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*Supplement of*

## **Can we safely go to 200 °C? An integrated approach to assessing impacts to the engineered barrier system in a high-temperature repository**

**Jens T. Birkholzer et al.**

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# Can We Safely Go to 200 °C? An Integrated Approach to Assessing Impacts to the Engineered Barrier System in a High-Temperature Repository

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# Bentonite Backfill in Repository Tunnels

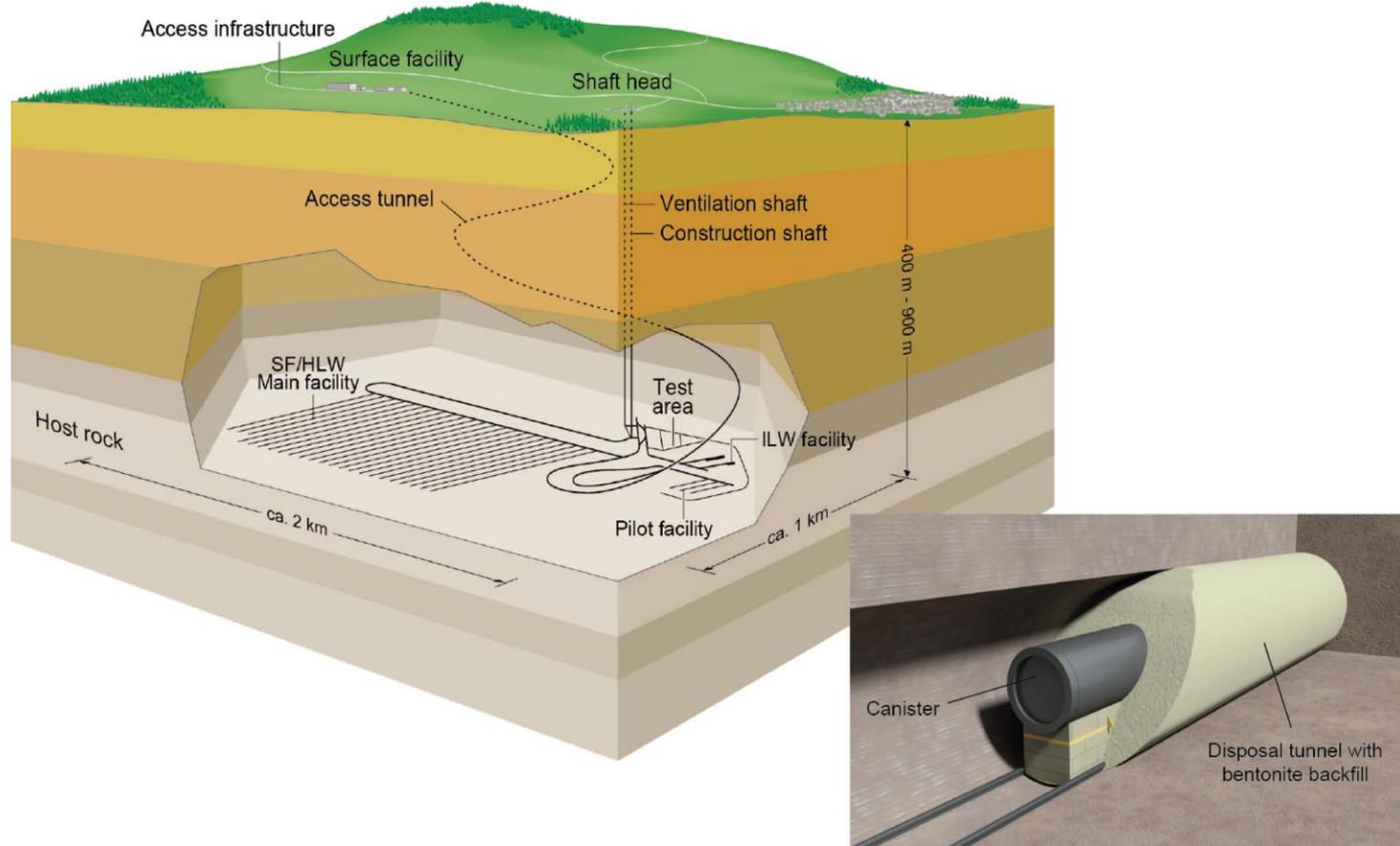
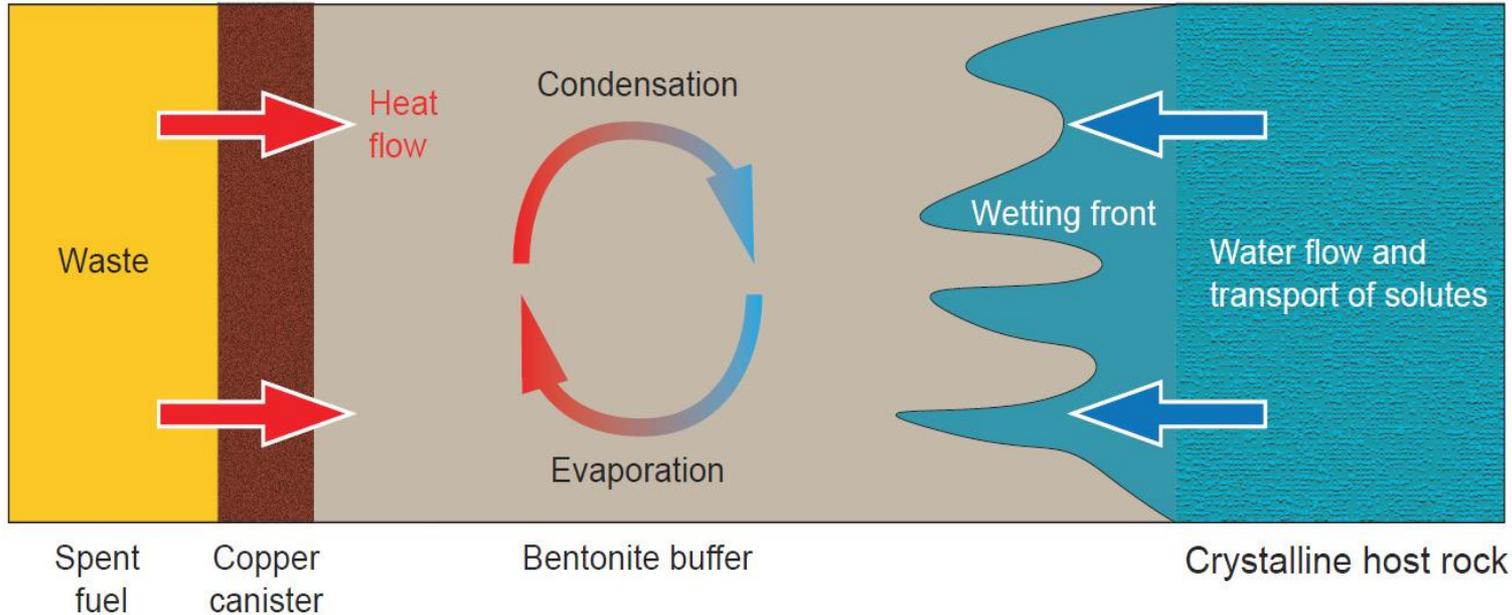


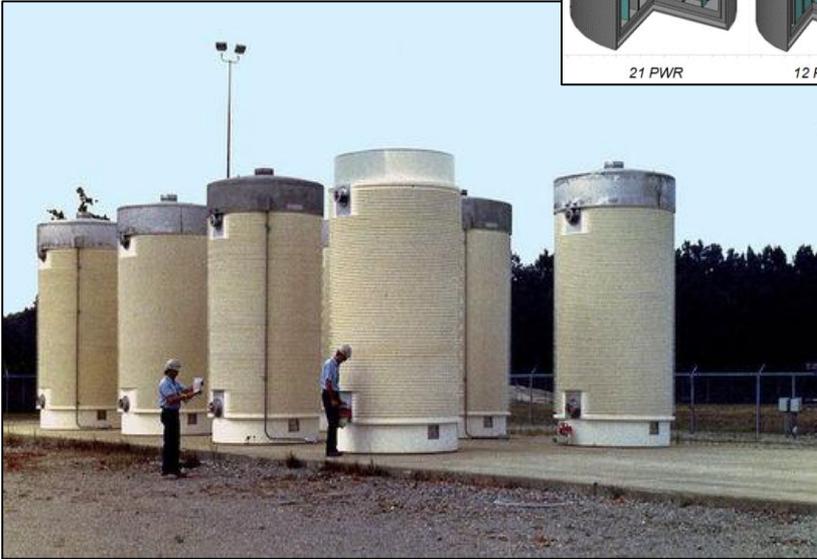
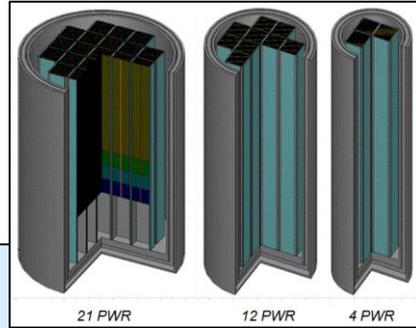
Figure: Courtesy of NAGRA

# THMC Perturbations in Bentonite Barrier

- **Thermal process:** Heat emission from waste and transport through barriers
- **Hydrological process:** Transient de-saturation and re-saturation of bentonite and host rock
- **Mechanical process:** Stress evolution from swelling and heat
- **Chemical process:** Mineralogical changes, solute transport, and radionuclide migration



# The Effect of Higher Temperatures: Going from 100°C Max to 200°C Max



**Dual Purpose Canisters: From Dry-Cask Surface Storage to Disposal in Geologic Repositories?**

- **Enhanced THM Effects:**

- Thermal pressurization (damage?) and complex moisture transport processes, including convection of vapor
- Delayed saturation of bentonite
- Strong gradient with heterogeneous time-dependent density evolution (differential swelling)

- **Possible Geochemical Effects:**

- Illitization and other alterations, possibly affecting swelling properties
- Possible impact on diffusion and sorption properties

# Integrated Research Plan: Going from 100 °C Max to 200 °C Max

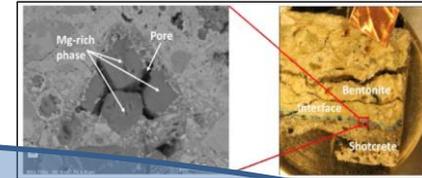
Can waste package and bentonite buffer temperature safely be raised to 200°C, without causing performance relevant alteration and damage in barrier behavior?

## Fundamentals of Physico-Chemical Alterations

- Laboratory imaging/characterization of heated samples
- Detailed coupled processes modeling of individual components



### Micro-structural analysis

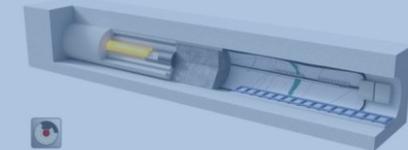


## Barrier System Behavior

- Laboratory or *in situ* testing of barrier systems
- Validation of predictive process models for system behavior
- Predictions of engineered and natural barrier perturbations
- Optimization studies (e.g., alternative backfill materials)



### Lab and field experiments (HotBENT)

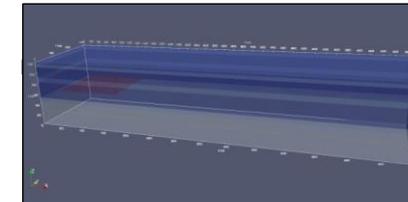


## Performance Assessment

- Include high temperature effects in performance assessment models
- Determine scenarios and parameters with significant impact on high-temperature repository performance
- Conduct performance assessment for different thermal designs



### Performance assessment modeling



**Can waste package and bentonite buffer temperature safely be raised to 200°C, without causing performance relevant alteration and damage in barrier behavior?**

- **FEBEX-DP: Long-term demonstration experiment at 100 °C**



- **Exploratory simulations for 200 °C: THMC modeling of generic repository**

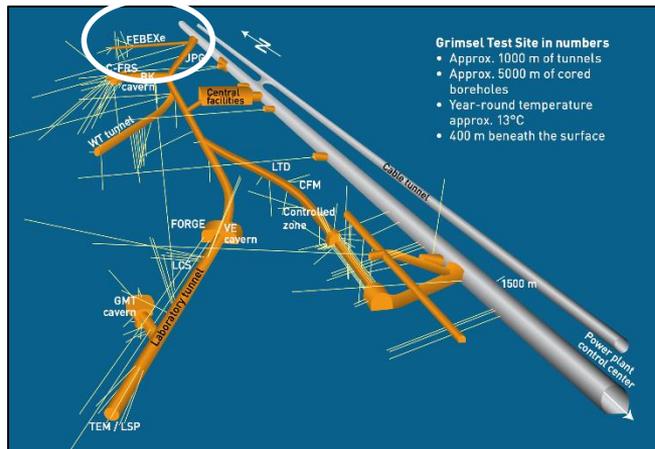


- **Laboratory testing: High-temperature column tests at 200 °C (ongoing)**



- **HotBENT: Long-term demonstration experiment at 200 °C (just started)**

# FEBEX Experiment at Grimsel Test Site in Switzerland

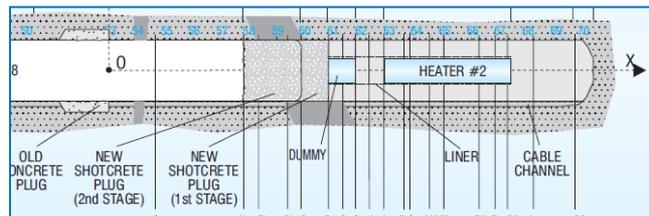


- What are the bentonite and near-field rock properties during & after hydration and thermal alteration?
- Will bentonite blocks become a homogeneous buffer material?
- Do we have suitable prediction tools for coupled behavior?

Heating Starts at 100°C in 1997



Heater #1 Dismantling in 2002

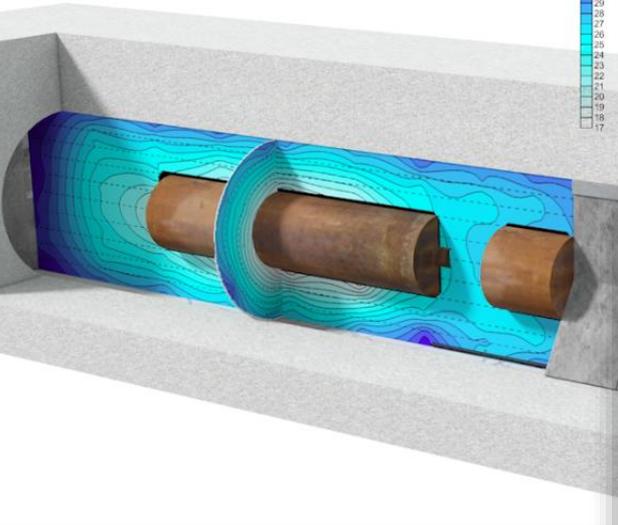


Heater #2 Dismantling in 2015

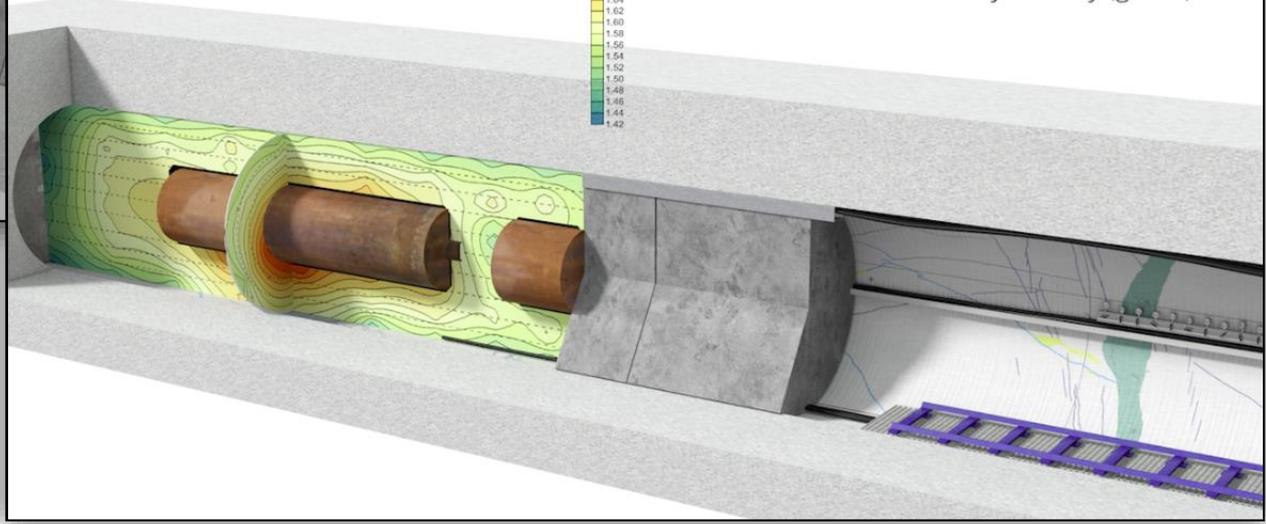
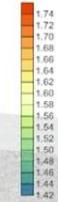


# 3D Moisture Distribution and Bentonite Density After 18 Years of Heating

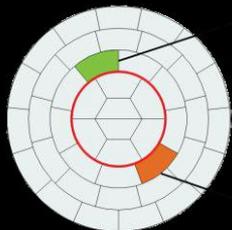
**RESULTS**  
Water Content (%)



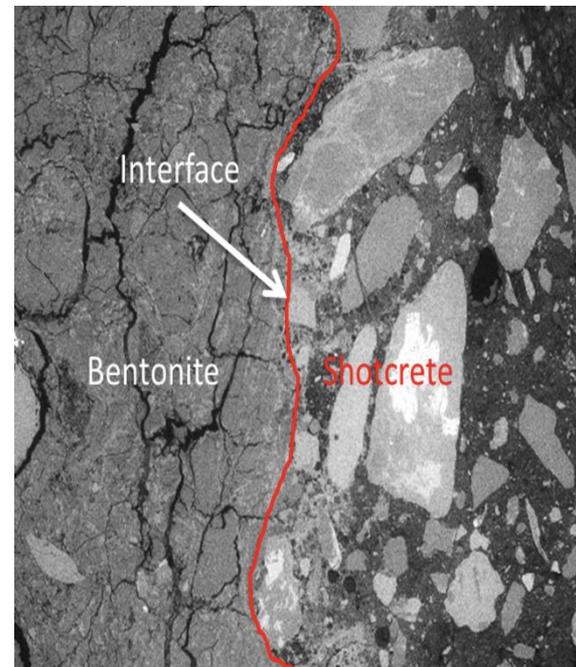
**RESULTS**  
Dry Density ( $\text{g}/\text{cm}^3$ )



## Bentonite-Canister Interface



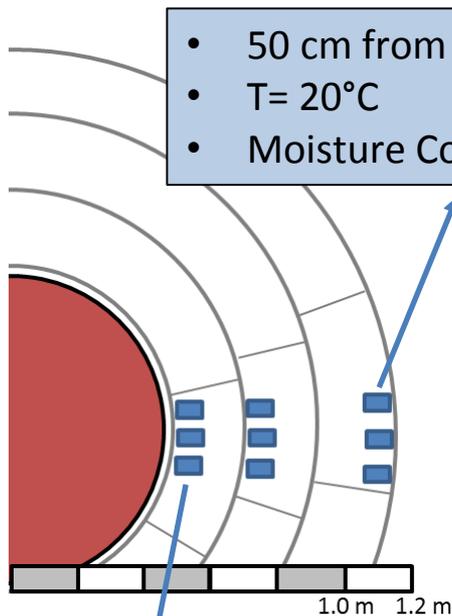
## Bentonite-Shotcrete Interface



# Transport Properties as a Function of Temperature Exposure

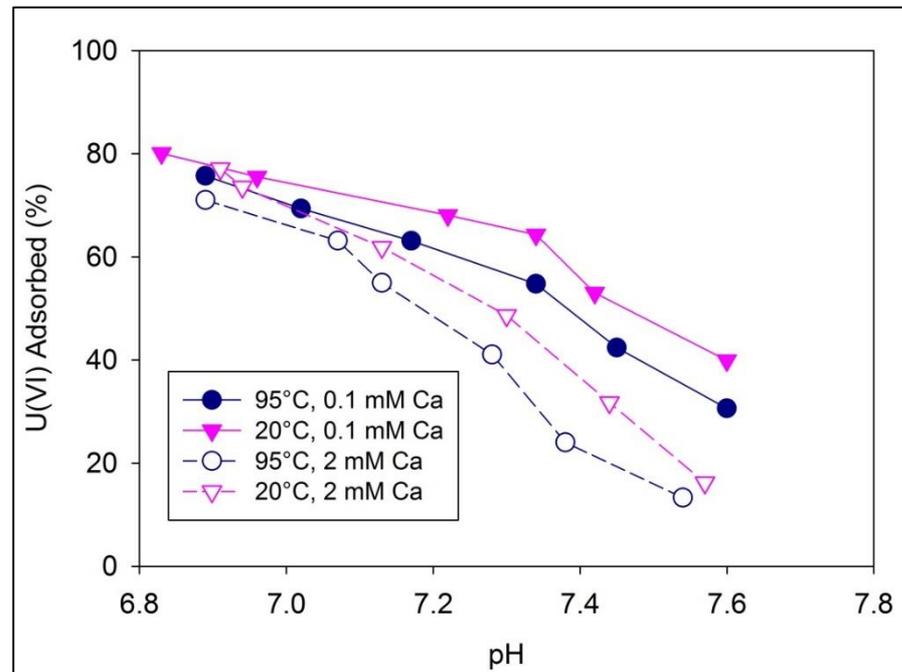
## Cold Zone:

- 50 cm from axis (Section 59)
- $T = 20^{\circ}\text{C}$
- Moisture Content  $\cong 25\%$



## Heated Zone:

- 50 cm from axis (Section 48)
- $T = 95^{\circ}\text{C}$
- Moisture Content  $\cong 18\%$



## Up to 10% lower U(VI) adsorption on altered bentonite

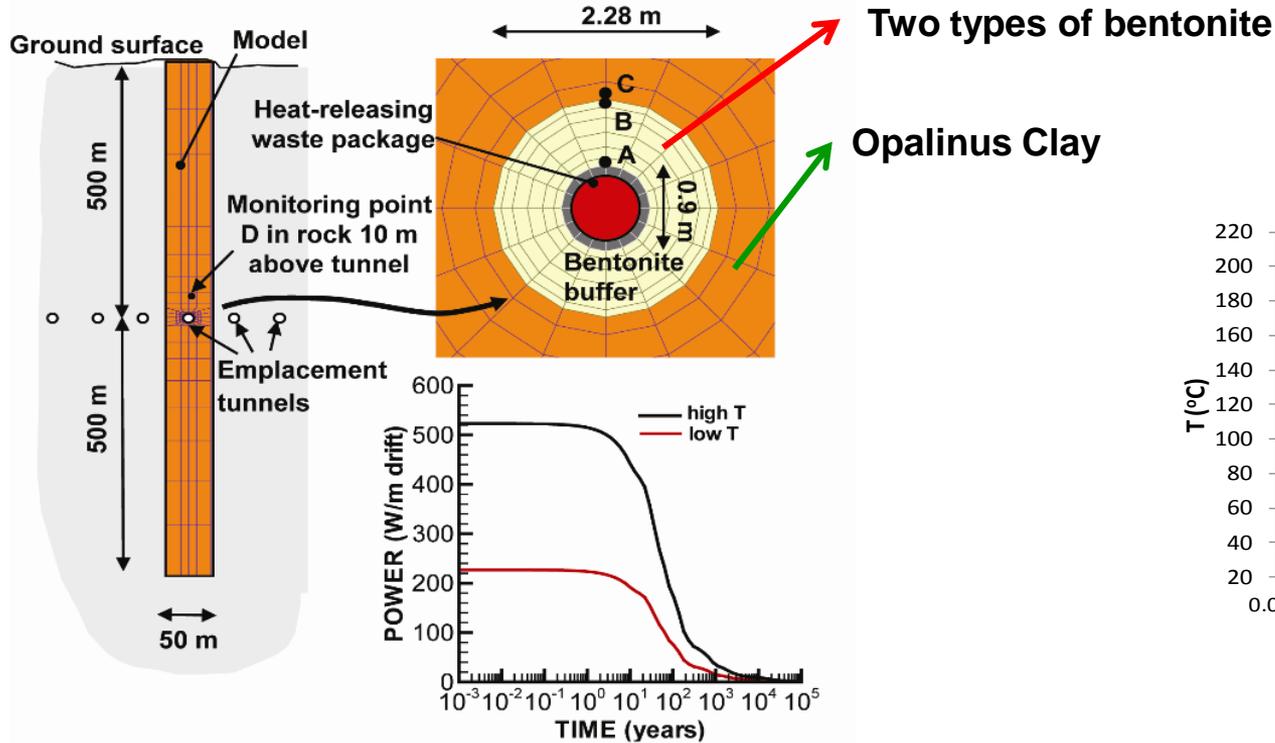
- Adsorption is lower in presence of 2 mM Ca compared to 0.1 mM Ca.
- Adsorption decreases as pH increases.

# Synthesis of FEBEX-DP Findings

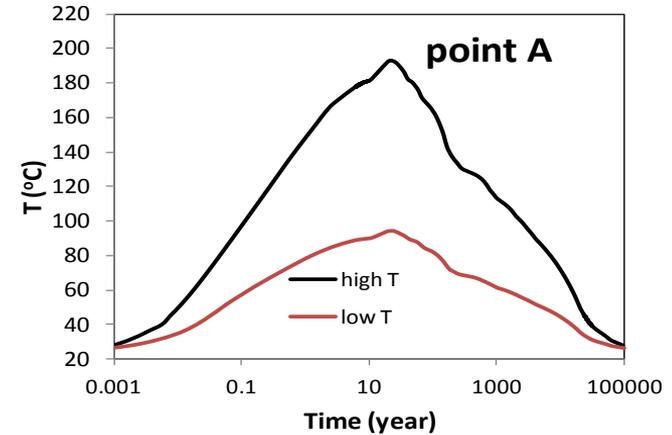
<b>Safety relevant attributes</b> (NTB 14-12; Leupin et al. 2014)	<b>FEBEX/FEBEX-DP contribution</b> (depending on organizational requirements)
Low hydraulic conductivity	Properties not altered, diffusion dominated (10 <sup>-12</sup> – 10 <sup>-14</sup> m/s)
Chemical retention of RN	Sorption properties unlikely altered (no mineral transformation)
Sufficient density	Density gradients evolved, mean 1.59 g/cm <sup>3</sup>
Sufficient swelling pressure (avoid EDZ extension)	~6 MPa (for 1.6 g/cm <sup>3</sup> ); lab-scale confirmed in 1:1 experiment
Mechanical support (canister sinking, stress buffering)	Sufficient support (↑ 6; ↓ -17; ↔ 6 mm - liner, with low confidence)
Sufficient gas transport capacity (gas transport without compromising hydraulic barrier)	not relevant
Minimise microbial corrosion	No indication of MIC on canister, No unambiguous indication of MIC on instruments
Resistance to mineral transformation	No significant transformations detected
Sufficient heat conduction	As predicted

(Kober et al., FEBEX-DP Dismantling Synthesis Report,)

# Exploratory THMC Simulations for Long-Term Evolution of Bentonite at 200 °C



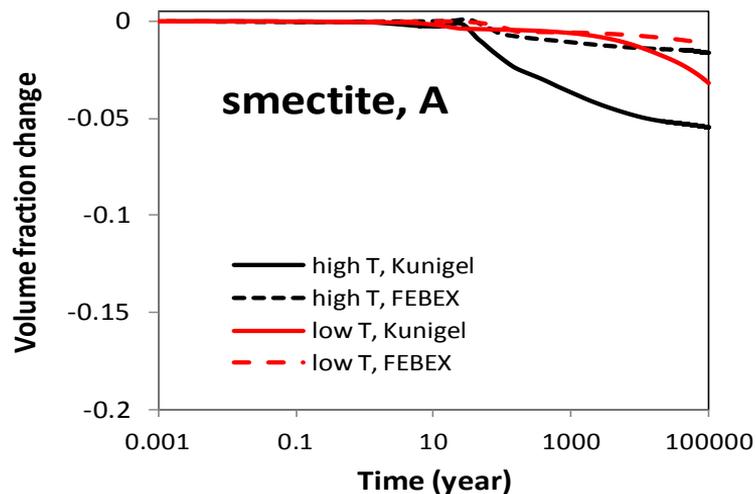
Two cases for comparison:  
"high T" and "low T"



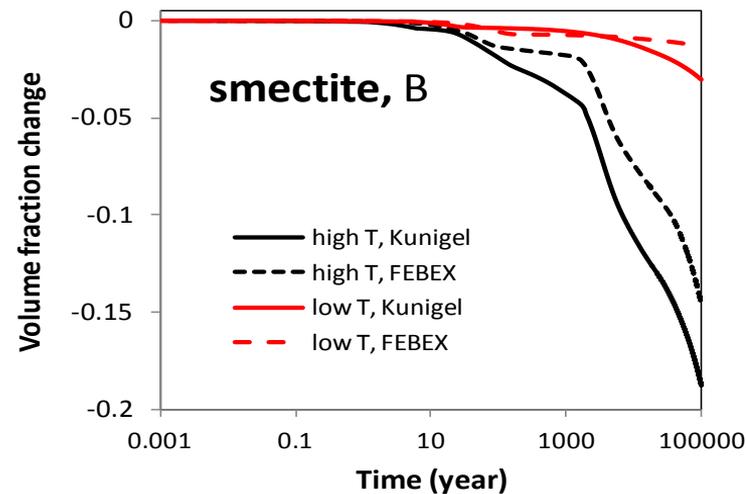
- Chemical model: aqueous complexation, minerals dissolution/precipitation and ion exchange
- Bentonite swelling model accounts for chemical alteration of clay minerals

# Exploratory Simulations for Long-Term Evolution of Bentonite

## A: Bentonite near waste package



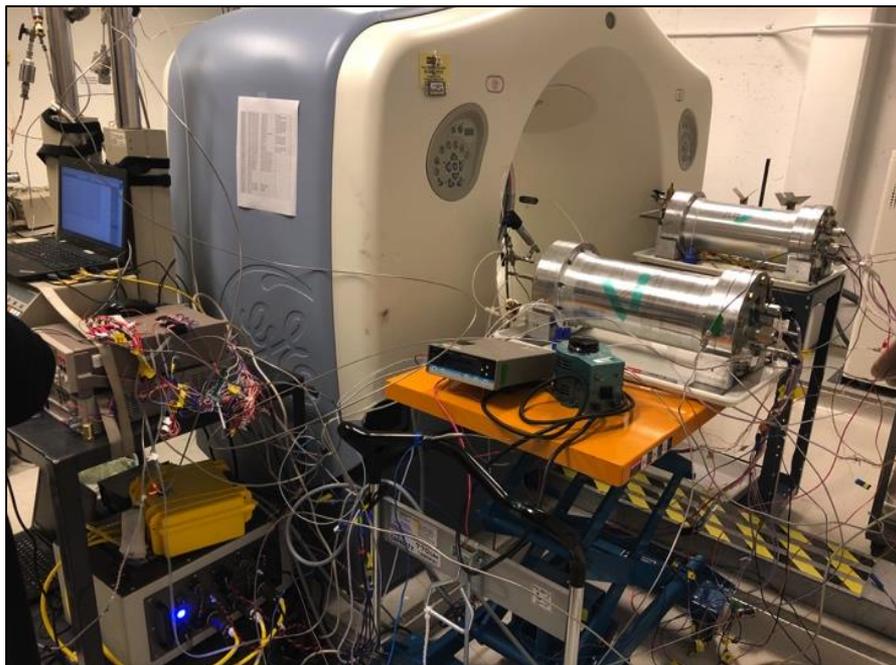
## B: Bentonite near host rock



## Key findings:

- Modest illitization occurs, temperature plays a key role, and bentonite-rock interaction is important
- Type of bentonite matters and supply of K and Al from surrounding groundwater is the key
- Swelling stress decreases as a result of chemical changes and such decrease varies case by case (our results suggest swelling stress decrease of up to 12% for FEBEX bentonite and 45% for Kunigel)

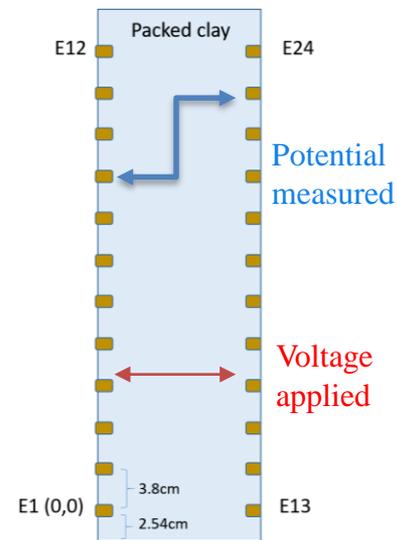
## CT Scanning



## Electrical Resistivity Tomography

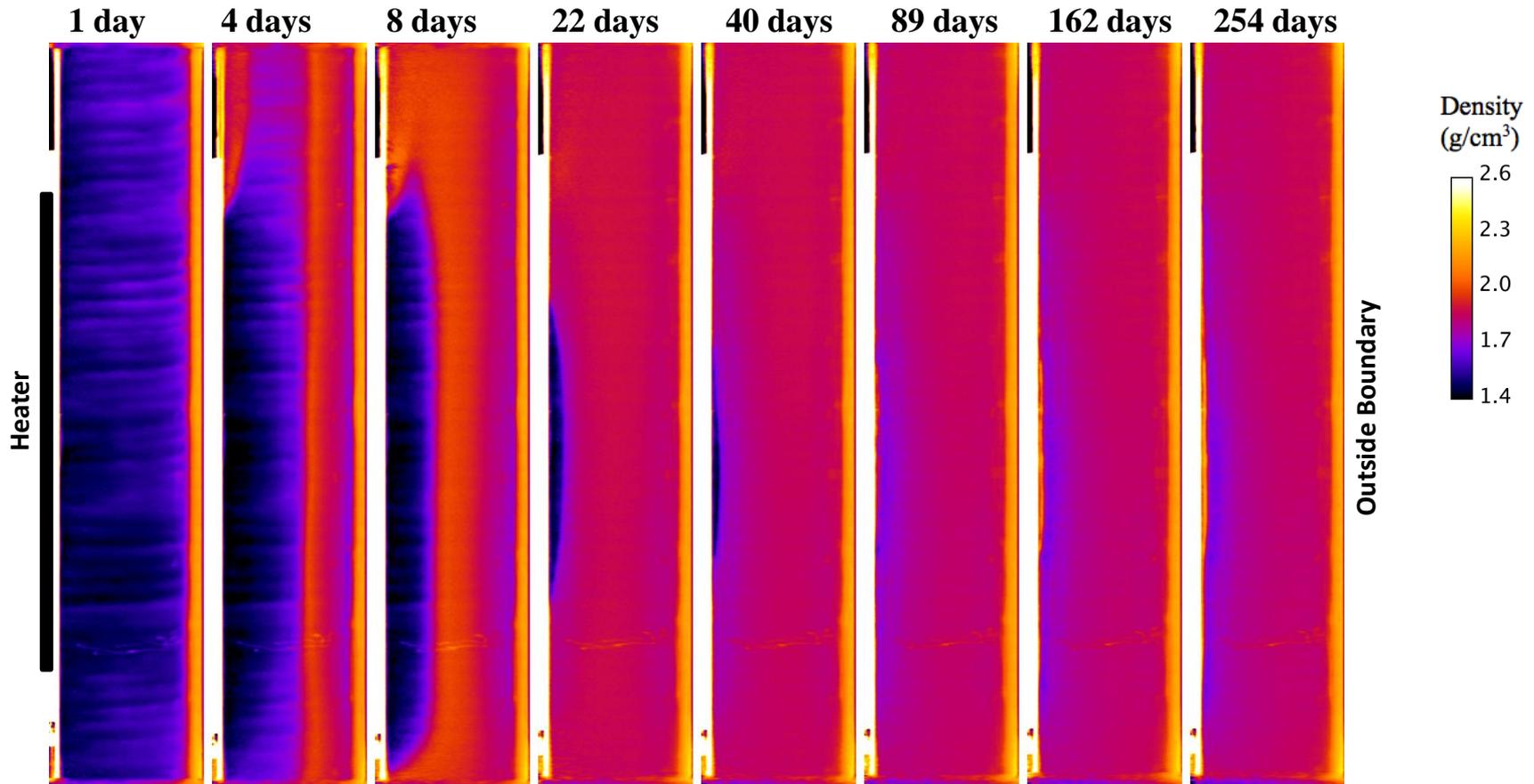
Packed-in cross-well electrodes array

Time-lapse in situ survey and inversion



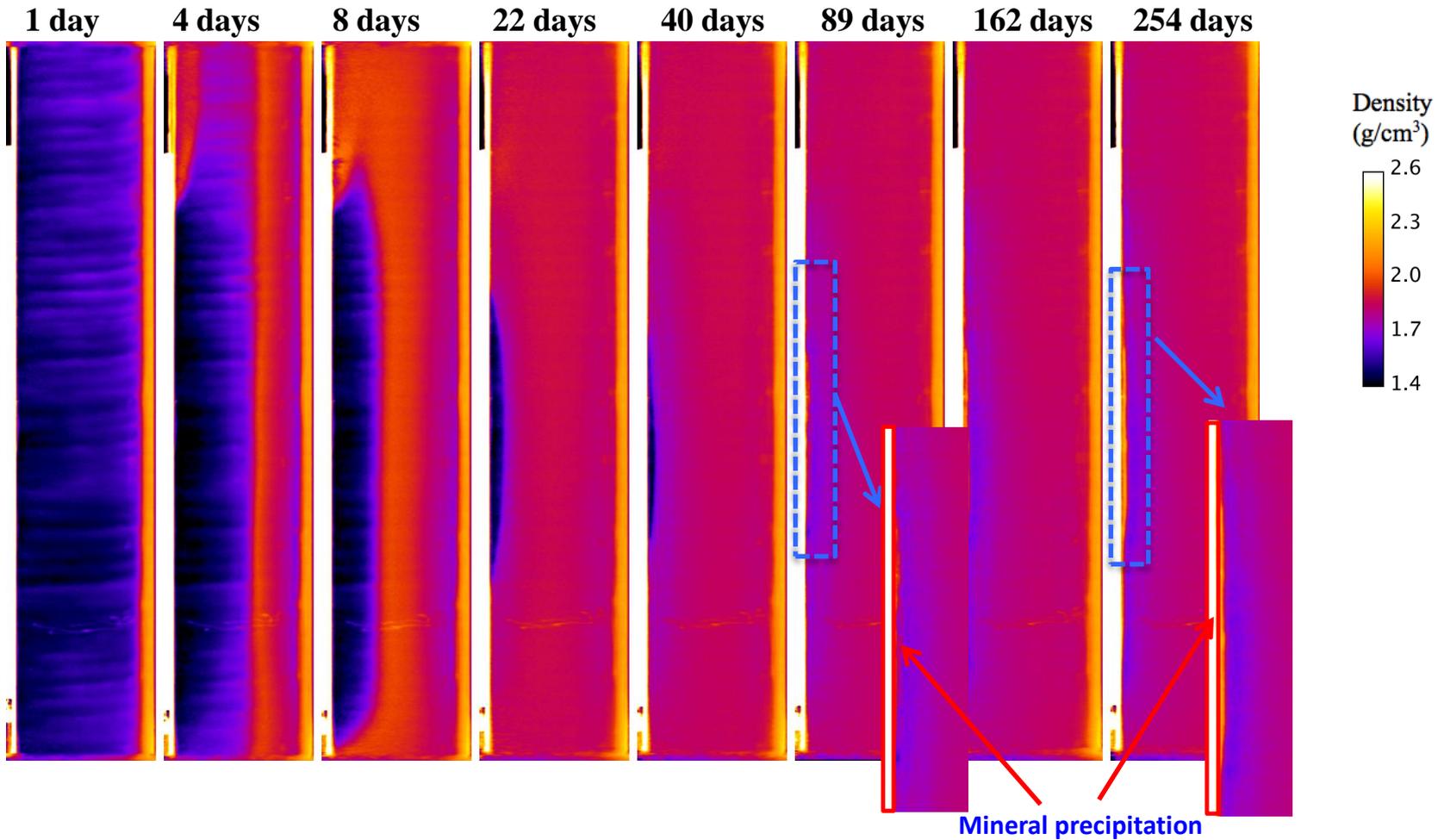
- Sensors for fluid pressure, temperature, stress
- Flow rates and chemistry of inflow/outflow
- Dismantling and sampling planned

# Long-term Density Evolution From CT Imaging

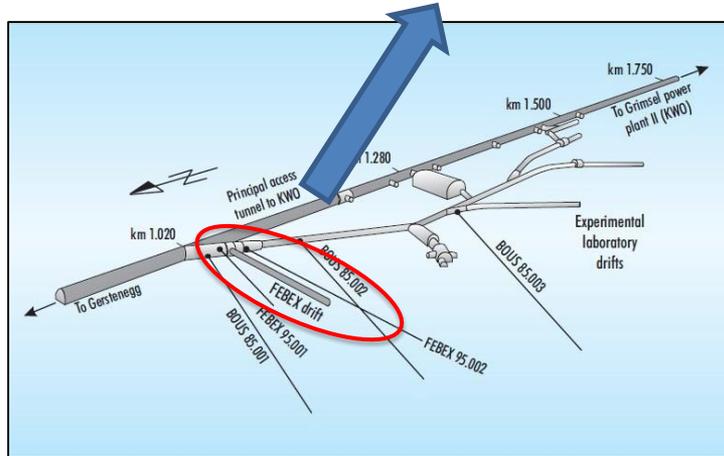


Radially averaged CT density map for the heated column. The heater in the center is shown on the left of each column.

# Long-term Density Evolution From CT Imaging



# High-Temperature *In Situ* Test at Grimsel Test Site: HotBENT



## Timeline Towards HotBENT:

- **2013:** first discussions with international partners about high-T emplacement
- **2015:** joint paper with NAGRA about research needs, including *in situ* test
- **2016:** international coalition building
- **2018:** official HotBENT partnership (five full & four associated partners)
- **Since 2019:** preparation and installation of *in situ* test (Nagra leadership)
- **2021:** HotBENT heaters turned on



## ANS 2015 International High-Level Radioactive Waste Management Conference

April 12-16, 2015 • Charleston Marriott • Charleston, SC

**“Real World Solutions for Achieving Disposal of Used Fuel and HLW through Integrated Management”**

**CALL FOR PAPERS – Abstract deadline: September 2, 2014**

THMC behavior of clay-based barriers under high temperature – from laboratory to URL scale

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\*\* Lawrence Berkeley National Laboratory, 1 Cyclotron Road, Berkeley CA, 94707, USA

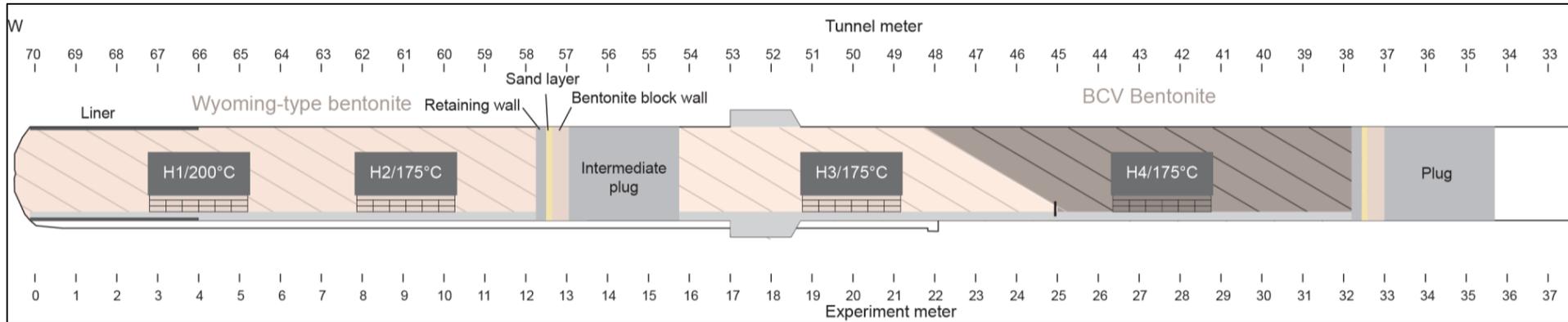
### ABSTRACT

International disposal programs have been investigating if clay-based barriers can withstand temperatures higher than the bentonite performance assumed in repository designs. For example, the Swiss disposal program is investigating geological disposal of large spent nuclear fuel canisters currently in dry storage.

advanced repository designs. For example, the United States disposal program is investigating the feasibility of direct geological disposal of large spent nuclear fuel canisters currently in dry storage. These canisters are currently in 32 PWR assemblies and recent projections mean that there is significant interest in these canisters. Projections indicate that by 2055, there will be more than 3,000 spent nuclear fuel canisters that will be cleared from the U.S. will be in

**Lead: NAGRA; Partners:  
DOE, SURAO, NUMO, RWM,  
BGR, Obayashi, ENRESA,  
NWMO**

# HotBENT – Modular Design



- **Four modules**
  - ✓ Differences in heating temperature, bentonite, time length and w/o concrete liner
- **Two experimental time lengths**
  - ✓ H3 and H4 will run for 5-10 years; H1 and H2 will run 15-20 years
- **Two bentonites**
  - ✓ Wyoming (MX-80); BCV (Czech Republic bentonite)

# HotBENT – Operational Phase Started September 9, 2021

Feb 2020



Mar 2020



Apr 2020



May 2020



Jun 2020



Jul 2020



Aug 2020



Sep 2020



Oct 2020



Nov 2020



Dec 2020



Jan 2021



Feb 2021



Mar 2021



Apr 2021 ...



# Summary: Going from 100 °C Max to 200 °C Max

- **Generic THMC modeling at higher temperatures (200 °C)**
  - Illitization is enhanced at higher temperature and affected by many chemical factors
  - Smectite dissolution leads to reduction in swelling stress, which varies spatially and temporarily  
Illitization and its effect on swelling stress differs for different type of bentonite
  - Bentonite with high swelling capability may safely sustain higher temperatures and raising maximum temperature to 200 °C seems feasible
- **High-temperature column tests**
  - Frequent CT-scans track well the hydration front and property changes in bentonite
  - Scans show evidence for precipitation of minerals near the heater
  - No evidence for excessive thermal pressurization and mechanical damage
  - Dismantling of hydration test is ongoing
- **Large scale *in situ* demonstration test just started operating at the Grimsel Test Site**

# Thank You !

Funding for this work was provided by the Spent Fuel and Waste Science and Technology Campaign, Office of Nuclear Energy, of the U.S. Department of Energy, under Contract Number DE-AC02-05CH11231 with Lawrence Berkeley National Laboratory.

# High-Temperature Column Tests (Started June 3, 2019)

