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

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

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It Is a Collaborative Effort !

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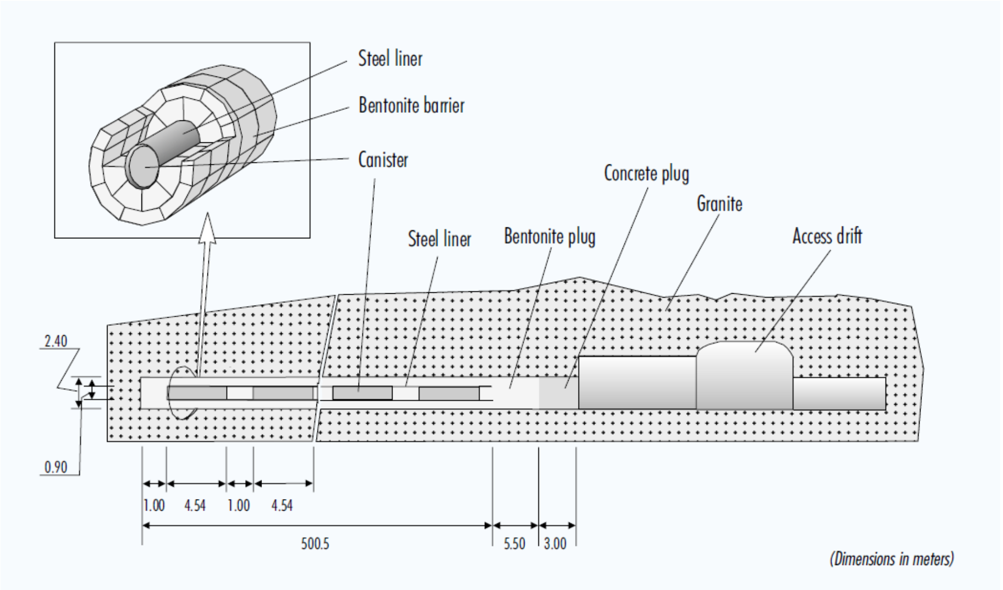
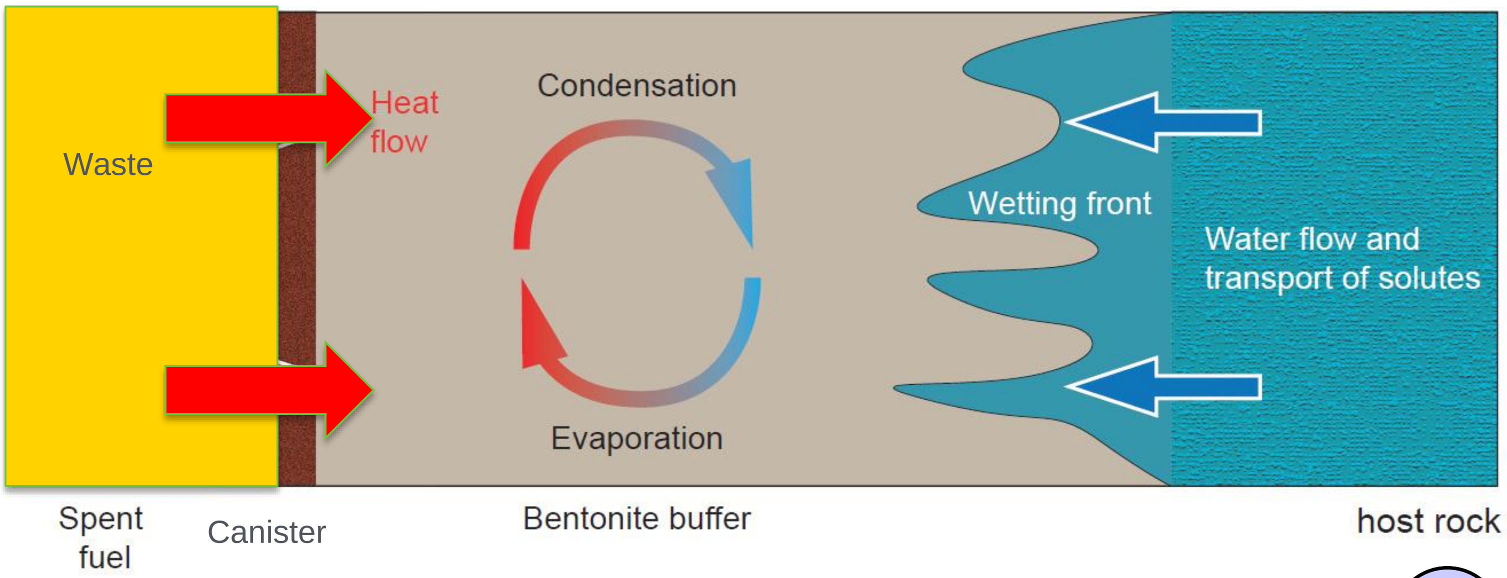
Engineered Barrier System (EBS) in a Geological Repository

- Material:
  - Bentonite granular/blocks
- Featured properties:
  - Swelling
  - Low permeability
  - High retardation capability
- 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



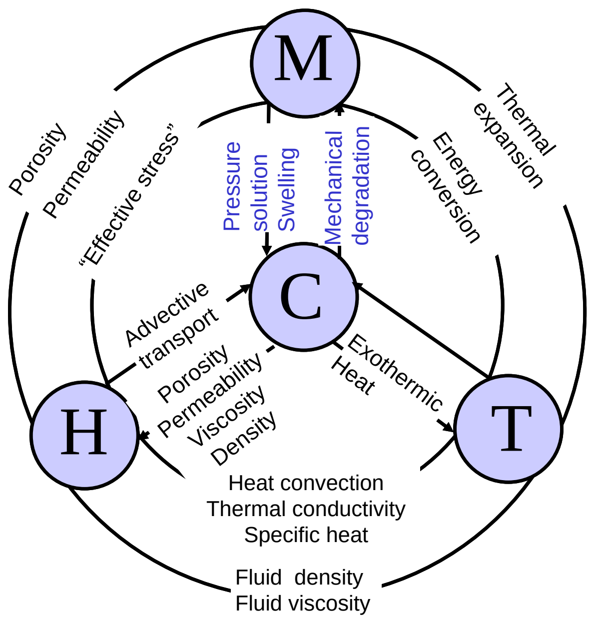
(ENRESA, 2000)

Processes Involved in EBS



(Modified from Leupin et al, 2014)

To ensure the favorable features of EBS in the long term, understanding and modeling of coupled thermal, hydrological, mechanical and chemical (THMC) processes are critical



Data Needed to Understand/Model EBS

Modeling THMC coupled processes in an EBS is very data-demanding:

- Detailed characterization of thermal (e.g. thermal conductivity), hydrological (e.g. permeability), mechanical (e.g. bulk modulus) and chemical (e.g. ion concentration in pore water and mineralogical composition) properties
- THMC properties evolve as EBS undergoes heating and hydration and usually are described by constitutive relationships in a model—experiments are needed to verify these relationships
- A coupled THMC model, which is composed of the basic THMC equations and constitutive relationships, has to be verified by laboratory experiments and ultimately large scale *in situ* tests
- Micro-scale experiments are needed to understand fundamental interactions of THMC processes

Roadmap to Study Long-term Stability of EBS

Developing simulators for coupled THMC processes

Evaluating key THMC processes with a large scale *in situ* experiment at 100 ^{o}C

Studying long-term alteration of bentonite at high temperature (200 ^{o}C) with exploratory generic models

Further testing THMC models with experiments at 200 ^{o}C, and improving understanding and reducing uncertainties by multi-scale experiments and modeling

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Developing THMC Simulators

T**OUGHREACT-FLAC3D** which combines **TOUGHREACT** and **TOUGH2-FLAC3D** has been developed and it is capable of conducting coupled THMC simulations.

TOUGHREACT — coupled THC code

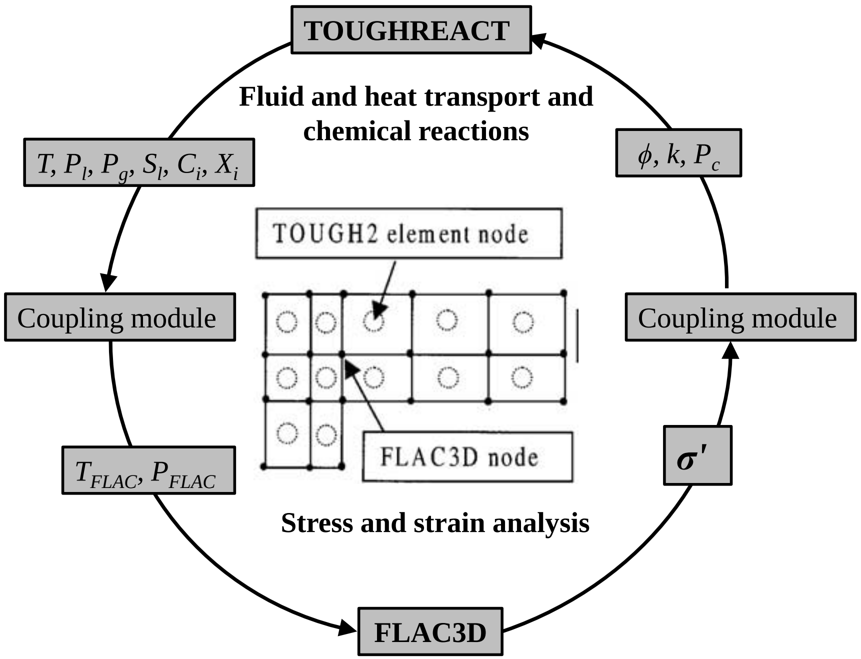
TOUGHREACT is a numerical simulator for chemically reactive nonisothermal flows of multiphase fluids in porous and fractured media *(Xu et al., 2014).*

TOUGH2-FLAC3D — coupled THM

TOUGH2-FLAC3D *(Rutqvist, et al., 2011)* sequentially couples the finite-difference geomechanical code FLAC3D with the finite-volume, multiphase fluid flow code, TOUGH2 *(Pruess et al., 2012)*.

Great effort has been invested to develop and verify mechanical-chemical coupling schemes and constitutive relationships.

THC Processes



Mechanical Process

TOUGHREACT-FLAC3D is used for all the simulations in this presentation

TReactMech, a parallel coupled THMC simulator, has been developed and used for geothermal applications. It will be upgraded and used for modeling EBS.

Roadmap to Study Long-term Stability of EBS

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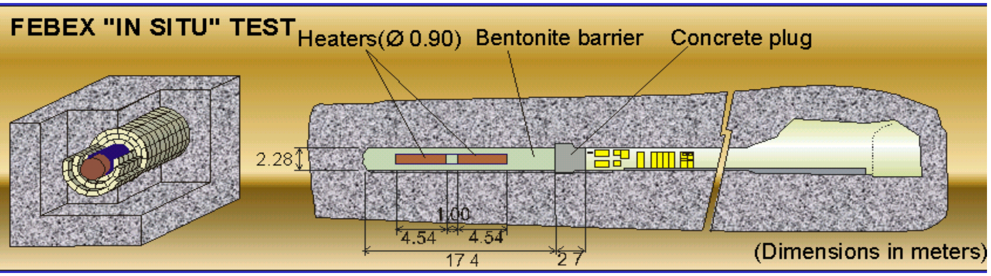
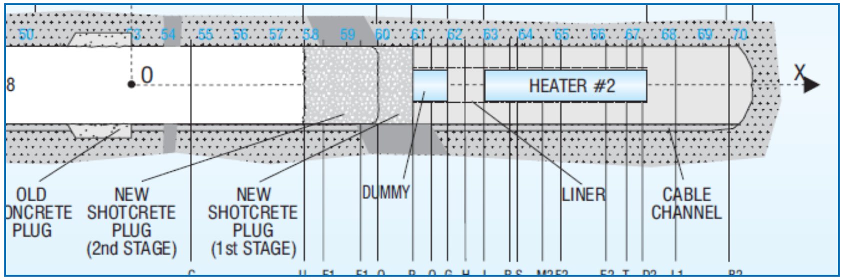
Evaluating Key THMC Processes with an *in situ* Test

Overview of the FEBEX *in situ* test

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

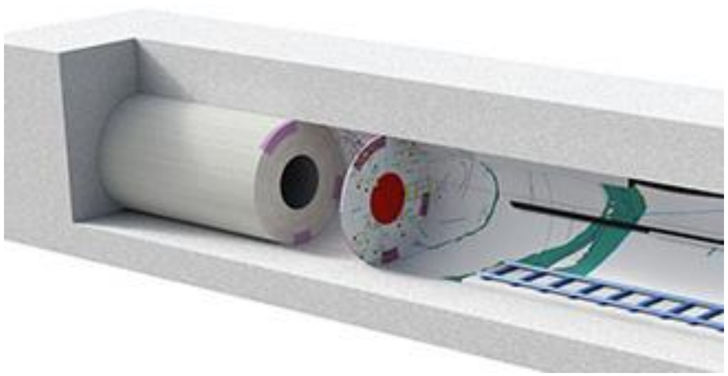


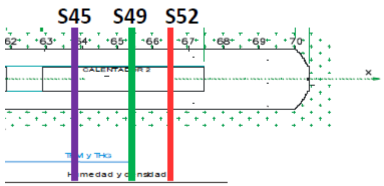
In 2002, Dismantling of Heater #1



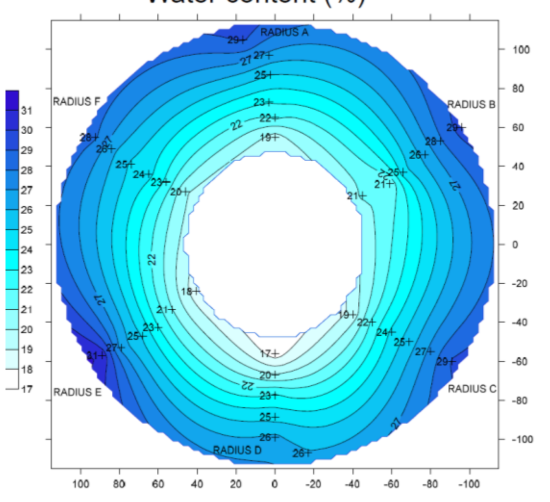
Extensive laboratory tests were carried out to characterize THMC properties of bentonite, concrete, steel liner and granite.

In 2015, Dismantling of Heater #2





Personal communication with M.V. Villar

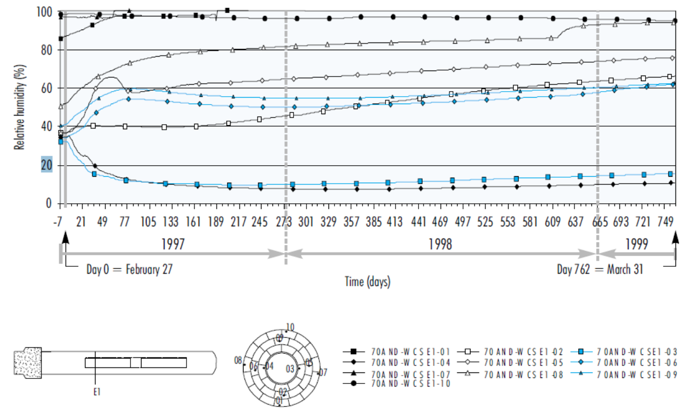


Evaluating Key THMC Processes with an *in situ* Test

THMC processes are implemented in the model and their relevance are evaluated by the goodness-of-fit between model and data

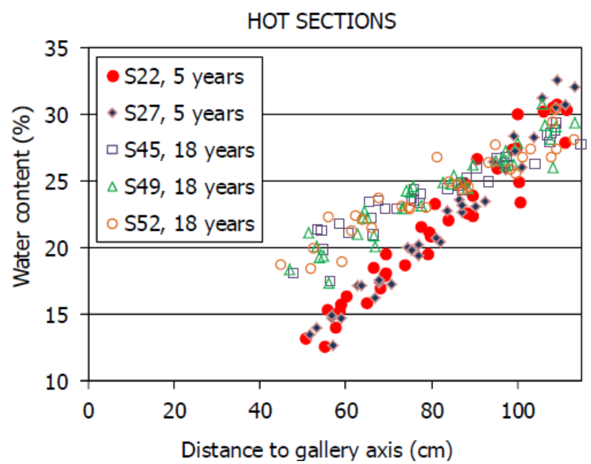
Available data:

- Temporal evolution of temperature, relative humidity, pressure and stress
- Spatial distribution of water content, dry density, ion concentration in pore water and mineralogical composition at two snapshots: dismantling of heater #1 and #2



(ENRESA, 2000)

Event	Date	Time (year)
Commencement of heating	2/27/1997	0.0
Shutdown of Heat #1	2/2/2002	5.0
Sampling after heat #1 was dismantled	5/2/2002	5.3
Shutdown of heat #2	4/24/2015	18.1
Sampling after heat #2 was dismantled	7/3/2015	18.3

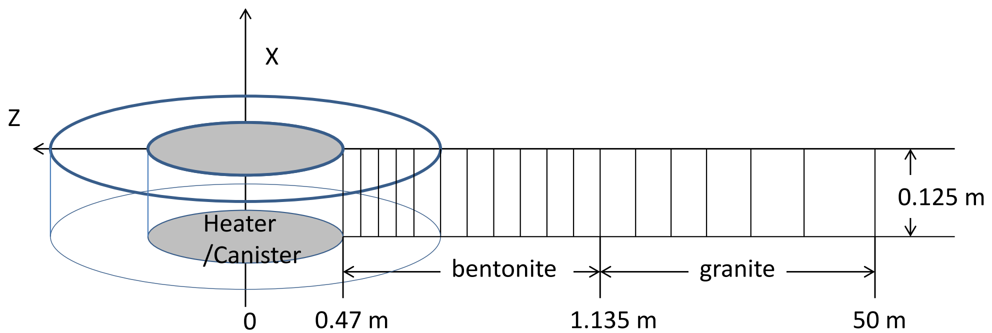


(Villar et al, 2018)

Evaluating Key THMC Processes with an *in situ* Test

Developing THMC model

1D axi-symmetrical model for “hot” sections



Thermal model: Heat convection and conduction
Flow model: Two-phase (gas and water) flow
Mechanical model:

State surface approach: $e = A + B\ln(-\sigma_m'') + C\ln(s+p_a) + D\ln(-\sigma_m'')\ln(s+p_a)$

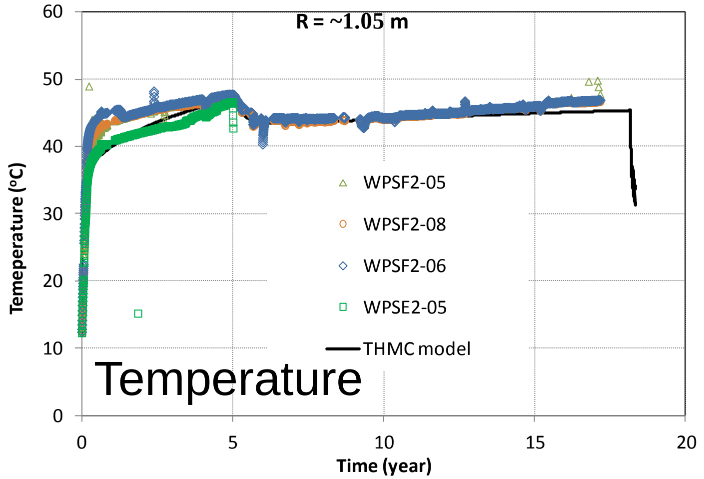
Permeability change : $\log k = (-2.96\rho_d - 8.57)/\alpha$

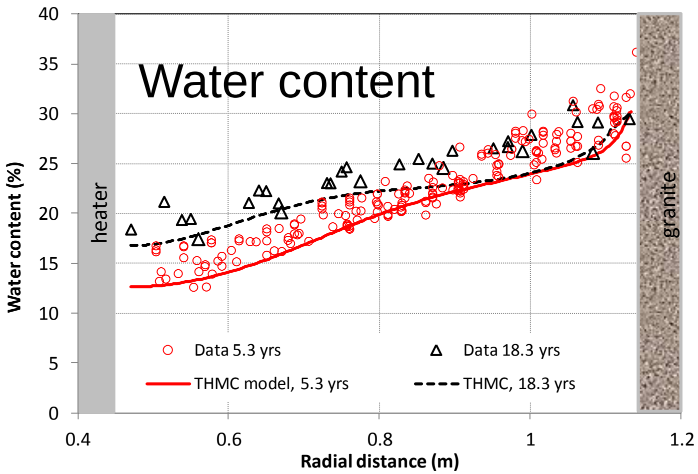
Thermal osmosis : $v_{to} = -k_T \nabla T$

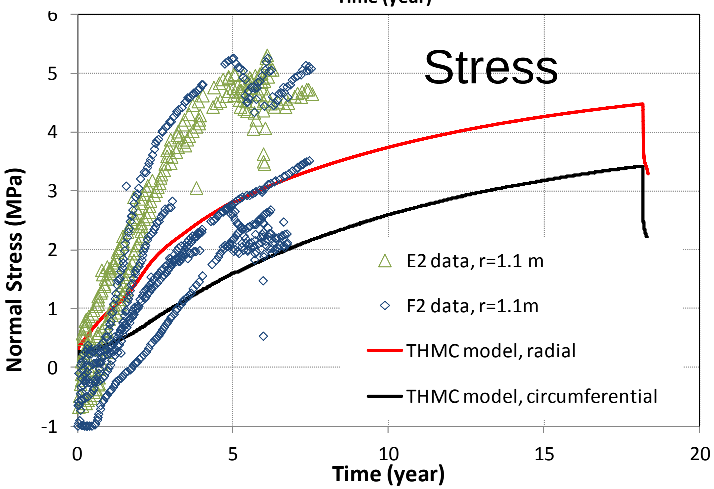
Chemical model: aqueous complexation, surface complexation, cation exchange and minerals dissolution/precipitation

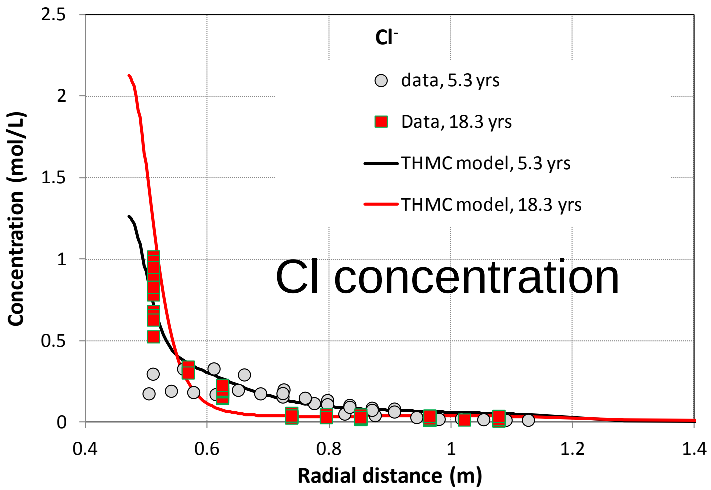
Evaluating Key THMC Processes with an *in situ* Test

Models evolved from TH model to THMC model; Matching between data and model were eventually achieved







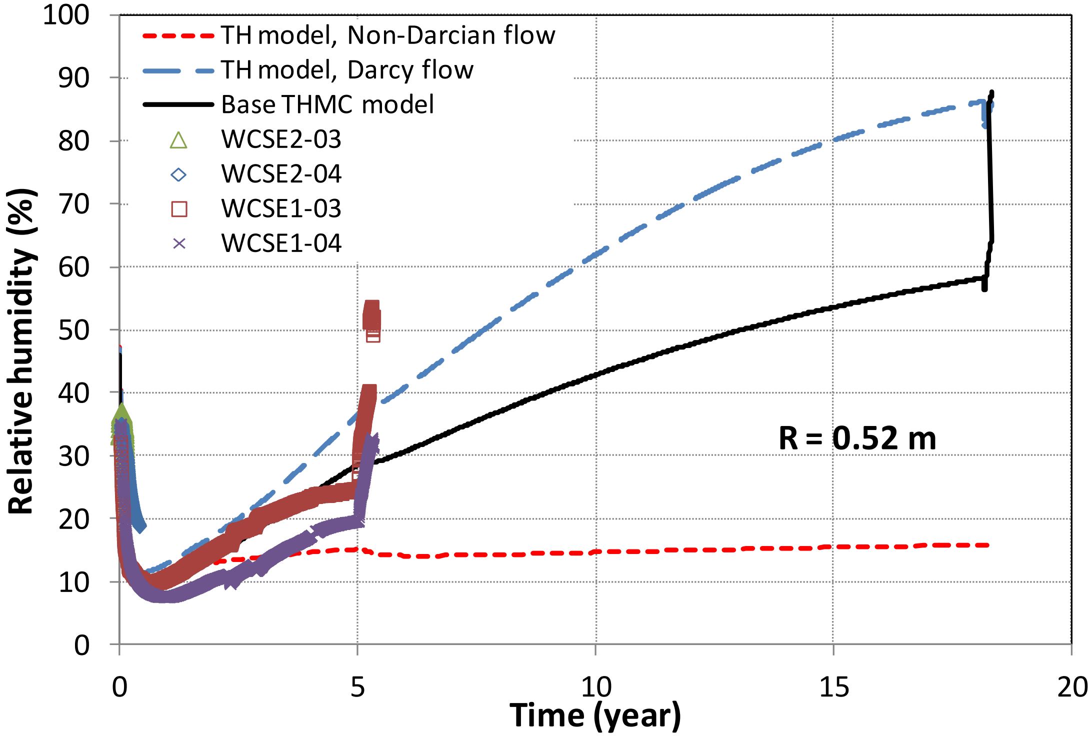


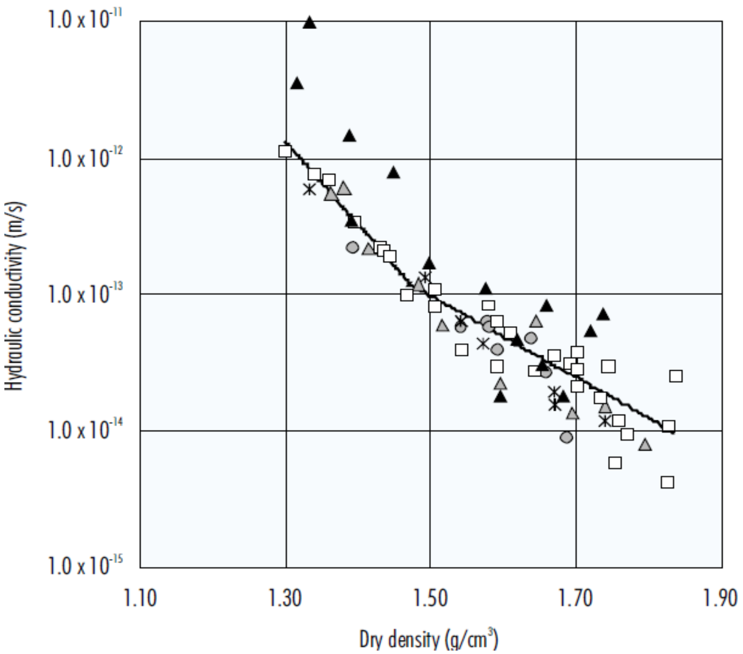
TH coupling has a moderate effect on thermal evolution in bentonite

Evaluating Key THMC Processes with an *in situ* Test

Key processes that affect hydrological behavior

- Two-phase (gas and liquid) Darcy type multiphase flow
- HM coupling (permeability and porosity changes as a result of swelling) has to be considered in the model
- The relevance of Non-Darcian flow is questionable



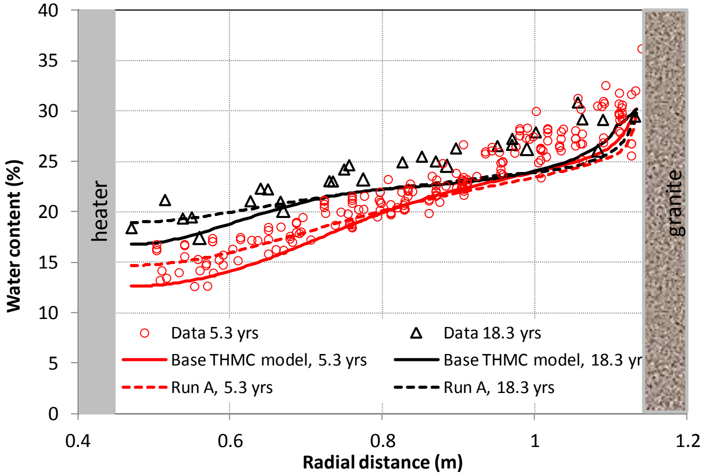


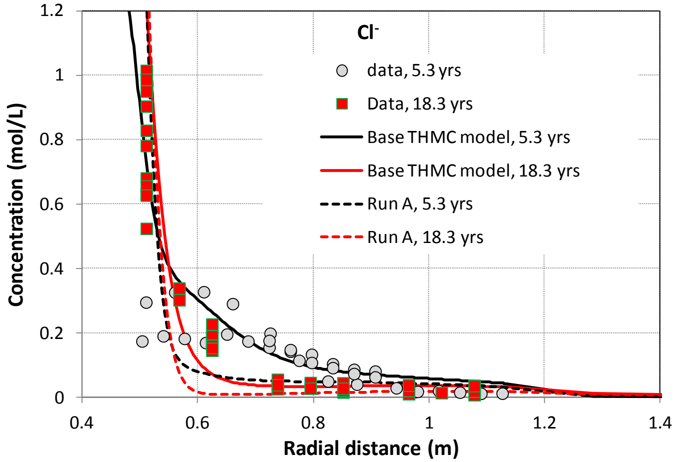
(ENRESA, 2000)

Evaluating Key THMC Processes with an *in situ* Test

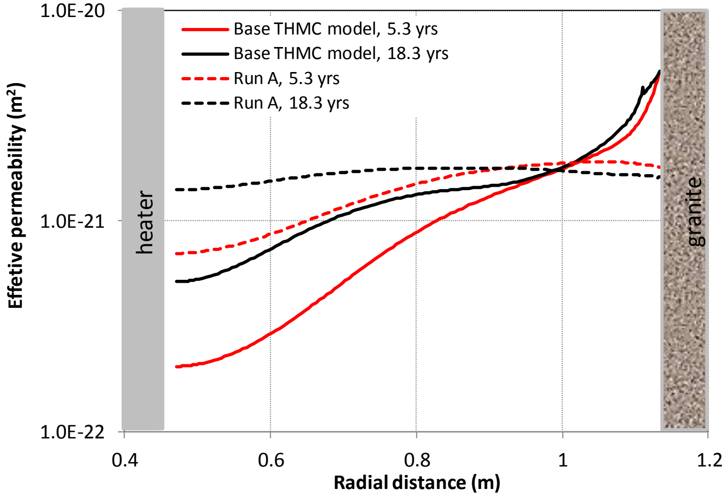
Key processes that affect hydrological behavior

The effect of different constitutive relationships for saturated permeability : k=f(dry density) in base model vs k=f(effective stress) in Run A





- The spatial distribution of permeability causes the differences between base model and Run A in Cl concentration profiles
- Chemical data provide relevant piece of information for calibrating THM model

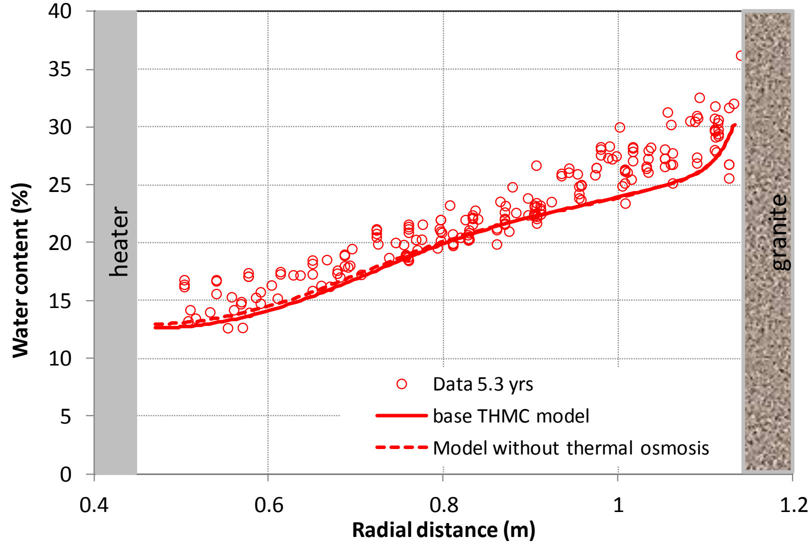


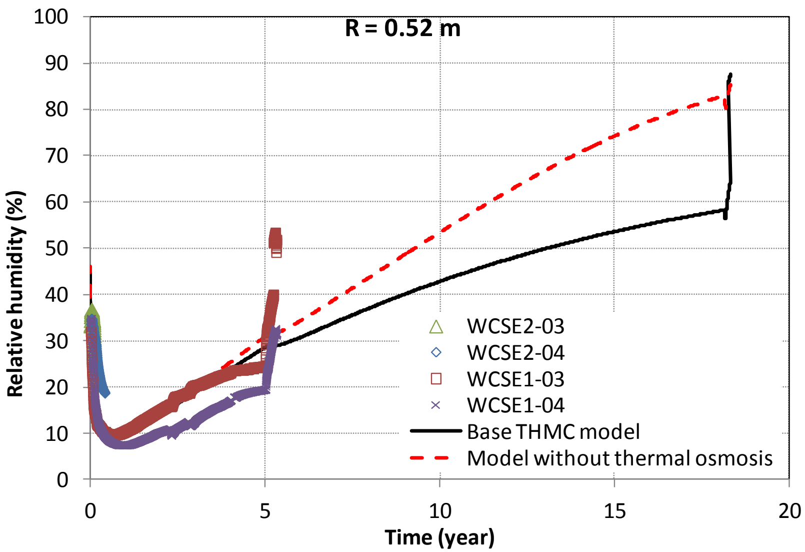
Evaluating Key THMC Processes with an *in situ* Test

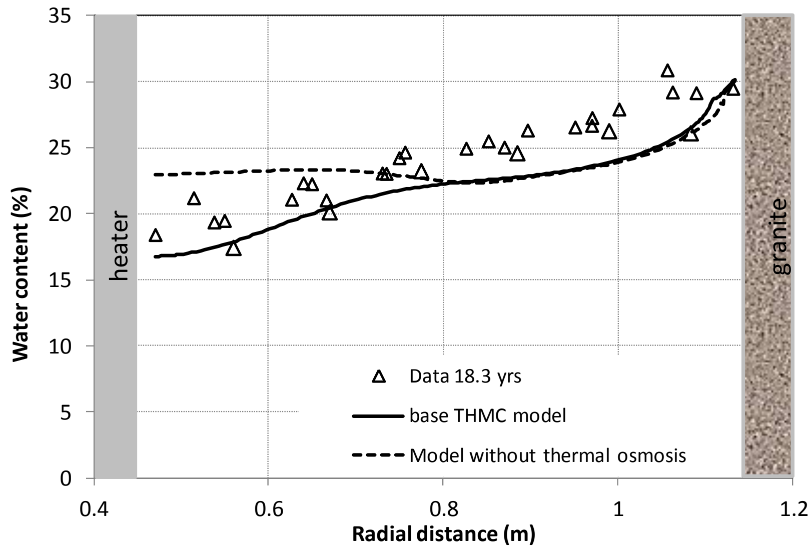
Key processes that affect hydrological behavior

Thermal osmosis might be relevant

Flux	Gradient		
	Hydraulic	Temperature	Chemical
liquid	Hydraulic flow Darcy's law	Thermo-osmosis	Chemical osmosis
Heat	Convective heat flow	Thermal conduction Fourier's law	Dufour effect
Solute	Hyperfiltration	Thermal diffusion or Soret effect	Diffusion Fick's law







Evaluating Key THMC Processes with an *in situ* Test

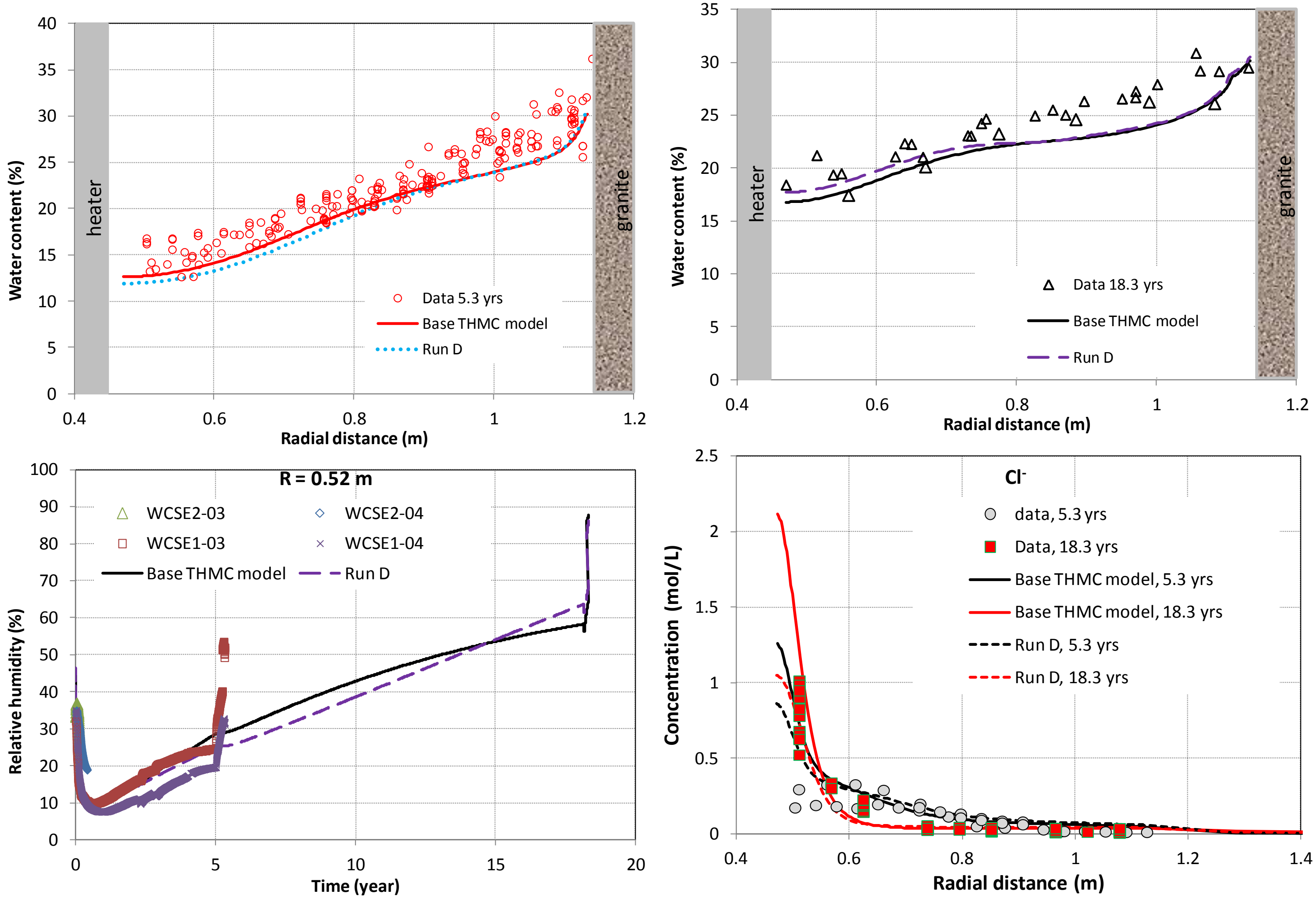
Key processes that affect hydrological behavior

An old but challenging issue for numerical modeling: Non-uniqueness of solution

Run D:

No thermal osmosis,

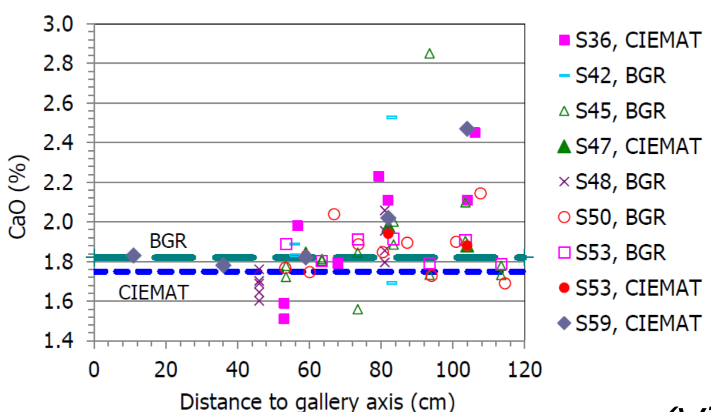
but permeability 30% lower than the base model



Evaluating Key THMC Processes with an *in situ* Test

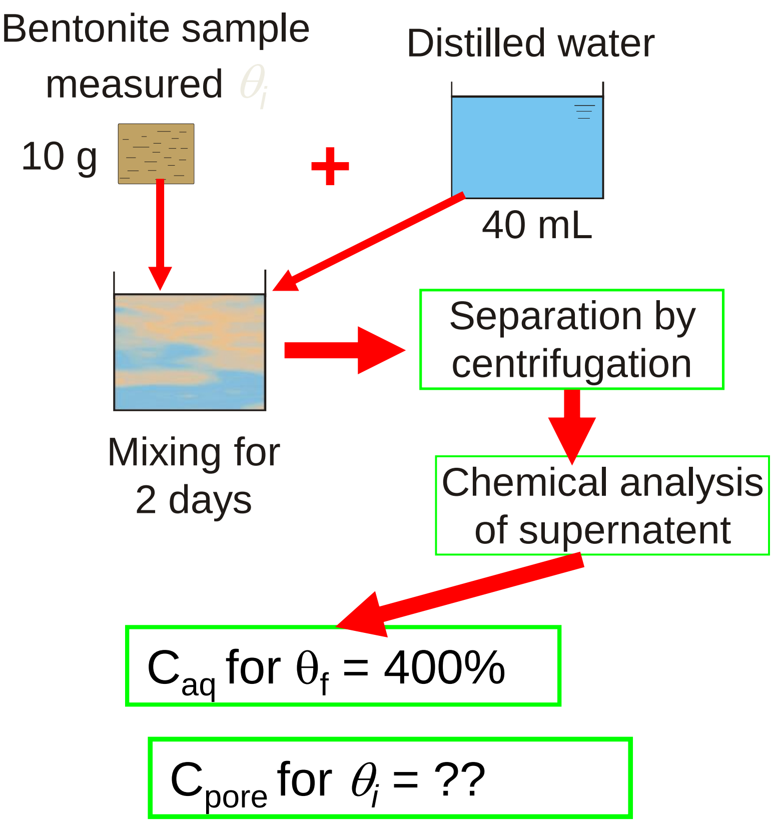
Understanding key chemical process: finding patterns while adequately accounting for uncertainties

- Data for solid phase are too scattered to show a clear spatial trend
- Data for exchangeable cations are problematic, but could be used with caution
- Ion concentrations in pore water were measured indirectly by aqueous extract method and need to be calibrated using models



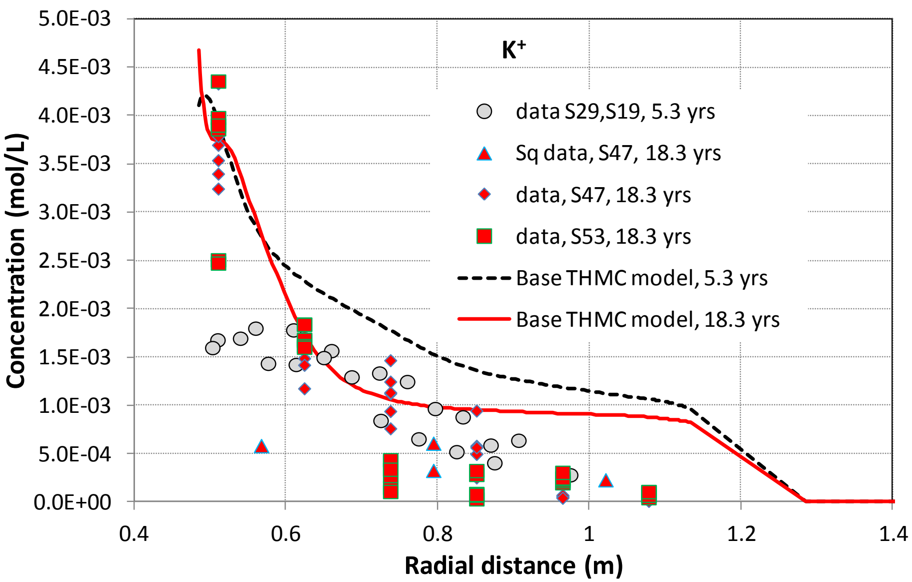
(Villar et al., 2017)

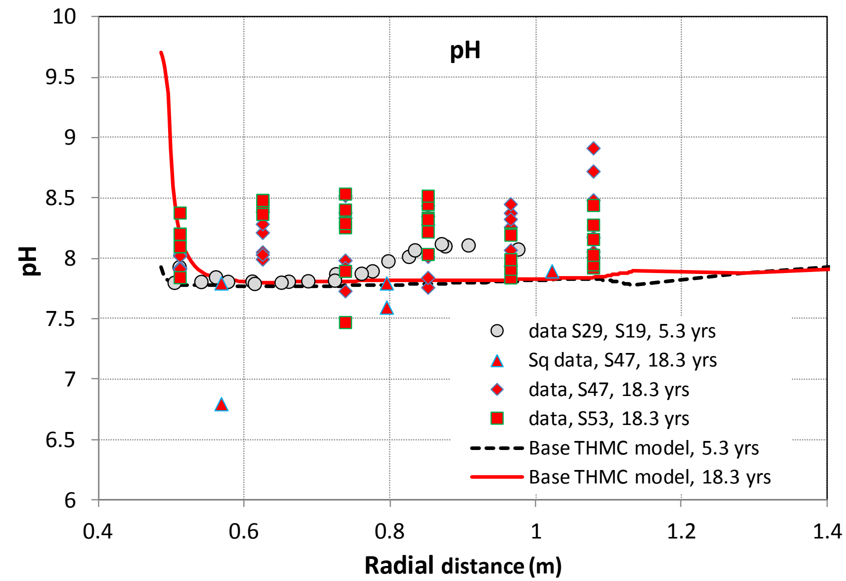
Measuring pore water chemistry using aqueous extract method



Evaluating Key THMC Processes with an *in situ* Test

Key chemical processes: chemical reactions are induced by the EBS-host rock interaction and thermal perturbation



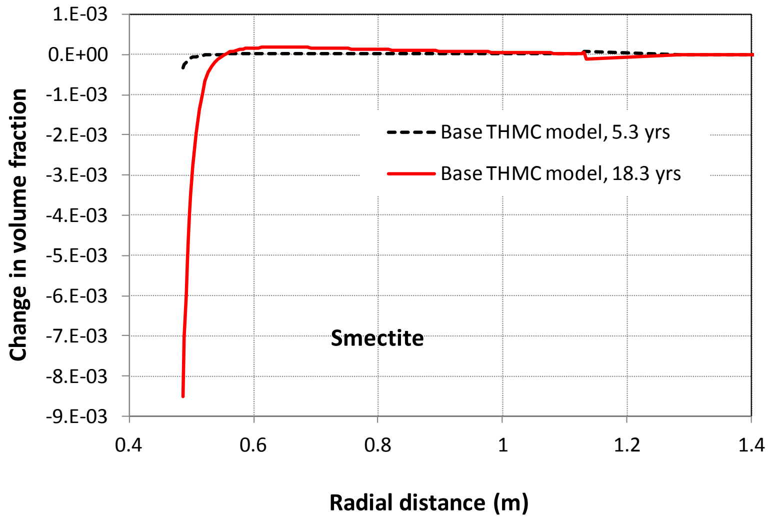


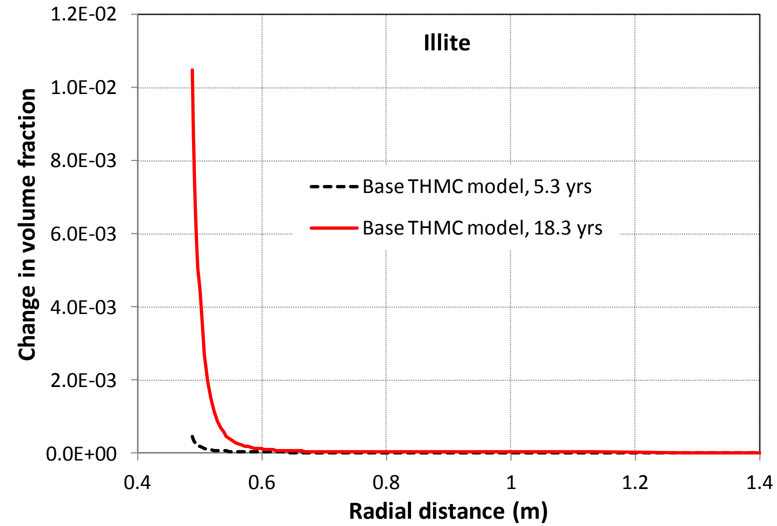
K concentration is affected by transport processes (advection and diffusion) and dissolution of K-feldspar and precipitation of illite

pH is buffered by surface protonation and affected by many chemical processes.

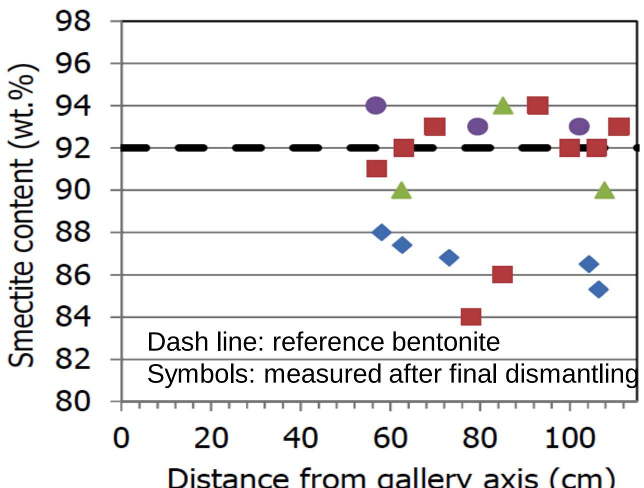
Evaluating Key THMC Processes with an *in situ* Test

Key chemical processes





Model shows dissolution of smectite and precipitation of illite at area near the heater. But the data of smectite content are too scattered to verify model results.



Evaluating Key THMC Processes with an *in situ* Test

Major concluding remarks from FEBEX project

- The project evolved from feasibility study to process understanding
- *In situ*, 1:1 experiment was proved very useful in terms of engineering aspects, monitoring, sampling and modeling *(Kober et al ., 2017)*
- Bentonite (EBS) was performed as expected *(Kober et al ., 2017)* in term of dry density, swelling pressure and geochemical properties
- Long-term, international collaboration was very beneficial *(Kober et al ., 2017)*
- Modeling capability was improved significantly, but there is room of improvement, especially in geochemical and coupled processes

Evaluating Key THMC Processes with an *in situ* Test

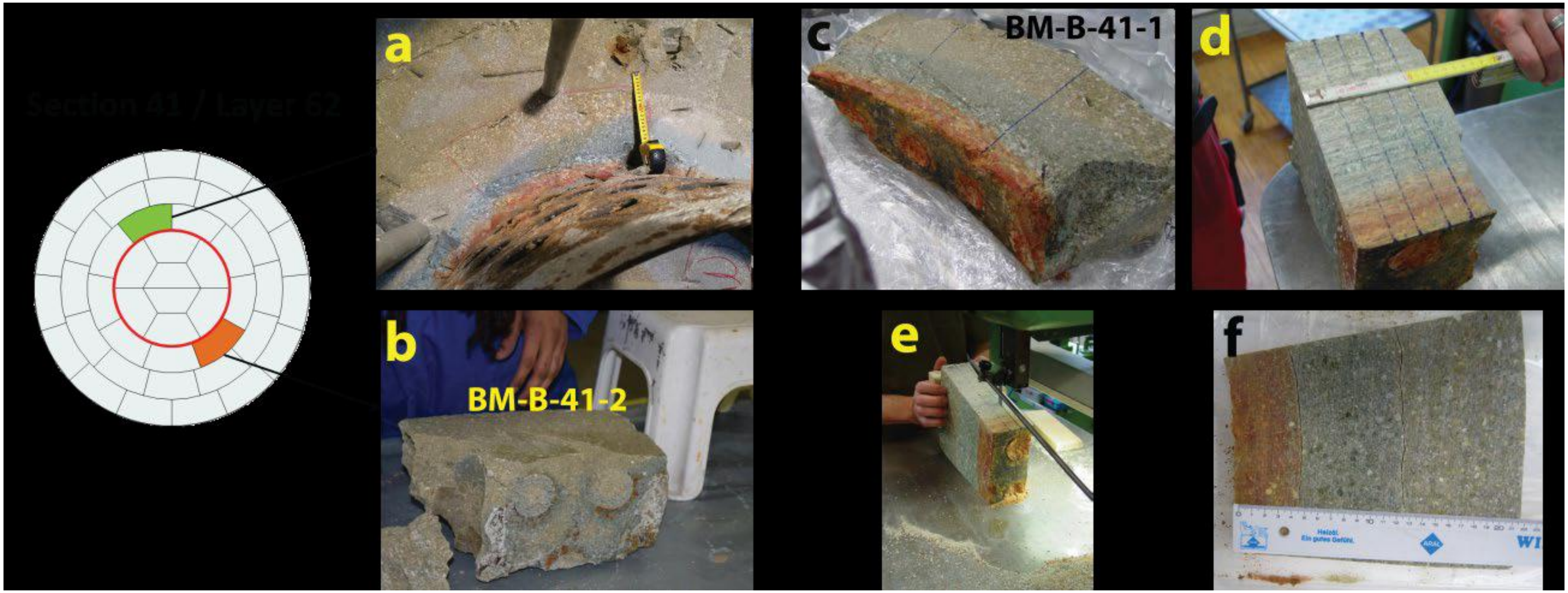
Summary of current modeling work

- The key processes needed to reproduce the THM data include thermal conduction and convection, multiphase flow, poro-elasticity, porosity and permeability changes due to swelling and thermal osmosis.
- Concentration profiles of cations were largely shaped by transport processes while their concentrations are also affected by mineral dissolution/precipitation and cation exchange; The concentration profile of pH, bicarbonate and sulfate were largely determined by chemical reactions.
- Measured mass fractions of smectite varied a great deal and were indistinguishable from the reference bentonite. The model results showed a small amount of illite precipitation and montmorillonite dissolution in the vicinity of the heater, which cannot be verified by the data.
- The robustness of model would be increased if the model is tested against long-term data and various types of data. Short-term data and single type of data may fail to reveal deficiencies of the model.
- Given the complexity of coupled THMC models, non-uniqueness is inevitable — different models can achieve similar goodness-of-fit for the same data set.

Evaluating Key THMC Processes with an *in situ* Test

Future modeling work

- Refining chemical model based on the syntheses of all geochemical data: pore water concentration, mineral phase, gas concentrations
- Understanding geochemical evolution at interfacial areas: canister/bentonite, concrete/bentonite, granite/bentonite



Wersin and Kober, 2017

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Exploratory Generic Models for EBS (100 ^oC vs 200 ^oC)

Motivation

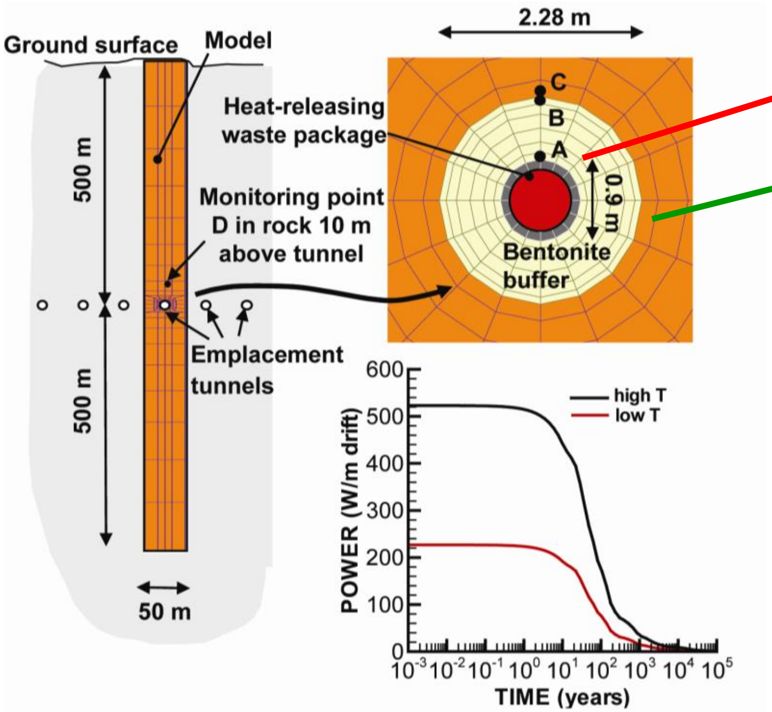
- Dual Purpose Canister disposal can lead to higher temperatures in the surrounding environment
- Thermal limit of 100 ^oC for small PWR canisters might be too limiting

Key questions to answer

- What is the long-term alteration of clay minerals, e.g. illitization? Although illitization is extensively evidenced from geological systems, laboratory experiments, field tests and modeling studies show no conclusive evidence that illitization will occur in bentonite.
- If illitization does occur, what are the key factors and how does it affect swelling capacity of bentonite?
- What is the long-term hydrological and mechanical evolution of bentonite?

Exploratory Generic Models for EBS (100 ºC vs 200 ºC)

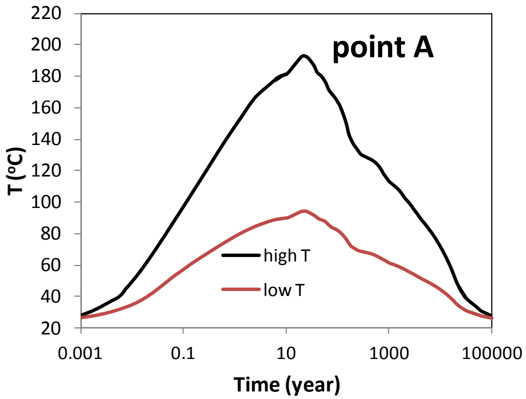
Model development



EBS Bentonite: Kunigel-VI or FEBEX bentonite

Clay formation : Opalinus Clay

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



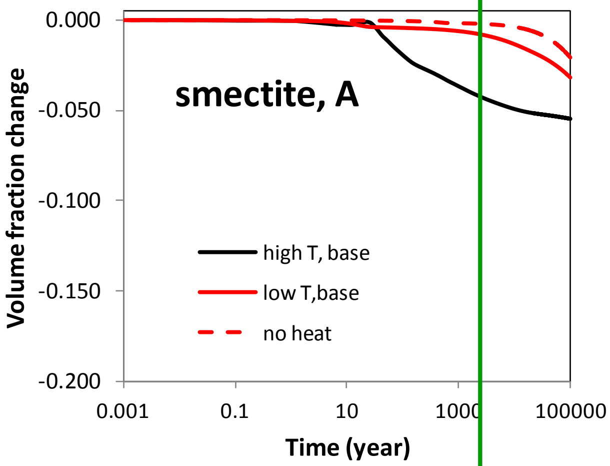
- Chemical model: 12 primary species, 97 aqueous complexes, 17 minerals and 5 exchangeable cations
- Illitization was modeled as smectite dissolution and precipitation of illite:

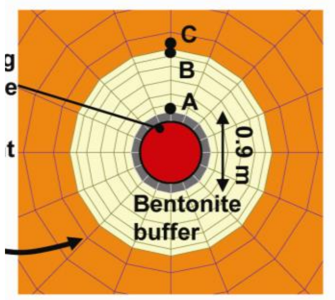
  Smectite + $0.52H^+$ + $0.63AlO_2^-$ + 0.6K = illite + $0.26H_2O$ + $0.08Mg^{+2}$ + $0.33Na^+$ + $0.5SiO_2$(aq)
- The reaction rate from 4.5e-14 to 2.4e-13 mol/g/s calibrated against data from Kinnekulle bentonite, Sweden *(Push and Madsen, 1995)*
- MC coupling was formulated via an extended linear swelling model or Dual structural Expansive clay Model (BExM)

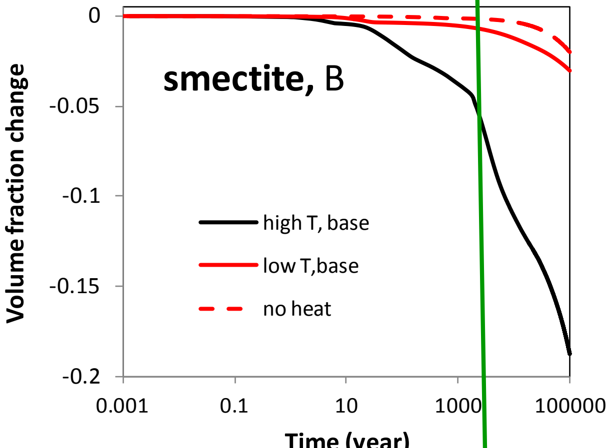
(Zheng et al., 2015; 2017)

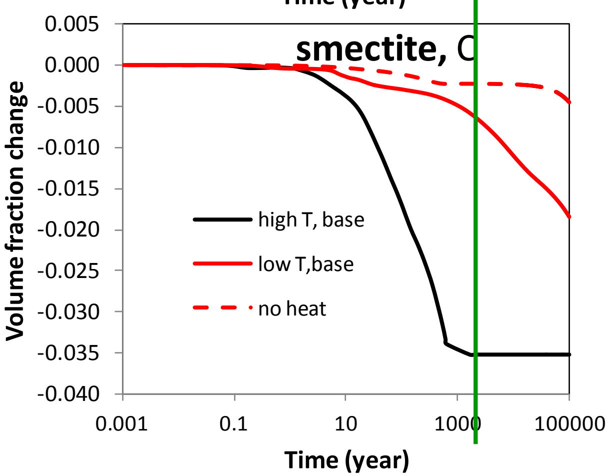
Exploratory Generic Models for EBS (100 ºC vs 200 ºC)

Key findings (1) : illitization occurs, T plays key role and bentonite-host rock interaction is important

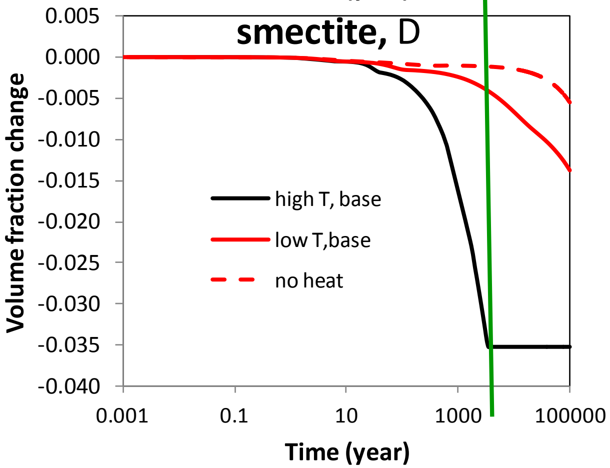








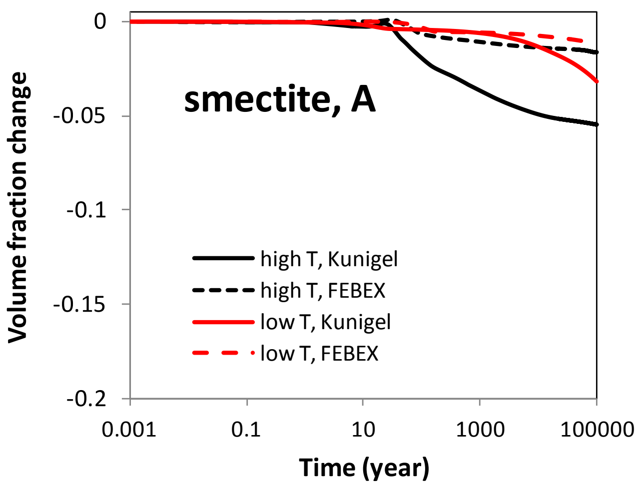
The temporal evolution of smectite volume fraction at points A, B, C, and D for Kunigel-VI bentonite.

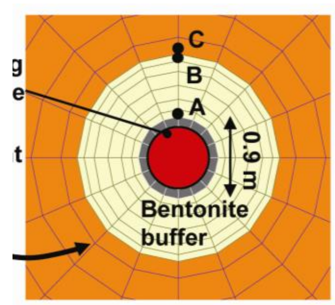


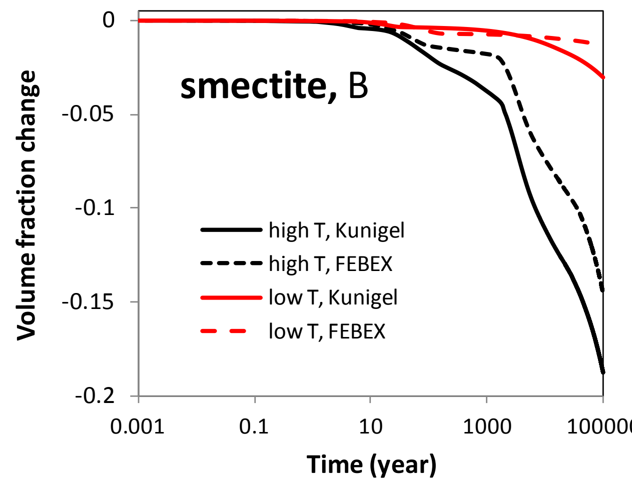
In early times, dissolution of k-felspar supplies K for illitization; after about 3000 years, illitization in host rock stops and K is transported into bentonite which leads to very different illitization at points A and B

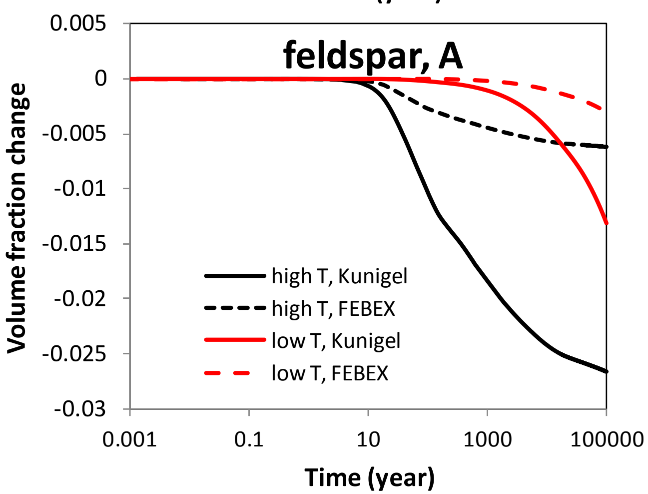
Exploratory Generic Models for EBS (100 ºC vs 200 ºC)

Key findings (2) :Type of bentonite matters and supply of K and Al is the key

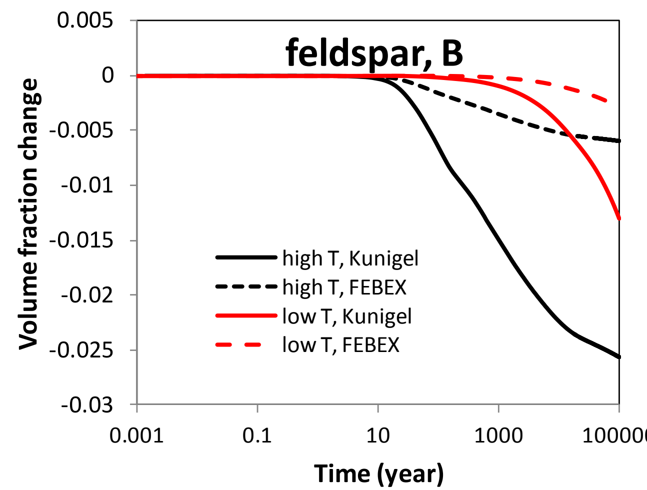






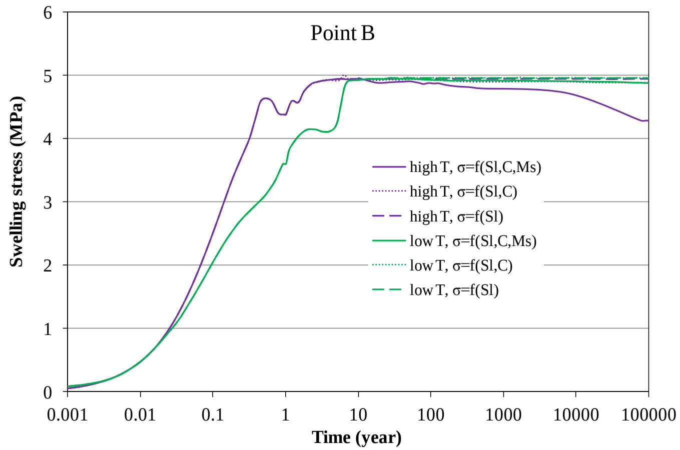


The temporal evolution of smectite and K-fledspar volume fraction at points A and B for Kunigel-VI and FEBEX bentonite.



Exploratory Generic Models for EBS (100 ºC vs 200 ºC)

Key findings (3) : Swelling stress decreases as a result of chemical changes and such decrease varies case by case

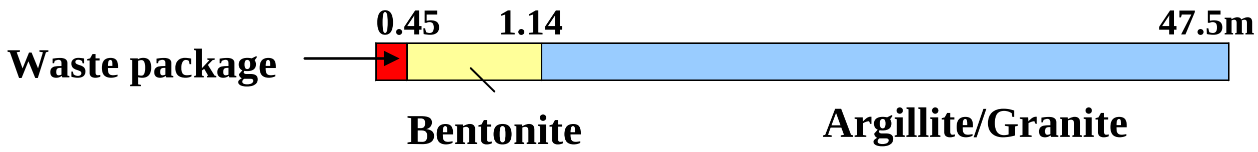


The geochemically induced swelling stress for Kunigel and FEBEX bentonite at points A and B for "high T" scenario

	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%

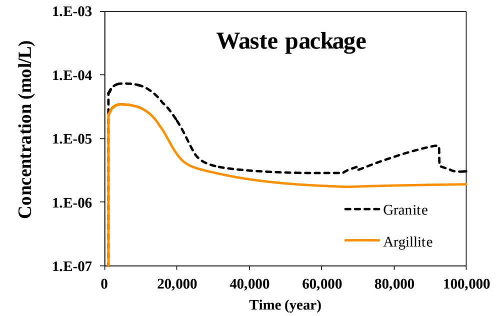
Exploratory Generic Models for EBS at 100 ºC (Argillite vs Granite)

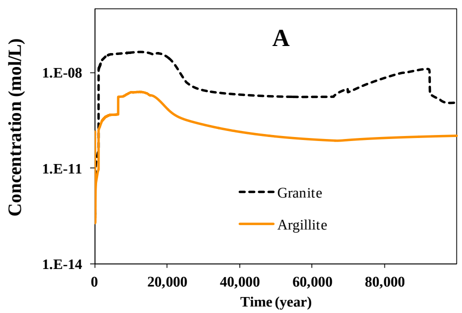
The importance of EBS-host rock interaction to migration of radionuclides — an example

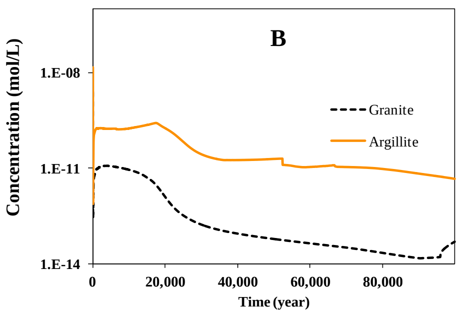


Key features:

- Two models with same EBS (FEBEX bentonite) but different host rocks: argillite (based on Opalinus Clay) and granite (based on Beishan granite, China)
- U(VI) is released by dissolution of Schoepite after 1000 years
- U(VI) undergoes diffusion, aqueous complexation (e.g. $Ca_2UO_2(CO_3)_3$) and adsorption via surface complexation in bentonite







Different host rocks → Different geochemical conditions in EBS → Different dissolution of Schoepite and adsorption of U

Roadmap to Study Long-term Stability of EBS

Developing simulators for coupled THMC processes

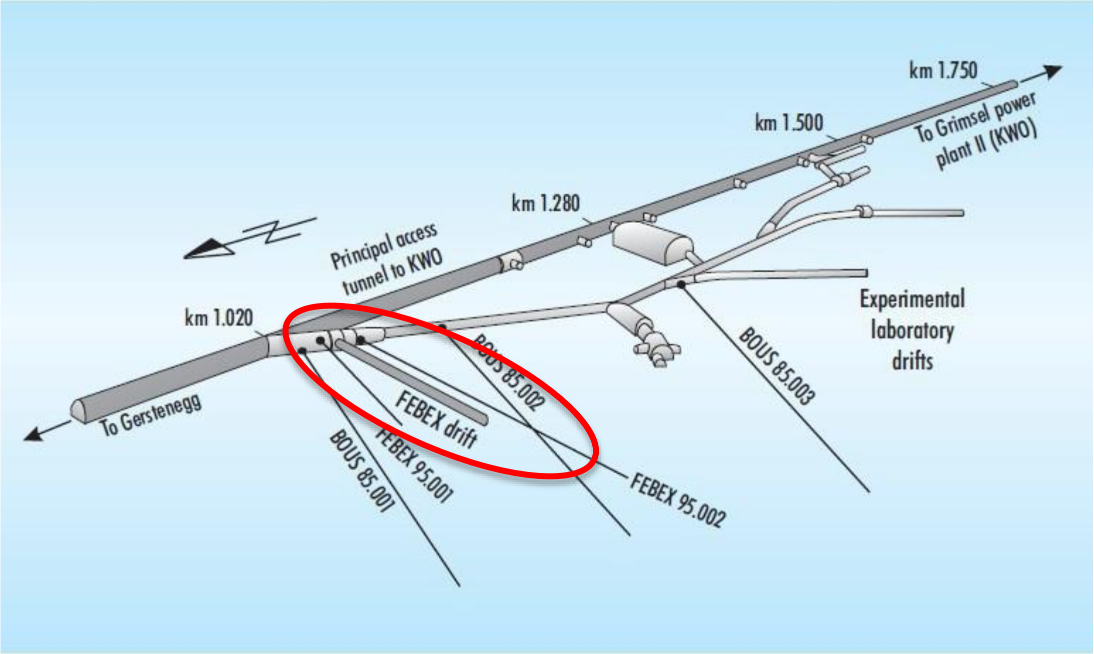
Evaluating key THMC processes with FEBEX "in situ" experiment at 100 ^{o}C

Studying long-term alteration of bentonite under high temperature (200 ^{o}C) with exploratory generic models

Further testing THMC model with experiments at 200 ^{o}C, and improving understanding and reducing uncertainties by multi-scale experiments and modeling

Further Testing THMC Models with Field Test HotBENT

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 ^{o}C to 200 ^{o}C.

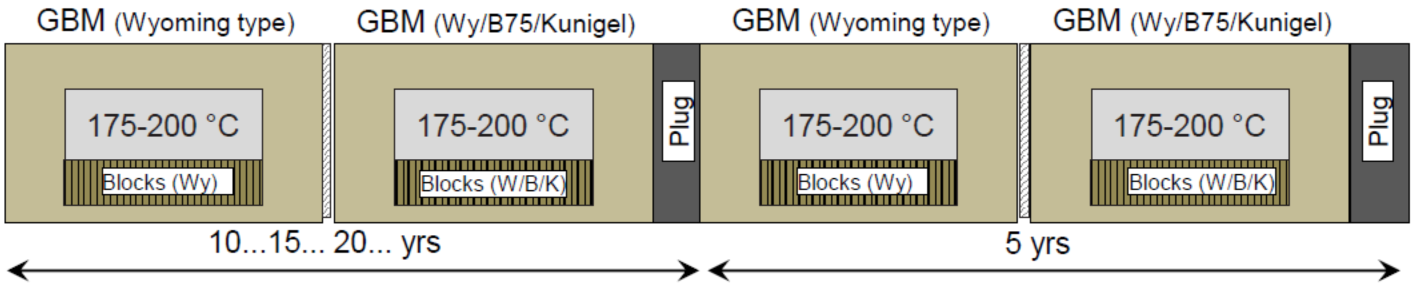


Participating organizations:

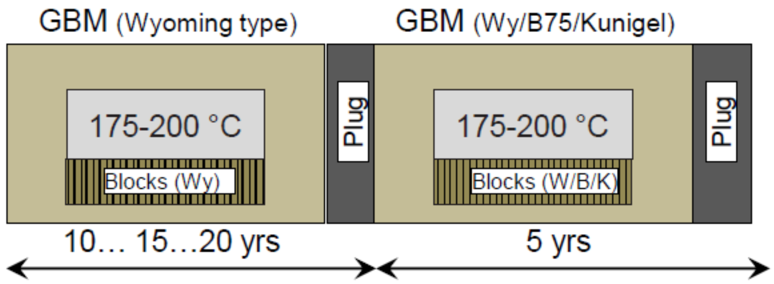
NAGRA (Switzerland), **DOE(**USA**), NWMO (**Canada**), NUMO (**Japan**), RWM (**UK**), SÚRAO (**Czech Republic)

Further Testing THMC Models with Field Test HotBENT

HotBENT modular design - example



Basis for detailed design

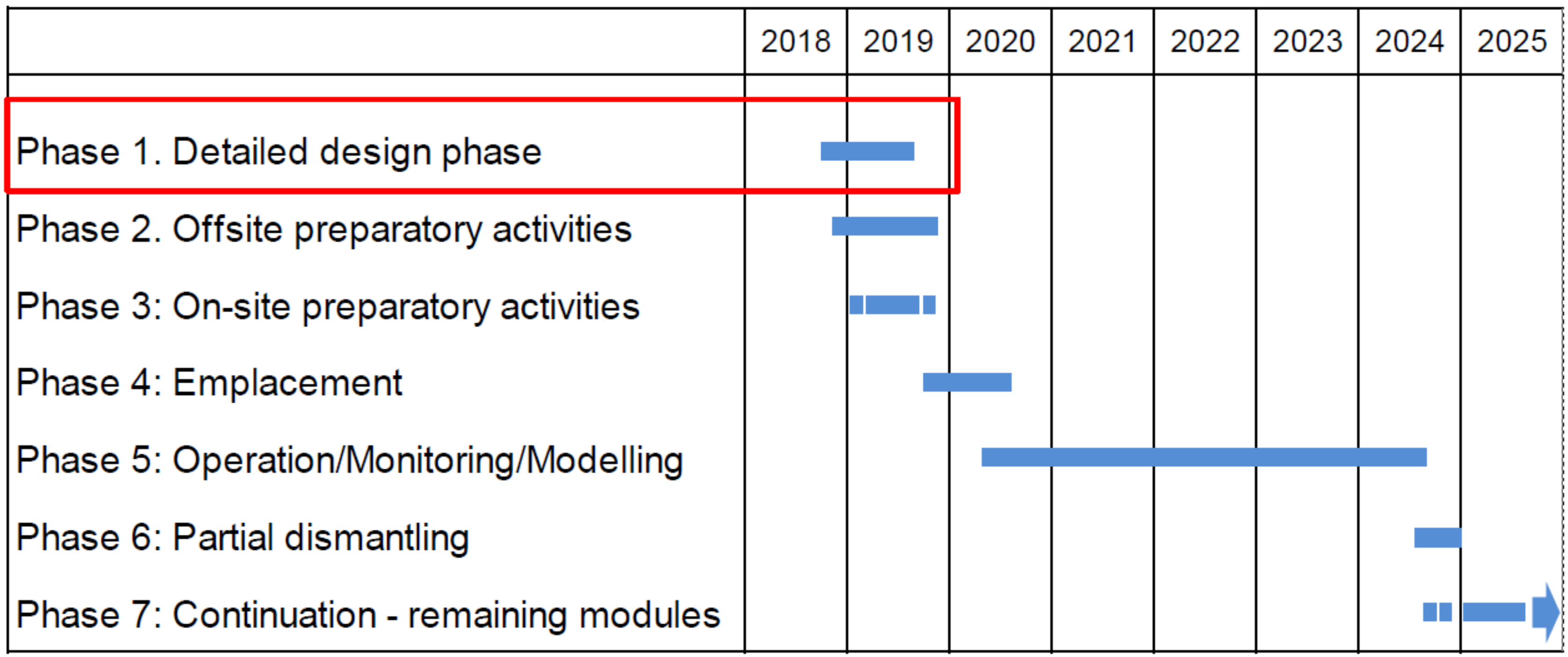


Total Budget (depending on number of modules)**: 5 – 8 Mio CHF**

App. costs per partner (depending on number of partners)**: 1 – 1.3 Mio CHF over 5 years**

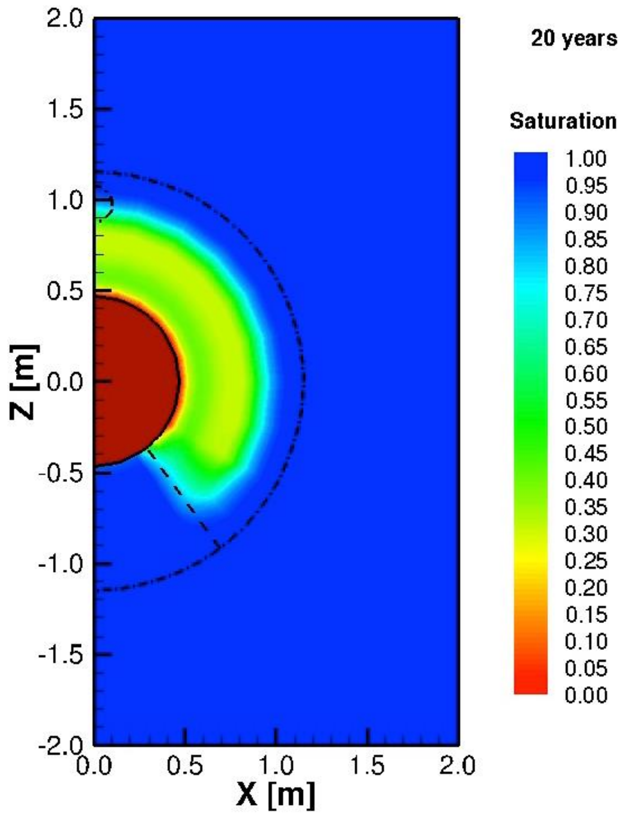
Further Testing THMC Models with Field Test HotBENT

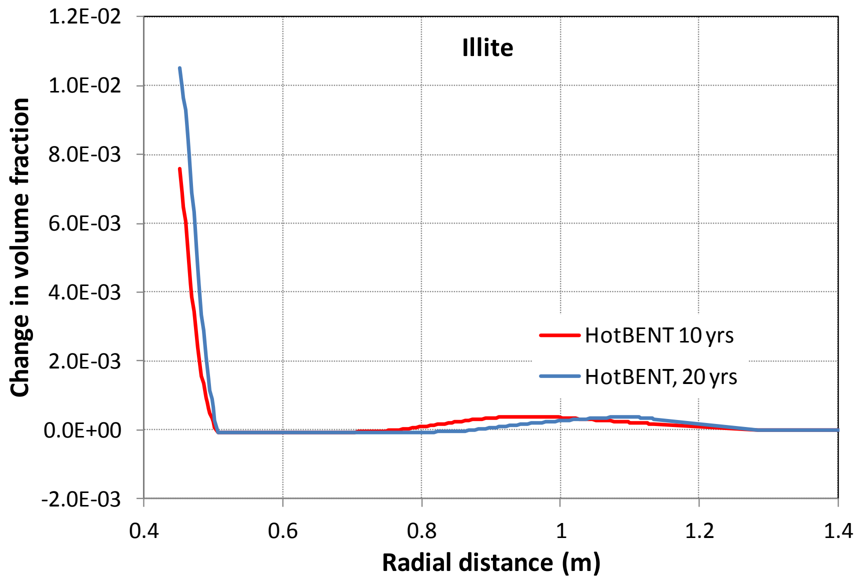
Timeline for the HotBENT experiment



Further Testing THMC Models with Field Test HotBENT

Scoping calculation results - example





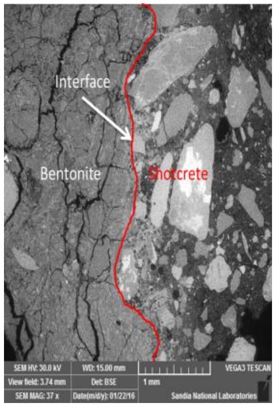
Liquid saturation evolution in vertical X-Z cross sections using Wyoming bentonite properties in a 3-D TH model *(Finsterle et al., 2017)*

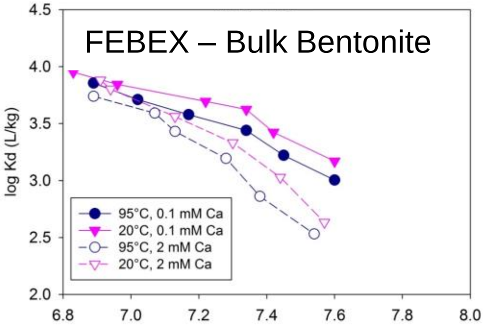
Smectite and illite profile from a 1-D model similar to the model for FEBEX *in situ* test but with temperature boundary of 200 ^{o}C.

If HotBENT with 200 ^{o}C uses FEBEX bentonite, only moderate increase in illitization is expected.

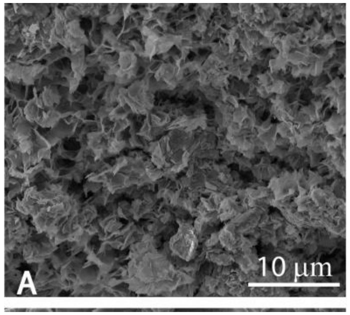
Multi-scale Experiments and Modeling for Better Understanding

- Studying the micro-scale structure of FEBEX bentonite
  - Back-Scattered Electron Image (BSEI) of Bentonite – Cement Interface
  - Bulk Bentonite Analyses using XRF, TGA and XRD
  - Synchrotron X-ray Microtomography Measurements
- U(VI) Sorption and diffusion experiments with FEBEX Bentonite





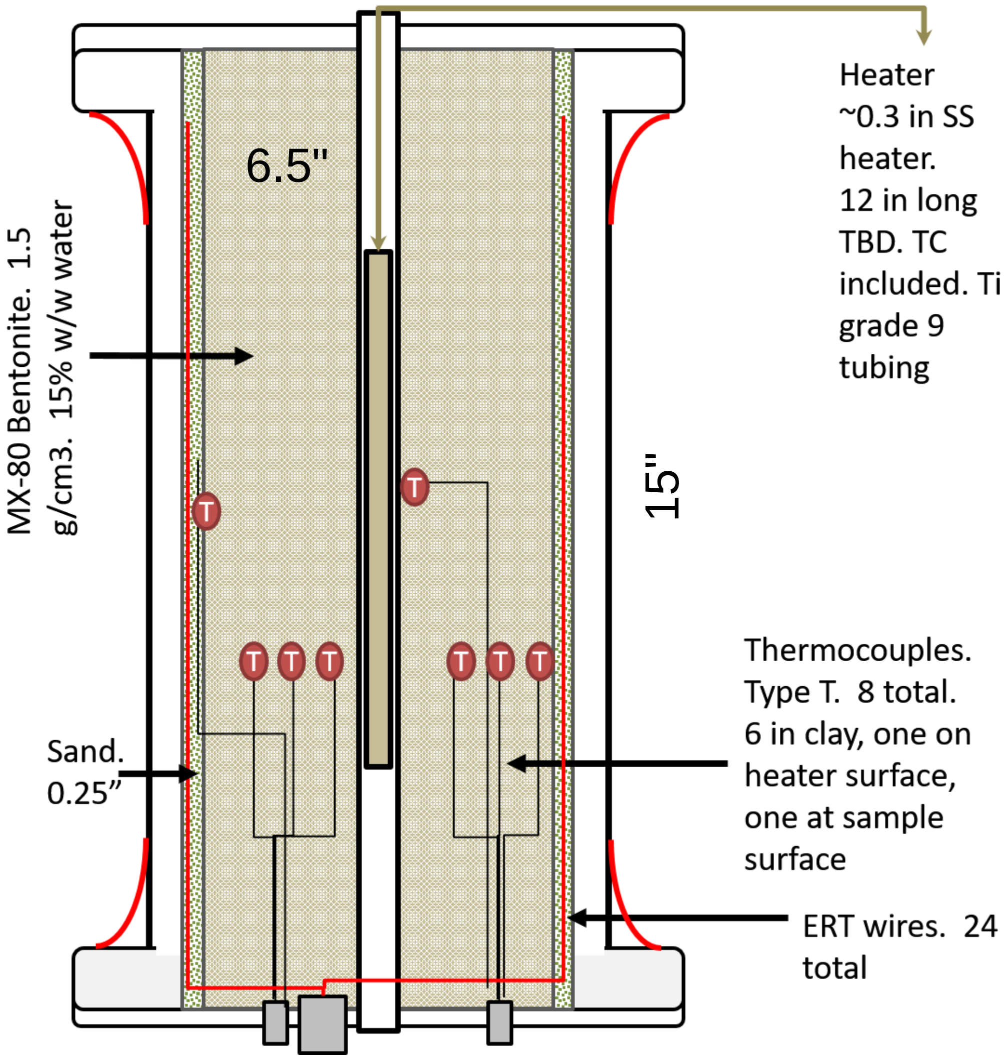
- Experimental studies of geochemical alteration of bentonite under higher temperature



Post-reaction SEM images showing montmorillonite foils displaying partially deteriorated edges from an isothermal 300 °C experiments (Cheshire et al., 2014)

Multi-scale Experiments and Modeling for Better Understanding

A cylindrical bentonite column with a 200 ^{o}C heater in the middle



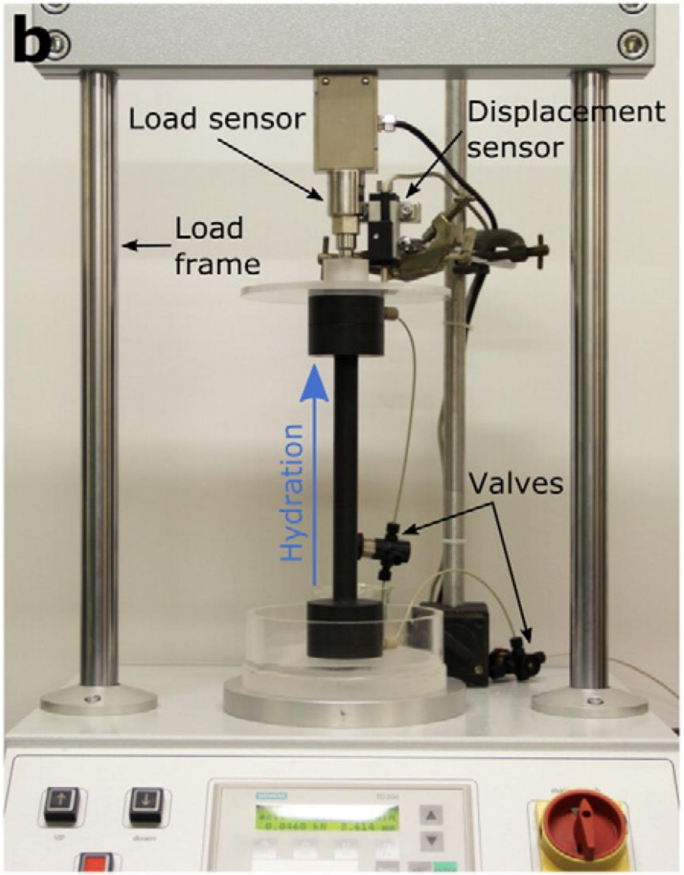
Experimental Summary:

- 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

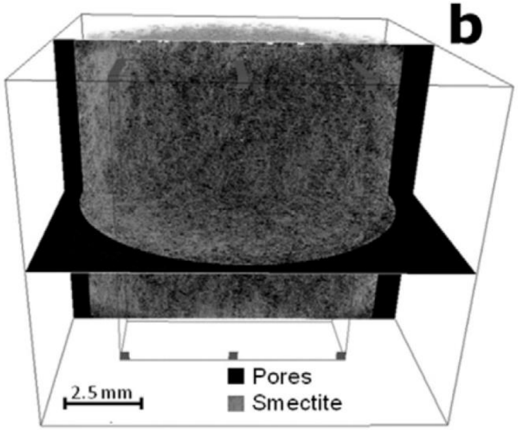
Multi-scale Experiments and Modeling for Better Understanding

Study chemical controls on smectite structure and swelling

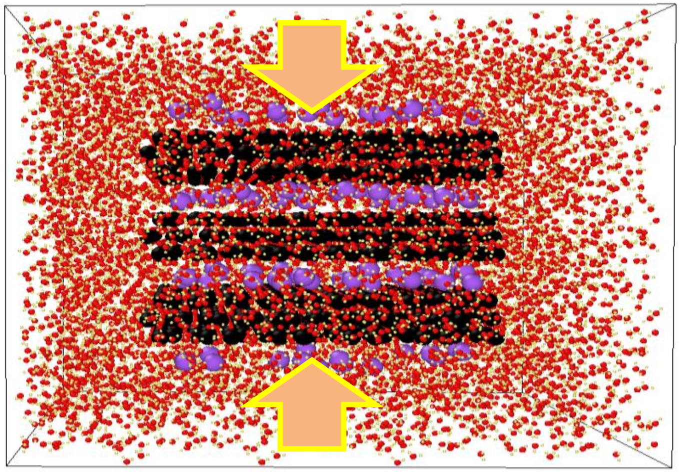
X-ray compatible oedometer



µXCT of pore development during hydration



Molecular predictions of swelling pressure



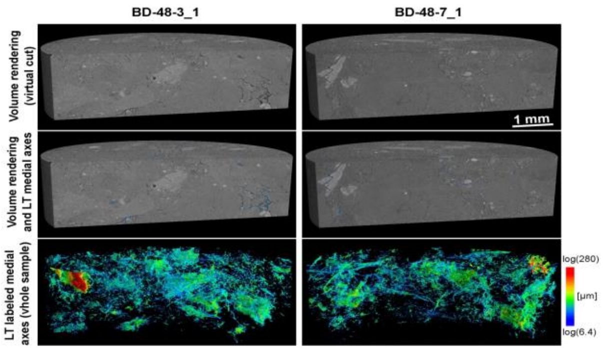
Integration with Generic Disposal R&D

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

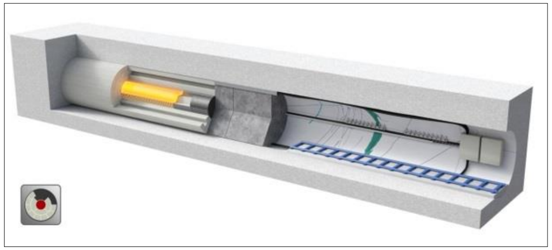
- Developing advanced modeling tools
- Constructing multi-physics coupled process models
- Testing models with large scale experiments

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

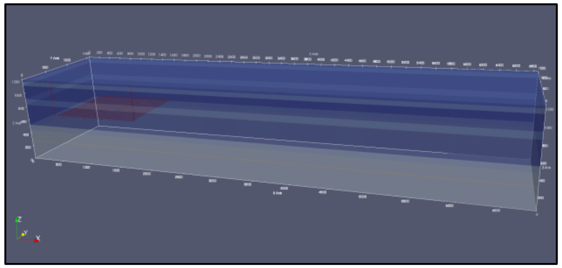
Micro-structural analysis



Field Experiments



Generic PA Modeling



Questions?

Clean. **Reliable.** **Nuclear.**

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