Argillite Host Rock: Reference Disposal Concept(s) and Key Considerations

Spent Fuel and Waste Science and Technology (SFWST)

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Outline

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• Knowledge Gaps & R&D Priorities
• Repository Relevant Processes
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Argillite/Shale Repository Concept

High-Level radioactive waste disposal (ANDRA) – COx Argillite (Bildstein and Claret 2015)

Swiss repository concept (Delage et al. 2010) - Opalinus Clay


Shale Attributes

- Low permeability / hydraulic conductivity
- Low diffusion coefficients
- Good retention capacity for radionuclides
Porewater Chemistry in Clay Formations

- Porewater compositions are highly variable
- Overall: Na-Cl brines with some Ca & carbonate

Sources: United States Geological Survey (USGS) produced water (Blondes et al. 2018); NATCARB (Bauer et al. 2018); WATSTORE (von Damm 1987)

Stein et al. (2020)
R&D Priorities – Argillite (Shale)

UZ = Unsaturated Zone
DPC = Dual Purpose Canisters
EBS = Engineered Barrier System
GDSA = Geologic Disposal Safety Assessment
Knowledge Gaps & R&D Priorities

Some Activities With Medium-High Score in 2019 R&D Roadmap Update

- **High-Temperature Behavior** - Chemical processes still under development, particularly at elevated temperature conditions
- **EBS High Temperature experimental data collection** - To evaluate high temperature mineralogical/geochemical changes
- **Analysis of clay hydration/dehydration and alteration** under various environmental conditions
- **Buffer/backfill dry-out and resaturation process**
- **THC processes in EBS** - High importance for design/construction arguments affecting disposal system design that utilize backfill-buffer as an engineered barrier
- **Argillite Coupled THM processes modeling** including host rock, EBS, and EDZ
- **Cement plug/liner degradation; Evaluation of ordinary Portland cement (OPC)**
Repository Phases and Relevant Processes: Cross-Cuts With International Partnerships

**Features:**
- Biosphere
  - Aquifer
- Host Rock (e.g., salt, clay, or granite)
- Disturbed Rock Zone (DRZ)
- Backfilled Drift

**Processes:**
- Biosphere (Aquifer, Receptor Well)
  - Dilution
  - Irrigation
  - Water Consumption
  - Dose Conversion Factors
- Far Field (NBS) (Host Rock, Interbeds)
  - Flow & Transport
  - Coupled Processes
  - Sorption
  - RN Decay and Isogrowth
- Near Field (EBS = DRZ) (Backfill, Shaft Seals, DRZ)
  - EBS & DRZ Evolution
  - Chemical Interactions
  - Thermal Effects
  - Mechanical Effects
  - Flow and Transport
  - Coupled Processes

**Events:**
- Seismic:
- Igneous:

**Key R&D Issues:**
- Near-Field Perturbation
- Engineered Barrier Integrity
- Flow and Radionuclide Transport
- Demonstration of Integrated System Behavior

**Abbreviations:**
- ALC = Full-scale Emplacement Experiment (France)
- BATS = Heated Brine Availability Test in Salt (USA)
- BRIE = Bentonite Rock Interaction Experiment (Sweden)
- FEBEX = Full-Scale Engineered Barrier Experiment (Switzerland)
- CFM = Colloid Formation Migration (Switzerland)
- TED = Thermal Experiment (France)
- FE = Full-scale Emplacement Experiment (Switzerland)
- HE-E = Heater Experiment in Micro-tunnel (Switzerland)
- HotBent = High-Temperature Heater Test (Switzerland)
- HLW = High Level Waste
- LTDE = Long-Term Diffusion Sorption Experiment (Sweden)
Argillite Reference Case

Stein et al. (2017)

Evaluation of disposal design concepts
- Thermal management in clay/shale repository
  - Waste package and drift spacing
  - Coupled Multiphase transport phenomena
Argillite Reference Case: Deterministic Simulations of Generic Disposal in Shale

Generic stratigraphic column for shale reference case

24-PWR = 24 SNF Pressurized Water Reactor Assemblies
drz = disturbed rock zone

Sevougian et al. (2019)
Highlights – Disposal in Argillite R&D

- **High temperature** experiments of bentonite interactions with barrier materials and host rocks: granodiorite & Opalinus Clay
- **Development of a preliminary GDSA reference case** for disposal in argillite media
- **Advances in thermal-hydrological-mechanical-chemical (THMC) modeling approaches** of bentonite barrier, argillite rock, and excavated disturbed zone (EDZ; fracture/damage behavior) & gas migration
- **Thermodynamic modeling** of bentonite – barrier material interactions & thermodynamic database development
- **Non-isothermal 1D-3D thermal-hydrological-chemical (THC) reactive transport modeling**
- **International collaborations:**
  - DECOVALEX19: PFLOTRAN hydrological-chemical (HC) modeling of barrier interactions
  - DECOVALEX2023: Gas transport in clays (just started!)
Past Experiments: Steel – Clay Interactions

**Experiment**
- $T = 300^\circ\text{C}$; STRIPA brine
- Wyoming Bentonite
- 316 & 304 stainless steel (SS)

**Corrosion products**
- Uniform corrosion (no pitting)
- Chromite passivation layer
- Fe-rich smectite (Fe-saponite), Chlorite
- Pentlandite ($\text{Fe}_9\text{Ni}_3\text{S}_8$)
- Millerite ($\text{NiS}$)

**Chemical Reaction**
$$
\text{Fe}^{1.22}\text{Cr}^{0.37}\text{Ni}^{0.22}(\text{SS}) + 2\text{O}_2 = 
\text{FeCr}_2\text{O}_4(\text{M}) + 5.60\text{Fe}^{4+} + 
1.19\text{Ni}^{3+} + 13.58\text{e}^- 
$$
$$
4.5\text{Fe}^{2+} + 4.5\text{Ni}^{2+} + 8\text{HS}^- + 2\text{e}^- = 
(\text{Fe,Ni})_9\text{S}_8(\text{pent}) + 8\text{H}^+ 
$$

**Pourbaix Diagram**
Thermodynamic modeling and database development

Cheshire et al. 2014, 2018
Barrier Material Interactions: Bulk Mineralogy Changes – Quantitative X-ray Diffraction (Q-XRD) Analysis

- **Opalinus Clay ± Wyoming Bentonite**
  - 300°C (6 months): Zeolite formation in clay and along cracks and edges on the Opalinus Clay fragments, plagioclase
  - 200°C (8 weeks): 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
  - Reduction in clay swelling
  - Amorphous material (gel?)

Sauer et al. (2019)
Thermo-Hydrological-Mechanical (THM) Processes in Clay URL Experiments and Simulation

Continuum model approach using TOUGH-FLAC
Discrete fracture model approach using TOUGH-RBSN

LBNL for modeling gas migration through clay associated with DECOVALEX-2019

Plan view of Mont Terri FE experiment – Opalinus Clay
Rutqvist et al. (2020)
Non-isothermal 1D-3D Thermo-Hydrological-Chemical (THC) reactive transport modeling

- Waste canister length: 4.7 m
- 12-PWR assemblies
- 50-year storage time

Evaluation of thermal effects on fluid/solid interactions
- Chemical reactions – mineral dissolution/precipitation
- Changes in bulk mineralogy
- Evaluate changes in porosity/permeability

Ho et al. (2019)
Section 49 Sampling

Bentonite Thermal Behavior

- Bentonite dehydration behavior is a function of the duration of hydration that precedes it.
- Appearance of a “shoulder peak” during dehydration suggests different energetics for swelling clay hydration and dehydration.
- Ideal for the thermal study of bentonite with additives.

Jove Colon et al. (2019)
3D Reactive Transport Simulations using PFLOTRAN simulation code

Focus: Shotcrete – groundwater interactions in the CTD
DECOVALEX19: 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 kinetic rate law parameters for various cement phases and volume fraction of mineral components
- Simulations have been conducted to evaluate the effect of shotcrete thickness effects

Jóve Colón et al. (2020).
Summary

• Development of a high temperature argillite reference case
  - Need to further disposal concepts for DPC’s, EBS design options (e.g., thermal management), and post-closure strategies

• Bentonite-metal-cement-Opalinus Clay interactions:
  - Reactions produces zeolites and with some swelling reduction in smectite as a result of interactions with alkaline solutions
  - Future Work: Study effects of host rock composition & other barrier materials (e.g. cement); expand 3D non-isothermal model to various waste packages

• DECOVALEX HC (GREET) modeling and Thermal Analyses on FEBEX-DP Bentonite:
  - 3D reactive transport model of shotcrete interactions in CTD experiment represent overall chemical trends
  - Cyclic thermal analysis (hydration/dehydration) experiments show reproducible results between cycles with slower dehydration rates
  - Future Work: Investigate hydrological-chemical (HC) model sensitivities to shotcrete thickness; expand cyclic thermal analyses & X-ray diffraction (XRD) methods to evaluate high temperature effects; maintain engagement with international programs (DECOVALEX2023; EBS Task Force)
References


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