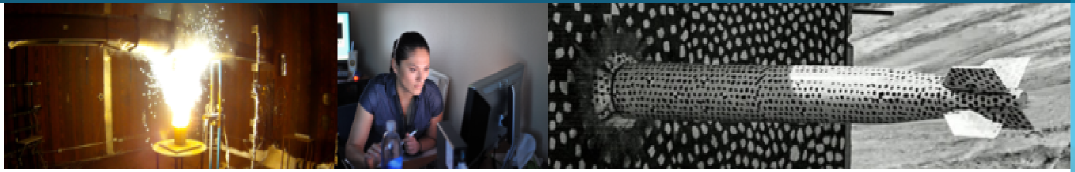
SAND2020-11910PE

Overview of SNL's R&D activities for Geologic Disposal: Argillite Studies



C. F. Jové Colón (SNL)

SAND20-XXXXX

SNL-NUMO Meeting Oct. 28, 2020

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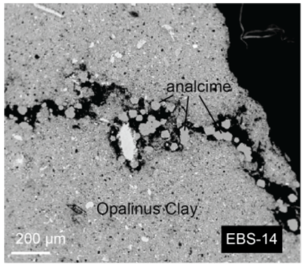
Contributors

C. Payne (SNL), Jessica Kruichak (SNL), Melissa Mills (SNL), A. Knight (SNL), H. Moffat (SNL), E. Coker (SNL), F. Caporuscio (LANL), K. Sauer (LANL), M. Cheshire (ORNL)

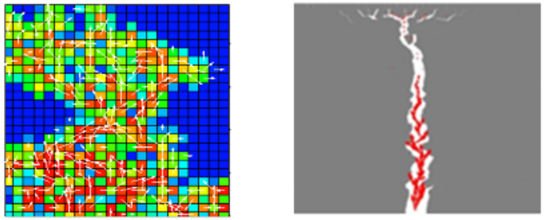
Highlights

- **High temperature** experiments of bentonite interactions with barrier materials and host rocks: granodiorite & Opalinus Clay
- Thermodynamic modeling of bentonite – barrier material interactions & thermodynamic database development
- Advances in THMC modeling approaches of bentonite barrier, argillite rock, and excavated disturbed zone (EDZ fracture/damage behavior) & gas migration
- Development of (non)isothermal 1D-3D THC reactive transport model
- Development of a preliminary GDSA reference case for disposal in argillite media
- International collaborations:
  - FEBEX-DP: bentonite sorption/structural/compositional/thermal studies
  - DECOVALEX Task C: PFLOTRAN HC modeling of barrier interactions

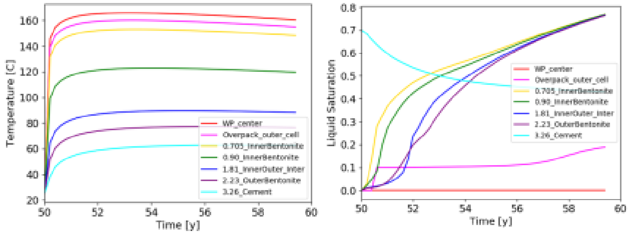


1) Continuum model approach using TOUGH-FLAC

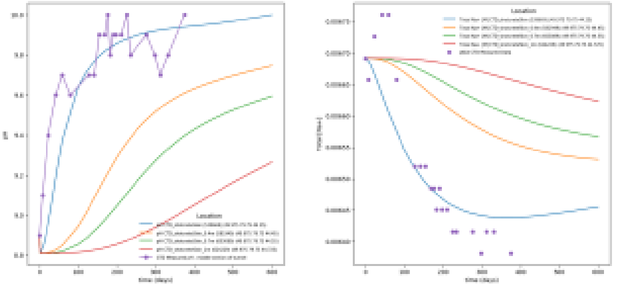
2) Discrete fracture model approach using TOUGH-RBSN



FLOTRAN 3D Single Waste Package



DECOVALEX19 Task C: FLOTRAN 3D HC model



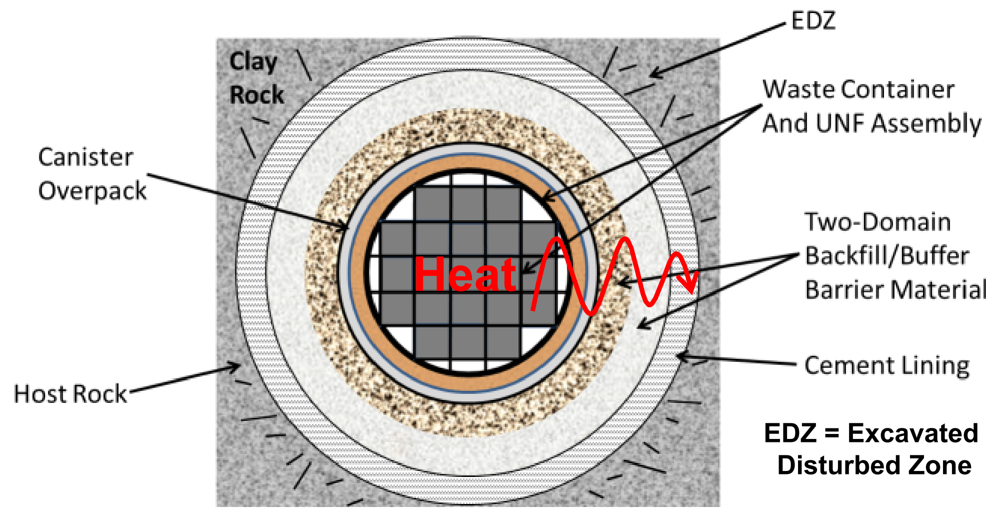
EBS Interactions & Canister Heating

- **Investigate the effects of temperature on bentonite clay barrier interactions:** clay phase change / degradation, smectite swelling, and structure / composition
  - ❖ Dual Purpose Canisters (DPC's) – High capacity canister (up to 37 SNF PWR assemblies); can generate peak T>200°C in disposal scenarios.

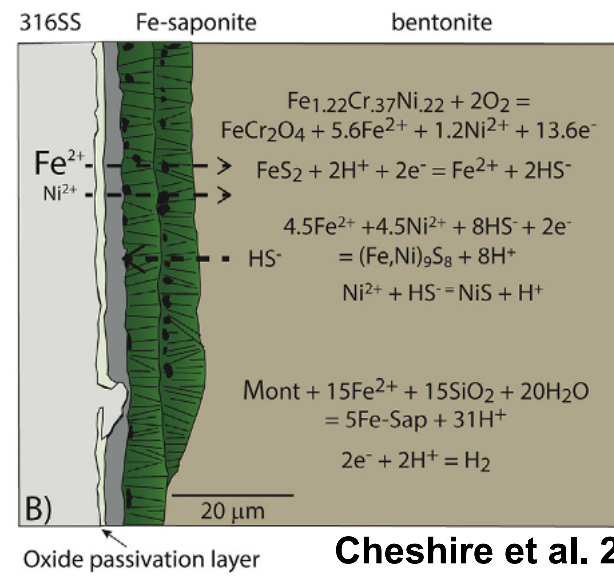
- **Inform fluid-solid chemical models** to assess barrier material interactions at elevated temperatures

- **Investigate effects of clay phase exposure to elevated temperatures** on sorption, diffusion, clay structure (e.g., FEBEX-DP)

- **Improve representation of barrier phase interactions at elevated temperatures** in sub-models that support performance assessment (PA) models for waste repositories, reduce uncertainty



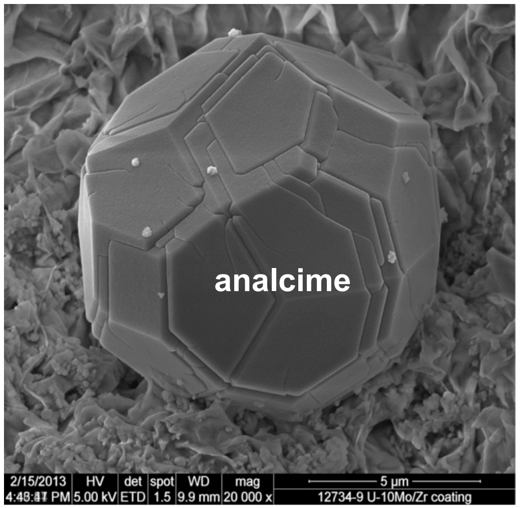
EDZ = Excavated Disturbed Zone

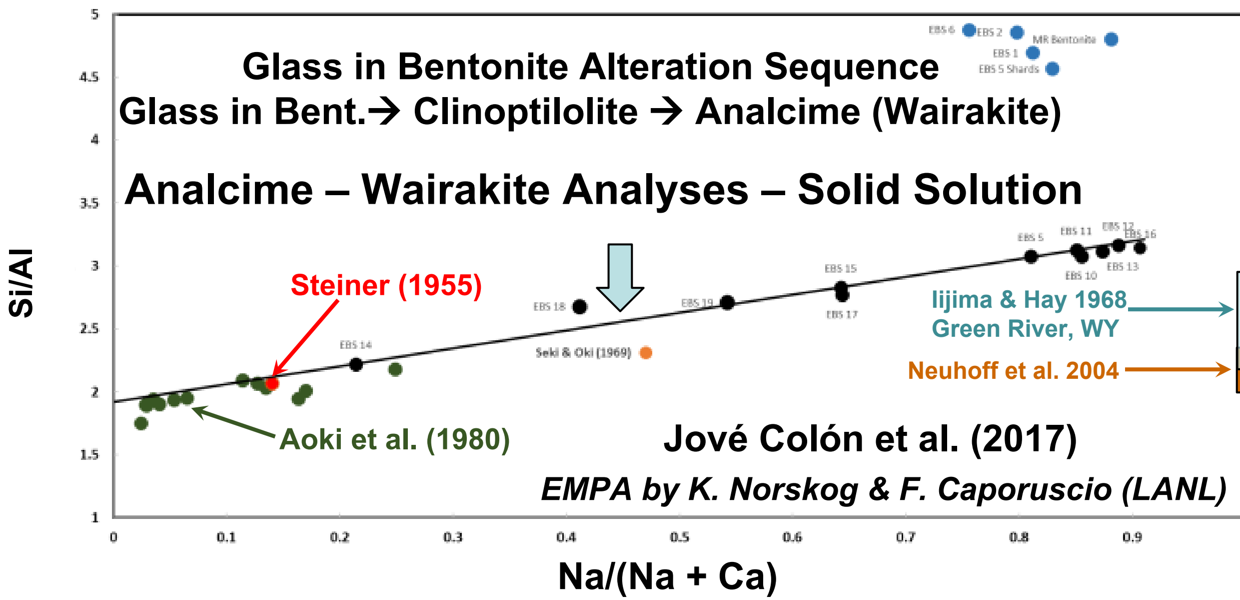


Cheshire et al. 2018

Authigenic zeolite produced from clinoptilolite / glass in bentonite

Analcime (Bentonite only)





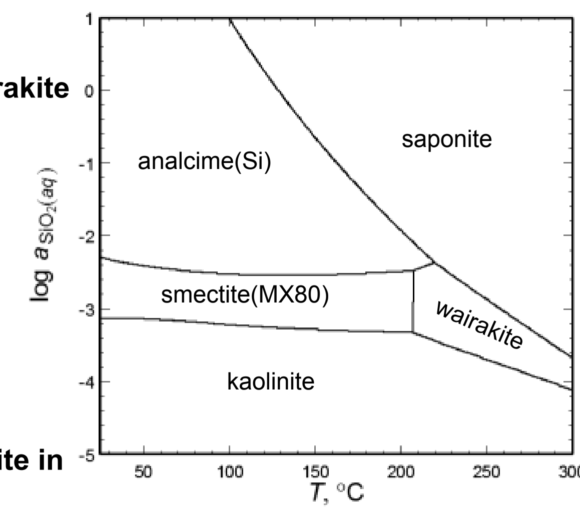
Wairakite-rich zeolite (Opalinus clay + Bentonite)

Bentonite Alteration and Zeolite Stability:

- Glass alteration in bentonite → high Si
- Formation of clinoptilolite, analcime – wairakite zeolites
- Analcime-wairakite solid solution
  - Expands zeolite stability?
- Little or no illite formation
  - Low K in solution
  - High Si in solution

Thermodynamic Analysis:

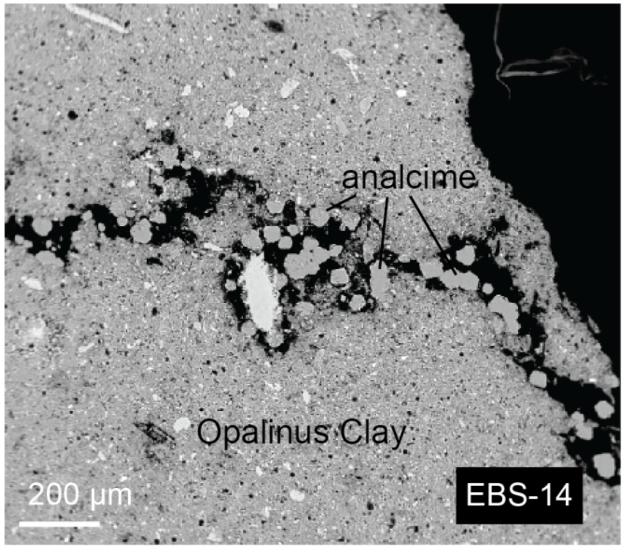
- Clay-zeolite phase equilibria (CHNOSZ)
- Constrain on aqueous activities of clay/zeolite in solution
- NEXT: Reaction path & solid solution modeling

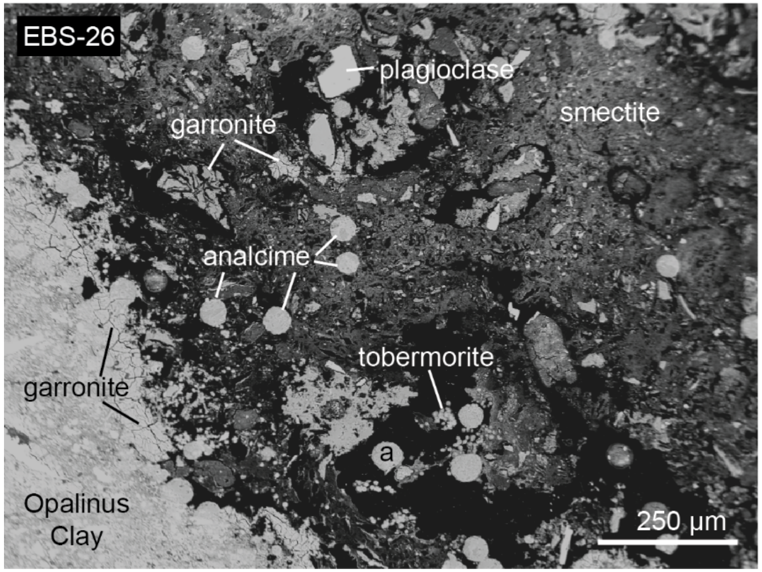


Jové Colón et al. (2017)

Bulk Mineralogy Changes (QXRD)

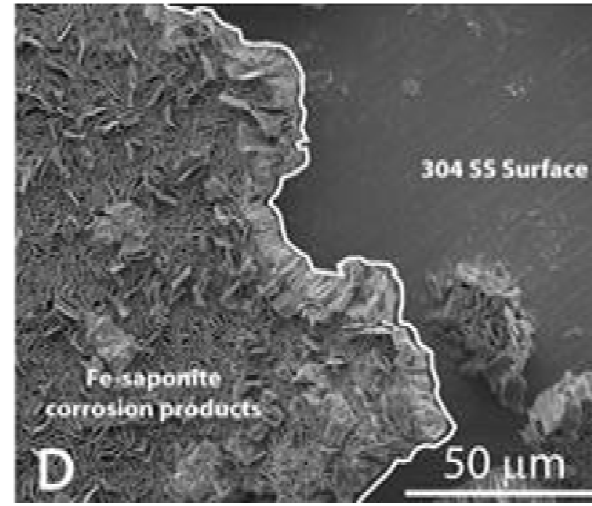
- Opalinus Clay ± Wyoming Bentonite
  - 300°C: Zeolite formation in clay and along cracks and edges on the Opalinus Clay fragments, plagioclase
  - 200°C: No zeolites or feldspar
  - Both: wt.% clay increases
- Opalinus Clay + Wyoming Bentonite + Portland Cement
  - Formation of calcium-silicate-hydrate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

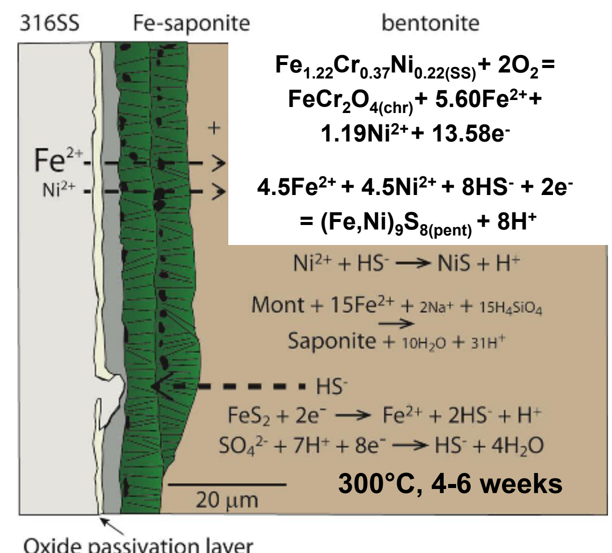




Waste Canister Degradation: 304 & 316L Stainless Steel – Clay Interactions

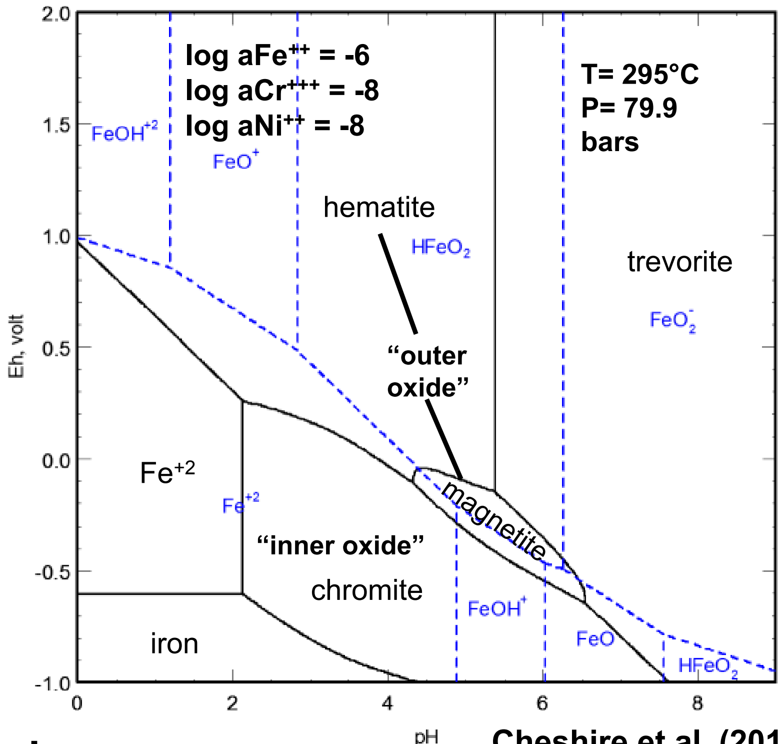
- **Experiment**
  - **T = 300ºC; STRIPA brine**
  - **Wyoming Bentonite**
  - **316 stainless steel (SS), 304 SS, low-C steel, copper**
- **Uniform corrosion – no pitting:**
  - **$Fe_{1.22}Cr_{0.37}Ni_{0.22(SS)} + 2O_2 = FeCr_2O_{4(chr)} + 5.60Fe^{2+} + 1.19Ni^{2+} + 13.58e^-$**
- **Corrosion products**
  - **Chromite passivation layer**
  - **Fe-rich smectite (Fe-saponite growth)**
  - **Chlorite**
  - **Early Pentlandite $(Fe,Ni)_9S_8$ formation**
  - **Millerite (NiS)**
- **316 SS - more extensive passive layer**



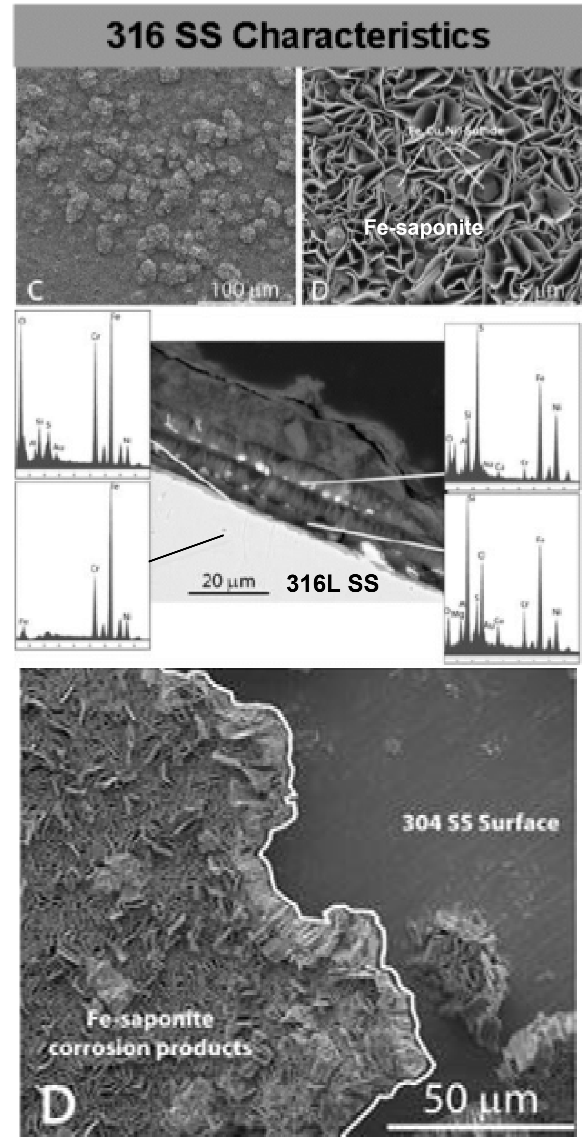


Cheshire et al. 2014, 2018

Waste Canister Degradation: 304 & 316L Stainless Steel – Clay Interactions



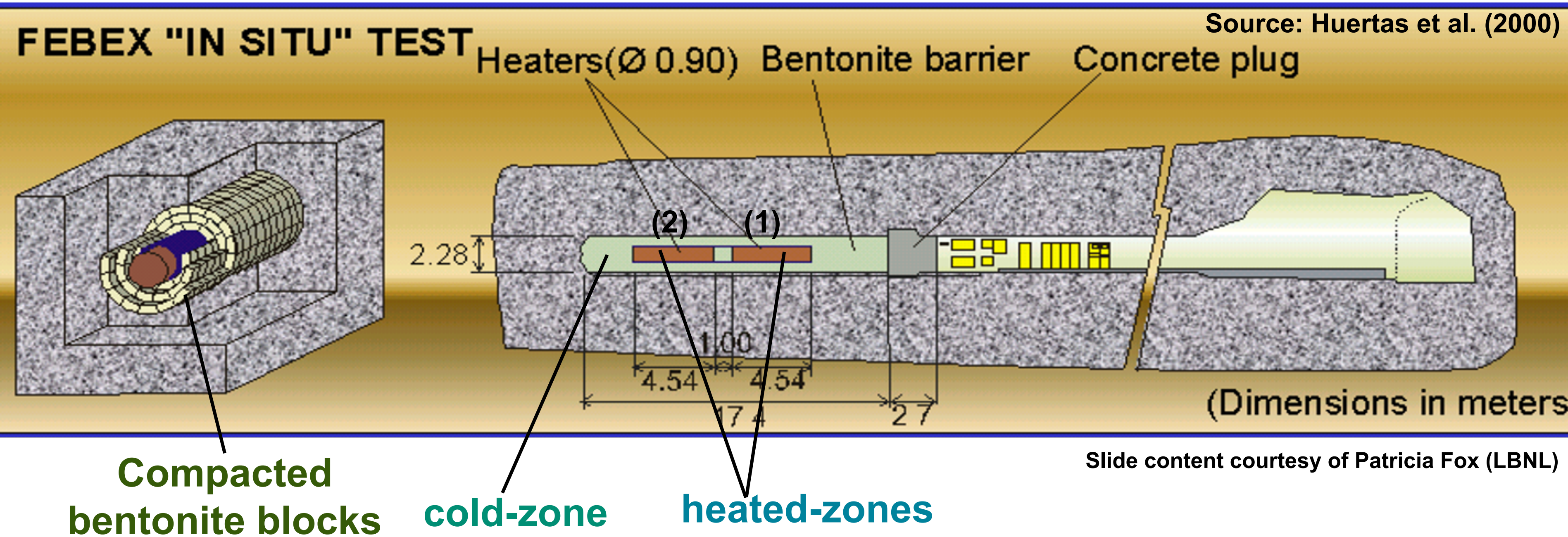
Cheshire et al. (2018)



Cheshire et al. (2014, 2018)

- **Remarks**
  - **Fe-Saponite growth perpendicular to metal substrate**
  - **S is generated from pyrite degradation in bentonite**
  - **Concurrent surface sulfide precipitation with Fe-saponite**

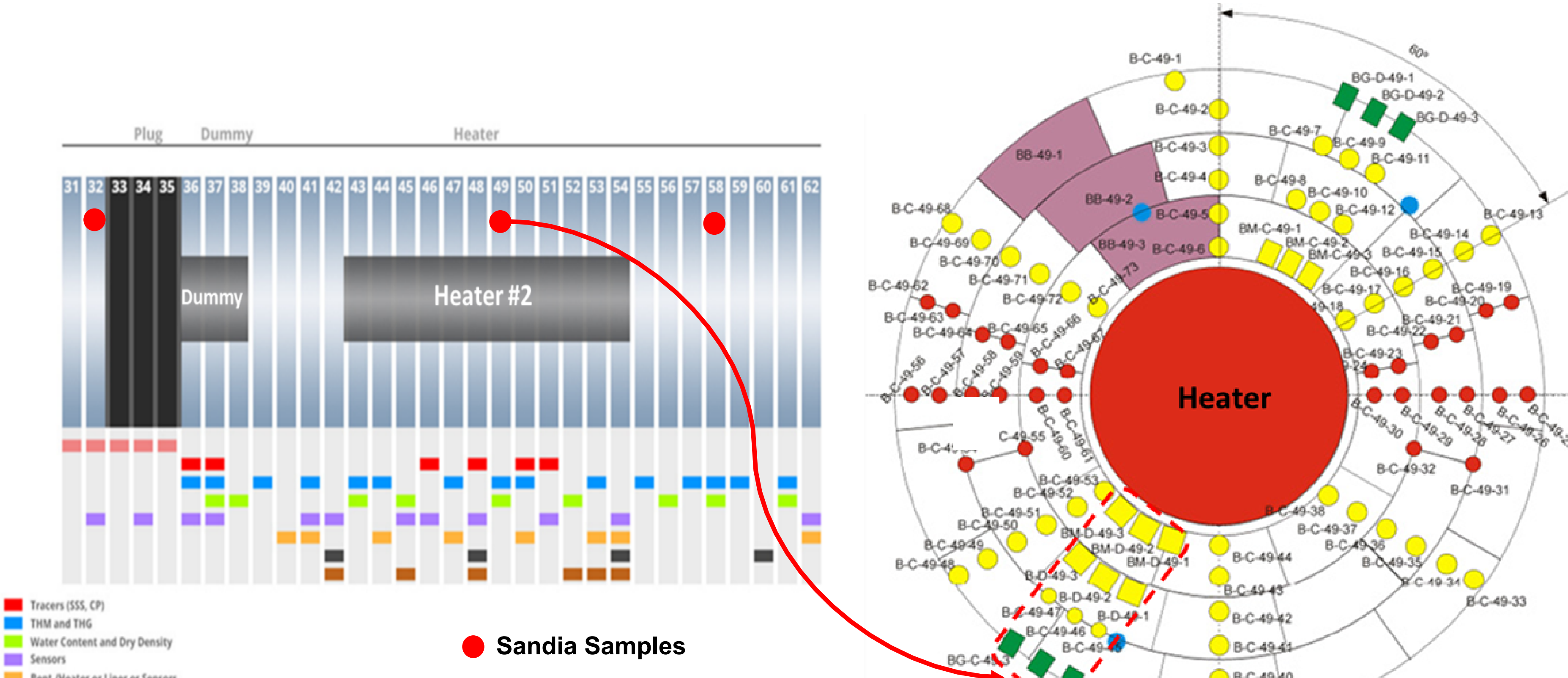
FEBEX Full Scale Heater Test Experiment



Slide content courtesy of Patricia Fox (LBNL)

- **Conducted by ENRESA under auspices of the EU at the Grimsel Test Site (GTS) in Switzerland**
- **Bentonite was compacted into blocks at 1650 kg/m^3 dry density and placed in a radial arrangement surrounding 2 heaters**
- **Heaters operated at a maximum of 100 °C – Heater 1 operated for 5 years; heater 2 operated for 18 years**
- **FEBEX-DP samples were obtained from heater 2 dismantling in 2015 after 18 years of heating**
- **Unique opportunity for long-term full-scale heater test and sample / data availability**

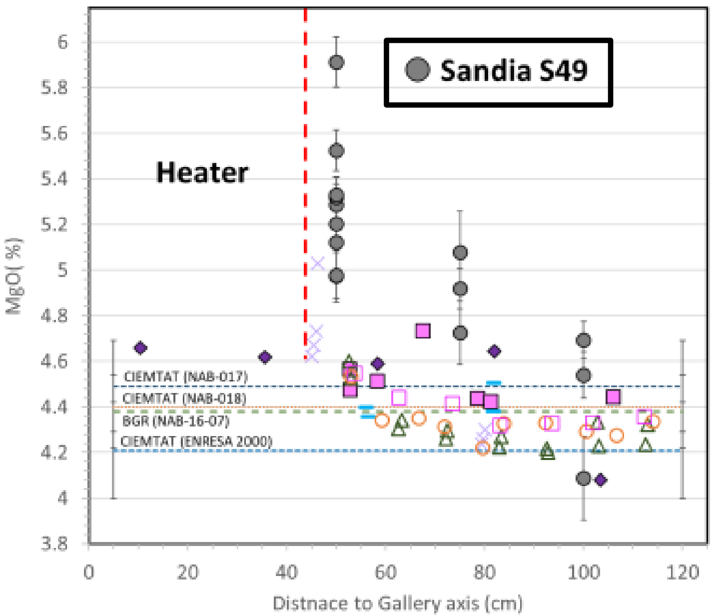
FEBEX-DP Experiment: Sampled Sections

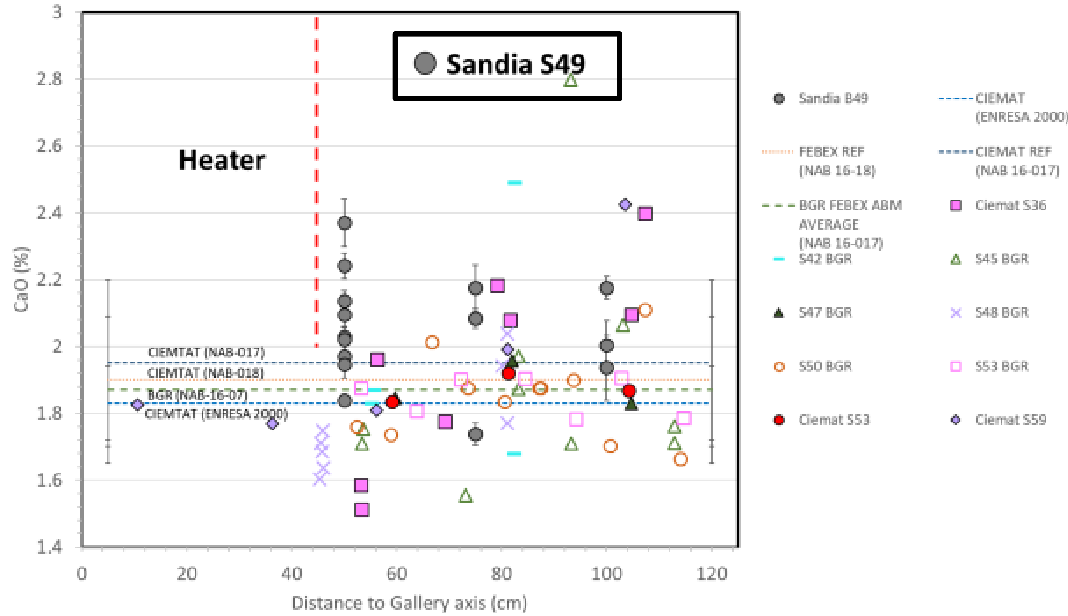


X-Ray Fluorescence (XRF) bulk composition, X-ray CT-scan, μ-XRF, SEM-EDS, X-Ray Diffraction (XRD), Thermogravimetric analysis (TGA)

Section 49

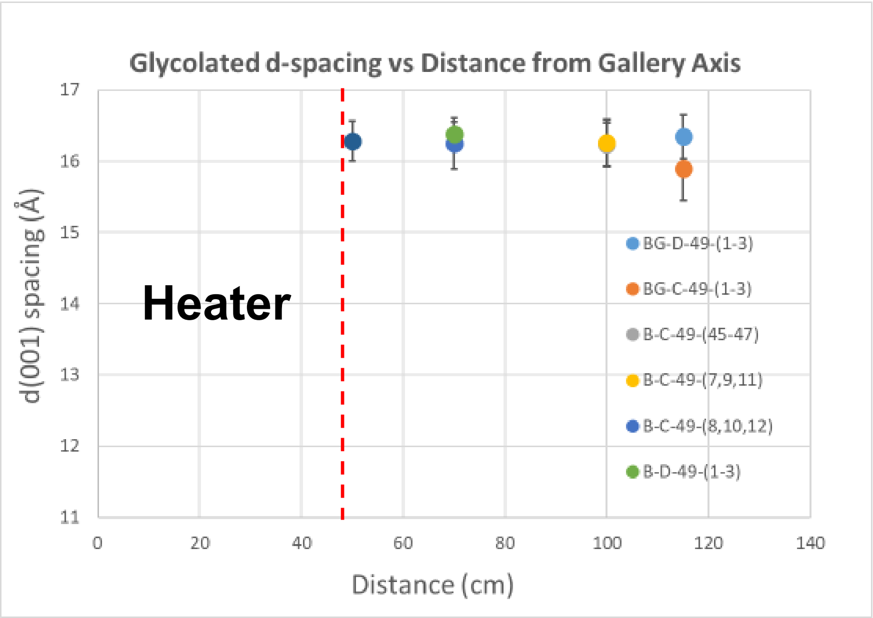
FEBEX-DP Bulk Bentonite Samples: X-ray Fluorescence (XRF)

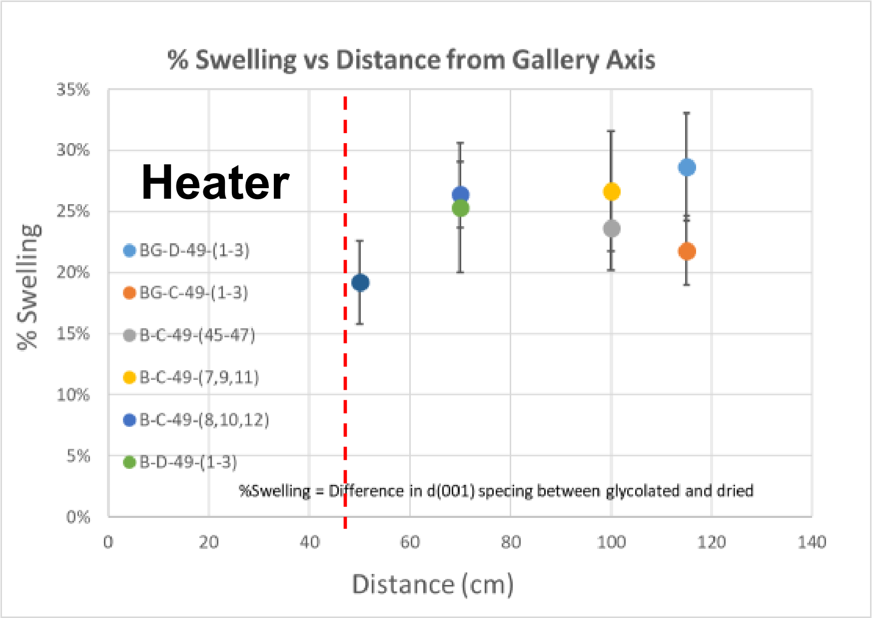




- Mg enrichment towards the heater surface – zones of increasing dry out conditions
- Bulk MgO content far from heater nominally within the bounds of other lab analyses
- Overall, CaO content is relatively variable close to the heater surface
- Mg enrichment(?):
  - Enhanced Mg content due to elevated temperatures?
  - SEM-EDS didn't reveal newly-formed Mg-bearing phases within the clay matrix

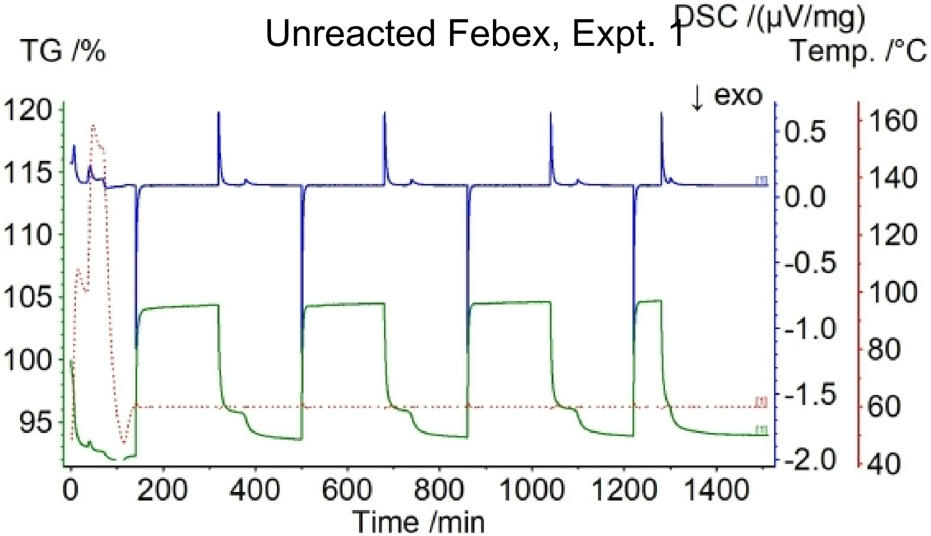
FEBEX-DP: Bentonite X-ray Diffraction (XRD)

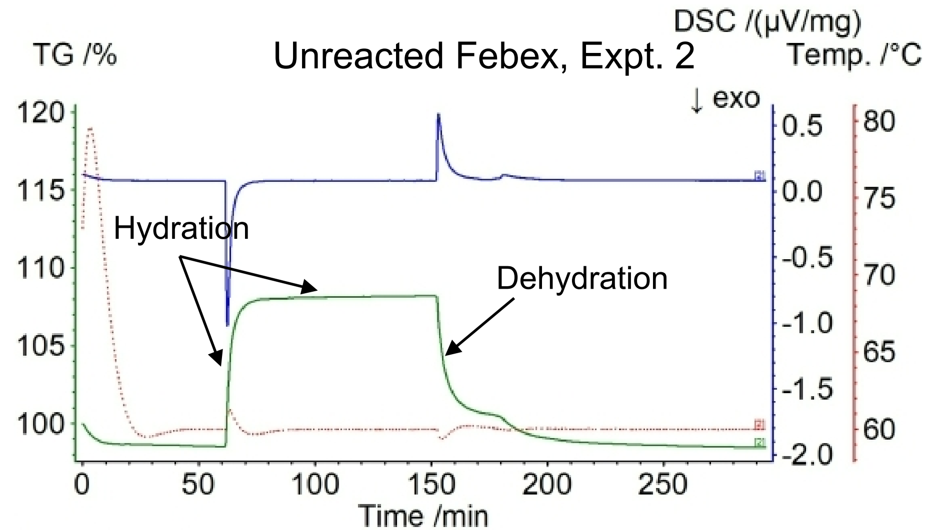




- No apparent effect of elevated temperatures on d(001) spacing for glycolated clay samples
- Slight decrease in swelling extent for samples in contact or close to the heater surface
- Prolonged exposure of bentonite to T = 95 – 100 ºC causes some changes in swelling
  - **Correlate with compositional changes in clay close to heater surface**

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

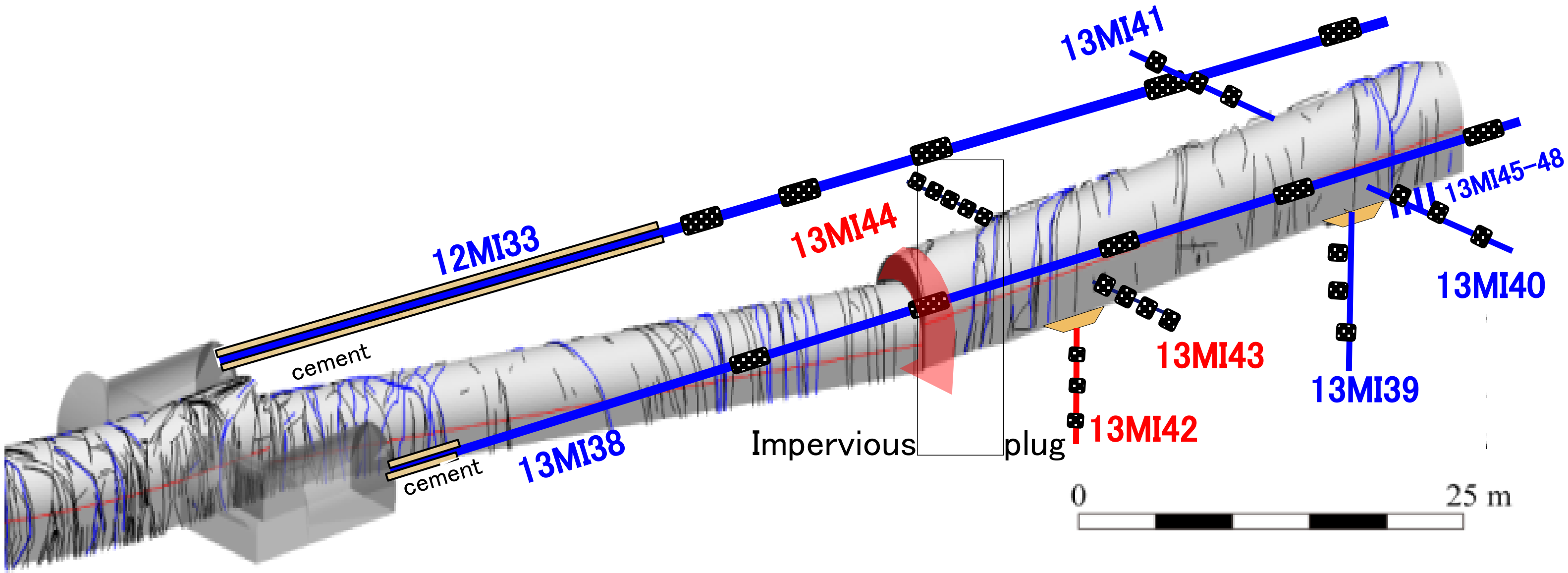




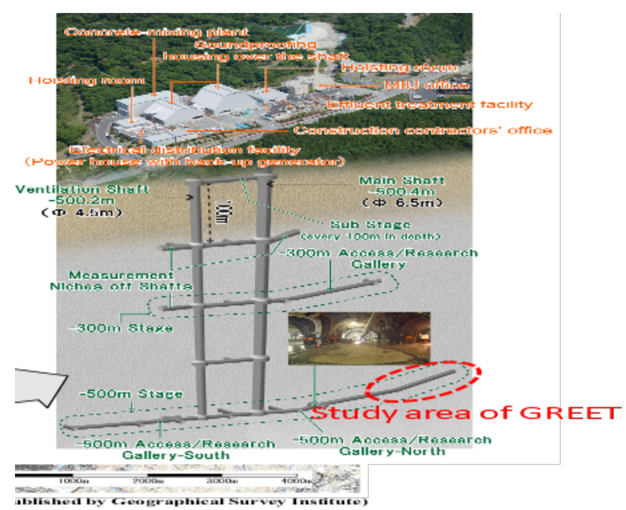
Expt. 1
1) Sample dehydrated under dry N_2 at 150 °C, then cooled to 60 °C.
2) 50 % RH N_2 for 180 min.
3) Dry N_2 for 180 min.
4) Steps 2 and 3 repeated twice.
5) 60 % RH N_2 for 60 min.
6) Dry N_2 for 240 min.

Expt. 2
Sample after expt. 1 subjected to:
1) Dry N_2, 60 °C, 60 min
2) 60% RH N_2, 60 °C, 90 min
3) Dry N_2, 60 °C, 150 min

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

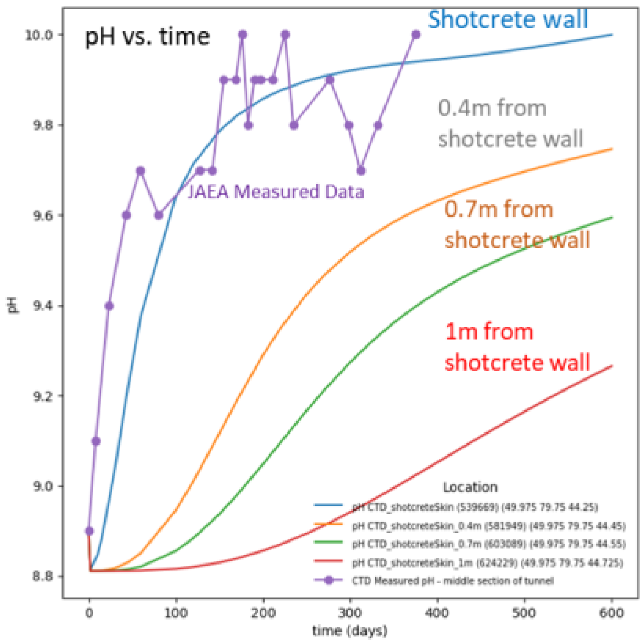


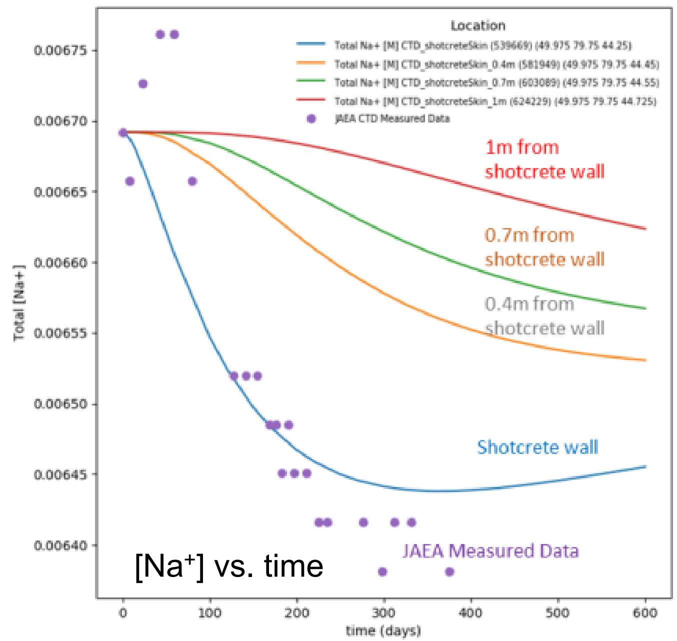
Schematic figure courtesy of Dr. Teruki Iwatsuki (JAEA)

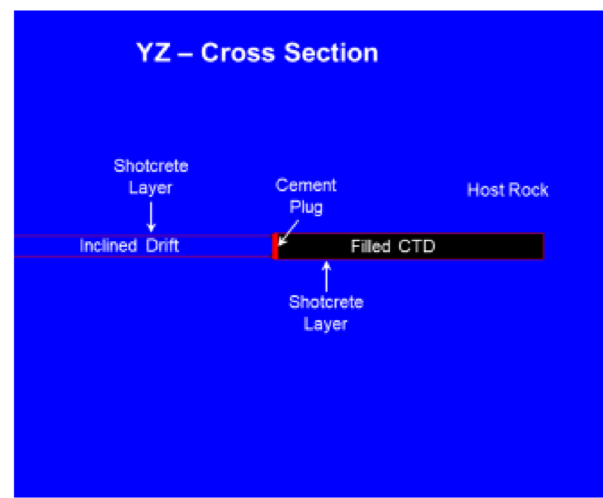


- 3D Reactive Transport Simulations using PFLOTRAN simulation code
- Focus: Shotcrete – groundwater interactions in the CTD

DECOVALEX19 Task C: 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 TST rate law parameters for various cement phases and volume fraction of mineral components
- More simulations have been conducted to evaluate shotcrete thickness effects

Backup Slides

1. Introduction, Purpose, and Context

2. Safety Strategy

2.1 Management Strategy

a. Organizational/mgmt. structure
b. Safety culture & QA
c. Planning and Work Control
d. Knowledge management
e. Oversight groups

2.2 Siting & Design Strategy

a. National laws
b. Site selection basis & robustness
c. Design requirements
d. Disposal concepts
e. Intergenerational equity

2.3 Assessment Strategy

a. Regulations and rules
b. Performance goals/safety criteria
c. Safety functions/multiple barriers
d. Uncertainty characterization
e. RD&D prioritization guidance

3. Technical Bases

3.1 Site Selection

a. Siting methodology
b. Repository concept selection
c. FEPs Identification
d. Technology development
e. Transportation considerations
f. Integration with storage facilities

3.2 Pre-closure Basis

a. Repository design & layout
b. Waste package design
c. Construction requirements & schedule
d. Operations & surface facility
e. Waste acceptance criteria
f. Impact of pre-closure activities on post-closure

3.3 Post-closure Bases (FEPs)

3.3.1 Waste & Engineered Barriers Technical Basis

a. Inventory characterization
b. WF/WP technical basis
c. Buffer/backfill technical basis
d. Shafts/seals technical basis
e. UQ (aleatory, epistemic)

3.3.2 Geosphere/ Natural Barriers Technical Basis

a. Site characterization
b. Host rock/DRZ technical basis
c. Aquifer/other geologic units technical basis
d. UQ (aleatory, epistemic)

3.3.3 Biosphere Technical Basis

a. Biosphere & surface environment:
- Surface environment
- Flora & fauna
- Human behavior

4. Disposal System Safety Evaluation

4.1 Pre-closure Safety Analysis

a. Surface facilities and packaging
b. Mining and drilling
c. Underground transfer and handling
d. Emplacement operations
e. Design basis events & probabilities
f. Pre-closure model/software validation
g. Criticality analyses
h. Dose/consequence analyses

4.2 Post-closure Safety Assessment

a. FEPs analysis/screening
b. Scenario construction/screening
c. PA model/software validation
d. Barrier/safety function analyses and subsystem analyses
e. PA and Process Model Analyses/Results
f. Uncertainty characterization and analysis
g. Sensitivity analyses

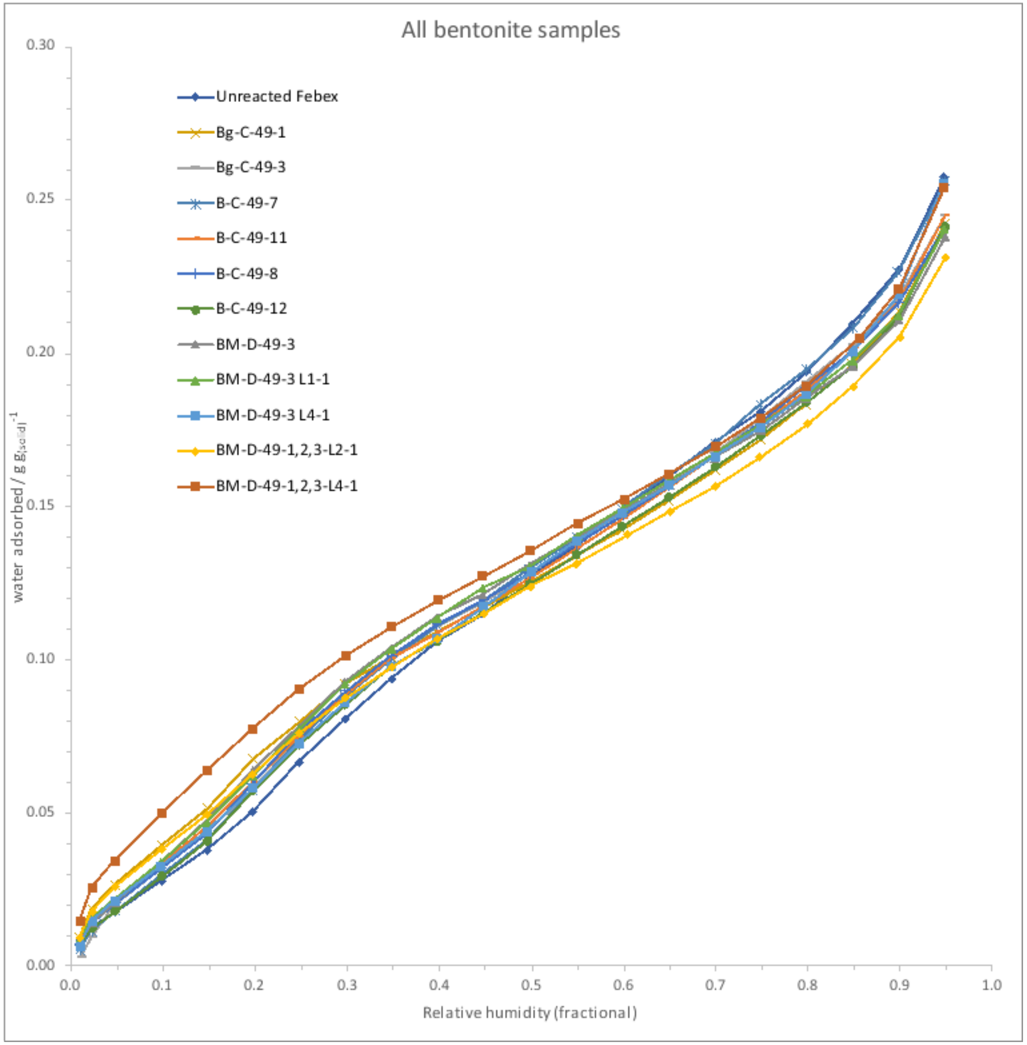
4.3 Confidence Enhancement

a. R&D prioritization
b. Natural/anthropogenic analogues
c. URL & large-scale demonstrations
d. Monitoring and performance confirmation
e. International consensus & peer review
f. Verification, validation, transparency
g. Qualitative and robustness arguments

5. Synthesis & Conclusions

a. Key findings and statement(s) of confidence
b. Discussion/disposition of remaining uncertainties
c. Path forward

FEBEX-DP: Volumetric Sorption



- All FEBEX-DP bentonite samples showed similar isotherms, except “BM-D-49-1,2,3 Location 4-1” which has slightly higher water uptake than others below 60% RH.
- H_2O capacity at 95% RH and 20 °C ranges from 0.23 to 0.26 $g_{(water)}\ g_{(solid)}^{-1}$

Summary

- Bentonite-metal interactions & 3D THC modeling at elevated temperatures:
  - Produces zeolites (analcime) and sulfide phases with Fe-saponite growth perpendicular to metal substrate
  - Little or no illite forms in the experiments and URL heater tests
  - Thermodynamic analysis of clay-metal and clay-zeolite equilibria is consistent with experimental observations
  - Advances in non-isothermal 3D modeling of waste package & EBS
  - Future Work: Study effects of host rock composition & other barrier materials (e.g. cement); expand 3D non-isothermal model to various waste packages
- *Post mortem* FEBEX-DP bentonite studies & DECOVALEX Task C HC modeling:
  - Slight decrease in bentonite swelling correlates with Mg-enrichment in clay close to the heated surface
  - Thermal analyses under controlled hydration/dehydration show no significant differences between samples close to and far from the heater surface
  - Future Work: Exploit cyclic thermal analyses & XRD methods to evaluate high T effects;
- UO_2 / metal corrosion modeling & thermodynamic data generation for UO_2 corrosion products
  - Progress in FMDM & Cantera/Zuzax electrochemical model development for UO_2/metal corrosion
  - Future Work: Applications to wasteform interactions and in-package chemistry modeling; use 1st principle approaches in the generation of thermodynamic data of UO_2 corrosion products (metaschoepite)