2013). A total amount of 7.61×10^{8} Bq of tritiated water (HTO) was injected from filter 13 and since then the HTO concentrations at all the filters on piezometer C (Fig. 8) have been regularly monitored. To build and validate the numerical model used to optimize the design of the NEMESIS experiment, the long-term tracer diffusion test is first analysed.

Model validation through the long-term HTO migration test. The HTO migration test was simulated by Ortiz *et al.* (2002) using diffusion of a point source in an infinite domain. This modelling, based on the first four years of diffusion, is hereafter referred to as ‘previous modelling’. This paper reassesses the HTO migration test with the objective to:

- extend the diffusion data to a measurement period longer than 20 years, including more accurate boundary conditions in 3D configurations;
- screen the possible HTO contamination levels of the filters to support the design of the new *in-situ* gas diffusion experiment; and
- validate the model used for interpreting the future gas diffusion data.

The HTO migration test is modelled in this paper using a 3D COMSOL advection–diffusion module with the Boom Clay (BC) block centred around injection filter 13. The domain is discretized into a total of 114 171 quadratic tetrahedral elements. The excavation of the first HADES URL galleries in 1982 is taken as the start of the drainage period. In 1998, after 16 years of drainage and when the HTO migration experiment started, a large drainage field of the order of tens of metres was formed around the MEGAS piezometer network. Initially, diffusion of HTO occurred only within a limited local region around injection filter 13. For such a flow-diffusion coupling problem, a sequential workflow is adopted to enhance the numerical calculation efficiency, with a larger domain for the hydraulic modelling (a block of 30 × 30 × 20 m) and a local domain for the diffusion modelling (a block of 10 × 10 × 10 m). Hydraulic modelling results indicate that the water flow reaches steady state after 16 years of drainage with an order of 10^{-4} m a^{-1}. The steady-state water velocity field is extracted from

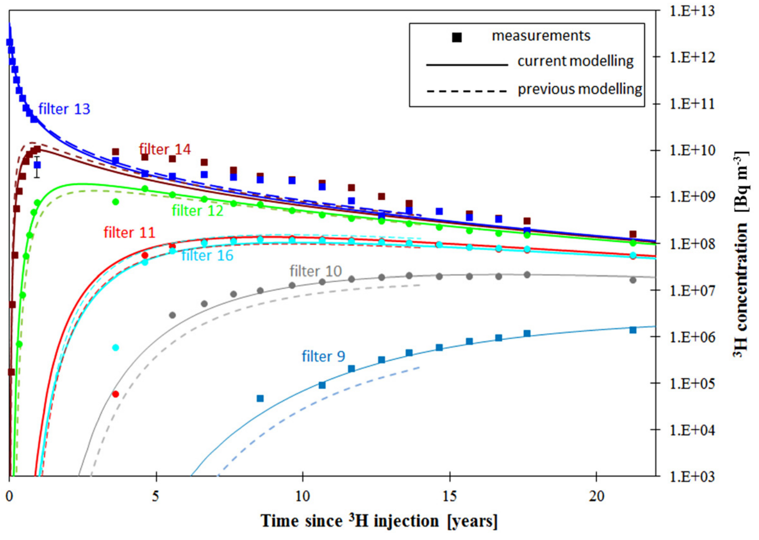

Fig. 16. Comparison between modelling results and field measurements at filters along piezometer C of the MEGAS E5 experiment.

the hydraulic modelling and serves as input in the subsequent diffusion modelling. In order to mimic the field measurements, dilution of the tracer in the filter dead volume (casing and tube) is considered. The dead volume is the total volume of the inner part of the filter and the tubing connecting this filter to the NEMESIS setup. In the modelling, filters are treated as porous cylinders with an equivalent porosity calculated from the dead volume. The external surface of the other parts of the piezometers is considered impermeable.

Figure 16 presents comparisons between modelling results and field measurements at filters along piezometer C (Fig. 8), ($D_{eff,H} = 1.6 \times 10^{-10}$ m^2 s^{-1} and $D_{eff,V} = 0.8 \times 10^{-10}$ m^2 s^{-1}), together with previous modelling results. The results show that the 3D modelling approach with boundary conditions more representative of *in-situ* conditions significantly improves the simulation outcomes. Although the comparison is good for most filters, it is clear that the measurements at injection filter 13 and its adjacent filter 14 are higher than the modelling results during years 3–14. Both filters have similar HTO concentrations, suggesting they are linked. This is presumably related to the foregoing gas breakthrough test of 1998, which is described by Ortiz *et al.* (2002).

After the first year, monthly sampling was stopped and when the sampling was resumed after 2.5 years, the HTO concentrations in filter 14 had increased more than expected, with HTO concentrations that were even higher than those measured at filter 13 until year 9. The higher concentration at filter 14 than at filter 13 can only be explained by some advective transport mechanism. During the sampling, it was observed that there were quite high initial flow rates when opening filter 14. This could indicate excessive gas pressures caused by occluded gas bubbles. This may be of interest for further re-interpretation of the gas breakthrough experiment carried out during the EC MEGAS project.

Numerical predictions of neon diffusion. A similar numerical tool as the one developed to analyse the HTO migration test is now used to assess the NEMESIS *in-situ* gas diffusion test and predict the amount of gas diffusing into the three monitoring filters. In the simulations, it is assumed that the external surface of injection filter 17 has a constant concentration of dissolved neon under a pressure of 1.58 MPa (measured *in situ* in October 2018). In addition, monitoring filters 9, 18 and 22 are considered as zero concentration boundaries. The other piezometers are considered impermeable. Anisotropic effective diffusion coefficients of the dissolved neon are used in the Boom Clay: $D_{eff,h} = 2.29 \times 10^{-10}$ (m^2 s^{-1}) and $D_{eff,h} = 1.75 \times 10^{-10}$ (m^2 s^{-1}) (Jacops *et al.* 2017*b*).

In order to get a suitable gas diffusion domain size, cubes with various side lengths of 2, 5 and 10 m were tested around injection filter 17. Numerical simulations show that results from the 5 m-cube are similar to those from the 10 m-cube. Therefore, a clay cube of 5 × 5 × 5 m around filter 17 is used for gas diffusion modelling. A total of 985 825 quadratic tetrahedral elements are used.

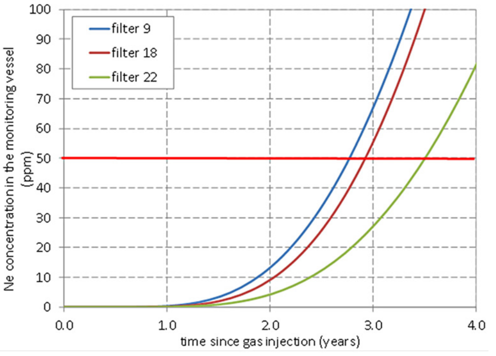

Fig. 17. Neon concentration in the gas phase of the monitoring vessel (in ppm) to indicate when the detection limit of 50 ppm is passed considering a total gas pressure of 1.58 MPa in the monitoring vessel.

Cumulative dissolved gas collected at neighbouring filters shows that the three filters closest to injection filter 17 are suitable as monitoring filters: filter 18 and 22 with horizontal distances of respectively 0.66 m (along the piezometer) and 0.71 m (parallel to the gallery) and filter 9 with the shortest vertical distance of 0.54 m. Knowing the total amount of dissolved gas entering each monitoring filter, the partial gas pressure of neon (MPa) and concentration of neon (ppm) in the gas phase can be calculated based on the equilibrium in the monitoring vessel, as shown in Figure 17. Taking into account a detection limit of 50 ppm, neon is estimated to be detectable after approximately 3 years.

Conclusions

Over the last 30 years, several experiments have been conducted to study gas transport in Boom Clay at lab scale and in the HADES URL. The lab-scale experiments showed that high gas entry pressures are required for gas to flow in a saturated clay sample. Gas flows when the injection pressure is higher than the local minimum effective stress. Once gas breakthrough occurs, volumetric deformation of the sample is observed, and correlates with the local enhancement of gas transport capacity. This coupling between geomechanical and hydraulic properties of the Boom Clay is confirmed by *in-situ* experiments. Borehole sealing experiments also pointed to the important role of interfaces in gas transport process.

The overview presented in this paper shows that various processes and phenomena play a role in the transport of gas in Boom Clay. In addition to consolidating these findings, the output of lab tests needs to be transposed to the conditions prevailing in a DGR. The transition from the laboratory to the *in-situ* scale is still challenging. It is the ambition of SCK CEN and ONDRAF/NIRAS to address these issues for Boom Clay, starting with the transport of dissolved gas by diffusion.

Over the last 10 years, SCK CEN obtained a large set of gas diffusion coefficients in Boom Clay from small-scale lab experiments (centimetre scale). In order to evaluate whether these results obtained from small-scale experiments can be used to inform *in-situ*/larger-scale experiments, an *in-situ* diffusion experiment with dissolved gas will be performed in the HADES URL using the same principle as the lab-scale tests. This experiment will use the existing MEGAS E5 boreholes, which were drilled and used in the 1990s for several gas injection experiments. 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. According to scoping calculations performed during the preparation phase, neon will be detected by the monitoring filters after about three years. By reusing the MEGAS E5 boreholes, the NEMESIS experiment will continue to provide new data for estimating *in-situ* gas diffusion properties for the next five years.

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Competing interests The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper, except from the agencies to which the authors are affiliated.

Author contributions **EJ**: conceptualization (lead), methodology (lead), writing – original draft (lead); **LY**: investigation (equal), writing – original draft (equal); **GC**: investigation (equal), writing – original draft (equal); **SL**: project administration (lead), resources (lead), writing – review & editing (lead).

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Data availability The data that support the findings of this study are available from SCK CEN and ONDRAF/NIRAS but restrictions apply to the availability of these data, which were used under licence for the current study, and so are not publicly available. Data are, however, available from the authors upon reasonable request and with permission of SCK CEN and ONDRAF/NIRAS.

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