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

Carlos F. Jové Colón, Sandia National Laboratories (Used Fuel Disposition Lead for EBS)
Jeffery Greathouse, Stephanie Teich-McGoldrick, Randy Cygan, Frank Hansen, Lupe Arguello, James Bean, Teklu Hadgu, Mario Martinez, Polly Hopkins, Dave Sassani, Philippe Weck (SNL)
Florie A. Caporuscio (LANL)
James Blink, Harris Greenberg, Thomas Wolery, Mark Sutton (LLNL)
Hui-Hai Liu, Jonny Rutqvist, Jens Birkholzer, Carl I. Steefel, James Davis, Ruth Tinnacher (LBNL)
William Ebert, Jim Jerden, Jeff Fortner, Terry Cruse (ANL)
Edgar Buck, Rick Wittman, Bruce McNamara, Frances Skomurski (PNNL)

Nuclear Waste Technical Review Board
Albuquerque, New Mexico
March 7, 2012

SAND2012-1304C
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 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.”

Source: http://www.sck.be
Used Fuel Disposition

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**

---

March 7th, 2012

NWTRB Meeting
Albuquerque NM
What has been done for EBS? (Cont.)

International examples:

- **Underground Research Laboratories (URLs):**
  - 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)

- **International Collaborations**
  - 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
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)

Modified After Olivella et al. (2011)
Work to Date: EBS Design Concepts – Backfilled Disposal Scenarios

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

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

EBS design concept A*

- One-Domain Backfill/Buffer
- Waste Container and PWR Assembly (arbitrary)

Increase in Complexity

EBS design concept A1*

- Two-Domain Backfill/Buffer Barrier Material
- Host Rock

*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
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 Fuel Disposition

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
  - **Goal**: Apply the analytical solution approach to multicomponent diffusion

*see Jové Colòn 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

*see Jové Colón et al. 2011*
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

---

Study Results

Clay swelling

(# of interlayer H₂O)

Relative humidity

Cations: Cs⁺, K⁺, Na⁺, Ca²⁺, Mg²⁺
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
**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)**

---

*After Gens & Alonso, 1992*
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
**Objectives:**
- Evaluate importance of Ru-Mo-Pd-Rh “noble metal particles” (NMP) as catalysts in the scavenging of oxidants (H₂ oxidation)
- Use Mixed Potential Model as base model for UO₂ fuel degradation (Shoesmith, 2003)

**Materials:** Ru-Mo-Pd-Rh alloys & UO₂ 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 UO₂ 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

Gariotte, B., and others, HE-E experiment - In situ Heater Test, Presentation given at 7th DECOVALEX 2011 workshop, April 2011, Helsinki.


THMC Processes and Interactions at EBS interfaces

Generic Host Environment (Clay, Granite, Salt, deep borehole)

Far-Field

Seal/Spacer (Cement)
Thin Metal Outer Shell
Multi-BARRIER EBS Bentonite/Sand Layers

Disposal Room (Near Field)

Seal/Plug (Cement)

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

THMC Processes and Effects in EBS Components and Interfaces, Degradation Processes, and Interactions in EBS Materials

TMC Characteristics of EBS Materials

Rn Chemistry and Transport; Colloid Formation, Stability, and Transport in EBS Materials

Not to scale
Used Fuel Disposition

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$^-$ will be converted into CO$_3^{2-}$, which is also a strong oxidant.
  - Formation of oxalate in a U-Oxide system may be more important.