Used Fuel Disposition in Crystalline Rocks

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Work package structure

UZ = Unsaturated Zone
DPC = Dual Purpose Canisters
EBS = Engineered Barrier System
GDSA = Geologic Disposal Safety Assessment
Acknowledgments

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### Characteristics of host rocks

<table>
<thead>
<tr>
<th>Attributes</th>
<th>Salt</th>
<th>Shale</th>
<th>Granite (crystalline rock)</th>
<th>Deep boreholes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal conductivity</td>
<td>High</td>
<td>Low</td>
<td>Medium</td>
<td>Medium</td>
</tr>
<tr>
<td>Permeability</td>
<td>Low</td>
<td>Low</td>
<td>Low (unfractured) to permeable (fractured)</td>
<td>Low</td>
</tr>
<tr>
<td>Mechanical strength</td>
<td>Low</td>
<td>Low</td>
<td>High</td>
<td>High</td>
</tr>
<tr>
<td>Deformation behavior</td>
<td>Visco-plastic</td>
<td>Plastic to brittle</td>
<td>Brittle</td>
<td>Brittle</td>
</tr>
<tr>
<td>Stability of cavity</td>
<td>Low</td>
<td>Low</td>
<td>High</td>
<td>Medium to high</td>
</tr>
<tr>
<td>Dissolution behavior</td>
<td>High</td>
<td>Very low</td>
<td>Very low</td>
<td>Very low</td>
</tr>
<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>
</tr>
<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>Retrievability of waste</td>
<td>Feasible</td>
<td>Feasible</td>
<td>Easily retrievable</td>
<td>Difficult</td>
</tr>
</tbody>
</table>

- **High mechanical strength and thermal limit**
  - Suitable for disposal of large and hot waste canisters
- **Fractured nature**
  - Engineered barrier system equally important as the nature barrier

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Geochemical characteristics of groundwater

Water type A: Dilute 0.5-2 g/L TDS; Δ18O = -11.7 to -9.6‰ SMOW; Na-HCO3; mainly Meteoric. 
Main reactions: Weathering, ion exchange, dissolution of calcite, redox reactions, microbial reactions. 
Redox conditions: Oxidising - reducing.

Water type B: Brackish 6-10 g/L TDS; Δ18O = -11.6 to -8.6‰ SMOW; Na(Ca,Mg)-Cl(SO4)2 to Ca-Na(Mg)-Cl(SO4)2; Marine (Strong Littorina Sea component); Meteoric; Glacial 1: Deeper Saline component. 
Main reactions: Ion exchange, ptn. of calcite, redox and microbial reactions. 
Redox conditions: Reducing.

Water type C: Saline 19-15 g/L TDS; Δ18O = -11.5 to -13.6‰ SMOW (only 3 samples); Na-Ca-CI to Ca-Na-CI; Glacial - Deeper Saline mixture. 
Main reactions: Ion exchange, microbial reactions. 
Redox conditions: Reducing.

Water type D: Strongly saline > 20 g/L TDS; Ca-Na-CI; Deep saline origin (Field observations). 
Main reactions: Long term water rock interactions. 
Redox conditions: Reducing.

Laaksoharju et al. (2008)
Disposal concept

Onkalo, Finland

https://spectrum.ieee.org/energywise/energy/nuclear/nuclear-waste-deep-storage-plans-approved

Forsmark, Sweden

https://www.wsj.com/articles/a-100-000-year-tomb-for-finlands-nuclear-waste-1485253831

Emplacement in tunnel boreholes (KBS-3 concept)
(modified from SKB, 2011)
Post-closure safety strategy

**Objectives:** Advance understanding of long-term disposal of used fuel in crystalline rocks (granitic or metamorphic rocks) and develop experimental and computational capabilities to evaluate various disposal concepts in such media.

- Assist the geologic disposal safety assessment (GDSA) team to develop a robust repository performance assessment model.
- Provide the GDSA with a basic “minimal” set of process models and model feeds to support the GDSA.
- Develop basis for process modeling that enables streamlined integration with system modeling resulting in feeds to GDSA.
- Consolidate model parameter data, especially thermodynamic data, to ensure more consistent usage of the data across the project.
- With the existence of different approaches taken by various researchers there is a need to understand how well the models are developed in terms of pedigree and rigor.
- Fully leverage international collaborations for data collection and model development and validation.
- Closely collaborate with other work packages, especially those on disposal in argillite and engineered barrier system design.

The current work focuses on: (1) better characterization and understanding of fractured media and fluid flow and transport in such media, and (2) designing effective engineered barrier systems (EBS) for waste isolation.
Technical gaps and priorities mapped to R&D roadmap

- Fuel matrix degradation model (FMDM). Account for the effect of metal corrosion (jointed with argillite work package) (ANL). (H: D-05, E-14)
- Radionuclide interactions with corrosion products, especially Pu sorption and incorporation into magnetite and green rust (LLNL). (H: D-05, E-14)
- Bentonite erosion and colloid generation and their impact on radionuclide transport (LANL). (H: C-15, M-H: E-20)
- Fluid flow in low-permeability media (SNL, LBNL). (H: I-08, M-H: C-11)
- Long-term (up to months) temperature-controlled (up to ~200°C) flow and mechanical (and chemical) experiments on multiple core-scale samples; radionuclide interaction with bentonite (SNL, LBNL). (H: C-15, M: C-08)
- New-generation buffer materials/waste package materials; understanding thermal limits of buffer materials (SNL). (H: C-15, C-16, E-11, E-17)
- Discrete fracture network (DFN) model; especially a reduced order model for GDSA (LANL). (M-H: C-01, P-02)
- Workflow for field data synthesis and flow modeling in fractured media (SNL). (M-H: C-01, M-H: C-13, P-02)
- Geophysical and well-testing techniques for characterizing fractures and inflows; uncertainty reduction of key flow parameters in the EDZ (LBNL). (M-H: E-03)
Integrated experimental & modeling activities for used fuel disposition in crystalline rocks

Disposal systems

Processes

Models

Laboratory Experiments & Field Tests

DECOVELAX = DEvelopment of COupled models and their VALidation against Experiments
DRZ = Disturbed rock zone
RN = Radionuclide
WF = Waste form
WP = Waste package
State of knowledge of process models

- Regional flow (topography, climate changes)
- EBS&DRZ thermal & hydrologic environments
- Buffer & backfill material performance
- RN transport through backfill
- RN transport through DRZ

EBS chemistry
- Waste package degradation
- RN release from WPs
- Flow & transport through access ramps/shafts

In-package chemistry
- Waste form degradation
- RN mobility & solubility
- RN release to biosphere
- Flow & transport through soil

Thermal pulse
Full saturation of buffer materials
- Failure of copper-shelled waste package
- WF degradation; RN release & transport

KBS-3 Overpack
- Failure of overpack
- WF degradation; RN release & transport

Manufacturing defects of Waste package
- WF degradation; RN release & transport

10^0 10^1 10^2 10^3 10^4 10^5 10^6 year

DRZ = Disturbed rock zone
RN = Radionuclide
THMC = Thermal-hydrologic-mechanical-chemical
WP = Waste package

- EBS = Effective Body System
- DRZ = Disturbed rock zone
- RN = Radionuclide
- THMC = Thermal-hydrologic-mechanical-chemical
- WP = Waste package
Waste form degradation

Fuel matrix degradation model
(Wang et al., 2020)

Nonlinear glass corrosion model
(Wang et al., 2016)
Three-electrode electrochemical cell

Wang et al. (2020)
Lead/lead-alloy as a corrosion-resistant outer layer packaging material

- Requirements
  - Longevity: >1000 years ✓
  - Avoid any detrimental impacts on other EBS materials. ✓
  - Retrievability ✓
  - Radiation shielding ✓
  - Reasonable structural strength (tensile strength 70 MPa for alloy) ✓
  - Availability ✓
- Lead
  - Good resistance in sulfide environments
  - $0.87/lb
  - RCRA: Already present as fission product

Passivation

Lead/lead alloy is much more stable than copper in the presence of H2S.
Development of next-generation buffer materials for harsh environments

Leverage materials science and engineering for engineered material development.

Saponite is more stable than Na-montmorillonite in alkaline and high temperature environments.
Thermal-hydrologic-mechanical-chemical (THMC) modeling of buffer materials

- **Figure 1**: Diagram showing a cross-section of a tunnel with labeled components such as ground surface, model, heat-releasing waste package, monitoring point D in rock 10 m above tunnel, and emplacement tunnels. The diagram also shows the distribution of volume fraction change over time for smectite A and B under different temperature conditions.

- **Graphs**:
  - Graph showing volume fraction change over time for smectite A with lines indicating high T, base, low T, base, and no heat.
  - Graph showing volume fraction change over time for smectite B with similar lines.

- **Legend**:
  - High T, base: solid black line
  - Low T, base: red line
  - No heat: dashed line

**Notes**:
- The graphs display the influence of temperature on the volume fraction change of smectite materials over extended periods.
Radionuclide transport through buffer materials: 
Pu interaction with goethite and clay

Figure 2-11. Both PuO$_2$ aggregates (blue) and dispersed Pu$_4$O$_7$ (red) on the goethite surface were observed at 25°C in high concentration samples. 8900 ppm Pu on goethite.

Kersting et al. (2012)

Figure 3-7. Summary of Pu sorption data at 25°C (green points) and 80°C (purple points). Shift between the 25 and 80°C isotherm is indicative of increasing Kd with temperature. Shaded zone is the reported total Pu concentration in equilibrium with PuO2(am, hyd) at pH 8. MDL – method detection limit.
Colloid-facilitated transport model and buffer material erosion

Multiple site sorption model

Figure 2-11. Model matches to the extraction breakthrough curves of test 08-01.

Multiple column experiments for interrogating radionuclide sorption parameters

Reimus et al. (2017)
Iodine-129 interaction with clays

Altmann, 2008

<table>
<thead>
<tr>
<th>Clay Mineral</th>
<th>Column $K_d$ Value (mL/g)</th>
<th>Batch $K_d$ Value (mL/g)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Opalinus (illite)</td>
<td>0.008-0.02</td>
<td></td>
<td>Van Loon et al., 2003</td>
</tr>
<tr>
<td>Montmorillonite</td>
<td>0.57</td>
<td></td>
<td>Sato et al., 1992</td>
</tr>
<tr>
<td>Callovo-Oxfordian (Interstratified illite/smectite)</td>
<td>0.15-0.37</td>
<td></td>
<td>Bazer-Bachi et al., 2006</td>
</tr>
<tr>
<td>Illite</td>
<td>27.7</td>
<td></td>
<td>Kaplan et al., 2000</td>
</tr>
<tr>
<td>Montmorillonite</td>
<td>-0.33</td>
<td></td>
<td>Kaplan et al., 2000</td>
</tr>
</tbody>
</table>

Iodide interacts with negatively charged clay interlayers through ion pairing induced by nanoconfinement.
Far-field flow and transport: Development of discrete fracture network model

Hadgu et al. (2017)
Development of a workflow for synthesizing field data into a fracture network model

- It is important to condition fracture network generation on actual fracture distribution (location, size) in tunnel and borehole.
- Statistical stability of fracture networks?
Technology for site characterization and monitoring: Disturbed rock zone (DRZ) characterization

Dual-sample triaxial rock testing system

Granite core samples from Grimsel, Switzerland

Rigid-body-spring network model for simulating fracture patterns
Fracture characterization and field monitoring

➢ The challenge of groundwater monitoring in fractured rocks is to design a system that captures sufficient number of particles.
➢ Technologically, this challenge is related to the ability for fracture characterization.
➢ High-resolution geophysical techniques are highly desirable.

<table>
<thead>
<tr>
<th>Relization</th>
<th>12M133</th>
<th>Upper Well</th>
<th>Lower Well</th>
</tr>
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<tbody>
<tr>
<td>5</td>
<td>1</td>
<td>3</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>9</td>
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<tr>
<td>10</td>
<td>5</td>
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<td></td>
</tr>
<tr>
<td>25</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

500 particles evenly distributed within the tunnel at time 0.

Step-rate Injection Method for Fracture In-situ Properties (SIMFIP) system
Current status of process models and total system integration

- Regional flow (topography, climate changes)
- EBS&DRZ thermal & hydrologic environments
- Buffer & backfill material performance
- RN transport through backfill
- RN transport through DRZ
- EBS chemistry
- Waste package degradation
- RN release from WPs
- RN release to biosphere
- Flow & transport through access ramps/shafts
- Flow & transport through far field
- In-package chemistry
- Waste form degradation
- RN mobility & solubility

Significant level of understanding achieved
Some level of understanding achieved
Work initiated
Next steps

- Develop a sensible GDSA model for sensitivity analyses.
  - Provide a minimum set of process models to GDSA
- Move model development more towards model validation with real data.
- Develop reduced order models for incorporation into the GDSA model.
- Continue with buffer material development.
- Develop and refine engineered barrier system (EBS) models, especially waste package (WP) degradation models.

Towards a more realistic perception (then representation) of fluid flows in crystalline rocks:
Crystalline rocks are generally quite impermeable.
References

- Wang et al. (2016) Nonlinear dynamics and instability of aqueous dissolution of silicate glasses and minerals, Scientific Reports, 6:30256; DOI: 10.1038/srep30256
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