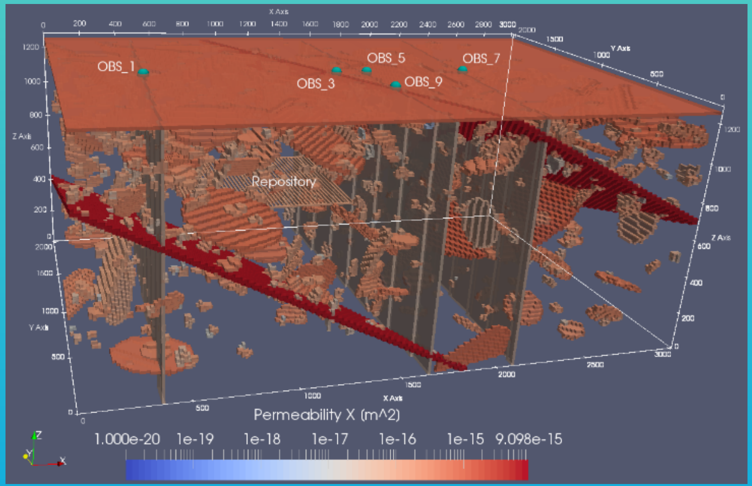
U.S. DEPARTMENT OF ENERGY | Office of NUCLEAR ENERGY

Spent Fuel and Waste Science and Technology (SFWST)

Clean. Reliable. Nuclear.



Carlos F. Jové Colón (SNL)

NWTRB Fall 2020 Fact-Finding Meeting

SAND2020-13315 PE

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- C. Payne (SNL), J. Kruichak (SNL), M. Mills (SNL), A. Knight (SNL), H. Moffat (SNL), E. Coker (SNL), T. Ho (SNL), Y. Wang (SNL), E. Stein (SNL), S. David Sevougian (SNL), F. Perry (SNL), C. M. Lopez (SNL), F. Caporuscio (LANL), K. Sauer (LANL), M. Cheshire (LANL), J. Rutqvist (LBNL), L. Zheng (LBNL)

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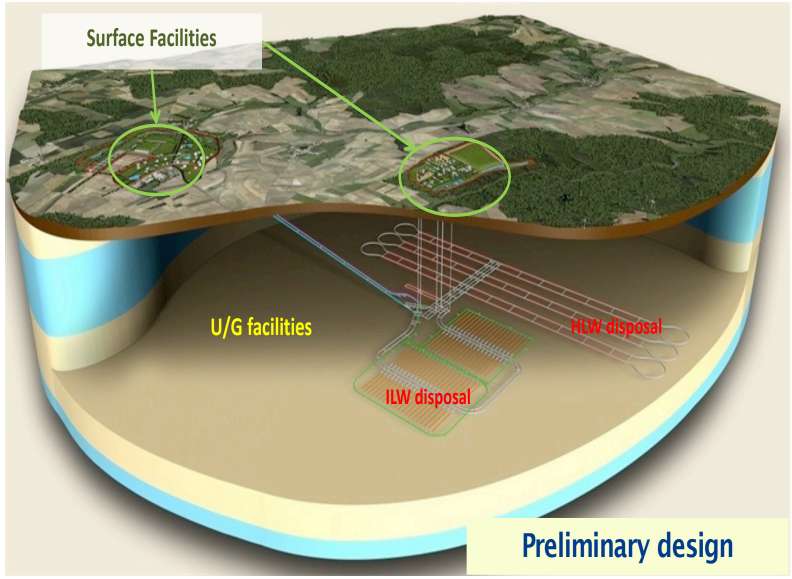
BERKELEY LAB

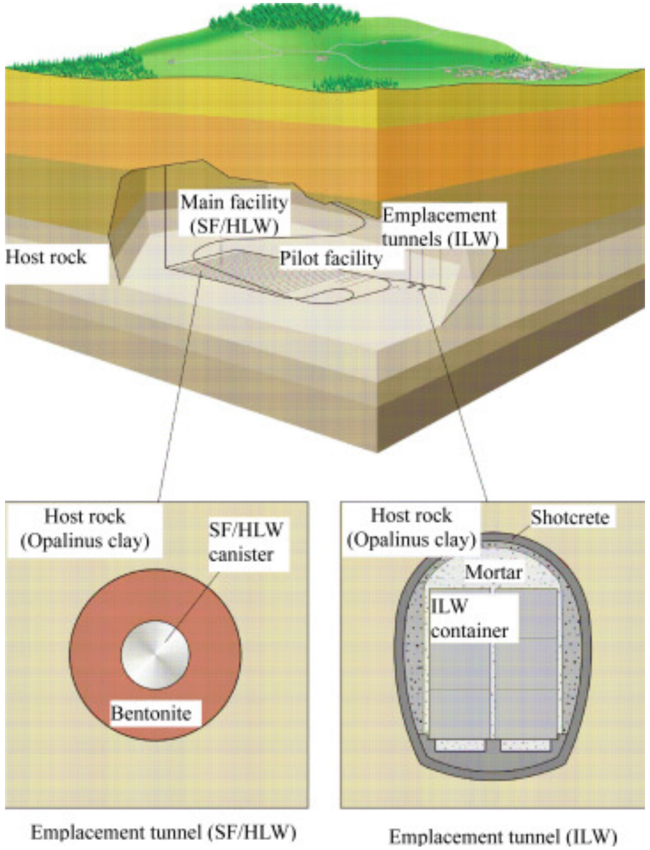
Outline

- Argillite Repository Concept
- Knowledge Gaps & R&D Priorities
- Repository Relevant Processes
- Argillite Reference Case
- Highlights – Disposal in Argillite R&D
- Summary

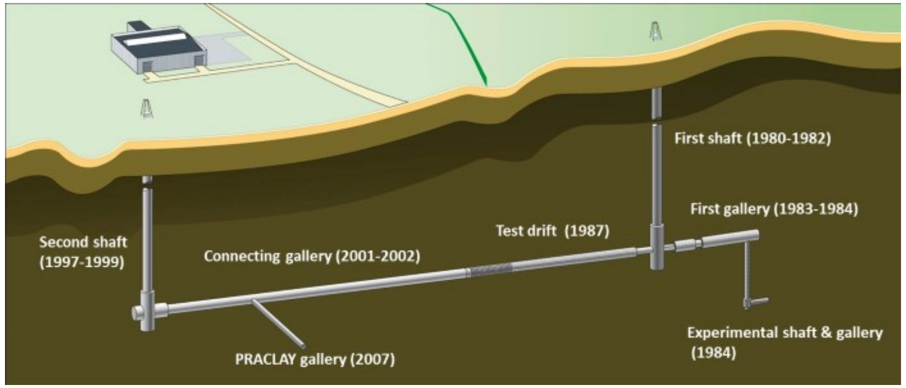
Argillite/Shale Repository Concept



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



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

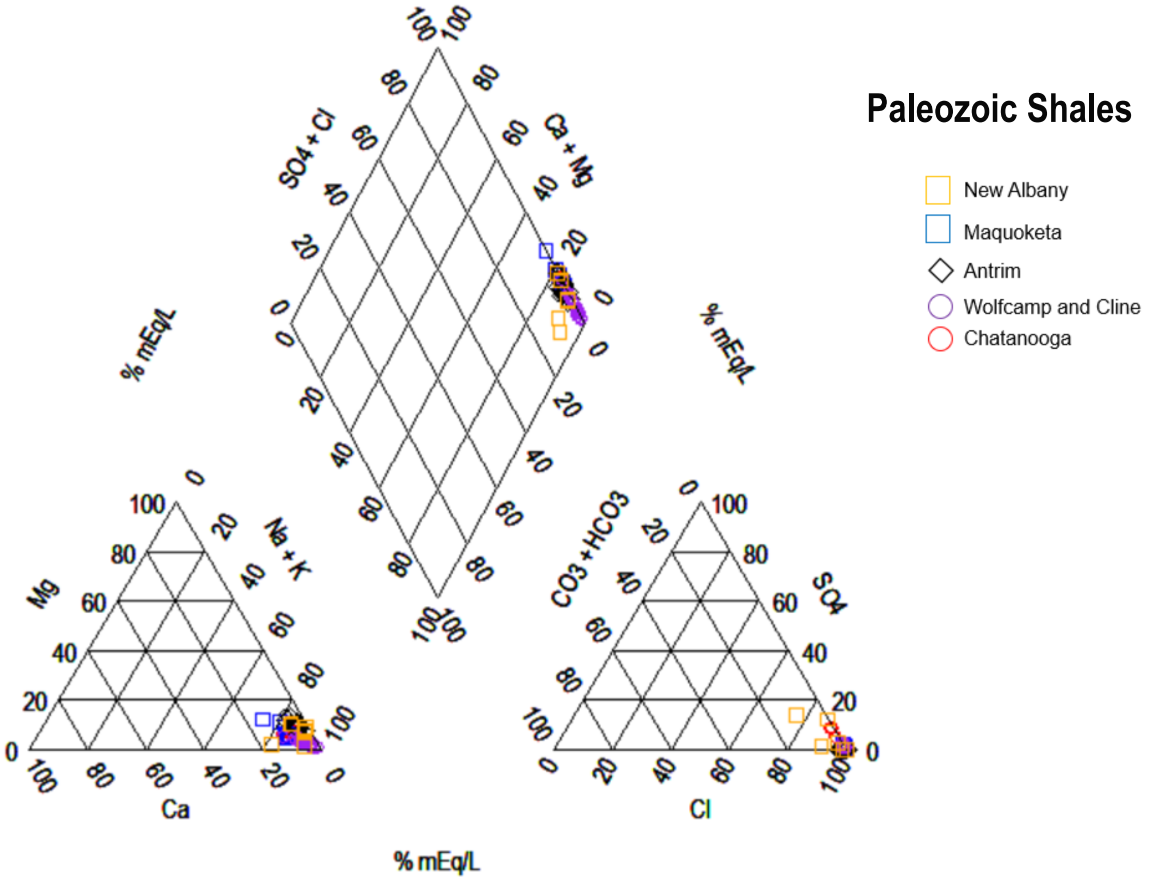


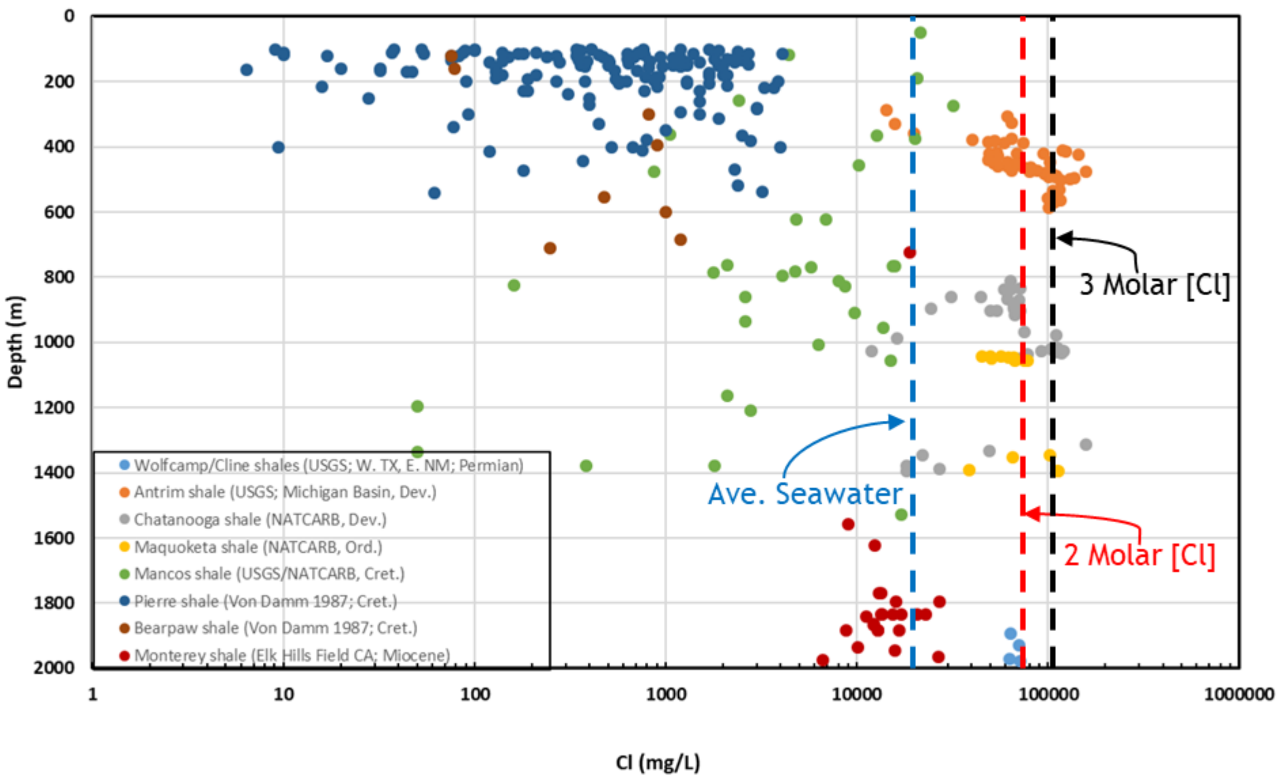
Belgian repository concept – HADES Underground Laboratory – Boom Clay (https://science.sckcen.be/en/Facilities/HADES)

Shale Attributes

- **Low permeability / hydraulic conductivity**
- **Low diffusion coefficients**
- **Good retention capacity for radionuclides**

Porewater Chemistry in Clay Formations



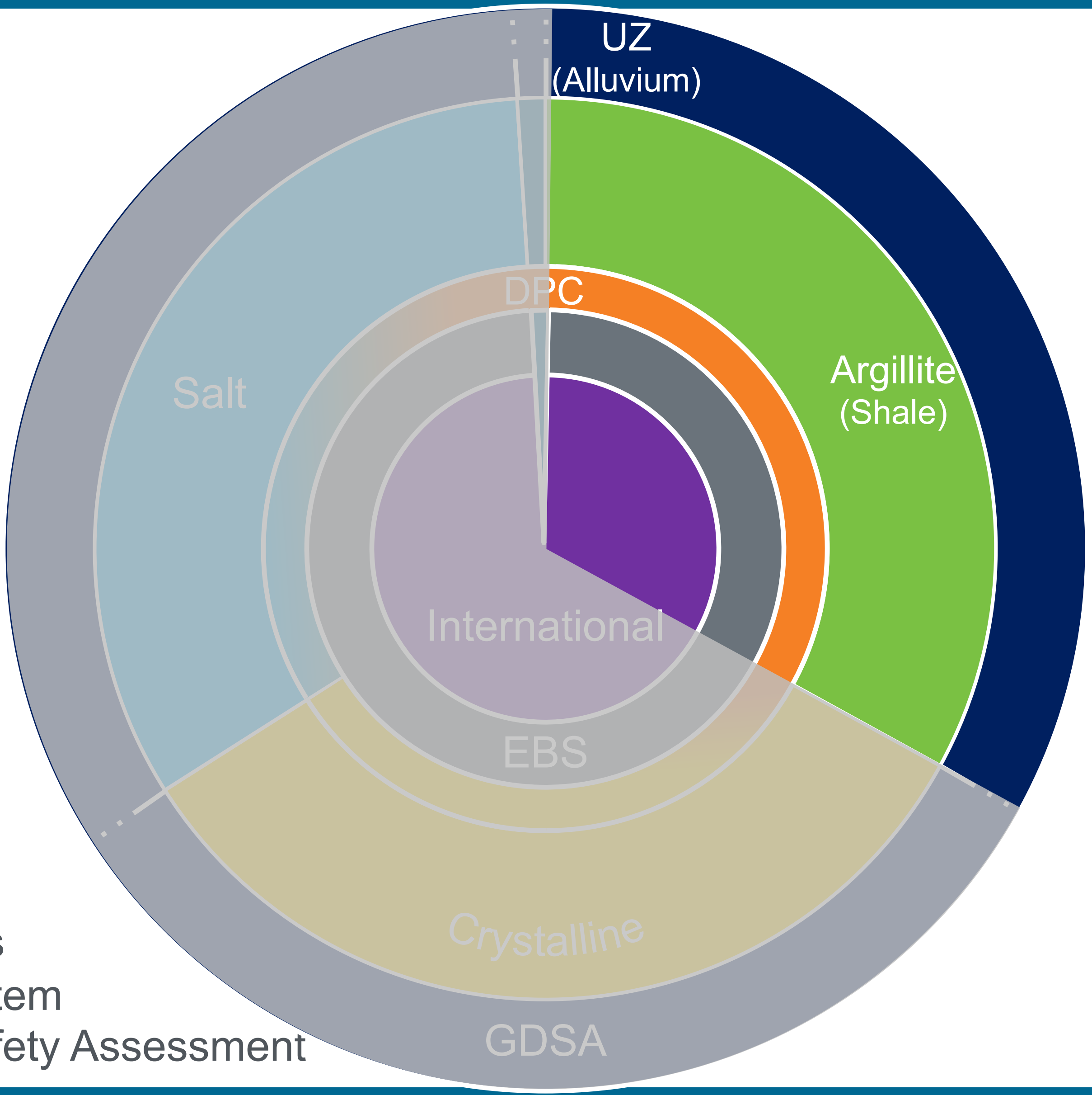


- Porewater compositions are highly variable
- Overall: Na-Cl brines with some Ca & carbonate

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

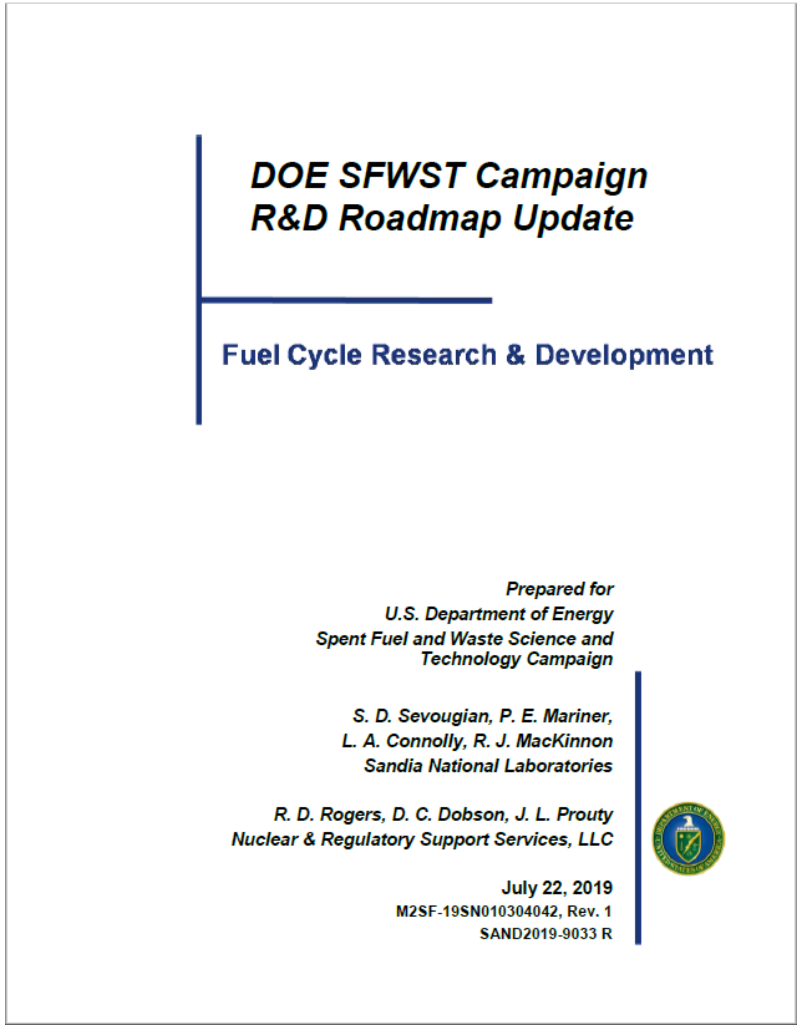
Stein et al. (2020)

R&D Priorities – Argillite (Shale)



UZ = Unsaturated Zone
DPC = Dual Purpose Canisters
EBS = Engineered Barrier System
GDSA = Geologic Disposal Safety Assessment

Knowledge Gaps & R&D Priorities



DOE SFWST Campaign R&D Roadmap Update

Fuel Cycle Research & Development

Prepared for
U.S. Department of Energy
Spent Fuel and Waste Science and Technology Campaign

S. D. Sevougian, P. E. Mariner,
L. A. Connolly, R. J. MacKinnon
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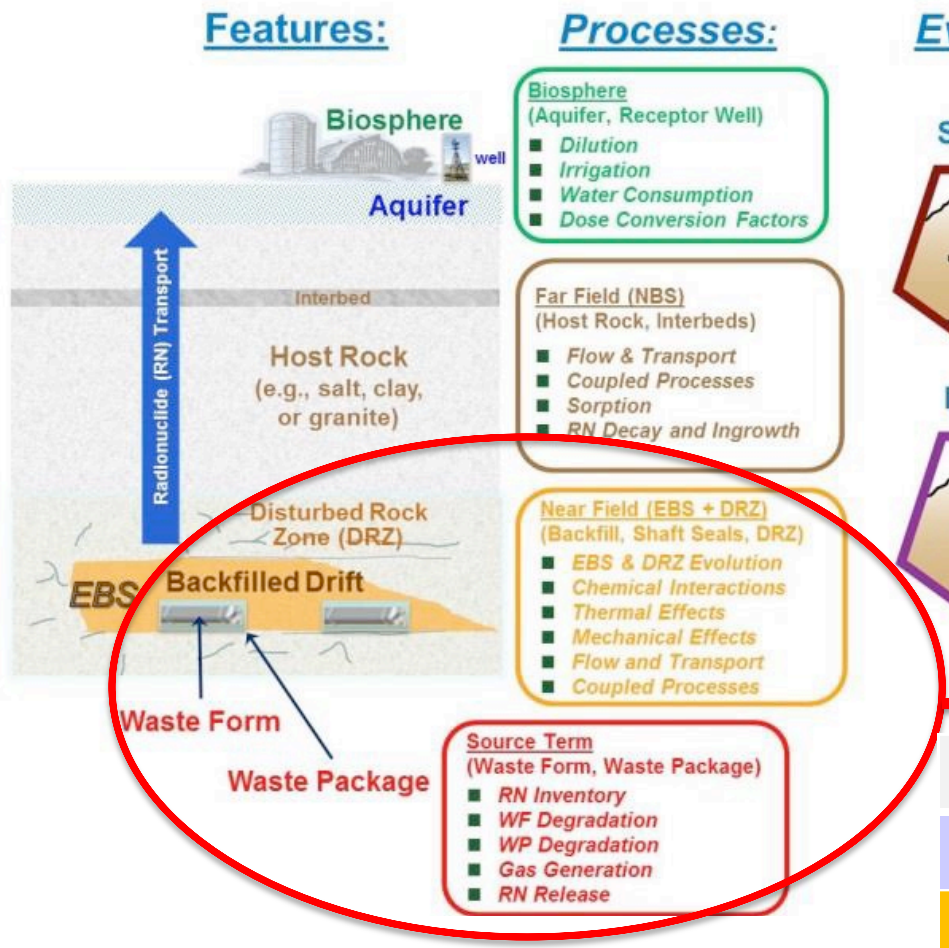
Nuclear & Regulatory Support Services, LLC

July 22, 2019
M2SF-19SN010304042, Rev. 1
SAND2019-9033 R

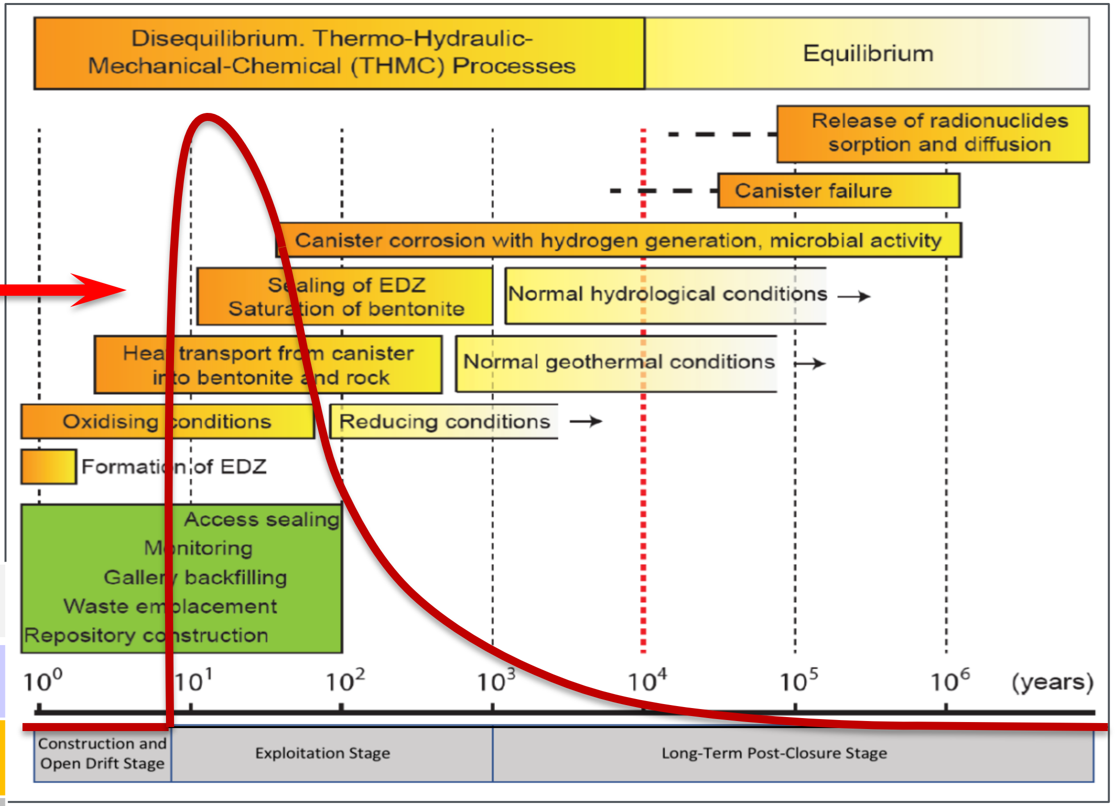
Some Activities With Medium-High Score in 2019 R&D Roadmap Update

- **High-Temperature Behavior** - Chemical processes still under development, particularly at elevated temperature conditions
- **EBS High Temperature experimental data collection** - To evaluate high temperature mineralogical/geochemical changes
- **Analysis of clay hydration/dehydration and alteration** under various environmental conditions
- **Buffer/backfill dry-out and resaturation process**
- **THC processes in EBS** - High importance for design/construction arguments affecting disposal system design that utilize backfill/buffer as an engineered barrier
- **Argillite Coupled THM processes modeling** including host rock, EBS, and EDZ
- **Cement plug/liner degradation; Evaluation of ordinary Portland cement (OPC)**

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



BATS	BRIE	CFM
FEBEX & HotBENT	Gas Migration	LTDE



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

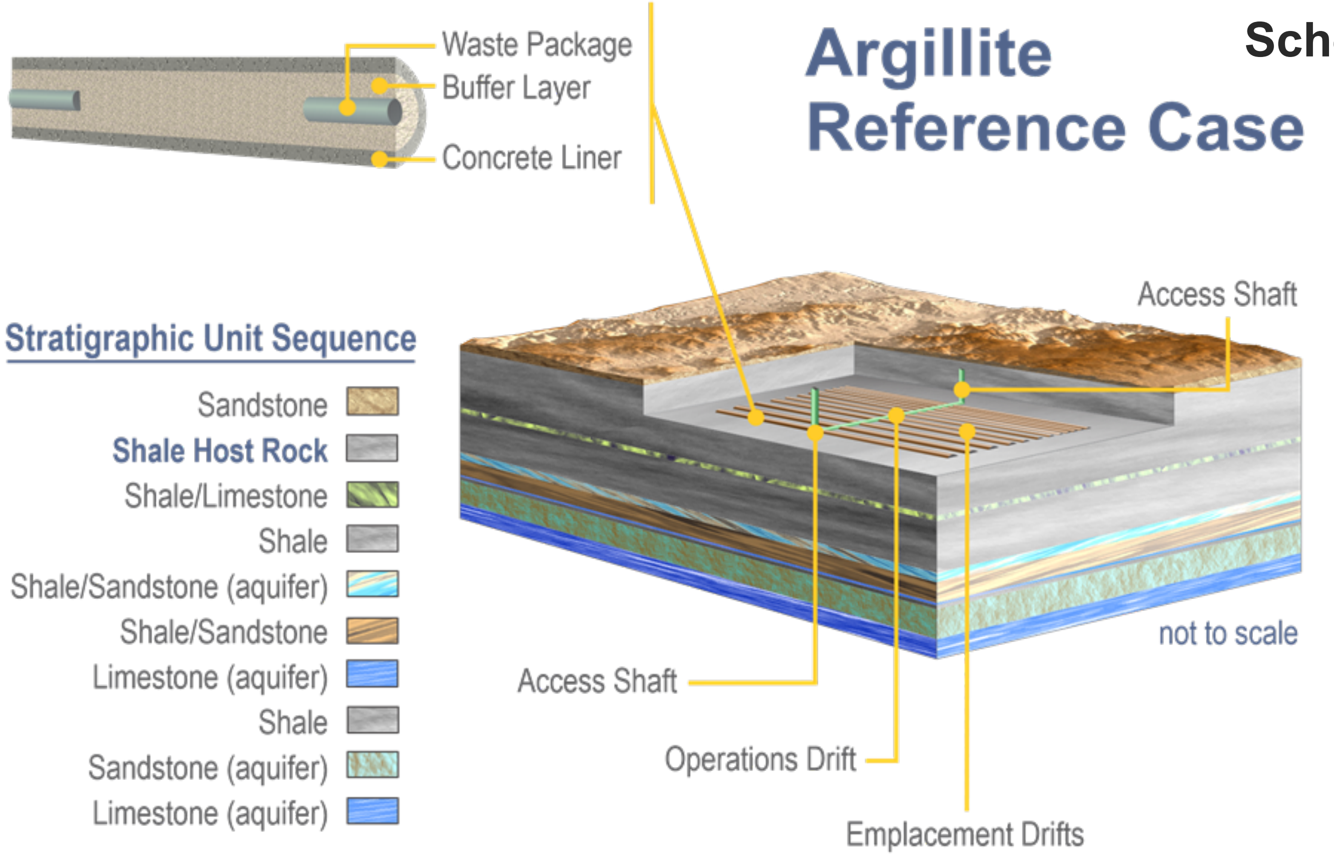
TED, ALC, HE-E

FE

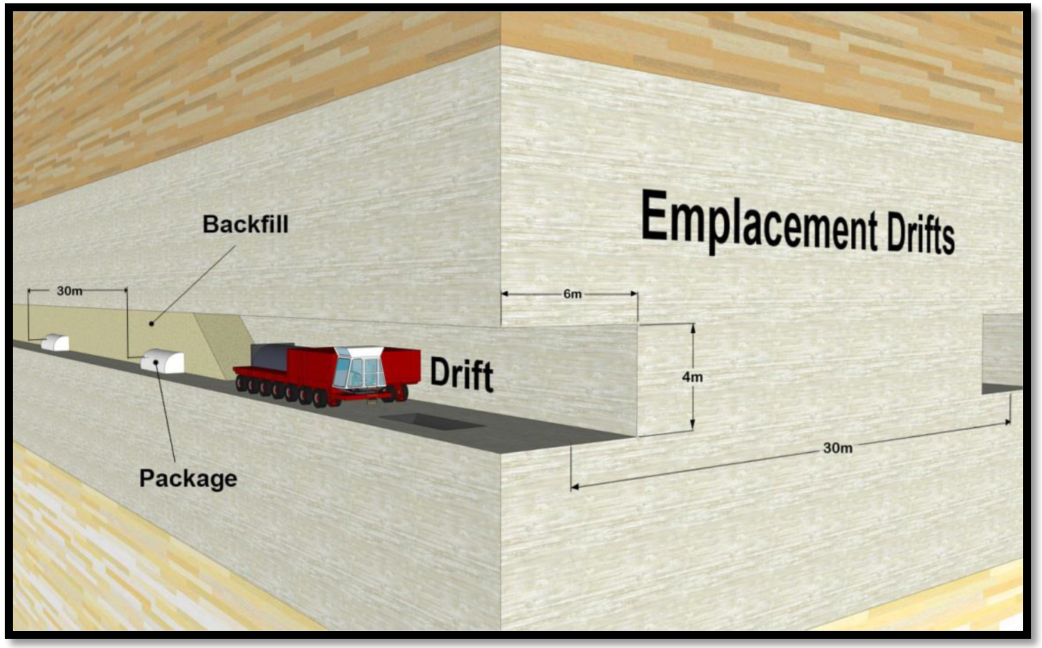
ALC = Full-scale Emplacement Experiment (France)
BATS = Heated Brine Availability Test in Salt (USA)
BRIE = Bentonite Rock Interaction Experiment (Sweden)
FEBEX = Full-Scale Engineered Barrier Experiment (Switzerland)
CFM = Colloid Formation Migration (Switzerland)

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)

Argillite Reference Case



Schematic Illustration of Package/Backfill Emplacement

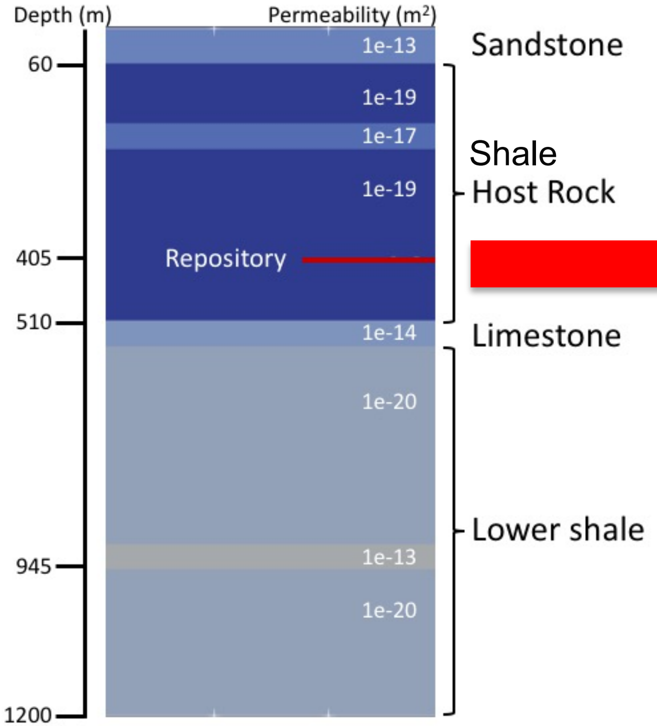


Stein et al. (2017)

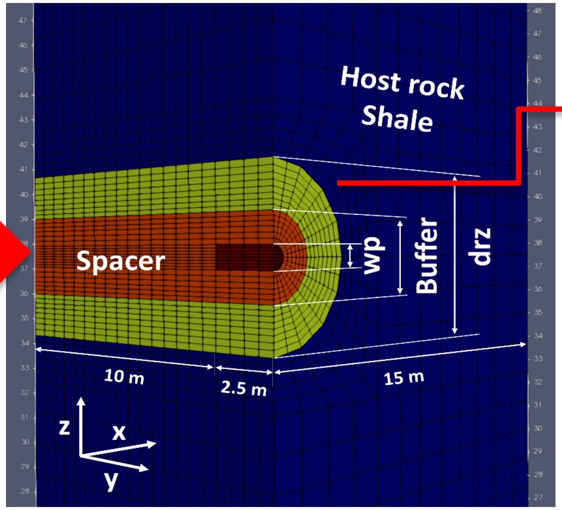
Evaluation of disposal design concepts

- Thermal management in clay/shale repository
  - Waste package and drift spacing
  - Coupled Multiphase transport phenomena

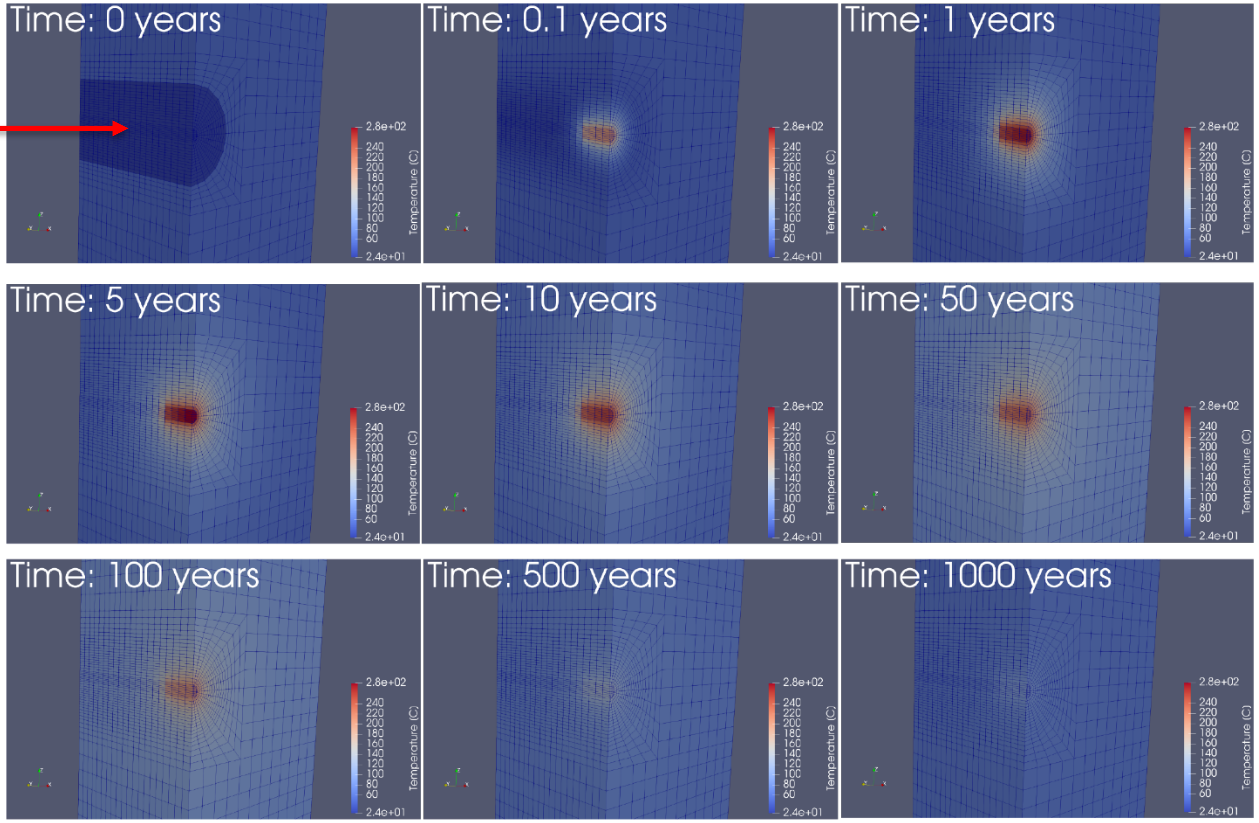
Argillite Reference Case: Deterministic Simulations of Generic Disposal in Shale



Generic stratigraphic column for shale reference case



Near-field model domain used in the simulation



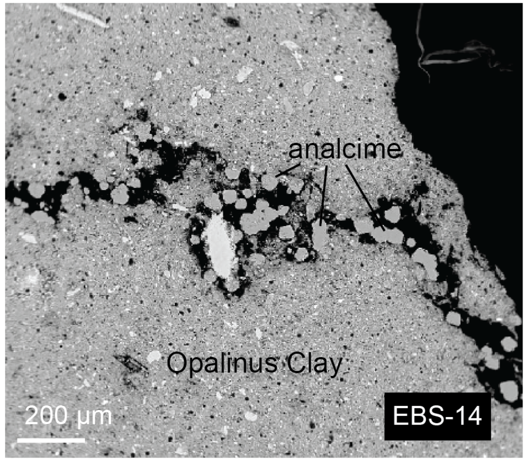
Simulation temperature for 24-PWR heat source

24-PWR = 24 SNF Pressurized Water Reactor Assemblies
drz = disturbed rock zone

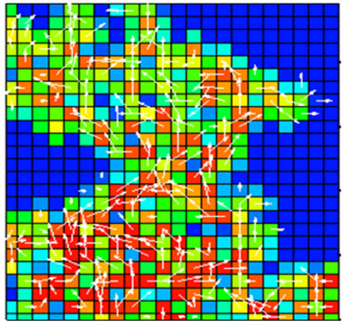
Sevougian et al. (2019)

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 thermal-hydrological-mechanical-chemical (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 thermal-hydrological-chemical (THC) reactive transport modeling**
- **International collaborations**:
  - DECOVALEX19: PFLOTRAN hydrological-chemical (HC) modeling of barrier interactions
  - DECOVALEX2023: Gas transport in clays (just started!)

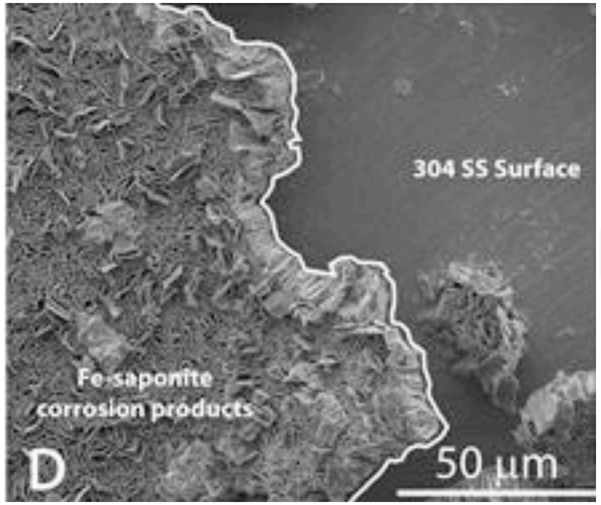


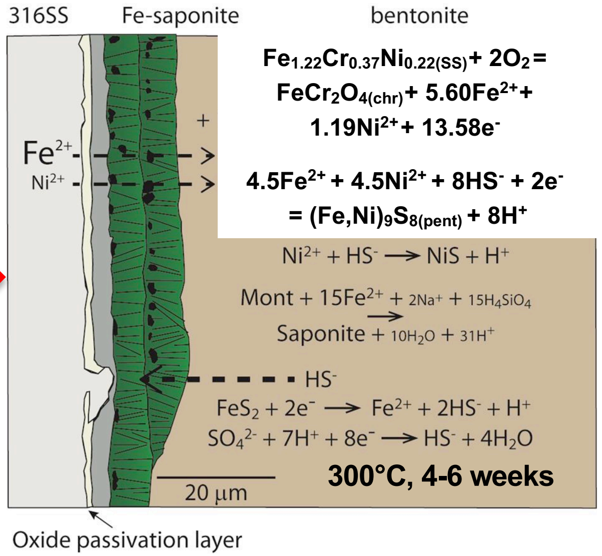
1) Continuum model approach using TOUGH-FLAC

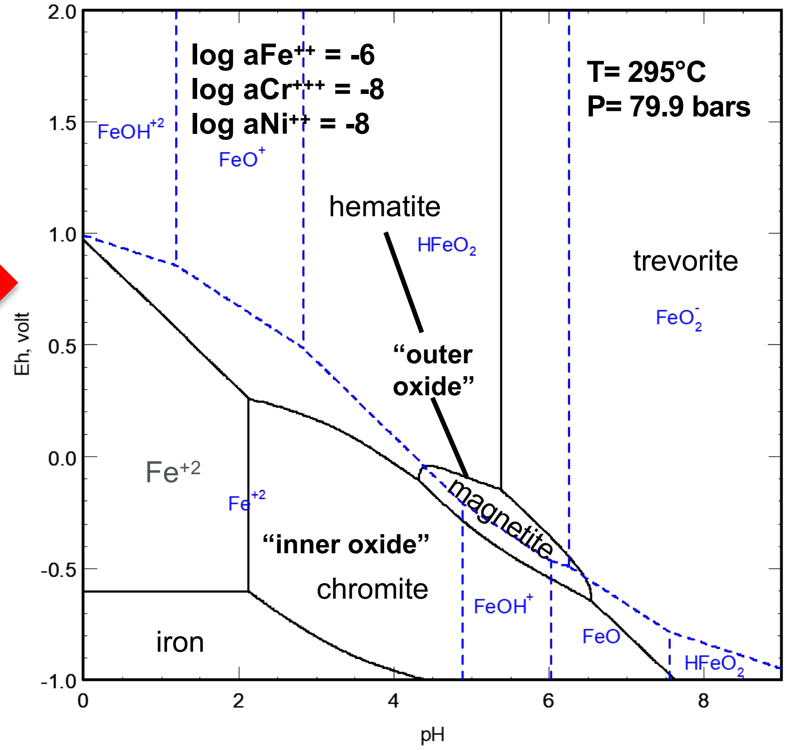


2) Discrete fracture model approach using TOUGH-RBSN

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)**

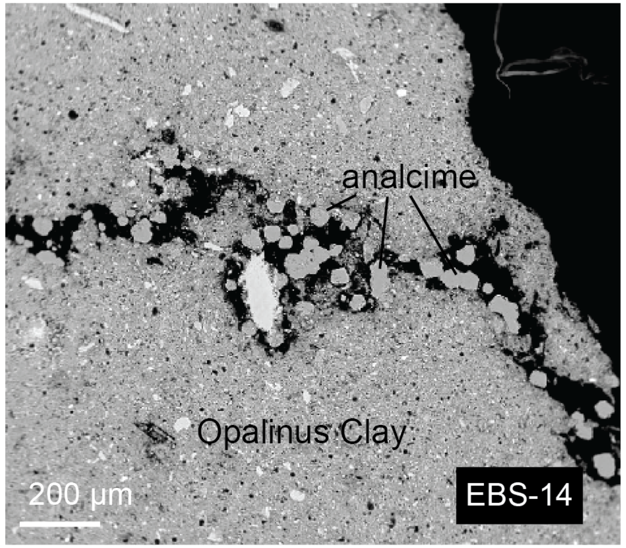
Pourbaix diagram

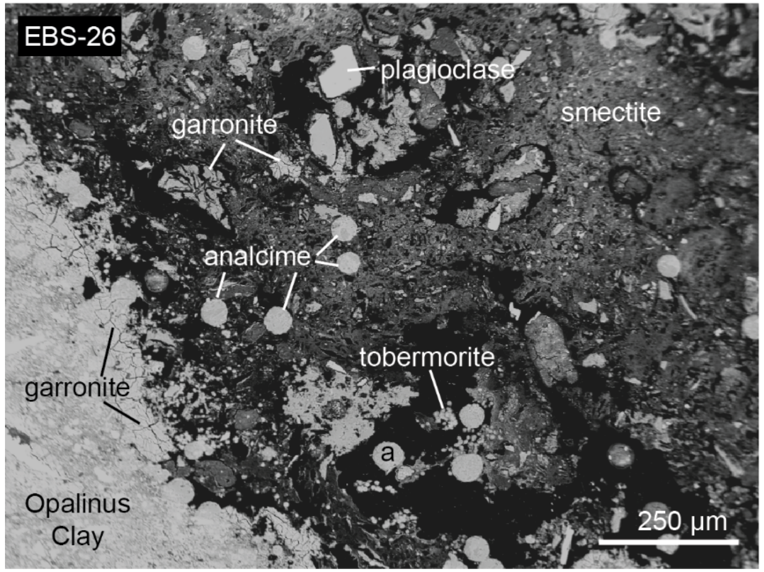
Thermodynamic modeling and database development

Cheshire et al. 2014, 2018

Barrier Material Interactions: Bulk Mineralogy Changes – Quantitative X-ray Diffraction (Q-XRD) Analysis

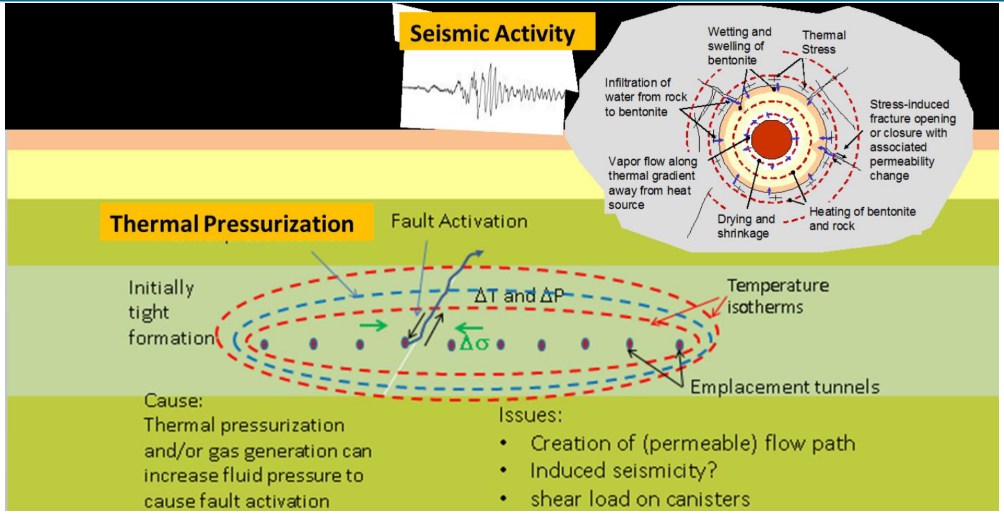
- **Opalinus Clay ± Wyoming Bentonite**
  - 300°C (6 months): Zeolite formation in clay and along cracks and edges on the Opalinus Clay fragments, plagioclase
  - 200°C (8 weeks): No zeolites or feldspar
  - Both: wt.% clay increases
- **Opalinus Clay + Wyoming Bentonite + Portland Cement**
  - Formation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



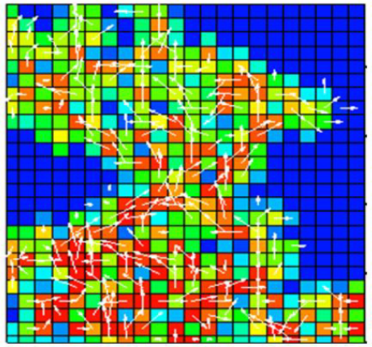


Sauer et al. (2019)

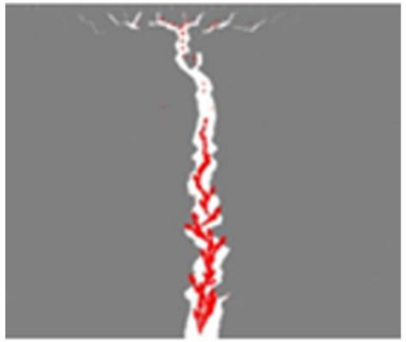
Thermo-Hydrological-Mechanical (THM) Processes in Clay URL Experiments and Simulation



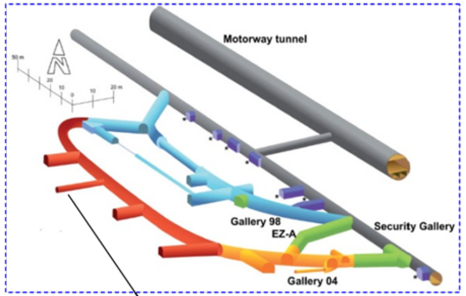
Continuum model approach using TOUGH-FLAC

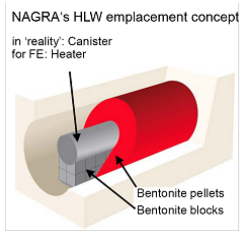


Discrete fracture model approach using TOUGH-RBSN



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

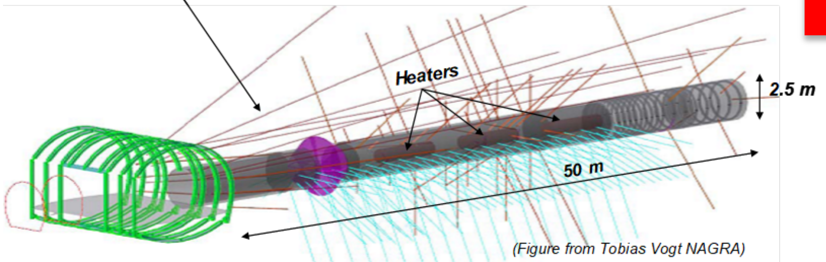




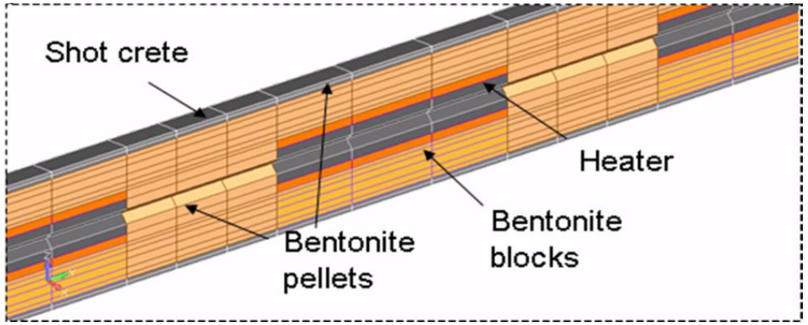
Full-scale

15-20 years of heating

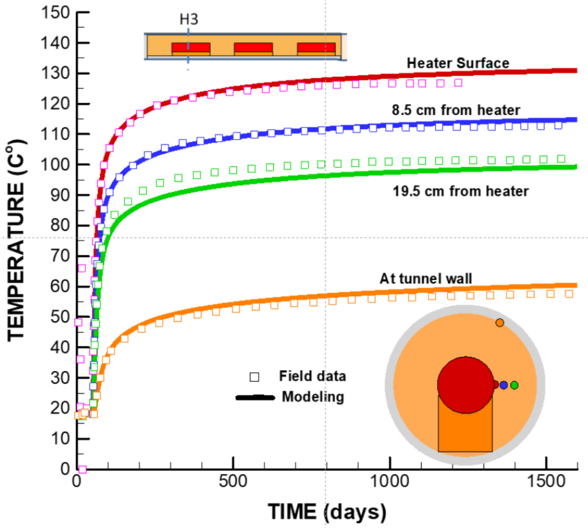
Granular bentonite emplacement



Plan view of Mont Terri FE experiment – Opalinus Clay

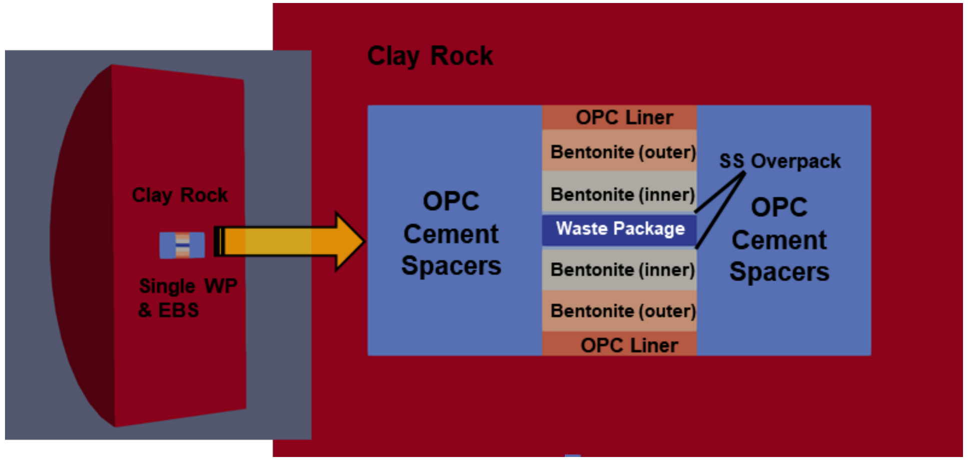


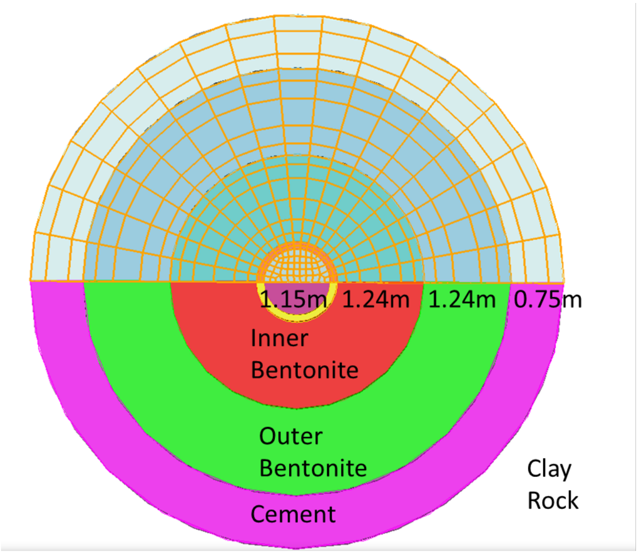
TOUGH-FLAC 3D numerical grid of the FE experiment



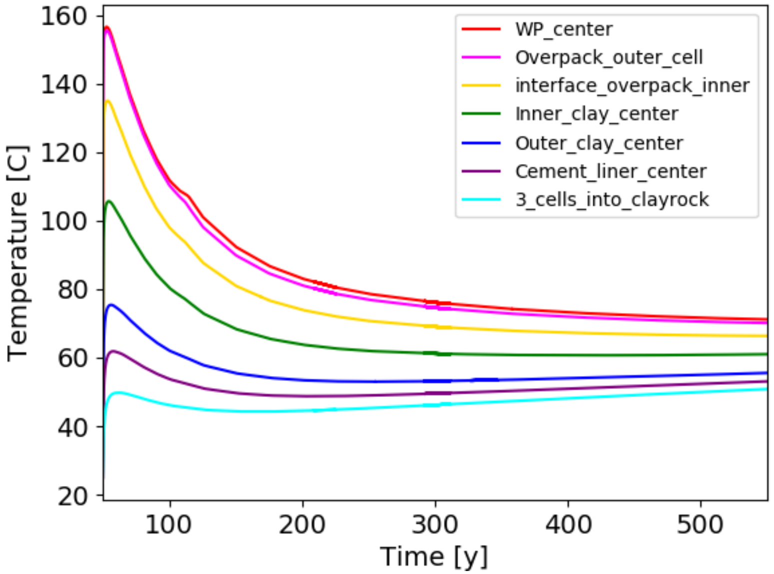
Rutqvist et al. (2020)

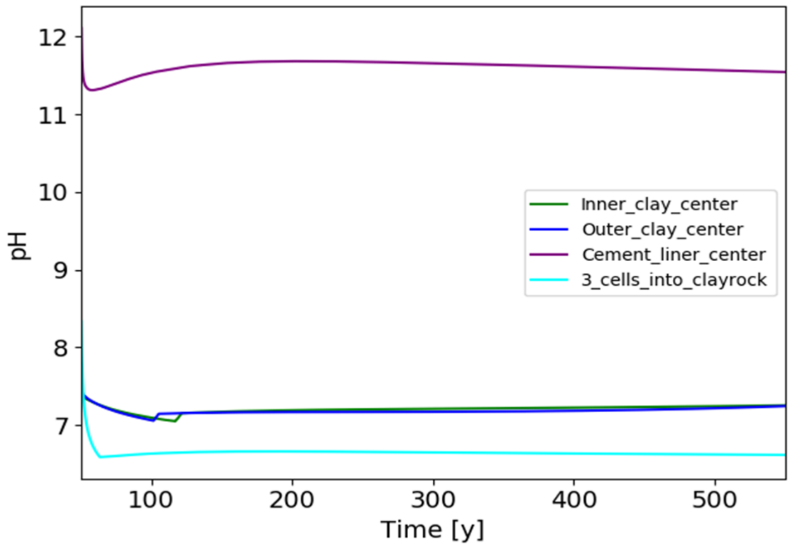
Non-isothermal 1D-3D Thermo-Hydrological-Chemical (THC) reactive transport modeling





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





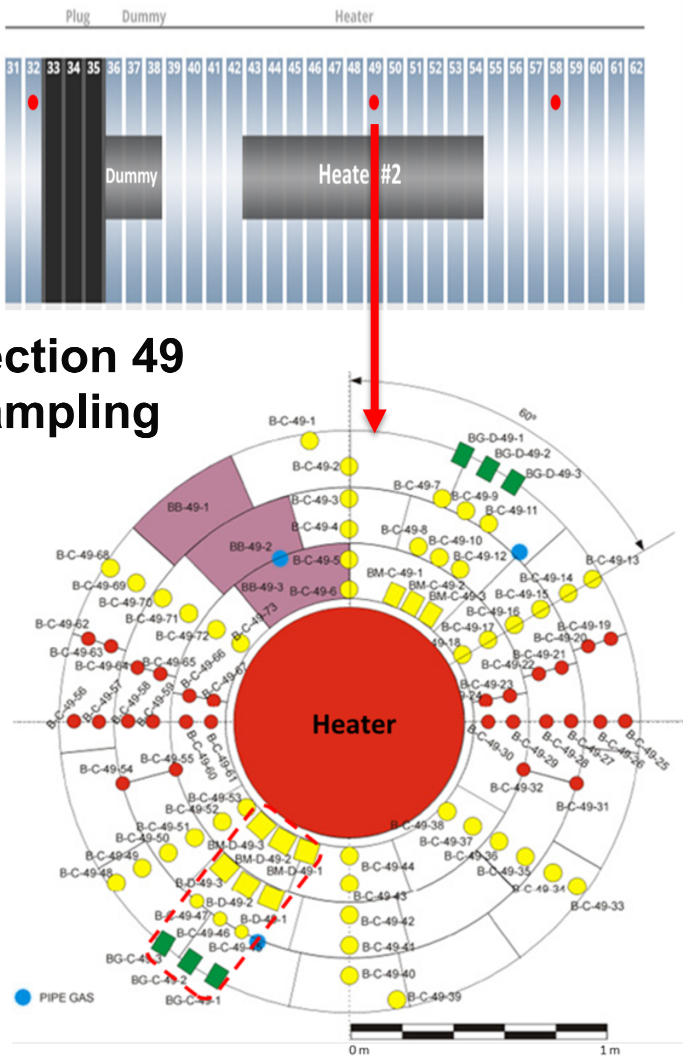
Evaluation of thermal effects on fluid/solid interactions

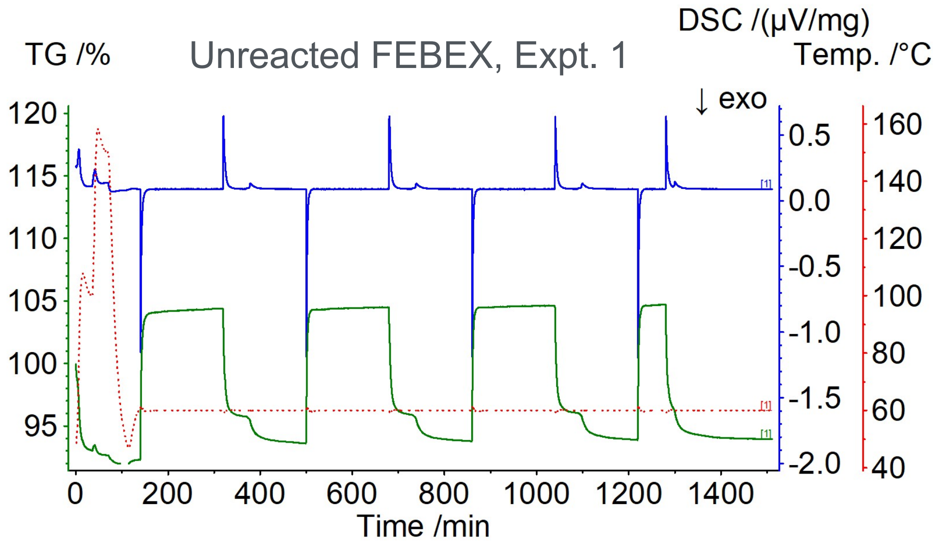
- Chemical reactions – mineral dissolution/precipitation
- Changes in bulk mineralogy
- Evaluate changes in porosity/permeability

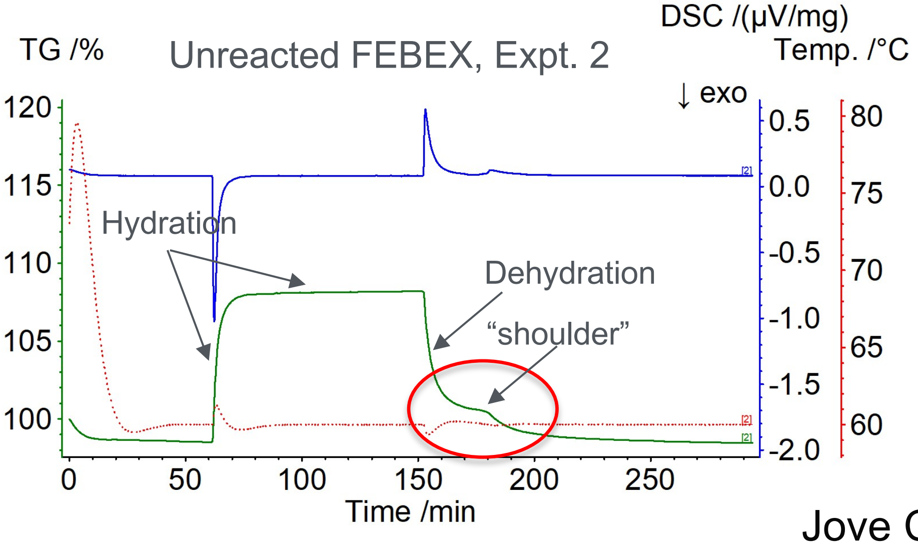
Ho et al. (2019)

FEBEX-DP: Thermal Analysis (TGA/DSC) Under Controlled Relative Humidity (RH) and Temperature

Section 49 Sampling





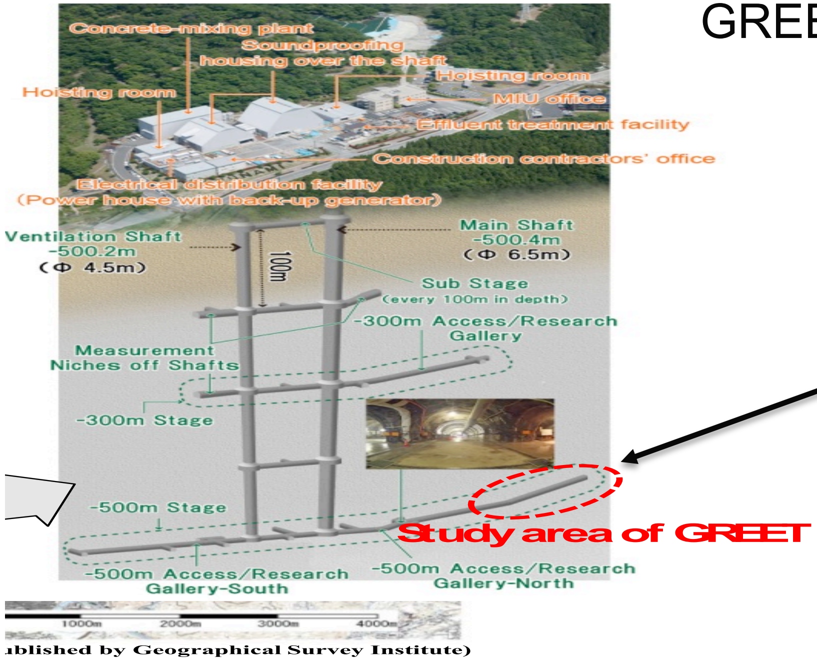


Bentonite Thermal Behavior

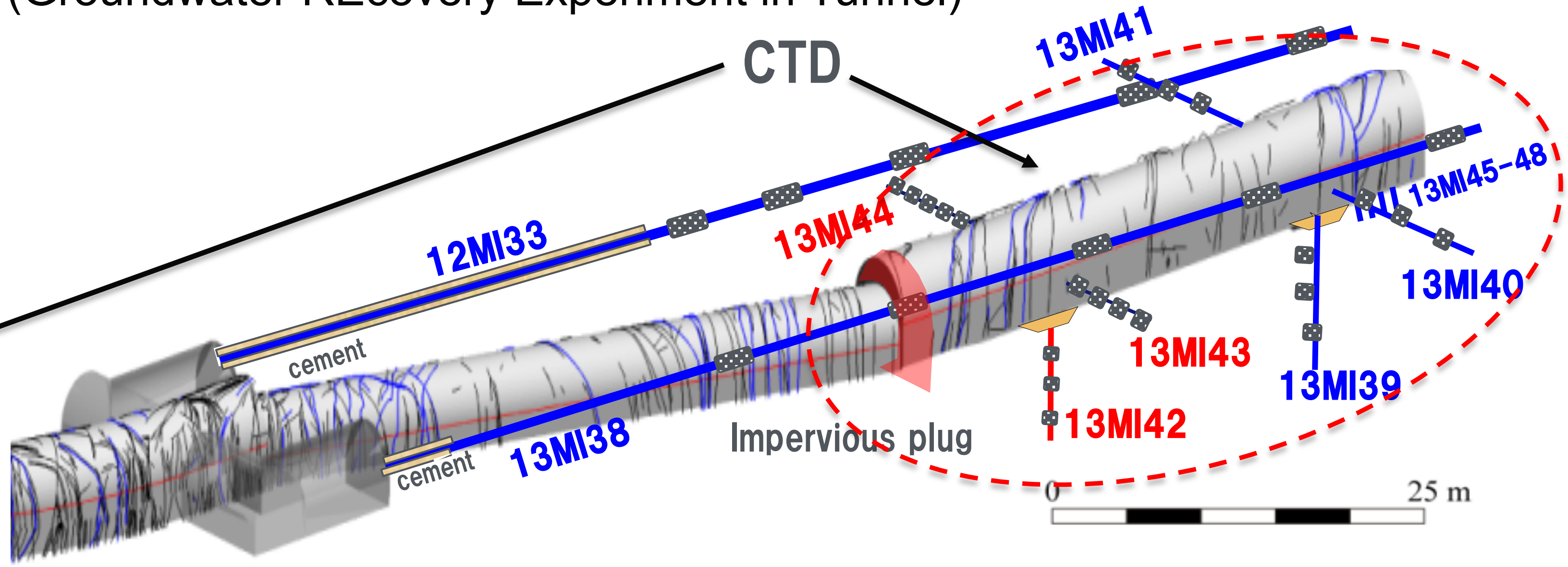
- Bentonite dehydration behavior is a function of the duration of hydration that precedes it.
- Appearance of a "shoulder peak" during dehydration suggests different energetics for swelling clay hydration and dehydration.
- Ideal for the thermal study of bentonite with additives.

Jove Colon et al. (2019)

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



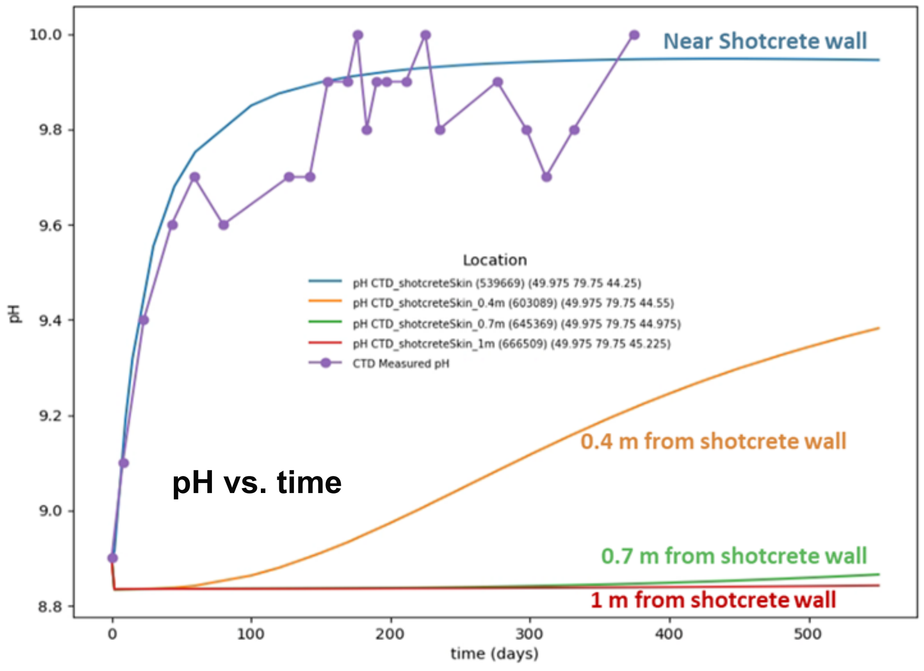
GREET (Groundwater REcovery Experiment in Tunnel)

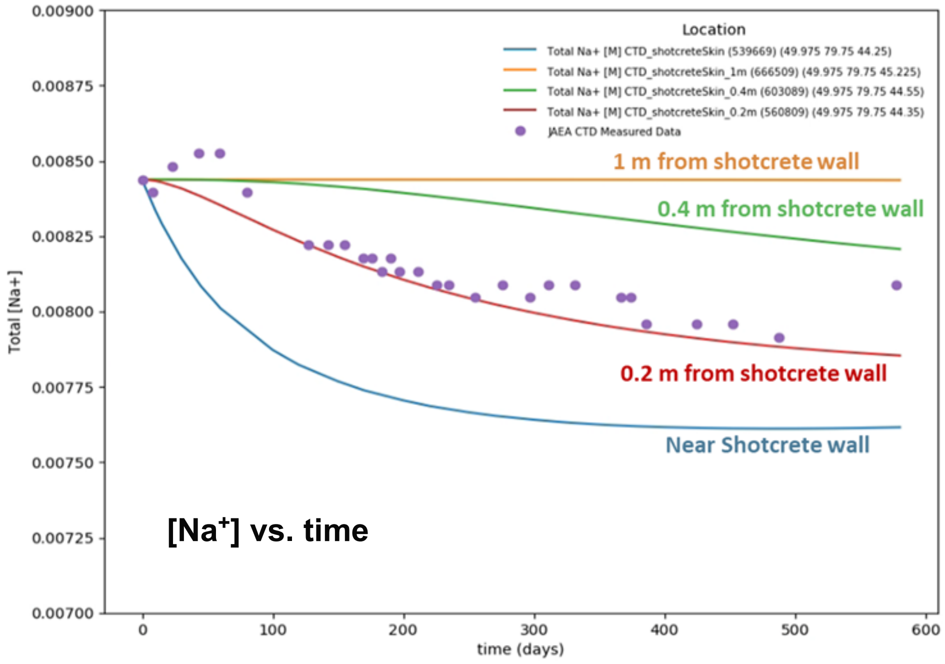


Schematic figure courtesy of Dr. Teruki Iwatsuki (JAEA)

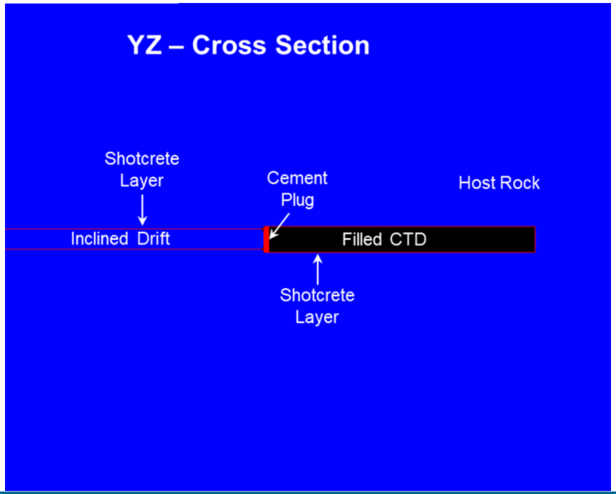
- 3D Reactive Transport Simulations using PFLOTRAN simulation code
- Focus: Shotcrete – groundwater interactions in the CTD

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 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 hydrological-chemical (HC) model sensitivities to shotcrete thickness; expand cyclic thermal analyses & X-ray diffraction (XRD) methods to evaluate high temperature effects; maintain engagement with international programs (DECOVALEX2023; EBS Task Force)

References

- Bauer, J., Rowan, C., Barkhurst, A., Digiulio, J., Jones, K., Sabbatino, M., Rose, K. and Wingo, P. (2018) NATCARB. National Energy Technology Laboratory-Energy Data eXchange; NETL, United States (doi:10.18141/1474110).
- Bildstein, O. and Claret, F. (2015) Stability of clay barriers under chemical perturbations, Developments in Clay Science. Elsevier, pp. 155-188.
- Blondes, M.S., Gans, K.D., Engle, M.A., Kharaka, Y.K., Reidy, M.E., Saraswathula, V., Thordsen, J.J., Rowan, E.L. and Morrissey, E.A. (2018) U.S. Geological Survey National Produced Waters Geochemical Database (ver. 2.3, January 2018), U.S. Geological Survey data release. U.S. Geological Survey Reston, VA; https://doi.org/10.5066/F7J964W8.
- Delage, P., Cui, Y.J. and Tang, A.M. (2010) Clays in radioactive waste disposal. Journal of Rock Mechanics and Geotechnical Engineering 2, 111-123.
- Ho, T.A. and Jové Colón, C.F. (2019) Non-isothermal Multiphase 3D Reactive Transport Modeling of an Engineered Barrier System in Clay Rock (SAND2019-5514 PE), Spent Fuel and Waste Disposition, Annual Working Group Meeting, Las Vegas, Nevada. Sandia National Laboratories, Albuquerque, NM USA.
- Jove Colon, C.F., Payne, C., Knight, A., Moffat, H., Coker, E., Caporuscio, F., Sauer, K., Cheshire, M., 2019, Disposal in Argillite R&D Work Package: Introduction & Key Accomplishments; Spent Fuel and Waste Disposition Annual Working Group Meeting, Las Vegas, Nevada, May 21-23; SAND2019-5596 PE.
- Jové Colón, C.F., Ho, T.A., Coker, E., Weck, P., Hadgu, T., Kalinina, E.A., Lopez, C.M., Sanchez, A., Rodriguez, M., Boisvert, L., Rutqvist, J., Guglielmi, Y., Xu, H., Tian, Y., Zarzycki, P., Deng, H., Li, P., Hu, M., Nico, P., Borglin, S., Fox, P.M., Sasaki, T., Birkholzer, J., Caporuscio, F.A., Sauer, K.B., Rock, M.J., Jerden, J.L., Thomas, S., Lee, E., Gattu, V.K., Ebert, W.L., Zavarin, M., Wolery, T.J., Deinhart, A., Genetti, V. and Shipman, S. (2020) Evaluation of Nuclear Spent Fuel Disposal in Clay-Bearing Rock - Process Model Development and Experimental Studies, Spent Fuel Waste Science and Technology (SFWST) Deliverable M2SF-20SN010301051. Sandia National Laboratories, Albuquerque, New Mexico USA, 551 pp.
- Rutqvist, J., Guglielmi, Y., Xu, H., Zarzycki, P., Deng, H., Li, P., Hu, M., Nico, P., Borglin, S., Fox, P.M., Sasaki, T. and Birkholzer, J. (2020) Investigation of Coupled Processes in Argillite Rock: FY20 Progress (LBNL-2001324), Spent Fuel and Waste Disposition. Lawrence Berkeley National Laboratory, Berkeley, CA USA, 206 pp.
- Sauer, K., Caporuscio, F.A., Rock, M.J. and Jové Colón, C.F. (2019) Hydrothermal interaction of Wyoming Bentonite and Opalinus Clay (LA-UR-19-24523), Spent Fuel and Waste Disposition Annual Working Group Meeting. Los Alamos National Laboratory, Los Alamos, New Mexico USA.
- Sevougian, D., Stein, E.R., LaForce, T., Perry, F.V., Nole, M. and Chang, K.W. (2019) GDSA Repository Systems Analysis FY19 Update, U.S. Department of Energy Spent Fuel and Waste Science and Technology Campaign. Sandia National Laboratories, Albuquerque, NM USA.
- Stein, E., Mariner, P., Hammond, G., Frederick, J., and Sevougian, S.D., Generic Disposal System Analysis: Argillite Reference Case and Performance Assessment, 2017 SWSFT Annual Working Group Meeting; May 24, 2017; SAND2017-5450 PE.
- Stein, E.R., Bryan, C.R., Dobson, D.C., Hardin, E.L., Jové Colón, C.F., Lopez, C.M., Matteo, E.N., et al. (2020), Disposal Concepts for a High-Temperature Repository in Shale, Sandia National Laboratories, Albuquerque, NM USA. SAND2020-12471R, 180 pp.
- von Damm, K.L. (1987) Geochemistry of shale groundwaters: survey of available data and postulated mineralogic controls on composition (Publication No. 2941). Environrnental Sciences Division, Oak Ridge National Lab., Oak Ridge, TN, USA.

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