Argillite Process Models and Testing

Carlos F. Jové Colón, Glenn Hammond, Emily Stein (SNL)

SFWST Campaign – Working Group Meeting, UNLV, Las Vegas, Nevada
May 23 – 25, 2017
Overview: Argillite Reference Case

Mariner et al. 2015

Dobson and Houseworth (2013)
R&D Priorities for Disposal in Argillite (1)

- **Experiments addressing FEPs:**
  - Chemical interactions with barrier materials and host rock at elevated temperatures
    - Information on stable phase mineralogy as a $f(P,T,X)$
    - Clay-metal interactions → corrosion processes
  - Radionuclide sorption onto clay
  - Radionuclide transport / diffusion in bentonite

- **Argillite process modelling in support of GDSA**
  - THMC – bentonite clay buffer
  - 1D reactive-transport (THC) modeling
    - Interactions at EBS interfaces (e.g., canister, seals / liners)
    - Temperature effects → phase transformation, secondary mineralogy
    - Porosity / permeability changes
  - Thermodynamic properties of barrier materials
Recognized gaps:
- THMC coupling implementation in PFLOTRAN → Emphasis on mechanical
- EDZ model representation in PFLOTRAN
  * THMC processes
  * Chemical interactions in the EDZ?
- Gas percolation through barriers & chemical interactions
- Treatment of concentrated pore solutions → Pitzer
- Canister corrosion representation

GDSA Integration
- Representation of EBS performance in GDSA → Multi-continuum approach
  * 1D EBS Reactive-Transport + 3D GDSA
- Waste package degradation (direct coupling)
  * Implementation within PFLOTRAN
  * Capture redox speciation dependencies
R&D Priorities for Disposal in Argillite (3)

Future Goals

- Integration of EBS process model work with GDSA-PA: PFLOTRAN implementation of 1D (EBS) + 3D (GDSA)
- PFLOTRAN implementation of canister degradation
- Expand non-isothermal reactive transport modeling of near-field chemical interactions to T>125°C
  - Thermal limits and the extent of sacrificial zones
  - PFLOTRAN simulation of multiphase transport
- Engagements and collaborations with international R&D activities (URL heater tests).
- Experiments on barrier material interactions at temperatures relevant of DWR and high-T waste disposal
  - Barrier / host-rock interactions
  - Effects of host-rock pore fluid chemistry on clay interactions
  - Assessment of thermal limits: experimental and code simulations
Chronological evolution of TC processes in argillite / shale repository

<table>
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<tr>
<th>Years</th>
<th>Temperature (T)</th>
<th>Chemical (C)</th>
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<td>Temperature Decrease</td>
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Coupled Processes in the Near- and Far-Field

- Far-Field Process Models
- Used Fuel Degradation Process Models
- Near-Field Environment
- Coupled Processes: Thermal, Hydrological, Mechanical, Chemical
- Wasterform Interactions
- EBS Interactions
- GDSA PA Level Of Integration

Modified After Sassani et al. 2013

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May 23-25, 2017
Coupled Processes in the Near- and Far-Field

GDSA PA Level Of Integration

Used Fuel Degradation Process Models

Far-Field Process Models

Barrier Domains
- Backfill / Buffer
- Canister / Liner Seals

Coupled Processes
- Thermal
- Hydrological
- Mechanical
- Chemical

Engineered Barrier System (EBS)

Geosphere

Biosphere

Coupled Thermal-Hydrologic-Chemical-Mechanical-Biological-Radiological Processes

Radionuclide Transport

Source Term

EBS Environment

Geosphere Environment

Biosphere Environment and Dose Factors

Wasterform Interactions

EBS Interactions

Modified After Sassani et al. 2013
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Coupled Processes in the Near- and Far-Field

Solution Chemistry: pH, T, [O₂], [H₂], [Fe], [SO₄], [Cl], [OH], [F]

Performance Assessment (Argillite & Crystalline)

Canister Degradation Rates

GDSA PA EBS
Level of Integration

Steel

PFLOTRAN
Canister Degradation

FMDM

Species Conc.

SO₄--

EDZ

Backfill

Canister

Backfill

EDZ

Rock

Clay Rock

Canister Overpack

Host Rock

Waste Container And UNF Assembly

Two-Domain Backfill/Buffer Barrier Material

Cement Lining

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Waste package degradation based on clay – metal interactions:
- Fe-rich clay parageneses
- Corrosion products
- Aqueous-Solid Equilibria
- Sulfide effects (e.g., pyrite decomposition)

Implementation within a reactive transport model:
- PFLOTRAN
  - Redox speciation
  - Solid phase assemblage
  - Model Conceptualization
High Level of Core Preservation During Extraction

Computerized Tomography (CT)

Overcoring Technique

Overcore Specimen

Mäder (2014); CI Report

X-Ray CT Scan Image

Pores

Shotcrete

Cracks

Interface

Bentonite

1.66 mm

Imaging by J. Eric Bower (SNL)

(a)

(b)

(c)

Crack

Bentonite

Pore

Shotcrete

Interface

Shotcrete

Bentonite

Cracks

Bentonite

1000 µm
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International: FEBEX-DP
Sampling Close to Heater

Bentonite With Corrosion Rind

Corroded Carbon Steel Liner
(Surrounding Heater)

X-Ray Elemental Maps

EDS Spectra Fe-Rich Clay?
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1D Waste Form-Canister-Buffer Model as Source Term within 3D Model

What concentrations to apply at buffer-host rock boundary?

GDSA 3D Model

Host Rock

Waste Form Cell
1D Waste Form-Canister-Buffer Model as Loosely Coupled Source Term within 3D Model

- Sequentially coupled feedback between 1D and 3D models.
- Similar to FMDM implementation

EBS 1D Model

GDSA 3D Model
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1D Radial+ 3D Domain Discretization at Waste Form Cells

- **Multi-Continuum approach to represent EBS in GDSA:**
  - 1D Radial (EBS) volumetric equivalent inside a 3D (GDSA) cube
  - Unstructured grid
  - High level of coupling
This work supported by the DOE-NE Spent Fuel Waste Science and Technology campaign, Fuel Cycle Technologies R&D program.
Overview: Coupled Processes in the Near-Field Environment

THMC Processes and Interactions at Repository interfaces

Generic Host Environment (Clay, Granite, Salt, deep borehole)

- Seal/Spacer (Cement)
- Thin Metal Outer Shell
- Multi-Border EBS Bentonite/Sand Layers
- Disposal Room (Near Field)
- Seal/Plug (Cement)
- EDZ
- Canister
- Not to scale

Mechanical damage modeling in a fractured EDZ

THMC Processes and Effects in EBS Components and Interfaces, Degradation Processes, and Interactions in EBS Materials

TMC Characteristics of EBS Materials

Rn Chemistry and Transport; Colloid Formation, Stability, and Transport in EBS Materials

T = Thermal
H = Hydrological
M = Mechanical
C = Chemical
FEBEX-DP: Bentonite – Concrete Interface Characterization

- No indication of strong elemental gradients beyond the interface region
- Cracks (desiccation?) tend to be abundant at the interface
- Portlandite mineralization at the interface?
- More elemental line-scans needed to resolve compositional gradients

Work in Progress!!!
Reactive transport modeling base case scenario(s):

- Interaction with EBS components gauged by anoxic hydrothermal experiments (e.g., Steel/copper corrosion in the presence of clay)
- Backfill-buffer composition, secondary phases (e.g., pyrite) influencing metal corrosion reactions (e.g., copper):
- Evaluate geochemical feedbacks (e.g., redox zones) and U transport and concentration profiles
Yifeng’s Pyramid for Crystalline Technology

![Diagram of Yifeng's Pyramid for Crystalline Technology](image-url)
Clay host-rock FEPs should map:
- Coupled THMC processes
- Rock hydraulic properties and associated heterogeneities
- Geometry and extent of surrounding units
- Hydraulic and diffusion gradients
- Migration pathways and EDZ structure
- Thermal rock properties
- Geomechanical damage and stability
- Fracture distribution
- Clay mechanical and swelling behavior
- Geological history (e.g., burial, tectonics)
- Etc…

Mariner et al. 2015
Shotcrete/bentonite interface sampling

Characterization studies cement/bentonite interactions

- Phase identification (SEM-EDS, XRD)
- X-ray CT Scan: micron-scale structures
UFD R&D Roadmap Priority Ranking

FEPs $\rightarrow$ Generic Disposal EBS Concepts

<table>
<thead>
<tr>
<th>Category</th>
<th>SNF</th>
<th>Glass</th>
<th>Ceramic</th>
<th>Metal</th>
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<td>WASTE PACKAGE MATERIALS $\rightarrow$</td>
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<td>BUFFER / BACKFILL (media type) $\rightarrow$</td>
<td>Clay</td>
<td>Salt</td>
<td>Crystalline</td>
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UFD R&D Roadmap Priority Ranking and Mapping FEPs → Generic Disposal EBS Concepts

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<tr>
<th>SEAL / LINER MATERIALS</th>
<th>Cement</th>
<th>Asphalt</th>
<th>Metal</th>
<th>Polymer</th>
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<td>Salt-Sat. Cements</td>
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</table>
Shale Distribution Map

Depth to Top of Shale in Meters

- < 100
- 101 - 200
- 201 - 300
- 301 - 400
- 401 - 500
- 501 - 600
- 601 - 700
- 701 - 800
- 801 - 900
- 901 - 1,000

Legend:

- 1,001 - 1,200
- 1,201 - 1,400
- 1,401 - 1,600
- 1,601 - 1,800
- 1,801 - 2,000
- 2,001 - 2,500
- 2,501 - 3,000
- 3,001 - 3,500
- 3,501 - 4,000
- > 4000
Experimental: Clay Interactions with EBS Materials -> Copper – Clay Interactions

- Sulfide-induced corrosion (anoxic):
  - Pyrite (FeS₂) decomposition
  - Primary corrosion product → Chalcocite (Cu₂S):
    - 13 μm thick chalcocite layer
  - Pitting corrosion

\[
\begin{align*}
\text{Cu}^0 + H_2S + CuCl_2^- & = Cu_2S(s) + 2Cl^- + H_2 + H^+ \\
\text{FeS}_2 + 2e^- & = Fe^{2+} + 2HS^- + H^+ \\
\text{SO}_4^{2-} + 7H^+ + 8e^- & = HS^- + 4H_2O
\end{align*}
\]

Cheshire et al. 2014
1D Waste Form-Canister-Buffer Explicitly Discretized within 3D Model
## General Attributes for Disposal in Shale

<table>
<thead>
<tr>
<th>Attributes</th>
<th>Salt</th>
<th>Shale</th>
<th>Granite (crystalline rock)</th>
<th>Deep boreholes</th>
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<tbody>
<tr>
<td>Thermal conductivity</td>
<td>High</td>
<td>Low</td>
<td>Medium</td>
<td>Medium</td>
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<tr>
<td>Permeability</td>
<td>Low</td>
<td>Low</td>
<td>Low (unfractured) to permeable (fractured)</td>
<td>Low</td>
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<tr>
<td>Mechanical strength</td>
<td>Low</td>
<td>Low</td>
<td>High</td>
<td>High</td>
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<tr>
<td>Deformation behavior</td>
<td>Viscous-plastic</td>
<td>Plastic to brittle</td>
<td>Brittle</td>
<td>Brittle</td>
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<td>Stability of cavity</td>
<td>Low</td>
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<td>High</td>
<td>Medium at great</td>
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<tr>
<td>Dissolution behavior</td>
<td>High</td>
<td>Very low</td>
<td>Very low</td>
<td>Very low</td>
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<tr>
<td>Chemical condition</td>
<td>Reducing; high ionic strength; relatively simple chemical system</td>
<td>Reducing; complex chemical system</td>
<td>Reducing; relatively simple chemical system</td>
<td>Reducing; relatively simple chemical system; moderate to high ionic strength</td>
</tr>
<tr>
<td>Radionuclide retention</td>
<td>Very low</td>
<td>High</td>
<td>Medium to high</td>
<td>Medium to high</td>
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<tr>
<td>Thermal limit</td>
<td>Relatively high</td>
<td>Relatively low (?)</td>
<td>No limit</td>
<td>No limit</td>
</tr>
<tr>
<td>Available geology</td>
<td>Wide</td>
<td>Wide</td>
<td>Wide</td>
<td>Wide</td>
</tr>
<tr>
<td>Geologic stability</td>
<td>High</td>
<td>High</td>
<td>High</td>
<td>High</td>
</tr>
<tr>
<td>Engineered barrier system</td>
<td>Minimal; waste package damage by room closure</td>
<td>Minimal; waste package damage by room closure</td>
<td>Needed. Able to fully take credit for the engineered barrier system</td>
<td>Borehole seal needed</td>
</tr>
<tr>
<td>Human intrusion/resource exploration</td>
<td>Relatively high</td>
<td>Relatively high</td>
<td>Low</td>
<td>Low</td>
</tr>
<tr>
<td>Retrievalability of waste</td>
<td>Feasible</td>
<td>Feasible</td>
<td>Easily retrievable</td>
<td>Difficult</td>
</tr>
</tbody>
</table>
1D EBS + 3D GDSA Discretization at Waste Form Cells
1D Radial+ 3D Domain Discretization at Waste Form Cells
Albany 3D FEM thermal model for single-drift multi-waste-package scenario (currently meshed for a multi-drift case)

Albany + DAKOTA coupling – Maybe use Trilinos+DAKOTA (i.e., TRIKOTA) for sensitivity analysis

Goal: Evaluate waste package and drift spacing to evaluate thermal limits at given heat loads.
Bullets, Numbers, Text, Pictures