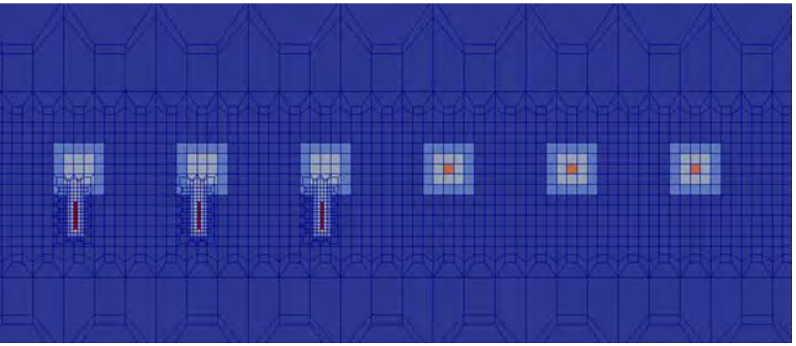
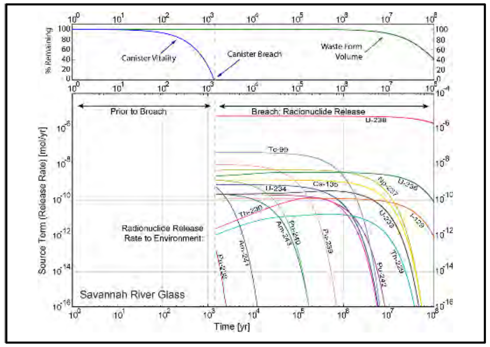
Exceptional service in the national interest

Sandia National Laboratories





MODELING WASTE PACKAGE DEGRADATION AND WASTE FORM DISSOLUTION FOR GEOLOGIC REPOSITORY PERFORMANCE ASSESSMENT IN PFLOTRAN

SAND2017-8898-C

Jennifer M. Frederick, Glenn E. Hammond, Paul E. Mariner, Emily R. Stein, S. David Sevougian

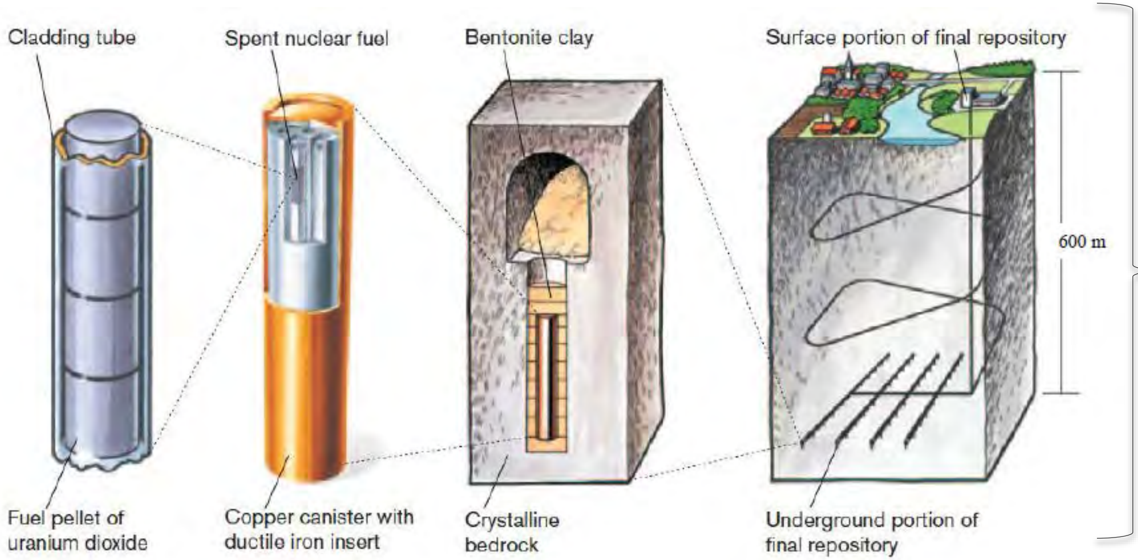
U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

A Generic, Geologic Repository

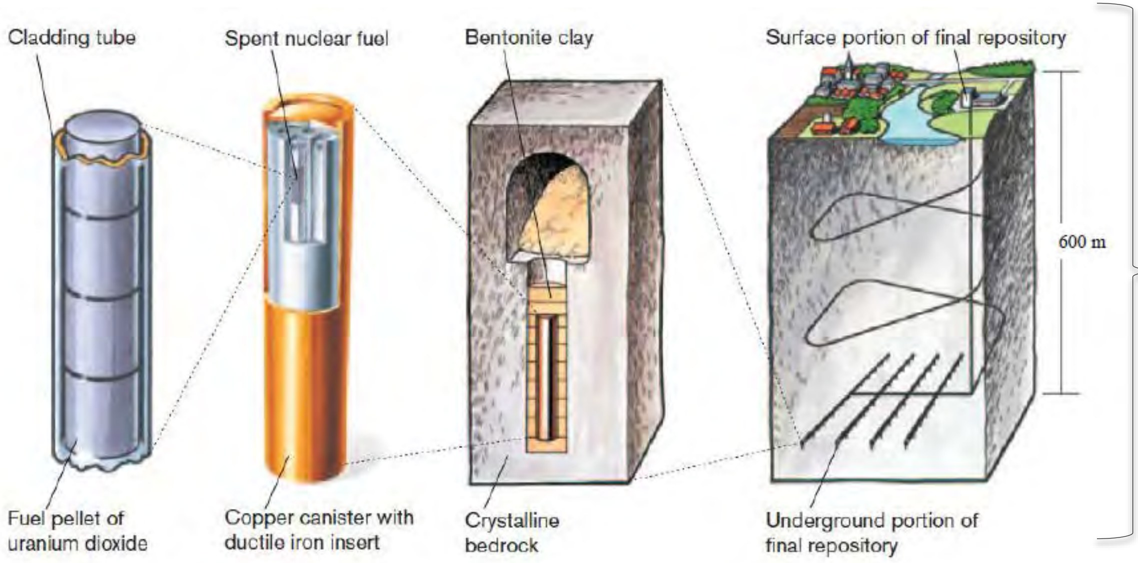
- A performance assessment (PA) for a nuclear waste repository requires a **comprehensive model** that can **simulate plausible scenarios and processes** that may affect **repository performance and safety**.



An example geologic repository in crystalline bedrock consists of a waste form within a copper canister that is surrounded by a bentonite clay 'buffer' and placed in the floor of mined drifts deep underground. The repository footprint at the surface is very small. Wang et al. (2014)

A Generic, Geologic Repository

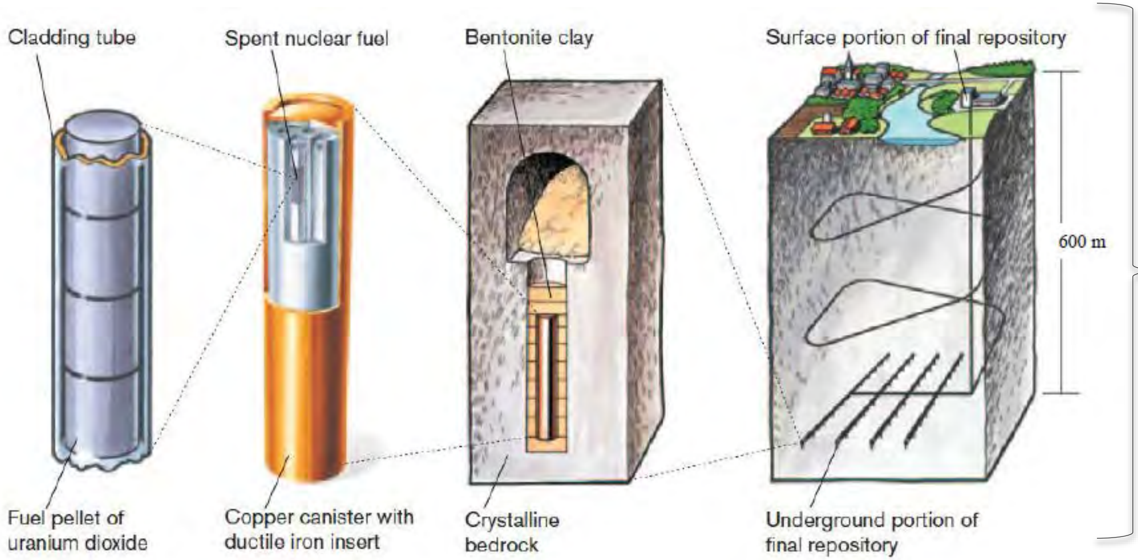
- Disposal options for spent nuclear fuel and high-level nuclear waste include:
  - mined repository concepts in salt, argillite, and crystalline rock
  - deep borehole disposal in crystalline rock.



An example geologic repository in crystalline bedrock consists of a waste form within a copper canister that is surrounded by a bentonite clay 'buffer' and placed in the floor of mined drifts deep underground. The repository footprint at the surface is very small. Wang et al. (2014)

A Generic, Geologic Repository

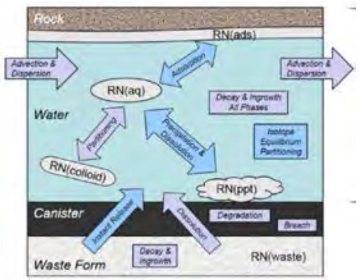
- A major modelling challenge is accurately representing processes across **multiple spatial and temporal scales**:



An example geologic repository in crystalline bedrock consists of a waste form within a copper canister that is surrounded by a bentonite clay 'buffer' and placed in the floor of mined drifts deep underground. The repository footprint at the surface is very small. Wang et al. (2014)

A Generic, Geologic Repository

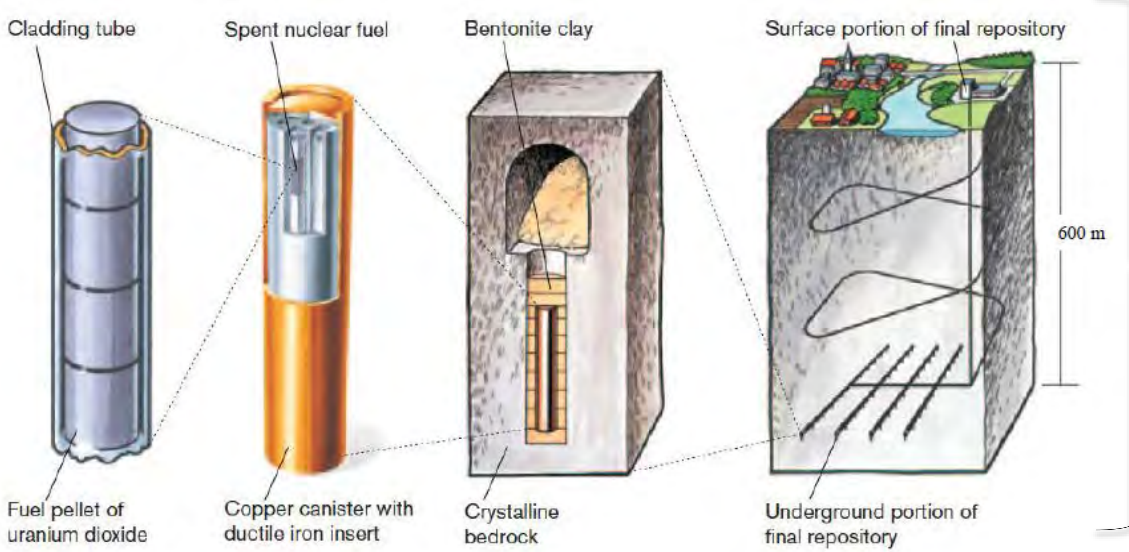
- A major modelling challenge is accurately representing processes across **multiple spatial and temporal scales**:



O(<<m^3), O(sec < t 1e8yr)

O(m^3)

O(km^3), O(t=1e8yr)



An example geologic repository in crystalline bedrock consists of a waste form within a copper canister that is surrounded by a bentonite clay 'buffer' and placed in the floor of mined drifts deep underground. The repository footprint at the surface is very small. Wang et al. (2014)

GDSA Framework

Sandia National Laboratories

GDSA = Geologic Disposal Safety Assessment

Input Parameters

Parameter database

DAKOTA

- Interface Control
- Uncertainty Sampling and Quantification
- Sensitivity Analysis

Computational Support

Processing (Python, etc.)

Mesh generation

Visualization

ParaView
Parallel Visualization Application

VisIt

PFLOTRAN

Multi-Physics Simulation and Process Integration

Source Term and EBS Evolution Model

- Inventory (decay/ingrowth)
- WF degradation
- WP degradation
- Radionuclide release
- Thermal, mechanical
- Gas generation

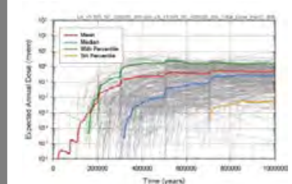
Flow and Transport Model

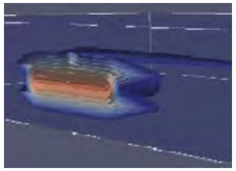
- Advection, diffusion, dispersion
- Sorption
- Precipitation, dissolution
- Decay, ingrowth
- Colloids
- Chemical reactions
- Heat transport

Biosphere Model

- Exposure pathways
- Uptake/ transfer
- Dose calculations

Output

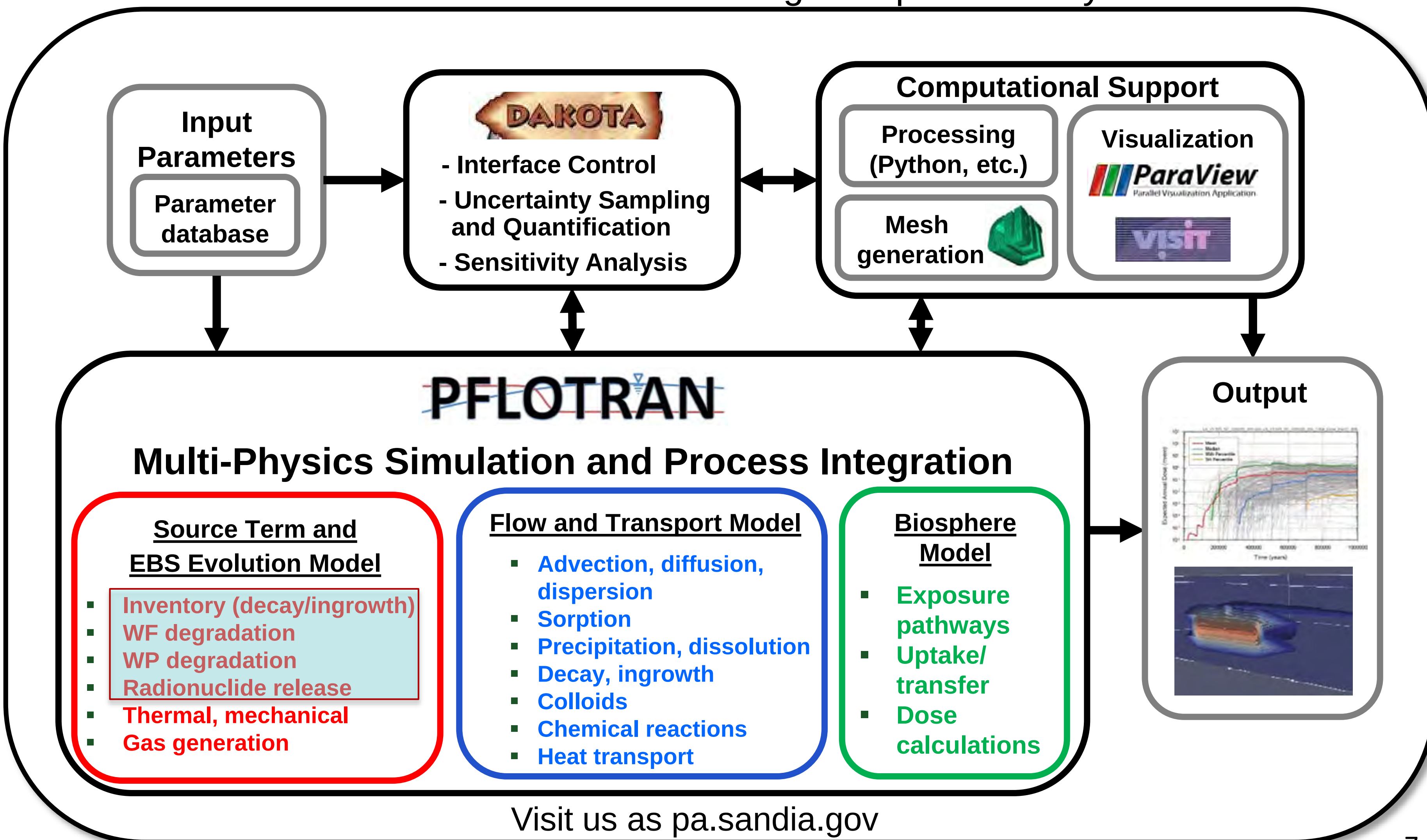




Visit us as pa.sandia.gov

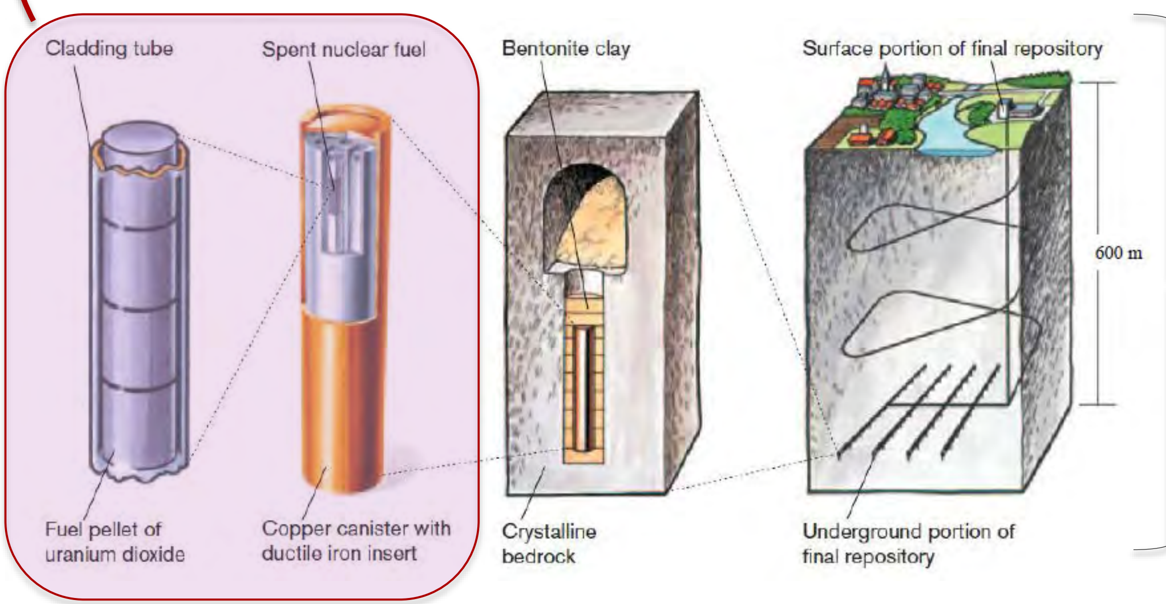
GDSA Framework

GDSA = Geologic Disposal Safety Assessment



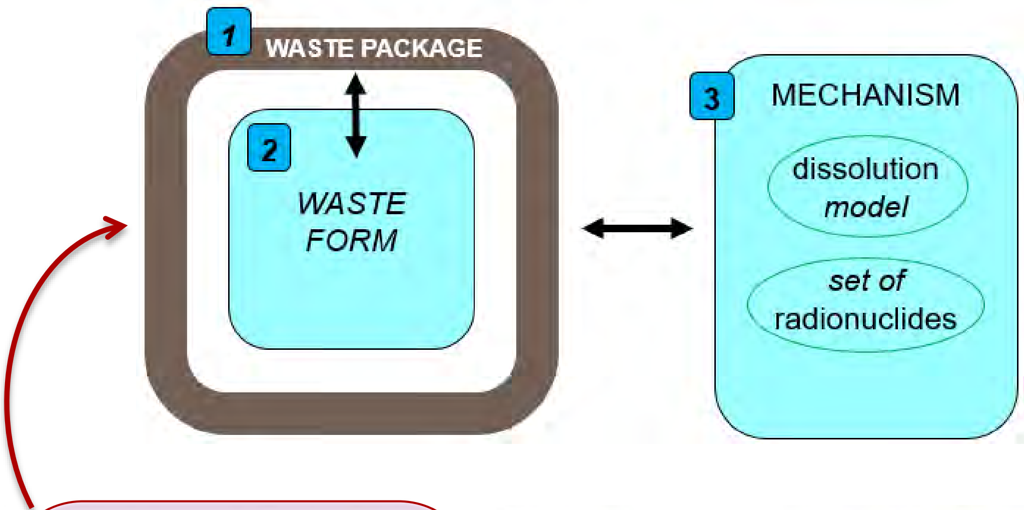
Modeling Waste Package/Form Degradation

- Waste package and waste form evolution involves:
  - Radionuclide decay and ingrowth
  - Waste package degradation processes and breach
  - Waste form dissolution



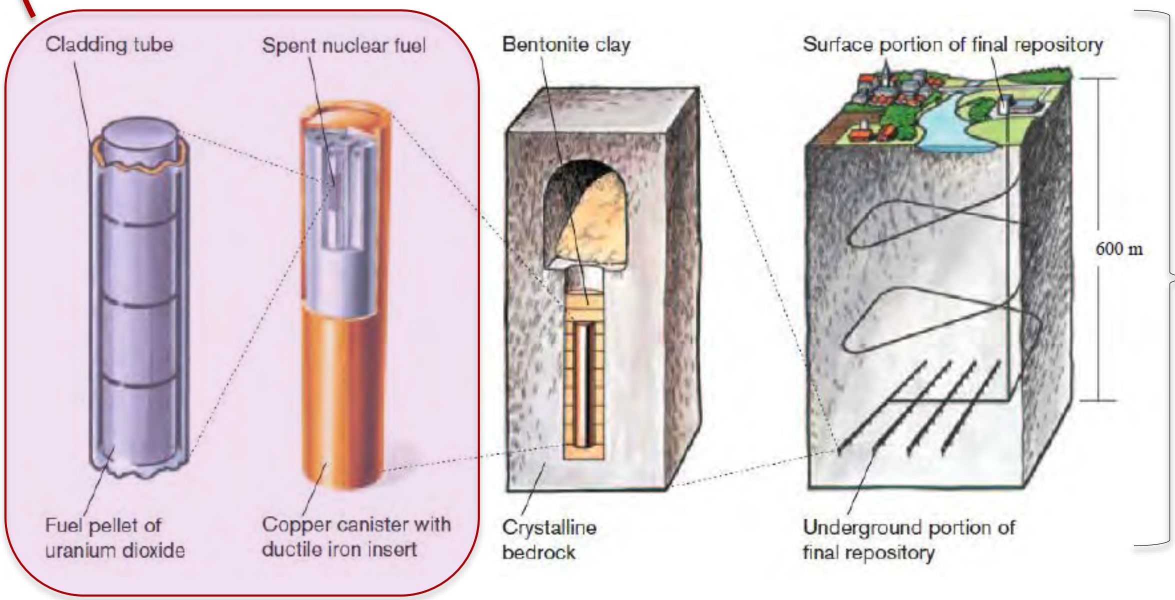
An example geologic repository in crystalline bedrock consists of a waste form within a copper canister that is surrounded by a bentonite clay 'buffer' and placed in the floor of mined drifts deep underground. The repository footprint at the surface is very small. Wang et al. (2014)

Modeling Waste Package/Form Degradation



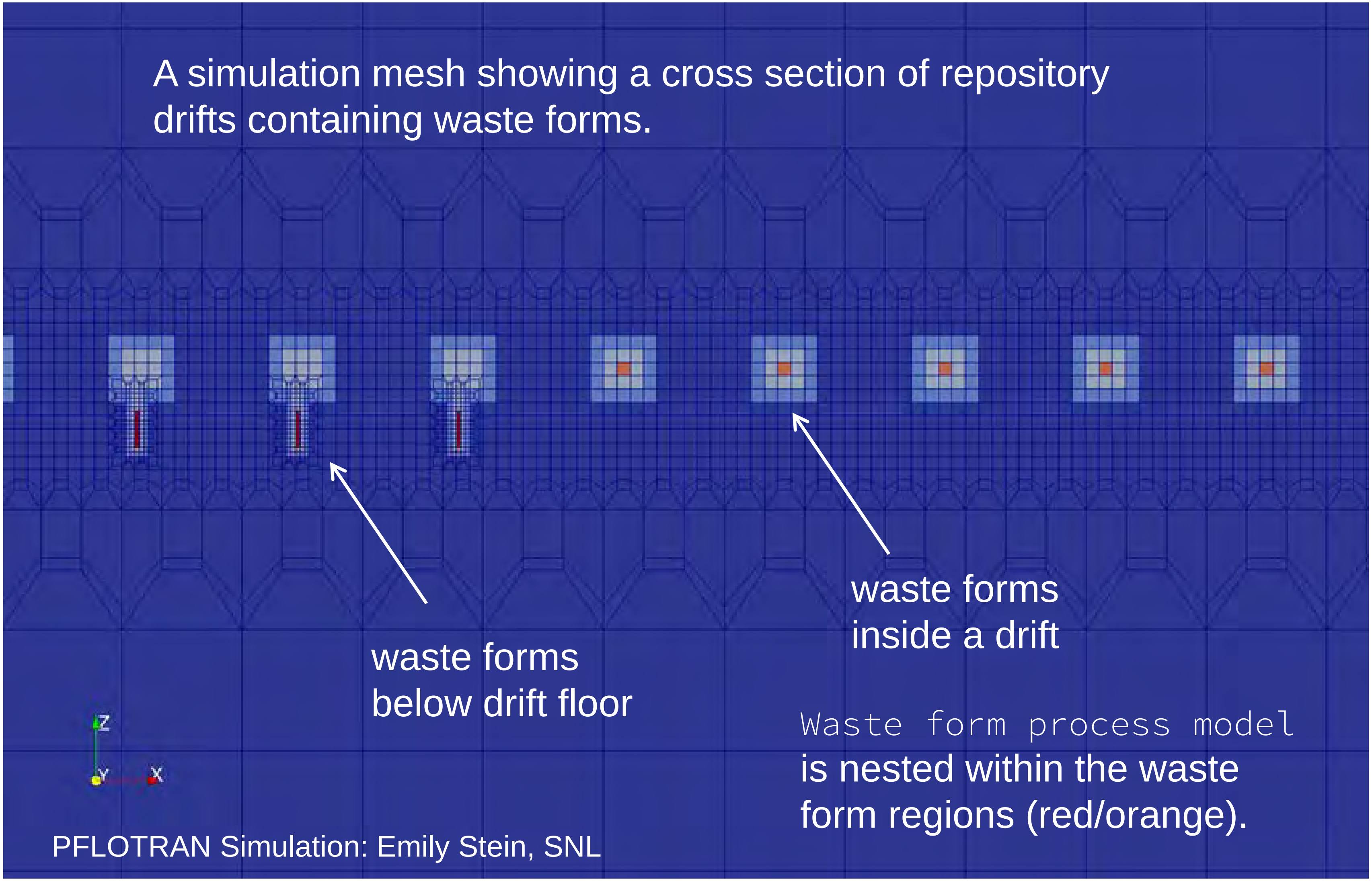
- We represent this in PFLOTRAN using a nested process model:

`waste form process model`

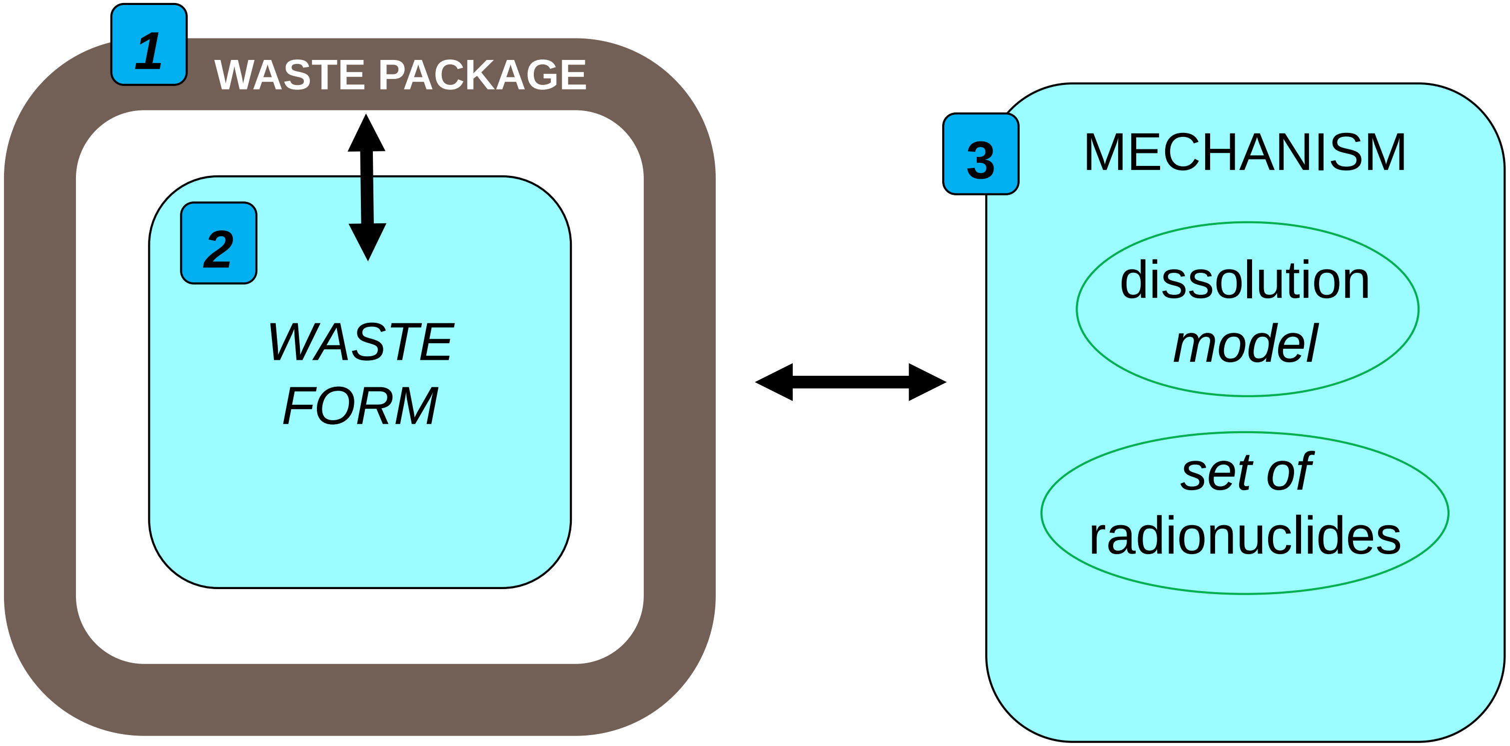


An example geologic repository in crystalline bedrock consists of a waste form within a copper canister that is surrounded by a bentonite clay 'buffer' and placed in the floor of mined drifts deep underground. The repository footprint at the surface is very small. Wang et al. (2014)

PFLOTRAN's Waste Form Process Model



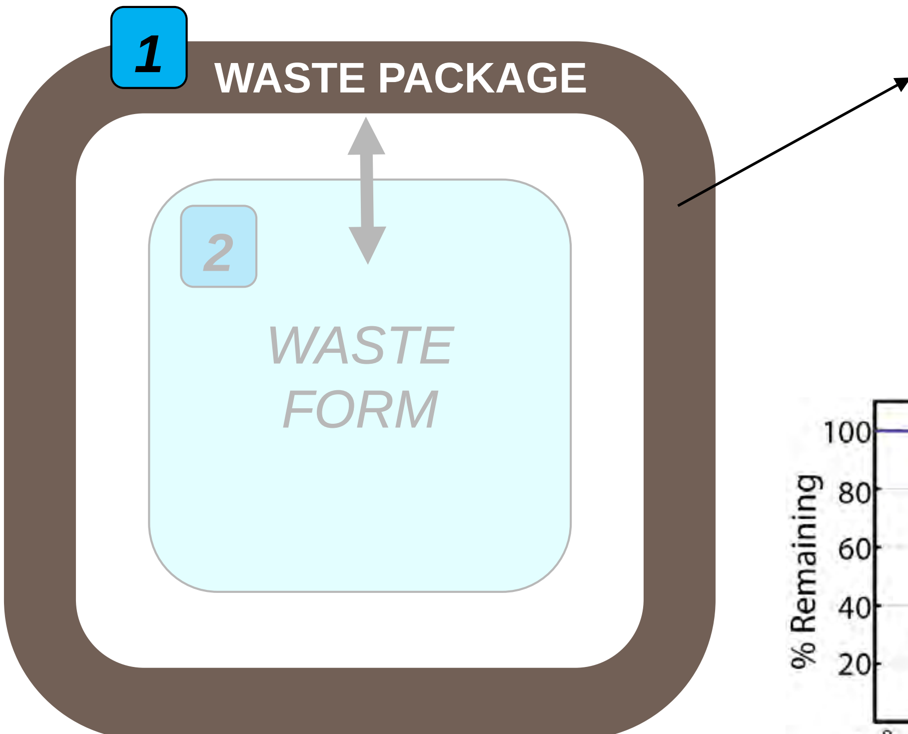
Consists of 3 Main Components:



"waste form object" "waste form type"

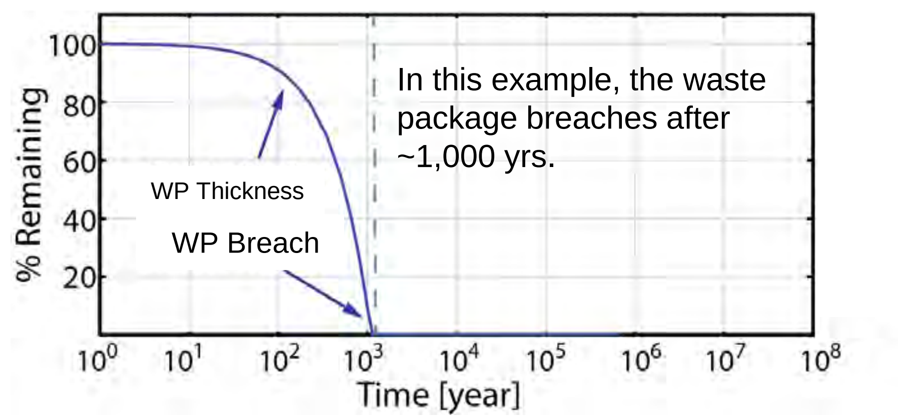
PFLOTRAN’s Waste Form Process Model

1. Waste Package Degradation Model

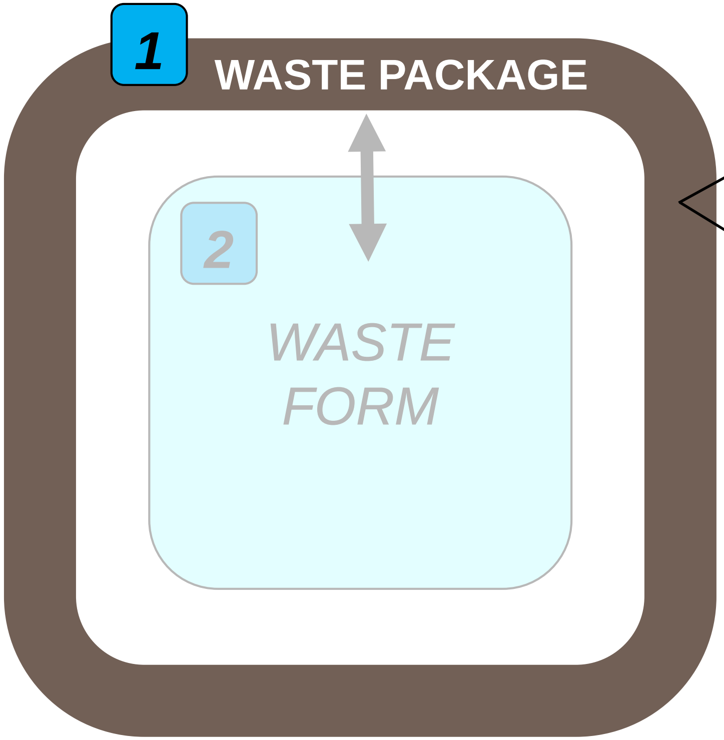


Remaining Canister Thickness

- A measure of how much ‘life’ the waste package has remaining
- Normalized waste package thickness
- Range: 100% - 0%
- Once thickness drops to 0%, the waste package breaches



1. Waste Package Degradation Model



Remaining Canister Thickness

- A measure of how much 'life' the waste package has remaining
- Normalized waste package thickness
- Range: 100% - 0%
- Once thickness drops to 0%, the waste package breaches

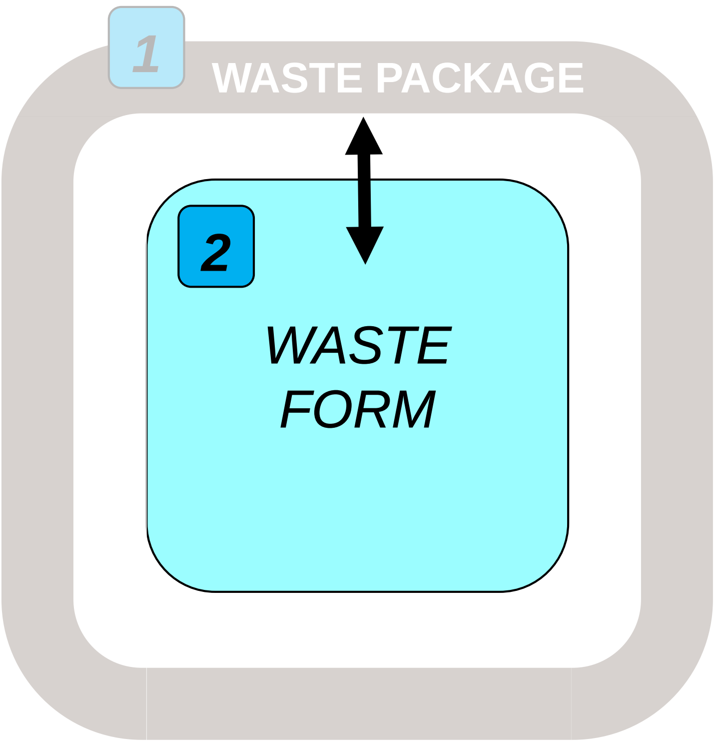
Waste Package Degradation Rate

- Rate at which waste package thickness decreases
- Unique to each waste form
- A base value is assigned via:
  - Directly as a user-provided value
  - 'Random' value from known distribution
- 'Effective' value is function of local conditions
- Provides a framework for future mechanistic processes for general corrosion

$$R_{eff} = R \cdot e^{\left[\frac{1}{60℃} - \frac{1}{T(x,t)}\right]}$$

PFLOTRAN's Waste Form Process Model

2. Waste Form Object



"waste form object"

Coordinate Point or Region
- Defines the location of a waste form
- Informs of local conditions (temperature, pressure, chemistry)

Volume
- Stores the remaining bulk volume
- Determines when source term "turns off"

'Mechanism' Pointer
- Points to the mechanism that defines the behavior of the waste form
- The mechanism determines the set of radioisotopes and the waste form dissolution equation

Radioisotope Concentrations
- Calculates isotope decay and ingrowth
- Stores isotope concentrations and mass fractions within the waste form

determines source term

Effective Dissolution Rate
- The rate of waste form dissolution after considering local conditions (temperature)
- Determines source term rate after breach

PFLOTRAN's Waste Form Process Model

All mechanisms can accommodate an "instant release fraction."

3. Waste Form Mechanism

Mechanism GLASS

- Waste form is a glass log, high level
- Dissolution equation (Kienzler et al. 2012): $R_g = 560e^{-7397/T(t,x)}$
- General dissolution equation:

$$R_g = K_0 10^{\eta pH} e^{-E_a/RT}(1 - Q/K)^{1/V} + K_{long}$$

Mechanism FMDM

- Waste form is used nuclear fuel (UO_2)
- Dissolution rate via Fuel Matrix Degradation Model

(J. Jerden *et al.*, Argonne National Lab)

Mechanism DSNF

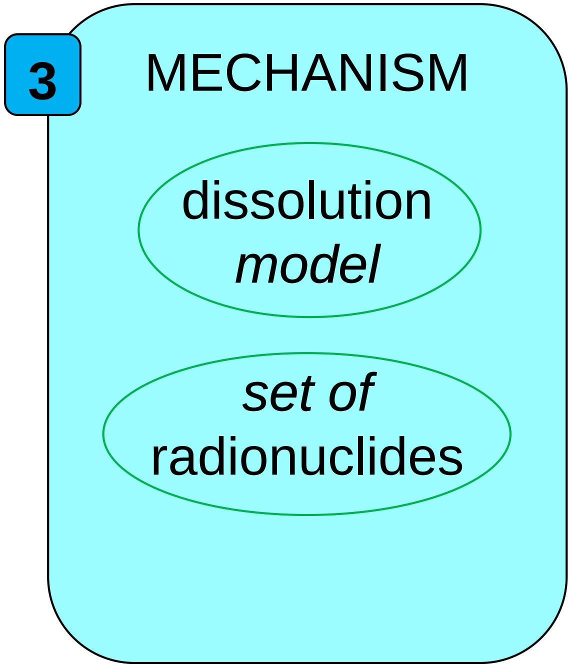
- Waste is defense-related spent nuclear fuel
- Dissolution rate "instantaneous" after w.p. breach

Mechanism WIPP

- Waste form is transuranic waste at WIPP
- Breach time and dissolution rate "instantaneous"

Mechanism CUSTOM

- User-defined specific surface area and dissolution rate
- Allows for flexibility if you need it



"waste form type"

PFLOTRAN's Waste Form Process Model

varying local physical and chemical conditions

waste package life (0-100%)

waste package degradation rate, R_{eff}

waste form 1
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

waste form 2
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

waste form 3
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

waste form 4
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

$$R_{eff} = R \cdot e^{\left[\frac{1}{60℃} - \frac{1}{T(x,t)}\right]}$$

PFLOTRAN's Waste Form Process Model

← varying local physical and chemical conditions →

waste package life (0-100%)

waste package degradation rate, R_{eff}

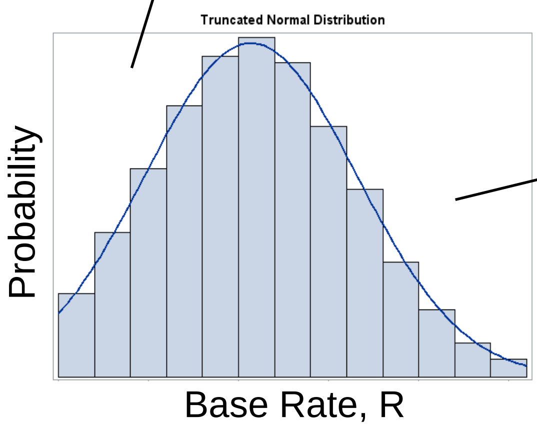
waste form 1
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

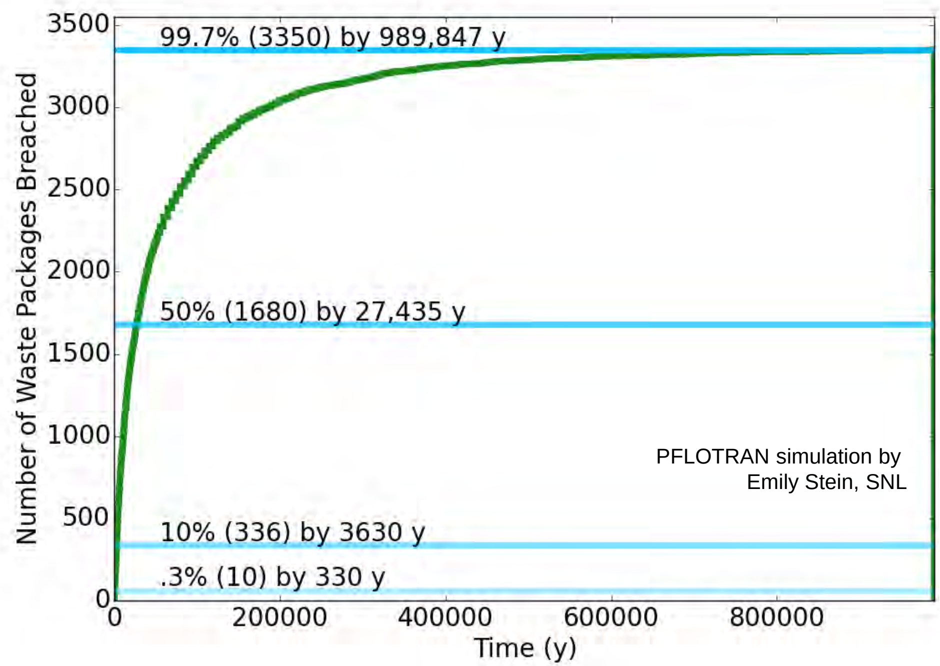
waste form 2
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

waste form 3
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

waste form 4
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

$$R_{eff} = R \cdot e^{\left[\frac{1}{60°C} - \frac{1}{T(x,t)}\right]}$$





PFLOTRAN's Waste Form Process Model

Sandia National Laboratories

← varying local physical and chemical conditions →

waste package life (0-100%)

waste package degradation rate, R_{eff}

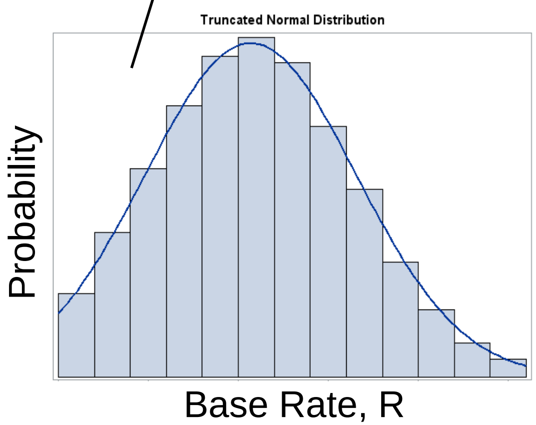
waste form 1
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

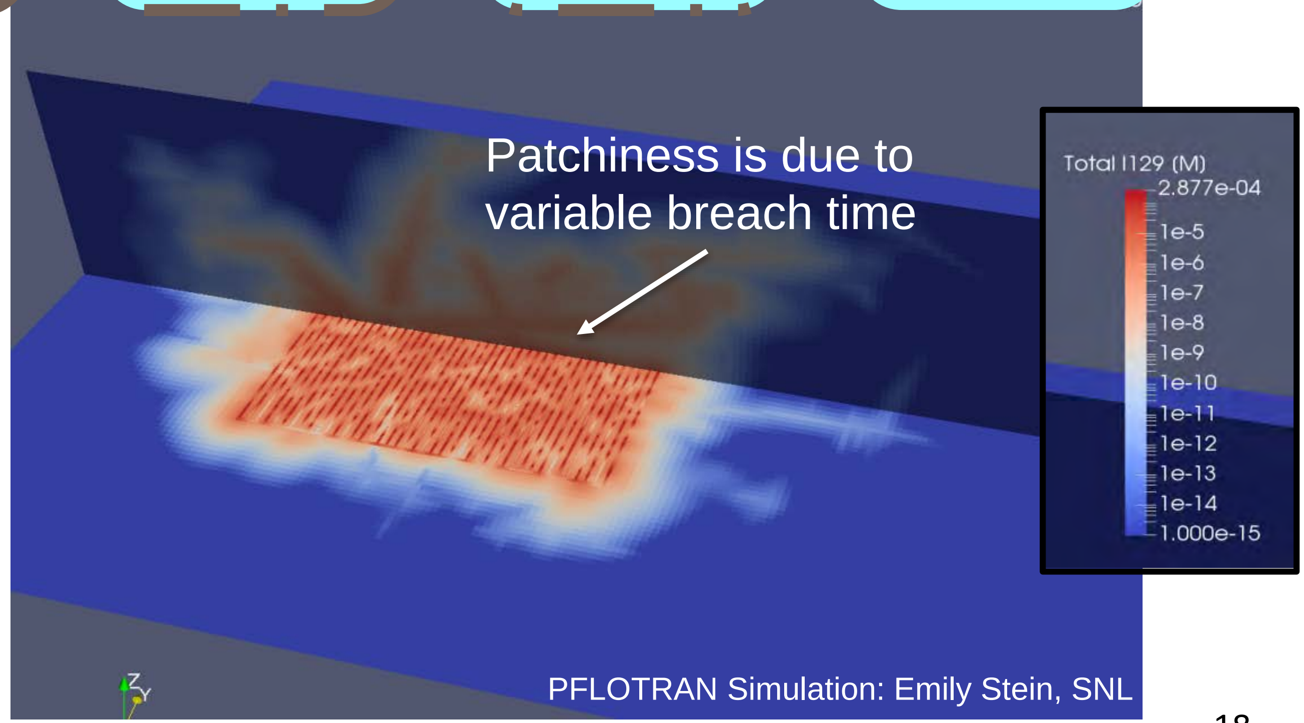
waste form 2
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

waste form 3
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

waste form 4
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

$$R_{eff} = R \cdot e^{\left[\frac{1}{60°C} - \frac{1}{T(x,t)}\right]}$$





PFLOTRAN's Waste Form Process Model

← varying local physical and chemical conditions →

waste package life (0-100%)

waste package degradation rate, R_{eff}

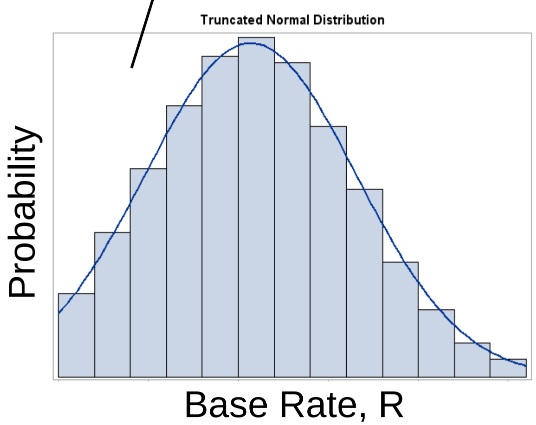
waste form 1
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

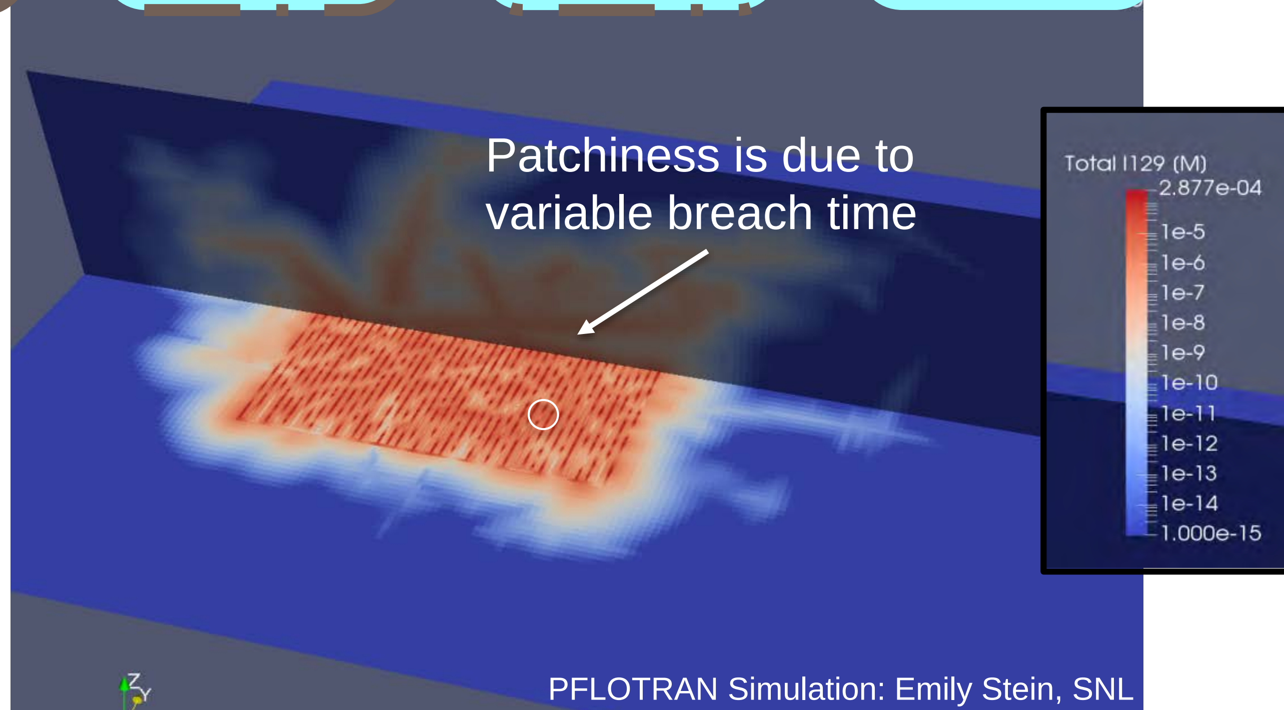
waste form 2
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

waste form 3
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

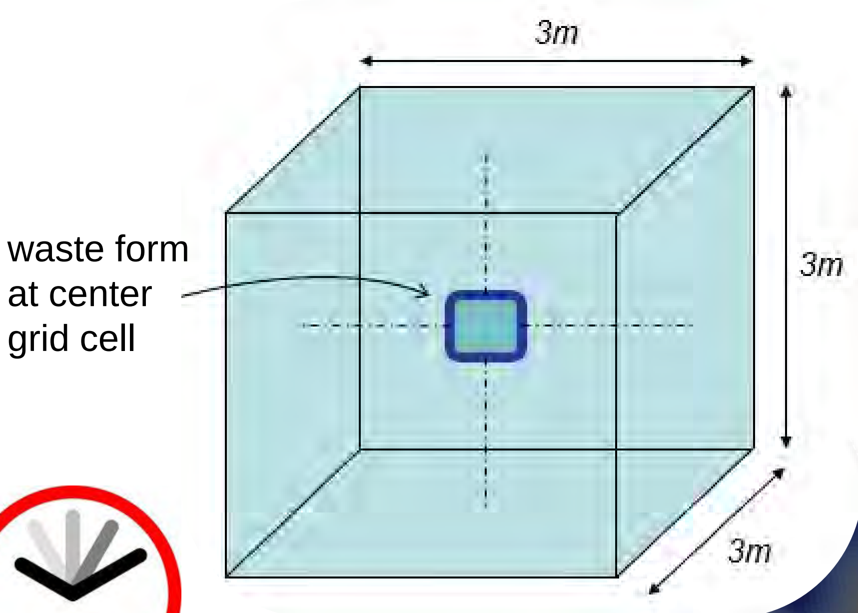
waste form 4
- region/coordinate
- RN concentrations
- eff. dissolution rate
- volume
- mech. pointer

$$R_{eff} = R \cdot e^{\left[\frac{1}{60°C} - \frac{1}{T(x,t)}\right]}$$





PFLOTRAN's Waste Form Process Model

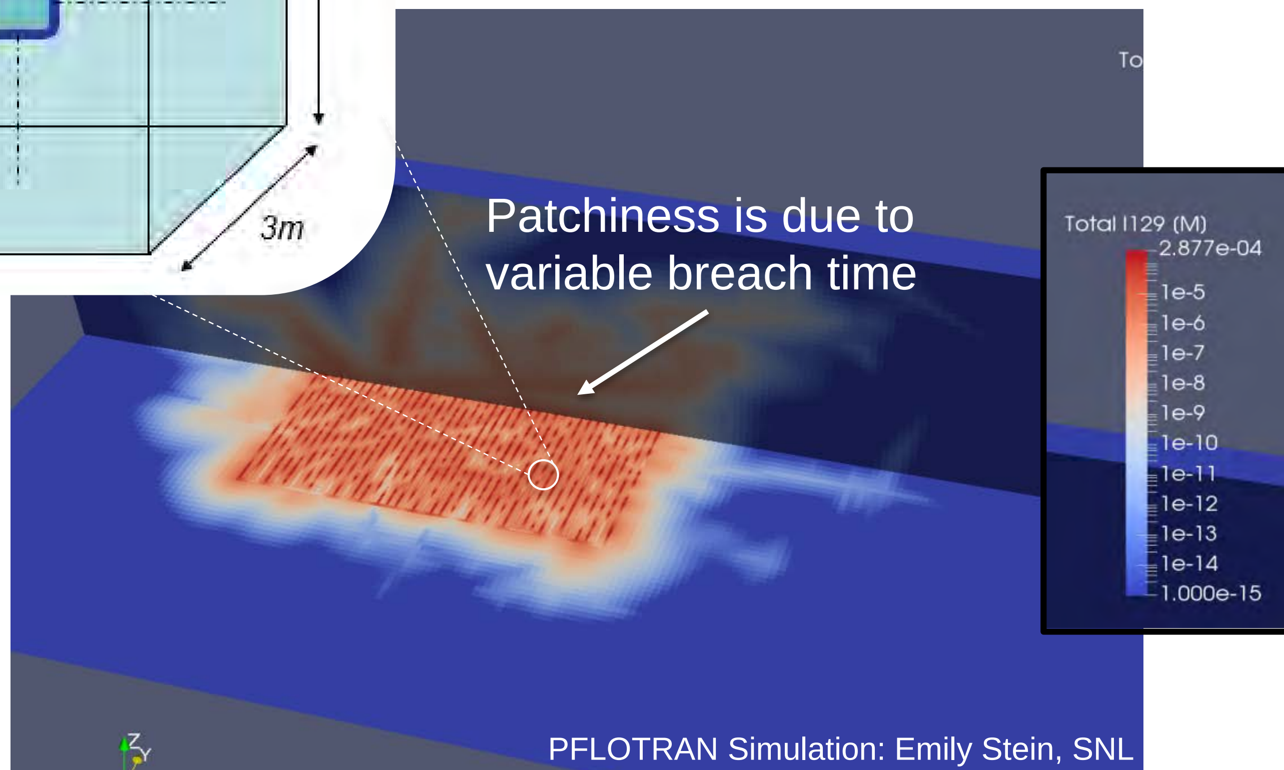


Mechanism GLASS

- Assumes waste form is a glass log type
- Dissolution equation (Kienzler et al. 2012):

$$R_g = 560e^{-7397/T(t,\mathbf{x})}$$

- no fluid flow
- no diffusive flux across boundaries
- 3x3x3 = 27 grid cells
- $1m^3$ grid cells



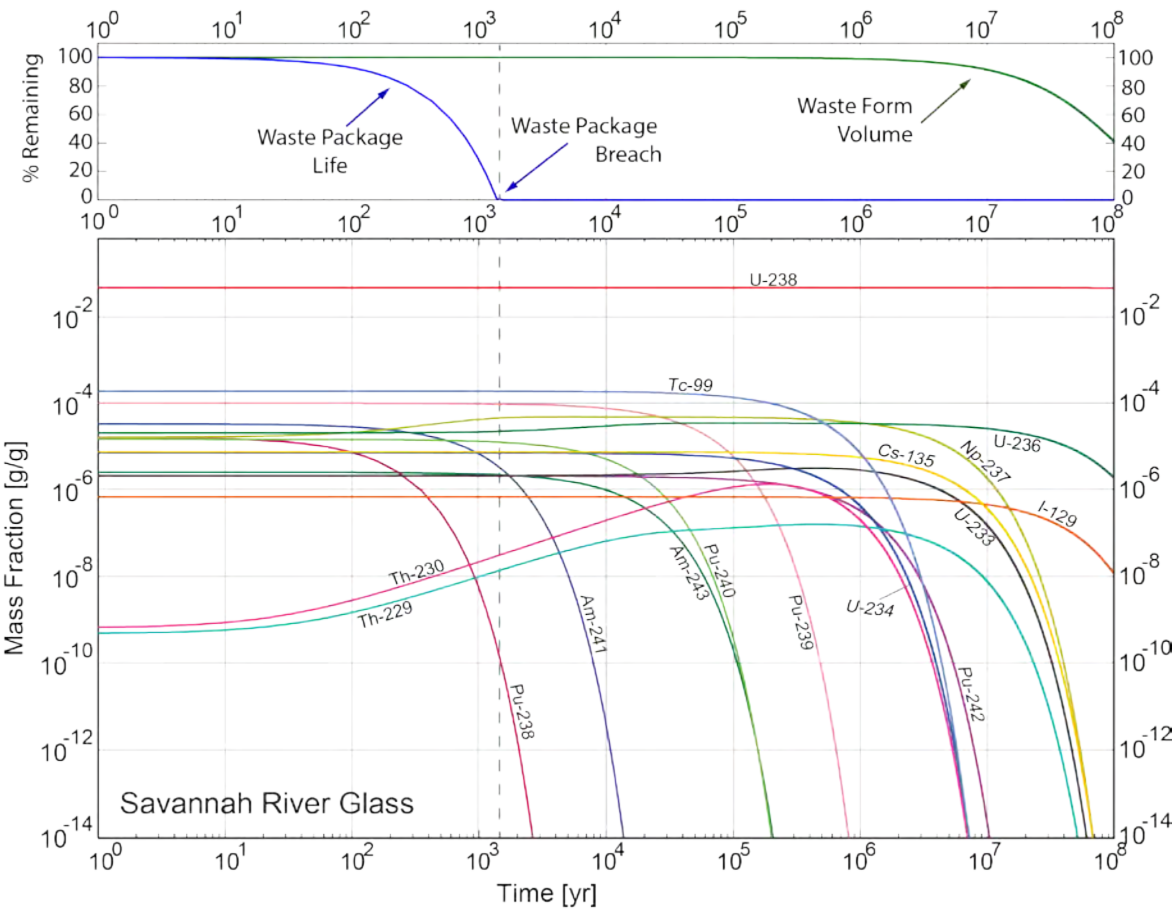
PFLOTRAN's Waste Form Process Model

- Waste package breach occurs at 1,000 yrs
- Waste form volume slowly decreases after breach

- Over time, the mass fractions of radionuclides evolve due to decay and ingrowth
- Mass fraction = g-RN/g-bulk

- The remaining mass fraction of each radionuclide and the glass dissolution rate determines its release rate

$$R_g = 560e^{-7397/T(t,x)}$$



Savannah River Glass Log Waste Form

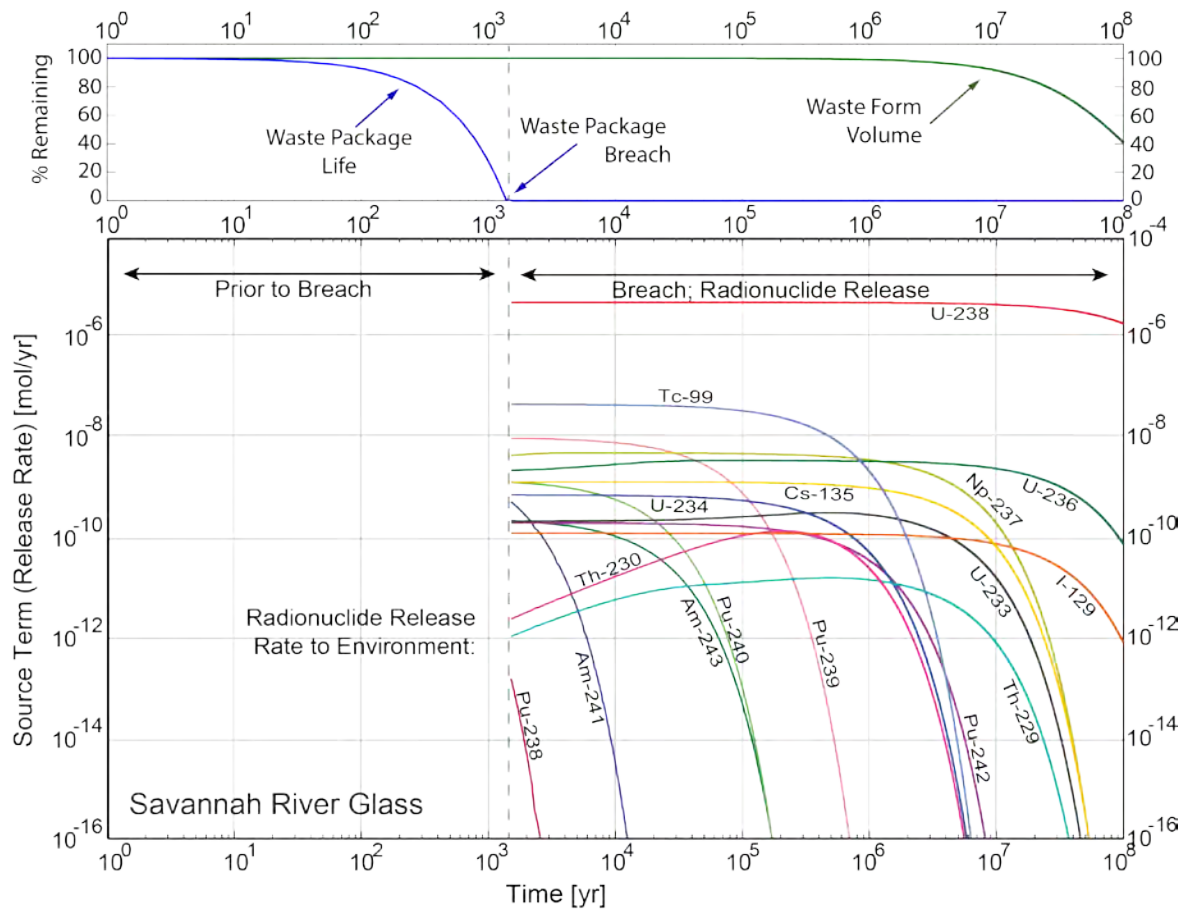
PFLOTRAN's Waste Form Process Model

- Waste package breach occurs at 1,000 yrs
- Waste form volume slowly decreases after breach

- Upon breach, radionuclides are released to the surroundings
- The source terms decrease over time, proportionally to the remaining inventory

- The remaining mass fraction of each radionuclide and the glass dissolution rate determines its release rate

$$R_g = 560e^{-7397/T(t,\mathbf{x})}$$

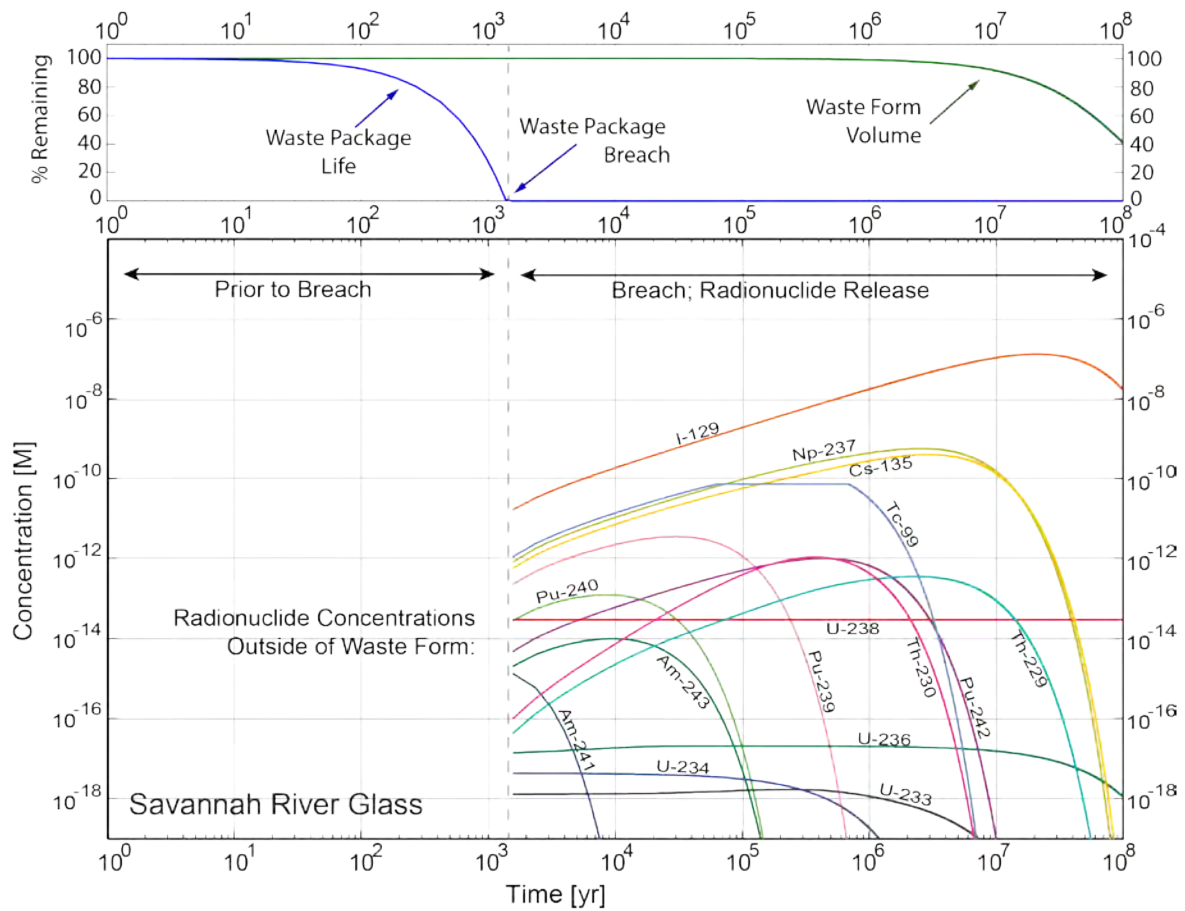


Savannah River Glass Log Waste Form

PFLOTRAN's Waste Form Process Model

- Waste package breach occurs at 1,000 yrs
- Waste form volume slowly decreases after breach

- The radionuclide concentrations outside of the waste form are influenced by:
  - Solubility
  - Sorption to host rock
  - Diffusion/advection
  - Decay and ingrowth

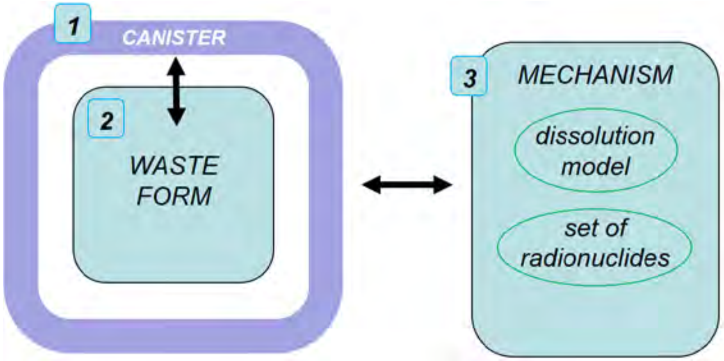


Savannah River Glass Log Waste Form

PFLOTRAN's Waste Form Process Model

Sandia National Laboratories

Future Development



PFLOTRAN

pflotran.org

GDSA Framework

Visit us as pa.sandia.gov

- **Waste form mechanisms:**
  - Add more mechanism types
  - Make dissolution models more mechanistic and interactive
- **Waste package degradation model:**
  - Include waste package degradation mechanisms like corrosion and damage models
- **PFLOTRAN's waste form process model is <u>open-source</u> and <u>modular</u>**
  - We invite collaboration to create new type of waste forms, mechanisms, etc.
  - We can work with you to get your functionality implemented

PFLOTRAN's Waste Form Process Model

Sandia National Laboratories

Check out the poster session for more applications!

PFLOTRAN

pflotran.org

GDSA Framework

Visit us as pa.sandia.gov

- *ISOTOPE PARTITIONING, DECAY, AND INGROWTH ACROSS MULTIPLE PHASES IN PFLOTRAN CODE OF GDSA FRAMEWORK*, P.E. Mariner, G.E. Hammond, and J.M. Frederick
- *RADON AND THE PFLOTRAN INGESTION DOSE MODEL OF GDSA FRAMEWORK*, P.E. Mariner, J.M. Frederick, and G.E. Hammond
- *ESTIMATING THE EFFECT OF FRACTURE CONNECTIVITY ON WASTE ISOLATION USING A HIGH-PERFORMANCE REACTIVE TRANSPORT SIMULATOR, PFLOTRAN*, D. Sevougian, E. R. Stein, G. E. Hammond, P. E. Mariner, and J. M. Frederick
- *MULTI-SCALE MODELING IN PFLOTRAN FOR GEOLOGIC REPOSITORY PERFORMANCE ASSESSMENT: AN ENHANCEMENT TO GDSA FRAMEWORK*, E. R. Stein, G. E. Hammond, and C. F. Jové Colón
- *PFLOTRAN REACTION SANDBOX: A FLEXIBLE, EXTENSIBLE FRAMEWORK FOR VETTING BIOGEOCHEMICAL REACTIONS WITHIN AN OPEN SOURCE SUBSURFACE SIMULATOR*, G. E. Hammond