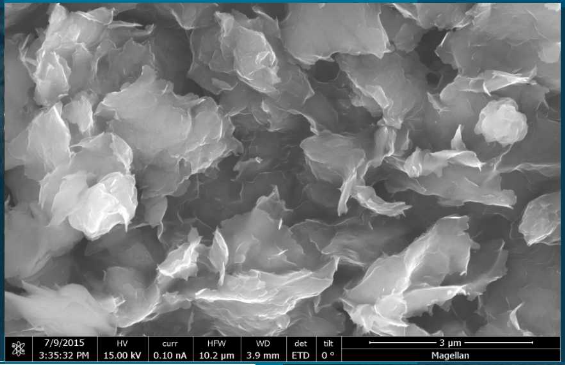
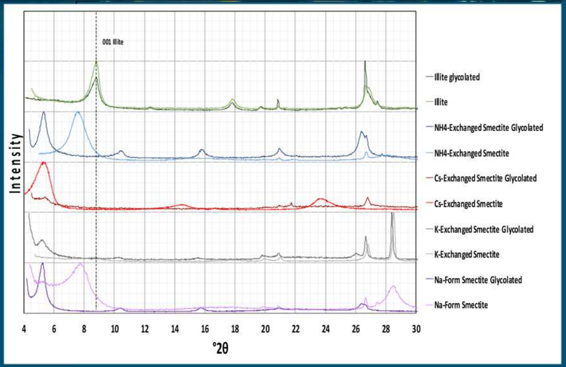
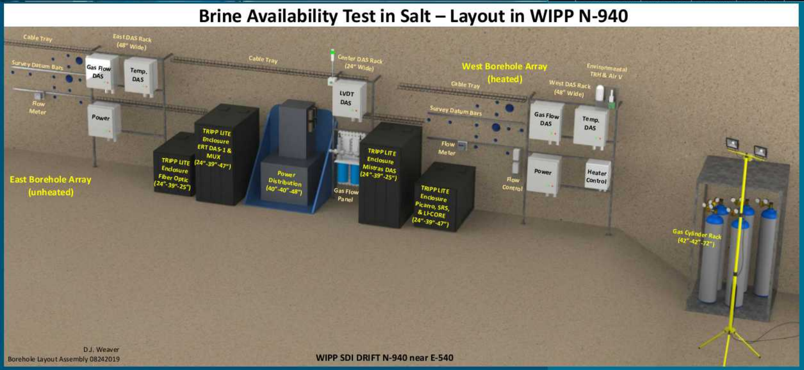
This paper describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the paper do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

SAND2020-2238C

Investigation and Characterization of Geotechnical Barrier Materials at Elevated Temperatures







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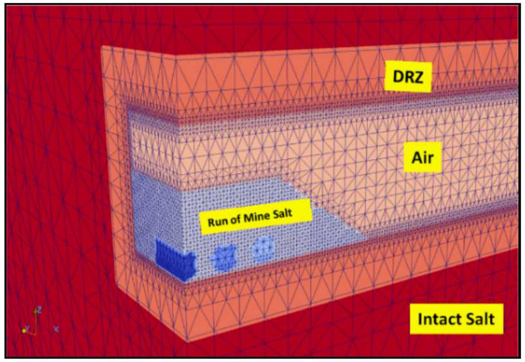
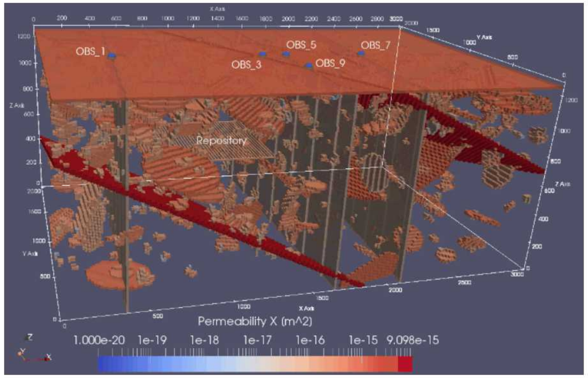
Overview

- Department of Energy Office of Nuclear Energy focused on research and development activities related to storage, transportation, and disposal of spent nuclear fuel and high-level nuclear waste.
- Within disposal campaign, various host rock media are being investigated to advance the understanding and performance of long-term isolation





Research presented here focuses on characterizing buffer materials proposed for geotechnical barriers with respect to heat-generating nuclear waste

Motivation

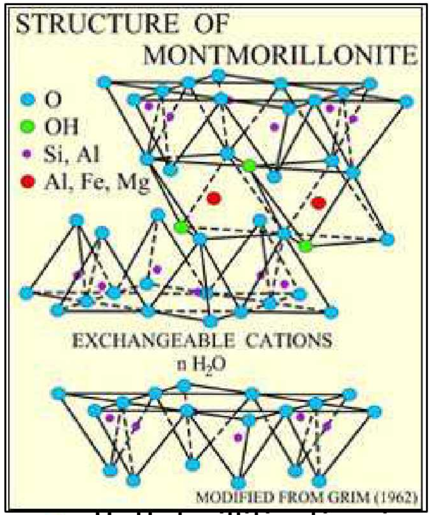
- Ongoing uncertainty in long-term performance of geologic repositories for nuclear waste disposal revolves around construction and temporal evolution of geotechnical barriers
  - In salt, barriers will have permeability and porosity approaching those of the surrounding host rock
  - In crystalline, barriers are critical for isolating emplaced waste given the properties of the surrounding host rock
  - Few large-scale, long-term in situ barrier experiments involve heat, especially including permeability and chemistry data

- Striving to add confidence in future nuclear waste repository design and performance with respect to heat generating waste

Clay Barriers: Illitization Studies

Background: Illitization

- To predict barrier functions over time within repository relevant temperatures, important to understand thermal alteration effects on smectite (montmorillonite), a main constituent of bentonite barriers
  - Illitization is a type of thermal alteration which weakens barrier functions



$2Na_{0.4}(Al_{1.47}Mg_{0.29}Fe_{0.18})Si_4O_{10}(OH)_2 + 0.85K^+ + 1.07H^+$

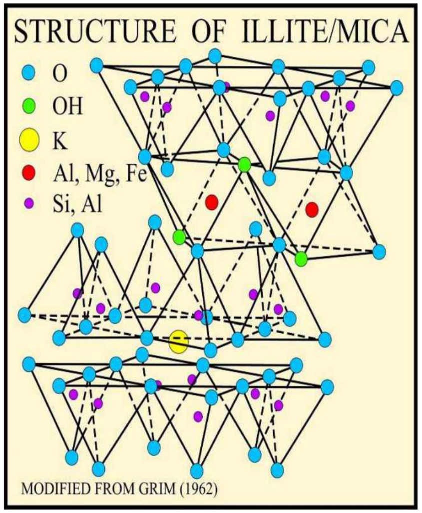
Na-Smectite

?

$1.065K_{0.8}(Al_{1.98}Mg_{0.02})(Si_{3.22}Al_{0.78})O_{10}(OH)_2$

Illite

$+ 4.6SiO_2 + 0.36Fe(OH)_3 + 0.56Mg^{2+} + 0.8Na^+ + 0.9H_2O$



- Path to illitization is complex
  - Dependent on Si, K, interlayer cations, temperature, solution chemistry, and time
- Important to safety case in crystalline repository since buffer break down is the main source of any release

Experimental Approach for Illitization

Exchanged Interlayer Cation

Liquid Chemistry

Liquid to Rock Ratio

Time

200 ° C

Volume limit for vessel: 150 mL

Current Sample Matrix

Temperature	200°C			
Time	1 Week	2 Week	2 Month	
Ratio of Liquid to Rock	100	500	1000	
Exchanged Cation	Sodium	Potassium	Cesium	Ammonium
Liquid Chemistry	1M KCl	1M Parent Cation	DI Water	

= 108 different samples

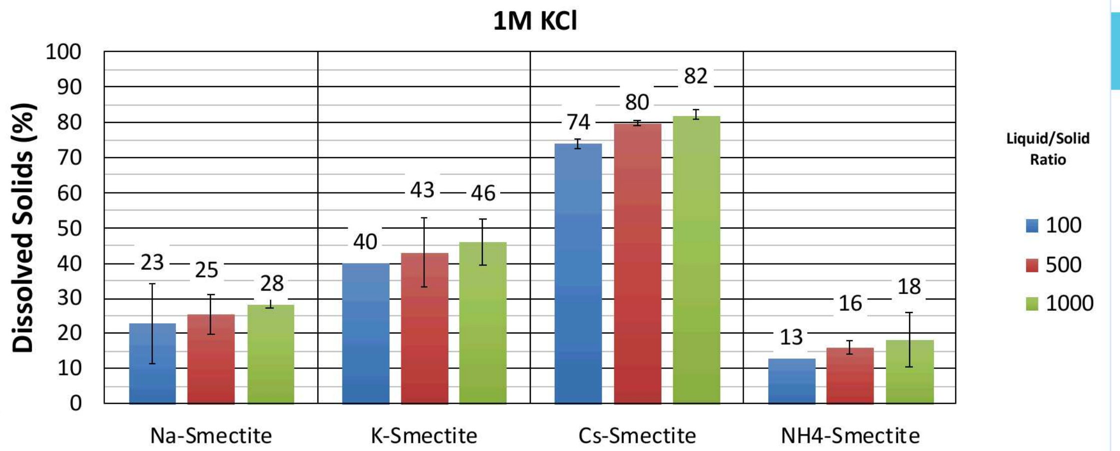
Experimental Techniques

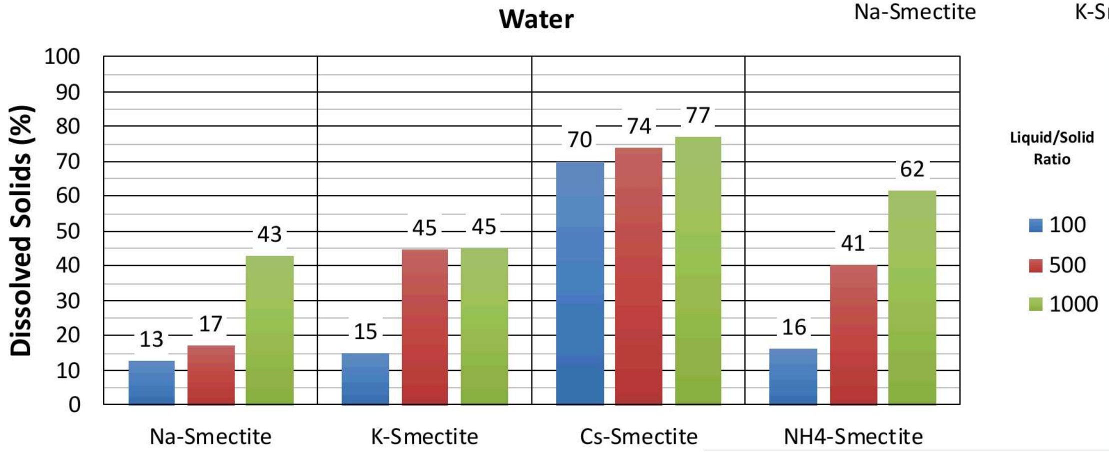
- XRD for basal spacing and lattice structure
- Cation exchange capacity (CEC)
- Recorded dissolved solid data and pH of reacted solutions
- Surface area by BET nitrogen adsorption
- Chemical analysis of solutions after reaction by IC, ICP-MS, ICP-OES to determine dissolved elements
- Particle size by Malvern Zetasizer
- XRF for composition
- SEM-EDS for morphology and compositional mapping

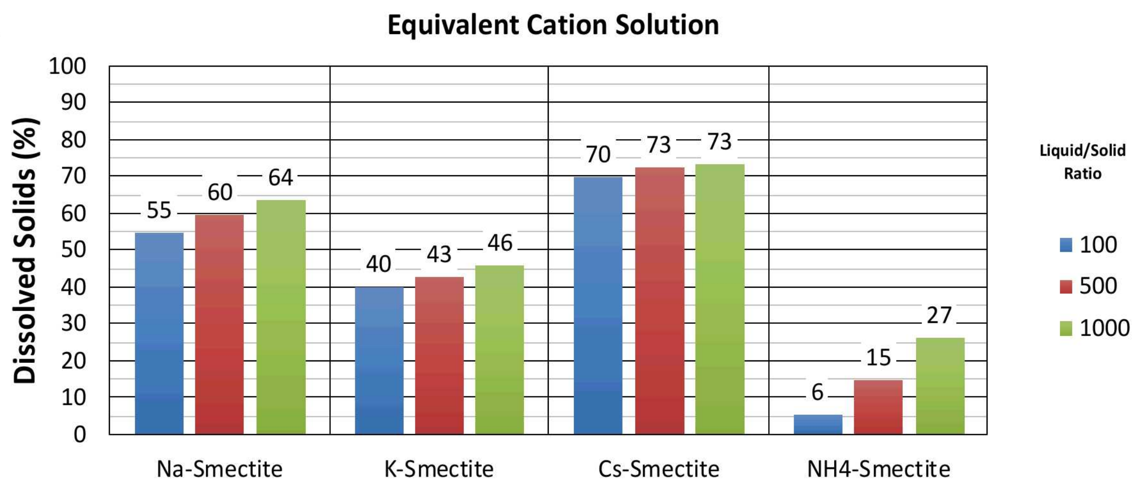
Observations

- Variable dissolution amounts
  - Solid in ≠ solid out
  - Dependent on liquid:rock loading and cation
- Basal spacing changes
  - XRD peaks shift
- Surface area and particle size increase in certain cases
- No significant change in pH
- Variability among concentration of Silica in solution

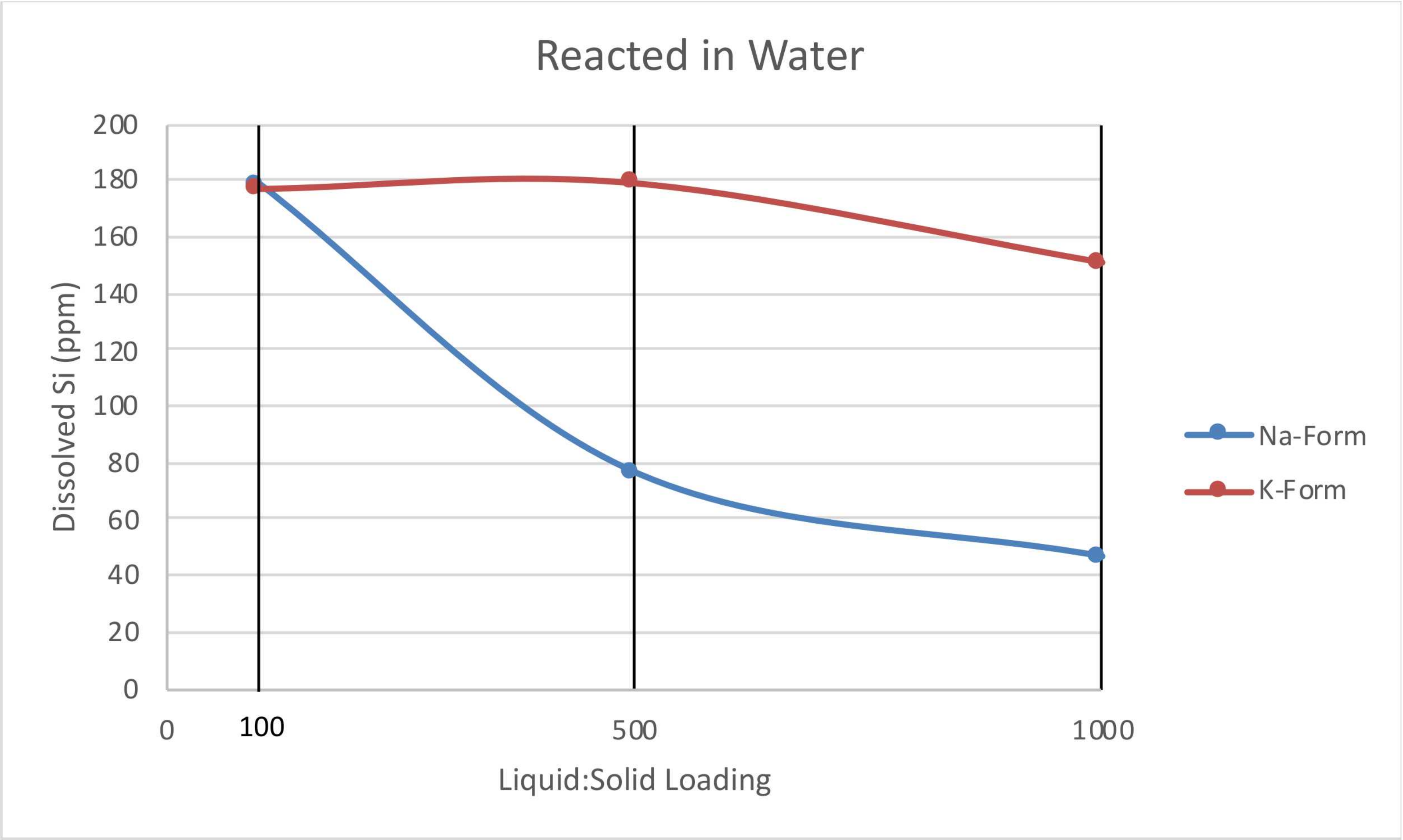
Results: Dissolution



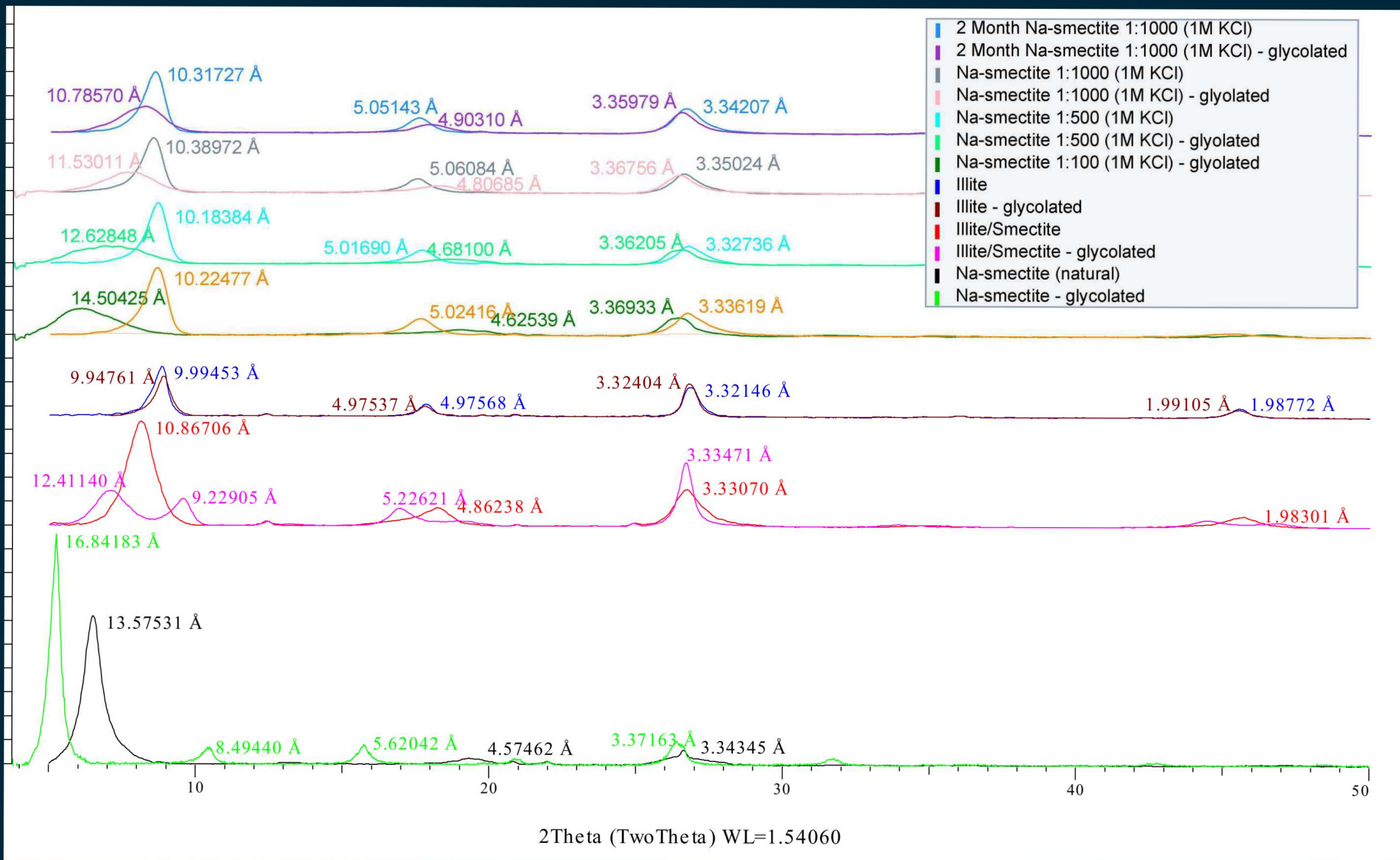




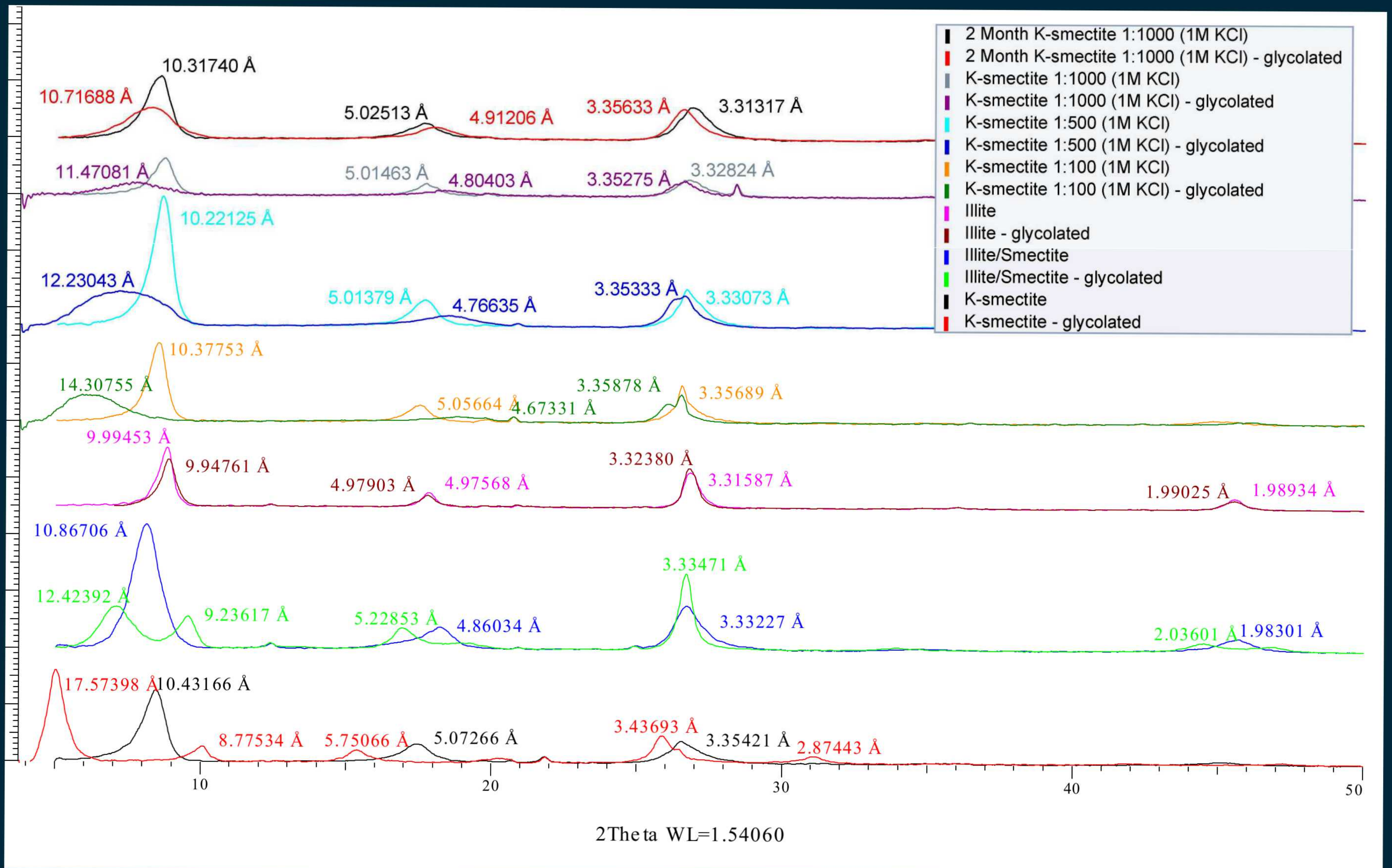
Dissolved Silica



Results: XRD- Sodium Form Smectite in 1M KCl

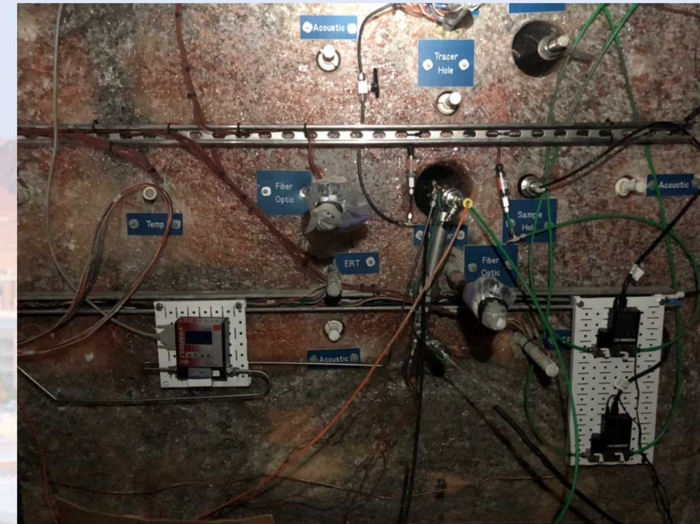


Results: XRD- Potassium Form Smectite in 1M KCl

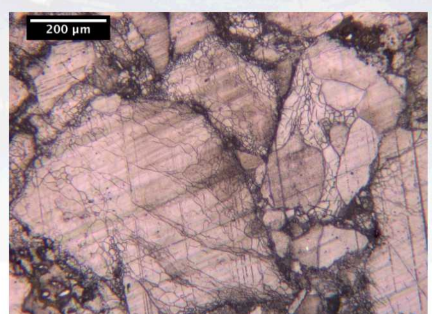


Remaining Research

- Explore different and longer time scales
- Continue analysis of solution chemistry
  - Effect of changes in liquid chemistry
- Continue analysis of composition (XRF)
- Changes in morphology by SEM
- Investigate behavior of iron in reaction
- Addition of mica to possibly escalate conversion
- Addition of quartz to inhibit conversion



DOE-NE Salt Seal System Research

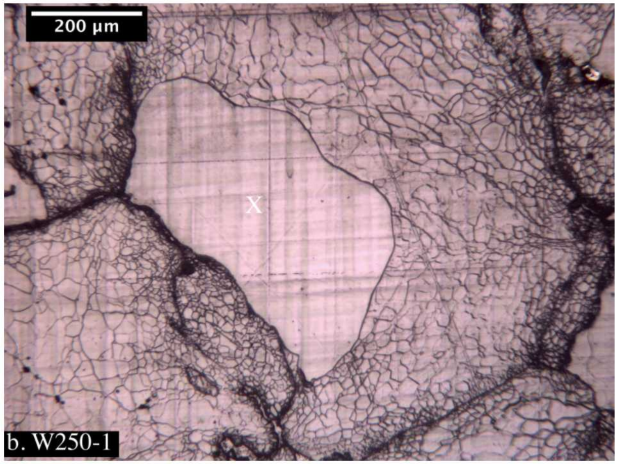


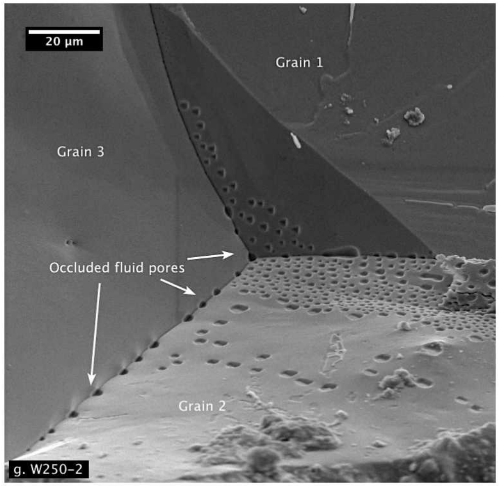
Barrier Systems in a Salt Repository

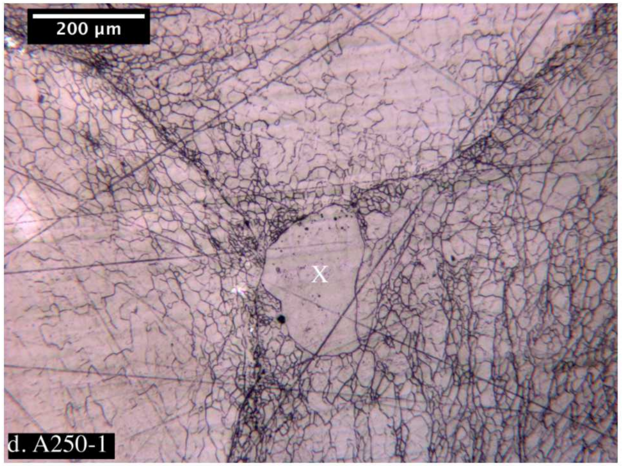
- Many programs on barriers systems in salt formations for repository applications
- Main types of barriers consist of either granular salt backfill or cementitious materials
  - Many different compositions, techniques of emplacement, and response of material depending on the formation (bedded, domal, or pillow)
- Focus will be on research conducted at elevated temperatures in relation to the Waste Isolation Pilot Plant (WIPP) in Carlsbad, NM.

Consolidated Salt Experiment at Elevated Temperatures

- Stormont et al. 2017: *Improving the Understanding of the Coupled Thermal-Mechanical-Hydrologic Behavior of Consolidating Granular Salt*
  - Granular domal and bedded salt consolidation at temperatures up to 250 °C, pressures up to 40 MPa, and addition of moisture
  - Porosity, permeability, and thermal conductivity was measured
  - Microscopic investigations were conducted to document the deformation mechanisms associated with reconsolidation
  - Results show samples exposed to higher temperatures of 250°C experience recrystallization regardless of moisture conditions







Previous Cement Seal Experiments related to WIPP

- 1977- ERDA No. 10 Test (near WIPP)

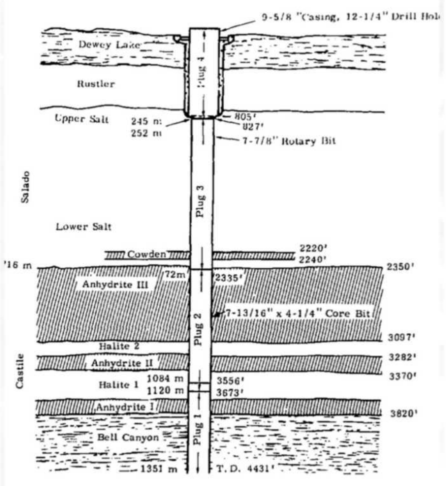


Figure 1. ERDA No. 10 Drill Hole

Gulick (1979)

- 1979- Bell Canyon Test (near WIPP)

- 1980's- Small-Scale Seal Performance Tests (at WIPP)
  - Used very specific formulation of "Expansive Salt Cor
  - Key ingredients are unavailable and potentially difficult reproduce

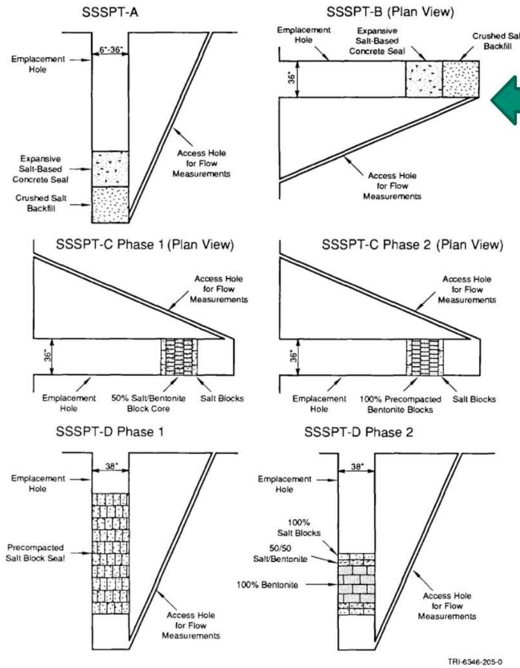
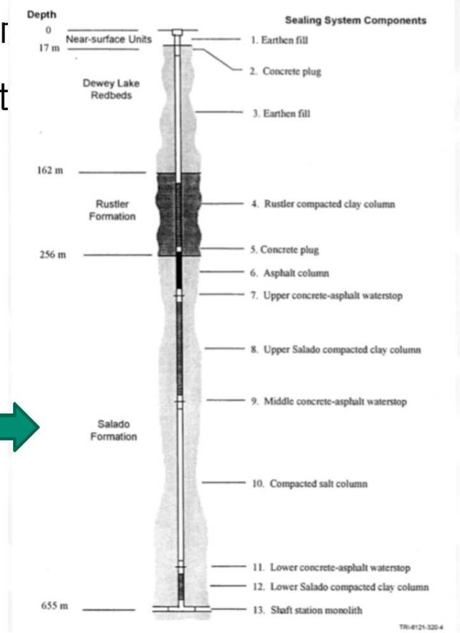


Figure 2. Generalized Test Configurations.

Finley et al. (1991)

- 1990's- Salado Mass Concrete
  - Design for closure



Hansen and Knowles (1999)

None focused on effects of elevated temperature exposure

Brine Availability Test in Salt (BATS)

- Heated field test in ~6-m horizontal boreholes at WIPP
  - Began heating: January 2020
  - Parallel heated/unheated tests
  - Temperatures up to 140°C
- Only active repository research underground test in the US
- Collaboration of Sandia, Los Alamos, and Lawrence Berkeley National Labs
- Includes cement seal components

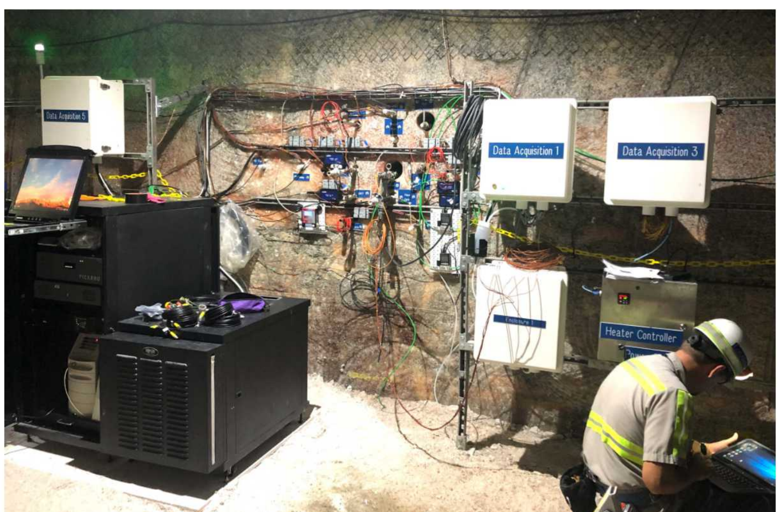
U.S. DEPARTMENT OF ENERGY
Office of Nuclear Energy

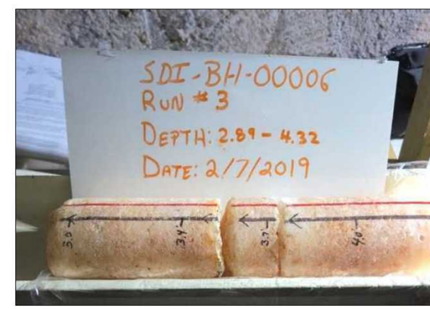
WASTE ISOLATION PILOT PLANT
WIPP
NWP • DOE • LANL • CTAC • SNL

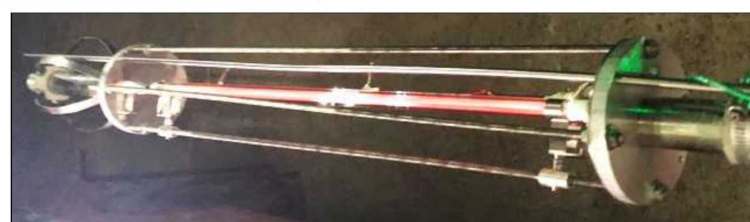
Sandia National Laboratories

BERKELEY LAB

Los Alamos NATIONAL LABORATORY EST. 1943

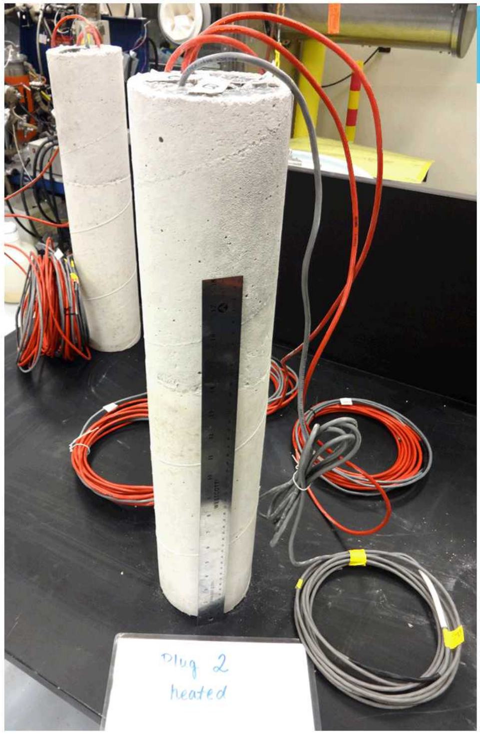


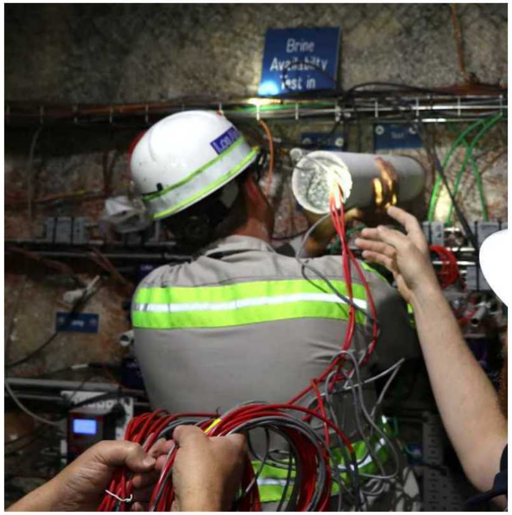




BATS: Cement Seal Borehole

- Two types: modified salt concrete and Sorel cement
  - Align with German programs
  - Complementary with laboratory tests
- Embedded strain gauges during fabricated
- Installed in borehole 1- 2 ft (0.3-0.6 m) from heated borehole
- Strain, temperature, moisture conditions, and brine composition monitored throughout the test
- Will overcore post-test to investigate salt/cement interface
- Understand the behavior of cement/salt/brine system



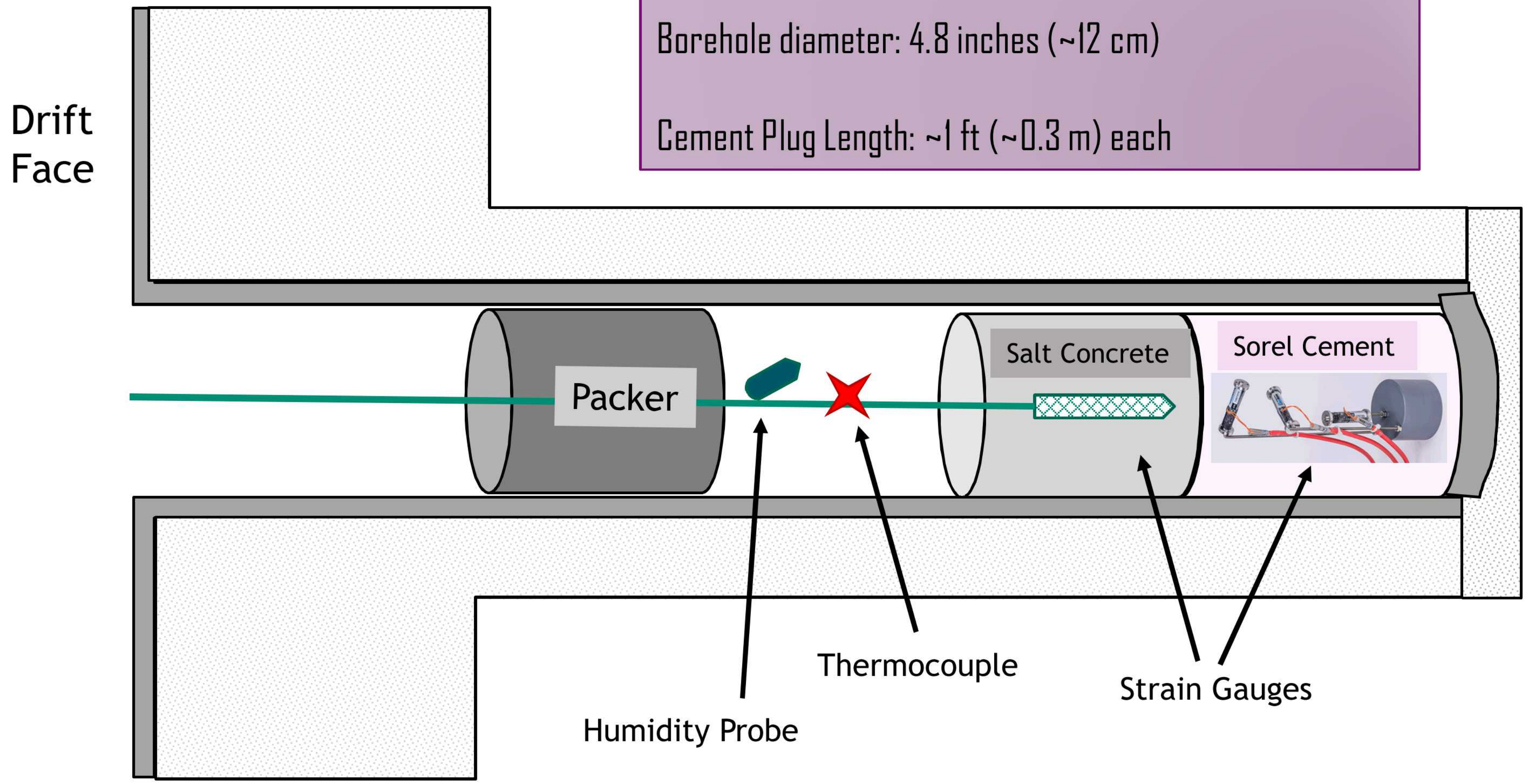


Side Profile View of Cement Seal Borehole

Overall Borehole length: 8 ft (2.4 m)

Borehole diameter: 4.8 inches (~12 cm)

Cement Plug Length: ~1 ft (~0.3 m) each



BATS: Recipes of Emplaced Plugs

Sorel Cement D4 (5-1-8 phase)

- Proportions similar to Popp et al. 2018
  - Density = 2240 kg/m^3
- Desired fast setting
- Used MgO currently emplaced with waste at WIPP
  - Crushed and sieved <75 um
  - reactivity = 272 s
- Aggregate: Run of mine WIPP salt <4 mm grain size

Sorel D4	Composition (mass-(mass-%)
5 M $MgCl_2$	18.3
< 75 um MgO	18.3
Salt Aggregate	63.4

Salt Concrete

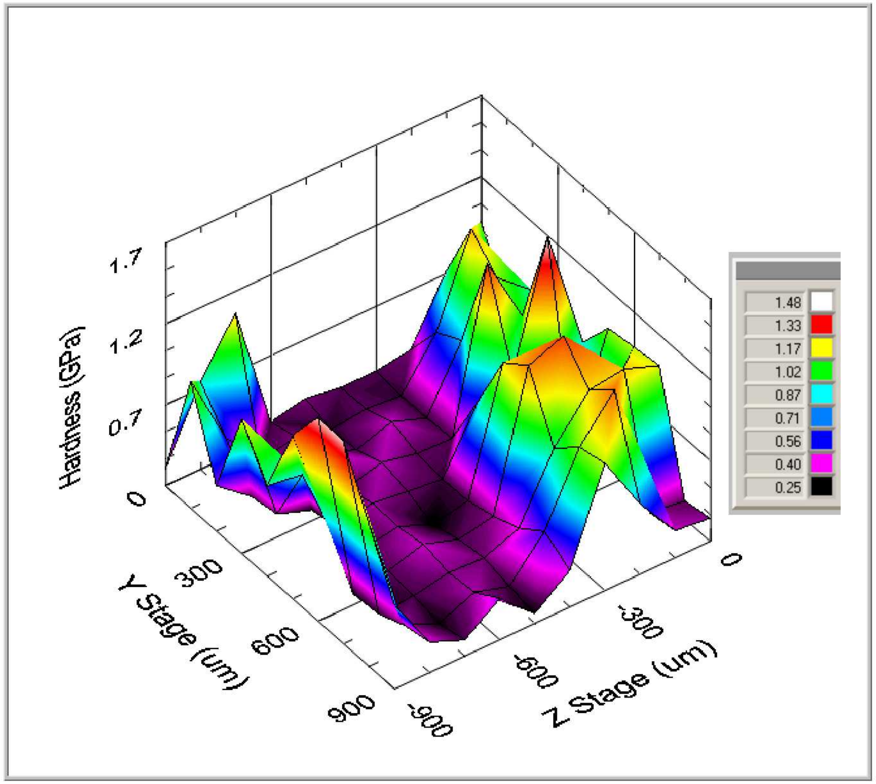
- Followed recipe from Muller-Hoppe et al. 2010 (LAVA2)
- Ground Blast Furnace Slag
- Aggregate: Run of mine WIPP salt <4 mm grain size
  - Impurities affected mixture
- Required to be mixed in glove box

Salt Concrete	Composition (mass-%) %)
Saturated NaCl	14.7
Blast Furnace Slag	28.4
Salt Aggregate	56.9

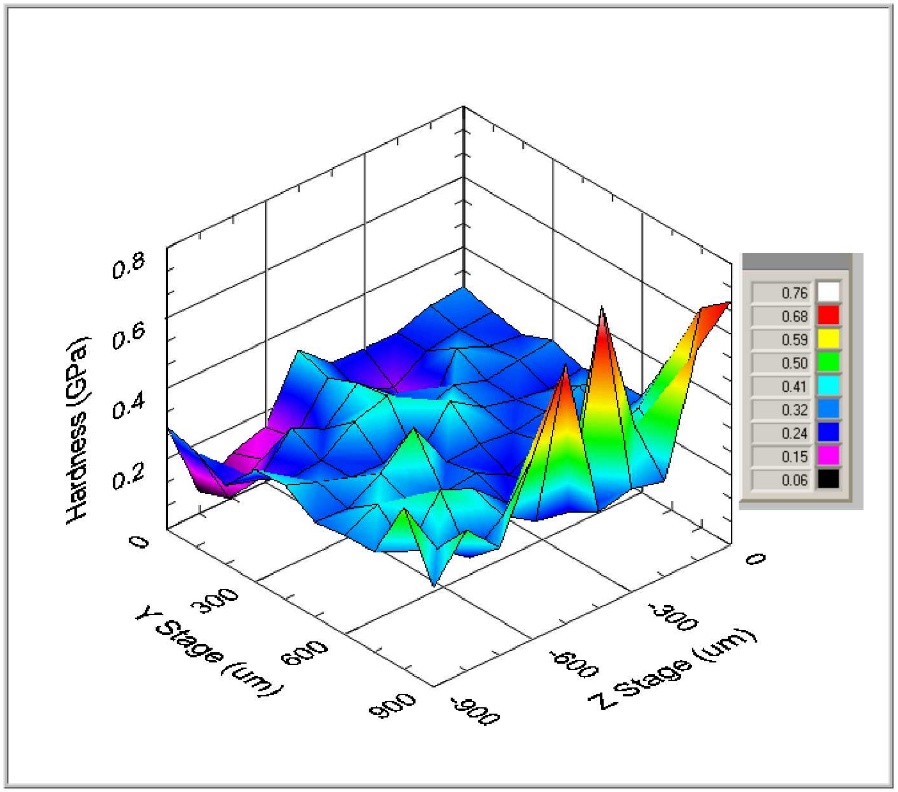
BATS: Characterization of Fabricated Plugs

- Currently analyzing mechanical and chemical properties of sub-samples from installed plugs
- Nano-indentation (250 mN constant load, 10x10 grid, 100 um spacing)

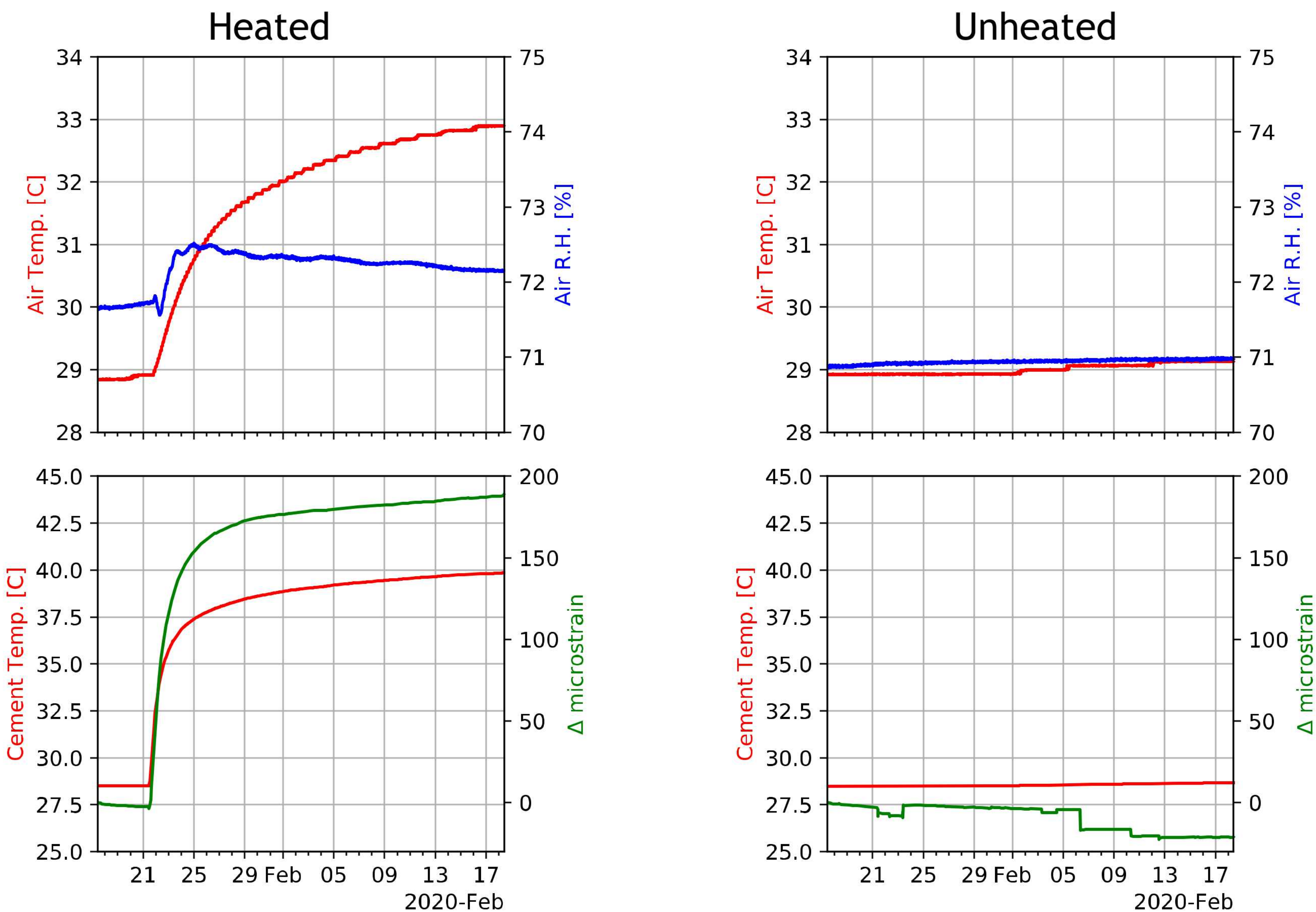
Sorel Cement



Salt Concrete



Preliminary In-Situ Data from Seal Borehole



Future Goals and Endeavors of Cement Seals

- Further investigation into other seal material recipes
- Lab-scale tests
  - Installing into hollowed bedded salt core (similar to DOPAS)
  - Effects of heat
- Analysis of over-cored plug
- Additional plugs deployed in other boreholes at WIPP
  - *In situ* permeability measurements desired

Questions?