U.S. Department of Energy
Office of Civilian Radioactive Waste Management

Assumptions and Results from Components of the Waste Form Degradation Model

Presented to:
Nuclear Waste Technical Review Board

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Management and Operating Contractor

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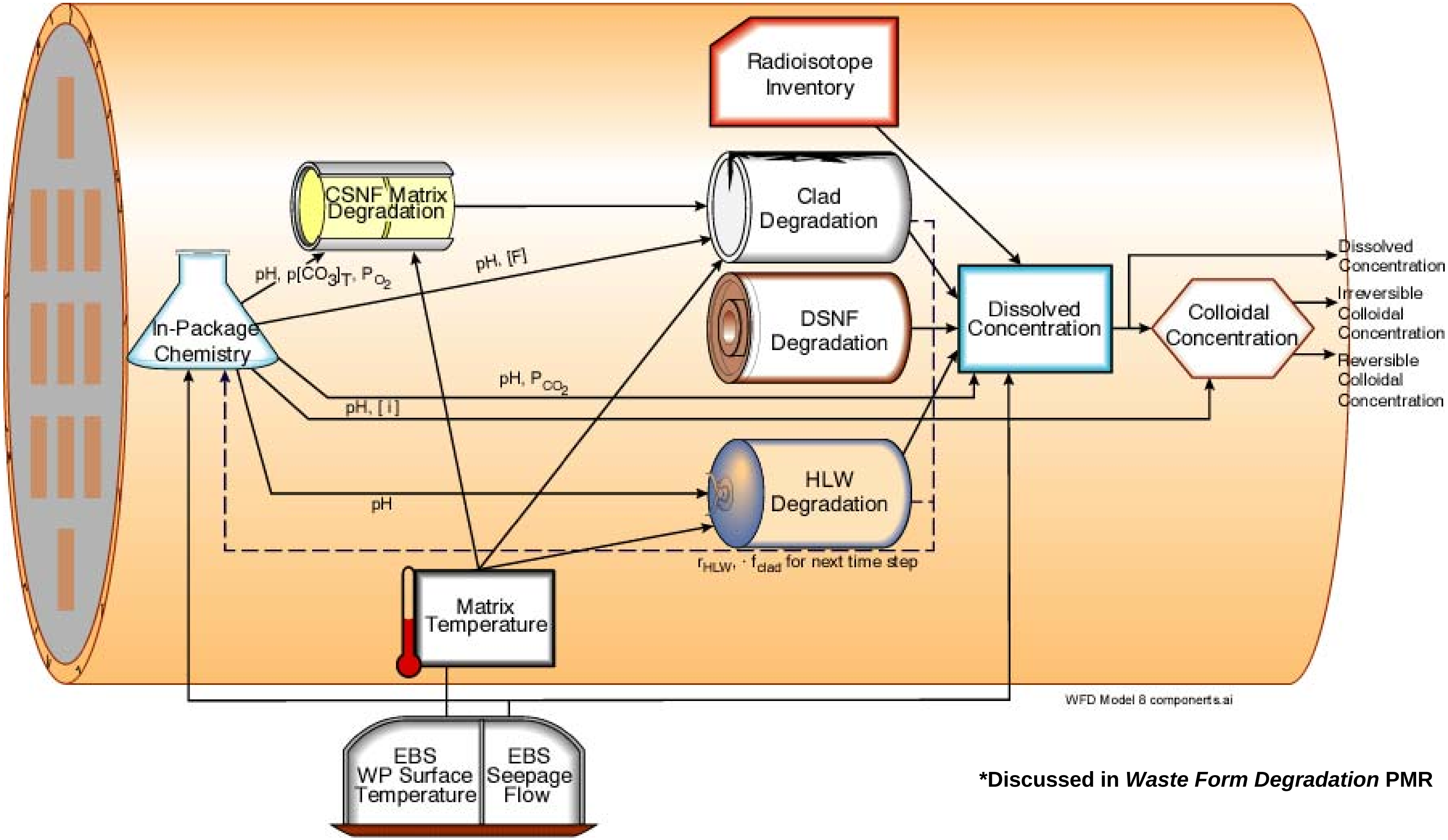
YUCCA MOUNTAIN PROJECT

Waste Form Degradation Model* has Eight Components



*Discussed in ***Waste Form Degradation*** **PMR**

Process Model Factors Affecting Radionuclide Release from Waste Form

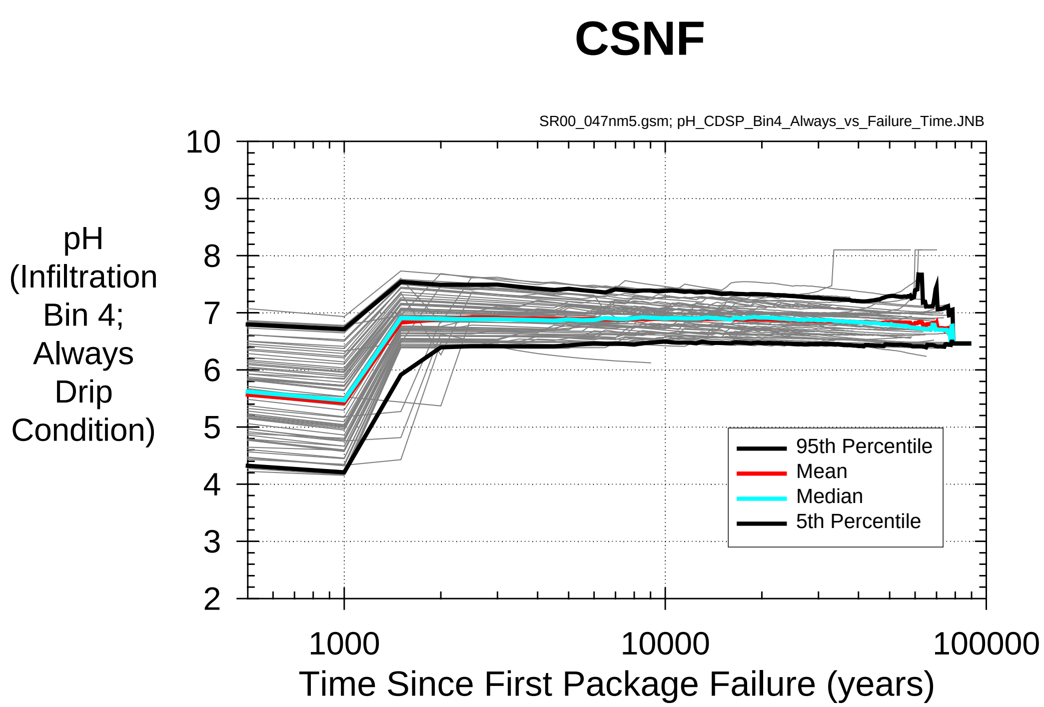
Key Attributes of Performance	Process Model Factor	TSPA-SR Input Parameters
Radionuclide Mobilization and Release from the Engineered Barrier System	In Package Environments	• pH – f (region, time) • Total dissolved carbonate (CO_3^{2}) – f (region, time) • Oxygen fugacity – f (region, time) • Ionic strength – f (region, time) • Fluoride – f(region, time) • CO_2 fugacity • Volume of water in the waste package/waste form cell
	Cladding Degradation and Performance	• Fraction of surface area of Zircaloy-clad CSNF exposed as a function of time
	CSNF Degradation and Performance	• CSNF intrinsic dissolution rate
	DSNF Degradation and Performance	• DSNF intrinsic dissolution rate
	HLW Degradation and Performance	• HLW intrinsic dissolution rate • Specific surface area
	Dissolved Radionuclide Concentration	• Concentration limits (solubilities) for all isotopes
	Colloid-Associated Radionuclide Concentrations	• Types of waste form colloids • Concentration of colloids • K_d and/or K_c for various colloid types • Fraction of inventory that travels as irreversibly attached onto colloids
	In-Package Radionuclide Transport	• Porosity of corrosion products – f (time) • Saturation of corrosion products – f (time) • Evaporation – f (temperature, relative humidity, composition)
	EBS (Invert) Degradation and Performance	• Thermally perturbed saturation in the invert – f (waste type, region, time, climate) • Porosity of the invert • Diffusion coefficient • Volumetric flux through the invert – f (climate, time) • Saturation in the invert after thermal pulse – f (time)

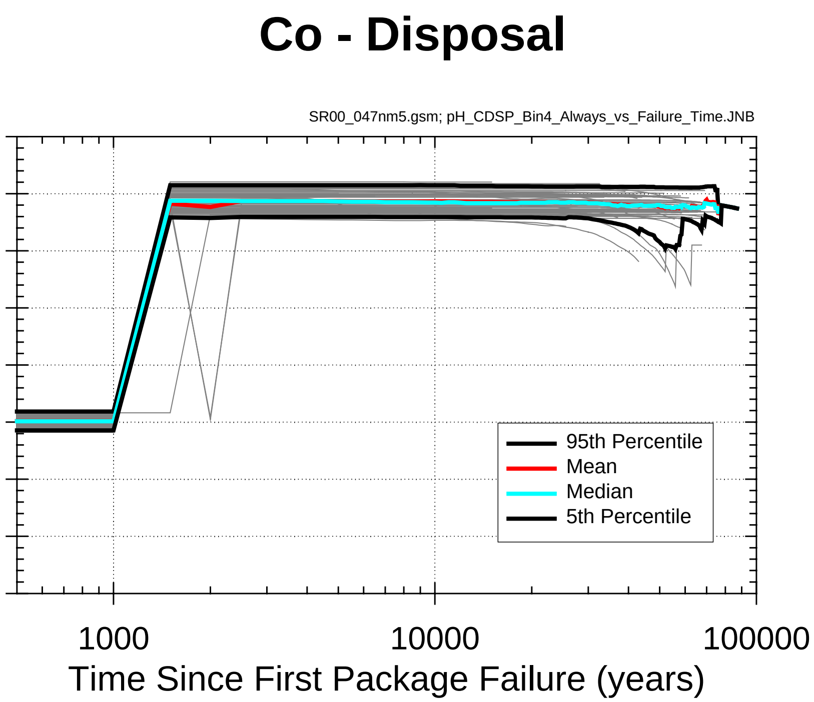
Assumptions of In-Package Chemistry Component*

- Cladding, HLW, and steel degradation rates, and fixed gas pressures (CO_2 and O_2) control bulk chemistry (pH, $[CO_3]_T$, [i], and [F])
- Bulk chemistry, in turn, influences five other components: CSNF degradation, HLW degradation, DSNF degradation, radionuclide solubility, and colloid stability
- Bulk chemistry approximated by well mixed, always oxidizing, full bathtub scenario
- Chemical condition in WP dominates; thus, J-13 well water can be used (e.g. no influence of evaporation assumed)

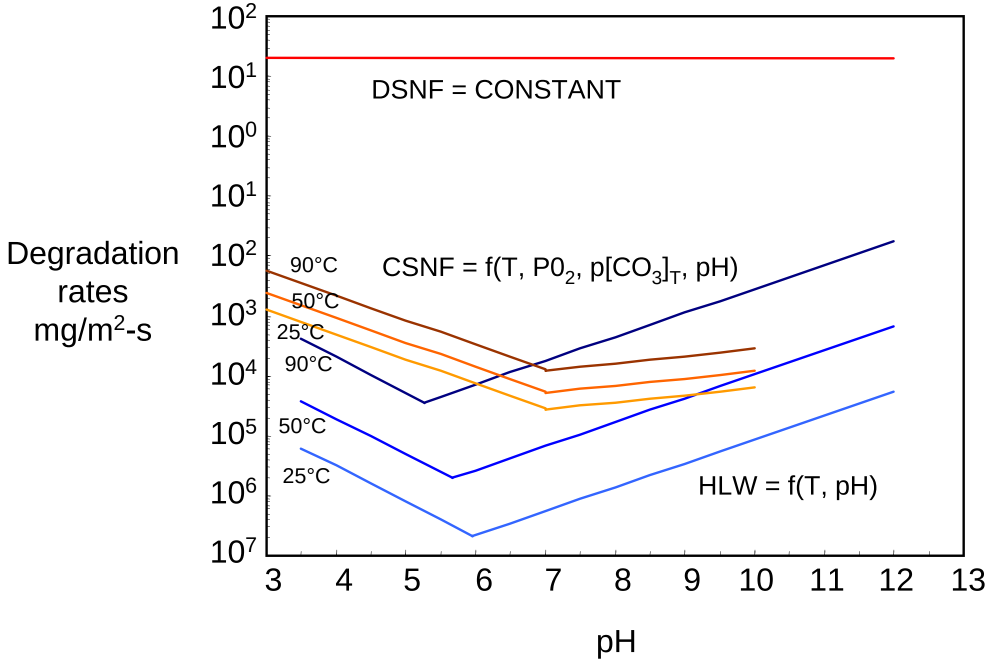
* Discussed in ***In-Package Chemistry Abstraction*** **AMR**

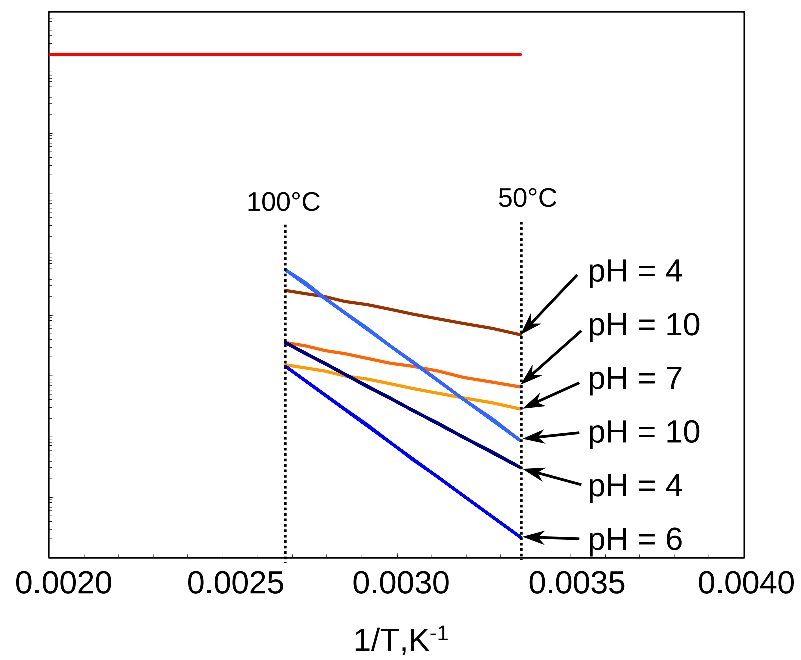
Uncertainty of pH
Greater in CSNF Waste Packages





Conservative Waste Form Degradation Rates Assumed for CSNF*, HLW^t, and DSNF☟



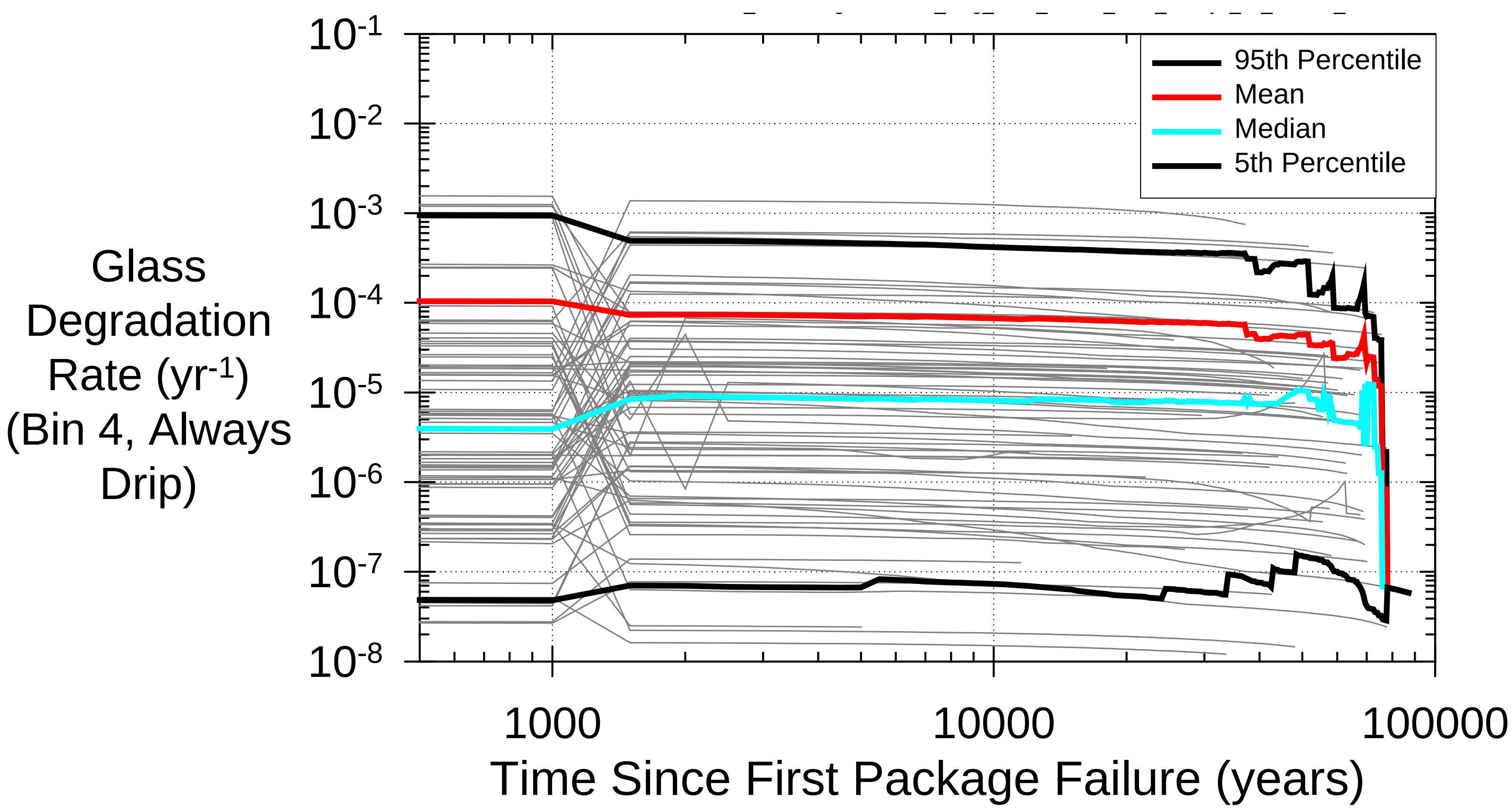


***Discussed in *CSNF Waste Form Degradation: Summary and Abstraction* AMR**

^{t}Discussed in *Defense High-Level Waste Glass Degradation* AMR

☟Discussed in *DSNF and Other Waste Form Degradation Abstraction* AMR

Glass Degradation Rate Has Very Large Uncertainty



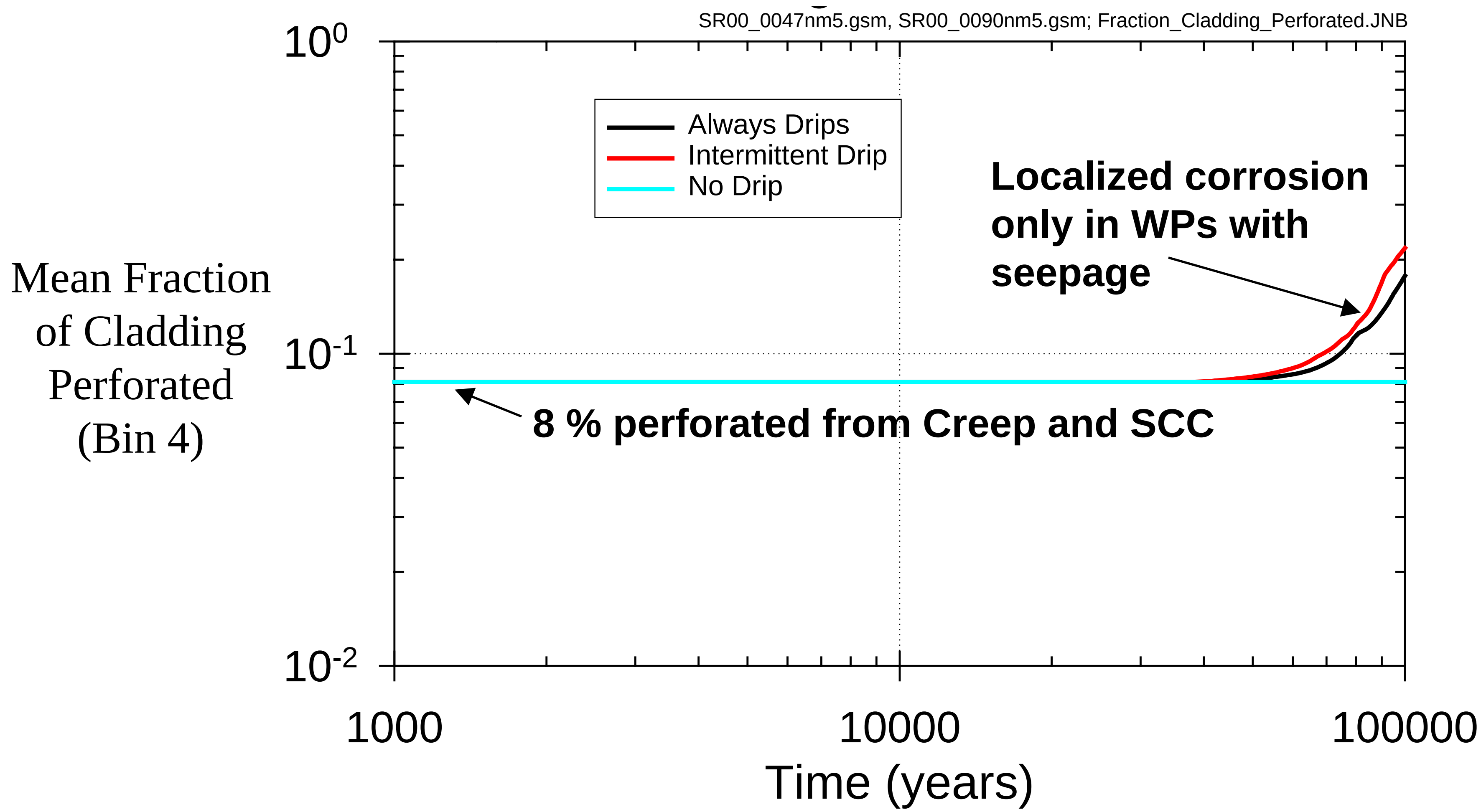
TSPA-VA: 1×10^{-2} yr^{-1} < 1000 yr after exposure; 1×10^{-6} yr^{-1} at 10,000 yr

Assumptions in Cladding Component*

- **Two steps: perforation, unzipping**
- **Four perforation mechanisms included**
  - initial
  - creep and SCC (average of 8%)
  - localized corrosion (after water in WP ~40,000yr)
  - seismic: (rare - frequency of 10^{-6}/yr)
  - Fast release = gap fraction + fraction of rod dissolved before unzipping starts
- **Unzipping assumed to occur between 1 and 240 times faster than CSNF degradation rate**
  - Inventory releases as cladding unzips (except fast release)

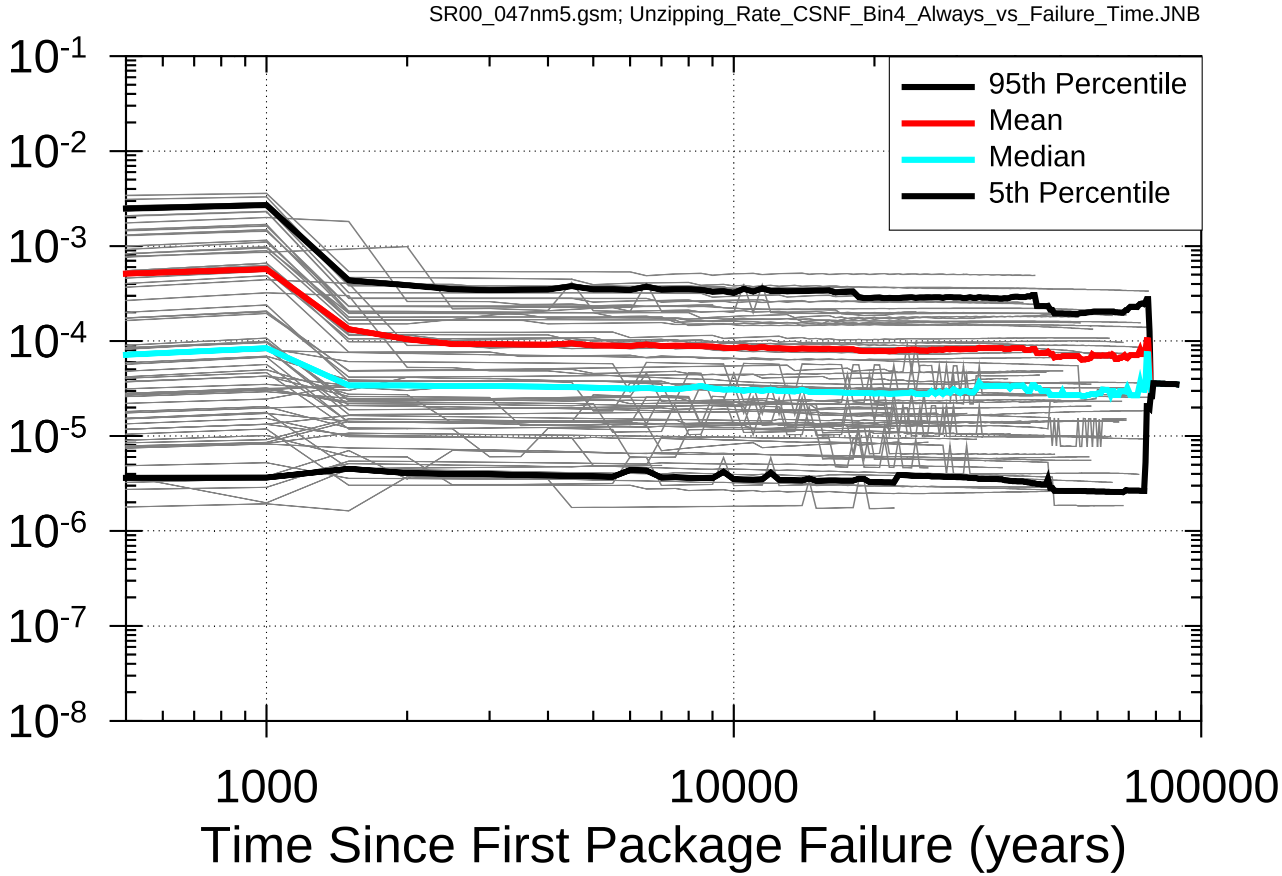
***Discussed in *Clad Degradation- Summery and Abstraction* AMR**

Prior to 50,000 yr Cladding Perforation Caused by creep rupture



Unzipping Rate has Large Uncertainty

Unzipping Rate for Cladding (yr^{-1}) (Bin 4, Always Drip)



TSPA-VA: CSNF degradation $2x10^{-2}$ yr^{-1} to $4x10^{-3}$ yr^{-1} over 10,000 yr

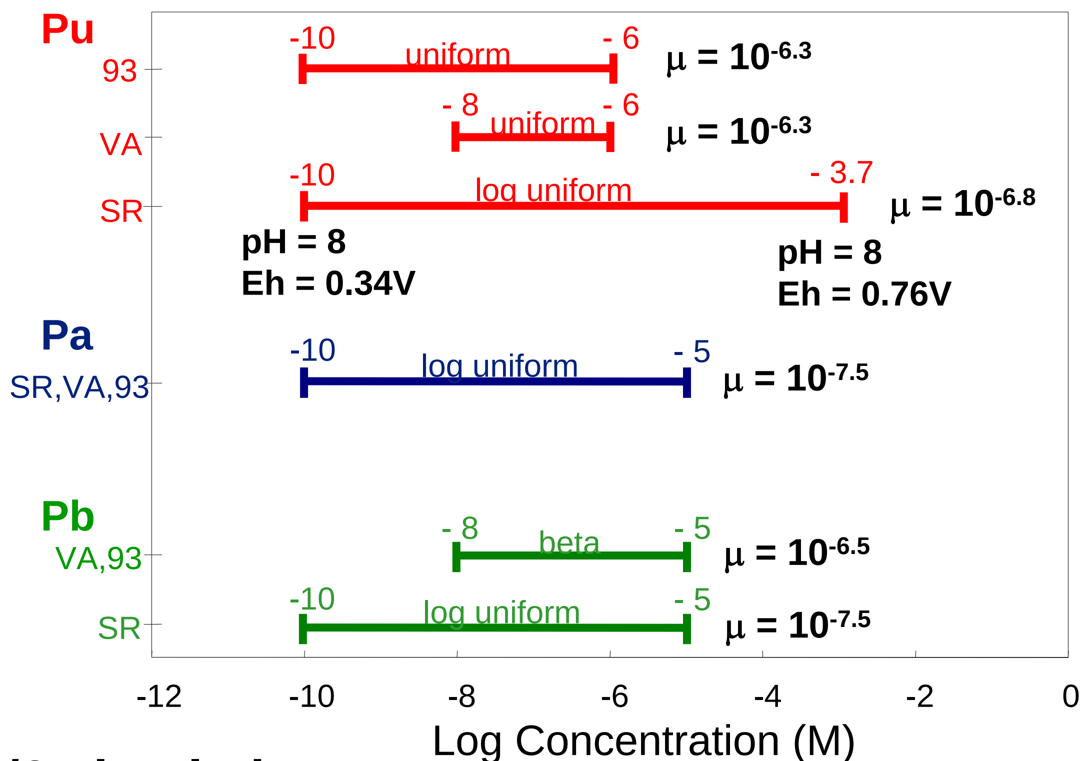
Assumptions in Solubility Component*

- Conservatively selected pure phases to control solubility
- Conservatively fixed gas pressures CO_2, O_2 at atmospheric conditions
- Conservatively neglect sorption or coprecipitation of radionuclides

*Discussed in ***Summary of Dissolved Concentration Limits*** AMR

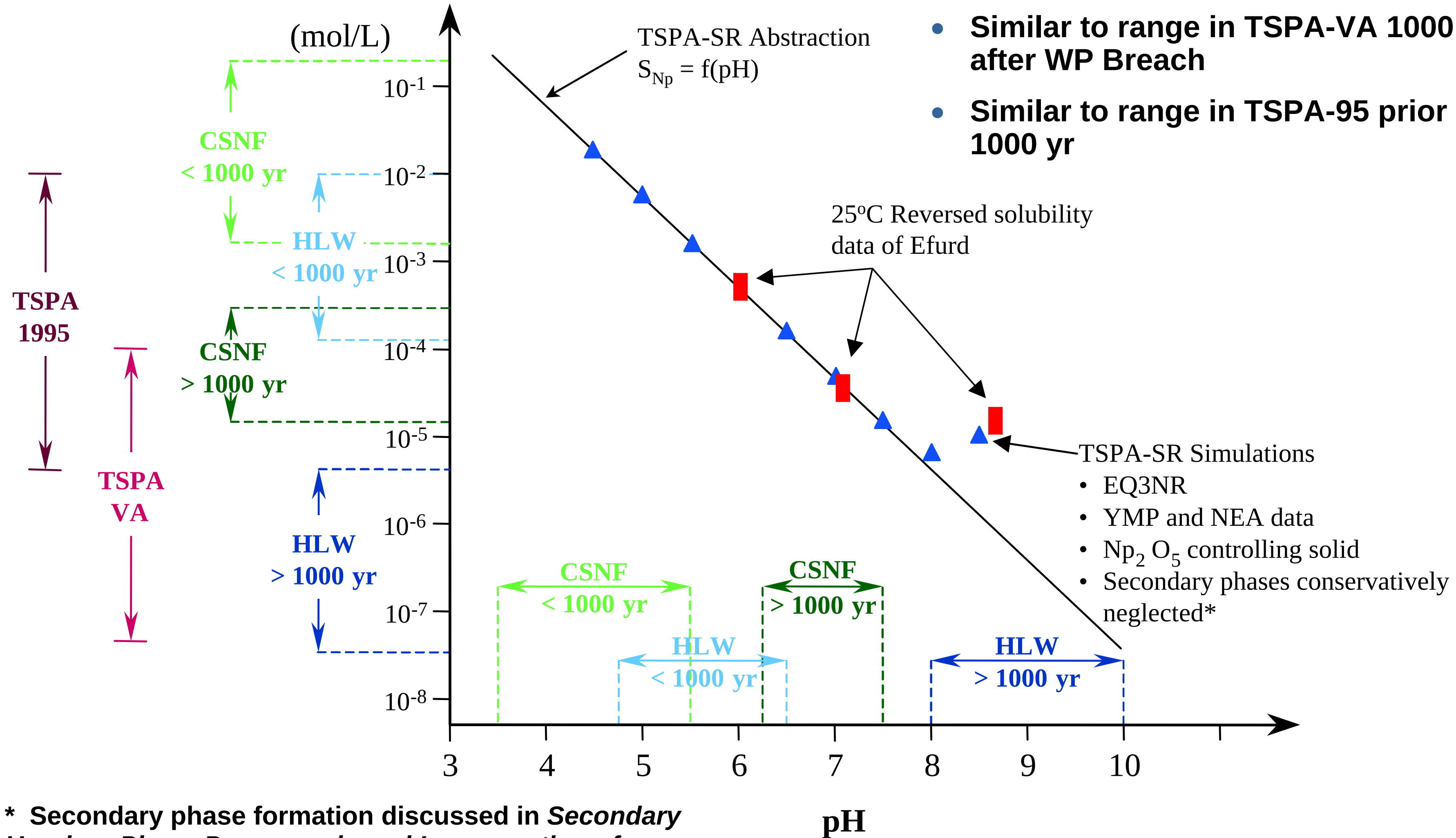
Solubility of Important Radioisotopes updated for TSPA-SR

- **Distributions**
  - Pu - Solid $Pu(OH_4)(am)$
  - Pa
  - Pb
- **Empirical functions**
  - Np = f (pH)
  - Am = f (pH, P_{co2})
  - U = f (pH, P_{co2}, T)
- **Constants**
  - Tc = I = Sr = Cs = C = 1M
- **Estimates based on EQ3/6 simulation**
  - Thermodynamic data used recent NEA and literature



Np Solubility Varies with pH

- **Similar to range in TSPA-VA 1000 yr after WP Breach**
- **Similar to range in TSPA-95 prior to 1000 yr**

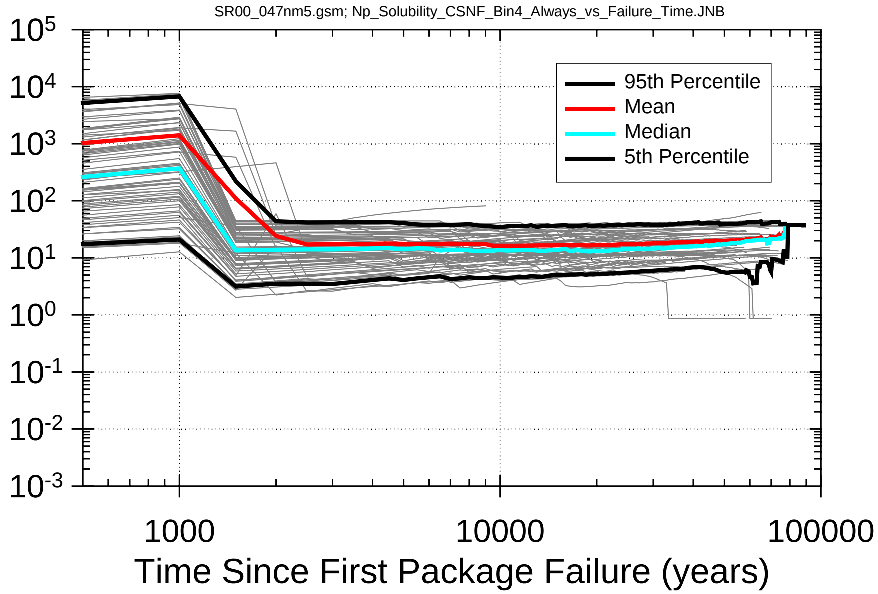


*** Secondary phase formation discussed in *Secondary Uranium-Phase Paragenesis and Incorporation of Radionuclides into Secondary Phases* AMR**

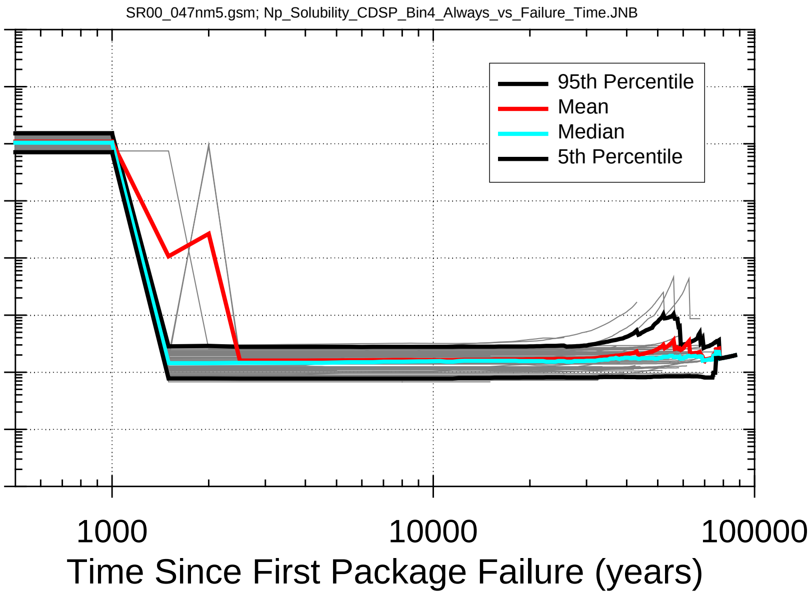
Uncertainty in Solubility of Np Determined by Uncertainty in pH

CSNF

Solubility of Np in Waste Form mg/L Bin 4, Always Drip Condition



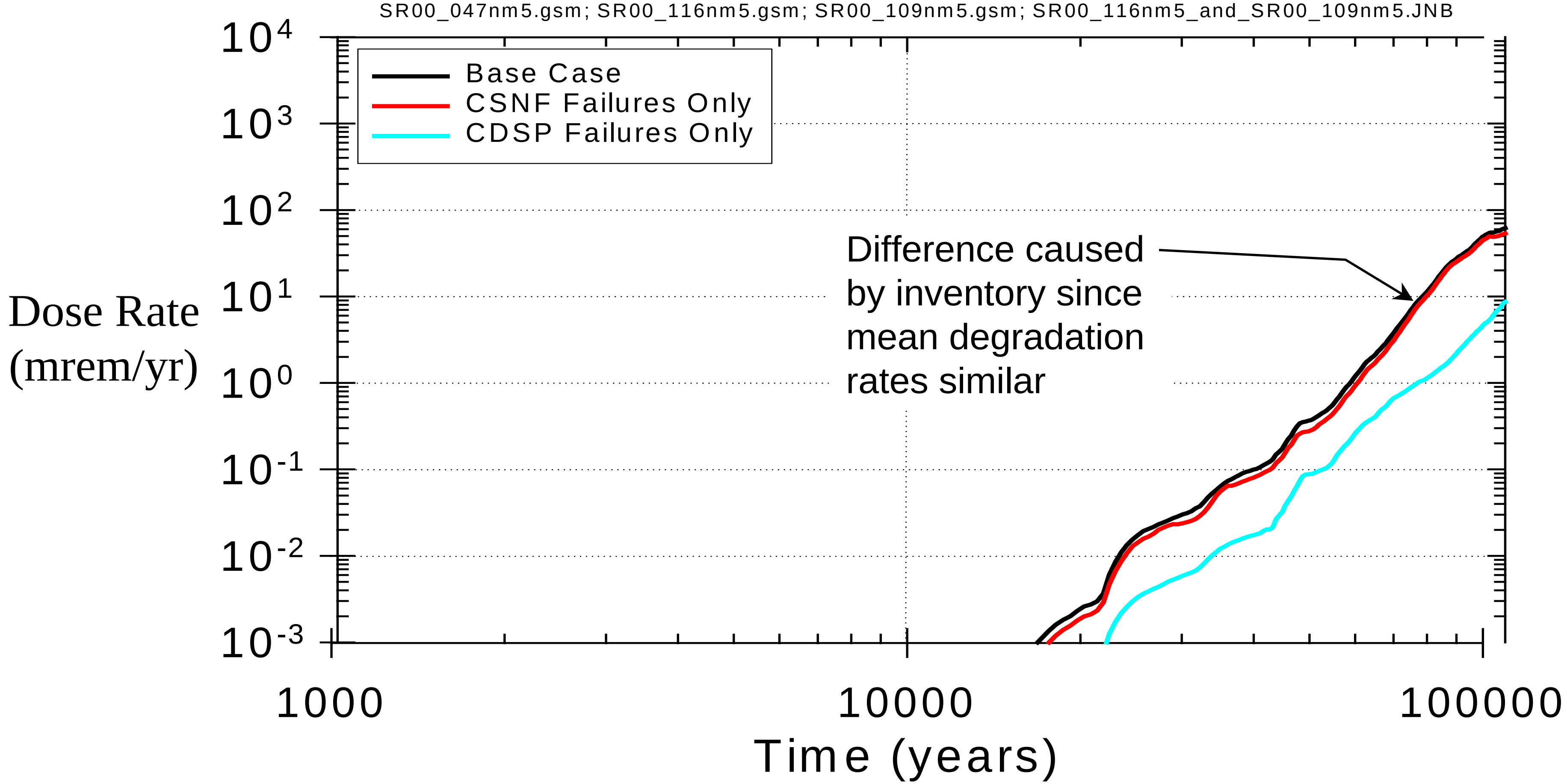
Co - Disposal



Assumptions in Colloid Component

- Calculated ionic strength and pH determines concentration
- *Irreversible* waste colloids from HLW
- Pu and Am transported as *irreversible* colloids
- *Reversible* colloids from groundwater, rust, and waste
- Pu, Am, Th (Ra, Pb), Pa (Ac), Sr, Cs transported as *reversible* colloids
- No filtration or sorption within package
- Diffusion coefficient for colloids very conservative (only 100 times less than aqueous diffusion)

Releases from CSNF Contribute Most to Dose



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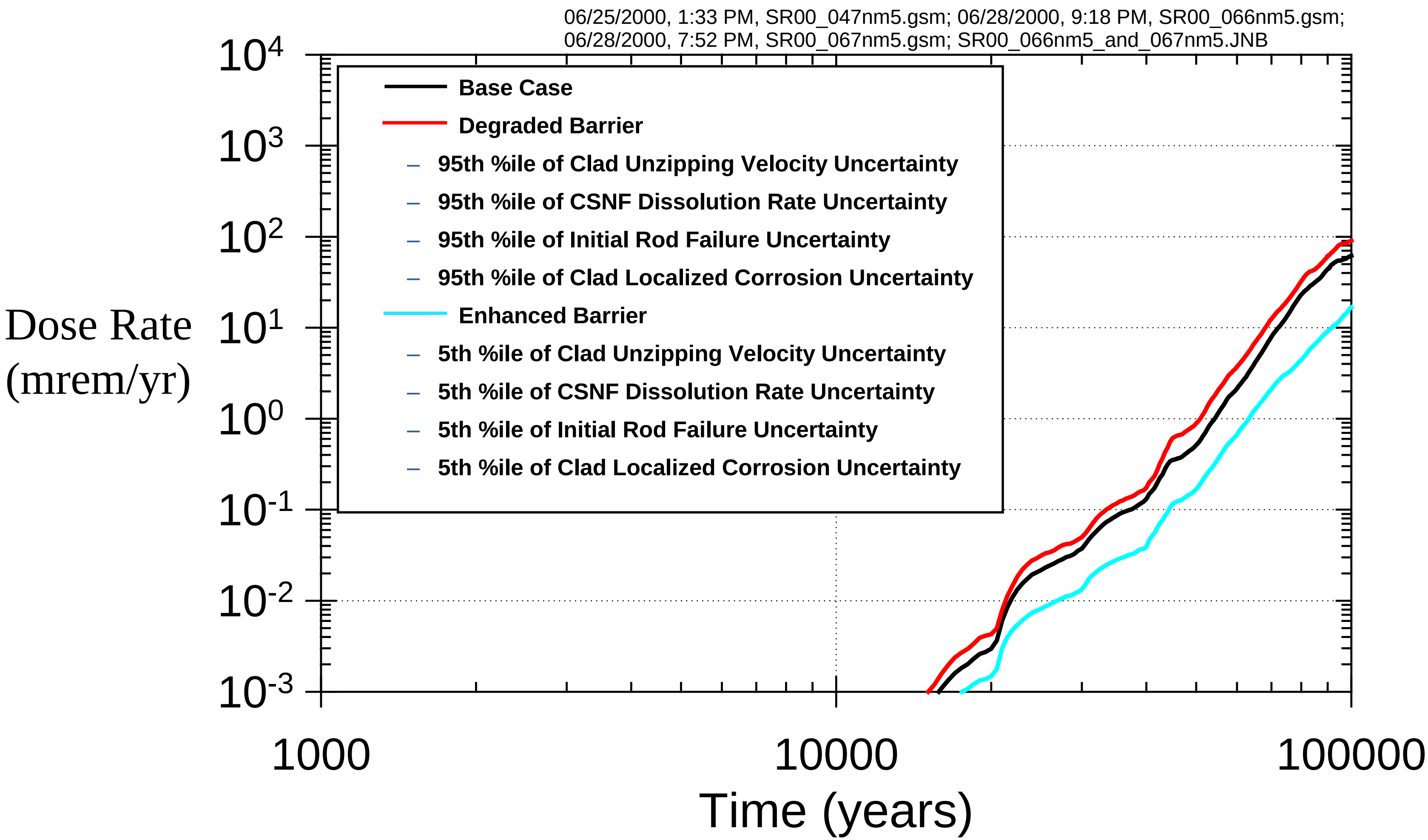
Degraded Cladding Condition Increases Dose by Factor of 4



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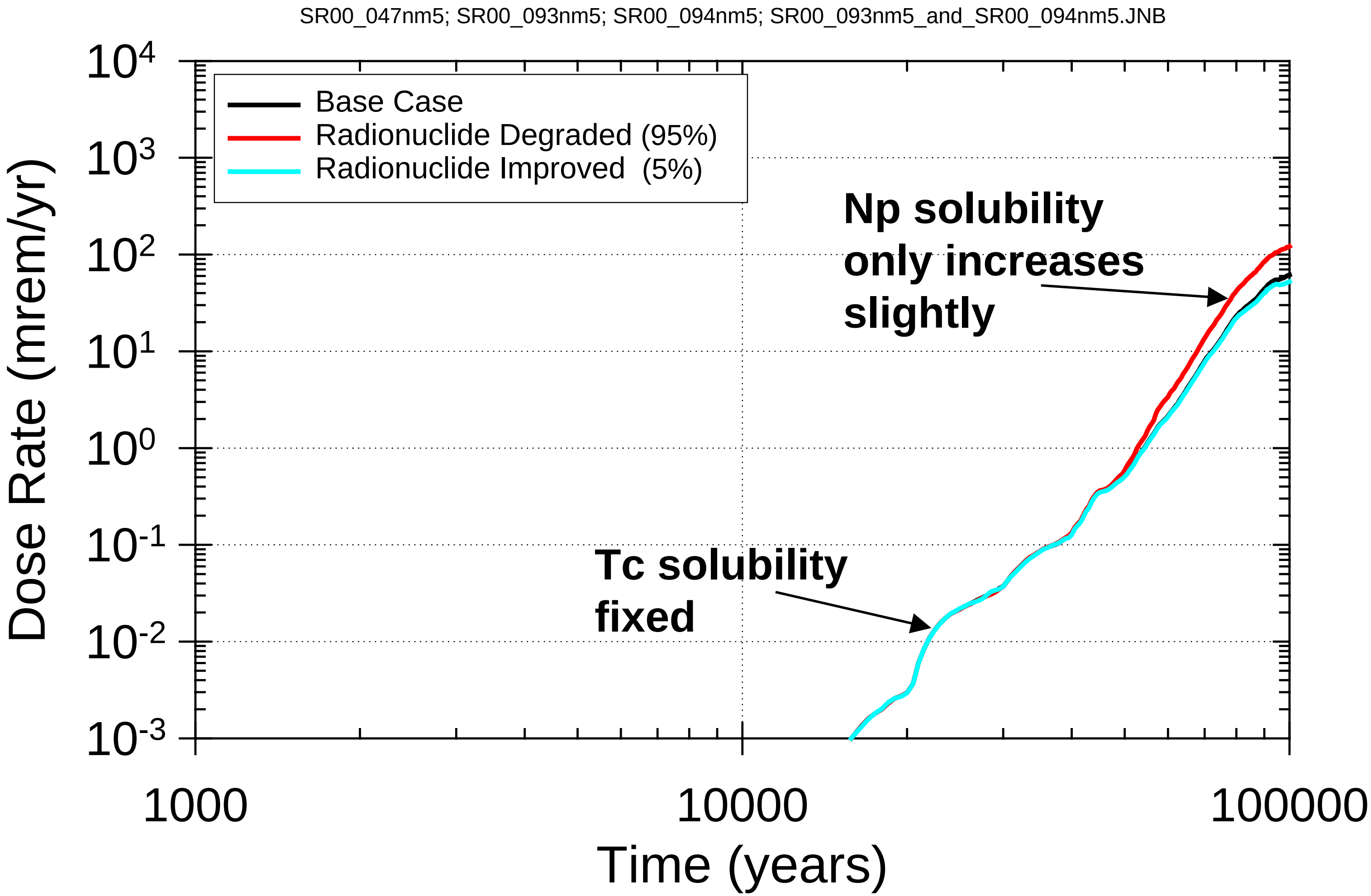
Because pH Range Narrow in EBS, Np Release Only Changes Slightly



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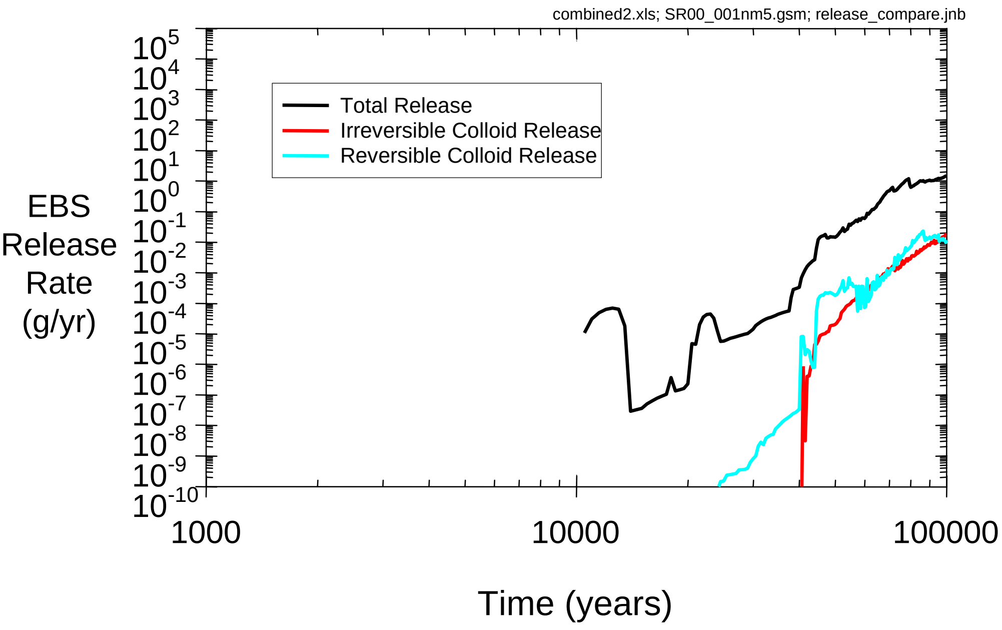
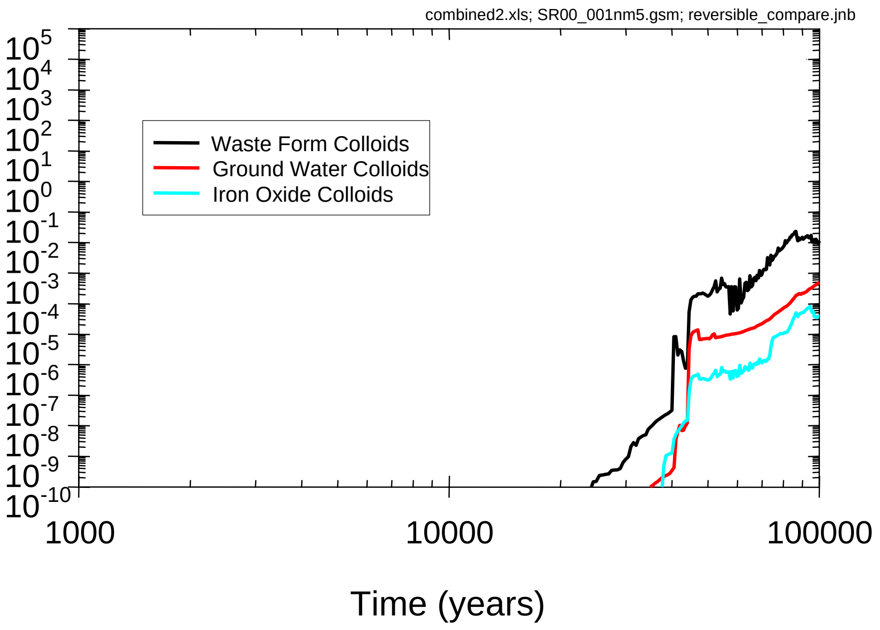
Colloids Only Contribute a Small Fraction to Release of ^{239}Pu

Total ^{239}Pu Release

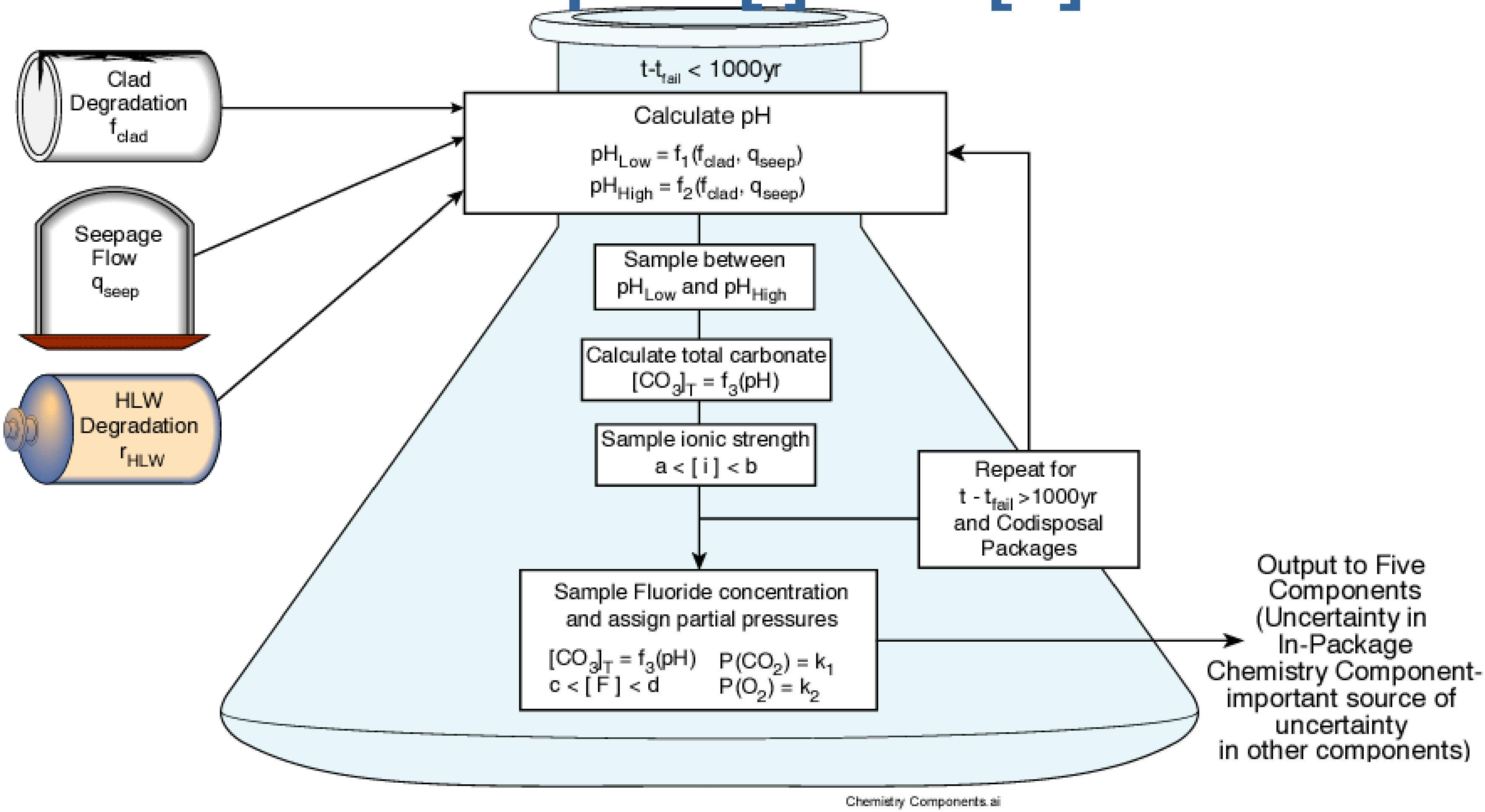


Source of Reversible Colloids

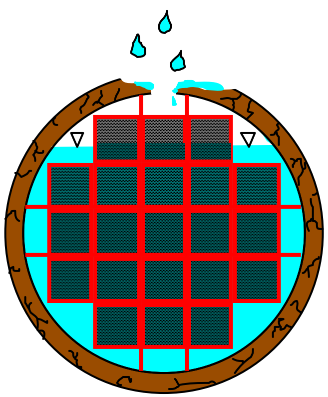


Backup Slides

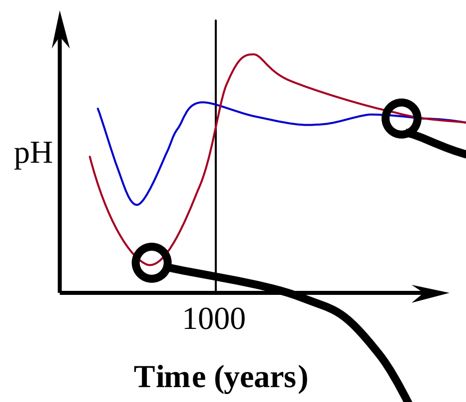
In-Package Chemistry Component Estimates pH, Calculates $[CO_3]_T$, and Samples [i] and [F]



In-Package Chemistry Component Developed from Regression Analysis on EQ3/6 Runs

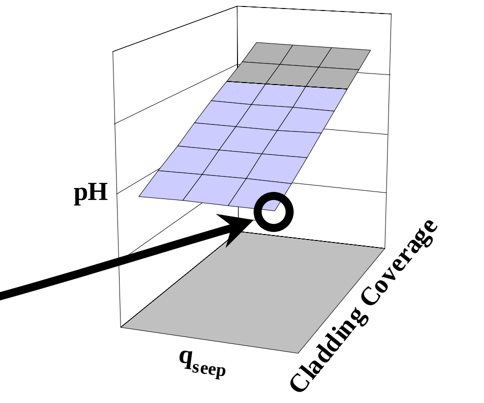


CSNF Conceptual Model

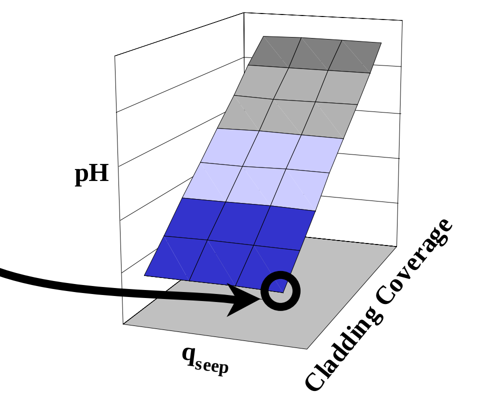


EQ3/6 Runs Varying:

- q_{seep}
- r_{steel}*
- r_{hlw}
- f_{clad}



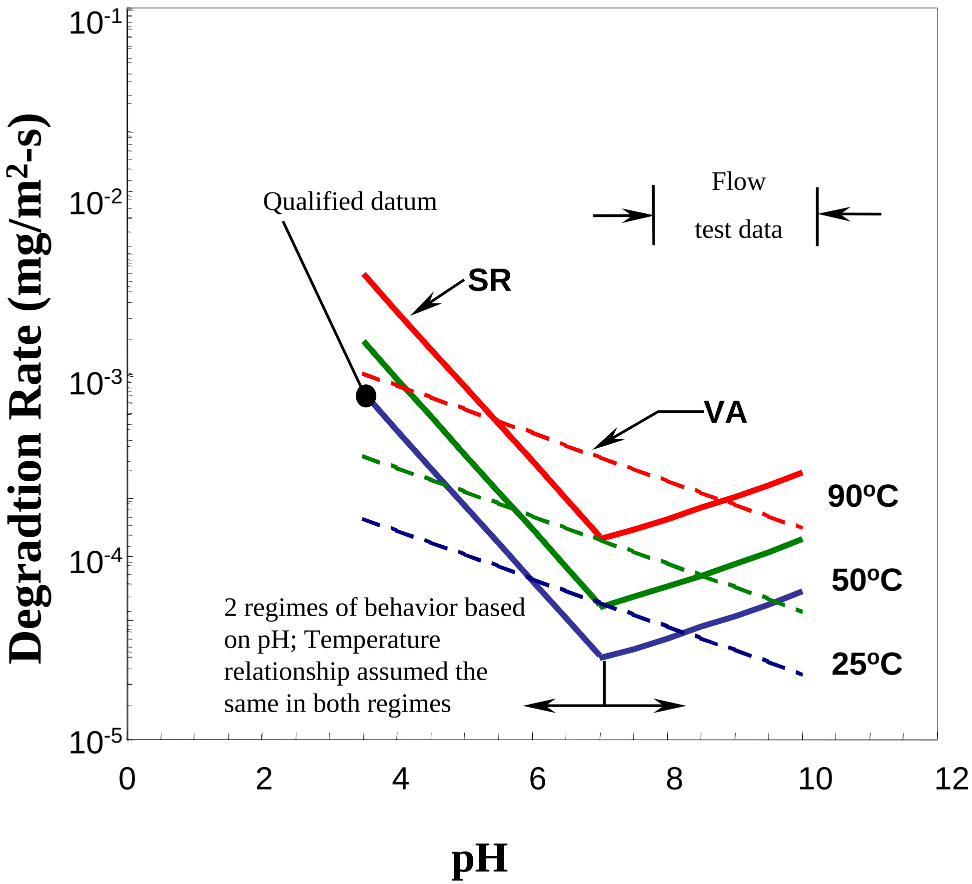
pH Abstraction for > 1,000 Years

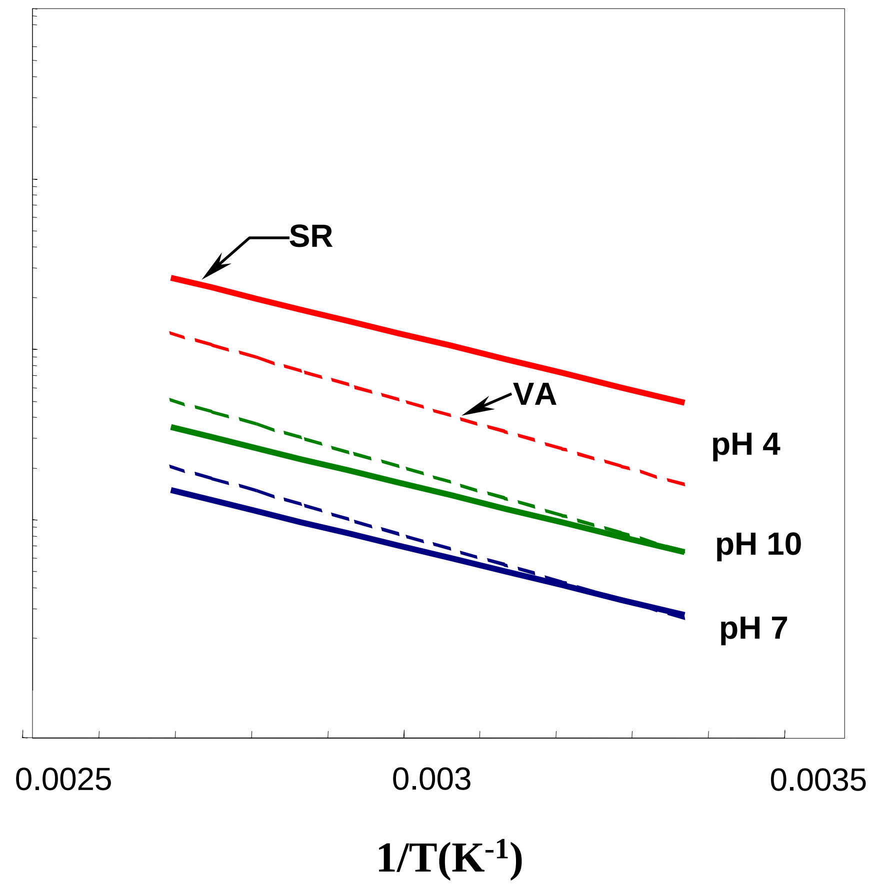


pH Abstraction for < 1000 Years

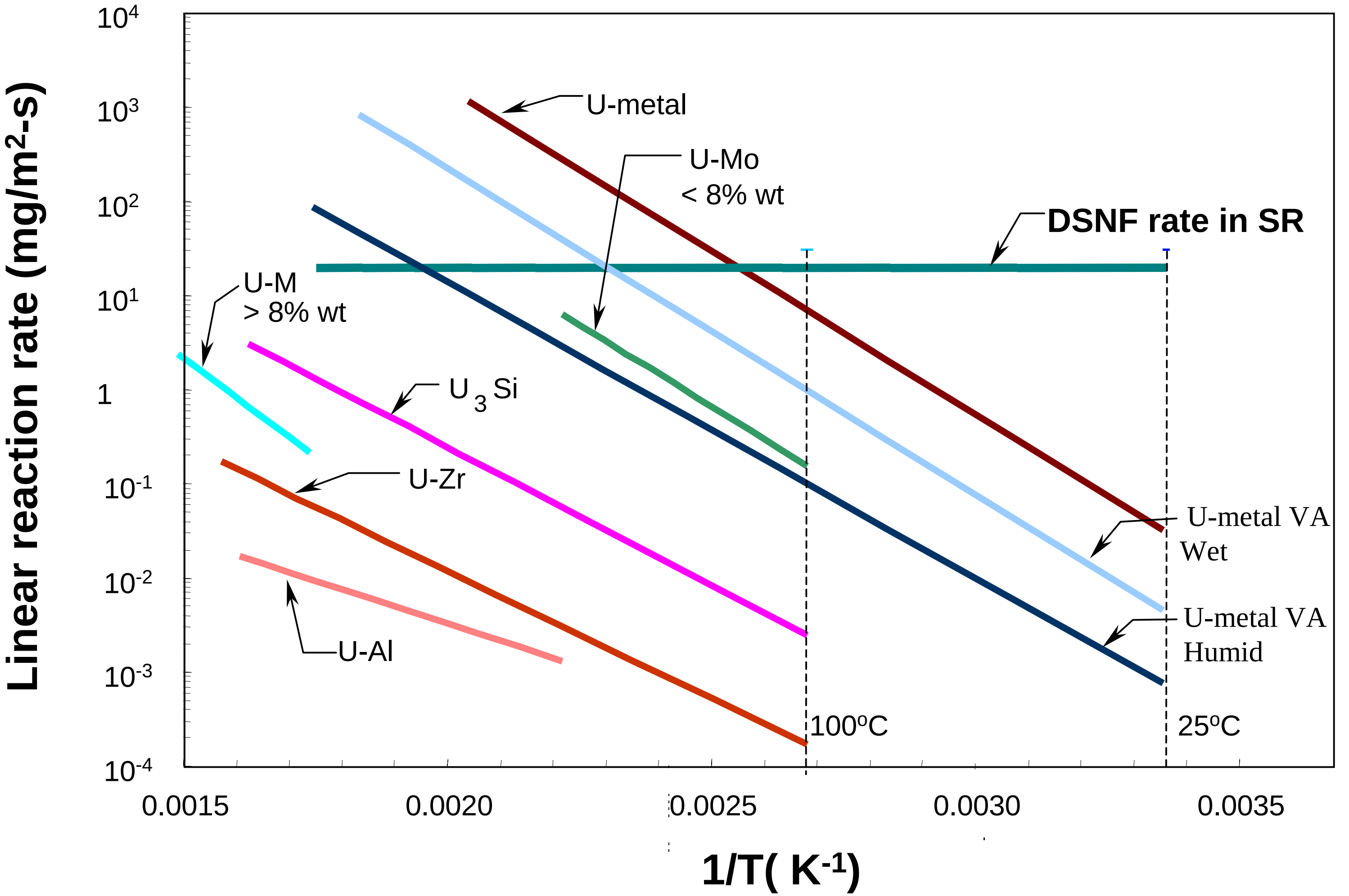
*** Corrosion of steel releases sulfur which can lower pH.**

CSNF Degradation Rates in SR Similar to Rates in VA



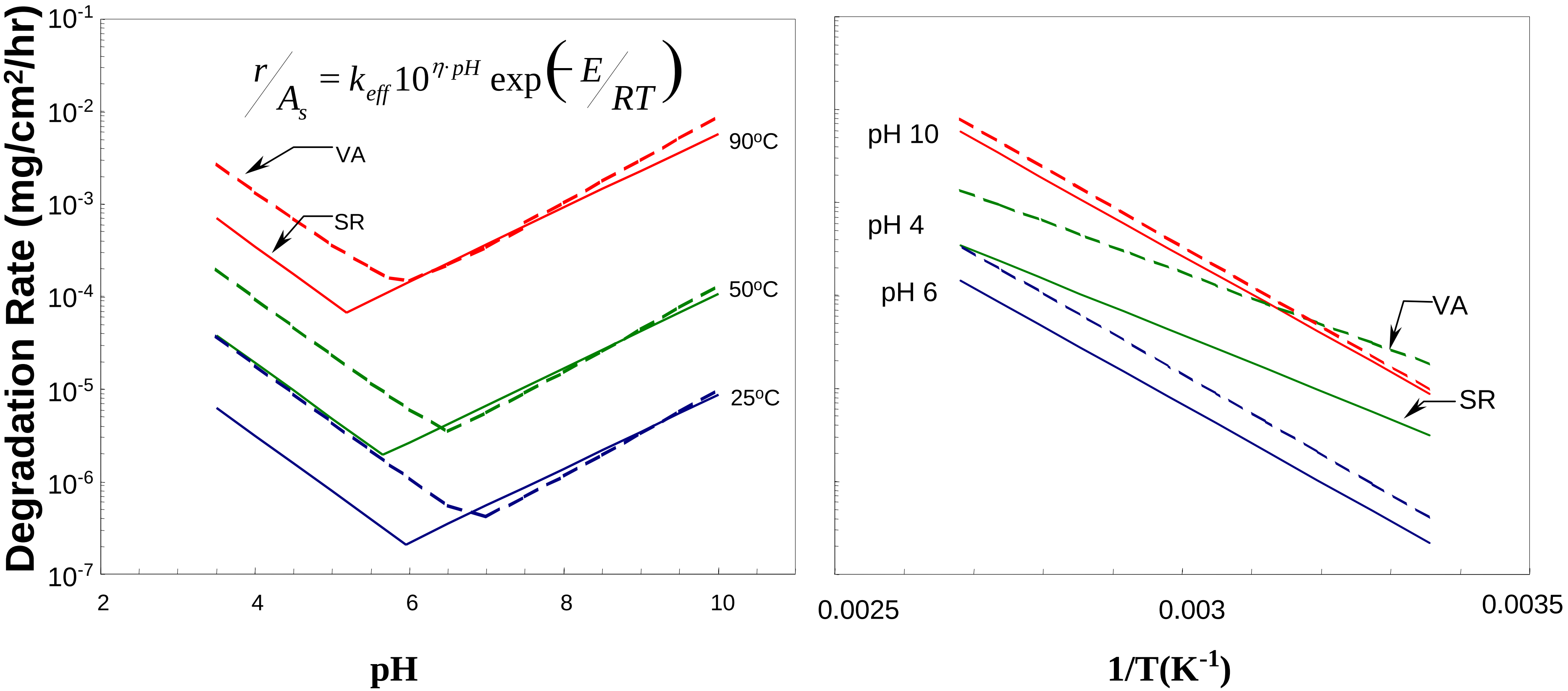


Constant Degradation Rate Used for DSNF Category Bounds all Measured Degradation Rates

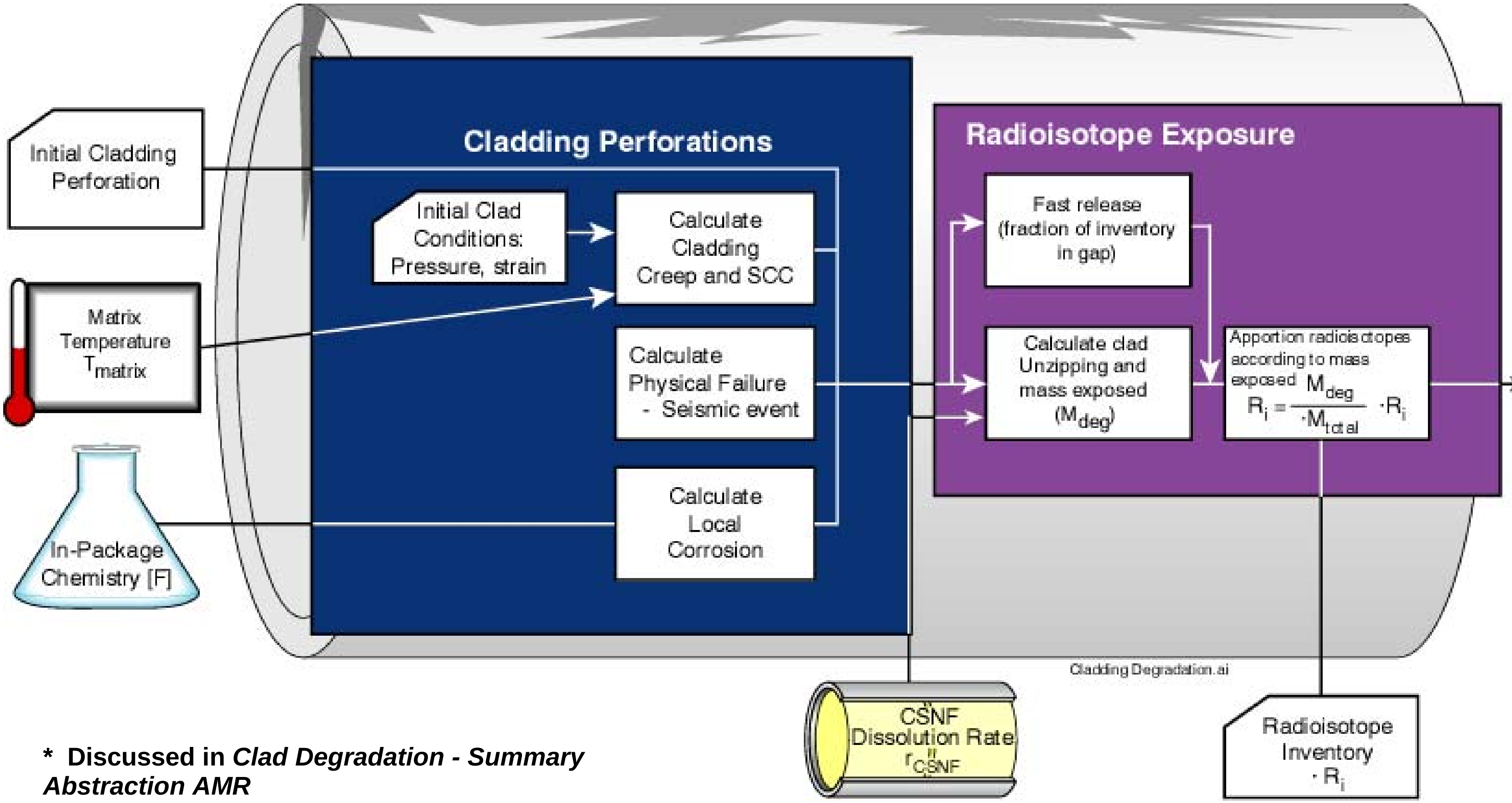


HLW Reaction Rates in SR similar to VA

- **HLW rate less than CSNF degradation except at high pH and high temperature**
  - **HLW rate bounds stage I, II, III degradation rates**

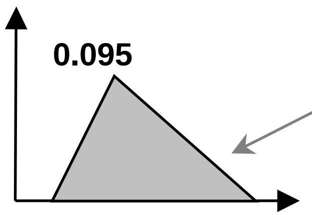


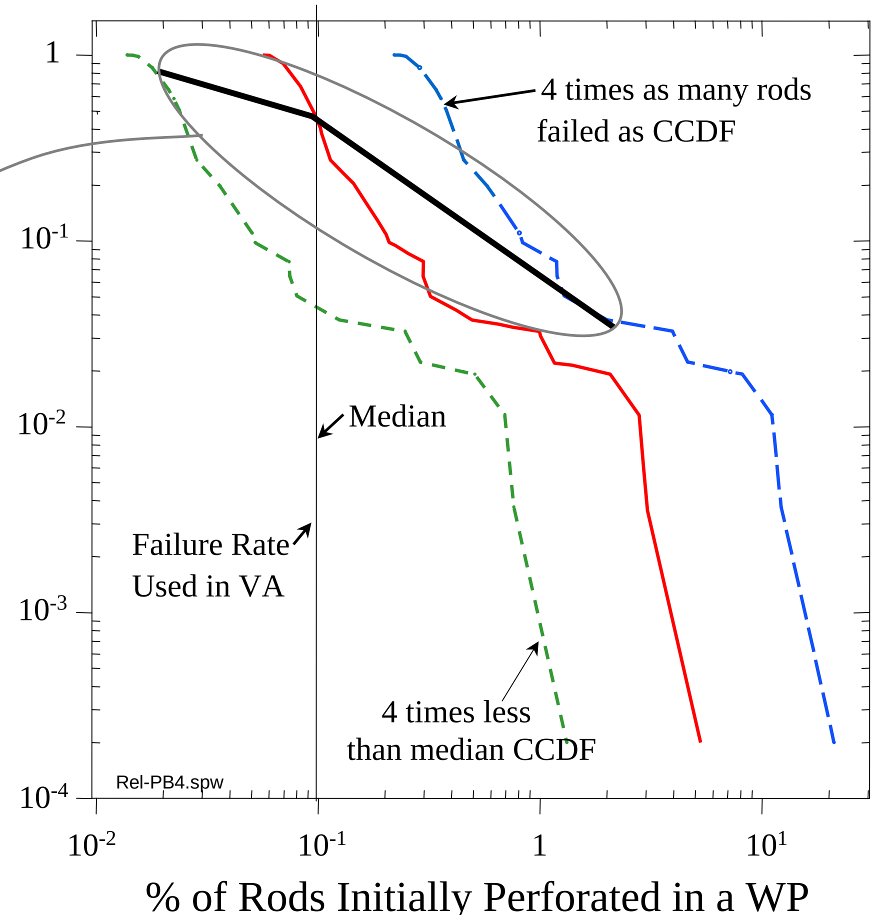
Cladding Degradation* Consists of Two Steps: Perforation and Unzipping



* Discussed in ***Clad Degradation - Summary Abstraction AMR***

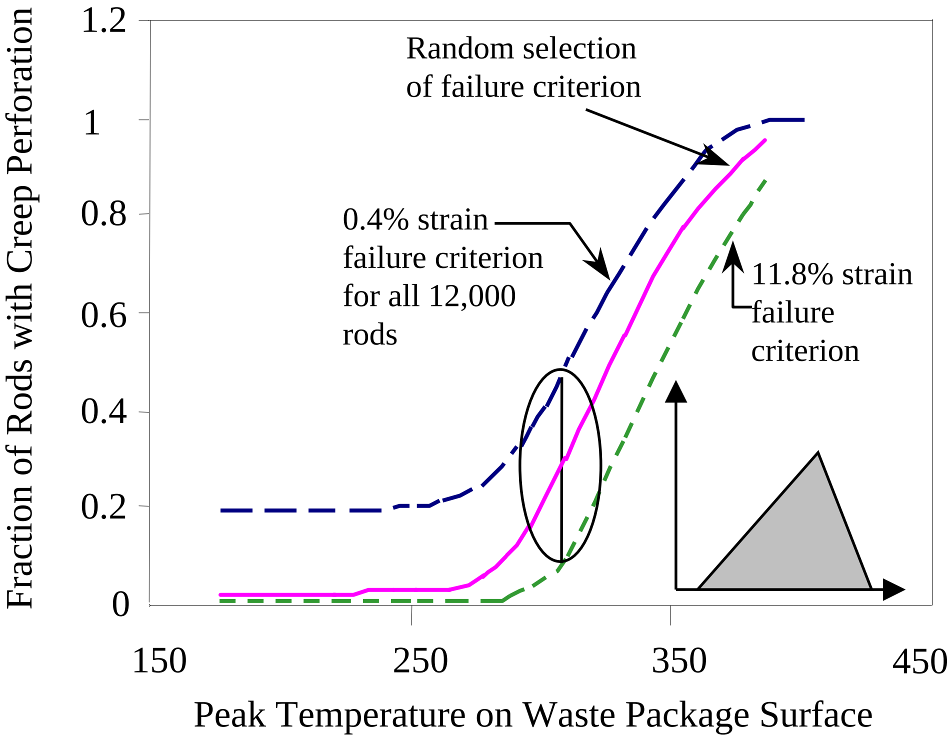
Cladding Perforations* before Receipt based on NRC Contractor Report (1969-1985) and Literature from 1985-1995

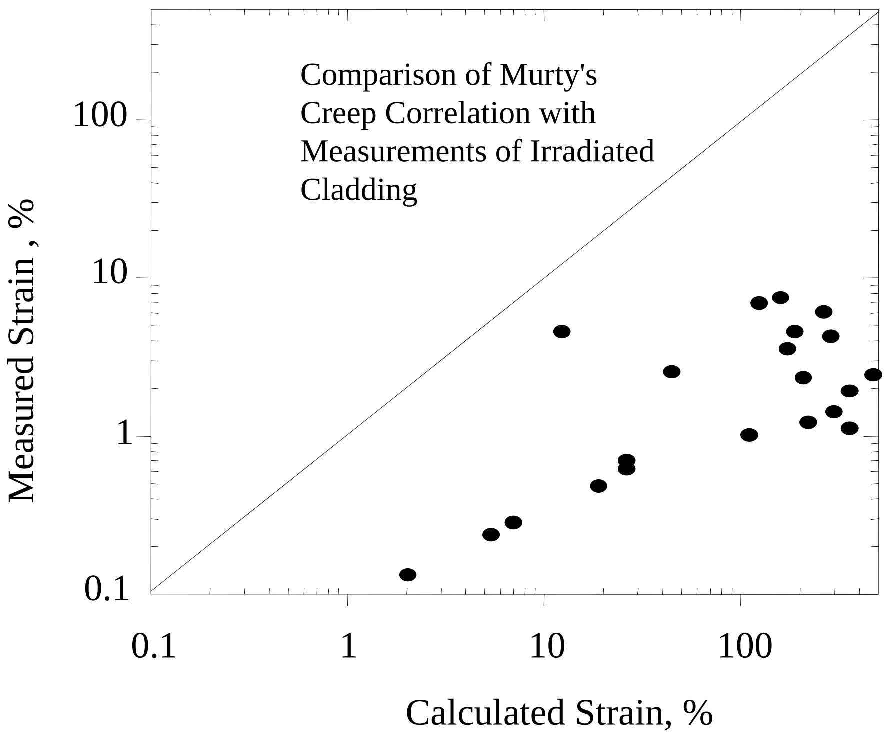




* **Discussed in Initial *Cladding Condition AMR***

In TSPA-SR, Perforation from Cladding Creep Sampled Between Analytical Estimates





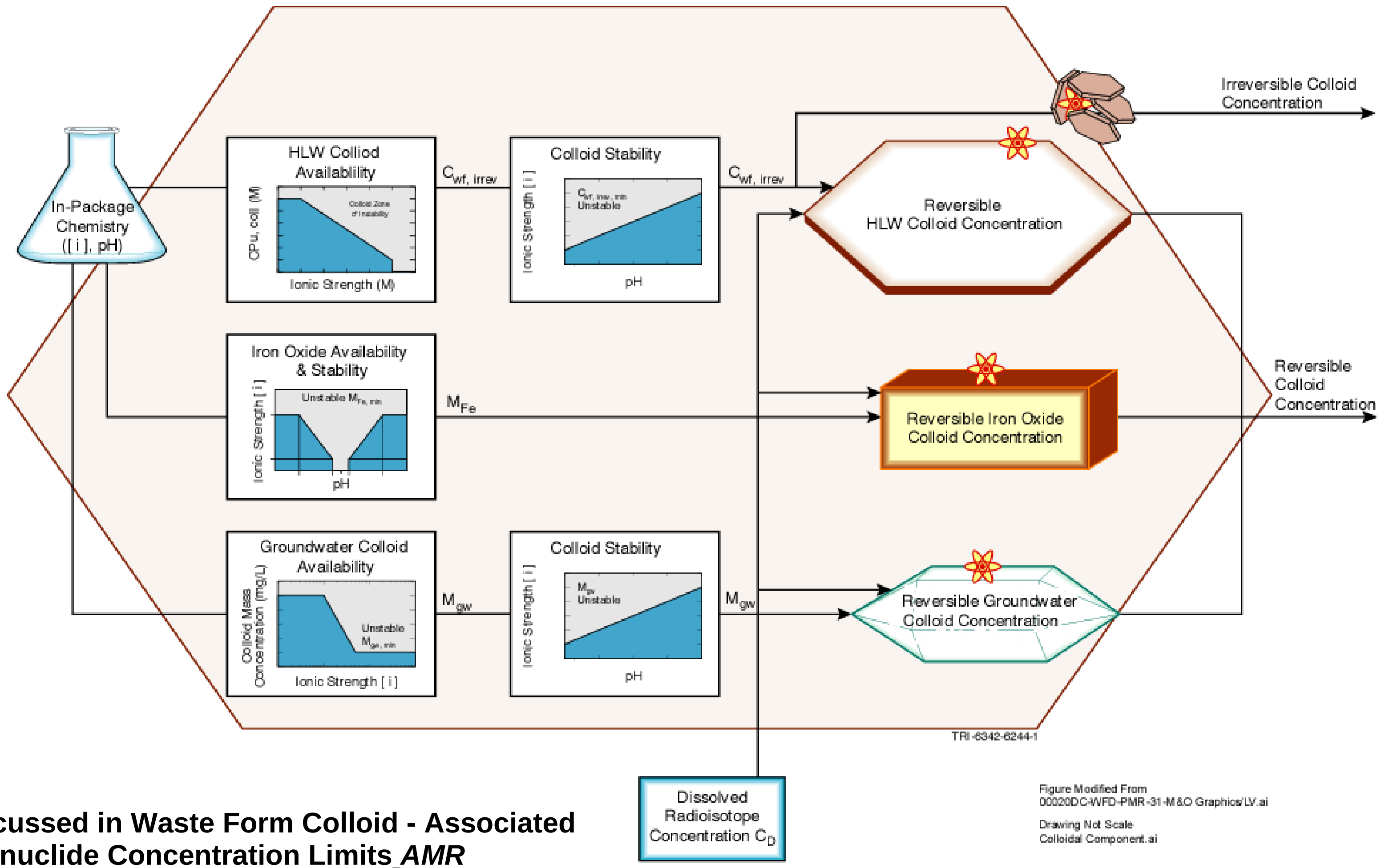
Perforation of Cladding* by Localized Corrosion

- To account for microvariation in chemistry, pitting is included since it is thought to be more likely to occur relative to other localized corrosion mechanisms
- Fraction of perforated rods conservatively assumed to be proportional to seepage of water into WP

$$f_{clad} = \frac{1}{2424 m^3} \bullet q_{seep} \Delta t_i$$

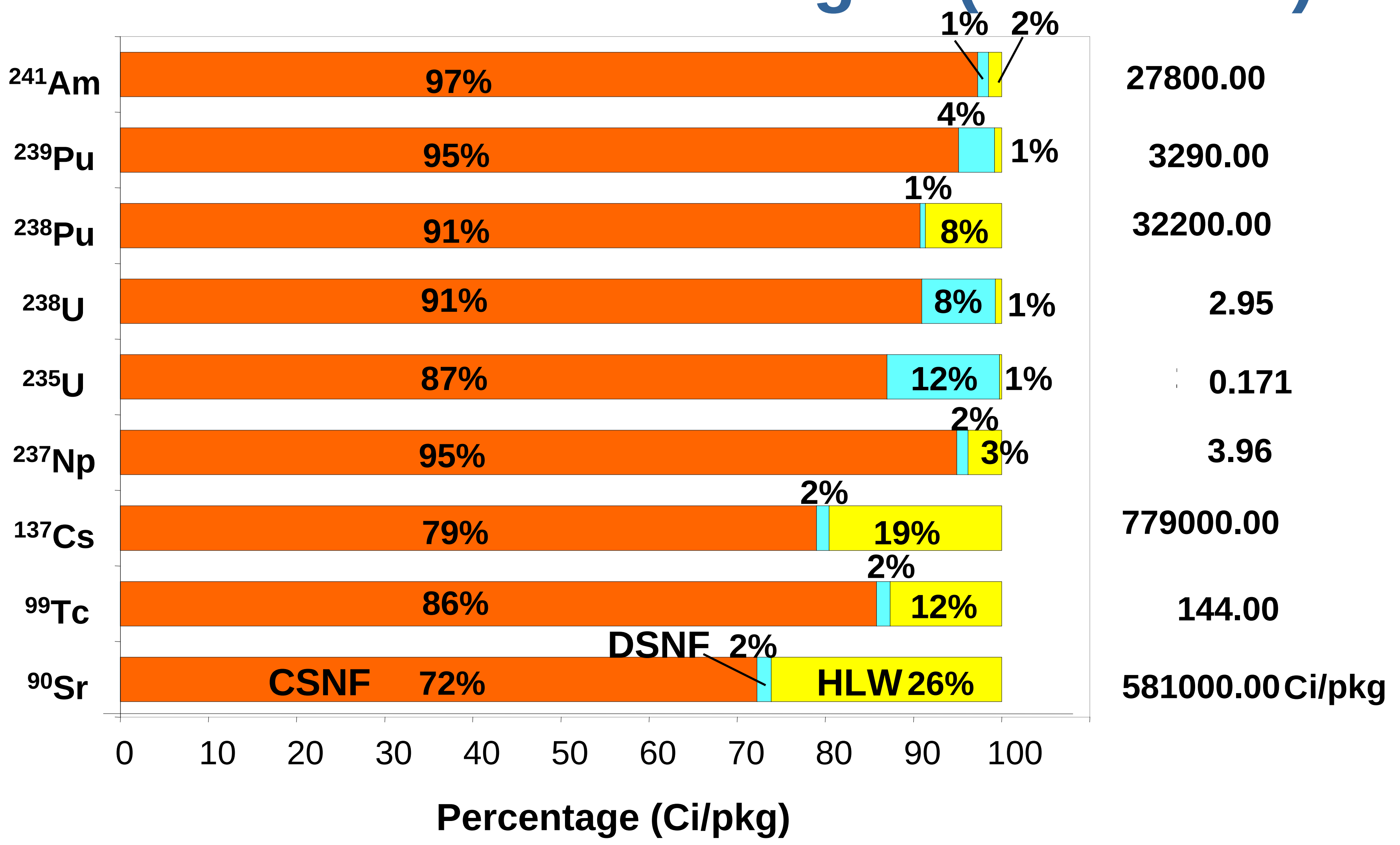
* Fluoride pitting discussed in ***Clad Degradation - Summary and Abstraction*** **AMR**

Colloidal Component* Evaluates the Colloid Concentration on Three Types of Colloids: Waste, Rust and Natural

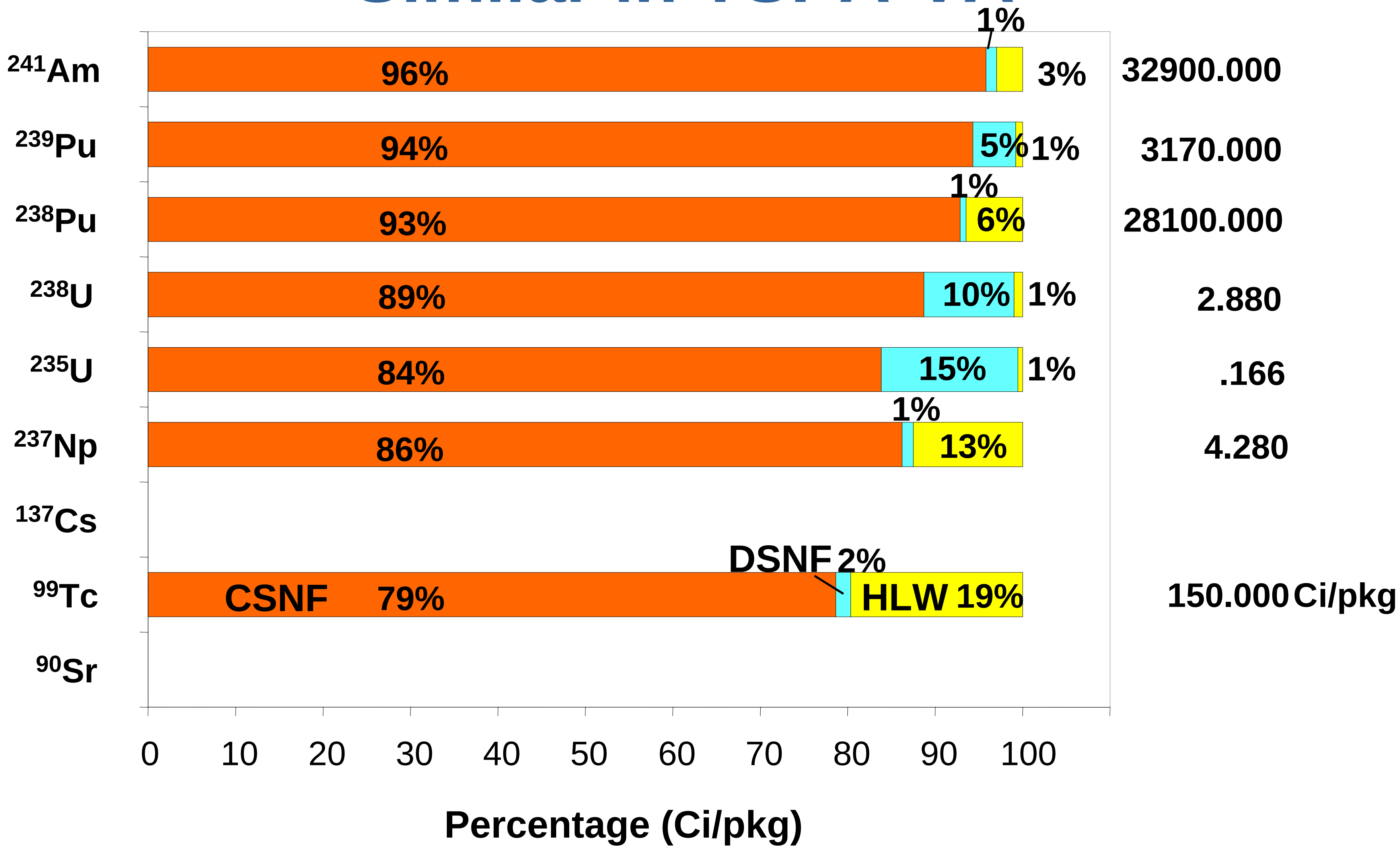


*** Discussed in Waste Form Colloid - Associated Radionuclide Concentration Limits *AMR***

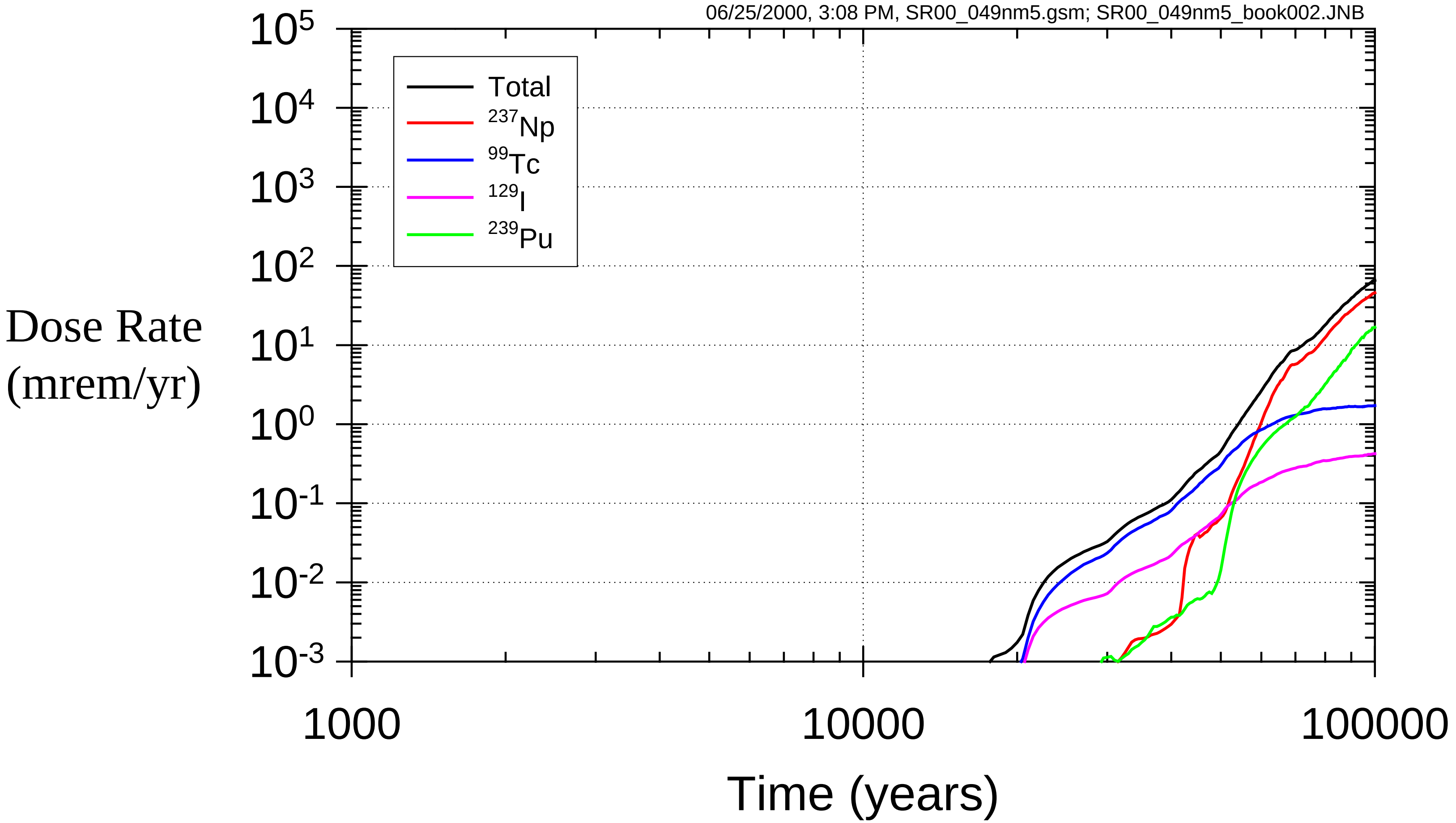
Highest Percentage of Radioisotopes Reside in CSNF Packages (TSPA-SR)



Percentage of Radioisotopes in Packages Similar in TSPA-VA



^{99}Tc and ^{237}Np Contribute Most to the Dose



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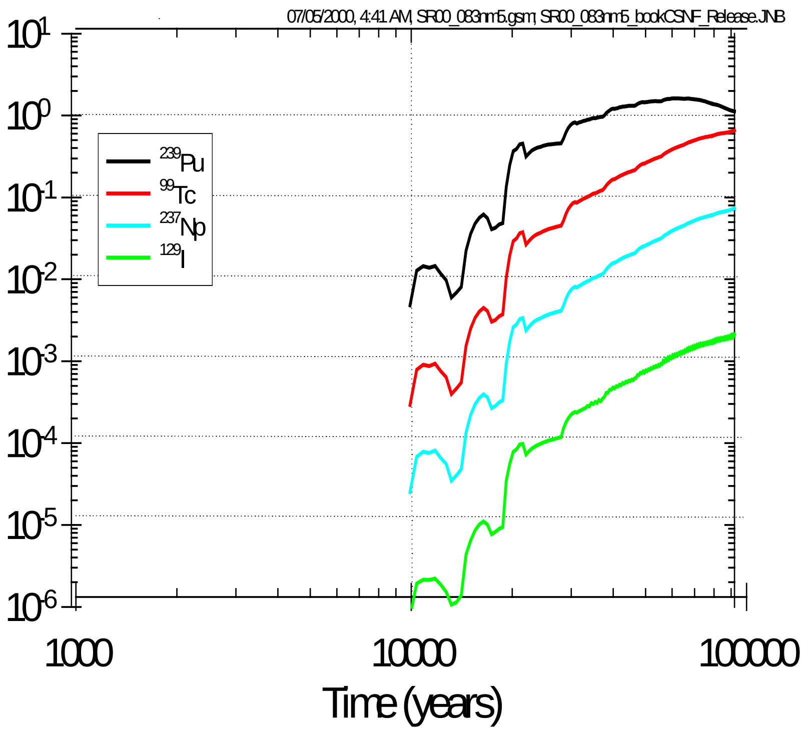
Rates of Release from CSNF Slightly Larger than from Co-Disposal Packages

Mean Release Rates from Waste Matrix (Ci/yr)

CSNF



Co - Disposal