



# **Deliverable 10.1: Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation**

Work Package 10

DOI: 10.5281/zenodo.18173404



Co-funded by the European Union under Grant Agreement n°101166718

**Document information**

Project Acronym	<b>EURAD-2</b>
Project Title	<b>European Partnership on Radioactive Waste Management-2</b>
EC grant agreement No.	<b>101166718</b>
Work Package Title	<b>ANCHORS - hydrAulic mechaNical CHemical evolution of bentOnite for barrieRs optimiSation</b>
Deliverable No.	<b>D10.1</b>
Deliverable Title	<b>Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation</b>
Lead Beneficiary	<b>SKB</b>
Contractual Delivery Date	<b>March 2025</b>
Actual Delivery Date	<b>March 2025</b>
Dissemination level	<b>PU</b>
Authors	Patrik Sellin (SKB), Xavier Pintado (Mitta), Jiri Svoboda (CTU), Arttu Miettinen (JYU), Eric Simo (BGE), Sebastia Olivella (UPC), Jean Talandier (ANDRA), Olivier Cuisinier (LEMTA), Ann Dueck (Clay Technology), Margit Fabian (HUN-REN EK), Stepane Gaboreau (BRGM), Matthew Kirby (NWS), Doncho Karastanov (GI-BAS), Olivier Leupin (Nagra), Zhanrong Liu (CNRS-Navier), Juan Carlos Mayor (Enresa), Miroslava Mecová (SÚRAO), Mathilde Métral (EPFL), Marvin Middelhoff (GRS), Nadia Mokni (ASNR), Jan Najser (CUNI), Thomas Nagel (TUBAF), Mika Niskanen (Posiva), Matteo Pedrotti (U. Strathclyde), Veli-Matti Pulkkanen (VTT), Nina Stoppe-Struck (BGR), Petr Vecerník (UJV), María Victoria Villar (Ciemat).

**To be cited as:**

Sellin P., Pintado X., Svoboda J., Miettinen A., Simo E., Olivella S., Talandier J., Cuisinier O., Dueck A., Fabian M., Gaboreau S., Kirby M., Karastanov D., Leupin O., Liu Z., Mayor J C., Mecová M., Métral M., Middelhoff M., Mokni M., Najser J., Nagel T., Niskanen M., Pedrotti M., Pulkkanen V-M., Stoppe-Struck N., Vecerník P., Villar M V., (2025): Title. Final version as of 31.03.2025 of deliverable D10.1 of the European Partnership EURAD-2. EC Grant agreement n°:101166718.

**Disclaimer**

All information in this document is provided "as is" and no guarantee or warranty is given that the information is fit for any particular purpose. The user, therefore, uses the information at its sole risk and liability. Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or European Atomic Energy Community. Neither the European Union nor the granting authority or the individual Colleges of EURAD-2 can be held responsible for them.

**Acknowledgement**

This document is a deliverable of the European Partnership on Radioactive Waste Management 2 (EURAD-2). EURAD-2 is co-funded by the European Union under Grant Agreement N° 101166718.

<b>Status of deliverable</b>		
	<b>By</b>	<b>Date</b>
Delivered (Lead Beneficiary)	Patrik Sellin (SKB)	21/02/2025
Verified (WP Leader)	Nadia Mokni (ASNR)	21/02/2025
Reviewed (Reviewers)	Nadia Mokni (ASNR) Vaidas Matuzas (JRC)	4/03/2025 10/03/2025
Verified (WP Leader)	Nadia Mokni (ASNR)	27/03/2025
Approved (PMO)	Vaidas Matuzas (JRC)	31/03/2025
Submitted to EC (Coordinator)	Andra	01/04/2025

## Executive Summary

The objective of the ANCHORS Work Package (WP) is to enhance the optimisation potential of bentonite barrier systems—including buffer, backfill, and seals—while strengthening the resilience of the Safety Case. This is achieved through:

1. Qualifying the Hydro Mechanical (HM) behaviour of various kind of bentonite types and mixtures through laboratory experimental programme focused on heterogeneity, chemical effects and friction at different scales and
2. Improving the numerical tools that are necessary to carry out performance assessment of bentonite barriers in a Thermo Hydro Mechanical Chemical (Gas) (THMC(G)) repository environment.

The aim of the current report is to describe the knowledge base on bentonite barrier systems in relation to the activities that will be performed in ANCHORS. The basis for this is:

- Issues regarding bentonite barriers identified in the EURAD strategic research agenda
- A description of how bentonite is used in national programs for radioactive waste management
- A description of a number of selected earlier projects/work packages that has involved bentonite studies
- The objectives, background and plans for the experimental work in ANCHORS
- A summary of the model development and testing that will be done in ANCHORS

A more detailed summary is given in the last chapter in this report.

## Keywords

Bentonite. Hydraulic Mechanical Chemical (HMC) Evolution, Engineered Barrier System (EBS), Deep Geological Repository (DGR), Swelling Pressure, Radioactive Waste Management, Performance Assessment, THMC(G) Modelling, Buffer and Backfill, Sealing and Plugs.

## Table of content

Executive Summary.....	4
Keywords .....	4
Table of content.....	5
List of figures .....	8
List of Tables .....	9
Glossary.....	10
1. Introduction .....	13
2. Strategic research agenda .....	14
2.1 Overview .....	14
2.2 Buffers.....	15
2.3 Backfills.....	15
2.4 Plugs and seals .....	15
2.5 Relation between the SRA and ANCHORS .....	16
3. Role of Bentonite in the Repository Concepts .....	16
3.1 Czech Republic.....	17
3.1.1 Role of bentonite.....	17
3.1.2 Current research needs .....	18
3.2 Finland .....	19
3.2.1 Use of bentonite.....	19
3.2.2 Current research needs .....	19
3.3 France.....	19
3.3.1 Use of bentonite.....	19
3.3.2 Current research needs .....	21
3.4 Germany .....	21
3.4.1 Use of bentonite.....	22
3.4.2 Current research needs .....	26
3.5 Spain.....	26
3.5.1 Use of bentonite.....	26
3.5.2 Current research needs .....	28
3.6 Sweden .....	28
3.6.1 Use of bentonite.....	28
3.6.2 Current research needs .....	29
3.7 Switzerland .....	30
3.7.1 Use of bentonite.....	31
3.7.2 Current research needs .....	31

3.8 United Kingdom .....	31
3.8.1 Use of bentonite.....	31
3.8.2 Current research needs.....	32
4. Bentonite materials.....	33
5. Examples of earlier projects related to bentonite .....	34
5.1 Febex .....	34
5.2 Prototype Repository .....	35
5.3 PEBS .....	37
5.4 BEACON.....	37
5.5 GAS .....	41
5.6 HITEC .....	41
5.7 SANDWICH .....	42
5.8 HotBENT.....	42
6. Laboratory testing.....	42
6.1 Phenomena investigated .....	43
6.1.1 Heterogenous materials .....	43
6.1.2 Effect of Salinity & Alkalinity (pH) .....	44
6.1.3 Vapour vs liquid saturation and hydration processes.....	45
6.1.4 Friction effect .....	45
6.2 Micro scale testing .....	46
6.2.1 BGR .....	47
6.2.2 BRGM .....	48
6.2.3 Ciemat .....	49
6.2.4 GI-BAS.....	49
6.2.5 GRS .....	50
6.2.6 HUN-REN EK .....	51
6.2.7 ASNR.....	52
6.2.8 Jyväskylä university .....	53
6.2.9 University of Lorraine (LEMTA) .....	53
6.2.10 University of Strathclyde .....	54
6.3 THMC(G) Laboratory testing .....	55
6.3.1 Ciemat .....	56
6.3.2 Clay Technology .....	56
6.3.3 CNRS-Navier .....	58
6.3.4 CUNI .....	58
6.3.5 CTU .....	59

6.3.6 EPFL.....	60
6.3.7 GI-BAS.....	61
6.3.8 GRS .....	61
6.3.9 ASNR.....	62
6.3.10 Mitta .....	63
6.3.11 TUBAF .....	64
6.3.12 UJV .....	65
6.3.13 University of Lorraine (LEMTA) .....	65
6.3.14 University of Strathclyde .....	67
6.3.15 VTT .....	67
6.4 Assessment of measures for better quality control and testing of bentonite .....	68
7. Bentonite Barrier modelling and Performance assessment .....	70
7.1 Enhancement of existing constitutive models and numerical tools .....	70
7.2 Application to assessment cases .....	71
8. Summary .....	72
References .....	74

## List of figures

Figure 1 Illustration of the disposal of SNF in the DGR

Figure 2 Diagram of the vertical disposal system

Figure 3 A canister in a deposition hole

Figure 4 Seal designs– (a) horizontal tunnels core is confined between two concrete abutment (b) horizontal tunnels core is in contact with backfill, (c) in a ramp, (d) in a shaft

Figure 5 Generic concept of a repository in claystone formations in Germany with illustration of the bentonite pedestal in the disposal gallery (Lohser, 2024)

Figure 6 Illustration of a gallery seal in a repository for radioactive waste in claystone formations in Germany (Lohser, 2024)

Figure 7 Schematic representation of a gallery seal in a low permeability area of the crystalline rock formation (Jobmann & Burlaka, 2021)

Figure 8 Schematic diagram of the geological repository in clay rock

Figure 9 Upper: Clay barriers and closure of the Spent Fuel Repository: buffer, backfill, closure and borehole seal. Lower, left: silo clay barrier and closure in SFR. Lower, right: clay barrier in the waste vault for legacy waste (BHA) in the Final Repository for Long-lived Waste (SFL)

Figure 10 An illustration of what a UK GDF could look like in an in-shore region

Figure 11 Schematic representation of the test cell and images of the block (upper right) and pellets (lower right). In the predictive test the block was placed in the bottom of the cell (Villar et al 2021)

Figure 12 Dry densities at saturation for the experiment used for predictive modelling

## **List of Tables**

Table 1 Mechanical constitutive models used in the benchmark

## Glossary

ANCHORS - hydrAulic mechaNical CHemical evolution of bentOnite for barrieRs optimiSation

ANDRA - National Agency for Radioactive Waste Management (France)

ARSN - National authority of radiation protection, safety, and nuclear safety (France)

BBM - Barcelona Basic Model

BACCHUS - BACKfilling Control Experiment for High level wastes in Underground Storage

BEACON - Bentonite Mechanical Evolution Project

BELBAR - Bentonite Erosion: effects on the Long term performance of the engineered Barrier and Radionuclide Transport

BExM - Barcelona Expansive Model,

BGE - Federal Company for Radioactive Waste Disposal (Germany)

BGR - Bundesanstalt für Geowissenschaften und Rohstoffe (Germany)

BRGM - French Geological Survey

CEC - cation exchange capacity

CIEMAT - Centre for Energy, Environmental and Technological Research (Spain)

Cigéo - Industrial Centre for Geological Disposal

CNRS - Centre national de la recherche scientifique (France)

COx - Callovo-Oxfordian Claystone

CTU - Czech technical university in Prague

CUNI - Charles University in Prague

DGR - Deep Geological Repository

DSM - Double Structure Model

EBS - Engineered Barrier System

EC - European Commision

ECOCLAY - Effects of Cement on CLAY barrier performance

EPFL - École polytechnique fédérale de Lausanne

EURAD - European Joint Programme on Radioactive Waste Management

EURAD-2 - European partnership on Radioactive Waste Management

FEBEX - Full-scale Engineered Barrier Experiment

FORGE - Fate of Repository Gases Project

GAS - Mechanistic understanding of transport processes in clay materials

GDF - Geological Disposal Facility

GI-BAS - Geological Institute at the Bulgarian Academy of Sciences

GRS - Gesellschaft für Anlagen- und Reaktorsicherheit (Germany)

HITEC - High-Temperature Effects on Clay Barriers

HM - Hydro-Mechanical

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

HC - Hydro-Chemical

HMC - Hydro-Mechanical-Chemical

HotBENT - High Temperature Effects on Bentonite Buffers

HUN-REN EK - Hungarian Center for Energy Research

ICFEP - Imperial College Finite Element Programme

KERAMOST - a Czech bentonite supplier

KBS-3V - Swedish/Finnish Nuclear Waste Disposal Concept

MIP - Mercury Intrusion Porosimetry

MX80 - A Type of Bentonite

NAGRA - National Cooperative for the Disposal of Radioactive Waste (Switzerland)

NF-PRO Near field Processes

NWS - Nuclear Waste Services (UK)

OPA - Opalinus Clay

PEBS - Project on the Effects of Post-Closure Evolution on the Performance of Engineered Barrier Systems

PM - Purified Montmorillonite

Posiva - Finnish waste management company

RESEAL - REpository SEALing

RD&D - Research, development and demonstration

SANDWICH - Vertical hydraulic sealing system based on the sandwich principle

SEM-EDX - Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy

SKB - Swedish Nuclear Fuel and Waste Management Company

SRA - Strategic Research Agenda

SNF - Spent Nuclear fuel

TBM - Tunnel Boring Machine

THM - Thermo-Hydro-Mechanical

THMCB - Thermo-Hydro-Mechanical-Chemical-Biological

THMC - Thermo-Hydro-Mechanical-Chemical

THMC(G) - Thermo-Hydro-Mechanical-Chemical (Gas)

TUBAF - Freiberg University of Mining and Technology

UJV - Nuclear Research Institute (Czech Republic)

VMP - Varied Multiplicative Processes Model

VTT Technical Research Centre of Finland

WAXS - Wide-Angle X-ray Scattering

WDP - Waste Disposal Packages

WMO - Waste Management Organisation

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

WP - Work package

XRD - X-ray Diffraction

$\mu$ CT - Micro-Computed Tomography

## 1. Introduction

The objective of the ANCHORS Work Package (WP) is to enhance the optimisation potential of bentonite barrier systems—including buffer, backfill, and seals—while strengthening the resilience of the Safety Case. This is achieved through:

1. **Qualifying the Hydro-Mechanical (HM) behaviour** of various bentonite types and mixtures via laboratory experiments, with a focus on heterogeneity, chemical effects, and friction across different scales.
2. **Improving numerical tools** required for performance assessment of bentonite barriers within a Thermo-Hydro-Mechanical-Chemical (Gas) (THMC(G)) repository environment.

Bentonite serves as the primary sealing material in most Deep Geological Repository (DGR) concepts. Therefore, a comprehensive understanding of its long-term THMC(G) behaviour is critical for improving safety, refining Engineered Barrier System (EBS) designs, and optimising repository performance. The specific objectives of this WP include:

- Investigating the chemical effects (e.g., alkaline and saline conditions) on the HM behaviour of bentonite and bentonite-based mixtures, particularly regarding sealing capacity under lower swelling pressures and water chemistry's influence on fluid permeability.
- Examining scale effects in laboratory tests (small vs. mock-up scales) and how laboratory results translate into full-scale repository modelling.
- Analysing the role of friction in laboratory tests and its implications for density distribution in EBS components constructed with bentonite and bentonite mixtures.
- Assessing the impact of initial heterogeneities in bentonite barriers and how hydration-induced changes influence long-term sealing performance under repository conditions.
- Studying the effects of different hydration conditions, including sodium vs. bivalent bentonites and hydration velocity, on density distribution and chemical interactions.
- Enhancing THMC(G) models by integrating chemical effects into HM behaviour, with improvements validated through experimental and numerical benchmarking.
- Conducting performance assessment test cases that account for thermal and chemical influences.

Additionally, this WP includes the development of a comprehensive database on the characterisation and THMC(G) behaviour of different kind of bentonite and mixtures based on the compilation of previous experiments and from new experiments to be carry out in this project. These efforts contribute to the optimisation of DGR designs, improved performance assessment, and more reliable simulations of repository evolution.

The work package is divided into four tasks:

**Task 1: Management and coordination.** This task also includes dissemination and quality control.

**Task 2: Knowledge Management.** The objective of this task is to:

- Capture knowledge relevant to the WP, gained prior to EURAD-2 and extended during this WPs progress,
- Deliver specific activities to transfer knowledge to interested parties. At least one face-to face-training in the form of a doctoral school will be arranged.
- Development of a database on the characterisation and THMC(G) behaviour of different kind of bentonite and mixtures based on the compilation of previous experiments and from new experiments to be carried out in ANCHORS

**Task 3: Laboratory testing.** Multiscale experimental characterization of a wide range of bentonite types (e.g., Na and Ca bentonites from different locations) and bentonite-based mixtures. Specific focus on chemical loadings, heterogeneity, role of friction in link with scale effects and mixture optimisation (influence of variations in the bentonite-based mixture components), laboratory characterization of “aged or matured” bentonite/mixtures coming from “in situ” tests or mock-up tests (if samples are available). Experimental groups will be asked to analyse a different kind of bentonites or bentonite mixtures in addition to the suggested materials to achieve the WP objective on material optimization and provide

supplementary data for the database. This task will also include an assessment of measures for better quality control and testing of bentonite.

**Task 4 Bentonite Barrier modelling and Performance assessment.** The first objective of this task is to improve existing constitutive models and numerical tools. This includes micro/macros coupling, consideration of heterogeneity and transient behaviour. Some constitutive models are going to be extended from THM to THMC for considering the salinity (saline water infiltration) and alkalinity (from cement components) of the porewater on deformations. Some hydromechanical models are going to be improved, the capability of the heterogeneity simulation analysed and implemented, and the friction effect will also be analysed to be implemented or improved in computer codes. Complex structure of bentonites and bentonite-granular mixtures can be studied using simpler models considering a single porosity or considering other more complex double or multiple structure approaches. The extension from HC to HMC models is going to be studied, considering the different degrees of coupling. The second objective is to enhance confidence in numerical tools for safety case applications by focusing on large-scale bentonite barriers components. This includes investigating the sensitivity of parameters in the long-term evolution of bentonite barriers, its quantification and propagation of uncertainties carrying out cases that will evolve during the project.

The objectives of this Deliverable are to present the state-of-the-art and compile existing data about bentonite materials, as well as the behaviour of bentonite materials in the context of radioactive waste disposal. Previous and ongoing national and Community-supported research programmes have led to detailed understanding of the various key thermo-hydro-mechanical and chemical (THM-C) processes taking place in the bentonite-based materials e.g. HITEC (Oline et al 2024) GAS (Levasseur et al 2024), FEBEX (Kober et al 2021), The prototype repository (Svemar et al 2016), BACCHUS (Neerdael et al 1992), RESEAL (Gens et al. 2009a), ECOCLAY (ECOCLAY II), PEBS (Schäfers et al 2014), DOPAS (White et al 2016), BELBaR (Shelton et al 2018), BEACON (Sellin 2022), NF-PRO (NF-PRO 2008)).

This report aims at presenting the state of the art of the issues that are planned to be studied within ANCHORS. It is not a comprehensive report that describes the bentonite concepts in national programs with safety functions and requirements in detail. Bentonite processes that are outside the scope of ANCHORS are also not covered by this report.

## 2. Strategic research agenda

### 2.1 Overview

The EURAD Strategic Research Agenda (SRA) (EURAD Bureau 2023) has been developed purposely with the aim to present a holistic, integrated view on identified needs of common interest that may require research, development and demonstration (RD&D), strategic studies (think tank), and/or knowledge management activities along the whole chain of radioactive waste management, from cradle to grave.

For domains primarily concerned with implementation of DGRs emphasis is reinforced on the continued optimisation of engineered barrier material and design concepts, particularly to enable industrialisation of these facilities. The term optimisation in the context of implementing geological disposal facilities has widened from focussing initially on post-closure nuclear safety to many other aspects (e.g., design, engineering, cost, environment, acceptance). Optimisation to implement geological disposal has thus become a multi-stakeholder and multi-objective challenge. The waste management concepts, and designs (for different types of waste categories) require huge amounts of manmade and natural materials and related logistics and installation aspects. The performance of the materials is the most important aspect and is handled generally in the safety case. Since disposal projects typically have very long timescales, the needs for updates in materials (innovative materials that become available and could decrease some uncertainties or current materials that will have limited availability in the future) are required in several steps in order to guarantee and/or increase the performance.

Roadmap Theme Overview 3 in the SRA provides background context and summarises existing knowledge in relation to the goal of developing an engineered barrier system, tailored to the characteristics of the waste and compatible with the natural (geological) barrier, that performs its desired functions, for the long-term disposal of radioactive waste. The relevant section for ANCHORS is 3.3 which covers buffers, backfills, plugs and seals and where the aim is to identify appropriate buffer, backfill and seal/plug materials and designs, and confirm their properties, behaviour and evolution for the selected repository concept.

## 2.2 Buffers

Characterised bentonite / clay-based material evolution under specific conditions to provide data on hydro-mechanical, thermal, gas transfer and chemical behaviour.

The expected outcomes and impact are an enhanced understanding of post-closure safety considerations, focussing on remaining open issues (chemical effects, THM effects) of bentonite and clay-based materials. Improved characterisation of different phenomena, including variations of properties arising from barrier installation, hydration history, elevated temperatures and chemical influences (including microbial processes) on long-term evolution behaviour and gas migration. Understanding the THM evolution of bentonite during the thermal phase helps to set requirements on the minimum and maximum density of bentonite near the primary container and better justify the maximum temperature in bentonite.

This is a mature domain, so optimisation and knowledge management of existing know-how are the main drivers. A wide range of bentonite/clay-based materials are available for potential use in deep geological repositories. To enable programmes to make best use of available resources, to optimise barrier designs and to inform the development of supply chain capabilities (material sourcing, demonstration testing) they require a good understanding of the critical properties and the full characterisation envelope, building on the existing understanding of bentonites / clay materials that are already well developed. During the early evolution (thermal phase) of a Spent Nuclear Fuel (SNF) repository, water and vapour redistribute in the vicinity of the canisters governing several processes linked to the sealing properties of bentonites, such as swelling, homogenisation of density differences, mass re-distribution, distribution of voids. Understanding these processes during the thermal phase means that we can better define the initial state and better predict the long-term evolution of the EBS.

## 2.3 Backfills

Improved understanding of mixtures bentonite/crushed rock, bentonite/sand, sand/crushed rock

The expected outcomes and impact for clay based backfills are improved data and understanding of design requirements for disposal systems that are compatible with the use of mixed-clay-based backfills. Increased confidence in simulations by reducing uncertainties in input data and understanding of key processes including hydromechanical behaviour.

Mixture with bentonite, crushed rock or coarse-grained soils (sands) material is one of the key candidates for backfilling of the access tunnels and shafts within the repository. Similar to the buffer material, they should present depending of the repository design, low permeabilities, swelling potential to mitigate development of inhomogeneities forming preferential radionuclide transport paths and to reduce microbial activity, stability under increased temperature and stability over the repository timescale, not gas-tight, low deformations under loading. To date, investigation has mainly focused on bentonite as a buffer material in the form of compacted blocks and pellets. Further investigation of mixtures, both experimental and numerical, is needed for the repository design optimisation.

## 2.4 Plugs and seals

Improved understanding of the performance of plugs and seals

The expected outcomes and impact are to further understand the coupled THMC behaviour of plugs and seals throughout the post-closure phase and to develop improved modelling capability to provide reassurance over the long-term. Establish link to technical feasibility: By means of linking THMC process understanding, safety functions, technical requirements and actual production / emplacement it has to be ensured that plugs and seals will perform as required.

The specific requirements of plug and seal components will depend on their location within a disposal system and site-specific conditions such as geochemistry, the groundwater flow regime, the mechanical stability and the thermal properties of the selected geological environment. Mature plug and seal designs exist for specific disposal systems with acceptable performance. Further understanding would enable tailoring of plug and seal designs for different system requirements or for the optimisation of existing concepts. Demonstrating the performance of plugs and seals is important to build/maintain trust about the feasibility of a safe closure of a disposal facility. Engaging with civil society about this topic is thus important.

## 2.5 Relation between the SRA and ANCHORS

The planned activities in ANCHORS cover many of the aspects discussed in the SRA.

- A number of bentonites with different origin will be studied. This will address the issues about significant variability of measured THM properties of buffer materials that is identified in the SRA.
- Tests will be performed with different geochemical boundary conditions. This will address issues about site-specific conditions geochemistry and chemical influences on barriers that is discussed in the SRA,
- Swelling, homogenisation of density differences, mass re-distribution and distribution of voids will be studied experimentally. In the SRA it is stated that the sealing properties of bentonites, such as swelling, homogenisation of density differences, mass re-distribution, distribution of voids. Understanding these processes during the thermal phase means that we can better define the initial state and better predict the long-term evolution of the EBS.
- Testing of sand-bentonite mixtures. An expected outcome from the SRA is improved data and understanding of design requirements for disposal systems that are compatible with the use of mixed-clay-based backfills. Increased confidence in simulations by reducing uncertainties in input data and understanding of key processes including hydromechanical behaviour.
- Models for the prediction of barrier evolution will be improved. The SRA states that to further understand the coupled THMC behaviour of plugs and seals throughout the post-closure phase and to develop improved modelling capability to provide reassurance over the long-term. Establish link to technical feasibility: By means of linking THMC process understanding, safety functions, technical requirements and actual production / emplacement it has to be ensured that plugs and seals will perform as required.
- Models will be tested on actual assessment cases. This is also covered in the previous bullet.

Although included in ANCHORS, less attention will be given to the effects of high temperature and gas, since that area was covered in the EURAD-1: HITEC (Oline et al 2024) and GAS (Levasseur et al 2024) work packages.

## 3. Role of Bentonite in the Repository Concepts

The purpose of this section is to give a brief overview on how bentonite is used by different waste management organisations and to give a summary of the current research needs in regard to the focus of ANCHORS.

The activities in ANCHORS are, in some respects, related to the activities in earlier EURAD Work Packages and EC and international projects of which many are listed in Section 1. More detail regarding bentonite studies can be found in the State-of-the-Art reports (SotAs) for HITEC (Villar et al., 2020) and BEACON (Wigger et al., 2017).

### 3.1 Czech Republic

#### 3.1.1 Role of bentonite

The current Czech deep geological repository (DGR) concept (Figure 1) is based on the disposal of waste disposal packages (WDP) containing spent nuclear fuel in crystalline rocks in a vertical position (Figure 2). Each WDP will be disposed of in a single vertical borehole, which will be located in disposal tunnels. Bentonite will be used for the buffer (the material that fills the space around the WDPs in the disposal boreholes) and backfill (the material that fills the disposal tunnels and the other underground openings). The Czech concept considers the use of local Ca-Mg bentonites (Hausmannová et al., 2018). Since 2017, calcium-magnesium bentonite, known as BCV (Bentonite Cerny vrch) bentonite, extracted from the Cerny vrch deposit (KERAMOST a.s., Czech Republic) has been considered to be the reference material in the Czech research programme (Laufek et al., 2021; Najser et al., 2023; Najser et al., 2024; Koubová et al., 2025).

The buffer primarily serves to prevent the contact of the WDP with the water and corrosion-active substances present in the local rock environment (chlorides, sulphides) so that, under normal DGR development conditions, both the groundwater and the various corrosion-supporting substances reach the WDP via diffusion only. The high density of the buffer will largely prevent the activity of microbes and thus limit the microbial corrosion of the WDP. The other buffer functions comprise the retardation of the mobility of radionuclides and ensuring both sufficient heat dissipation and the continued mechanical stability of the WDP and the repository as a whole. The main functions of the backfill are to seal the disposal tunnels and other underground openings and to prevent the movement of the buffer so that it maintains its safety functions.

The key properties of bentonite comprise its low hydraulic conductivity, high swelling capacity (the limitation of radionuclide migration, microbial activity and the contact of water with the WDPs and the sealing of the so-called technological gaps), sufficient thermal conductivity (heat dissipation from the WDP) and high sorption capacity (the limitation of radionuclide transport).

The currently preferred design of the buffer comprises a combination of compacted bentonite blocks as the main filling material and pelletised material for the filling of the technological gaps and the levelling of the bottom of the borehole. The target dry density of the blocks is  $1.7 \text{ Mg/m}^3$ , which was selected so that even in the space around the WDP, following the filling of the technological gaps, the required average buffer dry density of  $1.6 \text{ Mg/m}^3$  will be achieved.

The currently preferred design of the backfill comprises compacted pelletised bentonite with a single pellet dry density of over  $2.0 \text{ Mg/m}^3$  thus guaranteeing the achievement of the minimum required average backfill dry density of  $1.4 \text{ Mg/m}^3$  (Hausmannová et al., 2023).

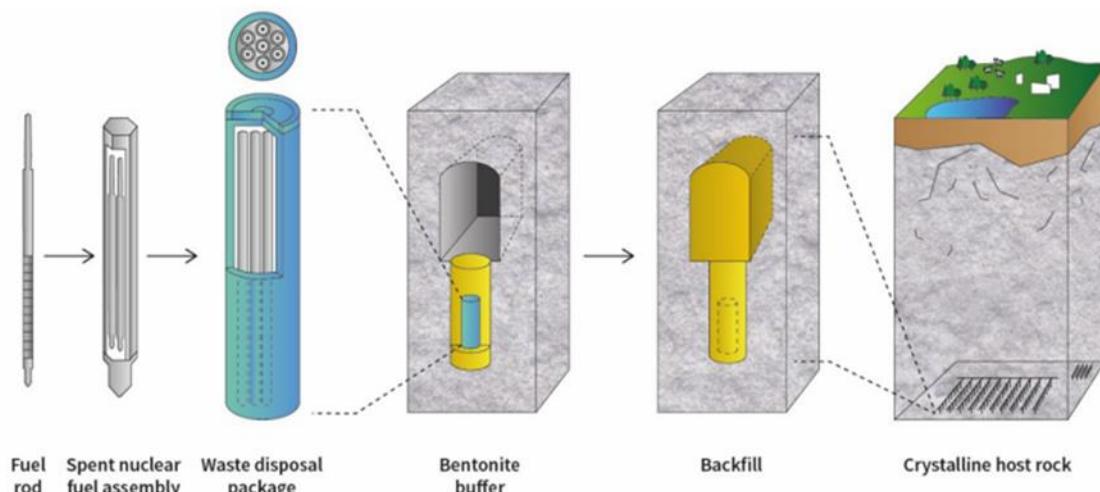


Figure 1 Illustration of the disposal of SNF in the DGR

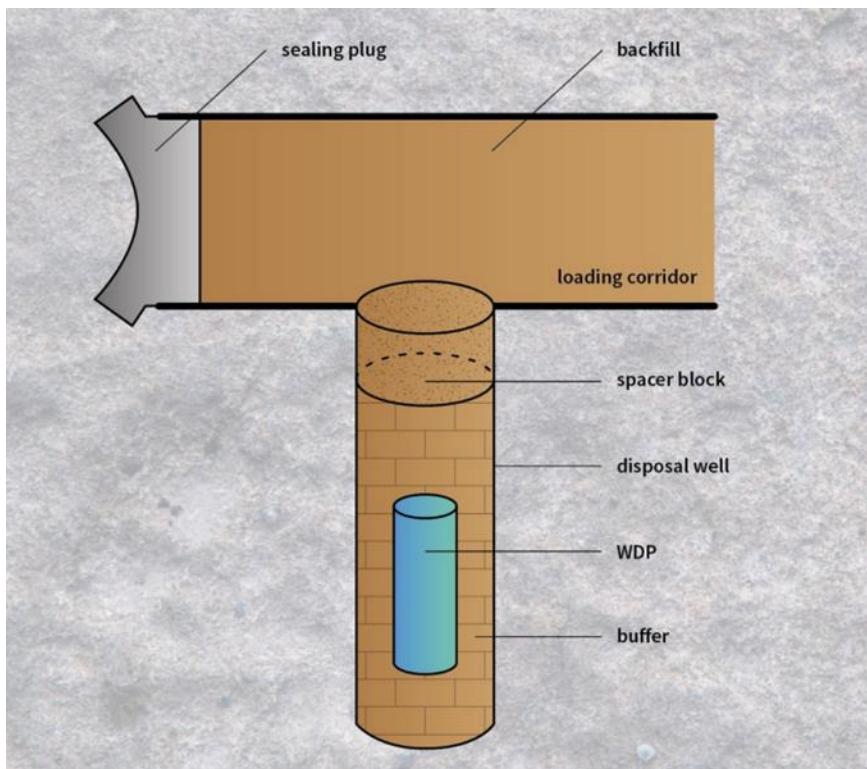


Figure 2 Diagram of the vertical disposal system

### 3.1.2 Current research needs

The current Czech disposal concept is based on the use of local bentonites for the construction of the engineered barriers. Due to the requirement for a robust dataset that accurately describes the stability and suitability of Czech bentonite for engineered barrier use, a range of complex national projects are underway concerning the analysis of existing data (Šachlová et al., 2022) and the conducting of laboratory and modelling experimental programmes aimed at obtaining the remaining data required for the assessment of the stability and suitability of Czech bentonite.

The laboratory programme of one of the related ongoing projects (Šachlová et al., 2022) is investigating the following topics:

- the complete characterisation of BCV bentonite loaded to various extents,
- the characterisation of alternative Czech bentonites,
- the analysis of the effect of pelletization on the hydromechanical properties of the bentonite,
- research into the potential microbial degradation of the bentonite.

Based on the results of the THM mathematical modelling (Krejčí et al., 2024), a number of important issues have been clarified, including the impacts of various initial bentonite moisture contents on the temperature of the outer casing of the WDP, the effect of differing variations of bentonite saturation from the host rock and various length of the spacer block on the uplift of the buffer, and the associated moisture and dry density redistribution processes that will act within the bentonite barrier both temporally and spatially.

The Engineered Barrier 200C project is investigating the hydromechanical properties of bentonite exposed to high temperatures of above 150°C (Svoboda et al., 2023). Proof that Czech bentonite is stable and performs its various functions at high temperatures will contribute to the optimisation of the dimensions of the repository and will help to enhance confidence in terms of its safety. The effects of high temperatures on bentonite-based barriers and their safety functions are also being investigated with concern to Czech bentonite as part of the HotBENT project (Kaufhold et al., 2024; Kober et al., 2024).

The phenomenon of the stress relaxation of bentonite at high temperatures that was observed in the HITEC WP of the EURAD project (Graham et al., 2023) requires further investigation. Enhancing the understanding of this phenomenon is important with respect to the long-term stability of the hydromechanical properties of the bentonite barrier.

Another key research topic that requires further investigation concerns the potential mechanical and chemical erosion of Czech Ca-Mg bentonite under the conditions expected in the future Czech DGR.

## 3.2 Finland

### 3.2.1 Use of bentonite

In Finland, spent nuclear fuel from current operating nuclear power plants will be embedded in Olkiluoto bedrock at a depth of 400-450 metres. Bentonite will be used in buffer that surrounds the vertically installed disposal canisters placed in deposition holes (Figure 3), backfilling of deposition tunnels, closure material composed of crushed rock-bentonite mixture that is used to fill central and access tunnels and for example as specific hydraulic plugs in closing of surface boreholes.

Bentonite is used to isolate the canisters from crystalline rock environment. In practice this means limiting transport of harmful substances from host rock to canister, protecting the canister in event of earthquake induced rock shear and limiting radionuclide transfer in case of canister failure. Additionally, the buffer needs to suppress microbial activity and conduct the decay heat from spent fuel to host rock.



Figure 3 A canister in a deposition hole

### 3.2.2 Current research needs

Posiva has submitted operating licence application for the spent fuel repository, Posiva SC-OLA (Posiva 2021). Current interests are in reducing leftover uncertainties and optimisation of the system with emphasis on robustness of manufacturing, installability and performance. Such work could include for example characterisation and model parametrisation of new bentonites and testing composition of closure material mixtures.

## 3.3 France

### 3.3.1 Use of bentonite

Cigéo (Industrial Centre for Geological Disposal) closure will happen after more than one hundred years corresponding to the operational phase. The objective of the closure system putting in place at this

moment is to isolate the radioactive waste from the biosphere. To assure this isolation, all the drifts will be backfilled, and several seals will be built at relevant locations.

Seals are installed locally in shafts, ramps, and galleries to limit water movement within the repository. This requirement is directly linked to the safety of the repository and contribute to the radionuclide confinement (Andra, 2022).

Function of backfill is to limit the development of the fractured zone after the rupture of the concrete lining strengthening the sides and roofs of the drifts in the underground facility. It helps to preserve the favourable characteristics of the Callavo Oxfordian clay (COx).

The main component of the seals is a core composed of a bentonite-based material. This material is used to achieve a low hydraulic permeability and must have a swelling capacity to fill the technological voids and support the surrounding clay. Two types of design are explored for the horizontal seals: (i) the core installed between two concrete blocks - reference configuration (Figure 4a), (ii) the two extremities of the core are in contact with the backfill (Figure 4b). For both horizontal seals, only a few parts of the concrete lining are removed due to the nature of the clay rock (high clay content). In shafts and ramps, the main difference compared to horizontal tunnels is that all the concrete lining is removed along the swelling core (Figure 4c and d). This is made possible by the high carbonate content in the upper part of the COx.

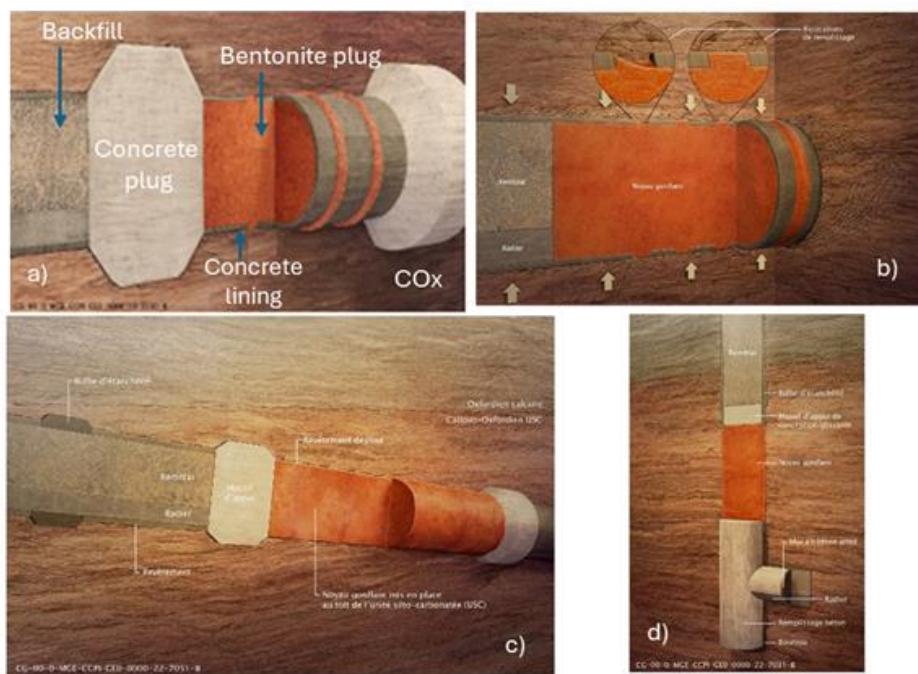


Figure 4 Seal designs – (a) horizontal tunnels core is confined between two concrete abutment (b) horizontal tunnels core is in contact with backfill, (c) in a ramp, (d) in a shaft

The seals and backfill are also designed to be gas-permeable, allowing for the dissipation of gas pressure without disrupting the hydraulic and mechanical functions of these closure components. The aim is to keep the gas pressure within the repository below fracturing levels that are defined by the minimum principal stress.

The two expected functions of seals are, a priori, antagonistic: limiting the flow of water and allowing gas to pass through. Pure or blended bentonites have low water permeabilities over a wide range of dry densities, making it easy to fulfil the sealing function regarding water flow. Improving the ability of seals to be 'gas-permeable' naturally leads to a move towards mixtures containing sand.

Sand is a variable for adjusting the properties of bentonite-based materials to the expected specifications (Zeng et al, 2022). The addition of sand generally makes it possible to control swelling

pressure, water permeability, gas transfer capacity and/or thermal conductivity. Tests carried out for mixtures ranging from 20% sand content to 70% lead to define a target sand/bentonite mixture with a sand percentage around 60%. This mixture fulfils the water/gas transfer requirement and keeps a high swelling potential. In order to achieve the right level of dry density for the bentonite/sand mixture and to obtain a good homogeneous distribution between the sand and the bentonite, it is envisaged at this stage to construct the seal in the form of pre-compacted blocks.

The same approach is retained for the backfill materials which is mainly composed with excavated crushed Callovo-Oxfordian claystone. COx is mixed with sand to improve the mechanical properties and to facilitate gas migration. Due to the large length of the tunnel to fill, the solution chosen at this stage, for reasons of efficiency and cost, is to compact the mixture directly in the drifts. Increasing the sand content, increases the dry density at the Proctor optimum, reduces the water content and increases the water permeability. Mixture with a sand content between 70% and 30% are investigated.

### 3.3.2 Current research needs

A large amount of work has been done on pure bentonite materials or mixture with a low sand content. For these materials, the behaviour is mainly driven by the clay matrix. Due to gas issues, bentonite/sand mixture with high sand content are investigated by several Waste Management Organizations (WMO). Consolidation of knowledge on these mixtures is needed, in particular concerning the properties to gas or the chemical reactivity of these sand/clay mixture (The clay can be either bentonite or claystone, depending on its intended use).

In Cigéo project, clay-based materials (bentonite/claystone) can be in contact with cementitious components. Knowledge on the evolution of these mixtures subjected to an alkaline plume needs to be consolidated, with short-term effects induced in particular by cation exchange, and long-term effects leading to mineralogical transformations. Mains issues concern the consequences on the swelling pressure and on both water and gas permeabilities.

The homogeneity of the material in terms of distribution of sand and clay in the compacted material can be impacted by the way of preparation of the mixture, the particle-size distribution of each component and the sand content. Material organization and phases distribution could have an influence on the hydromechanical properties. This effect needs further investigation related to the quality control of the final material in industrial context. The choice of clay sand mixture with, in some cases, a high sand content (more than 50%) could lead to an amplification of these effects particularly as regards the role of chemistry on long-term hydromechanical behaviour or material homogeneity.

Modelling swelling clay components and their hydromechanical evolution integrating gas migration or chemical evolution is still a great challenge. In the last decade, an important effort has been made in terms of model developments to represent chemical perturbation in bentonite-based materials. One of the main issues is to model how bentonite may be impaired by the geochemical interaction and especially alkaline plume or water salinity.

Most of the models developed consider pure bentonite hydromechanical evolution. The use of bentonite sand mixture with a high sand content can lead to revisit these models to take into account the arrangement of these two components and their consequences on the hydromechanical-gas behaviour.

## 3.4 Germany

The enforcement of the Site Selection Act (StandAG), enacted in 2013 and revised in 2017, marked the restart of the site selection process for a repository for high-level radioactive waste in Germany (StandAG 2013, 2017). In accordance with § 1(3) StandAG, the Federal Company for Radioactive Waste Disposal (BGE) considers three host rock types for the repository: rock salt, claystone, and crystalline rock.

Beginning with a blank map, the site selection procedure is conducted in three sequential phases that systematically narrow down the considered areas. During the first step of Phase 1, BGE identified sub-

areas across Germany exhibiting favourable geological conditions for hosting a high-level waste repository (BGE, 2020). This evaluation identified 90 sub-areas, encompassing approximately 54% of Germany's total land area, still taking into consideration the three host rock types.

In the second step of Phase 1, BGE is conducting representative preliminary safety analyses on each of these 90 sub-areas to evaluate and give recommendations to narrow down to suitable regions for further exploration in Phases 2 and 3. As part of Phase 1 and in accordance with German Safety Requirements § 6(4) EndlSiUntV (Ordinance on Requirements for Conducting Preliminary Safety Analyses in the Site Selection Procedure for the Disposal of High-Level Radioactive Waste), a preliminary repository design is under development. This design must include key aspects, such as the description of essential barriers in accordance with § 4(3) EndlSiAnfV (Ordinance on Safety Requirements for the Disposal of High-Level Radioactive Waste), their basic characteristics, spatial extent, and additional components of the repository system, including potential sealing and backfill measures.

BGE is currently developing generic repository systems and sealing concepts for the three host rock types as part of the representative preliminary safety analyses. According to § 4(2) EndlSiAnfV, the designated repository system must ensure the safe containment of radioactive waste over an assessment period of one million years by means of a robust, multi-barrier system with different safety functions. Following § 4(3) EndlSiAnfV, the safe containment of radioactive waste can fundamentally be achieved through two types of essential barriers (BGE, 2022):

- Repository System Type 1: Safe containment is ensured by one or more containment-providing geological formations. This containment type is applicable in all three host rocks.
- Repository System Type 2: In crystalline host rock, where no containment-providing geological formation can be identified, safe containment is achieved through technical and geotechnical barriers tailored to the geological environment. If a containment-providing geological formation is identified, both containment types may be applied (see Ch. 8.3.1 BGE, 2022).

An important element of the repository concepts in claystone and crystalline rock is the use of bentonite in closure and sealing operations. The following section addresses the use of bentonite in these concepts.

### 3.4.1 Use of bentonite

Bentonite in a claystone formation

In Germany, a final repository concept for a claystone formation has not yet been established, and ongoing work is being conducted on a generic basis, taking into account advanced international concepts like NAGRA. At the moment, clays are considered for the backfilling of various underground structures, including disposal galleries, crosscuts, access galleries, infrastructure areas, as well as in sealing structures (see Figure 5). Primarily, swellable clay minerals, such as bentonite, or processed, site-specific excavation material, are used. These materials can be applied in their pure form (e.g., in disposal galleries) or in mixtures with other materials such as sand.

In the disposal galleries, highly compacted bentonite blocks are foreseen to form a pedestal for the placement of the waste packages. The remaining space in the disposal galleries can be backfilled with a granular material made of swellable clay minerals. The primary function of this backfill is to reduce or close the space between the excavation lining and the waste package. This facilitates the transfer of heat from the container's surface through contact surfaces. In this regard, the thermal properties of the backfill plays an important role as the temperature at the surface of the waste container should not exceed 100°C. Thus, backfill with higher thermal conductivities are preferred. The backfill should also minimise fluid movement toward or away from the waste package. When groundwater slowly infiltrates, the bentonite begins to swell, further reducing any remaining voids. The resulting swelling pressure compacts the material, thereby reducing its permeability to fluids. This prevents or limits advective transport and slows diffusive transport. Additionally, microbial activity is reduced as the swelling

bentonite minimises pore space within the bentonite and restricts water availability, thereby minimising the suitable habitats for microbial growth and thereby potential microbial corrosion of the canisters.

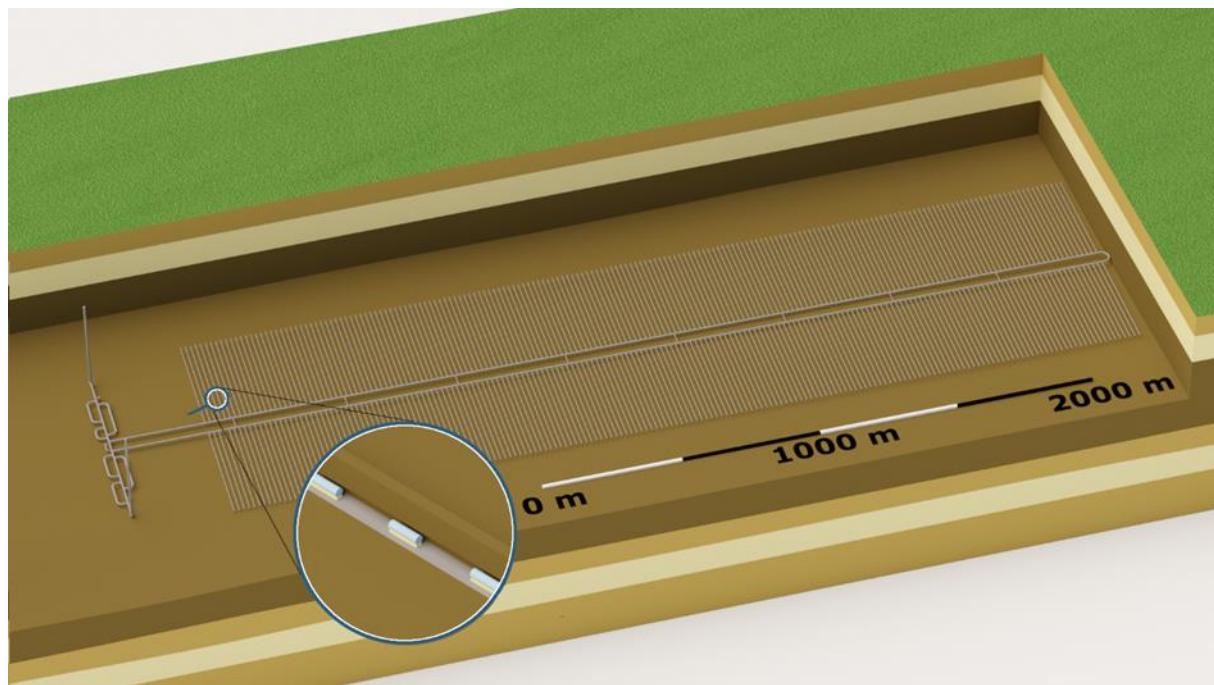


Figure 5 Generic concept of a repository in claystone formations in Germany with illustration of the bentonite pedestal in the disposal gallery (Lohser, 2024).

To prevent damage to the host rock and the waste packages, the maximum swelling pressure must be controlled by adjusting the density, type, and form of the bentonite material used. This will help to mechanically stabilise and support the excavated areas after the closure of the repository. At the same time, the bentonite is designed to create and maintain a stable, reducing chemical environment, thus protecting the repository container from corrosion.

Sealing structures are planned at the end of each disposal drift to prevent potential radionuclide transport pathways and limit fluid movement along the excavations. Figure 6 illustrates a possible sealing structure at the edge of a disposal drift and near the repository shaft.

To ensure direct contact between the host rock and the sealing material, the excavation lining will be (partially) removed in the area of the sealing element to ensure a direct contact with the surrounding rock. This measure aims to eliminate potential pathways along the contact zone between the lining and the host rock, ensuring a long-term sealing effect. If necessary, additional structural support elements, such as steel rings, may be incorporated.

As mentioned before, bentonite will be used for the sealing elements within the sealing structures. The swelling behaviour of bentonite, combined with the creep properties of claystone, enhances the contact between the sealing material and the host rock. This interaction ensures a durable and effective seal, providing long-term containment of radionuclides.

At this stage, no specific design or type of bentonite has been selected for the backfill and the sealing structures. Possible options include fine-grained materials, granulates, pellets, or blocks. Mixtures of these materials, e.g. to increase gas permeability to prevent high gas pressures, are also under consideration as potential solutions.

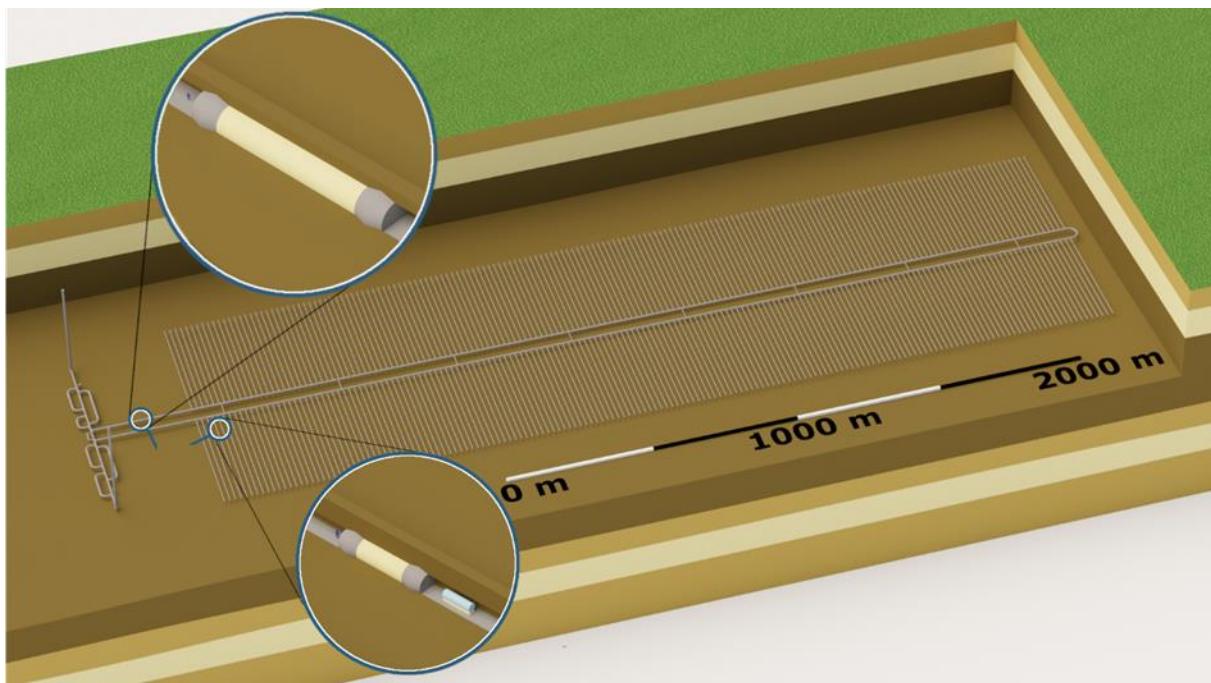


Figure 6 Illustration of a gallery seal in a repository for radioactive waste in claystone formations in Germany (Lohser, 2024).

To prevent the ingress of solutions from the overlying rock into the repository, which could facilitate the release of radionuclides, a sealing element made of bentonite will be placed within the shafts, similar to the sealing structures used in disposal galleries. Depending on specific requirements, this sealing element can be made from granulates, pellets, or blocks.

The bentonite sealing elements will be anchored between two structural supports. This design ensures that, upon contact with infiltrating solutions, the bentonite will swell, reducing permeability, and thus creating the desired sealing effect against fluid movement and radionuclide transport.

As with other sealing structures, the excavation lining along the shafts in the area of the sealing element installation will be (partially) removed to ensure direct contact between the bentonite and the surrounding rock.

#### Bentonite in a repository in crystalline rock

Within the scope of the representative preliminary safety analyses, a single repository concept is considered for both Repository System Type 1 and Repository System Type 2 in crystalline host rock. The backfilling and sealing measures are partially based on the requirements for Repository System Type 2, given the higher demands on the disposal container and surrounding buffer. Additionally, the repository concept considers the geotechnical barriers for the sealing of the containment-providing geological formation as these barriers must meet particularly high long-term sealing requirements. Thus, the sealing and backfilling measures are therefore described from the perspective of the Repository System Type with the more stringent requirements.

The repository concept for disposal in crystalline formation is still under development by BGE. The current working hypothesis follows the approach of Posiva Oy and is based on the KBS-3 concept (see sections 3.2 and 3.6). The KBS-3 concept was further modified to better account for safety concept and regulations in Germany (Jobmann & Burlaka, 2021). The repository is designed for vertical borehole disposal, where the disposal container is surrounded by bentonite blocks, and the borehole galleries are filled with a mixture of bentonite and excavation material (Lohser, 2024).

For backfilling around the disposal container, both rings and plates made of compacted bentonite are considered. The bentonite plates are placed above and below the disposal canister, which is itself

surrounded by bentonite rings. A filter layer may be applied to the top bentonite plate to facilitate even distribution of infiltrating fluids. The voids between the disposal canister, bentonite blocks, and the surrounding rock can be filled with bentonite pellets. Over time, as groundwater infiltrates, the material will swell, sealing remaining voids and potential fractures. The resulting swelling pressure compacts the material, reducing permeability to fluids, thereby hindering advective transport and slowing diffusive transport of radionuclides. The retardation of radionuclides migration is further supported by the high sorption capacity of bentonite.

Gallery seals are implemented within the repository to minimize groundwater flow and radionuclide transport along the gallery network. Their long-term safety contribution is particularly valuable in low-permeability rock formations, where the surrounding rock acts as a natural barrier. In such cases, gallery openings should be sealed to maintain this barrier function with emphasis on the retardation of radionuclides migration. In higher-permeability rock, gallery seals may be subject to fluid bypassing, serving primarily to slow down flow processes within the former mine infrastructure and protect backfill material from erosion caused by advective flows.

Figure 7 illustrates a schematic representation of a gallery seal designed for long-term containment. It consists of series of abutments, sealing elements made of asphalt and bentonite and of gravel elements serving as water reservoirs. The gallery seal is made of five individual sealing structures to ensure redundancy. Each sealing structure consist of one (bentonite) or two sealing (bentonite and asphalt) elements emplaced between two abutments. According to Jobmann & Burlaka (2021), the gallery seal is secured on both sides by a double abutment system. The abutments serve to stabilize both the backfill material and sealing elements until the swelling pressure is high enough to maintain the bentonite elements in place. The asphalt seals provide immediate fluid tightness upon installation. As the adjacent bentonite seals saturate, they exert pressure on the asphalt sealing element. Due to its low viscosity, the asphalt is pressed against the drift walls, and depending on its bitumen content, it may also penetrate into the excavation-damaged zone, effectively sealing it. The saturation of bentonite elements occurs via saturation chambers filled with gravel, which distribute incoming water evenly over the filter layers. The modular design of the drift seal allows for flexible adaptation to different drift lengths depending on spatial constraints.

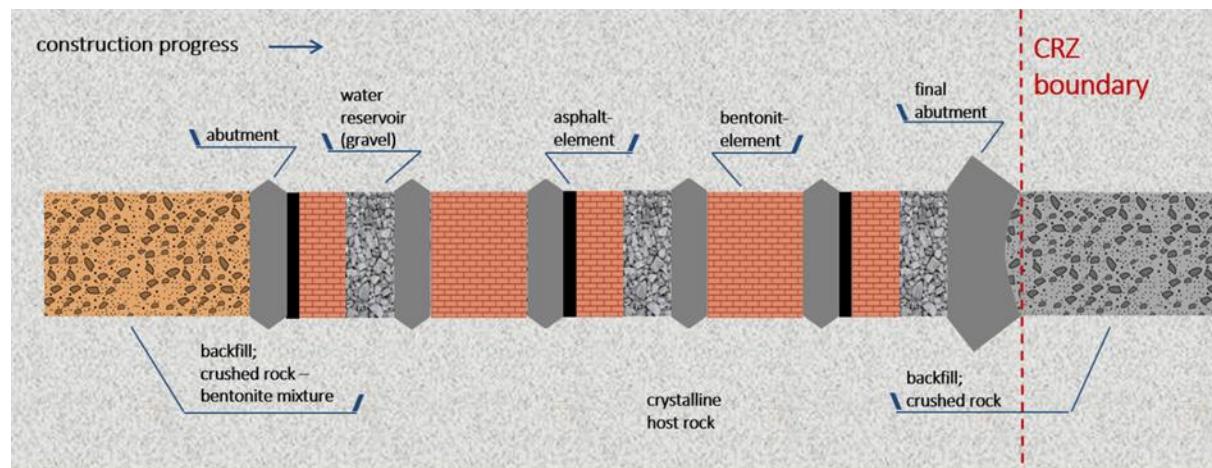


Figure 7 Schematic representation of a gallery seal in a low permeability area of the crystalline rock formation (Jobmann & Burlaka, 2021)

For backfilling in the underground repository (emplACEMENT galleries, crosscuts, and access galleries), a mixture of processed excavation material and bentonite can be used in all voids where no sealing elements are planned or where larger fractures are encountered. A key advantage of using site-derived excavation material is its consistent availability in homogeneous quality and its chemical similarity to the host rock.

### 3.4.2 Current research needs

In Germany, research on the THMCB behaviour of bentonite was less in focus, primarily due to a historical long-standing preference of salt rock repository concepts. However, as repository designs evolve, critical questions arise regarding the long-term behaviour of bentonite, particularly its potential degradation due to thermal effects or erosion over time periods extending up to one million years – the verification time for safety case of repositories for radioactive waste in Germany.

In current repository concepts for claystone and for crystalline rock, a temperature limit of 100°C at the surface of the waste package has been defined by BGE in accordance with the StandAG regulation (BGE, 2024) based on several research projects on the thermal compatibility of clay materials. Because bentonite will be subjected to high temperature, a better understanding of the THMC coupled processes occurring in bentonite is required. This includes the development of advanced numerical tools and constitutive models capable of accurately predicting the long-term behaviour of bentonite under repository conditions and support the safety analyses.

The specific type of bentonite or bentonite mixtures to be used remains an open question. Targeted research and development are necessary to inform the design and optimisation of bentonite barriers.

## 3.5 Spain

### 3.5.1 Use of bentonite

The Spanish repository concept in plastic clay rock is based on the disposal of spent fuel in carbon steel canisters in long horizontal disposal galleries. Canisters are disposed of in cylindrical disposal cells constructed with pre-compactated bentonite blocks of 1.7 Mg/m<sup>3</sup> dry density (to achieve a final dry density of 1.7 Mg/m<sup>3</sup>). The blocks are initially non-saturated (degree of saturation of 66%). The disposal galleries of 580 m in length and 2.4 m in diameter are located at a depth of 250 m in the host formation. A 0.3 m thick concrete liner is required to deal with the plastic nature of the clay host rock. The separation between canisters is determined mainly by thermal constraints. Separations of 1 m between canisters and 50 m between disposal galleries have been established, in order not to exceed a temperature of 100°C in the bentonite. Actual separation is a function of the properties of the host rock. Once a disposal gallery is completed, it is sealed with a 6-m long seal made of bentonite blocks and closed with a concrete plug at its entry. After completion of all the disposal galleries, the main drifts, ramp, shafts and other remaining rock cavities will be backfilled with compacted clay from the excavation of the repository, and subsequent projection of clay pellets in the remaining openings. The concept is shown in Figure 8.

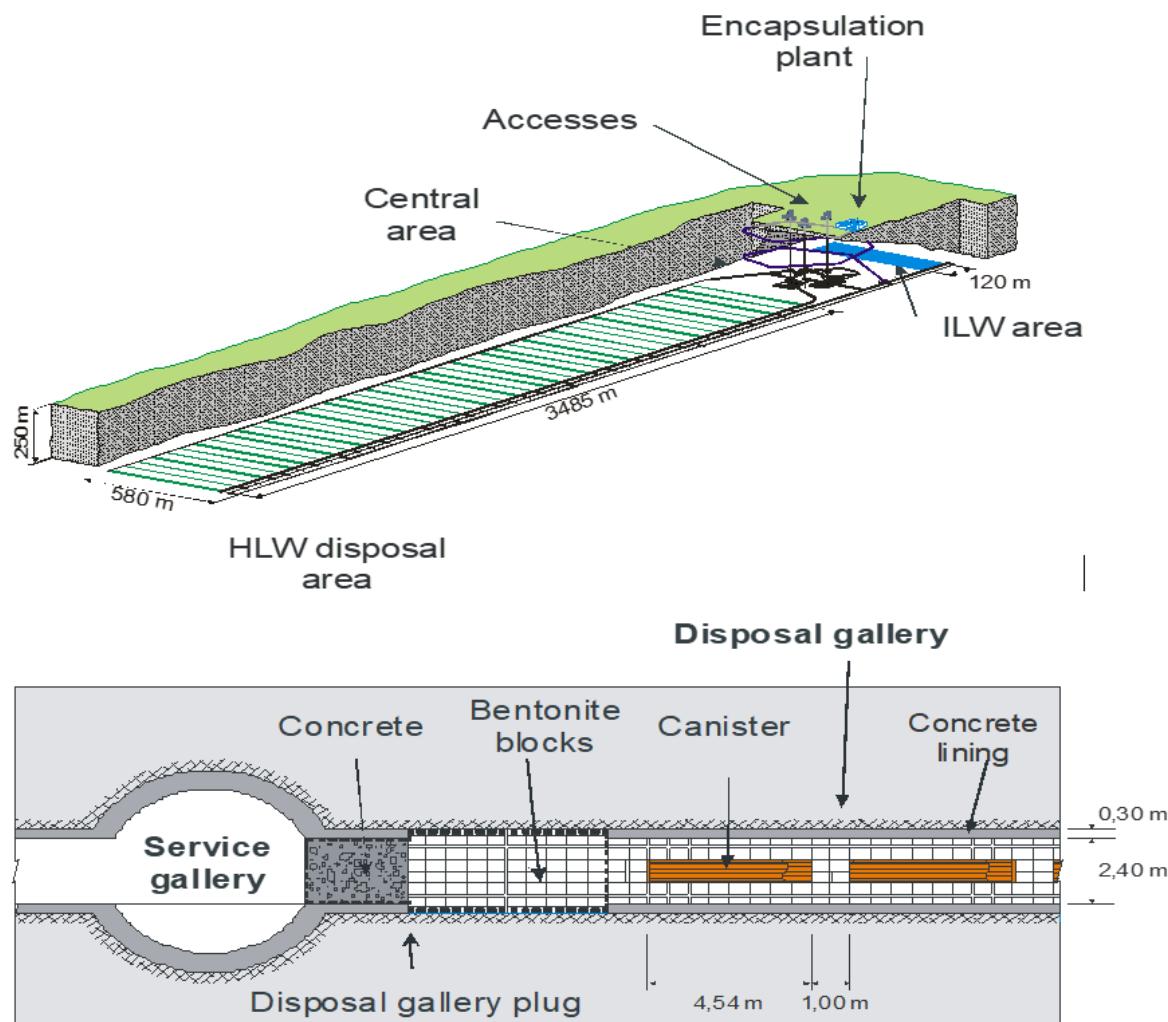


Figure 8 Schematic diagram of the geological repository in clay rock

The bentonite buffer is required to maintain a large diversity of safety functions, which can only be fulfilled once the bentonite saturates and swells, tightly closing the construction gaps between the bentonite blocks and the liner or the canister wall on the one hand and between the blocks themselves on the other. Nevertheless, there are no safety functional requirements applicable during the time when the canister provides absolute containment. During the re-saturation of the buffer, the main concern is the preservation of the favourable properties of the buffer material. As the safety functions assured by the buffer are assumed for the full duration of the quantitative safety assessment (on the scale of a million years), its properties have to be preserved at a sufficient level for commensurate periods of time.

The long-term safety functions of the bentonite buffer are to:

- Isolate the waste package from the geosphere by limiting advective transport of corrodng agents to the canister.
- Avoid canister sinking in the disposal drift that could result in direct contact of the canister with the rock, hence short-circuiting the buffer.
- Avoid excessive swelling pressures that could contribute to total pressures that the canister cannot withstand.
- Avoid excessive temperatures ( $>100^{\circ}\text{C}$ ) that could result in chemical alteration of the bentonite and jeopardize its safety functions.
- The buffer is a containment barrier by itself, as it retains radionuclides based on its properties:

- Low hydraulic conductivity, which makes radionuclide transport by advection negligible
- Sorption of many radionuclides, especially actinides
- Filtration of colloids and large complex molecules because of the small size of the pores
- Avoid the build-up of excessive gas pressure in the near-field, without undue impairment of the safety functions.
- Reduce microbial activity to minimize microbial corrosion of the canister

### 3.5.2 Current research needs

Enhancement of existing constitutive models and numerical tools by focusing on two novel aspects for the advancement of knowledge of bentonite as well as the improvement of the predictive capabilities of the models. Specific characteristics are:

- Greater understanding and improvement of dual structure models based on the concept of micro porosity and macro porosity. One of these models is the BExM model on which continuous improvements have been made.
- Study of the heterogeneity of materials through dry density measurements that allow a better understanding of the percolation processes of both liquid water and vapor and gas.
- Geochemical coupling focused on two main themes: the effect of salinity and the effect of alkalinity on the behaviour of bentonites as an impermeable barrier element in the repository.

Increase of confidence in numerical tools for safety case applications by focusing on large scale bentonite barriers experiments.

## 3.6 Sweden

### 3.6.1 Use of bentonite

The main function of the clay barrier is to limit the water flow around the canister and in the deposition tunnels in the Spent Fuel Repository, around the waste transport casks and in the plugs for the silo in the Final Repository for Short-lived Radioactive Waste (SFR) and in the vault for long-lived historical waste (BHA) in the Final Repository for Long-lived Waste (SFL) (Figure 9) and to limit advective transport in the deposition tunnels in the Spent Fuel Repository. This is achieved by means of low hydraulic conductivity in the clay, so that diffusion is the dominant transport mechanism, and by means of a swelling pressure that makes the buffer self-sealing. In the Spent Fuel Repository, the buffer will also hold the canister in place in the deposition hole, mitigate the shear movements of the rock and retain its properties during the period being analysed. In addition, the buffer should limit microbial activity on the canister surface and filter colloidal particles. An important function of the backfill in the Spent Fuel Repository's deposition tunnels is also to keep the buffer in place in the deposition hole. The clay barriers must not significantly impair the function of the other barriers.

Closure includes plugs, material installed in boreholes and the material used for sealing all underground openings outside the waste vaults (the waste vaults in SFR) and SFL as well as the deposition tunnels in the Spent Fuel Repository).

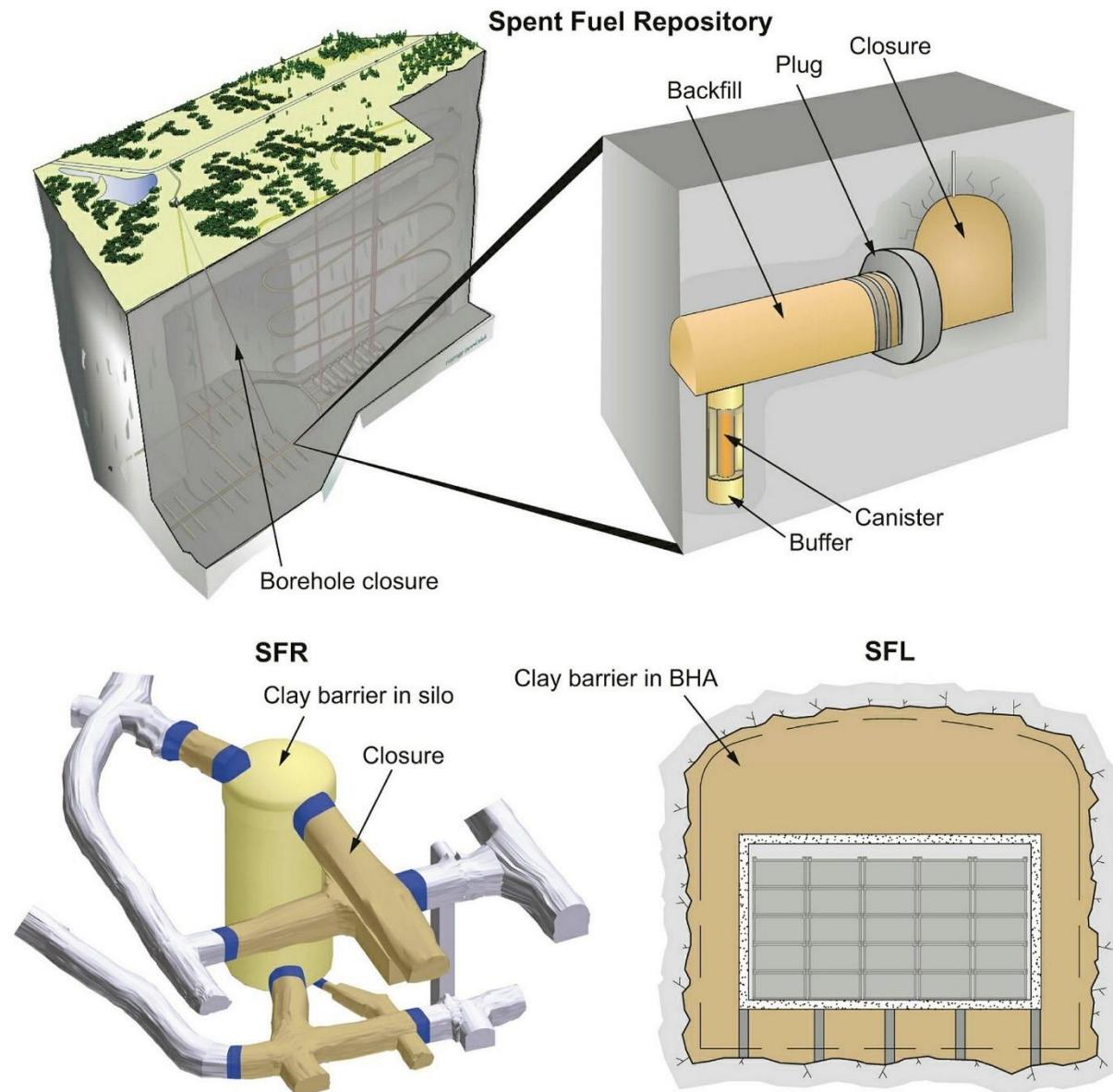


Figure 9 Upper: Clay barriers and closure of the Spent Fuel Repository: buffer, backfill, closure and borehole seal. Lower, left: silo clay barrier and closure in SFR. Lower, right: clay barrier in the waste vault for legacy waste (BHA) in the Final Repository for Long-lived Waste (SFL).

### 3.6.2 Current research needs

The main focus for the activities concerning the long-term performance of bentonite barriers at SKB is currently on licence application for the operational licence spent fuel repository at Forsmark. This means that all the assessment cases from the licence application for the construction licence (SKB 2022a) needs to be updated. The cases presented in SKB 2022 were to a large extent taken from the SR-Site (SKB 2011) safety case. The actual modelling was done around 2007-2008, which means that the models used are outdated today. In a first stage the models that will be used will be further developed, tested and verified. Some activities that are planned are:

- The conceptual description and the modelling tools used for water uptake predictions will be updated, focusing on both the hydraulic properties of the rock and the ability to represent channel formation in the bentonite.
- Continued studies of moisture redistribution from the buffer to the backfill will enhance understanding of the natural convection's contribution to the redistribution of moisture from warm to cold parts of the bentonite in the buffer and backfill. This work primarily involves model validation.

- The implementation of the so-called "Hysteresis-based material model" (HBM) in Comsol Multiphysics, along with the development and generalization of this model, will also include testing its performance against experimental data. This work will continue within frameworks such as ANCHORS.
- Research on long-term stability regarding creep, hysteresis, and friction aims to improve understanding of hydromechanical processes in bentonite over extended periods. This includes the impact of ion exchange ( $\text{Na}^+ \rightarrow \text{Ca}^{2+}$ ) on long-term stability, particularly regarding the stability of residual density gradients after homogenization and/or those arising from wall friction. Canister settlement may also be of interest. This work will involve laboratory tests, modelling, and theoretical studies.
- Water uptake modelling within the Task Force EBS and as part of the evaluation of the Prototype Repository aims to develop and validate the modelling tools used for water uptake predictions by applying them to various experiments.
- Analysis of the Prototype Repository will include studying the swelling of the buffer into the backfill and the homogenization of the buffer. These analyses will contribute to validating the material model for bentonite, particularly concerning hydromechanical process.

The idea is also to test the models on the earlier assessment cases to ensure that all cases can be handled with the models that are available today.

### 3.7 Switzerland

The concepts for implementing deep geological disposal of high-level waste (HLW) and low- and intermediate-level waste (L/ILW) in Switzerland have been developed over many years of research, development, and demonstration (RD&D). These efforts, led by Nagra, have included extensive studies on safety, design, cost, and site evaluation, as documented in reports such as Nagra (2024).

#### Protection Objective

Switzerland's protection objective for deep geological disposal is to ensure the long-term safety of humans and the environment from ionising radiation. This must be achieved without imposing undue burdens or obligations on future generations. To meet this goal, the repository design relies on a system of multiple safety barriers, complemented by additional protective and preventive measures.

#### Post-Closure Barrier System

The post-closure barrier system consists of several interdependent components, each contributing to the containment and isolation of radioactive waste:

- **Overall Geological Situation:** The natural geological setting serves as the primary containment structure.
- **Waste Matrices:** The physical and chemical forms of the waste are designed to limit radionuclide release.
- **Spent Fuel/HLW Disposal Canisters:** Robust canisters ensure the containment of high-level waste over long periods.
- **Buffer in the Emplacement Drifts:** Compacted bentonite provides a protective layer surrounding the canisters, ensuring mechanical stability and limiting water and gas movement.
- **Backfill of Underground Structures and Individual Seals:** Backfilling materials prevent open pathways for radionuclide migration and contribute to the stability of the repository.
- **Host Rock and Confining Rock Units:** The host rock, along with additional rock layers, forms the containment-providing rock zone (CRZ), offering substantial barrier efficiency.

The interactions between these barriers are influenced by the spatial arrangement of the waste and the repository's architecture. While each barrier evolves over time, all are designed to collectively provide a robust and integrated safety function by limiting radionuclide release across the timescales required for safety assessments.

### 3.7.1 Use of bentonite

In the provisional repository concepts, compacted bentonite plays a key role in ensuring safety due to its favourable properties, such as low permeability, swelling capacity, and chemical stability. Its applications include:

- **Buffer Surrounding SF/HLW Canisters:** Compacted bentonite blocks and granular bentonite create a buffer to stabilize canisters and limit water flow.
- **Sealing Elements in L/ILW Repository:** A sand/bentonite mixture is used to enhance sealing performance in the L/ILW sections.
- **Sealing and Closure Structures:** Granular bentonite and/or bentonite blocks are used in various repository sealing and closure components.

This comprehensive use of bentonite contributes to the effectiveness of the multiple-barrier system, ensuring long-term containment and isolation of radioactive waste.

### 3.7.2 Current research needs

In the current provisional repository design, the understanding of bentonite behaviour is considered sufficient. However, less attention has been given to optimizing the backfill materials for construction and operation tunnels. The backfill in these tunnels plays a significant role in the overall post-closure safety of the repository, primarily by influencing the storage and transport of corrosion gases.

Currently, crushed Opalinus Clay is proposed as the primary material for backfilling these tunnels. However, this material has undergone only limited characterization. To enhance its performance, particularly with respect to gas management, the addition of bentonite and/or sand may be considered. Such modifications could improve its properties, including its ability to handle gas-related challenges.

In a clay-rich host rock such as Opalinus Clay, gas diffusion through the host rock is inherently slow. Engineering solutions, including gas-permeable sealing sections, are planned to address potential risks associated with gas overpressures. In the current repository concept, the backfill of construction and operation tunnels is integral to gas mitigation. High porosity and gas accessibility in the backfill allow gases to be stored and transported within these tunnel sections, reducing the risk of compromising the host rock's integrity.

To ensure long-term reliability, the backfill must maintain these properties over the entire assessment period. Therefore, further research and optimization are required to improve the material's gas-handling capabilities and ensure its suitability as a backfill in the repository. The goal of the study is to define a mixture of sand, bentonite and crushed Opalinus Clay that meet the expectations listed above.

## 3.8 United Kingdom

### 3.8.1 Use of bentonite

UK policy incorporates a consent-based approach, working in partnership with communities, to determine whether hosting a Geological Disposal Facility (GDF) is right for them. A GDF will only be built where both a willing community and a suitable site can be demonstrated (Department for Business, Energy and Industrial Strategy, 2018), (Welsh Government, 2019). At the time of writing (January 2025) three Community Partnerships have been established (Mid Copeland, South Copeland and Theddlethorpe). All three communities are exploring the potential for inshore siting of a GDF. This will likely entail onshore surface facilities connected to the underground facility within the inshore region (within 12 nautical miles of the coast) via a series of accessways (Figure 10).

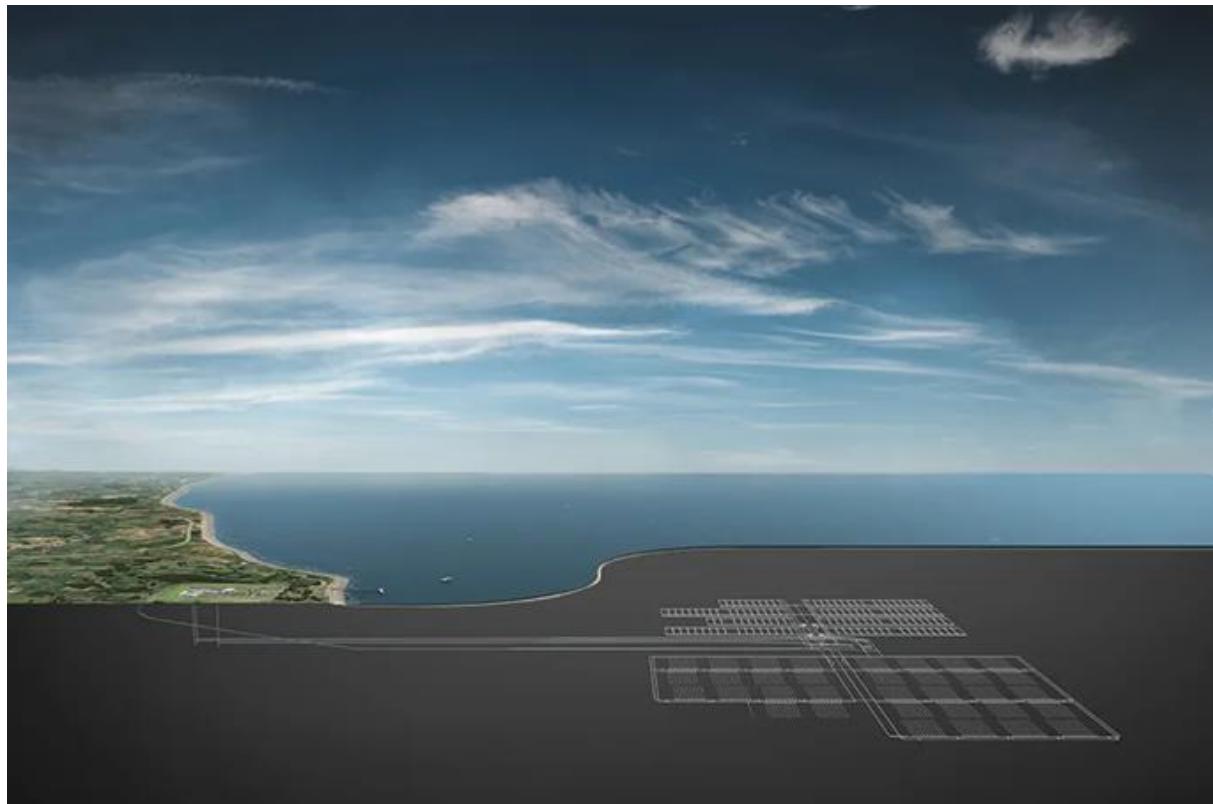


Figure 10 An illustration of what a UK GDF could look like in an in-shore region.

Nuclear Waste Services (NWS) is evaluating GDF construction, operation and long-term behaviour in potential host rocks present in Community Partnership areas, focussing initially on disposal concepts developed by other waste management organisations and their possible adaptation to UK-specific conditions. These disposal concepts are illustrated captured in the NWS' 2016 generic Disposal System Safety Case (Radioactive Waste Management, 2017). In some of these concepts, bentonite-based materials are used as a buffer around High Heat Generating Waste (i.e., High Level Waste and Spent Nuclear Fuel), as a hydraulic seal material placed strategically throughout the GDF, as gas permeable seals following closure of the Low Heat Generating Waste vaults (i.e., Intermediate Level Waste), and as mass backfill. Bentonite is proposed as a potential material option because of its low hydraulic conductivity, self-sealing capability, ability to minimise microbial activity and for its potential durability in the environmental conditions expected within a GDF and host rock (Sellin and Leupin 2013).

### 3.8.2 Current research needs

Over the next five years, the focus of NWS' bentonite research programme is to undertake targeted research to evaluate the suitability of this material to potentially form part of the engineered barrier system of a UK GDF. This will assist NWS' recommendations to the UK government for a campaign of deep borehole drilling, guiding further characterisation of the site(s).

The potential host rock for a GDF in Mid and South Copeland is the Mercia Mudstone Group, a mixed mudstone-evaporite sedimentary sequence. At Theddlethorpe, the potential host rock is the Ancholme Group, a clay-based sedimentary sequence. These rock types are classified as lower strength sedimentary rock in the UK generic Disposal System Safety Case (Radioactive Waste Management, 2017). Each of these rock types has an associated geochemical environment. Of particular relevance to bentonite is the salinity of groundwater present at each site, estimated to range from approximately 5-35 g/L in the Ancholme Group to approximately 190-320 g/L in the Mercia Mudstone Group, predominantly as sodium chloride (Smedley et al 2023). Data on bentonite performance at salinities estimated for the Mercia Mudstone Group is limited. Research is needed to understand the impact that

salinity has on the hydraulic-mechanical properties of bentonite, as part of a systematic programme to evaluate the suitability of this material for use in engineered barrier components in a highly saline environment.

To demonstrate bentonite feasibility under anticipated site conditions, NWS is undertaking a programme of experiments to build an understanding of swelling pressure and hydraulic conductivity for both a sodium and a calcium bentonite across a range of dry densities and sodium chloride concentrations. These data are being collected in shorter-term, smaller-scale experiments carried out in constant volume cells. Experiments are also underway to understand the evolution of suction and swelling pressure as bentonite re-saturates with porewaters of varying salinity.

Efforts are ongoing to couple the chemical component with the hydraulic-mechanical components within the Imperial College Finite Element Programme (ICFEP) to demonstrate process understanding and facilitate future upscaling in space and time.

#### 4. Bentonite materials

As stated in the SRA, a wide range of bentonite/clay-based materials are available for potential use in deep geological repositories. Commercial bentonites are normally divided into Na- and Ca-bentonite. Na-Bentonite is dominated by sodium ions ( $\text{Na}^+$ ) on the clay's exchange sites. Ca-Bentonite is dominated by calcium ions ( $\text{Ca}^{2+}$ ) on the exchange sites. Calcium bentonite is soda-activated to enhance its swelling and absorption properties, making it more similar to sodium bentonite. The process involves treating calcium bentonite with sodium carbonate (soda ash,  $\text{Na}_2\text{CO}_3$ ). Soda activation is widely used in industries where sodium bentonite's characteristics are needed, but natural sodium bentonite is unavailable or too costly. The primary difference between calcium (Ca-) and sodium (Na-) bentonite lies in their chemical composition and resulting physical properties, which determine their distinct applications. Sodium bentonite is favoured for applications requiring swelling and water retention, whereas calcium bentonite is preferred for adsorption and purification tasks. The choice depends on the specific physical and chemical requirements of the application (e.g. Odom, 1984).

The application of bentonite in a nuclear waste repository is somewhat different from most other bentonite applications. Bentonite is generally used as a low-density material, e.g. drilling mud, or as an additive, e.g. in foundry sand or iron ore pellets. A barrier in a nuclear waste repository will, in most cases, have a high dry density. At high density there is less difference in properties (e.g. swelling pressure) between Ca- and Na-bentonite than at low density. There is no oblivious advantage with either type and both may very well be used. Other considerations than the counter-ion may be more important. This is also seen in the national program where the focus was on Na-bentonite from Wyoming at an early stage, but later more attention has been given to Ca-bentonites and materials from a number of different locations have been studied, for example Milos, Černý vrch and Cabo de Gata. A soda-activated bentonite is of less interest for nuclear waste applications, since the activation offers no real advantages and the material will contain additional calcium carbonates. It may also be difficult to argue that an activated bentonite can be considered to be a "natural material", especially in the respect of long-term stability.

The favourable properties of a bentonite barrier, self-sealing ability (swelling pressure), and low hydraulic conductivity, are dependent on the montmorillonite content in the bentonite. A higher montmorillonite content will give more favourable properties. However, for the range of montmorillonite contents found in commercial bentonites this may not be important since a lower montmorillonite content can easily be compensated for by a higher installed density for some requirements. There are also other factors, apart from montmorillonite content, density and counter-ion, in the bentonite that may affect the hydromechanical properties. These are not fully understood. The properties expected of bentonite-based materials in the context of underground disposal can also be adjusted using additives such as sand.

## 5. Examples of earlier projects related to bentonite

The THMC(G) laboratory investigation is based on previous knowledge gained. Past projects FEBEX, (Enresa, 2000; Lloret et al. 2003, 2004; Gens et al. 2009b; Rodriguez-Dono et al. 2018; Villar et al. 2020; Kober et al. 2021), Prototype Repository (Mata et al. 2005; Villar, 2005; Chen and Ledesma, 2009; Svermar et al. 2016; Goudarzi, 2023), PEBS (Gaus et al. 2014; García-Siñeriz, et al. 2015; Schäfers et al. 2014), BEACON (Sun et al. 2019; Harrington et al. 2020; Sellin et al. 2020; Bosch et al. 2021; Narkuniene et al. 2021; Talandier et al. 2022; Leupin et al. 2022) EURAD WP HITEC (Villar et al. 2020; Vasconcelos et al. 2024; Graham et al. 2024), EURAD WP GAS (González-Blanco et al. 2022; Li et al. 2023; Gowrishankar et al. 2023; Owusu et al. 2023; Zheng et al. 2023) and FORGE (Alcoverro et al. 2014; Yamamoto et al. 2015; Bennet et al. 2015; Gutierrez-Rodrigo et al. 2021) among others, provided important information on bentonite and clay behaviour in EBS and investigation techniques. Other projects related to clay barriers are still ongoing: SANDWICH project (Wieczorek et al. 2024), where the conditions of a sealing system design are performed. HotBent is also ongoing as full-scale test at high temperatures (Finsterle et al. 2023; Kober et al. 2023). WP ANCHORS will further develop the topic by evaluating long-term experiments and investigating systems with aggregates and chemical loading. A variety of materials will be investigated using previously developed and new experimental methods. Multiscale approach is planned to study upscaling. Strong link to microscale investigation is envisaged.

### 5.1 Febex

The FEBEX project (Enresa, 2000; Kober et al. 2021) had multiple objectives: demonstrating the feasibility of manufacturing, handling and constructing the engineered barriers and of developing codes for the thermo-hydro-mechanical and thermo-hydro-geochemical performance assessment of a deep geological repository for high level radioactive wastes. The project integrated theoretical and experimental development work. The experimental work had three parts: an “in situ” test performed in Grimsel HRL (Alps, Switzerland), a “mock-up” test still ongoing in Madrid (Spain), in these two tests the maximum temperature at the heaters was 100°C, and a series of laboratory tests at small scale. The experiment was based on the Spanish reference concept for crystalline rock (granite and lamprophyre in Grimsel), in which the waste capsules are placed horizontally in drifts surrounded by high density compacted bentonite blocks. The bentonite used in this project was Calcium bentonite, named FEBEX bentonite, whose properties were studied in large laboratory tests campaigns, where swelling pressure, hydraulic conductivity and retention properties were measured for carrying out the parametrization of the material. Chemical properties were also measured because THC modelling was performed in that project. It was shown the dependency on the temperature of the retention capacity of bentonites, where the larger temperature, the smaller capacity (Villar and Lloret, 2004). This influence of the temperature was also measured by Tang and Cui (2005) in MX-80 bentonite. It was also shown the retention capacity of bentonites dependency on the dry density, in this case, the larger density the larger retention capacity (Villar, 2007) and it has also been observed in MX-80 (Seiphoori et al. 2014) and added in constitutive models (Della Vecchia et al. 2014). Gas permeability was measured in FEBEX bentonite and compared with the hydraulic conductivity and a large difference was observed when the intrinsic permeability was calculated (Villar and Lloret, 2001). Although the gas transport is not a main issue in ANCHORS, this work points out the difference between the structure and fabric of the bentonites in unsaturated conditions, with large voids (macrostructure) for air flowing and when it is saturated and the size of pores is smaller due to the growth of the microstructure that fills the macrostructure although the total porosity could not change. The double structure of the FEBEX bentonite and the different implications that it has in its behaviour like the differences during the hydration process when the intra- (microstructure) and inter- (macrostructure) aggregates govern the suction changes. During the hydration process, three different zones were observed: Zone 1 (swelling), Zone 2 (collapse) and Zone 3 (swelling) related to the yield surfaces described in the Barcelona Expansive Model (Gens and Alonso, 1992). These zones were also observed in MX-80 (Yigzaw et al. 2019). Due to the presence of very large voids between pellets, a triple structure porosity has also been considered (Navarro et al. 2020).

No significant mineralogical alteration was observed after dismantling the test (Kober et al. 2021). It is an important point because it proofs something expected, that the temperature of the test was not enough for starting mineralogical alterations. Although the salinity was not considered a main issue in FEBEX due to the granitic water in Grimsel has low content of solutes, the influence of the salinity in swelling pressure and hydraulic conductivity was measured in FEBEX bentonite and as it happens in other bentonites, the increase of the salinity reduces the swelling pressure and increases the hydraulic conductivity (Castellanos et al. 2008).

FEBEX project allowed to study the interaction between bentonite and concrete due to the plug was constructed with this material. The analysis performed did not allow to know the influence of the temperature in the concrete-bentonite interaction (Kober et al. 2021; Alonso et al. 2017a, 2017b). From the results obtained, it could be concluded that no safety relevant properties of the bentonite were affected and the concrete-bentonite interaction had no safety relevant impact although some mineralogical changes in bentonites were observed such as limited smectite dissolution and cation change at the exchange sites (Kober et al. 2021). Tests in FEBEX bentonite related to alkalinity has been performed (Sánchez, 2006; Fernández et al. 2008; Kaufhold et al. 2019; Gaboreau et al. 2020; González-Santamaría et al. 2021).

In the "in situ" and mock-up tests, the thermal effects of the spent nuclear fuel were simulated by means of heaters; hydration was natural in the "in situ" test and controlled in the "mock-up" test. The constitutive models already developed when the project started were improved and implemented in computer codes. The project was divided in two parts, FEBEX I (the most important) where the tests were started, switching on the heaters in 1997, and FEBEX II, where one of the two heaters in the "in situ" test was shut down and removed (2002). The second heater was shut down in 2014 and the test was finally dismantled in 2015, followed by an extensive laboratory program for carrying out the post-mortem analysis.

After the dismantling, some conclusions concerning the behaviour of a bentonite barrier in crystalline host rock can be performed (Villar et al. 2020): all the gaps between blocks were sealed and the opening carved in the blocks were also filled. The granite-bentonite contact was tight at all locations. Water content increased from outside to inside and the dry density increased from inside to outside. The saturation process was concentric and the differences between sections were due to the presence of the heaters.

## 5.2 Prototype Repository

The Prototype Repository project was also performed in crystalline rock (Äspö Hard Rock Laboratory (HRL), Sweden) where the rock was gneiss and granite (Baltic Shield). This test was full-scale where the production of blocks and pellets (made of MX-80) and the backfilling of the deposition tunnel with a mixture of crushed rock (made of tunnel boring machine (TBM) muck) and bentonite (Milos soda activated bentonite, 30% of weight) compacted "in situ" (Svemar et al. 2016) were tested as part of the design work of clay barriers. The canisters were emplaced in vertical position, following the Swedish and Finnish design (KBS-3V, Posiva 2021; SKB 2022). Six heaters were emplaced in six deposition holes separated in two sections with four and two heaters respectively. The section with four heaters was installed in 2001 and dismantled in 2023-2024 (Goudarzi, 2023), the section with two heaters was installed in 2003 and dismantled in 2010-2011. Mock-up and laboratory tests were performed in parallel with modelling work. It should be taken into account that the dismantling process finished late 2024-beginning 2025 and the reports with the analysis of the samples are still not available, so the information concerning the post-mortem analysis of this tests comes basically from the dismantling of the section with two heaters (Olsson et al. 2013; Svemar et al. 2016).

The mixture of bentonite and crushed rock was investigated and although from laboratory tests it was concluded that it was suitable for backfilling (Mata and Ledesma, 2003), the compaction process "in situ" showed heterogeneity and low densities close to the rock (Svemar et al. 2016).

Due to the groundwater in Äspö HRL contents solutes dissolved (12 g/L although with certain variability), large work related to salinity and its influence on the hydraulic conductivity and swelling pressure have been performed. These tests have confirmed the reduction of the swelling pressure and the increase of the hydraulic conductivity (Karnland et al. 2006). Tests under constant axial stress changing the water composition have also been performed with other bentonites like the GMZ (Chen et al. 2017). The presence of salts in the clay and how this water interacts with the groundwater during the saturation process has required larger efforts when the groundwater has salts dissolved and changes in groundwater salinity are expected once the bentonite is saturated (e.g. intrusion of fresh water after a glaciation, Jonsson et al. 2018). The study of the water and ions dissolved has given rise to develop different models and studies that consider the salt presence (Tournassant and Appelo, 2011). The definition of the chemical potentials (Karnland et al. 2003a; Hedström et al. 2011; Karnland et al. 2011; Navarro et al. 2015; 2017) and the extension of this concept to effective stress (Navarro et al. 2018; Tuttolomondo et al. 2021; Navarro and Asensio, 2023). In the case of the Prototype laboratory test, no large variation in swelling pressure were detected although the hydraulic conductivity was smaller in field-exposed samples than in reference samples (Olsson et al. 2013).

The effect of shearing is also an important issue in spent nuclear fuel constructed in places where it is possible to have glacial periods in future due to the activation of fractures due to the post-glacial rebound (Ojala et al. 2019), like it is the case of the rock where this test was performed. Tests in unsaturated conditions have been carried out (Pintado et al. 2019, 2023) where the suction effect increases the shear strength and shear stiffness. Saturated triaxial tests have also been performed at different salinities, where it has been proofed that the calcium bentonites have larger shear strength than sodium bentonites (Börgesson et al. 1995; Dueck and Nilson, 2010; Dueck et al. 2010). A combined chemo-mechanical work has been performed saturating samples of sodium bentonite in waters with  $\text{CaCl}_2$ , forcing the cation exchange and checking the increase of the shear strength (Kim et al. 2024). Other shear tests have been performed with similar results, checking the reversibility of the paths fresh-saline-fresh water (Di Maio, 1996). The shear rate has also been checked (Börgesson 1986; Dueck et al. 2010), proofing that the larger shear rate, the larger shear strength probably due to viscosity effects. The triaxial tests performed in samples coming from the dismantling of the first section of the Prototype Repository test indicated a larger deviator stress at failure than the reference samples and with the uniaxial confined tests, larger brittle (Olsson et al. 2013).

Due to the presence of cement in grouts that seal fractures, the behaviour of the bentonite when it is in contact with alkaline waters had to be checked (Karnland et al. 2003b). The bentonite alteration was measured in concrete plug – backfill contact. It was not identified any alteration in montmorillonite (Olsson et al., 2013; Svermar et al. 2016) but small changes in bentonite were detected, like exchangeable Mg and Ca had been replaced primarily by Na and the CEC of <0.5 mm fraction had increased.

The combination of studies of swelling pressure development, salinity effect and alkalinity has shown that these hydro-chemo-mechanical processes ongoing in clays proceed over different time scales. For instance, in samples with oedometric dimensions (20-70 mm diameter and 5-25 mm height), the hydration and its associated swelling pressure development might take some weeks, while cation exchange processes might take longer. Mineral dissolution in bentonite due to alkalinity proceeds over different time scales as well. While the dissolution of readily soluble accessory minerals (gypsum, halite, few carbonates) occurs over a comparable time scale as cation exchange processes, dissolution of other accessory minerals such as alumino-silicates typically take some months and even years depending on the type of mineral (dissolution of accessory silica such as cristobalite has been observed in experiments). Montmorillonite dissolution is an even slower process and short-term experiments hardly detect this dissolution.

An important result in Prototype Repository test is that contrary to FEBEX “in situ” test, the saturation process was not concentric, so the rock heterogeneity played a role in the saturation process in this test.

Another important result is that no piping or erosion was observed neither in buffer nor in backfill (Svemar et al. 2016).

### 5.3 PEBS

The 7th Framework EURATOM project PEBS (Schäfers et al 2014) was initiated in 2010 to study the complex interaction of thermal, hydraulic, mechanical and chemical (THMC) processes in clay-based EBS for geological repositories. During the four-year project, investigations were performed by 17 partners from Europe, China and Japan, including waste management organisations, research institutes and consultants.

A broad range of laboratory and in-situ experiments dealing with the early evolution of the EBS were performed in the framework of PEBS. The main achievements of the experimental studies performed in included:

- Confirmation, integration and improvement of existing knowledge of the THM-C processes in the early evolution of bentonite barriers
- Compilation of data for future interpretation of large-scale experiments
- Providing a reliable data base for the validation and enhancement of numerical models
- Providing more distinct criteria to be conservatively applied in the Performance Assessments of engineered barriers

Through the comprehensive numerical simulations performed within the following progress and achievements were made:

- Simulation of the laboratory and in-situ tests helped to interpret the experimental results
- Enhancements of existing models, including threshold gradient, thermo-osmosis and double porosity, allow for the simulation previously unexplained phenomena in the EBS evolution
- Development of new model concepts based on thermo-mechanical continuum mixture theory, allowing for an alternative description of the EBS behaviour
- Enhancements of THMC models for the simulation of corrosion and effects at the bentonite-concrete interface, considering porosity change by swelling and incorporating reactive gaseous species
- Quantification and reduction of uncertainties in the model predictions by implementing an inverse framework
- Improvement of the basis for long-term extrapolation of the EBS behaviour and prediction of long-term repository evolution.

Significant progress was made in the evaluation of uncertainties in process understanding of the EBS. The work was based on and integrated the outcomes of the other experimental and modelling work within PEBS as well as findings from outside the project. This permitted the integration of a broad range of knowledge and put the gained information into the context of performance assessment.

The China-Mock-Up experiment, is an important milestone of the buffer material study for HLW disposal in China. The observed THMC processes taking place in the compacted bentonite buffer improve the knowledge of the early phase evolution of HLW repositories. Furthermore, they provide a reliable database for the validation of the numerical models, which in turn are used for the extrapolation to the long-term behaviour of GMZ bentonite under relevant repository conditions.

### 5.4 BEACON

The HORIZON 2020 project BEACON (Bentonite mechanical evolution) (Sellin 2020, Claret et al 2022) aimed at developing, testing and validation of numerical models against experimental results to predict the evolution of the hydromechanical properties of bentonite during the saturation process.

The scientific part of Beacon was divided into five work packages. The first dealt with the application of the results from the project in safety case and design. The purpose of the second was to collate and share knowledge on the available information about the bentonite mechanical evolution. The third

handled development of models. The objective of the fourth was to provide experimental data to support the model development and testing. The core activities of Beacon were, however, performed in the fifth work package where numerical models describing the mechanical evolution of bentonite barriers were tested, verified, validated and finally applied in relevant assessment cases. The originality of work performed in this work package was to propose test cases in which heterogeneities in the bentonite-based component were present initially or to revisit large scale experiments in the perspective to follow heterogeneities evolution and the capacity of the model to predict the final state.

At the onset of the Beacon project, there were very few examples of the application of mechanical models of bentonite in a safety case. Many teams had the mechanical formulations included in their THM-codes, but the level of testing and verification of those formulations was, in general, rather limited.

To overcome this issue, modelling activities were carried out either in a test case that was built on experiments performed within Beacon and included a blind prediction of an experiment or on assessment cases proposed by waste management organizations (WMO) based on specific components taken from actual repository designs.

In detail, calibration/validation of the models was carried out using three sets of laboratory tests. These latter were chosen based on the initial heterogeneity of the materials or the introduction of perturbations during the test to induce some heterogeneities. These tests were complementary and represented relevant situations encountered in a repository when installing an EBS. The dispersion of the numerical results was rather large. This was especially true regarding the calculated stresses. For the final dry density in the test, the calculated values were in better agreement with the measured values. At this point, many teams were also rather inexperienced with this type of issue.

Regarding large scale experiments, to show the capacity of the models to reproduce in situ experiments, three experiments were selected:

- EB - Engineered Barrier Emplacement Experiment (EB experiment),
- FEBEX - Full-scale Engineered Barrier Experiment in Crystalline Host Rock,
- CRT - Canister Retrieval Test (CRT)

This modelling activity was much more difficult than the previous one due to complexity of the geometry, the uncertainties on the boundary and initial conditions and sometimes in the analysis of the information given by the sensors. Moreover, for two of the tests (CRT, FEBEX), it was necessary to consider the temperature and the couplings between the thermal part and the hydro-mechanical behaviour. Despite this, many teams managed to get a rather good estimate of the final state of the barrier in all three tests. One reason for this may be that the uncertainties in the initial conditions allow for more freedom in the setup of the models.

One of the main challenges in modelling swelling materials is the capacity of the models to perform predictive simulations. The presence of initial heterogeneities in these materials or heterogeneities due to external conditions increases the complexity of predicting the evolution of swelling clay materials. Tests performed within the Beacon project were simulated to evaluate the ability of the models to predict hydromechanical evolution of bentonite. Two tests were already finished at the beginning of project. All the data available on these tests were given to the partners. The purpose was to have a first calibration step. For these tests, the bottom part of the cell was filled with bentonite pellets with an average dry density close to 1.30 g/cm<sup>3</sup> and the top part with a bentonite block with a dry density of 1.60 g/cm<sup>3</sup> (Figure 11). Hydration with deionized water took place through the bottom. In the first case, a constant pressure was imposed and in the second a constant flow was imposed. One test was selected for predictive modelling. The results of this test were not given to the participants. The conditions of the test were similar to the first test except that the pellets layer was located in the upper part of the cell and block in the lower part. Predictive simulations of water intake, dry densities, gravimetric water content and stresses were expected on this test case. The results from the predictions of dry densities as well as the experimental results can be found in Figure 12.

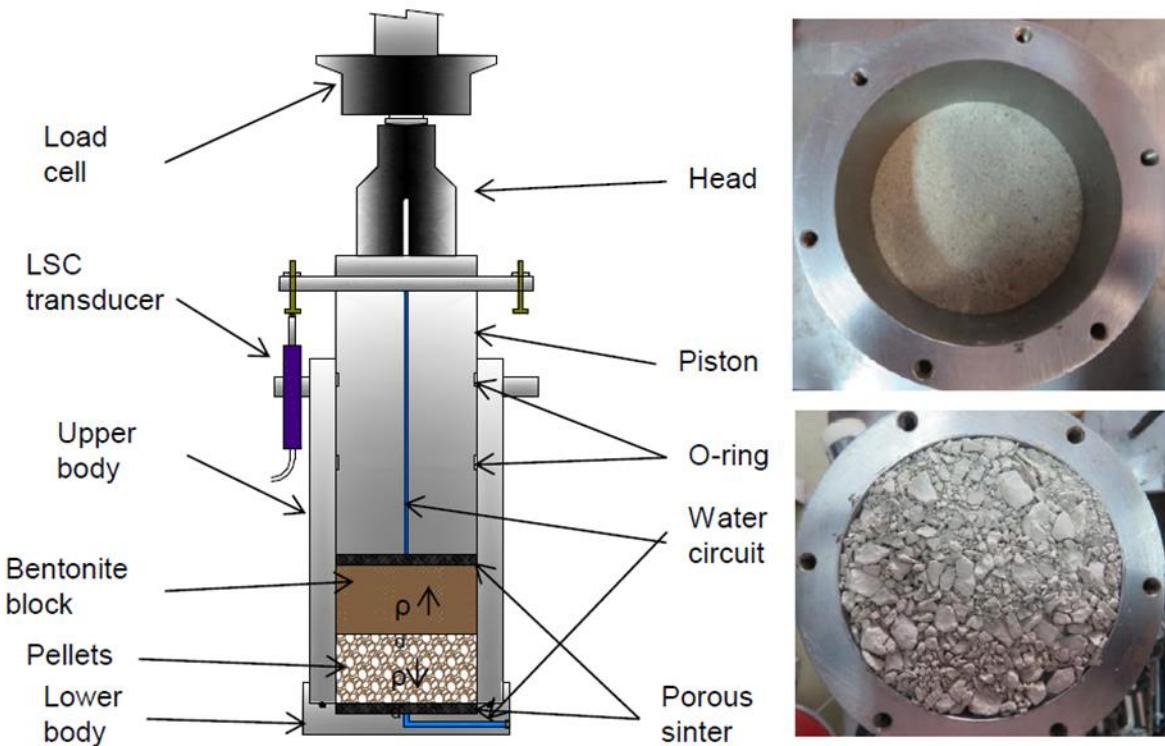


Figure 11 Schematic representation of the test cell and images of the block (upper right) and pellets (lower right). In the predictive test the block was placed in the bottom of the cell (Villar et al 2021).

The water intake with time was well modelled by most teams. However, one may observe a number of variations or divergences. Moreover, prediction is less easy for early times of hydration. These globally good results may be surprising considering the large range of permeability used in the simulations by the different teams. As seen in Figure 12, the final (at saturated state) dry densities are well reproduced by many of the models. They do not depend significantly on the mechanical models (including law and friction aspects). It was very difficult to predict the stress's final value and time evolution. Stress evolution showed much variation between teams. Comparing mechanical behaviour is not an easy task. Each team has a different conceptual model, and few parameters are comparable. A short synthesis of the mechanical constitutive models used by the different teams is presented in Table 1.

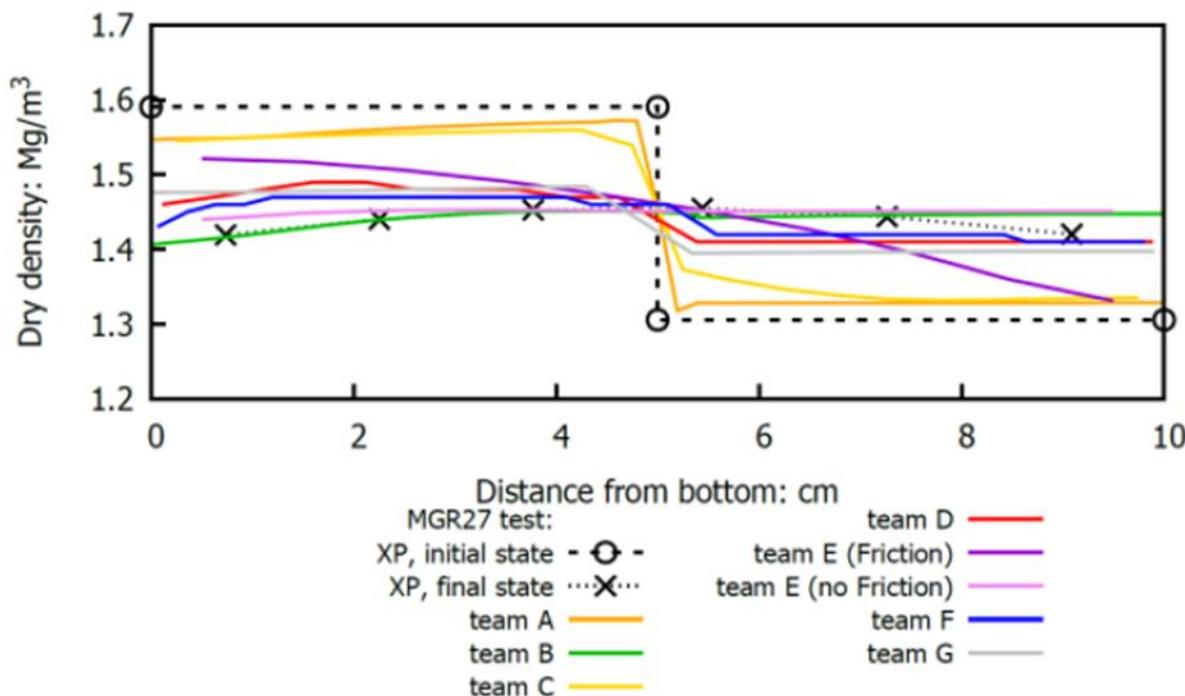


Figure 12 Dry densities at saturation for the experiment used for predictive modelling.

Table 1 Mechanical constitutive models used in the benchmark

Team	Model
A	Bishop effective stress; modified CamClay
B	Hysteresis Based Material (HBM) model
C	Double-structure hypoplastic model for expansive clays
D	ACMEG – TS elastoplastic model
E	Internal Limit Model (ILM)
F	Modified Barcelona Basic Model (BBM) elastoplastic model
G	Elastic with modulus depending on water saturation
H	Modified BBM with double structure ICDSM
I	Modified Barcelona Expansive Model BExM

Two teams modelled friction at the cell wall–bentonite interface. These teams managed to satisfactorily blind-predicted the evolution of axial stress in the test.

Direct application to real assessment cases in actual repository systems has also been tackled. A few cases from relevant repository systems were therefore selected as test examples. Three cases were proposed: (1) a tunnel plug based on the Andra design, (2) a disposal cell from the Nagra concept, and (3) the KBS-3 deposition tunnel backfill (SKB, POSIVA). These are representative of the primary areas of uncertainty in density homogeneity. Here, the teams divided the cases, and only 3–4 teams modelled a particular case. The results from the modelling showed a rather strong divergence in results for the final dry density distribution for all three cases. For the Andra and SKB cases, the calculated values were still within the range acceptable for repository performance, but that was less true for the Nagra case. This shows that there are challenges to moving from modelling laboratory and field experiments, where results are available, to simulations of repository performance.

## 5.5 GAS

The main objectives of the EURAD GAS WP (Lavasseur et al., 2024, Marschall et al., 2024) were to improve the mechanistic understanding of gas transport processes in engineered and geological clayey materials and to assess the gas transport mechanisms that may be active at the scale of a geological disposal system and their potential impact on barrier integrity and repository performance.

Task 3 of EURAD-GAS WP was dedicated to the hydro-mechanical phenomena and processes, associated with the gas-induced failure of clay barriers and to the effectiveness of self-sealing processes along gas-induced pathways in the clay barriers of a geological repository. The evaluation of achievements has been accomplished by model-supported data analyses, predictive modelling and the application of newly developed modelling tools on in-situ experiments.

Experimental activities on combined gas / water transport in engineered barrier materials (Czech Ca-Mg bentonite, Na-smectite based Wyoming bentonite) and gas invasion and self-sealing experiments on samples of clayey host rocks (Boom Clay, Callovo-Oxfordian, Opalinus Clay) were performed.

Several tests performed on bentonite showed no significant change in hydraulic conductivity and swelling pressure after one year of cyclic loading with high build-up rates. The integrity of the EBS appears to be preserved despite extreme stresses. An interesting detail of the results with the heterogeneous test samples was the formation of distinct preferential gas pathways even after long resaturation times. This may be important for heterogeneous materials investigated in ANCHORS.

Experimental setup developed for fractured clay investigation showed that the gas transport through the fractured clay does not limit the self-sealing ability of the material. This ability was observed on large fractures that could be associated with the initial EDZ (Zhang and Talandier, 2023; Agboli et al., 2024).

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

## 5.6 HITEC

The EURAD-HITEC WP (Villar et al, 2024) of the EURAD programme addressed the effect of high temperatures on the behaviour of bentonite and clay. It focused on THM understanding of clay-based materials (host rock and buffer) exposed to elevated temperatures (>100°C) for prolonged periods.

For bentonite buffer, proving that higher temperatures than presently accepted are suitable is very relevant even for current concepts. It increases safety margin and gives greater credibility to the design (e.g., if it is proven to work for 130°C then for 100°C it is likely to be safe). Also, this type of optimisation could be used to increase thermal limits on the bentonite buffer, reducing the footprint of the facility. So potentially significant cost savings and improved environmental sustainability. No new processes were identified for bentonite, but swelling, swelling stress formation and water conductivity were studied in detail over 100° degrees C temperatures. Relationships include: (i) the observation of swelling pressure and permeability as a function of temperature for various dry densities, swelling strains, chemical states and conditions and (ii) water retention curves, as function of temperature. For the materials and conditions tested, an influence of elevated temperature on water retention capacity has been observed. Multiple test programmes, in both Ca- and Na-bentonite have also found evidence that, whilst changes to hydraulic permeability are not very significant, swelling pressure can be substantially impacted by elevated temperatures under certain conditions. Further work to investigate and consider the mechanisms and consequences of this behaviour for repository design are recommended as a result.

## 5.7 SANDWICH

The SANDWICH project is currently ongoing presents an alternative of sealing systems. The sealing system consists of alternating sealing segments of bentonite and equipotential segments that are characterized by a high hydraulic conductivity. Within the equipotential segments fluid is evenly distributed over the cross section of the seal. Water bypassing the seal via the excavation damaged zone, or penetrating the seal inhomogeneously, is contained and a more homogeneous hydration and swelling of the sealing segment is obtained.

The functionality of the system has already been proven in semi-technical scale experiments. It was the aim of this project to install a large-scale in-situ experiment that addresses the interaction between the sealing system and the host rock. The experiment at the Mont Terri rock laboratory consists of two experimental shafts in which Sandwich sealing systems have been installed. The sealing systems can be saturated from pressure chambers located at the shaft bottoms via inclined lateral feeding boreholes. The seals and the surrounding rock are intensely monitored (Wieczorek et al. 2024). More information about this project can be found in Emmerich et al. (2019).

## 5.8 HotBENT

HotBENT is a project currently ongoing in Grimsel HRL laboratory. The project will provide information and data for repository optimisation with respect to design, space, emplacement strategies and costs. The experiment aims to evaluate current accepted safety functions by investigating the effects of high temperatures on bentonite-based barriers and their safety functions. More information concerning this test can be found in Finsterle et al. (2023) and Kober et al. (2023).

The performance of bentonite barriers in the < 100°C temperature range is underpinned by a broad knowledge base built on laboratory and large-scale in-situ experiments. Bentonite parameter characterization above 100°C is sparser (especially for pelletized materials), although up to about 150°C no significant changes in safety-relevant properties are indicated. Information on higher temperatures on buffers is desirable for repository optimization with respect to design, space and costs (e.g., footprint, layout) and to enable more options with respect to the required interim storage time periods. HotBENT corresponds to the initial highest thermal-output period of geological disposal, the data can be used to evaluate strategies for the performance confirmation period.

HotBent has an “in situ” test ongoing at the same gallery where the FEBEX was carried out in past, so the host rock is well characterized (granite with a dike of lamprophyre, crystalline rock), and it will help to perform the test simulations. In parallel, laboratory tests are ongoing with the strong handicap of the temperature, which is larger than 100°C in parts of the clay barriers. The test is running in two sections with two heaters each one, where two different types of bentonites were emplaced (Wyoming in the first section and Wyoming and Czech at the second). The design follows the Swiss design.

## 6. Laboratory testing

Laboratory testing has different purposes:

- validation of the constitutive models that are used for carrying out the simulations of deep geological disposals. In particular, the simulation of clay barriers;
- provide information about the behaviour of the clay components and the changes in chemical, mineral and structure due to the processes expected during the warranty time of the repositories;
- provide parameters of the constitutive models. These parameters are necessary for obtaining accurate predictions of the clay components behaviour;
- better understanding of the complex behaviour of bentonites.

There is a large number of issues that should be investigated in the design and construction of deep geological disposals for storing spent nuclear fuel. This section describes some of them related to the laboratory tests that are going to be performed in ANCHORS.

## 6.1 Phenomena investigated

### 6.1.1 Heterogenous materials

Several heterogeneous materials are currently proposed for use in EBS systems. They can offer several advantages: fine-tuned properties, lower cost, easy handling, compatibility with the environment.

Depending on the DGR concept, several heterogeneous materials are envisaged:

- Bentonite as compacted blocks or pellets (buffer and backfill, seals)
- Granulated bentonite (crushed pellets) (backfill, buffer)
- Mixtures of bentonite, claystones, crystalline rocks (backfill, main tunnels and shafts)
- Bentonite-pellet and bentonite-sand mixtures (backfill, shafts, plugs, seals)

Bentonite as compacted blocks and pellets

Compacted blocks and pellets are the main components in the KBS-3 disposal design developed in Sweden that it is going to be used in this country and in Finland (Posiva, 2021; SKB, 2022). Compacted blocks fill the deposition holes, where the canisters will be emplaced, and the deposition tunnels, the access tunnels to the deposition holes. Pellets fill the gaps due to the contact between blocks and rock cannot be assured and certain tolerance is needed in the emplacement of the canister in the block rings' void. Pellets can be replaced by granulated bentonite (crushed pellets). Compacted blocks are also used as cradle for emplacing on top the canisters in Swiss design (Nagra, 2021).

There is heterogeneity at the emplacement, which will not disappear due to the large difference in densities between the two components in Finish and Swedish disposal design (Olsson et al. 2013; Kristensson and Börgesson, 2015) and in Swiss design (García-Siñeriz et al. 2015) but the final state should be known. FEBEX project only had compacted blocks and gaps between blocks and between blocks and walls and certain heterogeneity still remained when the test was dismantled (Villar et al. 2020; Kober et al. 2021). Laboratory tests contribute to check the swelling process of both components and the final state once they are saturated or the gap sealing capacity (Harrington et al. 2020, Sellin et al. 2020; Villar et al. 2021; Dueck et al. 2022a). Results showed that the fully homogenization was not reached.

Granulated bentonite

Granulated bentonite material is envisaged for using in several repository designs: Swiss design (Nagra, 2021) and possible change in backfill design in Finland, where the blocks and pellets (Keto et al. 2013) will be changed by granulated bentonite. Handling, ease of placement, uniform overall mass distribution and the absence of large joints (as between compacted blocks) are among the main advantages.

Granulated material is inhomogeneous on a small scale (different sizes of granules, voids between granules). Once the material has access to water, the process of homogenisation begins as the voids are filled by swelling granules. The process of homogenisation needs to be studied, general material properties determined, and surrogate homogeneous material models developed.

Mixtures of bentonite and claystones or crystalline rock

French and Swiss concepts for backfill materials consider the reuse of excavated claystone, such as Callovo-Oxfordian claystone (COx) and Opalinus clay (OPA). The main advantages of this approach are: (i) a reduction of the environmental impact of the repository, (ii) minimisation of costs, and (iii) better physico-chemical compatibility with the host rock.

The barrier materials must fulfil essential performances and safety requirements defined by the two WMOs in their fully hydrated state such as (i) a secured exit of gas from the repository, (ii) adequate mechanical properties to sustain the mechanical loads in the long-term, and (iii) limited water permeability (only for sealing).

In more detail, this entails defining several levels of requirements for the following parameters: hydraulic conductivity, gas entry pressure, gas permeability after the breakthrough and under nearly saturated conditions, swelling pressure, and free swelling capacity. The criteria will be defined as a range of values to meet the needs of both agencies (Nagra and Andra) and allow more design flexibility.

Crushed rock (crystalline rock) might be used for filling the main tunnels in Finland and Sweden. These tunnels are far away from the canister containing the spent nuclear fuel and the requirements will be less strict.

#### Bentonite-pellet and bentonite-sand mixtures

Bentonite-pellet and bentonite-sand mixtures have recently been considered by several European countries for using as part of engineered barrier systems. Depending on the components proportion, the swelling pressure, water and gas permeabilities of the mixture can either decrease or increase.

Research on these mixtures focuses on optimising the bentonite content as well as sand and pellet sizes to achieve relevant properties. It also aims to understand the contribution of pellets and sand fraction to the mechanical and hydromechanical behaviour of the mixture. This is essential for the validation of models used to assess the behaviour of these heterogenous mixtures. A key aspect of the study is to determine the range of applications for these materials based on their thermal-hydraulic-mechanical and chemical (THMC) properties.

##### 6.1.2 Effect of Salinity & Alkalinity (pH)

Once emplaced at the repository, the initial unsaturated bentonite-based materials will be re-saturated gradually by infiltration of groundwater, which could introduce saline ions dissolved. Some deep geological repositories will be emplaced in rock formations with an expected transient phase with saline groundwater, such as those in Finland and Sweden, or with certain presence of salts dissolved, like in Switzerland (Pearson, 2002) or in France (Agnel, 2021). However, these conditions may change over time due to e.g., the potential intrusion of freshwater from glacial melt, anticipated in the coming millennia (Thölix et al. 2016; Jonsson et al. 2018, Ojala et al. 2019, SKB, 2020). Deep geological disposals will have concrete for lining reinforcement or for construction of certain units like plugs (Saanio et al. 2013) and in crystalline rocks, the fractures might be sealed with grout (Karnland et al. 2003b; Keto et al. 2013). Degradation of cements will increase the alkalinity of the surrounding groundwater (Jenni et al. 2019). Consequently, the development of swelling pressure upon hydration of bentonite will be associated not only with changes in the microstructure of the bentonite-based material due to swelling but also will be changed, e.g., through dissolution and precipitation of accessory minerals or cation exchange processes. In this regard, the effect of chemistry on swelling pressure and the long-term evolution of microstructure of bentonite-sand mixture remains unclear despite recent advances and requires further investigation.

The effect of salinity on the swelling capacity of expansive materials has been known for long. It is usually assessed by means of oedometer tests (swelling pressure, swelling capacity or consolidation tests) in which the concentration and predominant cation of the saturating solution are changed (Barbour and Yang, 1993; Karnland et al. 2006; Zhu et al. 2013; Dutta and Mishra, 2016; Jenni and Mäder, 2018; Kiviranta et al. 2018). These laboratory tests systematically show a decrease of the swelling capacity of the bentonite with the increase of salinity (Karnland et al. 2006; Kiviranta et al. 2018), which becomes less evident for high vertical loads and high bentonite densities. The extent of this decrease depends on the predominant cation in the solution and is more important for sodium than for calcium. These chemically-induced deformations are largely reversible (Di Maio, 1996). The explanations given to these observations usually resort to the effect of the solution salinity on the DDL thickness and to cation exchange processes (Di Maio, 1996). However, less attention has been paid to the latter, although the effect of the exchangeable cations on the swelling capacity is generally acknowledged.

These studies are considered to be of relevance for the assessment of the performance of the engineered barrier system in nuclear waste repositories, in which the salinity of the groundwater can

vary over time. However, it is unclear if the results of laboratory small-scale tests can be transferred to the real case, because of the boundary conditions differences, particularly the scale one.

The properties of expansive materials will also be influenced by alkaline plumes from concrete components (e.g., NaOH, KOH). During the operational phase of a repository, the pH of water in contact with the bentonite-based seal—initially neutral at around 7.5—may rise due to the release of hydroxides from cementitious porewater, which have an initial pH as high as 13.5. These highly alkaline environments are known to alter smectite in the long term (e.g., Tachibana et al. 2017; Leupin et al. 2014; Fernandez et al. 2014; Cuisinier et al. 2008; Sánchez et al. 2006). For bentonite-sand mixtures exposed to highly alkaline conditions, smectite alteration might be accompanied by the dissolution of sand grains. Both processes might induce an increase of hydraulic conductivity and compromise the sealing material's mechanical stability and swelling capacity (Laviña et al. 2024; Idiart et al. 2020a; 2020b).

#### 6.1.3 Vapour vs liquid saturation and hydration processes

The bentonite barrier is installed in an unsaturated state and undergoes hydration by the groundwater until becoming saturated. During the saturation process, the bentonite is in a non-uniform state in water content, dry density, and stress due to non-uniform swelling, in some cases enhanced by possible drying near hot waste canisters. The non-uniform dry density has been observed to remain even in the fully saturated state both in small- and large-scale tests. The changes in dry density and water content result in modification of the bentonite microstructure, such as changes in the pore size distribution, specific surface area, and interlayer thickness.

During the saturation process, part of the water enters the bentonite in liquid form and part as water vapour. The importance and the coupling of the advective and diffusive transport processes are not entirely clear, but it has been observed that the rate and mode of hydration significantly influences the homogenisation behaviour of bentonite. Notably, hydration from liquid water and vapour at 100% relative humidity often led to different final states, despite identical water activity in both cases (Yigzaw et al 2019). This still-unexplained phenomenon has parallels in other systems, such as lipid bilayers, gelatine, and swelling polymer membranes, where it is commonly referred to as the vapour pressure paradox or Schroeder's paradox (Vallieret et al. 2006).

In the scope of hydration and transport processes, direction-dependent behaviour has been observed in many critical parameters such as diffusion rate and swelling pressure. This anisotropy is attributed to the directional dependence of physical properties in bentonite and bentonite-sand mixtures, influenced by particle orientation, compaction direction, external stresses, sand fraction, and size. During hydration, anisotropy evolves as water is absorbed, leading to changes in microstructure, swelling behaviour, and mechanical properties, but the way in which anisotropy affects swelling and homogenization performance is still under debate.

Reliably predicting the degree of bentonite homogenisation during and after full saturation is crucial for safety assessments. Swelling and homogenisation of the buffer are key processes to ensure the buffer meets its functional requirements in a deposition hole. Understanding the final state of the buffer, following initial saturation and potential material loss (e.g., due to erosion), is essential for predicting long-term repository performance.

#### 6.1.4 Friction effect

Friction might play a certain role during the development of the swelling pressure in certain parts of the repositories like close to buffer – backfill contact (Kristensson, 2022; Pintado, 2022) and it has been considered in the simulation of “in situ” tests focused on buffer-backfill interaction (Leoni et al. 2018). It was considered as an important issue in modelling (Harrington et al. 2020; Narkuniene et al. 2021). Experimental work has been performed for studying the behaviour of the interfaces between the materials (Sinnathamby et al. 2014) in dry conditions and between the cells and the bentonite (Dueck et

al. 2018). Friction effect was considered in BEACON EU project (Bernachy-Barbe et al. 2022; Talandier et al. 2022).

Friction might also have influence in tests where the ratio surface/contour length is low or in tests where the swelling of bentonites will produce relatively large displacements between the sample and the cell. These situations occur often in micro-scale in situ tests, where the size of the sample assembly is a compromise between the requirements of the specific measurement technique and the phenomena being studied (e.g.,  $\mu$ CT or X-ray scattering techniques). There, the effect of friction must be carefully considered when interpreting the results or scaling the experiments up, and the results of such experiments should always be interpreted in conjunction with larger scale laboratory tests.

## 6.2 Micro scale testing

Bentonite exhibits structure in various scales. Montmorillonite, the main element of bentonite, consists of lamellar platelets packed together. Platelet spacing (interlayer spacing) is in the single-digit nanometer range. Platelets form montmorillonite particles whose size is in the 100 nm range, and they are typically accompanied by accessory mineral particles with wide size distribution up to tens of micrometers (Richards 2010). In some engineering cases relevant for the final deposition of radioactive waste, even more heterogeneous material systems are used, e.g., bentonite granulates or mixtures of bentonite and sand. There, inhomogeneity can be found also on larger length scales. Although traditional (soil-)mechanical testing is essential to produce information about the performance of the materials, it often does not show the intricate interplay of physical and chemical processes ongoing in the materials, but only their effects on the bulk behaviour. Studies in the small scales are necessary for understanding the physics and chemistry of bentonite-based materials and making judgements about the long-term safety of their use in waste deposition.

Bentonite-based materials can be characterized in comprehensive ways, including both destructive methods for detailed analysis, and non-destructive methods to observe the processes in situ during experiments. X-ray microtomographic ( $\mu$ CT) imaging has been performed on bentonite, bentonite-sand, and bentonite-claystone samples at various conditions to reveal  $\mu$ m-scale structure in 3D and its changes in several different treatments and processes. For example, the formation of cracks and changes in microporosity can be probed with this technique (Meng et al. 2023). In-situ oedometer cells allowing monitoring of both mechanical properties and macroporosity at the same time have been developed, and image analysis enables probing of the local water content and dry density of bentonite both in  $\mu$ CT and neutron imaging (McFarlane et al. 2021, Harjupatana et al. 2022). In-situ volume imaging studies are most feasible for slow processes, but luckily these are very common in bentonite studies (e.g. long-term homogenization and various transport processes (Zaidi et al. 2024)).

The limitation of volume imaging methods is the resolution that is typically constrained to  $\mu$ m-level. In the case of pore size distribution measurements, this can be overcome by Mercury intrusion porosimetry (MIP) techniques, whereby the dynamic range in pore size begins from single-digit nanometers and extends up to hundreds of  $\mu$ m. However, the technique assumes a well-connected pore network that might not be a good approximation of some materials in all scales (Houben et al. 2014).

X-ray diffraction (XRD) and scattering techniques (SAXS & WAXS) allow for the analysis of microstructural changes in different bentonites and bentonite-based mixtures from a different viewpoint. These techniques probe crystallinity and periodicity of the samples in different scales, and in the case of X-ray microdiffraction, even localized measurements are possible. Quantities available from these techniques include the evolution of interlayer water distribution, separation of the contributions of crystalline and osmotic swelling, and evolution of anisotropy during swelling (Chaaya et al. 2023). Similarly to  $\mu$ CT, miniaturized oedometer cells and other arrangements for studying processes in situ can be taken advantage of.

Information about the elemental and chemical composition of bentonite-based materials is available through X-ray fluorescence signals and Raman, IR, optical, and NMR spectra. All of these can be performed with spatial resolution if suitable equipment is available (e.g., SEM-EDX,  $\mu$ -Raman, or FTIR

microscope), and can therefore reveal localized chemical information of the various material phases. Although these techniques are superior to volume imaging and diffraction in both contrast and resolution, they are able to only image in 2D. The samples must be typically prepared into polished cross-sections. In the polishing process, focused ion beam (FIB) and broad ion beam (BIB) milling techniques are useful for high-quality cross-sections without disturbing the structures of interest (Desbois et al. 2009). In the interpretation of the results, care is required to account for the representativeness of the polished regions and especially stereological considerations arising from studying 3D structures through 2D sections. In these cases, in-situ measurements are usually not done due to the destructive sample preparation process.

In Anchors, in-depth microstructural characterization of different kind of bentonite types and bentonite-based mixtures will be performed (subtask 3.1) to improve understanding of physical and chemical mechanisms governing the material properties, improve possibilities for optimizing the EBS, to obtain more reliable data for constitutive modelling and to contribute to the bentonite THMCG database.

The following section outlines the contributions of the partners involved in Subtask 3.1. Each participating organisation provides a summary of its relevant previous studies, along with key references, as well as the objectives and planned activities to be carried out within the ANCHORS project.

### 6.2.1 BGR

#### Summary of earlier studies

Broad-Ion-Beam (BIB) milling combined with SEM has advanced the study of fine-grained materials by enabling the preparation of large, polished cross-sections with minimal artifacts. This is particularly important for defining the representative elementary area (REA), which is an area sufficiently large to be statistically representative of the composite and should include all microstructural heterogeneities present (Kanit et al. 2003).

Using box counting of mineral phases, Houben et al. (2013, 2014) determined a REA for the shaly facies of Opalinus Clay (OPA) to be only  $100 \times 100 \mu\text{m}^2$  in size. The authors argue that the OPAs' porosity has a strong spatial correlation to its mineralogy and that this REA is hence representative for both mineralogy and porosity.

Furthermore, the SEM-visible pore area follows a power-law distribution, aligning with bulk porosity measurements, and confirming a porosity of 15.3% in the shaly OPA facies (Houben et al. 2013, 2014). Mineral grains and pores are predominantly aligned parallel to bedding, further influencing permeability and mechanical behaviour (Houben et al. 2014).

Moreover, structural anisotropy plays a crucial role, as compaction-induced pore pressure response, dilation, and failure behaviour are controlled by the preferred orientation of mineral grains and pores, leading to different deformation mechanisms depending on loading direction (Winhausen et al. 2023). Of particular interest to our work is the previous finding by Philipp et al. (2017), who provided a microstructural explanation for the higher permeability of sandy OPA compared to shaly OPA, despite its porosity being only half that of shaly OPA. He proposed a force-chain of larger quartz grains, shielding areas from compaction and hence allow for preserved, interconnected fluid channels. We speculate to find similar force-chains in the here studied mixtures, where more rigid OPA aggregates might shield softer bentonite volumes (see also Zeng et al. 2023).

#### Objectives

The objectives of BGR in ANCHORS are to characterize the microstructure of crushed claystone-bentonite mixtures using reflected light microscopy to select areas for in-depth BIB-SEM analysis. If methodologically successful, this procedure allows insights in the microstructural variability, foremost in spatial distribution and morphology of pores. A subsequent key objective is to determine whether the produced samples exhibit a representative elementary area (REA) that is both homogeneous and

spatially invariant within the sample. This is crucial for ensuring material consistency in engineered barrier systems.

Furthermore, it is going to analyse microscale properties such as porosity, local void ratio, and particle contacts under different compaction and stress states, which is essential to understand the mechanical behaviour of the mixtures.

These findings will be evaluated in the context of real-world applications, offering valuable insights into the long-term performance and reliability of claystone-bentonite barriers.

#### Plans

The used materials are processed Opalinus Clay (sandy facies, Mont Terri URL, CH), Ca-bentonite, and their mixtures with varying bentonite content (0–40% in 10% increments). The preliminary phase focuses on defining an appropriate grain size for crushed OPA and optimizing sample preparation at hygroscopic water content to ensure specimen cohesion. Previous trials indicate that water content adjustment is necessary when using a maximum grain size of 4 mm.

For sample preparation, specimens will be produced through oedometric compaction, with a sample diameter of 10 cm. Two different compaction states will be applied, leading to different bulk densities and degrees of saturation, while maintaining a consistent initial water content. This approach allows to assess the influence of compaction on microstructural properties and porosity distribution.

For BIB-SEM analysis, specimens ( $l = 10$  mm,  $h = 7$  mm,  $w = 4$  mm) will be extracted and prepared with minimal disturbance to preserve their microstructure. A key challenge is developing a handling method that ensures the structural integrity of the compacted material during processing.

### 6.2.2 BRGM

#### Summary of earlier studies

BRGM has participated to project such as CEBAMA, HITEC and in partnership with ANDRA during which development was done in the miniaturization of oedometer set up coupled to X-ray techniques ( $\mu$ CT, WAXS) (Massat, 2016; Chaaya, 2023) that enable to go deeper in the characterization of hydration processes at lower scale. These new developments allow us to depict the impact of initial microstructure on swelling behaviour (Massat, 2016; Massat et al., 2016) and the relation between interlayer water distribution and swelling pressure development according to cation valency (Chaaya, 2023; Chaaya et al., 2023).

#### Objectives

The first objective of BRGM is to quantify interlayer hydration (water layer distribution) and also anisotropy effect and swelling behaviour for different dry densities, different cation saturation (monovalent and divalent) and pore water salinity. This information is of prime importance to understand the variability in swelling pressure capacity of bentonites. This data will provide input data and parameters for development and validation of predictive modelling.

The second objective is correlation of above with the new raw bentonite characterization (smectite content, sand proportion, cation saturation) to evaluate the influence of predictive parameters on swelling capacity.

#### Plans

Quantification of the water layer distribution and anisotropy as a result of hydration and swelling and to quantify osmotic and crystalline contribution by using miniaturized oedometric devices enabling in situ/operando measurements of swelling pressure coupled with wide angle X-ray scattering (WAXS). This work will be done in exchanged purified montmorillonite (PM) (Na, Ca) and PM/sand mixtures with different size of sand and smectite to control the orientation of the particles. The plans also include the characterisation of new raw bentonites (mineralogy, geochemistry and physico-chemical properties) and the measurement of the swelling capacity of new raw bentonites in the framework of ANCHORS.

### 6.2.3 Ciemat

#### Summary of earlier studies

CIEMAT has participated in the related projects FEBEX, BEACON and PEBS and also in the work packages of EURAD HITEC and GAS, during which specific laboratory equipment has been developed and improved. Thermo-hydro-mechanical, physico-chemical and geochemical characterisation of various bentonites and bentonite/sand mixtures, in the form of compacted blocks and as pellets mixtures has been performed. Materials coming from long-term field tests, with a high degree of maturation (10-18 years) under repository conditions (such as the FEBEX or the EB tests), have been analysed.

#### Objectives

The objectives are to reduce uncertainties about conditions and phenomena influencing bentonite microstructural evolution during hydration and swelling, focusing on bentonite initial variability (e.g. exchangeable cations), providing input data and parameters for development and validation of models and the identification of the characteristics of aged materials and how they compare with those of materials tested in the laboratory for shorter periods of time under similar conditions.

#### Plans

The plans include the analysis of the microstructural changes that take place as a result of hydration and swelling, trying to identify osmotic and crystalline mechanisms using techniques such as nitrogen adsorption, XRD and MIP to determine pore size distribution, interlayer swelling, specific surface area. This will be done in raw bentonites, perhaps exchanged bentonites (Na, Ca) and aged bentonites coming from thermo-hydraulic tests (laboratory and “in situ”) with the characterisation of new raw bentonites (mineralogy, geochemistry, physico-chemical properties).

The study of samples coming from long term tests is going to be done: Microstructural characterisation of bentonites (blocks and pellets) hydrated under room temperature for long periods of time (tests started during BEACON project and dismantled in the framework of ANCHORS), review of data available from long-term, large-scale tests (EB, FEBEX) and finally, the comparison with the characteristics of the bentonite coming from shorter laboratory tests (started during the BEACON project), which will give an insight on temporal evolution.

### 6.2.4 GI-BAS

#### Summary of earlier studies

The general geological and geochemistry of the Kardjali bentonite deposits have been elucidated in the early 1960s (Atanassov and Goranov, 1961/62, 1963/64) when started its industrial exploitation of bentonite quarries. Later the structure, chemical and mineral composition of the Kardjali bentonite have been studied in connection with the producing of bentonite for preparation of moulding materials in metallurgy, drilling usage and sorbents in the chemical industry (Atanassov and Goranov, 1986; Bozhinov and Bechev, 1979). In terms of particle size distribution, Kardjali bentonite consists of three fractions mainly: clay fraction (<2 µm) - about 90%; silt fraction (2-63 µm) – 5%, and sand fraction (>63 µm) – 5% (Bozhinov et al. 1984). The clay fraction is composed mainly by montmorillonite, accompanied by beidellite, illite, kaolinite, as well as some members of kaolinite-halloysite group (Ileva, 2006). The silt fraction is dominated by feldspars, calcite, and cristobalite although some montmorillonite is also present. The sandy fraction consists of quartz, calcite, feldspars and clinoptilolite. Traces of calcite and cristobalite are present in the three fractions. This bentonite expands a lot and the formation of gel at the front of expansion has been studied by Daniels et al. (2021).

It should be mentioned that this bentonite is using in FISST test in Finland as part of the buffer (Tsitsopoulos et. 2023).

## Objectives

The objectives are to know the sealing characteristics of a Ca-bentonite from Bulgaria and understanding the effects of chemical features on the swelling pressure and mechanical behaviour of bentonite-sand mixtures. So far, the Kardjali bentonite is not investigated for the purposes of isolation of radioactive waste disposal.

## Plans

The plans are to characterise mineral and chemical composition of the Ca-bentonite from the Kardjali bentonite deposit (Bulgaria), to investigate microstructure of this Ca-bentonite and bentonite: quartz sand mixture (70/30 % in dry mass) and finally, to assess the effect of geochemical properties on swelling pressure and mechanical behaviour of bentonite and bentonite-sand mixture tested in oedometers and triaxial cells.

### 6.2.5 GRS

#### Summary of earlier studies

Only little information on effects of pore water chemistry, particularly of alkalinity, on the time-dependent volume change and water transfer behaviour of claystone-based materials is available in the literature.

Middelhoff et al. (2020) and Middelhoff et al. (2023) present comprehensive experimental studies. They aimed to analyse the effect of pore water chemistry on the evolution of swelling pressure and hydraulic conductivity of samples which were composed of crushed Callovo-Oxfordian (COX)-claystone and a crushed COX-claystone mixed with MX80-bentonite in a ratio of 70/30 in dry mass. Swelling pressure and saturated hydraulic conductivity were determined applying constant-volume and constant-head methods. In the course of experiments, samples were saturated with deionized/ distilled water, synthetic formation water, and synthetic concrete leachate. Upon their termination, hydraulic conductivity experiments were complemented by performing post-mortem analyses, with particular focus on (the spatial distribution) of mineralogy as well as physico-chemical and structural properties. Middelhoff et al. (2020) reported that pore water chemistry hardly affected the evolution of swelling pressure, probably on account of the low ionic strength of solutions and the short duration of experiments of 7 days. Moreover, the effect of pore water chemistry vanished as the initial dry density of materials increased. Although the duration of experiments exceeded 1 year, Middelhoff et al. (2023) also observed only a negligible effect of pore water chemistry on the hydraulic conductivity. Post-mortem analyses indeed revealed that saturation-induced swelling provoked clogging of hydraulic conductive macropores, but mineralogy and physico-chemical remained stable. Eventually, their observations were attributed to the low ionic strength of solutions and to low kinetics of geochemical reactions.

The experimental study presented by Zeng et al. (2022) was similarly comprehensive. It aimed to analyse the effects of not only pore water chemistry but also of mineralogy and dry density on the evolution of swelling pressure and hydraulic conductivity of compacted samples. They were composed of crushed COX-claystone that was mixed with MX80-bentonite or Sardinia-bentonite in ratios of 70/30 in dry weight. Also, they applied constant-volume and constant-head methods and saturated with deionized/ distilled water, synthetic formation water, and synthetic concrete leachate. Mercury intrusion porosimetry complemented their experiments upon termination. Zeng et al. (2022) reported that pore water chemistry affected the evolution of swelling pressure and hydraulic conductivity of crushed COX-claystone/ Sardinia-bentonite-samples more considerable than that of crushed COX-claystone/ MX80-bentonite-samples. The effect intensified by reducing the initial dry density. They attributed the observations to interactions of hydroxide ions with montmorillonite in macropores causing a decrease in swelling capacity and an increase in macropore volume.

## Objectives

The work principally aims to improve the understanding with regard to time-dependent effects of pore water chemistry on the (spatial distribution of) physical, physico-chemical, mineralogical and structural

## **EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

properties of claystone-based materials and on their volume change and water transfer behaviour using state-of-the-art (non-) destructive experiment methods. Overall, the work aims to link changes in volume change and water transfer behaviour which are measured at milli- and centimeter scale to changes in physical and physico-chemical properties, mineralogy and structure. The linkage can be used to improve interpretative approaches for other experiments, and to improve numerical analyses by integrating it in existing constitutive models.

### Plans

It is planned to analyse the time-dependent response of the volume change and water transfer behaviour of claystone-based materials to the chemistry of different pore waters. Materials used in this experiment program are excavated Opalinus clay from the sandy facies of Opalinus clay (Mont Terri URL, Switzerland) as well as Georgian and Calcigel bentonites. Materials are classified and then mixed in a pre-defined ratio. The grain size distribution represents a pre-defined bi-Fuller-type curve. In parallel, GRS and JYU perform combined swelling pressure/ hydraulic conductivity-experiments on compacted claystone/bentonite-mixture samples applying constant-volume and constant-head methods. Samples are saturated with synthetic pore water and synthetic concrete leachate.

In the course of experiments, the analyses are complemented by JYU by performing CT-scans and by GRS by analysing the composition and properties of effluents. The time when analyses are performed is determined by the volume of pore water exchanged. It is expected that the experiments last more than two years. The comparability of experiment programs accounts for the transferability of findings of GRS to JYU and vice versa. The experiment programs of GRS and JYU are complemented through the performance of post-mortem analyses by BGR.

### 6.2.6 HUN-REN EK

#### Summary of earlier studies

HUN-REN EK has participated in the EURAD program in the ACED (Fabian et al. 2023; Tolnai et al. 2023), FUTURE (Czompoly et al. 2023, 2024) and SFC WPs.

Swiss-Hungarian cooperation program: Development of a macro and microscopic approach to investigate the geochemistry of radioactive waste disposal systems (Keri et al. 2016; Gergeli et al. 2016; Breitner et al. 2015; Osan et al. 2014).

Industrial R&D project: Characterization of simulated liquid radioactive waste in a new type of cement mixtures (Fabian et al. 2022).

R&D project with Eötvös Lorand University, Assessing the immobilization capacities of new cement paste mixtures containing natural and B-10 enriched boric acid (Rostamiparsa et al 2023).

#### Objectives

The objectives are the microstructural characterization of compacted bentonite-claystone mixtures, B75/BCF ratios ranging from 0.33 to 1.00 and experimental testing of different bentonite/claystone ratios to obtain an optimal mixture for buffer material.

#### Plans

HUN-REN EK interest focuses on the evaluation of the physico-chemical properties of bentonite-based mixtures that may be used as buffer material in repositories. Mixtures of B75 bentonite and BCF claystone at different proportions are planned to be tested experimentally. These mixtures could exhibit a very complex physico-chemical and mechanical behaviour governed by the strong heterogeneity on the microscale. Physico-chemical, mineralogical characteristics and porosity will be studied on the microscale on compacted samples of the proposed mixtures. The evaluation of the microstructural properties provides a scientific basis to understand the physical (strength, porosity, deformation, mineralogy) behaviour which should be supplemented with chemical characterization. Microstructural analysis will be performed using scanning electron microscopy coupled with energy dispersive

spectroscopy (SEM-EDS) to study the morphology, texture, and composition of bentonite/claystone mixtures. To characterize the porosity of the mixtures, the most important factor is to measure the density and to perform X-ray microtomography, which would provide crucial information for composition optimization. This provides information about overlapping layers, possible aggregates of bentonite/claystone grains, and the heterogeneity of the mixtures, so that the compactness of the structure can be optimized.

### 6.2.7 ASNR

#### Summary of earlier studies

Zaidi et al. (2025) investigated gas migration within a heterogenous mixture of MX80 bentonite pellets and powder with a ratio of 80/20 in dry mass by X-ray  $\mu$ CT observations. A novel X-ray transparent constant volume cell has been developed to assess the effect of gas pressure, material heterogeneities, and water vapor gas saturation on breakthrough pressure and gas pathways. Gas injection phase was followed by a long resaturation phase restoring material homogeneity as observed at  $\mu$ CT resolution scale (16  $\mu$ m). X-ray  $\mu$ CT results also revealed the opening of the specimen/cell wall interface during gas passage. This opening expanded as the injection pressure increased.

Barakat (2023) investigated the chemical-hydro-mechanical behaviour of Opalinus Clay from the Lower Sandy Facies (*LSF*) of Mont Terri site. This claystone has the advantage of having a very complex mineralogy which allows studies of the contribution of the various clay and non-clay minerals (quartz and calcite), as well as the coupled chemical-hydro-mechanical behaviour. The effects of salinity and alkalinity on the microstructure, mineralogy, water retention properties, swelling properties and compressibility of the investigated claystone were determined with regard to the high mineralogy variability. In addition, to complete the investigation at micro scale, a dedicated cell for X-ray computed microtomography was developed and scans were realized at different exposure times.

#### Objectives

The objective is investigating the effect of pH increase on the structural distribution of a bentonite sand mixture under chemical and gas loadings (optional) (e.g creation of macropores, heterogenous distribution of the mixture with zones with bentonite partially filling the voids between the sand particles). Particular attention will be paid to the capacity of the bentonite to self-seal the possible pathways that might be created due to the dissolution of sand under alkaline environments.

#### Plans

The investigated material will be a mixture of MX80 bentonite and sand with a proportion of 40/60 (in dry mass). However, several proportions might be investigated by increasing the sand proportion. At the microstructural level, the mixture with high sand content is probably characterized, by a sand skeleton with bentonite filling partially the remaining voids. Particular attention must be paid to the capacity of the bentonite to self-seal the possible pathways that might be created due to the dissolution of sand under alkaline exposure. The mixtures will be tested within an X Ray  $\mu$ -CT transparent cell at constant volume condition. The cell allows to perform high resolution X-ray computed micro-tomography (X-ray  $\mu$ CT) scans to track microstructural changes during different phases of saturation. The cell consists of hollow cylinder with a volume of approximately 9 cm<sup>3</sup> (23 mm in diameter and 22 mm in height). To ensure X-ray transparency, the cylinder is made of a high-performance semi-crystalline thermoplastic material. The cell was validated by Zaidi et al (2025). The compacted mixtures will be hydrated with an hyperalkaline solution of pH=13.5. The solution has the same chemical components and pH as for the concrete pore water ( $NaOH - KOH - Ca(OH)2$ ). As a reference, a synthetic water, with a chemical composition similar to that of the pore water in Collavo-Oxfordian (COX) clay will be used. Regular micro-CT scans will be taken upon sample exposure to the alkaline solution. Initial and post-mortem analysis will be performed to determine microstructural changes (by mercury intrusion porosimetry, nitrogen adsorption, etc.). Gas testing will be performed at full saturation. Regular scanning will be performed during gas injection tests.

## **EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

In addition, the aged sand bentonite mixture extracted from SEALEX in situ tests (if test dismantling occurs during EURAD-2) will be characterized through a set of microstructural analysis including destructive and non-destructive methods (e.g. X Ray micro-CT testing, mercury intrusion porosimetry, nitrogen adsorption, etc.). Cores can be shared with interested partners when available.

### 6.2.8 Jyväskylä university

#### Summary of earlier studies

Recently, an X-ray tomography method for measuring the local water content and local displacements of clay samples non-destructively has been developed at JyU (Harjupatana et al. 2015; Miettinen et al. 2016; Harjupatana et al. 2022; Tanttu et al. 2024). The method is based on placing the sample in an X-ray transparent cell, hydrating it with water or other fluid, taking consecutive X-ray tomographic images of it during the resaturation process, and finally employing image analysis techniques to find local deformations, local water content, and local dry density in the sample. The measurement process has been validated through gravimetric measurements (post-mortem slicing the sample) and applied in several previous projects such as BEACON, EURAD, and many national projects to study to bentonites of different compositions in varying chemical conditions, pressures, and temperatures. Although the method produces rich hydromechanical data of the sample, its applications to inhomogeneous materials such as granular bentonites has not been explored yet.

#### Objectives

The main objective is to produce spatiotemporal data on moisture content and dry density of different bentonites and inhomogeneous (10-100 µm scale) bentonite-based materials (other materials mixed with bentonite or granular bentonite) in constant-volume hydration experiments. The produced data will reduce uncertainties in water content evolution measurements done with slicing-based techniques, improve understanding of the water transport mechanisms such as the roles of non-linear diffusion and Darcian flow, and be useful for model calibration and validation.

#### Plans

In this project, it is planned to perform bentonite hydration experiments in constant-volume conditions. Georgian and Italian bentonites will be used and to study the possibilities of applying the X-ray tomography method to granular Georgian bentonite in order to see the effect of the inhomogeneous structure on the hydration process.

Further, there will be collaboration with GRS and BGR, also partners in ANCHORS, and apply the X-ray tomography method to study the hydration of a mixture of claystone and Georgian bentonite saturated with synthetic pore water and synthetic concrete leachate. In the collaboration, GRS will analyse the effluents, and BGR will perform post-mortem analysis other than tomography.

Finally, the participation in the post-mortem analysis of samples from THMC experiments by studying their µm-scale structure through X-ray tomographic imaging is going to done. Here, the contribution is to check if differences in the properties of samples tested in benchmarks performed in ANCHORS can be attributed to the µm-scale structures.

### 6.2.9 University of Lorraine (LEMTA)

#### Summary of earlier studies

*Swelling pressure development and inter-aggregate porosity evolution upon hydration of a compacted swelling clay*

A specific oedometer cell has been set up to measure the swelling pressure of compacted montmorillonites at constant volume and to concomitantly visualize the evolution, upon wetting, of how the microstructure is organised through X-ray microtomography (Massat et al., 2016; Yigzaw et al., 2016). The swelling pressure experiments were conducted with solvents of various natures. In addition to conventionally used water and saline solutions, an organic solvent (methyl methacrylate – MMA). to

## **EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

explore the effect of its different physical and chemical properties, and to differentiate the respective roles of crystalline and osmotic pressures on macroscopic swelling behaviour. The results, which combined both swelling pressure measurements and quantification of microstructure evolution upon hydration for the two different solutes, give sound understanding on the development of osmotic and/or crystalline swelling and their relative impact both on the microstructure and on the magnitude of the macroscopic swelling pressure of compacted montmorillonites.

### *Sand-claystone mixtures*

Several studies have investigated the behaviour of crushed claystone and bentonite-claystone mixtures (Kaddouri et al., 2019; Middelhoff et al., 2023, 2021, 2020) Investigating the impact of sand proportions on hydromechanical behaviour. Properties of these backfill materials must satisfy a number of requirements regarding their hydromechanical properties to, for instance, limit the convergence of the deep galleries or to limit gas pressure build up in the repository. The contribution of sand fraction to the mechanical behaviour of the mixture is still poorly understood (Kouakou et al., 2020; Wang et al., 2021). Moreover, compression may alter the microporosity of the mixture and thus alter the gas permeability. In this context, work is being carried out on the effect of sand on the hydromechanical behaviour of the material. Mixtures with different proportions of sand are thus studied at different scales in order to provide a better understanding of the mechanisms involved. This includes saturated odometer tests for hydraulic conductivity and compressibility measurements as well as X-ray microtomography imaging under loading.

### Objectives

The objectives are to monitor the sand skeleton development and evolution upon mechanical loading, with the main purpose of understanding how the sand presence lower the compressibility of the clay matrix, to determine the deformation of the clay matrix upon mechanical loading and infer how it will alter the transfer properties of the mixture, to analyse the impact of suction of loading on the above-mentioned processes and finally, to determine the best sand proportion to achieve a minimum compressibility of the mixture

### Plans

To better describe these mechanisms at the grain scale, four-dimensional X-ray microtomography scans will be performed on mixtures of crushed argillite and sand in various proportions (Massat et al., 2016; Yigzaw et al., 2016). The acquisitions will be carried out using an oedo-tomometer developed as part of this research to include a direct control of vertical stress. The device will be modified during the project to include suction control with the osmotic technique. This device enables odometer tests, including saturation and loading, to be conducted within a laboratory tomograph. Successive scans at different stress states were obtained for several mixtures. A volumetric correlation approach will be then used to closely monitor microstructural rearrangements induced by loading and to observe the evolution of internal structures.

### 6.2.10 University of Strathclyde

#### Objectives

Understanding microstructural evolution upon wetting at different pore water chemistry.

#### Plans

It has been planned to perform XRD analysis on Wyoming MX-80 and Ca Bentonite (source to be confirmed), LS particle size distribution analysis on Wyoming MX-80 and Ca Bentonite (source to be confirmed).

Several microcells, as described in Seiphoori et al. (2014) will be manufactured in order to perform the wetting at constant volume. The microcells, a 30mm brass container, will allow to compact, wet at constant volume, measure the swelling pressure, measure suction at target water contents and perform

freeze-drying, without the need to remove the sample from the cell, assuring zero disturbance. Given the simple design and the low costs, multiple cells will be prepared. This will allow multiple experiments to run simultaneously. As the wetting fluid will have high salt content, wetting cannot be performed by controlling the RH as suggested in Seiphoori et al. (2014). Wetting will be performed direct exposure to the wetting fluid. At each wetting step, a controlled amount of solute will be provided to the sample and let equilibrate. Suction measurement will be performed without removing the sample from the microcell, by using a chilled-mirror dew-point potentiometer.

MIP and SEM will be performed at the Advanced Materials Research Laboratory at University of Strathclyde. Dehydration of the samples will be performed by means of freeze-drying technique according to the experimental procedures already developed. For the samples created with each microcell will be directly submerged into liquid nitrogen as recommended in Seiphoori et al. (2014).

### 6.3 THMC(G) Laboratory testing

Tests at different scales in oedometric conditions are going to be performed for checking the scale effect. These tests are going to be performed with different bentonites, sand-bentonite mixtures and crushed rock-bentonite mixtures.

The samples will be saturated with deionized/distilled water, saline waters and alkaline waters for studying the effects of salinity and alkalinity in bentonites and mixtures of sand or crushed rock with bentonite.

Tests ongoing during some years can be dismantled and the aged samples analysed using the techniques described in this section and in previous section.

The difference in saturation with vapour and liquid water will be studied.

The friction effect can be assessed with small scale tests in order to know its influence in swelling pressure tests.

#### Benchmarks

The Strategic Research Agenda points out that past projects have shown significant variability of measured THM properties of buffer materials (such as swelling capacity, water retention properties, compressibility), which are key inputs needed for establishing correct EBS functionality. This difference was obtained even if tested on nominally identical samples in different soil mechanics laboratories. In addition, full-scale measurements in mock-up tests often show results inconsistent with laboratory measurements on small-scale samples. BEACON was one of the past projects where a comparison between results obtained in different laboratories was performed (tests from Ciemat, SKB and CTU) although the results were not reported.

The experimental benchmark exercise is described in Milestone MS32. The goal is to conduct simple tests that can be carried out by as many partners as possible. The tests will be constant volume tests and conventional oedometric tests with a simple loading-unloading path and hydration with chemical solutions (alkaline/saline).

The benchmark tests' results will be used in Subtask 4.1 for models' validation (Milestone MS33) and also for obtaining models' parameters needed for carrying out the simulations.

The following section outlines the contributions of the partners involved in Subtask 3.2. Each participating organisation provides a summary of its relevant previous studies, along with key references, as well as the objectives and planned activities to be carried out within the ANCHORS.

### 6.3.1 Ciemat

#### Summary of earlier studies

CIEMAT has participated in the related projects BEACON, FEBEX, PEBS and EURAD work packages HITEC and GAS, during which specific laboratory equipment has been developed and improved (see Villar, 2002; Villar et al. 2016, 2021, 2022a, 2022b, 2023, 2024; Gutiérrez-Rodrigo et al. 2021; Gramegna et al. 2023). Namely, decimetric cells to test the behaviour of buffer materials in conditions close to the repository ones have been used for decades, the more recent developments including online testing of the evolution of relative humidity, temperature and axial and radial pressures. These cells allow to impose thermo-hydraulic gradients representative of the field ones, and also to test at a representative scale the interaction between different buffer materials (e.g. pellets and blocks). Gap closure and gas transport have also been analysed. Expertise on postmortem characterisation of treated materials including physico-chemical and geochemical characterisation (mercury intrusion porosimetry, total and external specific surface area, water sorption isotherms, soluble salts, exchangeable cations, access to XRD facilities, etc.).

#### Objectives

The objectives are to reduce uncertainties about conditions and phenomena influencing bentonite swelling development and its eventual homogenisation: the scale effect, the salinity effect and the differences/similarities among bentonites with different exchangeable cations and to reach a conceptual understanding of the evolution of bentonite fabric and microstructure upon hydration (micro/macro interactions) and swelling development and of the factors affecting it. These activities, in connection with those in Subtask 3.1, will allow identifying the factors causing a larger heterogeneity in the barrier as a result of operation.

#### Plans

Laboratory work performed in decimetric cells intended to reduce uncertainties about conditions and phenomena influencing bentonite swelling development and its eventual homogenisation. In particular, the scale effect, the salinity effect and/or the differences/similarities among bentonites with different exchangeable cations will be analysed. These studies provide input data and parameters for development and validation of models.

Small-scale swelling pressure tests are going to be performed for checking the influence of water salinity/alkalinity on different bentonites as a function of dry density. Results of small-scale swelling pressure tests obtained previously on FEBEX and Wyoming MX-80 bentonites using NaCl and CaCl<sub>2</sub> solutions will be analysed and reported.

Tests to analyse the impact of water salinity on the fabric and microstructure evolution of initially inhomogeneous bentonite (compacted blocks and/or pellets) or of different bentonites. These tests will be performed in large-scale cells (10-15 cm) with measurement of axial and radial pressures and RH. Similar tests were performed during the BEACON project with deionised water and these will be used as baseline to compare the effect of salinity. Dismantling, analysis and reporting of several tests started during BEACON with FEBEX and Wyoming MX-80 bentonites, all of them using deionised water, will be performed. In particular, the connection between the water intake and the development of swelling pressure will be analysed, as well as the relation between axial and radial pressures and the possible friction effects.

### 6.3.2 Clay Technology

#### Summary of earlier studies

Extensive experimental and analytical work on bentonite homogenisation has been conducted at both laboratory scale (Dueck et al., 2019; Dueck and Börgesson, 2021) and full-scale in situ tests (Sandén et al., 2017). Ongoing studies, including a decades-long series of experiments, focus on factors such as the role of friction in the homogenisation process (Dueck et al., 2022, 2018, 2016, 2014, 2011).

## **EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Properties related to homogenisation, such as swelling pressure, water retention curves, vapour transport, and moisture redistribution, have been extensively examined in various projects (Åkesson et al., 2012; Dueck, 2008, 2007; Dueck and Nilsson, 2010; Villar et al., 2023). The role of slow re-saturation has also been discussed (Sellin et al., 2017).

Closely related to slow re-saturation is hydration under reduced water activity. Studies using solutes such as PEG to control water activity and explore water uptake in bentonite, where hydration of liquid water is compared to hydration of steam at equivalent water activity (RH < 100%), have been reported by several authors (Cuisinier and Masroui, 2005, 2004; Delage et al., 2008, 1998; Delage and Cui, 2008; Vandoorne et al., 2022; Wang et al., 2023; Yigzaw et al., 2016; Zhang et al., 2020).

Evidence suggests that hydration by liquid water leads to a different final state than hydration by vapour (Birgersson and Goudarzi, 2013; Villar et al., 2023). Notably, Na-montmorillonite exhibits indefinite swelling in liquid water (Birgersson et al., 2009; Hedström et al., 2016), whereas swelling in vapour appears limited, as observed using XRD (Brindley and Brown, 1980; Fu et al., 1990).

These observations are consistent with findings in related fields. Materials such as gelatin, ion-exchange membranes, swelling polymers, and oriented lipid bilayers also exhibit larger swelling in contact with liquid water than with saturated vapour (Davankov and Pastukhov, 2011; Podgornik and Parsegian, 1997; Rand and Parsegian, 1989; Roldughin and Karpenko-Jereb, 2016; Vallières et al., 2006). While discrepancies in hydration are sometimes attributed to experimental challenges in maintaining 100% RH (Beers et al., 2014; Katsaras, 1998; Nagle and Katsaras, 1999), differences in water uptake have also been confirmed at water activities below 1 (Bass and Freger, 2008).

### Objectives

The primary objective of the proposed laboratory tests is to enhance understanding of the processes of swelling, swelling pressure development, and homogenisation following hydration by water vapour compared to hydration by aqueous solution.

The study aims to interpret the results in the context of previous research on homogenisation under varying conditions, including geometry and water accessibility. Swelling pressure data will also be compared with findings from other studies involving aqueous solutions with reduced water activity.

Where applicable, parallels will be drawn to similar phenomena observed in other systems. Finally, the results will be linked to repository conditions, contributing to improved predictions of long-term repository performance.

### Plans

The study will combine a literature review, examining differences between hydration by vapour and by aqueous solution, with experimental investigations. Experiments will focus on refining and developing methodologies suitable for both hydration modes. To address challenges associated with 100% relative humidity (RH), the work will primarily use lower RH levels, with corresponding water activity in aqueous solutions regulated by solutes such as PEG.

The tests will be conducted on MX-80 bentonite using a constant-volume test cell (cylindrical, inner diameter 50 mm, height 10 mm) with aqueous solution or vapour circulated through the filters. Continuous pressure measurements will be recorded during the tests. Post-test analysis will include determination of water content and density.

Benchmark swelling pressure tests, which may also include bentonites other than MX-80, will utilise similar equipment. For these tests, swelling pressure will be measured only from above, and a smaller test cell diameter (35 mm).

### 6.3.3 CNRS-Navier

#### Summary of earlier studies

The hydro-mechanical behaviour of bentonite pellets (Mokni et al. 2018; Molinero-Guerra et al. 2016, 2018a, 2018b; 2019a; 2019b; Yang et al. 2023a, 2023b, 2024a, 2024b, 2024c, Zhang et al. 2022a, 2022b, Zeng et al. 2021) and bentonite/sand mixtures (Cui et al. 2008; Cui and Tang, 2013; Cui, 2017; Saba et al. 2013, 2014a, 2014b, 2014c, Wang et al. 2013a, 2013b, 2013c) have been investigated by both experimental and numerical methods. Effects of initial dry density (Cui and Tang, 2013; Cui, 2017; Mokni et al. 2018; Molinero-Guerra et al. 2016, 2018a, 2018b; 2019a; 2019b; Saba et al. 2013, 2014a, 2014b, 2014c, Wang et al. 2013a), initial structure (Mokni et al. 2018; Molinero-Guerra et al. 2018b, 2019b), axial/radial technological voids (Bian et al. 2019; Cui et al. 2008; Cui and Tang, 2013; Cui, 2017; Mokni et al. 2018; Molinero-Guerra et al. 2016, 2018a, 2018b; 2019a; 2019b; Saba et al. 2013, 2014a, 2014b, 2014c, Wang et al. 2013a, 2014b) and chemistry Cui and Tang (2013) on the swelling pressure, hydraulic conductivity and microstructure of such material were revealed. Based on experimental results, constitutive models were developed and validated against the measured data. It is recognized that the swelling pressure increased with the increase of effective dry density of bentonite. The axial swelling pressure depends on global dry density while the radial swelling pressure depends on local dry density nearby the sensors. The development mode of swelling pressure depends highly on the initial structure and wetting path. With presence of axial/radial technological voids, swelling and compression zones coexist in the wetting sample, accompanied by significant evolution of microstructure of the sample. As for the influence of chemistry, generally the swelling pressure decreases with increasing concentration and pH of the solutions. However, for in-depth understanding the buffer/sealing performance of bentonite-sand mixtures, further studies on the effects of chemistry on the swelling pressure and homogenization process of bentonite-sand mixtures are still required.

#### Objectives

The objectives are to characterise two aged bentonite pellet/powder mixtures after long-term (9 & 8 years) mock-up infiltration tests, in terms of density, water content/suction and microstructure, to investigate the chemistry (salinity and alkalinity) on the swelling pressure of bentonite/sand mixture and to investigate the chemistry (salinity and alkalinity) on the homogenisation process of bentonite/sand mixture by mock-up test.

#### Plans

The plans are to measure the distributions of density, water content/suction and microstructure of two aged bentonite pellet/powder mixtures in two long-term (9 & 8 years) mock-up infiltration tests will be characterized.

The effect of chemistry (salinity and alkalinity) on swelling pressure of compacted MX80 bentonite/sand mixture will be investigated by performing swelling tests under constant-volume condition using different solutions of different concentrations/pHs.

Mock-up infiltration tests using saline and alkaline solutions will be performed on compacted MX80 bentonite/sand mixture with the presence of technological voids (axial or radial), with monitoring of relative humidity/suction and swelling pressure at different positions.

### 6.3.4 CUNI

#### Summary of earlier studies

The Charles University soil mechanics laboratory has been involved within a number of international (BEACON, EURAD) and national research projects in the past. These projects led to a development of constitutive model for bentonite (THM hypoplastic double structure model) and to a detailed description of the behaviour of reference material (BCV bentonite), which is going to be studied in the present project. In particular, the constitutive model has been developed by Mašín (2013, 2017). Microstructure of the predecessor material to BCV bentonite (named B75 bentonite), from the same geological deposit,

## **EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

has been studied by Sun et al. (2019, 2020a, 2020b). Quantification of properties of BCV bentonite exposed to high temperatures has been investigated at laboratory temperatures by Laufek et al. (2021), Svoboda et al. (2022) and Najser et al. (2023), who also targeted the capacity of BCV bentonite to homogenisation. More recently, Najser and Mašín (2024) studied time-dependent properties of BCV bentonite (in particular, swelling pressure relaxation) at elevated temperatures.

### Objectives

The main objective will be to determine the range of applicability of the mixture materials (BCV bentonite with Strelec sand admixture) as buffer material. The research outcome will be quantification of mixture key properties (swelling pressures and permeability) at different percentages of coarse-grained fraction. These results will be compared with previously performed measurements on pure bentonite (in the form of both compacted blocks and pellets), which were done at Charles University and partner institutions during the past research projects. Potential deterioration of the key properties with coarse admixture will be studied to quantify the maximum amount of coarse-grained fraction under which the material keeps its required properties.

### Plans

The work in ANCHORS will focus on behaviour of mixtures of the Czech Ca-Mg bentonite BCV with granular material (Strelec sand) at different percentages. The study will explicitly target the performance backfill material to be used within the planned nuclear waste repository. In particular, the work will be focused on the determination of ranges of usability of materials/mixtures in terms of HM properties the repository needs and the optimization of the filling materials based on Czech Ca-Mg bentonite (pellets, powder) and sand mixtures.

Verification of self-healing capacity/integrity of heterogenous materials will also be performed.

### 6.3.5 CTU

#### Summary of earlier studies

CTU has participated in previous related studies within the EURAD (WP HITEC, WP GAS, Olin et al, 2024, Villar et al 2024, Marschall et al., 2024), BEACON (Fabian et al. 2022), FORGE, FEBEX projects. Within these projects, procedures and laboratory equipment have been developed which will be used in ANCHORS. Standard geotechnical techniques such as oedometric cells and constant volume cells for measuring swelling pressure and hydraulic properties have been used for a long time. These cells have been upgraded within WP HITEC to allow the measurement of total pressure from top&bottom, thus allowing the investigation of friction. Prior to the WP GAS project (and validated within) a laboratory equipment&methodology was designed and tested for rapid gas breakthrough testing. The results of the rapid gas breakthrough tests can be used to assess the integrity (resistance to gas pressure) of the material.

### Objectives

The main objective of this research is to determine the range of application of different materials and mixtures based on their thermal-hydraulic-mechanical (THM) properties. A key focus is the optimisation of fill materials, particularly those based on Czech Ca-Mg bentonite. This includes the development and evaluation of bentonite pellets and bentonite-rock mixtures to improve their performance in sealing and stabilisation applications. In addition, the study will provide an experimental basis for scaling up numerical models that simulate the behaviour of heterogeneous materials. By investigating the effects of friction and structural variability, the research will support the transition from small-scale laboratory experiments (centimetres) to larger, more realistic applications (decimetres to metres). This will help to improve the accuracy and reliability of predictive models used in repository design and safety assessment. Determination of ranges of usability of materials/mixtures in terms of THM properties

#### Plans

The plans are determination of thermophysical properties of bentonite sand mixtures and pellets, determination of swelling/compressibility parameters (oedometers), determination of hydraulic conductivity and swelling pressure of bentonite sand mixtures and pellets and finally, to perform mini mock-up – medium scale experiments

#### 6.3.6 EPFL

##### Summary of earlier studies

In the French concept, extensive research has been carried out on the crushed Callovo-Oxfordian claystone (COXc) for use as backfill. Studies, including those by Tang et al. (2011a, 2011b) and Middelhoff et al. (2021), have demonstrated that the hydro-mechanical behaviour and microstructural characteristics of COXc are significantly influenced by the initial material properties. The compaction method and the initial dry density have been shown to have a significant impact on swelling potential. The performance of COXc/bentonite mixtures has been widely studied due to their enhanced swelling capacity and hydraulic properties (Middelhoff et al. (2020, 2021, 2023), Robinet et al. (2021), Zeng et al. (2019, 2020b, 2020a, 2021, 2022), Zhang et al. (2014, 2021)).

In contrast, research on crushed Opalinus Clay (OPAc), the host rock selected for the Swiss concept, remains more limited. Zhang (2021) has examined the applicability of the results from COXc to OPAc and OPAc/bentonite. More recently, EPFL has carried out a study on OPAc and improved the understanding of the relationship between microstructure, hydro-mechanical and two-phase flow properties of OPAc.

Previous studies at EPFL have focused on characterising bentonite/sand mixtures and have indicated that a mixture in which less than 50% of bentonite is combined with sand yields promising results (Manca et al. 2016).

##### Objectives

The primary objective of the proposed laboratory tests is to enhance the understanding of the hydro-mechanical behaviour of crushed claystone-based material and its mixture with bentonite and sand as potential backfill and seal. The results will be set in parallel with the evolution of fabric and microstructure. Interpretation of the results will be undertaken in the context of previous research while considering the requirements of the WMOs.

#### Plans

Potential backfilling and sealing are a mixture of crushed claystone/MX-80 bentonite/sand. To be able to compare the results with the existing literature, the type of materials was set and are as follows:

- Crushed claystone: Callovo-Oxfordian
- Bentonite: MX-80
- Sand: Quartz sand

If there is a need for a specific mixture composition, hydro-mechanical tests will be carried out and include optimum compaction properties, stiffness modulus, gas entry pressure and permeability, swelling capacity (pressure and strain), and water permeability. These variables will be adjusted according to the target performance of the backfilling and sealing

(T)HM(C) experiments will be conducted in the advanced experimental facilities already developed at EPFL, such as standardized or high-pressure oedometric and isochoric cells. A preliminary geotechnical characterization of the selected mixtures in their compacted state will be conducted.

### 6.3.7 GI-BAS

Summary of earlier studies

Presented in microscale testing section.

Objectives

- The objectives are to study the Kardjali Ca-bentonite from Bulgaria and its mixture with quartz sand. In particular, to characterise the index properties, to investigate the swelling characteristics when mixed with quartz sand at 70/30 weight ratio, to assess the shear strength of the bentonite and the mixture with quartz sand, and finally to understand the effect of chemical solutions on the swelling pressure and mechanical behaviour of Ca-bentonite and bentonite-sand mixture.

Plans

- It is planned to determine the grain size composition, density of solids and plasticity limits; to perform oedometric tests (sample size - 50 mm diameter / up to 20 mm height) to determine the free swelling strain and the swelling pressure under saturation with different chemical solutions.
- Triaxial tests at low confining pressures (less than 1.5 MPa) will be carried out to assess shear strength – sample size 38 dia./76 mm height or 50 mm dia./100 mm height.

### 6.3.8 GRS

Summary of earlier studies

Information on binary bentonite systems (or two-component bentonite buffer systems) was generated in the European research “BEACON” project (Bernachy-Barbe et al., 2022). A binary system was created by combining powder/pellet or powder/block or pellet/block or block/block. The project had multiple objectives, including experimentally analysing 1. the propagation of the saturation front through a binary system and 2. the homogenization of bentonite in the binary system at micro, milli- and centimetre scales. Experiments were carried out using specially designed oedometer cells in isochoric conditions and considered binary systems, in which one component was stacked over the other. They revealed 1. a dependency of homogenization on the direction and kinetics of hydration and 2. a mostly non-monotonous increase in swelling pressure.

Currently, the international “Sandwich VP” and “Sandwich HP” projects generate information on vertical hydraulic sandwich sealing systems (Wieczorek et al., 2024). The major component of projects is a large-scale in-situ experiment that is installed in the Rock laboratory Mont Terri (Switzerland). The in-situ experiment is complemented with experiments at centi- and decametre scales and numerical simulations. The projects had multiple objectives, including monitoring the propagation of saturation front through alternating segments of bentonite and sand (or a sandwich system). Sand segments facilitate a homogeneous hydration of bentonite segments which generally demonstrates the functionality of the sandwich system. Moreover, results show a non-monotonous increase in axial stress indicating the swelling of bentonite segments upon artificial and natural hydration.

Objectives

In the context of the alternative concept, this study generally aims to experimentally analyse the volume-change and fluid transfer behaviour of a system composed of a sealing core and backfill that are in direct contact. The major objective is the analyses of interactions between sealing core and backfill at decimetre scale, particularly the analyses of swelling-induced displacements at the interface and their effect on the total behaviour of the system. Overall, the data generated in this study can reduce concept-related uncertainties and improve the performance assessment.

## Plans

It is planned to analyse 1. the volume-change and water/ gas transfer behaviour of an abstracted sealing system, 2. the propagation of the saturation front through components (sealing core, backfill) and 3. the interactions at the interface between the components at decimetre scale. Materials are crushed Callovo-Oxfordian (COX) claystone, Wyoming bentonite and Fontaine-Bleu quartz-sand. Backfill material is composed of a mixture of crushed claystone and sand, whereas bentonite and sand are mixed in order to prepare the material for the sealing core. A special oedometer cell is used to carry out combined swelling pressure/ hydraulic conductivity-experiments by applying constant-volume and constant-head methods. Its configuration is special on account of its two confining rings and base. The confining rings are 280 mm in diameter. By separately compacting backfill material and sealing core in confining rings, and then stacking one over the other, an abstracted sealing system including an interface is created. The base is equipped with non-contact-linear position sensors that are adjusted to the interface. They allow to measure its displacements along the centre axis. Upper and lower confining rings are laterally equipped with hygrometers. They allow to measure the relative humidity in materials along the centre axis. The device is complemented with a load frame and two volume-pressure-control units at the in- and outflow sides.

It is planned to determine the integral water/ gas hydraulic conductivity of the abstracted sealing system by injecting gas and then saturating with synthetic pore water. Effluent composition and properties are analysed in the course of experiments. Their duration is expected to exceed one year. Experiments are eventually complemented with post-mortem analyses (e.g. water content, density).

### 6.3.9 ASNR

#### Summary of earlier studies

ASNR has conducted within SEALEX (SEAling EXperiment) and VSeal (Vertical Sealing) projects extensive experimental research at both laboratory and full-scale in situ levels on MX80 bentonite-sand mixtures (70/30 in dry mass) to assess the impact of technological voids, as well as on MX80 bentonite pellet-powder mixtures (80/20 in dry mass) to investigate mixture homogenization (Yang et al., 2024a; Mokni et al., 2016,2020,2023; Molinero-Guerra et al., 2018a,2019a,2020). Comparison of the results in term of relative humidity evolution of different tests conducted on compacted sand bentonite mixture with various technological gaps showed that their presence constituted new hydration sources and flow paths, which impacted the saturation kinetics. The effect of technological voids was also evident when comparing the swelling pressure kinetics (Mokni et al., 2016).

Bentonite pellets mixture is one of candidate material envisaged in France for sealing systems. This mixture consists of high-density compacted bentonite pellets with a quasi-spherical shape and bentonite powder obtained by crushing pellets. Initially, the mixture exhibits significant heterogeneity, with the degree and distribution of heterogeneities evolving during hydration (Molinero-Guerra et al., 2018). Before the launch of the European BEACON project, few studies had investigated the impact of initial dry density gradients or those induced during hydration, and their influence on the hydro-mechanical properties of the mixture (Bernachy-Barbe et al. 2022).

To address this issue, ASNR launched a multi-scale characterization campaign to assess the hydro-mechanical behaviour of the mixture and its components. These analyses aimed to achieve a detailed understanding of the underlying processes under controlled boundary conditions, thereby enabling a more precise interpretation of in situ observations. Various experiments were conducted on bentonite powder, pellets, and the mixture to evaluate permeability, water retention properties, free and constrained swelling, and hydration/loading-induced damage on pellets (Yang et al., 2022; 2023b,2024d; Molinero-Guerra et al., 2019a; 2020). Micro-scale characterizations were performed to examine the evolution of the microstructure of the mixture and its components during hydration. Subsequently, several modelling approaches were developed to quantify the effects of microstructural distribution and initial heterogeneities on the hydro-mechanical behaviour of the mixture during hydration (Mokni et al., 2020).

## **EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

More recently, bentonite-sand mixtures with high sand content have been considered as engineered barriers in several European countries, including France. Depending on the bentonite-to-sand ratio, the mixture's water and gas permeability, as well as its swelling pressure, may either increase or decrease. These properties are also influenced by alkaline plumes resulting from concrete degradation.

### Objectives

The objectives of this work are to evaluate:

- The pH impact on the swelling potential, compressibility and permeability of a reference sand/bentonite mixture with high sand content.
- The Impact of sand proportion on the HMC behaviour of the mixture (testing of alternative mixtures).
- Finally, the aging effect by characterization of bentonite sand mixture samples coming from Sealex in situ that run over 13 years (if samples are available).

### Plans

The investigated reference material will be a mixture of MX80 bentonite and sand with a proportion of 40/60 (in dry mass). However, several proportions will be investigated by increasing the sand proportion. Swelling tests under various mechanical boundary conditions will be carried out on the compacted mixtures hydrated with a typical Portland concrete porewater (obtained with KOH, NaOH and Ca(OH)<sub>2</sub>). Various chemical boundary conditions will be considered including progressive increase of pH, and chemical cycles. Additional testing boundary conditions might be considered. For example, the mixture might be hydrated initially with a synthetic pore water and then exposed to an alkaline solution at high pH. This kind of testing conditions, allows an easy identification of the involved processes. In parallel, a reference test will be performed with a synthetic pore water solution for comparison. Upon saturation, water permeability tests will be carried out.

Aged sand bentonite mixture (70/30 in dry mass) samples dismantled from SEALEX in situ test from the Tournemire URL running over 13 years will be characterized for comparison (if dismantling occur during EURAD-2). Six SEALEX in-situ experiments were installed between 2010 and 2013 aim to quantify the impact of intra-core geometry, specifically technological voids, on the hydraulic properties of sealing systems and test the long-term hydraulic performance of sealing systems under normal conditions using different clay core compositions (pure bentonite or bentonite/sand mixtures) and various conditionings (pre-compacted blocks or in-situ compacted). In depth characterization of the dismantled sample will allow to study bentonite core homogenization and the role of friction at large scale.

#### 6.3.10 Mitta

##### Summary of earlier studies

The hydro-mechanical behaviour of different bentonites has been studied with different tests which includes swelling pressure tests (Kivirana and Kumpulainen, 2011; Kiviranta et al. 2018; Martikainen and Schatz, 2011; Pintado et al. 2013a; Schatz and Martikainen, 2013). Tests indicates the strong dependency on the dry density of the swelling pressure and the hydraulic conductivity. The presence of salts in water might reduce the swelling pressure and increase the hydraulic conductivity. The combination of suction controlled oedometer tests and the study of the double porosity of bentonites shows different behaviour when the hydration affects only the microstructure and when the macro structure is also hydrated (Lloret et al. 2003; Pintado et al. 2009). Mineralogy and chemical analysis have been performed (Kivirana and Kumpulainen, 2011; Kiviranta et al. 2018; Kumpulainen and Kiviranta, 2010), showing different percentages of smectite in bentonites. Although the smectite is the active component, the swelling capacity of certain bentonite depends also on other parameters.

Water retention curve has been measured with the relative humidity technique in powder (Pintado et al. 2022) and under constant volume conditions (Kiviranta et al. 2018; Pintado et al. 2013a). The air entry value reduces when the dry density decreases. The combination of the relative humidity technique with the osmotic technique can help to obtain the full-range water retention curve although the difference in

hydration when it is performed with vapour (relative humidity technique) or with liquid water (osmotic technique) must be assessed.

Mock-up tests have been performed for studying the effect of the mechanical erosion (Bendito and Pintado, 2015; Martikainen et al. 2019; Pintado et al. 2013b). These tests are going to be used for studying the scale effect in swelling pressure tests.

Shearing tests have been performed with different devices in unsaturated conditions out (Pintado et al. 2019, 2023) and in saturated conditions (Kim et al. 2024). The salinity increases slightly the shear strength of Na-bentonites although the dry density is considered the main parameter. The shear strength is larger in unsaturated samples because of the suction effect.

### Objectives

The objectives are to study the scale effect testing with different sample sizes, to investigate the chemistry (salinity) on the swelling pressure and hydraulic conductivity of different bentonites, to investigate the friction effect with alternative methods (pistons in both sides of the cell measuring the radial pressure) and finally, to participate in the experimental benchmark for clarify drawbacks pointed out at the Strategic Research Agenda of EURAD.

### Plans

Oedometric tests at different scales will be performed to investigate the scale effect and to analyse the hydration process with saline and cementitious waters. Tests will be performed with bentonite mixtures.

Mock-up tests will be performed in fully instrumented steel cells (samples of 350 mm diameter x 800 mm length). Mixtures, proportions and type of waters will be agreed by comparing the different resources available in the ANCHORS partners laboratories. The proposal is to test granular filling material made with Georgian bentonite.

Part of the tests performed in oedometric conditions (constant volume and axial stress paths in water with different salinities and alkalinites) will be carried out as part of the experimental benchmarking as well. These tests are going to be used for models' validation.

Tests in cells with pistons in both sides measuring the axial pressure in each one and the radial pressure.

#### 6.3.11 TUBAF

##### Summary of earlier studies

The experimental setup, performed measurements, analysis procedures and results of the first mock-up experiment called HTV-1, performed 2005-2006, are available in Emmerich et al. (2009). The cell has an inner diameter of 80 cm and an inner length of 1.90 m. It is designed for fluid pressures up to 20 MPa and can be rotated by 90°, so that shaft and drift seals can be studied. Aside from water content / moisture / saturation, salt, swelling pressure and dry density distributions also mineralogical changes and microstructural effects were analysed in the experiments. Since HTV-1, the individual experiments have been varied to investigate the effect of changes in the hydration fluid's composition (4 M NaCl solution vs. Pearson Water), the segment heights, installation dry density, type of bentonite (Secursol variants, Calcigel), hydration conditions, and other parameters (Aurich et al 2022). To demonstrate the functional role of the equipotential segments, defects were deliberately installed in some of the experiments to create potential pathways for the fluid to bypass a particular sealing segment. In all experiments, the homogenized hydration of the sealing segments from the adjacent equipotential segments could be demonstrated. Conclusions to minimum installed dry densities and the related manufacturing targets for bentonite pellets could be established in relation to the required swelling pressures (Wieczorek et al. 2024; Aurich et al. 2022). More information on recent HTV runs is available in the form of the final report of the Sandwich-HP project (Wieczorek et al. 2024).

### Objectives

The objectives are:

- Providing well-controlled data for models at a larger spatial scale in the form of spatially resolved time series (pore pressures, swelling pressures, dry density, water content, displacement/strain). Mainly HM coupling will be investigated, although different salinity solutions could be used (linking the data to C coupling).
- Improving post-mortem characterization (micro-structure).
- Testing the set-up with other types of bentonites.

#### Plans

It is planned to extend the setup by special types of displacement sensors, since currently available measurements during the test (excluding post mortem) include fluid flow rates, pore water pressures, total stresses in axial and radial directions at various positions as well as TDR sensors. The signal of the latter is sensitive to both water-content and density changes. A better evaluation of the experiment requires a direct observation of the internal displacements of the individual segments as a result of inhomogeneous swelling and stiffness contrasts. It is planned to propose to other ANCHORS groups for participating in the post-mortem analyses and testing the bentonites that are going to be used in ANCHORS project.

### 6.3.12 UJV

#### Summary of earlier studies

UVJ was participating in European project focused on studies and application of bentonite, like FORGE, DOPAS, EURAD HITEC, EURAD GAS such as national project supported by Ministry of trade and Industry (Trpkošová et al., 2013), Technology Agency of the Czech Republic (Večerník, 2018; Černá et al., 2022) or SÚRAO (Hofmanová et al., 2019; Šachlová et al., 2022; Večerník et al., 2022; Svoboda et al., 2024). Performed work was focused mainly on bentonite materials characterisation (chemical, physical and mineralogical properties), interaction with other components of DGR and testing of radionuclide interactions and migration.

#### Objectives

In cooperation with Czech technical University and Charles University propose the design and testing of bentonite-sand mixture as an alternative material for DGR backfill.

Study and understand saturated bentonite-sand interactions and their effects on chemical, mineralogical and transport properties of Ca-Mg bentonite and bentonite-sand mixture.

Study and understand the effect of bentonite pellets heterogeneity (existence of preferential pathway, bentonite "microstructural memory") on transport properties.

#### Plans

The plans are as follows:

- Design and testing of bentonite-sand mixture as an alternative material for DGR backfill.
- Saturation of compacted bentonite in standard diffusion cells (restriction of the maximum size of pellets).
- Upscaling of standard experimental cells to cover the maximum grain size of the pellets – mid(long)-term saturation (36-48m) of bentonite pellets/mixtures.
- Performing of mid(long)-term diffusion experiments.
- Determination of chemical, mineralogical and transport characteristics of bentonite / bentonite pellets / bentonite-sand mixtures.

### 6.3.13 University of Lorraine (LEMTA)

#### Summary of earlier studies

*Role of different suction components on swelling behaviour of compacted bentonites.* It was focused on the investigation of the development of swelling pressure in compacted bentonites during wetting under confined conditions (Devineau et al., 2006; Leroy et al., 2025; Massat et al., 2016; Yigzaw et al.,

2016). The main swelling pressure testing program has been performed using a new constant-volume oedometer especially designed to apply suction using the osmotic method. The device allows to continue to determine swelling pressure under constant-volume conditions during wetting close to saturation (suction below 8.5 MPa). Different wetting modes, water or vapor wetting, resulted different equilibrium swelling pressure values. Three stages of swelling pressure development were identified during suction reduction. In the first stage, following large suction reduction, swelling pressure increases up to a peak value; then, in second stage, in suction range comprised between 8.5 and 3.5 MPa, swelling pressure tends to decrease to a minimum value; and finally, upon further suction reduction to low values, swelling pressure increases again and eventually stabilizes.

*Impact of Hydro-mechanical behaviour of claystone-based backfill materials under geo-environmental conditions.* The main purpose of this study was to analyze how variations in geo-environmental conditions affect the performance of potential claystone-based backfill materials, in particular their volume change and hydraulic conductivity behaviour (Middelhoff et al., 2021, 2020). The hydro-mechanical behaviour of processed COX-claystone spoil and its mixture with MX80-bentonite were analyzed by means of constant-volume swelling pressure, free-swell potential, one-dimensional compression/ oedometer and constant-head hydraulic conductivity experiments.

*Impact of high pH fluid circulation on long term hydromechanical behaviour and microstructure of compacted clay from the laboratory of Meuse-Haute Marne (France).* The main object of the study was to depict couplings between the hydromechanical behaviour of compacted clayey soil and the mineralogical and microstructural transformations induced by high pH water (Cuisinier et al., 2014; Kaddouri et al., 2019; Middelhoff et al., 2023). Compacted MHM-clay samples were subjected to the circulation of high-pH water or natural site water over an 18-month period. The hydromechanical properties, microstructural characteristics and some physicochemical properties of the samples were then analysed (i.e., shear strength, swelling properties, retention curve, mineralogy and microstructure) showing the impact of the expansive clay fraction on the reactivity of the mixture.

### Objectives

The objectives are to study the swelling pressure evolution of compacted bentonites under controlled suction close to saturation (matric suction lower than 2 MPa) and to highlight the combined impact of ionic strength and matric suction on the swelling pressure development of sand/bentonite/crushed argillite.

Finally, to contribute to the benchmark experiments that will be undertaken in the framework of EURAD-2.

### Plans

In this context, the study will be focused on the relationship between swelling pressure development and suction. We will use different experimental systems, including suction-controlled oedometer cell with suction control based on the osmotic method to specifically investigate the swelling pressure development in the low-suction range. We will use homo-ionic water solutions with two ionic strengths and a polar organic solvent were considered to analyse the relative impact of osmotic and crystalline swellings on the swelling pressure development upon wetting. Emphasis was put on the development of swelling pressure in compacted bentonites under suction control and under constant-volume conditions. The material that will be used in this project are:

- Wyoming MX-80 bentonite;
- Sand;
- Crushed callovo-oxfordian rock.

It is going to contribute to the benchmarks that will be organised throughout the EURAD-2 project, using the bentonite that will be selected in the project for this purpose.

### 6.3.14 University of Strathclyde

#### Summary of earlier studies

Water retention curves for MX-80 samples at constant density (1.8 g/cc) were performed on samples with DI water.

Sedimentation tests on bentonite samples (MX-80 and Ca-bentonite) were conducted at different NaCl Molarities (ranging from 0.001M to 5M) (Pedrotti, 2016). Following sedimentation, water NaCl concentration was increased stepwise to 5M and then decreased back. On MX-80 samples increase in NaCl concentration and dilution showed a reversible volumetric behaviour. On Ca-bentonite the same chemical path after sedimentation showed an increase in swelling upon dilution, probably as a consequence of cation exchange.

Similar mechanical path was also tests on samples previously compacted at 1.8 g/cc and then hydrated in free swelling conditions. Results were similar to what reported with the sedimented samples.

Liquid limits of different bentonites exposed to different NaCl concentration were measured and showed that the liquid limit reduces as NaCl concentration increases (Pedrotti, 2016).

1-D consolidation tests in oedometer cells were performed on MX-80 samples starting from a moisture content of 300% and 200% (Pedrotti and Tarantino, 2018).

#### Objectives

Understanding the macroscopic behaviour of different bentonite upon different hydro-chemo-mechanical paths.

#### Plans

Experiment 1: Liquid limit: this is a standard geotechnical index. The experimental procedure is described in detail by British Standard. Pore fluid will be modified following the experimental procedure already developed.

Experiment 2: Consolidation from slurry state at different salinity. Different chemical path at different vertical stress will be also explored. Relevant work on the topic was carried out by Di Maio (1996).

Experiment 3: Sedimentation test of samples prepared at different salinity. Subsequent dilution or further increase in the concentration will test for the final volume upon different chemical paths.

Experiment 4: Wetting tests at zero confinement (sample 25 mm in diameter) will be carried out as preliminary tests with different pore water chemistry.

Experiment 5: Water retention curves of samples at constant void ratio will be measured for Na and Ca bentonites prepared with pore water at different salinity

Experiment 6: Series of bench mock experiments where bentonite swelling will be tested when in contact with halite rock will be performed. Water chemistry will be then replicated and used to create control samples.

Experiment 7: Constant vertical stress wetting and subsequent consolidation ton 2.2 MPa will be performed at varying pore water chemistry and vertical stress.

Experiment 8: Constant volume wetting test (sample size to be confirmed). Wetting fluid will have different salinity. Samples will be compacted at different density and initial water content. Fluid circulation pressure to be decided.

### 6.3.15 VTT

#### Summary of earlier studies

Bentonite volumetric compressibility and triaxial shear test procedures with complete sample history control have been developed in earlier projects (e.g. Graham et al. 2023, Svensson et al. 2023).

## **EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Bentonite volumetric compressibility experiments coupled to design of experiments methods and advanced data processing have been performed (Forsman, 2025). Similar but more extended work including also triaxial shear tests is on-going.

Earlier studies using the same cylinder are reported in Holt et al. (2011) and Holt et al. (2013). In these studies, instead of granular bentonite, small-scale bentonite buffer blocks and different forms of bentonite as a gap filling material were used. The purpose was to study whether the buffer bentonite could be artificially wetted after installation in order to speed up the time required for the gap between the buffer bentonite and the rock surface of the deposition hole to seal.

### Objectives

The objective of the volumetric compaction and triaxial shear tests is to produce experimental information of the elastoplastic behaviour of the recently suggested bentonite types and forms that is needed for developing models for such bentonites.

The objective of the mock-up tests is to study behaviour of granular bentonite during wetting. In particular, saturation phenomena, including homogeneity, and development of swelling pressure are of interest.

### Plans

The volumetric compression experiments starting from bentonite powder are performed using a self-built 100 MPa volumetric compression apparatus. The triaxial shear tests are performed for 50 mm diameter x 100 mm height samples (machined from pre-samples got from the volumetric compression tests) using GDS Instruments triaxial apparatus with 250 kN axial load capacity and 32 MPa triaxial cell. The material and condition combinations are based on the Experimental Designs created in Task 4.1.

The mock-up tests planned by VTT can be described as wetting experiments of granular bentonite. It is important to study how efficiently and homogeneously the granular form of bentonite saturates and its swelling pressure develops. The mock-up tests will be carried out using larger test cells than normally in laboratory conditions, providing information also on the scaling effects.

The mock-up experiments are carried out in a steel cylinder with a height of 380 mm and an internal diameter of 360 mm. Different materials and chemical compositions of water are used for the tests. In all tests, a granular form of bentonite is used. The monitoring setup includes continuous radial pressure measurements (6 sensors at different points on the cylinder wall) and electrical resistivity tomography (ERT) measurements at regular intervals (e.g. every week).

## **6.4 Assessment of measures for better quality control and testing of bentonite**

The waste management organizations have varying readiness for quality control (QC) of bentonite for use in final disposal. Posiva is already following a quality control plan for the material that will be used in the first deposition tunnel and deposition holes. Other organizations have general idea of likely needs for the quality control.

There are different approaches to the quality control. Commercially available bentonite can be ordered as-is with the analyses and analysis reports from the supplier or a more comprehensive quality control plan can be agreed on with the supplier. Such plan could have analyses from 3<sup>rd</sup> party or the waste management organization for the raw bentonite or bentonite product.

Examples of quality control analyses relevant to bentonite are mineralogical composition, chemical composition, cation exchange capacity, exchangeable cations, amount of carbon and sulfur, water content, swelling pressure and hydraulic conductivity. Some of the analyses are lengthy and some may use custom-built equipment.

A questionnaire was sent to the Waste Management Organizations to identify the tests that could be performed for material testing and QC. The tests were divided in two categories, the first one related to

the characterization of a new material (bentonite or mixture of bentonite with crushed rock or sand). These tests are performed when a new bentonite or material mixture is planned to be used as a sealing material in any of the engineered barriers. The second category is related to the QC of the material batches during the operational phase, meaning that testing is done for material that the Waste Management Organization receives. In this case testing can be composed of tests for raw material or ready processed bentonite, mixture or components. Finally, QC tests can take place also for emplaced components. Summarizing, the detailed testing phases include:

- Material (bentonite mine) selection
- Characterization for long term safety assessments
- QC testing of “raw material” for manufacturing
- QC of ready material / components
- QC of emplaced material / components

The QC tests will be compared in an attempt to have a more unified way to do quality control for bentonite. Moreover, possibilities to speed up slower analyses by alternate method, changes to equipment or changes to test methodology will be discussed by the waste management organizations.

A wide range of tests for QC have been identified and are related to:

- Mineralogical composition (Butler et al. 2021; Svensson et al. 2019; SS-EN 12698-2:2007)
- Chemical composition (SFS-EN 16170:2016, ISO 11466:1995, SFS-EN 16171:2016, SS-EN ISO 13196:2015, SS-EN ISO 12677:2011, Svensson et al. 2019)
- Amount of organic carbon (LECO-method (ISO 10694:1995), DIN EN 15936:2022, DIN 19539:2016)
- Amount of sulphur (LECO-method (ISO 15178:2000) or ISO 15350:2000, SM 4500-S2-D method)
- Amount of sulphide (LECO-method (ISO 15178:2000) or ISO 15350:2000, Rodrigues and Iemma, 2015; Box et al. 2015)
- Required dry density to prevent microbial activity (Bengtsson et al 2017)
- Cation exchange capacity (Kiviranta et al. 2018; Meier and Kahr, 1999; Ammann et al. 2005)
- Exchangeable cations (Kiviranta et al. 2018; Polubesova et al. 1997; Jackson, 1975; Sawhney, 1970)
- Uniaxial compression test / unconfined compressive strength (ASTM D2166-06/D2166M-16; Svensson et al. 2019; Dueck et al. 2022b)
- Triaxial tests and direct shear tests (ASTM D7012-14e1; ASTM D4767-11; ASTM D7181-20; ASTM D3080)
- Macroscopic particle size distribution of material/mixture (ISO 17892-4:2016; Svensson et al. 2017)
- Water content (CEN ISO/TS 17892-1:2014; DIN EN 14346)
- Atterberg limits (ASTM D4318-17e1; EN ISO 17892-12)
- Retention capacity (water retention curve) (Pintado et al. 2022)
- Thermal conductivity (ASTM D5334-22a)
- Swelling index (ASTM D 5890-19)
- Swelling capacity (ASTM D 4546-03)
- Swelling pressure (ASTM D 4546-21; Karnland et al. 2006; Svensson et al. 2019)
- Hydraulic conductivity (ASTM D 5084-16a; Karnland et al. 2006; Svensson et al. 2019)
- Grain density / density of solid particles (Karnland and Birgersson, 2006; Kiviranta and Kumpulainen, 2011; Kiviranta et al. 2018)
- Soluble salts (dissolved inorganic solutes)
- Specific surface area (Keeling et al. 1980)
- Vapour sorption isotherms and specific surface area (external) (ISO 9277:2010 / ISO 9277:2022; Brunauer et al. 1938; Newman, 1983)
- Pore size distribution (ISO 15901-1:2016)
- Beidellite content (Kiviranta and Kumpulainen, 2011; Kiviranta et al. 2018)
- Stability versus colloid formation and erosion. No standard method available.
- Gas permeability. Breakthrough testing.

- Homogeneity, void ratio, degree of compaction, installed density, location of blocks.

Some test methods have been identified. In some cases, there are standards available from ASTM, from EU countries and others that are standards in the EU (EN standards).

The research of other methods will continue. The current plan is:

- Milestone MS72 (month 18): Quality control stage. Report on state of knowledge.
- Deliverable D10.2 (month 54). Report on assessment of measures for better quality control.

## 7. Bentonite Barrier modelling and Performance assessment

### 7.1 Enhancement of existing constitutive models and numerical tools

This description and development plans of material models are briefly described in this section. The focus is on the description and development of material models concerning thermo-hydro-mechanical-geochemical (THMC) coupled processes. For each team, the formulation and model are summarised and a development plan is proposed.

In the model descriptions, which are specific for bentonite materials, special attention is given to mechanical model characteristics, such as elasto-plasticity and double-structure consideration. Porosity, strain, temperature and saturation-dependent model parameters, including the water retention curve, water and gas permeabilities, and thermal conductivity are considered. Geochemical formulation coupled to THM can be considered. An additional feature of formulations is the consideration of heterogeneity.

The reported definitions of material models and the development plan include:

**AMPHOS21 – Extended Barcelona Basic Model:** The presented model considers the elastic regime of the Barcelona Basic Model (BBM), combined with geo-chemical coupling under isothermal conditions. An osmotic suction term was introduced to reproduce the impact of water salinity. The development plan includes refining geo-chemical coupling, implementing a double-porosity approach, extending the model to the plastic regime, and incorporating thermal processes.

**BGE, TUBAF, UFZ, CTU, CU – THM Model – Hypo-plasticity:** The teams adopt a coupled THM model developed by CU based on hypo-plasticity principles, combined with the double-structure concept. The current model does not include geo-chemical coupling. The development plan involves evaluating the model's performance for bentonite mixtures and implementing necessary improvements in the constitutive model based on simulations of laboratory test results. The model does not account yet for geochemical effects, but first steps towards a geochemical coupling will be explored during the project.

**BGR - Double Structure Model:** The DSM model is based on coupled Richards flow in deforming porous media. It does not incorporate thermal or geo-chemical coupling. The development plan includes reformulating the governing equation and constitutive models, with thermodynamic consistency as a potential enhancement.

**CIMNE-UPC – Barcelona Basic and Expansive Models:** A fully coupled THMC-G elasto-plastic model (BBM) is presented, considering porosity- and strain-dependent flow parameters. The BExM model incorporates double-structure behaviour. The development plan includes improving geo-chemical coupling in BBM, implementing geo-chemical coupling in BExM, introducing alkalinity into the model formulation, and application of heterogeneity to Bentonite materials.

**Clay Technology – Hysteresis Based Material Model:** A hydro-mechanical model with double-structure behaviour was implemented. The current model lacks thermal formulations. The development plan includes incorporating salinity effects, thermal formulations, and enhancing micro-macro coupling.

## **EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

EPFL – BEACON Model: A fully coupled THM-C model is considered. The development plan focuses on adapting the model for mixture materials (double-structure) and revising the water retention curve equations.

Imperial College – Double Structure Model: The presented model is a fully coupled thermo-hydro-mechanical model and considers double-structure behaviour. The development plan includes extending the model to incorporate chemistry (C) and gas (G) interactions (THMC-G).

IGN – Thermodynamically Consistent Model: The thermodynamically consistent, is a fully coupled poro-elastoplastic model designed for swelling clays. The current model does not include gas transport or geo-chemical coupling. The development plan includes distinguishing between structurally bound and free water, particularly for mixtures and granular materials.

LEI – BEACON Model: This hydro-mechanical model describes water flow through unsaturated material under constant gas pressure, using elastic deformation to represent wetting-induced swelling. Geo-chemical coupling is not currently included. The development plan includes incorporating plastic strains and geo-chemical coupling.

PSI: It is planned to enhance and extend thermo-hydraulic (TH) model coupling by integrating a mechanical component, thereby improving the applicability and accuracy of the numerical models. Geochemical coupling is already implemented and available.

UBERN – ORCHESTRA: The Orchestra model is a geochemical reactive transport code that does not include mechanical processes, except for the impact of mineral reactions on aqueous species transport. The development plan includes extending mechanical processes.

UCLM – XMm: The mechanical model XMm considers three structure levels: the megastructure (X), macrostructure (M), and microstructure (m). It is a fully coupled THMC-G model. The development plan includes better conceptualization of chemo-mechanical couplings, implementation of spatial heterogeneity, and improvements in HM-C models.

ULIEGE, TU-DELFT: The mechanical model, based on a modified BBM, is a fully coupled THM-G model. It does not currently consider geo-chemical coupling. The planned developments focus on integrating chemical effects of pore water onto the THM-G part.

VTT - Varied Multiplicative Processes Model: The developed VMP model is a large deformation chemo-elastoplastic model for partially saturated swelling clays. Improvement consists on the improvement of the model parametrisation (function forms and the model parameters) based as directly on experimental results as possible. The aim is especially to extent the model to various bentonite types and forms (e.g. granules) as wells chemical conditions.

The details of the contents briefly described here will be presented in Milestone MS34 entitled “Bentonite Constitutive Modelling Report: Model Description and Development” A reference section will be included in that report.

## **7.2 Application to assessment cases**

A repository system will evolve over time. Future states will depend on

- the initial state,
- internal processes, i.e. a number of radiations related, thermal, hydraulic, mechanical, chemical and biological processes acting internally in the repository system over time, and
- external factors acting on the system.

Internal processes are e.g. the decay of radioactive material, leading to the release of heat and the subsequent warming of the fuel, the engineered barriers and the host rock. Groundwater movements and chemical processes affecting the engineered barriers and the composition of groundwater are other examples. External factors include effects of future climate and climate-related processes, such as glaciations and land uplift and erosion.

The interactions of these interconnected factors can never be fully described or understood. As a result, various uncertainties affect all aspects of repository evolution and, consequently, the assessment of its safety. Therefore, an essential part of any safety assessment methodology is the systematic management of these uncertainties. This involves identifying, classifying, and describing them, as well as ensuring they are consistently addressed in the evaluation of repository evolution and its radiological consequences.

To be able to describe the repository evolution and to assess the importance of the uncertainties in a safety case as set of numerical tools are needed. These should be able to describe how the internal processes and the external conditions affects the properties of the barriers.

The objective of ANCHORS in this respect is to enhance confidence in numerical tools for safety case applications by focusing on large-scale bentonite barriers components. This includes investigating the sensitivity of parameters in the long-term evolution of bentonite barriers, its quantification and propagation of uncertainties carrying out cases that will evolve during the project. The definition of the cases will consider the concerns of WMOs, the ability of partners to model them, and will be based on a logic of increasing complexity. More coupling between the main processes is to be considered. The role of wall friction and its influence on the movement of bentonite components, or of heterogeneities on the bentonite evolutions related to fulfilling the functions expected of these components will also be examined through these test cases. One of the main challenges will be to consider the influence of the chemical composition of the water on the HM evolution of the bentonite components. The objective is to identify the most important parameters and the processes or coupling that have the most significant influence on the bentonite components behaviour firstly during the saturation/swelling phase, and then by considering the long-term evolution. The expected outcome of the simulations is the prediction of the evolution of the bentonite barriers over the course of the repository evolution for the different assessment cases.

The concept of studying assessment cases originates from the Horizon 2020 project BEACON, where three distinct cases were examined. The results from this study are documented in BEACON Deliverable D1.3 Final assessment report. The assessment cases that were modelled in Beacon was selected already in the proposal for the project. At that time, it was not clear how much work that would be needed to handle these cases. In hindsight, it was clear that the number and complexity of the modelling tasks in Beacon, including the assessment case benchmarks, had been over-ambitious. The modelling teams have been able to produce results for all tasks, but not enough time was assigned to the evaluation and interpretation of the results.

## 8. Summary

The EURAD-2 Deliverable 10.1 provides an initial state-of-the-art review on the hydraulic, mechanical, and chemical (HMC) evolution of bentonite, aimed at optimizing its use in engineered barrier systems (EBS) for deep geological repositories (DGR) of radioactive waste. The report is part of the ANCHORS Work Package, focusing on research to enhance the performance and resilience of bentonite barriers.

### Key Objectives

- Investigate HMC Behaviour: Study the impact of alkaline and saline conditions on the hydro-mechanical properties of bentonite, including swelling capacity and permeability.
- Laboratory and Modelling Studies: Evaluate bentonite performance at different scales, incorporating experimental and numerical models for long-term assessments.
- Barrier Optimization: Improve quality control, address heterogeneity in bentonite barriers, and enhance constitutive models used in performance assessments.
- Material Characterization: Develop a comprehensive database of THMC(G) properties for various bentonite types and mixtures.

### **Strategic Research Agenda & Examples of European National Waste Management Programs**

The EURAD Strategic Research Agenda (SRA) provides an integrated approach to research, development, and knowledge management for optimizing radioactive waste disposal, particularly focusing on engineered barrier systems within deep geological repositories. Key areas of research include improving bentonite/clay-based buffer materials, optimizing mixed-clay backfills to enhance repository stability, and advancing plug and seal technologies to ensure long-term safety and performance. These efforts aim to enhance scientific understanding, reduce uncertainties in simulations, and refine disposal designs to support the safe, industrial-scale implementation of geological waste repositories.

Bentonite is a key material in national waste disposal programs across Europe. Each National program may have specific applications, such as:

- Finland, Sweden and Czech Republic: Concepts for spent fuel repositories in crystalline rock, using bentonite as buffer and backfill.
- France & Germany: Sealing and backfill materials in repositories with clay host rock.
- Spain & Switzerland: Bentonite for waste isolation and minimizing microbial activity.
- UK: Research on bentonite's performance in high-salinity environments.

### **Experimental and Modelling Approaches**

The THMC(G) laboratory investigation is based on previous knowledge gained. WP ANCHORS will further develop the topic by evaluating long-term experiments and investigating systems with aggregates and chemical loading. A variety of materials will be investigated using previously developed and new methods. Another objective is to enhance confidence in numerical tools for safety case applications by focusing on large-scale bentonite barriers components. This includes investigating the sensitivity of parameters in the long-term evolution of bentonite barriers, its quantification and propagation of uncertainties carrying out cases that will evolve during the project

### **Key Challenges and Planned Research**

- Heterogeneity & Friction Effects: Addressing inconsistencies in bentonite density and swelling behaviour.
- Chemical Stability: Studying long-term effects of alkaline plumes and saline conditions.
- Upscaling Laboratory Results: Bridging the gap between lab-scale experiments and full-scale repository conditions.
- Model Improvements: Enhancing THMC(G) models for reliable long-term predictions.

The research under ANCHORS is crucial for optimizing bentonite barrier designs, ensuring safe, long-term containment of radioactive waste, and addressing uncertainties in repository performance assessment.

## References

Agboli, M., Grgic, D., Moumni, M., Giraud, A. 2024. Study under X-ray tomography of the impact of self-sealing process on the permeability of the Callovo-Oxfordian claystone. *Rock mechanics and rock engineering* 57: 4213-4229. <https://doi.org/10.1007/s00603-023-03350-y>

Agnel, M. 2021. Experimental protocol for the synthesis of poral water in the Callovo-Oxfordian claystone at 25°C. Andra technical report CGRPASTR210001. France.

Åkesson, M., Olsson, S., Dueck, A., Nilsson, U., Karnland, O., Kiviranta, L., Kumpulainen, S., Lindén, J., 2012. Temperature Buffer test. Hydro-mechanical and chemical/mineralogical characterizations. SKB Report P-12-06.

Alcoverro, J., Olivella, S., Alonso, E.E. 2014. Modelling fluid flow in Opalinus Clay excavation damage zone. A semi-analytical approach. Geological Society, London, Special Publications 415: 143 - 166. <https://doi.org/10.1144/SP415.7>

Alonso, M.C., García Calvo, J.L., Cuevas, J., Turrero, M.J., Fernández, R., Torres, E., Ruiz, A.I. 2017a. Interaction process at the concrete-bentonite interface after 13 years of FEBEX-plug operation. Part I: Concrete alteration. *Physics and Chemistry of the Earth, Parts A/B/C* 99: 38-48.

Alonso, M.C., García Calvo, J.L., Cuevas, J., Turrero, M.J., Fernández, R., Torres, E., Ruiz, A.I. 2017b. Interaction process at the concrete-bentonite interface after 13 years of FEBEX-plug operation. Part II: Bentonite contact. *Physics and Chemistry of the Earth, Parts A/B/C* 99: 49-63.

Ammann, L., Bergaya, F., Lagaly, G. 2005. Determination of the cation exchange capacity of clays with copper complexes revisited. *Clay Minerals* 40: 441–453.

ASTM D2166-06/D2166M-16. Standard Test Method for Unconfined Compressive Strength of Cohesive Soil.

ASTM D3080. Standard Test Method for Direct Shear Test of Soils Under Consolidated Drained Conditions.

ASTM D4318-17e1. Standard Test Methods for Liquid Limit, Plastic Limit, and Plasticity Index of Soils.

ASTM D 4546-03. Method A, Standard Test Methods for One-Dimensional Swell or Settlement Potential of Cohesive Soils.

ASTM D5334-22a. Standard Test Method for Determination of Thermal Conductivity of Soil and Rock by Thermal Needle Probe Procedure.

ASTM D 4546-21. Standard Test Methods for One-Dimensional Swell or Collapse of Soils.

ASTM D4767-11. Standard Test Method for Consolidated Undrained Triaxial Compression Test for Cohesive Soils.

ASTM D 5084-16a. Standard Test Methods for Measurement of Hydraulic Conductivity of Saturated Porous Materials Using a Flexible Wall Permeameter.

ASTM D 5890-19. Standard Test Method for Swell Index of Clay Mineral Component of Geosynthetic Clay Liners.

ASTM D7012-14e1. Standard Test Methods for Compressive Strength and Elastic Moduli of Intact Rock Core Specimens under Varying States of Stress and Temperatures.

ASTM D7181-20. Standard Test Method for Consolidated Drained Triaxial Compression Test for Soils.

Atanassov, G., Goranov A. 1961/1962. Bentonitic clays from the region of the town of Kardzhali. Ann. de L'Univ. de Sofia, 56, 2-Geologie, 149-188. (in Bulgarian).

Atanassov, G., Stefanov D. Goranov A., 1963/64. Additional data on the mineralogy of bentonite clays from Kardzhali. Ann. de L'Univ. de Sofia, 58, 1-Geologie, 135-143. (in Bulgarian).

Atanassov, G., Goranov, A. 1986. Bentonites in Bulgaria. Ann. de L'Univ. de Sofia, 80, 1-Geologie, 226-237. (in Bulgarian).

Aurich, J., Gruner, M., Kudla, W., Hofmann, M., Königer, F., Schuhmann, R., & Emmerich, K. (2022). Technikumsversuche zu Bentonitdichtungen mit Äquipotenzialsegmenten. Bergbau, 6.

Barbour, S.L., Yang, N. 1993. A review of the influence of clay-brine interactions on the geotechnical properties of Ca-montmorillonitic clayey soils from western Canada. Canadian Geotechnical Journal, 30(6): 920–934. <https://doi.org/10.1139/t93-090>

Barakat, Y., Cui, Y.-J., Mokni, N., Delage, P., Bernier, F. 2022. Effects of pH and exposure time to alkaline solutions on the mineralogy of the Opalinus Clay from the lower sandy facies of Mont Terri site. Engineering Geology 306: 106766.

Bass, M., Freger, V., 2008. Hydration of Nafion and Dowex in liquid and vapor environment: Schroeder's paradox and microstructure. Polymer 49: 497–506. <https://doi.org/10.1016/j.polymer.2007.11.054>

Beers, K.M., Yakovlev, S., Jackson, A., Wang, X., Hexemer, A., Downing, K.H., Balsara, N.P., 2014. Absence of Schroeder's Paradox in a Nanostructured Block Copolymer Electrolyte Membrane. Journal of Physical Chemistry B 118: 6785–6791. <https://doi.org/10.1021/jp501374r>

Bendito, E., Pintado, X. 2015. Monitoring of swelling pressure in bentonite. Environmental Geotechnics. 3(5): 334-345. <https://doi.org/10.1680/envgeo.14.00007>

Bengtsson, A., Blom, A., Hallbeck, B., Heed, C., Johansson, L., Stahlén, J., Pedersen, K. 2017. Microbial sulphide-producing activity in water saturated MX-80, Asha and Calcigel bentonite at wet densities from 1 500 to 2 000 kg m<sup>3</sup>. SKB report TR-16-09. Solna, Sweden.

Bennett, D.P. Cuss, R.J., Vardon, P.J., Harrington, J.F., Sedighi, M., Thomas, H.R. 2015. Exploratory data analysis of the Large Scale Gas Injection Test (Lasgit) dataset, focusing on 'second-order' events around macro-scale gas flows. Geological Society, London, Special Publications 415: 225 - 239. <https://doi.org/10.1144/SP415.14>

Bernachy-Barbe, F., Bosch, J.A., Campos, G., Carbonell, B., Daniels, K.A., Ferrari, A., Graham, C.G., Guillot, W., Gutiérrez-Álvarez, C., Harrington, J.F., Iglesias, R.J., Kataja, M., Kröhn, K-P, Kröhn, M., Mašín, D., Najser, J., Pingel, J.L., Real, E., Schäfer, T., Svoboda, J., Sun, H., Tanttu, J., Villar, M.V., Wieczorek, K. 2022. Experimental work on bentonite evolution in the frame of BEACON – final report of WP4. Deliverable D4.3. BEACON EU Project.

BGE (2020). Sub-areas interim report pursuant to section 13 StandAG as per 28/09/2020. Technical report, Bundesgesellschaft für Endlagerung mbH, Peine

BGE (2022): Konzept zur Durchführung der repräsentativen vorläufigen Sicherheitsuntersuchungen gemäß Endlagersicherheitsuntersuchungsverordnung, Bundesgesellschaft für Endlagerung mbH,(BGE), Peine.

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

BGE (2024): Auslegungstemperaturen ab Schritt 2 Phase I des Standortauswahlverfahrens: Einordnung zum Umgang mit der Grenztemperatur. Grundlagenbericht, BGE-2024-GB-1, Bundesgesellschaft für Endlagerung mbH,(BGE), Peine.

Bian, X., Cui, Y. J., Li, X. Z. 2019. Voids effect on the swelling behaviour of compacted bentonite. *Géotechnique* 69(7): 593-605. <https://doi.org/10.1680/jgeot.17.p.283>

Birgersson, M., Börgesson, L., Hedström, M., Karnland, O., Nilsson, U., 2009. Bentonite erosion Final report. SKB TR-09-34. Stockholm, Sweden.

Birgersson, M., Goudarzi, R., 2013. Studies of vapor transport from buffer to tunnel backfill (Sauna effects) (R No. 13-42). SKB Report R-13-42. Stockholm, Sweden.

Börgesson L, Johanesson LE, Sandén T, Hernelind J. 1995. Modelling of the physical behaviour of water saturated clay barriers. Laboratory tests, material models and finite element application. SKB Technical report TR-95-20, Stockholm, Sweden. <https://skb.com/publication/12027>

Börgesson, L. 1986. Model shear tests of canisters with smectite clay envelopes in deposition holes. SKB technical report 86-26. Stockholm, Sweden. <https://skb.com/publication/3238>

Bosch, J., Ferrari, A., Laloui, L. 2021. Coupled hydro-mechanical analysis of compacted bentonite behaviour during hydration. *Computers and Geotechnics* 140: 104447.

Bozhinov, K., Bechev, D. 1979. Roentgenostructural and chemical characterization of inactivated and activated bentonite from the deposit near Kardzhali. *Rew. Bulg. Geol. Soc.*, 40, 2, 174-180. (in Bulgarian)

Bozhinov, K., Bechev, D., Zhelyazkova-Panayotova, M. 1984. On the mineral composition, properties and some modifying factors of the bentonite clays from the Kărdžaldali deposit. *Ann. de L'Univ. de Sofia*, 78, 1-Geologie, 145-173. (in Bulgarian).

Box, G. E. P.; Hunter, J. S.; Hunter, W. G. (2005): Statistics for Experimenters. Design, Innovation, and Discovery. 2. Aufl. Hoboken, NJ: John Wiley & Sons Inc. ISBN 978-0471-71813-0.

Breitner, D., Osan, J., Fabian, M., Zagvai, P., Szabo, C., Dahn, R., Marques Fernandes, M., Sajom I.E., Mathe, Z., Torok, S. 2015. Characteristics of uranium uptake of Buda Claystone Formation as the candidate host rock of high level radioactive waste repository in Hungary. *Environmental Earth Sciences* 73: 209.

Brindley, G.W., Brown, G. (Eds.). 1980. Crystal Structures of Clay Minerals and their X-ray Identification. Mineralogical Society.

Brunauer, S., Emmett, P.H., Teller, E. 1938. Adsorption of gases in multimolecular layers. *Journal of the American Chemical Society* 60: 309–319.

Butler, B., Hiller, S. 2021. Automated full-pattern summation of X-ray powder diffraction data for high-throughput quantification of clay-bearing mixtures. *Clays and Clay Minerals* 69(1): 38-51.

Castellanos, E., Villar , M.V., Romero, E., Lloret, A., Gens, A. 2008. Chemical impact on the hydro-mechanical behaviour of high-density FEBEX bentonite. *Physics and Chemistry of the Earth* 33: S516-S526. <https://doi.org/10.1016/j.pce.2008.10.056>

CEN ISO/TS 17892:1 2014. Geotechnical investigation and testing. Laboratory testing of soil. Part 1: Determination of water content.

Černá, K., Šachlová, Š., Bedrníková, E., Barták, D. S., Kašpar, V., Říha, J., Hlaváčková, V., Dobrev, D., Večerník, P., Zuna, M. 2022. Limitní faktory pro přežití a proliferaci mikrobiálních společenstev

významných pro korozí bariér hlubinného úložiště radioaktivních odpadů, Závěrečná zpráva projektu TAČR TK02010169

Chaaya, 2023. PhD. Couplage entre processus mécanique et chimique lors de l'hydratation d'une bentonite. Université d'Orléans, 235 p. France.

Chaaya, R., Gaboreau, S., Milet, F., Maubec N., Tremosa, J., Raimbourg, H., Ferrage, E. 2023. In-operando X-ray scattering characterization of smectite swelling experiments. *Applied Clay Sciences* 245(1): 107124. <https://doi.org/10.1016/j.clay.2023.107124>

Chen, G.J., Ledesma, A. 2009. Coupled thermohydromechanical modeling of the full-scale in situ test “Prototype repository”. *Journal of geotechnical and geoenvironmental engineering* 135 (1), 121-132.

Chen, Y.-G., Jia, L.-Y., Li, Q., Ye, W.-M., Cui, Y.-J., Chen, B. 2017. Swelling deformation of compacted GMZ bentonite experiencing chemical cycles of sodium-calcium exchange and salinization-desalinization effect. *Applied Clay Science*, 141: 55–63. <https://doi.org/10.1016/j.clay.2017.02.016>

Claret F, Dauzeres A, Jacques D, Sellin P, Cochebin B, De Windt L, Garibay-Rodriguez J, Govaerts J, Leupin O, Mon Lopez L, Montenegro V, Montoya V, Prasianakis N, Samper J, Talendier J (2022) Modelling of the long-term evolution and performance of engineered barrier system. *EPJ Nuclear Sci Technol* 8:41

Cui, Y. J., Tang, A. M., Loiseau, C., Delage, P. 2008. Determining the unsaturated hydraulic conductivity of a compacted sand–bentonite mixture under constant-volume and free-swell conditions. *Physics and Chemistry of the Earth, Parts A/B/C*, 33: S462-S471. <https://doi.org/10.1016/j.pce.2008.10.017>

Cui, Y. J., Tang, A. M. 2013. On the chemo-thermo-hydro-mechanical behaviour of geological and engineered barriers. *Journal of Rock Mechanics and Geotechnical Engineering*, 5(3): 169-178. <https://doi.org/10.1016/j.jrmge.2013.05.001>

Cui, Y. J. 2017. On the hydro-mechanical behaviour of MX80 bentonite-based materials. *Journal of Rock Mechanics and Geotechnical Engineering*, 9(3): 565-574. <https://doi.org/10.1016/j.jrmge.2016.09.003>

Cuisinier, O., Masrouri, F. 2004. Microstructure et comportement hydromécanique d'un sol gonflant non saturé. *Revue Française de Géotechnique* 47–56. <https://doi.org/10.1051/geotech/2004108047>

Cuisinier, O., Masrouri, F. 2005. Hydromechanical behaviour of a compacted swelling soil over a wide suction range. *Eng. Geol.*, *Issues in Nuclear Waste Isolation Research* 81: 204–212. <https://doi.org/10.1016/j.enggeo.2005.06.008>

Cuisinier, O., Masrouri, F., Pelletier, M., Villieras, F., Mosser-Ruck, R. 2008. Microstructure of a compacted soil submitted to an alkaline PLUME. *Applied Clay Science* 40: 159–170. <https://doi.org/10.1016/j.clay.2007.07.005>

Cuisinier, O., Deneele, D., Masrouri, F., Abdallah, A., Conil, N. 2014. Impact of high-pH fluid circulation on long term hydromechanical behaviour and microstructure of compacted clay from the laboratory of Meuse-Haute Marne (France). *Applied Clay Science* 88–89: 1–9. <https://doi.org/10.1016/j.clay.2013.12.008>

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Czompoly, O., Szabo, F., Fabian, M., Kolonits, T., Fogarassy, Zs, Zambo, D., Aertsens, M., Osan, J. 2024. Retention of Nickel and Cobalt in Boda Claystone Formation. *Minerals* 14: 1299.

Czompoly O, Fabian M, Koranyi TI, Nagy G, Horvath ZE, Zizak I, Pollastri S, Aertsens M, Osan J. 2023. Adsorption and diffusion of selenite on Boda Claystone Formation, *Applied Clay Science* 241: 106997. <https://doi.org/10.1016/j.clay.2023.106997>

Damiani, L., Coene, E., Nieves, A. (2020a). Reactive transport modelling of a low-pH concrete / clay interface. *Applied Geochemistry* 115: 104562. <https://doi.org/10.1016/j.apgeochem.2020.104562>

Daniels, K., Harrington, J., Milodowski, A., Kemp, S., Mounteney, I., Sellin, P. 2021. Gel Formation at the Front of Expanding Calcium Bentonites. *Minerals* 11(2): 215. <https://doi.org/10.3390/min11020215>

Davankov, V.A., Pastukhov, A.V. 2011. Paradoxes of Thermodynamics of Swelling Equilibria of Polymers in Liquids and Vapors. *Journal of Physical Chemistry B* 115: 15188–15195. <https://doi.org/10.1021/jp208209c>

Delage, P., Howat, M.D., Cui, Y.J. 1998. The relationship between suction and swelling properties in a heavily compacted unsaturated clay. *Engineering Geology* 50: 31–48. [https://doi.org/10.1016/S0013-7952\(97\)00083-5](https://doi.org/10.1016/S0013-7952(97)00083-5)

Delage, P., Romero, E., Tarantino, A. 2008. Recent developments in the techniques of controlling and measuring suction in unsaturated soils, in: *Unsaturated Soils. Advances in Geo-Engineering*. CRC Press.

Delage, P., Cui, Y.J., 2008. An evaluation of the osmotic method of controlling suction. *Geomechanics and Geoengineering* 3(1): 1–11. <https://doi.org/10.1080/17486020701868379>

Della Vecchia, G., Dieudonné, A.-C., Jommi, C., Charlier, R. 2014. Accounting for evolving pore size distribution in water retention models for compacted clays. *International Journal for Numerical and Analytical Methods in Geomechanics* 39(7): 702–723. <https://doi.org/10.1002/nag.2326>

Department for Business, Energy and Industrial Strategy, 2018, *Implementing Geological Disposal – Working with Communities*.

Desbois, G., Urai, J.L., Kukla, P.A. 2009. Morphology of the pore space in claystones - Evidence from BIB/FIB ion beam sectioning and cryo-SEM observations. *eEarth*, 4(1): 15–22. <https://doi.org/10.5194/ee-4-15-2009>

Devineau, K., Bihannic, I., Michot, L., Villiéras, F., Masrouri, F., Cuisinier, O., Fragneto, G., Michau, N. 2006. In situ neutron diffraction analysis of the influence of geometric confinement on crystalline swelling of montmorillonite. *Applied Clay Science* 31: 76–84. <https://doi.org/doi.org/10.1016/j.clay.2005.08.006>

Di Maio, C. 1996. Exposure of bentonite to salt solution: osmotic and mechanical effects. *Géotechnique*, 46(4): 695–707. <https://doi.org/10.1680/geot.1996.46.4.695>

DIN EN 14346. Characterization of waste - Calculation of dry matter by determination of dry residue or water content.

DIN EN 15936. Soil, waste, treated biowaste and sludge - Determination of total organic carbon (TOC) by dry combustion.

DIN 19539:2016-12. Investigation of solids - Temperature-dependent differentiation of total carbon.

Dueck, A. 2008. Laboratory results from hydro-mechanical tests on a water unsaturated bentonite. *Engineering Geology* 97: 15–24. <https://doi.org/10.1016/j.enggeo.2007.11.001>

Dueck, A. 2007. Results from Suction Controlled Laboratory Tests on Unsaturated Bentonite – Verification of a Model, in: Schanz, T. (Ed.), *Experimental Unsaturated Soil Mechanics*. Springer, Berlin, Heidelberg, pp. 329–335. [https://doi.org/10.1007/3-540-69873-6\\_33](https://doi.org/10.1007/3-540-69873-6_33)

Dueck A, Börgesson L, Johannesson LE. 2010. Stress-strain relation of bentonite at undrained shear. Laboratory tests to investigate the influence of material composition and test technique. SKB Technical report TR-10-32, Stockholm, Sweden. <https://skb.com/publication/2189444>

Dueck, A., Nilsson, U. 2010. Thermo-Hydro-Mechanical properties of MX-80. Results from advances laboratory tests. SKB report TR-10-55. Stockholm, Sweden. <https://skb.com/publication/2223073>

Dueck, A., Goudarzi, R., Börgesson, L., 2011. Buffer homogenisation, status report. SKB Technical Report TR-12-02. Stockholm, Sweden.

Dueck, A., Goudarzi, R., Börgesson, L., 2014. Buffer homogenisation, status report 2. SKB Technical Report TR-14-25. Stockholm, Sweden.

Dueck, A., Goudarzi, R., Börgesson, L., 2016. Buffer homogenisation Status report 3. SKB Technical Report TR-16-04. Stockholm, Sweden.

Dueck, A., Goudarzi, R., Börgesson, L. 2018. Buffer homogenization – status report 4. SKB technical report TR-17-04. Solna, Sweden.

Dueck, A., Börgesson, L., Kristensson, O., Malmberg, D., Åkesson, M., Hernelind, J., 2019. Bentonite homogenisation Laboratory study, model development and modelling of homogenisation processes. SKB Technical Report TR-19-11.

Dueck, A., Börgesson, L., 2021. Bentonite homogenisation Three studies based on laboratory test results. SKB Report P-21-05.

Dueck, A., Goudarzi, R., Jensen, V., Börgesson, L. 2022a. Buffer homogenisation - status report 5. SKB technical report TR-21-14. Solna, Sweden. <https://skb.com/publication/2492350/TR-17-04.pdf>

Dueck, A., Nilsson, N., Jensen, V., Börgesson, L. 2022b. Compressive strength of bentonites Factors influencing results from unconfined compression test. SKB technical report TR-21-13. Solna, Sweden.

Dutta, J., and Mishra, A.K. 2016. Consolidation behaviour of bentonites in the presence of salt solutions. *Applied Clay Science* 120: 61–69. <https://doi.org/10.1016/j.clay.2015.12.001>

ECOCLAY II (2005) Effect of cement on clay barrier performance. European Report EN 21921.

EndlSiAnfV: Endlagersicherheitsanforderungsverordnung vom 6. Oktober 2020 (BGBI. I S. 2094)

EndlSiUntV: Endlagersicherheitsuntersuchungsverordnung vom 6. Oktober 2020 (BGBI. I S. 2094, 2103)

Emmerich, K., Kemper, G., Königer, F., Schlaeger, S., Gruner, M., Gaßner, W., Hofmann, M., Nüesch, R., & Schuhmann, R. 2009. Saturation Kinetics of a Vertical Multilayer Hydraulic Sealing

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

System Exposed to Rock Salt Brine. Vadose Zone Journal 8(2): 332–342. <https://doi.org/10.2136/vzj2008.0094>

Emmerich, K., Schuhmann, R., Königer, F., Bohac, P., Delavernhe, L., Wieczorek, K., Hesser, J., Shao, H. 2019. Joint project: Vertical hydraulic sealing system based on the sandwich principle - preproject (Sandwich-VP): Joint final report: 01.07.2017-30.06.2019 (24 months). Karlsruhe, Germany. <https://doi.org/10.2314/KXP:1692488228>

Enresa. (2000). FEBEX project. Full-scale engineered barriers experiment for a deep geological repository for high level radioactive waste in crystalline host rock. Final report. Enresa technical publication 1/2000. Madrid, Spain.

EURAD Bureau. (2023): Update of the EURAD Strategic Research and Knowledge Management Agenda (SRA), of deliverable D1.9 of the HORIZON 2020 project EURAD. EC Grant agreement no: 847593.

Fabian, M., Tolnai, I., Kis, Z., Szilagyi, V. 2022. Characterization of Simulated Liquid Radioactive Waste in a New Type of Cement Mixture. ACS Omega 7: 36108–36116.

Fabian, M., Czompoly, O., Tolnai, I., De Windt, L. 2023. Interactions between C-steel and blended cement in concrete under radwaste repository conditions at 80°C, Scientific Reports 13: 15372.

Fernández, M.A., Melón, A., Sánchez, D.M., Galán, M.P., Morante, L., Gutiérrez-Nebot, L., Turrero, M.J., Escribano, A. 2008. Changes on the Mineralogical and Physical Properties of FEBEX Bentonite Due to Its Contact With Hyperalkaline Pore Fluids in Infiltration Tests. MRS Online Proceedings Library 1107, 483. <https://doi.org/10.1557/PROC-1107-483>

Finsterle, S., Kober, F., Vomvoris, S., Lanyon, B., Kowalsky, M.B. 2023. Data-worth analysis for the design of the HotBENT monitoring system. Geoenergy 1(1). <https://doi.org/10.1144/geoenergy2023-020>

Forsman, S. 2025. Experimentally based surrogate models for volumetric compressibility of partially saturated bentonite clay. Master's thesis. Aalto University. Espoo, Finland.

Fu, M.H., Zhang, Z.Z., Low, P.F. 1990. Changes in the Properties of a Montmorillonite-Water System during the Adsorption and Desorption of Water: Hysteresis. Clays and Clay Minerals 38: 485–492.

Gaboreau, S., Rodríguez-Cañas, E., Maeder, U., Jenni, A., Turrero, M.J., Cuevas, J. 2020. Concrete perturbation in a 13-year in situ concrete/bentonite interaction from FEBEX experiments. New insight of 2:1 Mg phyllosilicate precipitation at the interface. Applied Geochemistry 118: 104624.

García-Siñeriz, J.L., Villar, M.V., Rey, M., Palacios, B. (2015). Engineered barrier of bentonite pellets and compacted blocks: State after reaching saturation. Engineering Geology 192(18): 33-45. <https://doi.org/10.1016/j.enggeo.2015.04.002>

Gaus, I., Garitte, B., Senger, R., Gens, A., Vasconcelos, R., Garcia-Sineriz, J.L., Trick, T., Wieczorek, K., Czaikowski, O., Schuster, K., Mayor, J.C., Velasco, M., Kuhlmann, U., Villar, M.V. 2014. The HE-E Experiment: Lay-out, Interpretation and THM Modelling. Nagra technical report 14-53. Wettingen, Switzerland.

Gens, A., Alonso, E.E. 1992. A framework for the behaviour of unsaturated expansive clays. Canadian Geotechnical Journal 26(6): 881-889. <https://doi.org/10.1139/t92-120>

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Gens, A. (Ed.), Filippi, M., Vallejan, B., Weetjens, E., Van Geet, M., Volckaert, G., & Bastiaens, W. (2009a). RESEAL II PROJECT. Final report on modelling (WP4). (0 ed.) (SCK•CEN Reports; No. ER-80). SCK CEN.

Gens, A., Sánchez, M., Guimaraes, L. do. N., Alonso, E.E., Lloret, A., Olivella, S., Villar, M.V., Huertas, F. 2009b. A full-scale in situ heating test for high-level nuclear waste disposal: observations, analysis and interpretation. *Géotechnique*, 59(4): 377-399.

Gergely, F., Osan, J., Szabo, B.K., Torok, S. 2016. Analytical performance of a versatile laboratory microscopic X-ray fluorescence system for metal uptake studies on argillaceous rocks. *Spectrochimica Acta Part B* 116: 75

González-Blanco, L., Romero, E., Marschall, P., Levasseur, S. 2022. Hydro-mechanical response to gas transfer of deep argillaceous host rocks for radioactive waste disposal. *Rock Mechanics and Rock Engineering* 55: 1159-1177. <https://doi.org/10.1007/s00603-021-02717-3>

González-Santamaría, D.E., Fernández, R., Ruiz, A.I., Ortega, A., Cuevas, J. 2020. Bentonite/CEM-II cement mortar INTERFACE EXPERIMENTS: A proxy to in situ deep geological repository engineered barrier system surface reactivity. *Applied Geochemistry* 117: 104599. <https://doi.org/10.1016/j.apgeochem.2020.104599>

Goudarzi, R. 2023. Prototype Repository – Sensor data report Period 2001-09-17 to 2023-01-01. SKB technical report P-23-12. Solna, Sweden.

Gowrishankar, A., Jacops, E., Maes, N., Verboven, P., Janssen, H. 2023. An experimental methodology to assess the impact of desaturation on gas diffusion in clay based materials. Proceedings of the 8th International Conference on Unsaturated Soils (UNSAT 2023). Milos, Greece. <https://doi.org/10.1051/e3sconf/202338220001>

Graham, C., Daniels, K., Harrington, J., Chaaya, R., Gaboreau, S., Tremosa, J., Villar, M.V., Gutiérrez-Álvarez, C., García-Herrera, G., Iglesias, R., Gimeno, N., Svoboda, J., Černochová, K., Najser, J., Mašín, D., Yliharju, J., Sayenko, S., Pulkkanen, V-M, Rauhala, O-P, Vettese, G., Pakkanen, N., Siitari-Kauppi, M. 2024. HITEC technical report on Material characterisation. Deliverable 7.8: HITEC Eurad-1 Work Package.

Gramegna, L., Villar, M.V., Collin, F., Talandier, J., Charlier, R. 2023. Friction influence on constant volume saturation of bentonite mixed pellet-block samples, a numerical analysis. *Applied Clay Science* 234: 106846. <https://doi.org/10.1016/j.clay.2023.106846>

Gutiérrez-Rodrigo, V., Martín, P.L., Villar, M.V. 2021. Effect of interfaces on gas breakthrough pressure in compacted bentonite used as engineered barrier for radioactive waste disposal. *Process Safety and Environmental Protection* 149: 244–257.

Harjupatana, T., Alaraudanjoki, J., Kataja, M. 2015. X-ray tomographic method for measuring three-dimensional deformation and water content distribution in swelling clays. *Applied Clay Science* 114: 386-394. <https://doi.org/10.1016/j.clay.2015.06.016>

Harjupatana, T., Miettinen, A., Kataja, M. 2022. A method for measuring wetting and swelling of bentonite using X-ray imaging. *Applied Clay Science* 221: 106485. <https://doi.org/10.1016/j.clay.2022.106485>

Harrington, J., Daniels, K., Wiseall, A., Sellin, P. 2020. Bentonite homogenisation during the closure of void spaces. *International Journal of Rock Mechanics and Mining Sciences* 136: 104535.

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Hausmannová L., Dohnálková M., Matušková E., Lahodová Z., Augusta J. (2023): Technical Design of the Deep Geological Repository 2023, SÚRAO technical report no. TZ 711/2023/ENG,

Hausmannová L., Hanusová I., Dohnálková M. (2018): Summary of the research of czech bentonites for use in the deep geological repository – up to 2018, SÚRAO technical report no. TZ 309/2018/ENG, Prague.

Hedström, M., Birgersson, M., Nilsson, U., Karnland, O. 2011. Role of cation mixing in the sol formation of Ca/Na-montmorillonite. Physics and Chemistry of the Earth 36: 1564-1571. <https://doi.org/10.1016/j.pce.2011.07.019>

Hedström, M., Ekvy Hansen, E., Nilsson, U. 2016. Montmorillonite phase behaviour Relevance for buffer erosion in dilute groundwater. SKB TR-15-07. Stockholm, Sweden.

Hofmanová, E., Červinka, R., Vopálka, D., Baborová, L., Brázda, L., Pecková, A., Vetešník, A., Viglašová, E., Vašíček R. 2019. Transport of radionuclides from the repository / Input parametres and process models for evaluation of radionuclide transport through engineered barriers – Final report, SÚRAO TZ 420/2019/ENG

Holt, E., Marjavaara, P., Löija, M. 2011. Artificial Wetting of Buffer Material – Small Scale. Posiva Working Report 2011-53. Posiva Oy, Eurajoki, Finland.

Holt, E., Löija, M., Fortino, S., Marjavaara, P. 2014. Experimental Studies of Buffer Gap Filling with Artificial Wetting. Posiva Working Report 2013-52. Posiva Oy, Eurajoki, Finland.

Houben, M.E., Desbois, G. & Urai, J.L. 2013. Pore morphology and distribution in the Shaly facies of Opalinus Clay (Mont Terri, Switzerland): Insights from representative 2D BIB-SEM investigations on mm to nm scale. Applied Clay Science, 71: 82-97. <https://doi.org/10.1016/j.clay.2012.11.006>

Houben, M.E., Desbois, G., Urai, J.L. 2014. A comparative study of representative 2D microstructures in Shaly and Sandy facies of Opalinus Clay (Mont Terri, Switzerland) inferred form BIB-SEM and MIP methods. Marine and Petroleum Geology, 49: 143-161. <https://doi.org/10.1016/j.marpetgeo.2013.10.009>

Idiart, A., Laviña, M., Kosakowski, G., Cochebin, B., Meeussen, J.C.L., Samper, J., Mon, A., Montoya, V., Munier, I., Poonoosamy, J., Montenegro, L., Deissmann, G., Rohmen, S., Damiani, L.H., Coene, E., Nieves, A. 2020a. Reactive transport modelling of a low-pH concrete / clay interface. Applied Geochemistry 115. <http://doi.org/10.1016/J.APGEOCHEM.2020.104562>

Idiart, A., Laviña, M., Pasteau, A., Cochebin, B. 2020b. Hydro-chemo-mechanical modelling of long-term evolution of bentonite swelling. Applied Clay Science 195: 105717. <https://doi.org/10.1016/j.clay.2020.105717>

Ilieva, Al. 2006. Crystal chemistry and structural properties of natural and modified montmorillonite from Bulgarian bentonite deposits. 2006. Central Laboratory of Mineralogy and Crystallography – BAS, ThD Thesis, 127 p. (in Bulgarian)

ISO 9277:2010 / ISO 9277:2022 Determination of the specific surface area of solids by gas adsorption — BET method.

ISO 11466:1995. Soil quality – Extraction of trace elements soluble in aqua regia.

ISO 10694. Soil quality – Determination of organic and total carbon after dry combustion (elementary analysis).

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

ISO 15178:2000. Soil quality – Determination of total sulphur by dry combustion.

ISO 15350:2000. Steel and iron — Determination of total carbon and sulfur content — Infrared absorption method after combustion in an induction furnace (routine method).

ISO 15901-1:2016. Evaluation of pore size distribution and porosity of solid materials by mercury porosimetry and gas adsorption Part 1: Mercury porosimetry.

ISO 17892-4:2016. Geotechnical investigation and testing — Laboratory testing of soil. Part 4: Determination of particle size distribution.

ISO 17892-12. Geotechnical investigation and testing — Laboratory testing of soil Part 12: Determination of liquid and plastic limits.

Jackson, M. L. 1975. Soil chemical analysis: advanced course. 2nd ed. Madison, WI: University of Wisconsin.

Jenni, A., Wersin, P., Thoenen, T., Baeyens, B., Ferrari, A., Gimmi, T., Mäder, U., Marschall, P., Hummel, W., Leupin, O. 2019. Bentonite backfill performance in a high-level waste repository: a geochemical perspective. Nagra technical report NTB 19-03. Wettingen, Switzerland.

Jenni, A., Mäder. 2018. Coupling of chemical and hydromechanical properties in bentonite. Applied Geochemistry 97:147-156.

Jobmann, M., Burlaka, V., CHRISTA II - Verfüll- und Verschlusskonzepte für Endlager im Kristallingestein in Deutschland, 2021. BGETEC 2021-15. BGE TECHNOLOGY GmbH. Peine

Jonsson, M., Emilsson, G., Emilsson, L. 2018. Mechanical design analysis for the canister. Posiva SKB Report 04. Eurajoki, Finland.

Kaddouri, Z., Cuisinier, O., Masrouri, F. 2019. Influence of effective stress and temperature on the creep behavior of a saturated compacted clayey soil. Geomechanics for Energy and the Environment. Geomechanics for nuclear waste storage 17: 106–114. <https://doi.org/10.1016/j.gete.2018.09.002>

Kanit, T., Forest, S., Galliet, I., Mounoury, V., Jeulin, D. 2003. Determination of the size of the representative volume element for random composites: Statistical and numerical approach. International Journal of Solids and Structures, 40, 13-14: 3647-3679. [https://doi.org/10.1016/S0020-7683\(03\)00143-4](https://doi.org/10.1016/S0020-7683(03)00143-4)

Karnland, O., Muurinen, A., Karlsson, F 2003a. Bentonite swelling pressure in NaCl solutions – experimentaly determined data and models calculations: In Alonso E.E., Ledesma, A., eds. Advances in Understanding Engineered Clay Barriers: Proceedings of the International Symposium on Large Scale Field Tests in Granite. Sitges, Barcelona, 12 – 14 November 2003, London: Taylor and Francis Group: 241-256.

Karnland, O., Nilsson, U., Olsson, S. 2003b. ECOCLAY II. Laboratory experiment concerning compacted bentonite contacted to high pH solutions: In Alonso E.E., Ledesma, A., eds. Advances in Understanding Engineered Clay Barriers: Proceedings of the International Symposium on Large Scale Field Tests in Granite. Sitges, Barcelona, 12 – 14 November 2003, London: Taylor and Francis Group: 307-321.

Karnland, O., Olsson, S., Nilsson, U. 2006. Mineralogy and sealing properties of various bentonites and smectite-rich clay materials. SKB technical report TR-06-30. Stockholm, Sweden. <https://skb.com/publication/1419144>

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Karnland, O., Birgersson, M. 2006. Montmorillonite stability with special respect to KBS-3 conditions. SKB report TR-06-11. Stockholm, Sweden.

Karnland, O., Birgersson, M., Hedström, M. 2011. Selectivity coefficient for Ca/Na ion exchange in highly compacted bentonite. Physics and Chemistry of the Earth 56: 1554-1558. <https://doi.org/10.1016/j.pce.2011.07.023>

Katsaras, J. 1998. Adsorbed to a Rigid Substrate, Dimyristoylphosphatidylcholine Multibilayers Attain Full Hydration in All Mesophases. Biophysical Journal 75: 2157–2162. [https://doi.org/10.1016/S0006-3495\(98\)77658-1](https://doi.org/10.1016/S0006-3495(98)77658-1)

Kaufhold, S., Dohrmann, R., Ufer, K., Kober, F. 2019. Interactions of bentonite with metal and concrete from FEBEX experiment: mineralogical and geochemical investigations of selected sampling sites. Clay Minerals 53(4): 745-763. <https://doi.org/10.1180/clm.2018.54>

Kaufhold S, Dohrmann R, Ufer K, Gröger-Trampe J, Kober Florian, Schneeberger R, Weber C. (2024). HotBENT experiment on quality control of bentonites used for granular bentonite material backfilling and block production. Clay Minerals. 1-22. 10.1180/clm.2024.25.

Keeling, P.S., Kirby, E.C., Robertson, R.H.S. 1980. Moisture adsorption and specific surface area. Journal of the British Ceramic Society 79: 36 – 40.

Keri, A., Osan, J., Fabian, M., Dahn, R., Torok, Sz. 2016. Combined X-ray microanalytical study of the Nd uptake capability of argillaceous rocks. X-ray Spectrometry 45: 54

Keto, P. (ed.), Hassan, M., Karttunen, P., Kiviranta, L., Kumpulainen, S., Korkiala-Tanttu, L., Koskinen, V., Jalonens, T., Koho, P., Sievänen, U. 2013. Backfill Production Line 2012. Design, Production and Initial State of the Deposition Tunnel Backfill and Plug. Posiva Working Report 2012-18. Eurajoki, Finland.

Kim, J., Kumpulainen, S., Ferrari, A., Pintado, X., Laloui, L., Heino, V. 2024. Salinity-dependent ultimate shear strength of compacted Wyoming-type bentonite investigated by triaxial tests. Applied Clay Science 261: 107576. <https://doi.org/10.1016/j.clay.2024.107576>

Kiviranta, L., Kumpulainen, S. 2011. Quality control and characterization of bentonite materials. Posiva working report 2011-84. Eurajoki, Finland.

Kiviranta, L., Kumpulainen, S., Pintado, X., Karttunen, P. Schatz, T. 2018. Characterization of bentonite and clay materials 2012-2015. Posiva working report 2016-05. Eurajoki, Finland.

Kober, K., García-Siñeriz, J.L., Villar, M.V., Lanyon, G.W., Cloet, V., Mäder, U., Wersin, P., Leupin, O., Sellin, P., Gens, A., Schneeberger, R. 2021. FEBEX-DP Synthesis Summary of the Full-Scale Engineered Barriers Experiment – Dismantling Project. Nagra technical report 17-01. Wettingen, Switzerland.

Kober, F., Schneeberger, R., Vomvoris, S., Finsterle, S., Lanyon, B. 2023. The HotBENT Experiment: objectives, design, emplacement and early transient evolution. Geoenergy 1(1). <https://doi.org/10.1144/geoenergy2023-021>

Kober, F., Schneeberger, R., Manukyan, E. & Vomvoris S. (2024): HotBENT As Built Report. Nagra Arbeitsbericht NAB 22-08.

Kouakou, N.M., Cuisinier, O., Masrouri, F. 2020. Estimation of the shear strength of coarse-grained soils with fine particles. Transportation Geotechnics 25: 100407. <https://doi.org/10.1016/j.trgeo.2020.100407>

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Koubová M., Laufek F., Hanusová I., Szczerba M. (2025): The reversibility of interlayer ion fixation in the smectite of BCV 2017 bentonite during long-term heating in air at 200 °C, a detailed mineralogical study. *Applied Clay Science*, Volume 270, 2025, 107773, ISSN 0169-1317.

Krejčí, T., Koudelka, T., Kruis, J., (2024): Mathematical models of buffer and backfill, SÚRAO technical report 807/2024. In Czech.

Kristensson, O., Börgesson, L. 2015. Caniser Retrieval Test. Final report. SKB technical report TR-14-19. Stockholm, Sweden. <https://skb.com/publication/2479354>

Kristensson, O. 2022. Modelling buffer upwards expansion. SKB technical report P-22-22. Solna, Sweden.

Kumpulainen, S., Kiviranta, L. 2010. Mineralogical and chemical characterization of various bentonite and smectite-rich clay materials. Posiva working report 2010-52. Eurajoki, Finland.

Laufek, F., Hanusová, I., Svoboda, J., Vašíček, R., Najser, J., Koubová, M., Čurda, M., Pticek, F., Vaculíková, L., Sun, H. and Mašín, D. 2021. Mineralogical, Geochemical and Geotechnical Study of BCV 2017 Bentonite - The Initial State and the State Following Thermal Treatment at 200 °C. *Minerals* 11(8): 871. <https://doi.org/10.3390/min11080871>

Laviña, M., Pelegrí, J., Idiart, A., Pasteau, A., Michau, N., Talandier, J., Cochebin, B. 2024. Long-term Evolution of Bentonite-Based Seals for Closure of a Radioactive Waste Repository in Claystone: A Hydro-Chemo-Mechanical Modelling Assessment. *Transport in Porous Media* 151: 287–317. <https://doi.org/10.1007/s11242-023-01989-3>

Leoni, M., Börgesson, L., Keto, P. 2018. Numerical modelling of the buffer swelling test in Äspö HRL Validation of numerical models with the buffer swelling test data. SKB technical report TR-17-03. Solna, Sweden.

Leroy, A., Cuisinier, O., Masrouri, F., Talandier, J. 2025. Sand-claystone mixtures: Investigating the impact of sand proportions on hydro-mechanical behavior at different scales. Presented at the 9th International Conference on Clays in Natural and Engineered Barriers for Radioactive Waste Confinement, Hanover, p. 2.

Leupin (ed.), O.X., Birgersson, M., Karnland, O., Korkeakoski, P., Sellin, P., Mäder, U., Wersin, P. 2014. Montmorillonite stability under near-field conditions. Nagra technical report 14-12. Wettingen, Switzerland.

Leupin, O., Sellin, P., Talandier, J. 2022. BEACON / WP1.3 / Final Assessment Report. Nagra technical report NAB 22-16. Wettingen, Switzerland.

Levasseur S., Collin F., Dymitrowska M., Harrington J., Jacops E., Kolditz O., Marschall P., Norris S., Sillen X., Talandier J., Truche L. and Wendling J. (2024). Achievements of EURAD-GAS. Work Package GAS (Mechanical understanding of gas transport in clay-based materials) of the HORIZON 2020 project EURAD. EC Grant agreement N°847593. Deliverable 6.6 (D6.6).

Li, X. L., Van Geet, M., Bruggeman, C. and De Craen, M. (eds). 2023. Geological Disposal of Radioactive Waste in Deep Clay Formations: 40 Years of RD&D in the Belgian URL HADES. Geological Society, London, Special Publications, 536, <https://doi.org/10.1144/SP536-2022-42>

Lloret, A., Villar, M.V., Sánchez, M., Gens, A., Pintado, X., Alonso, E. 2003. Mechanical behaviour of heavily compacted bentonite under high suction changes. *Géotechnique* 53(1): 27-40. <https://doi.org/10.1680/geot.53.1.27.37258>

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Lloret, A., Romero, E., Villar, M.V. (2004). FEBEX II Project. Final report on thermo-hydro-mechanical laboratory tests. Enresa technical publication 10/2004. Madrid, Spain.

Lohser, T (2024): Arbeitsstände zur vorläufigen Endlagerauslegung, 3. Forum Endlagersuche – AG 8, Würzburg, BGE.

Manca, D., Ferrari, A., Laloui, L. 2016. Fabric evolution and the related swelling behaviour of a sand/bentonite mixture upon hydro-chemo-mechanical loadings. *Géotechnique* 66(1): 41-57. <https://doi.org/10.1680/jgeot.15.P.073>

Marschall, P., Talandier, J. and Kolditz, O. (editors). (2024). Barrier integrity: gas-induced impacts and model-based interpretation. EURAD-GAS: Work Package GAS (Mechanical understanding of gas transport in clay-based materials) of the HORIZON 2020 project EURAD. EC Grant agreement N°847593. Task 3. Final Technical Report. Deliverable 6.8 (D6.8).

Mašín, D. 2013. Double structure hydromechanical coupling formalism and a model for unsaturated expansive clays. *Engineering Geology* 165: 73-88. <https://doi.org/10.1016/j.enggeo.2013.05.026>

Mašín, D. 2017. Coupled thermohydromechanical double structure model for expansive soils. *ASCE Journal of Engineering Mechanics* 143(9).

Massat L. 2016. PhD. Influence de la chimie sur les propriétés multi-échelles du gonflement d'une bentonite compactée. Université de Lorraine, 272 p.

Massat, L., Cuisinier, O., Bihannic, I., Claret, F., Pelletier, M., Masrouri, F., Gaboreau, S. 2016. Swelling pressure development and inter-aggregate porosity evolution upon hydration of a compacted swelling clay. *Applied Clay Science* 124–125: 197–210. <https://doi.org/10.1016/j.clay.2016.01.002>

Martikainen, J., Schatz, T. 2011. Laboratory tests to determine the effect of Olkiluoto bounding brine water on buffer performance. Posiva working report 2011-68. Eurajoki, Finland.

Martikainen, J., Marjavaara, P., Leino, T. 2019. Testing backfill performance under early inflow in a 1:6 scale deposition tunnel test device. Posiva working report 2018-10. Eurajoki, Finland.

Mata, C., Ledesma, A. 2003. Permeability of a bentonite-crushed granite rock mixture using different experimental techniques. *Geotechnique* 53(8): 747-758. <https://doi.org/10.1680/geot.2003.53.8.747>

Mata, C., Guimarães, L.N., Ledesma, A., Gens, A., Olivella, S. 2005. A hydro-geochemical analysis of the saturation process with salt water of a bentonite crushed granite rock mixture in an engineered nuclear barrier. *Engineering geology* 81(3): 227-245. <https://doi.org/10.1016/j.enggeo.2005.06.018>

McFarlane, J., Anovitz, L. M., Cheshire, M. C., DiStefano, V. H., Bilheux, H. Z., Bilheux, J. C., Qualls, L. M. 2021. Water Migration and Swelling in Engineered Barrier Materials for Radioactive Waste Disposal. *Nuclear Technology* 207(8): 1237–1256. <https://doi.org/10.1080/00295450.2020.1812348>

Meier, L. P., Kahr, G. 1999. Determination of the cation exchange capacity (CEC) of clay minerals using the complexes of copper(II) ion with triethylenetetramine and tetraethylenepentamine. *Clays and Clay Minerals* 47: 386–388.

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Meng Y., Wang Q., Su W., Ye W., Chen Y. 2023 Experimental evidence on the cracking and sealing mechanisms of compacted bentonite by using microfocus X-ray computed tomography, *Engineering Geology* 322: 107153. <https://doi.org/10.1016/j.enggeo.2023.107153>

Miettinen, A., Harjupatana, T., Kataja, M., Fortino, S., Immonen, K. 2016. Time-resolved X-ray microtomographic measurement of water transport in wood-fibre reinforced composite material. In B. Madsen, A. Biel, Y. Kusano, H. Lilholt, L. Mikkelsen, L. Mishnaevsky, & B. Sørensen (Eds.), 37th Risø International Symposium on Materials Science (Article 012037). Institute of Physics Publishing Ltd.. IOP Conference Series: Materials Science and Engineering, 139. <https://doi.org/10.1088/1757-899X/139/1/012037>

Middelhoff, M., Cuisinier, O., Masrouri, F., Talandier, J., Conil, N. 2020. Combined impact of selected material properties and environmental conditions on the swelling pressure of compacted claystone/bentonite mixtures. *Applied Clay Science* 184: 105389. <https://doi.org/10.1016/j.clay.2019.105389>

Middelhoff, M., Cuisinier, O., Masrouri, F., Talandier, J. 2021. Hydro-mechanical path dependency of claystone/bentonite mixture samples characterized by different initial dry densities. *Acta Geotechnica* 16: 3161–3176. <https://doi.org/10.1007/s11440-021-01246-1>

Middelhoff, M., Cuisinier, O., Gaboreau, S., Masrouri, F., Talandier, J., Michau, N. 2023. Hydraulic conductivity, microstructure and texture of compacted claystone/ bentonite mixtures saturated with different solutions. *Applied Clay Science* 241: 106982. <https://doi.org/10.1016/j.clay.2023.106982>

Mokni, N., Barnichon, J.-D., Dick, P., Nguyen, T.S. 2016. Effect of technological macro voids on the performance of compacted bentonite/sand seals for deep geological repositories. *International Journal of Rock Mechanics and Mining Sciences* 88: 98-104. <https://doi.org/10.1016/j.ijrmms.2016.07.009>

Mokni, N., Guerra, A.M., Cui, Y.-J., Bornert, M., Tang A.-M. 2020. Modelling the long-term hydro-mechanical behaviour of a bentonite pellet/powder mixture with consideration of initial structural heterogeneities. *Géotechnique* 70(7): 563–580. <https://doi.org/10.1680/jgeot.18.P.110>

Mokni, N., Cabrera, J., Deleruyelle, F. 2023. On the installation of an in situ large-scale vertical SEALing (VSEAL) experiment on bentonite pellet-powder mixture. *Journal of Rock Mechanics and Geotechnical Engineering* 15(9): 2388–2401. <https://doi.org/10.1016/j.jrmge.2023.04.008>

Molinero-Guerra, A., Mokni, N., Delage, P., Cui, Y. J., Tang, A. M., Aimedieu, P., Bernier, F., Bornert, M. 2016. In-depth characterisation of a mixture composed of powder/pellets MX80 bentonite. *Applied Clay Science* 135: 538 – 546.

Molinero Guerra, A., Cui, Y.-J., Mokni N., Tang, A.M., Bernier, F. 2018a. Investigation of the hydro-mechanical behaviour of a pellet/powder MX80 bentonite mixture using an infiltration column. *Engineering Geology* 243: 18–25. <https://doi.org/10.1016/j.enggeo.2018.06.006>

Molinero-Guerra, A., Aimedieu, P., Bornert, M., Cui, Y.J., Tang, A. M., Sun, Z., Mokni, N., Delage, P., Bernier, F. 2018b. Analyses of the structural changes of a pellet/powder bentonite mixture upon wetting by X-ray computed microtomography. *Applied Clay Science* (165): 164 - 169.

Molinero Guerra, A., Cui, Y.-J., He, Y., Delage, P., Mokni, N., Tang, A.-M., Aimedieu, P., Bornert, M., Bernier, F. 2019a. Characterization of water retention, compressibility and swelling properties of a pellet/powder bentonite mixture. *Engineering Geology* 248: 14–21. <https://doi.org/10.1016/j.enggeo.2018.11.005>

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Molinero-Guerra, A., Mokni, N., Cui, Y.J., Delage, P., Tang, A. M., Aimedieu, P., Bernier, F., Bornert, M., (2019b). Impact of initial heterogeneity on long term swelling behavior of MX80 bentonite pellet/powder mixtures. Canadian Geotechnical Journal, 57(9), 1404 – 1416.

Molinero-Guerra, A., Delage, P., Cui, Y.-J., Mokni, N., Tang, A.M., Aimedieu, P., Bernier, F., Bornert, M. 2020. Water-retention properties and microstructure changes of a bentonite pellet upon wetting/drying; application to radioactive waste disposal. Géotechnique 70(3): 199–209. <https://doi.org/10.1680/jgeot.17.P.291>

Nagle, J.F., Katsaras, J. 1999. Absence of a vestigial vapor pressure paradox. Phys. Rev. E 59: 7018–7024. <https://doi.org/10.1103/PhysRevE.59.7018>

Nagra. 2021. The Nagra Research, Development and Demonstration (RD&D) Plan for the Disposal of Radioactive Waste in Switzerland. Nagra technical report 21-02. Wettingen, Switzerland. <https://nagra.ch/wp-content/uploads/2022/08/NTB-21-02.pdf>

Nagra 2024. Post-closure safety report. Nagra Technical Report NTB 24-10.

Narkuniene, A., Poskas, P., Justinavicius, D. 2021. The Modeling of Laboratory Experiments with COMSOL Multiphysics Using Simplified Hydromechanical Model. Minerals 11: 754. <https://doi.org/10.3390/min110707>

Najser J., Mašín D., Svoboda J., Vašíček R., Hanusová I., Hausmannová L., Kruis J., Krejčí T., Sun H. (2023): The homogenisation behaviour of BCV bentonite – A laboratory and numerical study. Applied Clay Science, Volume 241, 2023, 106969, ISSN 0169-1317.

Najser, J., Mašín, D. 2024. An experimental study on thermal relaxation of BCV bentonite. Applied Clay Science 254: 107374. <https://doi.org/10.1016/j.clay.2024.107374>

Navarro, V., Asensio, L., De la Morena, G., Pintado, X., Yustres, A., 2015. Differentiated intra- and inter-aggregate water content models of MX-80 bentonite. Applied Clay Science 118, 325-336.

Navarro, V., Yustres, A., Asensio, L., De la Morena, G., González-Arteaga, J., Laurila, T., Pintado, X. 2017. Modelling of compacted bentonite swelling accounting for salinity effects. Engineering Geology 223: 48-58. <http://dx.doi.org/10.1016/j.enggeo.2017.04.016>

Navarro, V., De la Morena, G., González-Arteaga, J., Yustres, A., Asensio, L. 2018. A microstructural effective stress definition for compacted active clays. Geomechanics for Energy and the Environment 15: 47-53. <https://doi.org/10.1016/j.gete.2017.11.003>

Navarro, V., Asensio, L., De la Morena, G., Gharbieh, H., Alonso, J., Pulkkanen, V-M. 2020. From double to triple porosity modelling of bentonite pellet mixtures. Engineering Geology 274: 105714. <https://doi.org/10.1016/j.enggeo.2020.105714>.

Navarro, V., Asensio, L. 2022. Effect of salinity on the effective stress of compacted bentonites. Géotechnique. <https://doi.org/10.1680/jgeot.22.00251>

Neerdael, B., Meynendonckx, P., & Voet, M. (1992). The Bacchus backfill experiment at the Hades underground research facility at Mol, Belgium: Final report. (Publications Office of the European Union; Vol. CD-NA-14-155-EN-C, No. EUR-14155). Publications Office of the European Commission. <https://op.europa.eu/s/y6UA>

Newman, A.C.D. 1983. The specific surface of soils determined by water sorption. Journal of soil Science: 3423-32

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

NF-PRO (2008) Understanding and Physical and Numerical Modelling of the Key Processes in the Near Field and their Coupling for Different Host Rocks and Repository Strategies (NF-PRO) European Report EUR 23730

Odom, I. E. (1984). Smectite clay Minerals: Properties and Uses. *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*. 311 (1517): 391–409. <https://doi.org/10.1098/rsta.1984.0036>

Ojala, A.E.K., Mattila, J., Ruskeeniemi, T., Palmu, J.-P., Nordbäck, N., Kuva, J., Sutinen, R. 2019. Postglacial reactivation of the Suasselkä PGF complex in SW Finnish Lapland. *International Journal of Earth Science* 108: 1049-1065. <https://doi.org/10.1007/s00531-019-01695-w>

Olin M., Grgic D., Svoboda J., Villar M.V., Graham C.C., de Lesquen C., Svensson D., Leupin O., Collin F. (2024) Final report of HITEC. Final version as of 21.05.2024 of deliverable D7.12 of the HORIZON 2020 project EURAD. EC Grant agreement no: 847593.

Olsson, S., Jensen, V., Johannesson, L-E, Hansen, E., Karnland, O., Kumpulainen, S., Kiviranta, L., Svensson, D., Hansen, H., Lindén, J., Akademi, Å. 2013. Prototype repository. Hydro-mechanical characterization of the buffer and tunnel backfill material from outer section of the Prototype Repository. SKB report TR-13-21. Stockholm, Sweden. <https://skb.com/publication/2477824/TR-13-21.pdf>

Osan, J., Keri, A., Breitner, D., Fabian, M., Dahn, R., Simon, R., Torok, Sz. 2014. Microscale analysis of metal uptake by argillaceous rocks using positive matrix factorization of microscopic X-ray fluorescence elemental maps. *Spectrochimica Acta Part B-Atomic Spectroscopy* 91: 12.

Owusu, J., Karalis, K., Prasianakis, N., Churakov, S. 2023. Diffusion and Gas Flow Dynamics in Partially Saturated Smectites. *Journal of Physical Chemistry* 127 (29): 14425-14438. <https://doi.org/10.1021/acs.jpcc.3c02264>

Pearson, F.J. (2002). PC experiment: Recipe of Artificial pore water. Mont Terri Technical Note, TN 2002-17. Switzerland Federal Office of Topography (Swisstopo). Wabern, Switzerland.

Philipp, T., Amann-Hildenbrand, A., Laurich, B., Desbois, G., Littke, R., Urai, J.L. 2017, The effect of microstructural heterogeneity on pore size distribution and permeability in Opalinus Clay (Mont Terri, Switzerland): Insights from an integrated study of laboratory fluid flow and pore morphology from BIB-SEM images. (In: Geological Society Special Publication). Bd. 454: 85-106. <https://doi.org/10.1144/SP454.3>

Pintado, X., Lloret, A., Romero, E. 2009. Assessment on the use of the vapour equilibrium technique in controlled-suction tests. *Canadian Geotechnical Journal* 46: 411-423.

Pintado, X., Md. Mamanul, H., Martikainen, J. 2013a. Thermo-hydro-mechanical tests of buffer material. Posiva report 2012-49. Eurajoki, Finland.

Pintado, X., Adesola, F., Turtiainen, M. 2013b. Downscaled tests on buffer behaviour. Posiva working report 2012-100. Eurajoki, Finland.

Pintado, X., Romero, E., Suriol, J., Lloret, A., Madhusudhan, B.N. 2019. Small-strain shear stiffness of compacted bentonites for engineered barrier system. *Geomechanics for Energy and the Environment* 19: 1-12.

Pintado, X., Kanerva, N., Leino, T. 2022. Determination of the water retention curve of bentonite from Georgia and from Wyoming (United States). Experimental results using the jar method and

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

assessment of available techniques for suction control and measurement. SKB report P-22-14. Solna, Sweden.

Pintado, X. 2022. Parameter study of buffer upwards expansion into backfill. Buffer swelling against the backfill. SKB technical report P-22-08. Solna, Sweden.

Pintado, X., Kumpulainen, S., Romero, E., Lloret, A., Weber, R.C., Ferrari, A., Villar, M.V., Abed, A., Solowski, W., Heino, V. 2023. Shear strength and shear stiffness analysis of compacted Wyoming-type bentonite. Geomechanics for Energy and Environment 34: 100648

Podgornik, R., Parsegian, V.A. 1997. On a Possible Microscopic Mechanism Underlying the Vapor Pressure Paradox. Biophysics Journal 72: 942–952. [https://doi.org/10.1016/S0006-3495\(97\)78728-9](https://doi.org/10.1016/S0006-3495(97)78728-9)

Polubesova, T., Rytwo, G., Nir, S., Serban, C., Marguiles, L. 1997. Adsorption of Benzyltrimethylammonium and Benzyltriethylammonium on Montmorillonite: Experimental Studies and Model Calculations. Clays and Clay Minerals 45: 834-841.

Posiva. 2021. Buffer, backfill and closure evolutions. Posiva Working Report 2021-08. Eurajoki, Finland.

Radioactive Waste Management, 2017, Geological Disposal Concept Status Report, NDA Report no. NDA/RWM/155.

Rand, R.P., Parsegian, V.A. 1989. Hydration forces between phospholipid bilayers. Biochim. Biophys. Acta BBA - Rev. Biomembr. 988: 351–376. [https://doi.org/10.1016/0304-4157\(89\)90010-5](https://doi.org/10.1016/0304-4157(89)90010-5)

Richards, T. 2010. Particle clogging in porous media - Filtration of a smectite solution. SKB Report TR-10-22. Stockholm, Sweden.

Robinet, J.-C., Tyri, D., & Djeran-Maigre, I. 2021. Hydro-mechanical response of crushed argillite and bentonite mixtures as sealing material. Engineering Geology 288: 106140. <https://doi.org/10.1016/j.engeo.2021.106140>

Rodrigues, M. I., lemma, A. F. 2015. Experimental Design and Process Optimization. Boca Raton: CRC Press. ISBN 978-1-4822-9956-4.

Rodriguez-Dono, A., Olivella, S., Mokni, N. 2018. Assessment of a high-level spent nuclear fuel disposal model. Environmental Geotechnics 7(1): 42-58.

Rostamiparsa, M., Tolnai, I., Czompoly, O., Fabian, M., Hegedus, M., Falus, Gy., Szabo, Cs., Ovari, M., Tobi, Cs., Konya, P., Völgyesi, P., Szabó-Krausz, Z. 2023. The geochemical role of B-10 enriched boric acid in cemented liquid radioactive wastes, Journal of Radioanalytical and Nuclear Chemistry 332: 2543.

Roldughin, V.I., Karpenko-Jereb, L.V. 2016. On the Schroeder paradox for ion-exchange polymers. Colloid Journal 78: 795–799. <https://doi.org/10.1134/S1061933X16060132>

Saanio, T., Ikonen, A., Keto, P., Kirkkomäki, T., Kukkola, T., Nieminen, J., Raiko, H. 2013. Design of the Disposal Facility 2012. Posiva working report 2013-17. Eurajoki, Finland.

Saba, S., Barnichon, J.D., Cui, Y.J., Tang, A.M. and Delage, P. 2013. Microstructure and anisotropic swelling behaviour of compacted bentonite/sand mixture. Journal of Rock Mechanics and Geotechnical Engineering 6(2), 126-132.

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Saba, S., Delage, P., Cui, Y.J., Tang, A.M., Lenoir, N. and Barnichon, J.D. 2014a. Further insight into the microstructure of compacted bentonite/sand mixture. *Engineering Geology* 168, 141–148.

Saba, S., Cui, Y.J., Tang, A.M. and Barnichon, J.D. 2014b. Investigation of the swelling behaviour of compacted bentonite–sand mixture by mock-up tests. *Canadian Geotechnical Journal* 51(12), 1399 – 1412.

Saba, S., Cui, Y.J., Barnichon, J.D. and Tang, A.M. 2014c. Infiltration column for studying the lateral swell behavior of expansive clay. *Geotechnical Testing Journal* 39(3):20150151.

Šachlová Š., Černochová K., Černá K., Svoboda J., Vašíček R., Macková D., Havlová V., Zuna M., Večerník P., Kolková K., Hlaváčková V. (2022): Analysis of Czech bentonites – Evaluation of data from the database, SÚRAO technical report 632/2022. In Czech.

Sánchez, L., Cuevas, J., Ramírez, S., Riuiz De León, D., Fernández, R., Vigil De la Villa, R., Leguey, S. 2006. Reaction kinetics of FEBEX bentonite in hyperalkaline conditions resembling the cement–bentonite interface. *Applied Clay Science* 33(2): 125-141.

<https://doi.org/10.1016/j.clay.2006.04.008>

Sandén, T., Börgesson, L., Nilsson, U., Dueck, A., 2017. Full scale Buffer Swelling Test at dry backfill conditions in Äspö HRL In situ test and related laboratory tests. SKB Technical Report TR-16-07. Solna, Sweden.

Sawhney, B.L. 1970. Potassium and cesium ion selectivity in relation to clay mineral structure. *Clays and Clay Minerals* 18(1): 47-52.

Schäfers, A., Gaus, I., Johnson, L., Liu, Y., Mayor, J.C., Sellin, P., Wieczore, K. (2014). PEBS Final Scientific Report Deliverable D5-16. European Commission. Grant agreement n° FP7-249681.

Schatz, T., Martikainen, J. (2013). Laboratory tests and analyses on potential Olkiluoto backfill materials. Posiva working report 2012-74. Eurajoki, Finland.

Seiphoori, A., Ferrari, A., Laloui, L. 2014. Water retention behaviour and microstructural evolution of MX-80 bentonite during wetting and drying cycles. *Géotechnique* 64 (9): 721-734.

Sellin, P., Leupin, O.X., 2013. The Use of Clay as an Engineered Barrier in Radioactive Waste Management: A review, *Clays and Clay Minerals*, 61, 477-498

Sellin, P., Åkesson, M., Kristensson, O., Malmberg, D., Börgesson, L., Birgersson, M., Dueck, A., Karnland, O., Hernelind, J. 2017. Long re-saturation phase of a final repository Additional supplementary information. SKB Technical Report TR-17-15. Solna, Sweden.

Sellin, P., Westermark, M., Leupin, O., Norris, S., Gens, A., Wieczorek, K., Talandier, J. Swahn, J. 2020. Beacon: bentonite mechanical evolution. *EPJ N Nuclear sciences and technologies* 6: 23. <https://doi.org/10.1051/epjn/2019045>

SFS-EN 16170:2016. Sludge, treated biowaste and soil. Determination of elements using inductively coupled plasma optical emission spectrometry (ICP-OES).

SFS-EN 16171:2016. Sludge, treated biowaste and soil - Determination of elements using inductively coupled plasma mass spectrometry (ICP-MS).

Shelton A, Sellin P, Missana T, Schäfer T, Červinka R, Koskinen K, 2018. Synthesis report: Colloids and related issues in the long term safety case. SKB TR-17-17, Svensk Kärnbränslehantering AB.

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Sinnathamby, G., Korkiala-Tanttu, L., Gallardo Forés, J. 2014. Interface shear behaviour of tunnel backfill materials in a deep-rock nuclear waste repository in Finland. *Soils and Foundations* 54(4): 777-788. <http://dx.doi.org/10.1016/j.sandf.2014.06.027>

SKB. 2020. Post-closure safety for the final repository for spent nuclear fuel at Forsmark Climate and climate-related issues, PSAR version. SKB report TR-20-12. Svensk Kärnbränslehantering AB

SKB, 2022. Post-closure safety for the final repository for spent nuclear fuel at Forsmark Main report, PSAR version. SKB Technical Report TR-21-01. Svensk Kärnbränslehantering AB

SM 4500-S D. Sampling water for dissolved sulphide (DS).

Smedley, P.L., Bearcock, J.M., Newell, A.J., et al. Guide to Reference Groundwater and Porewater Compositions in Support of the UK GDF Programme, NWS-CR-23-007, 2023.

SS-EN ISO 12677:2011. Chemical analysis of refractory products by X-ray fluorescence (XRF) — Fused cast-bead method.

SS-EN 12698-2:2007. Chemical analysis of nitride bonded silicon carbide refractories - Part 2: XRD methods.

SS-EN ISO 13196:2015. Soil quality - Screening soils for selected elements by energy-dispersive X-ray fluorescence spectrometry using a handheld or portable instrument.

StandAG: Standortauswahlgesetz vom 5. Mai 2017 (BGBl. I S. 1074), das zuletzt durch Artikel 8 des Gesetzes vom 22. März 2023 (BGBl. 2023 I Nr. 88) geändert worden ist

Sun, H., Masin, D., Nasjer, J., Nedela, V., Navratilova, E. 2019. Bentonite microstructure and saturation evolution in wetting-drying cycles evaluated using ESEM, MIP and WRC measurements 69(8): 713-726. <https://doi.org/10.1680/jgeot.17.P.253>

Sun, H., Mašín, D., Najser, J., Neděla, V., Navrátilová, E. (2020a). Fractal characteristics of pore structure of compacted bentonite studied by ESEM and MIP methods. *Acta Geotechnica* 15: 1655-1671. <https://doi.org/10.1007/s11440-019-00857-z>

Sun, H., Mašín, D., Najser, J., Scaringi, G. (2020b). Water retention of a bentonite for deep geological radioactive waste repositories: high-temperature experiments and thermodynamic modeling. *Engineering Geology* 269: 105549. <https://doi.org/10.1016/j.enggeo.2020.105549>

Svemar, Ch., Johannesson, L-E, Grahm, P., Svensson, D., Kristensson, O., Lönnqvist, M., Nilsson, U. (2016). Prototype Repository Opening and retrieval of outer section of Prototype Repository at Äspö Hard Rock Laboratory Summary report. SKB technical report TR-13-22. Stockholm, Sweden.

Svensson, D., Lundgren, C., Johannesson, L.-E., Norrfors, K. 2017. Developing strategies for acquisition and control of bentonite for a high level radioactive waste repository. SKB report TR-16-14. Solna, Sweden.

Svensson, D., Eriksson, P., Johannesson, L.-E., Lundgren, C., Bladström, T. 2019. Development and testing of methods suitable for quality control of bentonites as KBS-3 buffer and backfill. SKB report TR-19-25. Solna, Sweden.

Svensson, D., Sellin, P., Kaufhold, S., Chaaya, R., Gaboreau, S., Tremosa, J., Villar, M-V., Melón, A-M, Zabala, A.B., Iglesias, R. J., Najser, J., Svoboda, J., Černochová, K., Sayenko, S., Zlobenko B., Bugera S., Fedorenko Y., Rozko A., Cuevas, J., Ortega, A., Ruiz, A. I., Kašpar, V., Šachlová, S., Pulkkanen, V-M., Rauhala, O-P. 2023: HITEC technical report on Material characterisation. Final

Svoboda, J., Mašín, D., Najser, J. Vašíček, R., Hanusová, I. and Hausmannová, L. 2022. BCV bentonite hydromechanical behaviour and modelling. *Acta Geotechnica* 18: 3193-3211. <https://doi.org/10.1007/s11440-022-01689-0>

Svoboda J, Vašíček R, Krejčí T, Mašín D, Laufek F, Franěk J. (2023) Engineered barrier 200C. In: *Proceedings of the 15th International Conference “Underground Construction Prague 2023”*. Prague: The Czech Tunnelling Association ITA-AITES ISBN 978-80-906452-5-7.

Svoboda, J., Vašíček, R., Rukavičková, L., Řihošek, J., Večerník, P. 2024. Interakční experiment – Průběžná zpráva č. 8, SÚRAO, TZ 799/2024

Tachibana, S., Ito, S., Iizuka, A., Owada, H., Hayashi, D. 2017. H-M-C coupling analysis considering several scenarios of long-term alteration in cement-bentonite system. In: Proc. 2nd Annual Workshop CEBAMA Project.

Talandier, J., Kristensson, O., Malmberg, D., Narkuniene, A., Justinavicius, D., Zdravkovic, L., Pulkkanen, V-M, Gharbieh, H., Ferrari, A., Bosch, J., Kumar, V., Beese, S., Gens, A., Charlier, R., Newson, R., Åkesson, M., Scaringi, G., Masin, D., Leupin, O. 2022. Synthesis of the results obtained from all tasks in WP5 Final report for WP5. BEACON EU project.

Tang, A-M, Cui, Y.-J. 2005. Controlling suction by the vapour equilibrium technique at different temperatures and its application in determining the water retention properties of MX80 clay. *Canadian Geotechnical Journal* 42(1): 1-12. <https://doi.org/10.1139/t04-082>

Tang, C. S., Tang, A. M., Cui, Y. J., Delage, P., Schroeder, C., Shi, B. 2011a. A study of the hydro-mechanical behaviour of compacted crushed argillite. *Engineering Geology* 118(3–4): 93–103. <https://doi.org/10.1016/j.engeo.2011.01.004>

Tang, C. S., Tang, A. M., Cui, Y. J., Delage, P., Schroeder, C., De Laure, E. 2011b. Investigating the swelling pressure of compacted crushed-Callovian-Oxfordian claystone. *Physics and Chemistry of the Earth, Parts A/B/C*, 36(17–18): 1857–1866. <https://doi.org/10.1016/j.pce.2011.10.001>

Tanttu, J., Harjupatana, T., Miettinen, A., Kataja, M. 2024. Monitoring semi-free swelling and water transport in bentonites using X-ray radiography. *Applied Clay Science* 256; 107443. <https://doi.org/10.1016/j.clay.2024.107443>

Thölix, L., Korhonen, N., Venäläinen, A., Korhonen, H. 2016. Sensitivity tests and glaciation scenarios of the future with CLIMBER-2-SICOPOLIS model system for Olkiluoto. Posiva working report 2016-04. Eurajoki, Finland.

Tolnai, I., Osan, J., Czompoly, O., Sulyok, A., Fabian, M. 2023. Glass/steel/clay interactions in a simulated radioactive waste geological disposal system. *Scientific Reports* 13: 20381.

Tournassant, Ch, Appelo, C.A.J. 2011. Modelling approaches for anion-exchange on clay minerals. *Geochimica et Cosmochimica Acta* 75(13): 3698-3710. <https://doi.org/10.1016/j.gca.2011.04.001>

Trpkošová, D., Dobrev, D., Gondolli, J., Havlová, V., Hokr, M., Marková, L., Vašíček, R. 2013. Metodiky celkového posouzení bezpečnosti hlubinného úložiště, Záverecná zpráva projektu FR-TI1/362

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Tsitsopoulos, V., Holton, D., Appleyard, P., Thompson, S., Baxter, S., Niskanen, M. 2023. Thermal hydraulic and mechanical modelling of the full-scale in situ test (FISST). *Engineering Geology* 322: 107165. <https://doi.org/10.1016/j.enggeo.2023.107165>

Tuttolomondo, A., Ferrari, A., Laloui, L. 2021. Generalized effective stress concept for saturated active clays. *Canadian Geotechnical Journal* 58: 1627-1639. <https://doi.org/10.1139/cgj-2020-0390>

Vallier, C., Winkelmann, D., Roizard, D., Favre, E., Scharfer, Ph., Kind, M. 2006. On Schroeder's paradox. *Journal of Membrane Science* 278(1-2): 357-364. <https://doi.org/10.1016/j.memsci.2005.11.020>

Vandoorne, R., Gräbe, P.J., Heymann, G. 2022. Polyethylene glycol and membrane processes applied to suction control in geotechnical osmotic testing. *International Journal of Geotechnical Engineering* 16: 103-122. <https://doi.org/10.1080/19386362.2021.1962109>

Vasconcelos, R., Rodríguez, C., Gens, A., Krejčí, T., Mašín, D., Koudelka, T., Kruis, J., Pulkkanen, V-M, Rauhala, O-P. 2024. Influence of Temperature on Clay-based Material Behaviour. Deliverable 7.10: HITEC – Modelling. HITEC Eurad-1 Work Package.

Večerník, P. 2018. Vývoj aparatur pro charakterizaci materiálů inženýrských bariér hlubinného úložiště radioaktivních odpadů a vyhořelého jaderného paliva, závěrečná zpráva projektu TAČR TA04021378

Večerník, P., Svoboda, J., Stiblíková, P., Pospíšková, I., Špinka, O., Dobrev, D., Havlová, V., Zuna, M., Vašíček, R., Černá, K., Hlaváčková, V., Vozář, M., Butovič, A. 2022. Zhodnocení konstrukčních prvků z hlediska vzájemných interakcí s materiály na bázi bentonitu v úrovni ukládacího horizontu, SÚRAO, TZ 616/2022

Villar, M.V., Lloret, A., 2001. Variation of the intrinsic permeability of expansive clay upon saturation. 259-266 in *Clay Science for Engineering*. ADACHI, K. & FUKUE, M. (editors), Rotterdam: Balkema. ISBN 90 5809 175 9

Villar, M.V. 2002. Thermo-hydro-mechanical characterisation of a bentonite from Cabo de Gata. A study applied to the use of bentonite as sealing material in high level radioactive waste repositories. *Publicación Técnica ENRESA* 01/2002. 258 pp.

Villar, M.V., Lloret, A. 2004. Influence of temperature on hydro-mechanical behaviour of a compacted bentonite. *Applied Clay Science* 26: 337-350. <https://doi.org/10.1016/j.clay.2003.12.026>

Villar (2005). MX-80 bentonite. Thermo-Hydro-Mechanical Characterisation performed at CIEMAT in the context of the Prototype project. *Informes Técnicos Ciemat* 1053. Madrid. Spain. [http://documenta.ciemat.es/bitstream/123456789/778/1/ciematito1053\\_prototype.pdf](http://documenta.ciemat.es/bitstream/123456789/778/1/ciematito1053_prototype.pdf)

Villar, M.V., 2007. Water retention of two natural compacted bentonites. *Clays and Clay Minerals* 55(3), 311-322.

Villar, M.V., Martín, P.L., Romero, F., Iglesias, R, Gutiérrez-Rodrigo, V. 2016. Saturation of barrier materials under thermal gradient. *Geomechanics for Energy and the Environment* 8: 38-51. <https://doi.org/10.1016/j.gete.2016.05.004>

Villar, M.V., Iglesias, R.J, García-Siñeriz, J-L 2020. State of the in situ Febex test (GTS, Switzerland) after 18 years: a heterogeneous bentonite barrier. *Environmental Geotechnics* 7(2): 147-159. <https://doi.org/10.1680/jenge.17.00093>

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Villar, M.V., Iglesias, R.J., Gutiérrez-Álvarez, C., Carbonell, B. 2021. Pellets/block bentonite barriers: laboratory study of their evolution upon hydration. *Engineering Geology* 292: 106272. <https://doi.org/10.1016/j.engeo.2021.106272>

Villar, M.V., Iglesias, R.J., Gutiérrez-Álvarez, C., Carbonell, B., Real, E., Brea, N. 2022a. Laboratory tests on bentonite homogenisation performed by CIEMAT: Saturation of block/pellets systems (Project BEACON). *Informes Técnicos CIEMAT* 1509. Madrid, 110 pp. <https://www.ciemat.es/portal.do?TR=A&IDR=1&identificador=998>

Villar, M.V., Gutiérrez-Álvarez, C., Campos, G. 2022b. Laboratory tests on bentonite homogenisation performed by CIEMAT: Gap filling (Project BEACON). *Informes Técnicos CIEMAT* 1510. Madrid, 72 pp. <https://www.ciemat.es/portal.do?TR=A&IDR=1&identificador=997>

Villar, M.V., Gutiérrez-Álvarez, C., Campos, G. 2023. Bentonite swelling into a void under suction or water flow. *Acta Geotechnica* 18(3): 1495-1513. <https://doi.org/10.1007/s11440-022-01702-6>

Villar, M.V., Iglesias, R.J., Gutiérrez-Álvarez, C. 2024. Saturation of block/pellets barriers of Wyoming bentonite. *Informes Técnicos CIEMAT* 1542. Madrid, 67 pp. Spain. <https://www.ciemat.es/portal.do?TR=A&IDR=1&identificador=1105>

Wang, Q., Cui, Y.J., Tang, A.M., Barnichon, J.D., Saba, S. and Ye, W.M. 2013a. Hydraulic conductivity and microstructure changes of compacted bentonite/sand mixture during hydration. *Engineering Geology* 164: 67-76.

Wang, Q., Tang, A. M., Cui, Y. J., Delage, P., Barnichon, J. D., Ye, W. M. 2013b. The effects of technological voids on the hydro-mechanical behaviour of compacted bentonite–sand mixture. *Soils and Foundations* 53(2): 232-245.

Wang, Q., Tang, A. M., Cui, Y. -J., Barnichon, J. D., Ye, W. M. 2013c. A comparative study on the hydro-mechanical behavior of compacted bentonite/sand plug based on laboratory and field infiltration tests. *Engineering Geology* 162: 79-87.

Wang, D.-W., Zhu, C., Tang, C.-S., Li, S.-J., Cheng, Q., Pan, X.-H., Shi, B. 2021. Effect of sand grain size and boundary condition on the swelling behavior of bentonite–sand mixtures. *Acta Geotechnica* 16: 2759–2773. <https://doi.org/10.1007/s11440-021-01194-w>

Wang, Y., Teng, J., Huang, Q., Wang, W., Zhong, Y. 2023. Insight on the Swelling Pressure–Suction Relationship of Compacted Bentonite during Hydration. *Materials* 16: 5403. <https://doi.org/10.3390/ma16155403>

Wieczorek, K., Emmerich, K., Nagel, T., Bakker, E., Diedel, R., Furche, M., García-Siñeriz, J. L., Glaubach, U., Hesser, J., Hinze, M., Hofmann, M., Jaeggi, D., Königer, F., Mayor Zurdo, J. C., Räbiger, L., Mazón, M. R., Rölke, C., Schädle, P., Schumann, R., ... Yeatman, R. (2024). Sandwich-HP. Vertical Hydraulic Sandwich Sealing System. Final Report. GRS. <https://www.grs.de/en/aktuelles/publikationen/grs-745>

Winhausen, L., Khaledi, K., Jalali, M., Bretthauer, M., Amann, F. 2023. The Anisotropic Behavior of a Clay Shale: Strength, Hydro-Mechanical Couplings and Failure Processes. *Journal of Geophysical Research: Solid Earth*, 128, 11. <https://doi.org/10.1029/2023JB027382>

Yamamoto, S., Kumagai, M., Koga, K., Sato, S. 2015. Mechanical stability of engineered barriers in a subsurface disposal facility during gas migration based on coupled hydromechanical modelling. *Geological Society, London, Special Publications* 415: 213 - 224. <https://doi.org/10.1144/SP415.11>

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Yang, J., Cui, Y.-J., Mokni, N., Zhang, Z. 2022. Elastic and cracking behaviour of MX80 bentonite pellet at various suctions in uniaxial compression. *Géotechnique* 74(11): 1143–1154. <https://doi.org/10.1680/jgeot.21.00142>

Yang, J.W., Cui, Y.J., Mokni, N. and Ormea E. 2023a. Investigation into the mercury intrusion porosimetry (MIP) and micro-computed tomography ( $\mu$ CT) methods for determining the pore size distribution of MX80 bentonite pellet. *Acta Geotechnica* 19: 85–97. <https://doi.org/10.1007/s11440-023-01863-y>

Yang, J.-W., Cui, Y.-Y., Mokni, N., Ormea, E. 2023b. Microstructural investigation into the damage behaviour of MX80 bentonite pellet under wetting/drying path using Mercury Intrusion Porosimetry (MIP) and Micro-Computed Tomography ( $\mu$ CT). *Engineering Geology* 322: 107168. <https://doi.org/10.1016/j.enggeo.2023.107168>

Yang, J.-W., Cui, Y.-J., Mokni, N., Ormea, E. 2024a. Investigation into the mercury intrusion porosimetry (MIP) and micro-computed tomography ( $\mu$ CT) methods for determining the pore size distribution of MX80 bentonite pellet. *Acta Geotechnica* 19(1): 85–97. <https://doi.org/10.1007/s11440-023-01863-y>

Yang, J.W., Cui, Y.J., Mokni, N. 2024b. Structural evolutions of MX80 bentonite pellet/powder mixtures under wetting and suction-controlled oedometer loading. *Canadian Geotechnical Journal* 61(3): 519 – 533. <https://doi.org/10.1139/cgj-2023-0033>

Yang, J.W., Cui, Y.J., Mokni, N., Zhang Z. 2024c. Elastic and cracking behaviour of MX80 bentonite pellet at various suctions in uniaxial compression. *Géotechnique* 74(11): 1143–1154. <https://doi.org/10.1680/jgeot.21.00142>

Yang, J.-W., Cui, Y.-J., Mokni, N., Wang, H. 2024d. A triple-microstructure hydro-mechanical constitutive damage model for compacted MX80 bentonite pellet/powder mixture. *International Journal for Numerical and Analytical Methods in Geomechanics* 48(6): 1654–1680. <https://doi.org/10.1002/nag.3670>

Yigzaw, Z.G., Cuisinier, O., Massat, L., Masrouri., F. 2019. Role of different suction components on swelling behavior of compacted bentonites. *Applied Clay Science* 120: 81-91. <https://doi.org/10.1016/j.clay.2015.11.022>

Zaidi, M., Mokni, N., Dymitrowska, M., Liu, K. 2024. Analyzing structural changes induced by gas migration in heterogeneous pellet/powder bentonite mixtures through X-ray computed micro-tomography. *Journal of Rock Mechanics and Geotechnical Engineering*. In press

Zeng, Z., Cui, Y.-J., Zhang, F., Conil, N., Talandier, J. 2019. Investigation of swelling pressure of bentonite/claystone mixture in the full range of bentonite fraction. *Applied Clay Science* 178: 105137. <https://doi.org/10.1016/j.clay.2019.105137>

Zeng, Z., Cui, Y.-J., Conil, N., Talandier, J. 2020a. Analysis of boundary friction effect on the homogenization process of compacted bentonite/claystone mixture with technological voids upon hydration. *Acta Geotechnica* 16(2), 525–533. <https://doi.org/10.1007/s11440-020-01048-x>

Zeng, Z., Cui, Y.-J., Conil, N., Talandier, J. 2020b. Experimental Investigation and Modeling of the Hydraulic Conductivity of Saturated Bentonite–Claystone Mixture. *International Journal of Geomechanics* 20(10). [https://doi.org/10.1061/\(asce\)gm.1943-5622.0001817](https://doi.org/10.1061/(asce)gm.1943-5622.0001817)

Zeng, Z., Cui, Y.-J., & Talandier, J. 2021. Compaction and sealing properties of bentonite/claystone mixture: Impacts of bentonite fraction, water content and dry density. *Engineering Geology*, 287: 106122. <https://doi.org/10.1016/j.enggeo.2021.106122>

**EURAD-2** Deliverable 10.1 – Initial State-of-the-Art on Hydraulic Mechanical Chemical Evolution of Bentonite for Barriers Optimisation

Zeng, Z., Cui, Y.-J., Talandier, J. 2022. Effect of water chemistry on the hydro-mechanical behaviour of compacted mixtures of claystone and Na/Ca-bentonites for deep geological repositories. *Journal of Rock Mechanics and Geotechnical Engineering* 14(2): 527–536. <https://doi.org/10.1016/j.jrmge.2021.09.016>

Zeng, H., Gonzalez-Blanco, L., Romero, E., Fraccica, A. 2023. The importance of the microstructure on hydro-mechanical behaviour of compacted granular bentonite. *Applied Clay Science*, 246. <https://doi.org/10.1016/j.clay.2023.107177>

Zhang, C.-L. 2014. Characterization of excavated claystone and claystone–bentonite mixtures as backfill/seal material. *Geological Society, London, Special Publications* 400(1): 323–337. <https://doi.org/10.1144/sp400.28>

Zhang, Z., Ye, W.-M., Liu, Z.-R., Wang, Q., Cui, Y.-J., 2020. Mechanical behavior of GMZ bentonite pellet mixtures over a wide suction range. *Engineering Geology* 264: 105383. <https://doi.org/10.1016/j.enggeo.2019.105383>

Zhang, C.-L. 2021. Deformation and water/gas flow properties of claystone/bentonite mixtures. *Journal of Rock Mechanics and Geotechnical Engineering*, 13(4): 864–874. <https://doi.org/10.1016/j.jrmge.2020.12.003>

Zhang, Z., Cui, Y. J., Yang, J., Mokni, N., Ye, W. M., He, Y. 2022a. Water retention and compression behavior of MX80 bentonite pellet. *Acta Geotechnica* 17(6): 2435-2447. <https://doi.org/10.1007/s11440-021-01428-x>

Zhang, Z., Cui, Y. J., Yang, J. W., Mokni, N. 2022b. Investigation into the hydro-mechanical behaviour and microstructural evolution of MX80 bentonite pellet upon wetting/drying. *Construction and Building Materials* 345: 128319. <https://doi.org/10.1016/j.conbuildmat.2022.128319>

Zhang, C.-L., Talandier, J. 2023. Self-sealing of fractures in indurated claystones measured by water and gas flow. *Journal of rock mechanics and geotechnical engineering* 15(1): 227-238. <https://doi.org/10.1016/j.jrmge.2022.01.014>

Zhu, C.-M., Ye, W.-M., Chen, Y.-G., Chen, B., Cui, Y.-J., 2013. Influence of salt solutions on the swelling pressure and hydraulic conductivity of compacted GMZ01 bentonite. *Engineering Geology* 166: 74–80. <https://doi.org/10.1016/j.enggeo.2013.09.001>