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

Spent Fuel and Waste Science and Technology (SFWST)

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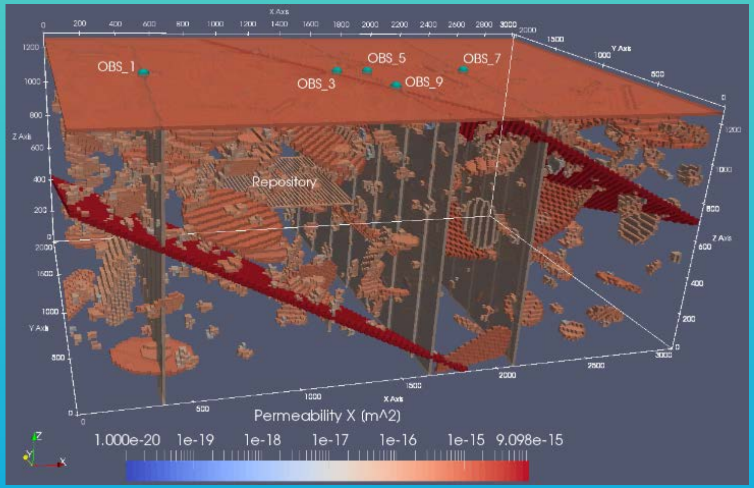

A Review of High Temperature Engineered Barrier Systems Experiments:
Part I: Modeling and Testing Activities of Bentonite Barrier Behavior (Sandia National Laboratories – SNL)

September 13th 2022 NWTRB Meeting
Arlington, VA

Carlos F. Jové-Colón (SNL)

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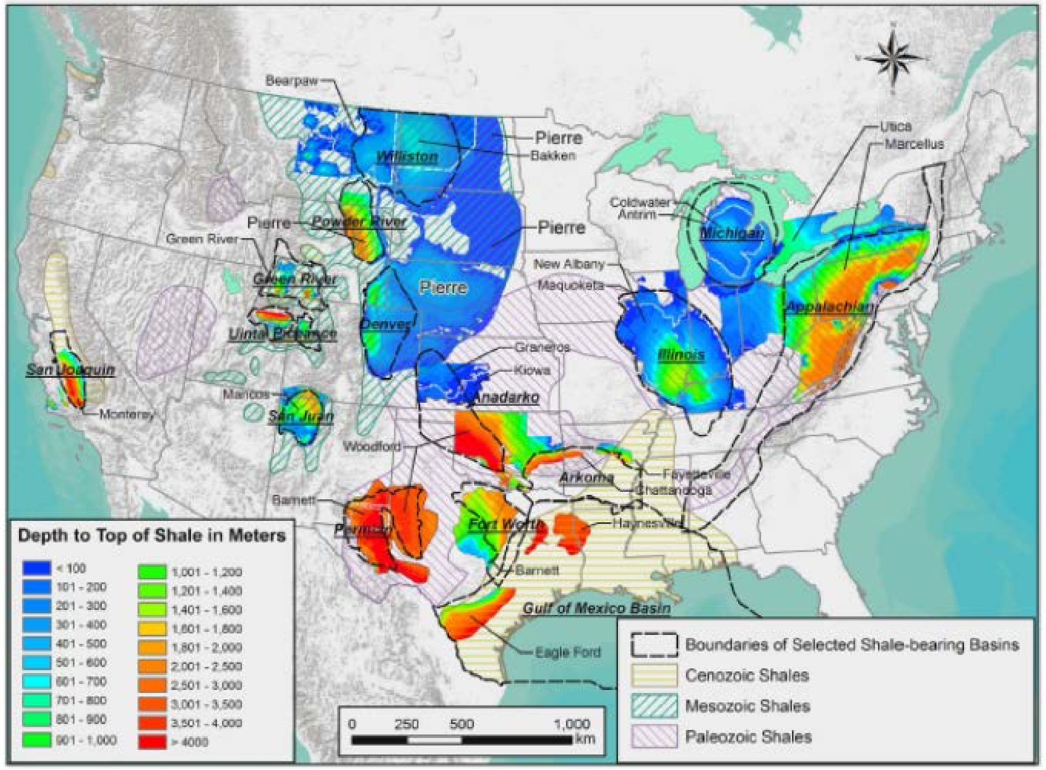
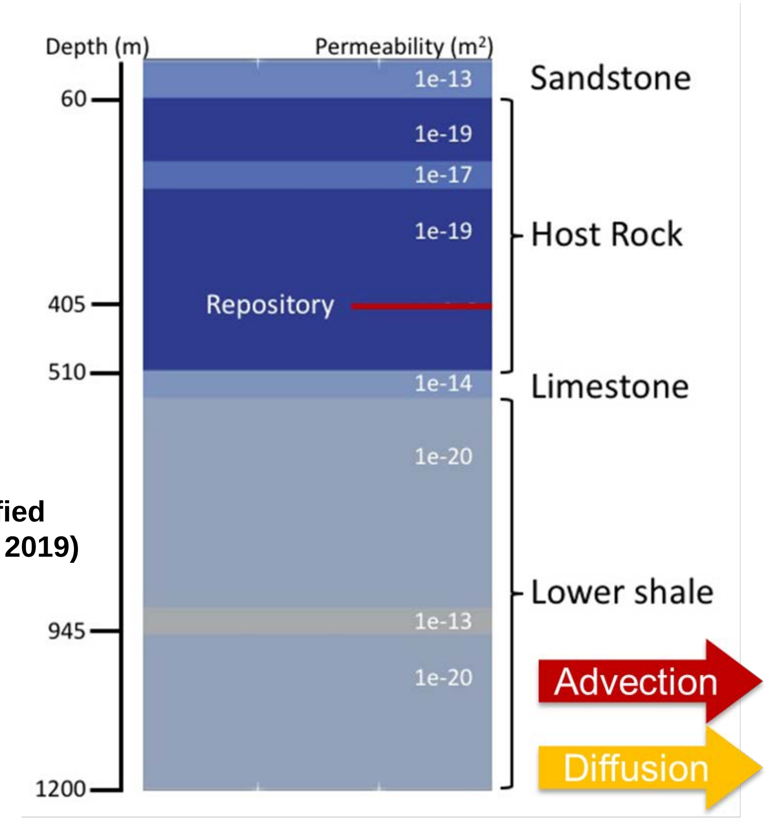
Argillite Host Rock Media



Source: Modified from Perry and Kelley (2014).

(Stein et al. 2020)

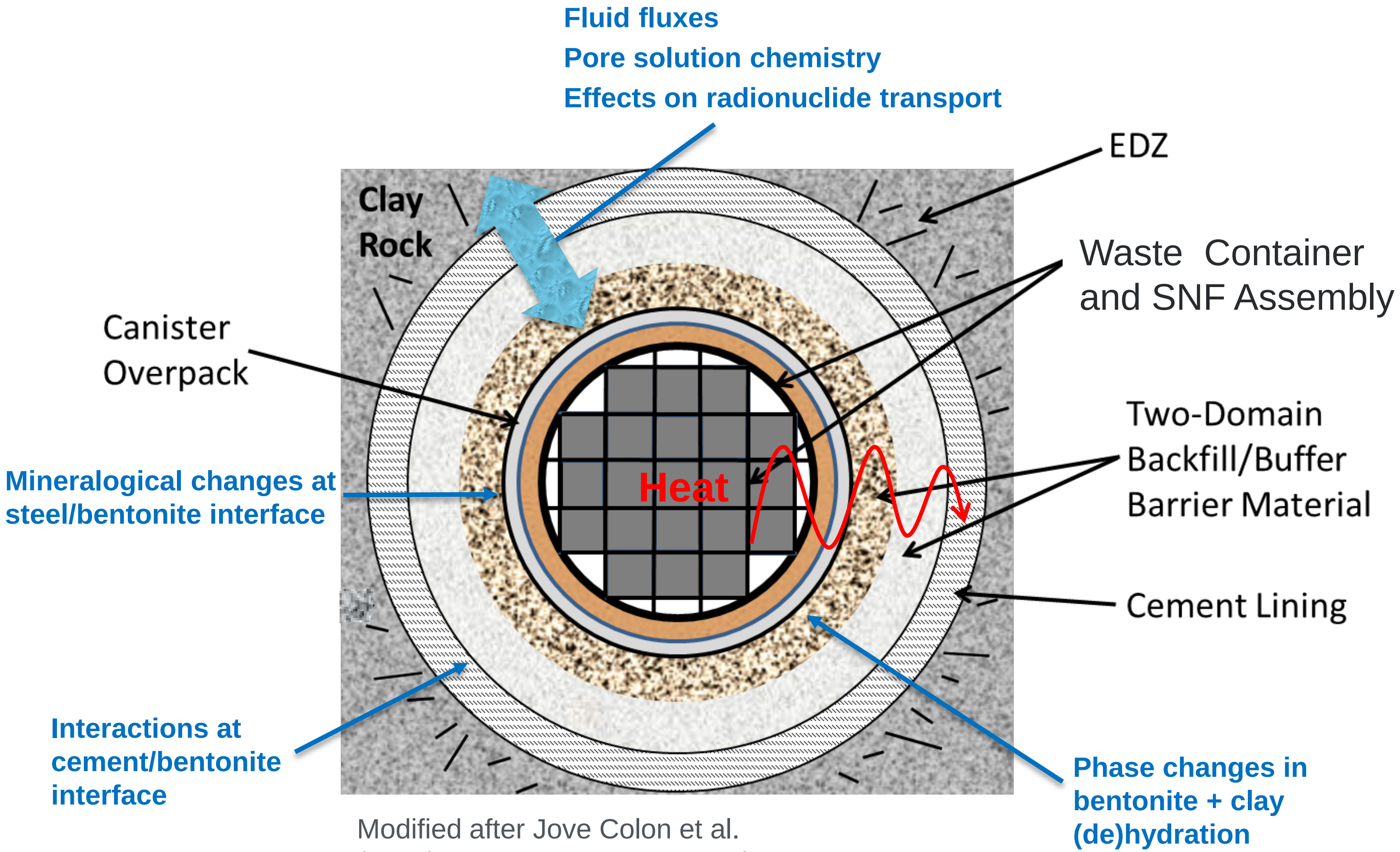
Distribution of Argillaceous Formation in the USA



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

- **Argillite Rock Characteristics**
  - Widespread geologic occurrence
  - Found in stable geologic settings
  - Appropriate thickness and depth for nuclear waste disposal concepts
  - Self-sealing properties
- **Highly effective retardation in host rock**
  - Low permeability
  - Low effective diffusion coefficient
  - High sorption capacity

Near-Field Processes



Modified after Jove Colon et al. (2019), DOE SFWST presentation.

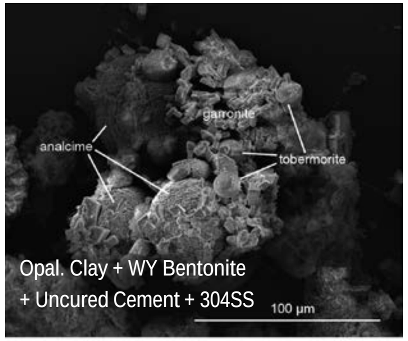
EDZ = Excavation Disturbed Zone
SNF = Spent Nuclear Fuel
SFWST = Spent Fuel Waste Science and Technology

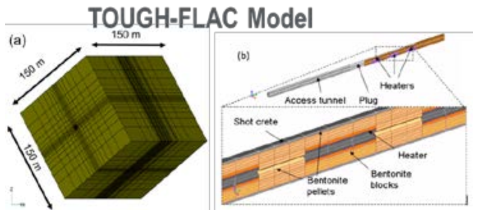
Thermally-driven processes

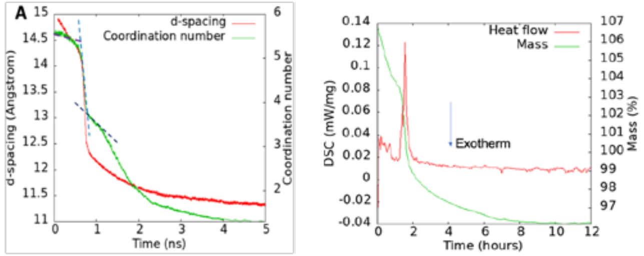
- **Clay barrier degradation**
- **Canister corrosion**
- **In-package chemistry**
- **Fluid transport in backfill**
  - **Bentonite swelling/shrinkage**
  - **Thermal phase stability**
  - **Pore solution interactions**

Highlights – Disposal in Argillite R&D: Experimental & Modeling Activities

- **Experimental Activities: Barrier Material Interactions at high temperatures (LANL)**
- **International Collaborations & Disposal R&D (SNL):**
  - **DECOVALEX2023: Modeling of THC processes in bentonite**
  - **SKB Task Force (TF): cement-bentonite interactions (Task 12; subtask A)**
  - **HotBENT (Grimsel site): Material characterization of column test bentonite**
- **Molecular dynamics (MD) simulation of water transport phenomena in smectite (SNL)**
- **Modeling of Ordinary Portland Cement (OPC) leaching experiments (SNL, Vanderbilt Univ.)**
- **Modeling of coupled THMC processes & shale creep in argillite repository (Int. Collaborations – LBNL)**
- **Machine-Learning (ML) approach for radionuclide-mineral interactions & surface complexation database development (LLNL)**
- **Thermodynamic database development (LLNL, SNL)**

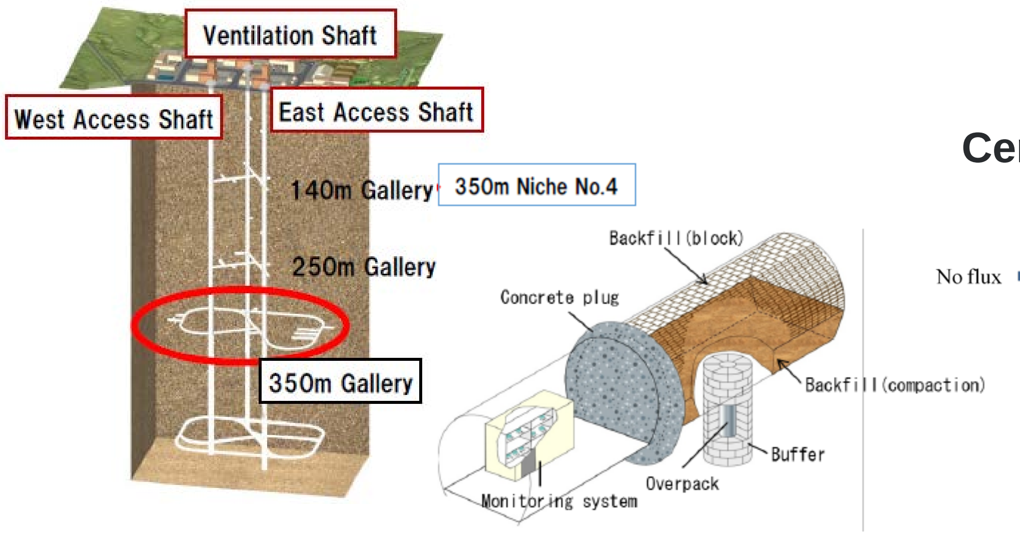






International Activities

DECOVALEX2023
(Task D – EBS Experiment)

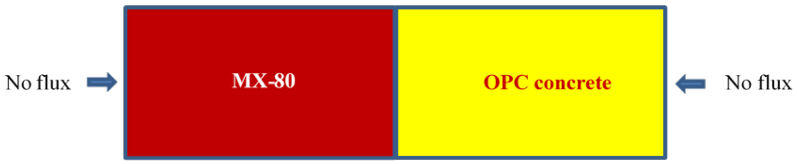


Honorobe URL (Japan)

Source: DECOVALEX2023, Task D presentation, Dr. Y. Sugita (JAEA)

SKB Task Force (TF)
Engineered Barrier System (EBS)
(Task 12)

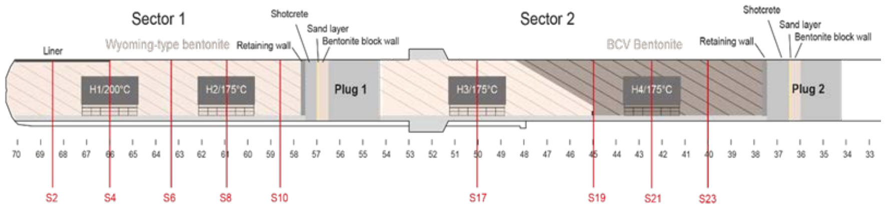
Cement-Clay Interaction Modeling



Source: SKB Task Force Description (2022)

EBS = Engineered Barrier System
JAEA = Japan Atomic Energy Agency
SKB = Swedish Nuclear Fuel and Waste Management Company
URL = Underground Research Laboratory

HotBENT



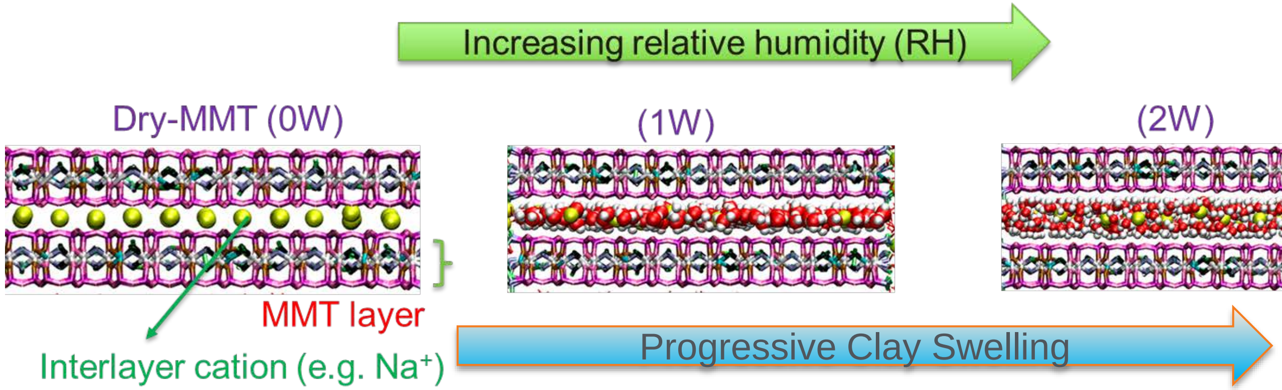
Source: HotBENT Modelling Platform Doc. (2022)

Bentonite (De)hydration Phenomena

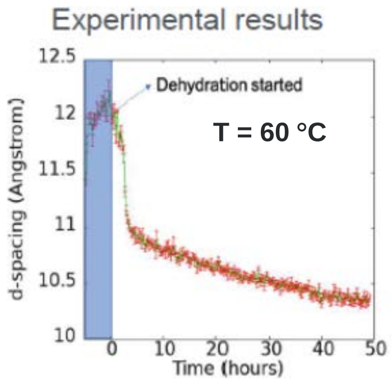
Research Questions:

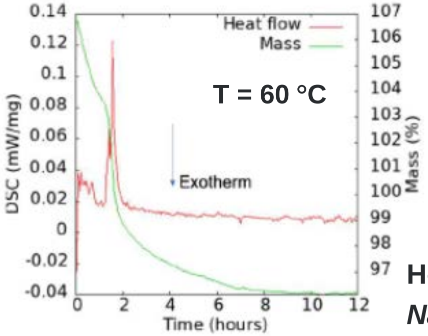
Water transport in smectite clay interlayers during clay dehydration?
Thermal stability of bentonite and effects on swelling performance?

Molecular Dynamics (MD) Simulations



Thermal (TGA/DSC)
and *in situ* XRD (RH, T)





Ho et al. (2022)
Nano Letters

Objectives

- **Elucidate mechanisms of bentonite (de)hydration and stability at elevated temperatures**
- **Moisture transport and bentonite behavior under unsaturated conditions**
- **Model comparisons with experimental observations**

International Collaboration Activity

Problem Overview

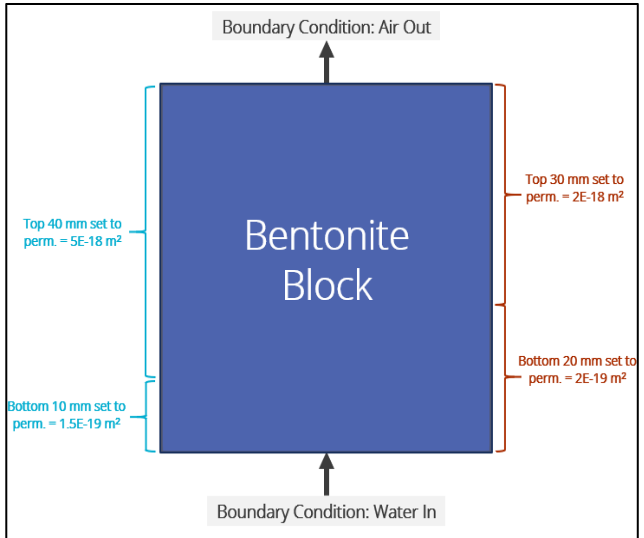
- A bentonite block is saturated vertically (from bottom to top) over 30 days
- Block is wetted with either deionized water (DW) or dilute groundwater (GW)

Computational Approach

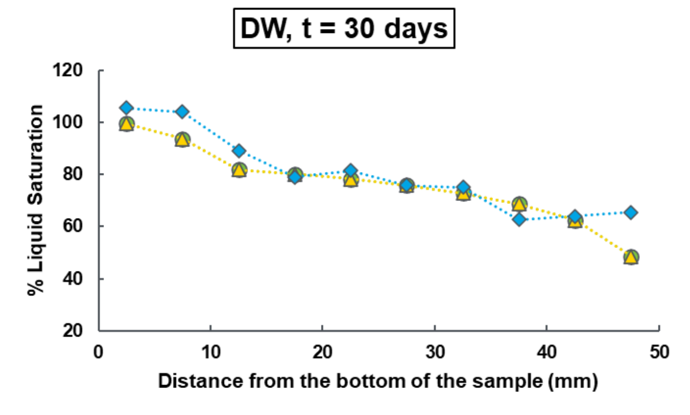
- Used PLFOTRAN to model laboratory experiments performed by JAEA
- 1D, saturation-driven, two-phase transport; chemistry off
- Permeability treated as variable input; heterogeneous
- Tested heterogeneous and homogeneous initial saturation profiles

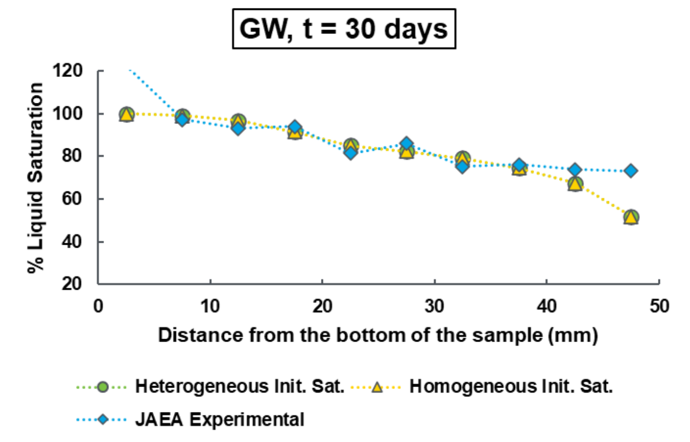
Findings

- General trends are well-represented by PFLOTRAN models
- Effects of initial saturation profile decrease with time



PFLOTRAN model inputs, including permeability structures for DW/GW cases.





Bentonite Block: Kunigel V1 bentonite + silica sand

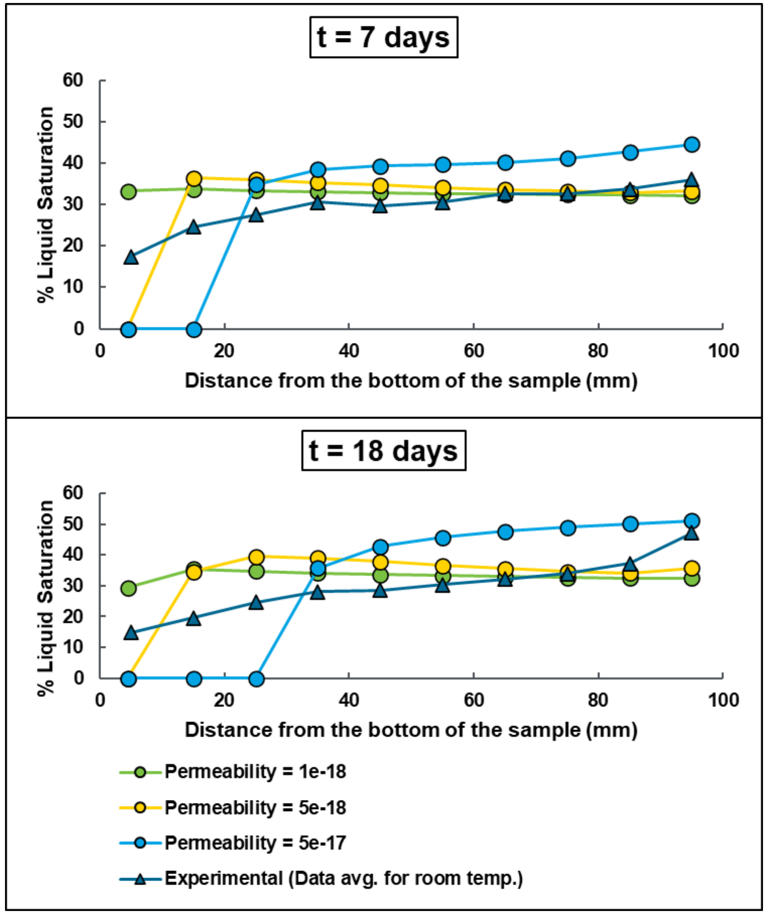
International Collaboration Activity

Problem Overview

- A temperature gradient is imposed on a bentonite block over 18 days
- Looking at evolution of saturation profile over time
- Constant temperature boundary conditions

Computational Approach

- Used PLFOTRAN to model laboratory experiments performed by JAEA
- 1D, saturation-driven, two-phase transport; chemistry off
- Permeability treated as variable input; homogeneous
- Uniform initial saturation
- Swelling not simulated
- Boundary conditions impose temperature gradient



Findings

- The model does not yet capture the trends of the experimental data.
- Results are very sensitive to permeability.

WORK IN PROGRESS!!!

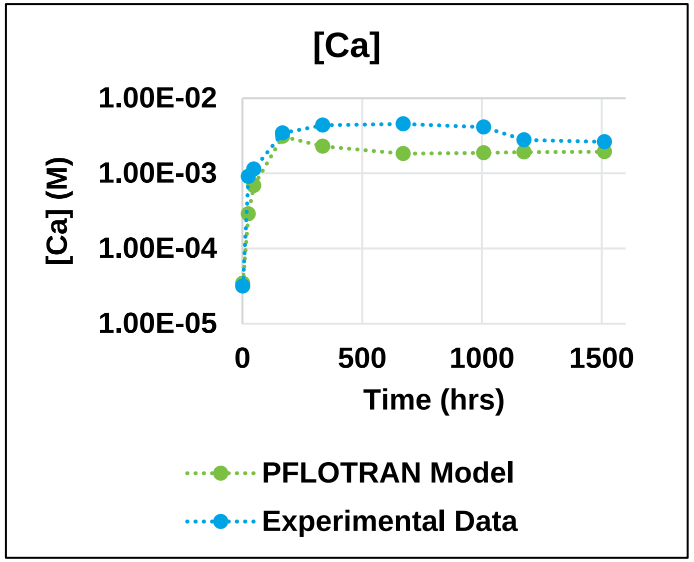
Bentonite Block: Kunigel V1 bentonite + silica sand

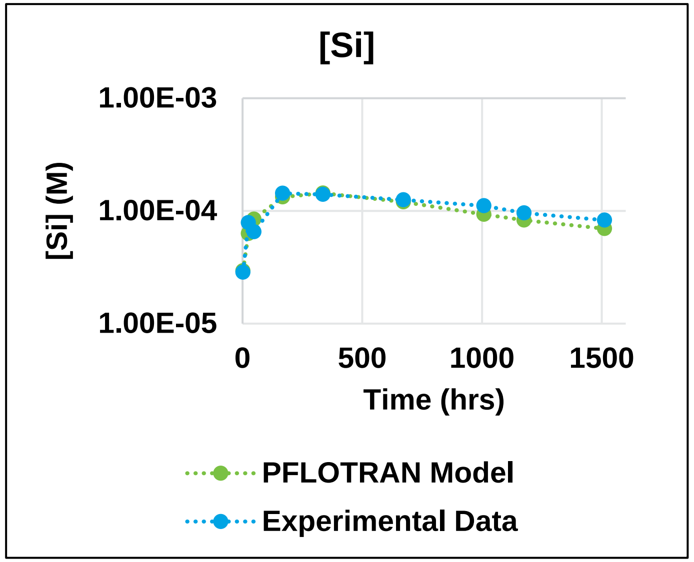
Modeling of OPC Leaching

Modeling leaching of OPC using PFLOTRAN, for comparison against experimental leaching data obtained by Vanderbilt University

- Diffusion-only 1D reactive transport model; isothermal (25°C)
- Reacting OPC with water over 1500+ hours.
- Experiments following EPA Method 1315 (Vanderbilt U.): leaching solution replenished with fresh water at specific time intervals.
- Initial cement composition uses prediction made by Vanderbilt's ORCHESTRA leaching model as a baseline.
- Anhydrous cement and sulfate salts are added to fit experimental data.

WORK IN PROGRESS





Ongoing and Future R&D Activities (SNL)

- PFLOTRAN THC modeling:
  - Variably saturated bentonite (TH) (isothermal / non-isothermal)
  - Reactive-transport modeling (HC) of OPC leaching experiments to evaluate chemical interactions at interfaces
    - Parameter evaluation, sensitivity analyses, mesh refinement
    - Reduced order model – development and implementation (e.g., bentonite swelling effects)
- LBNL HotBENT Heated/Unheated Column Experiments
  - Thermal analyses of bentonites from column experiments
  - Compositional and mineralogical characterization
- Cyclical thermal analyses at higher temperatures and controlled moisture conditions
  - In situ XRD analyses under controlled moisture and temperature conditions
  - Close examination of calorimetric data
- MD simulations on dehydration phenomena of the clay interlayer
  - Exploratory studies of H_2(gas) adsorption and transport/mobility at the clay interlayer
  - Analysis of thermodynamic parameters of clay dehydration from MD simulations
- Thermodynamic database evaluation / expansion / development
  - Feeds to geochemical and reactive-transport modeling of water/rock interactions
- Nuclear Energy University Partnership (NEUP) Project (U. of Nebraska-Lincoln; Texas A&M)
  - Multiscale and multiphysical testing-modeling of inorganic microfiber-reinforced engineered barrier materials (IMEBM) for enhancing repository performance

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