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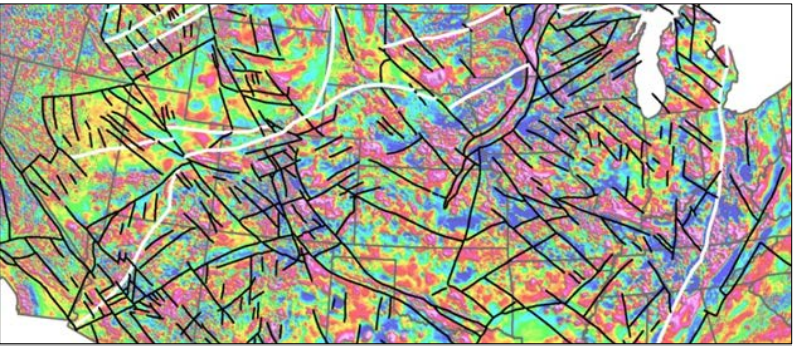

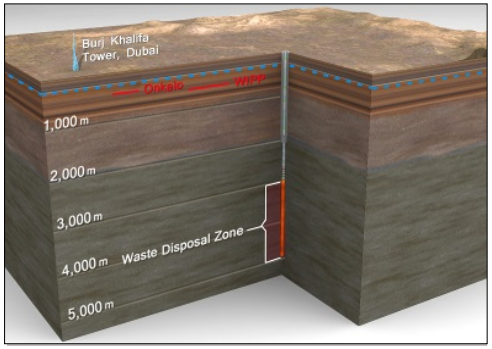

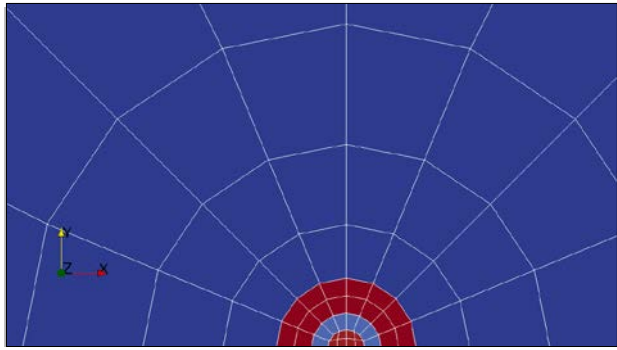

Fluid-Rock Processes Driving Isolation of Crustal Fluids in Crystalline Basement Systems

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Sandia National Laboratories

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U.S. DEPARTMENT OF ENERGY | NNSA National Nuclear Security Administration

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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

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- 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?

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- 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

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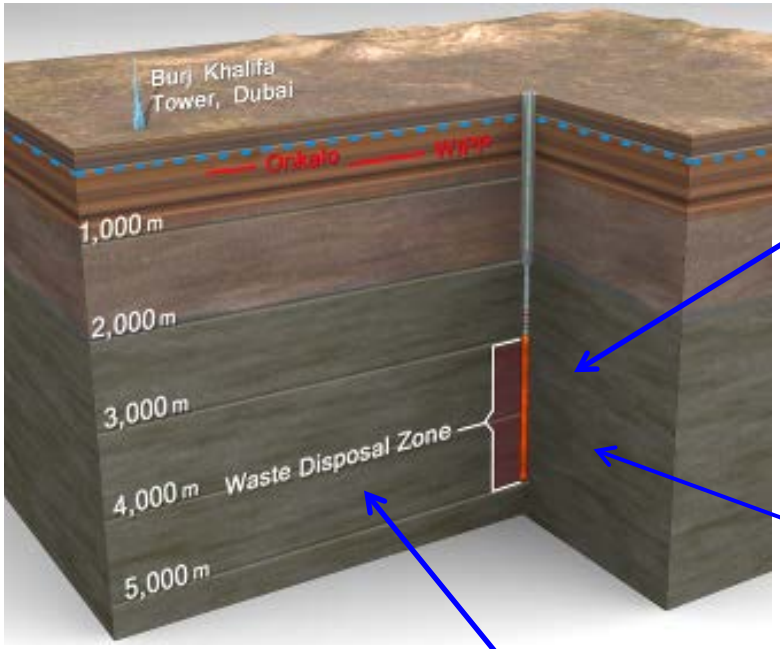
Long-Term Waste Isolation (hydrogeochemical characteristics)



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

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- 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): 21.6 cm [8.5”] @ Total Depth
  - Field Test Borehole (FTB): 43.2 cm [17”] @ Total Depth
- Evaluate our Ability to:
  - Drill deep, wide, straight in crystalline rocks (CB + FTB)
  - Characterize bedrock via geophysics (CB)
  - Conduct tests in basement ≤150^{o}C & 50 MPa (CB)
  - Collect geochemical profiles (CB)
  - Emplace/retrieve surrogate waste packages (FTB)

Geoscience Guidelines Considerations

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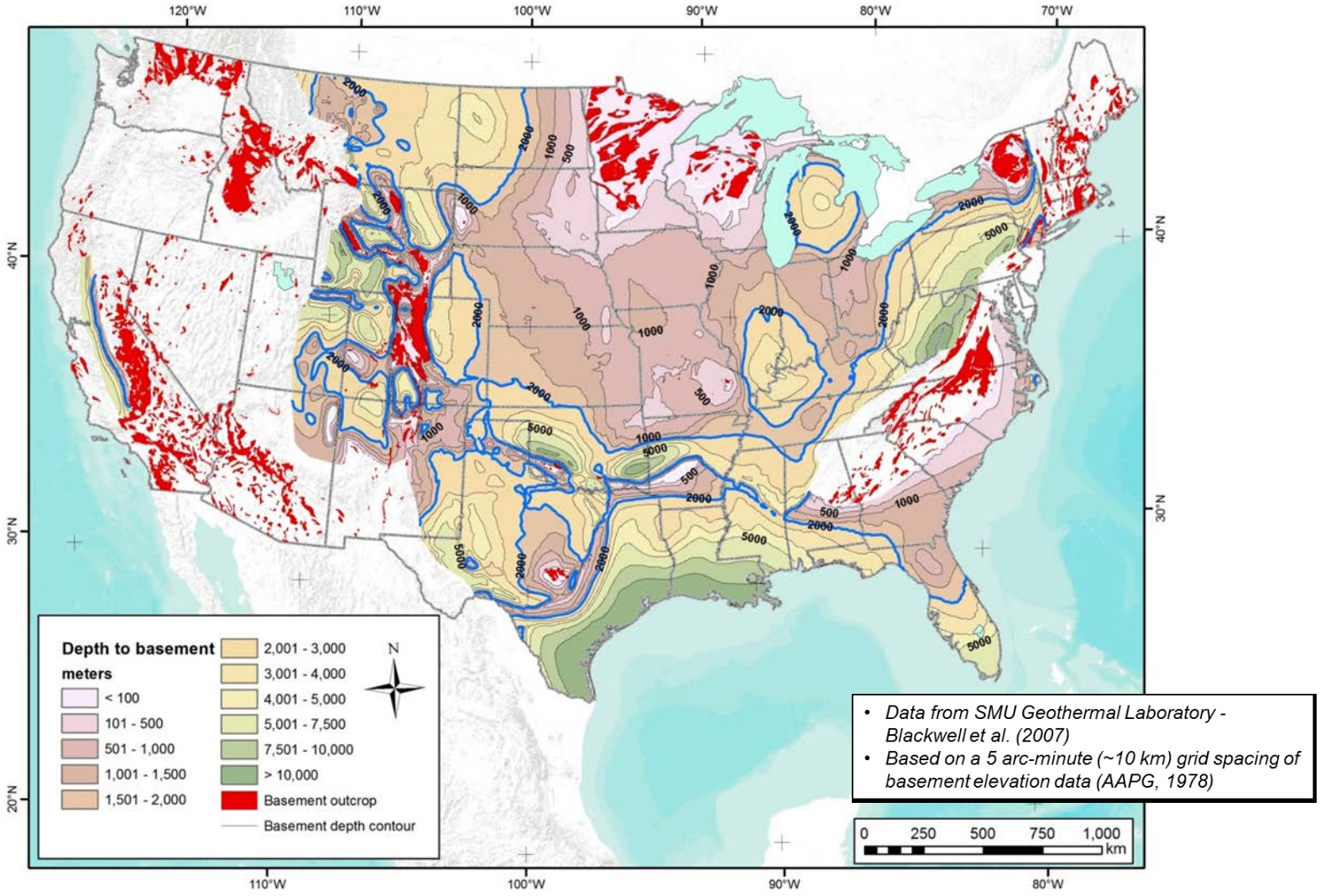
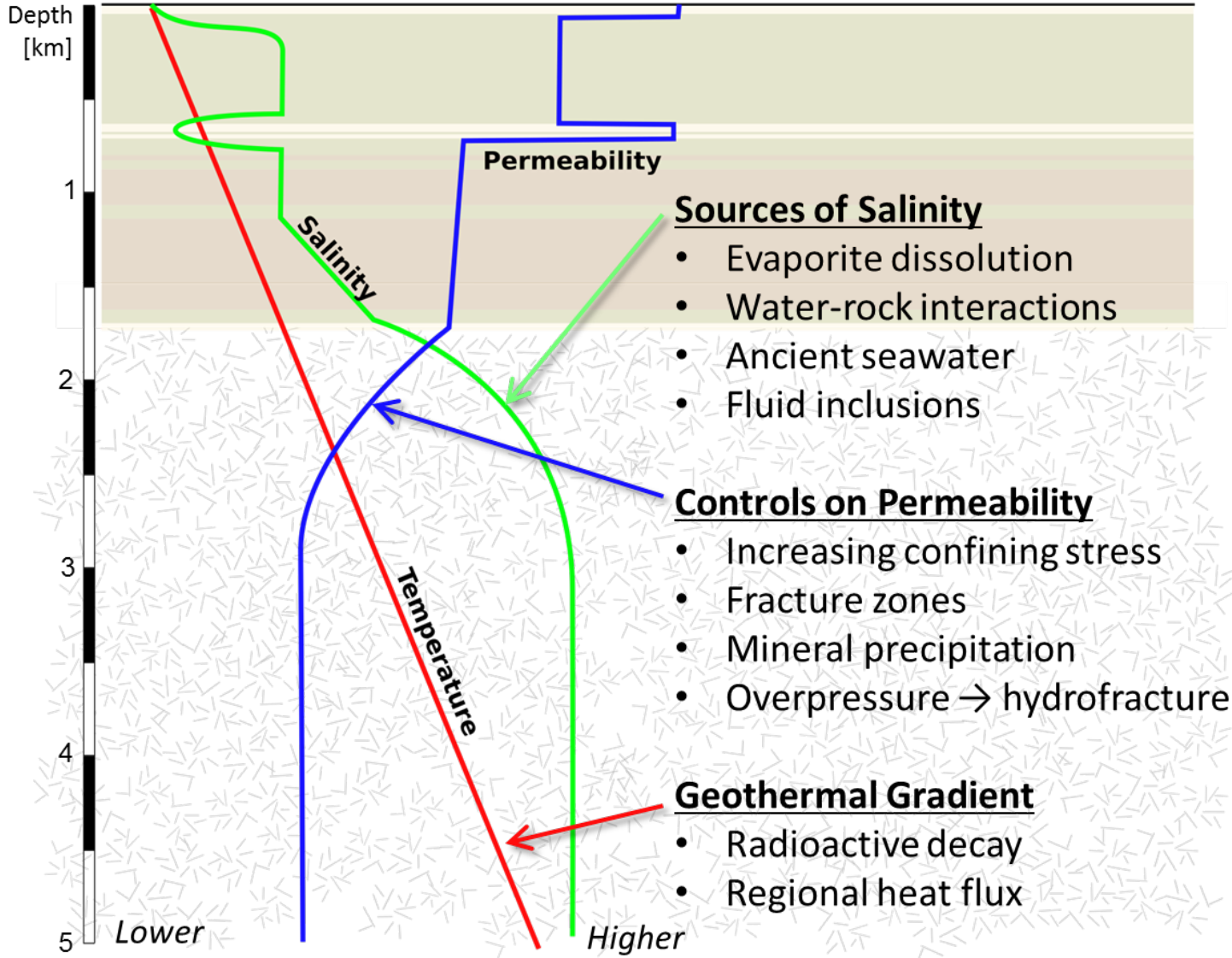
- Crystalline Basement
  - Depth
  - Rock Fabric & Stress State
  - Regional Structure(s)
  - Hydrology and Geochemistry
- Heat Flow
- Recent Seismicity/Volcanism
- Resources
- Anthropogenic Contamination

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)

Deep Borehole Conceptual Profiles



Sources of Salinity
- Evaporite dissolution
- Water-rock interactions
- Ancient seawater
- Fluid inclusions

Controls on Permeability
- Increasing confining stress
- Fracture zones
- Mineral precipitation
- Overpressure → hydrofracture

Geothermal Gradient
- Radioactive decay
- Regional heat flux

Sedimentary Overburden ≤ 2 km

Crystalline Basement ≥ 3 km

Observed Profiles

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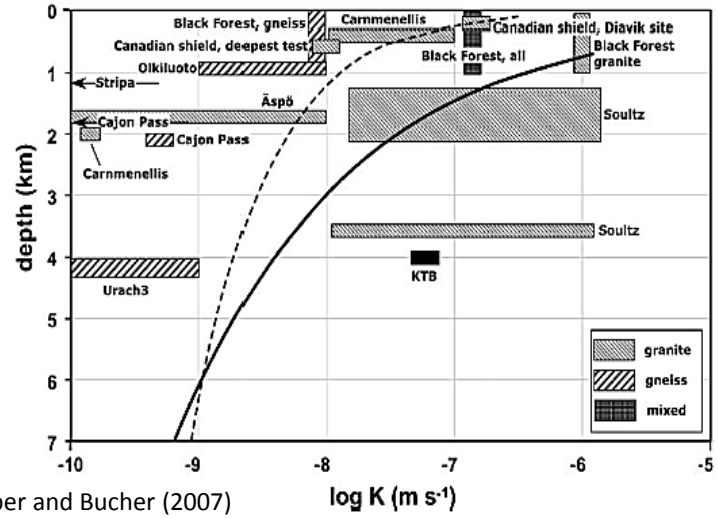
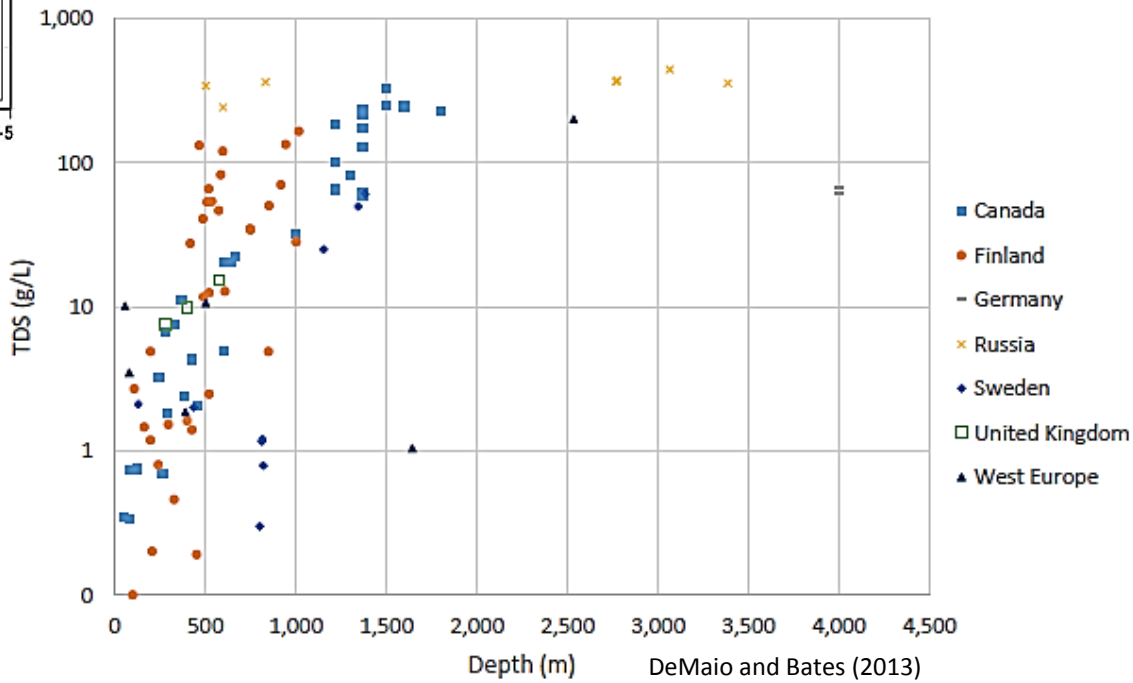
Bulk Permeability Decreases with Depth



Stober and Bucher (2007)

Salinity Increases with Depth



DeMaio and Bates (2013)

How Much of a Role Does Fluid-Rock Reaction Play in Driving Increased Salinity?

Fluid-Rock Reaction Evaluations

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- 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 (H_2O 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^{o}C, P_{sat}
    - PHREEQC Reaction Path Calculations

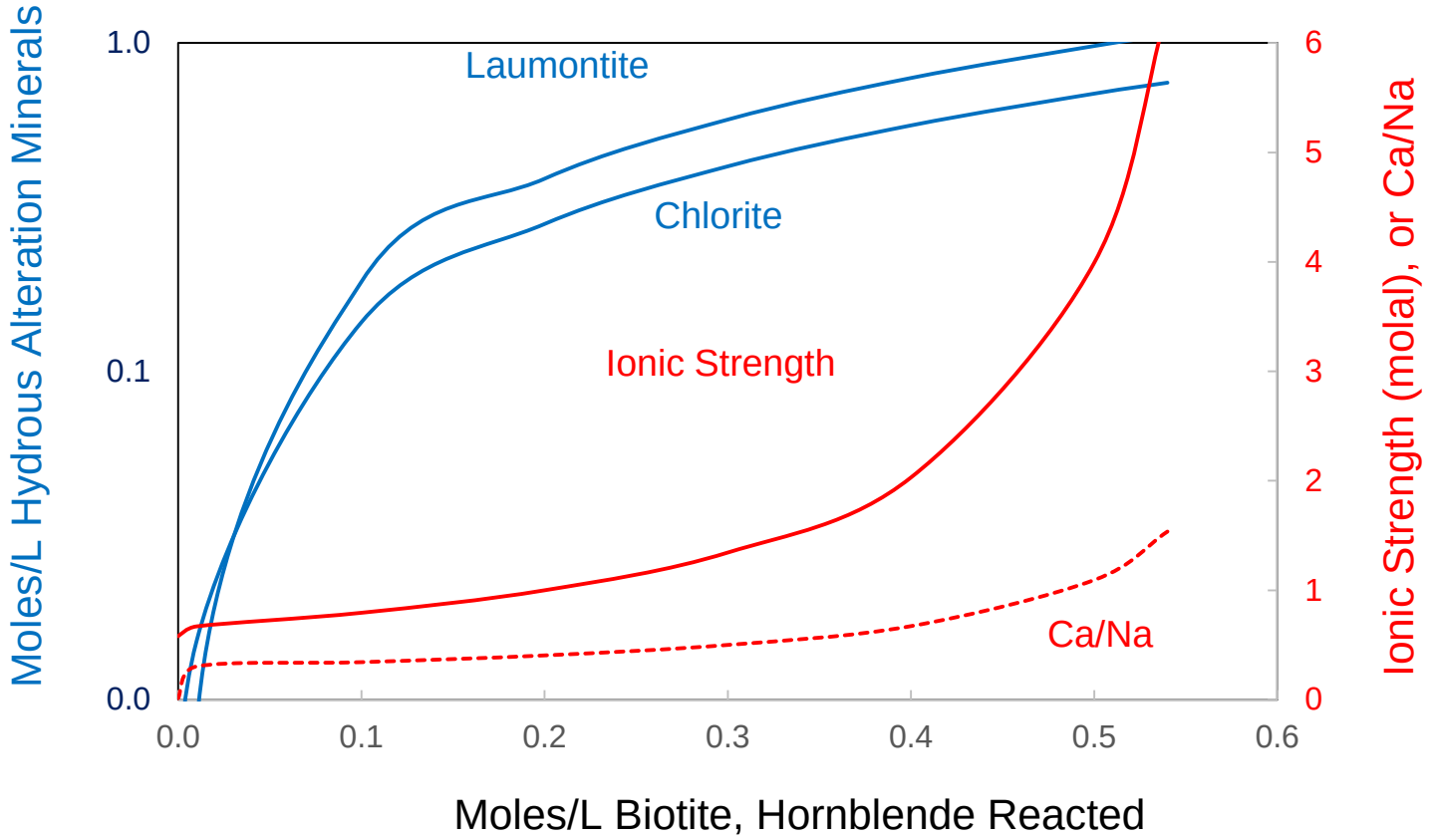
Hypothetical Granite

- 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^{o}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
    - Low Mg concentration

Solution and Mineralogic Evolution



Summary and Conclusions

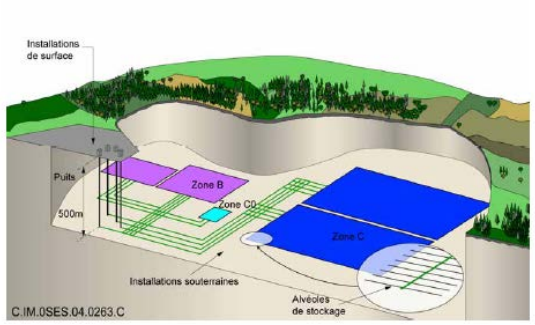
- Planned DBFT Project is Ending this Fiscal Year
- Many Sites within U.S. with Functional Geology
  - Multifaceted Objectives of DBFT Provide Opportunities for Success
  - Choosing Any Site would be based on Uncertain Geologic Information
    - Generally Regions Lacking Exploration
  - Each Site will 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

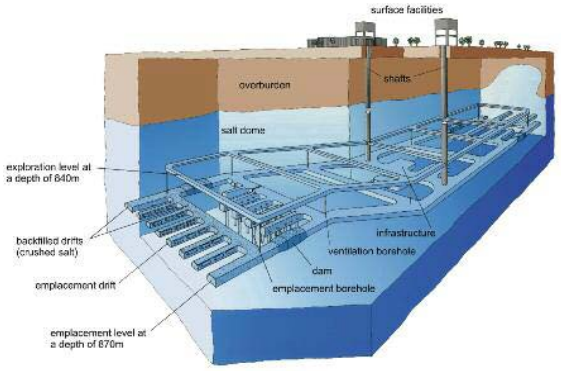
Backup Materials

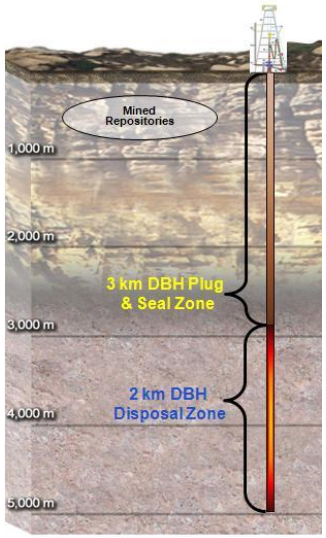
Deep Geologic Disposal Remains an Essential Element of Nuclear Waste Management

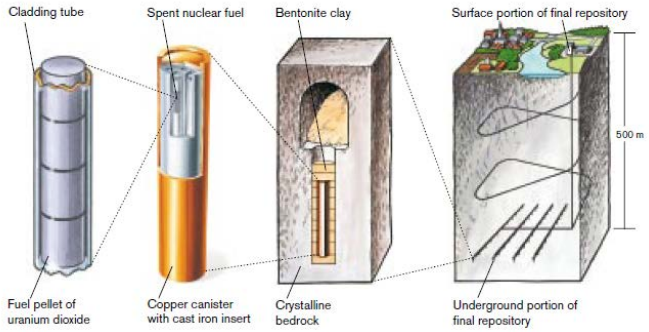
"The conclusion that disposal is needed and that deep geologic disposal is the scientifically preferred approach has been reached by every expert panel that has looked at the issue and by every other country that is pursuing a nuclear waste management program."

Blue Ribbon Commission on America's Nuclear Future, 2012









Deep Borehole Field Test Objectives

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- 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} m^2 (intact rock); ~10^{-14} to 10^{-17} m^2 (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: ^{81}Kr, ^{129}I, ^{36}Cl
        - » Radiogenic isotopes/ratios from rock: ^{81}Kr, $^{87}Sr/^{86}Sr$; $^{238}U/^{234}U$
        - » Noble gases (^{4}He, Ne) & stable isotopes (^{2}H, ^{18}O) compositions from deep water: (e.g., Gascoyne and Kamineni, 1993)

Preferred Geologic Conditions (Continued)

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- 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 fO_2 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

References

AAPG, 1978, Basement map of North America: Am. Assoc. Petroleum Geologists, scale: 1:5,000,000

Blackwell, D.D., P. Negraru, and M. Richards, 2007. Assessment of the enhanced geothermal system resource base of the United States, Natural Resources Research, DOI 10:1007/s11053-007-9028-7.

Bucher K, and Stober I (2000) Hydrochemistry of water in the crystalline basement. In: Hydrogeology of Crystalline Rocks (eds Stober I, Bucher K), pp. 141-75. Kluwer Academic Publishers, Dordrecht.

Bucher, K. and Stober, I. (2002) Water-rock reaction experiments with Black Forest gneiss and granite, Water-Rock Interaction. Springer, pp. 61-95.

Bucher, K. and Stober, I. (2010) Fluids in the upper continental crust. Geofluids 10, 241-253.

DeMaio, W., and Bates, E. (2013). Salinity and Density in Deep Boreholes. Massachusetts Institute of Technology. UROP REPORT: October 29, 2013. 14 p.

Frape, S., Fritz, P. and McNutt, R.t. (1984) Water-rock interaction and chemistry of groundwaters from the Canadian Shield. Geochimica et Cosmochimica Acta 48, 1617-1627.

Gascoyne, M. and Kamineni, D. C. (1993) The hydrogeochemistry of fractured plutonic rocks in the canadian shield. In: Hydrogeology of Hard Rocks, 440- 449. Banks, S. B. and Banks, D. (editors) Geol. Survey of Norway: Trondheim.

Park, Y.-J., E.A. Sudicky, and J.F. Sykes (2009), Effects of shield brine on the safe disposal of waste in deep geologic environments, Advances in Water Resources 32: 1352-1358.

Perry, F., Kelley, R., Houseworth, J., and Dobson, P., 2015. A GIS Database to Support the Application of Technical Siting Guidelines to a Deep Borehole Field Test. FCRD-UFD-2015-000603. LA-UR-15-22397. Los Alamos , NM: US Department of Energy Used Fuel Disposition Campaign.

Sassani, D.C., 1992. Petrologic and Thermodynamic Investigation of the Aqueous Transport of Platinum-Group Elements During Alteration of Mafic Intrusive Rocks, Ph.D. Dissertation, Washington University, St. Louis, University Microfilms.

Sassani, D.C., and Pasteris, J.D., 1988. Preliminary investigation of alteration in a basal section of the southern Duluth Complex, Minnesota, and the effects on the sulfide and oxide mineralization, in G. Kisvarzanyi and S.K. Grant, eds., North American Conference on Tectonic Control of Ore Deposits and the Vertical and Horizontal Extent of Ore Systems, Proceedings Volume, University of Missouri-Rolla, 280-291.

SNL (Sandia National Laboratories), 2014. Evaluation of Options for Permanent Geologic Disposal of Spent Nuclear Fuel and High-Level Radioactive Waste in Support of a Comprehensive National Nuclear Fuel Cycle Strategy (2 Volumes). FCRD-UFD-2013-000371. Albuquerque, NM: US Department of Energy Used Fuel Disposition Campaign.

Stober I, and Bucher K.,1999, Origin of salinity of deep groundwater in crystalline rocks, Terra Nova, 11, p. 181-189.

Stober I, and Bucher K (2000) Hydraulic Properties of the upper Continental Crust: data from the Urach 3 geothermal well. In: Hydrogeology of Crystalline Rocks (eds Stober I, Bucher K), pp. 53-78. Kluwer Academic Publishers, Dordrecht.

Stober I and Bucher K (2004) Fluid sinks within the earth's crust. Geofluids 4(2): 143–151.

Stober, I. & K. Bucher, 2007. Hydraulic properties of the crystalline basement. Hydrogeology Journal, 15:213 224.