U.S. DEPARTMENT OF ENERGY | Office of NUCLEAR ENERGY

Clean. Reliable. Nuclear.

DOE's Engineered Barrier Integrity Activities: Understanding EBS Coupled Processes and Mineral Alterations at High Temperatures: From FEBEX-DP to HotBENT

U.S. Nuclear Waste Technical Review Board Workshop
April 24-25, 2019
Burlingame, CA

Liange Zheng
Staff Scientist
Energy Geosciences Division
Lawrence Berkeley National Laboratory

It Is a Collaborative Effort !

Acknowledgment

- Lawrence Berkeley National Laboratory

  Jens T. Birkholzer, Sharon Borglin, Yiwei Cheng, Chunwei Chou, Chun Chang, Patricia Fox, Benjamin Gilbert, Peter Nico, Matthew Reagan, Joseph Saba, Eric Sonnenthal, Marco Voltolini, Yuxin Wu, Hao Xu

- Sandia National Laboratories

  Carlos Jove-colon, Andrew Knight, Clay Payne

- CIEMAT (Centro de Investigaciones Energéticas Medioambientales y Tecnológicas), Spain

  María Victoria Villar, Ana María Fernández

- NAGRA (Switzerland)

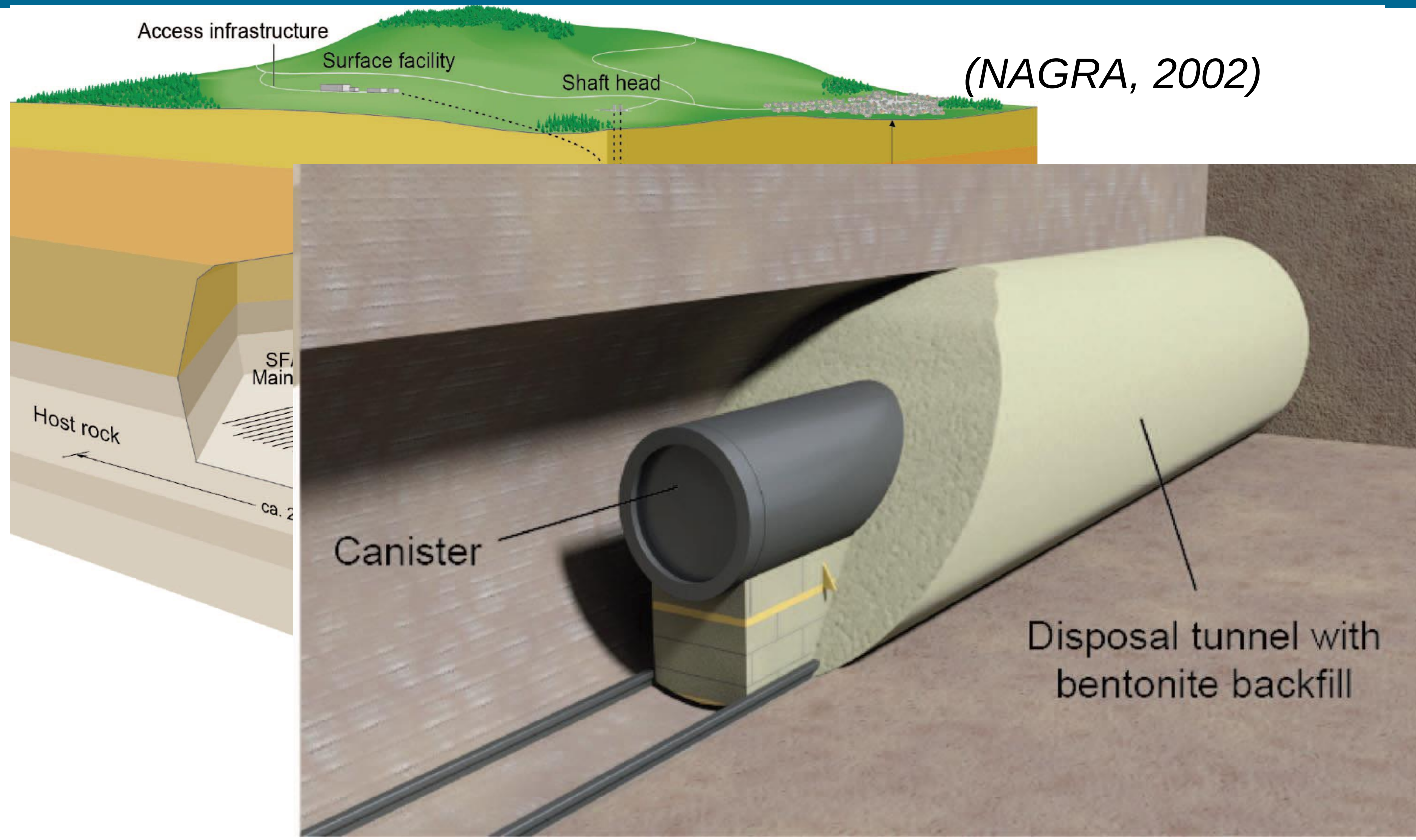
  Florian Kober, Stratis Vomvoris

Engineered Barrier System (EBS) in a Geological Repository

Material:
Partially saturated Bentonite

Featured properties:

- Swelling
- Low permeability
- High retardation capability



(NAGRA, 2002)

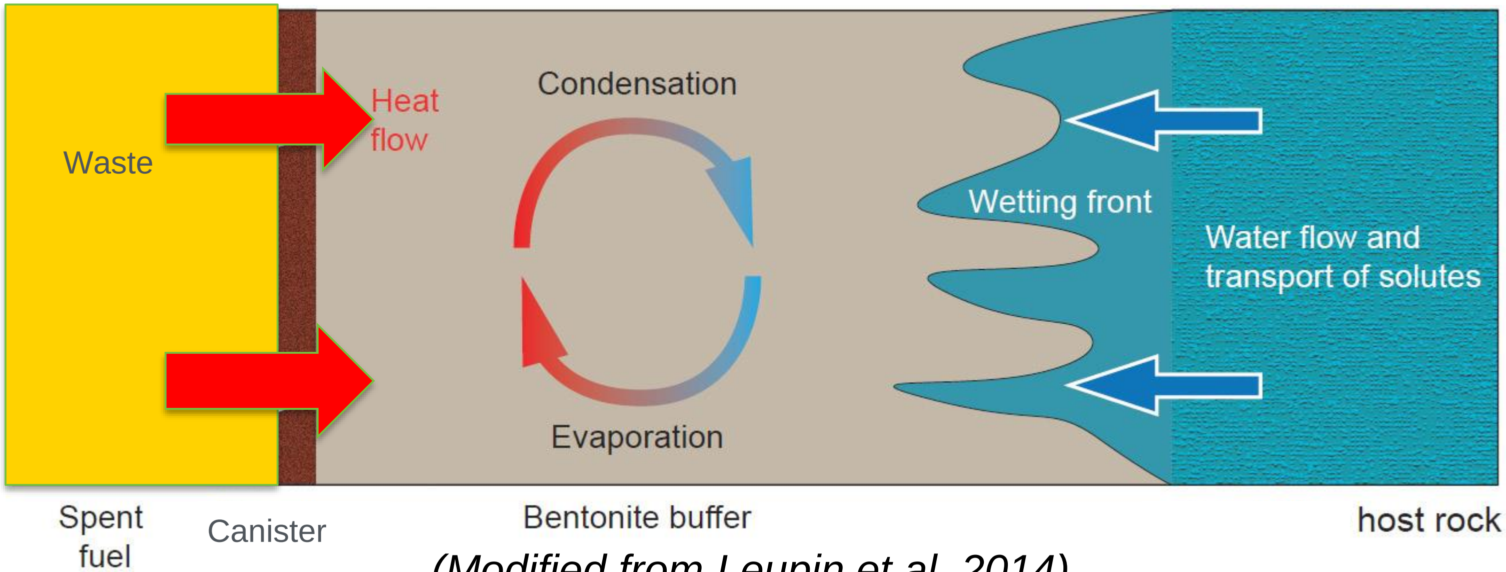
Safety functions:

- Limiting flow and transport in the near field
- Mechanical support including damping rock-shear movement, preventing canister sinking and limiting pressure on canisters
- Reducing microbial activity
- Retarding migration of radionuclides

Processes Involved in Bentonite Evolution (1)

To ensure favorable features of the EBS in the long term, understanding and modeling of early-time thermal, hydrological, mechanical and chemical (THMC) perturbations is critical:

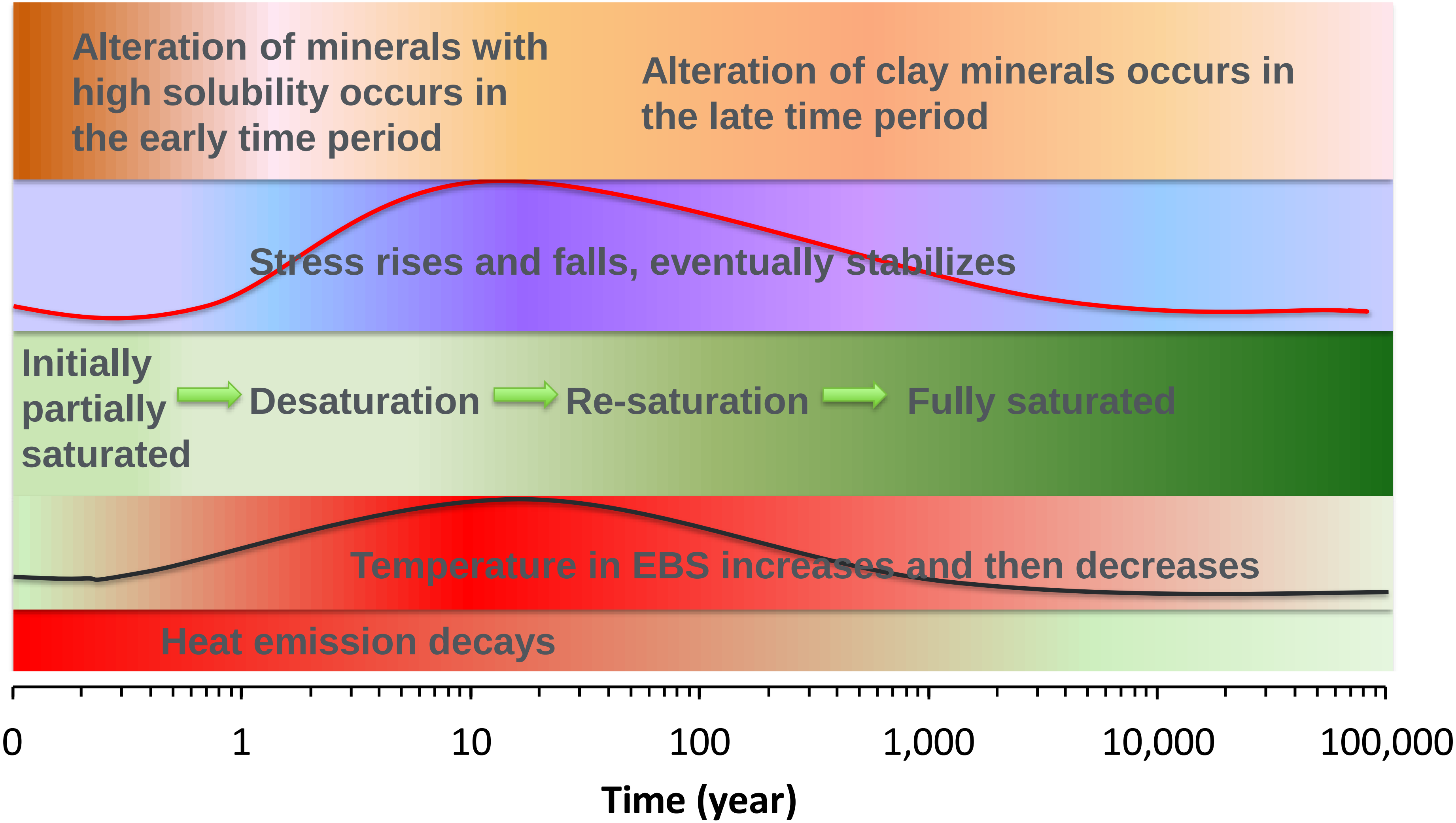
- **Thermal process:** Heat emission from waste and transport through EBS
- **Hydrological process:** Partially saturated bentonite becomes fully saturated after transient de-saturation and re-saturation
- **Mechanical process:** Stress evolution, possibly leading to damage
- **Chemical process:** solute transport, radionuclide migration and mineralogical change



(Modified from Leupin et al, 2014)

Processes Involved in Bentonite Evolution (2)

THMC processes are coupled and evolve temporally and spatially



Key Unknowns and Uncertainties in Understanding and Modeling EBS Evolution

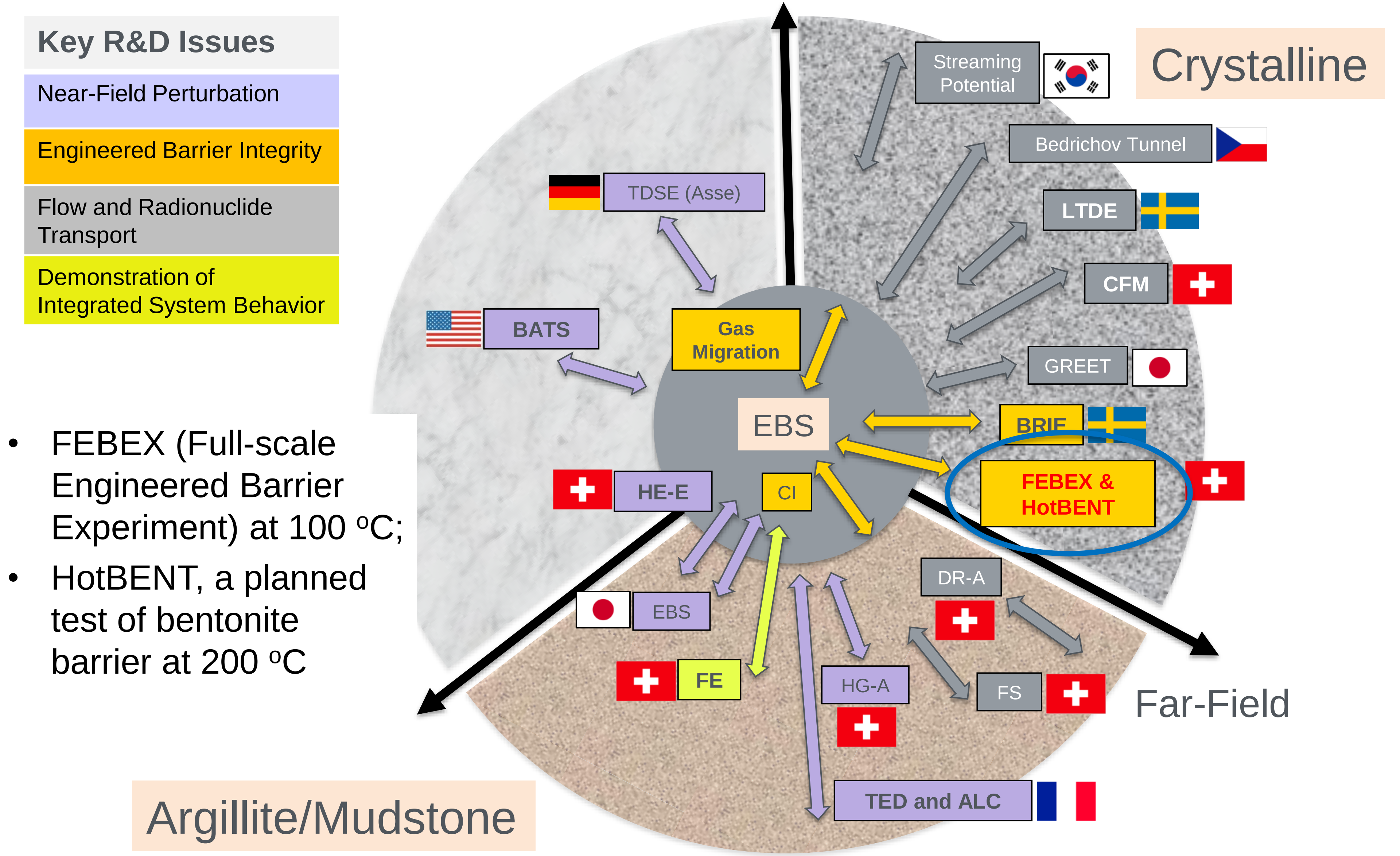
Being able to predict EBS processes is essential for long-term disposal safety evaluation. To build such models, we need to know:

- **What are the key processes that have to be included in the model?**
- **Do we have reliable constitutive relationships and parameters to describe THM processes?**
  - Porosity and permeability changes
  - Stress evolution
- **Do we have reliable chemical models and parameters to describe chemical processes?**
  - Evolution of pore-water geochemistry in bentonite
  - Mineralogical changes in bentonite
  - Retardation capability
  - Interactions between canister/bentonite/host rock

While laboratory experiments are helpful, large scale *in situ* tests are essential for answering unknowns and reducing uncertainties

- **Exploring processes and parameters at full scale of an emplacement tunnel**
- **Testing the "system", with all coupled processes incorporated at scale**
- **Confidence enhancement and ultimate demonstration of modeling capability**

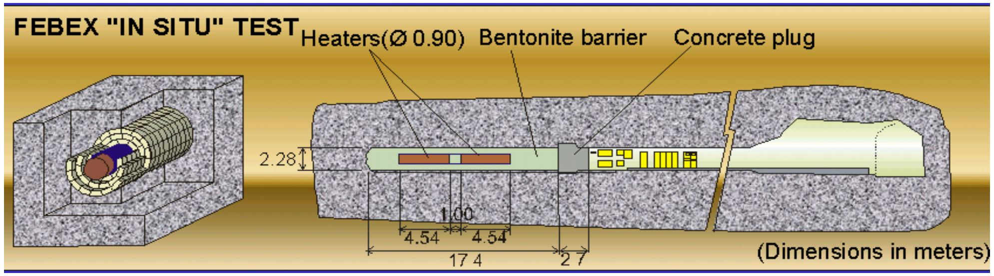
Where FEBEX & HotBENT Fit in the DOE URL Portfolio



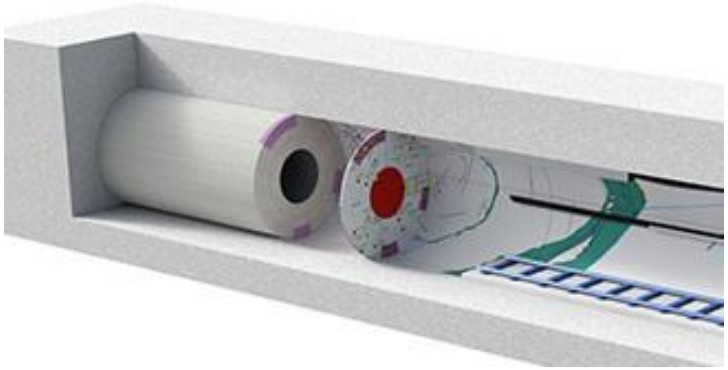
- FEBEX (Full-scale Engineered Barrier Experiment) at 100 ^{o}C;
- HotBENT, a planned test of bentonite barrier at 200 ^{o}C

FEBEX *In Situ* Test

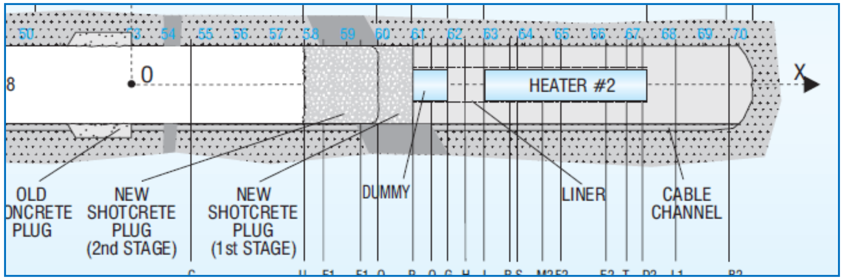
The full-scale *in situ* test is located in Grimsel, Switzerland, heating started in 1997 at 100 °C, as part of FEBEX (Full-scale Engineered Barrier Experiment) *(ENRESA, 2000)*.



In 2015, Dismantling of Heater #2



In 2002, Dismantling of Heater #1



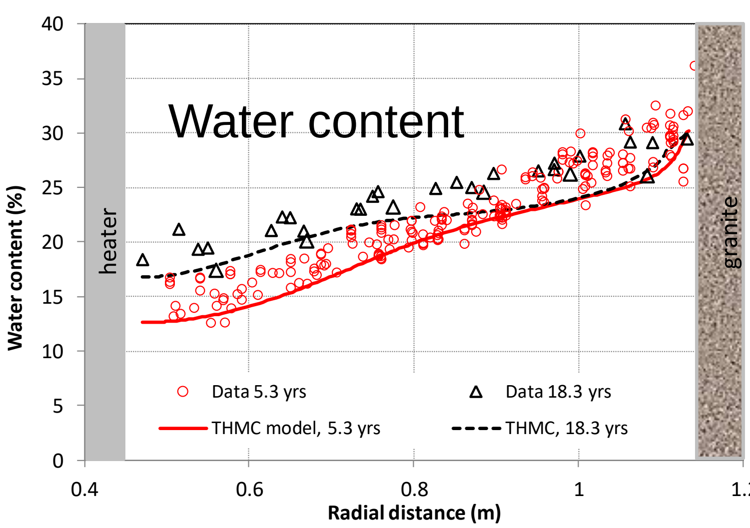
Extensive laboratory tests were carried out to characterize THMC properties of bentonite, concrete, steel liner and granite.

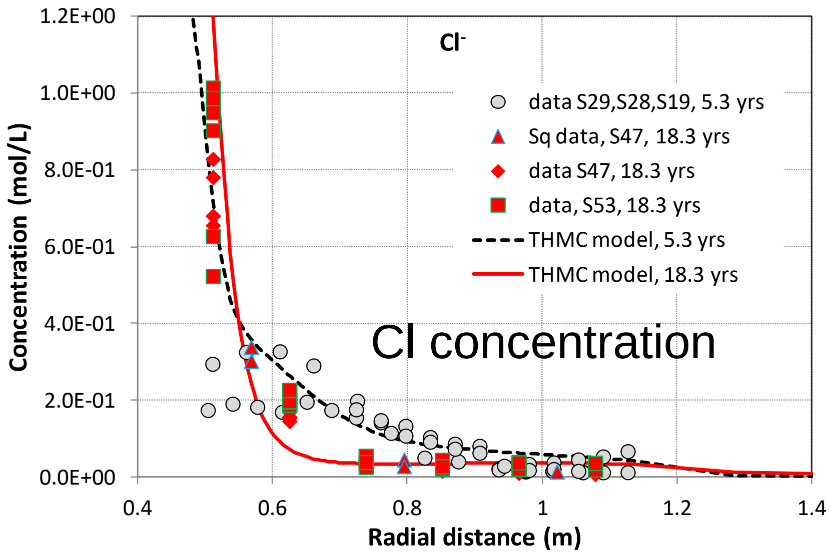
Developing THHC Models for the *In Situ* Test

Developing the model

- **Thermal model:** Heat convection and conduction
- **Flow model:** Two-phase (gas and water) flow
- **Mechanical model:** Poro-elastic using state surface approach
- **Chemical model:** Aqueous complexation, surface complexation, cation exchange and minerals dissolution/precipitation

Testing the model with THMC data





Details are given in the poster by Zheng et al.

Lessons Learned from the *In Situ* Test

- **The *in situ*, 1:1 scale experiment proved to be very useful in terms of engineering aspects, process understanding, monitoring, sampling and modeling** *(Kober et al ., 2017)*
- **Bentonite performed as expected:**
  - Full saturation of entire bentonite barrier was not achieved in 18 years
  - Final dry density varies around 1.6 g/cm^3 depending on water content
  - Where bentonite became fully saturated, the swelling pressure reached the design value (around 5 MPa)
  - Clay minerals underwent minimal mineralogical changes
- **International collaboration among several partner organizations was very beneficial** *(Kober et al ., 2017)*

Lessons learned from Modeling the *In Situ* Test

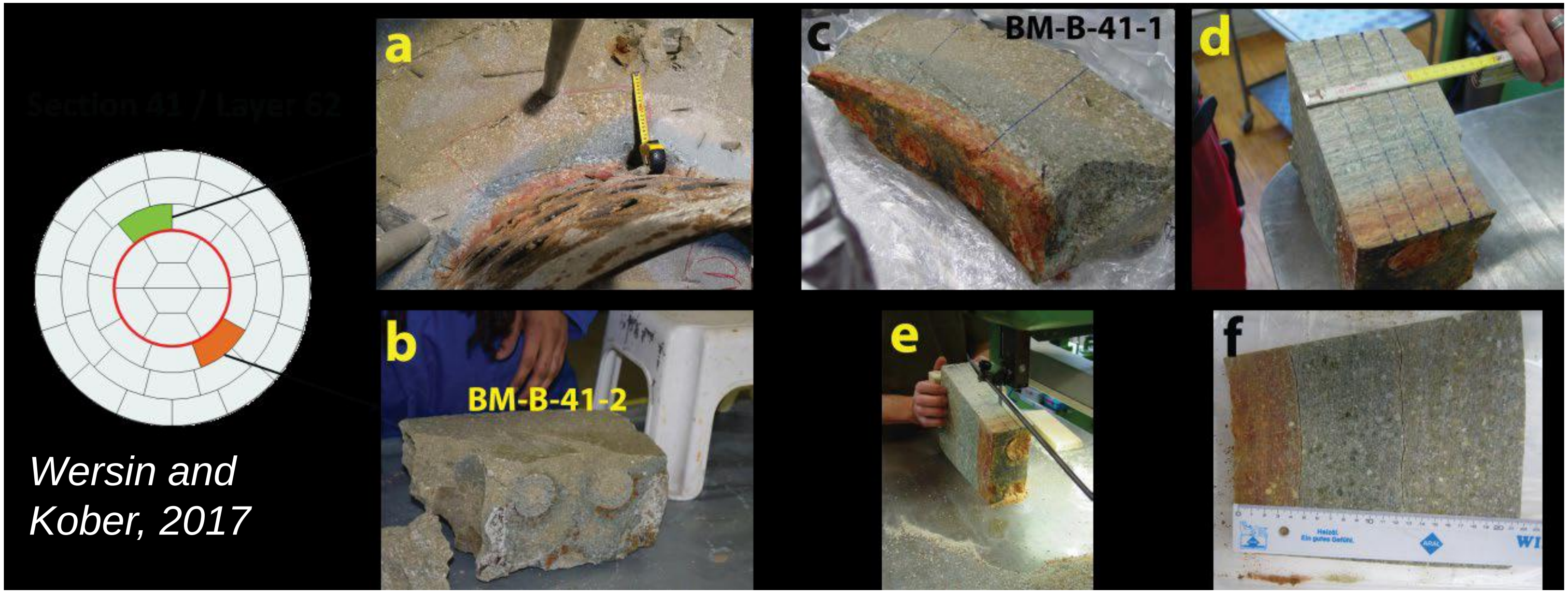
Understanding deepened and modeling capabilities improved

- **Processes needed for modeling bentonite THM evolution:**
  - Thermal conduction and convection
  - Multiphase flow
  - Poro-elasticity
  - Porosity and permeability changes due to swelling
- **About geochemical evolution:**
  - Ion concentrations in pore water are high near the heater, which were largely shaped by transport processes, but also affected by minerals and cation exchange
  - Alterations to carbonate minerals and gypsum happened in the entire bentonite barrier
  - Alterations to clay minerals were moderate and mostly occurred near the heater, which cannot be verified by the data that have large measurement uncertainties
- **Key to increase the robustness of our predictive models for bentonite:**
  - long-term measurements
  - Multiple types of data

More Work is Ahead

Knowledge gaps narrowed, but improvements are certainly warranted:

- Understanding geochemical evolution at interfacial areas: canister/bentonite, concrete/bentonite, granite/bentonite



- Constitutive relationships need to be tested with other conditions (e.g., higher temperature or different clays).
- Understanding could be deepened by multi-scale studies: pore-scale, laboratory and field scale

The Effect of High Temperature (200 ^{o}C)

Motivation

- Dual Purpose Canister disposal can lead to higher temperatures in the engineered and near-field natural barrier system
- Thermal limit of 100 ^{o}C for small PWR canisters might be too limiting

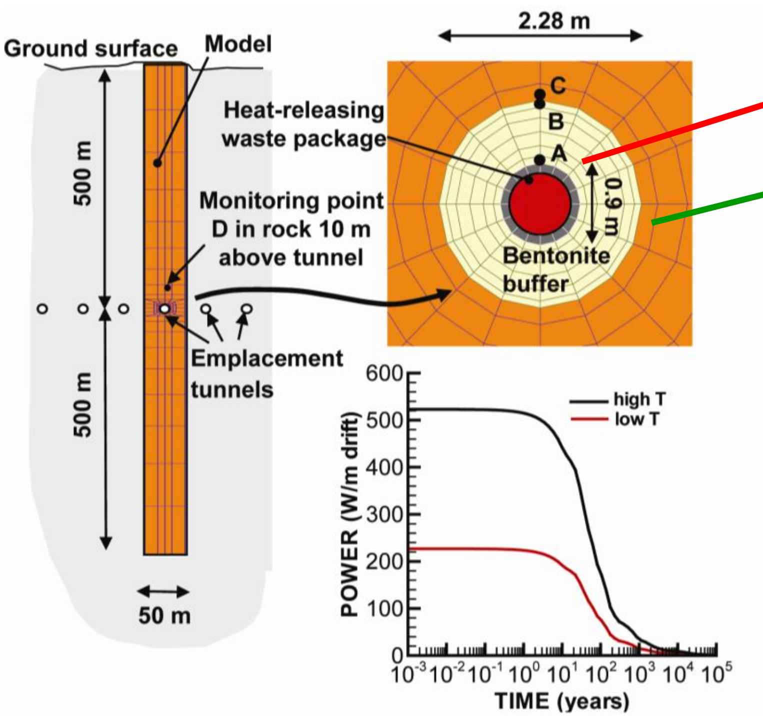
Key knowledge gaps to be narrowed

- When bentonite evolves from partial saturation to full saturation at temperatures up to 200 ^{o}C, how does bentonite change hydrologically and mechanically (e.g., boiling temperatures, high pore pressure, high stress, gas transport, etc.)?
- What are the mineralogical alterations of bentonite in the short-term and long-term (e.g., illitization and loss of swelling capacity)?
- Are the models (including processes, constitutive relationships and parameters) developed for 100 ^{o}C suitable for high temperature conditions?

Using explorative generic models, multi-scale experiments and field tests to address these questions

Exploration with Generic Models (100 ºC vs 200 ºC)

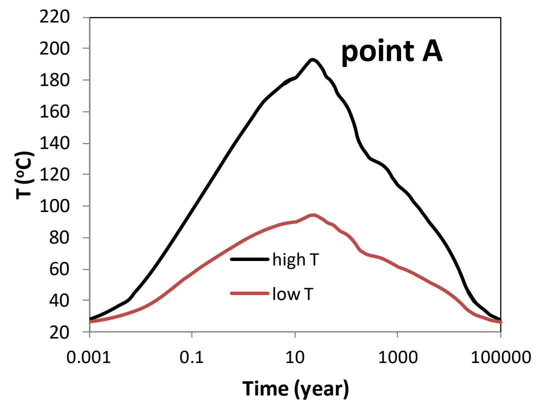
Model development



EBS Bentonite: Kunigel-VI or FEBEX bentonite

Clay formation : Opalinus Clay

Two cases for comparison: a “high T” case and a ”low T” case



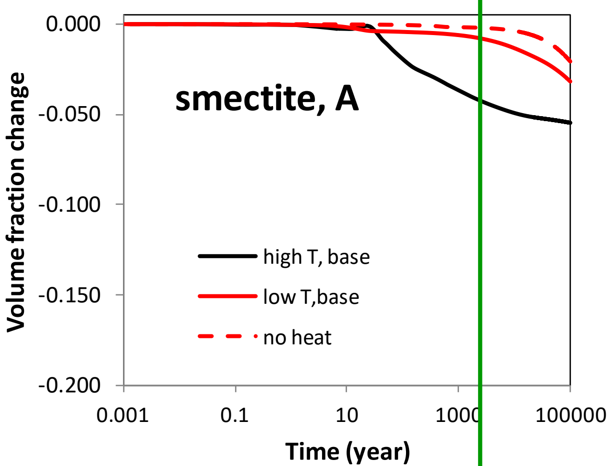
- Chemical model: aqueous complexation, minerals dissolution/precipitation and cation exchangeable
- Illitization was modeled as smectite dissolution and precipitation of illite:

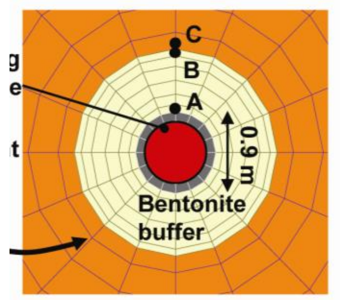
  Smectite + 0.52H^+ + 0.63AlO_2^- + 0.6K = illite + 0.26H_2O + 0.08Mg^{+2} + 0.33Na^+ + 0.5SiO_2(aq)
- The reaction rate was calibrated against data from Kinnekulle bentonite, Sweden *(Push and Madsen, 1995)*
- Mechanical-chemical coupling was formulated via an extended linear swelling model or Dual structural Expansive Clay Model (BExM)

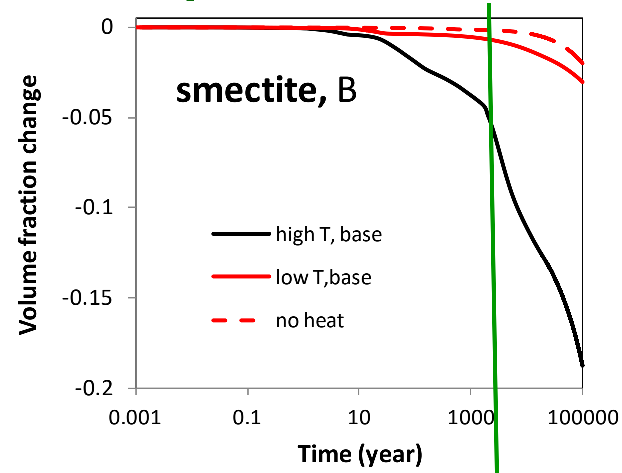
(Zheng et al., 2015; 2017)

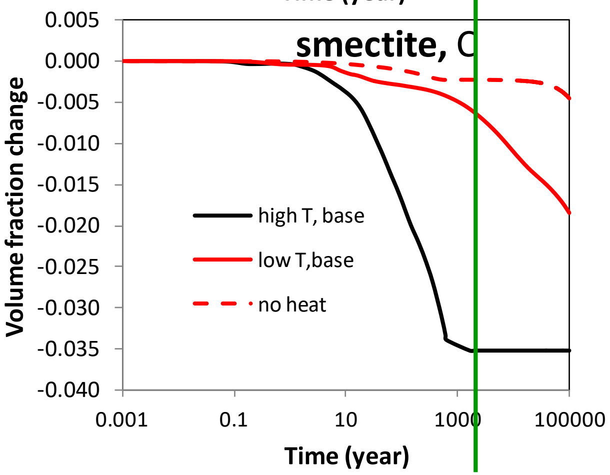
Exploration with Generic Models (100 ºC vs 200 ºC)

Key finding (1): illitization occurs, temperature plays a key role and bentonite-host rock interaction is important

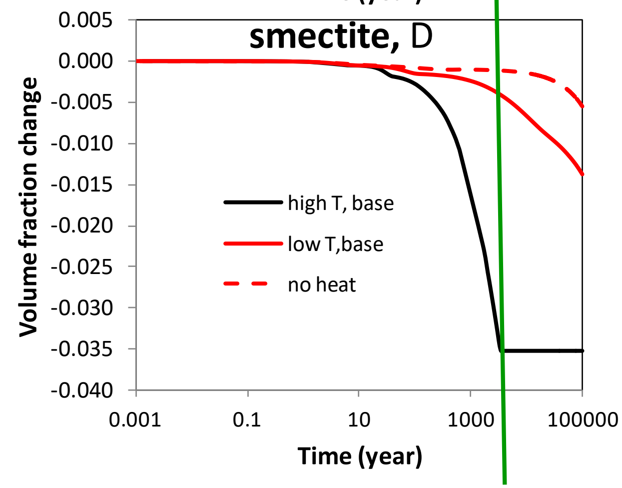








The temporal evolution of smectite volume fraction at points A, B, C, and D for Kunigel-VI bentonite.

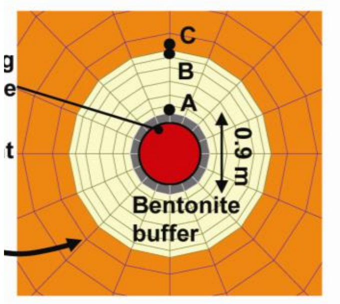


At early times, dissolution of k-feldspar supplies K for illitization; after about 3000 years, illitization in host rock stops and K is transported into bentonite which leads to very different illitization at points A and B

Exploration with Generic Models (100 ºC vs 200 ºC)

Key finding (2): Swelling stress decreases as a result of chemical changes and such decrease varies case by case

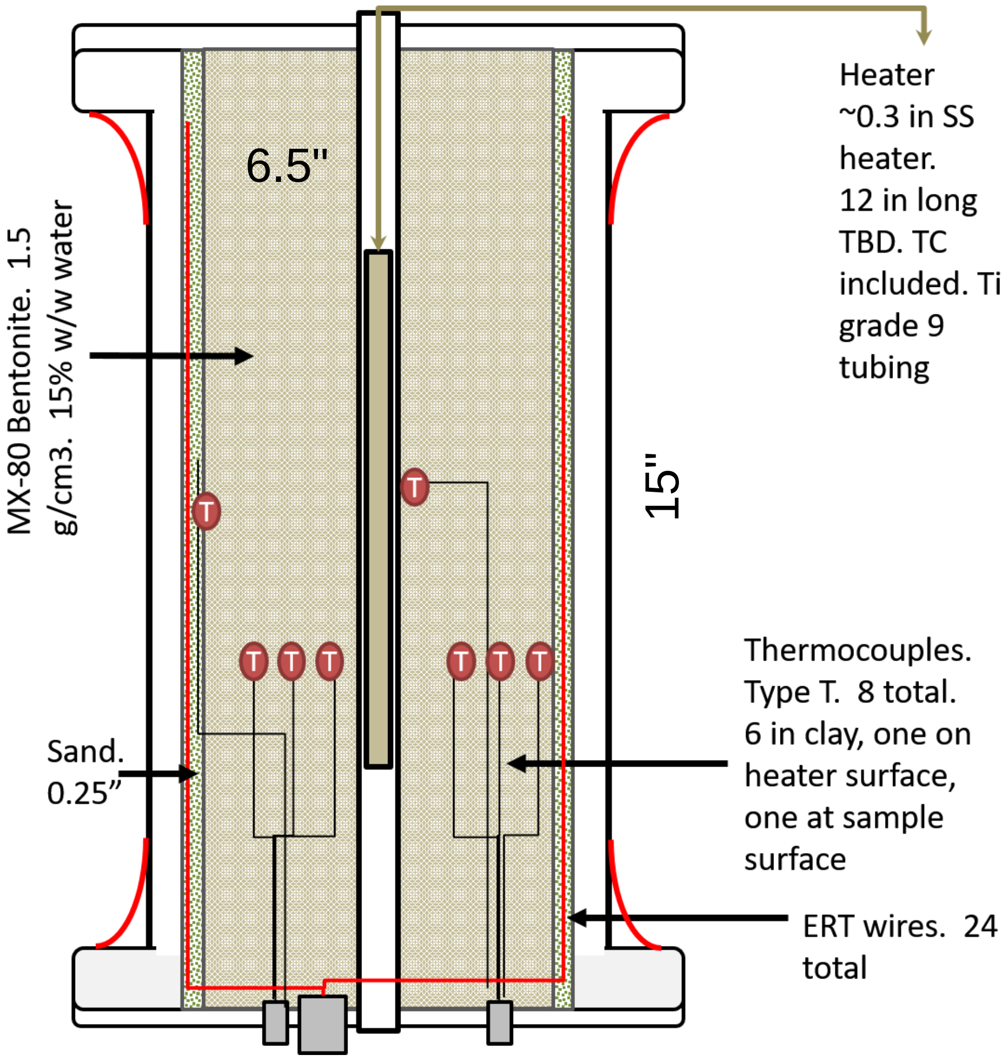
The geochemically induced swelling stress for Kunigel and FEBEX bentonite at points A and B for a “high T” scenario



	Kunigel-VI bentonite				FEBEX bentonite			
	Stress reduction by ion concentration		Stress reduction by smectite dissolution		Stress reduction by ion concentration		Stress reduction by smectite dissolution	
	MPa	%	MPa	%	MPa	%	MPa	%
Point A	0.07	7%	0.09	9%	0.006	0.1%	0.17	3.4%
Point B	0.08	8%	0.45	45%	0.06	1.1%	0.6	12%

Multi-scale Experiments and Modeling for Better Understanding (1)

A cylindrical bentonite column with a 200 °C heater in the middle



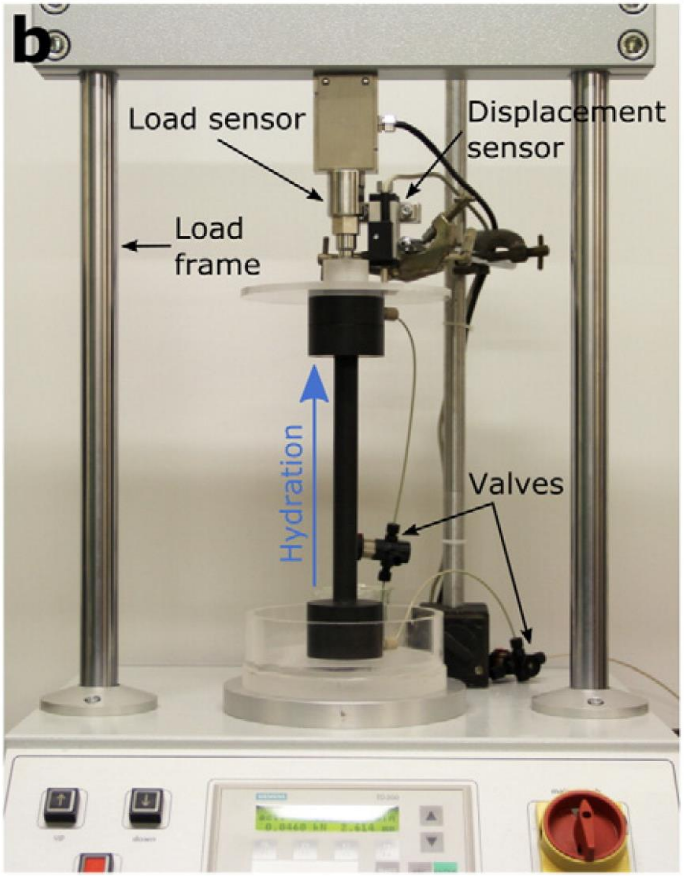
Experimental Summary:

- Laboratory analog of HotBENT experiment
- 2 parallel columns (heated and control)
- Pre-molded clay column with embedded sensors
- Circular hydrological boundary condition
- Temperature and ERT sensors embedded
- Time lapse CT scans

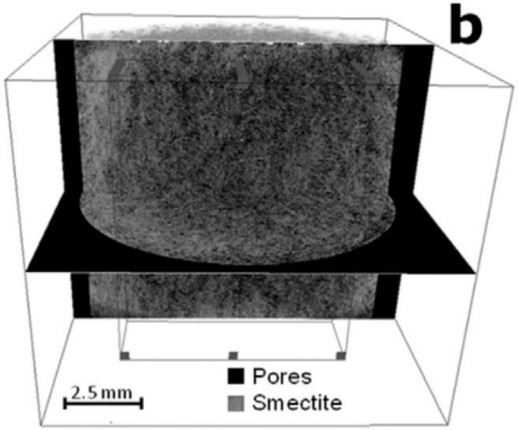
Multi-scale Experiments and Modeling for Better Understanding (2)

Study chemical controls on smectite structure and swelling

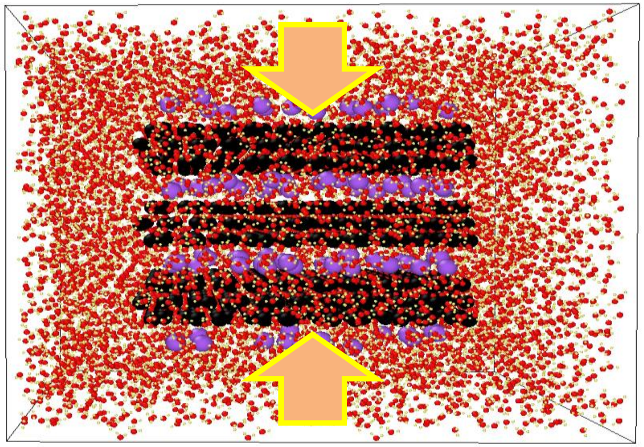
X-ray compatible oedometer



µXCT of pore development during hydration

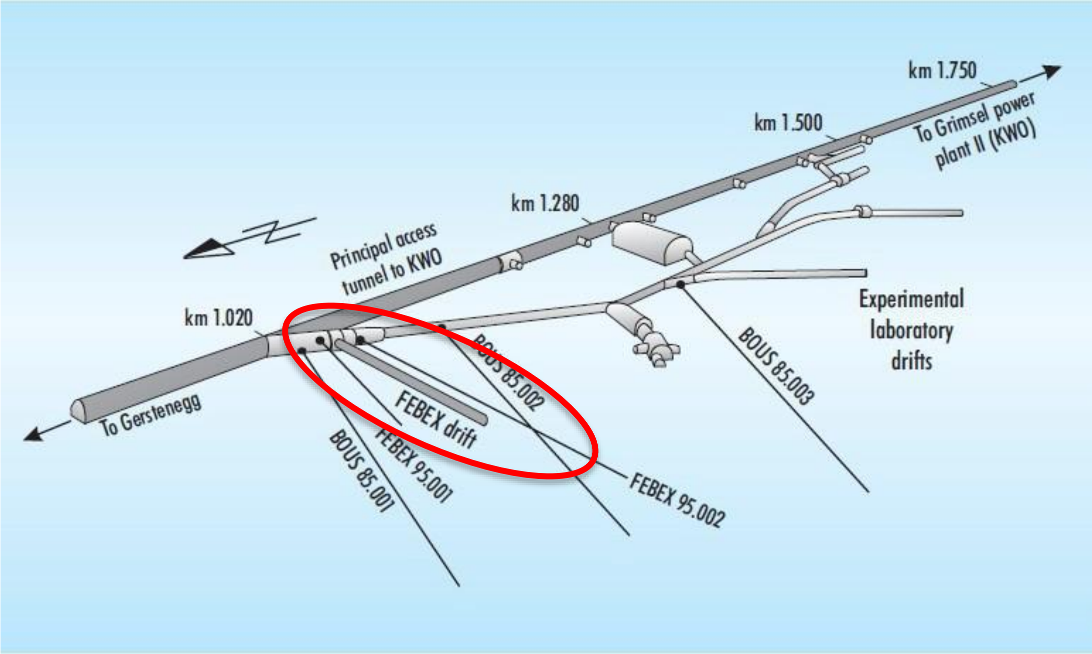


Molecular predictions of swelling pressure



A Planned Field Test: HotBENT (1)

A planned collaboration project, HotBENT, led by NAGRA (Switzerland), will conduct a joint experiment integrated with lab and modeling studies to evaluate buffer behavior at 150 °C to 200 °C.

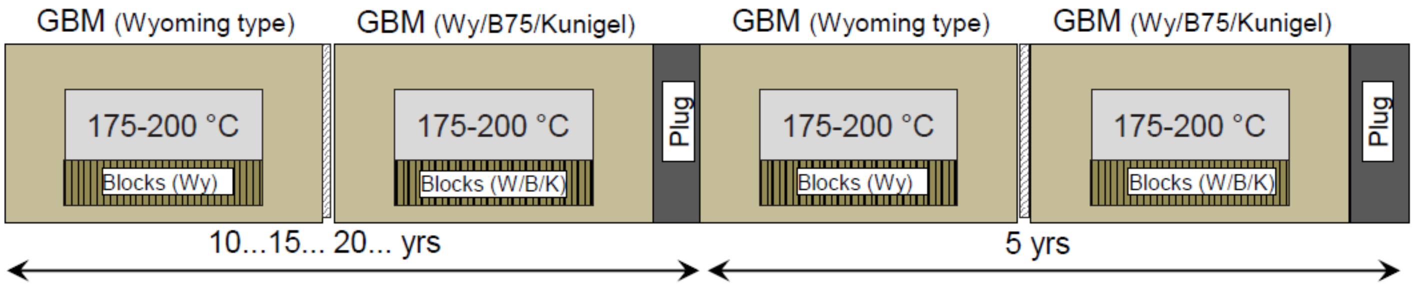


Participating organizations:

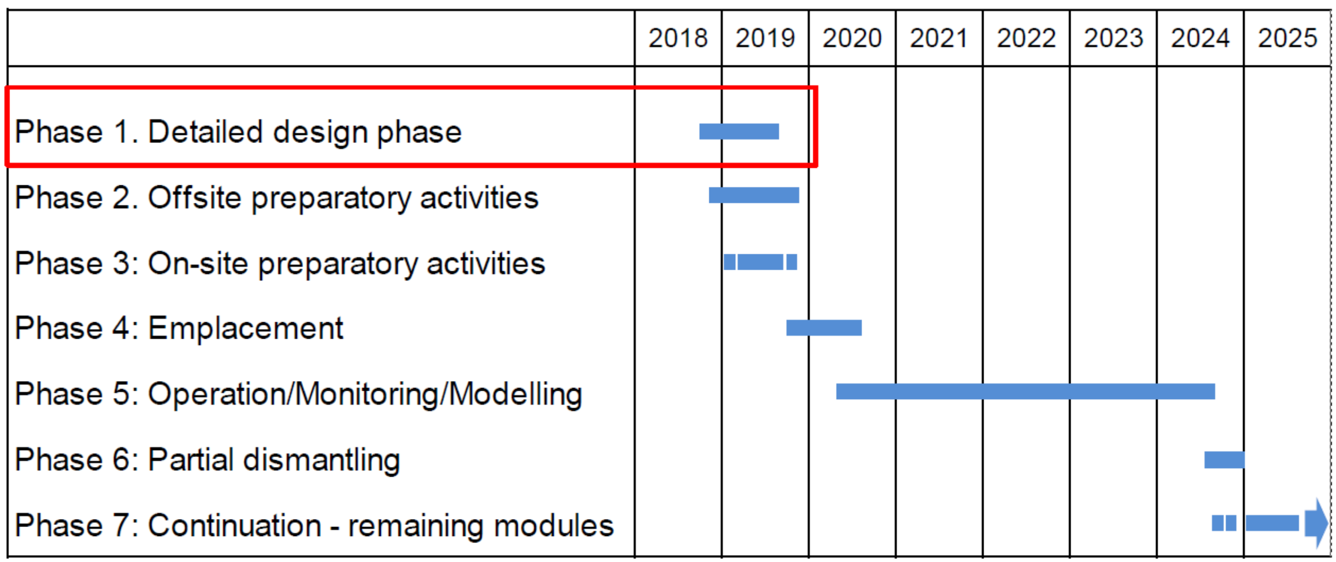
NAGRA (Switzerland), **DOE(**USA**), NWMO (**Canada**), NUMO (**Japan**), RWM (**UK**), SÚRAO (**Czech Republic)

A Planned Field Test: HotBENT (2)

HotBENT modular design - example



Timeline for the HotBENT experiment



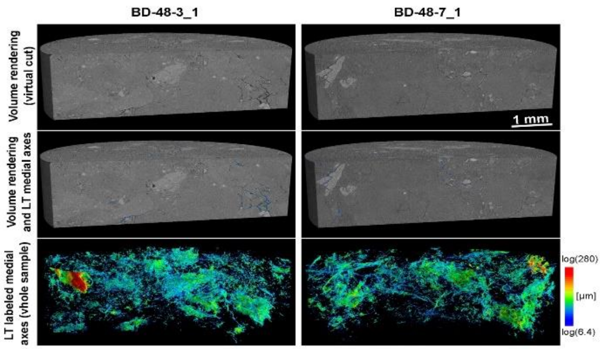
Integration with Generic Disposal R&D

- Fundamental understanding of coupled processes at multiple scales
- Building robust constitutive relationships for coupled processe**s**

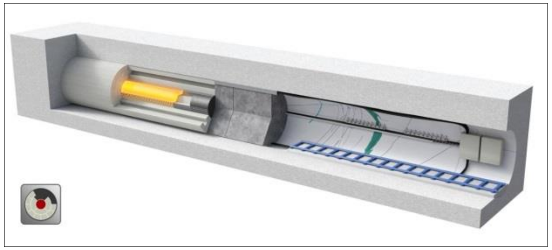
- Developing advanced modeling tools
- Constructing multi-physics coupled process models
- Testing models with large scale experiments

- Supplying generic Performance Assessment (PA) models with reliable conceptual model and parameters
- Providing generic PA models with well-tested constitutive relationships
- Integrating process models into PA

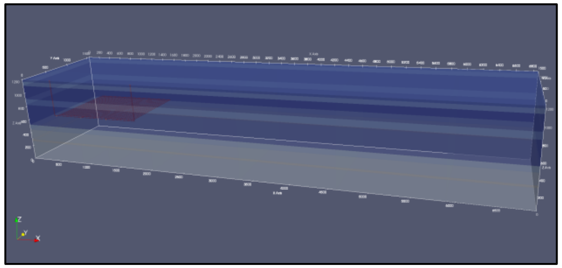
Micro-structural analysis



Field Experiments



Generic PA Modeling



Summary

- Participating large scale *in situ* tests (e.g. FEBEX *in situ* test) conducted by international collaborators significantly enhanced the understanding of the alteration of EBS and improved modeling capability
- Knowledge gaps narrowed, but more work is needed:
  - Models and experiments at higher temperature condition (e.g., HotBENT)
  - Multi-scale experiments and models
  - Integration with PA models

Questions?

Clean. Reliable. Nuclear.

References

ERESA, 2000. Full-scale engineered barriers experiment for a deep geological repository in crystalline host rock FEBEX Project. EUR 19147 EN, European Commission.

NAGRA, 2002. Project Opalinus Clay Safety Report: Demonstration of disposal feasibility for spent fuel, vitrified high-level waste and long-lived intermediate-level waste (Entsorgungsnachweis) NAGRA Tech. Rep. 02-05, NAGRA, Wettingen, Switzerland.

Kober F., Villar M.V., Turrero M., Wersin P., Gaus I & FEBEX-DP Consortia, 2017. FEBEX-DP: Dismantling FEBEX after 18 years – activities, results, conclusions. Clay Conference 2017, Davos, 25.09.2017.

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

Wersin P. and Kober F., 2017. FEBEX-DP Metal Corrosion and Iron-Bentonite Interaction Studies, NAB 16-16

Zheng L., Rutqvist, J. Birkholzer J. T. and Liu, H.H., 2015. On the impact of temperatures up to 200 °C in clay repositories with bentonite engineer barrier systems: A study with coupled thermal, hydrological, chemical, and mechanical modeling. Engineering Geology 197: 278-295.

Zheng L., Rutqvist J., Xu H. and Birkholzer J. T., 2017. Coupled THMC models for bentonite in an argillite repository for nuclear waste: Illitization and its effect on swelling stress under high temperature. Engineering Geology 230: 118-129.