SAND2016-5392PE

Used Fuel Disposition Campaign

Isotope Chemistry and Source Term

Paul Mariner, Glenn Hammond, and Jennifer Frederick

Sandia National Laboratories

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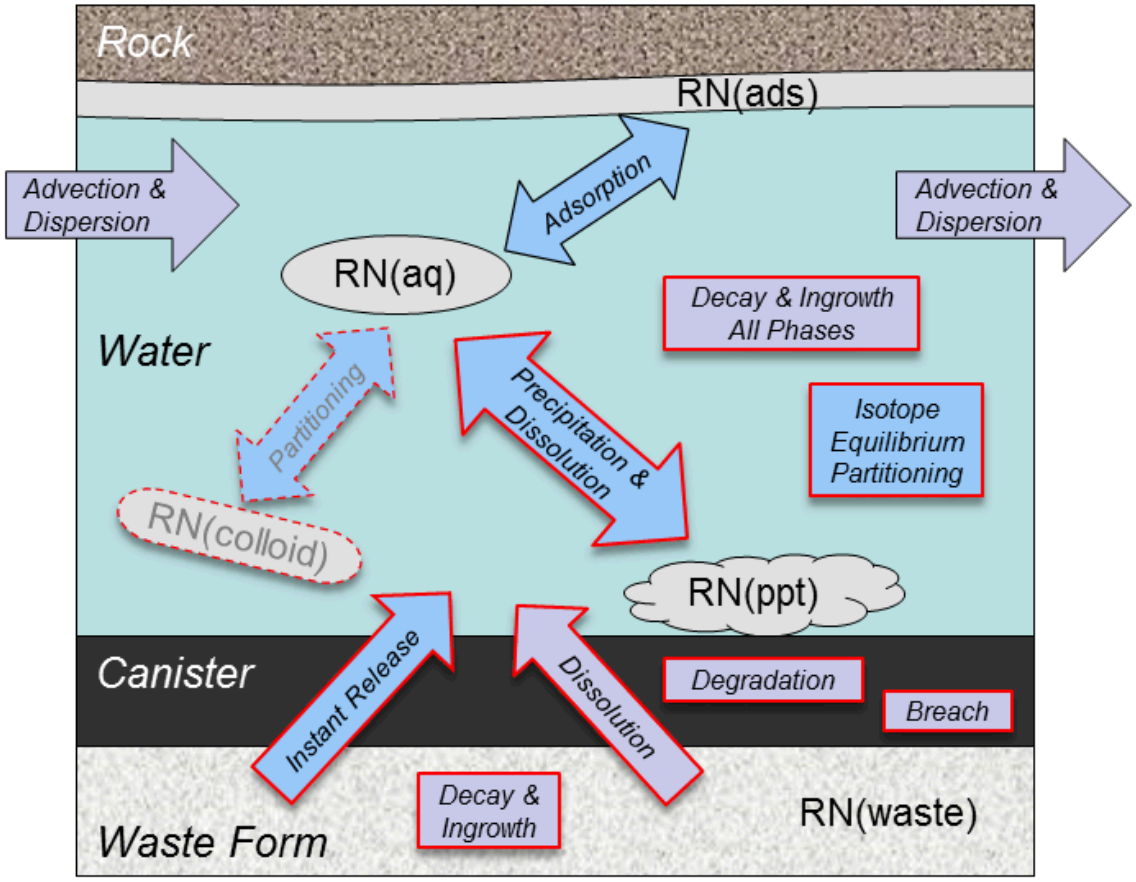
Presentation Outline

- Isotope chemistry conceptual model
- Decay and ingrowth
- Isotope partitioning
- Source term framework

Isotope Processes

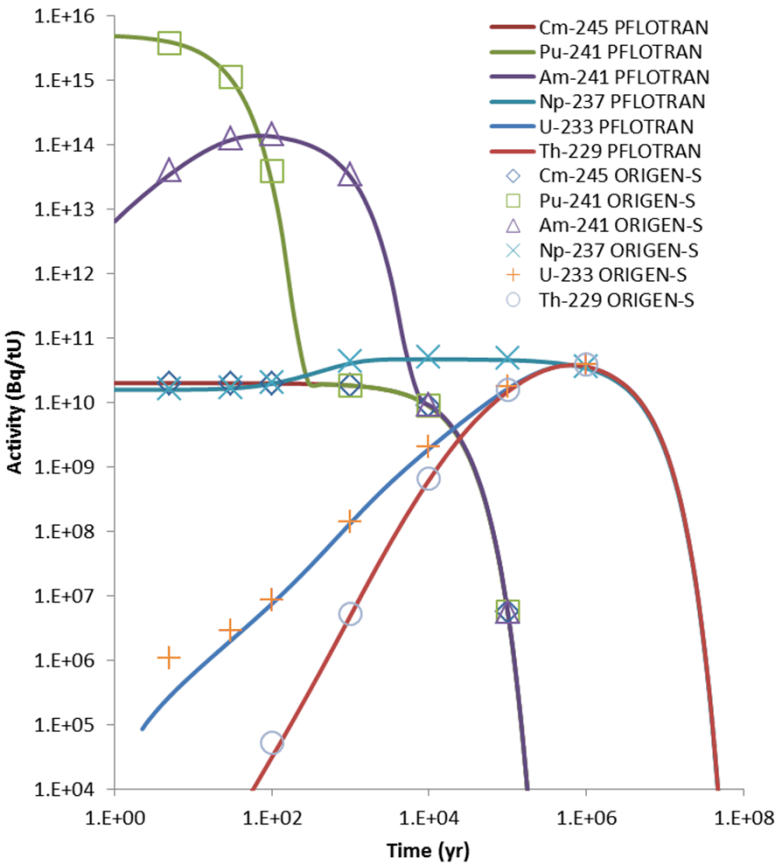
- **Isotope chemistry processes**
  - Decay and ingrowth
    - *Precipitation, sorption, aqueous*
  - Solubility
    - *Element-based*
  - Partitioning
    - *Maximum entropy*
    - *Precipitation, sorption, aqueous*
- **Source term**
  - Canister
    - *Degradation framework*
    - *Breach*
  - Waste form
    - *Decay and ingrowth*
    - *Instant release*
    - *Multiple dissolution models*
  - Isotope release
    - *Multiple isotopes*
    - *Congruent release of RNs*

Red = area of major advances this year
Underline = new feature this year



Decay and Ingrowth

- **3-Generation Analytical Solution**
  - Concentration of daughter is a function of:
    - Decay constants and initial concentrations of daughter, parents, and grandparents
    - Duration of time step
  - Accounts for multiple parents and grandparents
  - Limitation
    - Grandparents may not experience significant ingrowth
- **Validation**
  - Tested 7-generation Np series from ^{245}Cm to ^{229}Th
    - Limitation required removal of short-lived ^{233}Pa
    - Concentrations of short-lived isotopes like ^{233}Pa may be calculated on the side assuming secular equilibrium
  - Results compare well to ORIGEN-S results of Anttila 2005 (pp. 152 and 158)
    - Limitation affects ^{233}U and ^{229}Th at early times due to significant ingrowth of ^{241}Pu and ^{241}Am, respectively

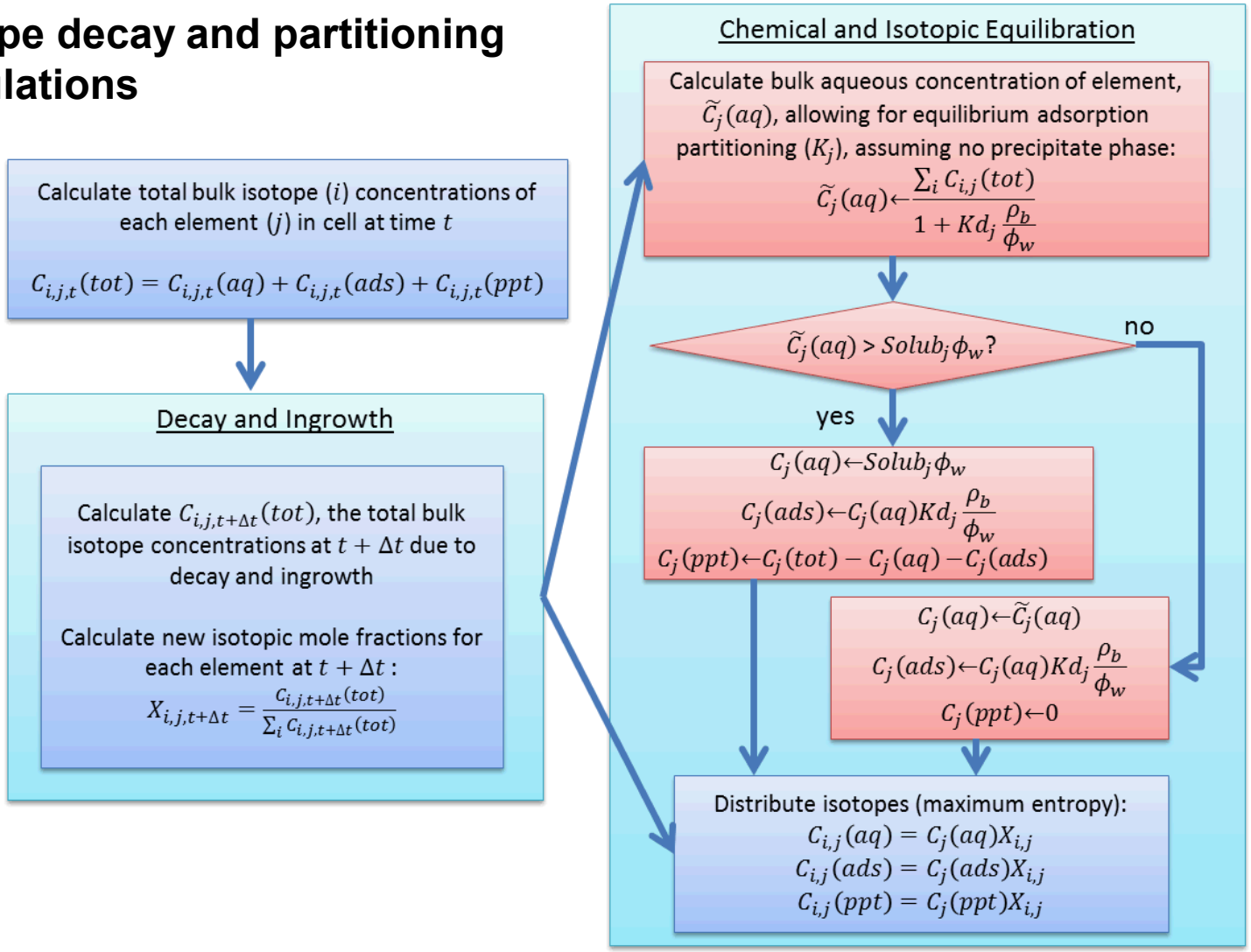


Isotope Modeling in GDSA

- **Physical and radioactive properties of isotopes of the same element are different**
  - Atomic mass, decay constant, daughter product
- **Chemical properties of isotopes of the same element are similar and are assumed to be identical**
  - Chemical reactions, Kd value, volume, volatility, etc.
- **Aqueous solubility is a property of the element**
  - The most abundant isotope(s) of each solubility-limited element must be included in the simulation to properly account for solubility-limited precipitation and dissolution
- **Thermodynamic equilibrium is enforced**
  - Precipitation and dissolution of solubility-limited elements
  - Sorption
- **Isotope concentrations are distributed to maximize entropy**
  - Isotope ratios of the same element in a given cell are forced to be identical in each phase (aqueous, sorbed, precipitate, etc.)
    - Done for simplification and for isotopic thermodynamic equilibrium

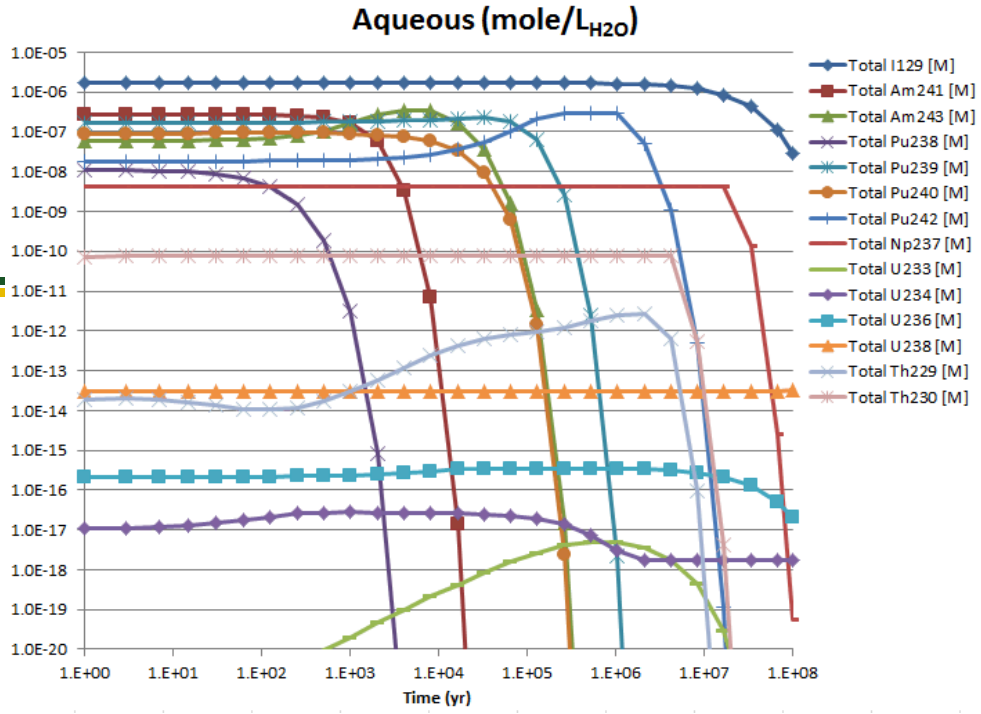
Isotope Partitioning Flow Chart

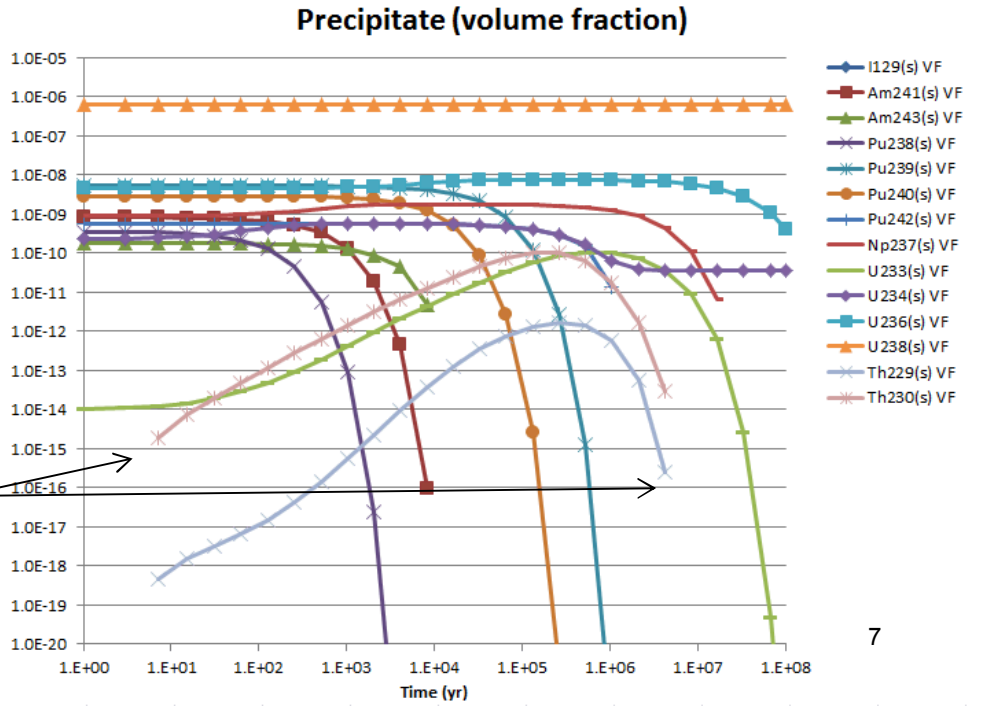
- **Isotope decay and partitioning calculations**



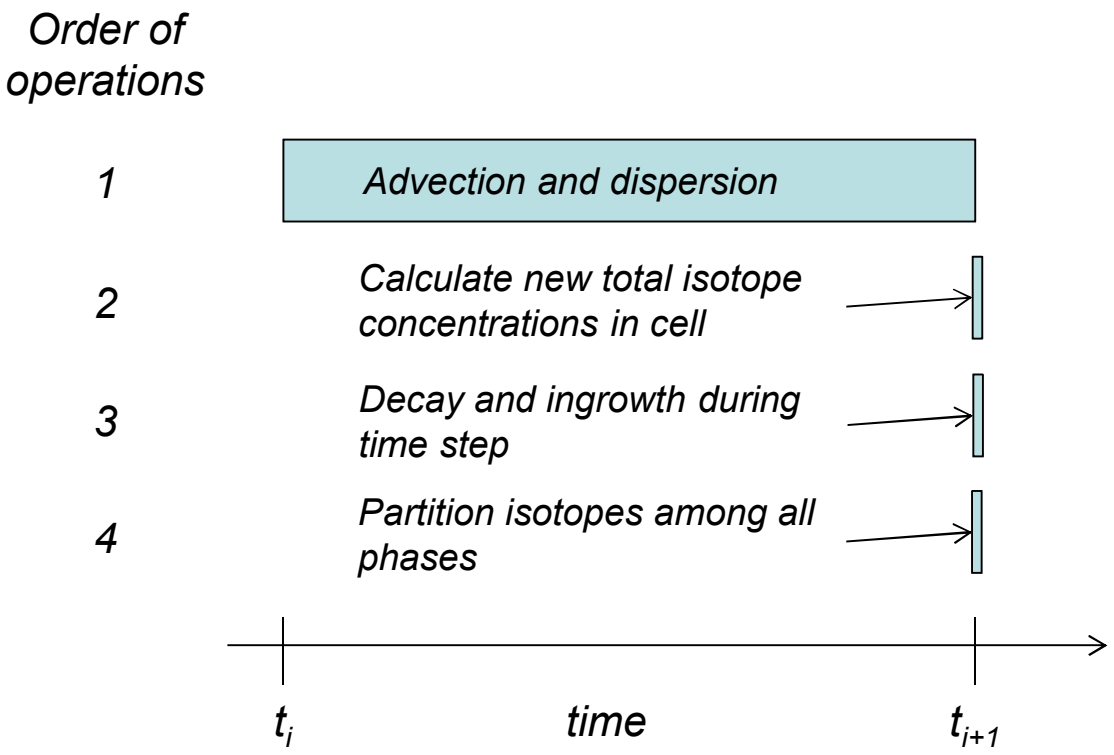
Used Fuel Disposition Isotope Modeling Demo

- **Isotope decay, ingrowth, and partitioning**
  - Example
    - Dissolve 1 gram UNF into 1 liter of water
    - Apply elemental solubility limits (M)
      - Th: 8×10^{-11}, Am: 3×10^{-7}, Pu: 3×10^{-7}, Np: 4×10^{-9}, U: 3×10^{-14}
    - Observe over time
  - Example shows
    - Ingrowth in aqueous and precipitate phases
    - Maximum entropy occurs (isotope ratios for a given element are the same in each phase)
    - Solubility constraints are honored
      - Th(aq) is below saturation prior to 7 yr and after 5 Myr





Isotope Modeling – Order of Operations



- **Operator splitting**
  - Needed to be able to decay and ingrow isotopes in the precipitate phase
- **Likely future improvements**
  - Numerical solver for decay and ingrowth (to address limitation of current analytical solution)
  - Colloid partitioning
  - Gas phase partitioning
- **Potential future improvements**
  - Solid solutions model for improved coupling

Source Term Framework

- **Canister degradation**
  - Degradation leading to breach
  - Degradation after breach
- **Waste form**
  - Decay and ingrowth
  - Instantaneous release
  - Multiple dissolution models
  - Radionuclide release
  - Track remaining volume

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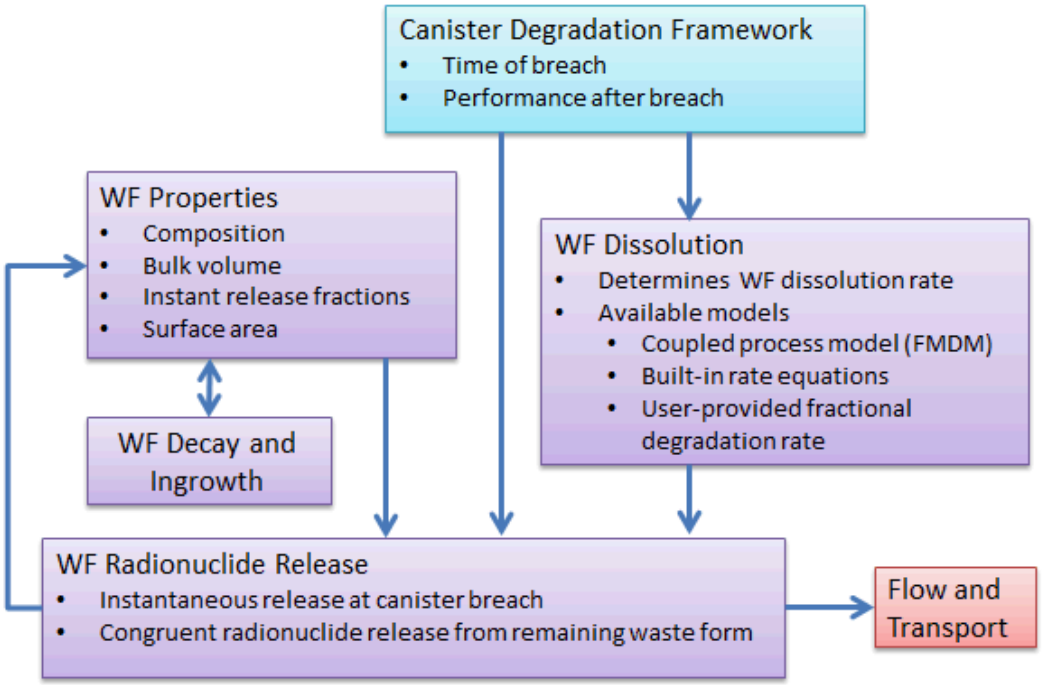
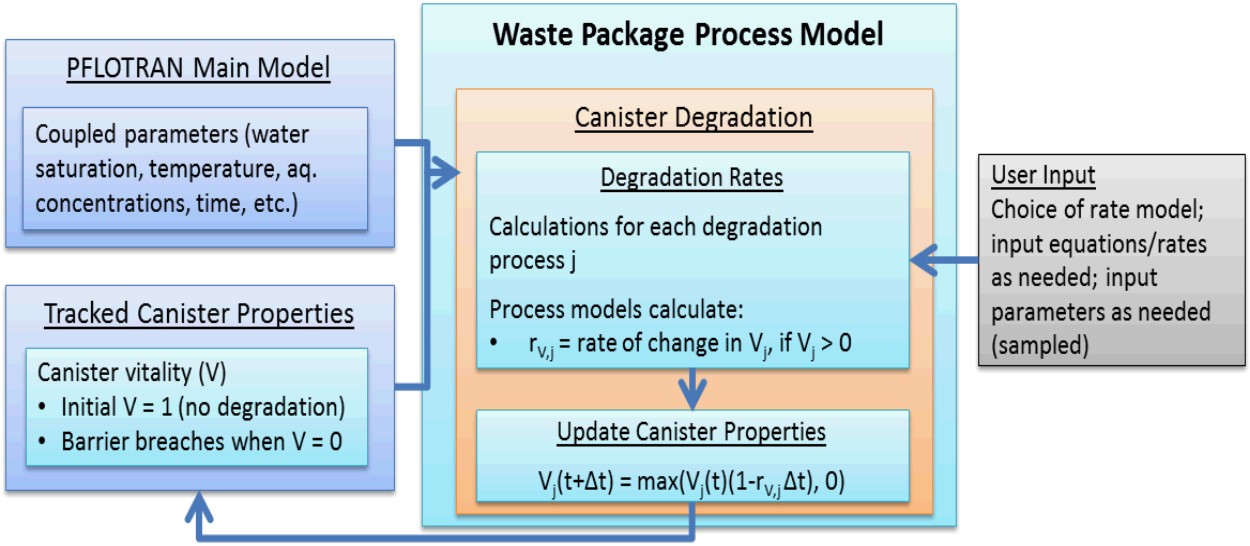


Canister Degradation Framework

- **Degradation leading to breach**
  - Framework allows for multiple processes to be modeled
    - General corrosion, localized corrosion, stress corrosion cracking (SCC), etc.
  - Each process will effectively provide a rate of degradation of canister *vitality*
- **Canister vitality, V**
  - V=1 prior to degradation
  - V=0 implies canister is breached
  - For general corrosion, V = fraction of remaining canister wall thickness
  - For SCC, V = fraction of canister thickness remaining between crack and breakthrough



- **Code**
  - Calculates dV/dt and V over time for each degradation process
  - Process models needed; current placeholder simply samples rates from a distribution

Future Work for Isotopes and Source Term

- **Isotope processes**
  - Partitioning to colloids
  - Partitioning to gas phase
  - Decay and ingrowth for 4+ generations
  - Solid solution modeling
- **Source term processes**
  - Canister vitality degradation process models
    - *Including early failure*
  - Canister performance process models for after breach
  - Improvements to waste form dissolution process models
  - Neutron activation process mode