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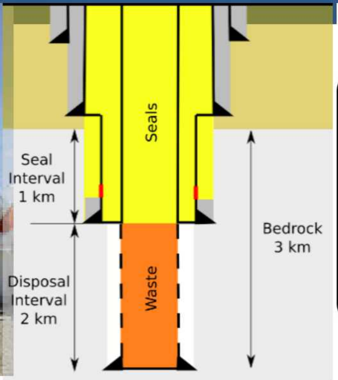
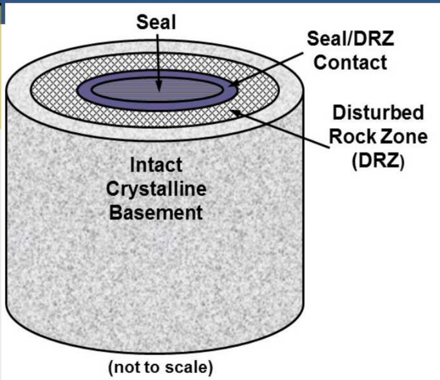
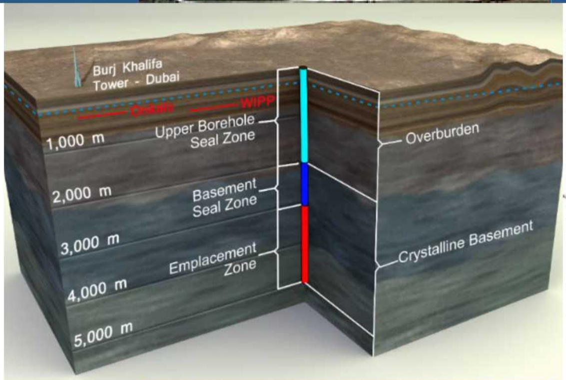
Sandia National Laboratories

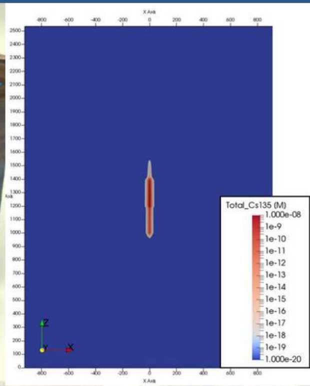
SAND2019-9371C

Evaluating Evolution of Crustal Fluids in Crystalline Basement Systems









Presented by

David Sassani, **Carlos Lopez, Patrick Brady, Kristopher Kuhlman, and Carlos Jove-Colon**

Goldschmidt 2019

August 22, 2019, Barcelona, Spain

Sassani et al., 2019
Goldschmidt Conference
August 18-23, 2019
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Outline

- Very Deep Borehole Disposal (VDBD) Overview
  - Recent R&D history
  - Disposal concept
  - Safety and feasibility
- Crystalline Basement Reaction Analyses
  - Observed conditions
  - Fluid-rock reaction evaluations
- Summary & Conclusions

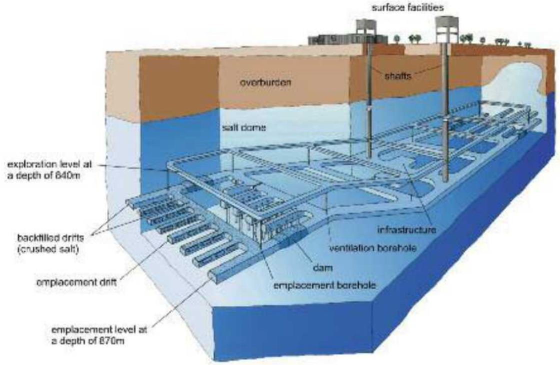
Deep Geological Disposal for Spent Nuclear Fuel and High-Level Radioactive Waste

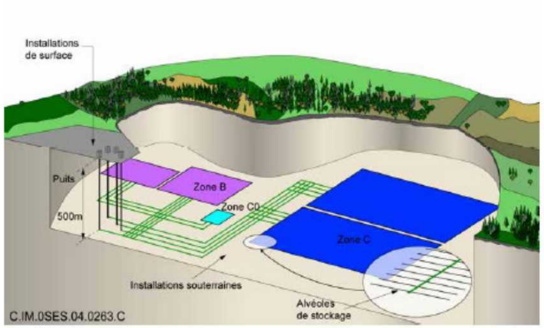
"There has been, for decades, a worldwide consensus in the nuclear technical community for disposal through geological isolation of high-level waste (HLW), including spent nuclear fuel (SNF)."

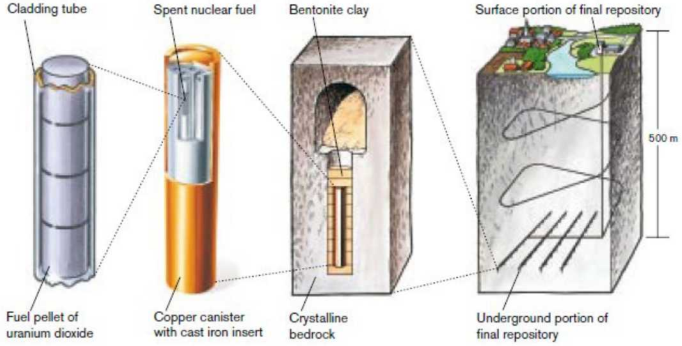
"Geological disposal remains the only long-term solution available."

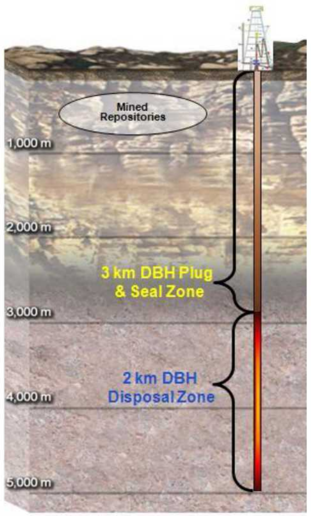
National Research Council, 2001

Deep geologic disposal has been planned since the 1950s







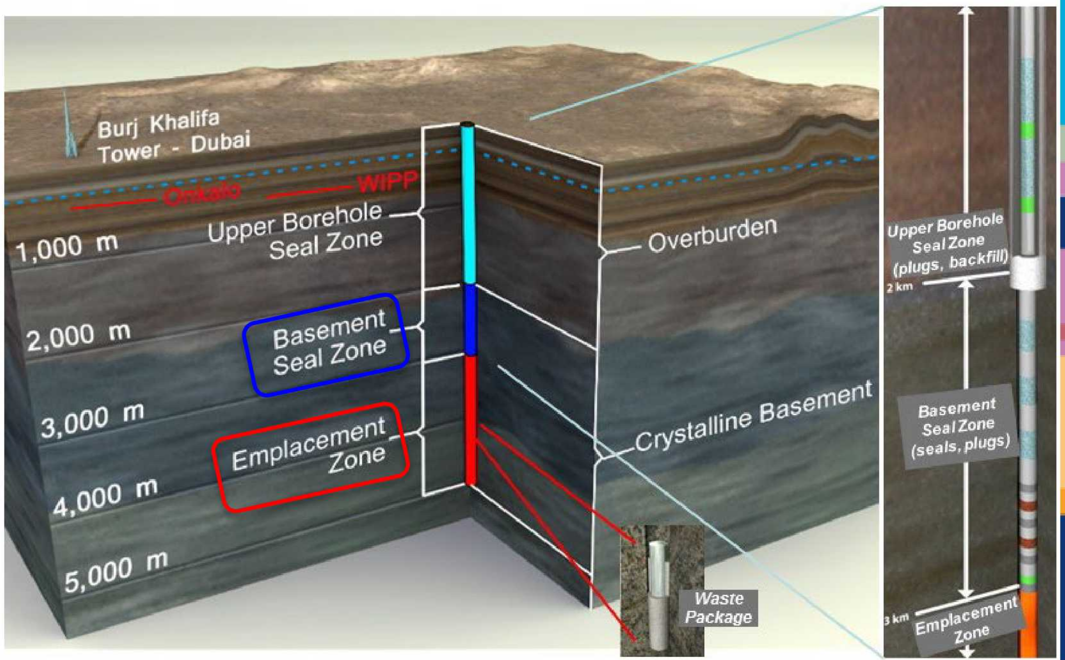


Research and Development (R&D) at Sandia National Laboratories (SNL)

- 2009 – 2012 (SNL internally funded)
  - DBD Consortium with Mass. Inst. of Tech. (MIT), U. of Sheffield, Industry (Brady et al. 2009, Arnold et al. 2011)
- 2012 – 2014 (U.S. DOE funded R&D)
  - Preliminary generic siting, design, and post-closure PA focused on SNF disposal (Arnold et al. 2013; Freeze et al. 2013)
  - DOE (2014) recommended consideration of DBD of smaller DOE-managed waste forms, such as Cs and Sr capsules
- 2014-2017 (U.S. DOE funded R&D)
  - Lead Lab for a planned 5-year Deep Borehole Field Test (DBFT) to evaluate the feasibility of siting and operating a DBD facility
    - Collaboration with other National Labs: LANL, LBNL, ORNL, PNNL, INL
    - DBFT to use "surrogate" waste packages (no radioactive waste)
    - DOE Project stopped at end of FY2017
    - Safety case (Freeze et al., 2019)
- SNL collaborating with other countries, IAEA
  - Special Issue of *Energies*, **2019**, *12*(11)

Very Deep Borehole Disposal Concept

- Drill a borehole or array of boreholes into deep, competent rock (e.g., crystalline basement)
  - ~ 5,000 m* total depth (TD)
  - up to 17” (43 cm) diam. at TD
    - 17” for SNF (1 PWR assembly)
    - ≥ 8.5” for some HLW
- Emplacement Zone (EZ)
  - Waste in lower ~ 2,000 m*
- Seal Zone (SZ)
  - Engineered seals and plugs above EZ
    - ≥ 1,000 m* robust seal in competent basement rock



* depths will be site and waste specific

Robust Isolation from Biosphere

Natural Barriers – deep, low permeability host rock
Engineered Barriers – redundant seals, possibility of long-lived waste forms and waste packages

VDBD Concept – Safety and Feasibility

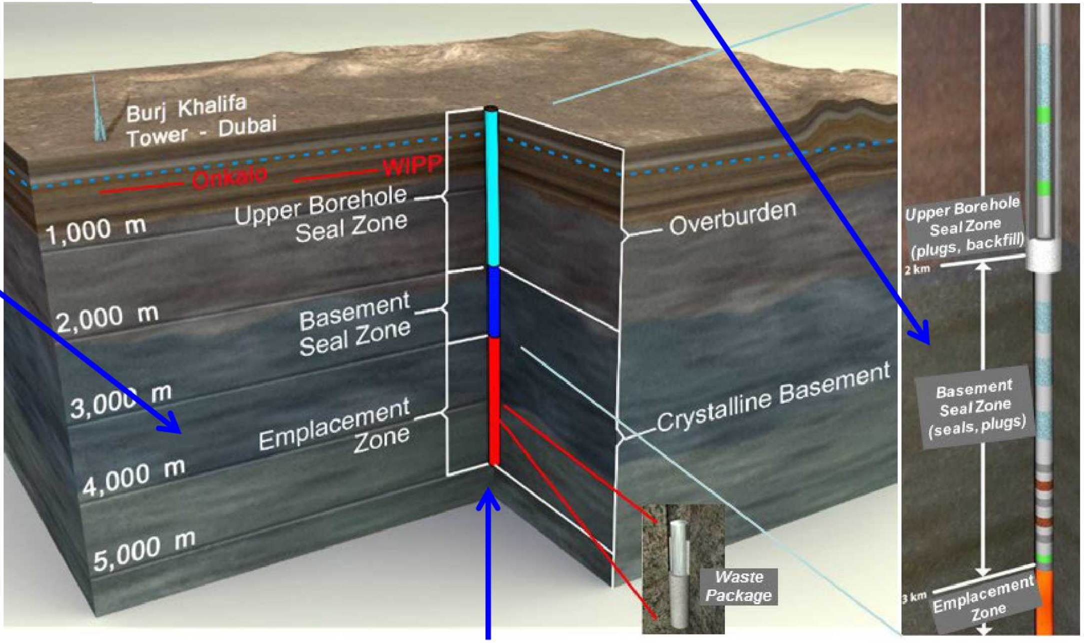
(Post-Closure Hydrogeochemical Waste Isolation)

Identify adequate host rock with sufficient depth and thickness

Deep basement systems can be/have:

- hydrologically isolated from shallow groundwater (low permeability and long groundwater residence time)
- density stratification (more saline brines underlying less saline fluids)
  - opposes upward flow
- geochemically reducing conditions at depth
  - limit the solubility and enhance the sorption of many radionuclides

Borehole Seals and Disturbed Rock Zone (DRZ) can be engineered/evolve to maintain a low-permeability barrier, at least over the time scale of thermally-induced upward flow



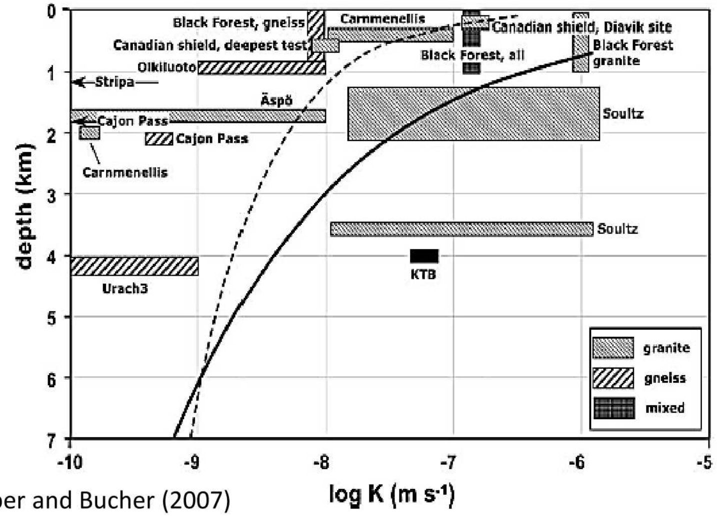
Waste is deep and isolated in basement rock

- well below typical depth of fresh groundwater ------
- with at least 1,000 m of basement rock (Seal Zone) overlying the Emplacement Zone

Safety Case Details in Freeze et al., 2019

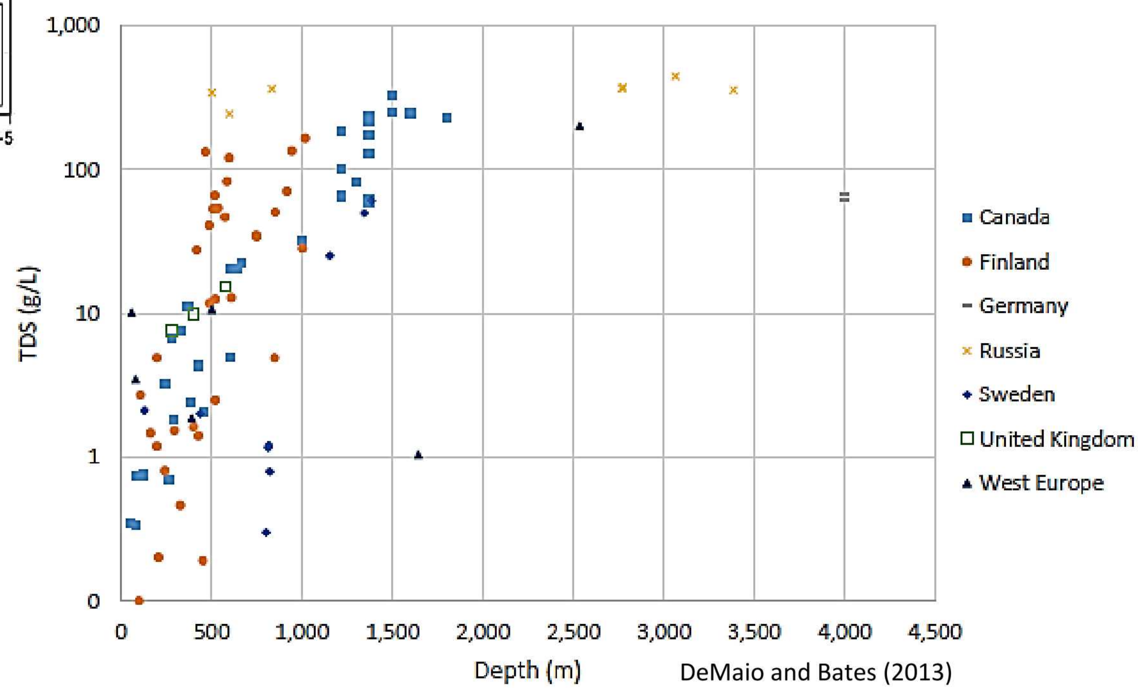
Observed Hydrogeochemical Profiles

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?*
- *Decreased Permeability?*

Fluid-Rock Reaction Evaluations

- 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, Ca/Na, Cl, Br)
    - Assessed leachate compositions from Black Forest crystalline basement rocks
  - Fluid inclusion contributions (soluble salts) considered
- Conditions comparable to ~ 3-5 km depth
  - Generic granite composition and sensitivity analyses
  - Starting fluid compositions: seawater and dilute groundwater
  - ~100 – 150^{o}C, P_{sat}
  - PHREEQC reaction path calculations

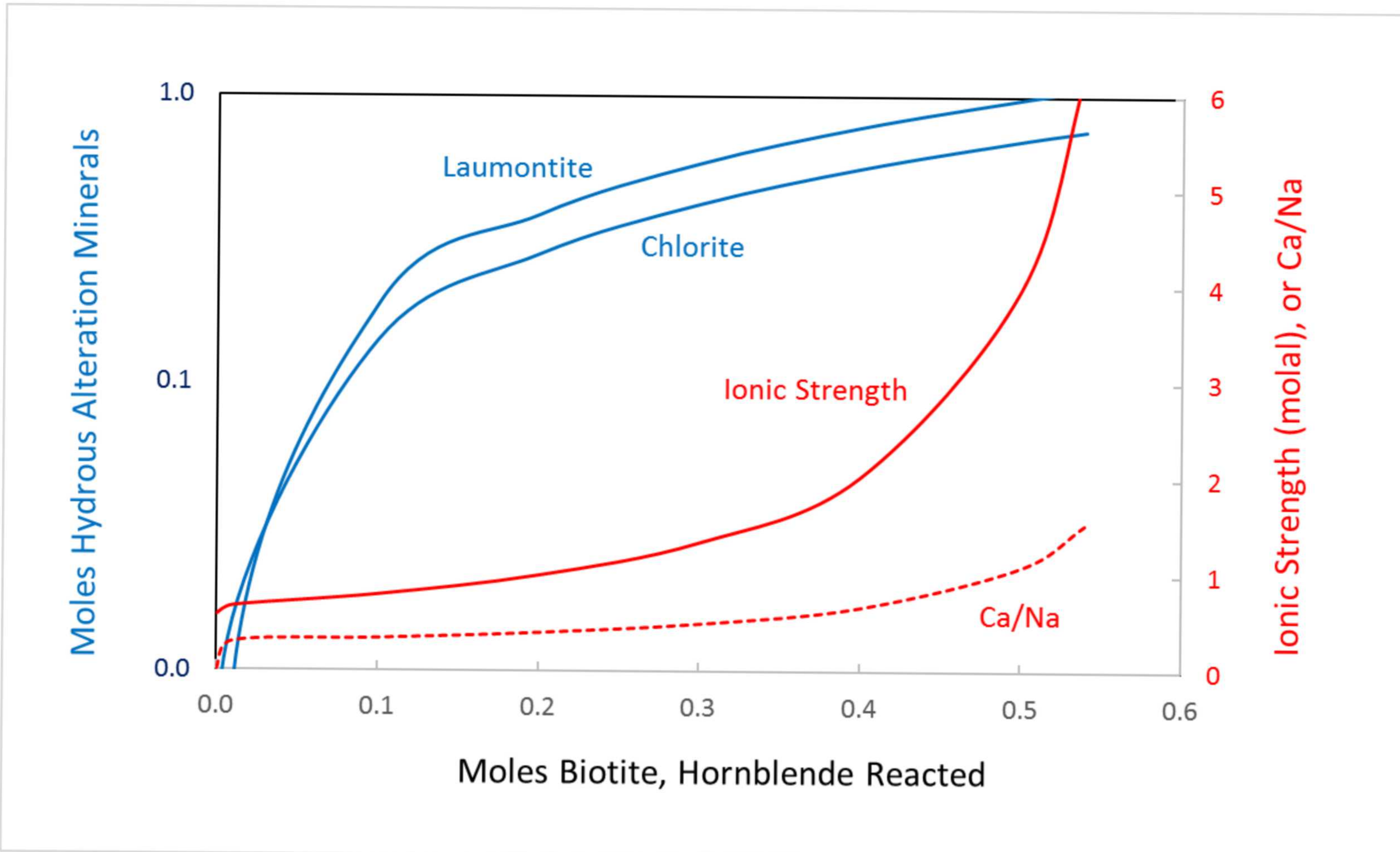
Hypothetical Granite

- Baseline Mineralogy (volume %)
  - 20% Quartz; 40% K-feldspar; 15% Plagioclase (Albite); 9% Muscovite; 8% Biotite; and 8% Hornblende
- Represented as a 10 kg (3.8 L) Block with
  - 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
- Reacted with 0.1 L 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

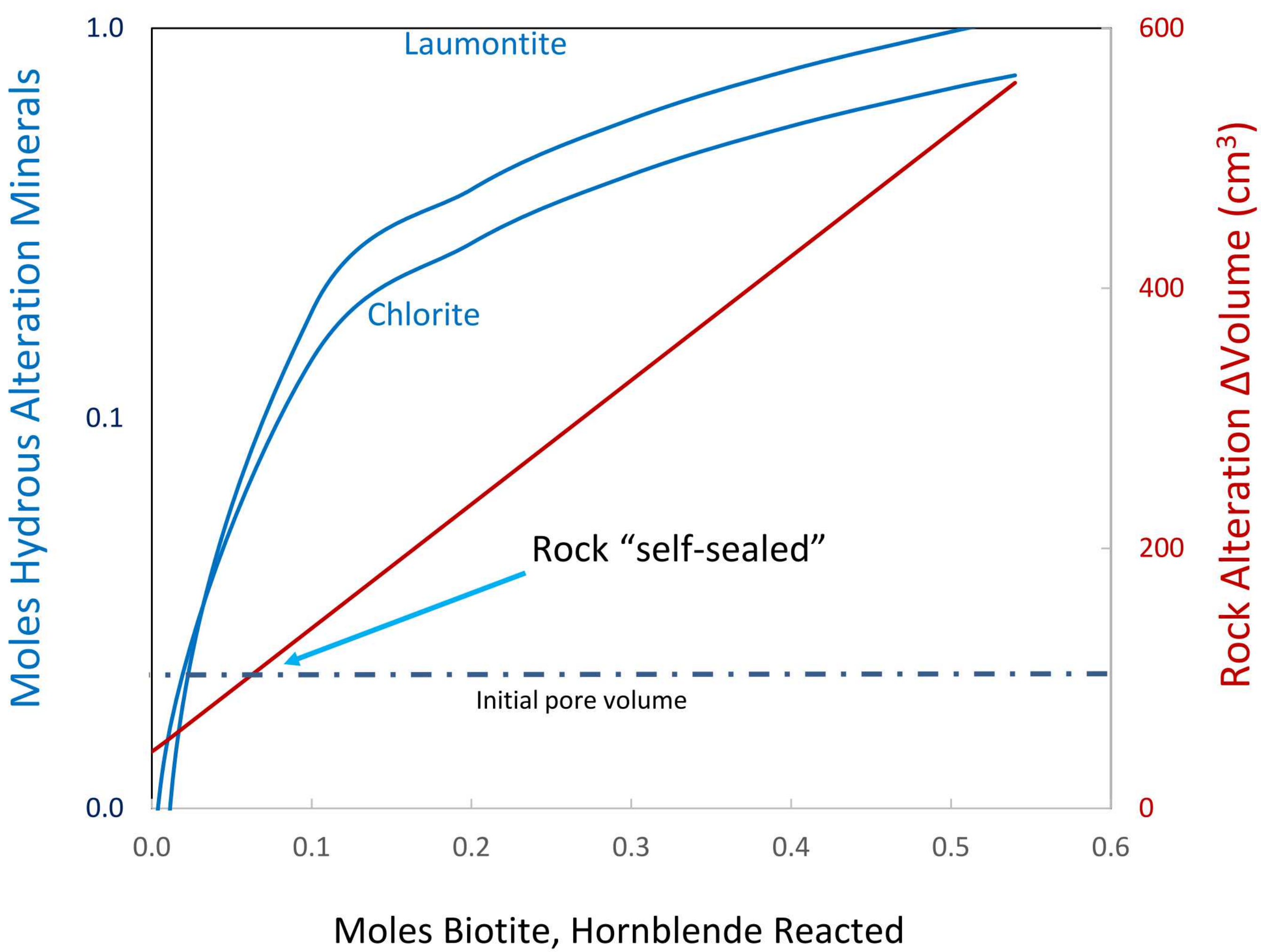
- Calculated Generic Granite Hydrologic Alteration Results
  - Reaction creates Albite + K-feldspar + Chlorite + Laumontite + Brine
    - Minor amounts (< 0.02 moles) of epidote, calcite, and gypsum
    - Albite and K-feldspar masses increase substantially
    - Almost all of the quartz is dissolved.
  - Fluid evolves to Ca-Na-Cl brine at pH of 6.8 (net water loss)
    - Initial ionic strength of 0.6 increases upwards to > 5 molal
    - The Ca/Na calculated for this 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
- Sensitivity Cases
  - Addition of anorthite (0.5 moles) to mineralogy
  - Dilute groundwater instead of seawater
  - Similar results to the baseline case

Mineralogic and Solution Evolution



Sassani et al., 2019 Goldschmidt, August 22, 2019

Mineralogic and Porosity Evolution



Sassani et al., 2019 Goldschmidt, August 22, 2019

Summary and Conclusions

- A Number of Countries are Considering this Disposal Concept
- Fluid-Rock Reaction Appears able to Drive the Evolution/Isolation of Crystalline Basement Systems
  - Brine evolution from seawater and dilute groundwater
  - Alteration mineralogy reducing already small porosity/permeability
- How Commonly are such Fluid Systems Isolated?
  - May be fundamental/intrinsic process, but still evaluating
    - Sensitivity of the PHREEQC calculations to
      - Initial water chemistry
      - Mineralogic variation
      - More detailed consideration of activity coefficient effects
    - Thermal gradient trajectories with depth
    - Comparative rates of reaction and H_2O diffusion at depth
  - Continue to advance comparison of predicted/observed alteration of both mineralogy and deep brine compositions

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