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

Spent Fuel and Waste Science and Technology (SFWST)

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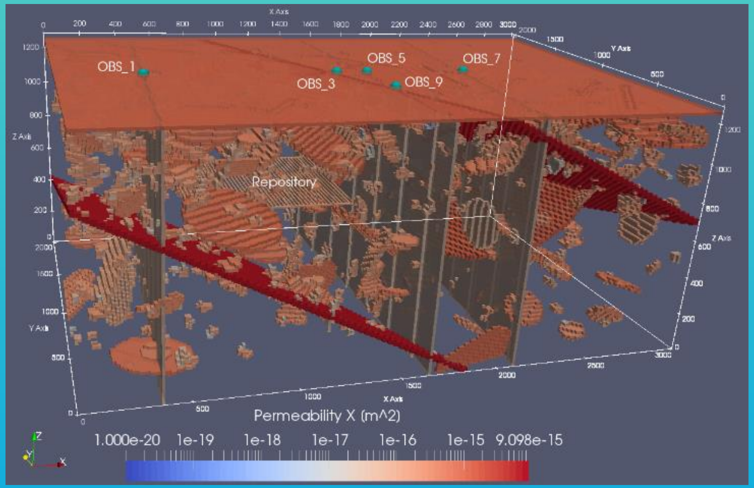

Argillite Host Rock: Disposal Concepts and Research & Development Activities

Nuclear Waste Technical Review Board
Fall 2020 Board Meeting
December 2 and 3, 2020
Online Virtual Meeting

Carlos F. Jové Colón
Sandia National Laboratories (SNL)
SAND2020-13133 PE

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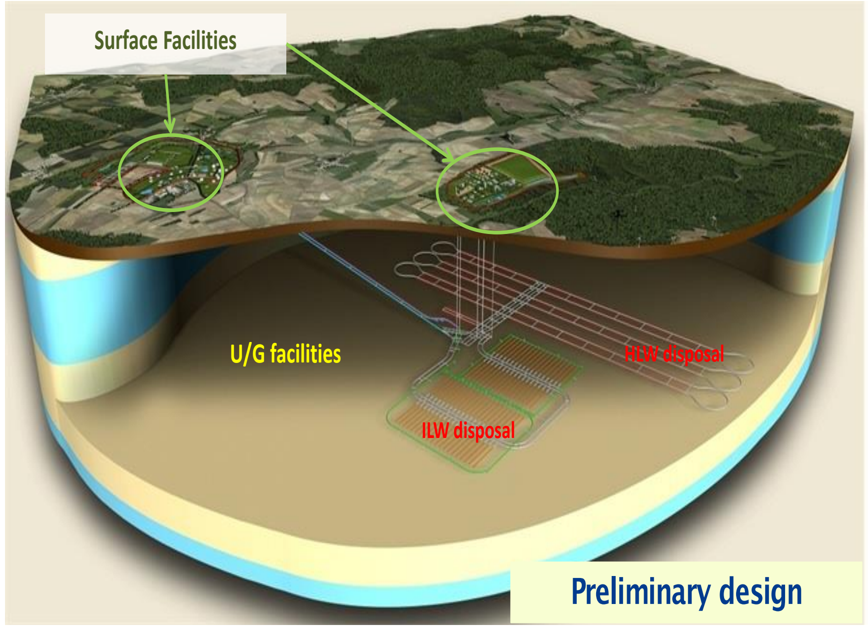
Outline

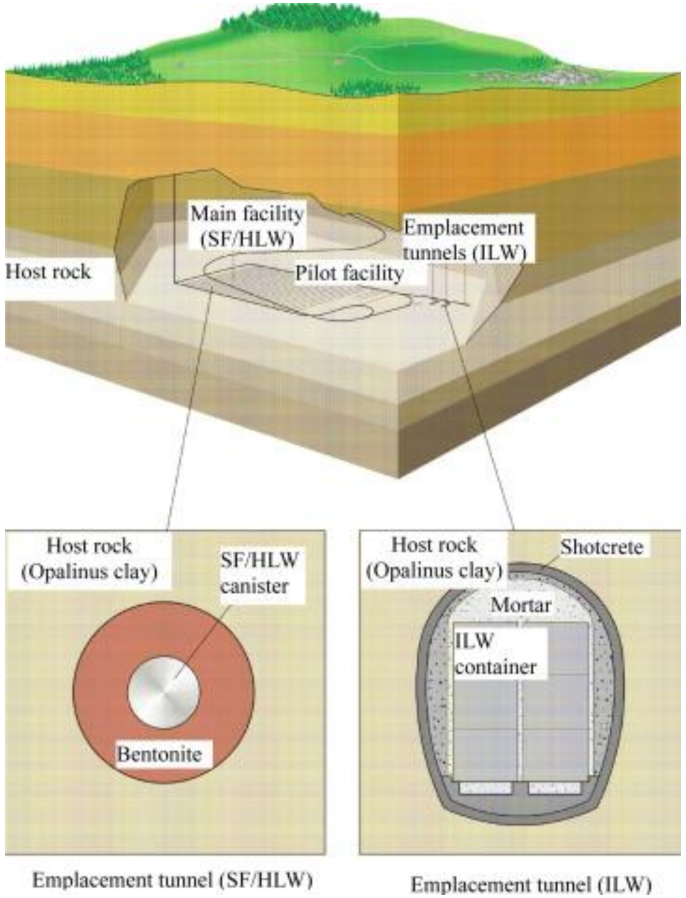
- Argillite Repository Concept
- Argillaceous Host Rock Characteristics
- Argillite Post-Closure Safety Strategy
- Waste Form and Engineered Barrier in Argillite
- Argillite Reference Case
- Knowledge Gaps & R&D Priorities
- Repository Relevant Processes
- Highlights – Disposal in Argillite R&D
- Summary

Argillite Repository Concept

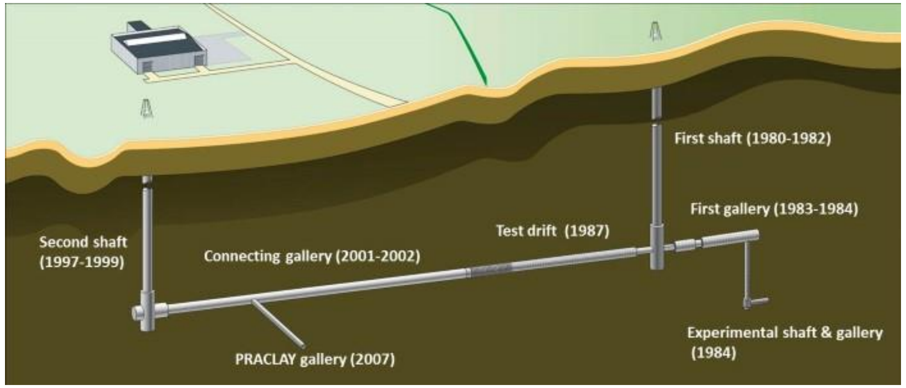


High-Level radioactive waste disposal (ANDRA) – **COx Argillite** (Bildstein and Claret 2015)

U/G = Underground
ILW = Intermediate Level Waste
HLW = High Level Waste
SF = Spent Fuel



Swiss repository concept (Delage et al. 2010) - **Opalinus Clay**



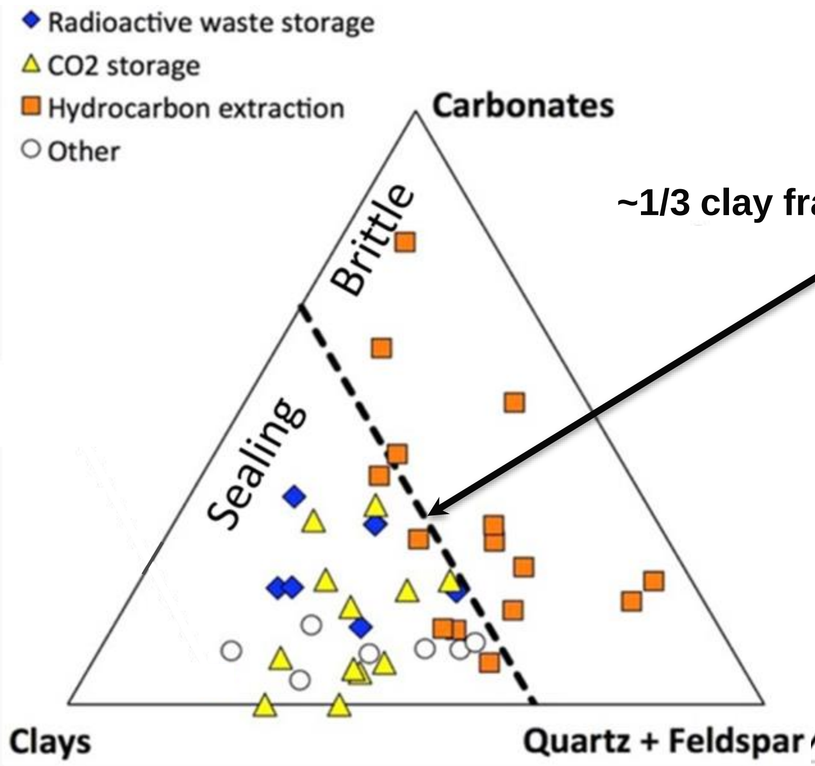
Belgian repository concept- HADES Underground Laboratory – **Boom Clay**
(https://science.sckcen.be/en/Facilities/HADES)

Argillite Host Rock Characteristics

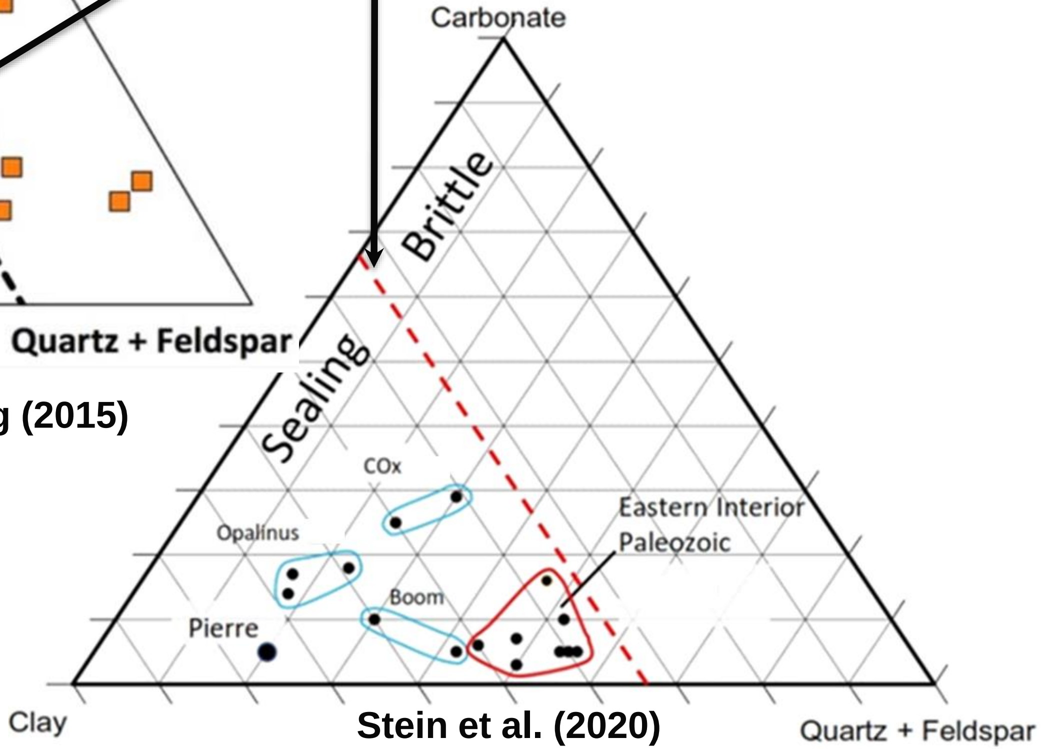
- Low permeability
- Low hydraulic gradients
- Low diffusion coefficients
- Good sorption capacity
- Widespread geologic occurrence
- Appropriate thickness and depth for nuclear waste disposal concepts
- Found in stable geologic settings
- Self-sealing properties

Bulk mineralogy: illitic clay, quartz, kaolinite, chlorite, some carbonate; minor feldspar, and pyrite

~1/3 clay fraction line after Bourg (2015)

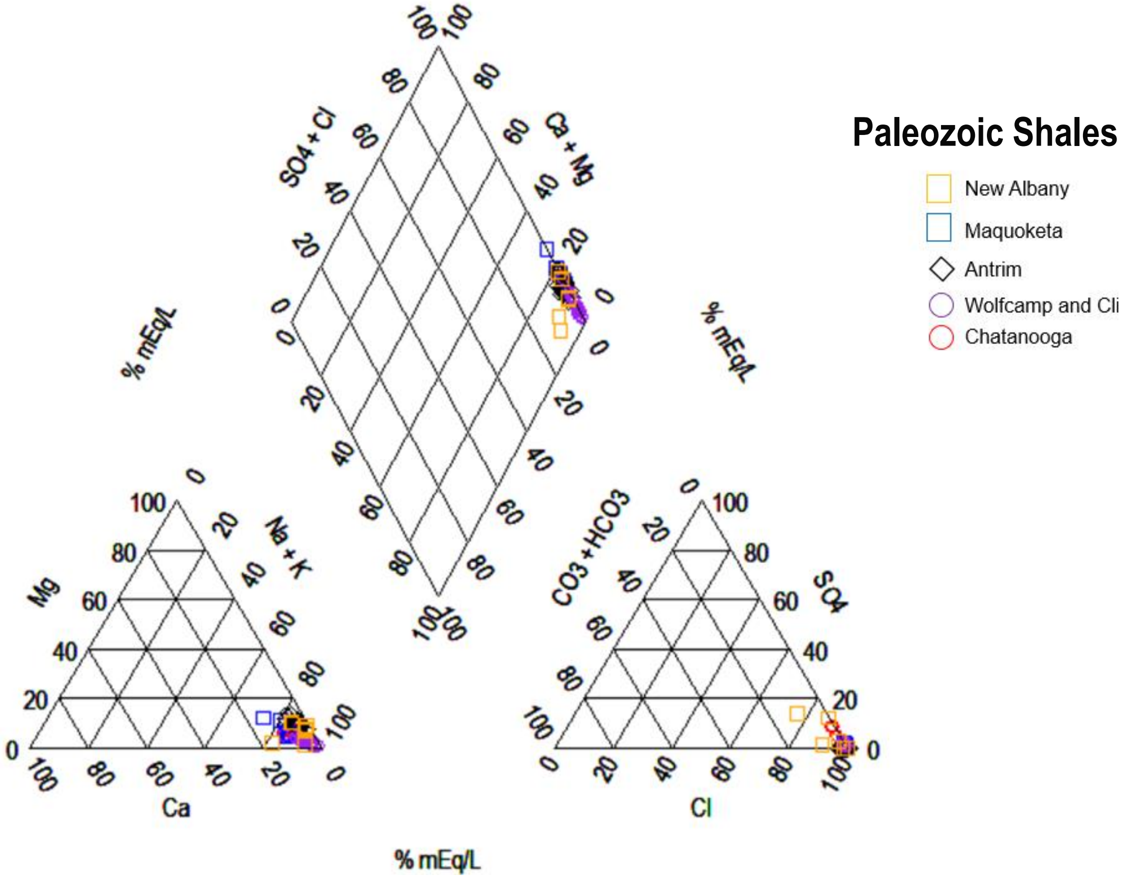


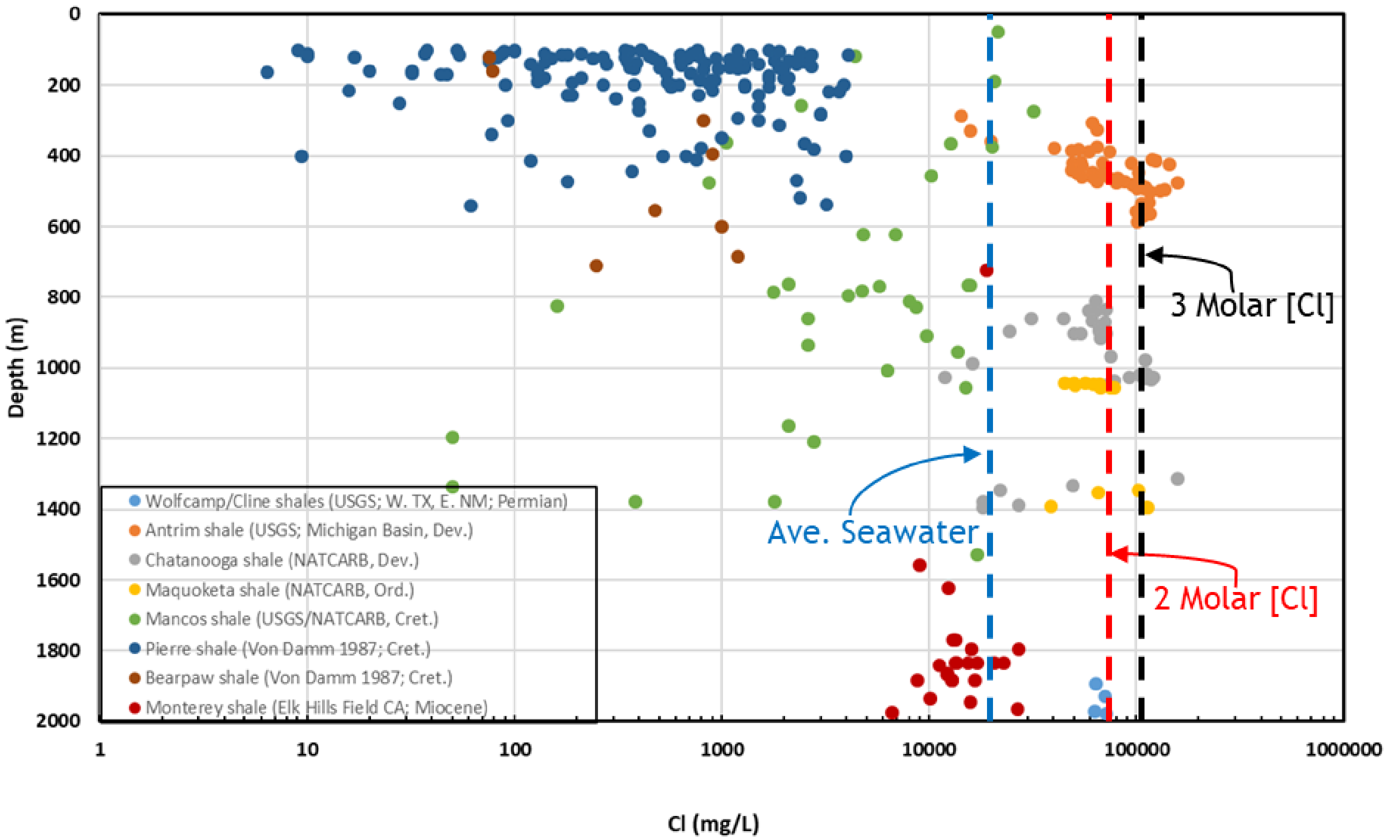
Modified after Bourg (2015)
Stein et al. (2020)



Stein et al. (2020)

Porewater Chemistry in Argillaceous Formations





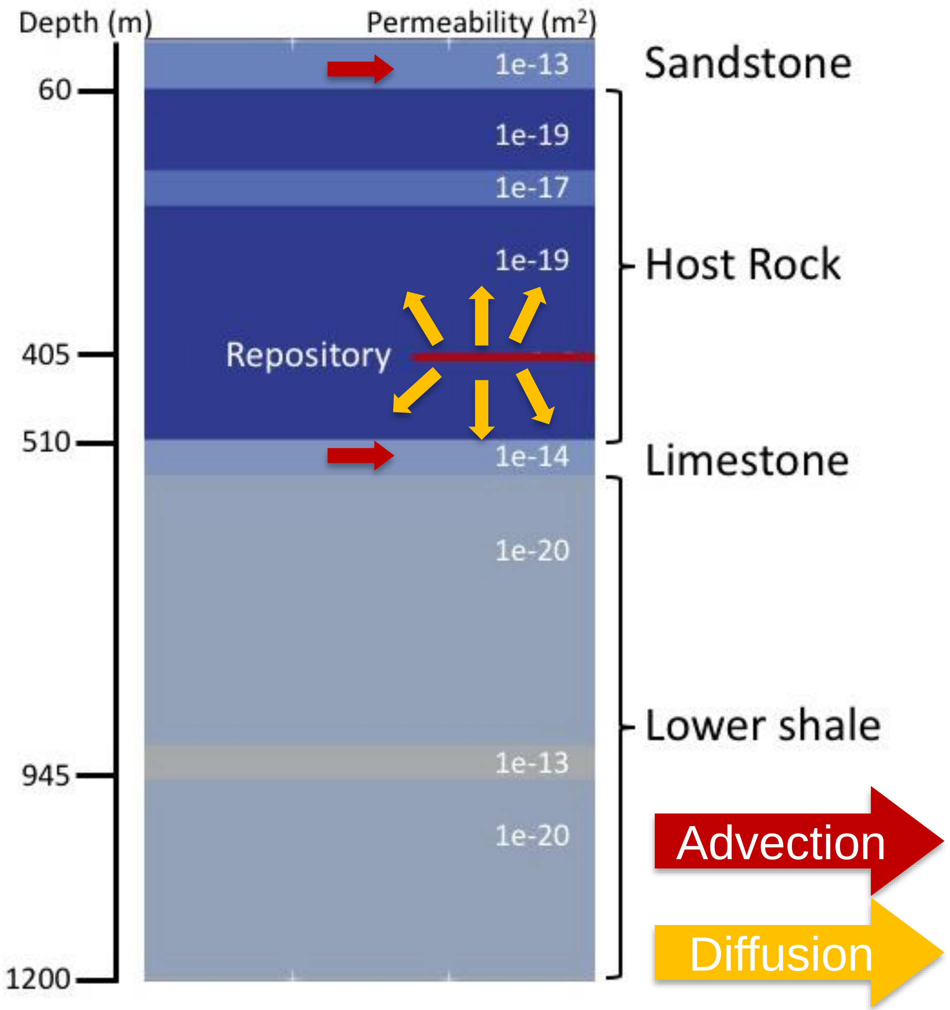
- Porewater compositions are highly variable
- Overall: Sodium chloride brines with some calcium and carbonate

Sources: United States Geological Survey (USGS) produced water (Blondes et al. 2018); NATCARB (Bauer et al. 2018); WATSTORE (von Damm 1987)

Stein et al. (2020)

Argillite Post-Closure Safety Strategy

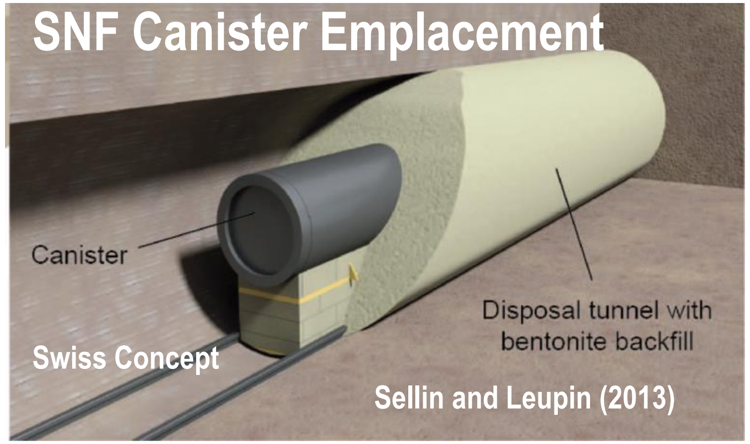
- Containment
  - Waste package is isolated by depth
  - Protected by buffer/backfill
  - Diffusion-dominated
  - Reducing conditions
  - Overpack integrity (100 yr to >10,000 yr)
- Limited Release
  - Fuel degradation / corrosion is slow in reducing environment
  - Highly effective retardation in host rock due to
    - Low permeability
    - Low effective diffusion coefficient
    - High sorption capacity



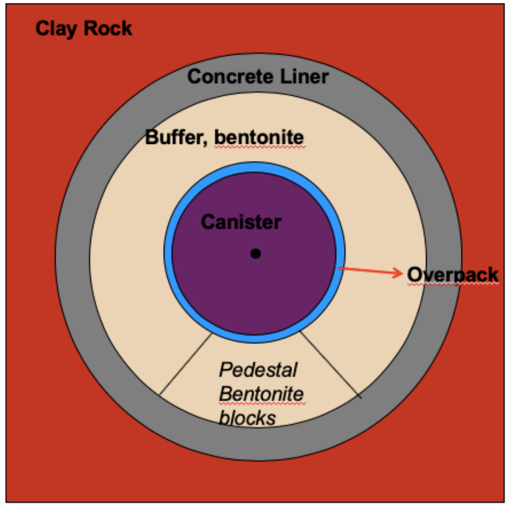
Generic stratigraphic column for argillite reference case (Sevougian et al. 2019)

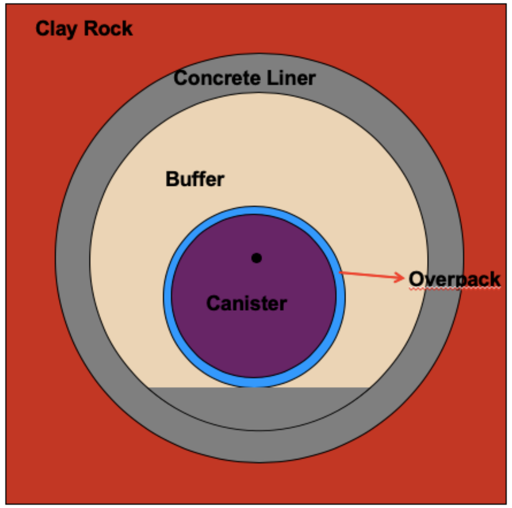
Waste Form and Engineered Barrier in Argillite

- Glass High-Level Waste
  - Vitrified glass log in waste package
  - Horizontal emplacement boreholes
  - Bentonite buffer
- Spent nuclear fuel (SNF) in 4-PWR waste package
  - Horizontal emplacement boreholes
  - With or without bentonite buffer
- SNF in 12-PWR waste package
  - In-drift axial emplacement
  - Bentonite buffer with or without additives
- SNF in 21-PWR to 37-PWR waste package
  - In-drift axial emplacement
  - Bentonite buffer with additives or crushed rock backfill
  - 4 kW per waste package thermal power limit



(21-37)-PWR

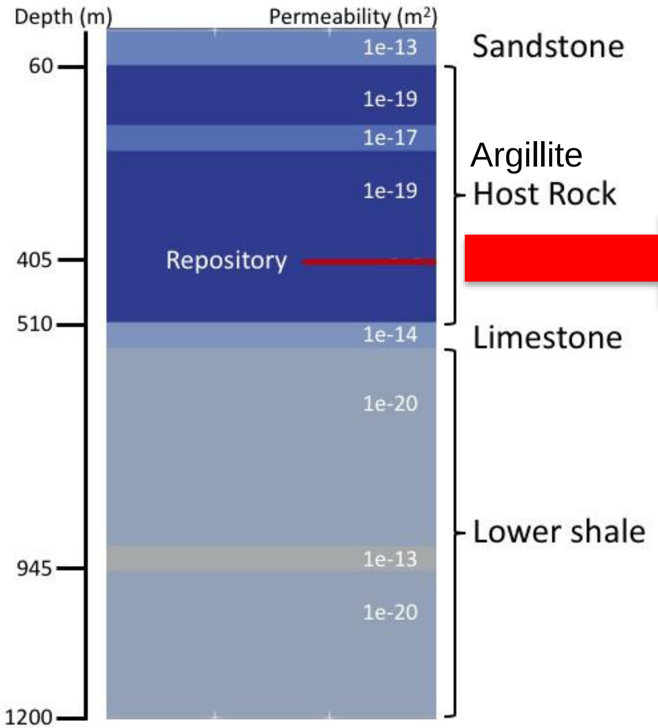




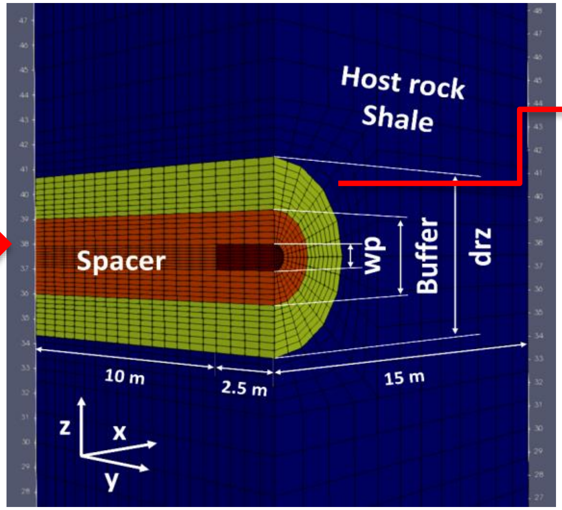
PWR = pressurized water reactor (assembly); represents waste package capacity

Jove Colon et al. 2014; Mariner et al. 2017; Sevougian et al. 2019; Stein et al. 2017; 2020

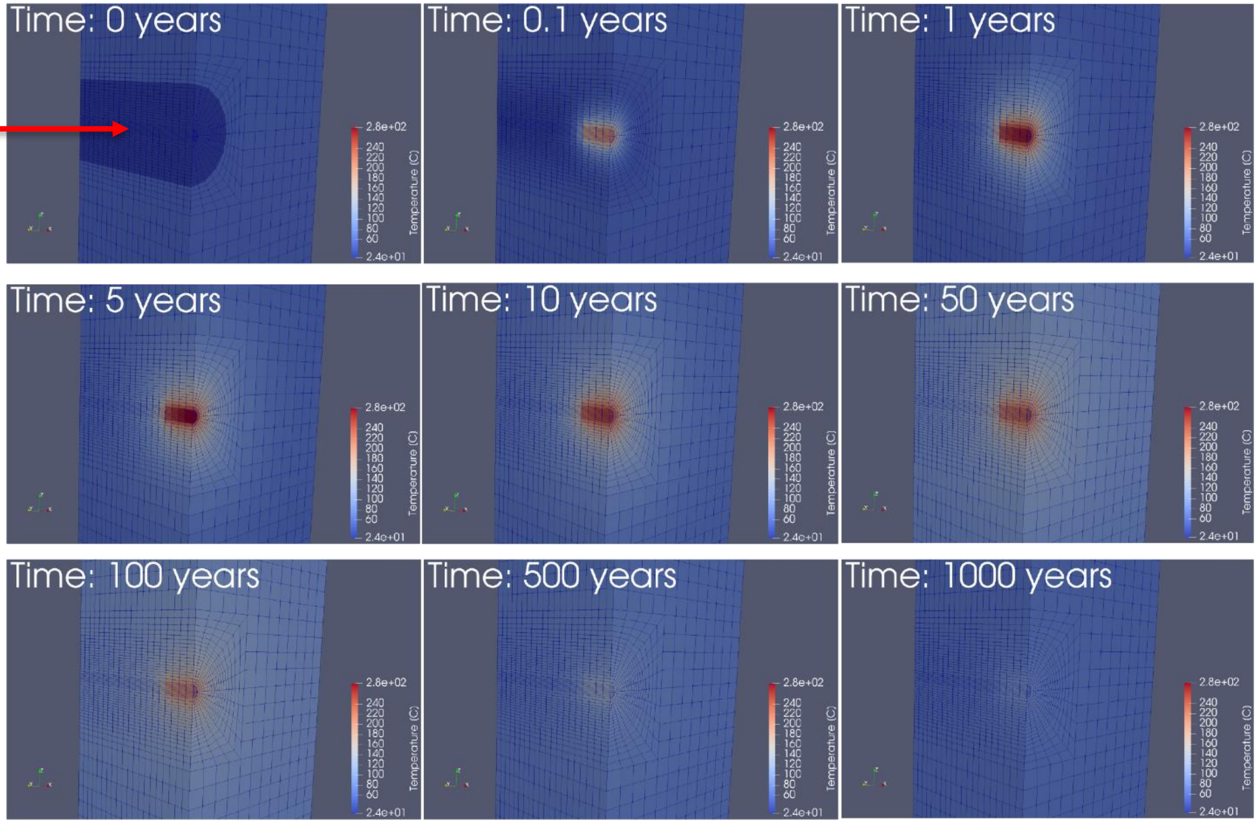
Argillite Reference Case: Deterministic Simulations of Generic Disposal in Argillite



Generic stratigraphic column for argillite reference case



Near-field model domain used in the simulation



Simulation temperature for 24-PWR heat source

Sevougian et al. (2019)

2019 Roadmap Update: High Impact Topic Groups with High and Medium-High Priority R&D Activities Scores

High Impact R&D Topics	High-Priority R&D Activities	Medium-High-Priority R&D Activities
High Temperature Impacts	D-1, D-4, I-4, I-6, I-16*, E-11, S-5	I-2, I-3, I-7, E-10
Buffer and Seal Studies	I-4, E-9, E-17*, A-8, C-15*	I-2, I-3, I-7, A-4, C-6, C-8, C-11
Coupled Processes (Salt)	S-1, S-3, S-4, I-12, I-13	I-14, S-2, S-7, S-8, S-11*
Gas Flow in the EBS	I-6, I-8, I-18*	I-9, P-17*
Criticality	D-1, D-3, D-4, D-5	
Waste Package Degradation	C-16*, P-12	E-4*, E-6
In-Package Chemistry	E-14*	E-2, E-20, P-15*, P-16*
Generic PA Models		P-1, P-2, P-4, P-11*, P-13*, P-14
Radionuclide Transport		C-11*, C-13*, C-14*, P-15*, P-16*
DFN Issues		I-21*, C-1, C-17*
GDSA Geologic Modeling		O-2, O-3
THC Processes in EBS		E-3

Activity Designator Legend:

A – Argillite
C – Crystalline
S – Salt
D – Dual Purpose Canisters
E – Engineered Barrier System
I – International
O – Other
P – Performance Assessment
* – indicates Gap Activity

2019 Roadmap Update: High-Priority R&D Activities

High Priority R&D Activities	
A-08	Evaluation of ordinary Portland cement (OPC)
C-15*	Design improved backfill and seal materials
C-16*	Development of new waste package concepts and models for evaluation of waste package performance for long-term disposal
D-01	Probabilistic post-closure DPC criticality consequence analyses Task 1 - Scoping Phase Task 2 - Preliminary Analysis Phase Task 3 - Development Phase
D-03	DPC filler and neutron absorber degradation testing and analysis
D-04	Coupled multi-physics simulation of DPC postclosure (chemical, mechanical, thermal-hydraulic) including processes external to the waste package.
D-05	Source term development with and without criticality
E-09	Cement plug/liner degradation
E-11	EBS High Temp experimental data collection- To evaluate high temperature mineralogy /geochemistry changes.
E-14*	In-Package Chemistry
E-17*	Buffer Material by Design

High Priority R&D Activities	
I-04	Experiment of bentonite EBS under high temperature, HotBENT
I-06	Mont Terri FS Fault Slip Experiment
I-08	DECOVALEX-2019 Task A: Advective gas flow in bentonite
I-12	TH and THM Processes in Salt: German-US Collaborations (WEIMOS)
I-13	TH and THM Processes in Salt: German-US Collaborations (BENVASIM)
I-16*	New Activity: DECOVALEX Task on Salt Heater Test and Coupled Modeling
I-18*	New Activity: Other potential DECOVALEX Tasks of Interest: Large-Scale Gas Transport
P-12	WP Degradation Model Framework
S-01	Salt Coupled THM processes, hydraulic properties from mechanical behavior (geomechanical)
S-03	Coupled THC advection and diffusion processes in Salt, multi-phase flow processes and material properties in Salt
S-04	Coupled THC processes in Salt, Dissolution and precipitation of salt near heat sources (heat pipes)
S-05	Borehole-based Field Testing in Salt

Activity Designator Legend:

A – Argillite

C – Crystalline

S – Salt

D – Dual Purpose Canisters

E – Engineered Barrier System

I – International

O – Other

P – Performance Assessment

* – indicates Gap Activity

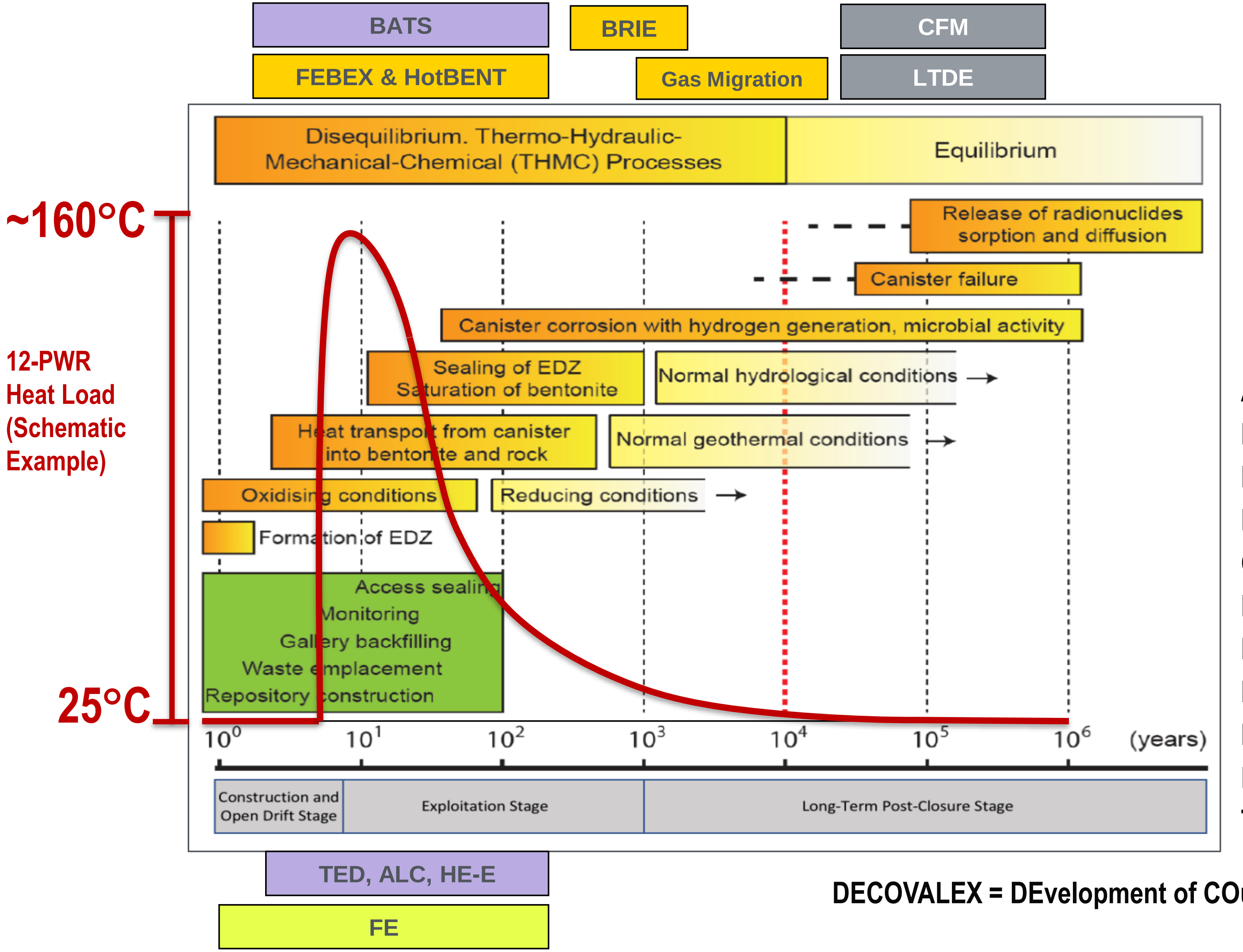
High Priority Argillite Activities

#	Description	Purpose	Crosscut	SFWST Argillite R&D Activity	Int’l Tie-in
A-08 E-09 C-15	Evaluation of Ordinary Portland Cement (OPC) Cement plug/liner degradation Design improved backfill and seal materials	**Evaluation of mineralogical alteration evolution in seals and liners**	**Crystalline, Engineered Barrier System (EBS)**	Experimentally verified cement-geomaterial 3D reactive transport model development in PFLOTRAN Experimental studies of barrier material interactions (cement-bentonite-metal)	DECOVALEX2019 – Task C EBS Task Force – Cement Task
E-11 E-17	EBS High Temperature Geochemistry/ Mineralogy Buffer Material by Design	**Evaluation of mineralogical alteration at buffer/waste package interface**	**Crystalline, Engineered Barrier System (EBS)**	Hydrothermal experiments evaluating host rock, buffer, buffer additives, and canister materials interactions at elevated temperatures	HotBENT

High Priority Argillite Activities (cont.)

#	Description	Purpose	Crosscut	SFWST Argillite Activity	Int'l Tie-in
I-06 I-08	Mont Terri Fault Slip Experiment Advective gas flow in bentonite	**Evaluation of transport effects and evolution in seals, and bentonite backfill**	**Crystalline, Engineered Barrier System (EBS)**	Fault activation and self-sealing in argillite Multiphase flow bentonite Studies at various scales	DECOVALEX 2019 - Mont Terri Fault Slip Experiment DECOVALEX 2023 • Task B: Modeling Gas Advection in Clay (MAGIC) • Task C, D: THMC Modeling, Heater Test Experiments
I-04 E-14	Experiments of Bentonite under High Temperature In-Package Chemistry (**Gap Activity**)	**Evaluation of barrier alteration, transport, & chemical effects in backfill & canister materials**	**Crystalline**	Benchtop High Temp Bentonite Column Test & modeling In-package material interactions modeling & experiments	HotBENT Column Test, EBS Task Force -

Repository Phases and Relevant Processes: Cross-Cuts With International Partnerships



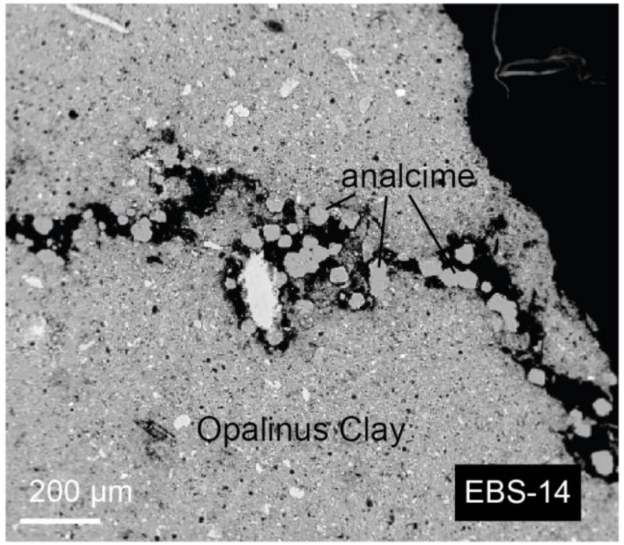
Key R&D Issues
Near-Field Perturbation
Engineered Barrier Integrity
Flow and Radionuclide Transport
Demonstration of Integrated System Behavior

ALC = Full-scale Emplacement Experiment (France)
BATS = Heated Brine Availability Test in Salt (USA)
BRIE = Bentonite Rock Interaction Experiment (Sweden)
FEBEX = Full-Scale Engineered Barrier Exp. (Switzerland)
CFM = Colloid Formation Migration (Switzerland)
FE = Full-scale Emplacement Experiment (Switzerland)
HE-E = Heater Experiment in Micro-tunnel (Switzerland)
HotBent = High-Temperature Heater Test (Switzerland)
HLW = High Level Waste
LTDE = Long-Term Diffusion Sorption Experiment (Sweden)
TED = Thermal Experiment (France)

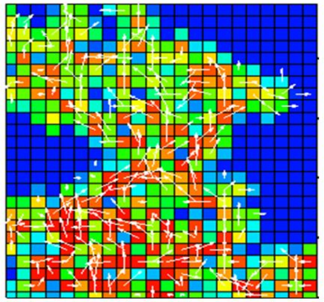
DECOVALEX = DEvelopment of COupled Models and their VALidation Against EXperiments

Highlights – Disposal in Argillite R&D

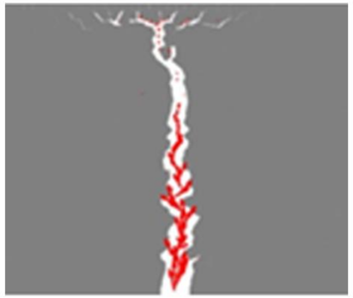
- **High temperature** experiments of bentonite interactions with barrier materials and host rocks: granodiorite & **Opalinus Clay**
- **Development of a preliminary GDSA reference case** for disposal in argillite media
- **Advances in Thermo-Hydrological-Mechanical (THMC) modeling approaches** of bentonite barrier, argillite rock, and excavated disturbed zone (EDZ fracture/damage behavior) & gas migration
- **Thermodynamic modeling** of bentonite – barrier material interactions & thermodynamic database development
- **Non-isothermal 1D-3D Thermo-Hydrological-Chemical (THC) reactive transport modeling**
- **International collaborations**:
  - **DECOVALEX19: PFLOTRAN Hydrological- Chemical (HC) modeling of barrier interactions**
  - DECOVALEX2023: Gas transport in clays, TH modeling (just started!)



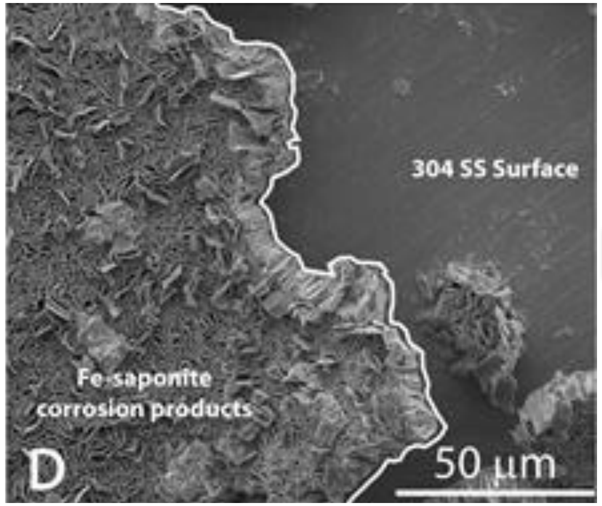
Continuum model approach using TOUGH-FLAC

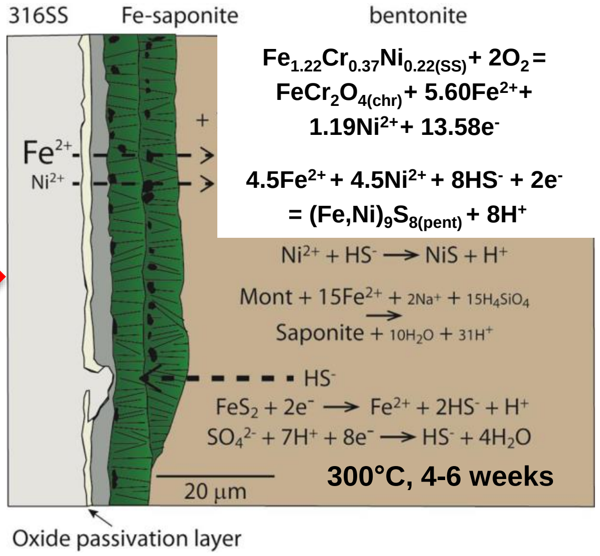


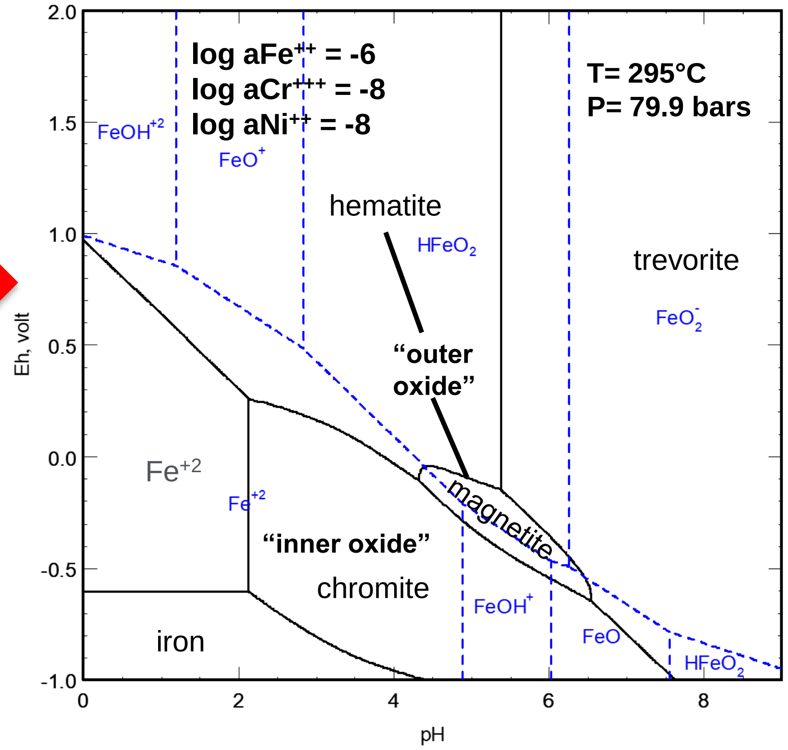
Discrete fracture model approach using TOUGH-RBSN



Past Experiments: Steel – Clay Interactions







- **Experiment**
  - **T = 300ºC; STRIPA brine**
  - **Wyoming Bentonite**
  - **316 & 304 stainless steel (SS)**

- **Corrosion products**
  - **Uniform corrosion (no pitting)**
  - **Chromite passivation layer**
  - **Fe-rich smectite (Fe-saponite), Chlorite**
  - **Pentlandite $(Fe,Ni)_9S_8$**
  - **Millerite (NiS)**

Pourbaix diagram

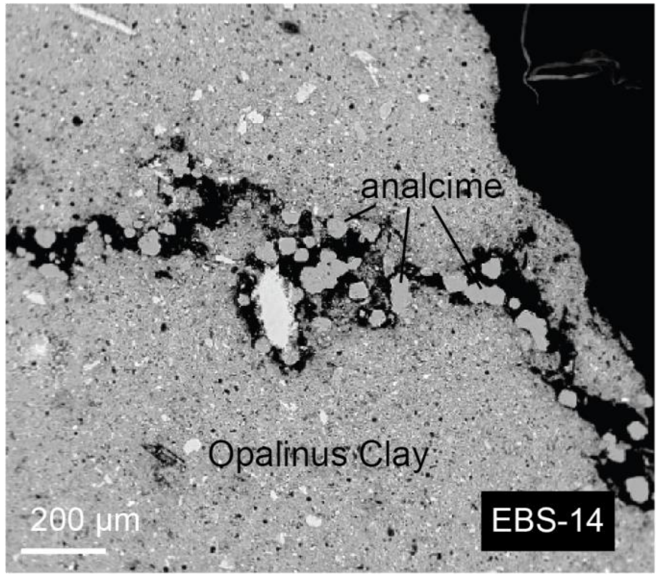
Thermodynamic modeling and database development

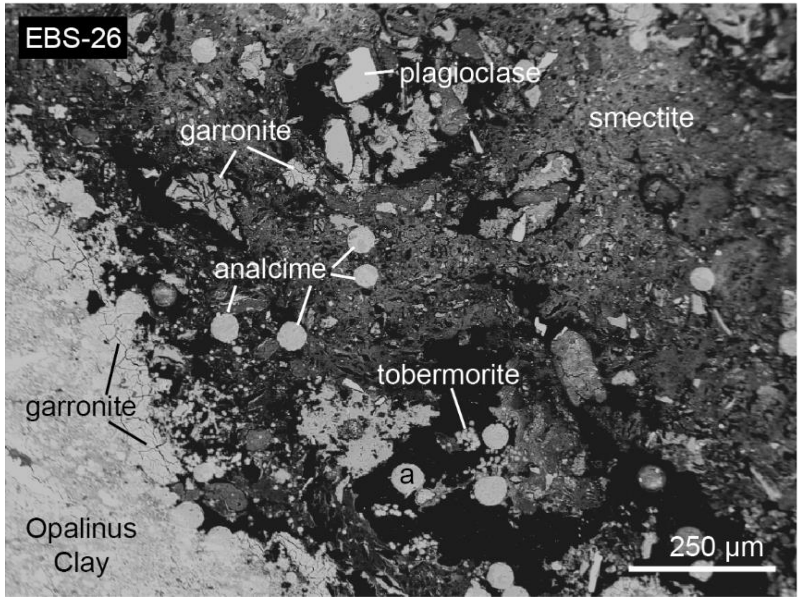
Cheshire et al. 2014, 2018

Barrier Material Interactions: Bulk Mineralogy Changes

- **Opalinus Clay ± Wyoming Bentonite**
  - Quantitative X-ray diffraction (Q-XRD) of experimental run products
  - 300°C (6 months): Zeolite formation in clay and along cracks and edges on the Opalinus Clay fragments, plagioclase
  - 200°C (8 weeks): No zeolites or feldspar
  - Both: wt.% clay increases
- **Opalinus Clay + Wyoming Bentonite + Portland Cement**
  - Formation of calcium-silicate-hydrate (CSH) minerals, zeolites, plagioclase feldspar at 200°C
  - Clay degradation
  - Reduction in clay swelling
  - Amorphous material (gel?)

Sauer et al. (2019)

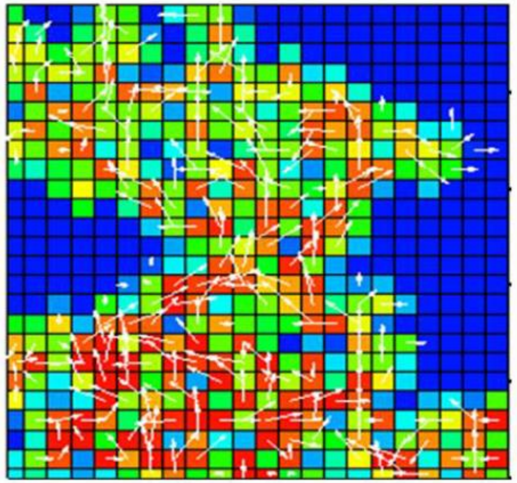




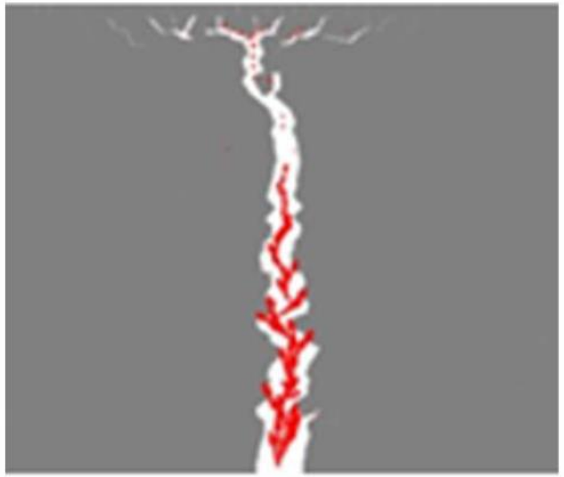
Thermo-Hydrological-Mechanical (THM) Processes in Clay Underground Research Laboratory Experiments and Simulation

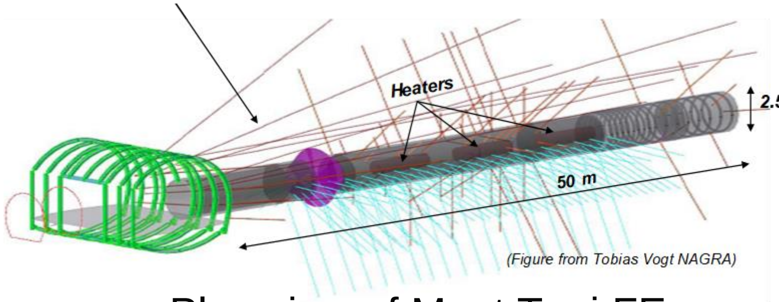
LBNL for modeling gas migration through clay associated with DECOVALEX-2019

Continuum model approach using TOUGH-FLAC

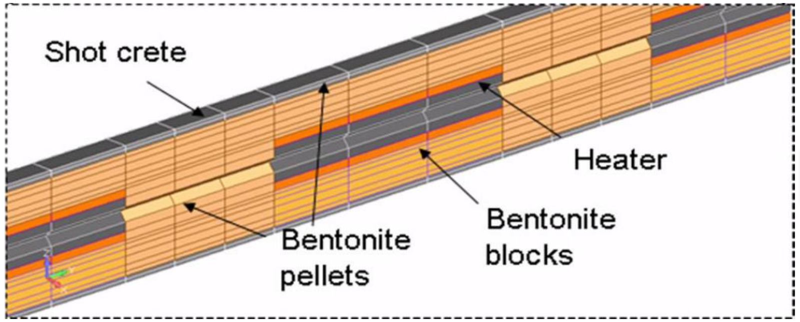


Discrete fracture model approach using TOUGH-RBSN



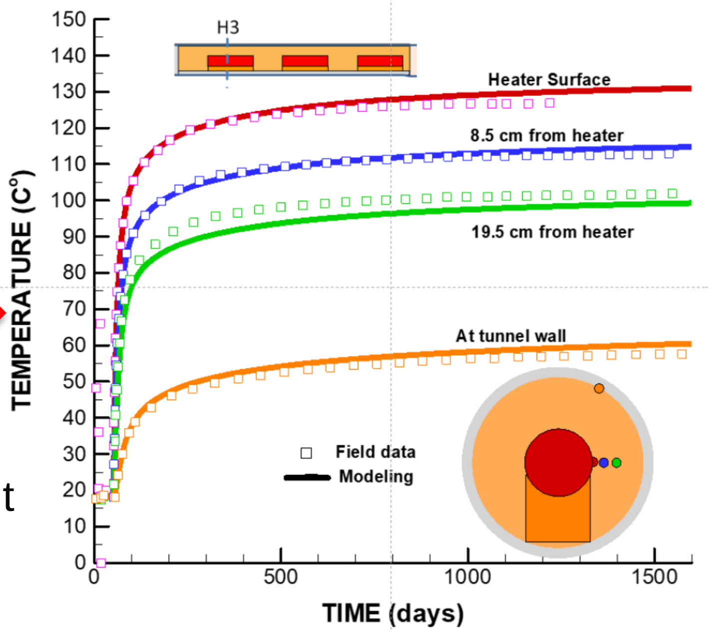


Plan view of Mont Terri FE experiment – Opalinus Clay

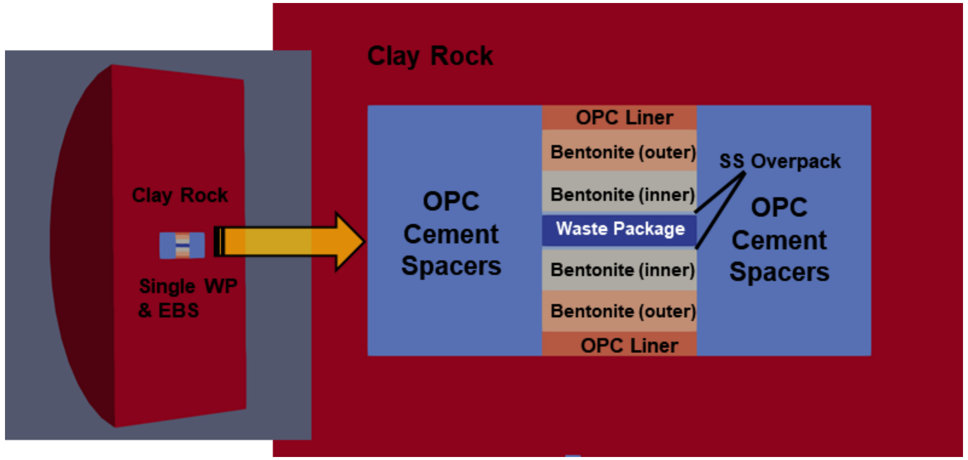


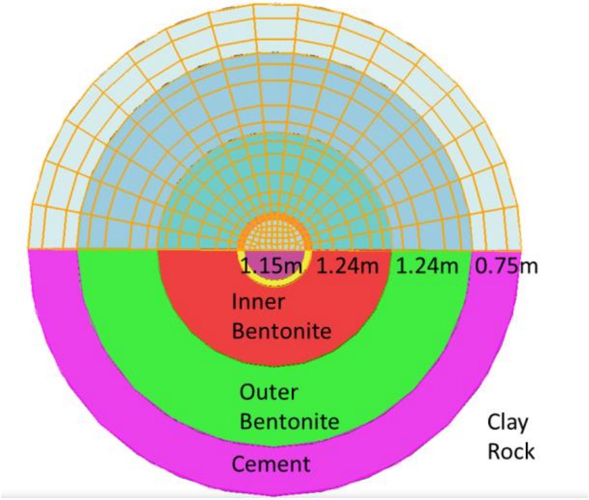
TOUGH-FLAC 3D numerical grid of the FE experiment

Rutqvist et al. (2020)

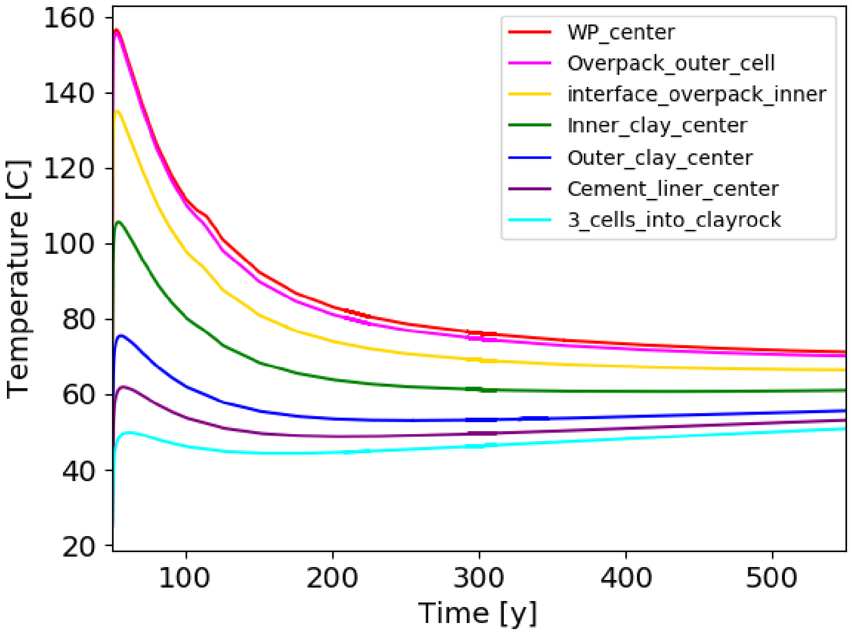


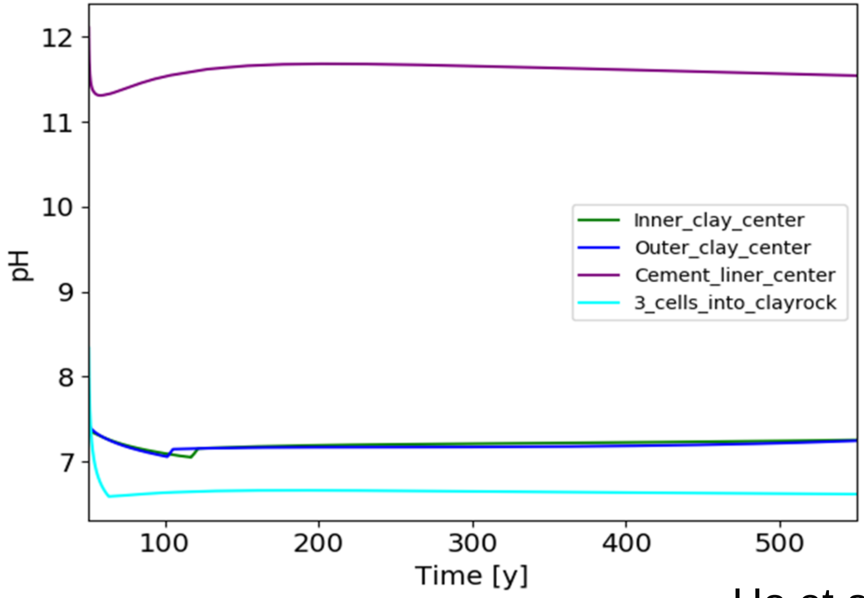
Non-isothermal 1D-3D Thermo-Hydrological-Chemical (THC) Reactive Transport Modeling





- **Waste canister length: 4.7 m**
- **12 PWR assemblies**
- **50-year storage time**





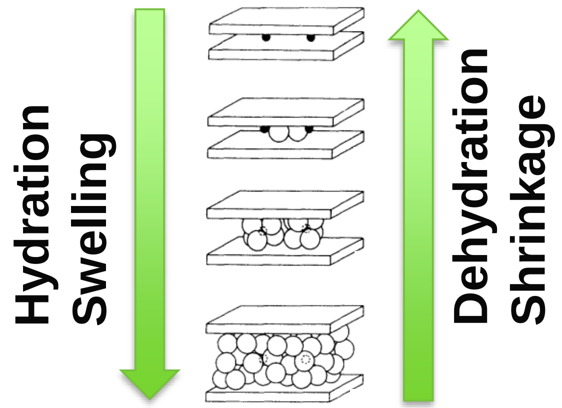
Evaluation of thermal effects on fluid/solid interactions

- Chemical reactions – mineral dissolution/precipitation
- Changes in bulk mineralogy
- Evaluate changes in porosity/permeability
- Next: Evaluate THC scenarios with higher thermal loads (24-PWR and 37-PWR).
- Next: Evaluate mesh resolution effects

Ho et al. (2019)

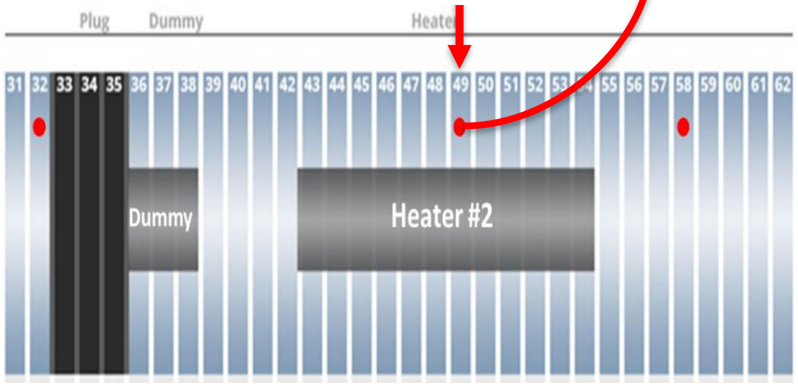
FEBEX-DP - Thermal Analysis: Thermogravimetric Analysis (TGA) & Differential Scanning Calorimetry (DSC) - Controlled Relative Humidity (RH) & Temperature

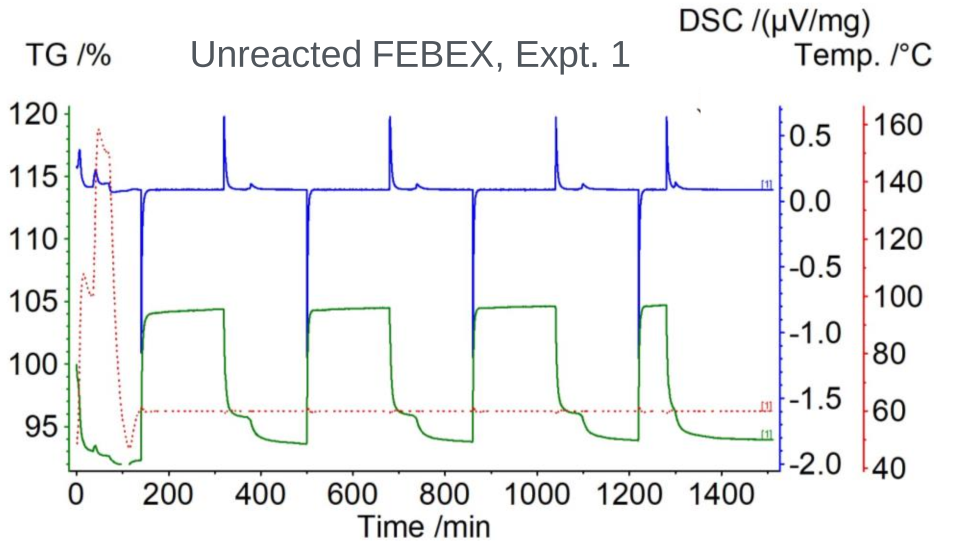
Montmorillonite

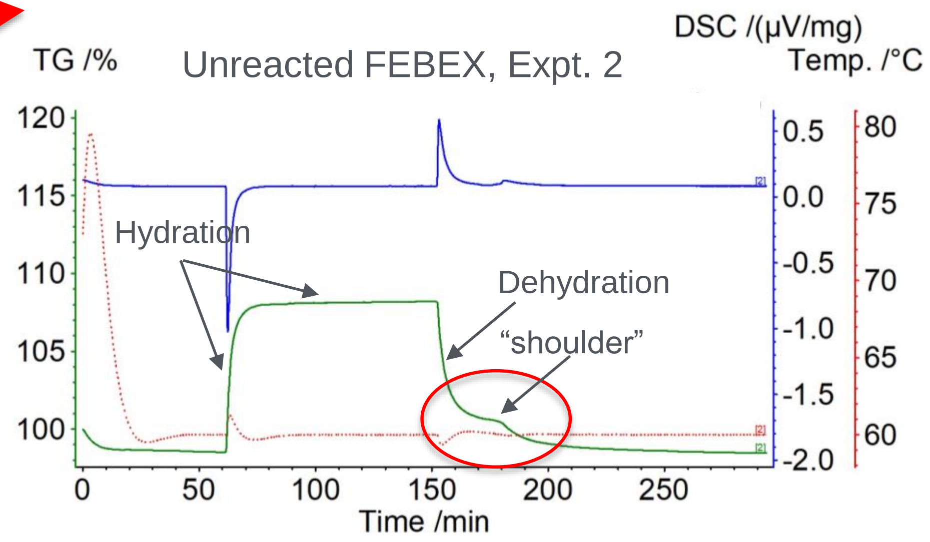


Modified after Madsen and Müller-Vonmoos (1989)

- **FEBEX Heater 2:**
  - **18+ years of heating**
- **Peak temperature: 100°C**
- **Sampling Section 49**





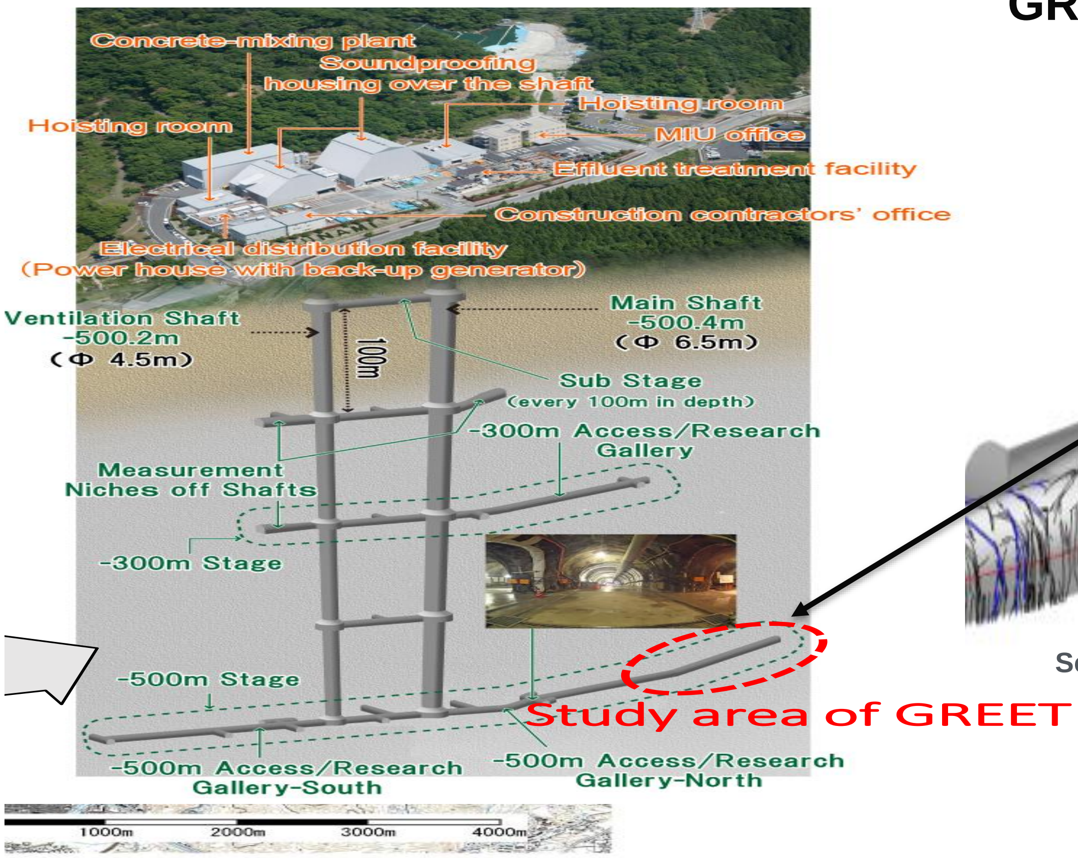


Jove Colon et al. (2019)

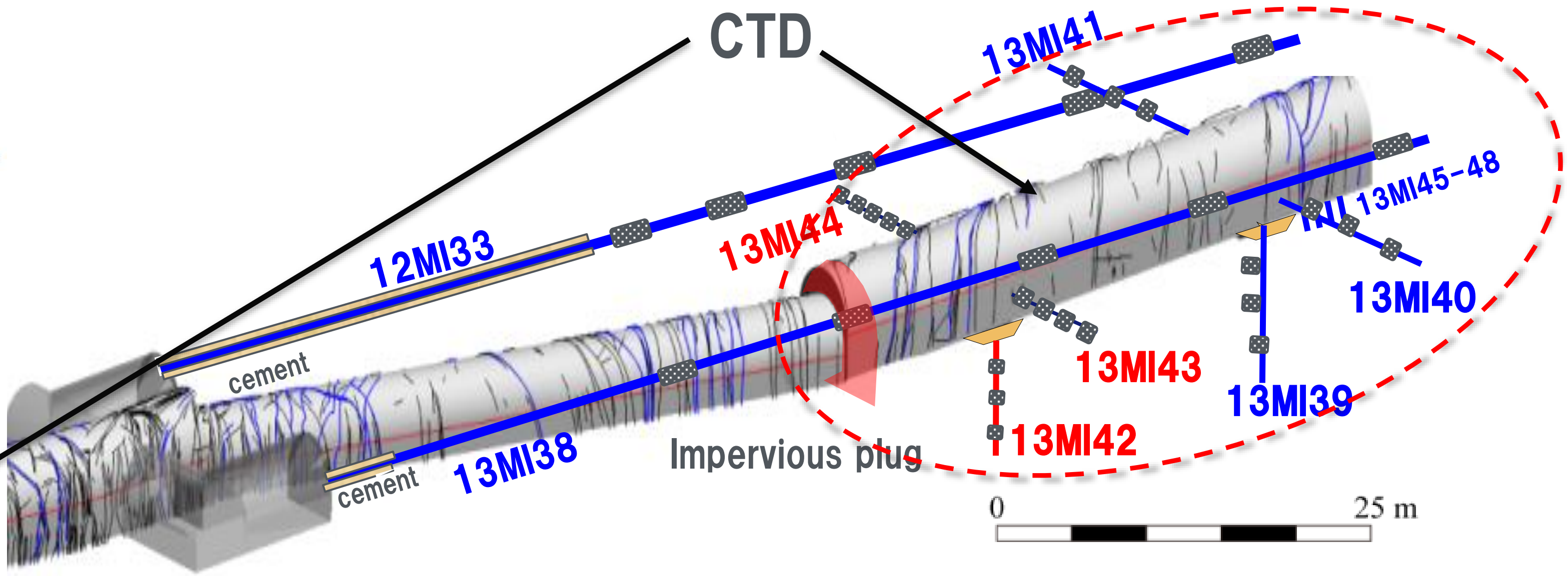
Bentonite Thermal Behavior

- Bentonite dehydration behavior is a function of the duration of hydration that precedes it.
- Appearance of a "shoulder peak" during dehydration indicating different energetics for swelling clay hydration and dehydration.
- Cyclical thermal analysis methodology will be used in the study of additives to bentonite.
- **NEXT**: Conduct thermal studies at temperatures above 100°C

DECOVALEX19: GREET Experiment at Mizunami Underground Research Laboratory Site (Japan) – Closure Test Drift (CTD) Geochemistry

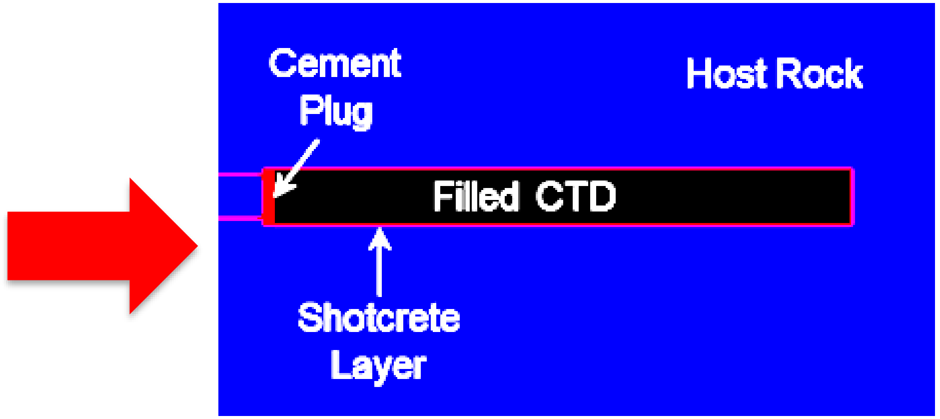


GREET (Groundwater REcovery Experiment in Tunnel)

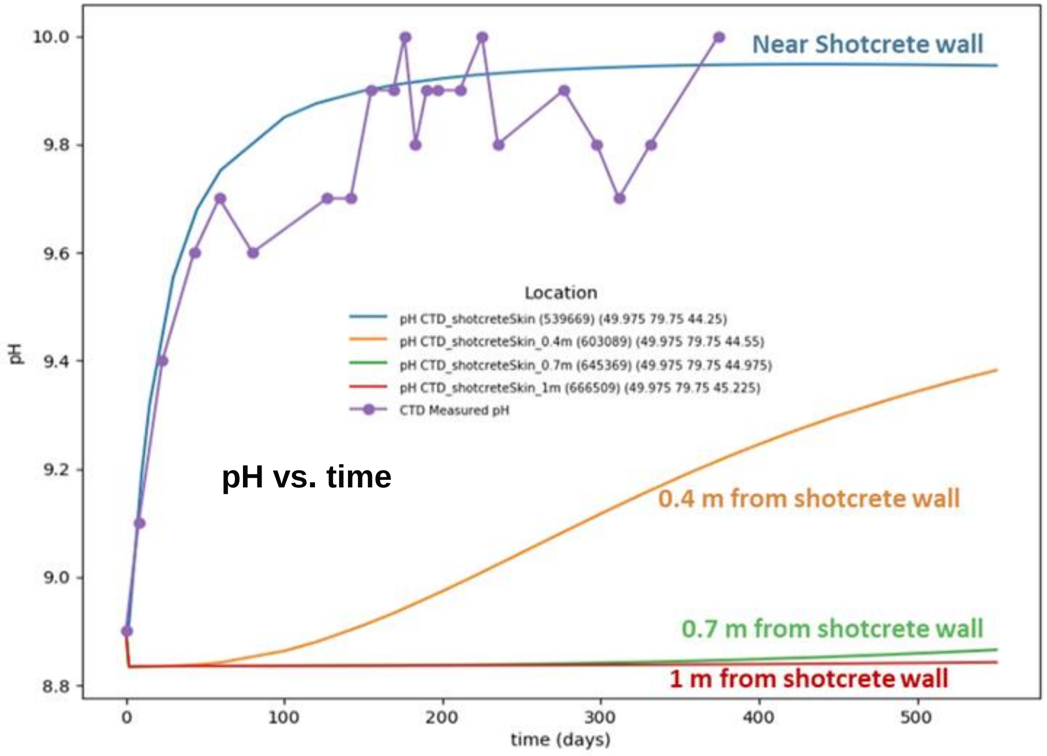


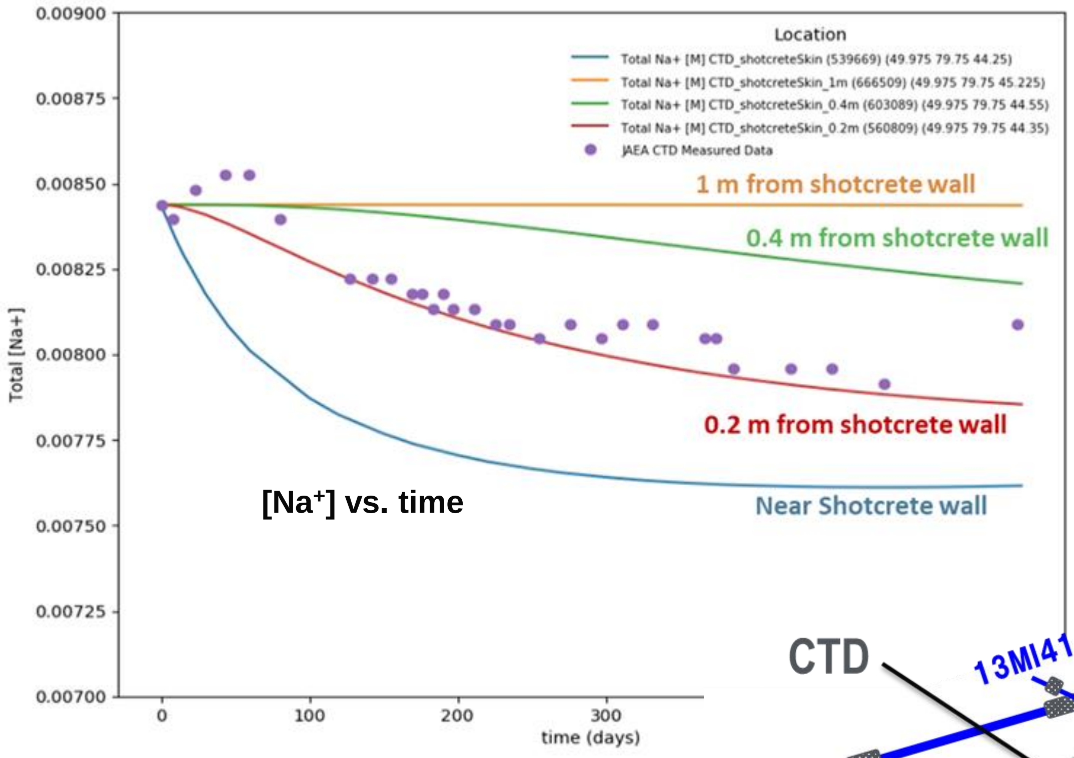
Schematic figure courtesy of Dr. Teruki Iwatsuki (JAEA)

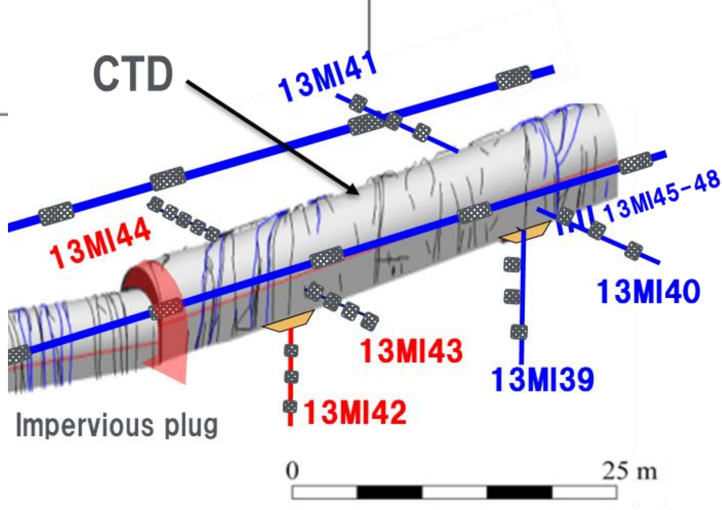
- 3D Reactive Transport Simulations using PFLOTRAN simulation code
- Focus: Shotcrete – groundwater interactions in the CTD



DECOVALEX19: PFLOTRAN 3D Reactive Transport (RT) Model of GREET URL Experiment (Mizunami Site, Japan)







- Model representation agrees with overall trend chemical trends
- Sensitivity analyses (SA) on kinetic rate law parameters for various cement phases and volume fraction of mineral components
- Simulations have been conducted to evaluate the effect of shotcrete thickness effects

Jové Colón et al. (2020)

Summary

- Development of a high temperature argillite reference case
  - Need to further disposal concepts for DPC's, EBS design options (e.g., thermal management), and post-closure strategies
- Bentonite-metal-cement-Opalinus Clay interactions:
  - Reactions produces zeolites and with some swelling reduction in smectite as a result of interactions with alkaline solutions
  - Future Work: Study effects of host rock composition & other barrier materials (e.g. cement); expand 3D non-isothermal model to various waste packages
- DECOVALEX Hydrological-Cchemical (HC) (GREET) modeling and Thermal Analyses on FEBEX-DP Bentonite:
  - 3D reactive transport model of shotcrete interactions in CTD experiment represent overall chemical trends
  - Cyclic thermal analysis (hydration/dehydration) experiments show reproducible results between cycles with slower dehydration rates
  - Future Work: Investigate HC model sensitivities to shotcrete thickness; expand cyclic thermal analyses & XRD methods to evaluate high temperature effects; maintain engagement with international programs (DECOVALEX2023; HotBent, EBS Task Force)

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