United States
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NRC'S APPROACH TO MODELING RADIONUCLIDE RELEASE FROM THE ENGINEERED BARRIER SYSTEM

ACNW Working Group on the Near-Field Environment and Performance of Engineered Barriers in the Yucca Mountain Repository

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OBJECTIVES

- Overview of general approach
- Discussion of Total-system Performance Assessment (TPA) waste form models
- Solubility considerations
- Engineered Barrier System (EBS) interactions
- Comparison between Department of Energy (DOE) and NRC approaches
- Conclusions

OVERVIEW OF GENERAL APPROACH

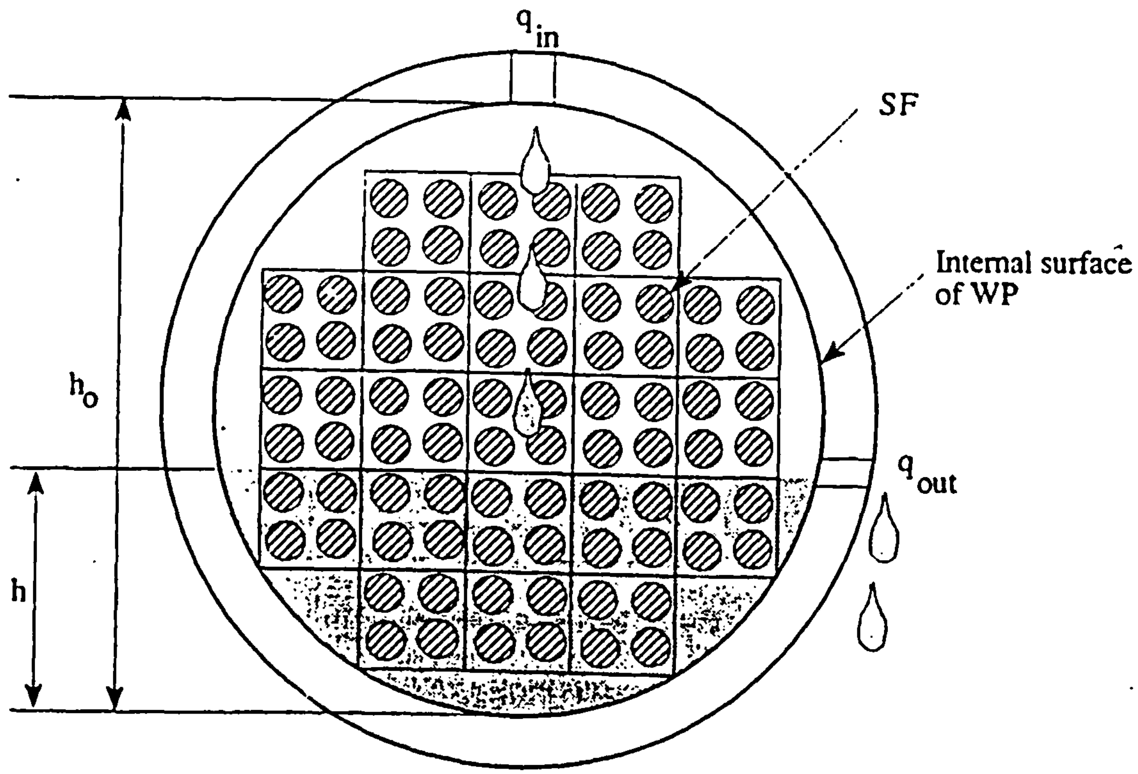
- Integrated, flexible and iterative
  - continual interactions between Container Life and Source Term, Evolution of the Near-Field Environment, and TSPAI teams
  - interplay between process-level and TPA level sensitivity studies
  - continual enhancements and modifications of the TPA code

- Radionuclide release from the EBS
  - assumes congruent dissolution of only the spent fuel waste form in a bathtub or flow-through configuration
  - solubility constraints applied on radionuclides released from waste form
  - transfer function approach to abstract other EBS material interactions

- NOTE: Use of a particular approach, model, or parameter in TPA 3.2 should NOT be construed as regulatory acceptance

- CAUTION: INSIGHTS AND ASSERTIONS ARE PRELIMINARY
  - Parameter and model development is continuing
  - Preliminary outputs based on limited analysis

BATHTUB MODEL



Schematic of bathtub model with incoming and outgoing water conduits

RADIOACTIVE RELEASE FROM WASTE FORM IN TPA 3.2

- Four options that are treated as alternative conceptual models
  - Fuel dissolution in the presence of carbonate (Ca and Si absent)
  - Fuel dissolution in the presence of Ca and Si
  - Constant rate of release
  - Secondary mineral (schoepite) dissolution

- Common assumptions
  - no colloidal release
  - time invariant chemistry
  - bathtub model that allows for flow-through calculations
  - radionuclides released at rate of fuel dissolution (congruent)
  - solubility constraints on radionuclides once released from fuel
  - option for cladding credit
  - release to interior of waste package
  - advective release from waste package
  - oxidizing environment

CARBONATE DISSOLUTION MODEL - MODEL 1

- Based on laboratory flow-through tests of Gray and Wilson (1995)

- Function of total carbonate concentration, oxygen partial pressure, temperature and pH

$$\log r = 9.310 + 0.142 \log [\Sigma CO_2] - 16.7 \log p_{O2} + 0.140 \log [H^+] - (2130/T) + 6.81 \log (T) \bullet \log p_{O2}$$

- Assumptions
  - no Ca and Si present
  - no secondary mineral formation
  - constant surface area

- Near field geochemical reactions influence on variables
  - volatilization from thermal pulse affects carbon and gas chemistry
  - interaction with cementitious materials affects carbonate and pH
  - reaction with metal and fuel could influence gas and water chemistry

CALCIUM AND SILICA DISSOLUTION MODEL - MODEL 2

- Based on laboratory batch tests of Wilson (1990)

- Function of temperature

$$r = (1.4 \sim 5.5) \times 10^4 \exp[-34.3/RT]$$

- Assumptions
  - Ca and Si always present at J-13 concentration
  - constant surface area

- Near field geochemical reactions influence on variables
  - reaction with fuel could reduce Ca and Si concentration, leading to formation of secondary minerals such as schoepite
  - interaction with cementitious materials affects silica and calcium
  - reaction with metal could influence water chemistry

CONSTANT RELEASE RATE MODEL - MODEL 3

- Fixed release rate; can be based on empirical or natural analog data

- Peña Blanca natural analog constraints
  - ore body mass and radiometric dating of period of oxidation
  - secondary U mineral solubility control on U release

- Maximum average value for U oxidation at Peña Blanca is calculated
  - scaled to the Yucca Mountain repository
  - yields a lower limit on the release from the proposed repository

- Assumptions
  - long-term release controlled by secondary uranium minerals
  - not dependent on surface area
  - release of other radionuclides proportional to U release

- Near field geochemical reactions influence on variables
  - reaction with metal and cementitious materials could influence water chemistry and may not be analogous to chemistry of reactions at Peña Blanca

SECONDARY MINERAL DISSOLUTION MODEL - MODEL 4

- Based on short-term (up to 10 yr) vapor phase and drip test observations, natural analog, and thermodynamic studies summarized by Murphy (1997)

- Schoepite is the controlling secondary U mineral - $UO_3 \bullet 2H_2O$

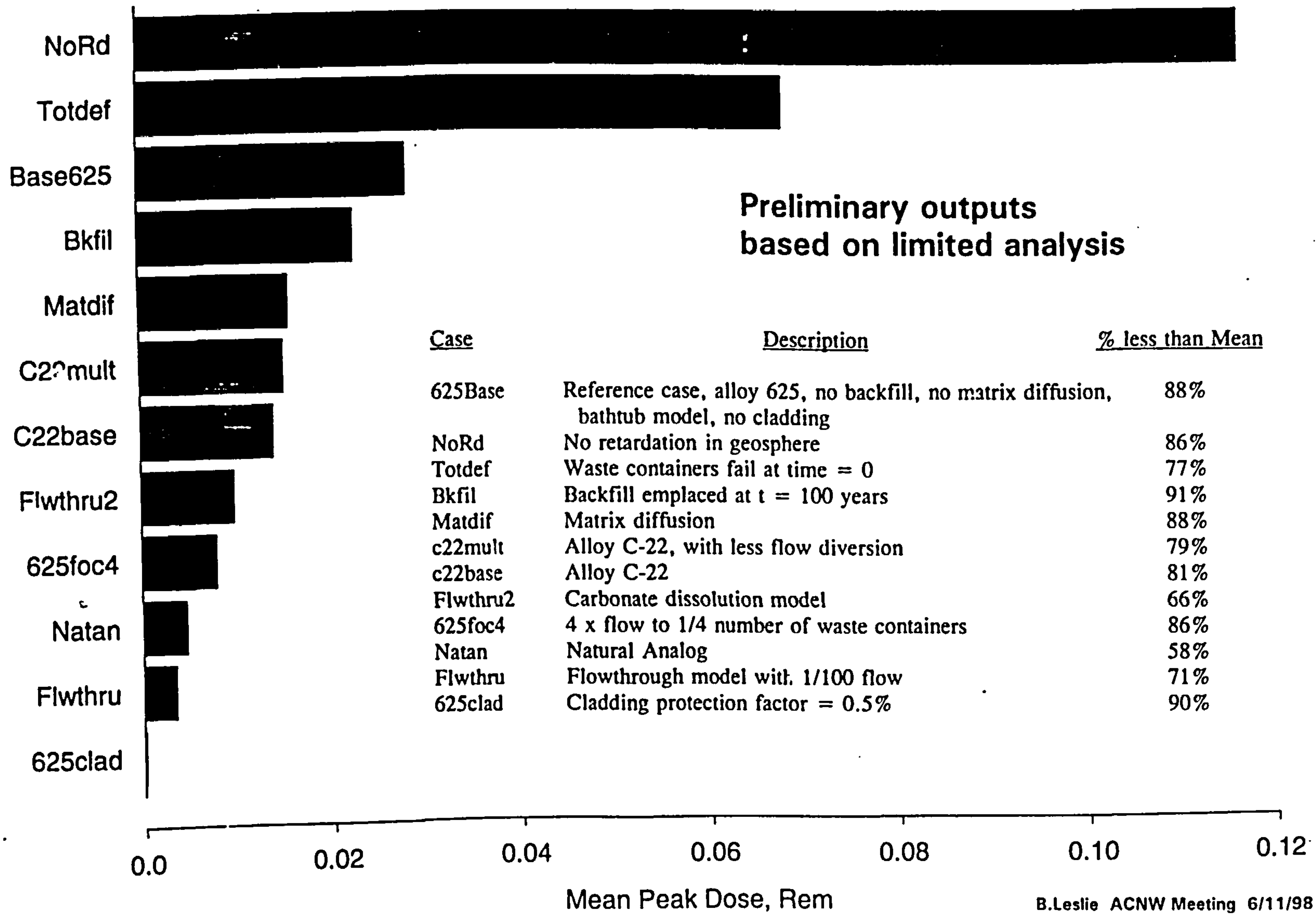
- Release is a function of temperature, pH, and total carbonate
  - schoepite exhibits retrograde solubility

- Assumptions
  - schoepite remains as dominant U secondary mineral
  - dissolution is not dependent on surface area (solubility limited)
  - complete rapid uptake of other radionuclides by schoepite (but this constraint can be relaxed; *e.g.*, all I_2 released as a pulse)
  - release of other radionuclides proportional to U release

- Near field geochemical reactions influence on variables
  - reaction with metal and cementitious materials could influence water chemistry and may not be analogous to laboratory conditions

PRELIMINARY OUTPUT BASED ON LIMITED ANALYSIS

Mean Peak Dose for 50,000 Years, Rem



Preliminary outputs based on limited analysis

Case	Description	% less than Mean
625Base	Reference case, alloy 625, no backfill, no matrix diffusion, bathtub model, no cladding	88%
NoRd	No retardation in geosphere	86%
Totdef	Waste containers fail at time = 0	77%
Bkfil	Backfill emplaced at t = 100 years	91%
Matdif	Matrix diffusion	88%
c22mult	Alloy C-22, with less flow diversion	79%
c22base	Alloy C-22	81%
Flwthru2	Carbonate dissolution model	66%
625foc4	4 x flow to 1/4 number of waste containers	86%
Natan	Natural Analog	58%
Flwthru	Flowthrough model with 1/100 flow	71%
625clad	Cladding protection factor = 0.5%	90%

RADIONUCLIDE SOLUBILITIES - OVERVIEW

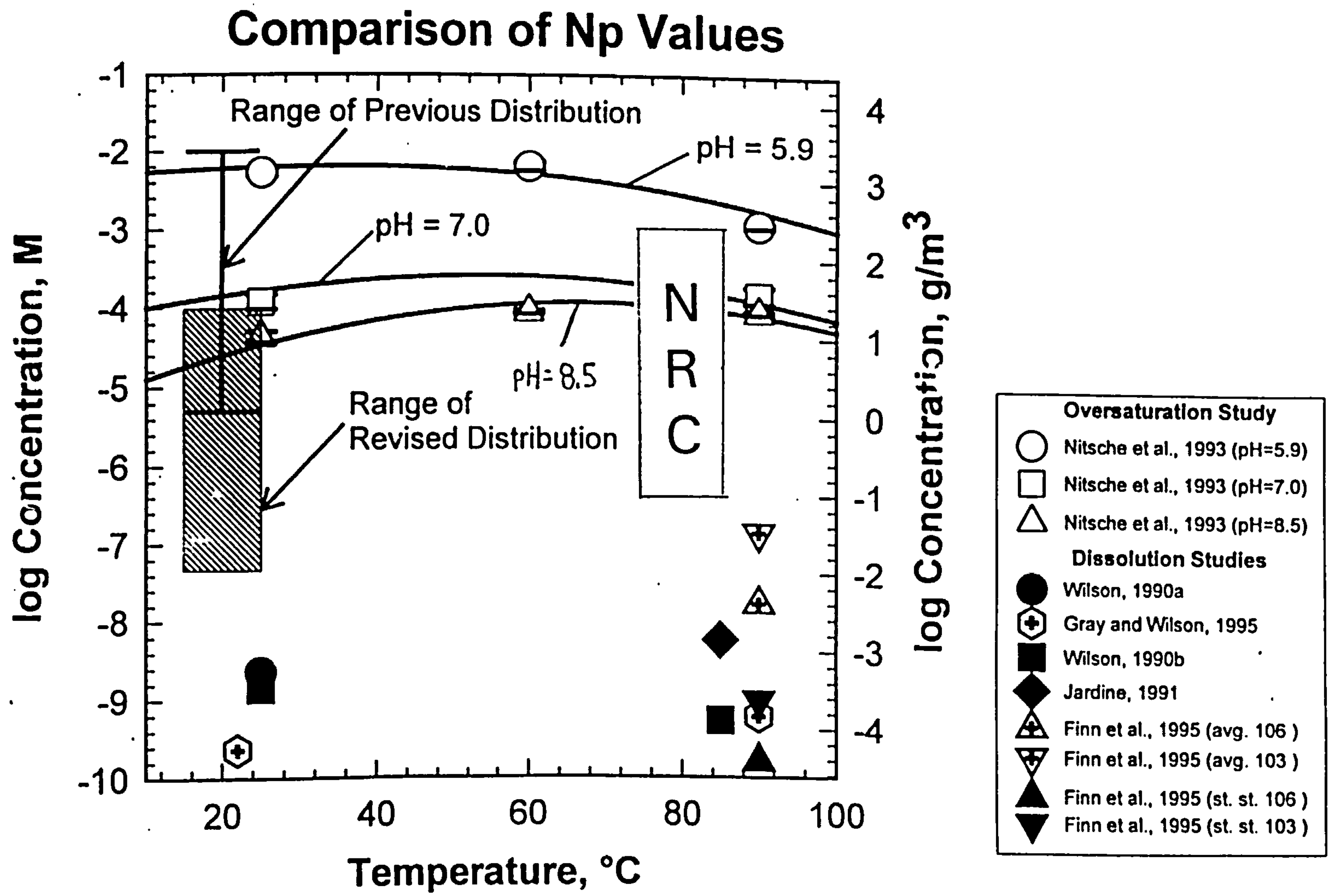
- Assumptions
  - Solubilities chosen for oxidative Yucca Mountain conditions
  - Solubilities controlled by pure mineral phases
  - Derived primarily from a poorly documented DOE "expert elicitation"

- Auxiliary analyses of solubilities
  - Murphy (1996) evaluated TSPA-1995 solubilities using EQ3 and a range of water chemistries reflective of Yucca Mountain fluids; found that TSPA-95 values were conservative
  - Ongoing study at CNWRA of "reacted waters" solubilities
  - Evaluating a correlated-solubility approach

- Doses are sensitive to solubilities - especially Np

- Inconsistency in approach to solubilities and release model needs to be considered; flexibility of range of solubilities for different release models

NEPTUNIUM SOLUBILITY - COMPARISON OF APPROACHES



(modified from McNeish, March 17, 1998)

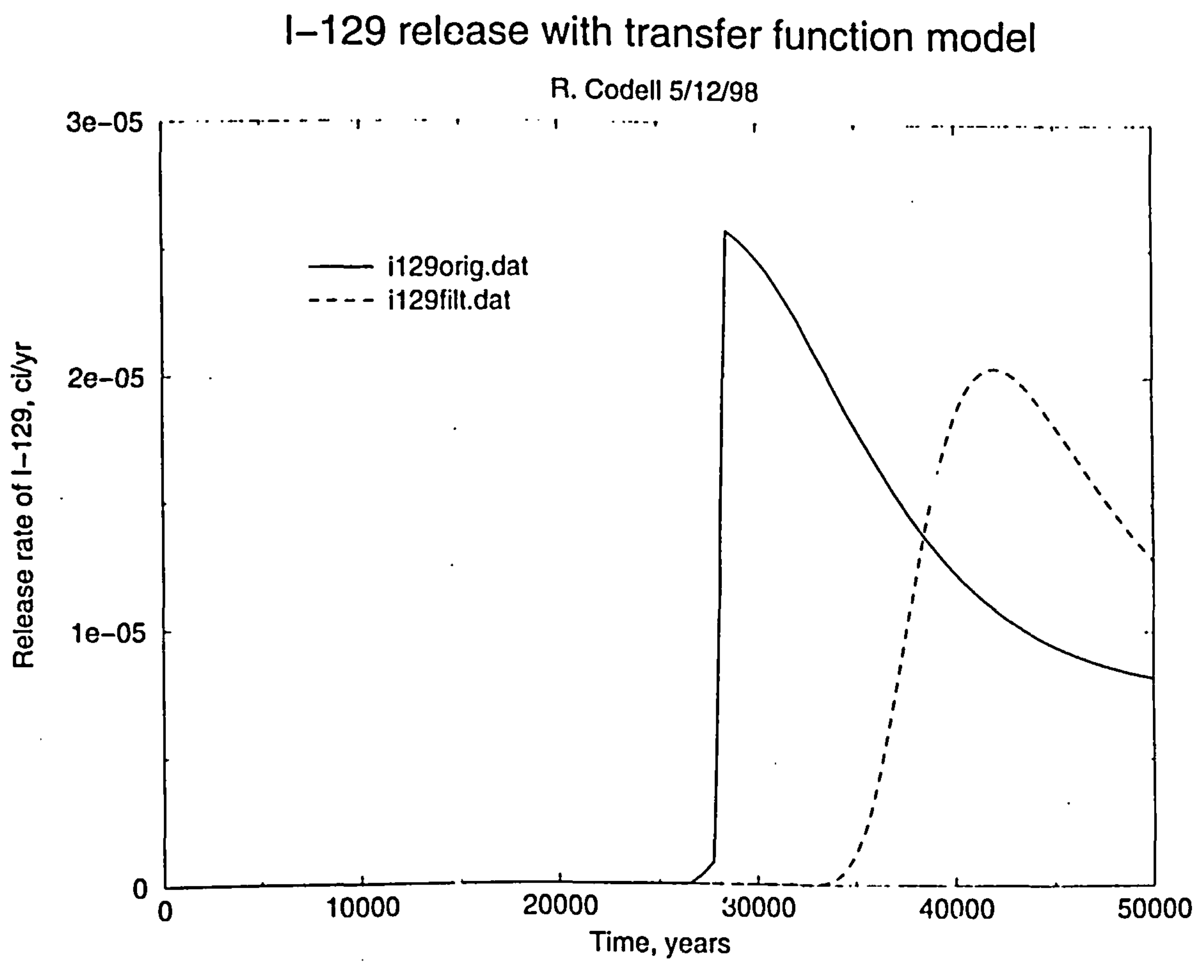
OTHER EBS INTERACTIONS - GENERAL

- Design specific
  - requires flexibility within TPA code to evaluate different design options
  - ability to evaluate sorption or co-precipitation

- No design specific model incorporated
  - will evaluate using transfer function approach in TPA 3.2
  - sufficient to evaluate DOE TSPA-VA base case

- Evaluate EBS interactions via process-level modeling (MULTIFLO) and sensitivity studies using TPA 3.2
  - metal specific K_ds, transfer function approach
  - cementitious specific K_ds, transfer function approach
  - alkaline solubilities tested via alternative solubility limits

TRANSFER FUNCTION - EXAMPLE



PRELIMINARY OUTPUT BASED ON LIMITED ANALYSIS

NRC'S TRANSFER FUNCTION APPROACH

- Used to model mass transport in the near field (*e.g.*, transport through the invert or backfill)

- Recently added to TPA version 3.2
  - allows flexibility to conduct sensitivity studies and assess the potential importance of other design options

- An option that is user chosen

- Requires time-invariant flux of water

- Requires appropriate radionuclide-specific input values to model anticipated sorption behavior

OVERALL COMPARISON OF APPROACHES

	Waste Form	Colloids	Release Models	Solubility	EBS approach
NRC	SF	No	Alternative Conceptual Models: Carbonate; Ca and Si; Constant Release Rate; and Schoepite	Base case is conservative, yet flexible for analysis under alternative conceptual model approach	Transfer function and sensitivity studies, advective only
DOE	SF and Glass	Yes	Carbonate; Secondary Minerals	Np ten times lower than NRC may affect dose	Advective, diffusive, and retardation

CONCLUSIONS

- Integrative and iterative approach to modeling radionuclide release from Engineered Barrier System, using process-level analyses, TPA code sensitivity studies, and continuous, controlled modification of the TPA code

- Assumption of time-invariant chemistry requires analysis of radionuclide release using alternative conceptual models

- Limitations include little data to support existing models for release, short duration of laboratory tests, and few experiments to evaluate EBS interactions and their impact on performance