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EURAD state-of-the-art report: thermo-hydro-mechanical behaviour of clay buffers at high temperatures

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Most safety cases for radioactive waste disposal consider a temperature limit of 100°C for the clay buffers. Given that being able to tolerate higher temperatures would have significant advantages, the work package HITEC of the EURAD project aimed at determining the influence of temperature above 100°C on buffer properties, trying to establish if the safety functions are unacceptably impaired. A synthesis of the state of knowledge on the thermo-hydro-mechanical and chemical behaviours of different buffer materials at different temperatures is presented, along with the progress made in this area during HITEC. The changes in the properties of the preheated material and the hydromechanical properties of bentonite at high temperatures were assessed. To cover the first instance, bentonite was heated at 150°C in dry and wet conditions for different periods of time up to 2 years. The clay mineralogy was significantly preserved. The slight changes observed in the other properties were opposite depending on the heating conditions: in the case of evaporation, the cation exchange capacity, specific surface area, sorption coefficients, and sometimes swelling pressure decreased. These changes likely resulted from the strong drying induced by the elevated temperatures. Bentonite was also subjected to hydration under a thermal gradient in field and laboratory tests. No post-mortem structural modifications of the smectite were observed; however, dissolution and precipitation of species occurred, conditioned by the type of bentonite and hydration water. These processes were accompanied by the modification of the exchangeable cation complex. Determination of the hydromechanical properties of expansive clay at elevated temperatures is challenging owing to experimental and interpretation issues. In most cases, a reduced swelling pressure was obtained when the temperature increased, particularly at higher dry densities. These results may have been affected by the experimental protocols, use of bentonite or purified smectite, and exchangeable cations. Even at the highest temperatures, bentonite can fill voids and develop large swelling pressures at high densities. Thermo-hydro-mechanical models were developed or upgraded during the project to include thermal phenomena and dependencies and were applied to the simulation of new laboratory thermo-hydraulic tests in cells.

KEYWORDS

bentonite, radioactive waste, thermo-hydraulic behaviour, swelling, hydraulic conductivity, buffer, geochemistry

1 Introduction

The work package "Influence of Temperature on Clay-based Material Behaviour" (WP7) of the European Union's Horizon 2020 EURAD project (https://www.ejp-eurad.eu/) aimed to develop and document improved thermo-hydro-mechanical understanding of clay-based materials (host rocks and buffers) exposed at high temperatures or having experienced high temperature transients for extended durations. The WP's raison d'être was to evaluate whether elevated temperature limits (up to 150°C for the clay buffer and ~90°C for the host rock) are feasible for a variety of geological disposal concepts for high heat generating wastes (HHGW). To dispose of HHGW, understanding the effects of the heat produced on the properties and long-term performance of natural and engineered clay barriers is crucial. Most safety cases for disposal concepts that involve clay currently consider a temperature limit of 100°C. The ability to tolerate higher temperatures, while ensuring appropriate performance, is an advantageous feature (e.g., shorter aboveground cooling times, more efficient packaging, fewer disposal containers, fewer transport operations, and smaller facility footprints). HITEC examined bentonite buffers and determined the influence of temperature on the physical properties, swelling pressure, hydraulic conductivity, water adsorption, mineralogy, and geochemistry of the buffer to establish whether the buffer safety functions are unacceptably impaired. Consequently, HITEC was a step toward optimizing the architecture of a deep geological repository, as the range of temperatures tested had not been considered systematically in previous research. The WP also studied the possible extent of elevated temperature damage in the near and far fields of clay-host rock formations and indicated the likely consequences of such damage (see companion paper, Villar et al., 2025). Within the framework of the EURAD project, state-of-the-art reports, which form the basis of the papers included in this Special Issue, were prepared for each work package, and this contribution and the companion paper are based on the updated state-of-the-art report of WP7 HITEC (Villar et al., 2024), which includes an overview of the knowledge gained during previous research activities and focuses on the progress made during the project.

Previous and ongoing national and European Community-supported research programmes led to detailed understanding of the various key thermo-hydro-mechanical and chemical (THM-C) processes taking place in the buffer material under temperatures of up to 100°C [e.g., BACCHUS (Neerdael et al., 1992); FEBEX (ENRESA, 2006); PROTOTYPE (Olsson et al., 2013); PEBS (Schäfers et al., 2014)]. Within the PEBS project (Long-term Performance of the Engineered Barrier System, seventh European FP 2007–2011), the conclusion was reached that the characterisation of bentonite performance and THM properties below 100°C was established (Johnson et al., 2014). Overall, the observed effects of temperature on the hydromechanical properties (decrease in swelling pressure, increase in permeability, and decrease in water retention capacity) can be qualitatively explained by considering the transfer of high-density interlayer water to macropores, which is triggered by an increase in temperature. The different behaviours of smectites exchanged with monovalent versus divalent ions can be explained by considering the different interlayer/diffuse-double-layer water ratios. However, a literature survey conducted during the PEBS project confirmed that information regarding bentonite at higher temperatures was less abundant. Specifically, lack of information was identified:

- for granular bentonite material (manufactured bentonite as grains or pellets);
- with respect to the water retention curve;
- concerning the effect of the degree of saturation on the thermal conductivity (low thermal conductivity leads to higher temperature) and water permeability; and
- regarding the effect of temperature on swelling capacity depending on exchangeable cations.

In the following sections, syntheses of the state of knowledge on the THM behaviour of different buffer materials at different temperatures are presented, along with the progress made during HITEC. To better contextualise this information, the common features of geological waste disposal concepts are summarised. The characteristics of the buffer materials analysed during the project are described, and their thermal, hydraulic, and mechanical properties are summarised in Section 3. Large-scale *in situ* experiments, in which buffer materials are subjected to elevated temperatures for long periods, are briefly described in Section 4. The samples retrieved from some of these tests were tested during the project, as were other raw bentonites that were either heated in the laboratory or tested at high temperatures. The

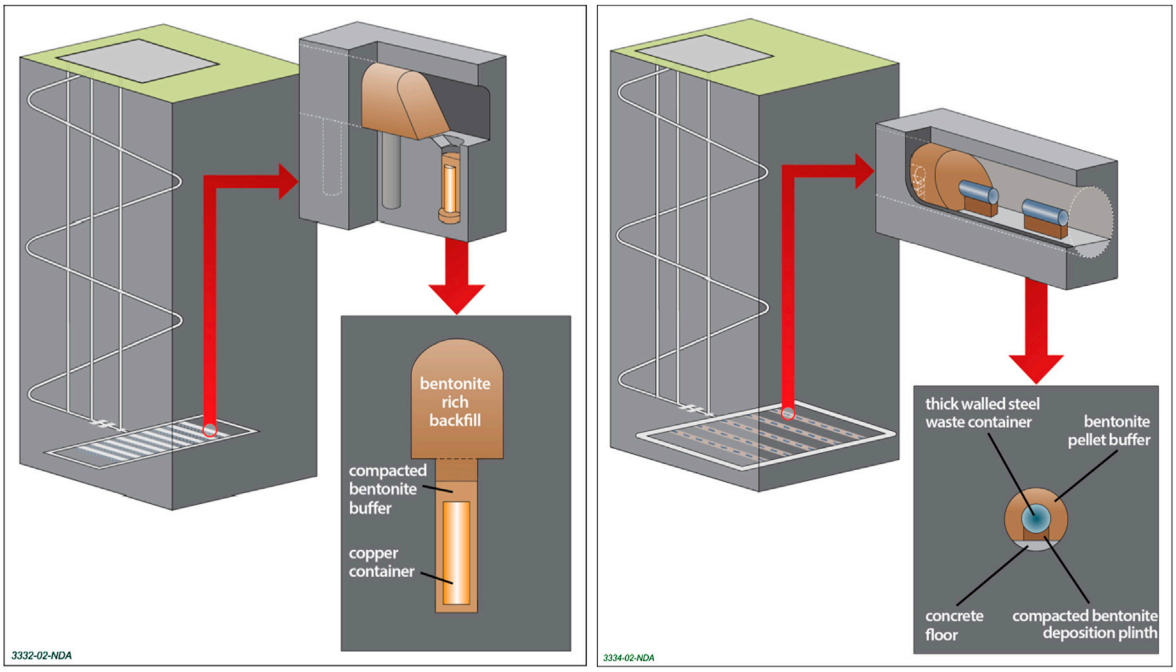

FIGURE 1
Illustrative designs for a higher strength rock (based upon KBS-3V, left) and lower strength sedimentary rock type (based upon NAGRA concept, right) after RWM (2016).

results obtained in this experimental campaign are summarised in Section 5, where also the experimental procedures and knowledge gained from previous investigations are reviewed. This section is organised according to the type of determination considered: the characterisation of preheated materials (Section 5.1), determination of HM properties at high temperatures (Section 5.2), and small-scale experiments reproducing the conditions of the barrier in the repository (Section 5.3). THM models previously available or upgraded during HITEC and their performances under conditions relevant to HITEC are briefly discussed in Section 6.

2 Deep geological repository concepts

Several common features exist in national concepts for HHGW disposal (e.g., spent fuel, SF, and/or high-level waste, HLW). Radioactive substances will be contained within overlapping protective barriers such that no deficiency in one barrier and no predictable changes will endanger isolation. The barriers include the physical state of the fuel, disposal canister, bentonite-based buffer, backfilling of the tunnels and surrounding rock (Figure 1), and other sealing elements such as plugs.

Supplementary Table S1 summarises the characteristics of the different national concepts. Their stages of development are diverse, and although some have selected the lithology and defined architecture, others are still considering designs for different potential host rocks. ANDRA (France) and ONDRAF/NIRAS (Belgium), both with concepts designed for clay host rock, do not consider using a clay buffer between the canister and host rock. However, NAGRA's concept (Switzerland), also designed for clay host rocks, considers using a clay buffer around canisters. The concepts of other waste agencies are based on crystalline host rocks (except ENRESA's). Among them, only ENRESA (Spain) and SÚRAO (Czech Republic) consider the use of bentonites with predominant divalent cations in the smectite interlayer, while the others consider using sodium bentonite.

The concepts presented assume that the maximum temperature anywhere in the buffer or host rock should remain below 100°C, except NAGRA, which allows for a maximum temperature in the bentonite of up to 130°C (see Villar et al., 2020 for more details). The maximum temperature is usually defined by considering the post-closure safety and economic aspects. In this sense, NAGRA, which relies on hundreds of meters of host rock and clay-rich geological units, is less conservative with respect to temperature limits in the buffer. This is different for geological repositories in other host rocks (e.g., crystalline) that could become fractured, potentially allowing for the fast transport of nuclides. Therefore, the relative significance of the technical barriers for post-closure safety becomes more relevant (e.g., POSIVA and SKB). Predictions of the thermal evolution of a repository have been obtained based on modelling exercises of generic repositories and have been used to optimize the repository design to reduce its thermal impact (Jobmann et al., 2017; Senger et al., 2014; NAGRA, 2008; NAGRA, 2015; Lanyon, 2019; Ikonen et al., 2018; Amec, 2016).

3 Bentonite materials used in HITEC

Three bentonites were identified as those that were better characterised and used by more participants in the HITEC project and in a wider variety of tests, including large-scale tests (Section 4). These were MX-80, FEBEX, and BCV, although several other bentonites such as Bara-Kade, Kunipia-G, PBA-22 Extra, and PBC were also used in the project. In addition, some of the materials used in the Alternative Buffer Material (ABM) large-scale test (e.g., DepCAN, Asha505, Calcigel) are briefly described below. Relevant references are provided for detailed information.

Supplementary Tables S2–S5 summarise the main properties of the buffer materials considered in HITEC. In some cases, the range of values was broad because they corresponded to different batches. All exhibited high smectite contents (>72%) and CEC (>64 meq/100 g), with sodium as main exchangeable cation in MX-80, Bara-Kade, and Asha505.

3.1 Wyoming-type bentonite

MX-80 bentonite is produced in Wyoming (USA) by American Colloid Co. It is a bentonite of volcanic origin. The deposit has been mined for decades, and the characteristics of the bentonite reported in the literature vary over time, depending on the batch. Some of the initial characterisations of the material in the context of radioactive waste disposal were performed by Müller-Vonmoos and Kahr (1983), Börgesson et al. (1988), Madsen (1998), and Pusch (2001). Dixon et al. (2023) presented a recent database of properties determined by various authors, including the thermal conductivity, hydraulic conductivity, and swelling pressure, highlighting their variability.

The material with the commercial name Bara-Kade is Wyoming sodium bentonite, which is similar to MX-80. The as-received water content was 8.2%.

In the HE-E and FE tests (Section 4), MX-80 was used in the form of irregular pellets of size <10 mm, dry density ~2.1 g/cm^3 and water content 6% (see de Book et al., 2009; Gaus et al., 2014 for manufacturing process). The Volclay (used by BGS), with a grain size of 16–200 µm, was characterised by Svensson et al. (2011).

The water retention capacity of MX-80 bentonite has been verified by various authors, and the effects of dry density (Pintado et al., 2013; Seiphoori et al., 2014) and temperature on the water retention curve have also been analysed (e.g., Jacinto et al., 2009). Jacinto et al. (2009) found that the influence of dry density on water retention capacity depends on the suction range; the limiting value is approximately 30 MPa. For suctions above this threshold value, the retention capacity in terms of water content was slightly higher as the dry density increased, whereas for lower suctions, the lower the dry density of bentonite, the higher its water content. The retention capacity decreased with temperature, more than predicted by the change in water surface tension, especially at high temperatures and low suction. Villar and Gómez-Espina (2008) reported suction measurements on compacted MX-80 bentonite with different water contents at temperatures of up to 120°C.

Villar (2005) and Karnland et al. (2008) systematically studied the dependence of swelling pressure and hydraulic conductivity on dry density.

The increase of thermal conductivity of the MX-80 bentonite with dry density, water content and temperature has been shown by Tang et al. (2008a) and Xu et al. (2019), with values measured between 0.2 and 1.2 W/m·K for ranges of dry density 1.0–1.8 g/cm^3, water content 0%–25% and temperature 5°C–90°C.

3.2 FEBEX bentonite

The FEBEX bentonite is a 900-t batch of bentonite extracted from the Cortijo de Archidona deposit and processed in 1996 for the FEBEX project. The processing consisted in homogenisation, air-drying and manual removal of volcanic pebbles on site and, at the factory, crumbling, drying in a rotary oven at temperatures between 50°C and 60°C and sieving through a 5-mm mesh. The initial characterisation can be found in the final report of the project (Enresa, 2006) and; Villar (2002); Fernández (2004). This material was also used for NF-PRO and PEBS projects (Villar and Gómez-Espina, 2009) and was distributed over the years to different laboratories for use in different projects [e.g., ABM (Svensson et al., 2011), ESDRED (Alonso et al., 2008)].

The smectite content of FEBEX bentonite exceeds 90 wt%. The smectitic phases are composed of a montmorillonite-illite mixed layer, with 10–15 wt% illite layers. It also contains variable quantities of other minerals, as shown in Supplementary Table S3. Although it predominantly contains divalent exchangeable cations, the amount of Na in the interlayer is also significant (Supplementary Table S4). The predominant soluble ions are chloride, sulphate, bicarbonate, and sodium. The other basic properties are listed in Supplementary Table S2.

The bentonite retention curve was determined at different temperatures for samples compacted to different dry densities and maintained at a constant volume (Lloret et al., 2004; Villar and Lloret, 2004). An empirical fit relating the water content to the suction, degree of saturation, porosity, and temperature was presented by Villar and Gómez-Espina (2009).

The saturated hydraulic conductivity of the compacted samples is exponentially related to their dry density and swelling pressure (Villar, 2002). For a dry density of 1.6 g/cm^3 the saturated permeability of the bentonite is approximately 5×10^{-14} m/s and the swelling pressure ~6 MPa at room temperature, either using diluted granitic or deionised water. An increase in temperature tends to increase permeability and decrease swelling pressure (Villar and Gómez-Espina, 2009). Some isothermal infiltration and heat flow tests at a constant overall water content were back analysed. The experimental data were fitted using a cubic law for the relative permeability and a value of 0.8 for the tortuosity factor (Pintado et al., 2002).

The thermal conductivity measured in samples compacted in the range of water content 0%–25% and dry density 1.49–1.75 g/cm^3 expands between 0.45 and 1.30 W/m·K, with a dependence on degree of saturation that could be fit to a sigmoidal relation (Villar, 2002).

3.3 BCV bentonite

Bentonite BCV is a Czech bentonite that formed via the in-situ alteration of Fe-rich tuffs and augite-biotite-type tuffites (Franče, 1992). These bentonites are enriched with Fe (especially in the octahedral positions) and accessory minerals (i.e., Fe carbonates and oxohydroxides).

BCV is industrially produced by Keramost Ltd. at the Obrnice plant. The bentonite treatment process commenced with the sieving of the coarse material through a 20 cm × 20 cm mesh. The bentonite is then homogenised in a wheeled mill and subsequently transferred to a rotary oven for drying (average temperature of 110°C) for 45 min. The drying process ensured a water content of approximately 10%, resulting in the preparation of the material for final milling and air sieving. The processing of the BCV material led to a mixture where 1% of the total weight of the grains can be larger than 0.315 mm and at least 70% of the grains are smaller than 0.063 mm. The BCV material was first tested in 2017, and its characteristics were reported by Červinka et al. (2018), Hausmannová et al. (2018), and Svoboda et al. (2019).

Some basic properties of this material and its mineralogy are listed in Supplementary Tables S2, S3. The amorphous-phase content can be extremely high, e.g., 10 wt% (Červinka et al., 2018). The main exchangeable cations are magnesium and calcium (Supplementary Table S3).

The retention curves of bentonite were determined at ambient temperature for constant-volume samples compacted at various dry densities. In the range of dry densities between 1.4 and 1.8 g/cm^3 no differences of gravimetric water content for a given suction were observed (Červinka et al., 2018).

The saturated hydraulic conductivity of compacted BCV bentonite samples decreased exponentially with dry density, whereas the swelling pressure increased exponentially (Hausmannová et al., 2018). For a dry density of 1.6 g/cm^3, the saturated hydraulic conductivity of the bentonite is approximately 1.14×10^{-13} m/s and the swelling pressure around 8 MPa at room temperature using deionised water.

The thermal conductivity of BCV bentonite compacted at a dry density of 1.6 g/cm^3 is ~0.6 W/m·K for w = 12% and 1.4 W/m·K for the nearly fully saturated sample (Červinka et al., 2018).

3.4 Other bentonites

The *in situ* large-scale test ABM, conducted by SKB at Äspö Hard Rock Laboratory, tested a variety of bentonites. They are all high-grade bentonites with a smectite content of approximately 80 wt% and high swelling pressures at high densities and are regarded as possible buffer or backfill candidates (Svensson et al., 2011). Upon dismantling the test parcel ABM5 (described in Section 4.1), some of these bentonites were analysed, and the results were presented in the context of HITEC, namely:

- DepCAN, a Ca-dominated bentonite, was obtained from Milos (Greece). The smectite is montmorillonite.
- Asha505 is from Kutch (India), sodic and high in iron (~13 wt % Fe_2O_3); hence, its colour is brown. The smectite is a montmorillonite/beidellite with a relatively higher charge.
- Calcigel is calcium Bavarian bentonite (Germany). The smectite is montmorillonite. Smectite structural iron is more sensitive to redox reactions than most of the other bentonites studied.

Ukrainian bentonite Powder Bentonite from Cherkasy region of Ukraine (PBC), provided by Dashukovskiy bentonites, was used in the research conducted at KIPT. It is a natural bentonite containing up to 60% of Na-montmorillonite and Ca, up to 20% of Mg-montmorillonite, and quartz and calcite. The bentonite was received ground to a grain size <0.16 mm, with a water content of 7.4%.

Bentonite from the Ukrainian deposit of Cherkasy was studied by SIIGNASU—in particular, the industrially milled powder called bentonite PBA-22 «Extra» delivered by Dashukovskiy (Zlobenko et al., 2023). It is a commercial calcium bentonite with a content of montmorillonite of 70%–85%. Its structural formula, based on the chemical analyses presented in Supplementary Table S5, is $(Al_{1.34}Fe^{3+}_{0.31}Fe^{2+}_{0.01}Mg_{0.34})[Al_{0.05}Si_{3.95}]O_{10}(OH)_2 + (Ca_{0.162}Na_{0.01}K_{0.07})$.

The Kunipia-G used in the studies by BRGM is a highly pure montmorillonite (>95%) with traces of quartz and carbonates provided by Kunimine Industries and issued from the purification of the natural bentonite Kunigel-V1. The structural formula of this montmorillonite $Na_{0.41}K_{0.01}Ca_{0.04}(Si_{3.92}Al_{0.08})(Al_{1.51}Mg_{0.37}Fe^{3+}_{0.07}\ Fe^{2+}_{0.05})O_{10}(OH)_2{\cdot}nH_2O$ with a negative layer charge of 0.5 per half unit-cell was determined by Massat et al. (2016). Other properties are listed in Supplementary Tables S2–S4. For the investigation conducted in HITEC, the smectite was washed and homionized in Na^+-and Ca^{2+}-exchanged forms. Details of the preparation and characteristics of these samples can be found in Chaaya (2023).

4 Relevant large-scale tests

Large-scale *in situ* tests are performed in underground research laboratories (URL) for demonstration purposes and to reproduce the representative conditions of the buffer and near field for long periods. Some large-scale tests performed in different URLs –namely, Äspö in Sweden, Mont Terri and Grimsel in Switzerland– and also a mock-up are briefly described in the following sections. In all cases, a bentonite buffer and heater(s) simulating the waste canister were used. In the in-situ tests, hydration was natural and, in some cases, with an additional artificial contribution. These tests were selected because of the relevance of the heater surface temperature (>100°C). Different host rocks, buffers, spatial configurations, and temperatures are represented. Some of these tests are still ongoing and provide online information. Others have already been dismantled (some parcels of the ABM and LOT tests, OPHELIE), and information on the post-mortem state and modification of properties is available. Material from the ABM test has been analysed in the context of HITEC, which is why this test is described in more detail.

A summary of the main characteristics of these large-scale tests and those relevant to their long durations is provided in Supplementary Table S6. However, this was not an exhaustive compilation. For example, the Temperature Buffer Test (TBT) ran in Äspö for 7 years with a heater temperature of 140°C using MX-80 bentonite as buffer material (Åkesson et al., 2012).

4.1 Alternative buffer materials (ABM, Äspö)

The ABM test parcels exposed different clay materials to conditions similar to the storage place (e.g., compacted clay, nonpowdered metallic iron, swelling pressure from the clay, large-scale, *in situ* placement at repository depth, and natural water sourced from the bedrock) and adverse conditions with

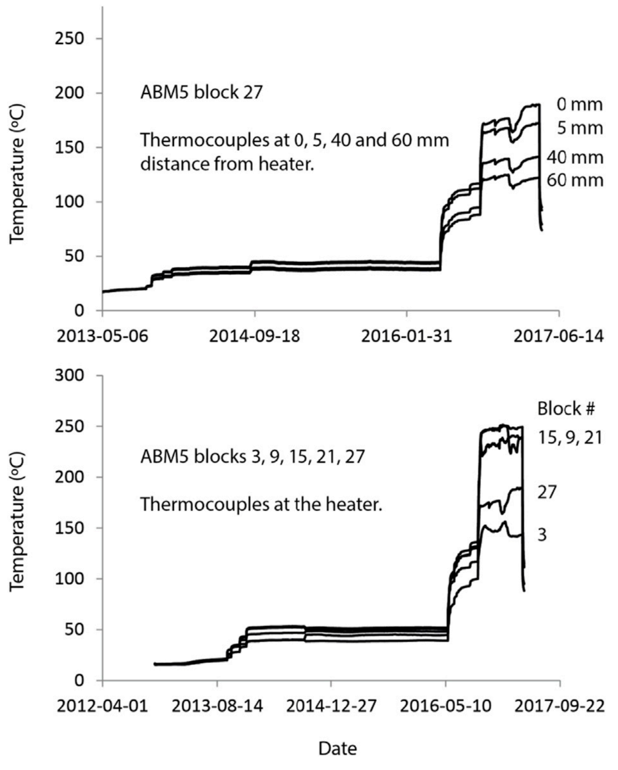

FIGURE 2
Overview of the thermal evolution of the ABM5 experiment.

respect to temperature (higher than expected in the current repository concepts). The tests offered valuable insights into the iron-bentonite interactions and the impact of temperature on the diversity of bentonites. The first ABM experiments (1–3) were conducted in 2006 (Eng et al., 2007), and three additional experiments were conducted within the ABM45 project in 2012 (Sandén et al., 2018). Each of these parcels was a vertical borehole with a depth of 3.2–3.7 m and a diameter of 0.3 m. For the experiments in general, the target temperature was 130°C (the only exception was ABM5), the heater was made of steel and different compacted bentonites were installed in direct contact with each other surrounding the central heater (Supplementary Figure S1). The following clays were used: Asha 505, Calcigel, Callovo-Oxfordian (Cox), IBECO, Deponit CA-N (Dep CAN), FEBEX, Friedland, Ikosorb Ca White, Ibeco Seal M-90, Kunigel VI, MX-80, and Rokle. A water saturation system, together with a sand filter, allowed for rapid water saturation. A summary of the test characteristics is presented in Supplementary Table S7.

ABM1 was excavated in 2009 (Svensson et al., 2011; Svensson and Hansen, 2013) and ABM2 was excavated in 2013 (Svensson, 2015). Upon dismantling, the bentonite exhibited local precipitates and iron corrosion products. The montmorillonites were fairly intact, with minor saponite formation. However, no impact on buffer performance was found (Svensson, 2015). The temperature in ABM5 was initially kept low at approximately 50°C (Figure 2), and in 2016, the temperature was increased stepwise to very high temperatures of up to 250°C at the heater interface, which were maintained for ~7 months. ABM5 was excavated in 2017, and some blocks appeared intact, whereas others were highly fractured and fragile.

4.2 Long-term test of buffer material (LOT, Äspö)

The LOT experiments were designed mainly to study the long-term stability of bentonite, mineral composition, and the impact on important properties such as swelling pressure and hydraulic conductivity. The overall design of the LOT was nearly identical to that of the ABM experiment (Supplementary Figure S1). However, the ABM used many different bentonites and steel instead of copper for the heater. The LOT experiments (7 installed, 6 retrieved, Supplementary Table S8) use Wyoming bentonite (MX-80) naturally saturated with groundwater, and run at 90°C (S-series) and 130°C (A-series). Copper heaters were not designed for the corrosion studies; however, separate copper coupons were installed in the bentonite for this purpose. The LOT experiments are ~4 m high, 0.3 m in diameter, and the central copper heaters are 0.1 m in diameter. The dismantling of the experimental packages A1 (adverse 130°C, 1 year), A2 (adverse 130°C, 6 years), and A0 (adverse 130°C, 1.5 years) is described in Karnland et al. (2000), Karnland et al. (2009a), Karnland et al. (2009b), and Karnland et al. (2011), respectively, and SKB TR-09-31.

LOT S2 and A3 were excavated in September 2019. Chemical analysis of block 15 in the A3 experiment showed maxima of Ca and S at the heater and approximately 20 mm from it, attributed to local accumulation of anhydrite ($CaSO_4$), which was also detected by X-ray diffraction (XRD). However, in neighbouring block 16, Ca and S showed only one maximum value. The reason for this is currently not understood; however, local variations in temperature, water saturation history, or fractures in the rock may give rise to minor local variations, demonstrating the importance of analysing several different profiles. However, anhydrite accumulation was expected and has been previously observed in other field experiments. A minor increase in Mg was observed toward the heater, as expected from earlier field experiments; however, no corresponding new phase was observed by XRD.

4.3 Heating experiment (HE-E, Mont Terri)

The Heating Experiment (HE-E) was a 1:2 scale heating experiment considering the natural re-saturation of the engineered barrier system (EBS) at a maximum heater surface temperature of 140°C (Gaus, 2011; Teodori and Gaus, 2012). This is part of a series of heater experiments, identified by the HE code, undertaken in the Mont Terri URL with the broad aim of investigating the THM processes occurring in Opalinus Clay and bentonite buffer (NAGRA, 2019). The experiment is conducted in a 50-m-long tunnel with a 1.3 m diameter (Gaus et al., 2014). The test section of the micro-tunnel has a length of 10 m and was characterised in detail during a ventilation experiment that took place in the same test section (Mayor et al., 2007). The HE-E focused on the early nonisothermal resaturation period and its impact on the THM behaviour of the EBS, namely:

- To provide the experimental data required for the calibration and validation of the existing THM models in the early resaturation phase.

- To increase the thermal conductivity of the partially saturated EBS from the laboratory to the field scale for bentonite and bentonite-sand mixtures.

The experiment comprises two independent heated sections, each 4 m in length. The heaters are placed in a steel liner supported by MX-80 bentonite blocks with a dry density of 1.81 g/cm^3 and a water content of 10.3%. The dimensions and materials of the two sections are the same, apart from the granular filling material: Section One was filled with a 65:35 granular mixture of sand and bentonite (Bentosund WH2), and Section Two was filled with MX-80 bentonite pellets. The use of two types of granular filling materials allowed a comparison of the THM behaviour of the two EBS materials under nearly identical conditions.

Heating began in June 2011, and the maximum temperature was reached in June 2012. Thereafter, the temperature has remained constant. The EBS is characterised by a high temperature gradient owing to its low thermal conductivity in response to drying from the introduction of the heaters. The temperature at the interface of the Opalinus Clay and the EBS is ~50°C.

4.4 Full-scale emplacement experiment (FE, Mont Terri)

The main aim of the FE experiment is to investigate the repository-induced THM coupled effects on the host rock at this scale and to validate existing coupled THM models (NAGRA, 2019). Further experimental aims were to 1) verify the technical feasibility of constructing a disposal tunnel using standard industrial equipment, 2) optimize the bentonite buffer material production, and 3) investigate (horizontal) canister and bentonite buffer emplacement procedures for underground conditions.

The FE experiment layout was designed to simulate the Swiss repository concept for SF/HLW conditions in a single tunnel at the Mont Terri URL (Müller et al., 2015). A 50-m-long experimental tunnel was constructed. At the deep end of the FE tunnel, an interjacent sealing section (ISS) was built using only steel arches for rock support, whereas the rest of the tunnel was supported by shotcrete. A 'bentonite block wall' was erected manually in a section of the ISS. In the FE tunnel, three heaters with dimensions similar to those of the waste canisters were placed on top of the bentonite block pedestals. The remaining space was backfilled with a highly compacted granulated bentonite mixture. Finally, the experiment was sealed (toward the FE cavern) with a concrete plug holding the bentonite buffer in place and reducing the air and water fluxes. The heating phase started in December 2014, with the heaters' surface temperature at values between 120°C and 130°C, although they were expected to eventually reach a temperature range of 130°C–150°C at the surface of the middle heater and approximately 60°C–80°C at the rock interface.

Even though the swelling potential of bentonite does not apply directly to the FE, as full saturation is not likely to occur within the 10–15 years planned for the experiment duration, the emplaced material must comply with safety-related requirements (Leupin and Johnson, 2013). This is because, in the FE, the focus was on the THM behaviour of the buffer, which should be as close as possible to that expected for real disposal conditions.

The experience and data from previous experiments, especially HE-E, were used to develop scoping estimates of the key parameters and variables of the FE experiment, including 1) the evolution of temperature on the heater surface, in the bentonite and Opalinus Clay, 2) the degree of saturation and relative humidity in the bentonite and Opalinus Clay, 3) the pore water pressure in the Opalinus Clay, and 4) the strain in the Opalinus Clay. A set of scoping calculations aimed at bracketing the main parameters and independent variables associated with the FE experiment was performed by Ewing and Senger (2012). This exercise supported the design of the FE experiment and monitoring system. Calibration and validation exercises, which included uncertainty analysis, were performed by different modelling teams (e.g., Senger, 2015; Garitte et al., 2014).

4.5 OPHELIE mock-up

The OPHELIE (On-surface Preliminary Heating simulation Experimenting Later Instruments and Equipments) mock-up, conducted at the EURIDICE EIG facilities in Mol (Belgium), simulated a section of a disposal gallery of the Belgian SAFIR-2 reference design (valid in the 1990s) concerning the buffer material and the disposal gallery (full-scale with respect to the diameter, with a length of 5 m). The initial objective of the mock-up was to verify some practical aspects, such as the robustness and performance of the sensors under harsh conditions over several years, and the specification, manufacturing, and placement procedures for the buffer material and its hydration process (Van Humbeeck et al., 2009). The mock-up also served as a preliminary investigation into the buffer THM behaviour and an observation of the evolution, during 4.5 years (1997–2002), of hydration and heating through the monitoring and postmortem analysis program.

The mock-up metallic structure simulated the disposal gallery. The hydration system was maintained at 1 MPa to saturate the buffer. The buffer material consisted of prefabricated blocks of a mixture of 60% FoCa bentonite, 35% sand, and 5% graphite, uniaxially compacted at 61 MPa.

The operational stage of the mock-up consisted of the following phases: hydration at ambient temperature, heating with continuous hydration, and cooling. The heating phase, at 170°C, started 6 months after the start of hydration (when the buffer was mostly saturated and high thermal conductivity was measured) and lasted 4.5 years. The mock-up was then cooled rapidly by switching off all heating elements 6 weeks before dismantling began.

Mineralogical changes observed in the exposed buffer material after 4.5 years of hydration and heating were very limited. The main modifications concerned the presence of calcium sulphate, where the buffer made contact with the central heater, at the interface with the stainless-steel liner at the periphery, and in the joints between the blocks. An enrichment of chemical species toward the central heater (chlorides) was identified, combined with the impoverishment of chemical species, such as bicarbonate or sulphate, toward it.

Overall, from a thermo-hydro-mechanical perspective, the buffer material fulfilled its role of retaining low hydraulic conductivity/permeability and high thermal conductivity. However, the hydraulic conductivity of the exposed material was slightly higher than that of the initial material. All technological gaps were filled with swelling. However, the swelling process was not homogeneous. It mainly occurred close to the liner, whereas the joints between the blocks, although closed, remained visible.

5 Effect of high temperature on buffer

Mineralogical, geochemical, and microstructural alterations in the bentonite buffer owing to thermal, hydraulic, and chemical gradients may impair the safety functions of the engineered barrier. These alteration processes may include the loss of montmorillonite mass, cementation due to the precipitation of secondary minerals, alteration of CEC, and dissolution of accessory minerals. A summary by Villar et al. (2023a) indicates that the most commonly reported alteration processes of bentonite are illitisation via potassium fixation in the interlayer, an increase in smectite layer charge, and the release of silicon (see Leupin et al., 2014, for a review). The main parameters triggering this transformation are temperature and potassium availability, whereas the presence of divalent cations in the interlayer and unsaturated conditions would suppress the conversion. The transformation to chlorite and saponite was also reported (Kumpulainen et al., 2016). In addition, the accessory minerals in bentonite (sulphates and carbonates) will also be affected by the thermal gradient; their dissolution, transport, and reprecipitation can lead to significant salt accumulation close to the canisters (Fernández and Villar, 2010; Kober et al., 2021) and silica precipitation, leading to potential cementation (Svensson and Hansen, 2013; Wersin et al., 2007). The study of these mineralogical and geochemical transformations is important as they may affect buffer performance by altering swelling capacity and retention ability. In particular, sorption occurs on clay (montmorillonite) surfaces, at the edges of clay layers, or in the interlamellar space. Interlamellar sorption sites outnumber edge sites, and therefore, changes in interlamellar sites, described by CEC, might affect both sorption properties and diffusion, as cations diffuse there via a surface diffusion mechanism.

Wersin et al. (2007) reviewed work assessing the performance of the bentonite barrier beyond 100°C. No significant changes in the hydraulic and mechanical properties had been reported for bentonite materials exposed to temperatures of at least 120°C under wet conditions, but the data suggested significant cementation and perhaps illitisation at 150°C and beyond. Natural analogue bentonite samples that showed substantial cementation and illitisation effects still displayed favourable hydraulic properties. Under dry conditions, bentonite was stable to higher temperatures, perhaps 350°C. Hence, the mineralogical transformation of bentonite minerals that are likely to occur at temperatures of relevance (between 100°C and 150°C) seems to be limited even over very long times. Because in repository-relevant conditions, temperatures above 100°C coincide with low water saturation (because of the slow groundwater inflow toward the engineered barrier and the existence of a thermal gradient across it), the already low reaction rates may further reduce.

For example, in the safety assessment of the SKB, the montmorillonite transformation in the KBS-3 repository is assumed to be small based on the following observations and arguments:

1. The time scale for significant montmorillonite transformation at repository temperatures in natural sediments is orders of magnitude longer than the period of elevated temperatures in the KBS-3 repository (e.g., Velde and Vasseur, 1992).
2. The bentonite material is close to mineralogical equilibrium to start with (e.g., Fritz et al., 1984).
3. Transformation is limited by a low transport capacity, principally with respect to potassium (Hökmark et al., 1997) and silica (Karnland and Birgersson, 2006).
4. All published kinetic models, based on both natural analogues and laboratory experiments, indicate that the transformation rate is very slow under repository conditions (e.g., Huang et al., 1993).

The montmorillonite transformation process can also be quantified by modelling. Based on the above description, no mineral transformation is expected to occur faster than illitisation at elevated temperatures. Consequently, the maximum temperature effect was modelled in the safety assessment of the SKB by the kinetic expression for illitisation proposed by Huang et al. (1993) and by using different but realistic potassium concentrations. As long as the maximum temperature is below 100°C, the montmorillonite in the buffer is assumed to be stable for the timescale of the repository.

However, a fully coupled THM-C 1000-year simulation of a nuclear waste repository in a clay formation with a bentonite buffer and a temperature close to the waste canister of 200°C showed some degree of illitisation in both the bentonite buffer and the surrounding clay formation (Zheng et al., 2015). Other chemical alterations detected included the dissolution of K-feldspar and calcite and the precipitation of quartz, chlorite, and kaolinite. In general, illitisation in the bentonite and the clay hostrock was enhanced at higher temperatures but was affected by many chemical factors, mainly the concentration of K in the pore water as well as the abundance and dissolution rate of K-feldspar; the less important factors were the concentration of sodium and the quartz precipitation rate.

Concerning geochemical evolution, Sena et al. (2010) developed a numerical model to simulate the thermo-hydraulic, transport, and geochemical processes observed in the LOT A2 test parcel (Section 4.2). The numerical model developed was a 1D axisymmetric model that simulated the water saturation of bentonite under a constant thermal gradient, transport of solutes, and geochemical reactions observed in the bentonite blocks. A case was modelled considering the highest temperature reached by the bentonite, 130°C and 85°C near the copper canister and near the granitic host rock, respectively. The transport of solutes during the bentonite water saturation stage is considered to be controlled by water uptake from the surrounding groundwater to the wetting front and, additionally, by a cyclic evaporation/condensation process (Karnland et al., 2009a). Once bentonite is water-saturated, solute transport is driven by diffusion. However, the transport of chloride was modelled considering advective, dispersive, and diffusive fluxes, which are believed to have occurred under high temperatures in the LOT A2 test. The computed evolution of bentonite saturation and the simulated transport of chloride were in good agreement with the data measured at the end of the test, reflecting the reliability of the defined conceptual model. The main geochemical processes developed during the LOT A2 test are (Arcos et al., 2006): 1) precipitation/dissolution of carbonate, sulphate, and silica minerals, and 2) cation exchange in the montmorillonite interlayer. The numerical results predicted the dissolution/precipitation of anhydrite, calcite, and silica in the heated bentonite, consistent with the data measured at the end of the LOT A2 test.

Sellin and Leupin (2013) identified the effects of long periods under unsaturated conditions and elevated temperatures on bentonite. The question of how this may influence the subsequent swelling properties after full saturation was discussed by Johnson et al. (2014) based on a literature review. The evidence found was not completely consistent, but showed that a significant reduction in swelling pressure could occur above 110°C–120°C, although in most cases in which swelling pressure was measured, values of more than 50% of initial values were observed. Fully saturated samples exposed to short-term laboratory heating at 150°C experienced small swelling pressure and hydraulic conductivity changes after cooling compared with untreated samples (Dueck, 2014). Furthermore, no significant changes were found after the 6-year LOT study, performed at Äspö (Section 3.2.2), in which the bentonite experienced temperatures up to 130°C (Karnland et al., 2009a).

The considerations exposed above show a need to reduce conceptual and modelling uncertainties, and studies were conducted to assess buffer alteration under thermal gradients and high temperatures with varying water saturation. The effect of temperature on bentonite properties can be studied by analysing changes in preheated material properties or determining bentonite properties at high temperatures. Both approaches were used in HITEC as they complemented each other. In the first instance, the material can be heated in the laboratory (usually in powder form) or obtained from the dismantling of a thermo-hydraulic (TH) cell or an *in situ* test, where samples for postmortem analyses are obtained. The properties of these preheated materials are usually studied at laboratory temperatures and compared with those of nonheated materials to assess changes during operation (Section 5.1). By contrast, the aim of the THM tests performed on nontreated material at high temperatures is to determine the parameters that define the behaviour of the buffer during operation, that is, under high temperatures. These, in turn, could be divided into those intended to determine specific parameters, such as swelling pressure, permeability, or water retention curves (Section 5.2), and those that try to simulate the conditions of the barrier in the repository with respect to the hydraulic and thermal gradients, while following the evolution of the buffer behaviour (Section 5.3). As indicated above, upon dismantling, the latter also provides a preheated material to analyse potential changes in the buffer characteristics or properties.

The following subsections summarise the relevant results obtained from the recent literature and the advances made during the HITEC project (reported in detail in Svensson et al., 2023a; Graham et al., 2023; Villar et al., 2023), grouped according to the classification detailed in the previous paragraph. Acronyms for the institutions performing the work are written along with the authors' affiliations.

5.1 Preheated material

The preheated materials used in this kind of study may come from field or laboratory experiments. Consequently, significant differences in spatial and temporal scales are expected. Furthermore, even among laboratory tests, the scales and treatment conditions may vary significantly. For example, in the studies reported below, bentonite was treated under a thermohydraulic gradient (laboratory and field tests, Sections 5.1.2 and 5.1.3) or heated without simultaneous hydration (Section 5.1.1). Some treatments were performed with dry bentonite and others with wet bentonite, either allowing evaporation during heating or not. These experimental conditions may have affected the observed changes and should be considered when assessing the results.

5.1.1 Laboratory treatment at constant temperature

5.1.1.1 Literature review

Tests in which the material is preheated are usually performed with powdered materials in a dry state or mixed with water at different solid-to-liquid ratios. Tests of this type concerning montmorillonite mineralogical stability were reviewed by Leupin et al. (2014) and classified as follows.

- Smectite-to-illite hydrothermal experiments with MX-80 bentonite at 270°C that reveal no evidence of any illitisation even with variable potassium activity conditions;
- Experiments on thermal stability of montmorillonite at 90°C–150°C, showing montmorillonite dissolving and releasing silica, altering the montmorillonite toward beidellite;
- Experiments on the effect of steam on the swelling capacity of bentonite at temperatures up to 200°C showing that high-temperature water vapour and unsaturated conditions do not cause significant reduction in the water uptake capacity of montmorillonite.

Steam heating is an extreme treatment performed in high-pressure vessels (autoclaves made of stainless steel, titanium, and PEEK) at temperatures much higher than those currently considered in repository concepts (Leupin et al., 2014; Heuser et al., 2014), and is usually carried out using low solid:liquid ratios and previously purified materials. These studies are designed to analyse the illitisation process (which requires high temperature and potassium), cementation (silica precipitation), and loss of swelling capacity (in water uptake or interlayer changes). Results depend on solid:liquid ratio, time, temperature, potassium concentration in pore water, smectite layer charge, or interlayer cations. Owing to the variety of factors affecting results, no general agreement on the effect of steam on bentonite properties exists, although none of the recent studies point to drastic changes, but to slow changes in smectite character (from montmorillonite to beidellite) or layer charge (Wersin et al., 2007; Leupin et al., 2014).

In the framework of the Safe Barriers project (RWM, 2017), the effects of corrosion products, ionic solutions, and heating on the behaviour of bentonite were explored (Davies et al., 2017). Experiments were conducted using 1:10 bentonite suspensions in NaCl, $CaCl_2$, and KCl solutions (up to 1 M) with steel coupons and in constant-volume test cells with compacted bentonite. Data for heated (up to 150°C) samples and those exposed to iron corrosion products yielded lower reductions in swelling indices than those exposed to saline solutions. However, these data do not reflect the

TABLE 1 Summary of changes observed on preheated bentonite.

Bentonite	Solid:liquid ratio[a]	T (°C)	Duration (months)	Observations
Bara-Kade	1:20	150	8–30	Increase of CEC, increase of Ca, reduction in Sr adsorption
Bara-Kade	Dry (open)	105, 150	6	No changes in volumetric compression after 6 months
PBC	Dry (airtight)	150	6, 12	No mineralogical changes, reduction of swelling pressure over time
PBA-22 Extra	Dry (open)	150	3, 6, 18	Decrease of CEC, SSA No changes in swelling pressure or hydraulic conductivity
BCV	Dry (open)	150	6–24	No mineralogical changes. Slight decrease of CEC, liquid limit, SSA, and swell index. Lower Cs distribution coefficients over time. Slightly lower swelling pressure for high-density samples and higher hydraulic conductivity, irrespective of treatment time. Lower water retention capacity
BCV	1:2	150	6–24	No mineralogical changes. Similar or slightly higher values for all the parameters indicated above. Increase of exchangeable Ca over Mg

[a] Dry refers to a solid material dried in an oven that allows evaporation (open) or in a closed vessel (airtight).

impact of highly compacted bentonites on swelling pressure, which is a more relevant safety property than the free-swelling index.

The tests mentioned above generally refer to materials with a low solid:liquid ratio, which may not be fully representative of the actual repository conditions. Tests using compacted bentonite with much higher solid:liquid ratios (up to 1:50) were performed by Valter and Plötze (2013), with MX-80 bentonite with different degrees of saturation stored at different temperatures in a closed system. They showed a high mineralogical stability but considerable changes in physicochemical properties, particularly above the critical temperature of 120°C. The CEC decreased during heating at 150°C by approximately 10%. The specific surface area decreased by more than 50%. The water vapor adsorption ability dropped by 25% already within 3 months at 120°C. These changes were mostly related to variations in the interlayer cation composition (a slight conversion from sodium to an alkaline-earth form of bentonite) and smectite aggregation processes.

5.1.1.2 Progress during HITEC

In the context of HITEC, several laboratories preheated bentonite under different conditions (detailed in Supplementary Table S9). The findings for the different bentonites analysed, reported by Svensson et al. (2023a), are briefly discussed below, and a summary is given in Table 1. With respect to previous investigations, the stress during HITEC has been on the differences between heating conditions, dry or wet.

5.1.1.2.1 Wyoming-type bentonite. The University of Helsinki studied the impacts of increasing heat load (150°C, up to 3 years) on a Na-Wyoming bentonite buffer material (Bara-Kade, used as supplied), heating vessels with solid:liquid ratio 1:20 and using reference water for the Onkalo site in Finland (Vuorinen and Snellman, 1998). Sr sorption isotherm experiments and extended X-ray absorption fine structure (EXAFS) analyses were combined to provide a quantitative and mechanistic description of the effects of elevated temperatures on bentonite/radionuclide interactions. Visible bentonite alterations with increasing heat treatment time were seen in the XRD patterns (Figure 3, left), namely, an increase in the basal reflection from 10.27 Å to 12.13 Å over 30 months, which is consistent with a decrease in exchangeable Na^+ and an increase in Ca^{2+}. The CEC increased after 22 months of heat treatment (from 91 to 98 meq/100 g), in contrast to previous studies (e.g., Kaufhold and Dohrmann, 2010). In Sr-90 sorption isotherm experiments (Figure 3, right), a large difference in Sr uptake in neutral, pH 8 (approx. 40%) vs alkaline, pH 13 (approx. 90%) regimes, with slightly lower uptake (~5%) on heat-treated bentonite than raw. Although the observed thermal alterations in bentonite—changes in basal spacing and cation exchange properties—can influence the strength and mode of radionuclide retention, Sr adsorption was largely unaffected following heat treatment (22 months, 150°C). This was further supported by EXAFS analyses, which showed that outer-sphere sorption dominated at pH 8, whereas inner-sphere sorption prevailed at pH 13. However, no observable changes in the Sr coordination environment were detected following heat treatment. These results suggest that despite measurable physicochemical alterations, the fundamental retention mechanisms remained stable under the tested conditions. Further insights into the site-specific sorption processes can be gained through thermodynamic surface complexation modelling.

The same type of bentonite was used by VTT to perform volumetric, isotropic compression and shear tests in powder samples preheated at 105°C and 150°C for 6 months. The treated samples were wetted either by water vapor equilibrium (to minimize dissolution of minerals and cation exchange) or by mixing with liquid water (much faster) to water content of 17%, which is the bentonite average installation moisture in the POSIVA disposal concept. Afterwards, the samples were isotropically compacted to a dry density of 1.6 g/cm^3 following compression-decompression cycles (ranging from 0 to 12 MPa) with simultaneous measurement of volume changes. This procedure provided data on volumetric elastoplastic behaviour. The results indicated that the heat treatment of the powder did not affect the volumetric compression properties of bentonite.

5.1.1.2.2 Ukrainian bentonites. The Ukrainian sodic bentonite PBC was heated at 150°C for 12 months with no water addition and under airtight conditions, for which reason the water content of the clay remained equal to the initial value during heating (7.4%). A

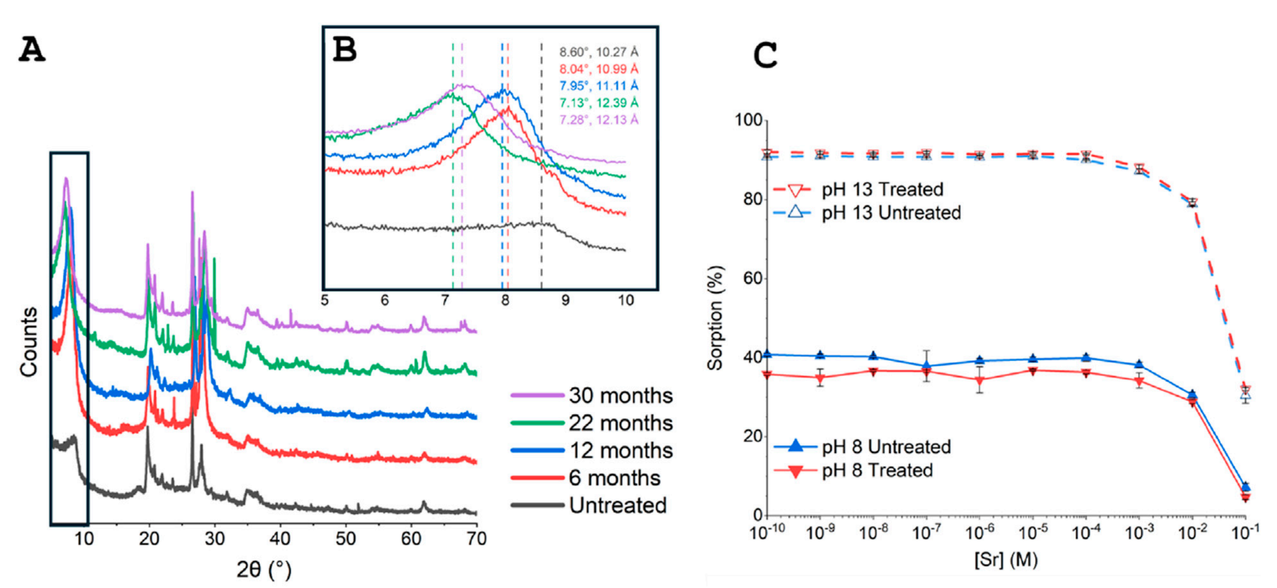

FIGURE 3
(A) Changes in XRD patterns obtained with a Cu Kα-radiation source for unaltered bentonite (black) vs 150°C heat-treated systems (coloured lines); **(B)** changes in the basal reflection (marked with vertical lines), values in Å given in colours; **(C)** Sr isotherms for treated (22 months, 150°C) and untreated bentonite at pH 8 and 13.

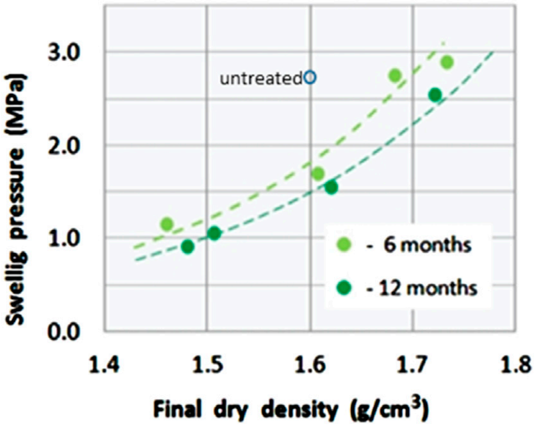

FIGURE 4
Swelling pressure of the Ukrainian PBC bentonite compacted to different dry densities after treatment at 150°C in dry conditions for different periods of time.

comparison of the XRD data for the initial bentonite and after heat treatment showed that no phase transformations occurred, and the weight ratios of the mineralogical phases (quartz, calcite, and montmorillonite) remained constant. The powder material was compacted after treatment at different dry densities with a water content of 7.5%, and the swelling pressure was measured at room temperature. A reduction in the swelling pressure of the preheated samples was observed, particularly for those heated for a longer time (Figure 4).

Another Ukrainian bentonite, of reference PBA-22 Extra, was also submitted to heating at 150°C under dry and wet conditions (Zlobenko et al., 2023). The samples heated under dry conditions lost the interlayer water (decrease in the (00l) reflections with heating time), which was restored when montmorillonite was saturated with water. After 3 and 6 months of heating, the CEC decreased to 79 and 73 meq/100 g, respectively, whereas the BET specific surface area decreased to 90–100 and 75–80 m^2/g, respectively. The influence of temperature on the swelling pressure and hydraulic conductivity of Ca-bentonite after dry processing was negligible within the limits of the measurement errors.

5.1.1.2.3 Czech bentonite. The Czech bentonite BCV was heated in powder form at 150°C for periods of time between 6 and 24 months under dry (with evaporation allowed) and wet (1:2 solid:liquid ratio in a hermetic vessel) conditions. This preheated material was subsequently analysed by CTU and ÚJV at room temperature. The dry material was allowed to cool down before testing, whereas the wet material was dried at 60°C for 2 weeks, ground into a powder similar to the dry material, and tested.

No significant changes in mineralogical composition were detected by ÚJV using XRD, the percentage of smectite in the illite/smectite mixed-layer remained the same (80% ± 5%) in all samples. After heating in wet conditions, the basal reflection changed from 14.63 to 15.19 Å. A slight decrease in the CEC of the dry samples and a slight increase in the wet samples were detected, which correlated well with the slight decrease in the specific surface area observed in the samples treated under dry conditions and the increase observed in the samples treated under wet conditions (Supplementary Table S10). In dry samples, Mg^{2+} prevailed as exchangeable cation over Ca^{2+}, whereas the opposite occurred in wet samples. Given that the mineralogical composition and illite/smectite ratio did not change significantly, these reduced values were attributed to changes within the montmorillonite layer

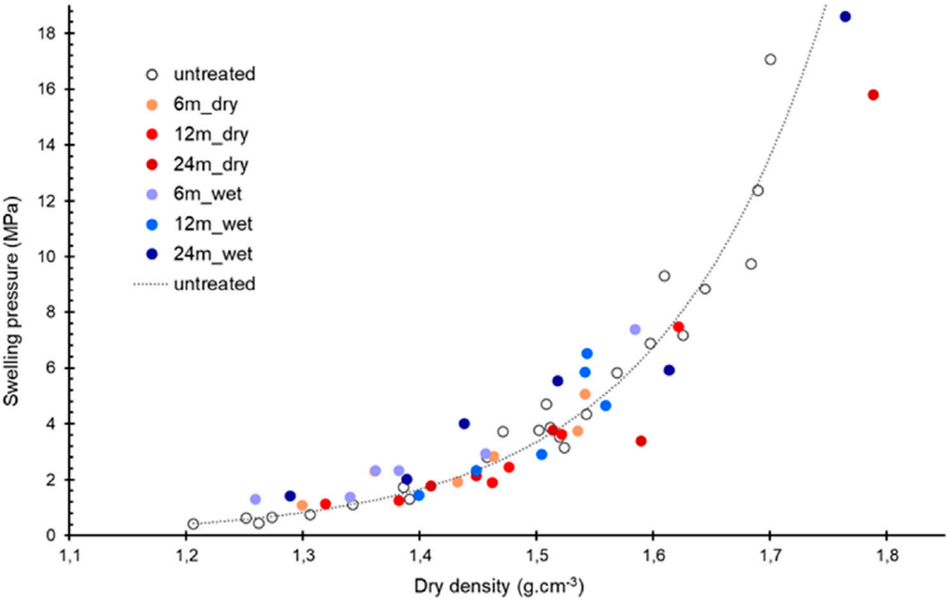

FIGURE 5
Swelling pressure of BCV bentonite preheated in dry and wet states for 6, 12, and 24 months.

structure, which were likely due to the collapse of some of the montmorillonite interlayer spaces. Lower Cs distribution coefficients were found for the dry-heated samples as the treatment time increased, which was attributed to an actual reduction in the sorption sites.

Swelling pressure and hydraulic conductivity were measured in samples compacted at different dry densities (from 1.4 to 1.8 g/cm^3) by CTU, CU and ÚJV. The swelling pressure of the dry treated bentonite was not affected by elevated temperature in the low dry density range, but lower swelling pressure was observed at dry densities higher than 1.6 g/cm^3, although possibly within the variability range (Figure 5). By contrast, the values measured for the wet-treated samples were in the upper range of the expected values. No decrease in the swelling potential due to thermal treatment was observed in the samples that were dry-heated for 12 months. The hydraulic conductivity increased with respect to the reference values for all dry densities in samples treated under dry conditions, irrespective of the treatment time, and remained approximately the same (or even lower) for the wet-treated samples (Supplementary Figure S2). Consistent with these results, the liquid limit, specific surface area, CEC, and swell index of the dry-treated bentonites were lower than those of the untreated bentonites, whereas they were similar or slightly higher for the wet-treated samples (Supplementary Table S10).

The water retention curve was determined for preheated samples compacted to dry densities of 1.4, 1.6, and 1.8 g/cm^3 in cells with perforated lids (30 mm diameter, 15 mm height) placed in a high-relative-humidity atmosphere. A sensor placed inside the sample allowed to check the suction variation as the sample was hydrated and couple it to the water content estimated from the cell weight changes. A drying path was followed by placing the samples in a desiccator containing silica gel. Figure 6 shows the obtained results. The samples heated under dry conditions had a lower water retention capacity than those heated under wet conditions, particularly at higher suctions. In addition, they exhibited a larger hysteresis between the wetting and drying paths. The thermal loading time had only a minor effect.

5.1.2 Material subjected to thermal gradient in laboratory cells

A closer approach to the actual conditions in the repository than the preheating tests described in the previous section is the thermohydraulic tests in cells. In these, the compacted buffer material is simultaneously subjected to heating and hydration in opposite directions to simulate the interaction of the water coming from the host rock and the thermal gradient generated by the heat emitted by the wastes in the canisters. After dismantling the tests, postmortem analyses of the treated buffer materials allow to observe changes in their properties and performance. In the framework of HITEC, TH tests were performed using Wyoming-type and BCV bentonites and heater temperatures above 140°C (see Section 5.3 for a more detailed description and Supplementary Table S11 for a summary of their characteristics). The modifications detected in the bentonite due to the TH treatment are summarised below.

Two sets of thermohydraulic tests were analysed by CIEMAT and UAM. The tests involved heating the base of a bentonite column while water was injected to the top for 2.5 and ~9 years. These durations were significantly longer than those of previous similar laboratory tests at temperatures higher than 100°C (described in Section 5.3.1); hence, the samples analysed at the end of the tests were treated for a much longer time. In one set of tests, blocks of compacted MX-80 powder (10 cm × 10 cm) were used and the heater was set at 110°C. The cells were hydrated using saline and dilute water (tests C4 and C5, respectively) (Villar et al., 2021). In the other set of TH tests (consisting only of test HEE-B), the height of the column was 50 cm, and the heater was set at 140°C (Villar et al.,

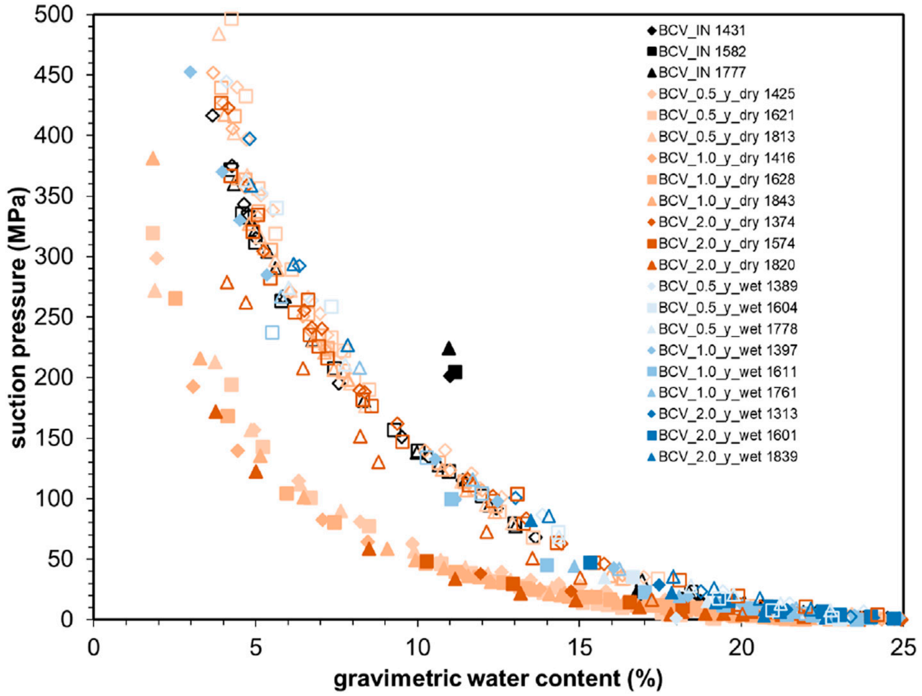

FIGURE 6
Water retention curves of the reference BCV bentonite (IN) and the bentonite preheated at 150°C under dry conditions for 6 and 12 months (0.5_y and 1_y) compacted at different dry densities (indicated in the legend in kg/m^3) [(]modified from Kašpar et al. (2021)].

2022; Villar et al., 2023a). It was saturated with saline water, and MX-80 pellets were used.

In tests C4–C5 no significant montmorillonite structural modifications occurred during the operation. Octahedral Mg increased near the heater and, in all samples, the layer charge decreased with respect to the values of the reference sample. Mineralogical changes were only observed in areas where the temperatures were higher than ~60°C and particularly in the few millimetres closest to the heater. Calcium sulphate (probably anhydrite) was detected in areas less than 4 cm from the heater. The total consumption of oxygen appeared to have occurred in both experiments, explaining the presence of pyrite and oxides in different oxidation states close to the heater in the case of C4. As a result of hydration with saline water in cell C4, carbonates and sulphates precipitated at various locations, NaCl-spotted areas were observed close to the heater, and corrosion occurred upon contact with the steel elements. None of these features were observed in cell C5 hydrated with glacial water. In this cell, calcium carbonate precipitated close to the hydration surface.

The changes in the pore water composition inferred from the aqueous extracts were affected by 1) the composition of the incoming water, 2) the dissolution of mineral species present in the initial bentonite as a result of the increase in water content, and 3) cation exchange processes. The ions coming with the hydration water and those coming from the dissolution of minerals were transported by advection and accumulated in the lower half of the columns at the bottom, precipitating closer to the heater. The overall ion concentration in the samples increased significantly with respect to the initial concentration. The maxima in sulphate concentration were accompanied by increases in soluble sodium and calcium content, indicating the precipitation of anhydrite at less than 4 cm from the heater. The precipitation of carbonates close to the heater was reflected in the decrease in bicarbonate content from the hydration surface toward the heater.

At the end of the HEE-B test, all samples continued to consist predominantly of dioctahedral smectite with no significant differences from the original with respect to the distribution of structural cations and layer charge. Sodium still predominated in the exchangeable cation complex, probably because of hydration with a predominantly sodic solution. However, some contributions from divalent cations toward the hydration surface were detected. Despite the lack of montmorillonite alteration at the structural level, the drier samples, those that were submitted to temperatures >60°C, had a significant resistance to rehydration under room relative humidity conditions. However, this was not an irreversible process because the samples hydrated normally when the relative humidity was high (97%) and expanded as expected when suspended in water.

Although hydration took place with highly saline water, which seemed to have inhibited the formation of colloids, the overall increase in water content allowed the dissolution of some species, and the solubilised ions were transported toward the heater and precipitated at two distinct areas: sulphate, sodium, and calcium peaked at ~18 cm from the heater, whereas chloride moved closer to the heater (accompanied by sodium and calcium), concentrating at 9 cm from it, coinciding with a vapor leak area. This leakage started at an undetermined moment during the test through a sensor inlet. This experimental artifact seems to have conditioned the surrounding processes, such as the movement of solubilised ions. The liquid water availability was probably also affected in areas with temperatures higher than 60°C, which would limit the reactivity there. Nevertheless, evidence of precipitation of calcite and calcium sulphates and dissolution of cristobalite and quartz were observed in areas where the temperature was higher than 100°C (Villar et al., 2023a).

Around this sensor inlet, the specific surface area of the bentonite was the lowest, indicating the aggregation of particles likely caused by shrinkage when water evaporated from the bentonite, whose water content was higher prior to the start of the leak. From this point toward the heater, the bentonite was disaggregated with specific surface areas smaller than the initial one. Lower external specific surface areas were systematically observed in other TH tests toward the heater. An extremely low value of 5 m^2/g was measured in test HT2 (described in Section 5.3), which was conducted with Bara-Kade bentonite under the same conditions and for the same duration as those of test C4 (Wyoming-type bentonite, hydration with saline water, 2.5°years) but at relatively higher heater temperatures (150°C vs 110°C).

The swelling pressure and hydraulic conductivity of the undisturbed samples were determined at the end of the TH tests. Both parameters were related to the dry density following the trend expected for untreated bentonite, irrespective of the type of hydration water or the duration of the TH test (Villar et al., 2021; Villar et al., 2022). The slightly lower swelling pressure after the TH treatment was attributed to the higher initial water content of the samples. The hydraulic conductivity values after the TH treatment were below the ones obtained for the untreated bentonite, which could be explained by the “maturation” of the microstructure during the thermo-hydraulic treatment, with the average pore size becoming smaller and more homogeneous over time. However, these observations only apply to samples that reached a high degree of saturation at the end of the TH treatment and were subjected to a maximum temperature of ~100°C (cells C4 and C5) and even lower (~34°C) for cell HEE-B. The BCV samples treated by CTU for much shorter times (<1 year) at temperatures of up to 150°C showed no changes in hydraulic conductivity and liquid limit with respect to the untreated bentonite, provided that the postmortem degree of saturation was high. By contrast, the samples that retained a low degree of saturation during the TH treatment period showed higher postmortem hydraulic conductivity and a lower liquid limit, in agreement with the results obtained for the oven-dried samples (section 5.1.1). No changes in the swelling pressure were observed.

5.1.3 Material subjected to thermal gradient in large-scale tests

The dismantling of large-scale *in situ* tests as those described in Section 4, provides relevant postmortem information regarding the modifications experienced by the barrier material and the processes that occurred during the operation. In the SKB field experiments at Äspö using various bentonites, at least the following mineralogical and geochemical processes related to the thermal gradient were identified:

- Stiffening of the buffer (LOT, Karnland et al., 2009a).
- Accumulation of non-exchangeable magnesium toward the heater (LOT, ABM, Prototype, e.g., Karnland et al., 2009a; Olsson et al., 2013; Svensson, 2015).

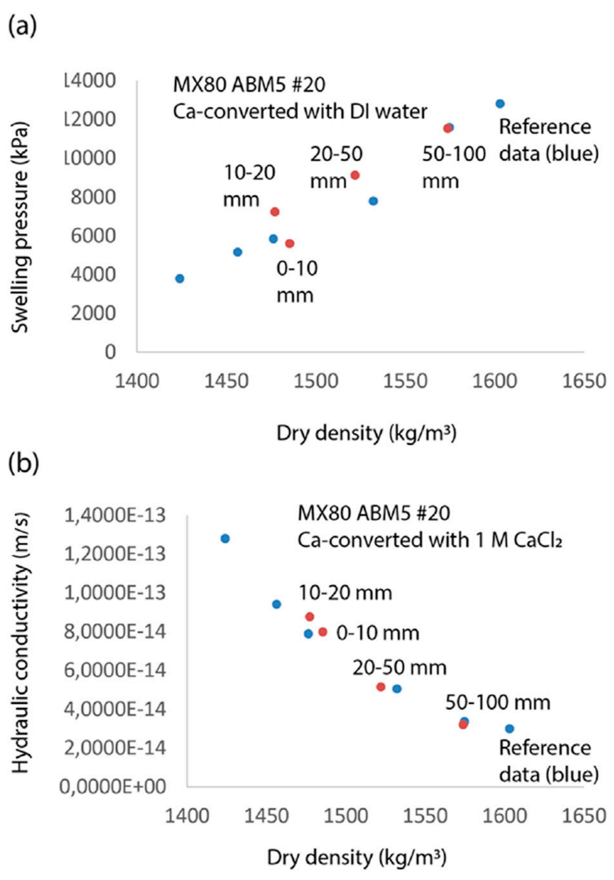

FIGURE 7
Swelling pressure and hydraulic conductivity of MX-80 samples of the ABM5 #20 block (red symbols, distances to the heater indicated in mm) and of the reference bentonite (blue symbols). The samples were first Ca-exchanged in 1 M $CaCl_2$ solution and then **(a)** tested with DI for swelling pressure, **(b)** tested with 1 M $CaCl_2$ for hydraulic conductivity.

- Dissolution of cristobalite close to the heater (e.g., Svensson and Hansen, 2013).
- Formation of oxygen-sensitive Fe(II) phases at the steel/bentonite interface (Svensson and Hansen, 2013).
- Formation of ferrosaponite/saponite associated with a large increase in Mg (ABM2 with FEBEX bentonite, Svensson, 2015).
- Accumulation of $CaSO_4$ (gypsum/anhydrite) (LOT, Prototype, and ABM).
- Local accumulation of NaCl (halite) (ABM2).
- Physical disintegration of bentonite blocks in some parts along with halite accumulation (ABM2).

Work during the HITEC focused on materials obtained from the ABM5 parcel, which was retrieved in 2017 after 4.5 years of heating. The temperature in the buffer was initially below 100°C, and subsequently stepwise increased to 100°C–150°C (depending on the location) and finally to 150°C–250°C for nearly1 year (Figure 2). This high temperature has not been achieved in any other related field test. The changes in the mineralogy, geochemistry, and hydromechanical properties of four different bentonites (Section 3.4) were analysed by various laboratories. The SKB made the following observations (Svensson et al., 2023b).

- No general significant alteration of smectite in the bentonite was observed.
- Minor changes were detected in the CEC. Cation-exchange reactions made the distribution of Na and Ca more homogenous after the experiment. However, these changes did not seem to be correlated with smectite transformation or have any significant impact on important properties of bentonite, such as swelling pressure or hydraulic conductivity.
- In fact, the swelling pressure and hydraulic conductivity were unaffected by extremely high temperatures in the ABM5 experiment. Figure 7 shows results for some samples retrieved after Ca homoionisation.
- Corrosion products were observed at the iron/heater interface. Dissolution precipitation of sulphate and cation-exchange reactions were observed.
- Minor Mg redistribution was occasionally observed; however, very small or no amounts of trioctahedral smectite were found.
- Unexpectedly, extensive physical breakdown of some of the bentonite blocks occurred, which was attributed to the extremely high temperature conditions during the experiment (Supplementary Figure S1).

Samples from ABM5 were also analysed by BGR. Elemental profiles were obtained (Kaufhold et al., 2021) and compared with previously measured results. Most of the ABM5 blocks exhibited a slight increase in sulphur content in the central part and some dissolution in the inner and outer parts. This behaviour was similar to the results found in the LOT A2 experiment (Karnland et al., 2009a) but was much less pronounced. The redistribution of S and C phases, probably caused by dissolution and precipitation, did not depend on the type of bentonite. The same bentonites installed at different depths showed different profiles, indicating that the local conditions were more important than the material properties. No general trend was detected when comparing the dry density, water content, and degree of saturation with the chemical and mineralogical changes. On average, more chloride was found in the more saturated lowermost part. Most of the bentonites had an initial Cl^- content of <0.1 mass% that increased in the reacted blocks because of groundwater input. Iron increased at the contact with the heater owing to corrosion, although the increase was smaller than that in previous tests, which could be partially explained by the differences in the sampling depths among the different tests (the sampling depth was larger in the samples from ABM5 because of the relatively small area close to the heater). Simultaneous thermal analysis (STA) proved that calcite was involved in corrosion, leading to the formation of siderite at the expense of calcite. STA also showed that pyrite was sometimes preserved and sometimes oxidised, depending on both the local conditions and the characteristics of the pyrite. Magnesium enrichment was observed close to the heater, as was the case in many other large-scale tests, regardless of whether an iron or copper heater was used. In previous tests, Mg enrichment was observed along with the formation of trioctahedral domains, which were detected based on the intensity of the d_{060} XRD reflection of smectite. However, in the ABM5 test, no sign of trioctahedralisation was found. For the first time, clay, originally containing trioctahedral smectite (saponite), was used in a large-scale test and exhibited a reduced Mg content at the heater.

In summary, based on the chemical and mineralogical analyses performed on the ABM5 material and the comparison with other tests, it was not possible to detect any specific high-temperature reaction. Moreover, most of the changes, for example, Mg enrichment, corrosion, and trioctahedralisation, were less pronounced compared to other tests conducted at lower temperatures but for longer times with higher water contents.

5.2 Determination of properties at high temperature

The determination of thermo-hydro-mechanical properties (permeability, swelling, water retention capacity, strength, etc.) at high temperatures generally involves technical challenges that may primarily arise from:

- The materials used to manufacture the test cells and their reactions upon heating involve calibration issues:
- Seals that are not compromised at high temperatures are required;
- Adequate sensors, which must be suitable not only for high temperatures but also, in most cases, for harsh, aggressive environments boosted by heat, are required.
- undesirable behaviour of the testing systems, such as vapor leaking.

Because of these concerns, testing of THM properties at temperatures higher than 100°C has rarely been performed or reported in the literature. Newly developed oedometers for testing soils are designed to work at temperatures of up to just 70°C (e.g., Kirkham et al., 2020). Daniels et al. (2017) identified limitations for assessing gas flow at temperatures above 150°C, and concluded that further development of the apparatus is required to ensure robust data collection at high temperatures.

Nevertheless, Yoon et al. (2023) recently reported measurements of thermal conductivity of a Ca-type compacted bentonite (Gyeongju, South Korea) with different water contents at temperatures of up to 150°C. The values obtained at this temperature are similar to those obtained at room temperature.

Other hydromechanical properties are discussed in the following subsections, particularly the advances made in the framework of HITEC (Graham et al., 2023), both in terms of equipment development and process understanding.

5.2.1 Permeability and swelling

5.2.1.1 Literature review

The effect of temperature on the permeability, swelling capacity, and swelling pressure of compacted bentonite has been tested for many years (see, for example, Villar and Lloret (2004) for an initial review and Chaaya (2023) for a recent one). This effect may differ depending on the type of material and even on the type of cations in the exchange complex. Temperature-induced transfers between intra-aggregate adsorbed water, which has a higher density than that of free water, and inter-aggregate free water have been invoked (see also Johnson et al., 2014).

Although most experimental evidence (including that obtained in HITEC and summarised below) reports a decrease in swelling pressure with temperature, molecular dynamics simulations of the effect of temperature on the swelling pressure of sodium smectite showed a gradual increase in swelling pressure with increasing temperature from 27°C to 327°C, which was attributed to the increase in the thermal motion of water molecules (Akinwunmi et al., 2019). In these simulations, the increase in swelling pressure was higher at high dry densities over the entire temperature range.

Jadda and Bag (2020) determined the swelling pressure of two Indian bentonites at elevated temperatures (up to 95°C) using three different procedures (isothermal, incremental, and decremental) and found that the effect of temperature on swelling depended on the methodology followed. In isothermal conditions, the swelling pressure increased for divalent bentonite with increasing temperature, whereas it decreased for Na-bentonite. The measured swelling pressure increased for both bentonites under increasing temperature conditions, whereas a significant decrease in the swelling pressure was observed under decreasing temperature conditions. According to these authors, the predominant exchangeable cations were responsible for the differences in the measured swelling pressures of the two bentonites.

Ruan et al. (2022) discussed different factors that may affect the swelling pressure values obtained at high temperatures, such as the liquid injection rate, system compliance, and side of water injection (from the top or bottom of the sample). They tested various bentonites compacted at a range of dry densities between 1.1 and 1.85 g/cm^3 at temperatures of up to 80°C. The swelling pressure evolution curves obtained followed the usual pattern observed by many authors at both room and high temperatures: an initial quick development followed by a decrease that is recovered and eventually ends at the equilibrium swelling pressure value, giving rise to peak, valley, and equilibrium swelling pressure values. They observed that the effect of temperature on the swelling behaviour depended on the clay fabric. For the powdered bentonite, the swelling pressure values at the end of different stages increased with temperature, whereas for the granular bentonite, these values decreased. The authors considered that these observations resulted from competition between interlayer swelling, interparticle swelling, free water expansion, and mineral thermal expansion.

Conversely, the temperature increases the hydraulic conductivity, although, in most cases, this increase cannot be explained solely by the increase in water kinematic viscosity; other mechanisms, such as microstructural or pore fluid chemistry changes, have been invoked (e.g., Villar and Lloret, 2004).

In a suite of isotropic and constant-volume tests with MX-80 bentonite in custom-made cells, researchers at the BGS found a trend for increasing intrinsic permeability by up to an order of magnitude with increasing temperature up to 120°C (Zihms and Harrington, 2015), which was reversible after cyclic thermal loading. The intrinsic permeability remained below 10^{-20} m^2. Above 150°C, large increases in permeability were seen which were attributed to sample thermal contraction enabling flow along the sides, but uncertainties remained in the expected amount of vessel expansion and steam generation (Daniels et al., 2017).

5.2.1.2 Progress during HITEC

Building on the work by Daniels et al. (2017) at BGS, during HITEC, constant volume cells allowed the observation of radial and axial swelling pressure evolution at multiple locations when compacted MX-80 bentonite was subject to high temperature. Testing began with samples very close to or fully saturated and constrained by the apparatus, either under a constant volume condition or with radial constraints and a limited degree of axial expansion permitted (to explore the impact of heat on density and water content homogenisation). Deionised water was used to prepare the samples and saturate them during the tests. Two types of tests were conducted: 1) swelling pressure tests, in which a constant water pressure of 4.5 MPa was applied to the sample for 50–100 days, and 2) combined hydraulic conductivity and swelling pressure tests, in which the sample was subjected to a water pressure gradient of 2000 kPa (with 4000 kPa injection pressure and 2000 kPa downstream pressure) and subjected to a series of thermal cycles. The key findings of the swelling pressure tests were as follows:

- Even at high temperature (90°C–100°C), the bentonite was able to swell and completely fill the void space when axial strain was allowed, exerting a measurable swelling pressure (Daniels et al., 2021), which was observed to be lower toward the lower density end and at higher temperature (Figure 8, left). Significant heterogeneity remained in most of the observed swelling pressure distributions, which were greater in samples that were allowed to swell axially. In agreement with this observation, no homogeneous state was reached in terms of dry density or water content by the end of the test.
- Measured swelling pressure was found to reduce with increasing temperature (100°C–200°C) for dry densities higher than 1.6 g/cm^3, irrespective of the axial constraint conditions, but for low dry densities (1.3 g/cm^3), swelling pressure was relatively insensitive to temperature. Furthermore, when the swelling pressure was reduced ($T >$ 100°C and higher dry densities), notable temporal degradation was apparent (Figure 8, right). Although both axial and radial stresses were still measurable at high temperatures, such as at 200°C, these values reduced substantially by ~5 MPa between 100°C and 175°C at a dry density of 1.7 g/cm^3.
- During the thermal cycling tests, the observed reduction in swelling pressure was hysteretic and not completely recovered on returning to the initial temperature of 100°C.

Permeability tests were performed in samples that had a relatively high saturation before being allowed to fully hydrate once exposed to a temperature of 100°C. Permeability was then measured at temperatures up to 200°C. A minimal change in the hydraulic conductivity was observed with increasing temperature, despite the measured impacts on the swelling pressure.

One possible explanation for the observed reduction in swelling pressure at higher dry densities would be the thermally induced yield of the sample between temperatures of 100°C–200°C (Graham et al., 2023). Consequently, contraction of the bentonite would result in a drop in stresses under a constant volume condition and is consistent with only a minimal increase in hydraulic conductivity, which can be explained by the flow of water along the sides of the sample. Nevertheless, they suggested that additional testing is necessary to further explore this process, as well as to observe the swelling pressure reduction with time.

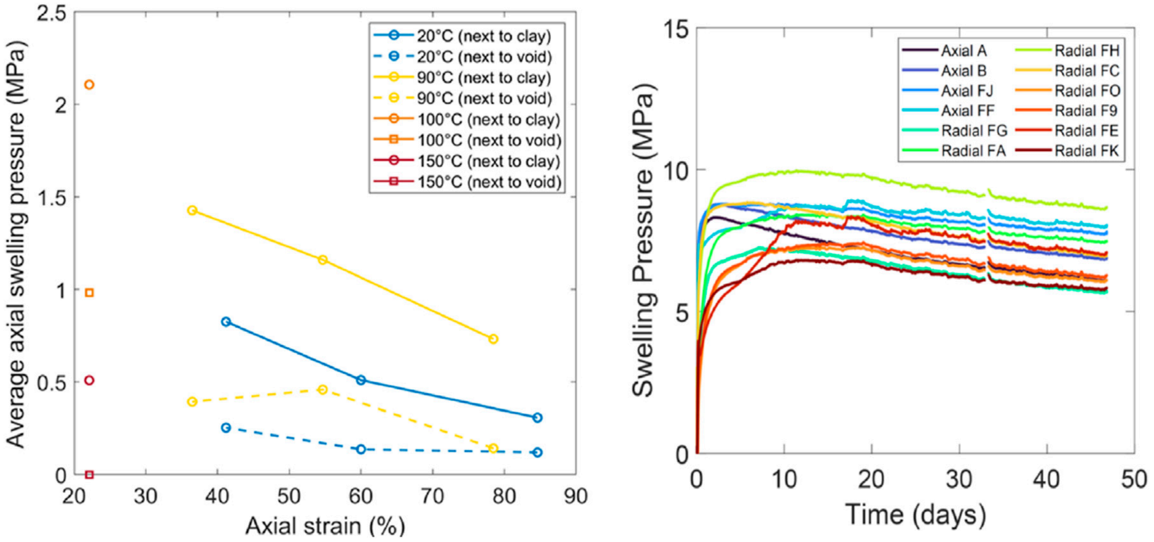

FIGURE 8
Average swelling pressure after 100 days, as a function of axial strain resulting from swelling into remaining void of MX-80 samples initially compacted at 1.7 g/cm^3 (left) and swelling pressure evolution of MX-80 bentonite compacted to dry density 1.7 g/cm^3 and tested at T = 175°C with a water injection pressure of 4 MPa as measured by sensors located at different sample locations (right) (results of BGS in Graham et al., 2023).

X-ray tomography was used at JYU and GTK to monitor wetting and swelling of temperature-controlled Bara-Kade bentonite samples (from 20°C to 130°C). The samples were held in a constant cylindrical volume and wetted with simulated Olkiluoto groundwater (Vuorinen and Snellman, 1998) from a container in the upper part of the cell. The water was not pressurised, but for the test at 130°C, the container was sealed, which would make the pressure in it increase to 270 kPa. The sample's diameter and height were 42 and 21 mm, respectively. Venting at temperatures above 100°C allowed evaporation of water through the venting valve, which inhibited saturation of the sample. Thus, the valve was closed during the tests at 130°C and was only occasionally opened. During the wetting process, the swelling pressure was monitored using force sensors, and X-ray tomography, combined with image analysis, was used to track water transport and deformation, enabling a detailed analysis of the 3D spatial and temporal evolution of the local partial densities of dry bentonite and water.

The X-ray tomography method is based on measurements of changes in the local values of linear X-ray attenuation coefficients. A key part of the analysis is the measurement of internal displacements based on digital image correlations, which is performed using a block-matching method based on phase correlation (Harjupatana et al., 2015). In practice, displacement field computations were performed using pi2 software (https://github.com/arttumiettinen/pi2). To determine an accurate quantitative estimate, a careful calibration procedure must be performed based on known values of the initial and final average water content of the samples. These initial and final average water contents were determined gravimetrically. The selection of voxel sizes (192 × 192 × 192 μm at JYU and 169 × 169 × 169 μm at GTK) aimed to optimize both displacement and density measurements. The voxel size must be sufficiently small to accurately measure displacements in clay as well as sufficiently large to ensure a good signal-to-noise ratio for density measurements during the wetting process.

Water transport was faster at higher temperatures, with saturation achieved more quickly as temperature increased. Figure 9 shows the spatial distribution of water content inside samples (some artifacts from analysis cannot be ruled out). The temporal evolution of saturation profiles suggested diffusion may contribute to transport alongside advection, as no clear waterfront was visible. In fact, effective diffusion coefficients were computed from the average degree of saturation, and the values obtained allowed for a reasonably good reproduction of the estimated profiles of the degree of saturation using a simple 1D diffusion model. However, these coefficients should depend on water content, and the roles of advection and water vapor diffusion should be considered.

The development of axial pressure on both the top and bottom of the samples was faster at higher temperatures and presented a non-monotonous pattern usually observed in bentonite, as described above: an initial increase followed by a decrease that eventually leads to the final steady values. The initial peak was sharper at higher temperatures. The preliminary results indicated that the effect of temperature was more significant for higher dry densities.

The swelling pressure and hydraulic conductivity of the Czech bentonite BCV were by CTU and CU. In the first case, the compacted samples were 30 mm in diameter and 20 mm high. They were initially saturated with distilled water injected at a pressure of 1 MPa. The permeability was measured once steady flow was reached, and after releasing the water injection pressure, the swelling pressure was measured. Afterward, the temperature was gradually raised up to 130°C and the measurement of permeability and swelling pressure was repeated for each temperature step. When the temperature increased from 20°C to 130°C, total pressure decreased from 1.5 to 0.8 MPa for a bentonite dry density of 1.39 g/cm^3, from 1.8 to 0.7 MPa for a bentonite dry density of 1.45 g/cm^3 and from 4.6 to 3.1 MPa for a bentonite dry density of

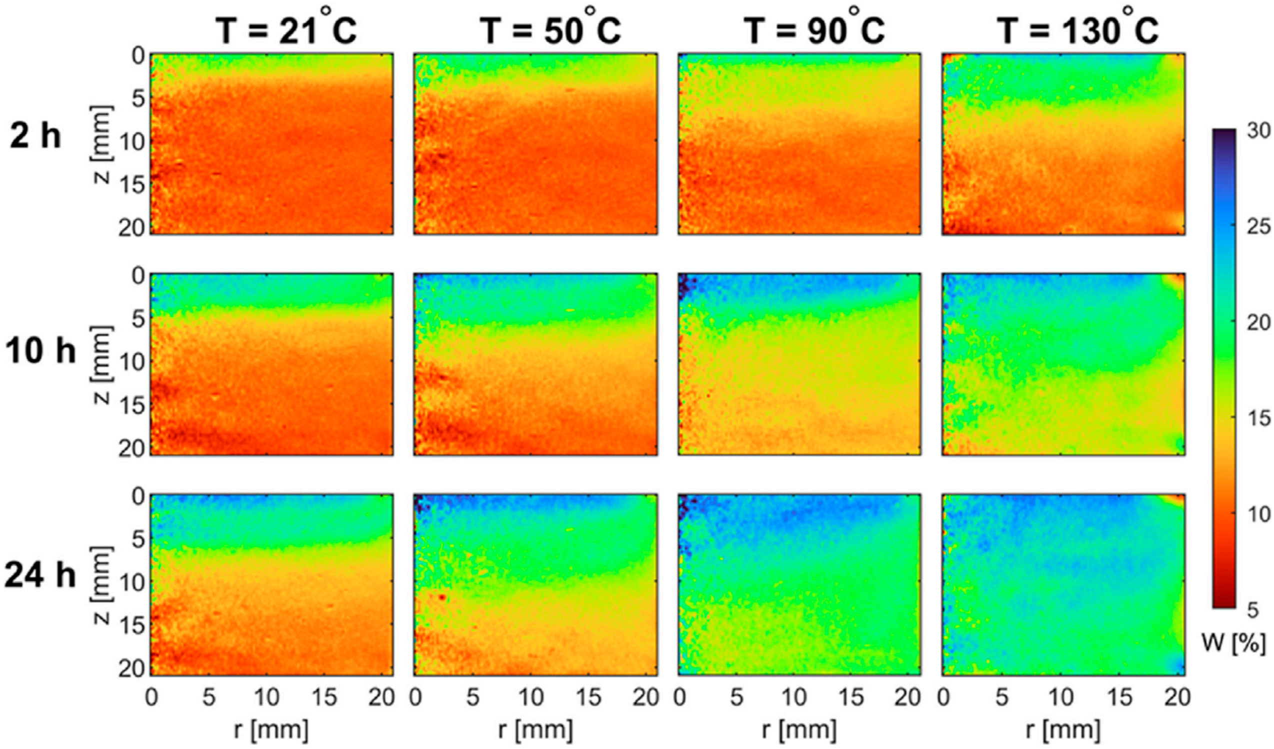

FIGURE 9
Azimuthally averaged water content (obtained by X-ray tomography) at four temperatures and three times after the start of wetting of a Bara-Kade sample compacted at dry density 1.65 g/cm^3.

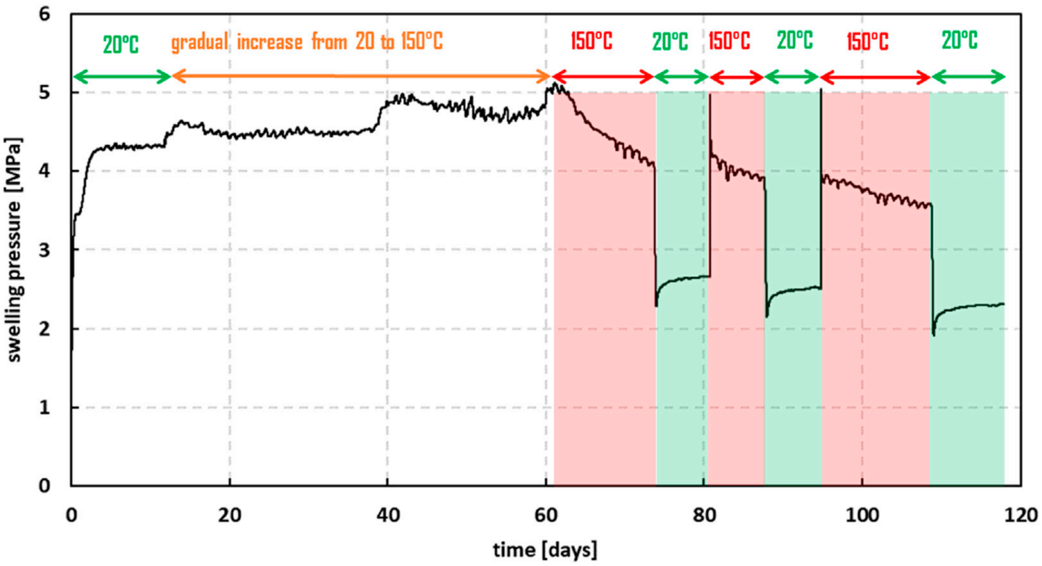

FIGURE 10
Evolution of axial stress measured during saturation and thermal cycles in a BCV sample compacted at dry density 1.6 g/cm^3 (CU).

1.54 g/cm^3. In all cases, the decrease became more significant as the temperature increased. Remarkably, for each temperature, the axial pressure did not reach a stable value but decreased continuously, particularly at the highest temperature. After cooling, the swelling pressure did not reach its original value. As expected, hydraulic conductivity increased with temperature. In contrast to the observations of CTU, no significant influence of temperature on swelling pressure was identified for temperatures of up to 150°C by

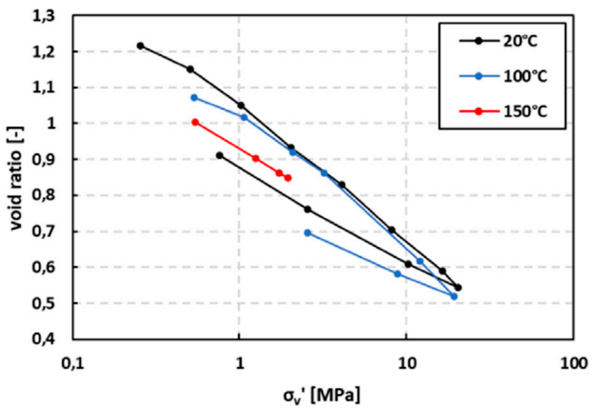

FIGURE 11
Results of oedometric tests performed with BCV bentonite compacted at dry density 1.6 g/cm^3 at different temperatures.

CU in samples that had been previously saturated at room temperature. Only small variations were measured, which could be attributed to the effects of the apparatus and rate of thermal loading. The reason for the qualitatively different results obtained by the two laboratories was the different evaluation methods used. The swelling pressure at the CU was determined shortly after heating started (on the order of hours), whereas at CTU, it was measured weeks after temperature was increased, and the measured value had contributions from the reduced swelling pressure caused by long-term high-temperature exposure. In both laboratories, the pressure responses after heating exhibited similar qualitative trends.

The effect of the cyclic temperature change was investigated on BCV samples compacted at 1.6 g/cm^3 saturated at 20°C, and later submitted to heating-cooling cycles up to 150°C by CU. The final swelling pressure measured in both the heating and cooling stages gradually decreased with increasing number of cycles (Figure 10). Furthermore, for temperatures higher than 50°C, a continuous decrease in swelling pressure during 30 days was observed for each temperature step. This decrease became more significant as the temperature was higher. Indeed, no cyclic temperature changes will occur in the repository, but these results show that under the conditions tested, the decrease in swelling pressure resulting from heating would be permanent, which was demonstrated by comparing the swelling pressure measured before and after the heating period (Najser and Mašín, 2024).

CU performed oedometric loading tests at 20, 100, and 150°C on BCV samples compacted to 1.6 g/cm^3. The samples were saturated with distilled water at room temperature, the backpressure was increased to 0.5 MPa, the temperature was set to the target value, and the samples were loaded incrementally. The slopes of the compression lines determined at different temperatures were similar. However, the positions of the compression lines shifted toward lower porosities with increasing temperature (Figure 11). Furthermore, the vertical strain of a sample saturated at room temperature under free swelling conditions was barely affected by the increase in temperature to 100°C.

A miniaturized oedometer set up (9 × 9 mm) made of PEEK was developed at the BRGM to perform tests at high temperatures using Kunipia smectite. The cells were equilibrated for 24 h at the selected temperature (inside an oven or by using a heating jacket) before hydration through the lower piston. For tests performed at 100°C, the solution was injected with a pressure of 1–500 kPa to avoid evaporation of the solution. The swelling tests were performed on Na$^+$ or Ca^{2+}-exchanged smectite at dry densities of 1.4, 1.5 and 1.8 g/cm^3 using solutions of NaCl or CaCl$_2$, respectively, of ionic strength 0.0001 and 0.1 M (Chaaya, 2023). The temperature accelerated the saturation process (Figure 12), likely because the hydraulic conductivity increased with temperature and decreased the final swelling pressure value of the Na-exchanged smectite more significantly as the density was higher. In contrast, the final swelling pressure of the Ca-exchanged samples was not affected by temperature (Supplementary Figure S3). Hence, it seems that under confined conditions, temperature only affected the crystalline swelling of sodic smectite, likely because of the different affinities to water of Na$^+$ and Ca^{2+}. The tests performed at 150°C showed a scarce influence of the sample density and very low swelling pressure values with respect to those measured at 100°C: 1.3–1.6 MPa for the Na-exchanged samples and 4.2–4.4 MPa for the Ca-exchanged ones.

5.2.2 Water retention curves: adsorption processes

The methodology most frequently used to determine water retention curves is to subject the samples to different relative humidities for sufficiently long periods of time for the material to reach equilibrium water content. This is known as the water transfer technique and is equivalent to the determination of vapor sorption isotherms. Using this technique, Sun et al. (2020) determined the water retention curves of the Czech bentonite B75 compacted at dry densities between 1.3 and 1.9 g/cm^3 at temperatures of up to 80°C. The water retention capacity decreases significantly at high temperatures, particularly at high RH (low suction). The authors predicted this dependence using a combination of the thermodynamic Clausius-Clapeyron equation and the Guggenheim–Anderson–de Boer model, which was also able to reproduce the independence of water retention from soil compaction (dry density) under high total suction. Ni et al. (2022) determined the water retention curves of the Chinese GMZ bentonite also using the vapor transfer technique for temperatures up to 80°C, and were able to fit the results obtained by considering a double-porosity structure in which the microstructural deformation depends lineally on temperature.

In the framework of HITEC the sorption isotherms of homogenised Kunipia smectite at free and constant volume conditions (dry density from 1.2 to 1.9 g/cm^3) were determined at BRGM in a temperature- and humidity-controlled chamber. Because of vapor condensation issues, the tests for RH higher than 70% could not be performed when the temperature was above 42°C. At higher temperatures, the limitations of this procedure become more significant. The results obtained under constant volume in samples compacted at dry densities ~1.5 g/cm^3 under 25°C and 42°C are shown in Figure 13. For the two materials, the water retention capacity decreased with temperature, but was much more significant in the case of Na-exchanged smectite.

Chaaya (2023) also monitored the apparent basal spacing of Na and Ca-Kunipia via XRD analyses of unconfined oriented powder slides at 25°C and 80°C, while increasing the relative humidity. Divalent smectites hydrate more rapidly than monovalent smectites

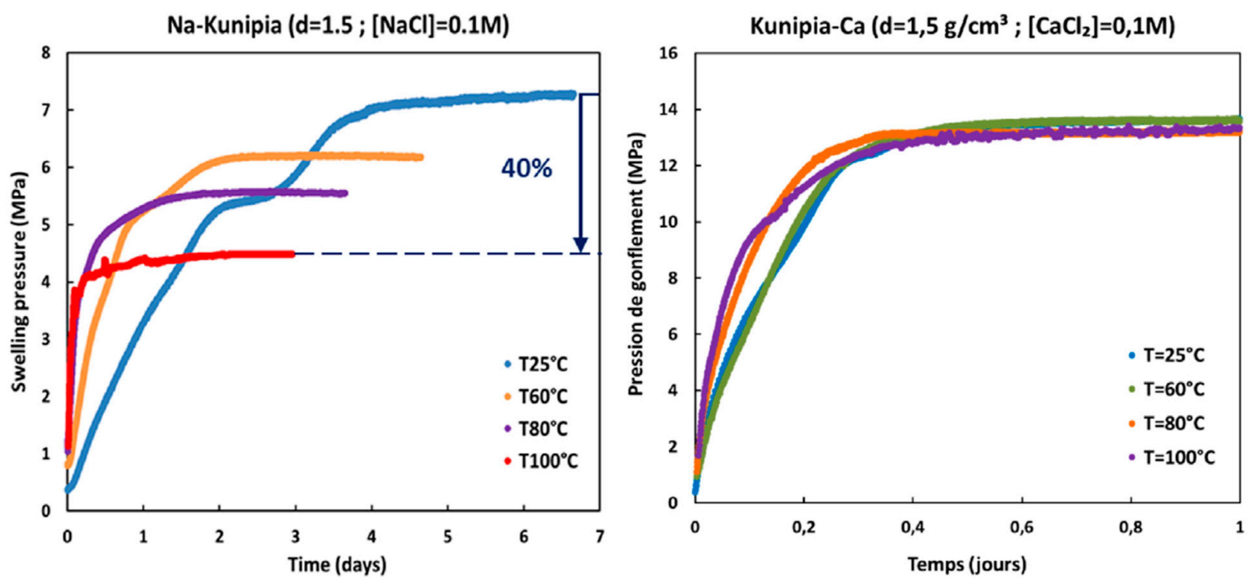

FIGURE 12
Swelling pressure evolution of Na and Ca-exchanged Kunipia smectite compacted at a dry density of 1.5 g/cm^3 and saturated with solutions 0.1 M (NaCl and $CaCl_2$, respectively) under different temperatures (Chaaya, 2023).

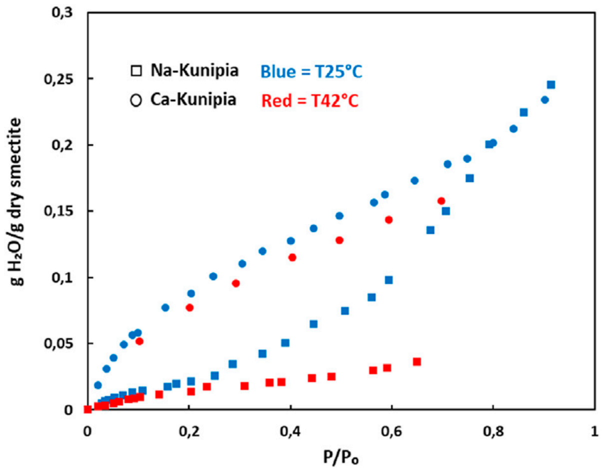

FIGURE 13
Water sorption isotherms of Na-exchanged and Ca-exchanged Kunipia smectite obtained under constant volume (dry density 1.5 g/cm^3) and two temperatures (Chaaya, 2023).

at both temperatures, given their higher ionic potential (Ferrage et al., 2005). The increase in temperature from 25°C to 80°C only negligibly affected the interlayer water retention capacity; thus, the basal spacing at 80°C was only slightly lower than that at 25°C (Supplementary Figure S4).

Another procedure for determining the water retention curve of compacted bentonite at high temperatures involves measuring the suction of samples with different water contents while maintaining the desired temperature. The compacted bentonite is maintained in a hermetic cell heated externally. The relative humidity and temperature inside the bentonite are measured (and converted into suction) using capacitive sensors (for the high-suction range) or psychrometers (for suction of <6 MPa). However, the determination of water retention curves in the low suction range at high temperature using thermocouple psychrometers presents the setback that they do not reach the equilibrium necessary for the measurement for temperatures higher than 60°C. This did not allow the curves to be defined within the low-suction range (Villar et al., 2020). The measurements showed the usual decrease in suction as the temperature increased, but the accuracy of the measurement worsened as the temperature increased because of the higher uncertainty in the relative humidity measurement for higher values. In addition, the suction measured using capacitive sensors was overestimated.

The water retention curves obtained at temperatures between 20°C and 100°C showed a sharper decrease in suction with increasing water content as the temperature was higher, particularly above 60°C (Figure 14). A comparison with results obtained in previous investigations for water content <20% confirmed the same trends. Suction reached null values at lower temperatures as the degree of saturation of the bentonite increased, because the increase in temperature triggers the transfer of water from the microstructure to the macrostructure, where it behaves as free water (Lloret and Villar, 2007). For temperatures above 40°C and in the range of water contents analysed, the water retention capacity of the FEBEX bentonite tended to be higher than that of MX-80, which is consistent with the results reported by Chaaya (2023) for Na- and Ca-exchanged smectite.

5.3 Small-scale simulation experiments

Laboratory tests in thermohydraulic (TH) cells that simulate the conditions of the buffer material in a radioactive waste repository are very useful for identifying and quantifying the processes occurring

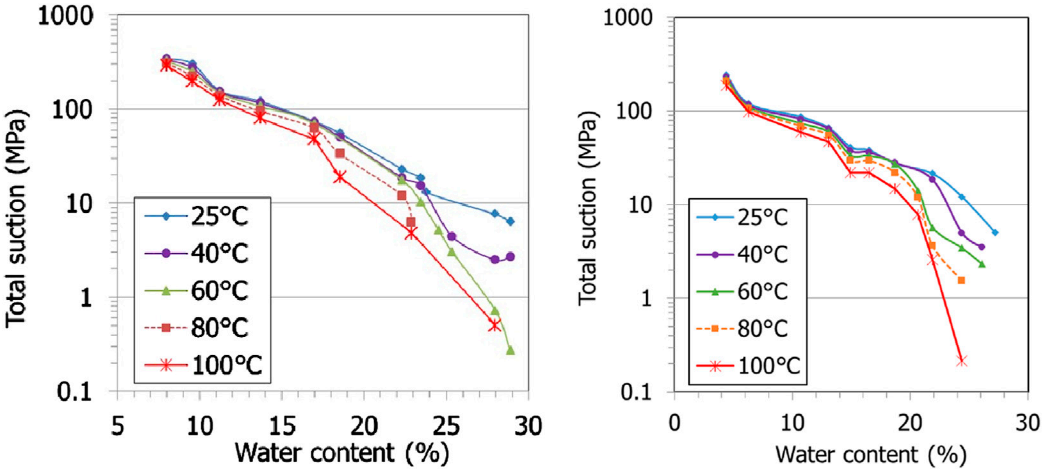

FIGURE 14
Water retention curves at different temperatures for FEBEX (left) and MX-80 (right) bentonites compacted at dry density 1.6 g/cm^3, obtained during HITEC and in previous projects (w < 20%).

in the engineered barrier. This type of test has evolved in the last decades from simple designs in which only temperatures inside the material were measured to current designs that involve the measurement of temperature and relative humidity, pressure, and water intake (Villar et al., 2012). These tests run for different periods (up to several years), and the analysis of the material upon dismantling includes mineralogical, geochemical, microstructural, hydromechanical, and chemical studies that provide insights into the time evolution of the properties of the barrier.

In the TH cells, the sealing material can be simultaneously subjected to heating and hydration (Supplementary Figure S5). They are cylindrical and designed to be hermetic and nondeformable. The heat generated by the radioactive decay of the waste in the containers is simulated using an electric heater, and the simulated groundwater is injected through a porous stone or stainless-steel sinter that ensures a uniform distribution of water over the sample surface.

5.3.1 Literature review

Two tests were performed to simulate the Äspö field test TBT (Åkesson et al., 2012), in which highly compacted MX-80 bentonite was used, and the heater temperature was set to 140°C. The laboratory cell tests lasted for 1.4 and 5 years, and the postmortem analyses focused on mineralogical, microstructural, and geochemical changes (Gómez-Espina and Villar, 2010; Gómez-Espina and Villar, 2015; ; Gómez-Espina and Villar, 2016). Problems in correctly measuring the water intake and vapor leakage through the sensor openings and the bottom of the cell were identified. These technological problems were solved, and in the framework of the PEBS project and to support the HE-E *in situ* test (Section 4.3), two tests in which the heater was set to 140°C were launched in 2012 (Supplementary Figure S4). Two sealing materials were tested: MX-80 bentonite pellets (Cell HEE-B) and a 65/35 sand/MX-80 bentonite mixture (Cell HEE-S/B). The two materials have very different gas and liquid permeabilities, as well as water retention capacities. This conditioned the water redistribution in the vapor phase triggered by heating as well as the liquid water intake, which were both much more restricted in the bentonite pellets. The HEE-S/B cell was dismantled after 2.8 years, and evidence of soluble species transport and precipitation close to the heater was found (Villar et al., 2016).

CEA performed two THM mockup tests in the laboratory on compacted MX-80 bentonite with two different initial water contents (Gatabin and Billaud, 2005). The samples had diameters and heights of 203 mm. Each test comprised two phases. In Phase 1 (lasting 4 months), heat was applied to one end of the column, while the temperature at the other end was maintained at 20°C; a maximum temperature of 150°C was applied. Phase 2 began after thermal equilibrium was achieved and involved gradual hydration of the sample for periods of time of 6 and 8 months. A constant water pressure was applied to the end opposite to that where the temperature variation was prescribed. Constant volume conditions were maintained during the two phases of the test. During the tests, the temperature, relative humidity, pore pressure, total axial stress, and total radial stress were measured. The tests provided interesting information regarding the temperature distribution, water movement, and pressure development, which were modelled as part of the SKB Task Force on Engineered Barrier Systems (Gens, 2019). The possible influences of the initial water content and sample length (scale effect) on the water transfer process were determined.

In the framework of the Safe Barriers project, experimental work by Cardiff University explored THM-C evolution under nonisothermal conditions (Tripathy et al., 2017). Column cells were configured using compacted bentonite cylinders with heater elements at one end to achieve temperatures of 85°C and 150°C, whereas the other end was maintained at 25°C. The relative humidity, temperature, and axial stress were monitored. Hydraulic tests were conducted in parallel using the same configuration in which water was injected into the cool end of the sample. The tests lasted for between 3 and 10 months. In the nonhydraulic tests, moisture redistribution occurred as a function of temperature, with greater relative humidity and water content measured further away from the heater. An increase in drying near the heater was also observed over time. However, difference in temperature between 85°C and 150°C and water injection pressures (5 and 600 kPa) appeared to have little impact upon axial stress over the experimental period.

More recently, a bench-scale (17 cm diameter, 46 cm length) experiment was conducted in an MX-80 bentonite compacted column (ρ_d = 1.2 g/cm^3) experiencing heating up to 200°C in the axis and hydration from a sand-clay boundary surrounding the column, where the temperature was 95°C (Chang et al., 2023). During the experiment, running for 1.5 years, frequent X-ray computed tomography (CT) scanning of bentonite provided insights into the spatiotemporal evolution of (1) hydration/dehydration, (2) clay swelling/shrinkage, and (3) displacement. Bentonite hydration was axisymmetrical despite the initial heterogeneity owing to packing, confirming the ability of bentonite to seal fast-flow/transport paths. Compared to the nonheated control experiment, the heated column showed greater CT density variations along the radial distance, indicating that the homogenisation of bentonite might be more difficult if a temperature gradient is maintained in the repository. The precipitation of an anhydrite layer occured at the inner hot zone.

5.3.2 Progress during HITEC

In the context of HITEC, the tests summarised in Supplementary Table S11 were performed or dismantled by CIEMAT and CTU. This lasted significantly longer than most of the tests reported in the previous section. Overall, the knowledge gained from these tests does not differ from that obtained in previous projects at lower temperatures.

Cell HEE-B, mentioned in Section 5.1.2, was dismantled during HITEC after 10 years of operation (Villar et al., 2022). The initial heating phase, lasting 7 months, showed that the thermal conductivity of the dry materials was low, which caused a large difference in temperature between the heater surface and the sensor located at 10 cm, generating a high thermal gradient near the heater and low temperatures in the rest of the column. The movement of water in the vapor phase as a result of the thermal gradient was evinced by the increase in the relative humidity recorded by the sensor closest to the heater, followed by a continuous decrease and a slower increase recorded by the two other sensors (see Supplementary Figure S4 for the exact location of the sensors). The temperature was not affected by hydration and remained approximately constant until the end of the test. The evolution of relative humidity inside the bentonite highlighted the low permeability of the pellets. The water content and dry density distribution measured after dismantling of the column showed uniform degrees of saturation between 92% and 99% in the upper half of the column and a sharp decrease toward the heater in the bottom half of the column, with values close to 0% in the 5 cm closest to the heater. Vapor leakage through the sensor located 10 cm from the heater may have contributed to the extremely low water content measured in this area.

Two other tests were carried out in cylindrical stainless steel cells of dimensions 10 cm × 10 cm with Bara-Kade bentonite compacted at dry density 1.55 g/cm^3 with water content 17%. The heating phase was similar in the two tests: the heater temperature was increased from room to 150°C in 9 days, and it remained in this value for 3 months. Thermal equilibrium was quickly reached. The water vapor moved away from the heater toward the upper part of the cell, and the bottom of the samples started to dry out. After quasi-steady hydraulic conditions were reached, hydration was started with glacial water for cell HT1 and saline water for cell HT2. The radial pressure sensors in the upper and middle parts of the cells exhibited increasing trends from the beginning of the hydration (faster in the upper sensors). The pressure values at the same location were higher for cell HT1 hydrated with glacial water, which was consistent with the salinity-induced reduction in swelling capacity. Radial pressures between 5 and 7 MPa were measured in cell HT1 after 2.5 years of hydration, attesting to the bentonite swelling capacity even at high temperatures.

In the two tests performed with BCV bentonite and a heater set at 150°C, the same vapor migration patterns, water content, and dry density changes were observed, both in the tests performed using powder or pellets.

6 Thermo-hydro-mechanical models

One of the objectives of HITEC was the development and validation of suitable THM models for clay buffer at temperatures higher than 100°C and the incorporation in them, if necessary, of the processes identified during the WP activities, mostly those in subtask 3.3 “Small-scale experiments.” A conclusion from the PEBS project (Johnson et al., 2014) was that the THM formulations developed and validated for temperatures below 100°C can be extended without modifications to temperatures above that value. The variations in the retention curve, water permeability, and surface tension with temperature can be considered without modifying the basic formulation.

In the SKB Task Force of Engineered Barrier Systems reported by Gens (2019), several teams modelled small-scale laboratory THM tests (some of which are introduced in Section 5.2). The conclusions related to the thermal problem are as follows:

- The main difference between the general formulations is their capacity to incorporate the air-balance equation. Without a gas equation, the usual assumption is that the gas pressure is constant. This may lead to nonphysical situations if the vapor pressure exceeds 1 atm when temperatures rise above 100°C.
- The only thermal effect considered on the mechanical behaviour was thermal expansion derived from a constant coefficient of thermal dilation.
- The dominant heat transfer mechanism identified was conduction. Hence, a good model of the thermal problem requires consideration of the changes in thermal conductivity with the degree of saturation.
- The differences between the computed and observed hydraulic results were more apparent in the nonisothermal tests. The reason for this may be the existence of unnoticed vapor leaks from the testing cells that were not accounted for in the models.

Prior to HITEC, Thatcher (2017) performed coupled THM modelling for the FEBEX *in situ* experiment carried out at the Grimsel URL (ENRESA, 2006) that was built upon an HM model, called the Internal Limit Model (ILM), which had been used for an MX-80 bentonite and sand mixture. Thatcher et al. (2017)

documented the first application of this model to an alternative bentonite with the addition of thermal coupling and tested its transferability in a large-scale experiment. The model was effective in describing the THM evolution of the experiment after minimal calibration, despite the addition of new thermal parameters and the different materials modelled. In addition, at high strains, the behaviour of bentonite was more similar to that of a fluid than that of an elastic material.

Ghiadistri (2019) developed a new constitutive framework for compacted, highly expansive clays that was implemented in the Imperial College Finite Element Program (ICFEP, Potts and Zdravkovic, 1999). The model was built on a single-structure model previously developed at the Imperial College (Georgiadis et al., 2005; Tsiampousi et al., 2013) by extending the formulation to capture the double-porosity structural characteristics of highly expansive clays. The model was validated using available experimental data, demonstrating substantial improvements in the numerical predictions of hydromechanically coupled processes in compacted bentonite compared with those of the single-structure model. In conjunction with the THM formulation of the governing finite element equations in ICFEP, the model successfully reproduced the behaviour of compacted bentonite clays both at the laboratory scale (in the simulations of a series of swelling pressure tests) and at the field scale (in the simulation of the FEBEX large-scale experiment). A key knowledge gap identified in this research was the lack of experimental data for quantifying the microstructure of compacted bentonites; the unavailability of such data influenced the calibration of the microstructural parameters of the new constitutive model.

During the HITEC project, the UPC developed and verified a double-structure constitutive model (BExM-T) that incorporates the effects of temperature, considers the hydromechanical coupling between the micro- and macropore levels, and defines the retention curves for each structural domain (Vasconcelos, 2021). In addition, the following features were formulated and verified: 1) the possibility of decoupling the elastic response at the micro and macro levels, 2) the dependence of the Bishop parameter (mechanical behaviour of the microstructure) on the effective saturation of the micropores, and 3) the evolution of water retention curves with structural changes at the micro and macro levels. This model was applied to the column test described in Section 5.3 with MX-80 pellets (HEE-B cell). The simulations incorporated a number of relevant processes observed in the laboratory tests, such as heat conduction, water phase changes, vapor diffusion, differential thermal expansion of the solid and liquid phases, and swelling behaviour induced by thermohydraulic actions. The model satisfactorily predicted the evolution of the temperature, relative humidity, water intake, and axial stress over the entire test duration. However, larger discrepancies were observed in the final states (dry density and degree of saturation) of the bentonite columns. Improvement of the constitutive model to include features, such as the dependence of the water retention curves on the evolution of the pore volume fractions in micro- and macromedia, may be necessary (Vasconcelos et al., 2023).

Mašín (2017) developed a coupled THM model based on hypoplasticity principles combined with the concept of a double structure by hierarchical enhancement of an earlier model by Mašín (2013), which did not consider the effects of temperature. To include the thermal component, an additional thermal dependency was introduced for the water retention curves, volumetric behaviour of the microstructure, and normal compression behaviour of the macrostructure. The updated model could predict the dependency of water content on suction at temperatures up to 80°C (Mašín, 2017). The complex behaviour of the volume strains resulting from the heating of MX-80 bentonite up to 80°C at various values of suction and mean total stress (described by Tang et al., 2008b) was predicted as follows:

- At high suctions (110 MPa), the model predicts swelling, whose magnitude is controlled by α_s (the dependency of microstructural volume strains on temperature). The heat-induced expansion at high suction is primarily reversible.
- At lower suction (9 MPa for a total stress of 0.1 and 39 MPa for a total stress of 5 MPa), the model predicts heating-induced contraction, which is irreversible and controlled by the offset of normal compression lines at different temperatures. However, for a stress of 0.1 MPa at a suction of 39 MPa, the state is well within the state boundary surface and heating-induced expansion is predicted, in agreement with experimental data. In principle, the model could also be calibrated using negative α_s to predict the heating-induced contraction observed by some authors.
- The model predicts the cooling-induced contraction, which depends on suction, such that it is the most pronounced at 110 MPa and the least significant at 39 and 9 MPa. These predictions are governed by the dependence of the double-structure coupling factor f_m on the macrostructural degree of saturation.

In the HITEC framework, this model was used to simulate two small-scale TH tests, in which the BCV bentonite was saturated under the temperature gradient imposed by a heater set at 150°C on one side of the columns (Villar et al., 2023a). Although the dry densities of the bentonite in the two tests were 0.9 and 1.4 g/cm^3, the initial material parameters were set for a dry density of 1.5 g/cm^3, because of convergence problems of the hypoplastic bentonite model for low dry densities. Good agreement between the calculated and measured temperatures was observed, and the evolution of the vertical pressure (corresponding to the swelling pressure) followed the gradual saturation of the bentonite and subsequent increase in water pressure. The model exhibits convergence issues during the increasing water pressure phase.

At the VTT, the varied multiplicative process (VMP) model was upgraded to include thermal phenomena and dependencies. The VMP is a large deformation model with terminology and concepts that deviate from those used in standard linear deformation theories (Pulkkanen, 2019). Bentonite is divided into a skeleton, which contains solid material and adsorbed water, and a free porosity, which is filled with free water or gas. Thermal phenomena were consistently added thermodynamically to the VMP model. The upgraded model consists of 1) the mass balance equation for adsorbed water transport, 2) the force balance equation for mechanical behaviour, and 3) the thermal balance equation, including thermal terms (Vasconcelos et al., 2023).

7 Summary and conclusions

The primary aim of the HITEC work package (WP) of the EURAD project was to ascertain whether temperatures higher than the limits currently established in most repository concepts (a maximum of 100°C for the buffer) would compromise the safety functions of engineered and geological barriers. The effect of temperature on bentonite properties was studied in HITEC by analysing the change in the properties of the preheated material and by determining the properties of bentonite at high temperatures. The bentonites under assessment included Wyoming-type (MX-80, Bara-Kade), BCV (Czech Republic), and FEBEX (Spain).

Although the impact of preheating on the physicochemical and hydromechanical properties of bentonite had been previously analysed, during HITEC the differences between heating the material under dry and wet conditions have been stressed. Different research groups determined the properties and characteristics of bentonite heated at 150°C in dry and wet conditions after different periods of time up to 2 years. While no drastic changes were observed in any case and for any bentonite sample, with remarkable preservation of the clay mineralogy, slight changes depending on the heating conditions were detected. If evaporation was allowed (dry conditions), then the changes were more important, generally involving reduced CEC, specific surface area, and sorption coefficients. However, heating under wet conditions had the opposite effect on these properties. Changes in these parameters could affect the performance of the buffer, particularly its radionuclide sorption ability, although experimental evidence indicated no detrimental effects. Some evidence was found for changes in the cation exchange complex resulting from heating under wet conditions, with increases in the amount of exchangeable calcium observed in both the Bara-Kade and BCV bentonites. In any case, given that the deterioration of properties was only noticeable in the samples treated under dry conditions, it can be presumed that this deterioration was caused not by the temperature itself, but by the loss of water induced by the elevated temperature. This is consistent with the changes observed in the physicochemistry, geochemistry, and mineralogy of bentonite heated under a thermal gradient for 10 years in areas where continuous evaporation was allowed (test HEE-B). The analysis of reversibility was not included in the test programs, but some observations suggest that the changes in physicochemical properties that occurred owing to strong drying, such as resistance to rehydration, were not definite and would simply require longer recovery times.

As for the hydro-mechanical properties of preheated material, the volumetric compression properties of Wyoming-type bentonite heated under dry conditions were not modified, and the swelling pressure and hydraulic conductivity deteriorated only in samples heated under dry conditions and compacted at high dry density. In addition, the water retention capacity was lower in samples that had been previously dry-heated than wet-heated, but this was expected given the hysteretic character of water retention.

For bentonite subjected to hydration under a thermal gradient from TH tests in cells in which the heater simulating the canister was set at temperatures higher than 100°C, no postmortem structural modification of the smectite was observed; however, dissolution and precipitation of species occurred and were conditioned by the type of bentonite and hydration water. These processes were accompanied by the modification of the exchangeable cation complex and were in line with those observed in similar tests performed at lower maximum temperatures. The hydromechanical properties were not negatively affected if the bentonite reached a high degree of saturation during the treatment, but the hydraulic conductivity slightly increased if the bentonite was heated allowing evaporation. The use of pellets instead of compacted blocks does not appear to be an additional source of uncertainty.

The material obtained from the field test ABM5 which was also hydrated under a high thermal gradient for ~5 years, did not present any noticeable high-temperature reaction, even though extremely high temperatures (such as 250°C, which were never reached before in a field experiment) were measured at the heater contact. Moreover, the modifications observed in the materials, such as magnesium enrichment, corrosion, and an increase in the trioctahedral character of smectite, were less pronounced compared to other tests conducted at lower temperatures but for longer times with higher water contents. This is consistent with previous laboratory test findings, indicating minor mineralogical transformations under unsaturated conditions.

The determination of properties of expansive clay at elevated temperatures, though started decades ago, still requires the development and improvement of testing equipment and methodologies, particularly for the application of experimental methods used for temperatures lower than 100°C to higher temperatures, which has been one of the novel contributions of the WP. Special materials such as invar have been used in the developments made during HITEC. Vapor leakage through various rig elements (e.g., sensor inlets) and gas tightness are among the most common issues. Calibration of the apparatus deformation is essential in hydromechanical tests, and failure to apply the correct correction of deformation and pressure may lead to contradictory conclusions regarding the effect of temperature on the behaviour of clay. Other aspects, such as the accurate consideration of the dry density of the samples during testing (instead of after testing), can also significantly affect the conclusions. Venting the testing cells to assist hydration may trigger sample drying at high temperatures. Precise measurement of water inflow/outflow, which is essential for correctly computing permeability, is also difficult, as is the selection of the right water pressures.

In addition, the procedures used to determine the properties at high temperatures may have significantly impacted the results. For example, heating under isochoric conditions a material that had been previously saturated at room temperature, entailed a progressive and permanent, not recovered after cooling, decrease in the swelling pressure with respect to that measured at the end of saturation. This phenomenon, which has been observed for sodic and divalent bentonites, did not affect hydraulic conductivity. However, saturating the material under high temperatures resulted in lower equilibrium but stable swelling pressure, which is frequently reported in the literature. By contrast, tests performed in purified homoionised smectite (using as saturating fluid solutions containing the same cation as the smectite interlayer) showed a decrease in swelling pressure with temperature for the sodic-exchanged material and no effect of temperature on the calcium-exchanged material. This observation points to a possible influence of cation exchange processes on the changes in swelling pressure with temperature measured in raw bentonite: the dissolution of accessory minerals at high temperatures could trigger changes in the cation exchange complex (or other chemical reactions), and consequently, swelling. However, the results consistently indicate that the effect of temperature is more significant at higher dry densities and temperatures. Even at the highest temperatures, bentonite has the ability to fill voids and develop large swelling pressures at high densities. The hydration and swelling pressure development were quicker as the temperature was higher, in accordance with the well-known increase in hydraulic conductivity with temperature.

Among the cutting-edge developments in HITEC, X-ray tomography was used to monitor the wetting and swelling of bentonite during temperature-controlled hydration, which allows subsequent analysis of bentonite deformation and spatial distribution of density and water content. The temporal evolution of the degree of saturation profiles suggests that diffusion may play a role in the transport process, in addition to advection.

The determination of water retention curves in the low-suction range at high temperatures has not been achieved, neither by using the vapor transfer technique (because of condensation issues) nor by measuring suction with thermocouple psychrometers. Nevertheless, as expected, the water retention capacity decreased with temperature in tests performed using different methodologies and materials, and this decrease was more significant for predominantly sodic materials. The increase in temperature from 25°C to 80°C had a negligible effect on the interlayer water retention capacity, and hence the basal spacing at 80°C was only slightly lower than that at 25°C.

Modeling activities focused on the development and upgrading of thermo-hydro-mechanical models to include thermal phenomena (heat conduction, water phase changes, vapor diffusion, differential thermal expansion of the solid and liquid phases, swelling behaviour induced by TH actions) and dependencies (water retention curve, volumetric behaviour of microstructure, normal compression behaviour of macrostructure). These models were applied to simulate laboratory thermohydraulic tests in cells performed during the project. The interaction between the micro- and macrostructures is essential for properly reproducing the behaviour of expansive materials at high temperatures.

The observations summarised above highlight open issues and aspects that require additional work:

- The effect of temperatures higher than 100°C has been considerably studied concerning mineralogical transformations, albeit not always in clearly representative conditions, namely, solid:liquid ratios, vapor pressure conditions, chemical composition of the saturating water, and the presence of accessory minerals. The effects of these factors on the observed processes should be assessed.
- Information on the hydromechanical properties in this temperature range is inadequate, partly because testing at high temperatures imposes additional challenges: sensor suitability, vapor leaks, and equipment calibration. Therefore, these technological issues must be addressed.
- Most studies concerning the impact of temperature on hydromechanical properties refer to saturated materials, and studies on unsaturated materials are scarce. In particular, the determination of the water retention curve at high temperatures and low suction presents experimental challenges that have not been solved.
- The impact of certain boundary conditions on the saturation behaviour of the barrier is still unclear (also at room temperature), as is the role played by advection and diffusion.
- Robust assessment of bentonite swelling pressure as a function of temperature is necessary to design barrier materials for repositories intended to operate at elevated temperatures (>100°C).
- Although most evidence shows a decrease in bentonite swelling pressure at high temperature, the measurements performed using purified smectite and avoiding cation exchange during saturation did not show any changes in swelling pressure for Ca-exchanged smectite at least until 100°C. The roles of swelling pressure development of the initial exchangeable cations and exchange processes (or other temperature-induced chemical processes) during bentonite saturation are not completely understood.
- In most cases, the increase in hydraulic conductivity with temperature cannot be explained solely by the increase in water kinematic viscosity. Other factors may play a role (microstructure, mineralogy, or pore fluid chemistry changes), which should be understood.
- Additional assessment of the long-term swelling behaviour of bentonite is necessary to inform numerical simulations, which may require modifications to allow for time-dependent swelling at elevated temperatures. The use of blind prediction modelling trials may be particularly beneficial for validating understanding over long timescales.

Author contributions

MV: Conceptualization, Data curation, Formal Analysis, Investigation, Visualization, Writing – original draft, Writing – review and editing. KC: Data curation, Investigation, Writing – original draft. JC: Data curation, Investigation, Writing – original draft. AG: Formal Analysis, Investigation, Software, Writing – original draft. NG: Visualization, Writing – review and editing. CG: Data curation, Investigation, Writing – review and editing, Writing – original draft. JH: Data curation, Investigation, Writing – original draft. VK: Data curation, Investigation, Writing – review and editing, Writing – original draft. SK: Writing – original draft, Data curation, Investigation. OL: Conceptualization, Investigation, Writing – original draft. DM: Investigation, Software, Writing – original draft. JN: Data curation, Investigation, Writing – original draft. MO: Project administration, Writing – original draft. HR: Data curation, Investigation, Writing – original draft. ŠŠ: Data curation, Investigation, Writing – original draft. SS: Data curation, Investigation, Writing – original draft. DS: Data curation, Investigation, Writing – original draft. JS: Data curation, Investigation, Writing – original draft. GV: Data curation, Investigation, Writing – original draft, Writing – review and editing. JY: Data curation, Investigation, Writing – original draft, Writing – review and editing. BZ: Data curation, Investigation, Writing – original draft.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Publisher's note

Supplementary material

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fnuen.2025.1436087/full#supplementary-material

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