Disposal Systems Evaluations & Tool Development

SAND2010-0361C

- Work Package 1
  - Initiate evaluation of natural system performance attributes and modeling, focusing on those systems still in the conceptual stage (e.g. deep boreholes) and other environment not studied in significant detail.
- Work Package 2
  - Continue FY09 WF Campaign Generic Disposal System Environment (GDSE) modeling Finalize granite & clay models, begin inclusion of conceptual engineered barrier systems, and initiate model of other concepts (deep borehole, enhanced confinement).
- FY10 Focus
  - Work planning: short term activities, long-term vision
  - Package 1: Current status overview, concept development, experimental design
  - Package 2: Modeling tool development, data integration
  - Integration of two activities at each lab

Team

- **ANL:**
  - Mark Nutt & others
- **INL:**
  - Michael Simpson
- **LANL:**
  - Shaoping Chu
- **LBNL:**
  - Hui-Hai Liu
- **LLNL:**
  - Susan Caroll
- **SNL:**
  - Yifeng Wang
  - Joon Lee
  - Carlos Jove Colon
  - Frank Hansen

Work Package 1: Specific activities for FY10

- Direct disposal of electrochemical refinery waste
  - **SNL:** Concept development & integration
  - **INL:** Support on the fuel reprocessing side
- Key natural system attributes related to near field
  - **SNL**
- Key natural system attributes related to far field hydrology and radionuclide transport
  - **LANL:** Literature survey, gap analysis for key attributes, technical approach development
  - **SNL:** Integrating LANL analysis into FY final report
- Detailed clay repository study: Mechanical-chemical-hydrological couplings in the near field
  - **LBNL:** Literature survey, gap analysis for key attributes
  - **SNL:** Integrating LANL analysis into FY final report
- Study of radionuclide sorption irreversibility
  - **LLNL:** Literature survey, gap analysis for key attributes, technical approach development
  - **SNL:** Integrating LANL analysis into FY final report
- Focus
  - Technical gap analysis
  - Long-term plan

Work Package 2: Specific activities for FY10

- Model development and simulation for a clay environment - ANL
- Model development and simulation for a granite environment - LANL
- Model development and simulation for a deep borehole environment and refinement of the model for a salt environment - SNL
- Clay swelling calculation to demonstrate modeling capability - LBNL
- Test plan for studying radionuclide sorption irreversibility - LLNL
- Decay heat-induced ambient rock melting in deep boreholes – SNL
- Focus:
  - Uniform assumption across all GDSEs (e.g. WFs)
  - Comparison among GDSEs
  - Feedback to WF development or other activities

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Nuclear Energy

Fuel Cycle Research and Development

Clay Generic Disposal System Modeling

Mark Nutt, Ted Bauer

Argonne National Laboratory

Used Fuel Disposition Campaign Working Group Meeting

January 29, 2010

Radioactive Waste Disposal and Clay Environments

- **Clay formations potentially have features conducive to waste isolation**
  - Stable
  - Low permeability
  - Self sealing
  - Sorptive
  - Reducing
  - Diffusive dominated
  - Buffering
- **Clay buffers (bentonite) are included as engineered barriers in several disposal system concepts**
- **Paraphrased quote: "If the intent is to reduce risk associated with disposal, I'll take a few meters of clay over a transmutation reactor any day."**
  - Source remains anonymous, but a geologist in this room

Radioactive Waste Disposal and Clay Environments

- **Cigar Lake ore deposit has survived roughly 1.3 billion years of geologic history, chiefly because of its natural clay buffer**
- **Clay reduces both the penetration of groundwater into and the diffusion of uranium out of the ore deposit**
- **The deposit has remained intact through several mountain-building episodes (the Rocky Mountains, the Appalachians), the trauma of continental drift, multiple ice ages, and significant uplift caused by the erosion of over 2.5 km of overlying sedimentary rock**
- **So stabilized in its position (430 meters below the surface) that no chemical or radioactive signature can be detected on the ground surface**

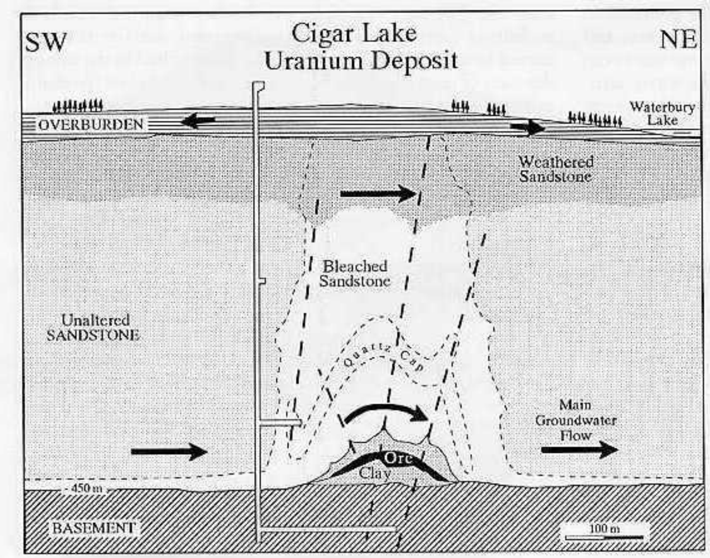


Fig. 3. Cross section through the Cigar Lake uranium deposit parallel to the direction of regional groundwater flow. Heavy dashed lines represent major faults, and heavy arrows indicate the general flow direction of groundwater.

[From J. Cramer, p. 40]

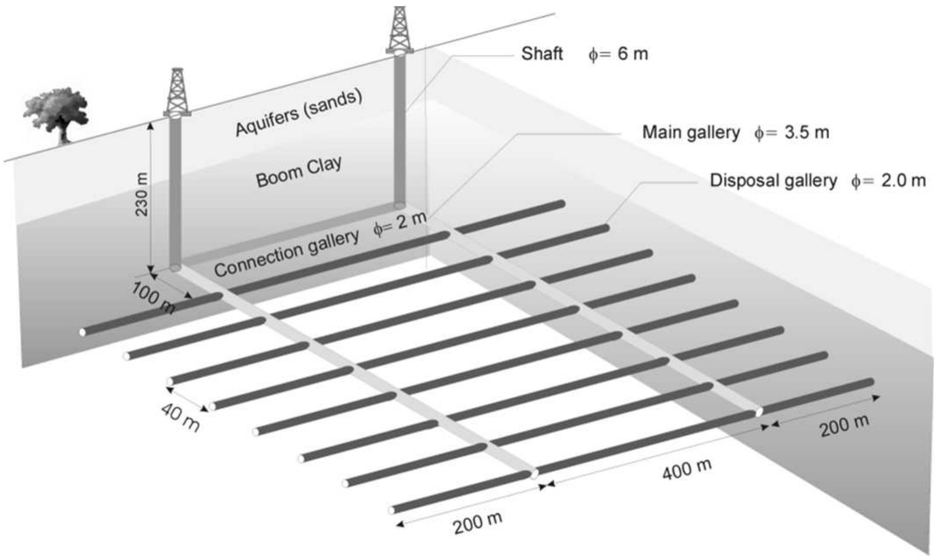
www.nuclearfaq.ca/cnf_sectionE.htm

Radioactive Waste Disposal and Clay Environments

- **Clay is being considered as a host geologic environment in Belgium (Boom Clay), France (Meuse/Haute Marne) and Switzerland (Opalinus Clay)**
- **Clay was not identified in the past as a potential environment for disposing used nuclear fuel or high level nuclear waste in the United States**
- **Clay formations have been identified as sites for locating low-level disposal facilities in the U.S.**
  - Surface disposal
  - Barnwell, Andrews County Texas, Illinois
- **Clay may be suitable for wastes generated from an advanced fuel cycle**
- **Issues**
  - Homogeneity, lateral extent, depth, thickness, thermal constraints

Modeling of a Clay GDSE

- **Initial model developed in FY09 for a clay generic disposal environment (GDSE)**
- **Clay model development based on existing studies**
  - Belgium - Boom Clay: SAFIR 2 Report
  - France – Meuse/Haute-Marne: Dossier 2005 Argile
- **General characteristics of modeled clay GDSE**
  - Saturated
  - Diffusion dominated
    - *Hydraulic conductivity and hydraulic gradients are very small*
  - Reducing conditions
    - *pH = 8.2; Eh = -0.250 (mV/SHE)*



Modeling of a Clay GDSE

Element	Solubility Limits in Boom Clay [Mol/L][1]				Retardation Coefficient[2]
	Best Estimate	Minimum	Maximum	Distribution Type	
Ac	2.0E-06	5.0E-08	5.0E-06	Log-Triangular	1000
Am	2.0E-06	5.0E-08	5.0E-06	Log-Triangular	1000
Nb	3.2E-06	1.0E-08	1.0E-03	Log-Uniform	50
Np	1.0E-06	1.0E-10	1.0E-05	Log-Triangular	1000
Pa	1.0E-05	5.0E-11	2.0E-05	Log-Triangular	400
Pd	1.0E-07	1.0E-09	1.0E-05	Log-Uniform	20
Pu	5.0E-07	1.0E-09	5.0E-05	Log-Triangular	1000
Ra	1.0E-09	1.0E-10	1.0E-05	Log-Triangular	50
Se	5.5E-08	1.0E-09	3.0E-06	Log-Uniform	1
Sn	5.5E-07	1.0E-09	1.0E-05	Log-Uniform	20
Tc	3.0E-08	4.0E-09	5.0E-08	Log-Triangular	1
Th	5.0E-07	1.0E-10	1.0E-06	Log-Triangular	400
U	3.2E-08	1.0E-10	1.0E-05	Log-Uniform	300
Zr	1.0E-06	1.0E-09	1.0E-03	Log-Uniform	400

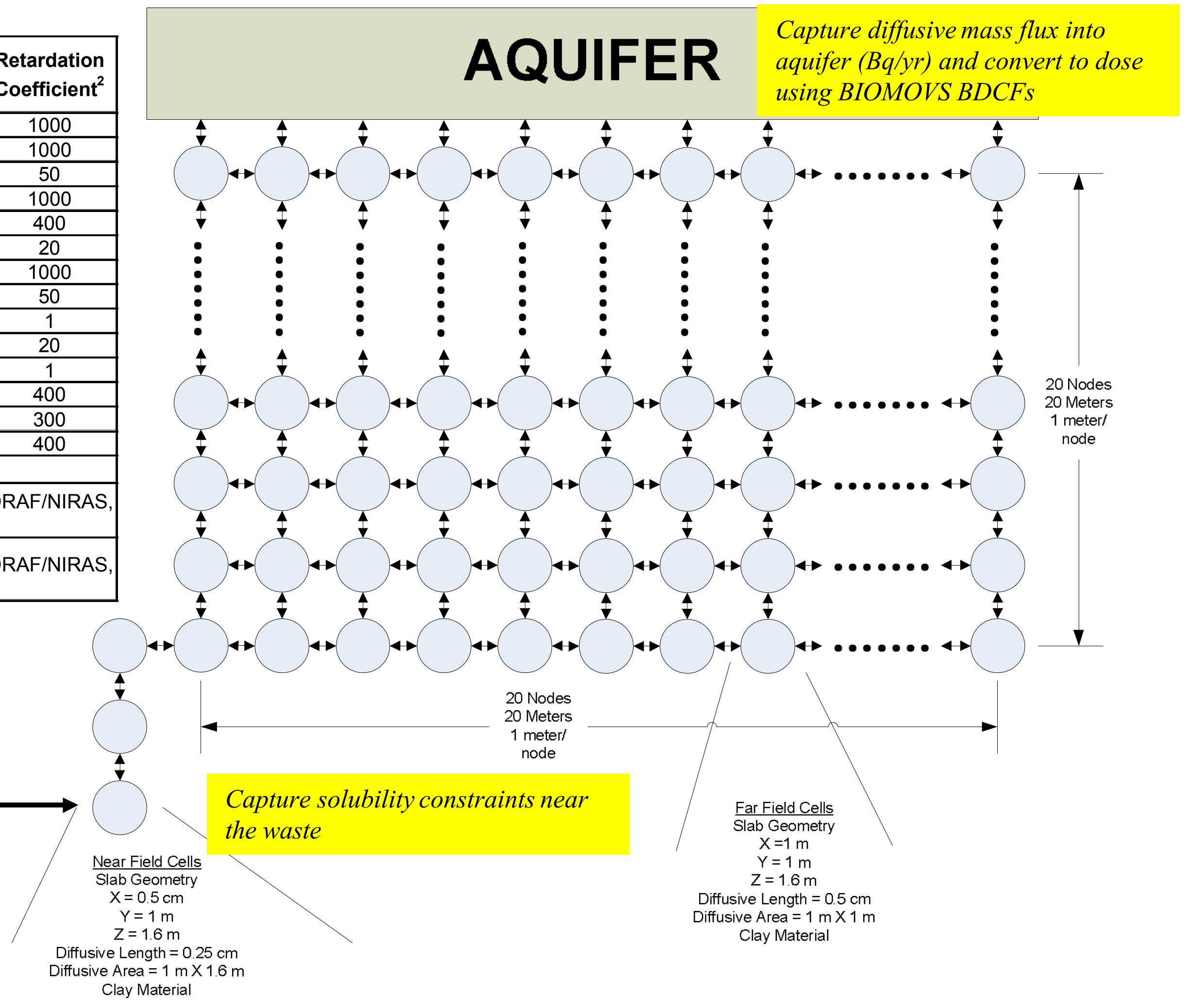
Diffusion Coefficient[2]: 2E-10 m^2/s, except for Cs - 3.6E-10 m^2/s

[1]SAFIR 2, Safety Assessment and Feasibility Interim Report 2, ONDRAF/NIRAS, NIROND 2001-06 E, December 2001, Table 11.3.8-3

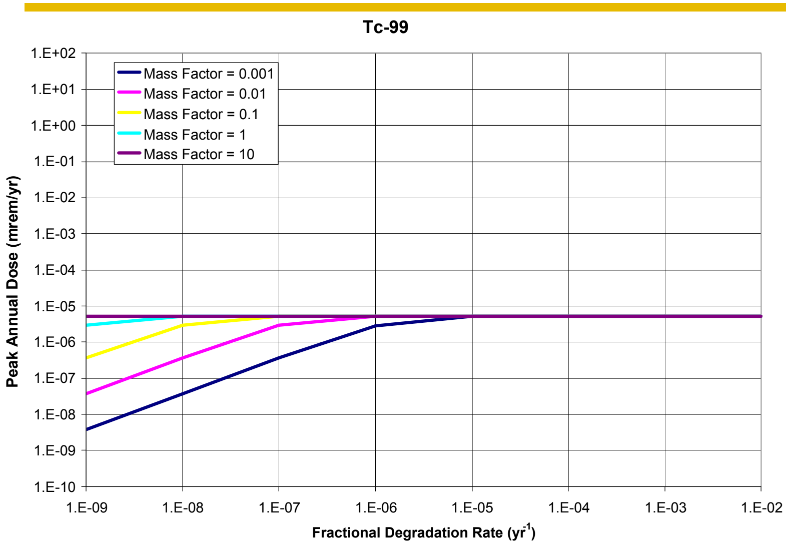
[2]SAFIR 2, Safety Assessment and Feasibility Interim Report 2, ONDRAF/NIRAS, NIROND 2001-06 E, December 2001, Table 11.3.8-4

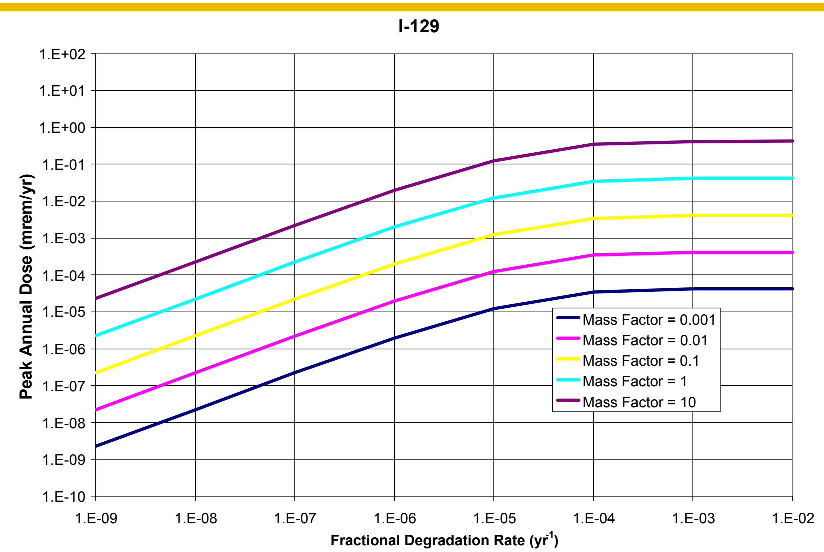
$$\dot{m}_j(t) = F_{wf} \bullet I_{WF,j}(t)$$

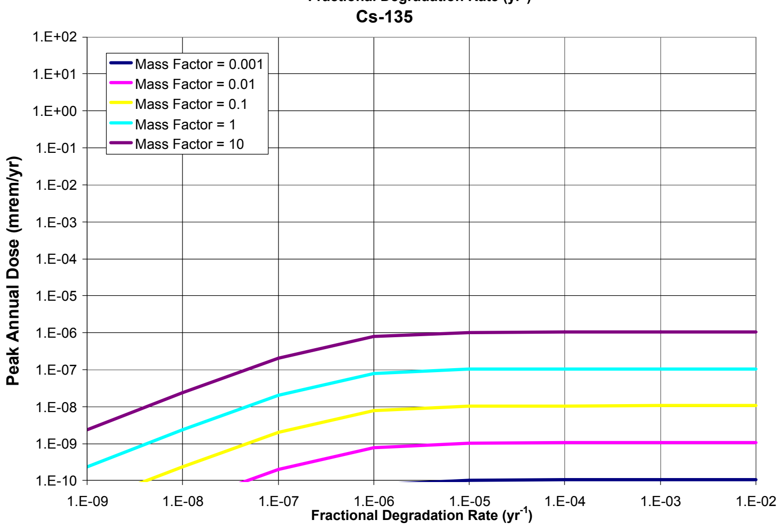
$$I_{WF,j}(t) = I_{WF0,j} \exp^{-(F+\lambda_j)t}$$



Modeling of a Clay GDSE

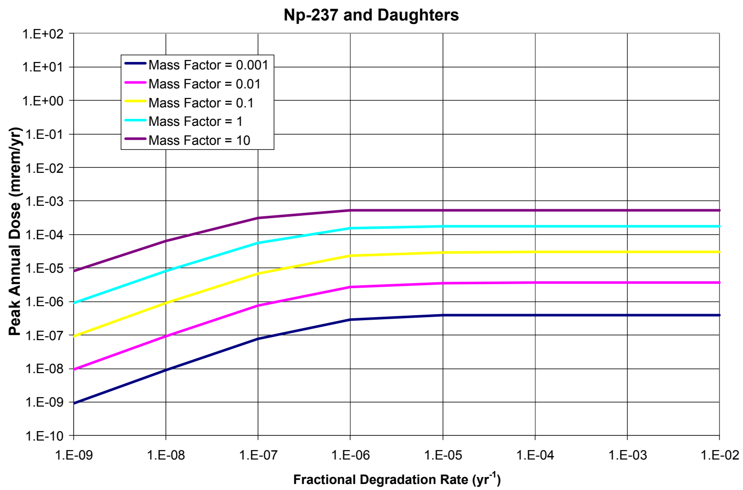


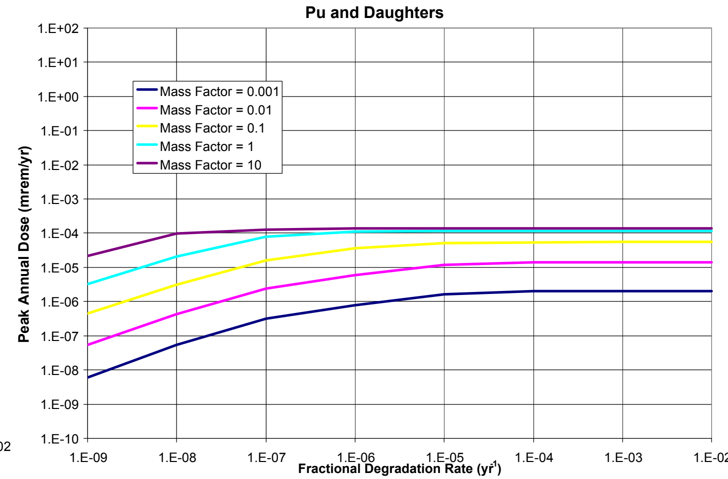


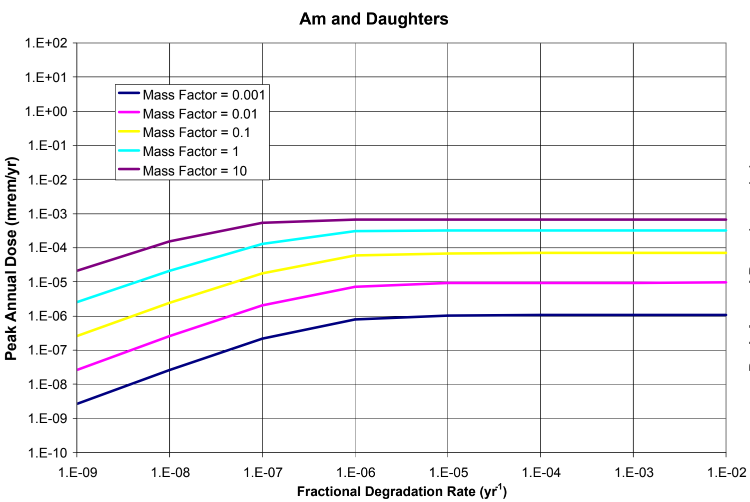


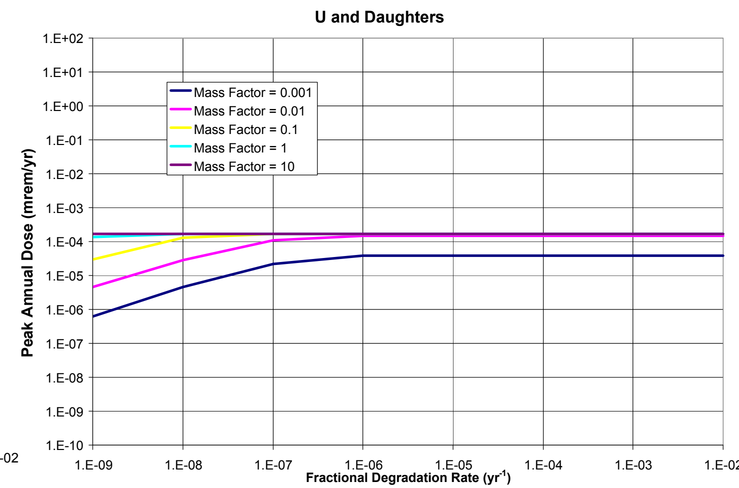
- **Tc solubility limited in clay**
- **I and Cs: Proportional release rate dependence until characteristics of natural system control**

Modeling of a Clay GDSE









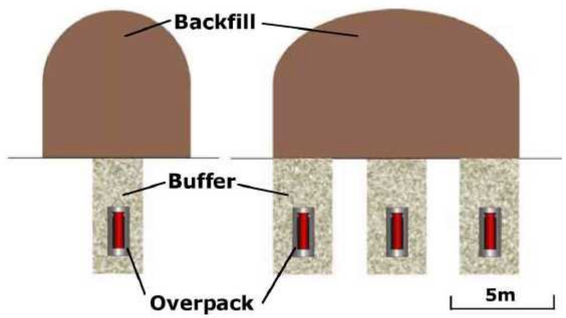
- Proportional release rate dependence except at lower degradation rates
- Solubility control and natural system breakthrough characteristics at higher degradation rates

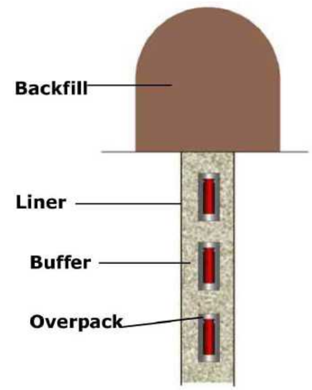
Potential FY10 Activities

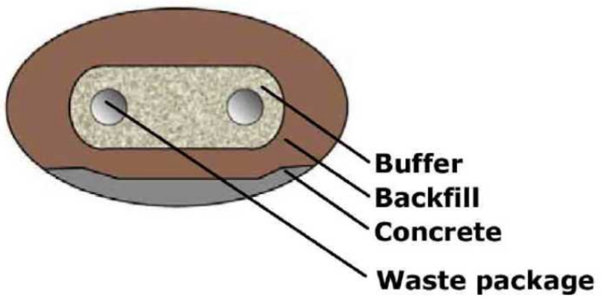
- **Peak of the mean instead of mean of the peak**
- **Sensitivity analyses with current model**
  - Thickness
  - Spacing
  - Properties
  - Advective transport
  - Suggestions?
- **Develop model for a "shallow" generic disposal concept**
- **Thermal Analysis**
  - Available information suggests a 100 degree C temperature limit in clay
  - Waste loadings affects thermal output – unknown
  - Different design alternatives for different thermal loadings
  - Design alternatives affect disposal system performance (boundary condition)

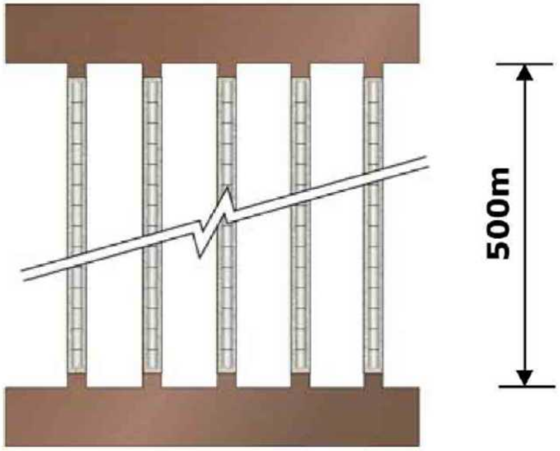
Potential FY10 Activities

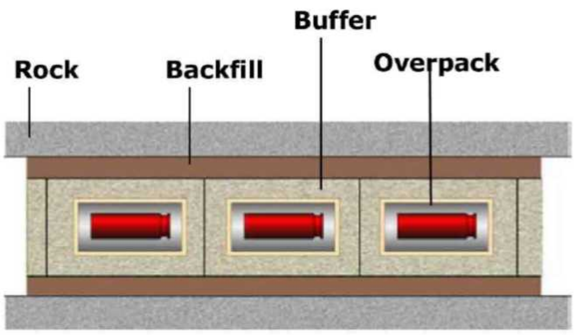
- Design Concepts Considered in Japan (NUMO-TR-04-03)

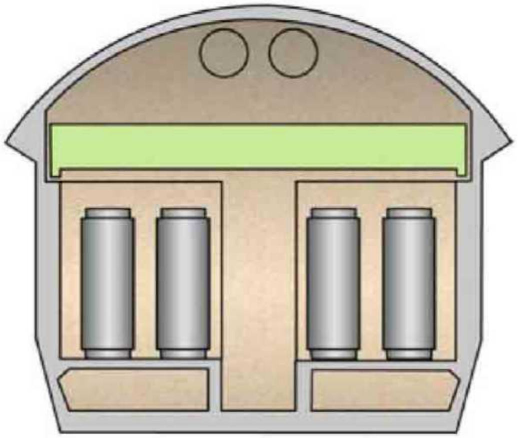












Potential FY10 Activities – General 3-D Thermal Modeling

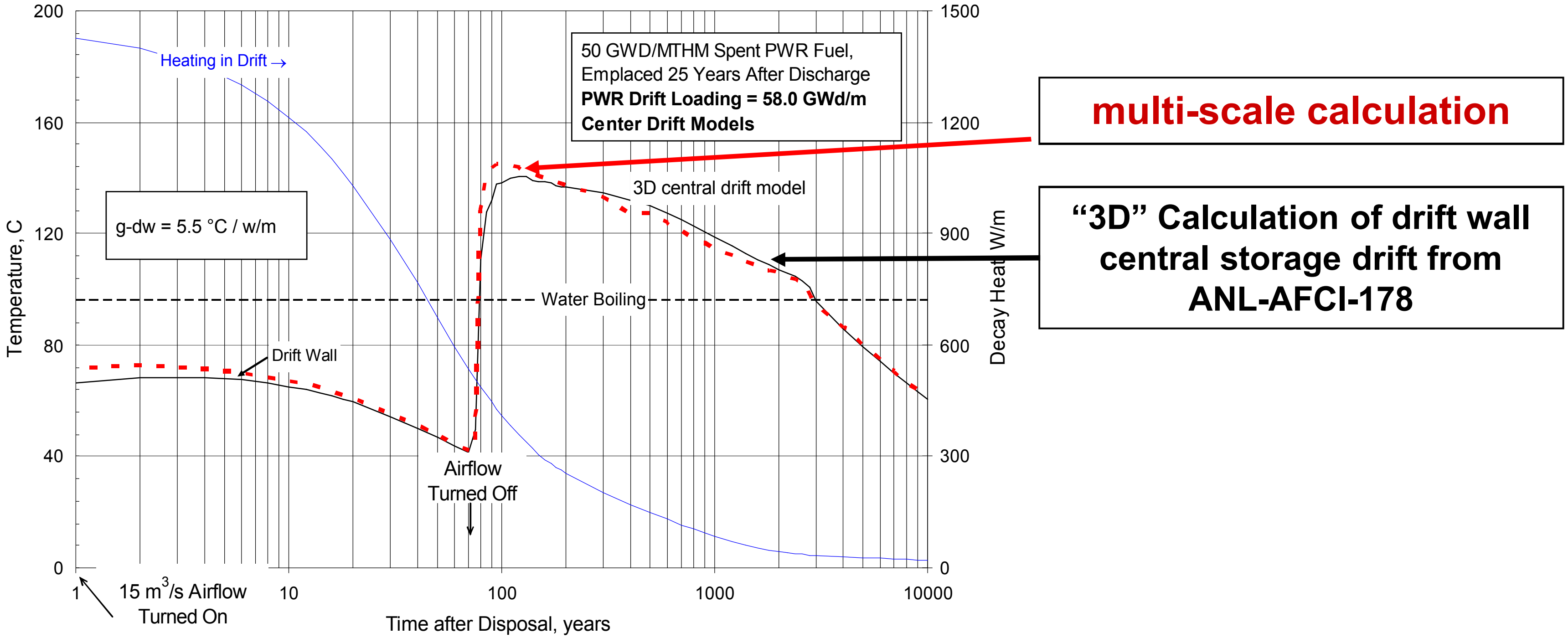
- **Develop a multi-scale technique for accurate and efficient computation of temperature fields within enclosed storage units and waste packages as well as over extended regions of host rock**
  - Utilize techniques and tools used in previous AFCI analyses
  - Capable of rapid evaluation of a variety of design concepts in different disposal environments – **<u>initial application for clay</u>**
- **Utilize coupled thermal models that separately address thermal behaviors at very different "length scales"**
  - Storage Unit Model: enclosed room, gallery, or tunnel that contains heat-generating waste packages. Unit may be externally cooled or ventilated.
  - Host Rock Model: the repository's surrounding host rock and its boundaries

Potential FY10 Activities – General 3-D Thermal Modeling

- **Modeling Assumptions**
  - Host rock nodes are sized so that storage units account for only a small volume fraction
  - Temperature fields in storage units respond to changes in waste package decay heat much more rapidly than those in surrounding host rock
- **Solution Method**
  - The transient host rock thermal model is solved directly on the basis of host rock material properties, boundary conditions, and the heat generated by any enclosed storage units. Generated heat is included through simple "point" or "line" sources within host rock nodes
  - Temperature fields within each storage unit are determined by a steady-state solution of the storage unit model assuming its known heat generation rate and using the temperature in the surrounding host rock (as determined from the host rock model) as a nearby boundary condition

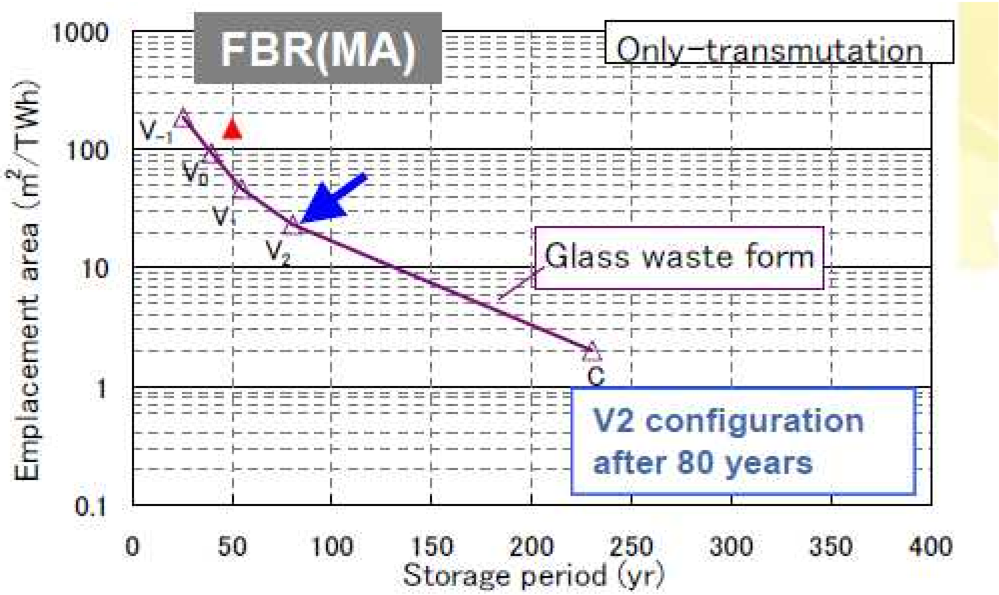
Potential FY10 Activities – General 3-D Thermal Modeling

- **Technique has been used and "verified" for tuff (Yucca Mountain)**



Potential FY10 Activities – General 3-D Thermal Modeling

- **Ability to parametrically evaluate design alternatives, repository loading density, and decay storage requirements for wastes with different radionuclide concentrations**
- **Applicable to all generic disposal environments – develop and demonstrate initially for clay GDSE**
- **Potential "follow-on" use as a systems analysis tool – similar to method developed by Tracy Radel (University of Wisconsin)**



From "Systems Analysis – An Example: Effects of separation of heat generating FPs and MAs on repository footprints", Kenji Nishihara, JAEA, Presented at the JNEAP Waste Management Working Group, December 8, 2009.

Scale Dependence of Effective Matrix Diffusion Coefficient and Modeling Coupled THMC Processes in Clay Repositories

H.H. Liu
LBNL

UFD Working Group Meeting
January 28-29, 2010

Outline

- **Scale dependence of effective matrix diffusion coefficient**
  - Model analysis results of field tests
  - Mechanisms
  - Implications

- **Modeling Coupled THMC Processes in Clay Repositories**
  - Numerical codes
  - Modeling approaches

Modeling Analysis of Alcove 8/Niche 3 Flow and transport Tests in Yucca Mountain: *A Surprise*

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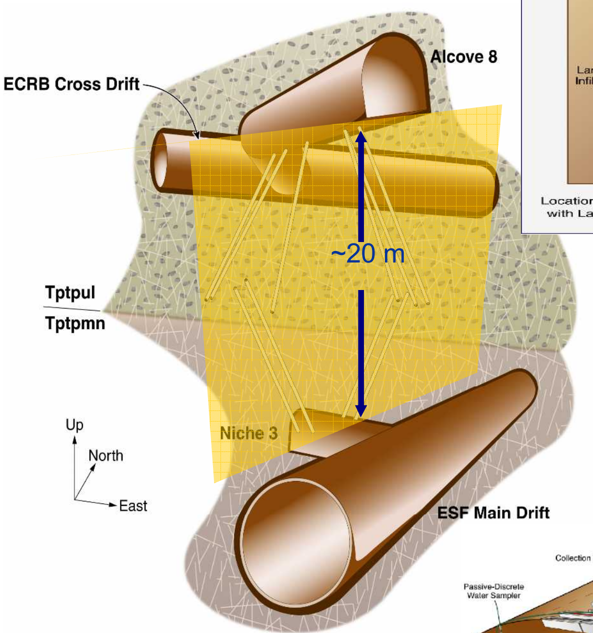
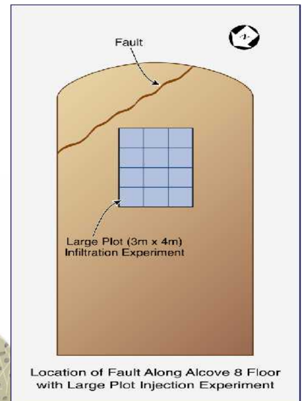
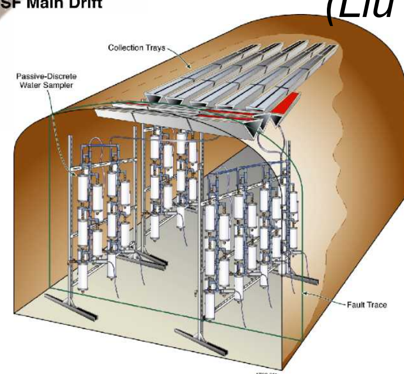
Field Tests

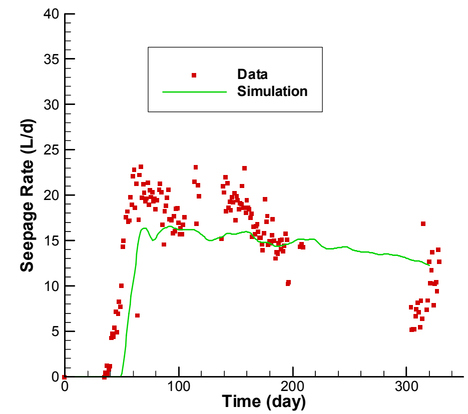


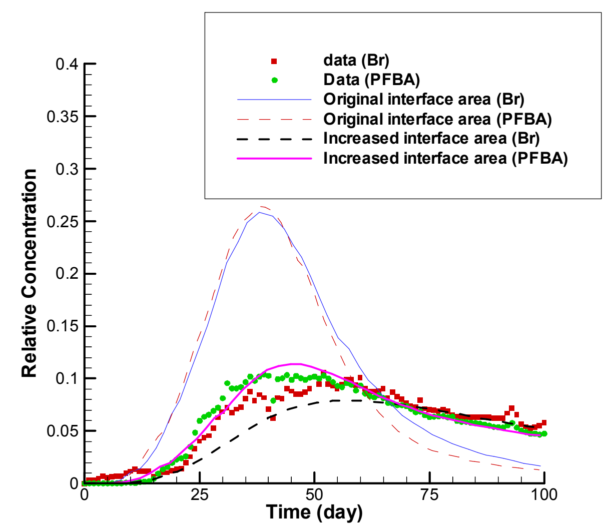




We need to increase the (lab-scale) matrix-diffusion coefficient value by about 40 times to match the observations (Liu et al. 2004).

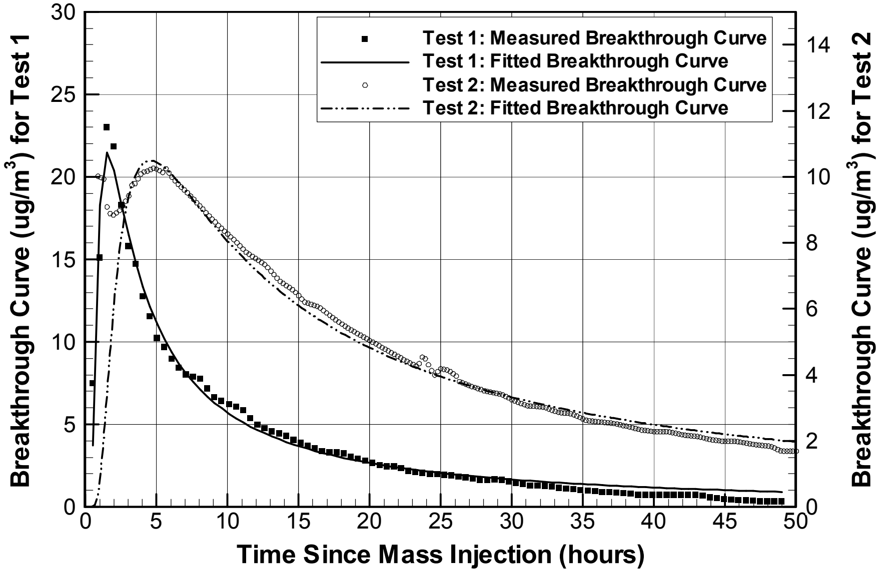
Modeling Results





Literature Survey:
Effective Matrix Diffusion Coefficient

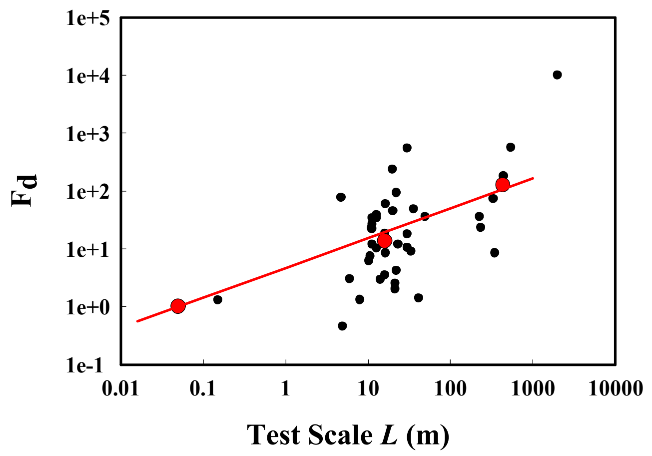
- **Reviewed 41 field tracer tests in fractured rock from 16 sites (Zhou et al. 2007)**
- **Determined field-scale effective matrix diffusion coefficients from**
  - published values
  - re-analysis of field tracer tests

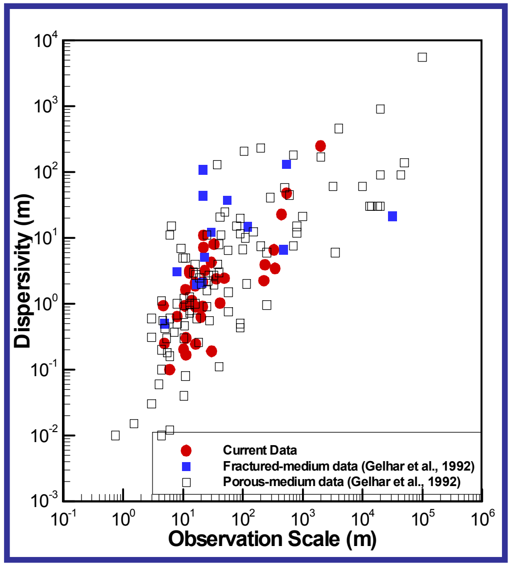


Literature Survey Results:

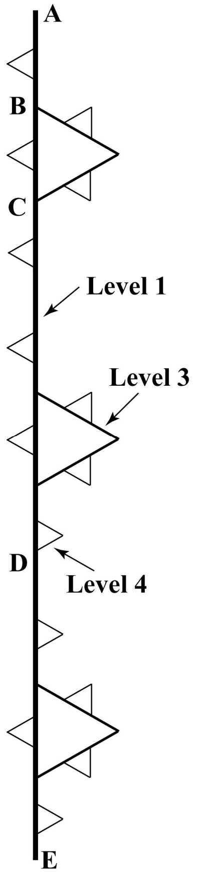
Effective Matrix Diffusion Coefficient and Dispersivity

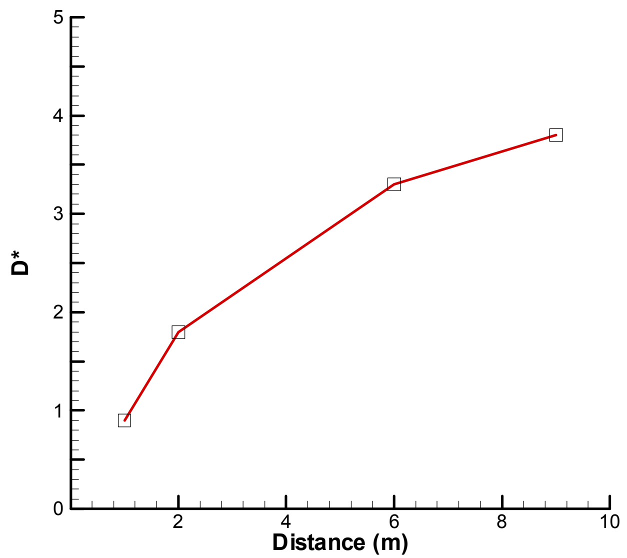
- Determined ratio of effective matrix diffusion coefficient to the lab-scale value for the same tracer.
- On average, the effective matrix diffusion is scale-dependent and increases with scale (Zhou et al. 2007).
- Estimated dispersivity is within the range of data reported in Gelhar et al. (1992).

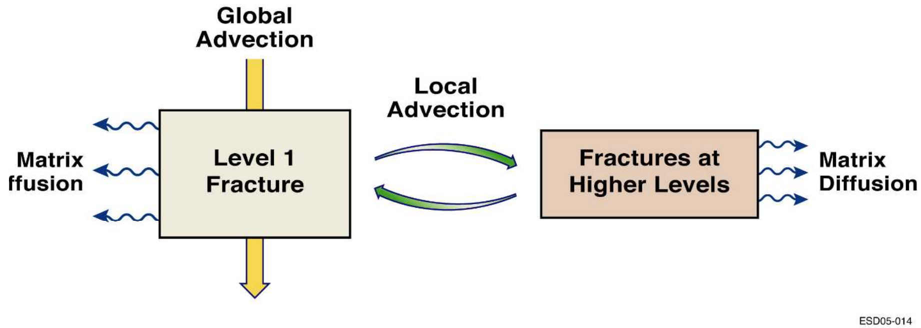




Mechanisms:
Complex Flow Path Geometry







The small-scale fractures may not considerably impact global flow, but significantly affect solute transport process and contribute to the scale dependence of the effective matrix diffusion coefficient (Liu et al. 2007a).

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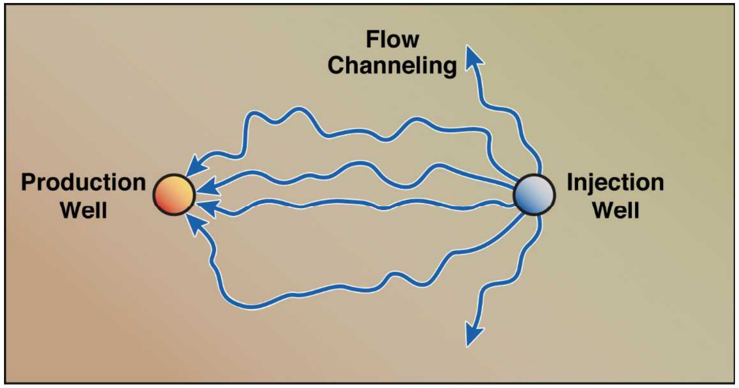
Mechanisms:
Subsurface Heterogeneity

- **An Illustrative Case (Liu et al., 2007b)**
  - The flow field is characterized by multiple flow channels, and each channel is represented by a single fracture system.
  - Each channel has homogeneous property distributions and does not mix with any other channels except at influent and effluent points.
  - Each channel has its own value for parameter *a*

$$a = \frac{\phi_m \sqrt{D_m}}{b}$$

$$f(a) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{\left(a - \bar{a}\right)^2}{2\sigma^2} \right]$$



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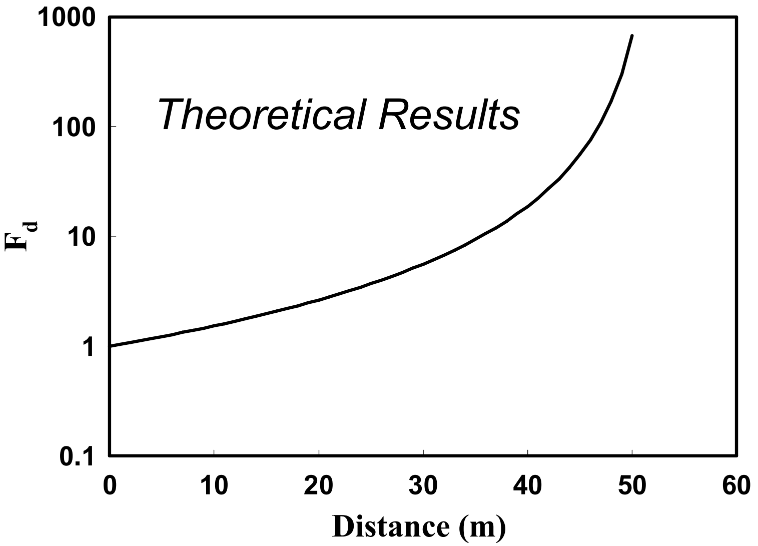

Some Thoughts

- **A "better" model must be based on a good understanding of field observations.** *Analyses of field tests are important for model development.*

- **Flow and transport processes share the same flow paths, but are sensitive to different properties of these paths.** *An integrated approach is required for developing a good modeling strategy for a given repository site.*

- **Almost all the rock properties (related to subsurface flow and transport) are scale dependent.** *We should be careful with transferring knowledge or data obtained on the laboratory scale to site-scale.*

Coupled THMC Processes in Clay Repositories

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- **Clay is one of the rock types under consideration for geological repository worldwide.**
- **Key features (characterized by the coupled processes):**
  - Induced fractures that are very dynamic (EDZ)
  - Swelling/shrinkage (depending on moisture, chemical composition and temperature)
  - Multiphase flow
  - Self sealing
- **Modeling coupled THMC processes is important for performance assessment (PA).**
- **Modeling study for natural system is closely related to other UFD activities (FEP evaluation and EBS)**

Mont Terri Site, Switzerland (Blumling et al., 2007)

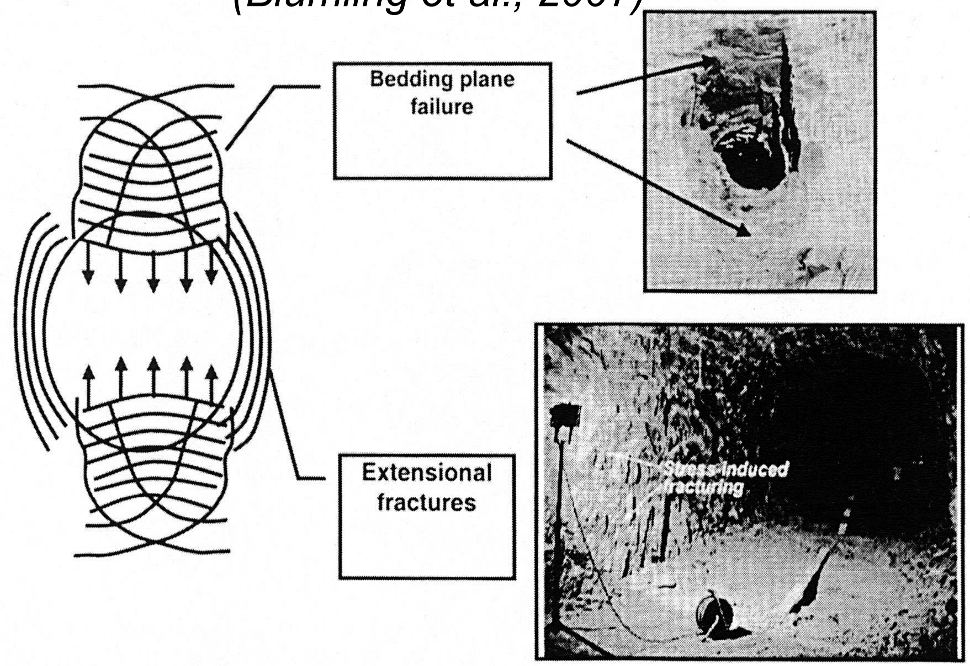


Fig. 1: Development of extensional fractures and bedding plane fractures in the case of horizontal bedding planes. (adopted from [1])

Modeling THM Processes: TOUGH2-FLAC3D

- "Fully" and "partially" coupled modeling approaches for hydro-mechanical processes.

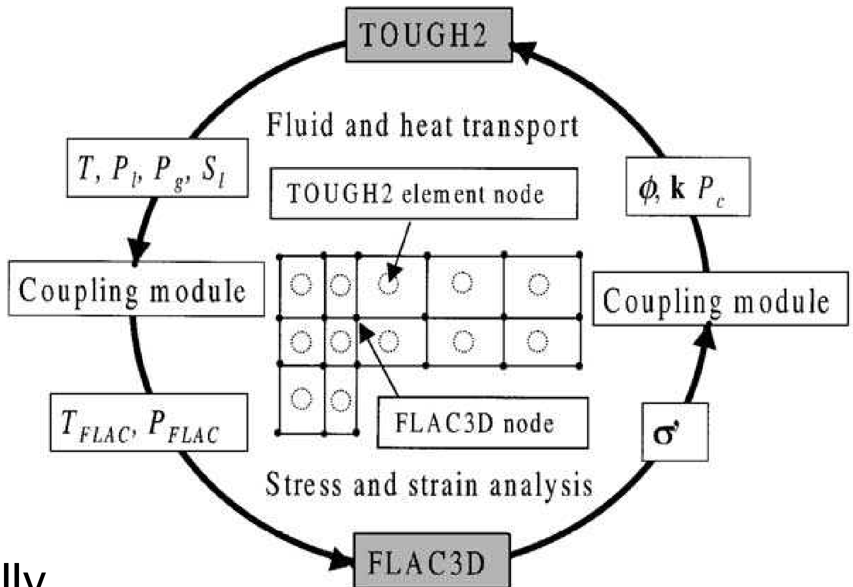
- Governing equations and/or constitutive relationships for flow need to be modified

$$\frac{\partial\left(M^{k}_{j,\psi}\right)}{\partial t}+\nabla\bullet \boldsymbol{q}^{k}_{j,r\psi}+\frac{M^{k}_{j,\psi}}{\phi_j}\left(\Phi_j\alpha_j\frac{\partial\varepsilon_{v,j}}{\partial t}-\frac{\partial\phi_j}{\partial t}\right)-\boldsymbol{Q}^{k}_{j,\psi}=0$$

Additional storage term

- TOUGH2-FLAC3D has been successfully used in different applications including geothermal energy development and CO2 geological sequestrations (Rutqvist et al., 2003).

TH Processes



Mechanical Process

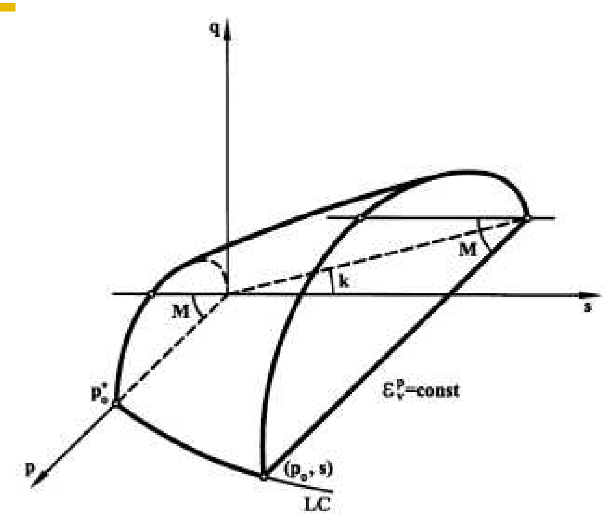
Modeling THM Processes: The Barcelona Basic Model

•Recently, the Barcelona Basic Model (BBM) (an advanced constitutive elasto-plastic model for unsaturated soils) has been implemented in the TOUGH-FLAC simulator for rigorous analysis of clay mechanical behavior.

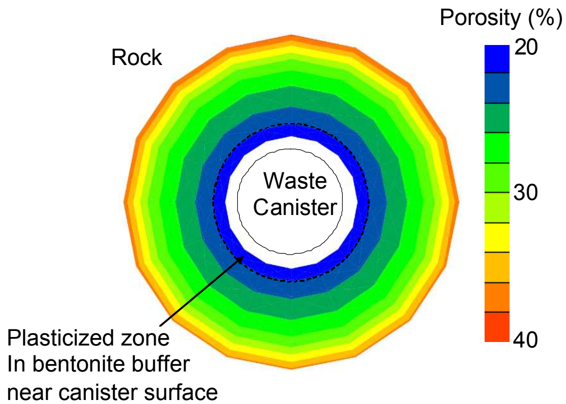
•The BBM can describe a large number of typical features of the mechanical behavior of unsaturated soils, including wetting-induced swelling.

•The BBM is one of the most advanced and accepted models currently adopted by European nuclear waste organizations for modeling of clay engineered and clay host rock systems.

•The implementation of BBM in TOUGH-FLAC has been verified and tested by modeling a number of laboratory scale experiments and on a DECOVALEX bench mark test of a bentonite back-filled repository.



Three-dimensional representation of the yield surface in the BBM (Gens et al., 2006).



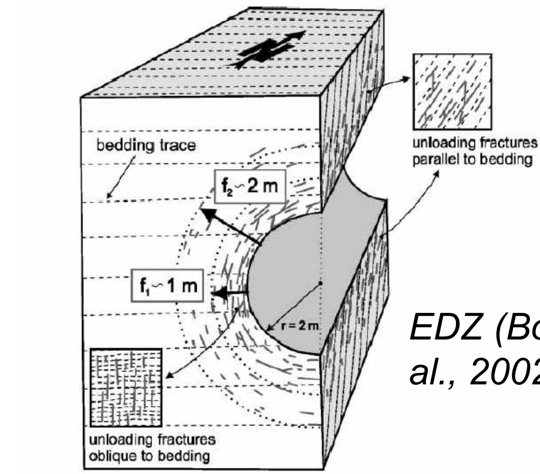
Calculated porosity distribution in the buffer at the 100,000 years showing a permanent pore collapse near the canister induced by drying shrinkage and shear failure.

Modeling THM Processes: The Barcelona Basic Model (Cont.)

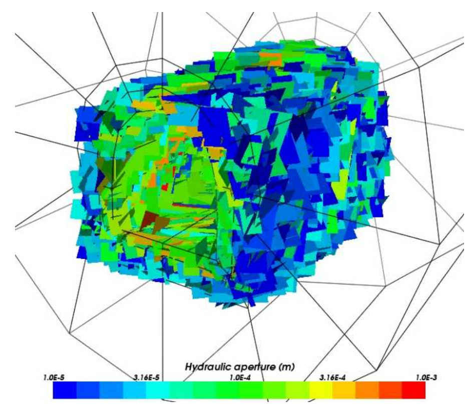
- **Further test and document the BBM model in TOUGH-FLAC (FY 10).**
- **Conduct new simulation studies of a bentonite back-filled repository in clay host rock in Generic Disposal System Environment (GDSE): We will test existing model capabilities and identify geomechanical performance and issues associated with the interaction between buffer and the clay host rock (FY 10).**
- **Extension of the current BBM model to so called double-structure behavior for modeling of expansive (swelling) clays. In this approach the material consist of two structural levels (macropore and micropore) : The double structure approach is especially useful when trying to incorporate the effects of chemical variables on the mechanical behavior of expansive clays (FY 10 and beyond).**

Modeling THM Processes: A dual-continuum model for EDZ

- **Excavation damaged zone (EDZ) may have a long-term impact on radionuclide transport.**
- **A dual-continuum model will be developed to consider the coupling between hydraulic and mechanical processes occurring within fractured EDZ.**
- **The focus will be on elastic deformation in FY 10 and more general behavior in the future.**
- **The model development will be based on both data analyses and mathematical derivation and incorporated into TOUGH2-FLAC3D simulator.**



EDZ (Bossart et al., 2002)



A fracture network model (Lanyon, 2007)

Modeling THC Processes: TOUGHREACT

Processes:

- Multiphase fluid and heat flow: TOUGH2 V2
- Transport: advection and diffusion in both liquid and gas phases
- Chemical reactions:
  - **Aqueous complexation**
  - **Acid-base**
  - **Redox**
  - **Mineral dissol./precip. (equilibrium and/or kinetics)**
  - **Gas dissol./exsol.**
  - **Cation exchange**
  - **Surface complexation**
  - **Linear Kd adsorption**
  - **Decay**

Special Features:

- **Changes in porosity and permeability, and unsaturated zone properties due to mineral diss./ppt. and clay swelling**
- **Gas phase and gaseous species are active in flow, transport, and reaction**
- **General: Porous and fractured media; 5 ϕ-k models; rate laws; any number of chemical species**
- **Wide range of conditions: P, T, pH, Eh, Salinity**

Modeling THC Processes: Simulations

- **Long-Term Objective: Fully coupled THMC model and simulation of THMC processes in clay repositories**

- **FY10: Evaluate potential changes in transport properties in Generic Disposal System Environment:**
  - Heat generated by waste package
  - Mass transfer between clay rock and bentonite (diffusion, rewetting) -- evaluation of multicomponent diffusion
  - Mineral-water dissolution/precipitation reactions in clay-rich host rock
  - Thermal expansion/contraction
  - Clay swelling/shrinkage owing to ionic strength changes (simple model later replaced by more comprehensive model developed for EBS buffer materials)

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LBNL Team Members for UFD Activities

- **Received funding at the beginning of 2010.**
- **Team Members**

H.H. Liu , Jens Birkholzer, Carl Steefel, Jonny Rutqvist, Chin-Fu Tsang, Eric Sonnenthal, Nic Spycher, Stefan Finsterle, and Jim Houseworth

- **Team expertise:**

Flow and transport in fractured porous media

Coupled Processes (THM; THC; THMC)

Reactive transport

Model development (Theoretical, mathematical, and numerical)

Uncertainty analysis

International collaborations (e.g., DECOVELEX)

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Fuel Cycle Research and Development

Identification and evaluation of key natural system attributes related to far field hydrology and radionuclide transport

&

Generic modeling for granite environment

Shaoping Chu

Los Alamos National Laboratory

Used Fuel Disposition Campaign Working Group

January 29, 2010

Introduction

- **Scope: Identify and evaluate the key natural system attributes related to far field hydrology and radionuclide transport.**

- **Formulation of conceptual models and parameter distributions for models**
  - Focus on granite systems, but evaluating other rock types in terms of far-field environment
  - Pros and cons of different geologic environments

- **Sources of conceptual models and parameters distributions**
  - U.S. and international programs (granite, salt, clay)
  - Enhanced geothermal systems
  - General scientific literature

- **Contributors: Shaoping Chu, Carl Gable, Schon Levy, Giday Woldegabriel, Frank Perry, Paul Reimus and Mei Ding**

Geologic Media and Environment

- **Media**
  - Crystalline Rock (e.g., granite, some metamorphic)
  - Volcanic (Tuff)
  - Volcanic (Basalt)
  - Salt/Evaporite
  - Clay
  - Sedimentary (e.g., Sandstone, Siltstone, Shale)
    Note: Limestone not considered

- **Environment**
  - Saturated vs. Unsaturated
  - Oxidizing vs. Reducing Geochemistry

Natural Systems Assessment: Attributes of Geologic Media and Environment

- Rock properties: physical, thermal-mechanical, hydrologic, and geochemical
- Structures: layering, zonation, and discontinuities
- Tectonic setting, stress field, seismicity, fracture system, deformation history
- Thermal regime
- Regional and local hydrology
- Shape, size, and boundaries of crystalline bodies
- Overburden and location relative to land surface
- Potential for exploitable economic resources

Natural Systems Assessment – Geologic Media pros and cons (transport/ geochemistry perspective)

Media	Pros	Cons
Crystalline Rock	■May be located in old, stable tectonic settings ■Geochemistry is often reducing, lowering radionuclide solubilities increasing sorption ■Low heterogeneity in geochemical/mineralogic properties ■Amenable to retrievability	■Hardness of rock and low matrix porosity and permeability leads to fracture flow, more rapid and less predictable than flow in other environments ■Fracture flow in low porosity environment results in less matrix diffusion and less surface area available for sorption ■Less sorptive mineralogies ■More reliance on engineered barriers
Volcanic (Tuff)	■Generally considerable mineralogic alteration, helps far-field sorption ■Generally higher matrix porosity than crystalline rocks or basalt ■Can be sited for reducing geochemistry ■Amenable to retrievability	■May be dominated by fracture flow ■More reliance on engineered barriers ■Tend to be located closer to recent volcanic/ tectonic activity

Natural Systems Assessment – Geologic Media pros and cons (transport/ geochemistry perspective)

Media	Pros	Cons
Volcanic (Basalt)	■Geochemistry is often reducing, lowering radionuclide solubilities increasing sorption ■Amenable to retrievability	■Dominated by fracture flow ■Generally less altered and lower matrix porosity than tuffs and thus have reduced diffusion and sorption capacity ■More reliance on engineered barriers ■Tend to be located closer to recent volcanic/ tectonic activity
Salt/Evaporite	■Old stable tectonic settings, limited water availability ■less reliance on engineered barriers because of lack of radionuclide transporting agent (water) ■Reducing geochemistry if water is available ■Relative homogeneity in salt deposit simplifies modeling	■Not amenable to retrievability (salt creep), particularly under heat load. ■Heat may cause available water to migrate to waste packages ■High corrosion rates if water is available ■More complex solution chemistry and sorption because of high ionic strengths ■Greater risk of human intrusion

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Natural Systems Assessment – Geologic Media pros and cons (transport/ geochemistry perspective)

Media	Pros	Cons
Clay	■Generally stable environments less vulnerable to seismic hazards (plastic deformation vs. rock breakage) ■Inherently low permeability and high sorptive capacity and typically reducing environment ■Easier to predict groundwater flow and radionuclide transport than in fractured rocks ■Less reliance on engineered barriers	■Effects of long-term heat more complex and have greater uncertainty than with other rock types ■Tend to be shallower and closer to aquifers than other rock types. More complex release scenarios and greater susceptibility to human intrusion and climate change ■Ground motions probably greater ■Less amenable to retrievability
Sedimentary (e.g., Sandstone, Siltstone, Shale) Note: Limestone not considered	■May be located in stable tectonic settings ■Geochemistry often reducing, lowering radionuclide solubilities increasing sorption ■Less fractured and higher matrix porosity than crystalline or volcanic rocks, enhancing flow porosity and matrix diffusion. Also tend to be more homogeneous mineralogically and hydrologically ■Possibly best rock type for natural analogs	■Tend to have less sorptive mineralogies for radionuclide retardation than many other rock types ■Sometimes have localized oxidizing and reducing zones (e.g., roll front deposits) which could tend to concentrate redox-sensitive radionuclides and make predictions more difficult ■Often associated with oil and gas or mineral deposits, so potentially greater risk of human intrusion

Natural Systems Assessment – Geologic Environment pros and cons (transport/ geochemistry perspective)

Environment	Pros	Cons
Saturated	■ Typically reducing and inherently easier to model and predict than unsaturated systems ■ Tend to have less transient behavior more steady flow and geochemical conditions ■ Less susceptible to climate change or other influences	■ If engineered barriers fail, groundwater travel times in saturated systems can be rapid
Unsaturated	■ Engineered barriers should last longer if water is not continuously present, as water is the main driver for waste package and EBS failure ■ EBS failure does not necessarily translate to rapid radionuclide release and transport in unsaturated systems, as abundant water is necessary for release and transport (with the exception of volatile radionuclides such as ^{14}C)	■ Oxidizing, higher radionuclide solubilities and less sorption for redox-sensitive radionuclides ■ Flow and transport in unsaturated systems, particularly under the influence of heating, is inherently more complex and difficult to predict than in saturated systems

Natural Systems Assessment – Geologic Environment pros and cons (transport/ geochemistry perspective)

Environment	Pros	Cons
Oxidizing vs. Reducing Geochemistry	■ Reducing conditions are desirable from the standpoint that they generally result in lower radionuclide solubilities and higher radionuclide sorption coefficients than oxidizing conditions ■ Waste forms and waste package materials will be more stable under reducing conditions than oxidizing conditions ■ Microbial processes are less likely to cause problems under reducing conditions (although microbes can also drive an oxidizing system reducing)	■ Disadvantage of reducing systems is that it may be necessary to consider the implications of a potential future transition to oxidizing conditions, if that can't be screened out

Model Development– far-field for granite environment

- 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

Radionuclides simulated for granite environment

32 Radionuclides simulated:

Actinides & Daughters:

Am (241,243)
Pu (238,239,240,242)
Np237
U (232,233,234,235,236,238)
Th (229,230,232)
Pa231
Ac227
Ra (226,228)
Pb210

Fission Products:

Tc99, Sn126, Cs135, I129, Se79, Sr90
Sb126, Zr93, Nb93, Pd107

Others:

C14

Transport parameters necessary for modeling granite environment

Geometric parameters:

- Aperture
- Fracture spacing
- Geologic framework (structure and stratigraphy)

Transport parameters:

- Matrix diffusion
- Matrix sorption
- Fracture sorption
- Thermal properties

Other parameters:

- Solubility
- Flow rate
- Porosity
- Permeability
- Rock density
- Anisotropy
- Specific Discharge
- Matrix compressibility
- Dispersivity
- etc.

Enhanced Geothermal Systems Projects as Analogues for Natural-Systems Information on Crystalline Rocks

- **Increase our conceptual 3D understanding of crystalline rock in the subsurface**
- **Help define the ranges of parameter values important for modeling**
- **Identify key technologies/techniques for site selection and characterization**

Analogue Sites:

- **Soultz-sous-forêts (Paleozoic granite), France/Germany**
- **Fenton Hill (Precambrian granodiorite), New Mexico, USA**
- **Rosemanowes (Carnmenellis granite), Great Britain**
- **Ogachi (granodiorite in volcanic setting), Japan**
- **Cooper Basin, Australia**
- **Fjällbacka (Bohus granite, shield environment), Sweden**

These projects are information sources for crystalline-rock properties of interest to the used-fuel disposition campaign. They provide some of the most detailed information about granites/crystalline rocks in a variety of geologic settings.

Enhanced Geothermal Systems Projects as Analogues for Natural-Systems Information on Crystalline Rocks

EGS projects target depths of 3 km or more, which is deeper than would be practical for a mined disposal site. However, information is collected from shallower depths. Information of interest includes:

- In-situ and laboratory thermal/mechanical properties
- Variations in local stress regimes/seismic activity
- Characteristics of fracture systems, including vein mineralization
- Effects of thermal cycling
- Hydrologic properties/systems
- Water chemistry

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Nuclear Energy

Fuel Cycle Research and Development

Near-Field Model and Analysis for Salt GDSE

Joon Lee, Yifeng Wang, and Carlos Jove-Colon

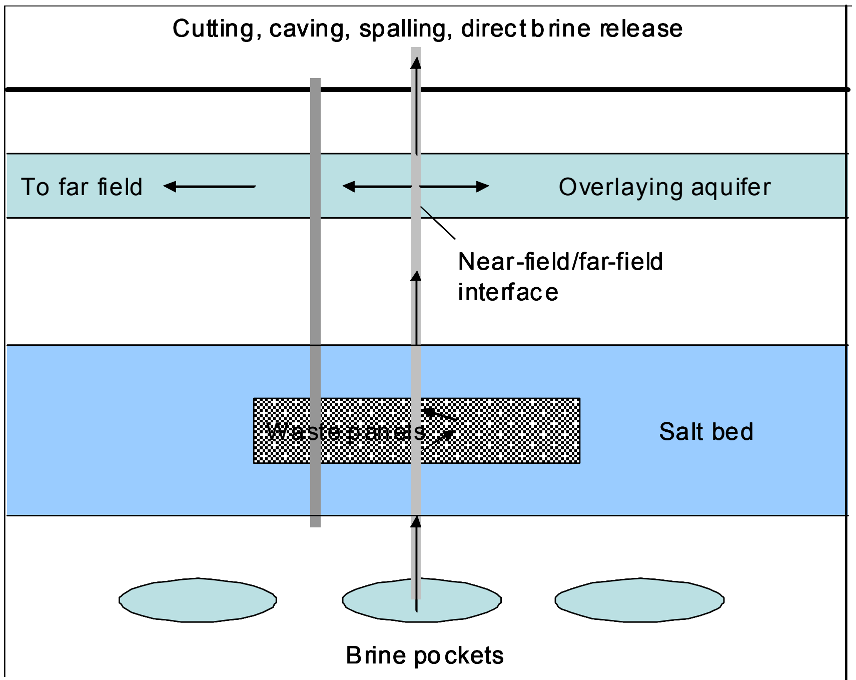
Sandia National Laboratories

UFD Campaign Working Group Meeting

Albuquerque, NM

January 29, 2010

Conceptual Description of the Salt GDSE Source-Term / Near-Field Model



- **Conceptual model for repository layout and waste disposal developed based on the WIPP**
- **Divide the repository into 10 isolated waste disposal rooms (or panels)**
- **Human intrusion is the dominant scenario leading to RN release**
- **WF is the only EBS component**
- **Assume RN release through the borehole and into the overlying aquifer is the major RN release pathway**
- **Include 32 RNs, accounting for decay daughter in-growth and isotopic mix among RNs**
- **Consider two different brine water chemistries and key RN elemental solubility in the waters**
  - Repository brine (more reducing and concentrated)
  - Near-field interface brine
- **Assume ambient temperature (no decay heat effect)**

Simulation Approach

		Initial Inventory Mass Factor				
		0.001	0.01	0.1	1	10
Waste Form Degradation Rate (yr^{-1})	1.E-09	Group 1	Group 2	Group 3	Group 4	Group 5
	1.E-08	Group 6	Group 7	Group 8	Group 9	Group 10
	1.E-07	Group 11	Group 12	Group 13	Group 14	Group 15
	1.E-06	Group 16	Group 17	Group 18	Group 19	Group 20
	1.E-05	Group 21	Group 22	Group 23	Group 24	Group 25
	1.E-04	Group 26	Group 27	Group 28	Group 29	Group 30
	1.E-03	Group 31	Group 32	Group 33	Group 34	Group 35
	1.E-02	Group 36	Group 37	Group 38	Group 39	Group 40

- **Reference initial inventory based on commercial LWR spent fuel with a burn-up of 50 GWd/MTHM and 20 years after reactor discharge**
- **Consider a wide range of WF fractional degradation rates and initial inventories**
- **100 realizations for each combination of degradation rate and inventory**
- **Evaluate fission products and actinide elements independently to limit solubility "competition"**
  - Uranium + daughters
  - Plutonium + daughters
  - Americium + daughters
  - Neptunium + daughters and fission products

Simulation Approach (cont'd)

- **LHS of uncertain parameters**
  - Number of bore holes through repository
  - Brine flow rate through a bore hole
  - Porosity of disposal room and overlying aquifer
  - RN elemental solubility (U, Pu, Np, Am, Th, Tc (repository brine only), and Sn)
- **Use the mean of peak doses at the near-field interface as the measure for the near-field performance**
  - Intermediate system performance analysis purpose only
  - No real performance implications
- **IAEA BIOMASS Example Reference Biosphere 1 (ERB 1) dose conversion factor applied at the near-field interface**

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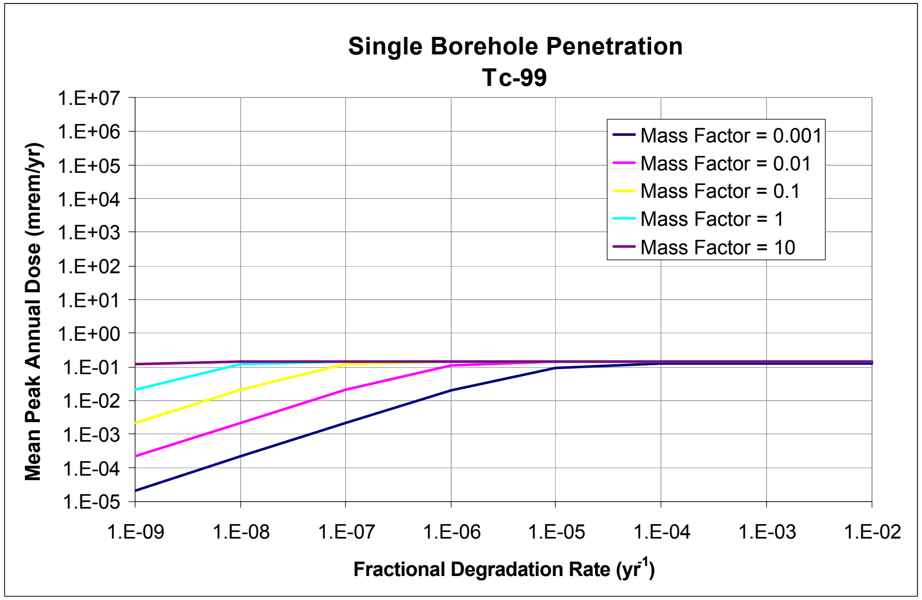
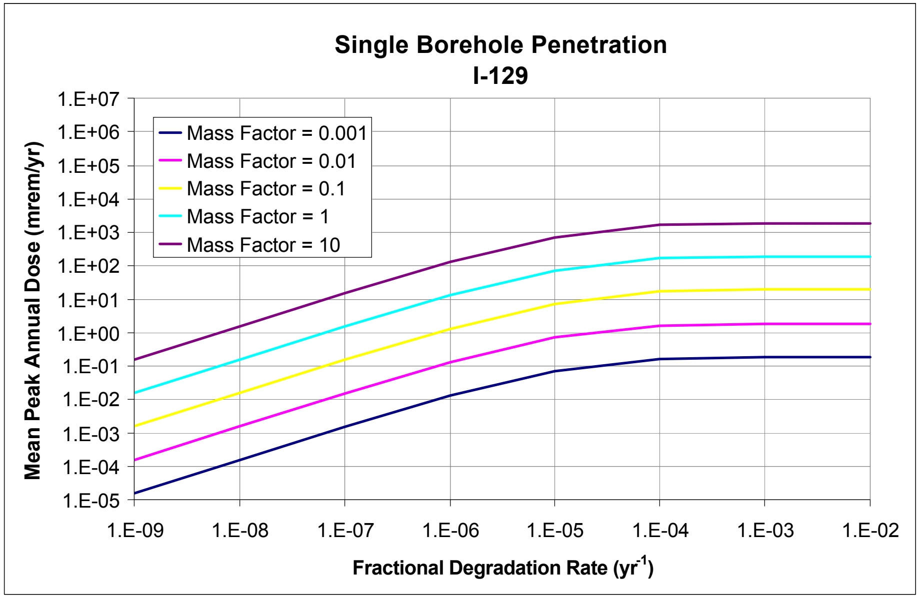
Example Simulation Results





- Peak dose of solubility-limited radionuclides is likely to be independent of waste form degradation rate
- Peak dose of no solubility-limited and non-sorbing radionuclides is proportional to the host waste form degradation rates and their inventories

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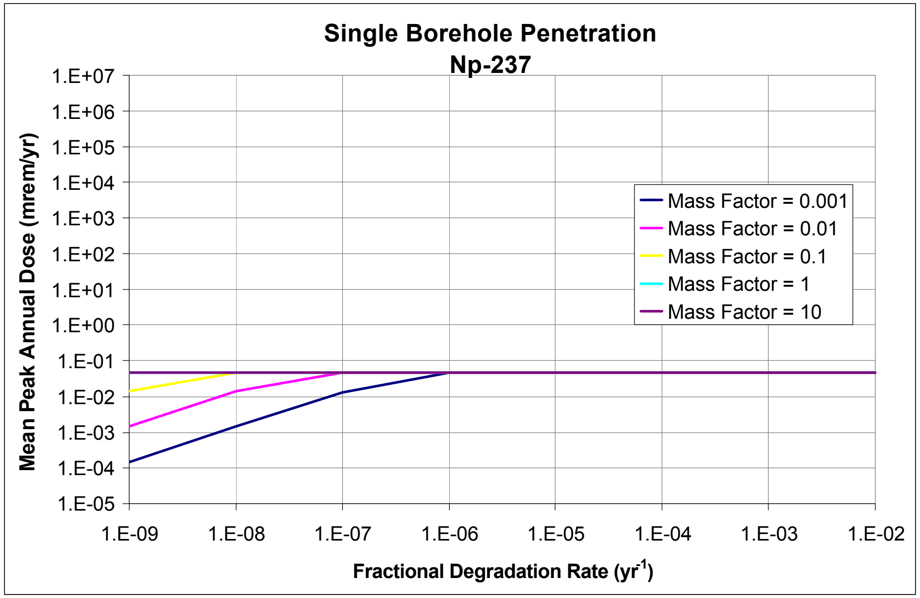
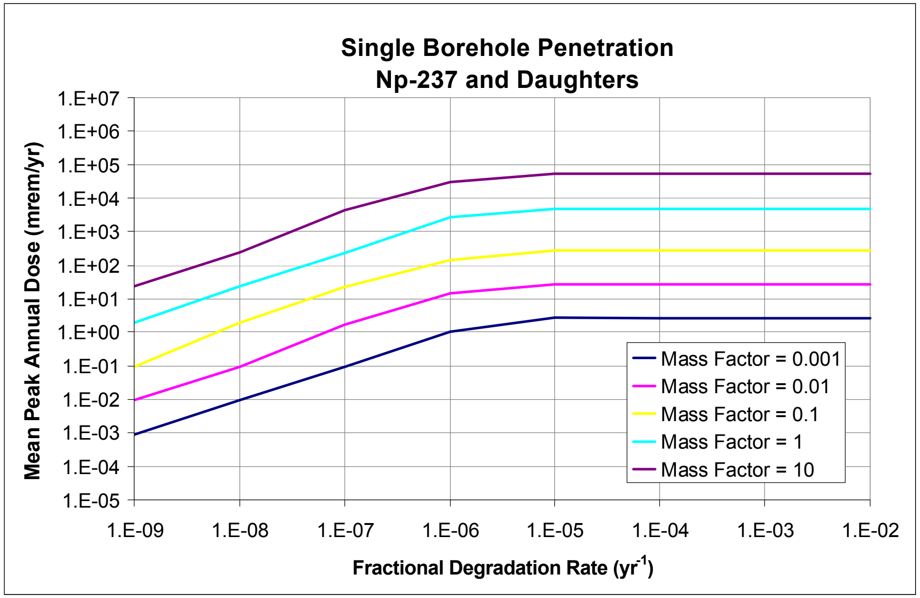
Example Simulation Results





- **Dose contribution of actinide decay daughters is important**
- **Become more significant for the combinations of high inventories and high waste form degradation rates**

Summary and Future Work

- **Peak dose of solubility-limited radionuclides such as Tc-99 is likely to be independent of waste form degradation rate**
- **Peak dose of no solubility-limited and non-sorbing radionuclides such as I-129 is proportional to the host waste form degradation rates and their inventories**
- **For actinides, the contribution of their decay daughters is important**
  - Become more significant for the combinations of high inventories and high waste form degradation rates
- **Consider alternative options for salt GDSE**
  - Repository and EBS design
  - Waste release pathway
- **Consider decay heat effects on source-term and near-field processes**
  - Waste form degradation
  - Radionuclide solubility
  - Water and RN movement
- **Consider waste form characteristics and waste inventories specific to reprocessing waste streams**

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Nuclear Energy

Fuel Cycle Research and Development

Direct Disposal of Electrochemical Refinery Wastes: The Concept

Yifeng Wang (SNL) & Michael Simpson (INL)

UFD Campaign Working Group Meeting

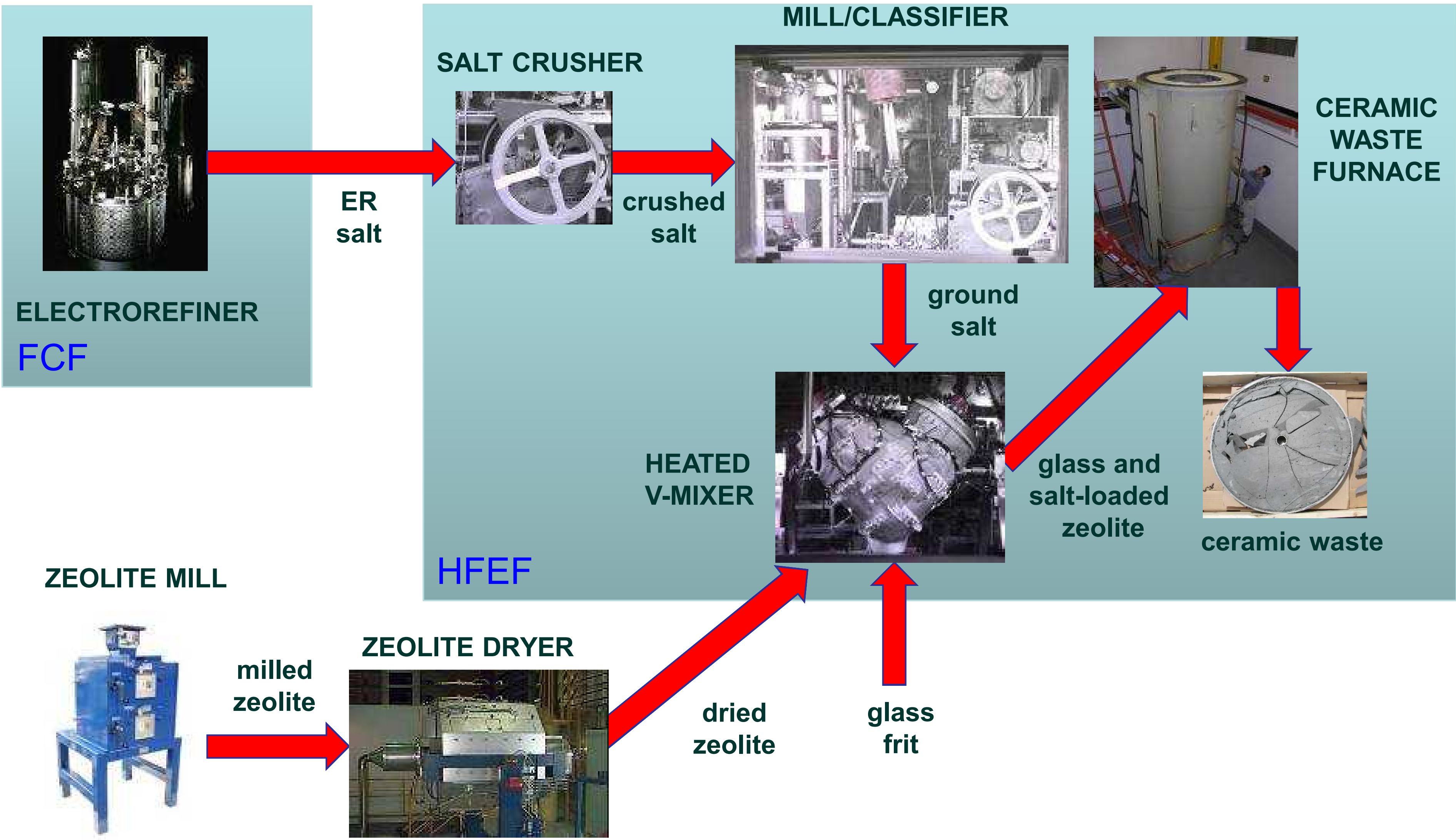
Albuquerque, NM

January 29, 2010

Problem Statements

- **Electro-refining (ER) as compared to aqueous processing**
  - Fewer steps
  - Simpler waste streams: salts
  - Less expensive
- **Existing disposition plan**
  - Separated by aqueous processes (?)
    - *Issues: Going back to "messy" aqueous processes*
  - Immobilized into ceramic, metallic or glass waste forms (WFs)
    - *Issues: Low Cl loadings, large waste form volume, durable but not stable WFs*

Ceramic Waste Process



Waste Salt from Electrorefiners

- **Salt characteristics**
  - Initially anhydrous
  - Solid (density ~1.8 g/cc)
  - Very hygroscopic
- **Baseline disposal option has been to convert into ceramic waste form and send to Yucca Mountain.**
- **Problems with this option:**
  - Uncertainty regarding Yucca Mountain
  - Excessive waste quantities and disposal cost
  - Scarcity of space for processing in hot cells

Mark-IV ER (driver treatment)

Salt	Mole fraction	Mass fraction
LiCl	0.348	0.199
KCl	0.252	0.253
NaCl	0.305	0.240
RbCl	3.06E-03	5.02E-03
$SrCl_2$	6.33E-03	1.37E-02
YCl_3	3.90E-03	1.03E-02
CsCl	1.54E-02	3.54E-02
$BaCl_2$	7.03E-03	1.98E-02
$LaCl_3$	5.43E-03	1.80E-02
$CeCl_3$	1.02E-02	3.41E-02
$PrCl_3$	5.07E-03	1.69E-02
$NdCl_3$	1.71E-02	5.78E-02
$PmCl_3$	1.01E-04	3.46E-04
$SmCl_3$	3.26E-03	1.12E-02
$EuCl_3$	1.98E-04	6.93E-04
$GdCl_3$	8.47E-05	3.00E-04
$NpCl_3$	3.47E-04	1.61E-03
UCl_3	1.01E-02	4.68E-02
$PuCl_3$	7.50E-03	3.50E-02
$AmCl_3$	1.00E-05	4.69E-05

Mark-V ER (blanket treatment)

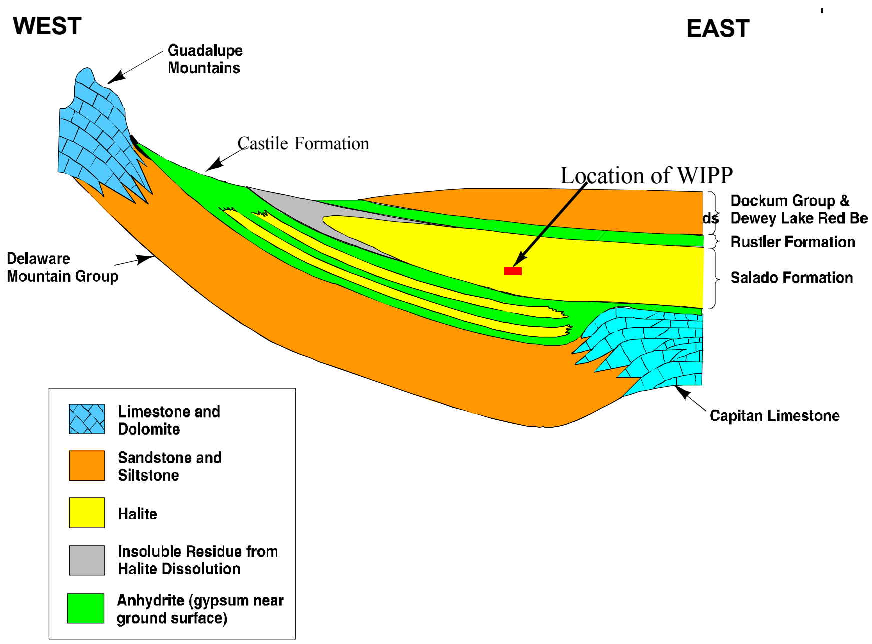
Salt	Mole fraction	Mass fraction
LiCl	0.458	0.290
KCl	0.327	0.365
NaCl	0.178	0.156
RbCl	5.12E-05	0.000
$SrCl_2$	7.50E-05	1.80E-04
YCl_3	6.16E-05	1.80E-04
CsCl	4.01E-04	1.02E-03
$BaCl_2$	2.35E-04	7.36E-04
$LaCl_3$	1.41E-04	5.17E-04
$CeCl_3$	2.59E-04	9.59E-04
$PrCl_3$	1.30E-04	4.83E-04
$NdCl_3$	4.26E-04	1.60E-03
$PmCl_3$	1.37E-06	5.21E-06
$SmCl_3$	1.14E-04	4.33E-04
$EuCl_3$	1.37E-05	5.30E-05
$GdCl_3$	9.98E-06	3.92E-05
$NpCl_3$	2.37E-05	1.22E-04
UCl_3	1.09E-02	5.60E-02
$PuCl_3$	2.43E-02	0.126
$AmCl_3$	9.15E-06	4.76E-05

Taking Advantage of Salt Repository: Direct disposal of ER Salt Waste

- Directly emplace ER salt waste in a salt repository with zero or minimal treatment.
- ER salt waste itself constitutes an ideal waste form for disposal in a salt repository, because any brine in the repository is already saturated with salt minerals in the waste.
- Such a waste form will essentially be *thermodynamically stable* in a salt repository environment.
- As long as radionuclides are encapsulated by the salt matrix, the release of these radionuclides from ER salt waste by dissolution will be minimal.
- This encapsulation will be further reinforced over time by salt creep accelerated by radioactive decay heat.

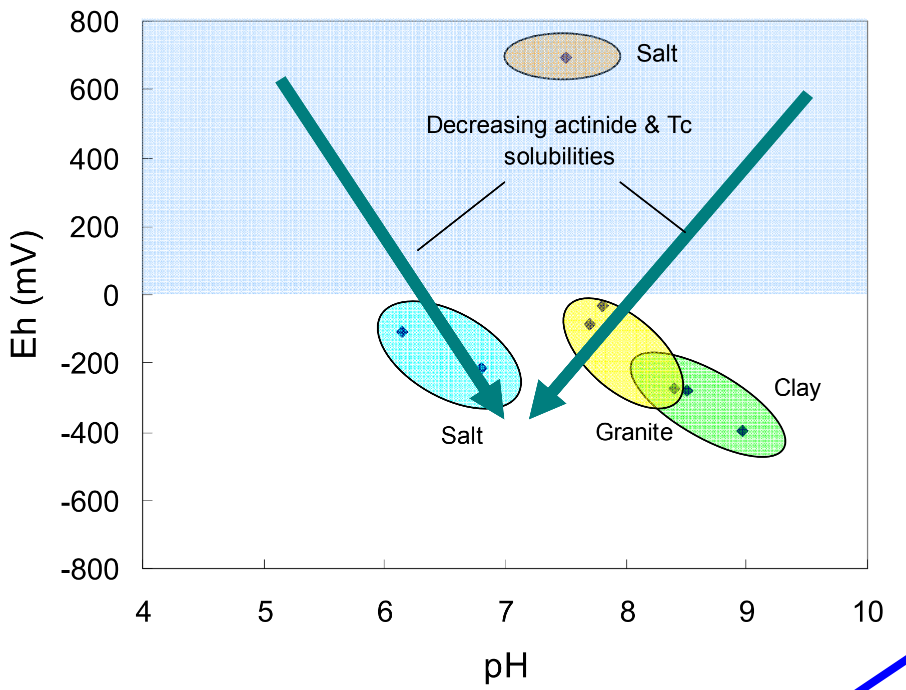
What's good about salt?

- Wide geographic distribution (many potential sites)
- Salt can be mined easily
- Salt has a relatively high thermal conductivity
- Salt is plastic *
- Salt is essentially impermeable *
- Fractures in salt are self healing *
- Salt has existed underground for millions of years *



* Attributes of Natural Barrier

Chemical Environment of Salt Repository



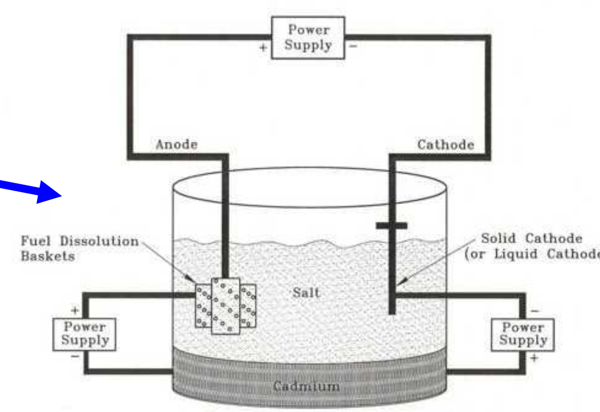
WIPP brineC	G-Seep	ERDA-6
B^{3+} (mM)	20	63
Br^- (mM)	10	11
Ca^{2+} (mM)	20	12
Cl^- (M)	5.35	4.8
K^+ (mM)	770	97
Mg^{2+} (M)	1.44	19
Na+ (M)	1.83	4.87
SO_4^{2-} (mM)	40	170
TIC (mM)	10	16
pH	6.5	6.17
Eh (mV)		-152

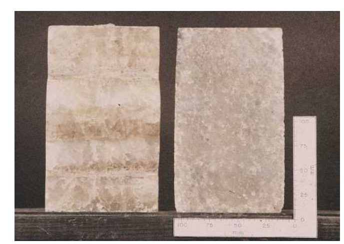
The effect of colloid-facilitated radionuclide release is minimal due to high ionic strength environments.

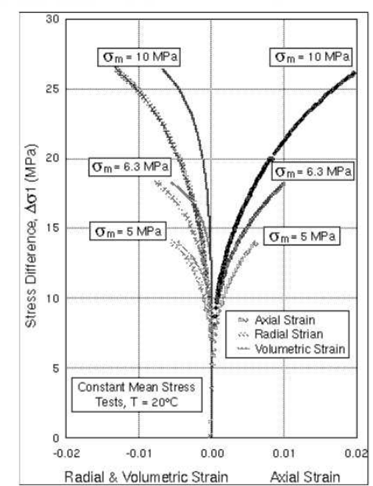
Multiple Level Radionuclide Encapsulation

- **Micro-scale encapsulation**
  - Radionuclide precipitates encapsulated inside salt matrix
  - Encapsulation done during ER process
- **Macro-scale encapsulation**
  - Encapsulation of individual waste packages
  - Disposal room closure
  - Salt creep and self-healing
  - Isolation of individual waste packages
  - Reducing radionuclide release during human intrusions

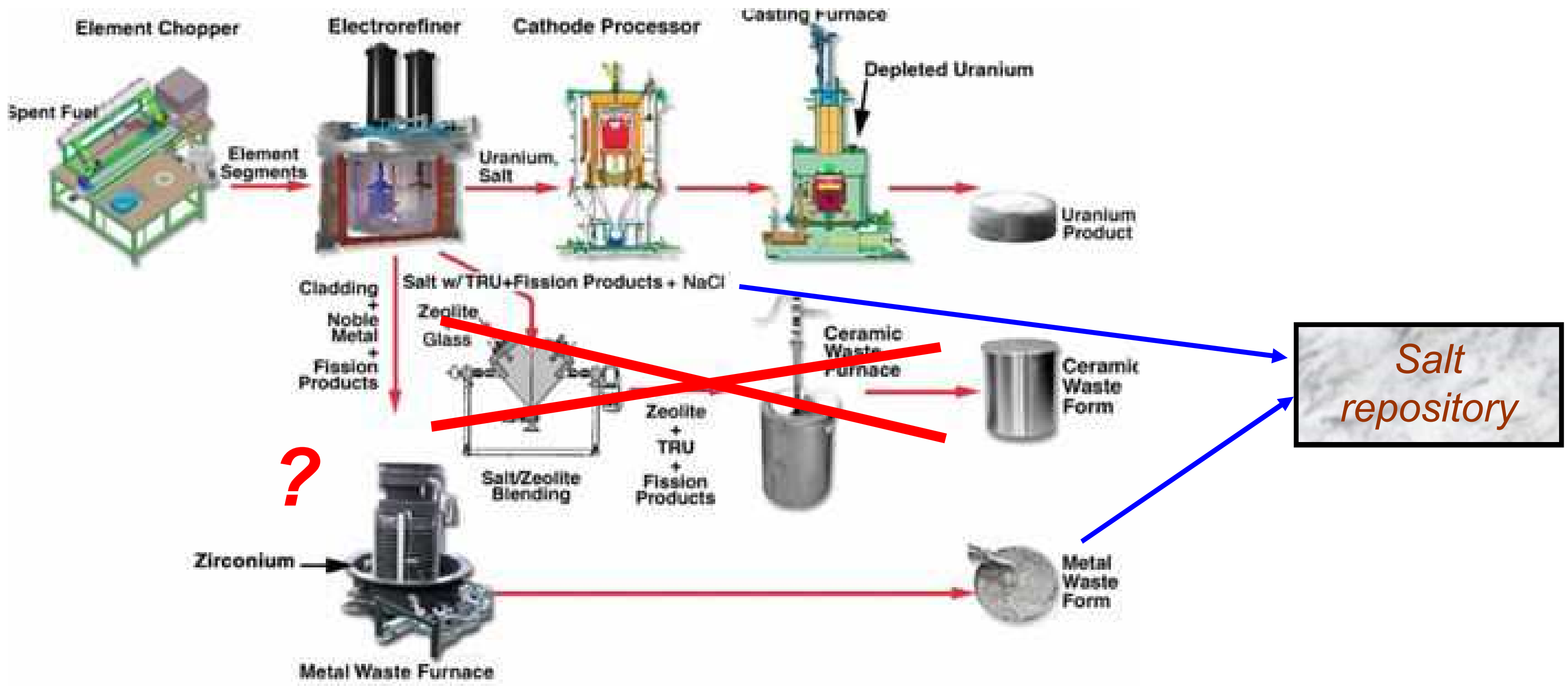
Schematic of Electrochemical Transfer Modes







Benefits of Direct Disposal Concept



- The direct disposal concept will practically eliminate all process steps currently proposed for ER salt waste treatment and immobilization.
- The volume of waste for disposal will be significantly reduced.
- No robust waste packages are needed.
- Save $$$$$.

Processing Options Currently Being Assessed for Impact on Waste Disposal

Mark-IV ER

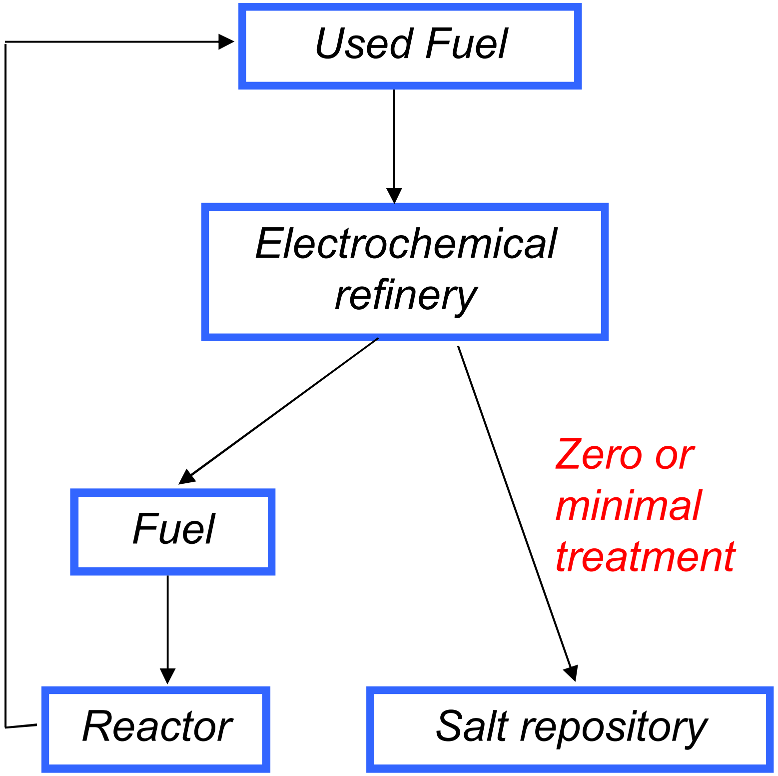
Strategy	Metal Waste (MT)	Ceramic Waste Loaded w/ Salt (MT)	Salt for Direct Disposal (MT)	Sodium Waste (kg)	U/TRU product (kg)
Throw-Away	2.73	14.3	1.0	0	0

Mark-V ER

Strategy	Metal Waste (MT)	Ceramic Waste Loaded w/ Salt (MT)	Salt for Direct Disposal (MT)	Sodium Waste (kg)	U/TRU product (kg)
Throw-Away	2.68	31.2	2.6	0	0
Na-Separation	2.45	30.8	2.56	174	0
LCC	2.68	13.0	1.08	0	545
Na-Sep.+LCC	2.45	7.18	0.60	174	626
Na-Sep.+DD	2.45	9.60	0.72	95	345

How to Move Forward?

- **Proposed activities**
  - Better understand phase transition & separation behavior of ER waste salt during the cooling process.
  - Better understand salt creep and encapsulation.
  - Test radionuclide release from salt matrix.
  - Develop long-term performance assessment tools for direct disposal of ER waste.
  - Evaluate the impacts of direct disposal on waste handling and transportation.
- **FY10**
  - Summarize the current status of ER process related to the direct disposal concept.
  - Synthesize the existing data related to salt creep and encapsulation on disposal room and waste package scales and identify the technical gaps related to the direct disposal.
  - Identify the potential impact of the direct disposal concept on waste handling and transportation.



FY10: From the Processing Side

- List processing options with impacts on disposal
- Assess effect of fission product decay heat on disposal
- Initial experiments to measure uptake of moisture from representative brine to surrogate salt waste
- Develop plan for out-year experiments
- Write narrative description of process, salt composition, options, and technical issues