Used Fuel Disposition Campaign

Radioisotope Source Term Degradation and Implementation in PFLOTRAN

NOT reviewed and approved for release outside of Sandia National Laboratories

Jennifer M. Frederick & Glenn E. Hammond
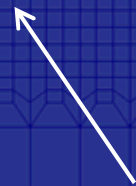
Sandia National Laboratories

2016 UFDC Annual Working Group Meeting

GDSA Session, June 8, 2016

Las Vegas, NV

PFLOTRAN's Waste Form Process Model

*Advances in PFLOTRAN's implementation of
the radioisotope source term due to degrading waste forms*

*waste forms
inside a drift*

*waste forms
below drift floor*

PFLOTRAN Simulation: Emily Stein, SNL

June 8, 2016

PFLOTRAN's Waste Form Process Model

- **The Waste Form Process Model is used to:**
  - track radioisotope decay and ingrowth inside the waste form
  - couple with a canister degradation model to determine breach time
  - track waste form dissolution and remaining volume
  - determine radionuclide source term to environment

Implemented in: **PFLOTRAN**
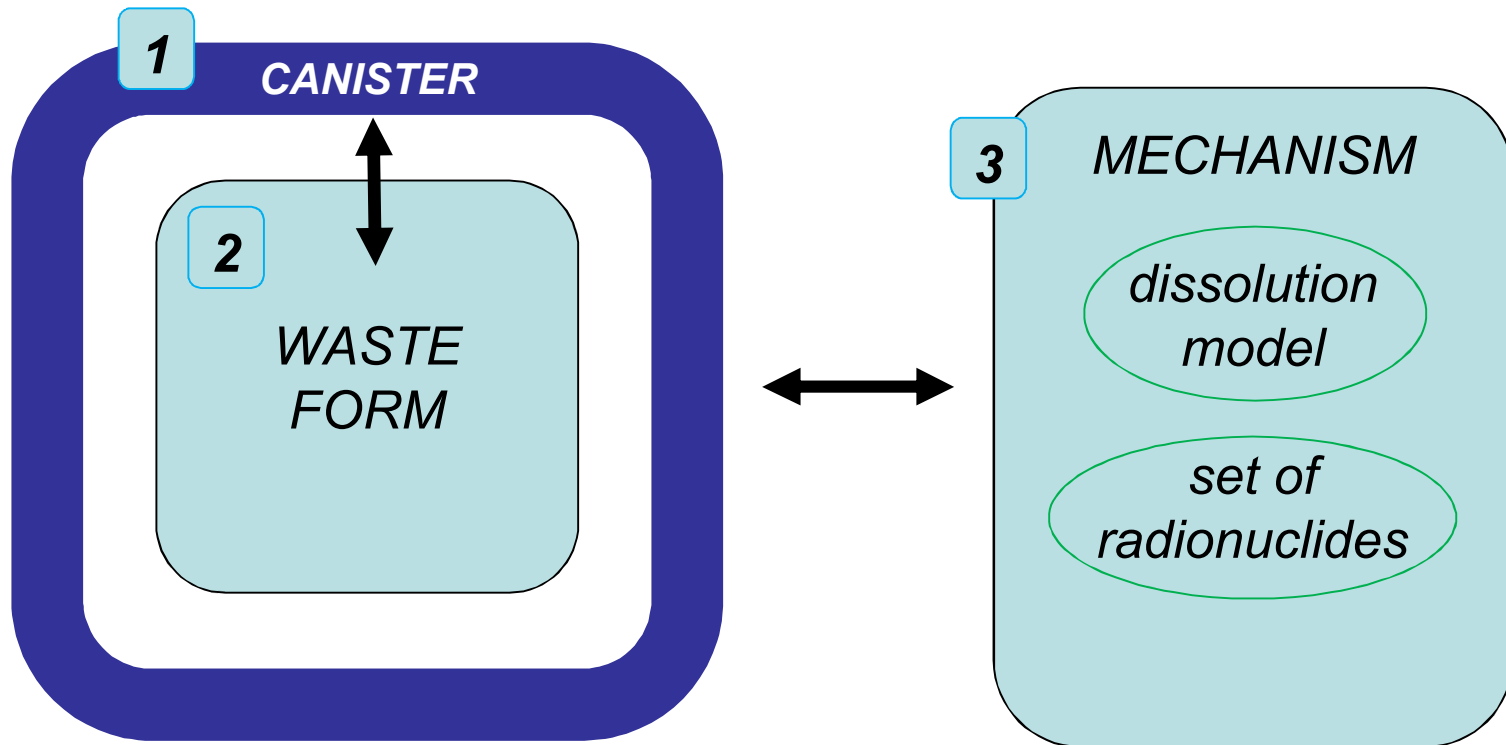
- **Development Team:**
  - Jennifer M. Frederick (Sandia National Laboratories)
  - Glenn E. Hammond (Sandia National Laboratories)
  - Paul Mariner (Sandia National Laboratories)
- **Major restructuring in FY16 now allows:**
  - variable canister breach time
  - multiple waste form "types" can be run in a simulation
  - instantaneous release fraction upon canister breach

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PFLOTRAN's Waste Form Process Model

Consists of 3 Main Components:

1 *CANISTER*

2 *WASTE FORM*

3 *MECHANISM*

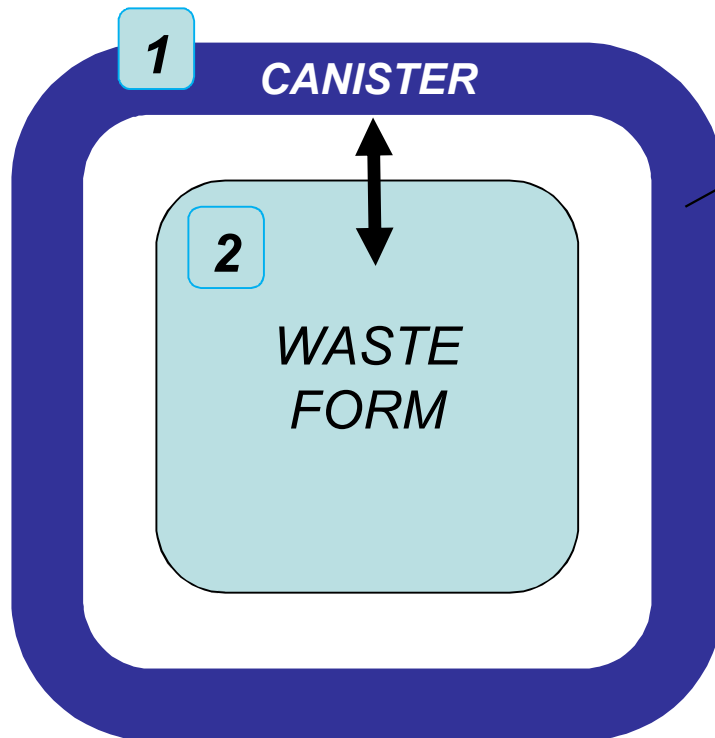*dissolution model*

set of radionuclides

"waste form object"
"fruit"

"waste form type"
"banana, apple, orange, etc."

PFLOTRAN's Waste Form Process Model

1. Canister Degradation Model

CANISTER

1

2

WASTE FORM

"waste form object"
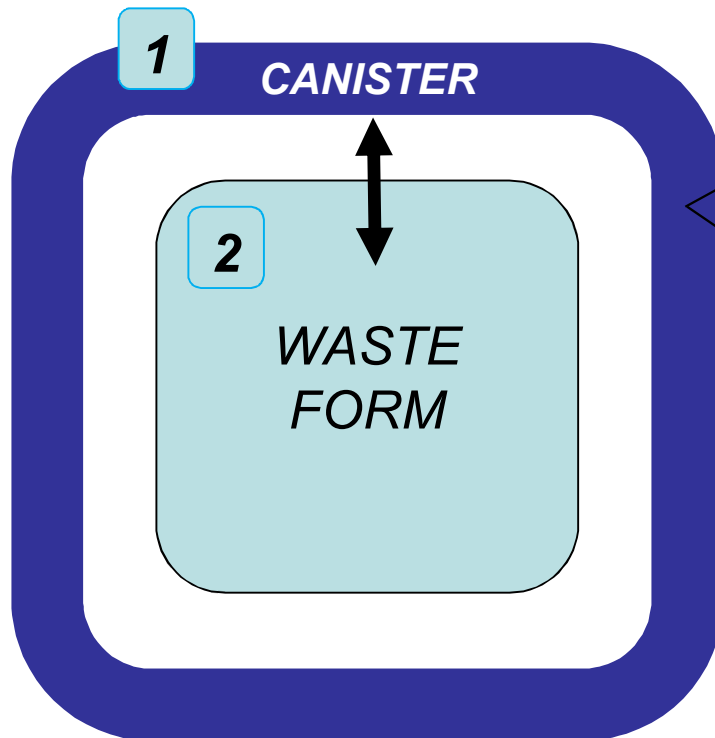*"fruit"*

Canister Vitality
- *A measure of how much 'life' the canister has remaining*
- *Range: 100% - 0%*
- *Once vitality drops to 0%, the waste form canister breaches*

In this example, the canister breaches after ~1,000 yrs.

PFLOTRAN's Waste Form Process Model

1. Canister Degradation Model

CANISTER

WASTE FORM

1

2

"waste form object"
"fruit"

Canister Vitality
- *A measure of how much 'life' the canister has remaining*
- *Range: 100% - 0%*
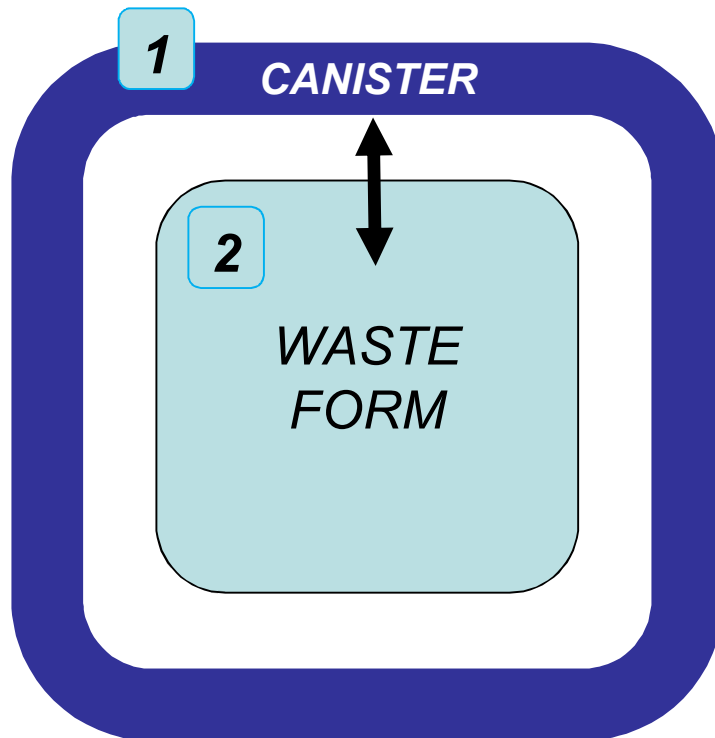- *Once vitality drops to 0%, the waste form canister breaches*

Canister Degradation Rate
- *The rate at which canister vitality 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 that can control vitality degradation*

PFLOTRAN's Waste Form Process Model

2. Waste Form

1 CANISTER

2 WASTE FORM

"waste form object"
"fruit"

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

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

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

determines source term

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

Mechanism Pointer
- *Points to the mechanism that defines the type of waste form*
- *The mechanism determines the set of radionuclides and the waste form dissolution equation*

PFLOTRAN's Waste Form Process Model

3. Waste Form Mechanism

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

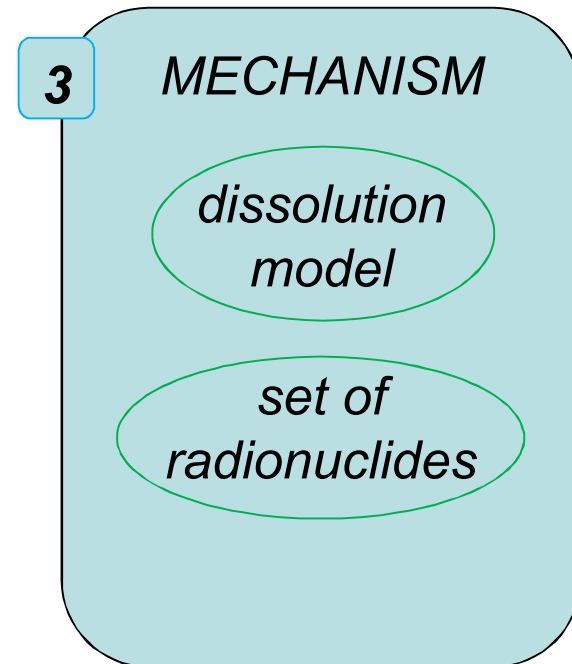$$r(T) = 560e^{\frac{-7397}{T(t,\bar{x})}}$$

Mechanism FMDM
- Assumes waste form is used nuclear fuel (UO_2)
- Dissolution rate via Fuel Matrix Degradation Model
  (J. Jerden et al, Argonne National Lab)

Mechanism DSNF
- Assumes waste form DOE spent nuclear fuel
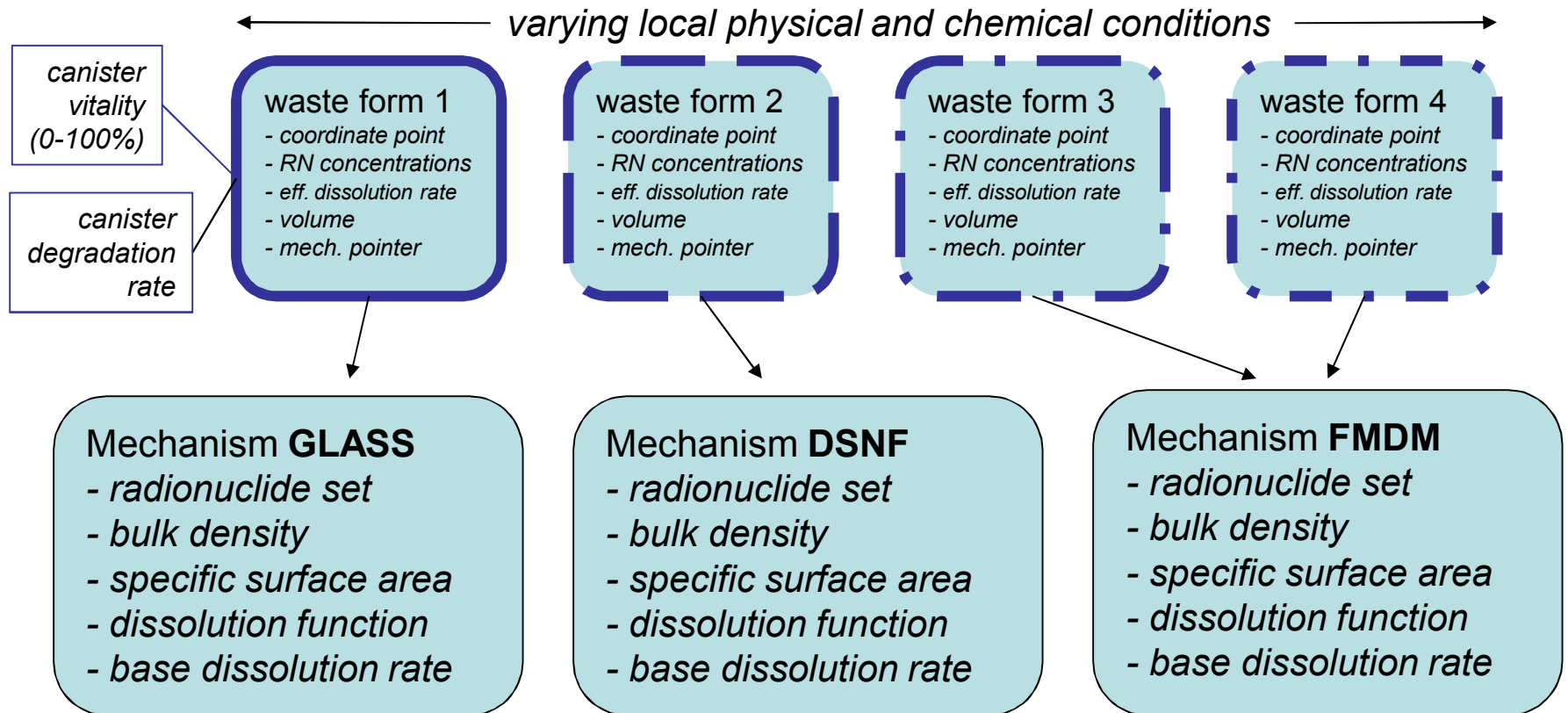- Dissolution rate "instantaneous" after canister breach

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

3   *MECHANISM*

dissolution model

set of radionuclides

"waste form type"
"banana, apple, orange, etc."

PFLOTRAN's Waste Form Process Model

varying local physical and chemical conditions

canister vitality (0-100%)

canister degradation rate

waste form 1
- *coordinate point*
- *RN concentrations*
- *eff. dissolution rate*
- *volume*
- *mech. pointer*

waste form 2
- *coordinate point*
- *RN concentrations*
- *eff. dissolution rate*
- *volume*
- *mech. pointer*

waste form 3
- *coordinate point*
- *RN concentrations*
- *eff. dissolution rate*
- *volume*
- *mech. pointer*

waste form 4
- *coordinate point*
- *RN concentrations*
- *eff. dissolution rate*
- *volume*
- *mech. pointer*

Mechanism **GLASS**
- *radionuclide set*
- *bulk density*
- *specific surface area*
- *dissolution function*
- *base dissolution rate*

Mechanism **DSNF**
- *radionuclide set*
- *bulk density*
- *specific surface area*
- *dissolution function*
- *base dissolution rate*

Mechanism **FMDM**
- *radionuclide set*
- *bulk density*
- *specific surface area*
- *dissolution function*
- *base dissolution rate*

Numerically efficient and modular design
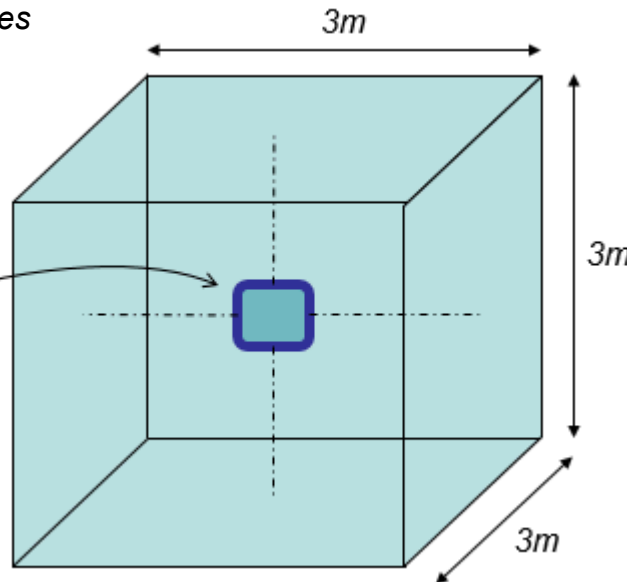
PFLOTRAN's Waste Form Process Model

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

waste form at center grid cell

10^8 yrs

Example portion of PFLOTRAN input file:

```
#========================= waste forms =========================
WASTE_FORM_GENERAL

  WASTE_FORM
    COORDINATE 1.5d0 1.5d0 1.5d0
    VOLUME 1.0d0 m^3
    MECHANISM_NAME fmdm_60
    CANISTER_VITALITY_RATE 0.0000042 1/day
  /

  MECHANISM FMDM
    NAME fmdm_60
    SPECIFIC_SURFACE_AREA 0.001 m^2/g
    MATRIX_DENSITY 10970. kg/m^3 #calculated assuming 100% UO2
    BURNUP 60 # MWd/MTHM
    SPECIES
    #name,    MW[g/mol],dcy[1/s], initMF,    inst_rel_frac,daughter
    Am-241  241.06d0  5.08d-11  8.7000d-4  0.0d0  Np-237
    Am-243  243.06d0  2.98d-12  1.8800d-4  0.0d0  Pu-239
    Pu-238  238.05d0  2.56d-10  3.4200d-4  0.0d0   U-243
    Pu-239  239.05d0  9.01d-13  5.1500d-3  0.0d0  #U-235 (not tracking)
    Pu-240  240.05d0  3.34d-12  2.8400d-3  0.0d0   U-236
    Pu-242  242.06d0  5.80d-14  5.6800d-4  0.0d0   U-238
    Np-237  237.05d0  1.03d-14  8.5900d-4  0.0d0   U-233
     U-233  233.04d0  1.38d-13  9.7000d-9  0.0d0  Th-229
     U-234  234.04d0  8.90d-14  2.1200d-4  0.0d0  Th-230
     U-236  236.05d0  9.20d-16  4.3300d-3  0.0d0  #Th-232 (not tracking)
     U-238  238.05d0  4.87d-18  6.3200d-1  0.0d0  #Th-234 (not tracking)
    Th-229  229.03d0  2.78d-12  4.4300d-12 0.0d0  #Ra-225 (not tracking)
    Th-230  230.03d0  2.75d-13  1.5800d-8  0.0d0  #Ra-226 (not tracking)
     I-129  128.90d0  1.29d-15  2.1800d-4  0.0d0  #Xe-129 (not tracking)
    Tc-99    98.91d0  1.04d-13  8.8700d-4  0.0d0  #Ru-99  (not tracking)
    /
    CANISTER_DEGRADATION_MODEL
      #VITALITY_LOG10_MEAN -4.5
      #VITALITY_LOG10_STDEV 0.50
      #VITALITY_UPPER_TRUNCATION -3.0
      CANISTER_MATERIAL_CONSTANT 1500.0
    /
  /

  PRINT_MASS_BALANCE

END
```
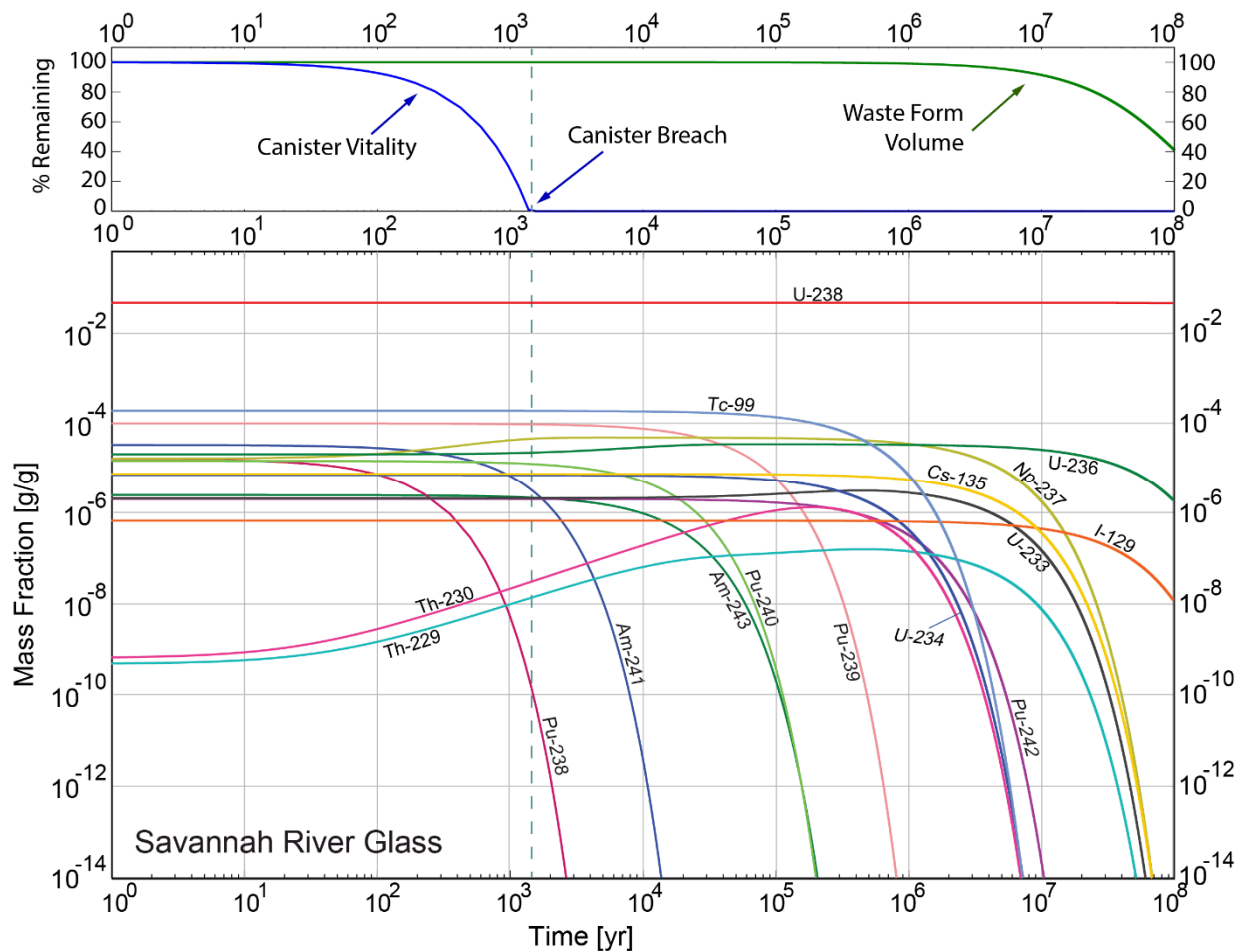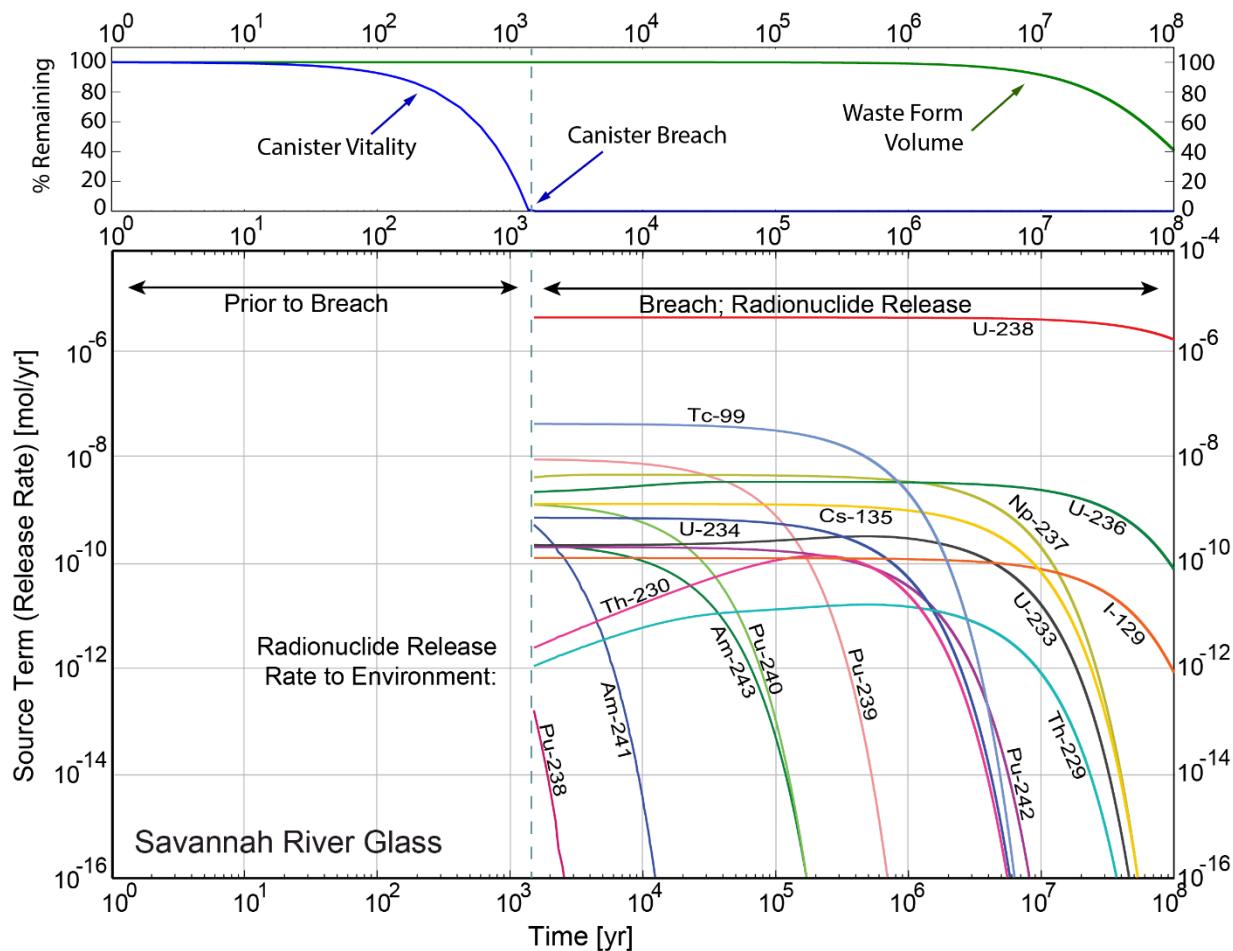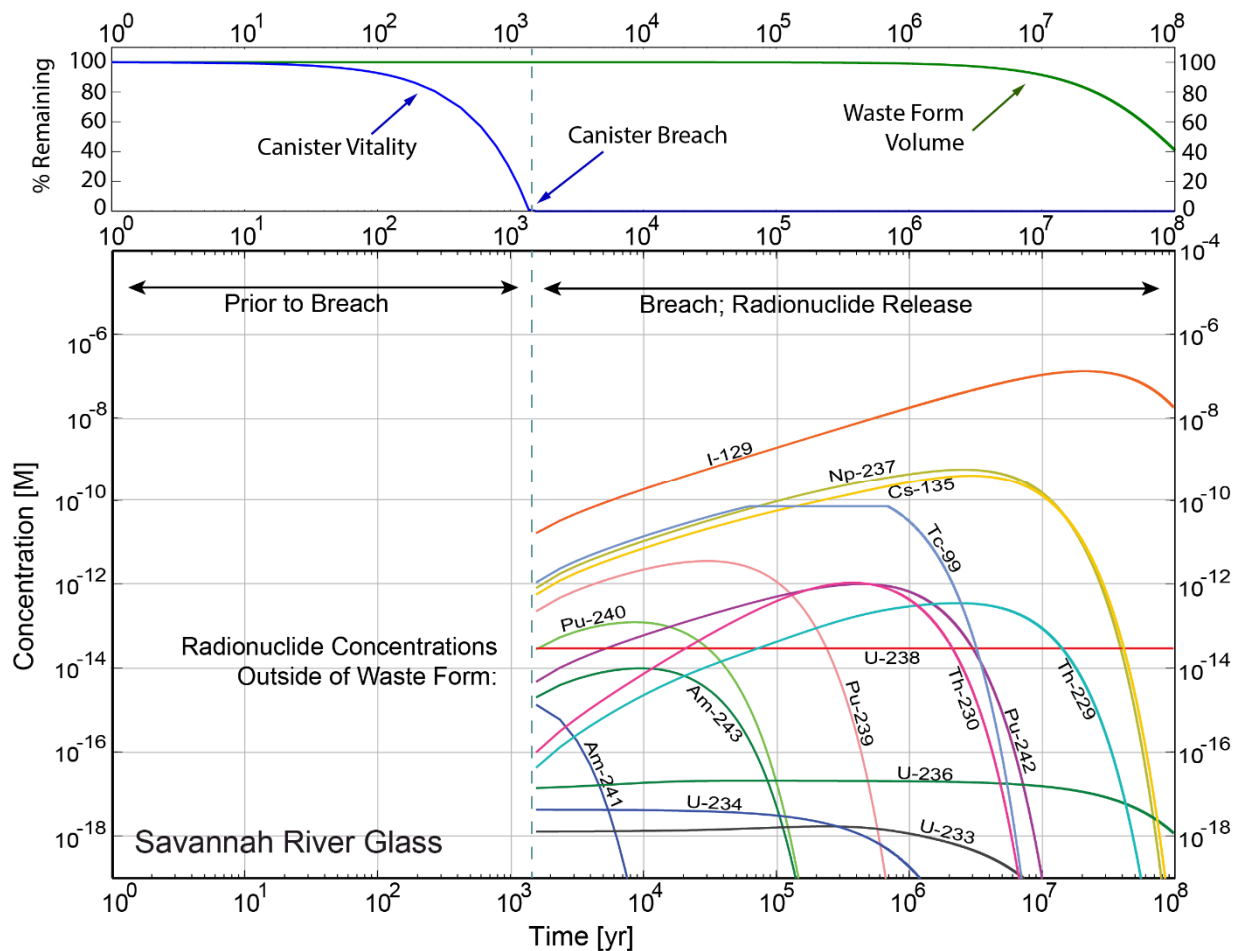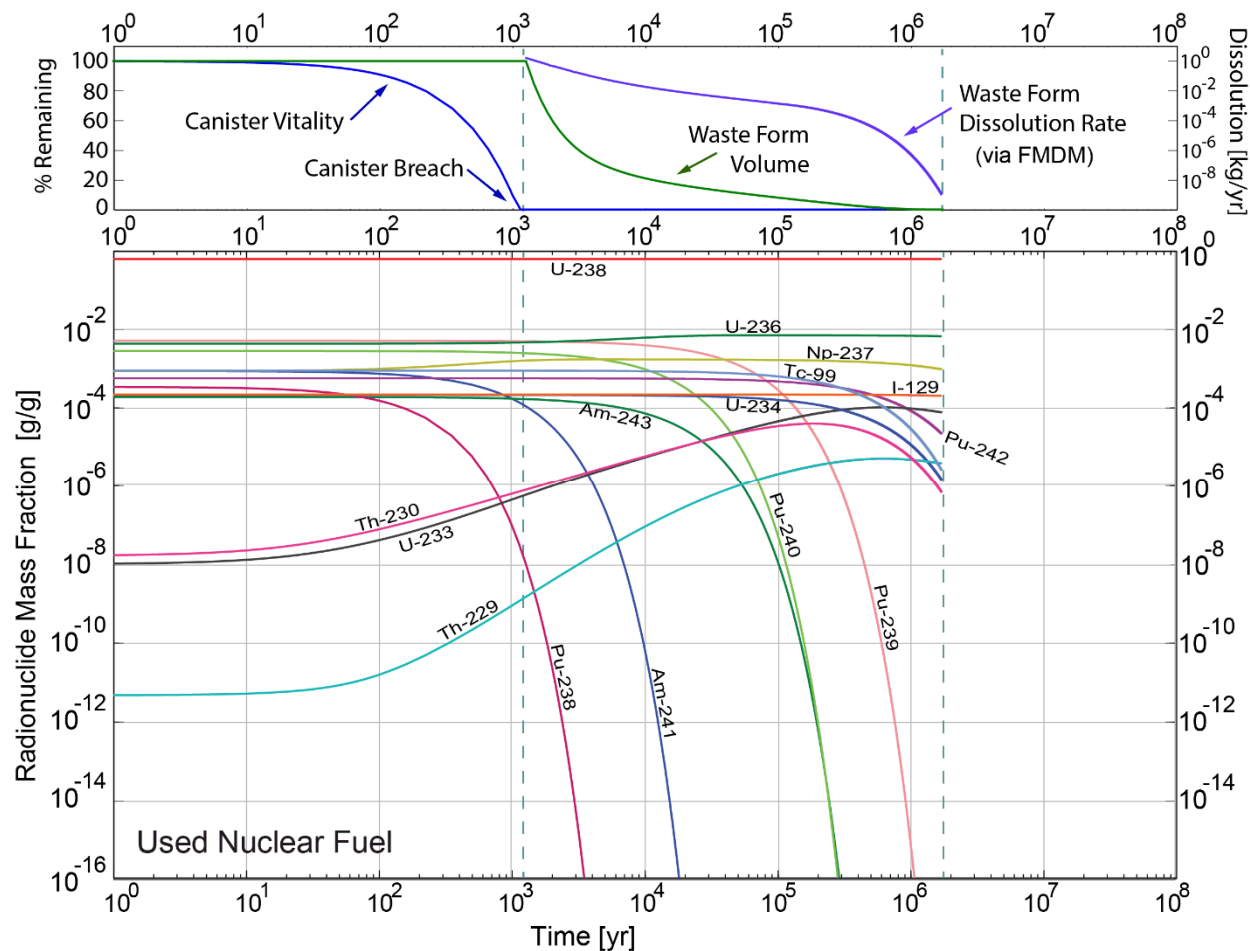
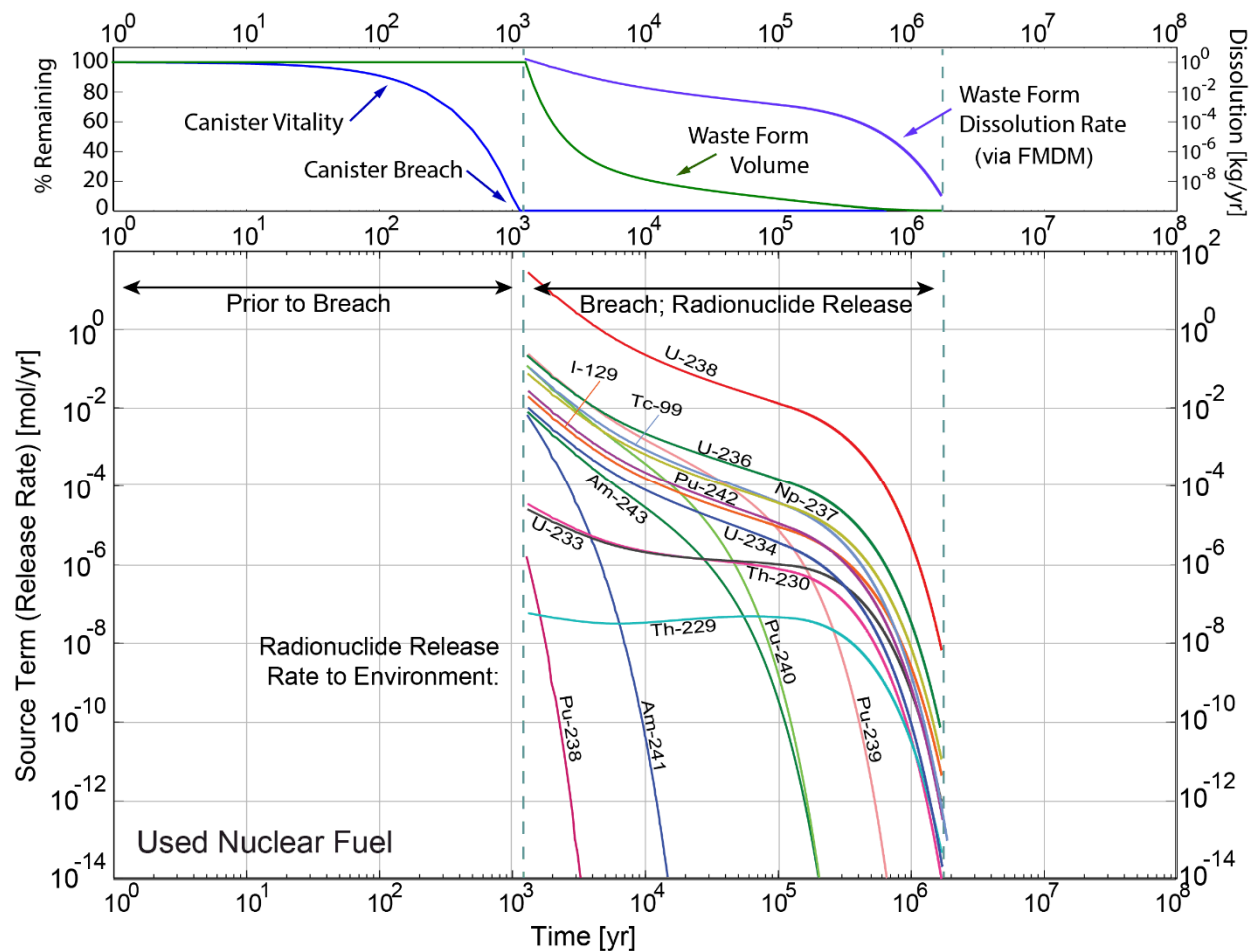# Savannah River Glass

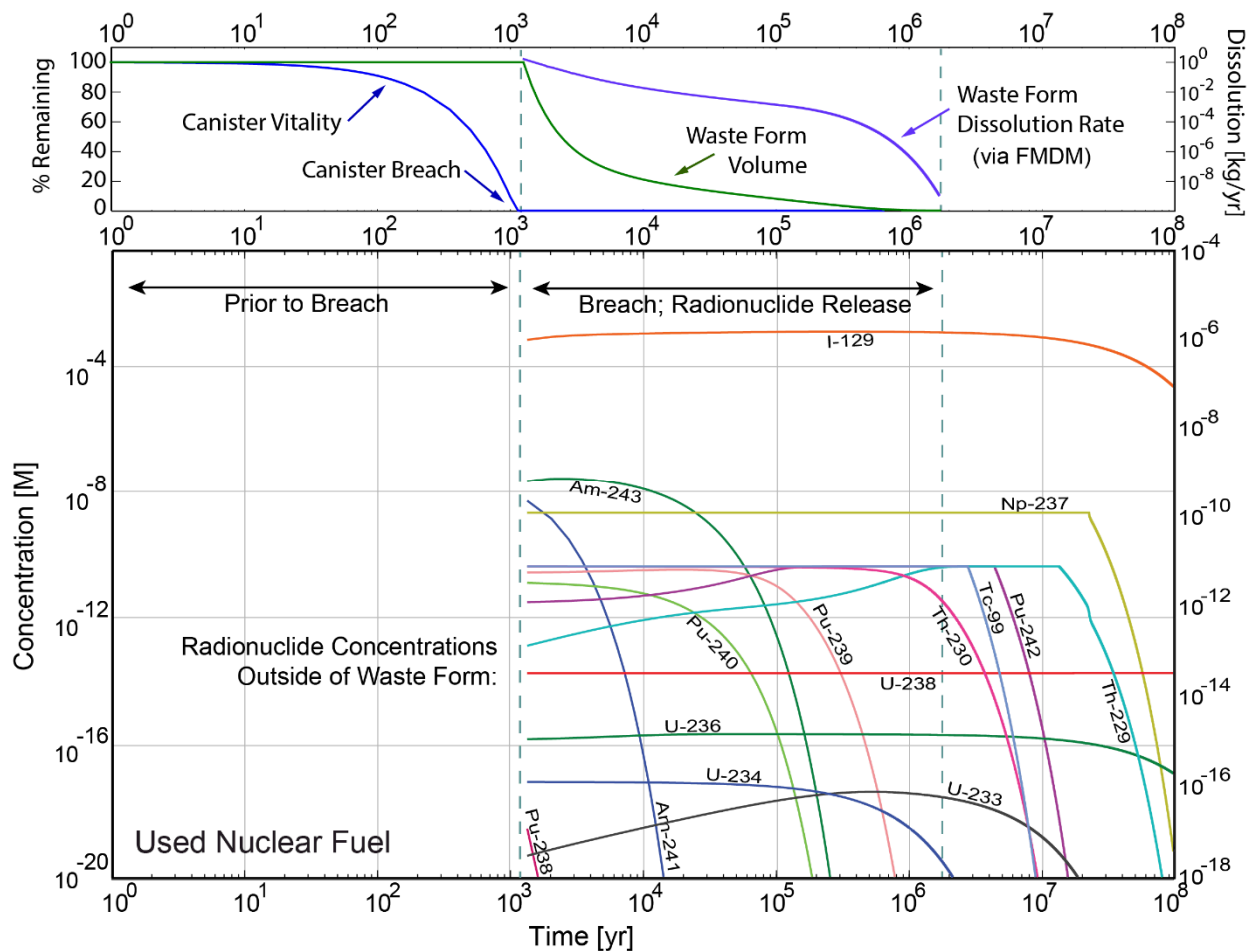# Savannah River Glass

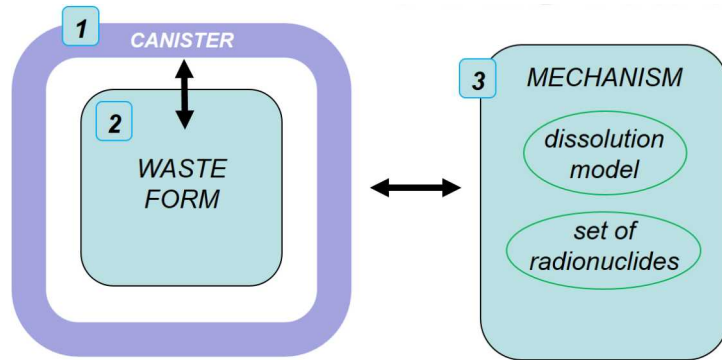# Savannah River Glass

Used Nuclear Fuel (FMDM)

Used Nuclear Fuel (FMDM)

Used Nuclear Fuel (FMDM)

PFLOTRAN's Waste Form Process Model

Future Development

PFLOTRAN

pflotran.org

- **Waste form mechanisms:**
  - *Add more mechanism types*
  - *Make dissolution models more mechanistic and interactive*

- **Canister degradation model:**
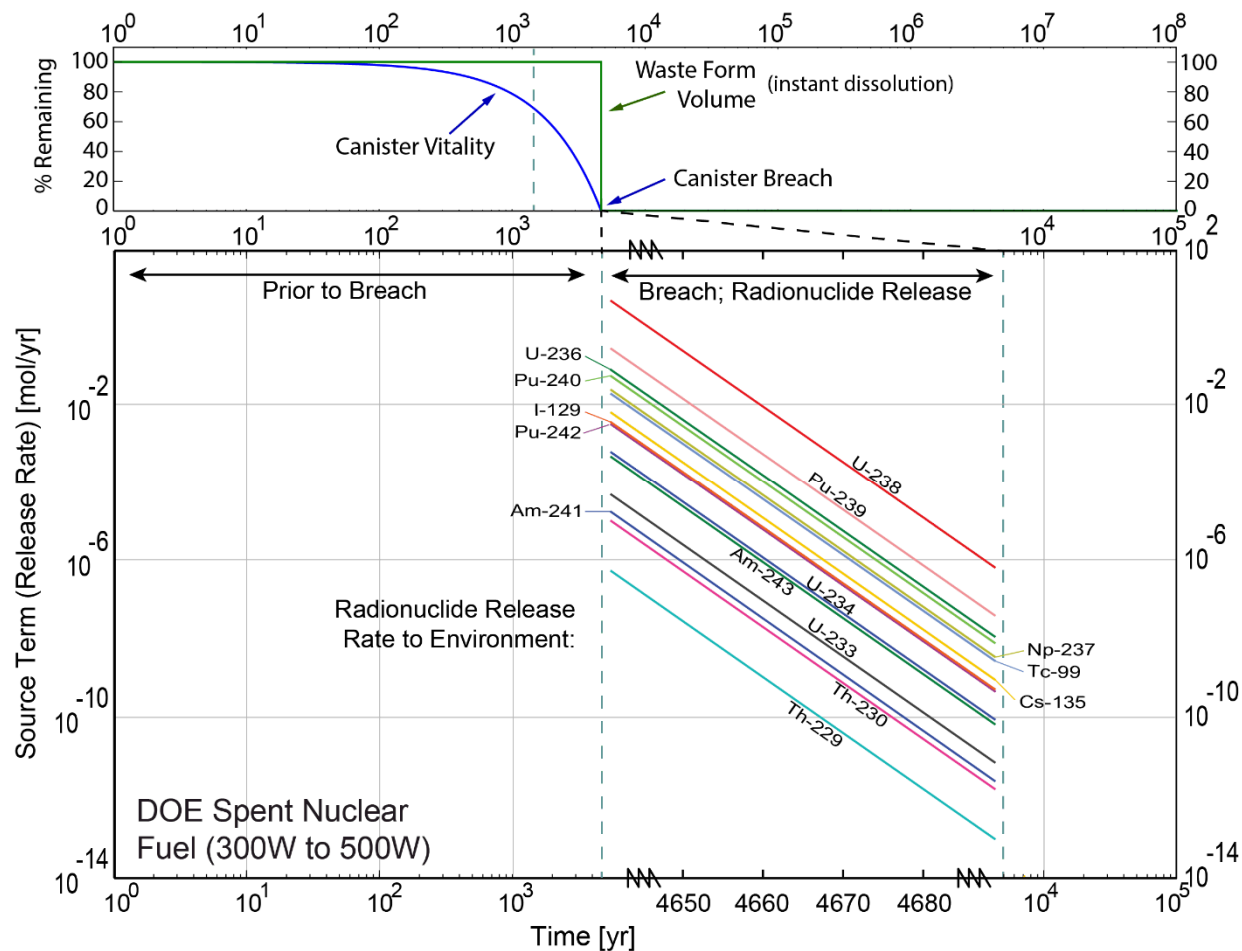 - *Include canister degradation mechanisms like corrosion and damage models*

- **Isotope decay and ingrowth algorithm:**
  - Fully implicit solution rather than 3-generation explicit

- **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 will work with you to get your functionality implemented

June 8, 2016
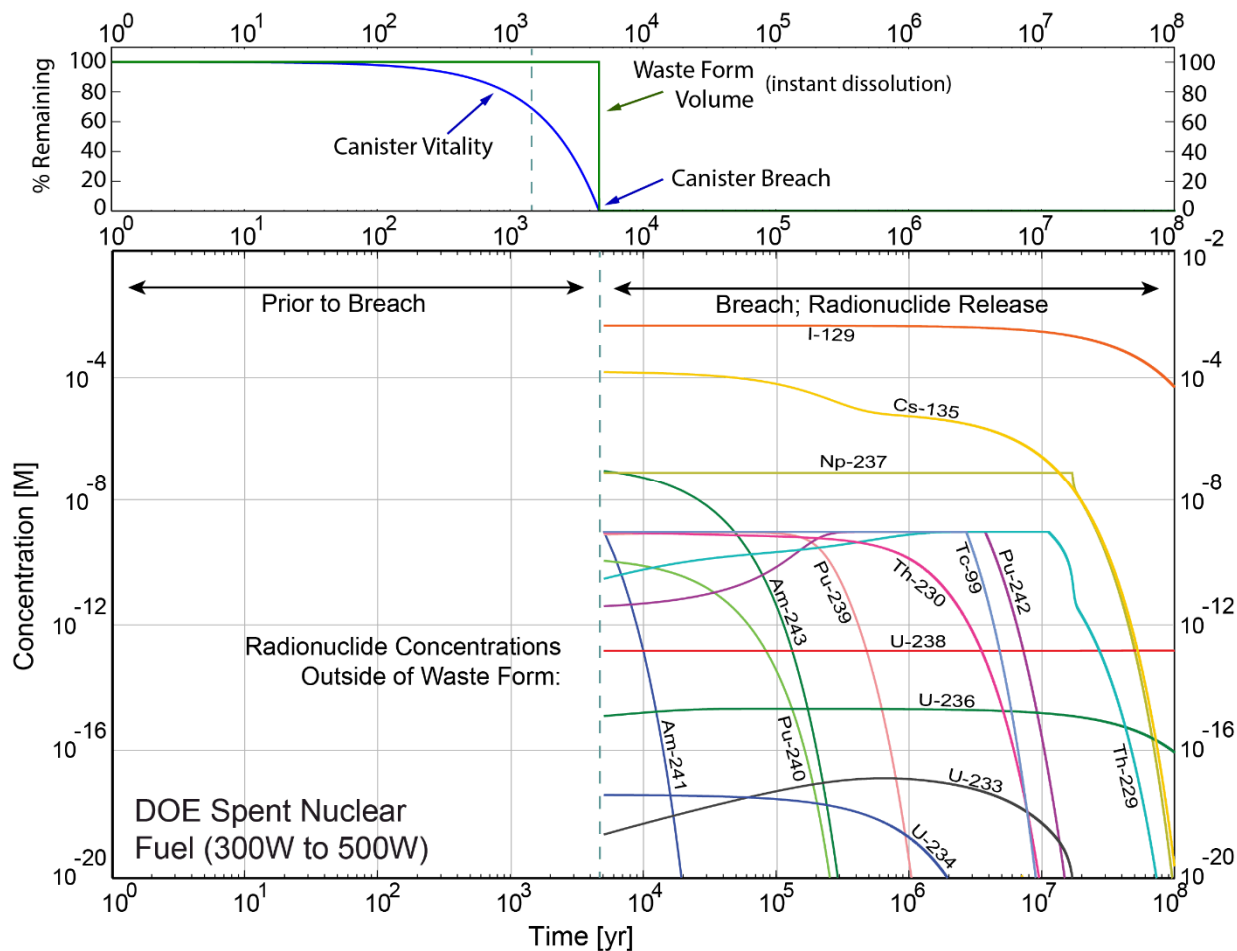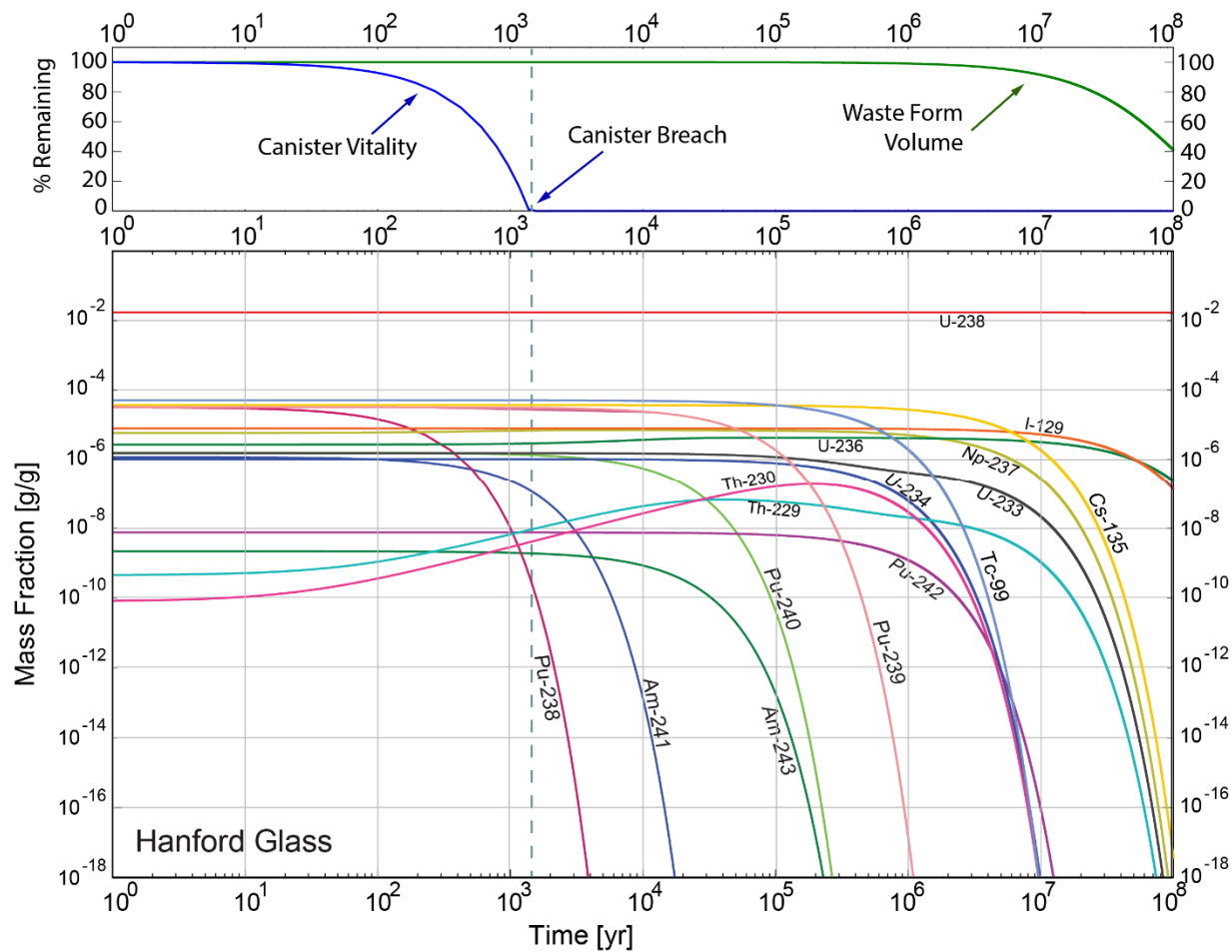
EXTRA SLIDES FOLLOW

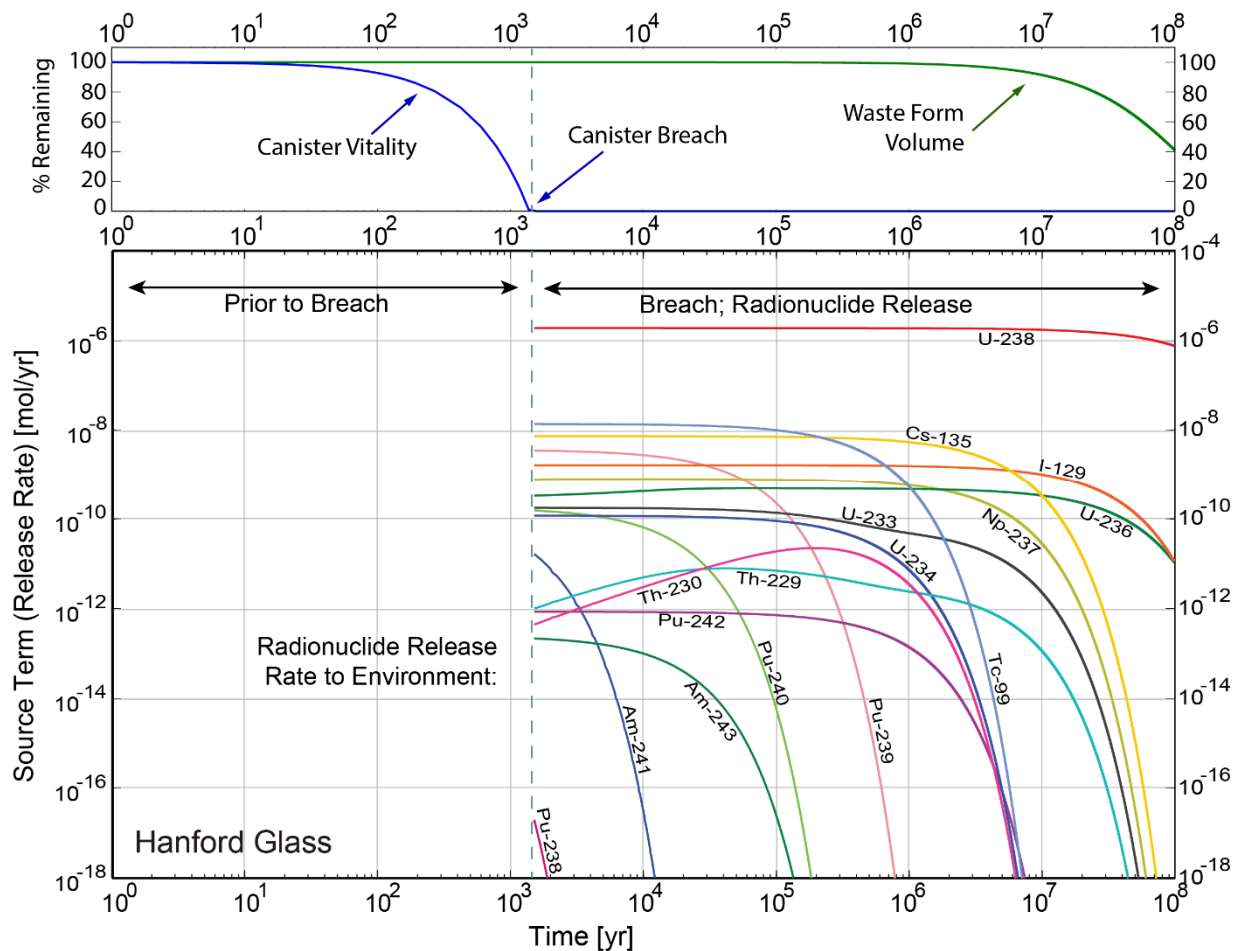# DOE Spent Nuclear Fuel

DOE Spent Nuclear Fuel

DOE Spent Nuclear Fuel

Hanford Glass

Hanford Glass

Hanford Glass