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Fluid-Rock Processes Driving Isolation of

Crustal Fluids in Crystalline Basement Systems

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Presentation Overview



- Deep Borehole Field Test (DBFT) Background
 - The Deep Borehole Disposal (DBD) Concept
 - Feasibility is being evaluated
 - DBFT Overview Science and Data to Evaluate DBD Concept
 - Current Project is Being Wrapped Up
 - Geoscience Guidelines for DBFT Site
 - Considered Characteristics
 - Crystalline Basement Characteristics/Conceptual Profiles
- Evaluation of Fluid-Rock Reactions in Crystalline Basement
 - Fluid-Rock Reaction Concepts and Geochemical Description
 - General Observations and Results
- Summary and Conclusions

Deep Borehole Disposal (DBD) Concept



- Deep borehole disposal of high-level radioactive waste has been considered in the U.S. and elsewhere since the 1950s and has been periodically studied since the 1970s
- Current DBD concept consists of drilling a borehole (or array of boreholes) into crystalline basement rock
 - Total depth about 5,000 m depth
 - Lower 3000 m in crystalline basement
 - Waste canisters would be emplaced in the lower 2,000 meters
 - Upper crystalline basement portion would be sealed with compacted bentonite clay, cement plugs, and cemented backfill
 - At least 1000 m
 - Upper borehole filled/sealed

Why Consider Deep Borehole Disposal?



- Potential for Robust Isolation
- Gives DOE the Flexibility to Consider Options for Disposal of Smaller Waste Forms in Deep Boreholes
 - Potentially earlier disposal of some wastes than might be possible in a mined repository
 - Possible reduced costs associated with projected treatments of some wastes
- Several DOE-managed Small Waste Forms are Potential Candidates for Deep Borehole Disposal (SNL 2014), e.g.,
 - 1,936 cesium and strontium capsules stored at the Hanford Site
 - Untreated calcine HLW currently stored at INL in sets of stainless steel bins within concrete vaults

Deep Borehole Disposal Concept – Safety and Feasibility Considerations



Long-Term Waste Isolation (hydrogeochemical characteristics)



Waste emplacement is deep in crystalline basement

- At least 1,000 m of crystalline rock (seal zone) overlying the waste disposal zone
 - Crystalline basement within 2,000 m of the surface is common in many stable continental regions
- Crystalline basement can have very low permeability
- limits flow and transport

Deep groundwater in the crystalline basement:

- Can have very long residence times isolated from shallow groundwater
- Can be highly saline and geochemically reducing enhances the sorption and limits solubility of many radionuclides
- Can have density stratification (saline groundwater underlying fresh groundwater) opposes thermally-induced upward groundwater convection

Deep Borehole Field Test (DBFT) Overview



- Assess DBD Feasibility Via Field Study of Site and Handling
 - NOTE: Project is Being Closed-out this Fiscal Year
- Construct Two 5-km Boreholes
 - Characterization Borehole (CB):
 - Field Test Borehole (FTB):
- Evaluate our Ability to:
 - Drill deep, wide, straight in crystalline rocks (CB + FTB)
 - Characterize bedrock via geophysics (CB)
 - Conduct tests in basement ≤150°C & 50 MPa (CB)
 - Collect geochemical profiles (CB)
 - Emplace/retrieve surrogate waste packages (FTB)

- 21.6 cm [8.5"] @ Total Depth
- 43.2 cm [17"] @ Total Depth

Geoscience Guidelines Considerations

- Crystalline Basement
 - Depth
 - Rock Fabric & Stress State
 - Regional Structure(s)
 - Hydrology and Geochemistry
- Heat Flow
- Recent Seismicity/Volcanism
- Resources
- Anthropogenic Contamination

Depth to Basement – National Scale



Distribution of crystalline basement at a depth of less than 2 km (tan shading) and granitic outcrop (red) in the contiguous US (from Figure 3-2 in Perry et al., 2015)



Observed Profiles





Fluid-Rock Reaction Evaluations



- Analyses for generic fluid-rock reaction systems in crystalline basement
 - Evaluate mechanisms in the crystalline basement to form deep, isolated brines
 - Reaction path models for granite mineral reactions with seawater
 - Alteration mineralogy hydrous phases (H₂O sinks)
 - Evolved brine compositions (major elements, Cl, Br)
 - Fluid inclusion contributions (soluble salts) considered
 - Calculating leachate compositions from Black Forest crystalline basement rocks
 - Conditions Comparable to ~ 5 km depth
 - Generic Granite Composition(s)
 - Seawater Starting Brine Composition
 - ~100 150°C, P_{sat}
 - PHREEQC Reaction Path Calculations

Hypothetical Granite



- 20% Quartz; 40% K-feldspar; 15% Plagioclase (Albite); 9% Muscovite; 8% Biotite; and 8% Hornblende (volume %)
- Represented as a 10 kg (3.8 L) block having a molar mixture of
 - 33.3 moles Quartz: 14.4 moles K-feldspar: 5.7 moles Albite: 2.2 moles Muscovite: 1.8 moles Biotite: 0.9 moles Hornblende
- Granite is "reacted" with 0.1 liter of seawater at 100°C.
 - This is a 38:1 rock:fluid ratio by volume, equivalent to a rock with a fluid-filled porosity of ~ 3%

General Observations



- Calculated Generic Granite Hydrologic Alteration Results
 - Reaction creates Albite + K-feldspar + Chlorite + Laumontite + Brine
 - Minor amounts (< 0.02 moles) of epidote, calcite, and gypsum form
 - Albite and K-feldspar masses increase substantially
 - Almost all of the quartz is dissolved.
 - Produces a residual Ca-Na-Cl brine at pH of 6.8
 - Net Loss of water causes the ionic strength of the solution to increase
 - From an initial ionic strength of 0.6 upwards to > 5 molal
 - The Ca/Na calculated for brine is 1.55
 - Low Mg concentration
 - End-member Canadian Shield brines from Frape et al. (1984) with highest salt contents of ~240 – 325 g/L
 - Have ionic strengths of 4.5 6.2
 - 0.7 < Ca/Na < 3</p>
 - Low Mg concentration

Solution and Mineralogic Evolution 🖻



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Summary and Conclusions



- Planned DBFT Project is Ending this Fiscal Year
- Many Sites within U.S. with Functional Geology
 - Multifaceted Objectives of DBFT Provide Opportunities for Success
 - Choosing Any Site would be based on Uncertain Geologic Information
 - Generally Regions Lacking Exploration
 - Each Site will have its own Geologic Challenges
 - Would Provide Substantial Direct Data and Understanding
 - Characterization methods
 - Feasibility of implementation
- Geochemical Processes in Generic Crystalline Basement Appear to be Able to Affect/Control Fluid System Isolation
 - Still to evaluate
 - Sensitivity of the PHREEQC calculations to input water chemistry
 - More thorough comparison of predicted/observed alteration
 - More detailed consideration of activity coefficient effects



Backup Materials



Deep Geologic Disposal Remains an Essential Element of Nuclear Waste Management

"The conclusion that disposal is needed and that deep geologic disposal is the scientifically preferred approach has been reached by every expert panel that has looked at the issue and by every other country that is pursuing a nuclear waste management program." Blue Ribbon Commission on

America's Nuclear Future, 2012













Deep Borehole Field Test Objectives



- The RD&D objectives for deep borehole disposal are being met with a borehole field test that is conducted to a depth of 5 km in a suitable location (without emplacement of radioactive wastes)
- The DBFT includes the following major activities:
 - Obtain a suitable test site
 - Design, drill and construct the Characterization Borehole (8.5" diameter) to requirements
 - Collect data in the Characterization Borehole to characterize crystalline basement conditions and evaluate expected hydrogeochemical conditions

Accommodate a subsequent Field Test Borehole (17" diameter)

- Design, drill and construct the Field Test borehole to requirements
- Design and develop surface handling and emplacement equipment systems and operational methods for safe canister/waste package handling and emplacement

Preferred Geologic Conditions



- Geohydrological Considerations
 - No large-scale connected pathways from depth to aquifer systems
 - No through going fracture/fault/shear zones that provide fast paths
 - No structural features that provide potential connective pathways
 - Low permeability of crystalline basement at depth
 - Urach 3: (Stober and Bucher, 2000; 2004)
 - ~10⁻¹⁹ m² (intact rock); ~10⁻¹⁴ to 10⁻¹⁷ m² (bulk: parallel to or across shears)
 - Decreasing with Depth
 - Evidence of ancient, isolated nature of groundwater
 - Salinity gradient increasing downward to brine at depth (Park et al., 2009)
 - Limited recharge/connectivity with surface waters/aquifers
 - Provides density resistance to upward flow
 - Major element and isotopic indication of compositional equilibration with rock
 - Crystalline basement reacting with water (Stober and Bucher, 2004)
 - Ancient/isolated groundwater
 - » Ages isotopes, paleoseawater (Stober and Bucher, 2000)
 - » Radiogenic isotopes from atmosphere lacking: ⁸¹Kr, ¹²⁹I, ³⁶Cl
 - » Radiogenic isotopes/ratios from rock: ⁸¹Kr, ⁸⁷Sr/⁸⁶Sr; ²³⁸U/²³⁴U
 - Noble gases (⁴He, Ne) & stable isotopes (²H, ¹⁸O) compositions from deep water: (e.g., Gascoyne and Kamineni, 1993)

Preferred Geologic Conditions (Continued)



- Geochemical Considerations
 - Reduced, or reducing, conditions in the geosphere (rock and water system)
 - Crystalline basement mineralogical (and material) controls
 - Magnetite-hematite buffer low oxygen potential
 - Oxides equilibria => T-low fO₂ paths (e.g., Sassani and Pasteris, 1988; Sassani, 1992)
 - Biotite common Fe⁺² phase (Bucher and Stober, 2000)
 - Rock-reacted fluid compositions water sink (Stober and Bucher, 2004)
 - More rock dominated at depth (Gascoyne and Kamineni, 1993)
 - Stratification of salinity increasing to brine deep in crystalline basement
 - Canadian Shield salinity increases with depth to ~350 g/L TDS; (Gascoyne and Kamineni, 1993; Park et al., 2009)
 - More Ca-rich brines with further reaction with deeper rock
 - Urach 3, Germany, ~70- g/L TDS NaCl brine (Stober and Bucher, 1999; 2004)
 - Subset of waste forms and radionuclides are redox sensitive
 - Lower degradation rates
 - Lower solubility-limited concentrations
 - Increased sorption coefficients
 - Higher salinity
 - Density gradient opposes upward flow
 - Reduces/eliminates colloidal transport

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