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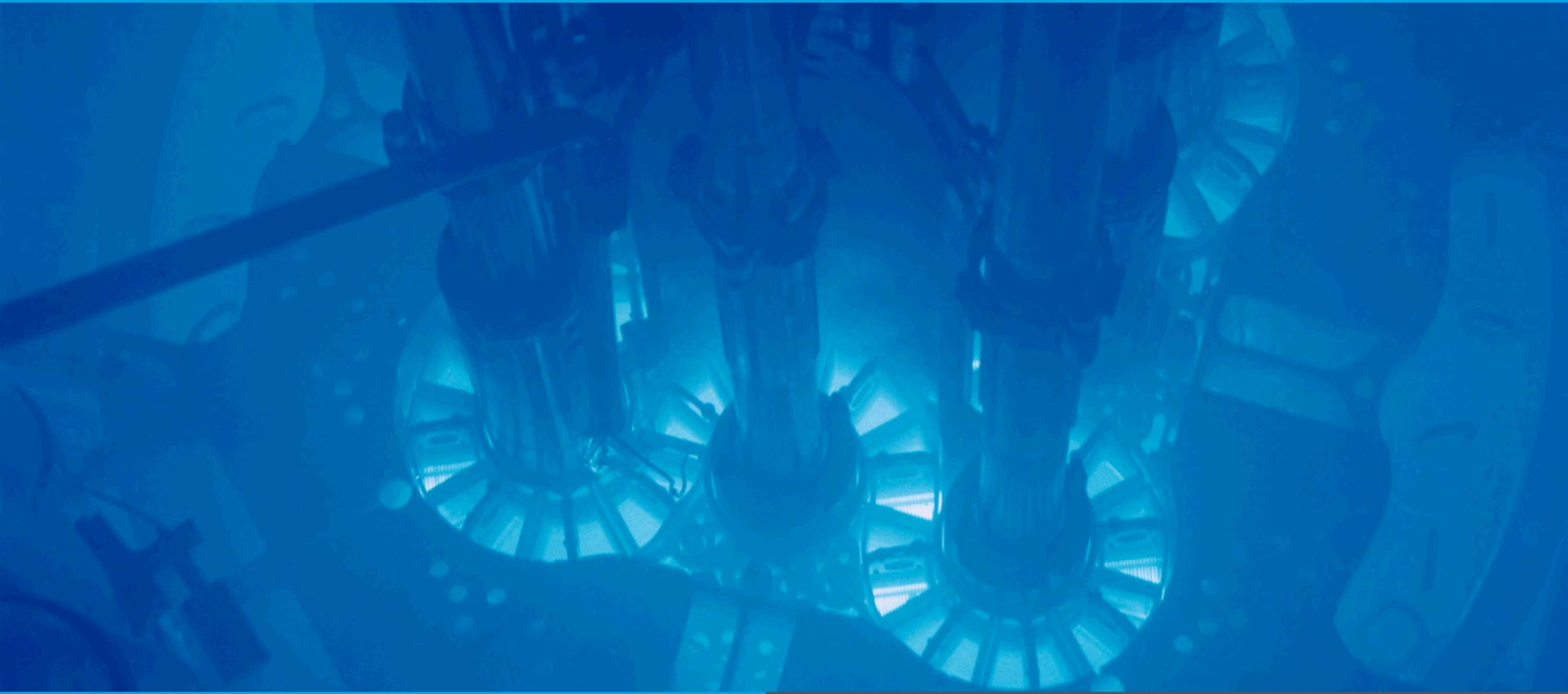
SAND2019-3793PE



Thermal Implications on Transport in Bentonite: Using Full-Scale Engineered Barrier Experiment-Dismantling Project (FEBEX-DP) Samples for Laboratory Studies and Model Testing

U.S. Nuclear Waste Technical Review Board Spring Meeting
Burlingame, California
April 24-25, 2019

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Disposal in Argillite SFWST Work Package

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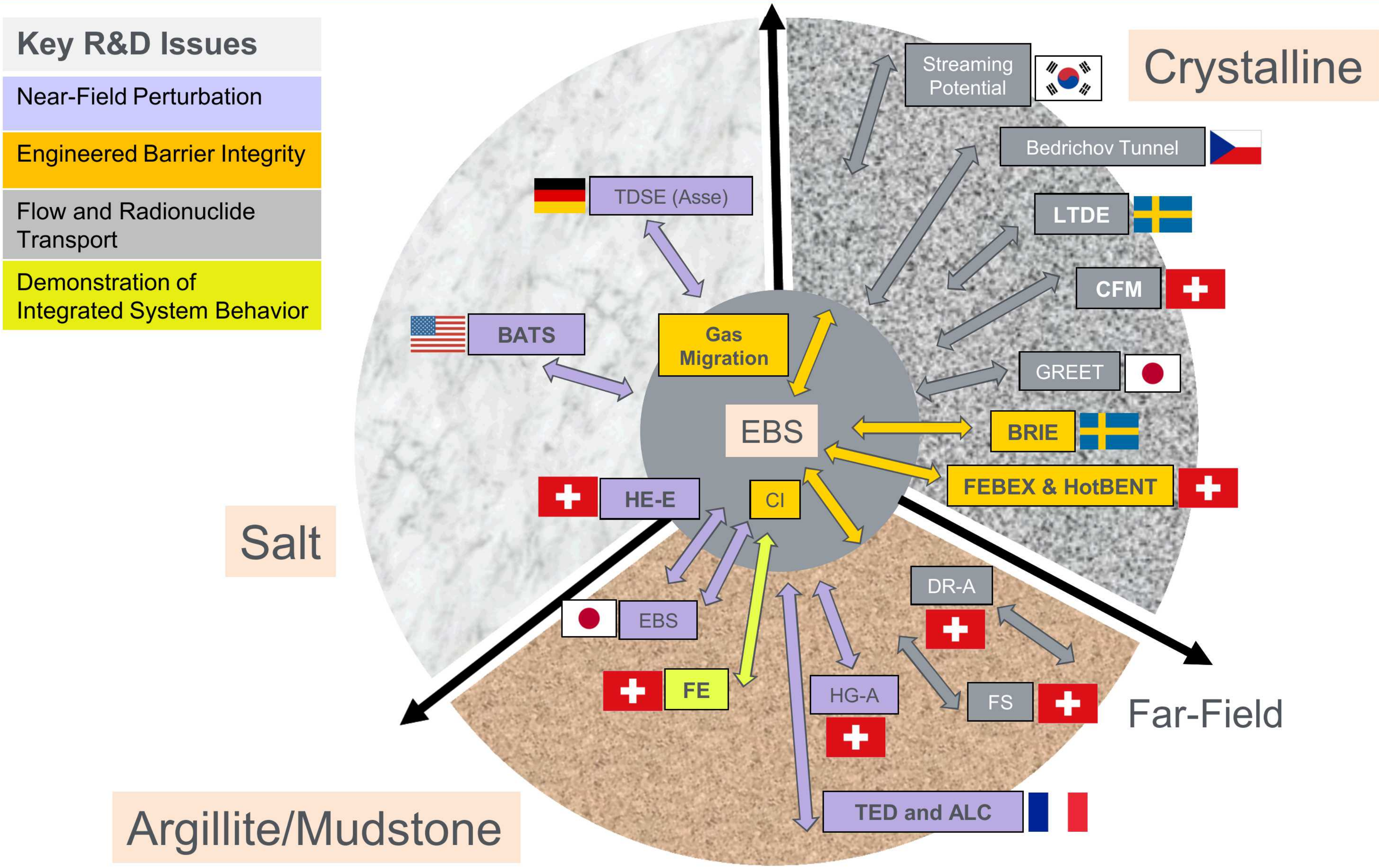
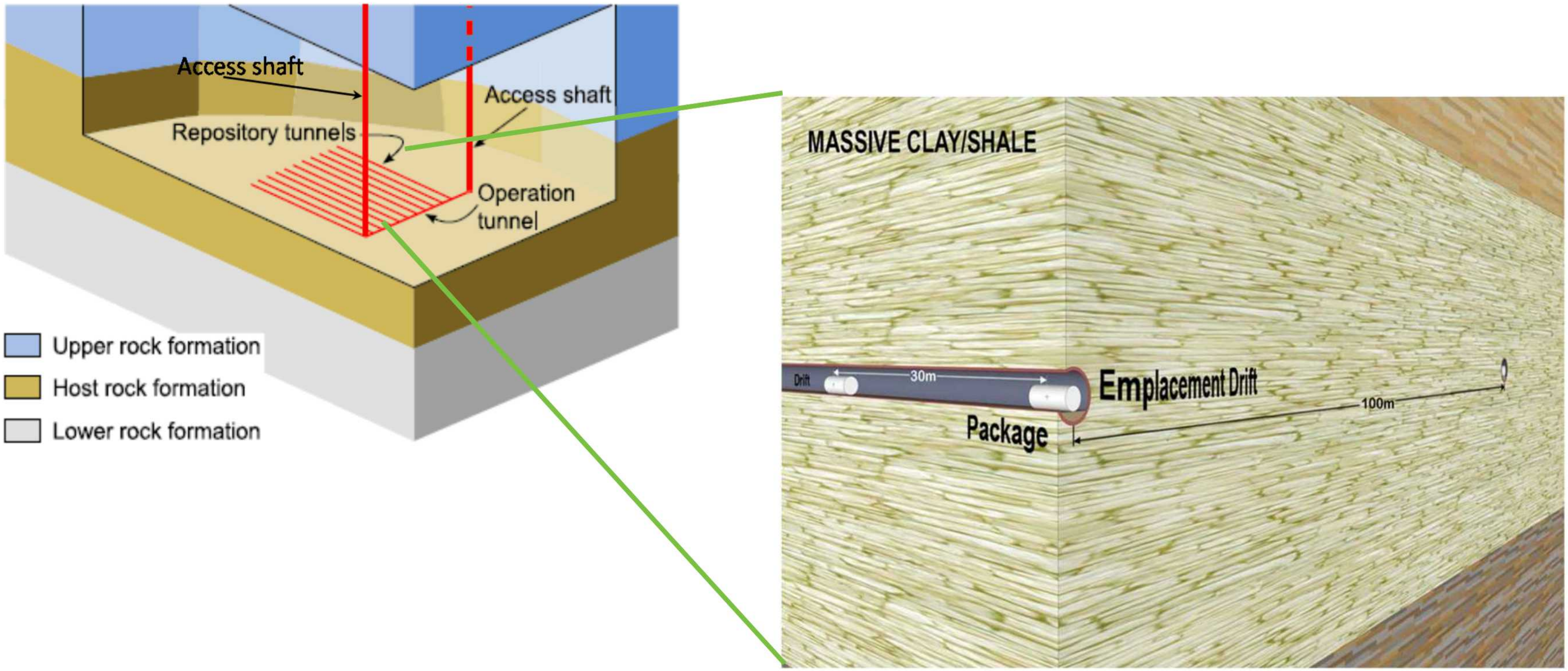
International URL Portfolio in a Nutshell



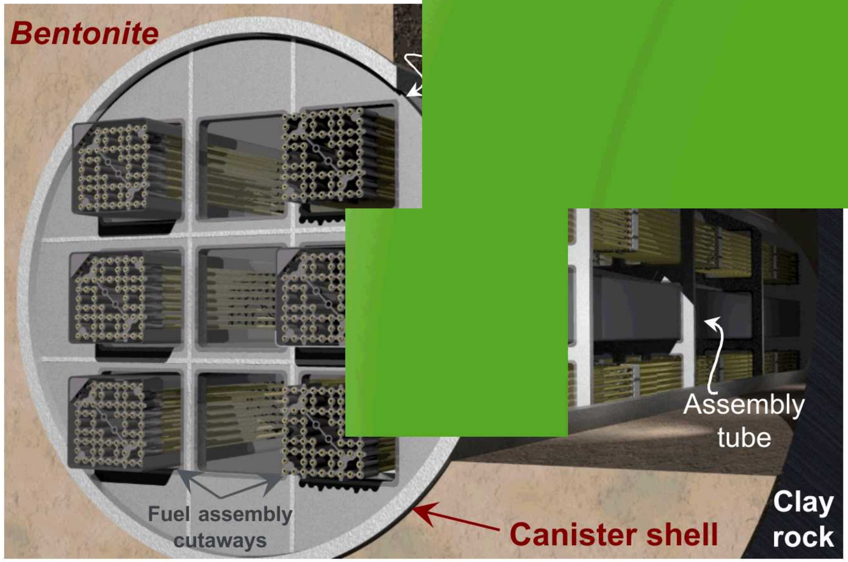
Spent Fuel Waste Science Technology (SFWST) Campaign: Disposal in Argillaceous Clay Rock

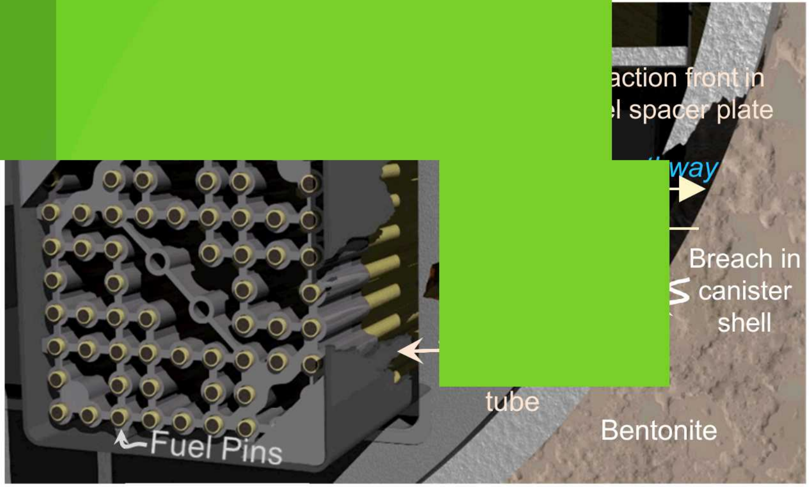


- Coupled process modeling of Thermal-Hydrological-Chemical-Mechanical (THCM) with decay heat effects
- Engineered barrier system (EBS) model integration with performance assessment (PA)
- Thermodynamic modeling of barrier material interactions (**clay, cement, metal) and thermodynamic database (TDB) development**
- Clay interaction experiments:
  - **High temperature mineral phase stability, clay – metal interactions (waste package material (steel) corrosion)**
  - **Low temperature radionuclide (RN) sorption/diffusion in bentonite & modeling**

Conceptual Model for Transport in Bentonite

Slide content: Jim Jerden (ANL) and Ruth Tinnacher (CSU)





- ⇨ **Bentonite (montmorillonite)** is the proposed backfill material in the EBS.
- ⇨ **Diffusion –** dominant transport mechanism in the EBS
- ⇨ **Temperature and chemical gradients (e.g., solution chemistry)** are expected over time and across the EBS.

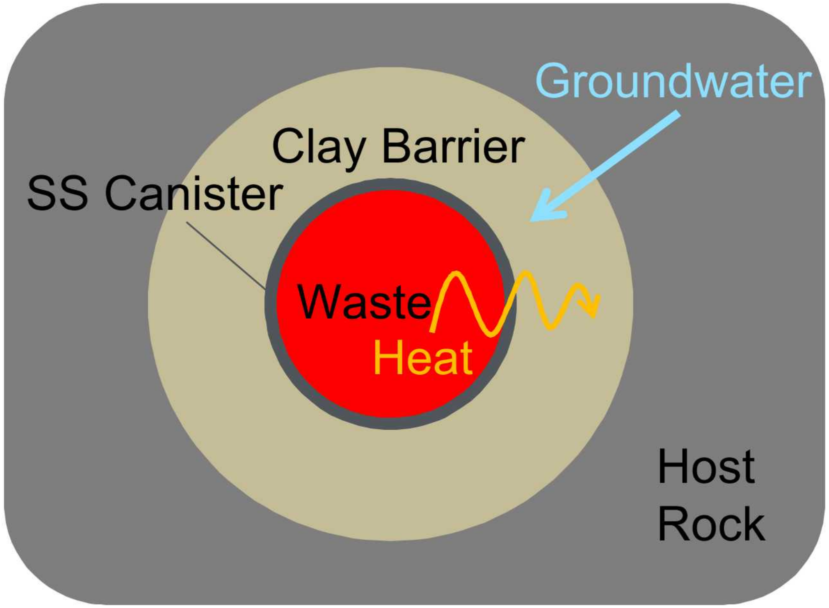
Goals:

- **Investigate the effects of temperature on bentonite clay barrier interactions:** clay phase change / degradation, smectite swelling, and structure / composition
- **Investigate the effects of changing chemical conditions and temperatures** on uranium(VI) sorption and diffusion.
- **Reduce the uncertainty** in actinide sorption / diffusion sub-models that are part of performance assessment (PA) models for waste repositories.

Understanding radionuclide adsorption to clay under realistic waste-disposal scenarios

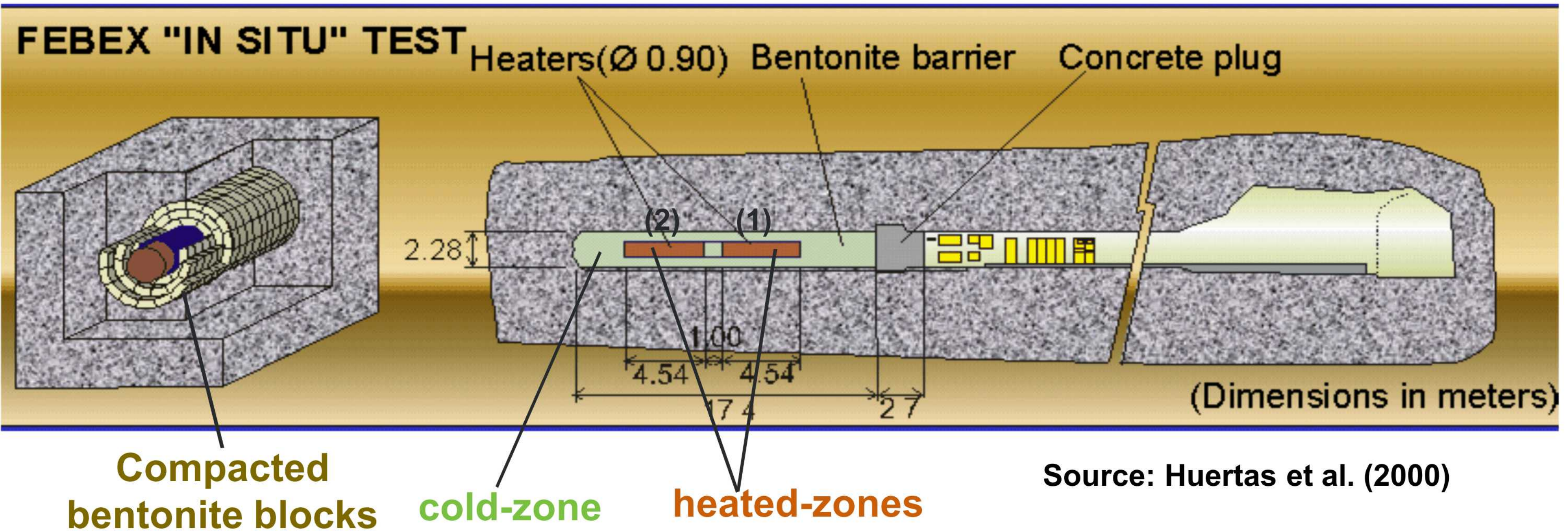
- Heat-generating waste canisters increase temperatures of surrounding engineered barriers
- Groundwater Intrusion from surrounding host rock

- Variable saturation across clay barrier
- Changes in pore water chemistry
- Changes in accessory mineral assemblage (e.g., calcite, ferrihydrite)
- Changes in clay structure/composition (e.g., illitization, ion exchange)



- Changes in aqueous radionuclide (RN) speciation
- Changes in mineral sorption capacity
- Changes in swelling behavior

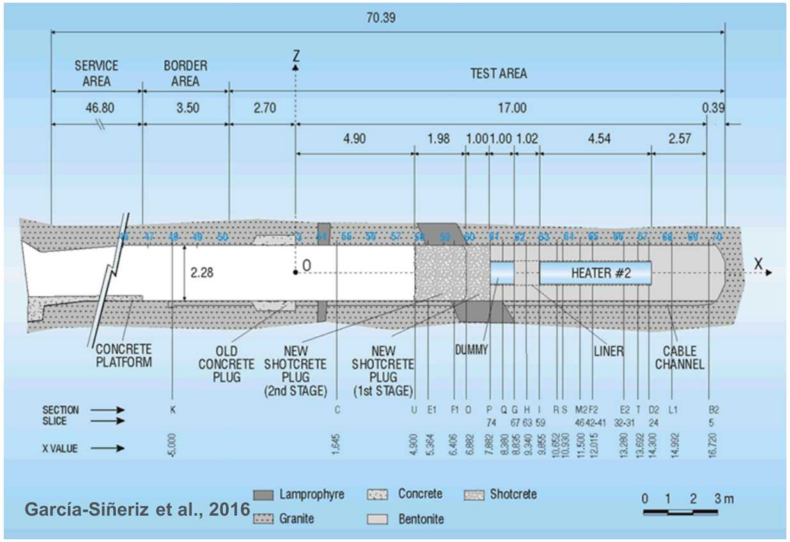
FEBEX Full Scale Heater Test Experiment



Source: Huertas et al. (2000)

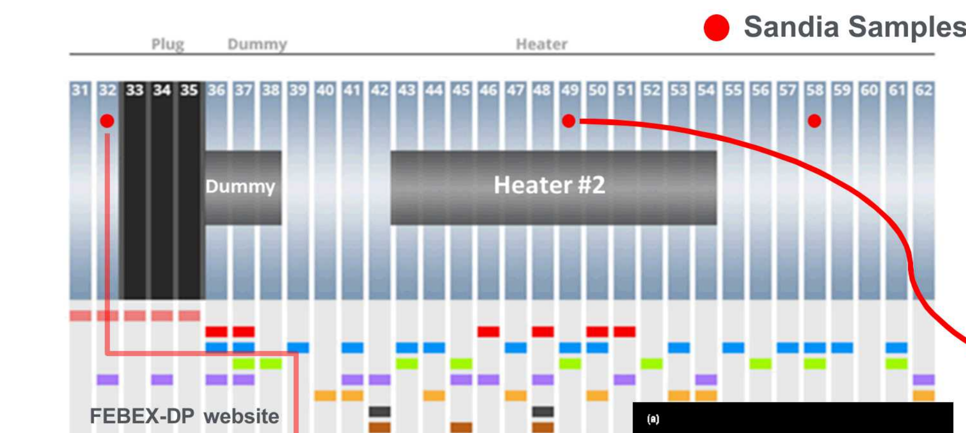
- Conducted by ENRESA under auspices of the EU at the Grimsel Test Site (GTS) in Switzerland
- Bentonite was compacted into blocks at 1650 kg/m^3 dry density and placed in a radial arrangement surrounding 2 heaters
- Heaters operated at a maximum of 100 °C – Heater 1 operated for 5 years; heater 2 operated 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

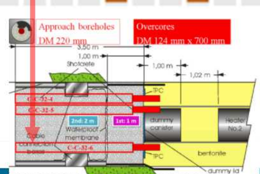


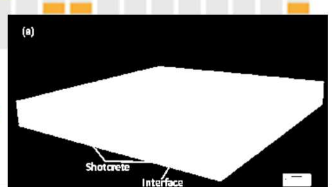
García-Siñeriz et al., 2016

- **FEBEX-DP**
  - **<u>Section 49</u> samples (near longitudinal central area of heater)**
  - **Bentonite samples from close to the heater towards the outer parts of the barrier**
  - **X-Ray Fluorescence (XRF) bulk composition, X-ray CT-scan, µ-XRF, SEM-EDS, X-Ray Diffraction (XRD), Thermogravimetric analysis (TGA)**

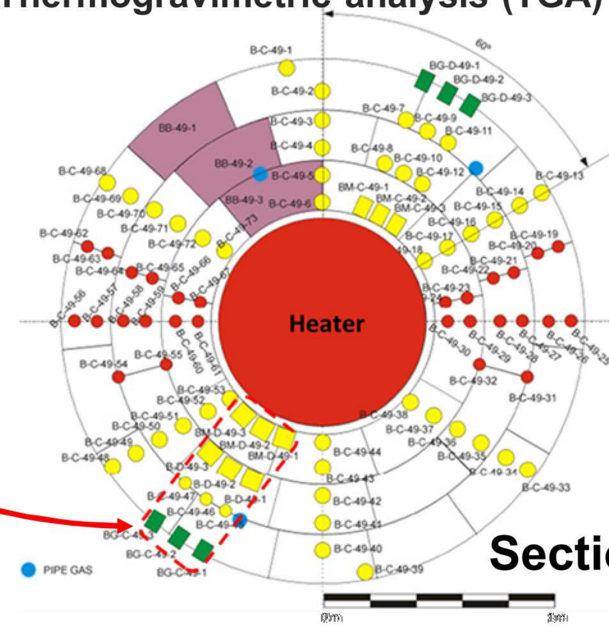


FEBEX-DP website



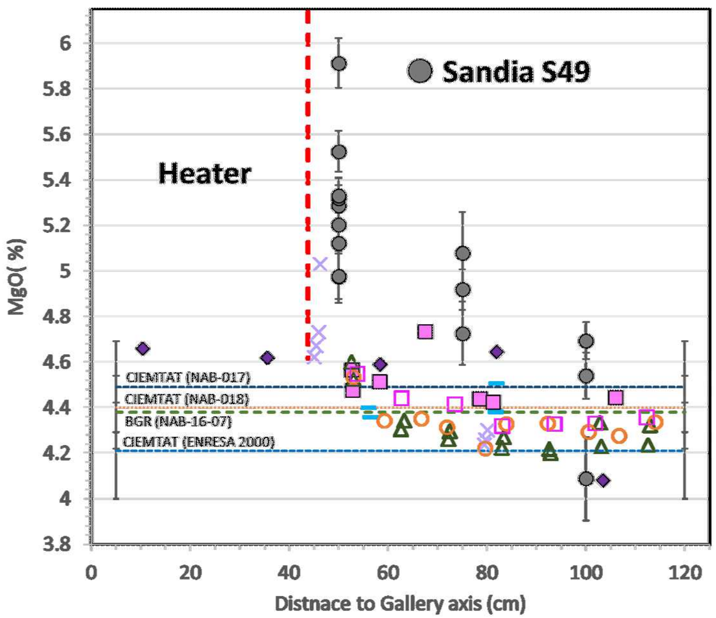


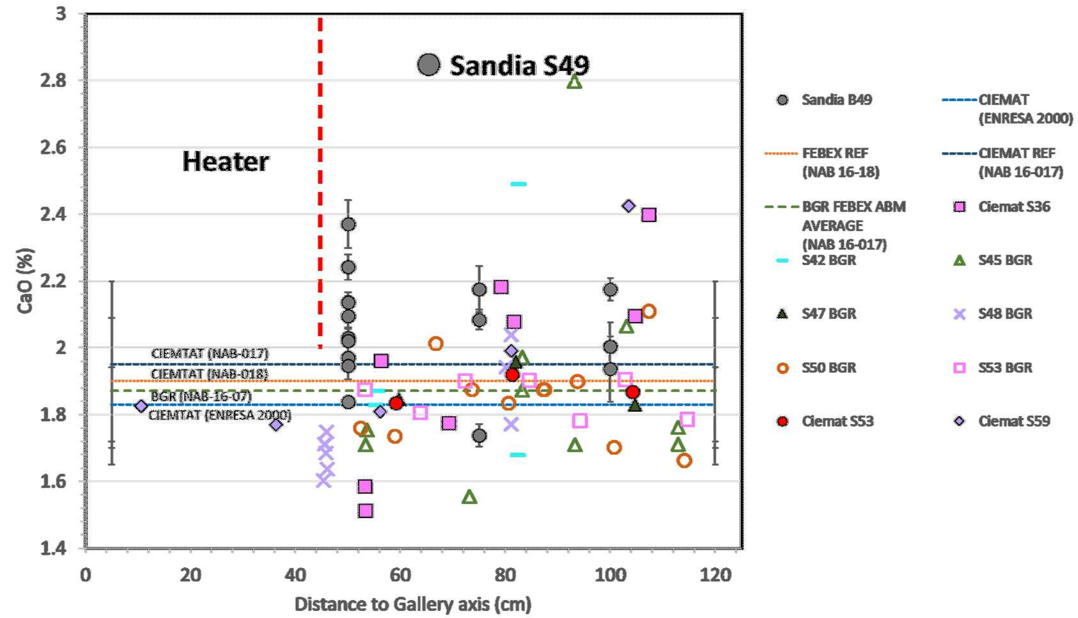
X-ray CT-Scan



Section 49

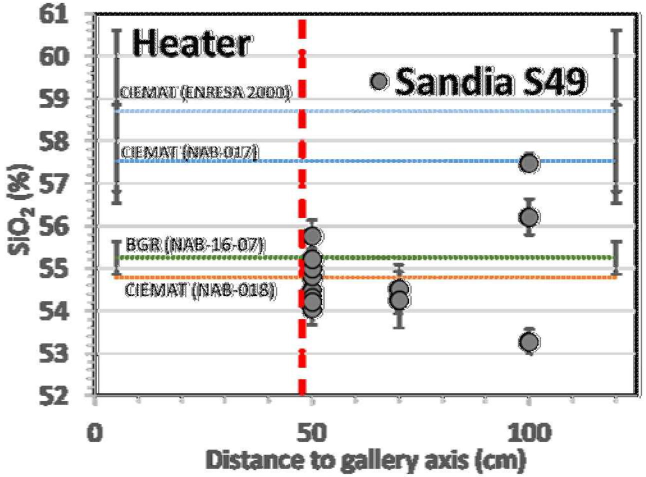
FEBEX-DP Bulk Bentonite Samples: X-ray Fluorescence (XRF)

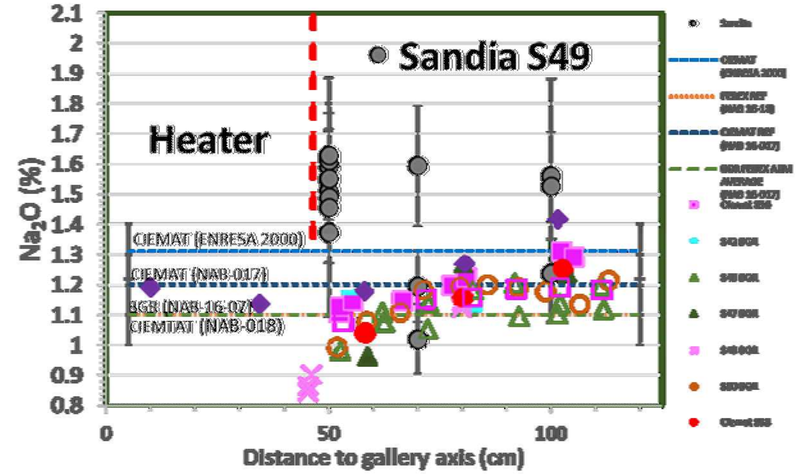


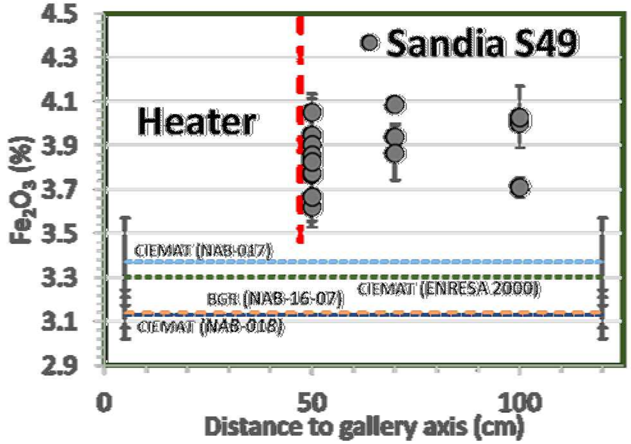


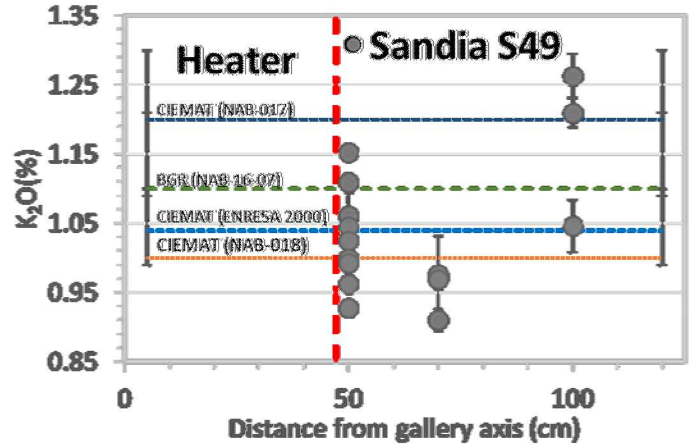
- Mg enrichment towards the heater surface – zones of increasing dry out conditions
- Bulk MgO content far from heater nominally within the bounds of other lab analyses
- Overall, CaO content is relatively variable close to the heater surface
- Mg enrichment(?):
  - Enhanced Mg content due to elevated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

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



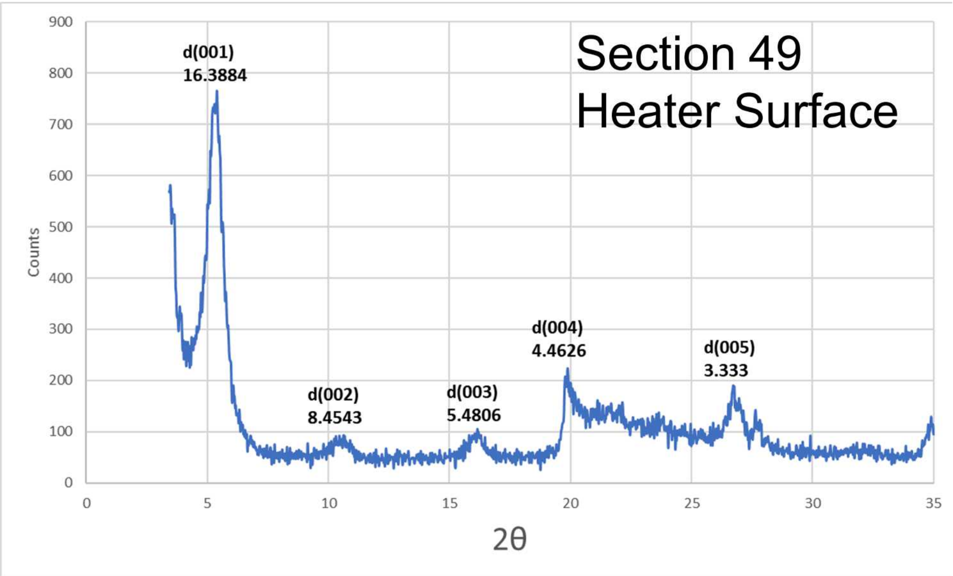


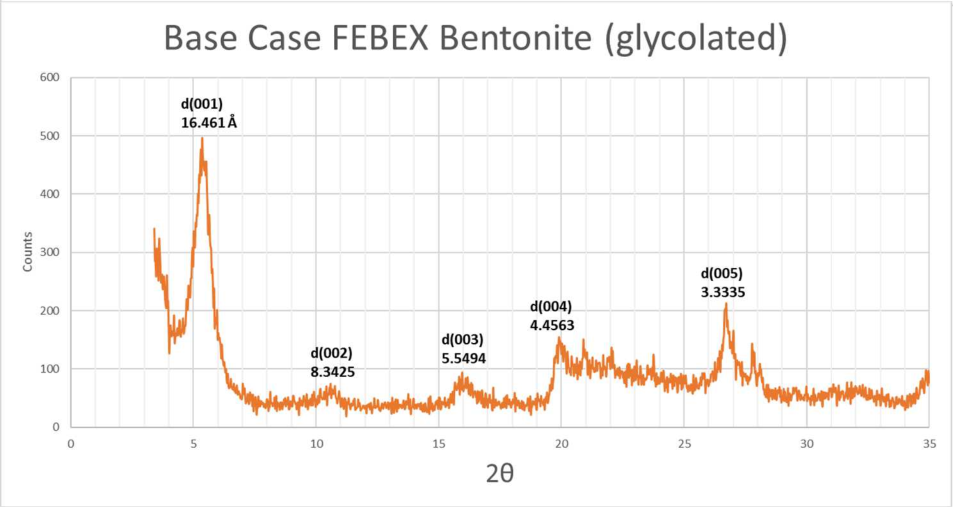


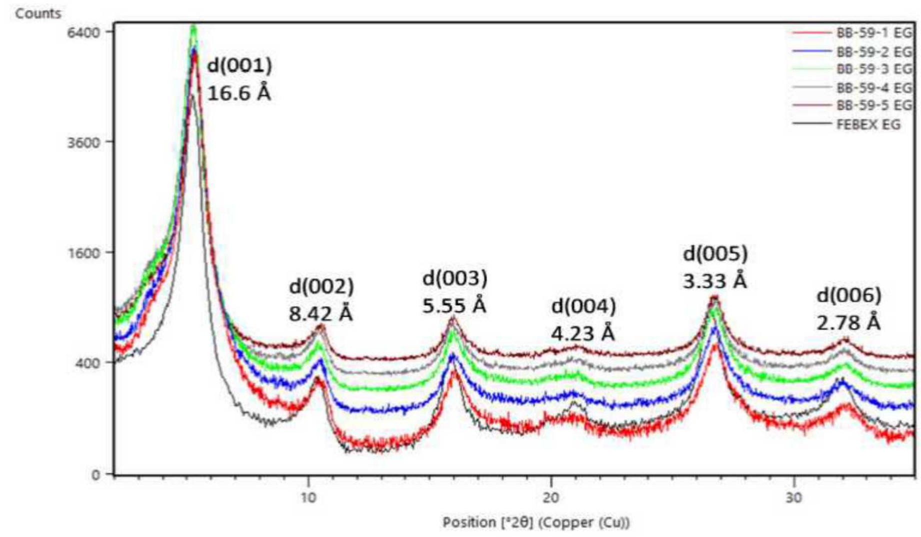


- Large uncertainties on Na_2O content – Issues with detection limits
- Slightly enriched in Fe_2O_3 relative to reference bentonite compositions
- Fe_2O_3, SiO_2, & K_2O fall within the range of reference bentonite compositions

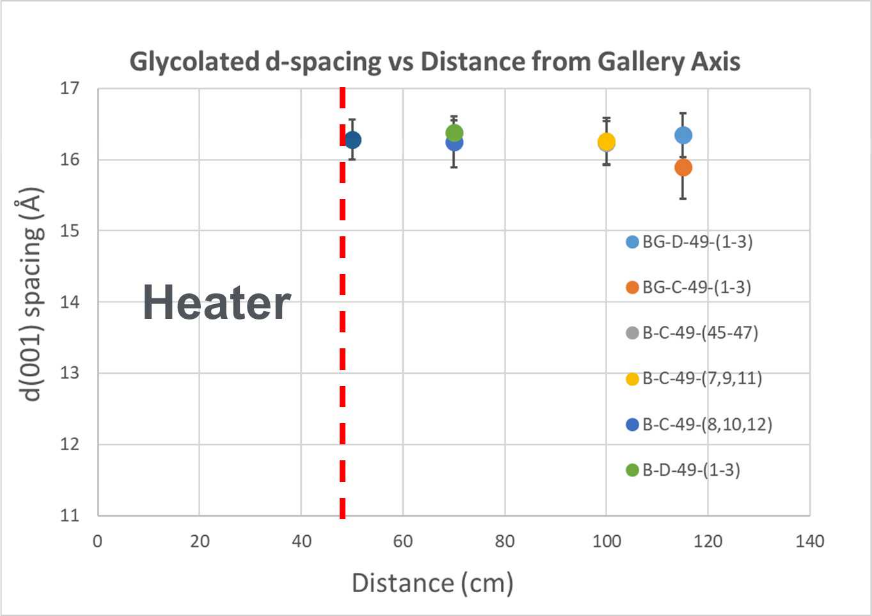
FEBEX-DP: Bentonite X-ray Diffraction (XRD)

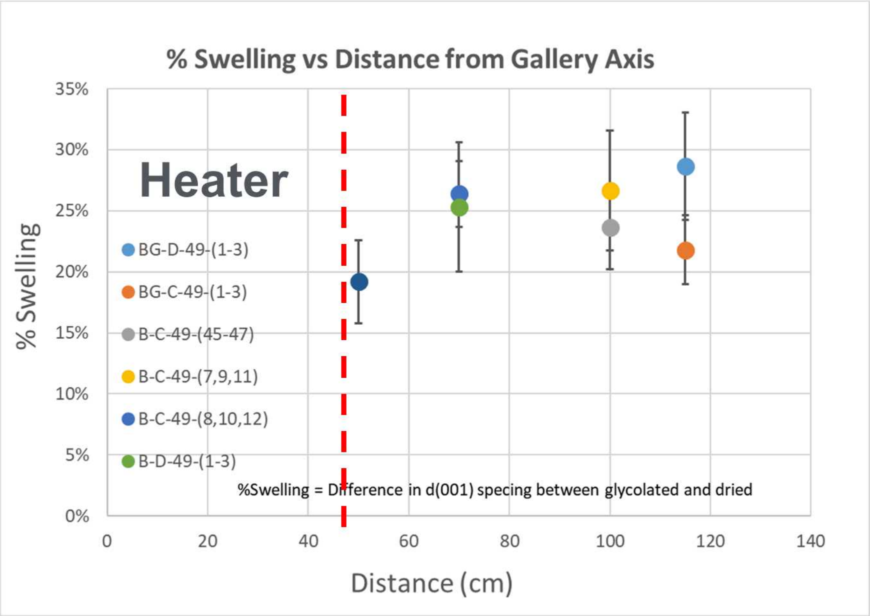






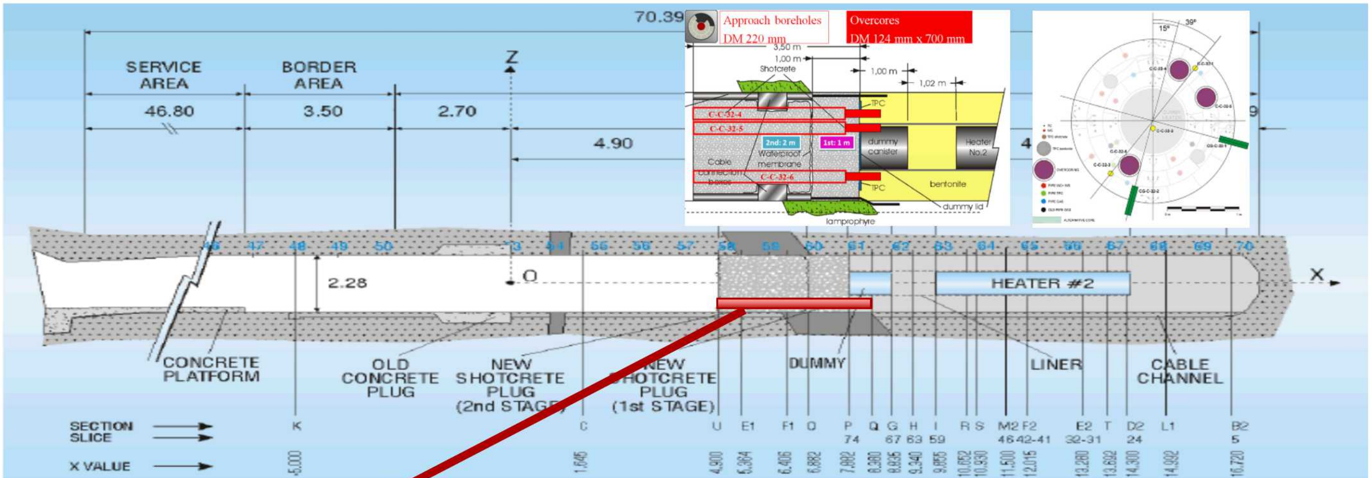
- Overall, XRD profiles are similar to those reported by FEBEX-DP project
- **Clay Structural Characterization:**
  - Comparison of XRD spectra across sampled domain
  - Evaluate d(001)-spacings as a function of distance from heater surface



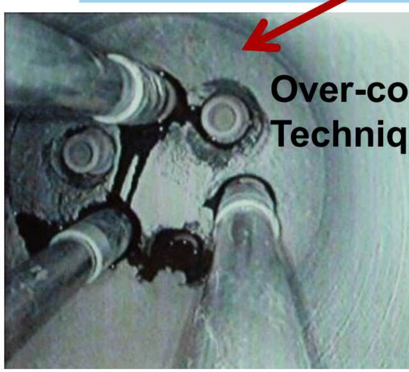


- No apparent effect of elevated temperatures on d(001) spacing for glycolated clay samples
- Slight decrease in swelling extent for samples in contact or close to the heater surface
- Prolonged exposure of bentonite to T = 95 – 100 ºC causes some changes in swelling
  - **Correlate with compositional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

FEBEX-DP: Shotcrete – Bentonite Interface Core Extraction



FEBEX-DP Sampling Plan



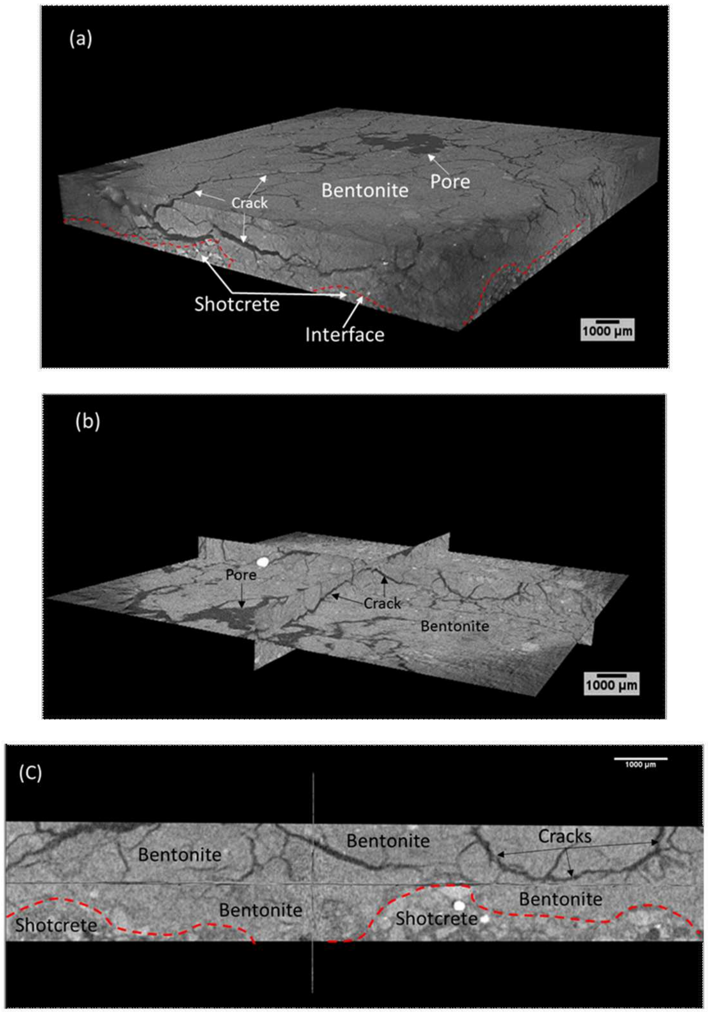
Over-coring Technique

Mäder et al. (2016)

C. F. Jove Colon (SNL)

- Shotcrete/bentonite interface sampling
- Characterization studies cement/bentonite interactions
  - Phase identification (SEM-EDS, XRD, μ-XRF)
  - X-ray CT Scan: micron-scale structures

Bentonite – Concrete Interface Characterization (X-ray CT Scan)



lain Features:

- **Occurrence of microcracks and pore spaces – connected in many cases**
- **“Craquelure” or “chickenwire” microcrack pattern (desiccation)**
- **Some embedded granular material in bentonite matrix with radiating cracks**
- **Heterogeneous microcrack spatial distribution → localized regions with no cracks**

rack – Pore pathways:

entonite:

Continuous and discontinuous pore-microcrack networks (2D & 3D)

Large pores tend to be connected to microcracks

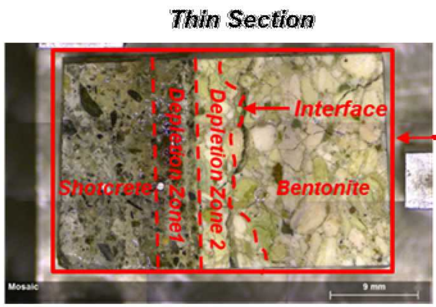
hotcrete:

Bentonite: Large pores tend to be connected to microcracks

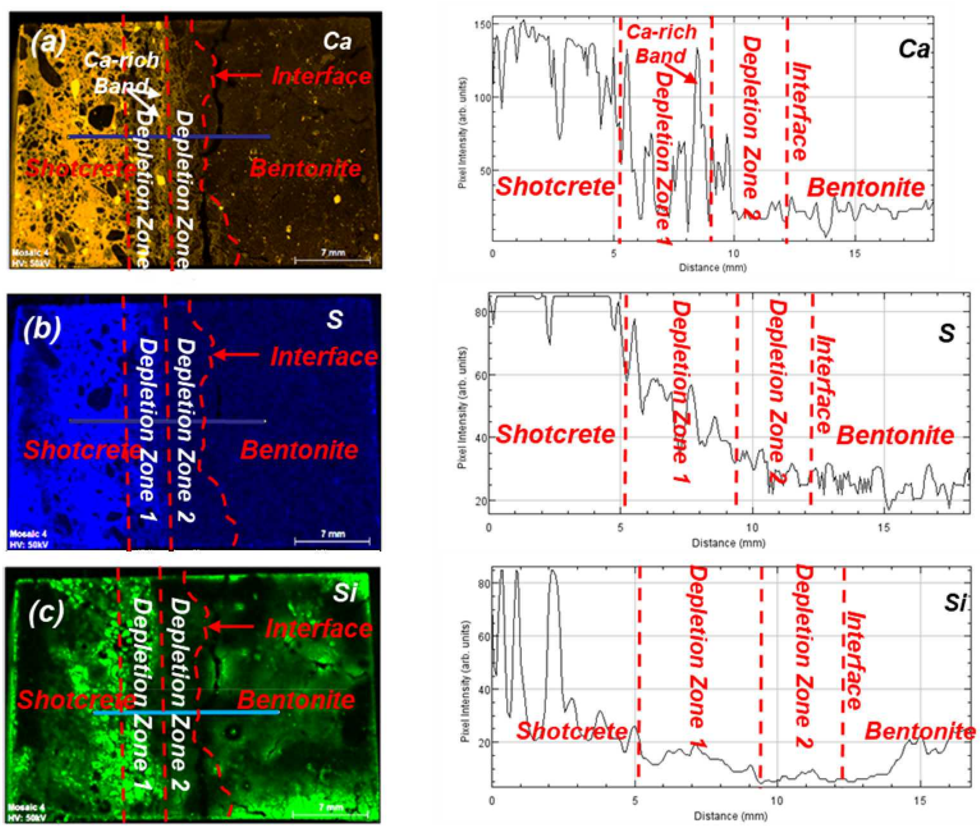
No or little microcracks

Isolated pores except at the interface

Shotcrete - Bentonite Interface Characterization (μ-XRF)



- 34.744 mm x 23.88 mm Scan Area,
- 30μm Spot to Spot Distance,
- 25μm Spot Size,
- X-ray Energy 50kV/200μA
- ~90 min to complete sample

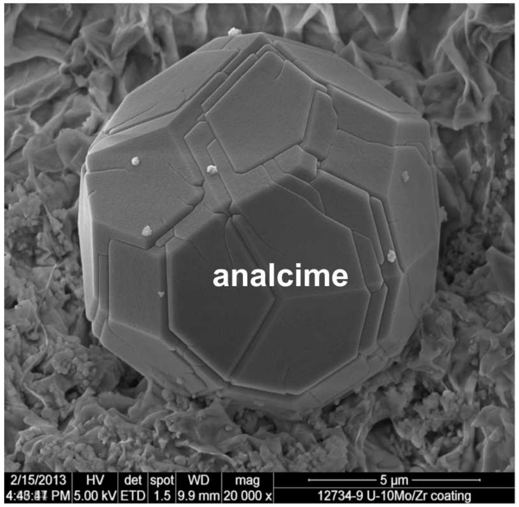


- **Main Features**
  - **Compositional map at thin section (mm) scale – Scanning at the μm scale**
  - **Sharp compositional changes at the bentonite-shotcrete interface**
  - **Consistent spatial correlation among various elements across interface**
- **Compositional Gradients**
  - **Depletion on shotcrete side of the interface → Leaching?**
  - **Bentonite seems compositional homogeneous at the interface**
  - **Limited reaction front?**

Jové Colón et al. (2017)

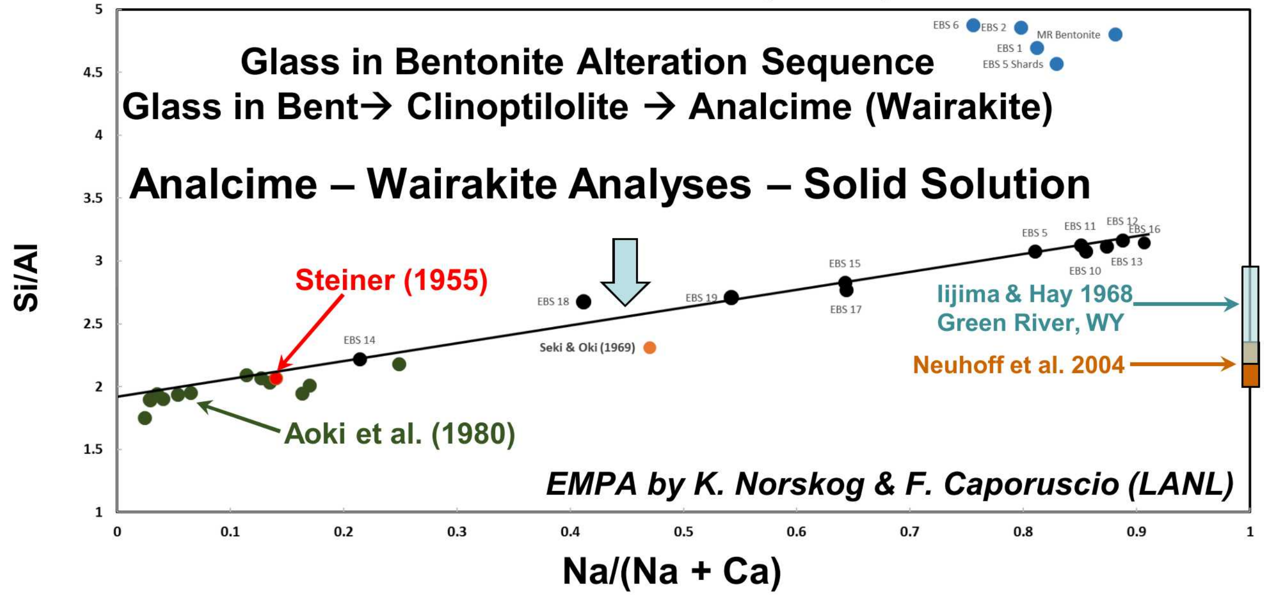
Authigenic zeolite produced from clinoptilolite / glass in bentonite interaction experiments

Analcime (Bentonite only)



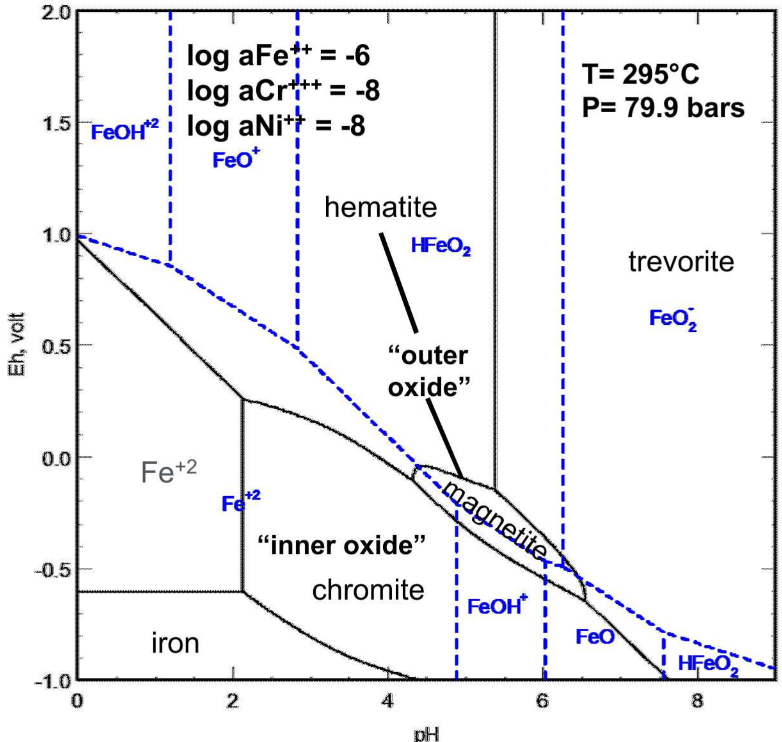
Wairakite-rich zeolite (Opalinus clay + Bentonite)

Jové Colón et al. (2017)

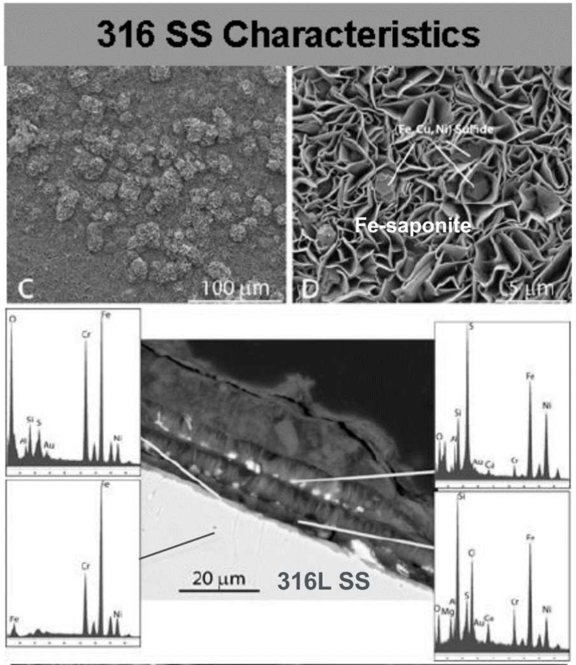


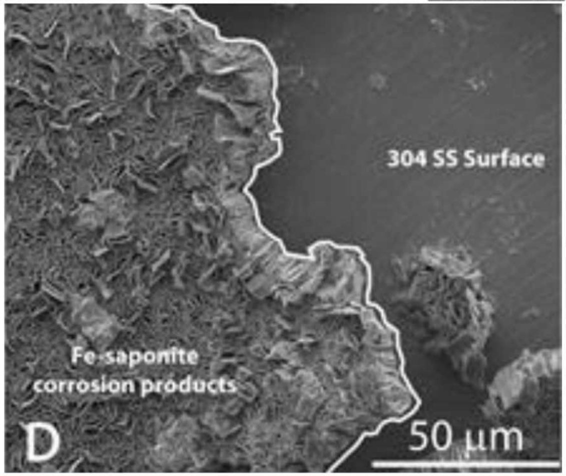
- **Bentonite Alteration and Zeolite Stability:**
  - **Glass alteration in bentonite → high Si**
  - **Formation of analcime – wairakite zeolites**
  - **Wairakite – analcime solid solution → widen range of zeolite stability**
  - **Little or no illite formation detected**
    - **High Si activities prevents illite stability?**

Bentonite – Steel Interaction Experiments



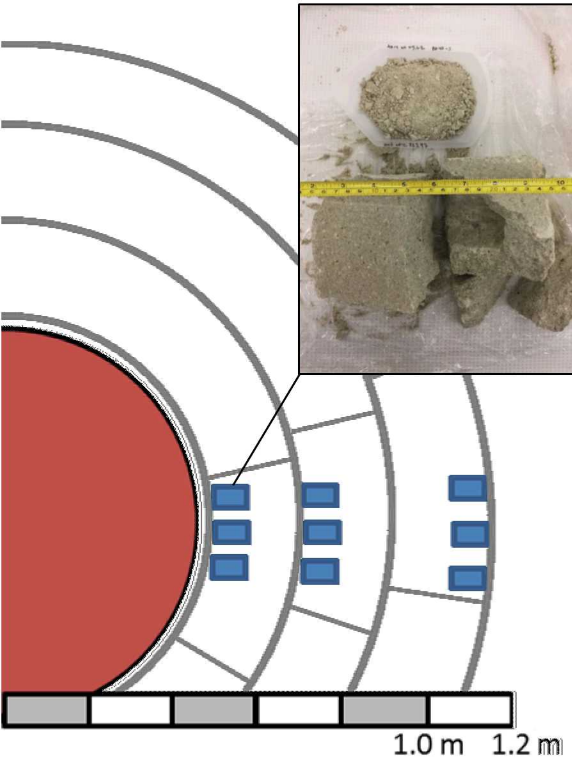
Cheshire et al. (2014)





- **Experiment**
  - **T = 300ºC; STRIPA brine**
  - **Wyoming Bentonite**
  - **316 Stainless Steel (SS), 304SS, low-C steel**
- **Results**
  - **Fe-Saponite growth perpendicular to metal substrate**
  - **S is generated from pyrite degradation in bentonite**
  - **Concurrent surface sulfide precipitation with Fe-saponite**
  - **More work needs to assess metal passivation effects**

U(VI) adsorption experiments: FEBEX-DP clay samples that experienced different temperature and moisture regimes



Heated Zone:

- 50 cm from axis (Section 48)
- T= 95°C
- Moisture Content≅18%

Cold Zone:

- 50 cm from axis (Sectio 59)
- T= 20°C
- Moisture Content ≅ 25%

Original FEBEX Bentonite Mineral Composition*

92 % smectite (illite-smetite mixed layer, with ~11% illite layers)
2% plagioclase
2% quartz
2% cristobalite
<1% potassium feldspar, calcite, trydimite, Fe- and Al-oxides

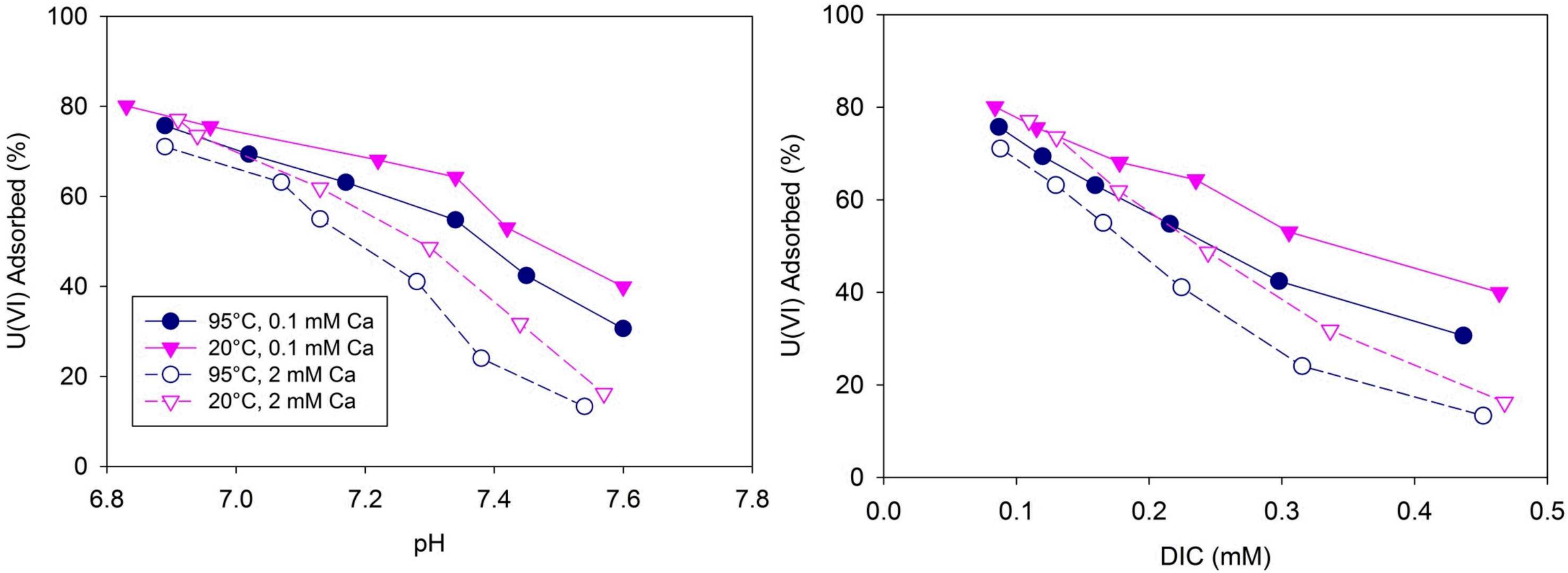
*Fernández et al. (2004)

Composite samples were created from 3 replicate blocks from each location, air-dried and sieved to < 63 µm.

Moisture content and temperature from Villar et al. (2018)

Lower U(VI) Sorption onto Heated Bentonite

< 63 μm fraction, bentonite composite samples, 0.5 g/L bentonite



Up to 10% lower U(VI) adsorption on heated bentonite.

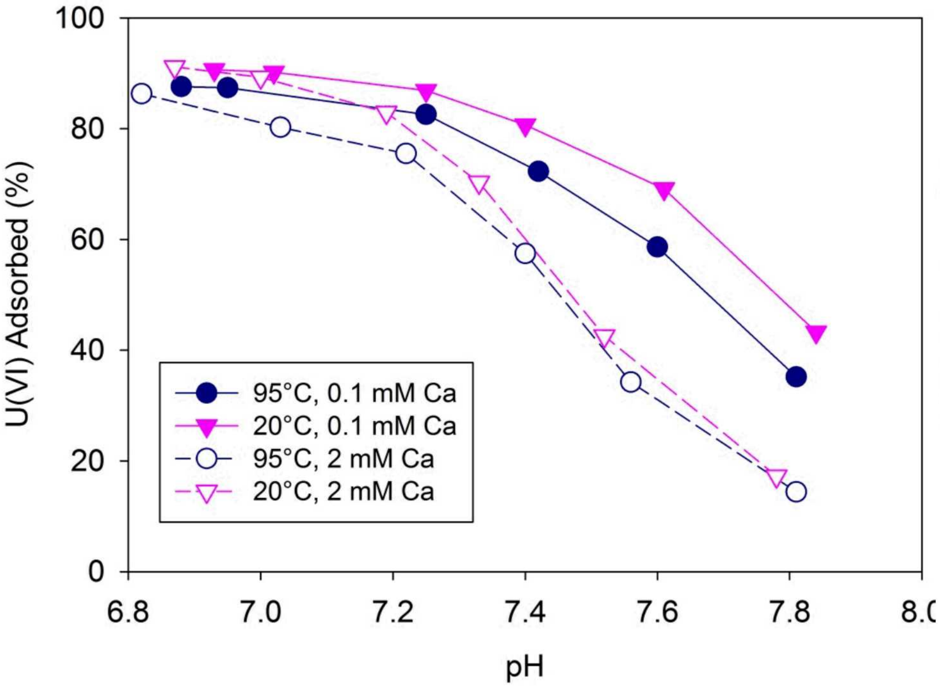
- Adsorption is lower in presence of 2 mM Ca compared to 0.1 mM Ca.
- Adsorption decreases as pH and DIC increase.

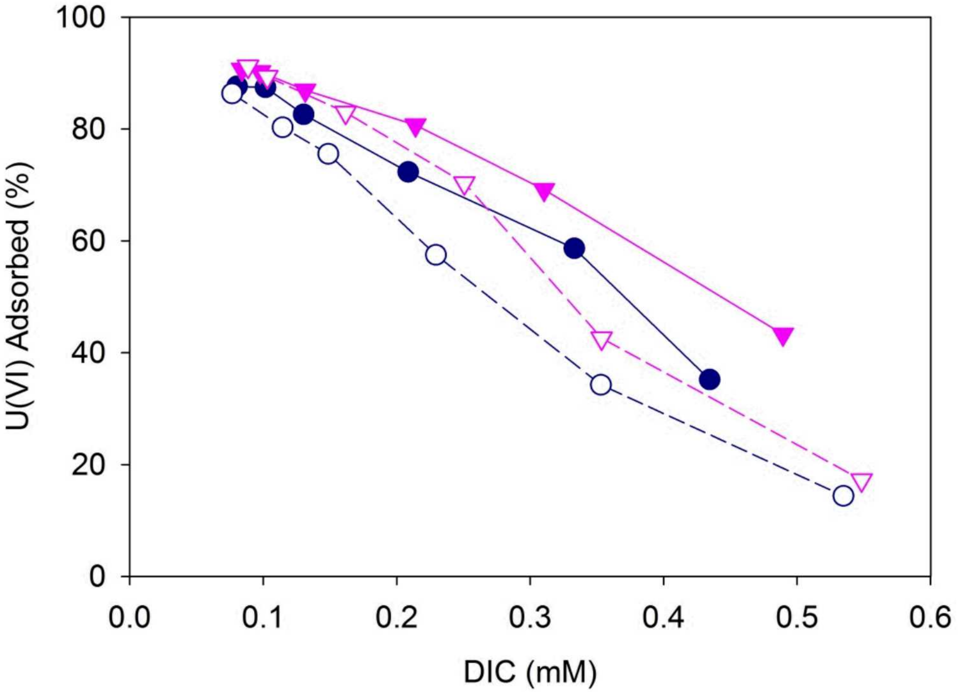
Possible reasons for lower U(VI) adsorption:

- aqueous U(VI) speciation
- relative fraction of clay (montmorillonite) mineral phase
- structure/composition of clay mineral fraction
- structure/composition of accessory mineral fraction (e.g., Fe-oxides)

U(VI) adsorption onto **purified** bentonite

< 2 μm fraction, carbonate minerals removed





Lower U(VI) adsorption on 95°C heated bentonite persists after purification.

- Consistently lower U(VI) adsorption onto 95°C heated sample in presence of 0.1 mM Ca
- Smaller difference in presence of 2 mM Ca
- As with bulk samples, U(VI) adsorption is lower at higher Ca concentration

Summary

- International collaborations on URL activities and partners provide unique opportunities for data and sample collection from heater tests
- Characterization and sorption studies of *post mortem* FEBEX-DP bentonite samples indicate:
  - Mg-enrichment in clay observed in bentonite close to the heated surface
  - Slight decrease in bentonite swelling also observed close to the heated surface
  - Lower U(VI) sorption for samples subjected to 95ºC relative to those exposed to ambient temperatures
  - Bentonite-cement interactions and cement leaching effects appear largely constrained to the interface
- Bentonite-metal interfacial interactions at elevated temperatures:
  - Produces zeolites (analcime) and sulfide phases
  - Fe-saponite growth perpendicular to the metal substrate
  - More work is needed to assess metal passivation effects

Acknowledgement

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Questions?

Clean. **Reliable. Nuclear.**

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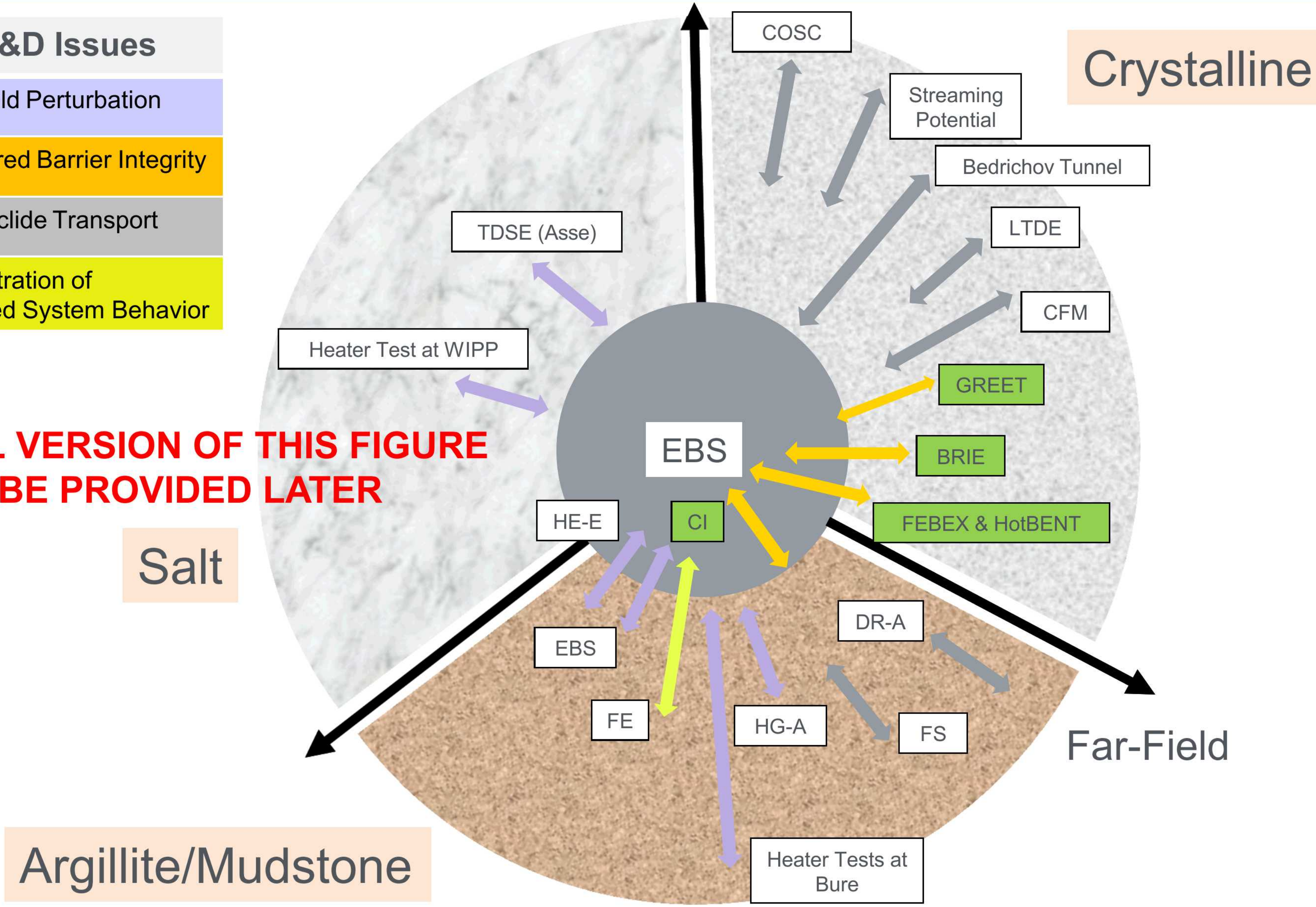
EXTRA SLIDES

EBS fits in the DOE Underground Research Laboratory (URL) Portfolio

Key R&D Issues

- Near-Field Perturbation
- Engineered Barrier Integrity
- Radionuclide Transport
- Demonstration of Integrated System Behavior

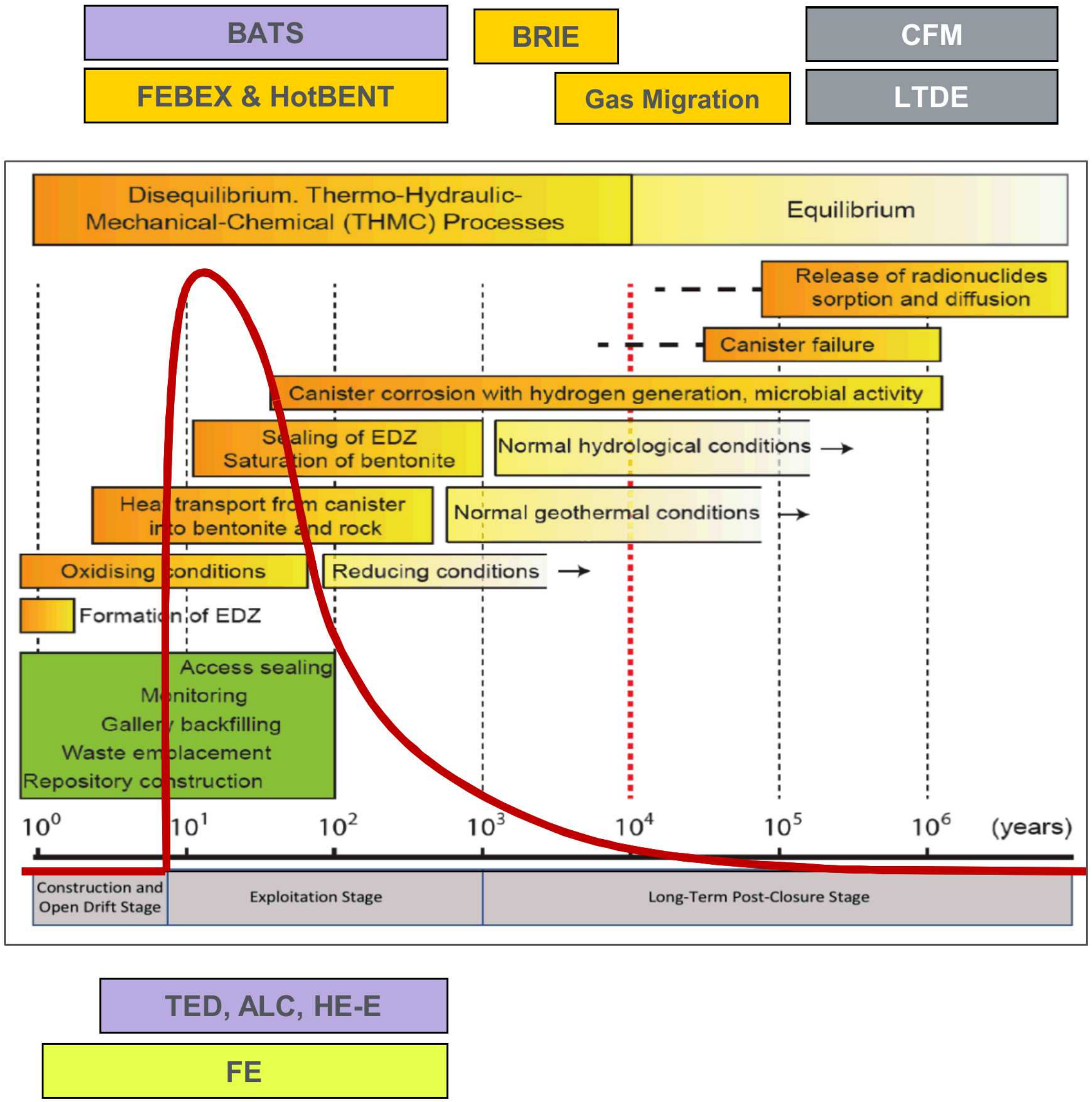
FINAL VERSION OF THIS FIGURE WILL BE PROVIDED LATER



Repository Phases and Relevant Processes

Key R&D Issues

- Near-Field Perturbation
- Engineered Barrier Integrity
- Flow and Radionuclide Transport
- Demonstration of Integrated System Behavior



Conceptual Model for Transport in Bentonite

- Location
  - Hosting nation or program
  - Host rock type
- Purpose of the test
  - E.g., provide basic in-situ data regarding brine flow in heated salt
    - Rock properties
    - Fluid composition, etc.
  - E.g., provide an opportunity for model validation/benchmarking with field data
- Why this is important in the context of the conceptual model for the medium (if this takes an extra slide, okay)

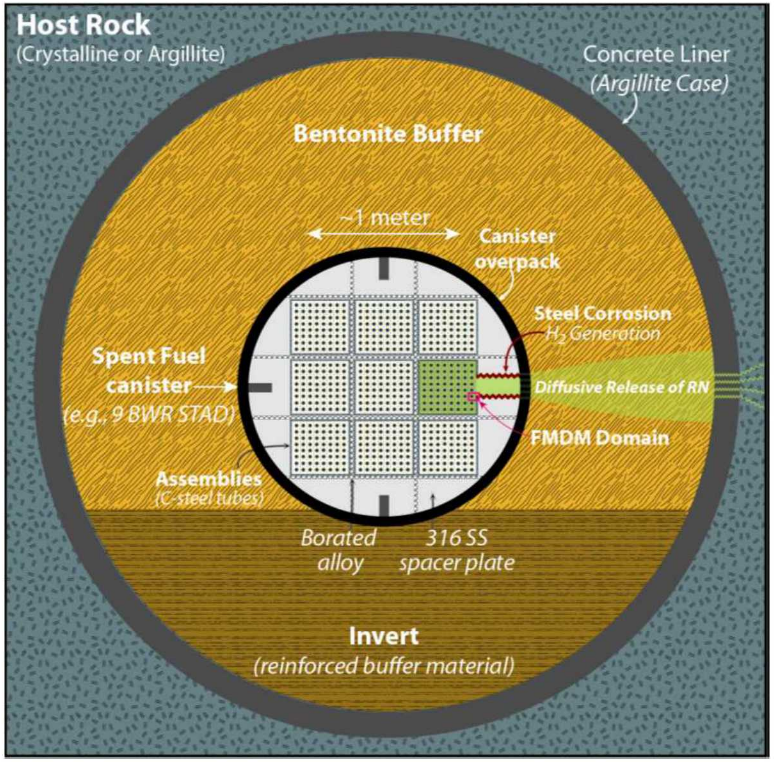
Body of the talk (~6-14 slides, depending on time available)

- Additional information on test design
- Results
- Analysis/interpretation: what have we learned?
- How results will be used in supporting the safety case/safety assessment

What is the Engineered Barrier System (EBS)?

- **EBS definition from the US Nuclear Regulatory Commission (10 CFR 60.2)**
  - "Engineered barrier system means the waste packages and the underground facility"

- **EBS definition from to the NEA/OECD EBS State-Of-The-Art Report (2003):**
  - "The "engineered barrier system" represents the man-made, engineered materials placed within a repository, including the waste form, waste canisters, buffer materials, backfill and seals."

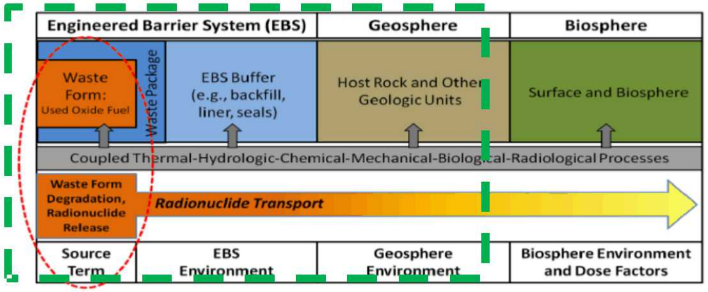


Generic EBS concept with bentonite barrier showing a canister breaching scenario (Jerden et al. 2019)

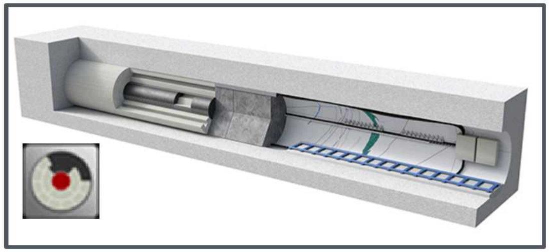
Argillite Disposal R&D

- Reactive-transport modeling (THC) with decay heat effects
  - 1D → 3D EBS Simulation Capability
- Engineered barrier system (EBS) model integration with performance assessment (PA)
- Thermodynamic modeling of barrier material interactions (**clay, cement, metal) and thermodynamic database (TDB) development**
- Clay interaction experiments:
  - **High temperature mineral phase stability, clay – metal interactions (waste package material (steel) corrosion)**
  - **Low-T RN sorption/diffusion in bentonite & modeling**
- High temperature coupled thermal-hydrological-mechanical-chemical (THMC) modeling
- Spent fuel matrix degradation model development
- International collaborations: **FEBEX-DP (GRIMSEL URL), DECOVALEX19, SKB EBS Task Force, Mont Terri URL (Switzerland)**

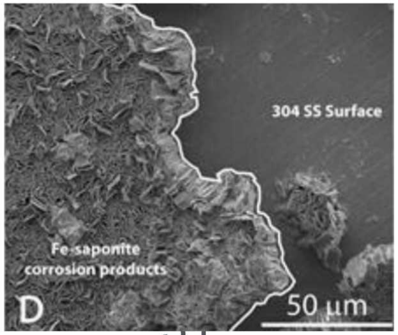
GDSA PA Level Of Integration



International Collaboration: FEBEX-DP



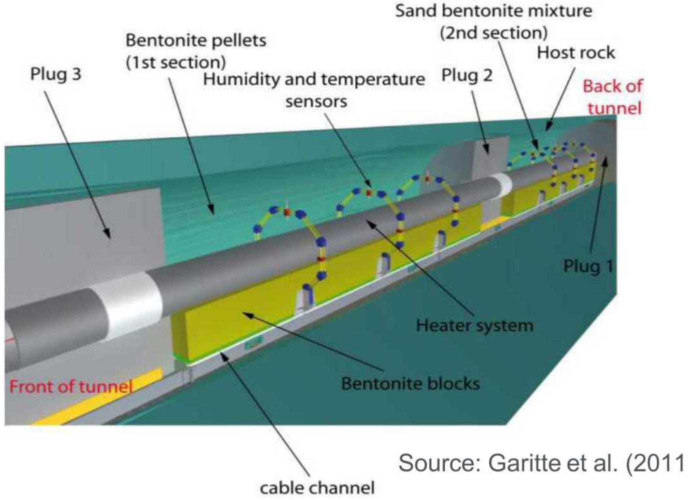
Clay-Metal Interactions



Steel Corrosion

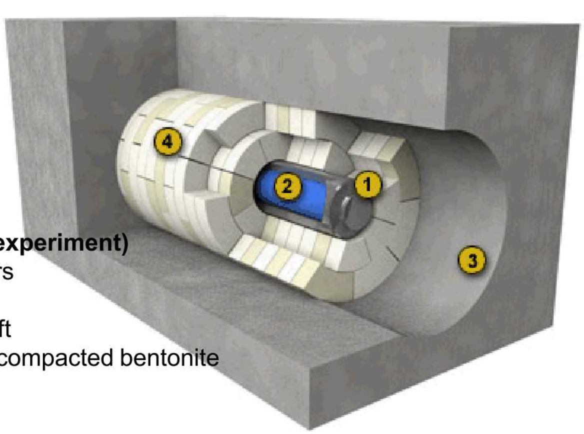
What has been done for EBS? (Cont.)

- International Activities:
  - **Underground Research Laboratories (URLs)**:
    - Mt. Terri (Opalinus Clay, Switzerland)
    - Grimsel (Granite, Switzerland)
    - Tournemire (Argillite, France)
    - Meuse/Haute-Marne (BURE) (Callovo-Oxfordian Clay, France)
    - FEBEX (Mock-Up, Spain; Granite, Site-Scale, Grimsel site, Switzerland)
    - KAERI/KURT (Granite, South Korea)
    - Horonobe (Mudstones) and Mizunami (Granite) Sites (Japan)
  - **International Collaborations**
    - DECOVALEX (Development of Coupled Models and their Validation Against Experiments, International Collaboration)
    - SKB EBS Task Force



Source: Garitte et al. (2011)

HE-E heater test at Mont Terri



FEBEX (in situ experiment)
(1) Steel canisters
(2) Waste
(3) Horizontal drift
(4) High-density compacted bentonite blocks

Source: http://www.grimsel.com

For more details, see Birkholzer et al. (2017)

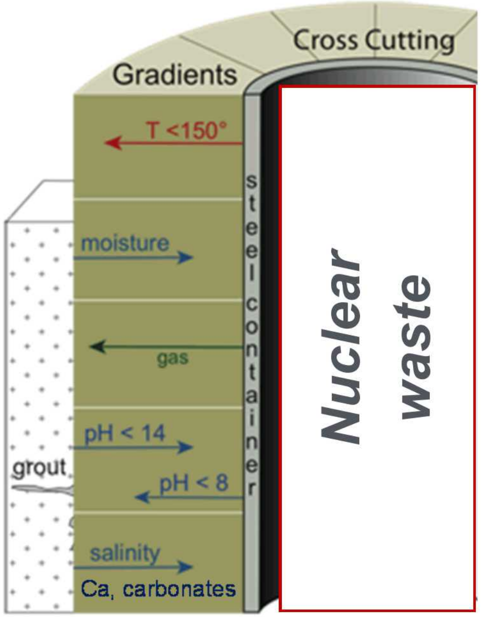
Conceptual Model for Transport in Bentonite

The long-term management of nuclear waste requires reliable predictions of radionuclide transport through engineered barrier systems (EBS).

⇨ **Compacted bentonite (montmorillonite)** is the proposed backfill material in EBS.

⇨ **Diffusion** will be the dominant transport mechanism in EBS that contributes to radionuclide dose in the environment.

⇨ **Gradients of chemical solution conditions and temperature** are expected over time and across EBS.

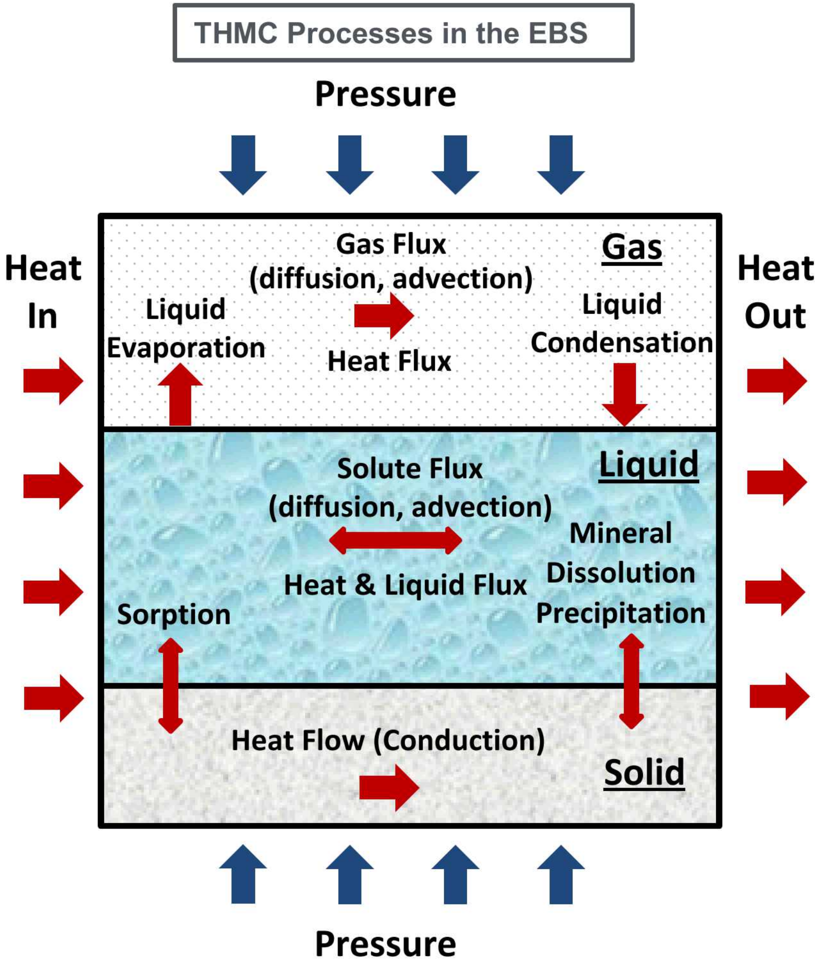


Goals:

- **Decrease the uncertainty** in actinide sorption / diffusion sub-models that are part of performance assessment models for waste repositories.
- **Investigate effects of changing chemical conditions and temperatures** on uranium(VI) sorption and diffusion.

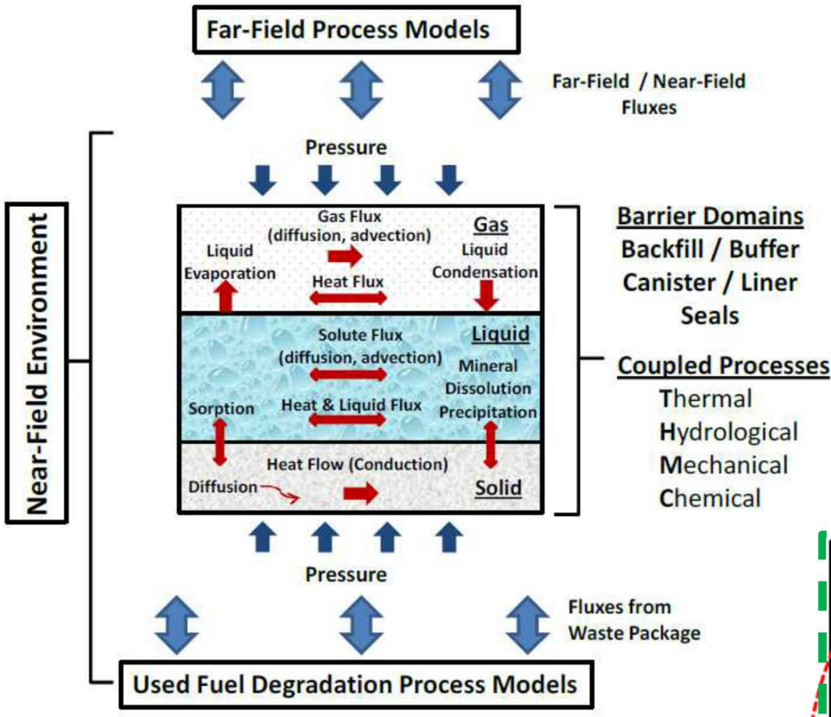
SFWST Needs for EBS?

- Highest ranked issues:
  - Waste Form
  - THM Processes
  - Waste Container
  - Radionuclide speciation and solubility
  - Buffer/Backfill material
- **THMC processes** relevant to interactions at **EBS interfaces:**
  - **Loci for important degradation processes in the near-field**
  - **Shares a boundary with far-field region**
  - **THMC models must assess the generic aspects of EBS design concepts**

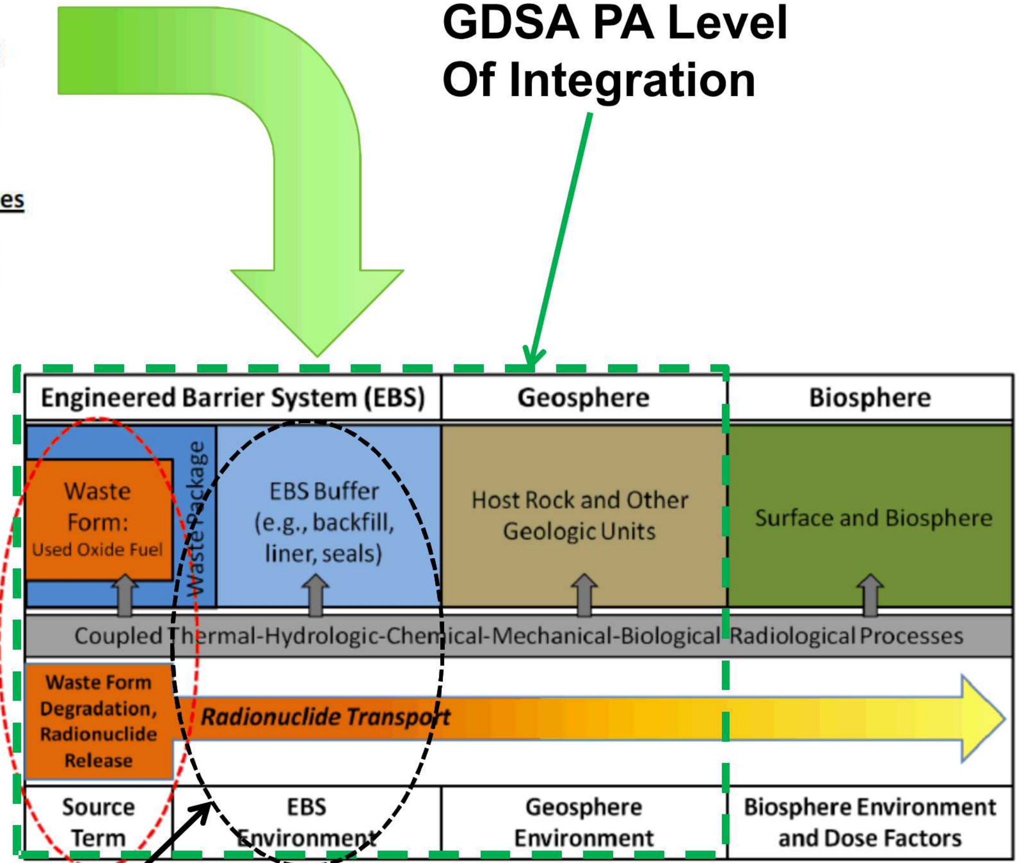


Modified After Olivella et al. (2011)

Coupled Processes in the Near- and Far-Field



GDSA PA Level
Of Integration

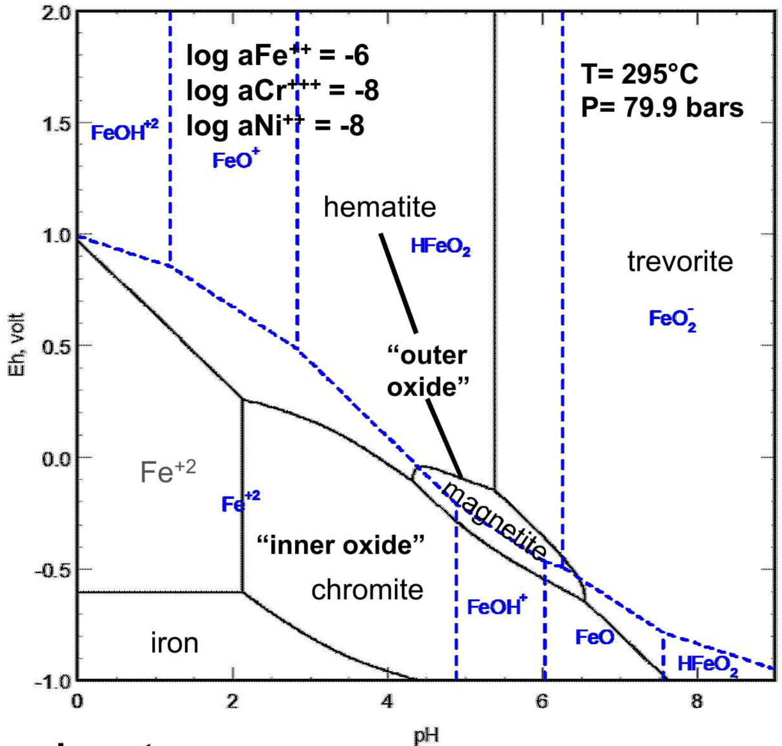


Wasterform
Interactions

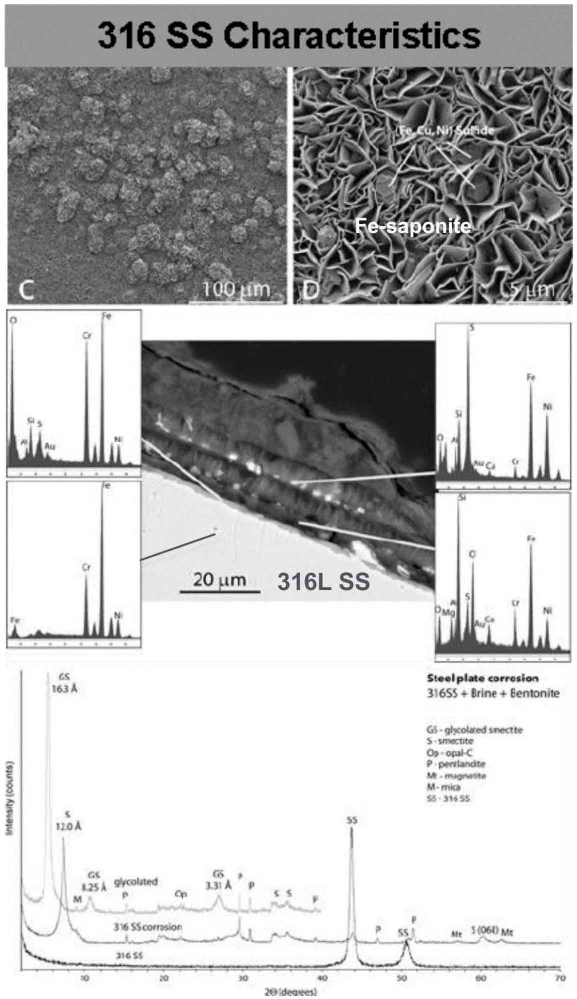
EBS
Interactions

Modified After Sassani et al. 2013

Bentonite – Metal Interaction Experiments



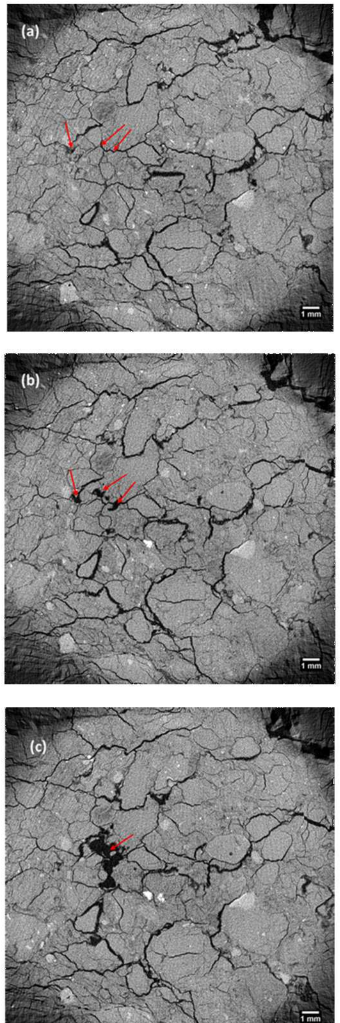
Cheshire et al. (2014)



- **Experiment**
  - **T = 300ºC**
  - **STRIPA Brine**
  - **Wyoming Bentonite**
  - **316 Stainless Steel (SS), 304SS, low-C steel**
- **Results**
  - **Fe-Saponite growth perpendicular to metal**
  - **Concurrent sulfide precipitation**
  - **Observations consistent with thermodynamic relations**

FEBEX-DP: Bentonite – Concrete Interface Characterization (X-ray CT Scan)

Red arrows: microcrack segments evolving into pores

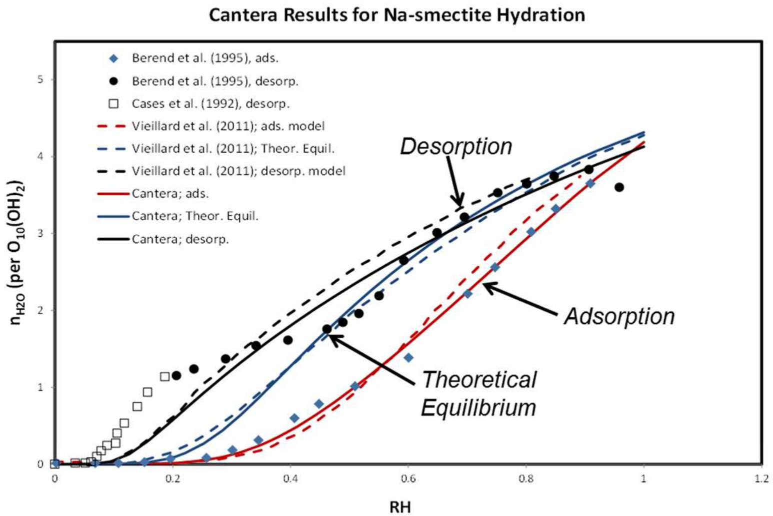


2D Slice Stack

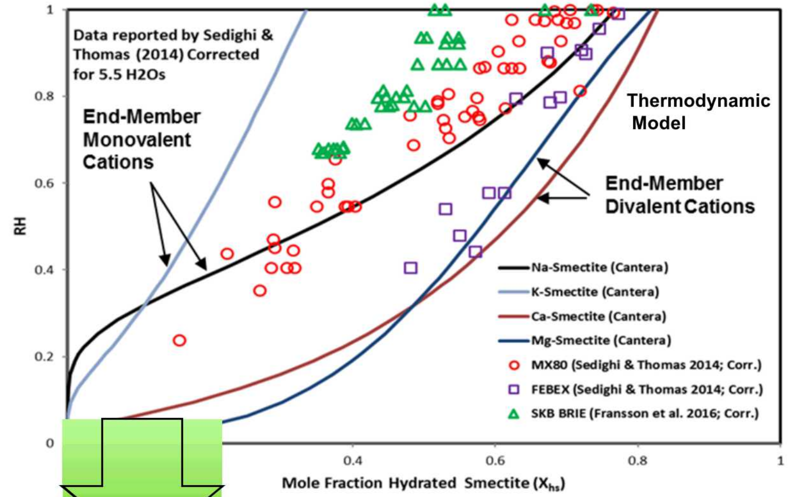
Z direction

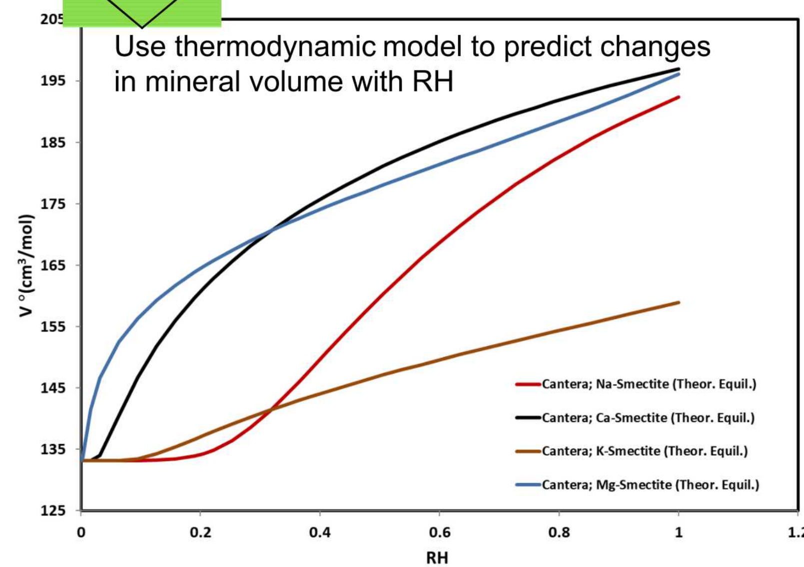
- **2D – 3D Stacked Image Evaluation:**
  - **Microcrack aperture enlargement/shrinking**
  - **Crack segments and junctions evolving into pores**
  - **Microcrack pathways can be highly heterogeneous**

Clay Hydration Modeling: Comparison with BRIE Water Retention Data + MX80 Bentonite



Fitting H_2O adsorption data for various smectite clay compositions





- **Relationships between clay composition and swelling behavior**
- **Data retrieval from URL and laboratory experiments**
  - **FEBEX**
  - **Bentonite MX80**
- **Trends for monovalent and divalent cationic composition consistent with thermodynamic model predictions for clay hydration**