

U.S. Department of Energy



#### Mass and Activity of Key Radionuclides Potentially Released From Waste Forms, Waste Packages and Drifts Over Time

Presented to: Nuclear Waste Technical Review Board

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#### General Engineered Barrier System (EBS) Design Features and Materials and Natural Processes





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### Yucca Mountain Radionuclide Inventory





Initial inventory from Total System Performance Assessment - Site Recommendation, adjusted for radioactive decay and ingrowth



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### **Yucca Mountain Radionuclide Inventory**

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Initial inventory from Total System Performance Assessment - Site Recommendation, adjusted for radioactive decay and ingrowth



### **Key Processes and Events**

Summary of Key Processes and Events that Affect the Estimate of Mass/Activity Flux from the Waste Packages and Engineered Barrier System

Model Component	Scenario Class					
	Nominal		Igneous		Seismic	
	Corrosion Failure	Early Failure	Intrusive	Eruptive	Mech Damage	Fault Displacement
Drift Seepage	Intact Partially Degraded	Intact Partially Degraded	No	N/A	Seepage for	Seepage for Degraded
	Drifts	Drifts	Diversion		Degraded Drifts	Drifts
Drip Shield Damage	GC Only; No Failures	Still Performs Flow Diversion	Assumed t	o Provide No	Still Performs	Loss of Function at
		Function	Barrier Fu	nction	Flow Diversion	$2 \times 10^{-7}$
					Function	Initiating Event
Waste Package Damage	GC, MIC, SCC; No Failures	Failed WPs Perform No	Assumed t	o Provide No	SCC Damage	Crimping Causes Opening
		Barrier Function; Poisson	Barrier Function		Function of	
		Distribution			Peak Ground	
					Velocity	
Waste Package Damage Due	Very Unlikely to Occur	Bounded by Early Failure	Due to Deg	gradation of	Very Unlikely	May Occur; Affects WPs
to Localized Corrosion	Because Drip Shields	Configuration	Waste Package During to		to Occur	Already Damaged by
	Function		Igneous Ev	vent		Crimping
Commercial Spent Nuclear	Initial Conditions Specify		Assumed to Provide No Function of Peak Ground Velocity			
Fuel Cladding Damage			Barrier Function			
Engineered Barrier System	Nominal Conditions		Basaltic	N/A	Temperature and Relative Humidity	
Thermal -Chemical			Chemistry		Adjusted for Col	llapsed Drifts
Environment			Conditions			

#### GC = Generalized Corrosion MIC = Microbially Influenced Corrosion SCC = Stress Corrosion Cracking





### **Key Processes in EBS Transport**

- Transport of dissolved radionuclides (RNs)
- Transport of RNs that are reversibly sorbed onto three types of mobile colloids: iron oxyhydroxide, waste form, and groundwater
- Transport of RNs irreversibly sorbed onto iron oxyhydroxide colloids
- Transport of embedded (irreversibly attached) RNs in waste form colloids
- Irreversible sorption of RNs (Pu and Am only) onto stationary corrosion products in the degrading waste package (no reversible sorption)





### Discretization of the EBS to Model Radionuclide Transport

#### • Spatial Domains:

- Waste form (WF) domain consisting of commercial spent nuclear fuel (CSNF) or high-level radioactive waste (HLW) glass
- Waste package corrosion products domain
- Invert domain composed of crushed tuff
- EBS- Unsaturated Zone (UZ) interface
   "domain" to establish
   a boundary condition for UZ transport
- Solving the mass transport equations for each domain requires specification of water volume, saturation, porosity, diffusive area, diffusive path length, diffusion coefficient, and advective flux; these vary by waste form type and environment (dripping and nondripping)







### Commercial Spent Nuclear Fuel Waste Form Transport

- CSNF WF domain represents breached and axial splitting of fuel that has degraded into a porous rind from alteration of UO<sub>2</sub>
- Rind (schoepite) is assumed to saturate quickly and completely for both dripping and non-dripping environments at temperatures below 100°C
- Continuous thin liquid film is assumed to exist at temperatures below 100°C
- Rind water volume is a function of time dependent fraction of degraded waste form, porosity, and saturation
- Rind (shoepite) porosity is treated as an epistemic uncertain parameter (sampled uniformly between 0.05-0.3 based on long term spent fuel test)
- Amount of each radionuclide mobilized is a function of rind water volume, waste form degradation rate and dissolved concentration of the radioelement in water
- No colloids present (RNs transported in dissolved state)





### **CSNF Waste Form Transport**

(Continued)

- CSNF WF diffusive area is the product of fuel rod length, split opening, and number of failed rods in a waste package
  - Split opening as a function of time differs among scenarios
- Diffusive Path length from CSNF WF domain to the corrosion products domain is the time varying thickness of the rind
- Diffusion Coefficient (all dissolved radionuclides):
  - Free water diffusion coefficient:  $2.3 \times 10^{-5}$  cm<sup>2</sup>/s at 25°C (from literature) represents upper bound
  - Modified by porosity and saturation using power law (i.e, form of Archie's Law)
- Advection occurs in Seismic Fault Displacement and Igneous Intrusion





### **Co-Disposed Waste Form Transport**

- Co-Disposed (CDSP) WF domain represents oxide and HLW glass logs degraded to a clay like alteration product
- HLW glass rind (clay) fuel is assumed to saturate quickly and completely for both dripping and non-dripping environments at temperatures below 100°C
- Continuous thin liquid film is assumed to exist at temperatures below 100°C
- HLW glass rind water volume is a function of time dependent fraction of degraded WF (porosity = 0.17, saturation = 1.0)
- Amount of each radionuclide mobilized is a function of rind water volume, waste form degradation rate and solubility of the radioelement in water
- Alteration of HLW glass includes embedded Pu and Am colloids





# Co-Disposed Waste Form Transport

- HLW glass WF diffusive area is the sum of the initial surface area of the five HLW glass logs in a CDSP waste package
- Diffusive Path length for HLW glass is the time varying alteration rind thickness
- Diffusion Coefficient (all radionuclides):
  - Free water diffusion coefficient:  $2.3 \times 10^{-5}$  cm<sup>2</sup>/s at 25°C (from literature) represents upper bound
  - Modified by porosity and saturation using power law (e.g. form of Archie's Law)
  - Reduced by a factor of 100 if radionuclide is bound to a colloid



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### **Corrosion Product Domain Transport**

- The corrosion product domain represents the internal components of a waste package (except WF) that have degraded after the waste package is breached
- Degradation of internals results in
  - Stationary iron oxyhydroxide corrosion products
  - Mobile iron oxyhydroxide colloids
- Time varying mass of corrosion products is a function of the mass of stainless steel and carbon steel available and their respective degradation rates
- There is no accounting for consumption of water by chemical reactions
- Water volume is the product of the pore volume and the saturation of the corrosion product mass
- Pore volume is a function of corrosion product mass, porosity, and iron oxyhydroxide density





### **Corrosion Product Domain Transport**

- Effective saturation in CSNF corrosion products is from adsorbed water and is a function of relative humidity and sampled specific surface area of corrosion products for non-dripping environments
- Saturation = 1 for dripping environments
- Diffusion coefficient modified by porosity and saturation using power law ( i.e., form of Archie's Law)
  - Reduced by a factor of 100 if radionuclide is bound to a colloid





## Corrosion Product Domain Transport

- Diffusive path length is represented as an epistemic uncertain parameter ranging from 0.02 m (thickness of the WP outer shell) to 0.859 m (outside radius of a representative CSNF WP) or 1.063 m (outside radius of a representative CDSP WP)
- Sorption:
  - Irreversible sorption to stationary corrosion products
  - Reversible and irreversible sorption to iron oxyhydroxide colloids
  - Reversible sorption to groundwater and WF colloids to groundwater and waste form colloids





### **Invert Domain Transport**

- RN concentration in the invert depends on radionuclide solubility limits, colloid stability, RN transfer between the corrosion product domain and the invert, and the boundary concentrations at the invert-UZ interface
- Advective flux sources for advective transport are defined by the EBS Flow abstraction and generally dominate diffusive releases
- Transport path along waste package pallet is not included





# Invert Domain Transport

- Diffusion coefficient modified by porosity and saturation using power law (i.e., form of Archie's Law)
  - Modified to account for temperature effects
  - Reduced by a factor of 100 if radionuclide is bound to a colloid
- Invert diffusive length is set to the average thickness of the invert (0.597 m)
- Invert diffusive area is equal to the product of the width of the invert top surface and the length of a CSNF or CDSP waste package





### **EBS-UZ Interface Transport**

- The EBS-UZ Interface domain establishes a boundary condition for calculating mass flux entering the UZ fractures and matrix continua
- A semi-infinite zero-concentration boundary condition is applied approximately three drift diameters below the invert
- The near-field UZ is modeled as a dual continuum consisting of overlapping UZ matrix and UZ fracture continua





### **Sorption in the EBS**

- Reversible sorption onto stationary corrosion products is not considered based on field observations and laboratory experiments
- Sorption onto corrosion products in the invert transport path is not considered
- Sorption coefficients (K<sub>d</sub>s) for reversible sorption of radionuclides (Pu, Am, Th, Pa, Cs) onto colloids
- Irreversible sorption rate constants (Pu, Am) for stationary corrosion products and iron oxyhydroxide colloids depend on:
  - Goethite sorption site density
  - Hydrous ferric oxide (HFO) sorption site density
  - Goethite surface area
  - Fraction of total iron oxide that is goethite
  - Percentage of high-affinity goethite sites
  - Percentage of high-affinity HFO sites
- Sorption coefficients (K<sub>d</sub>s) for reversible RN sorption onto crushed tuff in the invert (Pu, Am, Th, Pa, Cs, U, Np, Ra, Sr, Ac)



### **Colloid Transport in the EBS**

- Sorption coefficients (K<sub>a</sub>s) for reversible radionuclide (Pu, Am, Th, Pa, Cs) sorption onto all three types of colloids (WF, iron oxyhydroxide, and groundwater)
- Rate constant for irreversible sorption (Pu, Am) onto iron oxyhydroxide colloids is dependent on uncertain parameters
- Rate constant for irreversible sorption (Pu, Am) onto WF colloids is a function of ionic strength and pH
- Colloid processes that are not considered:
  - Physical filtration of colloids in the drift
  - Colloid retardation due to sorption at the air-water interface
  - Interaction of colloids with organic components



#### Gravitational settling of colloids

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### Key Parameters Determining EBS Releases In Nominal, Igneous Intrusion, and Seismic Ground Motion Modeling Cases

Nominal Early Failure Modeling Case	Igneous Intrusion Modeling Case	Seismic Ground Motion Modeling Case		
Number of Early     Waste Package     Breaches	<ul> <li>Number of Waste Packages and Drip Shields Disrupted</li> </ul>	<ul> <li>Waste Package Damaged Area</li> <li>In-Package Solubility (e.g.,of Uranium and Neptunium)</li> </ul>		
<ul> <li>In-Package Solubility (e.g., of Uranium and Neptunium)</li> </ul>	<ul> <li>In-Package Solubility (e.g., of Uranium and Neptunium)</li> </ul>			
<ul> <li>Irreversible Sorption (e.g., of Plutonium and Americium) onto Corrosion Product Stationary Phases</li> <li>In-Package Diffusion Characteristics</li> </ul>	<ul> <li>Irreversible Sorption (e.g., of Plutonium and Americium) onto Corrosion Product Stationary Phases</li> <li>Colloid Concentration Limits</li> </ul>	<ul> <li>Irreversible Sorption (e.g., of Plutonium and Americium) onto Corrosion Product Stationary Phases</li> <li>In-Package Diffusion Characteristics</li> </ul>		

