SAND2011-0291C

Used Fuel Disposition Campaign

Generic Disposal System Models

Clifford Hansen (SNL)
Mark Nutt (ANL)
Shaoping Chu (LANL)
Joon Lee (SNL)
Liange Zheng (LBL)

Used Fuel Disposition Working Group
January 20, 2011

Outline

- **Work Package Goals and Deliverable**
- **Interface with FEPs**
- **Waste Inventories and Repository Scenarios**
- **Summary Status of GDSE Models**
  - Clay
  - Granite
  - Deep Borehole
  - Salt
- **Improved Diffusive Transport Models for Clay**

Work Package Goals

- **Prepare to perform system assessments for waste form/repository geology combinations**
- **Ready when we have:**
  1. Baseline models for salt, clay, crystalline, deep borehole repositories
  2. Models described in terms of FEPs
  3. Parameter baselines (in cooperation with other work packages)
  4. Waste form/package concepts (from other UFD and Waste Form campaign work packages)

     and

     Baseline repository designs (accounting for geotechnical, thermal and operational perspectives)
  5. Configuration management practices

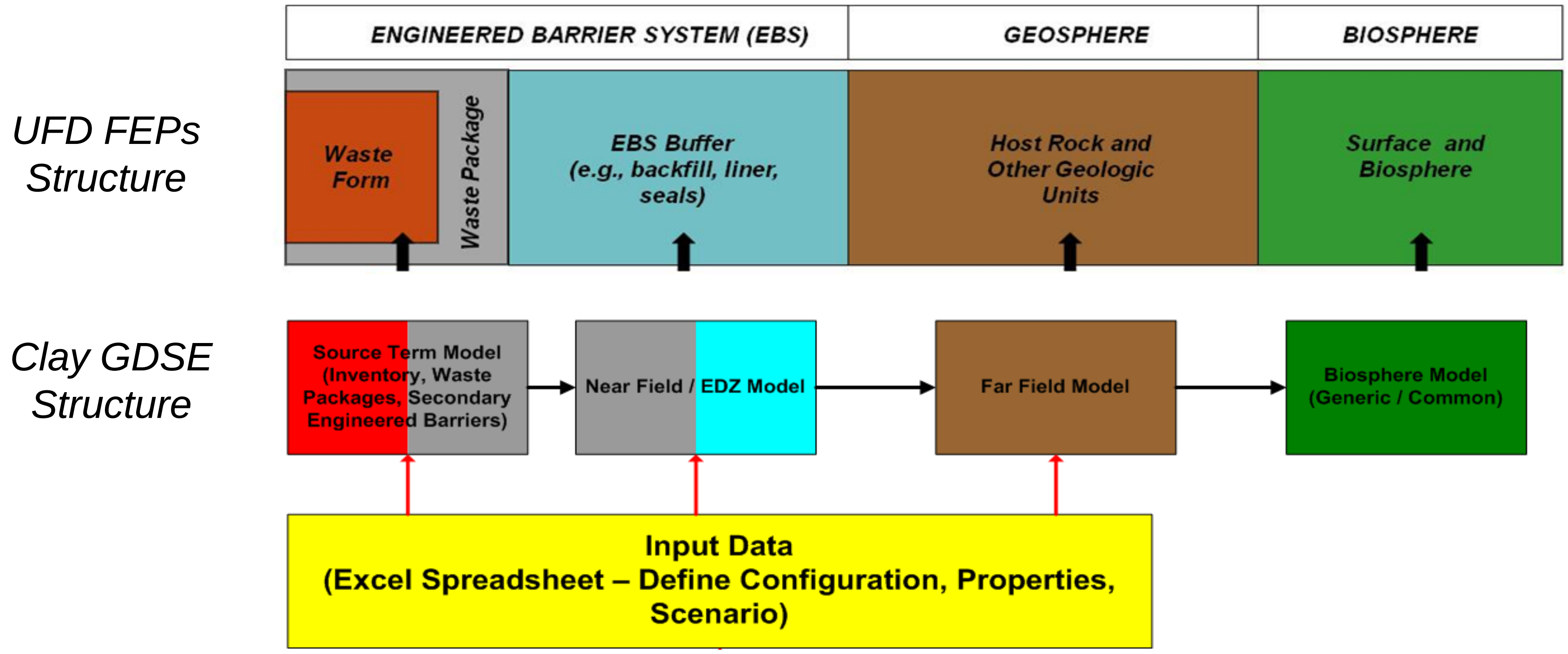
FY11 Work Package Actions

1. **Complete baseline models**
   - Describe each model in terms of common set of FEPs
   - Use UFD FEPs report as template
   - Compile list of parameters for each model

2. **Bring models into common architecture**
   - Streamline implementation in GoldSim
   - Use of common model components (e.g., inventory, biosphere)
   - Common input/output interfaces
   - Facilitate configuration management

3. **FY11 Milestone report**
   - Level 2 Deliverable (Aug 2011)
   - Descriptions of each model (FEPs, parameter catalogs)
   - Illustrative results

GDSE Models: Conceptual Structure and Linkage with FEPs



FY11 Scope: Cross-Mapping UFD FEPs to GDSE Models (same task for all geologies)

Used
Fuel
Disposition

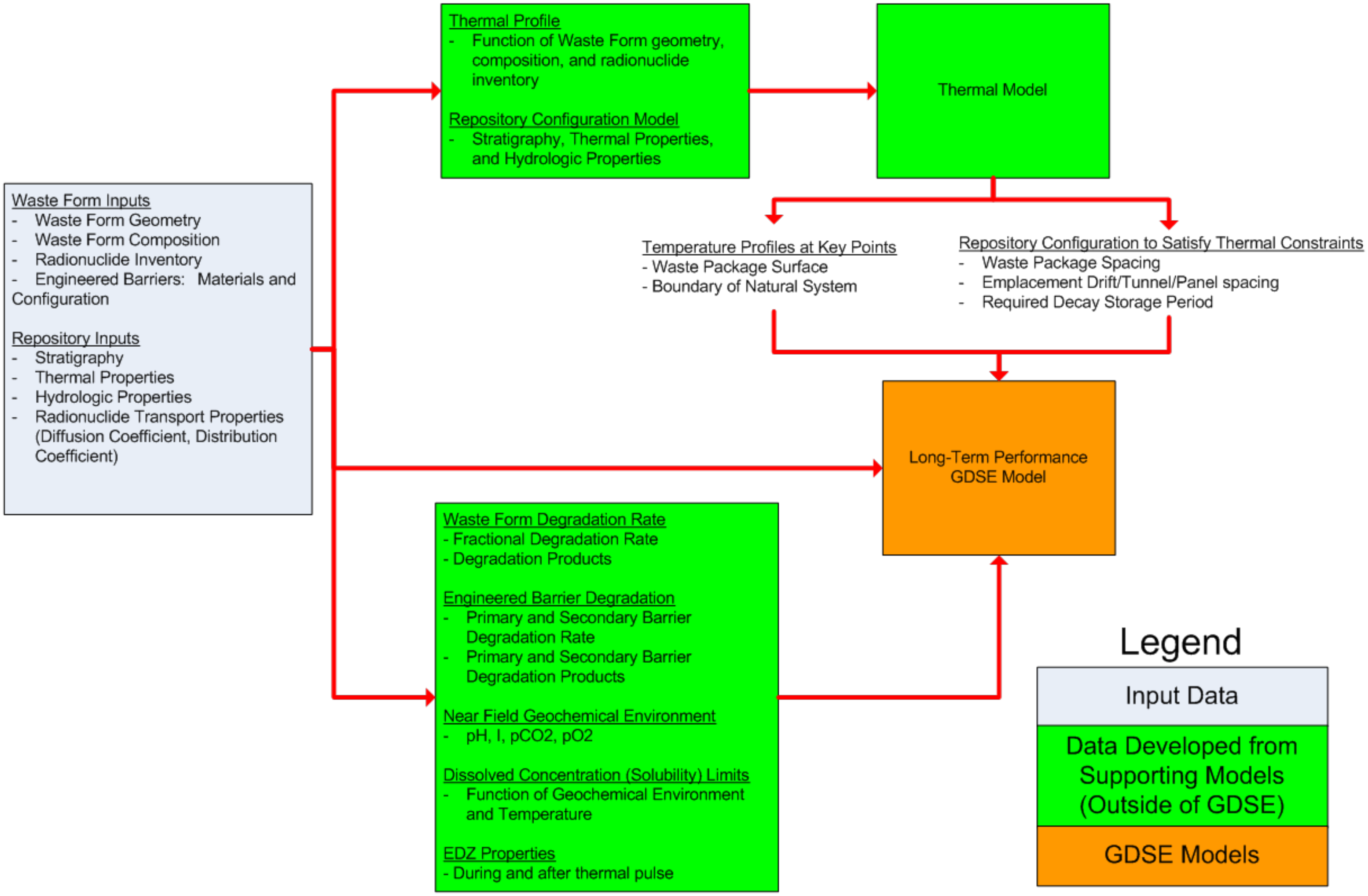
Model Descriptions using FEPs

Objective	Feature	Process [Issue]			Capability Included in Salt GDSE Model		Capability Included in Crystalline GDSE Model	
		UFD FEP ID	UFD FEP Title	Process/Issue Description	Yes / No	Discussion	Yes / No	Discussion
		0.0.00.00	0. ASSESSMENT BASIS					
All	System	0.1.02.01	Timescales of Concern		No		No	
All	System	0.1.03.01	Spatial Domain of Concern		No		No	
All	System	0.1.09.01	Regulatory Requirements and Exclusions		No		No	
All	System	0.1.10.01	Model Issues	-Conceptual model -Mathematical implementation -Geometry and dimensionality -Process coupling -Boundary and initial conditions	No		No	
All	System	0.1.10.02	Data Issues	-Parameterization and values -Correlations -Uncertainty	No		No	
		1.0.00.00	1. EXTERNAL FACTORS					
		1.1.00.00	1. REPOSITORY ISSUES					
Limited Release - Natural Barriers	Natural System - Geosphere	1.1.01.01	Open Boreholes	-Site investigation boreholes (open, improperly sealed) -Preclosure and postclosure monitoring boreholes -Enhanced flow pathways from EBS	No		No	
Containment; Limited Release Engineered Barriers	Engineered Barriers	1.1.02.01	Chemical Effects from Preclosure Operations	-Water contaminants (explosives residue, diesel, organics, etc.) -Water chemistry different than host rock (e.g., oxiding) -Undesirable materials left -Accidents and unplanned events	No		No	
Containment; Limited Release Engineered Barriers	System	1.1.02.02	Chemical Effects from Preclosure Operations -In EBS -In EDZ -In Host Rock	-Creation of excavation-disturbed zone (EDZ) -Stress relief -Boring and blasting effects -Rock reinforcement effects (drill holes) -Accidents and unplanned events -Enhanced flow pathways [see also Evolution of EDZ in 2.2.01.01]	No		No	
Containment; Limited Release Engineered Barriers	System	1.1.02.03	Mechanical Effects from Preclosure Operations -In EBS -In EDZ -In Host Rock	-Creation of excavation-disturbed zone (EDZ) -Stress relief -Boring and blasting effects -Rock reinforcement effects (drill holes) -Accidents and unplanned events -Enhanced flow pathways [see also Evolution of EDZ in 2.2.01.01]	No		No	

Waste Inventories and Repository Scenarios

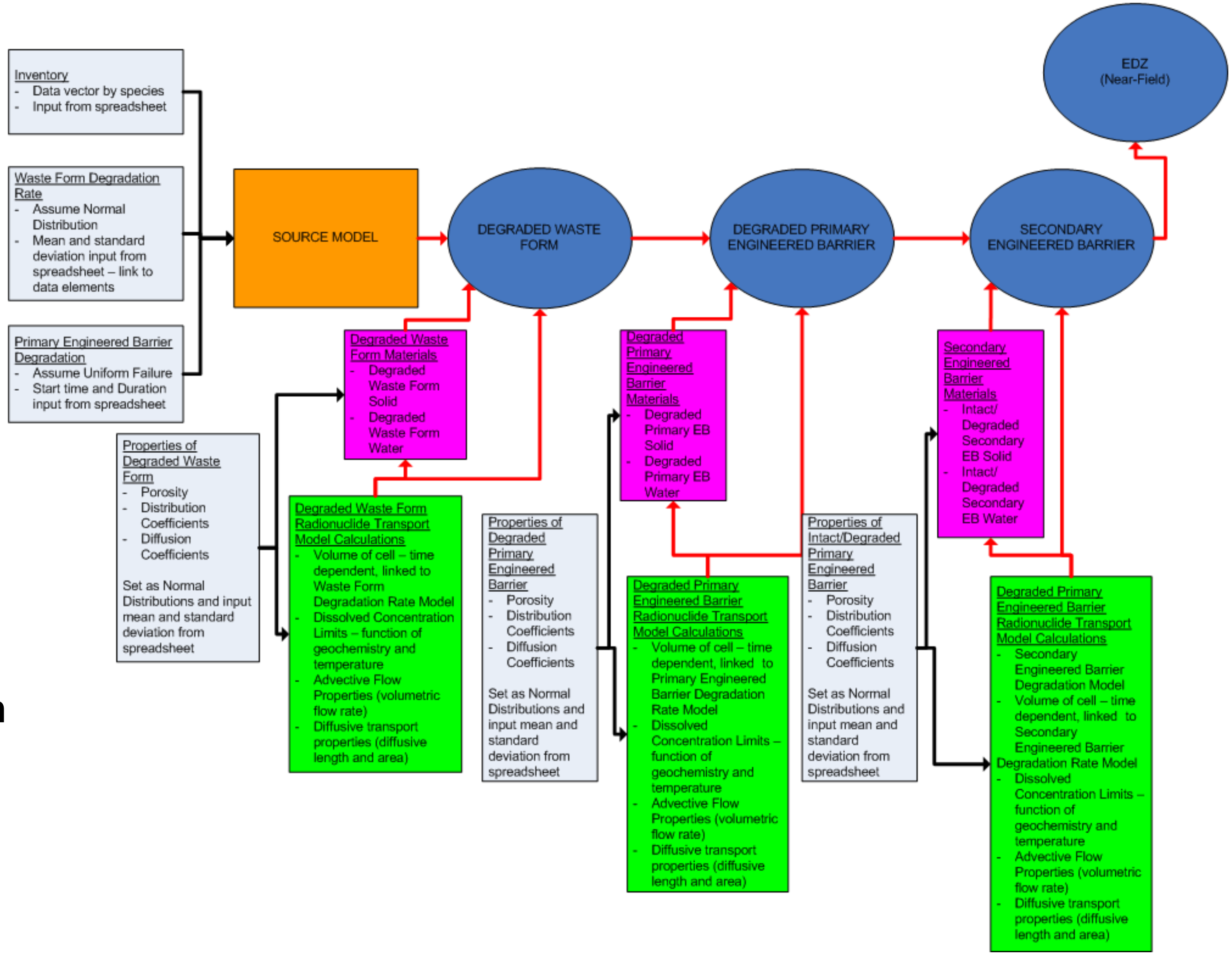
- **Waste types considered**
  - Commercial used nuclear fuel (UNF)
  - Existing DOE high-level radioactive waste (HLW)
  - HLW from reprocessing of commercial UNF
    - *Consider the reprocessing waste inventories in a recent UFD report (Carter and Luptak 2010)*
- **Repository Scenarios**
  - Undisturbed: Natural processes without disruptive events
  - Disturbed: Effects of human intrusion (i.e., drilling)

Clay GDSE Model Structure: Overall



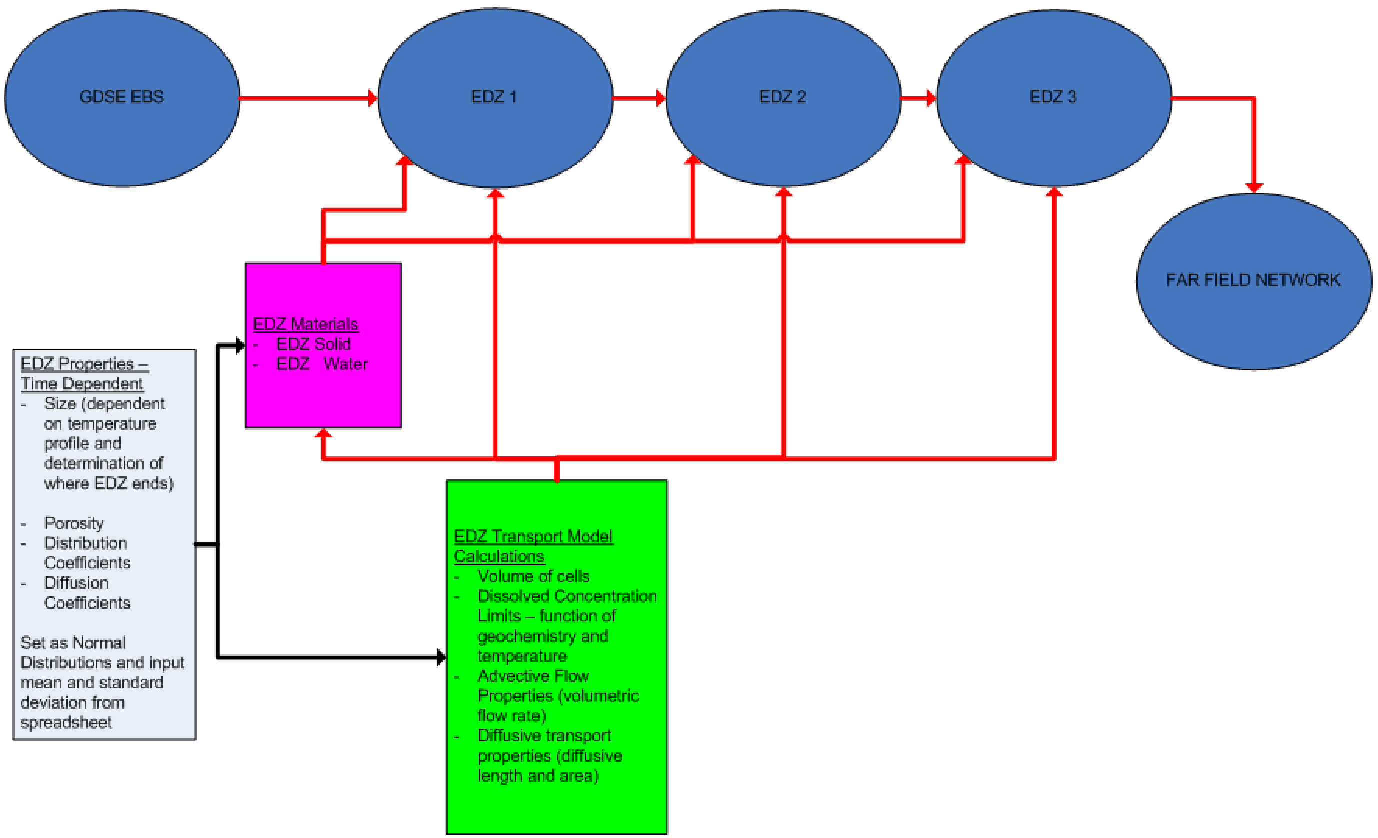
Clay GDSE Model Structure: Source Term and EBS

- Isothermal
- Fractional waste form fractional degradation rate
- Defined time for "primary" engineered barrier failure
- Diffusion and advection – Dissolved only
- Solubility limits
- Reversible sorption
- No temporal changes in properties



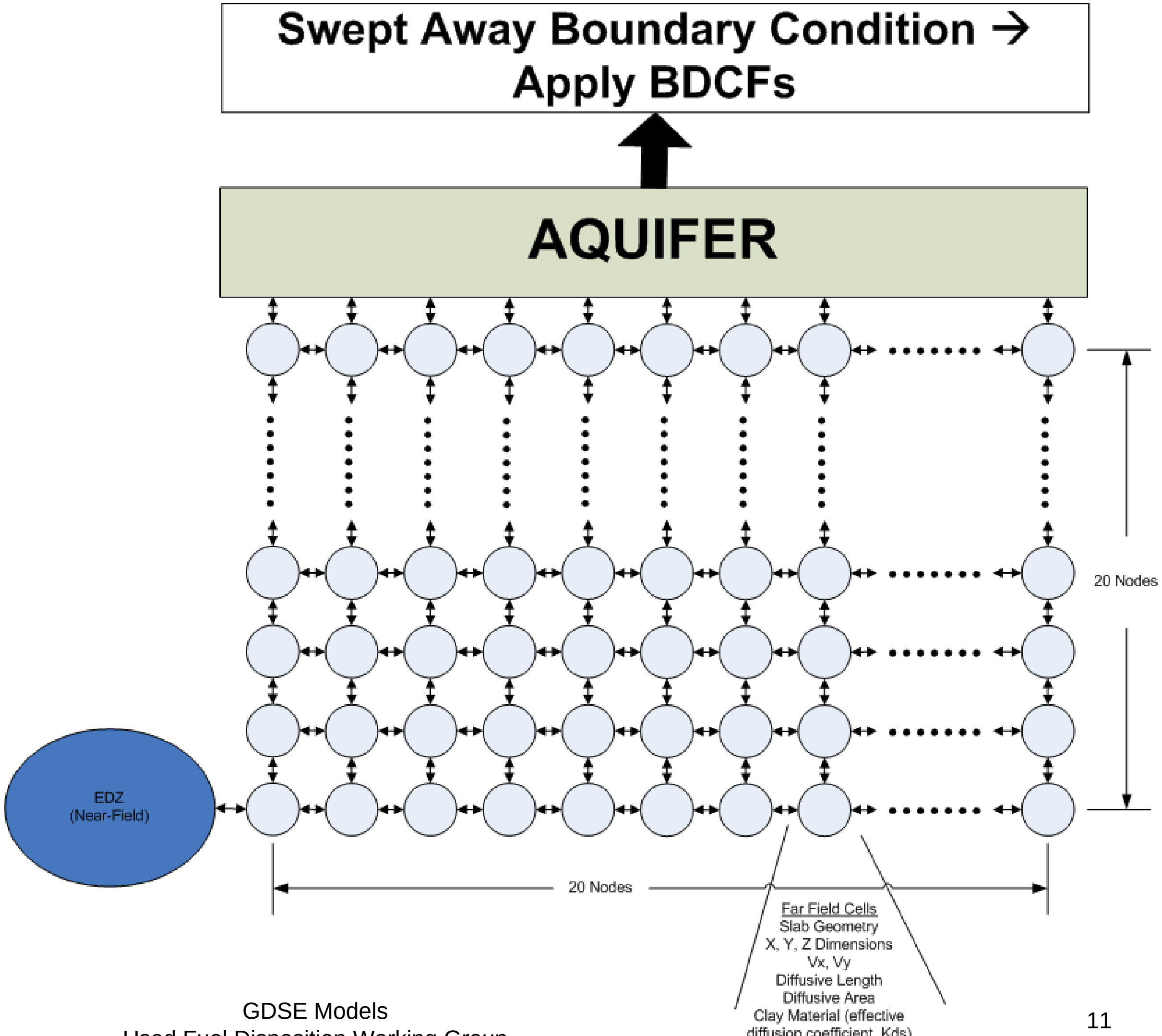
Clay GDSE Model Structure: Excavation Damage Zone

- Isothermal
- Diffusion and advection – Dissolved only
- Solubility limits
- Reversible sorption
- No temporal changes in properties



Clay GDSE Model Structure: Far Field

- **Isothermal**
- **Diffusion primarily – dissolved radionuclides only**
  - Capability for advection
- **Solubility limits**
- **Reversible sorption**
- **No temporal changes in properties**

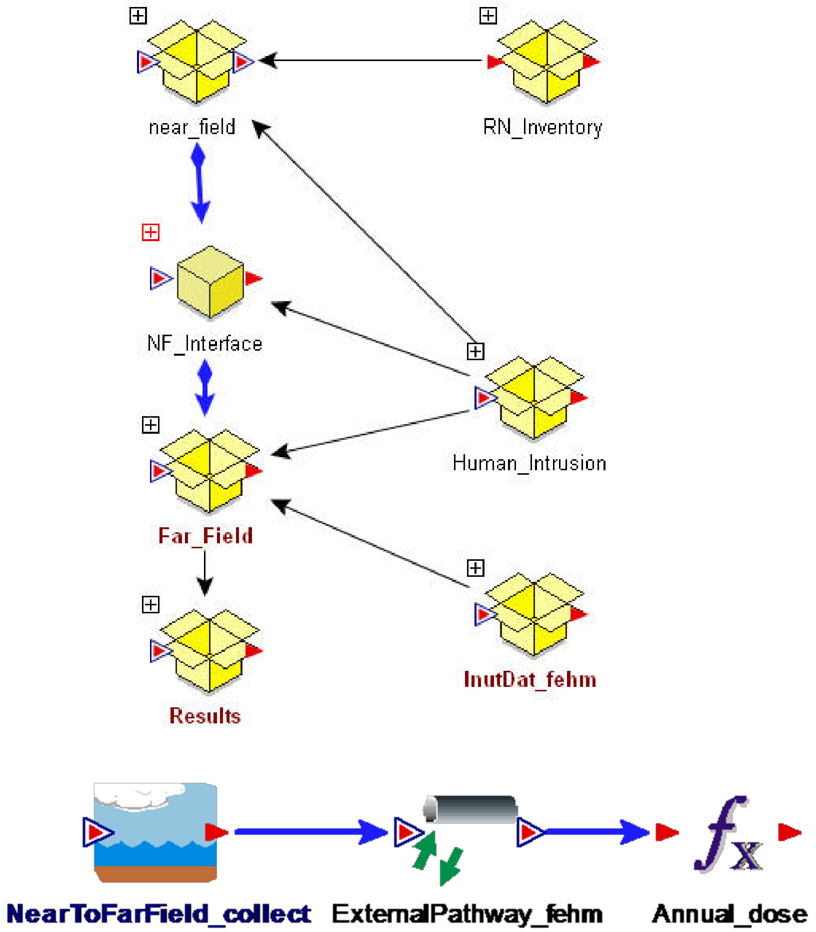


Clay GDSE Model: Status and Future Work

- **Status**
  - No significant model changes since FY10
    - *Minor clean-up*
    - *Awaiting completion of other GDSE models; will be merged into overall GDSE architecture*
    - *FEP cross-walk underway*
- **FY11 remaining work**
  - Complete re-structuring of model to prepare for merging into common GoldSim architecture
  - Complete FEPs cross-walk; identify if any key FEPs are missing and implement
  - Identification of needed parameters from ES and NS areas
  - Develop common GoldSim architecture (Primary effort)
- **FY12 and out-year proposed work**
  - Test unit-cell approach for design concepts developed
  - Integrate thermal analysis/thermal loading tools/output
  - Incorporate improved model for diffusive transport (LBNL lead effort)
  - Sensitivity analyses; support to FCT program efforts

Granite GDSE Model: Structure and Simulation

- Combined Near Field and Far Field model for Generic Disposal System Environments (GDSE)
- Determination of appropriate input boundary condition
- Treatment of daughter product in-growth
- Isotopic mix of key elements
- Development of response surfaces for all key radionuclides
- Demonstration simulations for helping develop waste form performance criteria
- Uncertainty and sensitivity analyses



Granite GDSE Model Development

Near field

- Encompasses the EBS and the interface with, and adjacent portion of, the host rock
- Repository layout
- Radionuclide inventory and waste form degradation
- Solubility control and radionuclide release from waste panels
- Solubility control at the near-field and far-field interface
- Human intrusion
- Diffusion through bentonite buffer (Add on to common GDSE source-term model)

Far field

- Represents 100s – 1000s of meters of natural system
- FEHM coupled with GoldSim (system level model) to represent far-field component (FEHM: The Finite Element Heat and Mass Transfer code)
- Radionuclide decay and ingrowth
- Advection (RTD residence time distribution-based transport model, enable study of potentially very heterogeneous domains)
- Matrix diffusion (GDPM generalized dual porosity model, diffusive exchange between flow porosity and surrounding rock matrix)
- Sorption
- Monte Carlo multi-realization probabilistic simulations with Latin Hypercube sampling
- Runtime input data altering program INPUTDAT

Granite GDSE Model: Source-Term

- **(Preliminary) Reference repository**
  - Consider the near-field as a large uniform mixing cell
  - Use degraded WP porosity and bedrock porosity to calculate total near-field void volume (i.e., total water volume)
  - Spacing between emplacement tunnels: 25; between WPs: 6 m
  - Repository layout will be finalized based on simplified repository thermal loading analysis

- **Inventory** (Based on Fuel Cycle Potential Waste Inventory by Joe Carter)

Commercial UNF	32154 WPs
Existing HLW (encapsulated in borosilicate glass)	5003 WPs
Reprocessing waste (RW, encapsulated in borosilicate glass)	4055 WPs

- **Repository waste inventories**
  - Case 1: UNF plus HLW
    - A total of 37,157 WPs (32,154 UNF WPs + 5,003 HLW WPs)
    - A square repository footprint with a side of 3,270 m
  - Case 2: HLW plus reprocessing waste (RW)
    - A total of 9,058 WPs (5,003 HLW WPs + 4,055 RW WPs)
    - A square repository footprint with a side of 1,615 m

Granite GDSE Model
- radionuclide release and transport scenarios

1. Undisturbed Scenario

- Assume barrier fail at beginning, release radionuclides by diffusion through bentonite buffer.
- Some failed waste packages intersect with fracture, which provide fast pathway for released RN to the aquifer.
- Waste packages affected (diffusion and intersect with fracture) sampled between 0.1 to 1 percent of inventory considered.
- **Inventory Case1**: UNF plus HLW
- **Inventory Case2**: Reprocessing waste plus HLW

2. Disturbed Scenario

- A single borehole penetrates through repository at 1,000 yrs, and provides a fast pathway for released radionuclides (RN) to the aquifer.
- Waste packages affected sampled between 1 and 5.
- **Inventory Case1**: Assume UNF WPs are affected by drilling intrusion. No HLW inventory is affected.
- **Inventory Case2**: Assume HLW WPs are affected by drilling intrusion. No reprocessing waste inventory is affected.

Granite GDSE Model: Future Work

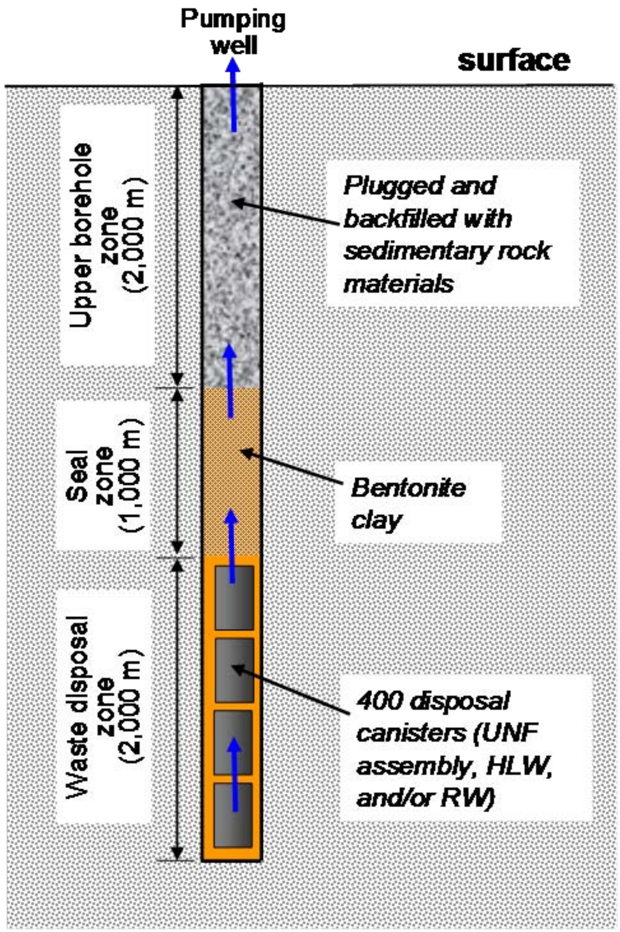
FY11 remaining work

- Consider different repository underground layout schemes
- Document model conceptual basis in terms of FEPs
- Compile list of parameters needed for granite model
- Continue/improve granite GDSE analysis
- Conduct sensitivity analysis
- Merge granite model into overall GDSE architecture

FY12 and out-year proposed work

- Continue to improve granite GDSE model to enhance flexibility and integration to address technical issues with minimal changes
- Continue to improve granite GDSE model by incorporating more detailed processes
- Develop thermal analysis tools for thermal loading and thermo-hydrologic response in generic granite repository
- Improve near-field chemistry for generic granite repository environment
  - *High ionic strength, elevated temperature, reducing condition*
  - *Solubility and sorption of RNs in near-field environments*
- Perform comparative studies among the different geologic disposal environments

Deep Borehole GDSE Model: Conceptual Model and Implementation



- **Conceptual model adopted from a recent study by Sandia National Labs (SAND2009-4401)**
- **Each borehole contains 400 disposal canisters**
- **Each canister contains one PWR assembly, HLW canister, or RW canister**
- **Assume the disposal zone temperature constant at 100°C**
- **Assume the borehole cross sectional area of 1 m^2**
- **Hypothetical accessible environment at the pumping well location**
  - Apply IAEA BIOMASS Example Reference Biosphere 1B (ERB1B) dose model

Deep Borehole GDSE Model: Waste Inventories and Scenarios

- **Waste inventories**
  - Case 1: UNF plus DOE HLW
  - Case 2: DOE HLW plus reprocessing HLW
- **Single scenario (Undisturbed)**
- **Not consider performance of disposal canisters**
- **Fractional degradation rate model for waste form degradation**
  - Waste form degradation and RN release at the beginning of the simulation
  - Canister treated as porous medium (3% porosity) representing corrosion products of canister and waste form

Deep Borehole GDSE Model: Mobilization and Transport

- **Radio-element solubility for two conditions**
  - Water in disposal and seal zones (reducing condition at elevated temperature (100°C))
  - Water in upper borehole zone (reducing condition at ambient temperature)
- **Analysis underway for water flows as a function of time and location in the borehole**
- **RN sorption in the three borehole zones**
  - Linear equilibrium sorption (Kd) model from the Sandia report and other sources
- **Performance measure matrix**
  - RN mass flux from major system components (e.g., disposal zone, seal zone, and upper borehole zone)
  - Mean dose at the hypothetical accessible environment

Deep Borehole GDSE Model: Future work

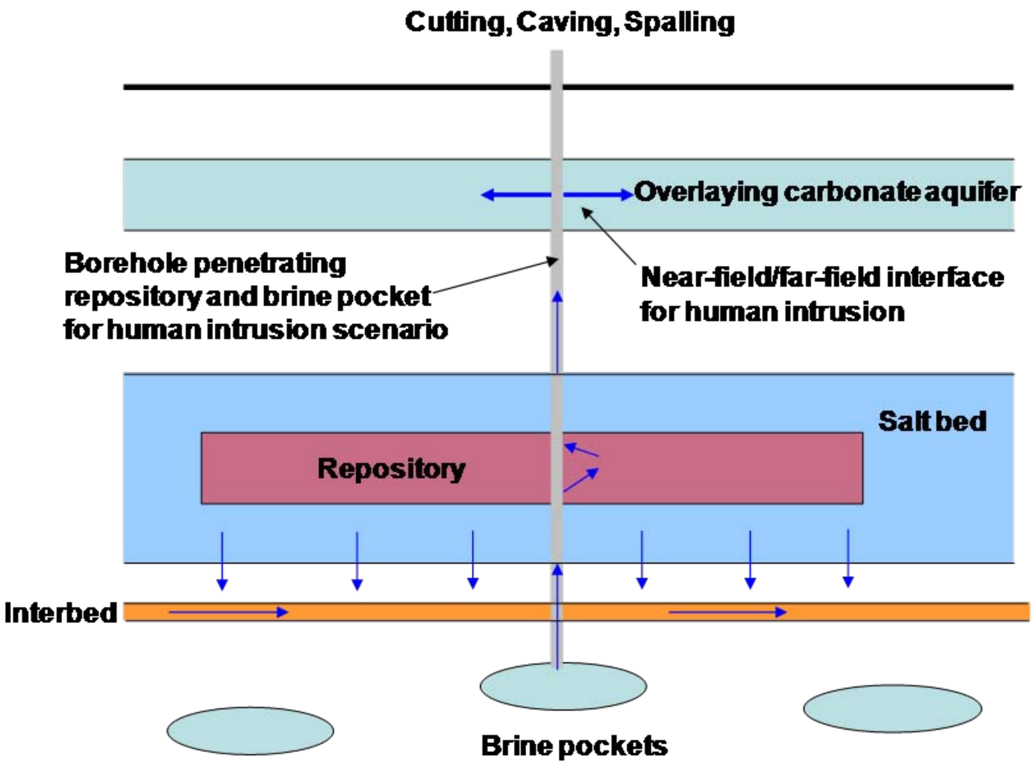
FY11 remaining work

- Complete abstraction of time- and location-dependent water flows in borehole and implementation in the deep borehole GDSE model
- Complete analysis and implementation of Kd models for the RNs with missing values
- Continue improvement of deep borehole GDSE analysis
- Conduct sensitivity analysis

FY12 and out-year proposed work (with other work packages)

- Improve analysis for thermal loading and thermal-hydrologic response in generic deep borehole repository
    - *Water movement and RN transport*
    - *Effect of neighboring boreholes*
- Improve near-field chemistry for generic deep borehole repository environment
    - *High ionic strength, elevated temperature, reducing condition*
    - *Solubility and sorption of RNs in deep borehole environments*
- Degradation of candidate waste forms in generic deep borehole repository environment

Salt GDSE Model: Conceptual Model and Implementation



- **Conceptual model developed using relevant salt site data**
  - Assume repository in a bedded salt formation
- **Undisturbed Scenario**
  - RN released into and transported in an interbed (1 m thick) below repository
- **Disturbed Scenario**
  - A single borehole penetration at 1,000 years
  - Sample the number of affected WPs (between 1 and 5)
  - RNs from affected WPs released directly to overlying carbonate aquifer by pressurized brines
- **Isothermal condition at ambient temperature**

Salt GDSE Model: Waste Inventories and Scenarios

- **Waste inventory cases for the Undisturbed Scenario**
  - Case 1: UNF plus DOE HLW
  - Case 2: DOE HLW plus RW
- **Waste inventory cases for the Disturbed Scenario**
  - Case 1: assume only UNF WPs affected
  - Case 2: assume only DOE HLW WPs affected
- **Fractional degradation rate model for waste form degradation**
- **Not consider WP performance**
  - Waste form degradation and RN release at the beginning of simulation
  - Treat the WP interior as porous medium of corrosion products of WP, internal components and waste form
- **Model the near-field as a mixing cell**

Salt GDSE Model: Mobilization and Transport

- **Radio-element solubility for two redox conditions**
  - Near-field brines (reducing condition)
  - Far-field brines (less reducing or slightly oxidizing condition)
- **Analysis underway for:**
  - Waste disposal area saturation
  - Brine flows from repository and in the interbed below repository
  - Analysis assumes initial dry-out zones around the waste disposal area
- **RN sorption in the near-field and far-field transport**
  - Linear equilibrium sorption (Kd) model
- **Performance measure matrix**
  - RN mass flux from major system components (e.g., near-field and far-field)
  - Mean dose at hypothetical accessible environment (5 km from the edge of repository)
    - *(IAEA BIOMASS Example Reference Biosphere 1B (ERB1B) dose model*

Salt GDSE Model: Future work

- **FY11 remaining work**
  - Complete brine flow analysis and abstraction for implementation in the salt GDSE model
  - Complete analysis and implementation of Kd models for the RNs with missing values
  - Continue/improve salt GDSE analysis
  - Conduct sensitivity analysis
- **FY12 and out-year proposed work (with other work packages)**
  - Develop thermal analysis tools for thermal loading and thermo-hydrologic response in generic salt repository, incorporating associated processes
    - *Salt creep and closure*
    - *Brine movement*
  - Improve near-field chemistry for generic salt repository environment
    - *High ionic strength, elevated temperature, reducing condition*
    - *Solubility and sorption of RNs in near-field environments*
  - Flow and RN transport in generic interbeds
  - Degradation of WP, candidate waste forms and other EBS components in generic salt repository environment
    - *Characterization and quantification of gases from corrosion under reducing condition*

Used Fuel Disposition Campaign

Development of an Improved PA Model for Diffusive Radionuclide Transport in Clay Rocks

Liange Zheng, Hui-Hai Liu, Jens Birkholzer, Carl Steefel (LBNL)
and Mark Nutt (ANL)

UFD Working Group Meeting

Jan 19-20, 2011

Background

- Performance assessment (PA) of nuclear waste disposal in a deep geological repository requires understanding and quantifying radionuclide transport through the hosting geological formation.
- Clay rocks are one important type of host formation studied in several countries. Examples include the Opalinus Clay at Mont Terri in Switzerland *(Wersin et al., 2007, Soler et al., 2008, Samper et al., 2006, Palut et al., 2003)*, the Toarcian clayey formation of the Tournemire experimental site in France *(Motellier et al., 2007)*, and the Callovo–Oxfordian clay at ANDRA's underground research facility in France *(Appelo et al., 2008)*.
- In the absence of fractures, diffusion is the main transport mechanism for radionuclide transport in these formations *(e.g. Motellier et al., 2007)*.
- Both mechanistic approaches, which couple diffusive and electro-chemical processes *(Revil and Leroy, 2004; Appelo et al., 2010; Bourg et al., 2003; Jougnot et al., 2009)* and phenomenological approaches which are based on Fick's law and retardation factors (*eg. Soler et al., 2008, Samper et al., 2006)* have been used to simulate diffusion through dense compacted clays.

Used Fuel Disposition

Mechanistic Approach

- From Appelo et al. (2008; 2010):

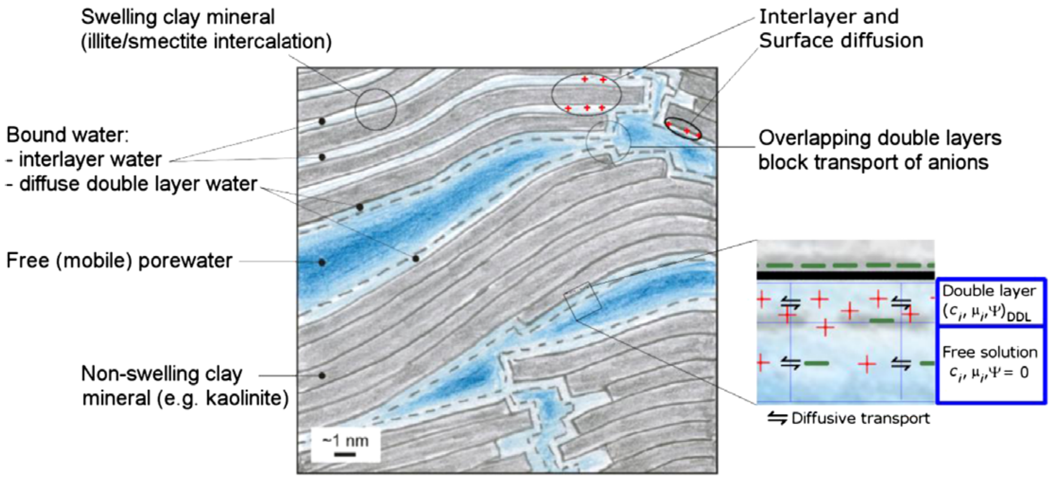


Fig. 1. A diagram of the porespace in Opalinus Clay, showing three water-types with associated diffusion domains (modified from NAGRA, 2002b). Right hand side: representation of a pore in PHREEQC.

- Diffusive flux of species i is the result of both chemical and electrical potential gradients:

$$J_i = -\frac{u_i c_i}{|z_i| F} \frac{\partial \mu_i}{\partial x} - \frac{u_i z_i c_i}{|z_i|} \frac{\partial \psi}{\partial x}$$

- The double diffusion layer (DDL) is explicitly incorporated. The concentrations in DDL are linked to the concentration in free solution by Boltzmann's equation, assuming Donnan approximation:

$$c_{i,\mathrm{DDL}} = c_i exp\left(\frac{-z_i F \psi_{\mathrm{DDL}}}{RT}\right)$$

Phenomenological Approaches: An Example

- Diffusive transport equation :

$$\phi_a R \frac{\partial C}{\partial t} = \nabla \cdot (\overline{D}_e \cdot \nabla C)$$

ϕ_a is accessible porosity, D_e is effective diffusion coefficient, R is retardation coefficient :

$$R = 1 + \frac{\rho K_d}{\phi_a}$$

- While this kind of approach cannot capture the detailed transport mechanisms, it is relatively simple, computationally efficient, and straight-forward to implement. It might be preferred in a PA model for large-scale diffusive transport in clay rock.

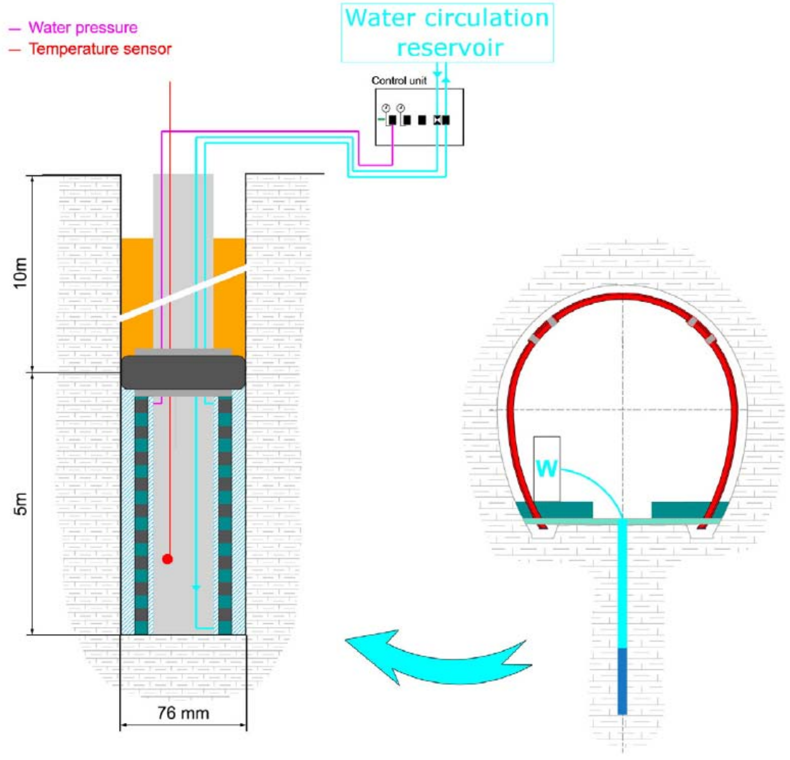
Work Plan

- Literature survey of diffusion test data and their analyses (FY11).
- Evaluation and comparison of mechanistic and phenomenological modeling approaches (FY11).
- Evaluation of the relative importance of spatial variability on diffusion processes
- Developing a particle-tracking numerical modeling approach for radionuclide transport in clay rock
- Performing radionuclide transport simulations for typical clay-rock systems

Literature Survey (1)

- Literature survey of diffusion test data --- finding diffusion test and using them to evaluate different diffusion model.

In situ test for Callovo–Oxfordian clay rock (Appelo et al., 2008) has been successfully interpreted with a complex model using a mechanistic approach --- Can the simple model using a phenomenological approach do an acceptable job?



Literature Survey (2)

- Literature survey of diffusion test data and their analyses --- the heterogeneity of diffusion coefficient.

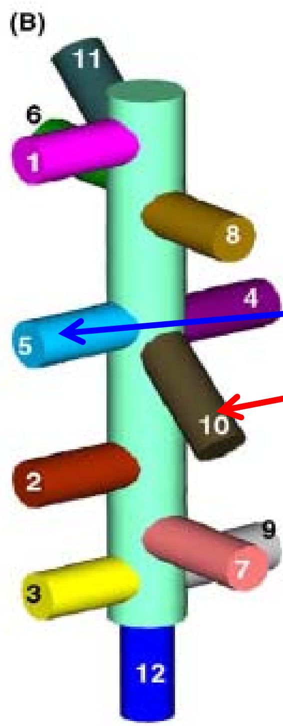
Tracer	Parameter $D_e \times 10^{-11}$ (m^2/s) K_d (L/kg)	In situ test DI-A2 this work	In situ test DI-A1[a]
HTO	D_e	6.0	5.4
	ε	0.15	0.18
I^-	D_e	3.0	1.3
	ε	0.08	0.09
Br^-	D_e	3.0	
	ε	0.10	
$^{85}Sr^{2+}$	D_e	7.0	
	ε	0.15	
	K_d	1.0	
Cs^+	D_e	20.0	30.0
	ε	0.15	0.18
	Freundlich isotherm	$S = 0.186\ C^{0.53}$	$S = 0.186 C^{0.53}$

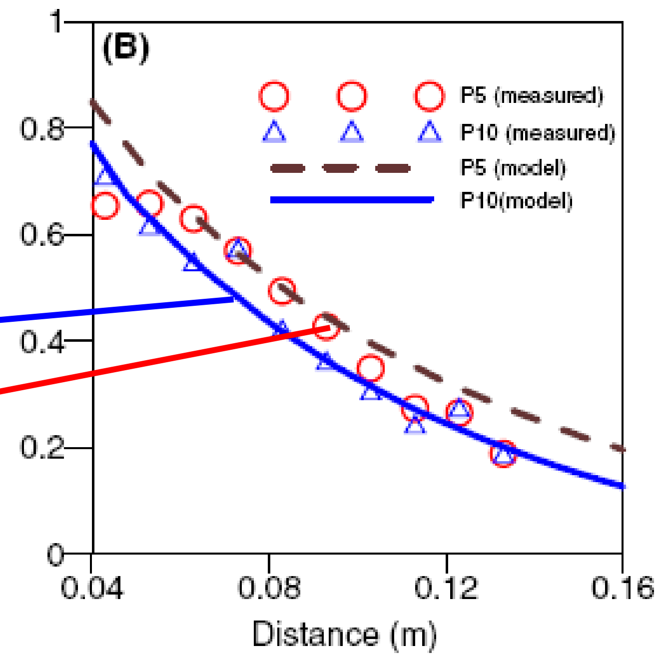
Locations of In situ tests DI-A2 (Wersin et al., 2008) and DI-A1 (Van Loon et al., 2004) are 1 m apart. The different diffusion coefficients for HTO, I^- and Cs^+ show heterogeneity. Is this important for large scale diffusion?

Literature Survey (3)

- Literature survey of diffusion test data and their analyses --- the anisotropy of diffusion.

DI-B test for Opalius clay (Samper et al., 2008, Soler et al., 2008) shows anisotropic diffusion --- how does anisotropic diffusion affect large scale transport of radionuclide in host clay rocks?





Deuterium diffusion in profile 5 and 10

Questions and Discussion