U.S. DEPARTMENT OF ENERGY | Nuclear Energy

Research and Development Activities Related to the Development of Engineered Barrier Systems for Different Geologic Media

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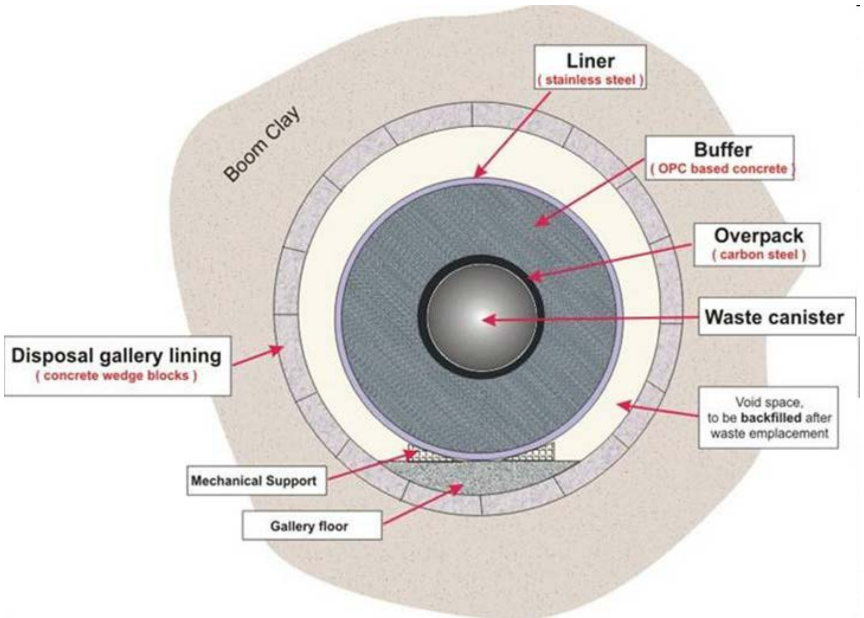
What is the Engineered Barrier System (EBS)?

- ***EBS definition from the US Nuclear Regulatory Commission (10 CFR 60.2)***
  - *"Engineered barrier system* means the waste packages and the underground facility"

- ***EBS definition from to the NEA/OECD EBS State-Of-The-Art Report (2003):***
  - *"The "engineered barrier system" represents the man-made, engineered materials placed within a repository, including the waste form, waste canisters, buffer materials, backfill and seals."*

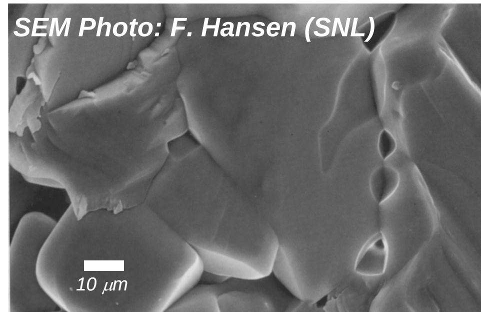


Multi-barrier concept for the Belgian radioactive waste repository concept

Source: http://www.sck.be

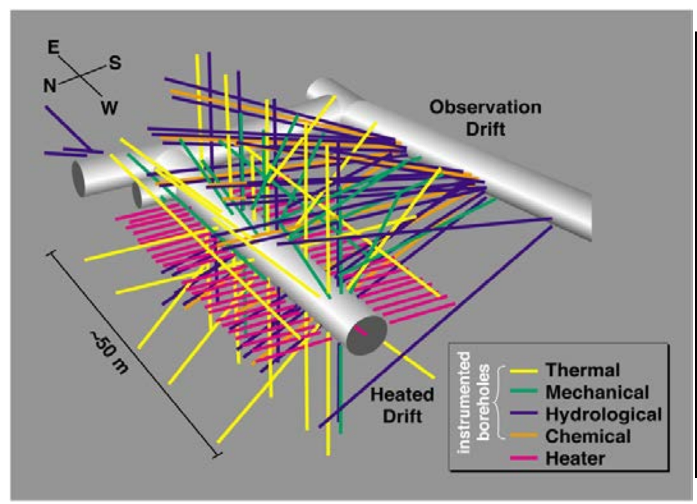
What has been done for EBS?

- **USA examples:**
  - **Disposal in Bedded Salt Media:**
    - *Deaf Smith (TX) Site Studies*
    - *WIPP (NM)*
    - *Coupled Thermo-Mechanical Studies (experiments and modeling):*
      - Intact Salt
      - Crushed Salt
  - **Nuclear Waste Encapsulation:**
    - *Glass Waste form – HLW (Borosilicate Glass)*
    - *Cementitious Waste forms – LLW (SRS Saltstone®)*
    - *Research on Novel Wasteforms (ceramic, mixed-phase glass-ceramic)*
  - **Drift-Scale Test Facility – Yucca Mountain Project (YMP)**
    - *Thermal environments in disposal drifts*
  - **Waste Package, Drip Shield, and TAD concepts for YMP**



Consolidated WIPP Crushed Salt
(Source: The Mechanical Behavior of Salt, 1996)

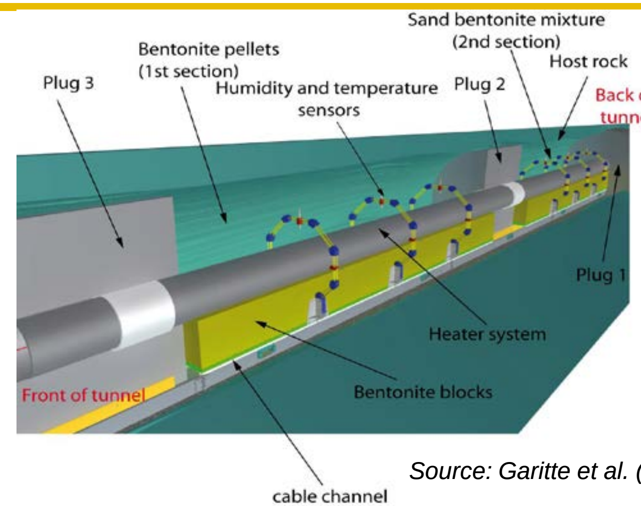
WIPP Waste in Panel one (Sacks of MgO Buffer on Top)



What has been done for EBS? (Cont.)

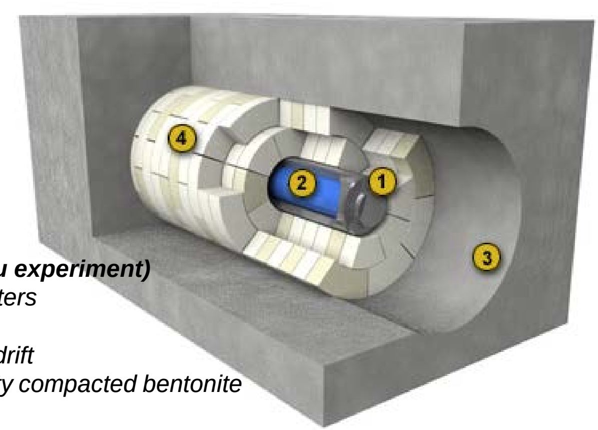
- **International examples:**
  - <u>**Underground Research Laboratories (URLs)**</u>:
    - *Mt. Terri (Opalinus Clay, Switzerland)*
    - *Grimsel (Granite, Switzerland)*
    - *Tournemire (Argillite, France)*
    - *Meuse/Haute-Marne (BURE) (Callovo-Oxfordian Clay, France)*
    - *HADES (Boom Clay, Belgium)*
    - *Äspö (Granitoids, Sweden)*
    - *Gorleben (Dome Salt, Germany)*
    - *FEBEX (Mock-Up, Spain; Site-Scale, Grimsel, Switzerland)*
    - *KAERI/KURT (Granite, South Korea)*
    - *Horonobe (Mudstones) and Mizunami (Granite) Sites (Japan)*
  - <u>**International Collaborations**</u>
    - *DECOVALEX (Development of Coupled Models and their Validation Against Experiments, International Collaboration)*
    - *NEA/OECD Integration Group for the Safety Case (IGSC) EBS project*

For more details, see Jové Colón et al. 2011



Source: Garitte et al. (2011)

HE-E heater test at Mont Terri



FEBEX (in situ experiment)
(1) Steel canisters
(2) Waste
(3) Horizontal drift
(4) High-density compacted bentonite blocks

Source: http://www.grimsel.com

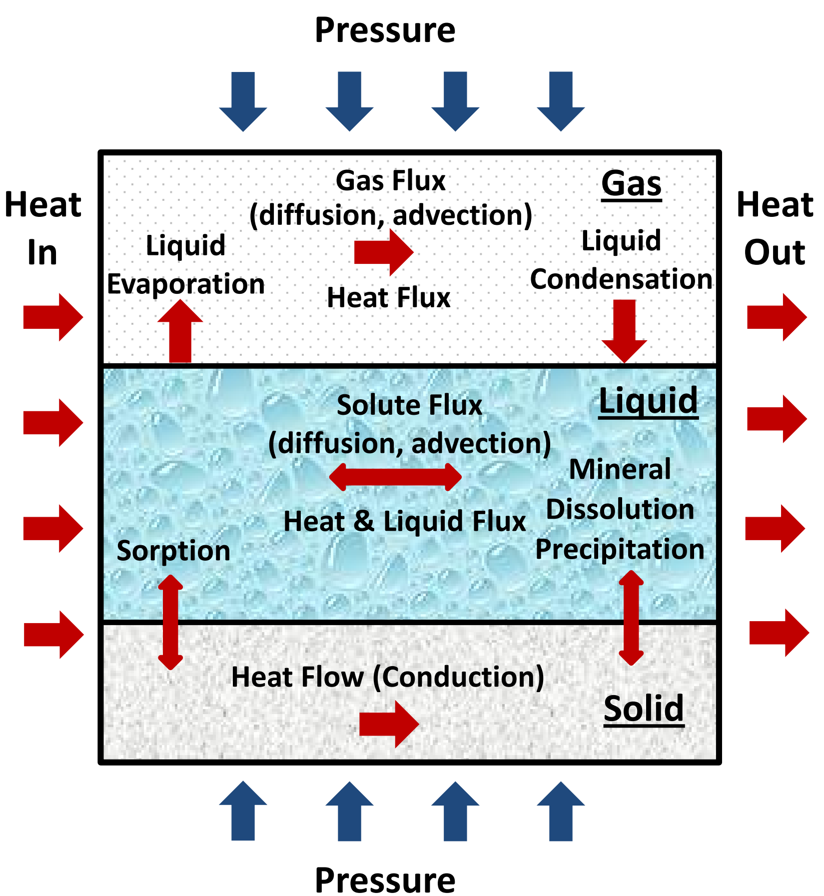
Used
Fuel
Disposition

UFD Needs for EBS?

- **Knowledge gaps and R&D prioritization in EBS (based on the UFDC Disposal R&D Roadmap, Nutt et al. 2011) :**
- **Highest ranked issues:**
  - *Waste Form*
  - *THM Processes*
  - *Waste Container*
  - *Radionuclide speciation and solubility*
  - *Buffer/Backfill material*
- **High rank of THMC processes is relevant to interactions at EBS interfaces:**
  - ***Loci for important degradation processes in the near-field***
  - ***Shares a boundary with far-field region***
  - ***THMC models must assess the generic aspects of EBS design concepts***

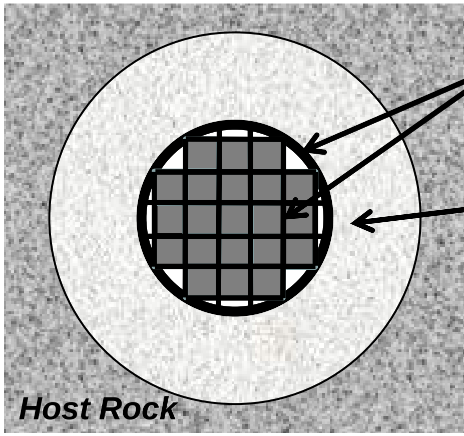
For more information, see Nutt et al. (2011)

THMC Processes in the EBS



Modified After Olivella et al. (2011)

Work to Date: EBS Design Concepts – Backfilled Disposal Scenarios

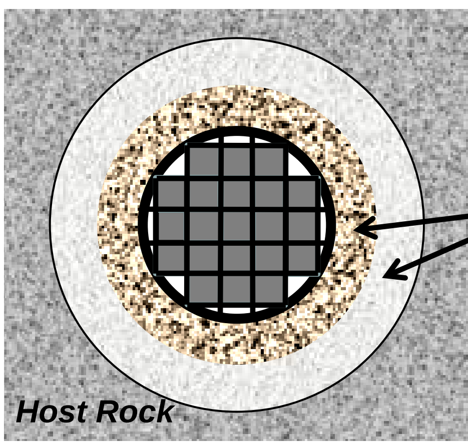


Waste Container and PWR Assembly (arbitrary)

One-Domain Backfill/Buffer

EBS design concept A*

Simplest EBS Design Concept: FEBEX EBS design concept (Spain) adopted in mock-up and field-scale experiments in Europe



Two-Domain Backfill/Buffer Barrier Material

EBS design concept A1*

Not To Scale

Increase in Complexity

- **Evaluation of generic EBS design concepts representative of various levels of complexities**
- **Provide the basis for a flexible EBS design optimization**

****see Jové Colón et al. 2011***

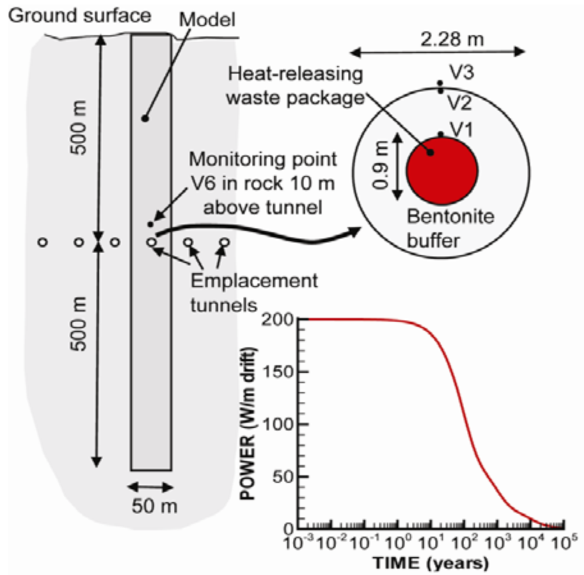
Work to Date: Databases

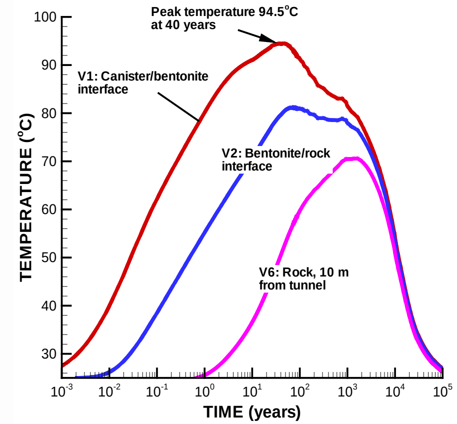
- **Compilation of subsurface hydrochemical data from various sites**
- **Expansion and maintenance of (qualified) thermodynamic databases that were developed for the Yucca Mountain Project**
  - Maintain a high level of thermodynamic internal consistency and transparency
  - Use similar tools and methods (e.g., temperature extrapolation)
  - Focus on data needed to evaluate the current set of UFD HLW disposal options
    - Clay thermodynamic data and hydration models
- **Evaluating fluid-solid interactions and thermodynamic data for cementitious phases:**
    - *Evaluation and comparisons between YMP cement thermodynamic database and CEMDAT07 (Matschei et al. 2007; Lothenbach et al. 2008; Blanc et al. 2010)*
    - *Expansion of existing thermodynamic data for cementitious material*
  - Studying model implementation of fluid-solid interactions of cement phases:
    - Modeling code tool identification: EQ3/6 (LLNL), Cantera-DAKOTA (Caltech, SNL)
    - Evaluation of solid solution models for cementitious phases

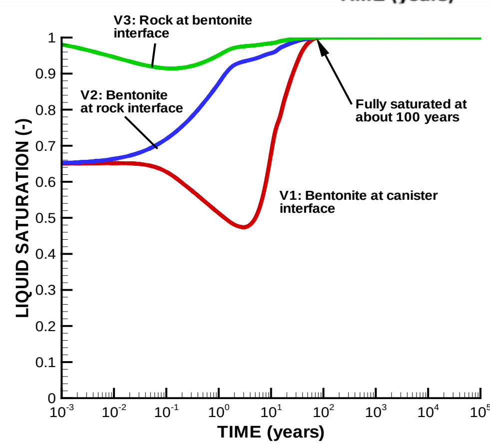
Used Fuel Disposition

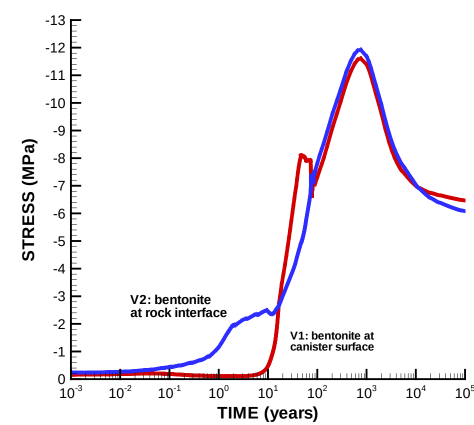
Work to Date: THM Modeling on Clay

- THM coupled behavior of bentonite clay evaluated with TOUGH-FLAC simulator
- New implementation of the TM Barcelona Basic Model (BBM) for clay
- Thermal management and peak temperatures:
  - Buffer saturation and thermal conductivity
  - Tunnel and canister spacing
  - Elevated peak temperature
- Resaturation and buffer swelling
- Rock failure of layered rock









Used
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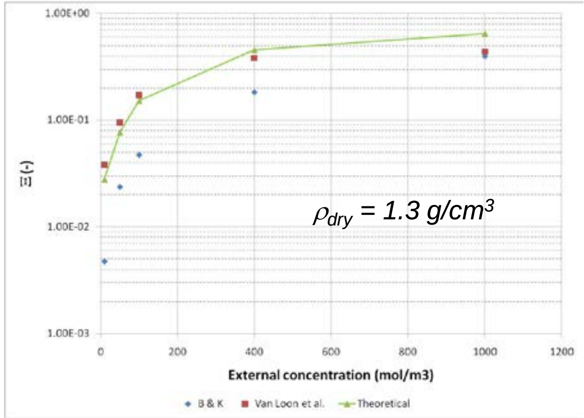
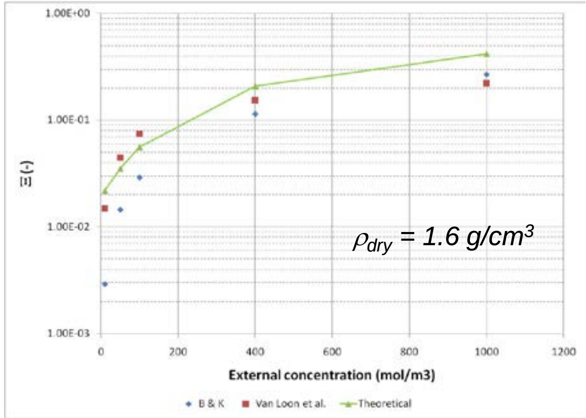
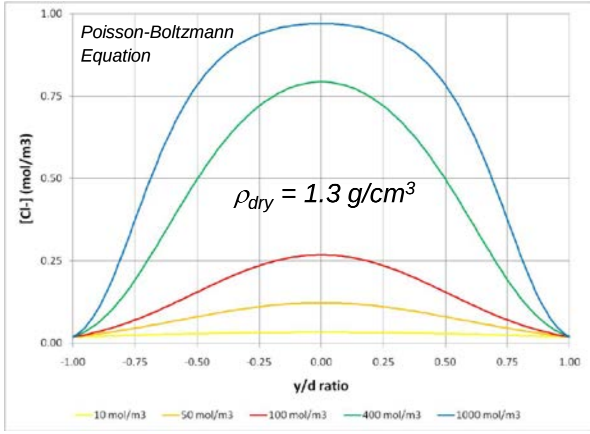
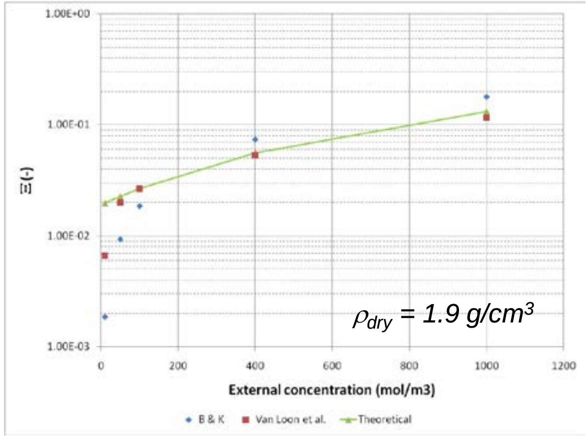
Work to Date: Reactive Diffusion on Clay

- **Reactive diffusion models for clay implemented in the CrunchFlow code:**
  - Single and double porosity models
  - Analytical Solution of Poisson-Boltzmann equation to resolve anion concentration in the pore space
  - **<u>Goal</u>**: Apply the analytical solution approach to multicomponent diffusion





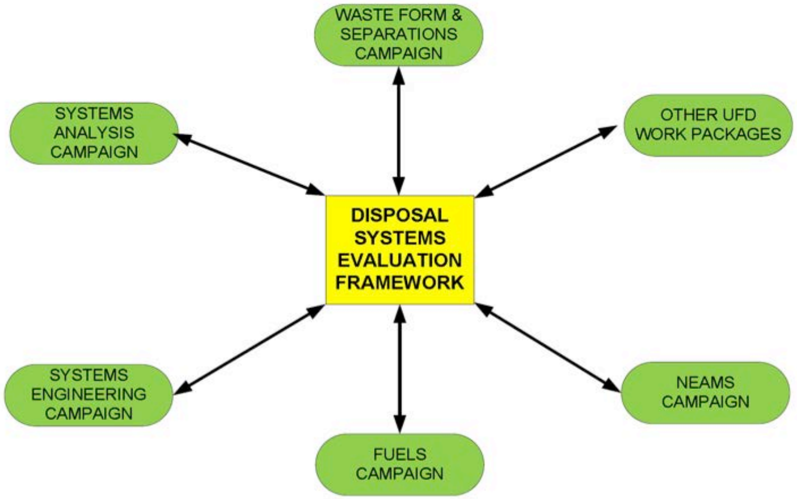




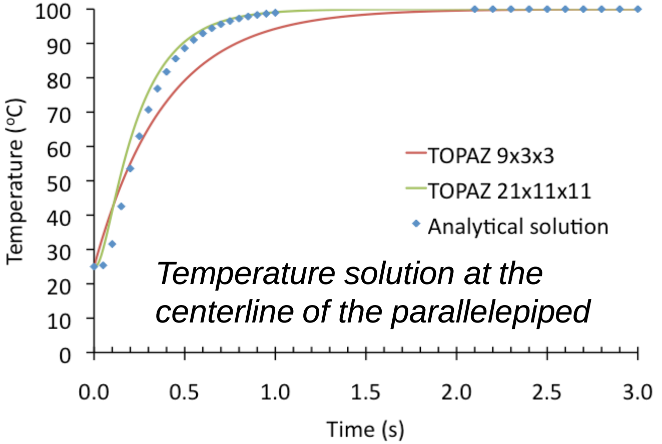
***see Jové Colón et al. 2011**

Work to Date: Disposal System Evaluation Framework (DSEF) and Thermal Analysis

- **DSEF:**
  - **Allows for efficient (albeit high-level) evaluations and comparisons between**:
    - *Fuel cycles (open, modified open and closed)*
    - *Disposal environments (granite, salt, clay/shale, and deep borehole)*
    - *Repository designs*
    - *EBS materials (bentonite, mixed clay/sand mixtures)*
    - *Fuel types with pre-emplacement aging times (short, moderate and extended)*
    - *Implemented in MS Excel and Access*



- **Thermal Analysis:**
  - *Interfaces with Analytic (Mathcad®) and finite element models (TOPAZ3D)*
  - *Analytic: point and line source geometries*
  - *Finite element: captures more accurately thermal transport and complex geometries*



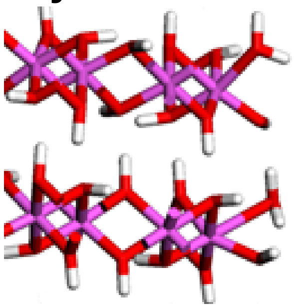
Temperature solution at the centerline of the parallelepiped

***see Jové Colón et al. 2011**

Used Fuel Disposition

Ongoing Research Work: Molecular Dynamic (MD) Studies on Clay

ClayFF force field to model clay-minerals



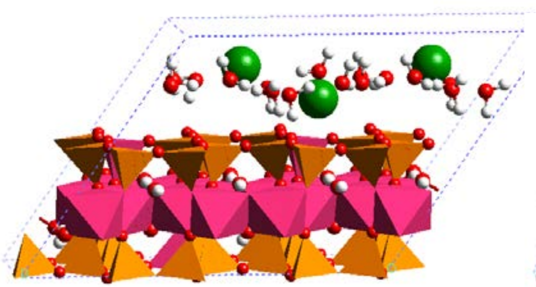
- Cygan *et al.*, 2004, cited 200 times
- Actively developing force field to model mineral edges

Molecular Dynamics (MD) simulation

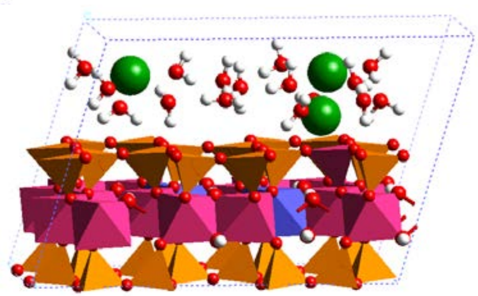
- **Redsky** – Sandia's supercomputer resource
- **LAMMPS** – massively parallel MD simulation code

- **Study clay swelling behavior as a function of relative humidity**
- ***Variation in clay behavior captured by end-member clays and cation species***
- **Comparisons with literature data**

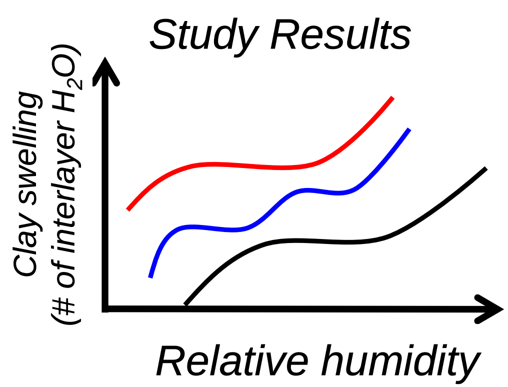
Beidellite



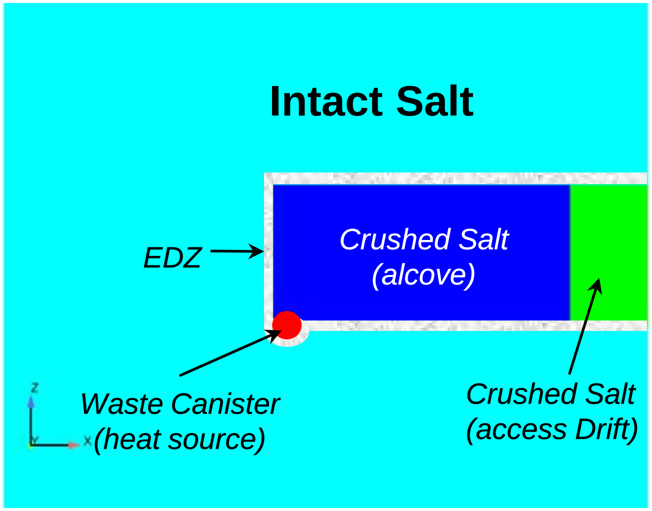
Montmorillonite

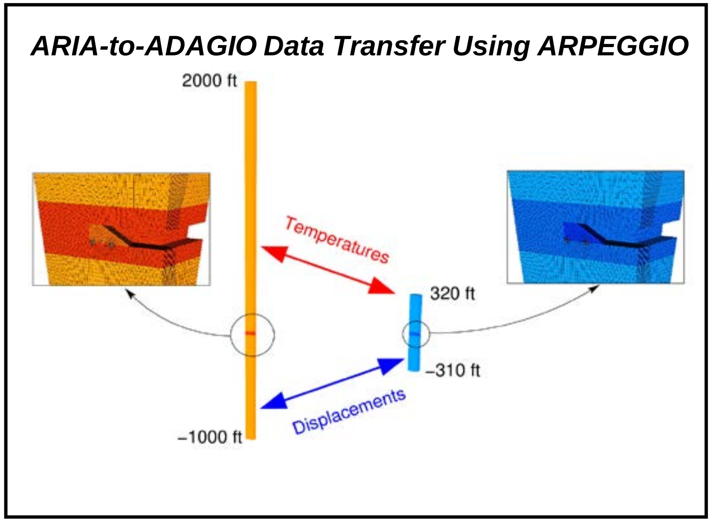


Cations : Cs^+; K^+; Na^+; Ca^{2+}; Mg^{2+}



Ongoing Research (Cont.): Coupled THM Calculations in Salt Media





- **Using Sandia's SIERRA Mechanics High Performance Computing capabilities**
- **Porous crushed salt backfill:**
  - ***Constitutive models to capture temperature and porosity dependencies***
  - ***Focus on moisture transport***
  - ***Coupling of salt permeo-porous properties with mechanical deformation***
  - ***ARPEGGIO Code: externally couples ARIA (thermal-hydrological) and ADAGIO (Lagrangian mechanical) codes***

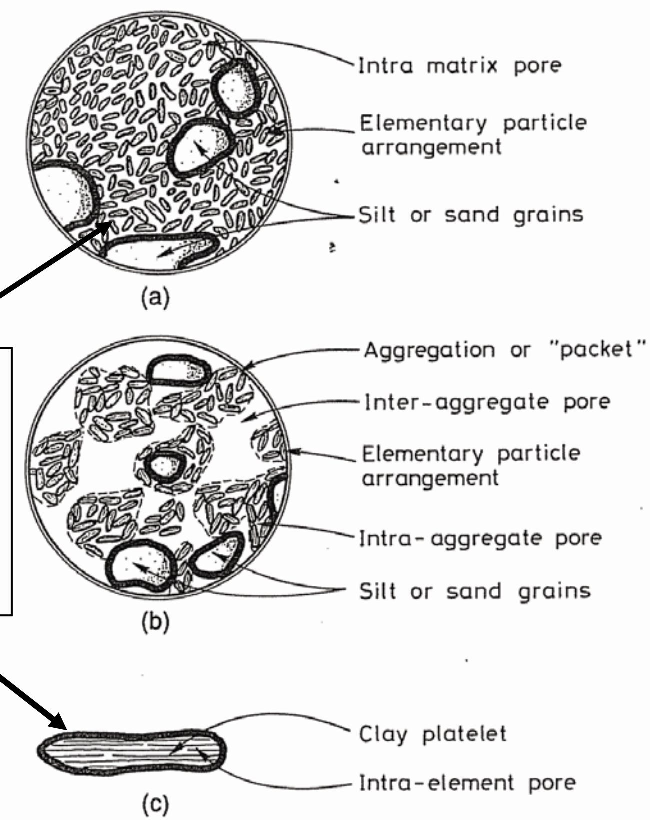
Ongoing Research (Cont.): Thermodynamic Model, Databases, Integration Activities

- **Thermodynamic Databases:**
  - **Cementitious materials**
    - *YMP database ported into Cantera code input format*
    - *Implementation of solid solution model for C-S-H (Margules type) using Cantera*
  - **Thermodynamics of clay phases**
    - *Review / update of available models and thermodynamic data for clay including clay hydration*
  - **Disposal System Evaluation Framework (DSEF)**
    - *Build the multi-sheet backbone of the Excel Workbook, focusing on user interface*
    - *Incorporate thermal algorithms and results from FY11 work*
    - *Develop cost algorithms using literature information as a starting point, and implement in the DSEF*
    - *Test case for a multi-layered EBS design optimization*
  - **Integration activities**
    - *Integration with other UFDC activities (e.g., GDSM)*
    - *Initiated development (with Natural System) of web-based information management tool for database cataloging*

Ongoing Research (Cont.): THM on Clay

- ***Modeling interactions between EBS and natural system***
- ***Extension of TOUGH-FLAC-BBM to Barcelona Expansive Model (BExM) to consider micro- and macro-structural interactions***
- ***This model enhancement will serve as a framework for further extension to coupled THMC behavior (i.e., coupling to chemistry)***
- ***Participation in DECOVALEX project to validate the THM model (HE-E heater test at Mont Terri URL)***

Chemo-Mechanical coupling through diffuse double layers theory (BExM)



After Gens & Alonso, 1992

Used Fuel Disposition

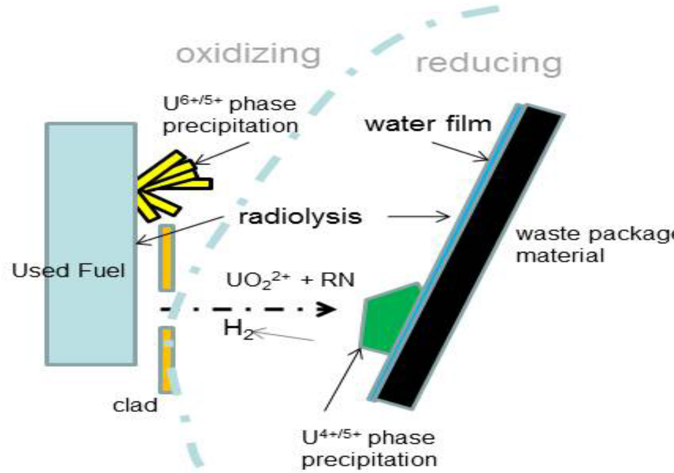
Ongoing Research (Cont.): Reactive Diffusion Through Bentonite and Clay Barrier Interactions (Modeling and Experiments)

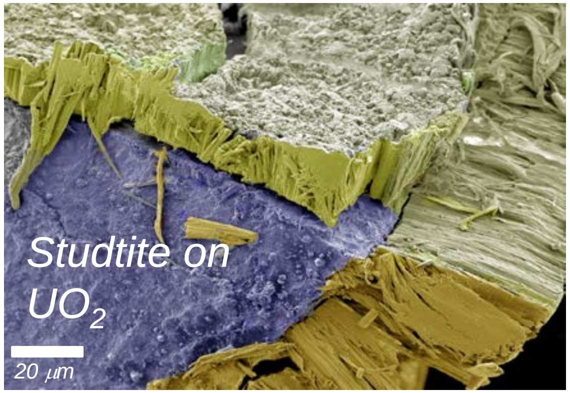
- **Experimentally characterize U(VI) sorption and diffusion behavior in terms of:**
  - Chemical solution conditions: pH, ionic strength, carbonate concentration
  - Degree of clay compaction
  - Experimental data for development of a reactive U(VI) diffusion model
- **Complete implementation of multicomponent Poisson-Boltzmann equation in reactive transport simulator:**
  - Test against full range of diffusion data (Van Loon et al. 2007)
  - Test against uranium transport experiments in smectite / bentonite cell
  - Developed a fractal, multiple pore size model to describe anion transport in compacted clays
- **Experimental Work on Clay Barrier Interactions:**
  - Waste container (304 SS, 316 SS, Copper), backfill (Wyoming bentonite), liquid (DI water, Stripa Brine)
  - Clay – water and Clay – Metal – water
  - Experimental conditions: 100, 200, 300 °C; 150 bars; buffered at Mt-Fe oxygen fugacity
  - Study phase changes in container material and clay, brine chemistry

Used Fuel Disposition

Ongoing Research (Cont.): Used Fuel Degradation – Experimental and Electrochemical and Studies

- **Objectives:**
  - **Evaluate importance of Ru-Mo-Pd-Rh "noble metal particles" (NMP) as catalysts in the scavenging of oxidants (H_2 oxidation)**
  - **Use Mixed Potential Model as base model for UO_2 fuel degradation (Shoesmith, 2003)**
- **Materials: Ru-Mo-Pd-Rh alloys & UO_2 electrodes:**
  - **Surrogates for NMP & used fuel matrix**
- **Full system characterization:**
  - **In-situ X-ray absorption spectroscopy (XAS)**
  - **Electron microscopy (SEM, TEM), XRD**
  - **Solution chemistry**
- **Radiolysis Model and Experiments on Used Fuel Degradation:**
  - **Evaluation of radiolysis models**
  - **Development of simulant fuels for Experiments**
  - **Quantum mechanical calculations for UO_2 oxidation**





Source: E. Buck, PNNL

Expected Future Work

- **Expand modeling activities for coupled processes (THMC)**
- **Expand experimental activities to research key processes in EBS performance and used fuel degradation**
- **Increase level of international collaboration with URLs involving field- and lab-scale experiments**
- **Continued enhancement of level of integration between UFD activities**
  - Continue support to Generic Disposal System Modeling
  - Increase DSEF integration with UFDC activities and other FCT campaigns

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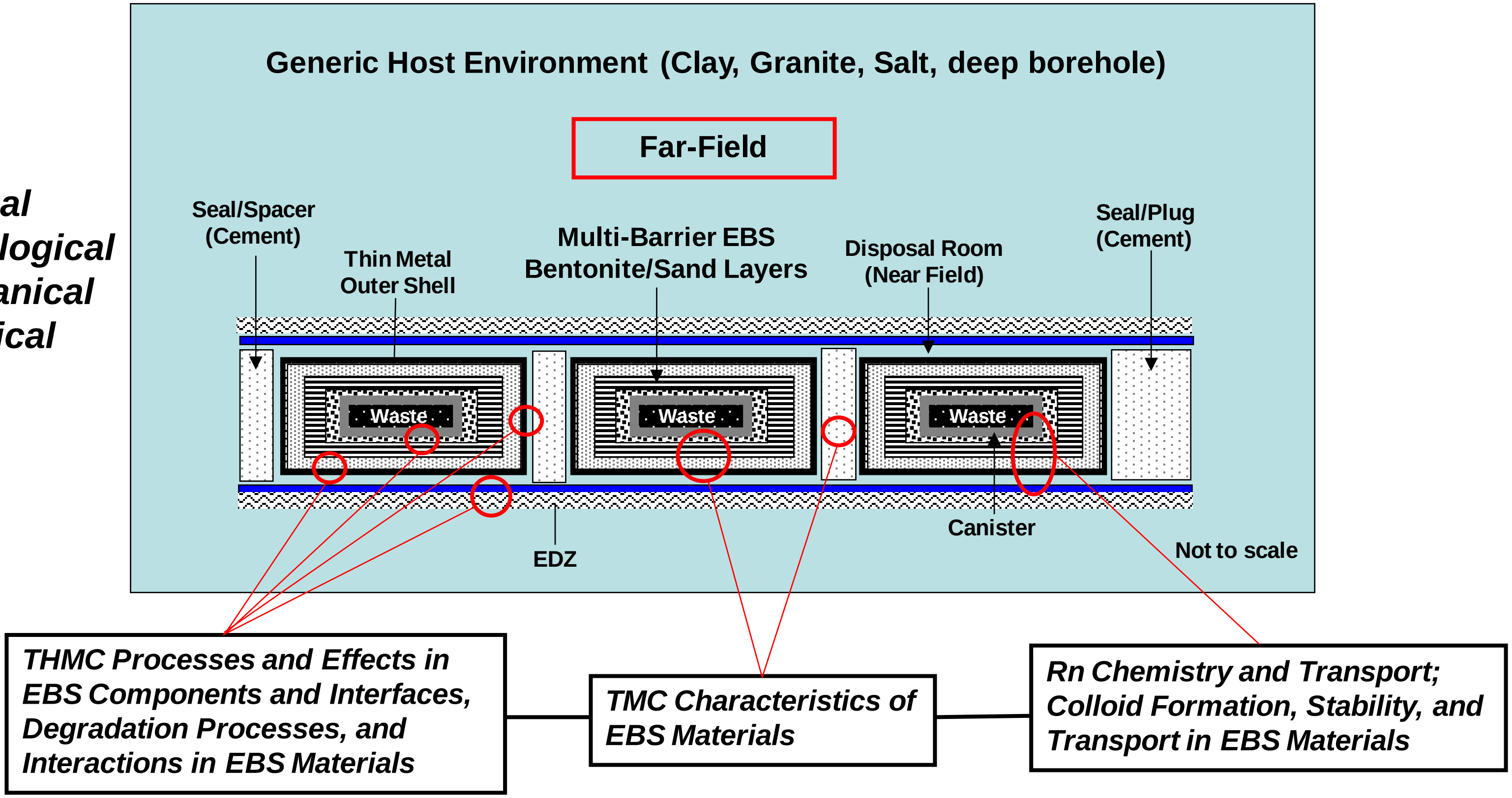
Backup Slides

UFD Needs for EBS? (Cont.)

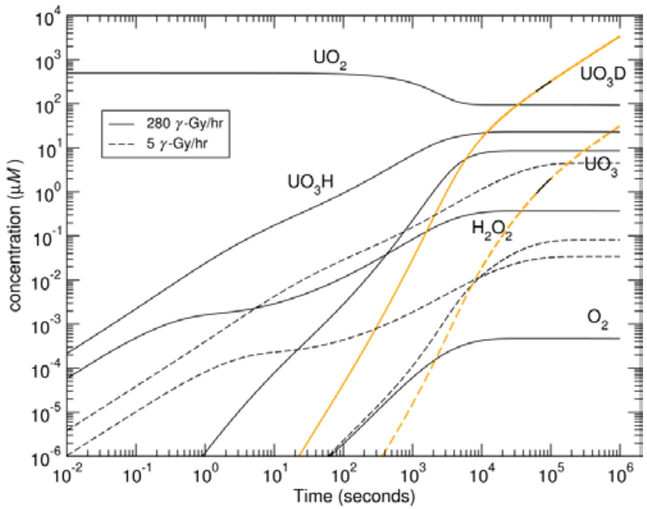
THMC Processes and Interactions at EBS interfaces

T = Thermal
H = Hydrological
M = Mechanical
C = Chemical



Ongoing Research (Cont.): Used Fuel Degradation: Radiolysis Model

- **Objectives:**
  - **Experimental investigation to elucidate long-term behavior of used fuel as a waste form**
  - **Evaluation of radiolysis models for used fuel degradation and radionuclide migration**
    - ***Useable program enables modeling of a three state system with UO_2, water and an atmosphere***
    - ***Reducing environments were examined***



- ***Preliminary Results:***
  - *Role of CO_2 in the system - $OH^\bullet$ will be converted into CO_3^-, which is also a strong oxidant.*
  - *Formation of oxalate in a U-Oxide system may be more important.*