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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

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Abstract. 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.

1 Introduction

Deep geological repositories for the disposal of radioactive waste generally rely on a multi-barrier system to contain and isolate the waste from the biosphere. This multi-barrier system typically comprises the natural geological barrier provided by the repository host rock and its surroundings and an engineered barrier system (EBS). The EBS represents the man-made, engineered materials placed within a repository, including, among others and depending on the disposal concept, waste canisters, buffer materials, backfill, concrete lining and seals. This multi-barrier principle creates the overall robustness of the system that enhances confidence that the waste will be successfully contained, as the natural barrier provides a stable environment that allows the EBS to function for hundreds to thousands of years, depending on the disposal concept. Moreover, the geological barrier itself can contribute to the confinement of radionuclides that would eventually be released after the degradation of the engineered barriers. Owing to their excellent properties for the confinement of contaminants, including very low permeability, high sorption capacity and swelling/self-sealing capacity, clay-based materials are often considered for use as part of the engineered barriers in about all repository designs under development. Clay formations are also considered as potential hosts for geological disposal in several European countries.

During the lifetime of the repository, clay barriers will be exposed to repository-induced perturbations, which result 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.

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Considerable amounts of gas can be generated in a repository containing radioactive waste. The largest fraction of the gas is expected to be hydrogen produced by the anaerobic corrosion of steel and reactive metals in the waste, their packaging, and the EBS. The degradation of organics and radiolysis also produce gas. Even though the gas production processes are generally slow, it is important to verify that these will not be detrimental to the good functioning of the disposal system. The low permeability of clays that is favourable with respect to the containment function of a repository also limits the evacuation of the generated gas. It is possible that gas could be generated at a faster rate than it can be removed through the engineered barrier components and the host rock by diffusion of dissolved gas, resulting in the development of a pressurised gas phase within the repository and its surrounding. The accumulated gas could then escape from the repository by creating discrete, gas-specific pathways (e.g., fracturing or pathway dilation) through the EBS and/or the host rock. In addition, the potential release to the biosphere of gas containing volatile radionuclides is also an issue that needs to be considered.

The heat produced by radioactive decay in the waste matrix will be transported, mainly by thermal conduction, to the bedrock surrounding the repository. In the beginning, the heat production is high, and therefore most concepts limit maximum disposal container surface temperatures to 100°C to protect clay-based buffer, backfill and host rock materials from undesirable evolution; the temperature gradients will also be high. When temperature increases in a low permeable media like clay host formations, the pore water is pressurised because of the difference between its thermal expansion coefficient and that of the solid skeleton of the clay. In the near field characterised by an excavation damaged zone, this could induce fracture re-opening or propagation, thus increasing the permeability. In the far field, this could induce damage and reactivate fractures/faults. On the other hand, the increased temperature in buffer bentonite may result in changes in buffer material, strong evaporation near the heater and vapour movement towards the external part of the buffer. Accepting higher temperature limits could have significant advantages, such as allowing the disposal of higher enrichment/burn-up spent fuels, shorter interim storage/cooling requirements, easier (re)packaging of the waste and a reduced disposal facility footprint, and so needs to be considered.

The WP HITEC and WP GAS of the European Joint programme EURAD aim to increase the understanding and predictability of the impact on clay barriers of the fundamental processes and their couplings associated with the thermal and gas perturbations. In this paper, the raisons d'être of these two WPs are first presented. It is then shown how the integration of experimental and modelling approaches can provide building blocks that can be used in multiple national programmes to support the conceptualisations of gas and heat transport through clay barriers and evaluate the robustness of the proposed repository concepts.

2 The GAS work package

2.1 Work package's raison d'être

Work Package 6 of the EURAD European Joint Programme 'Mechanistic understanding of gas transport in clayey materials' (WP GAS) focuses on gas transport in natural and engineered clayey materials. It aims:

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

The first raison d'être of this WP is to provide results that apply to a wide range of national programmes in clayey host rock. This is possible because the results of previous efforts on the identification and characterisation of the possible gas transport processes suggest that the mechanisms at play in different clays are generally similar, while the conditions (gas pressure, stresses/deformations, saturations, ...) for the transition from one transport regime (diffusion, two-phase flow, pathway dilation and fracturing) to another strongly depend on the specific properties of a given clayey material.

The second raison d'être of this WP is to transfer knowledge gained from laboratory and in situ experiments to configurations that are commonly found in current repository designs in clays to address key questions from the end-users:

- how could gas be transported throughout the clayey disposal system, and which water-soluble and volatile radionuclide transport could be associated with it?
- how and to what extent could the hydro-mechanical perturbations induced by gas affect clayey barrier integrity and long-term repository performance in clayey host formations?

This WP builds strongly on the return of experience, results and conclusions from the past FORGE EC project [27]. The experimental investigation of gas transport in clayey materials in FORGE revealed complex mechanisms, e.g., the development of discrete, unstable pathways controlled by the mechanical behaviour of the porous media. However, it was also suggested that this complexity could be addressed if one can bound the effects of these mechanisms using simpler and robust descriptions for evaluation purposes. The WP GAS of EURAD aims then at increasing the confidence in the overall understanding of gas behaviour in clayey materials gained from FORGE and improving its integration in the conceptualisation process for the different clayey components of a disposal system. This should, in turn, support and justify the use of robust evaluation approaches and confirm the expert judgment at the end of FORGE that gas is not a showstopper for geological disposal but a question of managing uncertainties [22].

2.2 Key objectives and work programme

The Work Package GAS aims to increase the understanding and predictability of gas transport in different clayey host rocks (Boom Clay, Callovo-Oxfordian claystone and Opalinus Clay) and clayey engineered barriers (MX-80, BCV and FEBEX bentonites) of geological disposal systems. It also aims to support the stepwise integration of the knowledge on gas transport and its effects in conceptualisations of the functioning of a disposal system in support of the safety case.

To reach these objectives, the work programme of the WP is defined through three scientific tasks: a first dealing with the 'transport mechanisms' of gas, a second dealing with the 'Barrier integrity' and a last dealing with 'Repository performance aspects'.

The WP GAS produces new data and develops new process-level models to improve mechanistic understanding of transport processes in natural and engineered clayey materials, including couplings with mechanical behaviour and impact on the material properties. Experimental work that is carried out determines, for each identified gas transport regime, the conditions under which that regime is possible in clay materials representative of host formation and clayey EBS components. Data obtained are pertinent from low (diffusion) to high (advection) gas transport rates.

This WP also illustrates how knowledge gained from laboratory and in situ experiments can be integrated into the conceptualisation of gas transport through different clayey components of a disposal system and how gas could affect (or not) the performance of the system. This work carried out involves (i) the development of phenomenological descriptions of gas transport and of its likely consequences at the relevant scale and (ii) the testing of different approaches to represent the effects of gas at the repository scale and bounding its consequences in terms of repository performance.

3 The HITEC work package

3.1 Work package's raison d'être

The Work Package 7 of the EURAD European Joint Programme 'Influence of Temperature on Clay-based Material Behaviour' (WP HITEC) aims to develop an improved thermo-hydro-mechanical (THM) understanding of clay-based materials (host rocks and bentonite buffers) exposed at high temperatures (>100°C) or having experienced high-temperature transients for extended durations. The WP's raison d'être is to evaluate whether or not elevated temperature limits (up to 150°C) are feasible for various geological disposal concepts for high heat generating wastes (HHGW). HITEC studies clay host rock formations, document and establish the possible extent of elevated temperature damage in the near and far field (e.g., from over-pressurisation of water due to temperature increase) and also indicates the likely consequences of any such damage. The WP also looks at bentonite buffers and determines the temperature influence on buffer swelling pressure, hydraulic conductivity, erosion, or transport properties and see where the buffer safety functions start to be unacceptably impaired.

For the disposal of HHGW, it is important to understand the consequences of the heat produced (temperature and temperature gradients) on the properties and long-term performance of the natural and engineered clay barriers. Most safety cases for disposal concepts that involve clay currently consider a temperature limit of 100°C [33]. Being able to tolerate higher temperatures while still ensuring an appropriate performance would have significant advantages, e.g., shorter above-ground cooling times, more efficient packaging, fewer disposal containers, fewer transport operations, and smaller facility footprints. This WP has the potential to effectively integrate with the parallel SFC R&D WP (i.e., interrogate the validity of the currently applied thermal limits and also the importance of the accuracy of the assumed radiological waste properties) and consequently is the first step toward optimisation of the design architecture of the deep geological disposal.

3.2 Key objectives and work programme

The overall objective of HITEC is to evaluate whether an increase in temperature is feasible and safe by applying existing and work package produced novel knowledge about the behaviour of clay materials at elevated temperatures:

- to improve understanding of the THM behaviour of clay rock and engineered clay material (buffer) under high temperature and provide suitable THM models both for clay host rock and buffer,
- to better assess the effect of water overpressures, build-up induced by the heat produced from the radioactive waste on the THM behaviour and properties of the clay host rock, and
- to identify processes at high temperature and the impact of high temperature on the THM properties of the bentonite buffer material.

In the host clay formation task, the aim is to deploy new knowledge of the mechanics of clay in order to better evaluate and model possible damage evolution during the temperature transient phase and better assess the consequences of possible damage. This includes experiments, model development and model benchmarks.

In the buffer bentonite task, the aim is to deploy new knowledge on the mechanical behaviour of the buffer at high temperatures. The temperature impact on important processes is measured either after a high-temperature exposure or while the clay is at a high temperature. Processes that may have a temperature dependence are swelling pressure, hydraulic conductivity, erosion properties and transport of solutes.

Finally, HITEC aims to document all the above to be utilised in Safety Cases studies.

4 Conceptualisation of gas and heat transport trough repository in clayey host formations as considered in Europe

From past evaluations, it is known that gas generated by corrosion and/or radiolysis in large quantities may result in the development of a gas phase within the existing porosity of the EBS, within the excavation damaged zone (EDZ) and, to some extent, within the host formation. Experimental evidence suggests that discrete, transient, gas-specific pathways may also, or alternatively, form through (or between) EBS materials, the EDZ and the clayey host formation in the form of subcritical (pathway dilation) or supercritical (fracturing) cracks. The transient nature of such phenomena is to be emphasised.

Desaturation of the existing porosity because of gas invasion can have a significant effect on soluble radionuclide migration: it may limit the extent of diffusion of soluble radionuclides but may also result in advective transport of radionuclides if groundwater is displaced one way or another by gas as a consequence of pressurisation or suction. High levels of desaturation may even affect the gas source term by decreasing the availability of water for gas production processes. Continuous gas-specific pathways, possibly evolving and unstable, may form from the deposition zones to the repository access. Even though most of the gas flowing through these pathways would be inactive, it could also carry volatile radionuclides. Finally, high gas pressures may possibly result in mechanical damage to the clayey engineered and natural barriers, including the host formation. It is important to assess if this could occur in practice and if such damage would be transient only or would have a lasting effect and how this would affect (or not) the global functioning of the repository.

Most high-level waste types, spent fuel or reprocessed fuel, produce heat during the early post-closure phase of the repository life. The heat production typically decreases quickly so that the highest temperatures occur in the nearby field only decades after closing the (part) of the repository. The highest temperature in most concepts is limited to about 100°C at the surface of waste canisters. This temperature requirement has a big impact on the dimensioning of the repository and, therefore, not only directly affects the size and costs of the repository but also sets requirements for interim storage before final disposal. Therefore, accepting higher temperatures has many benefits, e.g., shorter above-ground cooling times, more efficient packaging, fewer disposal containers, fewer transport operations, and smaller facility footprints. However, applying higher temperatures even during the short period of time at the beginning of final disposal might have detrimental effects on the material properties of EBS and/or change the mechanical behaviour of those materials.

To guarantee that the migration of radionuclides will be delayed as long as possible for each type of waste, no alteration of the properties of clay host rock or clayey materials of the engineering barrier has to be induced by either the gas production & evacuation or by the temperature increase. To that goal, most concepts adapt the general architecture of the repository to:

i. have a sufficient expansion volume available in the porosity of the EBS, buffer & backfill for the gases generated after closure and a continuous release and evacuation through dissolution and diffusion or along existing pathways through the EBS, EDZ or interfaces by visco-capillary two-phase flow;
ii. limit temperature increase and its impact on the EBS in the near & far field host rock by adjusting the spacing between disposal packages and between galleries, respectively.

The current conceptualisations of gas and heat transport through repositories as considered in Europe are detailed in [17] and [32], respectively by the radioactive waste management organisations (WMO) and the regulatory technical safety organisations (TSO) working with the clayey host formations and/or bentonites considered in the WPs GAS and HITEC. Communication of these conceptualisations developed in several national programmes is based on 'storyboards'. The storyboard elements are then compared to highlight the commonalities and explain the rationale for differences between the repository development approaches and the expected evolutions considered by the end-users within their respective national programmes in relation to gas and heat generation and transport. All these aspects are summarised in the following.

4.1 Comparison of repository design approaches

Unlike in crystalline and halite host rocks, there is still no geological disposal facility in operation or even in construction in a clayey host rock in Europe. For the moment, only France has selected a site in a clayey host rock, so it has been possible for Andra to define a complete repository concept for that site [2]. National programmes of other countries considering clay host rocks are at various stages of searching for a site and/or pre-design of a repository concept (e.g., [25,31]). Nevertheless, all those countries are targeting clay layers at a depth of a few hundred metres in their search for a site, and all participating WMOs are planning single-level disposal with separate zones for high-level waste disposal and long-lived intermediate-level waste disposal.

For all participating end-users, the concepts are different for high-level waste and long-lived intermediate-level waste. These differences relate mainly to (i) the geometry of the primary waste packages, (ii) the management of the thermal phase for high-level waste and (iii) the need to delay the migration of radionuclides as long as possible for each type of waste [17].

All participating WMOs include in their repository concept for high-level waste a disposal overpack made of steel. To delay the release and attenuate the migration of radionuclides out of the waste, two main approaches are envisaged:

- placing the steel overpack in a highly alkaline environment for as long as possible to slow down generalised corrosion and to guarantee the longest possible tightness of the overpack. The counterpart is that once the

tightness is lost, in the case of disposal of vitrified high-level waste, the glass containing the waste may degrade more rapidly if the alkalinity of the water is still elevated and release all the radionuclides over a relatively short period of time.
- Guarantee tightness only during the operating phase (possible safe handling of the waste packages, includes reversibility/retrievability period) and maintain the pH of the water around the packages at values that guarantee, in case of disposal of vitrified high-level waste, slow degradation of the glass once the tightness of the package is lost. The counterpart is that these low-alkaline waters imply faster corrosion of the steel of the packages and thus an earlier loss of tightness.

As far as gas generation is concerned, the first approach rather implies a lower gas source term (low corrosion rate) but longer in time, whereas the second approach implies a higher gas source term (higher corrosion rate) but of shorter duration.

Regarding long-lived intermediate-level waste, the primary waste packages that have to be dealt with in all national programmes generally have a wider variety of geometries (e.g., [23]) than primary packages of high-level waste, which are generally cylinders of about a few decimetres in diameter and between one and a few metres in length (e.g., [24]). In contrast, for long-lived intermediate-level waste, the primary packages may be drums of more diverse sizes. More than this, for high-level waste, considerations like criticality and thermal criteria have to be considered. As no damage to the clay host formation should be caused by thermal loading, it results in:

- high-level waste disposal cells are generally smaller in diameter, and the waste disposal packages are disposed of horizontally with only one package per cell section;
- long-lived intermediate-level disposal cells are generally larger in diameter, and waste disposal packages are disposed of either horizontally or vertically, with several waste packages per cell section, the optimal arrangement being partly driven by cost consideration.

In addition to the above elements that are important for the general architecture of the repository and, therefore, the expansion volumes available for the gases generated after closure, the following points related to the repository concepts are directly related to the generation of post-closure gases and are common to all programmes:

- high-level waste overpacks are made of steel, which represents an important source of hydrogen generation by anoxic corrosion.
- The lining of the galleries includes metallic elements, mainly reinforcing bolts and/or concrete reinforcement.
- Some long-lived intermediate-level waste contains (mainly or partially) metallic elements.
- Some long-lived intermediate-level waste (e.g., organic waste) may produce significant quantities of gas by radiolysis and/or (bio-)chemical degradation processes.

The sealing system is also an important component concerning gas migration after repository closure. For most participating WMOs and TSOs, it is dimensioned (usually by the definition of a water permeability below a certain value) to limit the flow of water containing dissolved radionuclides but with an induced effect on the transport of gases which are also fluids and for which it may represent a bottleneck for gas transport across the system. Therefore, some designs take explicitly into account the gas component in the dimensioning and choice of materials of the components of the sealing system [21].

4.2 Common phenomenological representations of the hydraulic-gas transient and the THM processes in clays

Regarding gas production mechanisms, other gases than hydrogen are produced in the repository, but the latter represents the main source term and evaluations are conducted assuming that only this gas is produced (except possibly in some sensitivity analyses).

The main mechanism producing hydrogen after the closure of the repository is corrosion of metals (principally steel) under anoxic conditions [7,26]. Radiolysis of organic matter and/or water is a secondary mechanism but not a negligible source term of hydrogen. Other production mechanisms (e.g., alpha decay which produces helium, or bacterial activity, which may produce additional gas or convert part of the hydrogen into methane) are studied but currently neglected in practice (except possibly in some sensitivity analyses). Gas consumption processes (like bacterial activity or pyrite oxidation) have been addressed in generic studies for gas-related repository optimisation, indicating a certain potential for reducing the build-up of gas overpressures in the backfilled repository structures [15]. However, due to the complexity of the associated microbiological processes, gas consumption is generally neglected for the sake of robustness of the assessment (except possibly in some sensitivity analyses).

The main mechanisms of transport of hydrogen at the repository scale (including the host rock) are advection for the part expressed in gaseous form and diffusion for dissolved gases. As detailed in [17], the main mathematical formulation used for advection is the Darcy formulation generalised to the water-gas two-phase flow. The relative permeability and retention curves are generally represented by van Genuchten/Mualem type relationships [20,30], but sensitivity analyses with other relationships are common as summarised in [17]. The main mathematical formulation used for diffusion is that of Fick generalised to water-gas two-phase transport. The impact of porosity and water saturation on diffusion coefficient is generally represented by Millington-Quirk type formulations [19]. Gas exchanges between the liquid and gas phases are generally represented by a Henry-type formulation (linear relationship between dissolved concentration and gas partial pressure, [14]).

Studies are in progress to increase support for using these gas transport process representations and refine these where needed, particularly in connection with phenomena associated with hydro-mechanical couplings (gas capillary thresholds, pathway dilation, degradation of concrete over time due to corrosion of the rebars, etc.) or hydro-chemical coupling (alkaline plume, effect of bacteria, etc.). The WP GAS of EURAD is expected to contribute to these efforts by extending the scientific bases about possible transport modes and couplings in clayey materials and gathering additional data in conditions relevant to the repository configurations considered in clayey host formations.

The thermal transient has been exhaustively investigated these last decades. For the clay host rocks and bentonite, previous knowledge indicates that an increase in temperature due to the presence of heat-emitting wastes will induce strong and anisotropic THM coupled responses within the clayey materials [18]. Although the effect of temperatures higher than 100°C has been studied concerning, for instance, mineralogical transformations of bentonites [16,28,29], less is known with respect to HM properties of clayey materials for this range of temperatures, mainly because of the testing experimental issues.

This relative scarcity of scientific bases at high temperatures has led to geological repository concepts for heat-emitting wastes that often limit maximum disposal container surface temperatures to about 100°C [33]. Higher temperature limits could have significant advantages, such as allowing the disposal of higher enrichment/burn-up spent fuels and shorter interim storage/cooling requirements. The WP HITEC of EURAD is expected to contribute to these efforts by extending the scientific bases on the THM behaviour of clays at high temperatures. Although the purposes and requirements of clay host rock and bentonite in repository concepts differ, their mechanical behaviour is studied both experimentally and numerically in a similar way.

4.3 Analysis of long-term gas-related repository transient

For countries that already have, if not a site, at least data on the two-phase behaviour of the target geological layer and that have carried out numerical evaluations of the long-term transient evolution of a repository under the action of gases, the overall evolution of the geological disposal facility with time includes similar structuring elements as detailed in [17] and illustrated in Figure 1 based on Andra concept.

At the time of closure, the remaining gases from the operational period are rapidly dissolved and diffused through the groundwater infiltrating the host rock (gas transport number 3 in Fig. 1).

After a certain period of time, which varies according to the repository concept and the properties of the host rock, dissolution and diffusion may no longer be sufficient to evacuate all the gas that is produced by anoxic degradation processes (mainly corrosion of metals) and a gas phase may develop in part or all of the repository (gas transport number 4 on Fig. 1).

This gas phase generally remains confined to the repository's system of disposal cells and galleries while the host clay layer remains almost saturated with water due to at least one of the following elements: very high gas entry pressure, high capillarity gradient once gas desaturates the media, very low permeability. Desaturation of the host rock, if it is considered to happen, would be restricted to the immediate vicinity of the galleries (metre scale), and saturation would not decrease by more than a few per cent in that zone.

The gas migrates towards the repository access structures (transport galleries, ramps, shafts) mainly by convection but also continues to dissolve into the surrounding groundwater along the way.

Along the way, gas flow is slowed down by the repository closure system, mainly by the seals.

While migrating, the gas is not expected to displace large quantities of water along the galleries mainly (i) because a limited desaturation of the EBS/EDZ is sufficient to obtain high enough transmissivities for gas and (ii) because water can be more easily pushed into the surrounding clay by gas than displaced along the galleries.

In some evaluations, a significant portion of the gas generated in the repository may reach the geological layer above the host rock under gaseous form through the shafts, ramps and/or the surrounding EDZ (gas transport number 6 in Fig. 1).

The duration of these phenomena linked to gas generation in the repository is of the order of several tens to several hundred thousands of years.

It cannot generally be excluded that gas pressure may reach values such that the transfer of gases will be controlled by hydro-mechanical couplings. This would materialise initially as an expansion of the connected pores ('dilatant pathway') and then possibly in the fracturing of the clay barrier material (in particular if this pressure exceeds the local minor mechanical stress). These effects are investigated in terms of gas transfer and material permeability.

In the frame of the WP GAS of EURAD, UPC/CIMNE geotechnical laboratory studies, for instance, the consequences of the passage of gas in Boom Clay and Opalinus Clay by analysing gas injection tests performed in oedometer cells at constant vertical stress [11,12]. They show that the bedding plane orientation plays a fundamental role in the volume change behaviour during gas migration (Fig. 2). Samples with bedding planes orthogonal to flow are less constrained and consistently displayed more significant expansions than samples with bedding planes parallel to flow. Estimation of the effective permeability to gas flow during the dissipation stages is, in general, higher than the measured for water flow, which indicated a possible opening of preferential pathways for the gas flow that controls the permeability. This opening of fissures or discontinuities due to gas transfer is confirmed by the pore size distributions obtained from mercury intrusion porosimetry (MIP) tests before and after the gas injection experiments.

As the host rock has self-sealing properties due to its high clay content, if a fracture occurs, it is expected to close as soon as the gas pressure decreases and the effects on dissolved element transfers are weak. To assess the self-sealing capacity of clays after the passage of gas, UPC/CIMNE geotechnical laboratory compares the water permeability

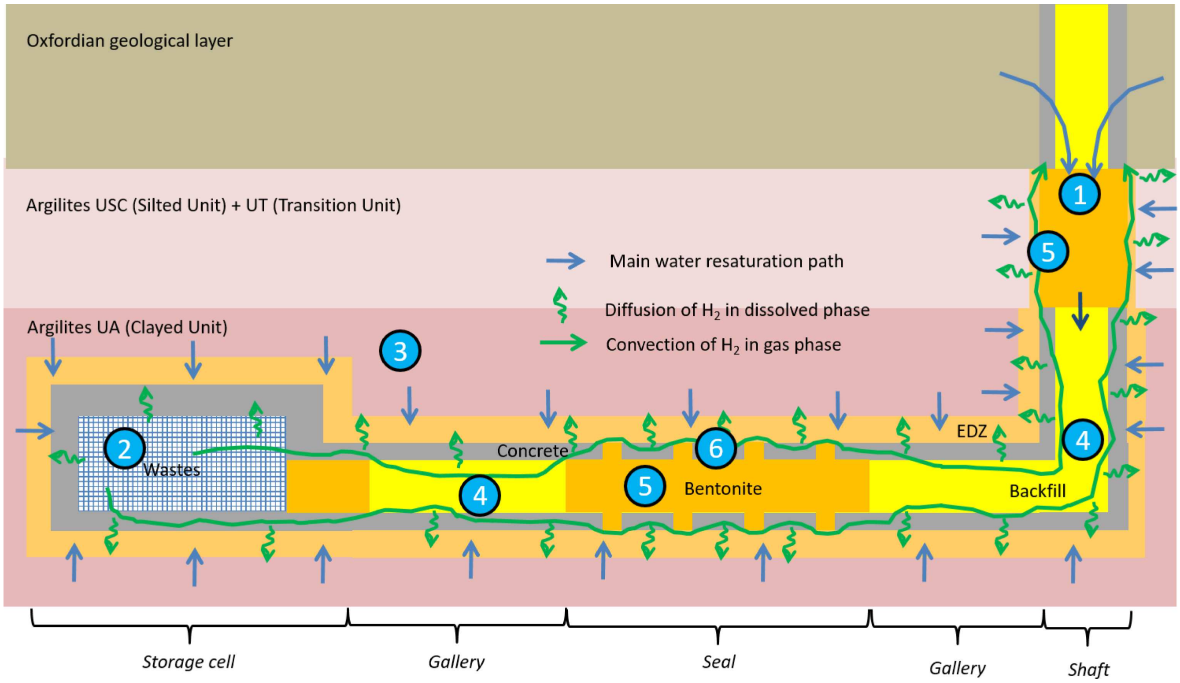

Fig. 1. Schematic representation of the Andra repository concept and the main phenomena structuring the hydraulic-gas transient [17]: 1 – resaturation of the shafts and ramps seals; 2 – gas production; 3 – diffusion of dissolved gas in clay host rock; 4 – transfer of gas by the components of the repository; 5 & 6 – low permeability of the bentonite of sealing cores favouring the transfer of gas by concrete annular rings, the EDZ and interfaces between the different materials.

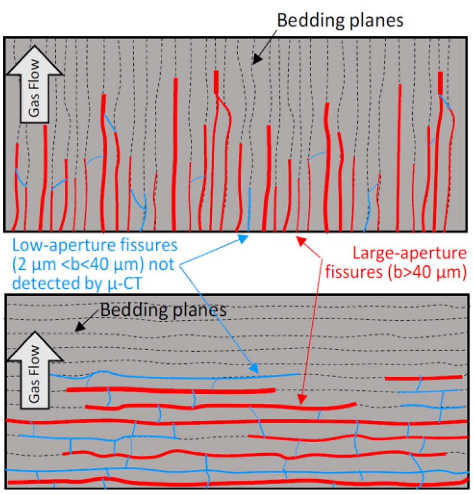

Fig. 2. Schematic representation of expected gas flow at different orientations of bedding planes: parallel to flow (top) and orthogonal to flow (bottom). Dashed thin lines represent bedding planes. Solid lines depict the volume occupied by gas: apertures larger than 40 µm and detected by µ-CT (in red); and apertures between 2 and 40 µm detected by MIP but not by µ-CT (in blue) [12].

before and after gas injection. Their experiments on Boom Clay indicate a progressive recovery of water permeability after re-saturation, which pointed to a good self-sealing capacity. The preferential pathways developed along the bedding direction after the gas injection was not observed on µCT images after the re-saturation. Nevertheless, a small pore volume at the macro-porosity scale was still detected by MIP and the gas permeability after re-saturation is slightly high, which could imply some memory of the previous gas pathways [13].

Such assessments at the scale of repository sub-systems are a good illustration of how experimental and modelling results of the WP GAS programme can help to determine whether hydro-mechanical couplings and possibly associated damages play a role in the functioning of the global repository system.

4.4 Analysis of temperature effects on repository

4.4.1 Clay host rock

The characterisation of in situ THM behaviour of the clay host rock is significant for the design and the long-term safety of the deep geological disposal facility. Obviously, the heat generated by the waste must not affect the favourable properties of the host rock, especially its chemical, mechanical and transport properties.

The temperature rise in a low permeability porous medium such as Callovo-Oxfordian (COx) claystone, Opalinus Clay or Boom Clay generates pore pressure increase essentially due to the difference between the thermal expansion coefficients of water ($\sim 10^{-4}\,K^{-1}$) and one of the argillaceous rock skeleton (e.g., $\sim 1.28 \times 10^{-5}\,K^{-1}$ for COx). Thermal pressurisation is the key mechanism for the potential damage induced by the waste emitted heat. There is a competition between excess pore pressure

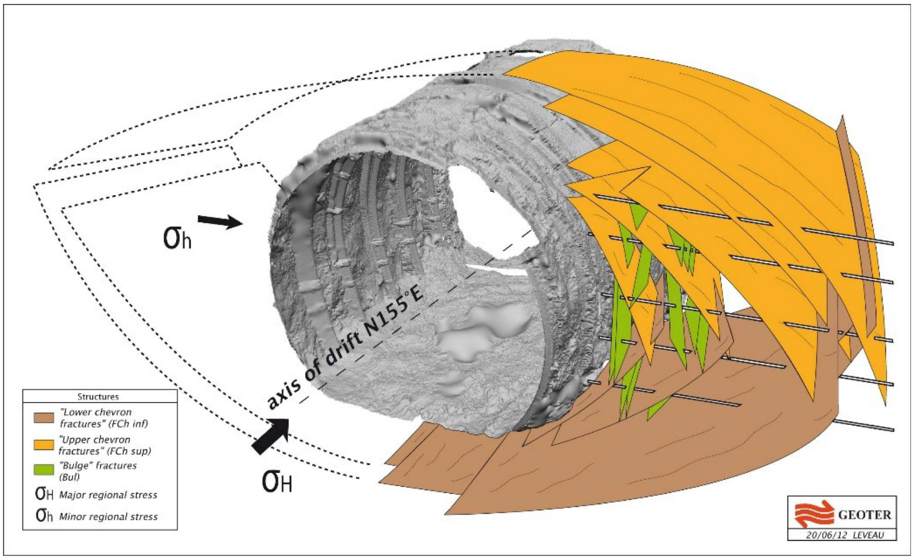

Fig. 3. Conceptual model of the excavation-induced fractures network in Callovo-Oxfordian claystone (Meuse/Haute-Marne URL, [32]).

due to thermal pressurisation and drainage due to pore pressure gradient increase. With the periodic distribution of an important number of similar parallel cells and their lengths, it can be expected that the induced pore pressure between cells could not be dissipated in the horizontal direction. In the near field, the excavation of micro tunnels induces a fractured zone around them ([3]; Fig. 3). In the far field, the pore pressure induced by the temperature increase leads to a decrease of the mean effective stress which can reach the tensile strength of the rock and induce damage, e.g., it could reactivate fracture/fault. The evolution of the effective stress field in the vicinity of the cell due to the temperature rise can induce the fracture opening or propagation in this fractured zone. The effect of the temperature on the behaviour of the excavation-induced fracture network around the HLW cell will affect the load evolution on the casing (emplaced for retrievability purposes) [4].

When modelling a nuclear waste repository, horizontal displacement, thermal flow and fluid flow are set to zero on the lateral boundaries of the model due to the symmetry conditions. The periodic distribution of parallel cells prevents lateral expansion of the rock and thus provokes compression thermal stresses in horizontal directions. The decrease of the total stress sets off the effect of the pore pressure build-up on the horizontal components of the effective stress tensor. However, the vertical effective stress increases and can, in some cases, reach the tensile strength of the rock (i.e. potential appearance of sub-horizontal cracks). One of the objectives of HITEC is to better assess the behaviour of clays under such stress path, and determine the potential damage appearance and the effect of a macro cracking over the pore pressure field and the permeability.

In that context, ULorraine studies, for instance, the effect of temperature on the behaviour of the Callovo-Oxfodian claystone. To achieve that goal, they perform short-term triaxial compression tests and long-term (creep) triaxial tests at different temperatures and confining pressures. The first results obtained from short-term triaxial compression tests [9] suggest that the peak strength decreases with the increase of temperature up to 100°C for both orientations, parallel and perpendicular to the bedding plane (Fig. 4).

Modelling the interaction between a structure, the casing and the rock mass is always a complex task (as illustrated in Fig. 5 by a schematic representation of coupled processes associated with vapour transport). Around the canister and the casing, the increase of the thermal stress and change in pore pressure increase the complexity of interaction between the rock mass and the structure. The higher temperature is reached in the surrounding of the casing/heater, meaning that the heat could affect the behaviour of the excavation fracture network around the cell. It is important to assess if clay host rock thermal compaction could affect the near field and far field.

4.4.2 Bentonite buffer

Although the effect of temperatures higher than 100°C has been considerably studied concerning mineralogical transformations (unfortunately not always in clearly representative conditions), less is known concerning hydro-mechanical properties for this range of temperatures, mainly because of the testing experimental issues. It has been shown that the effect of temperature on hydro-mechanical properties of bentonite has been systematically studied for temperatures of up to 100°C and is quite well established with respect to safety functions: temperature modifies some properties, but they keep in values acceptable for complying with the safety functions. Although temperature increases the hydraulic conductivity, the effect on swelling capacity seems to depend on the predominant exchangeable cations [6,34]). Less work has been done on the effect of temperature on the water retention curve and thermal conductivity. Likewise, most

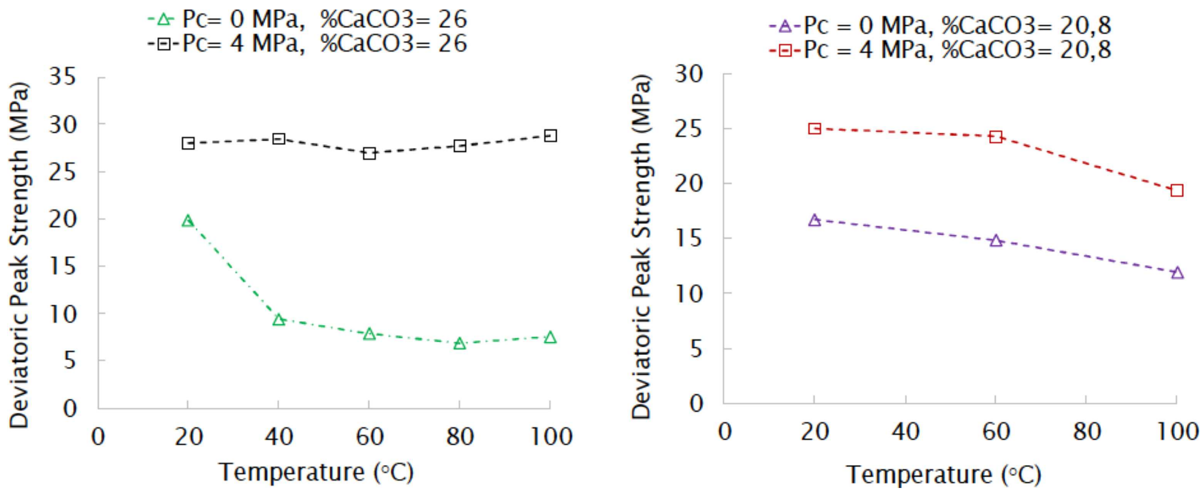

Fig. 4. Evolution of the peak strength of COx claystone as a function of temperature: at left the parallel orientation, at right the perpendicular orientation (from [9]).

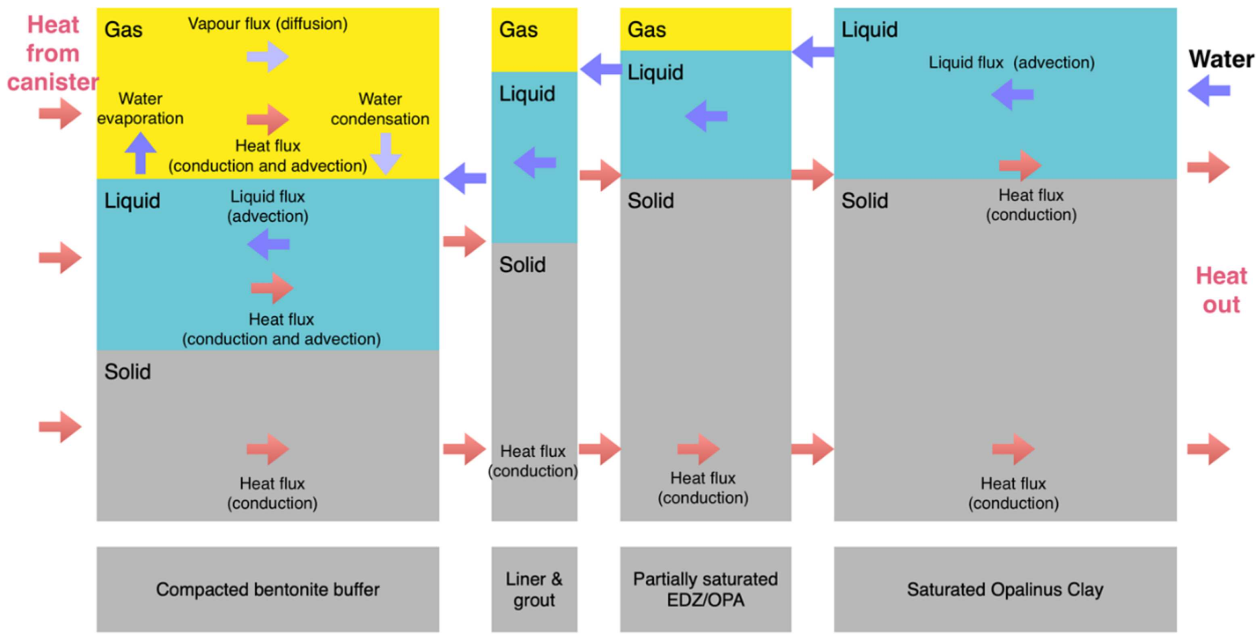

Fig. 5. Simplified schematic illustration of coupled processes associated with vapour transport [32].

laboratory studies have focused on compacted bentonite; therefore, it cannot be stated if the effect of temperature on some properties is affected by the initial fabric (compacted powder, grains, pellets) or not. Concerning the modelling of the buffer behaviour, it is considered that the THM formulations developed and validated for temperatures below 100°C can be extended without large modifications to temperatures above that value [10].

In the frame of the WP HITEC of EURAD, a Czech consortium consisting of the CTU, CU, and UJV is focusing, for instance, on determining changes in the hydro-mechanical, geochemical and mineralogical characteristics of Czech BCV bentonite due to elevated temperatures. The initial results of the analysis of the dry-treated bentonite show a slight degradation with a temperature higher than 100°C in terms of hydraulic conductivity and water retention capacity, while the effect of temperature on the swelling pressure is not significant. Similar tests carried out on wet treated material did not allow to put in evidence of such changes. [5,8].

4.5 Accounting for gases and temperature in performance assessments

Total system performance assessment models in most national programmes considering clayey host formations and/or barriers do not currently directly represent the effects of gases on radionuclide transport – at least on soluble radionuclide transport – and thus their potential radiological impact. Indeed, it is assumed that the transport rate of solutes, among which soluble radionuclides, is higher if the 'repository/host rock' system remains saturated with water. It is thus deemed conservative in estimating the transport of radionuclides in water-saturated conditions.

It is also recognised that inactive gas produced in large quantities in a repository (essentially H_2) and transported through it could act as a carrier for radionuclides that would be present in gaseous form. This can be taken into account via a numerical evaluation based on the two-phase phenomenological conceptualisation, which is currently used as a reference (Darcy and Fick laws generalised for water and gas transfers and Henry's law for exchange between liquid and gas phases). Alternatively, gaseous radionuclides might be represented as dissolved in the pore water but with specific hypotheses concerning their transport (increased migration parameters values and privileged passage via the interfaces between materials, instantaneous transport from the galleries up to the accesses, etc.).

As far as gas fracturing is concerned, if this mechanism is considered, it is generally under the form of an altered evolution scenario or even under the form of a low probability or 'What-If' type scenario. This kind of scenario generally assumes that the system is fully saturated but incorporates the presence of a drain (local or more extensive) connecting the repository galleries to the nearest aquifer above and/or below the clayey host rock.

For the disposal of HHGW, performance assessment models require to well understand the consequences of the heat produced on the properties and the long-term performance of the natural and engineered clay barriers. For now, most of those developed for disposal concepts that involve clay consider a temperature limit of 100°C. In such conditions, it is known that the performance of the system in clay host rock should not be affected negatively by the thermal evolution of the EDZ around a radioactive waste repository. However, in the case of higher temperatures, process understanding is currently insufficient to guarantee that this will still be true.

Being able to tolerate higher temperatures while still ensuring an appropriate performance would have significant advantages in terms of repository optimisation. The WP HITEC contributes to assessing how higher thermal loads, changes in canister pitch and/or tunnel spacing affect the likely performance of geological barriers. This includes an assessment of the consequences of locally surpassing the threshold criteria that are now put forward to ensure the integrity of the geological barrier in safety cases.

Thanks to its excellent properties for radionuclide containment and confinement, clays are exploited in developing most geological disposal systems in Europe either as host rock or as materials for engineering barriers. One of its particular favourable properties is its self-sealing capacity which allows fractures to close rapidly. The impact of temperature or gas on the self-sealing capacity of clays is one of the key questions addressed by both WP HITEC and GAS of EURAD. ULorraine investigates, for instance, the self-sealing capacity of the Callovo-Oxfordian claystone with the same test configurations after both thermal loading and/or gas passage. Their first self-sealing tests performed on the Callovo-Oxfordian claystone under triaxial condition and at various temperature shows that the self-sealing process significantly reduces the fracture permeability [1]. This mechanism is faster for samples oriented in parallel to the bedding plane than samples oriented perpendicularly to the bedding plane. Similar tests are ongoing in the frame of the WP GAS after gas injection to characterise all the (T)HM(-C)(+gas) processes associated with self-sealing mechanisms in clayey materials.

5 Expected impacts and key challenges of these two WPs

5.1 Expected impacts of the WP GAS

During the course of the WP GAS, conceptualisations of gas transport through repositories in clayey host formations will be improved by the integration of the findings of the tasks on transport mechanisms and barrier integrity. The phenomenological description improvements will be evaluated for transfer to the repository scale via updates to the storyboards. Uncertainties about which gas transport modes will effectively be active or about other gas-related aspects of the evolution of the system will be identified and evaluated in terms of possible scenarios to be investigated. This task will also benefit 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 facilities in clayey host formations and (ii) a better characterisation of hydro-mechanical coupling in swelling clayey materials (from the installation of materials to their evolution over the long term).

In that sense, the WP GAS is expected to provide building blocks to implementers that may inspire their storyboards and, in turn, design measures that further reduce the gas impact on the disposal system and/or the uncertainties associated with the gas flow through engineered and natural clayey barriers in geological disposal systems. It will allow testing of various approaches for the treatment of gas in repository-scale models, identifying the inherent strengths and limitations of each approach and assessing its suitability in different contexts, as this may depend on the disposal system that is being evaluated (host formation/concept) or even the advancement of the (national) programme.

5.2 Expected impacts of the WP HITEC

While both clay host rock and bentonite materials have a long history of studies, investigations of thermo-hydro-mechanical behaviour at elevated temperatures have not been common. Carrying out experiments at higher than 100°C requires typically large modifications in the equipment and measuring system. The same is true for models, which might require rather many modifications (parameters and constitutive relations) to be functional at higher temperatures. In that sense, the WP HITEC is expected to increase the scientific and technical knowledge of the mechanical behaviour of clay host rocks and bentonites at temperatures higher than 100°C.

This knowledge will interest radioactive waste management, implementation, and safety cases. Being able to better accommodate higher temperatures than presently accepted 100°C while ensuring similar safety standards can have significant advantages with respect to disposing of higher enrichment/burn-up fuels, interim storage requirements, (re)packaging of the waste and reducing the footprint of the disposal. It could also allow for evaluating limits of temperature and the overall impacts higher temperatures cause to materials and systems. In addition, proving temperatures higher than presently considered acceptable is very relevant even for current concepts: it increases safety margin and gives greater credibility to the design.

5.3 GAS and HITEC common challenges

The main challenges of the two R&D work package, GAS and HITEC of the European Joint Programme EURAD, lies in the stepwise integration and contextualisation of experimental results and modelling approaches. On the one hand, it sought to increase the understanding and predictability of the impact on clay barriers by studying fundamental mechanisms and their couplings related to gas and heat transport, respectively, observed at the lab scale or via past and present in-situ experiments. On the other hand, models of different complexity are built to support the understanding of the observed mechanisms and transfer it to the configurations and conditions of the different clayey components and the context of the functioning of a disposal system. To that aim, it is also essential to be transparent about the context in which any piece of data has been collected, or any model has been developed, key uncertainties, and clearly communicate all elements of scientific consensus.

These two WPs strongly build on the results, return of experience and conclusions from past EC projects, for instance, FORGE, TIMODAZ, BELBaR, and BEACON. These projects revealed complex mechanisms but also hinted that these could probably be addressed using a combination of simple and robust (e.g., bounding) approaches. By providing the end-users with a collection of data, tools and building blocks for storyboards that may be of use in the conceptualisation of the functioning of their specific geological disposal system, GAS and HITEC will contribute to the development of such approaches and support the justification of their use.

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Conflict of interests

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Séverine Levasseur: conception, execution, interpretation, writing. Xavier Sillen: interpretation, revision. Paul Marschall: interpretation, revision. Jacques Wendling: Paul Marschall: interpretation, revision. Markus Olin: conception, execution, interpretation, writing. Dragan Grgic: interpretation, revision. Jiri Svoboda: interpretation, revision.

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