U.S. DEPARTMENT OF ENERGY | Nuclear Energy

SAND2014-19251C

Experimental Program for Used Fuel Disposition in Crystalline Rocks

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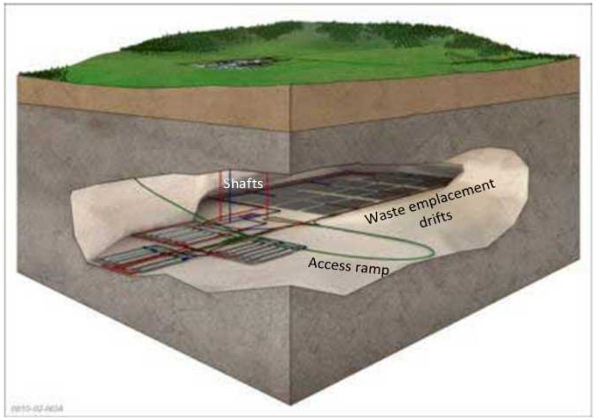
Idaho Falls, ID

Objectives of Crystalline Disposal R&D Work Packages

- **Directly support the following UFDC objectives**
  - Develop a fundamental understanding of disposal system performance in a range of environments for potential wastes that could arise from future nuclear fuel cycle alternatives through theory, simulation, testing, and experimentation.
  - Develop a computational modeling capability for the performance of storage and disposal options for a range of fuel cycle alternatives, evolving from generic models to more robust models of performance assessment.
- **Focus on two key components of deep geologic repository in crystalline rocks**
  - Better characterization and understanding of fractured media and fluid flow and transport in such media
  - Designing effective engineered barrier systems for waste isolation
- **Fully leverage international collaborations**
  - Korean Atomic Energy Research Institute
  - Äspö Hard Rock Laboratory (Sweden)
  - Bedrichov Tunnel Tests (Czech)
  - Colloid Formation & Migration Project (Switzerland)
  - Others



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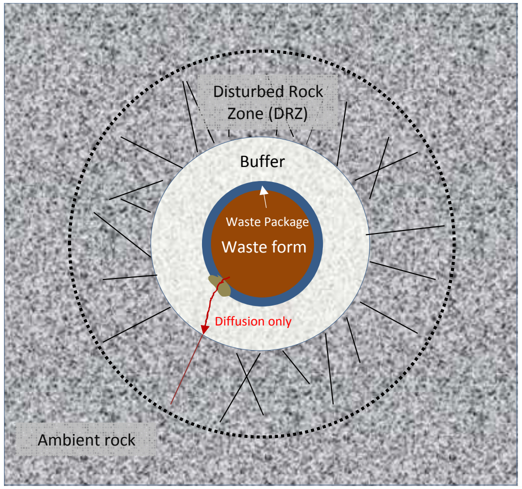
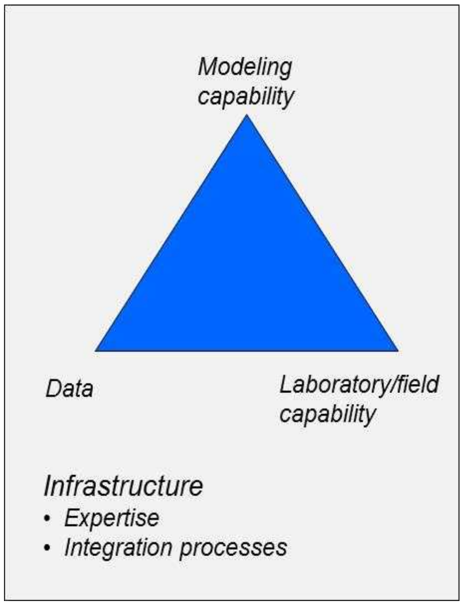
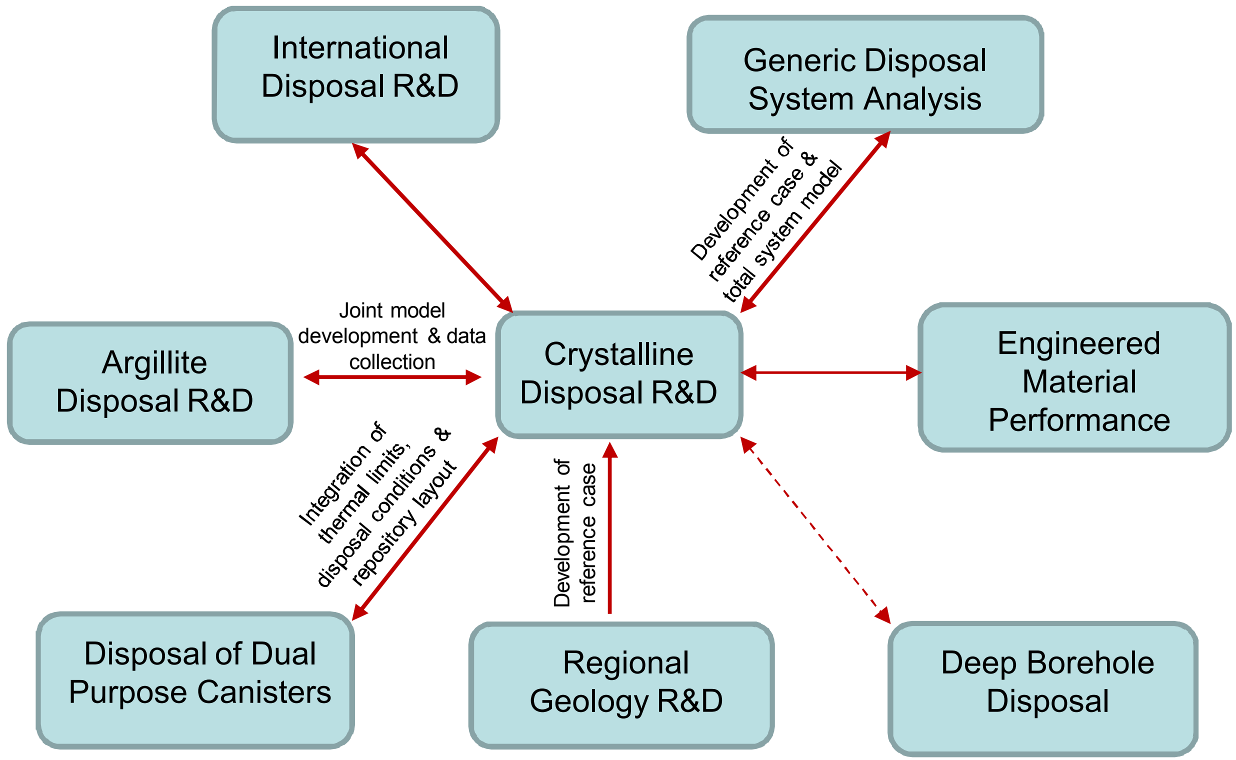


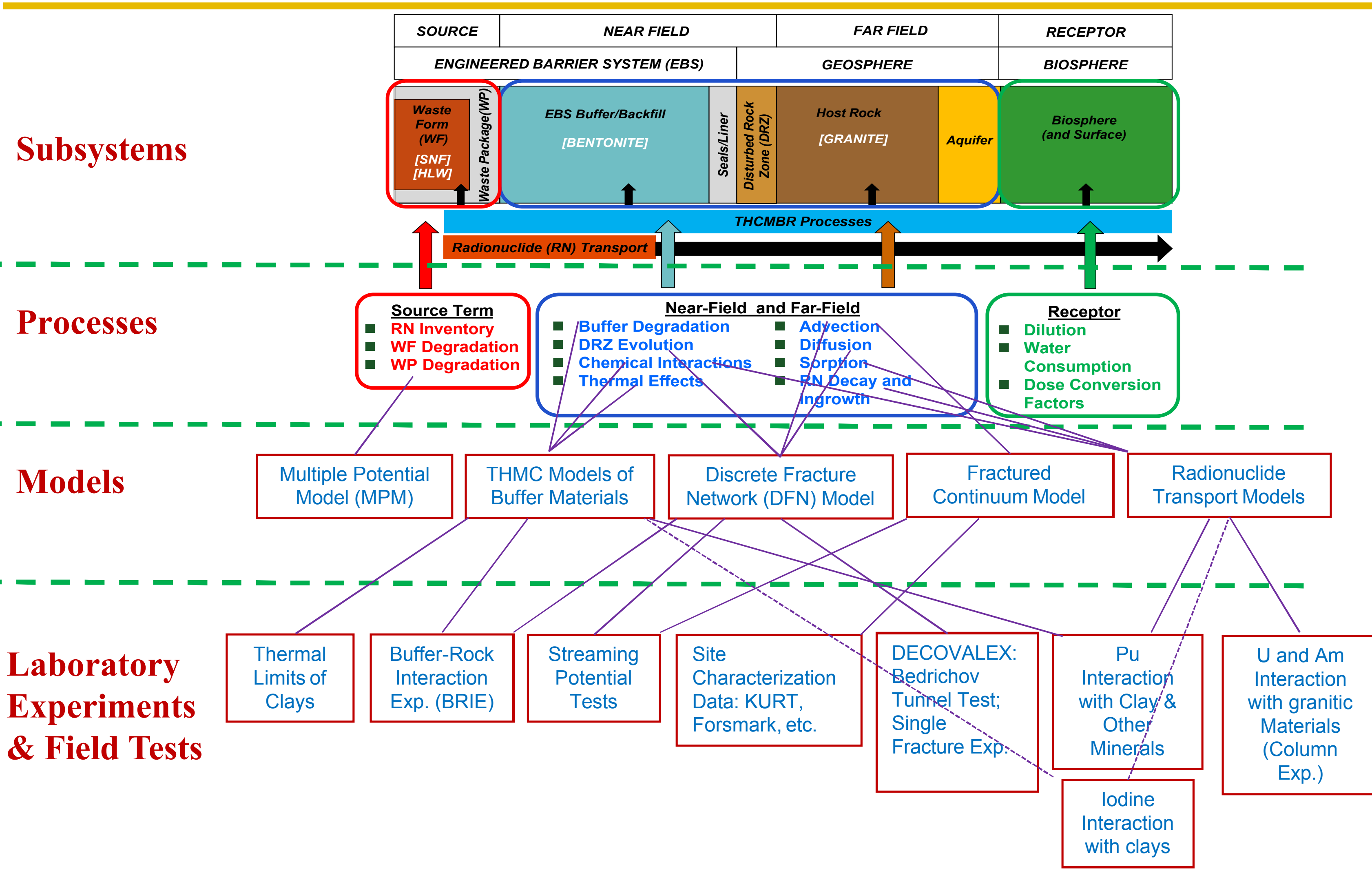
Integration & coordination





Institutions involved: ANL, LANL, LBNL, LLNL, SNL

Experimental program for used fuel disposition in crystalline rocks



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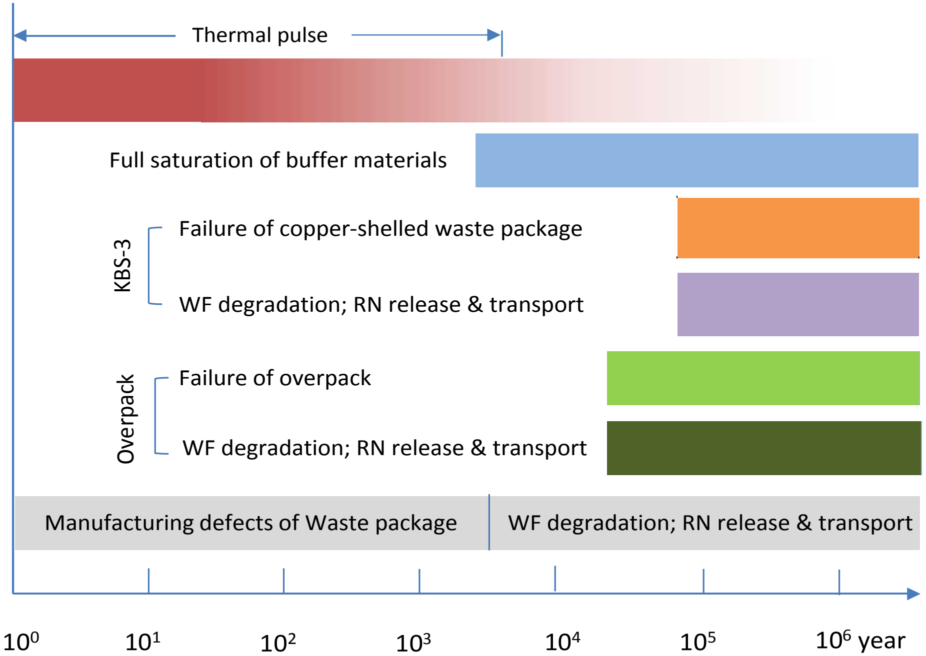
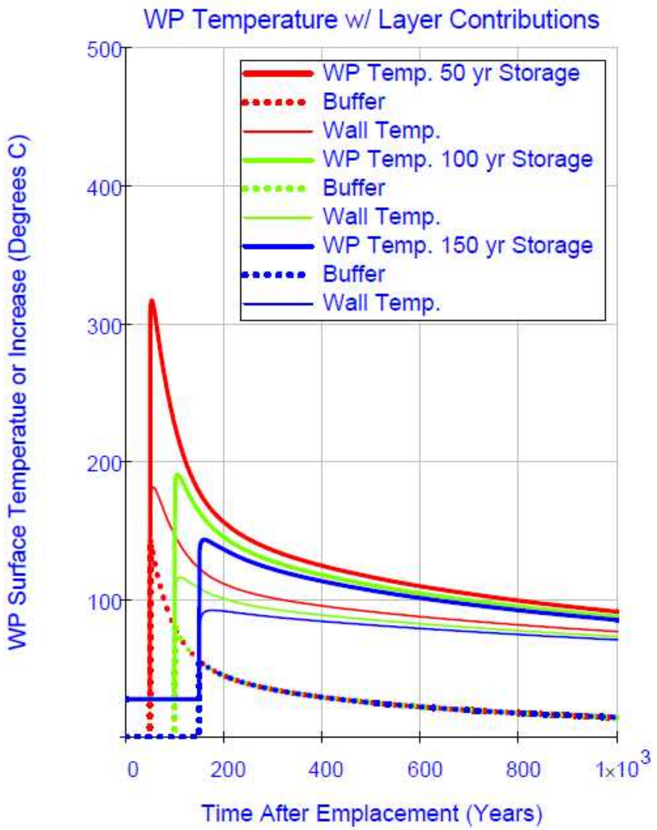
Thermal limit tests of buffer materials



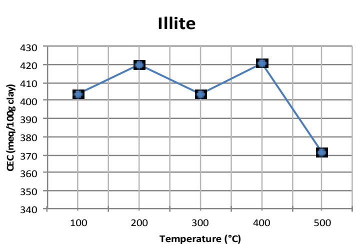


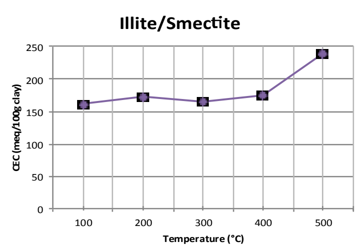
UOX fuel (40 GW-d/MT), 32-PWRs (Hardin & Voegele, 2013)

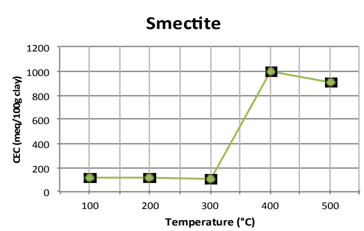
- **Key issues to be addressed:**
  - Can the existing thermal limit (100 ^oC) of bentonite materials be relaxed or even eliminated?
  - How will the thermal perturbation affect material's mechanical and chemical performance? (beneficially or detrimentally?)
- **Relevant conditions:**
  - Dry heating or heating with limited moisture

Thermal limit tests of buffer materials: Preliminary results

- **Materials tested:**
  - Smectite, illite, -mixed-layer illite-smectite
- **Testing conditions:**
  - 100 – 800 °C; dry heating (open), heating with limited moisture (closed); 1 – 8 hours
- **Material characterization:**
  - BET, CEC, TGA, XRD, Zeta sizing and settling measurements







Potential enhancement of cation exchange capacity (CEC)?

Enhanced instability of colloid suspension?

FY15: More systematic testing; swelling testing

Pu sorption on montomorillonite

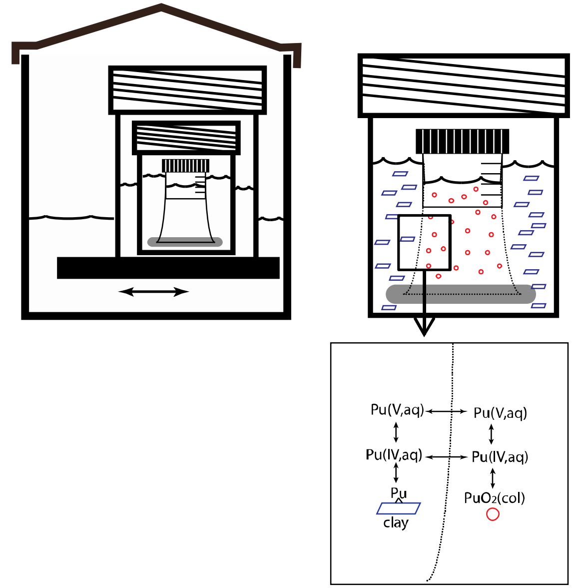
- **Key issues to be addressed:**
  - Stability of intrinsic Pu colloids
  - Formation of Pu pseudo-colloids
- **Experimental setup**
  - Dialysis bag
  - Control temperature
  - Presence or absence of montomorillonite

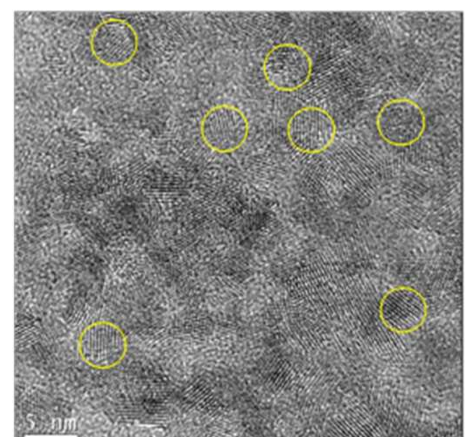
Limiting step

$$PuO_2(s)\,|\,inside \xrightarrow{k_1} Pu(aq)\,|\,inside$$
$$Pu(aq)\,|\,inside \xrightarrow{k_2} Pu(aq)\,|\,outside$$
$$Pu(aq) + clay\,|\,outside \overset{K,k_3,k_{-3}}{\Longleftrightarrow} Pu - clay\,|\,outside$$

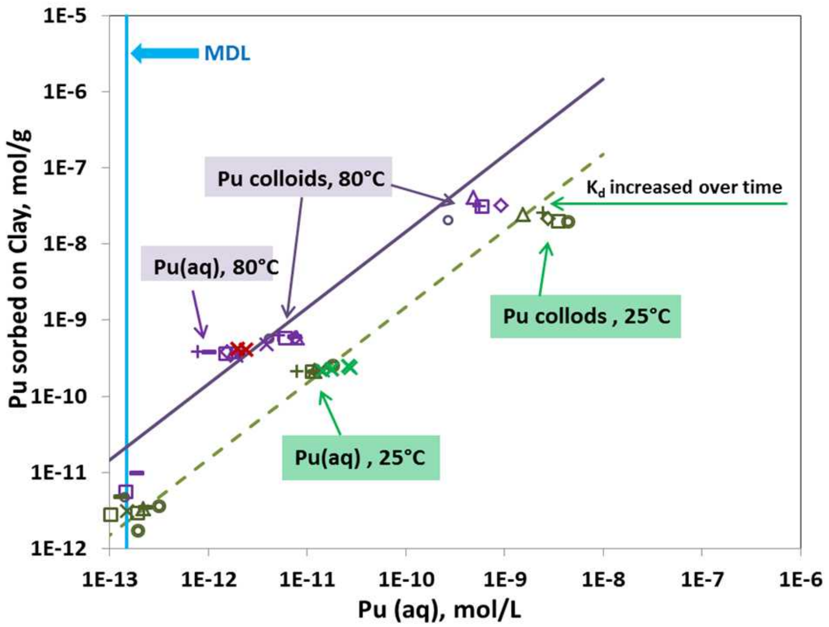
The kinetic constants for dissolution are one to two orders of magnitude lower than that of diffusion of aqueous Pu species.

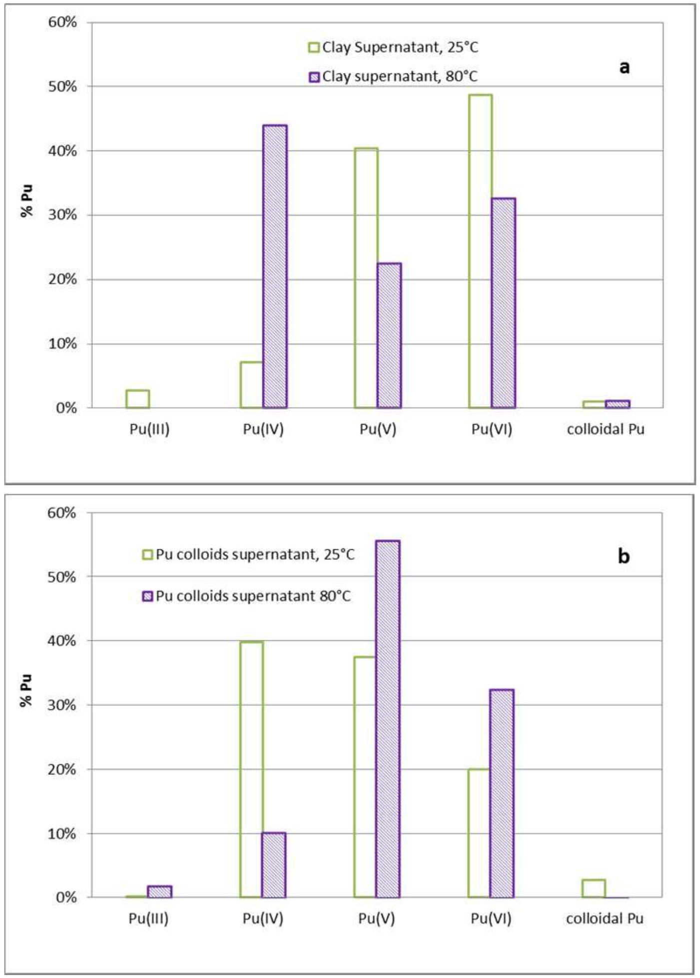
Zhao et al. (2014)





Pu sorption on montomorillonite: Temperature effect & oxidation state change





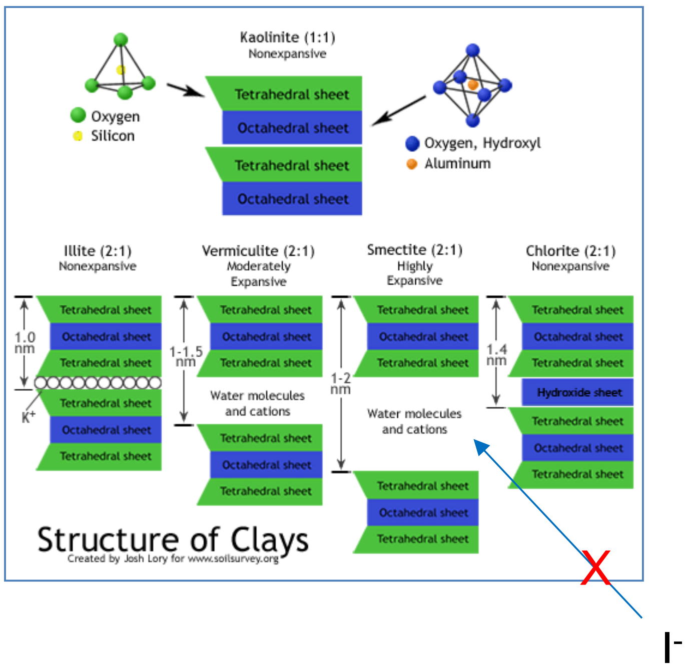
- Presence of clay stabilize dissolved Pu species and drives the formation of more stable Pu pseudo-colloids.
- Affinity of Pu for clay increases with temperature.
- Elevated temperatures may lead to more reducing solution conditions?

Zhao et al. (2014)

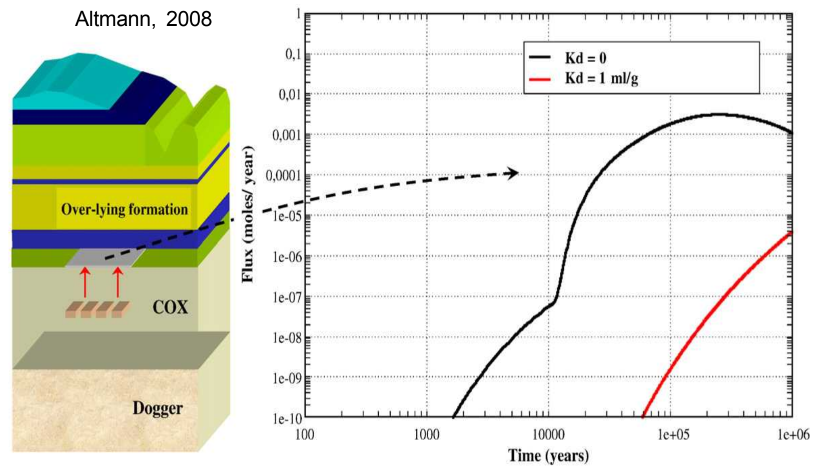
Iodide interaction with clays

- **I-129 a top contributor to total dose release. The questions are:**
  - Are the measured small Kd values real?
  - What is the mechanism for iodide interactions with clays?

Clay Mineral	Column K_D Value (mL/g)	Batch K_D Value (mL/g)	Ref.
Opalinus (Illite)	0.008-0.02		Van Loon et al., 2003
Montmorillonite	0.57		Sato et al., 1992
Callovo-Oxfordian (Interstratified illite/smectite)		0.15-0.37	Bazer-Bachi et al., 2006
Illite		27.7	Kaplan et al., 2000
Montmorillonite		-0.33	Kaplan et al., 2000



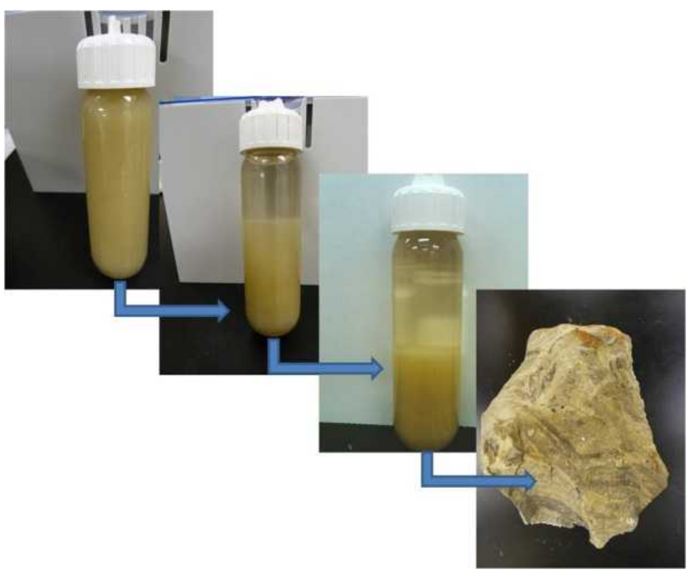
Altmann, 2008



Iodide interaction with clays (cont.)

7 clays under consideration: All clays obtained from the clay bank repository (Purdue Univ.)

- Kaolinite
- Ripidolite
- Illite
- Illite/Smectite
- Montmorillonite
- Palygorskite
- Sepiolite

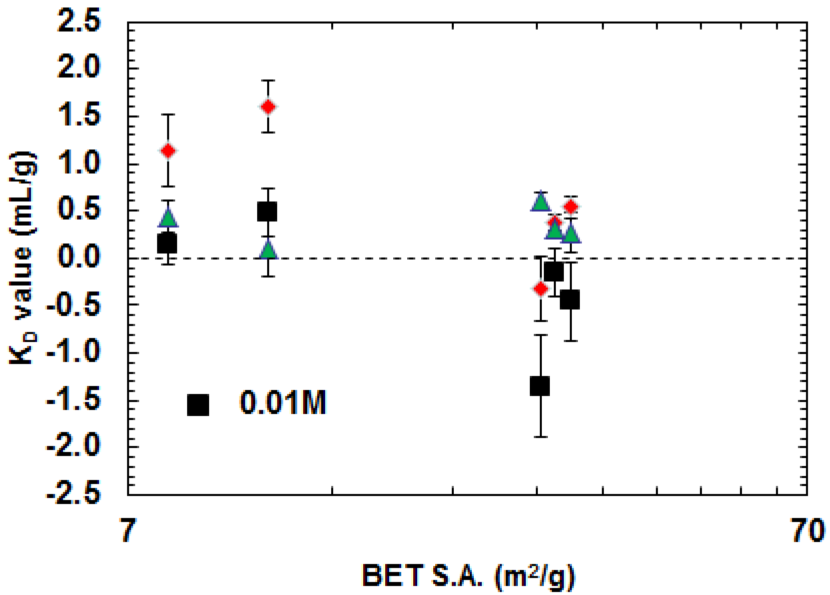


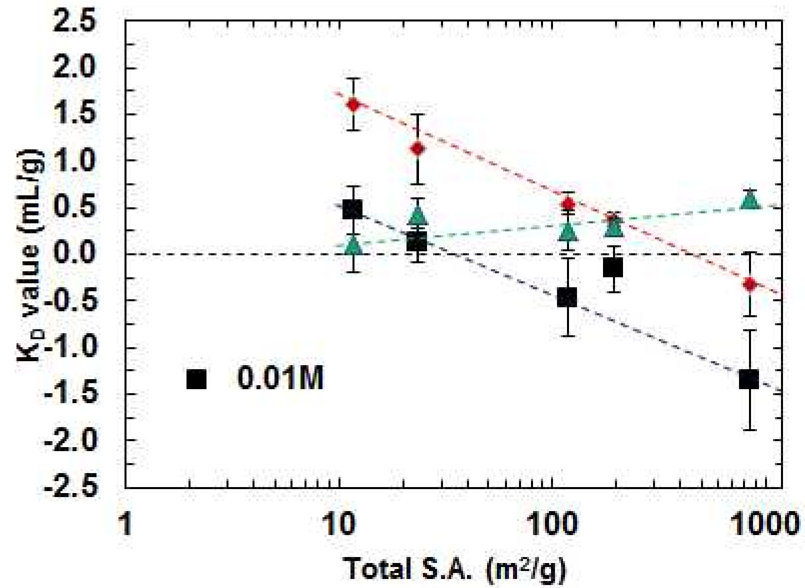
Sorption experiments:

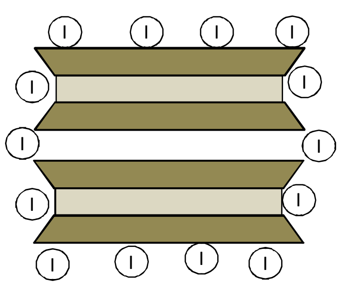
- **N_2 BET**
- **Methylene Blue (MB)**
  - Na-exchanged clays
  - Variable amounts of MB were added until clay surface was saturated
- **$BaCl_2$ Exchange**
  - Excess of barium displaces native cations
  - Measure native cation release
- **Iodide**
  - Solid:Liquid ratio: 100g/L
  - No specific pH control; 'natural' pH of clay
  - Seven day reaction time

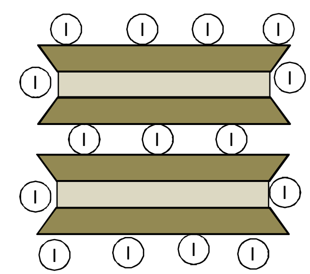
Concentration (M)	NaCl	NaBr	KCl
1.0	X		
0.1	X	X	X
0.01	X		

K_D values trend with total surface area, suggesting interactions with negatively charged interlayer sites.









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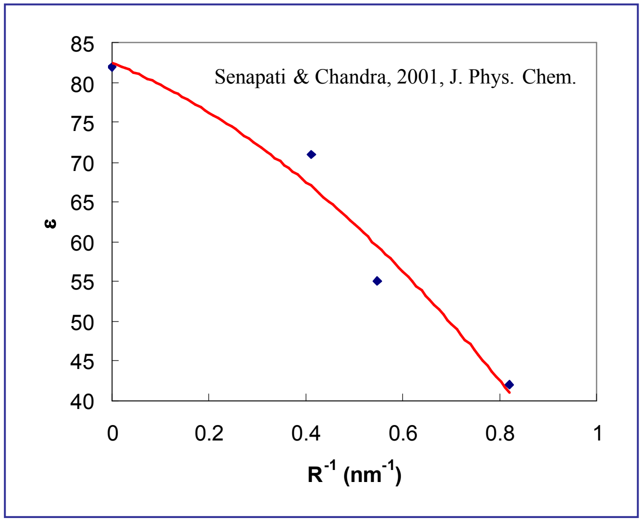
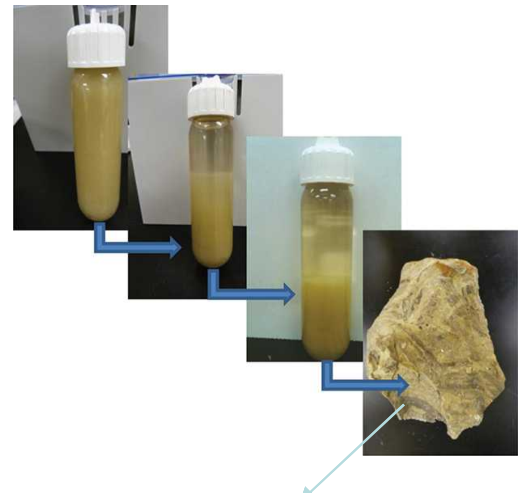
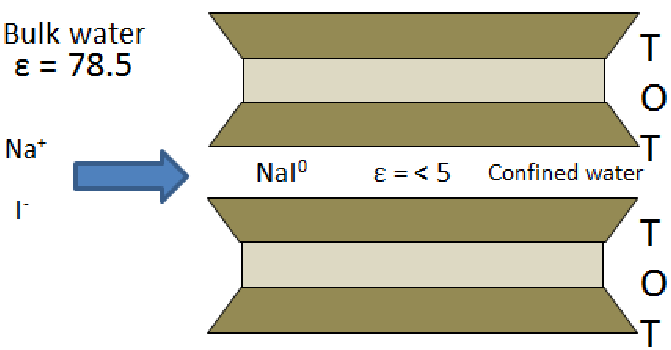
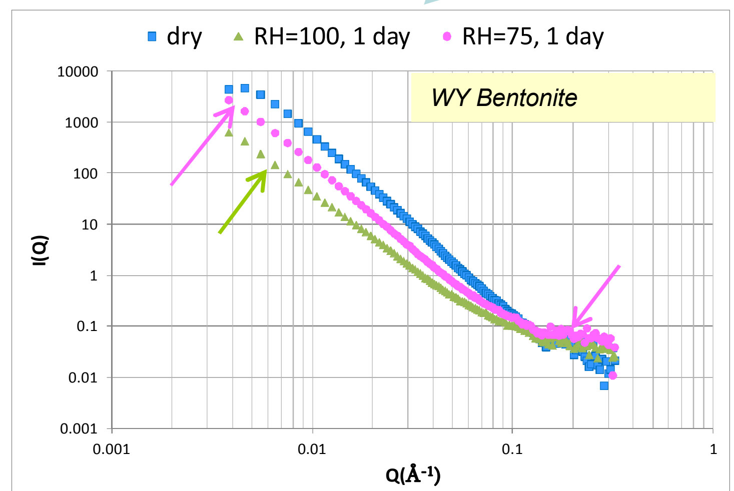
Data is consistent with ion pair formation caused by reduced dielectric constant of confined water





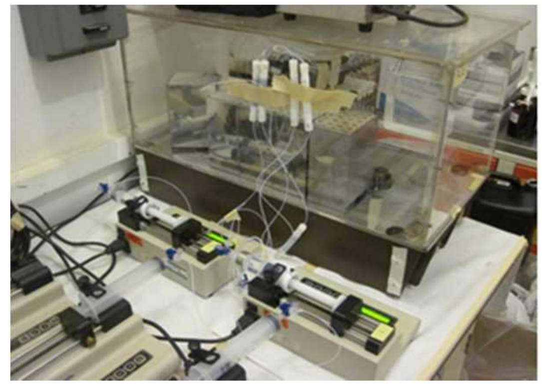


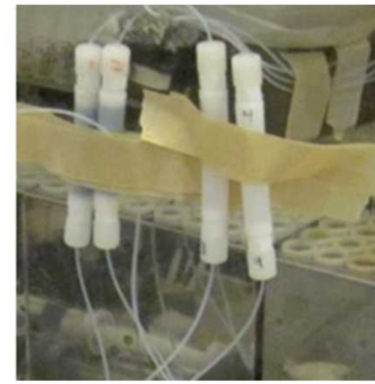


Ding (2013)

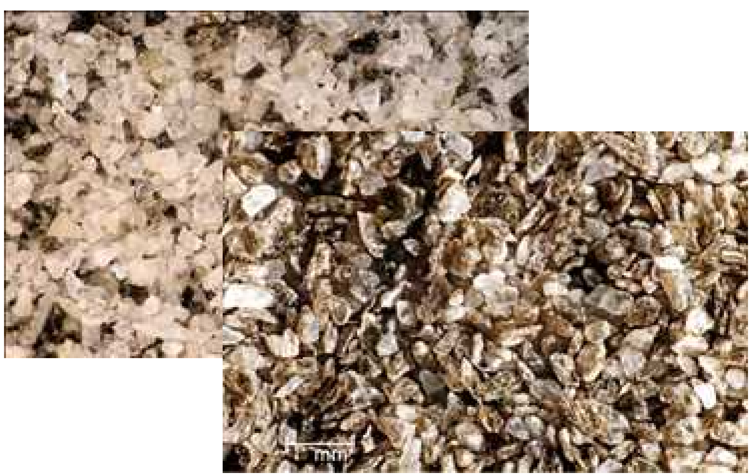
U interaction with granite: Column experiments

- **Key issues to be addressed:**
  - Traditional Kd approach over-predicts radionuclide migration.
  - How to interrogate the kinetics of multiple site radionuclide sorption and desorption?
  - How to upscale radionuclide sorption and desorption?





Grimsel granodiorite



Grimsel fracture filling material

$$\frac{\partial s_i}{\partial t} = k_{fi} c \left(1 - \frac{s_i}{s_{mi}}\right) - k_{ri} s_i$$

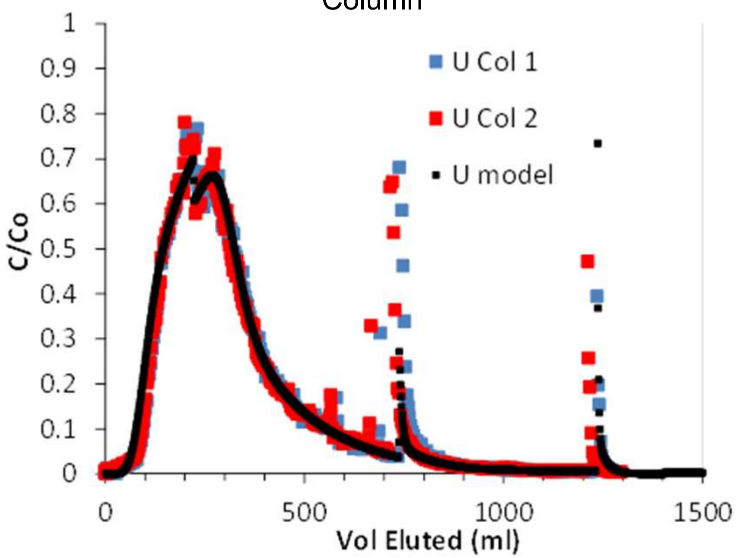
$$\frac{\partial c}{\partial t} = \frac{M}{V}\left[-\sum_i k_{fi} c \left(1 - \frac{s_i}{s_{mi}}\right) + \sum_i k_{ri} s_i\right]$$

$$\frac{\partial c}{\partial t} - v\frac{\partial c}{\partial x} + D\frac{\partial^2 c}{\partial x^2} + \frac{\rho_B}{\emptyset}\left[-\sum_i k_{fi} c \left(1 - \frac{s_i}{s_{mi}}\right) + \sum_i k_{ri} s\right]$$

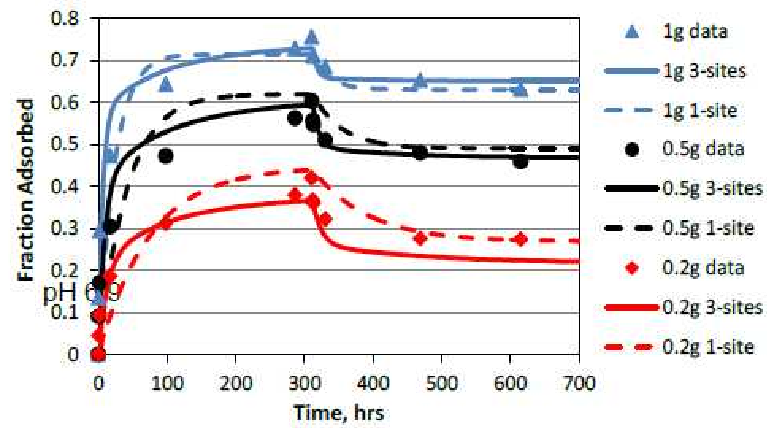
Multi-site reactive transport model

U interaction with granite: Column experiments (cont.)

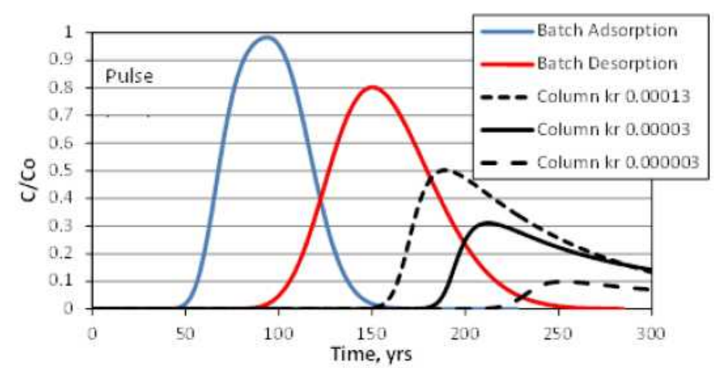
Column



Batch



Model simulation for a large scale system

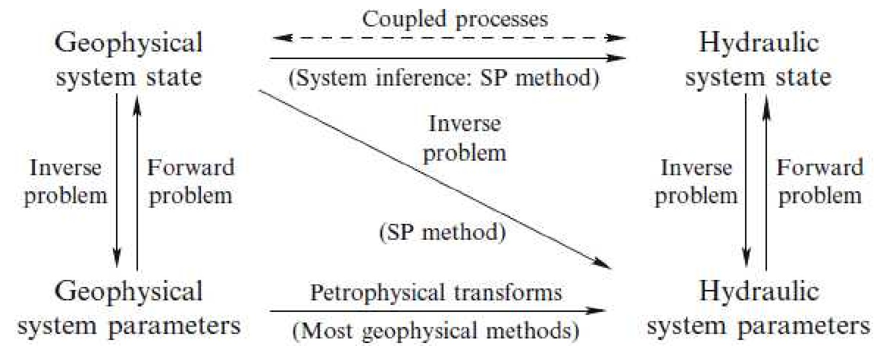


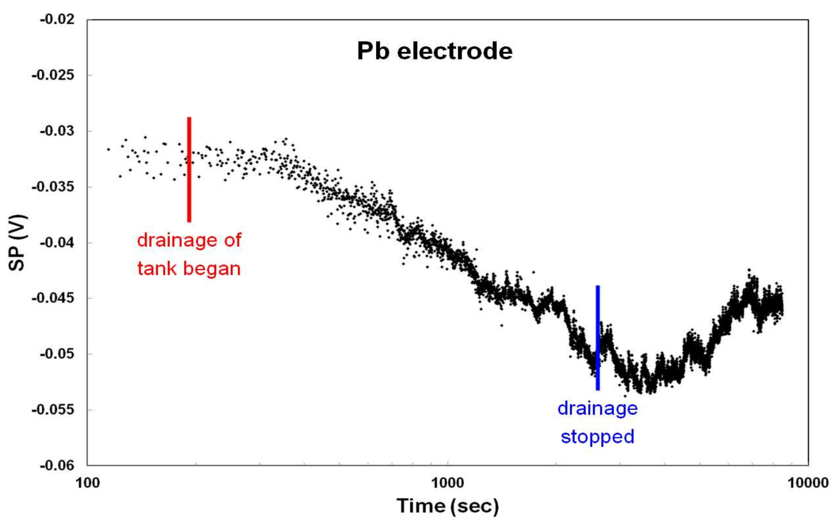
- It seems that column and batch experiments are transferable under relatively high solid/solution ratios.
- Different models may result in a significant difference in the prediction of long-term radionuclide retardation.

Development of site characterization techniques

Focus

- In-situ measurements in boreholes
- Multiple signal integration



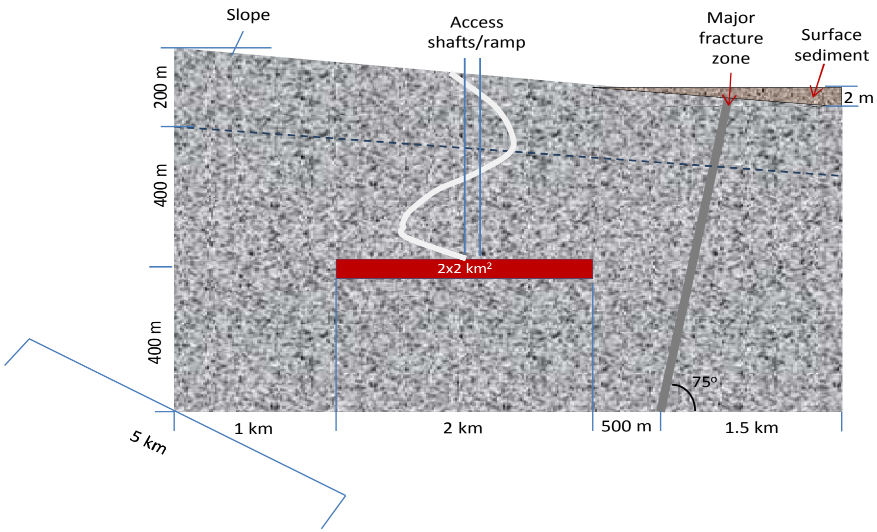


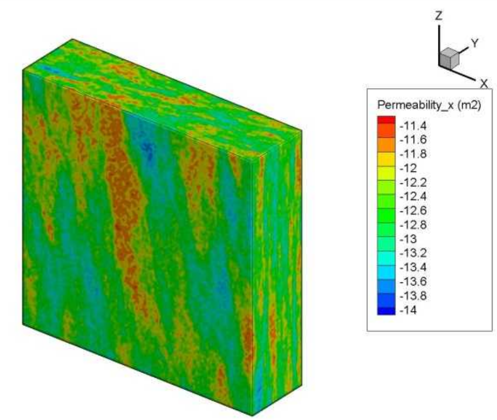
Darcy's flux $$\vec{q} = \overbrace{\frac{k}{\nu}\left(\nabla p - \rho_f \vec{g}\right)}^{\text{Darcy's law}} - \overbrace{\sigma C \nabla \varphi}^{\text{electro-osmosis}}$$

Current flux $$\vec{j} = \underbrace{\sigma C \left(\nabla p - \rho_f \vec{g}\right)}_{\text{electro-kinetic}} + \underbrace{\sigma \nabla \varphi}_{\text{Ohm's law}}$$

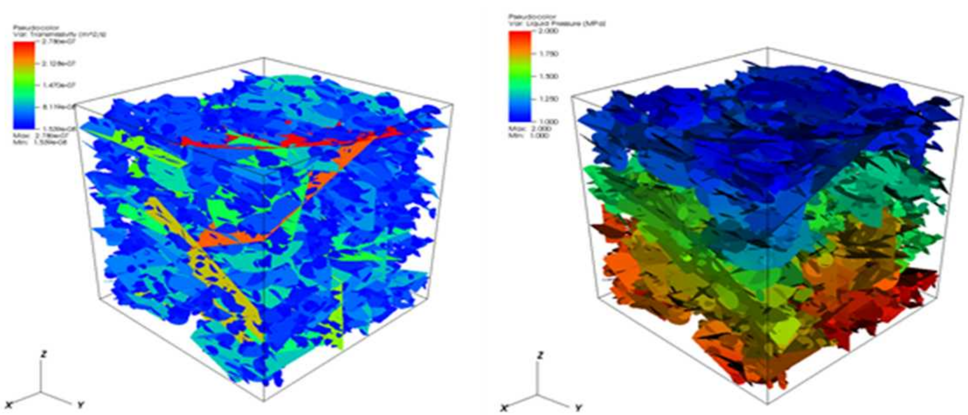
KAERI (2014)

Model development & demonstration using site characterization & tracer testing data

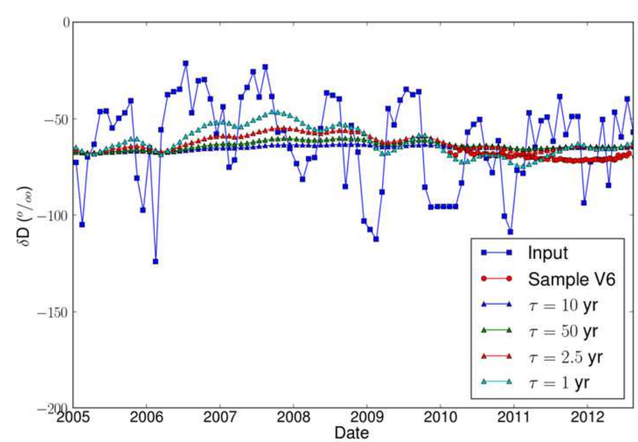




Fractured Continuum Model



Discrete fracture network model demonstration with Forsmark site data

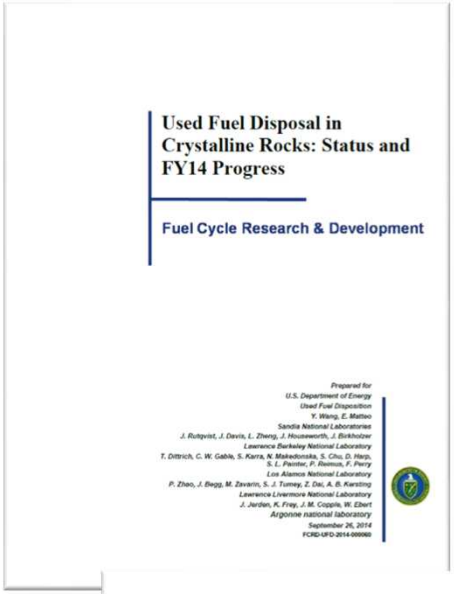


Modeling of tracer transport in Bedrichov Tunnel

Perspectives

FY15 work

- Better characterization and understanding of fractured media and fluid flow and transport in such media;
- Designing effective engineered barrier systems (EBS) for waste isolation. Specific attention will be given to the development of scientifically sound thermal limits for various buffer materials.
- Model demonstration using site characterization data obtained through international collaboration;
- Close coordination with deep borehole disposal activities.


Used Fuel Disposal in Crystalline Rocks: Status and FY14 Progress

Fuel Cycle Research & Development

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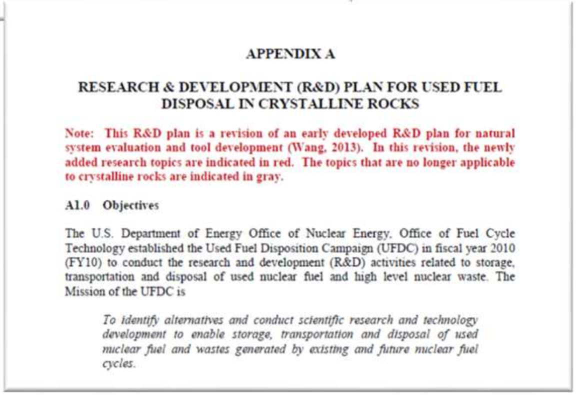
APPENDIX A

RESEARCH & DEVELOPMENT (R&D) PLAN FOR USED FUEL DISPOSAL IN CRYSTALLINE ROCKS

Note: This R&D plan is a revision of an early developed R&D plan for natural system evaluation and tool development (Wang, 2013). In this revision, the newly added research topics are indicated in red. The topics that are no longer applicable to crystalline rocks are indicated in gray.

A1.0 Objectives

The U.S. Department of Energy Office of Nuclear Energy, Office of Fuel Cycle Technology established the Used Fuel Disposition Campaign (UFDC) in fiscal year 2010 (FY10) to conduct the research and development (R&D) activities related to storage, transportation and disposal of used nuclear fuel and high level nuclear waste. The Mission of the UFDC is

To identify alternatives and conduct scientific research and technology development to enable storage, transportation and disposal of used nuclear fuel and wastes generated by existing and future nuclear fuel cycles.