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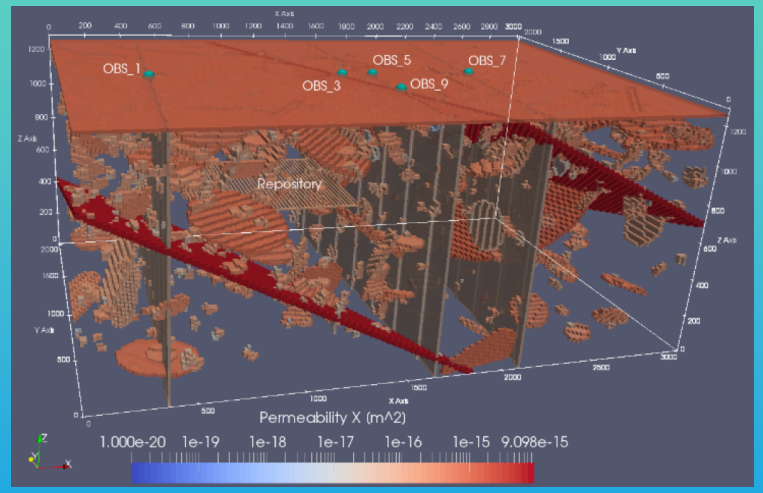

Session 1 (Tuesday): Introduction and overview of Argillite activities

May 2021 SFWD Campaign Virtual Meeting

SAND2021-XXXX PE

Carlos F. Jové Colón, Carlos M. Lopez, Amanda C. Sanchez
Sandia National Laboratories

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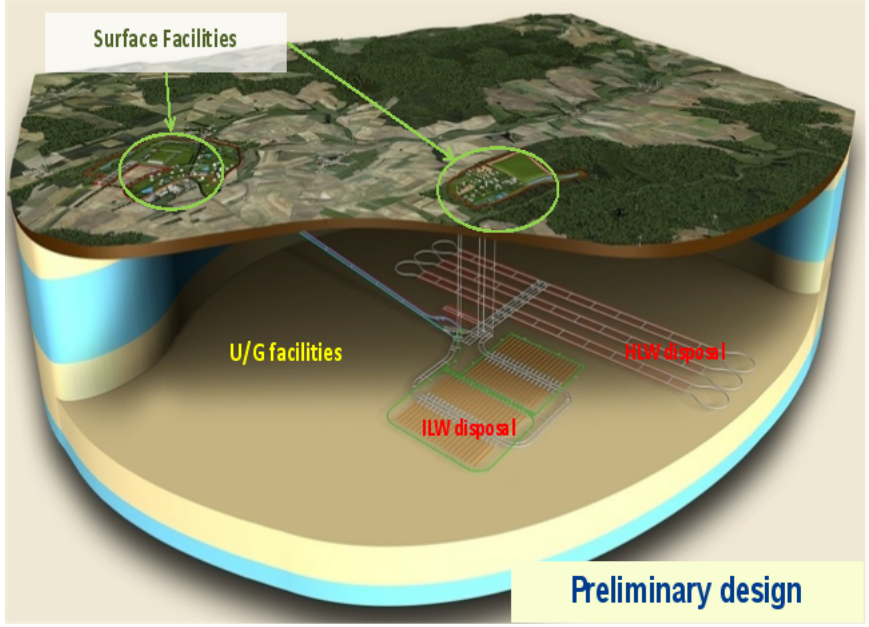
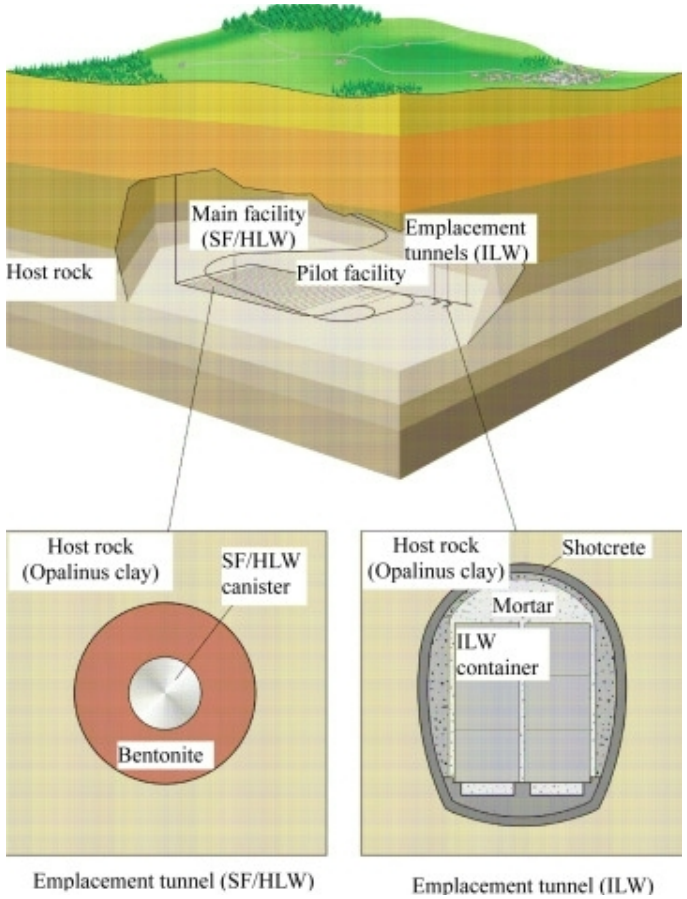
Argillite Repository Concept

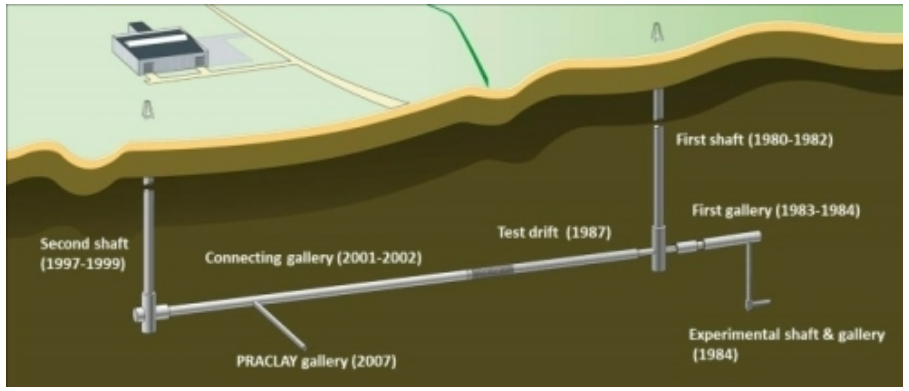


High-Level radioactive waste disposal (ANDRA) – **COx Argillite** (Bildstein and Claret 2015)

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



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



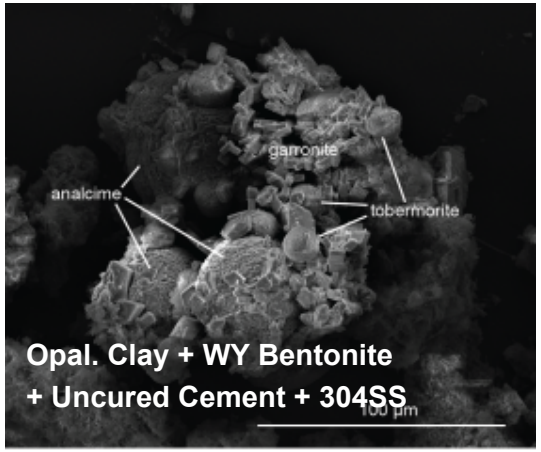
Belgian repository concept- HADES Underground Laboratory – **Boom Clay** (https://science.sckcen.be/en/Facilities/HADES)

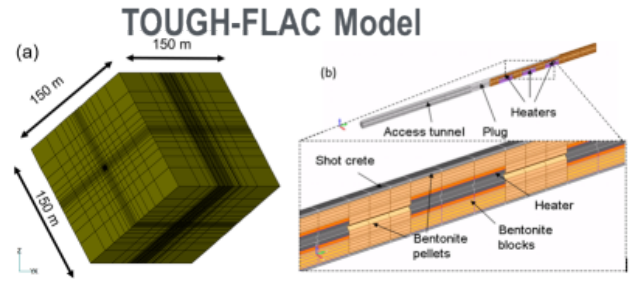
Attributes:

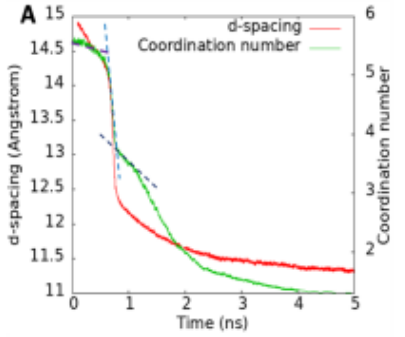
- **Low permeability**
- **Low hydraulic gradients**
- **Low diffusion coefficients**
- **Good sorption capacity**
- **Widespread geologic occurrence**
- **Appropriate thickness/depth for nuclear waste disposal concepts**
- **Found in stable geologic settings**
- **Self-sealing properties**

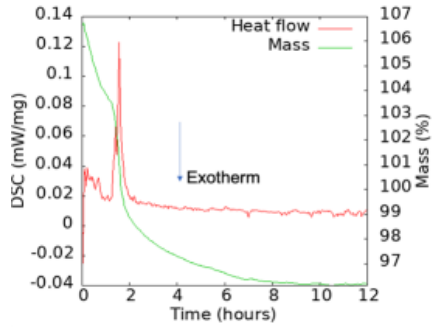
Highlights – Disposal in Argillite R&D

- **Experimental Activities: Barrier Material Interactions at high temperatures (LANL)**
- **Experimental/characterization studies: Thermal and XRD analyses of bentonite hydration/dehydration at elevated temperatures (SNL)**
- **Molecular dynamics (MD) simulation of water transport phenomena in smectite (SNL)**
- **Modeling of coupled THMC processes in argillite repository (Shale Creep, Int. Collaborations - LBNL)**
- **In situ fault activation experiments (LBNL)**
- **Radionuclide immobilization and the role of corrosion (LLNL)**
- **Update on thermodynamic database (LLNL et al.)**



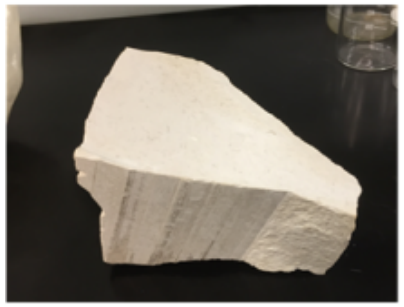






Marl Leaching: 1D Reactive-Transport Modeling of Sedimentary Rock Leaching

a



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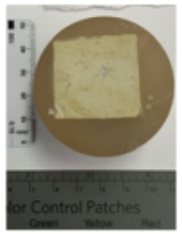
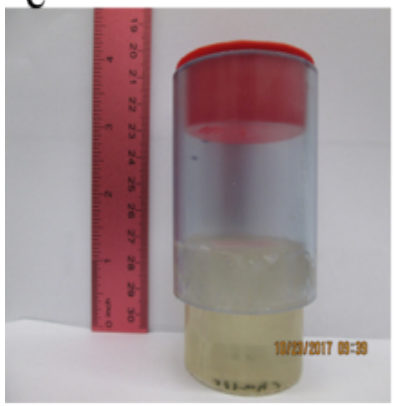


Marl

c

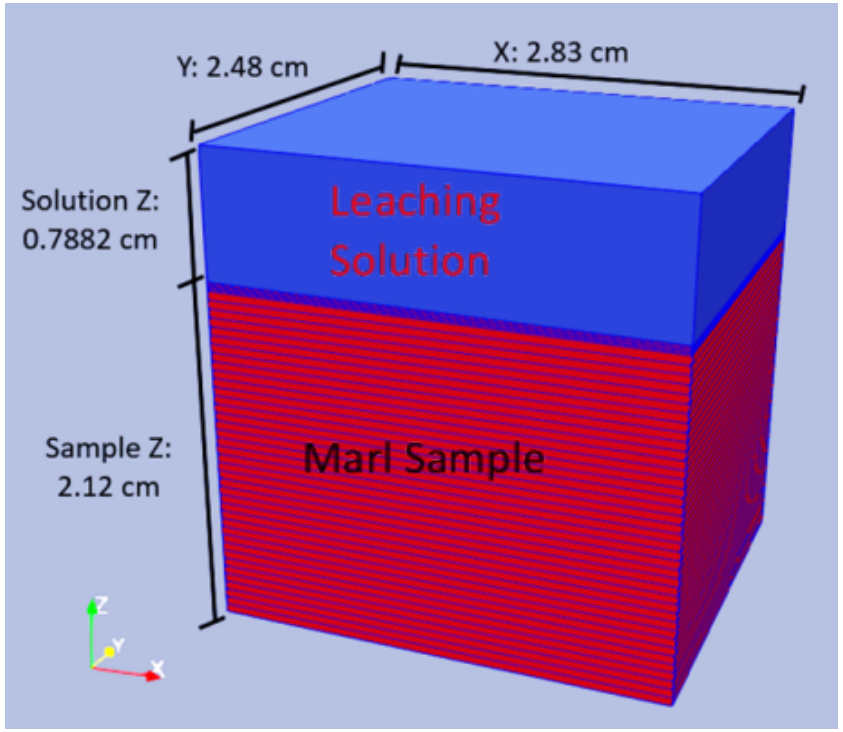


Source: Dr. C. Gruber & Dr. David Kosson (Vanderbilt Univ.)

- Leaching Solution: Dilute LiBr Solution; pH 9.4 – 10.8
- Marl Pore Solution: Sample saturated with H_2O prior to experiment; very diluted starting solution for the reactive-transport simulation

EPA 1315 test: Tank leaching procedure to quantify mass release rates of compacted solid materials

PFLOTRAN 1D Reactive-Transport Model Setup



- Marl porosity (~32%)
- Marl minerals (vol. fractions):
  - Calcite (0.64) – Constrained from XRD
  - Kaolinite (0.021)) – Constrained from XRD
  - Quartz (7×10^{-3})) – Constrained from XRD
  - Halite (7.5×10^{-4}) – Constrained from $[Na^+]$ profile
  - Sylvite (2.7×10^{-5}) – Constrained from $[K^+]$ profile

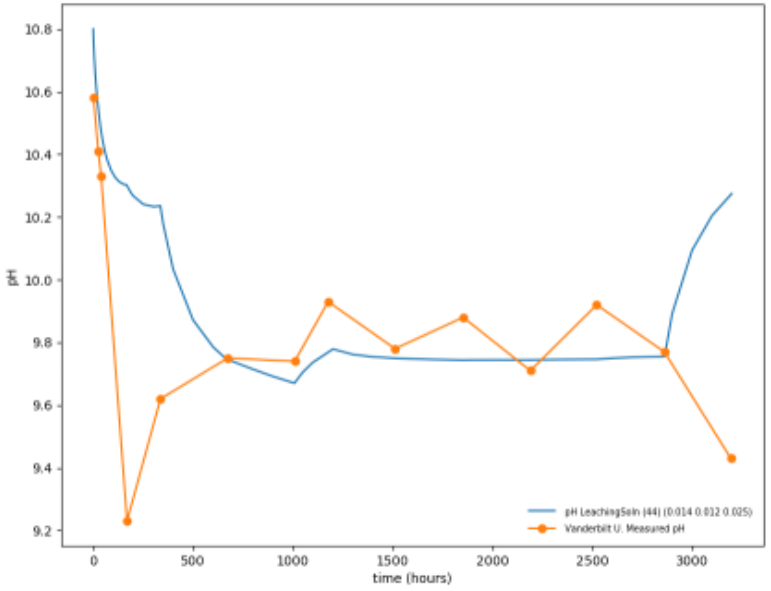
Objectives

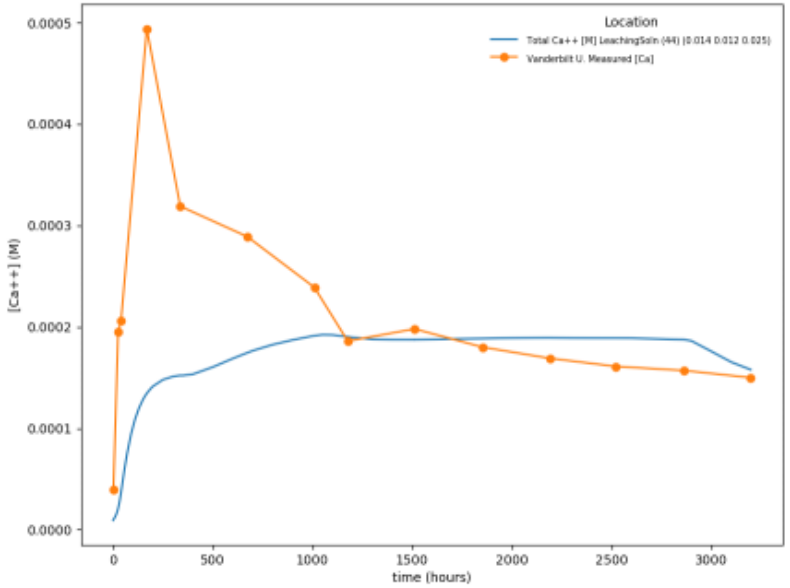
- To represent a leaching experiment (EPA Method 1315) of marl rock sample with a 1D reactive transport model using PFLOTRAN
- EPA 1315 test: Tank leaching procedure to quantify mass release rates of compacted solid material
- Capture episodic changes of solution chemistries in leaching intervals
- Evaluate model parameter sensitivities through comparisons between model and experimental leaching data

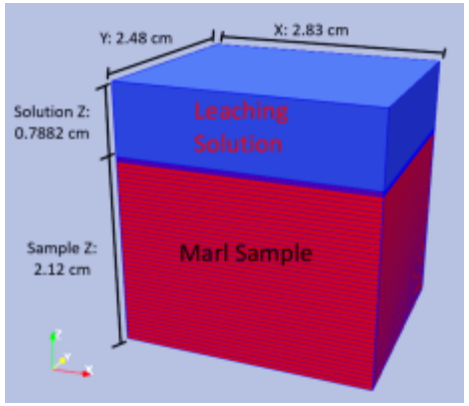
Work is conducted in concert with the EBS work package.

Preliminary Results: [Ca], pH vs. time profiles

Work in Progress!!!





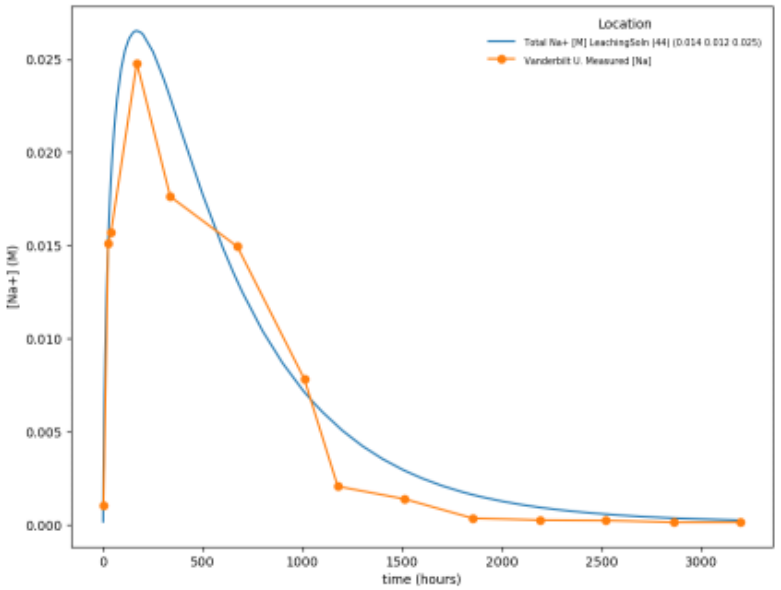


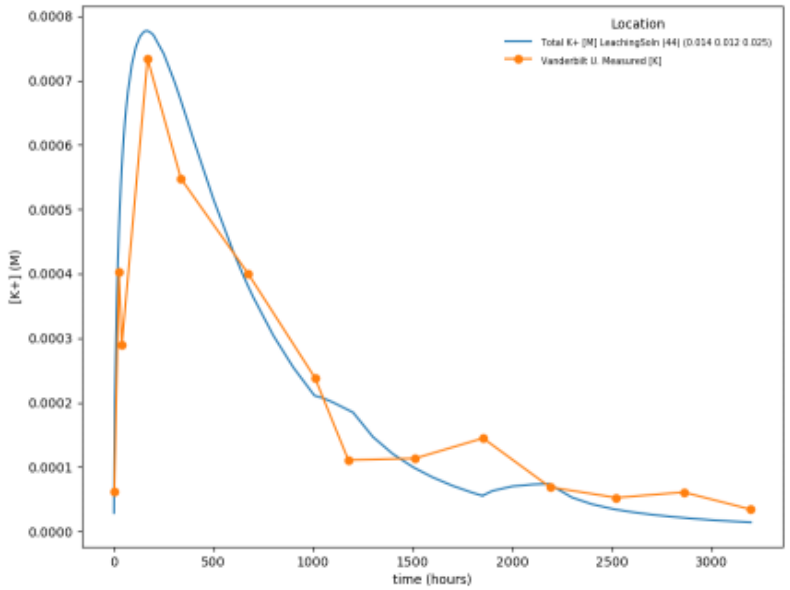
Observation point: Center of leaching volume cell

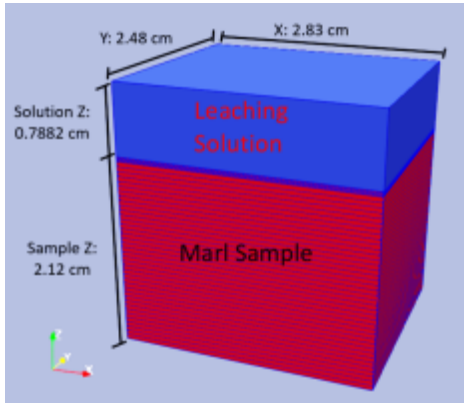
- Overall trends are captured at later times:
- Discrepancies in the initial and final drops in pH
- Discrepancy in the initial [Ca] peak but stabilize after 1000 hours
  - Further evaluation of TST rate law parameters for calcite
  - Surface area effects on solute profiles

Preliminary Results: [Na], [K] vs. time profiles

Work in Progress!!!







Observation point: Center of leaching volume cell

- The overall [Na^+] and [K^+] profile is well represented
  - Using halite (NaCl) & sylvite (KCl) as reactant phases – suggested by the Vanderbilt Univ. group
- Both profiles were used to constrain the diffusion coefficient to a value of 2.5×10^{-10} cm^2/s

FY22 and Ongoing FY21 Activities (SNL)

- Continue (1D) PFLOTRAN reactive-transport modeling of leaching of rock (monolith) and cement experiments to evaluate chemical interactions at interfaces
  - Parameter evaluation, sensitivity analyses, mesh refinement
  - This activity is done in concert with the EBS work package
- LBNL HotBENT Heated/Unheated Column Experiments
  - Continue XRD analyses of both column experiments
  - Plans to conduct X-ray fluorescence (XRF) compositional analysis
- Continue cyclical thermal analyses at higher temperatures and controlled moisture conditions
  - in situ XRD analyses under controlled moisture and temperature conditions
- Expand MD simulations on dehydration phenomena of the clay interlayer
  - Expand the work on montmorillonite dehydration in the presence of K^{+} and Ca^{++}
  - Evaluate the effects of surface charge density on montmorillonite dehydration and water transport
  - Exploratory studies of H_2(gas) adsorption and transport/mobility at the clay interlayer (wet, dry)
  - Analysis of thermodynamic parameters of clay dehydration from MD simulations