



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

Development of Components of the Waste Form Degradation Model and Relationship with NRC Key Technical Issues

Presented to:

**NRC/DOE Technical Exchange on Total System
Performance Assessment (TSPA) for Yucca Mountain
San Antonio, Texas**

Presented by:

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June 6, 2000

YUCCA
MOUNTAIN
PROJECT



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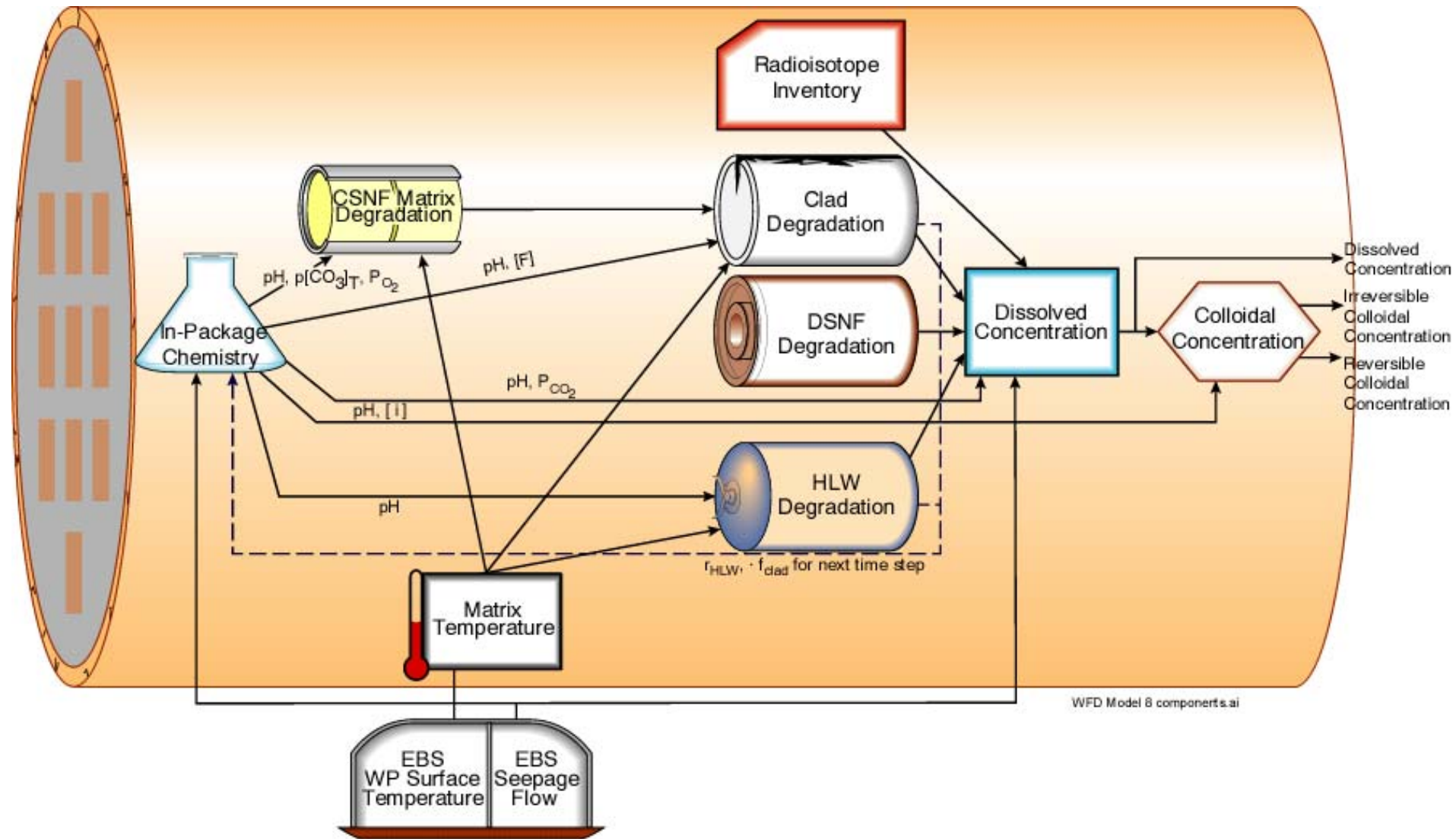
Waste Form Degradation Model

- Used for both nominal and volcanic scenarios in TSPA
- CLST IRSR primarily related to this model (October 12, 2000)
 - Subissue 3 (CLST3): CSNF degradation
 - Subissue 4 (CLST4): HLW degradation
- ENFE IRSR (September 7, 2000)
 - Subissue 3 (ENFE3): Coupled thermal-hydrological chemical coupling
- TSPAI IRSR
 - Subissue 3 (TSPAI3): Model abstraction
 - ♦ ENG3: Chemistry of water contacting waste
 - ♦ ENG4: RN release rates and solubility limits
 - Subissue 2 (TSPAI2): Scenario analysis



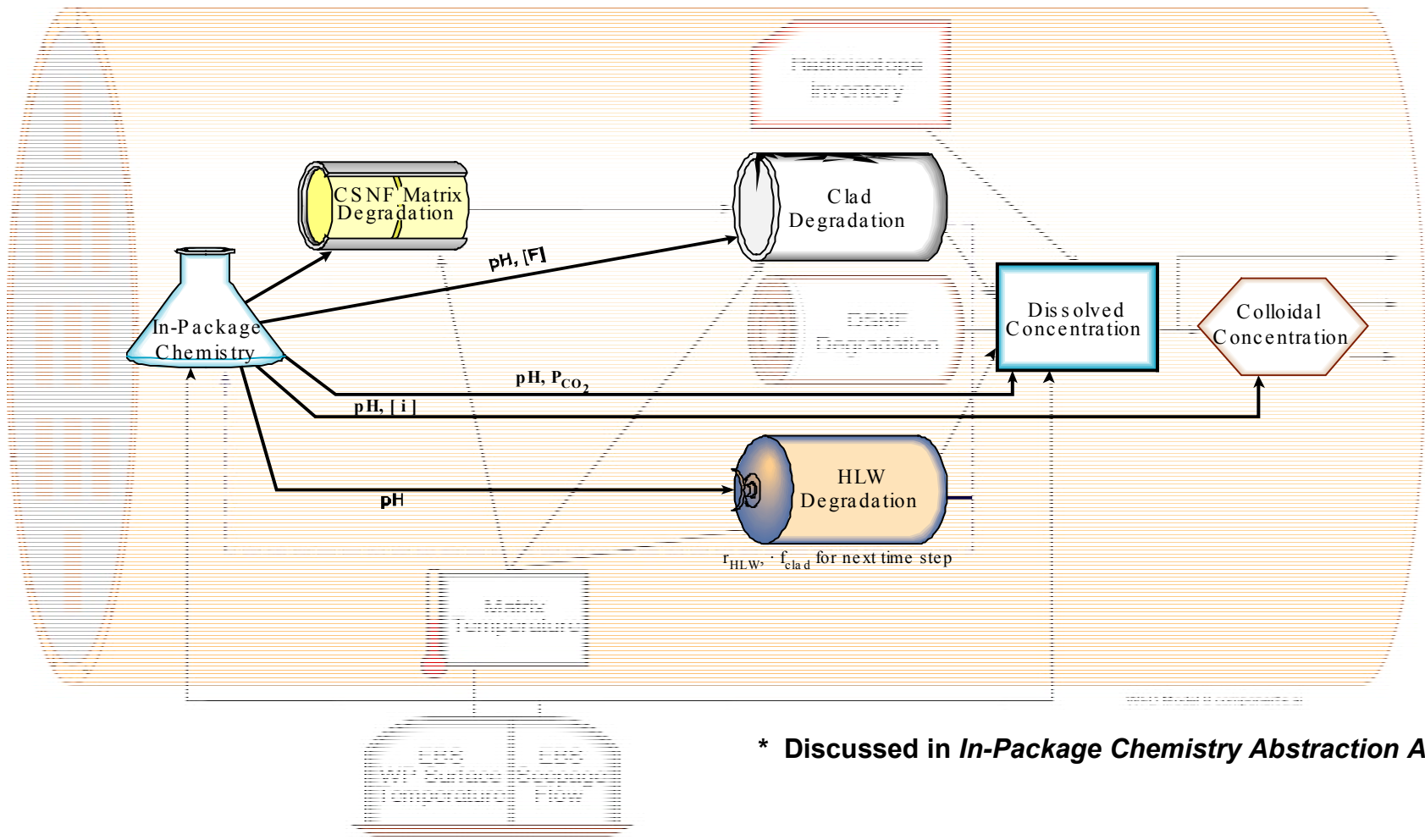
* Discussed in *Waste Form Degradation PMR*

Waste Form Degradation Model has Eight Components



In-Package Chemistry Component Couples to Five Other Components

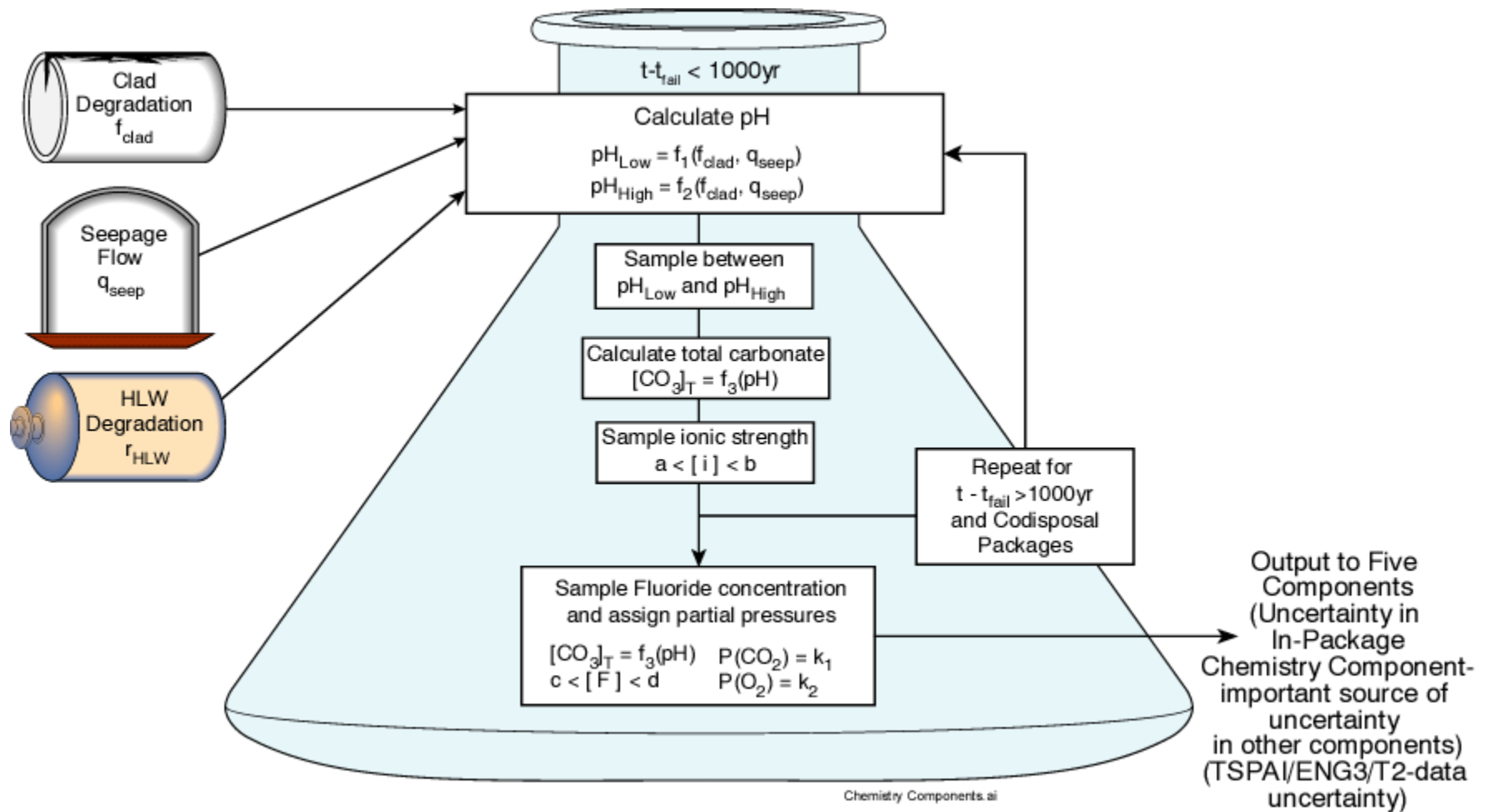
- Addresses thermal-hydrological-chemical coupling issue in ENFE3
- Addresses T5 (integration criterion) of TSPAI/ENG3 model abstraction
- Uncertainty in chemistry important source of uncertainty in 5 other components (addresses T2-data uncertainty)



* Discussed in *In-Package Chemistry Abstraction AMR*

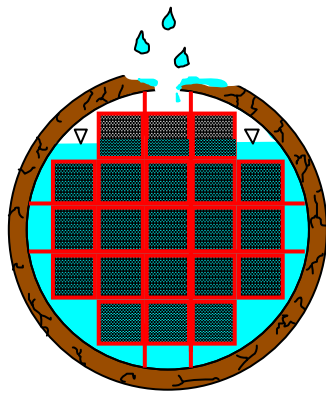
In-Package Chemistry Component

Estimates pH, Calculates $[\text{CO}_3]_T$, and Samples $[i]$ and $[F]$

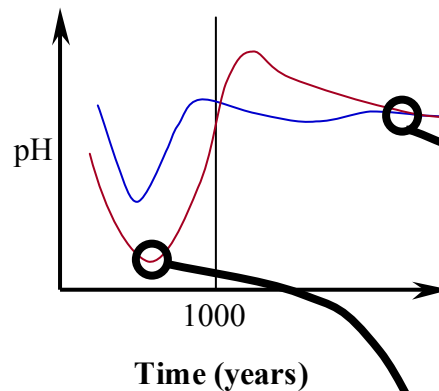


In-Package Chemistry Component Developed from Regression Analysis on EQ3/6 Runs

- Addresses TSPA13/ENG3 and ENG4/TI, T4, and T5
- Addresses CLST3 and its concern that influence of corrosion products on waste degradation be evaluated

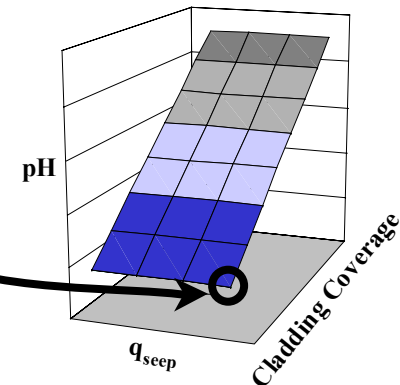
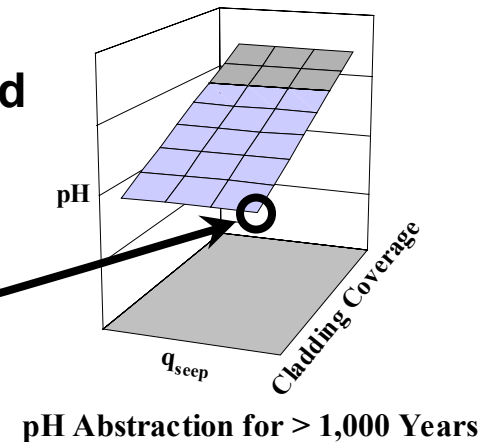


CSNF Conceptual Model



EQ3/6 Runs Varying:

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



* Corrosion of steel releases sulfur which can lower pH; the CLST IRSR specifically notes influence of corrosion products on CSNF and HLW matrix degradation

CSNF Matrix Degradation Component* Based on Regression of Laboratory Experiments

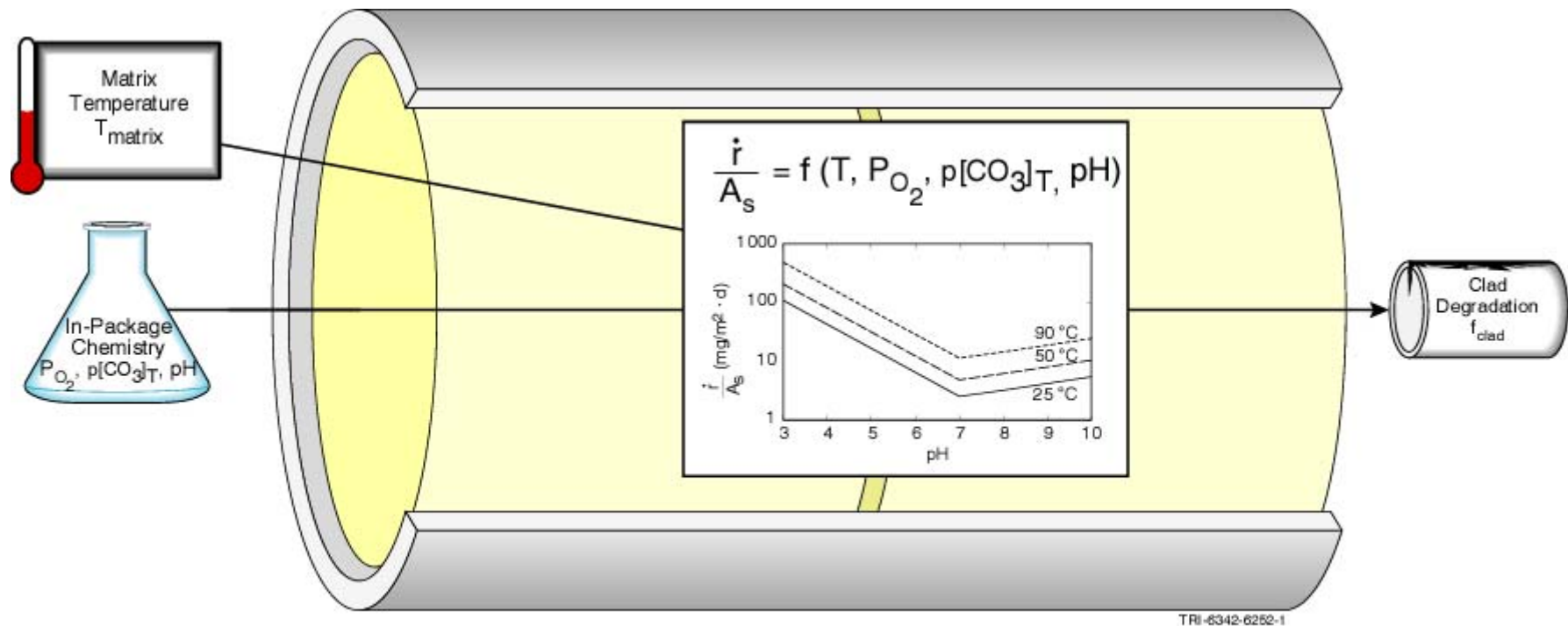
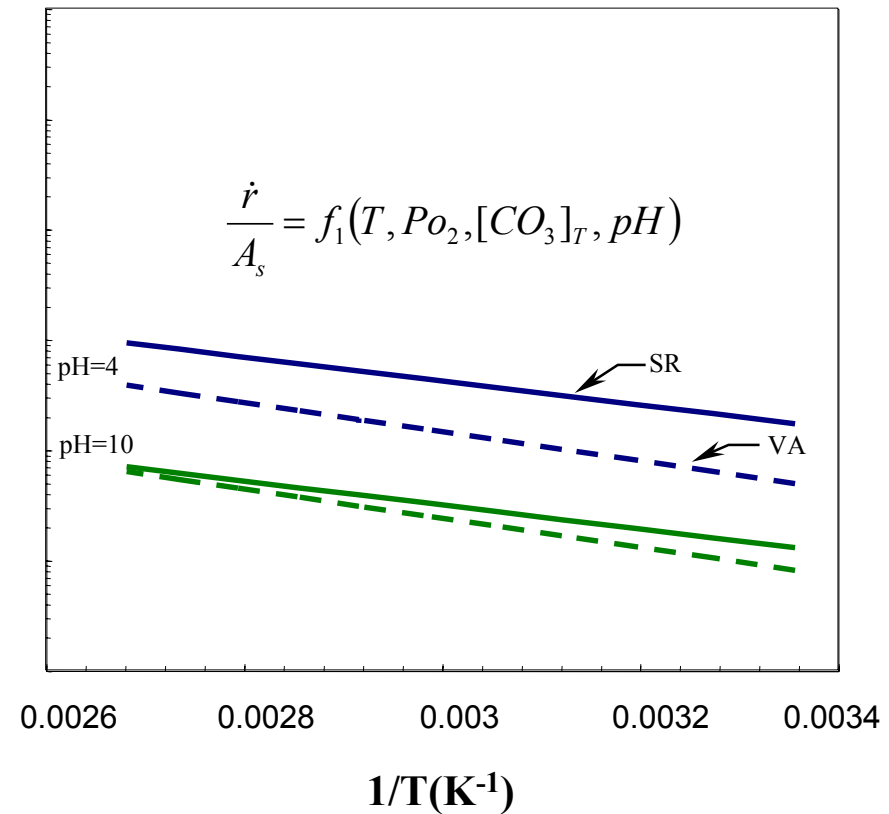
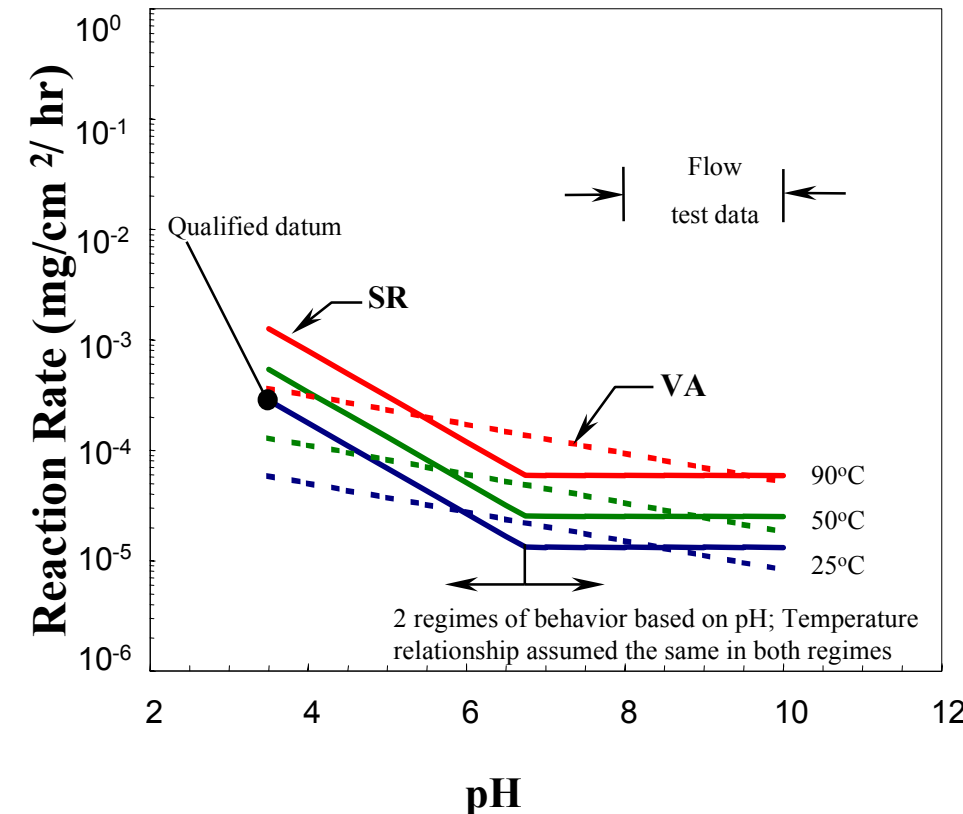


Figure Modified From
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Drawing Not To Scale
CSNF Degradation Model.ai

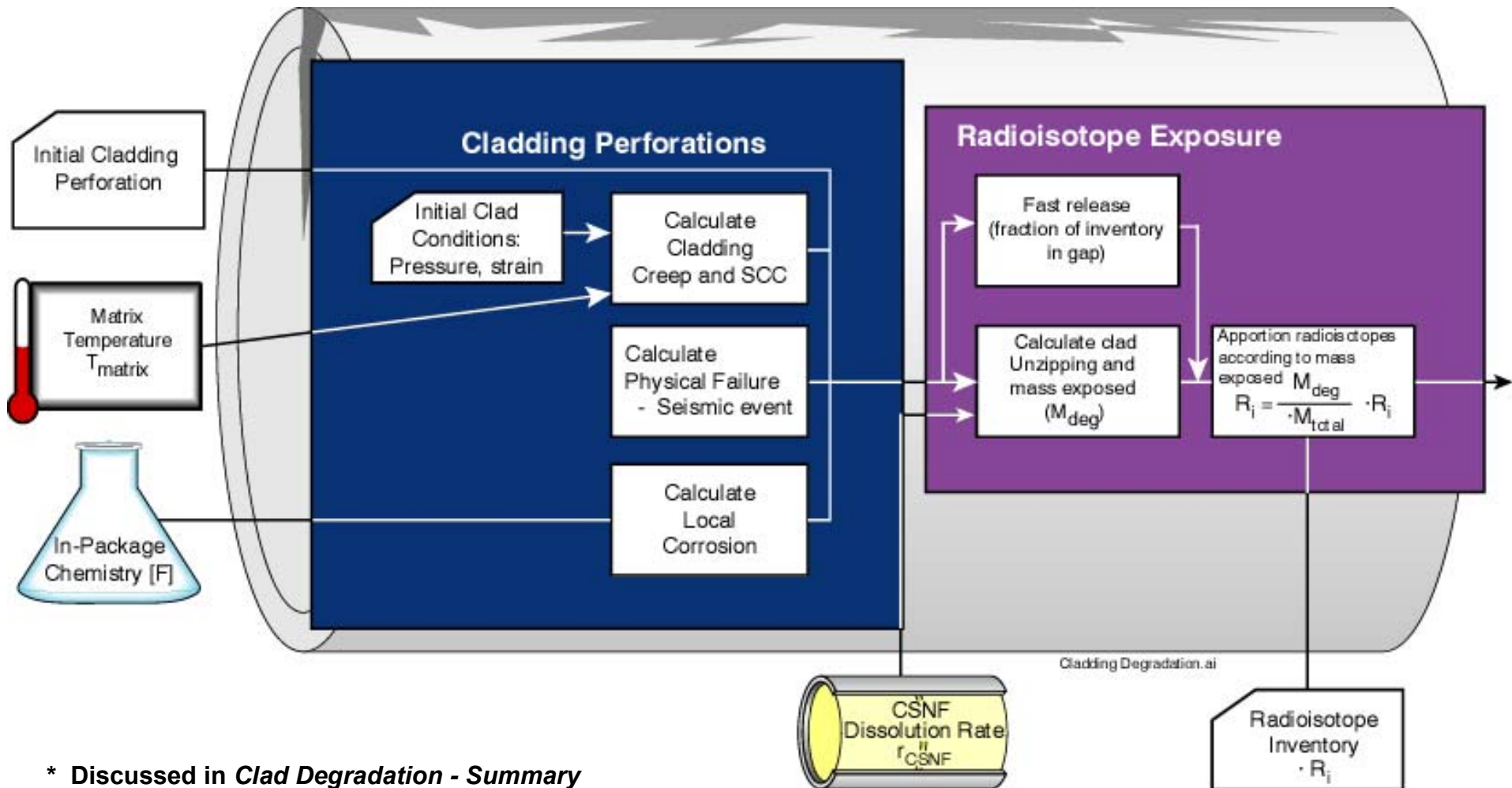
* Discussed in CSNF Waste Form Degradation Summary and
Abstraction AMR

CSNF Degradation Rates in SR Similar to Rates in VA

- Addresses CLST3: CSNF degradation and influence of T, pH, $[CO_3]_T$
- Addresses TSPAI/ENG3/T1 and T4 (not T2 - data uncertainty - since in chemistry)

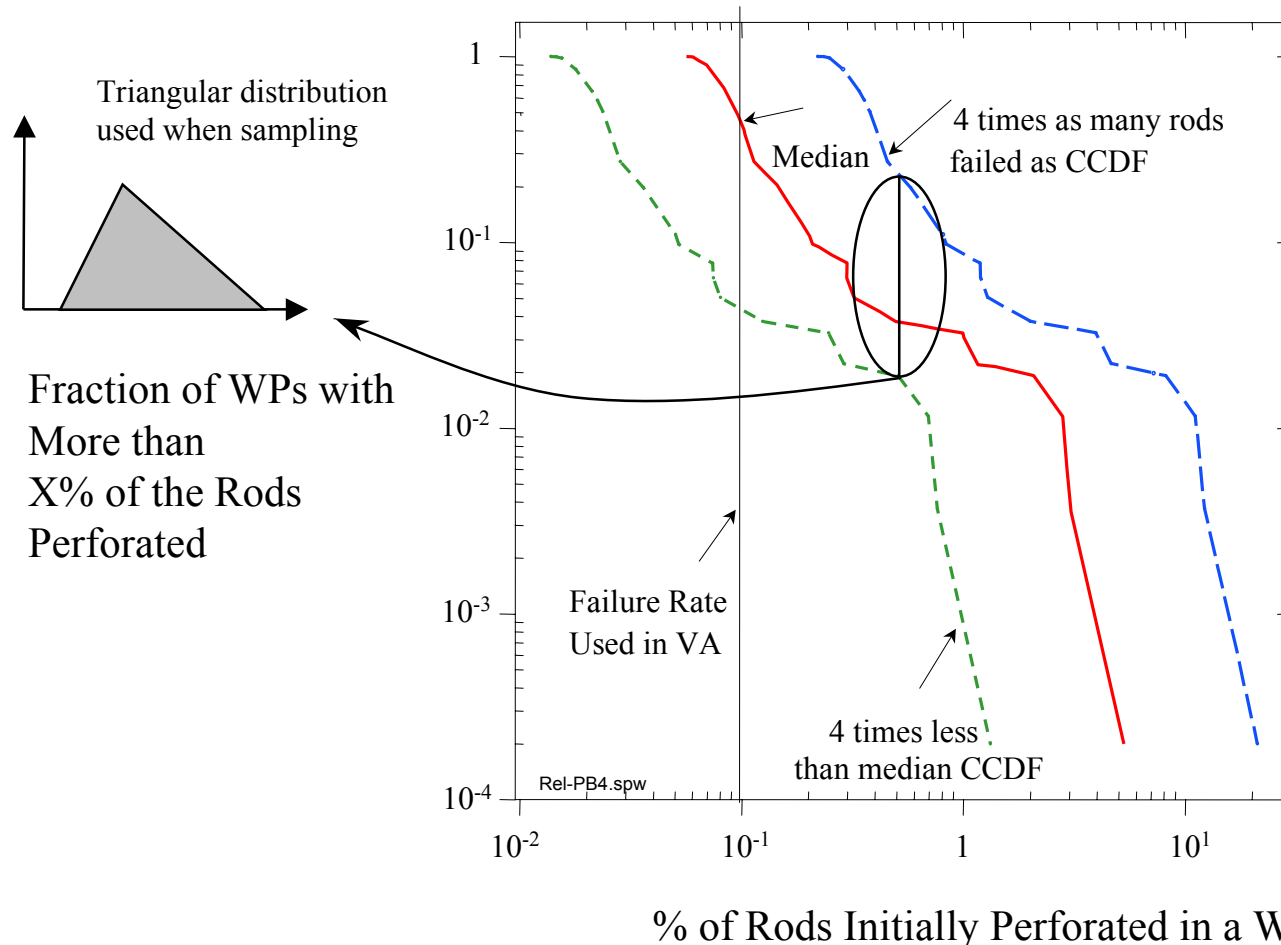


Cladding Degradation* Consists of Two Steps: Perforation and Unzipping



* Discussed in *Clad Degradation - Summary Abstraction AMR*

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

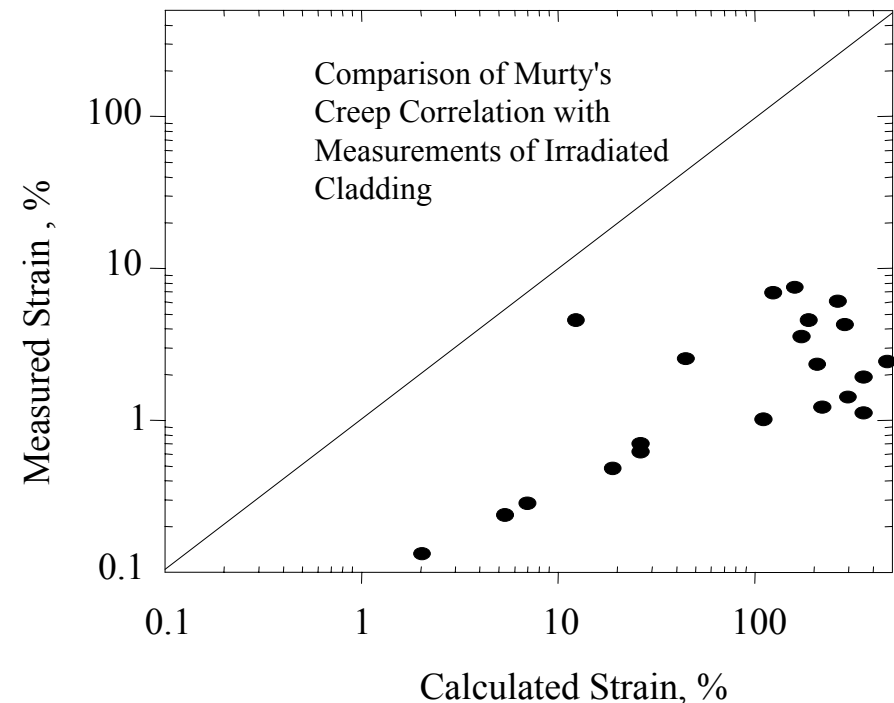
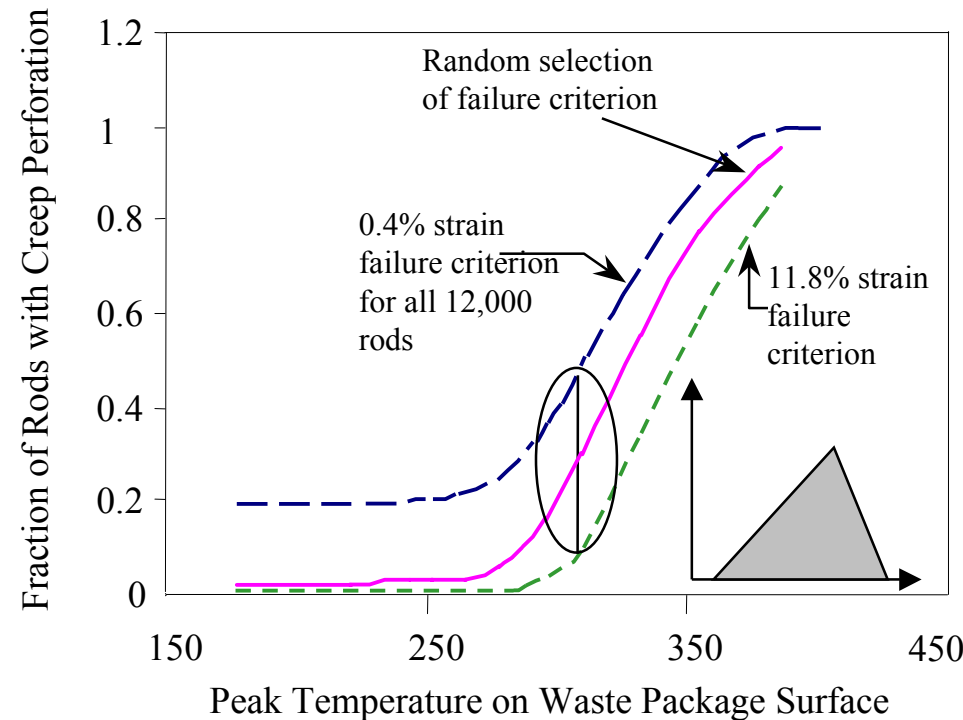


- Addresses CLST3 request for evaluation of cladding damage during operation, storage, and transportation
- Related to TSPAI/ENG4 issue on RN release

* Discussed in Initial *Cladding Condition* AMR

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

- Addresses request in CLST3 for evaluation of creep rupture models (alternate DCCG creep model addressed as FEP)
- Related to TSPA/ENG4



Perforation of Cladding* by Localized Corrosion

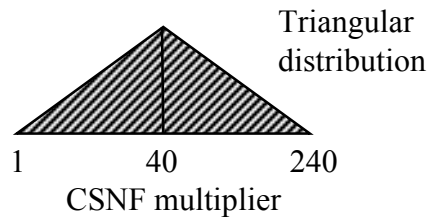
- To account for microvariation in chemistry, pitting is included since it is thought to be more likely to occur relative to other localized corrosion mechanisms
- Fraction of perforated rods conservatively assumed to be proportional to seepage of water into WP

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

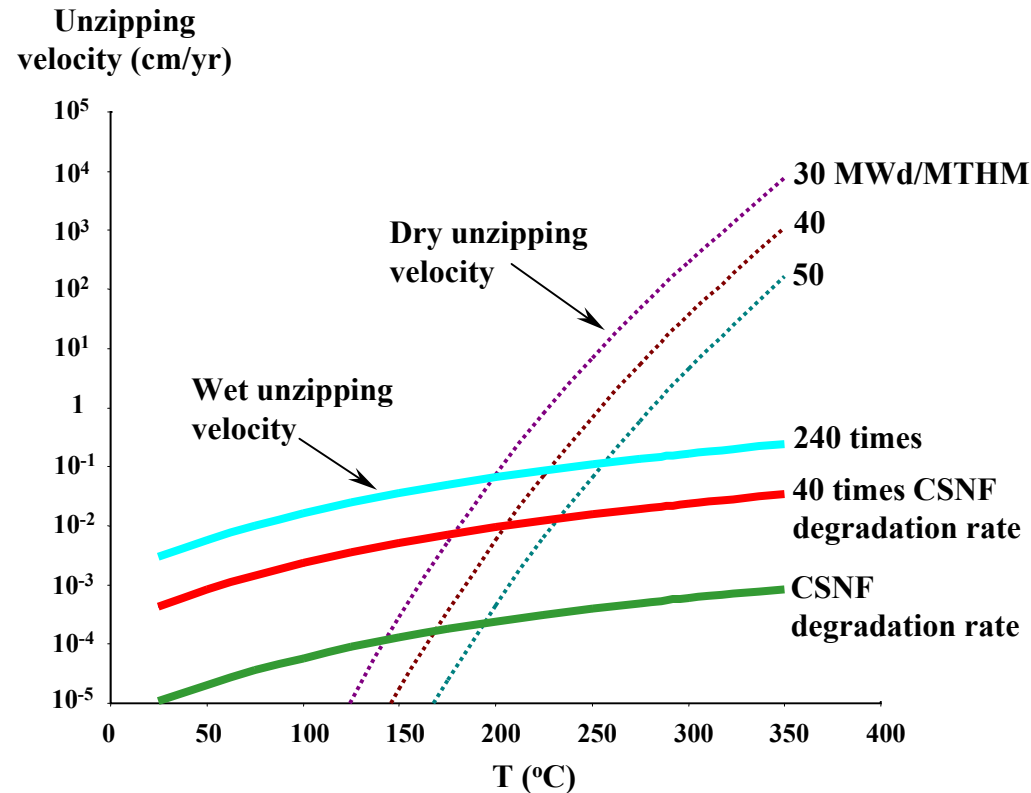
- Both CLST3 and TSPAI/ENG4 ask for evaluation of potential for pitting corrosion

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

Cladding Unzipping* Follows Perforation



- Addresses request in CLST3 to develop models for clad splitting
- Related to TSPAI/ENG4 discussion
- Unzipping rate in dry environment bounded by wet unzipping rate at temperatures anticipated in WP and thus included indirectly (no release, however, until water seeps into the container)
- Radioisotope releases from unzipping conservatively bound diffusive releases out of pinhole perforations



* Discussed in *Clad Degradation - Wet Unzipping*, *Clad Degradation - Dry Unzipping*, and *Clad Degradation - Summary and Abstraction AMRs*

Cladding Perforation from Several Mechanisms Evaluated (TSPA2-FEP analysis)

1. **Perforation from delayed hybrid cracking and hydrogen embrittlement screen out** - (discussed in *Hydrogen-Related Degradation of SNF cladding under Repository Conditions* AMR)
2. **In general, localized corrosion not predicted to occur for CSNF WP; thus, localized corrosion FEPs generally screen out**-(discussed in *Clad Degradation - Local Corrosion of Zirconium and its Alloys under Repository Conditions* AMR)

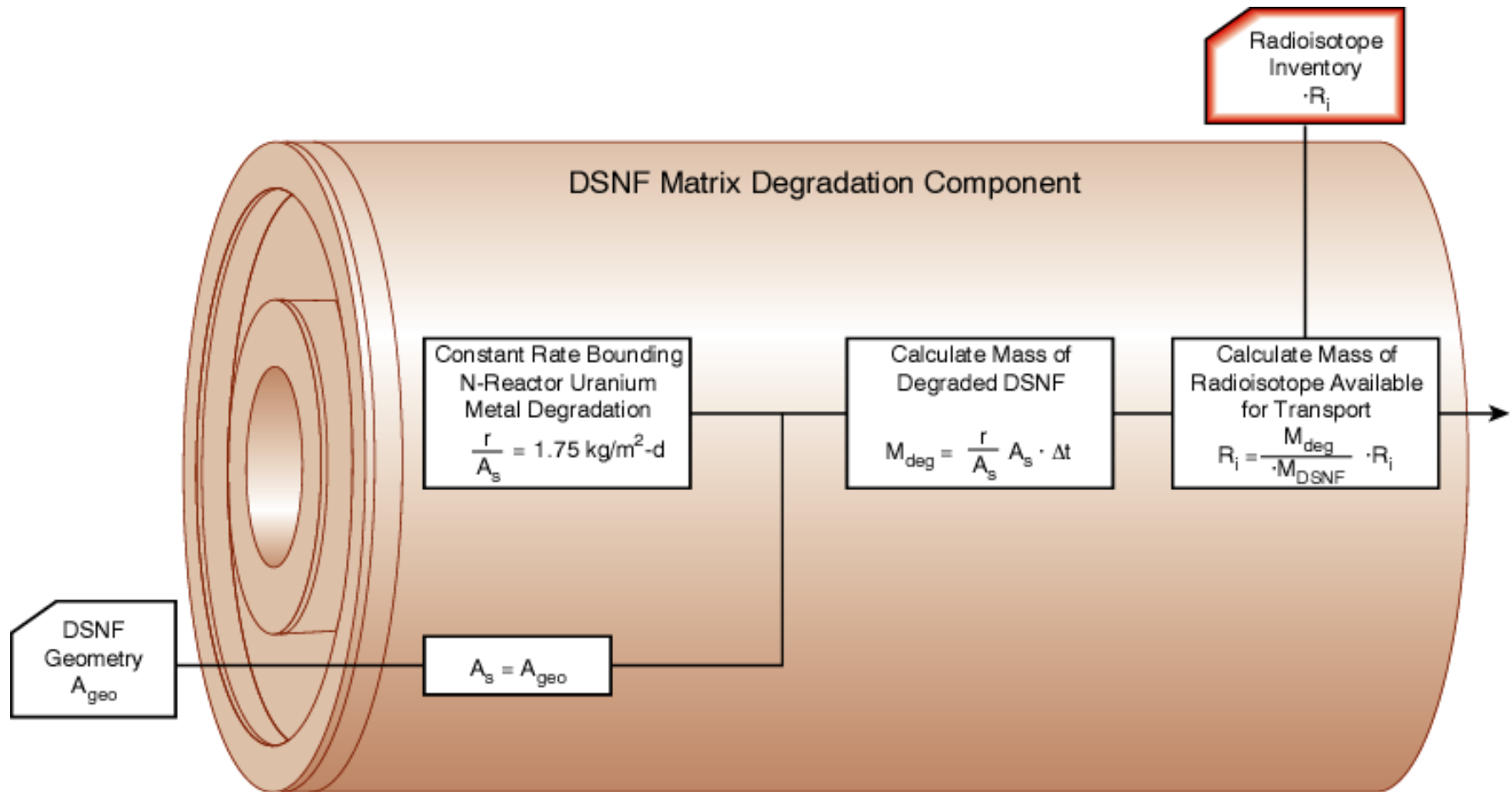
$$1 \times 10^{-4} < [F] < 9 \times 10^{-4} \text{ m}$$

$$[Cl] = 2 \times 10^{-4} \text{ m}$$

$$3.6 < \text{pH} < 8.1$$

3. **Generalized corrosion screened out** - (discussed in *Clad Degradation - FEPs Screening Arguments* AMR)
4. **Unlike TSPA-VA, cladding failure from rockfall screened out since other perforation mechanisms more likely now that WP life has been extended**
5. **TSPA-SR does include perforation from severe earthquake (frequency of $10^{-6}/\text{yr}$)** - (discussed in *Clad Degradation - Summary and Abstraction* AMR)

DSNF Degradation Component* uses Constant Degradation Rate

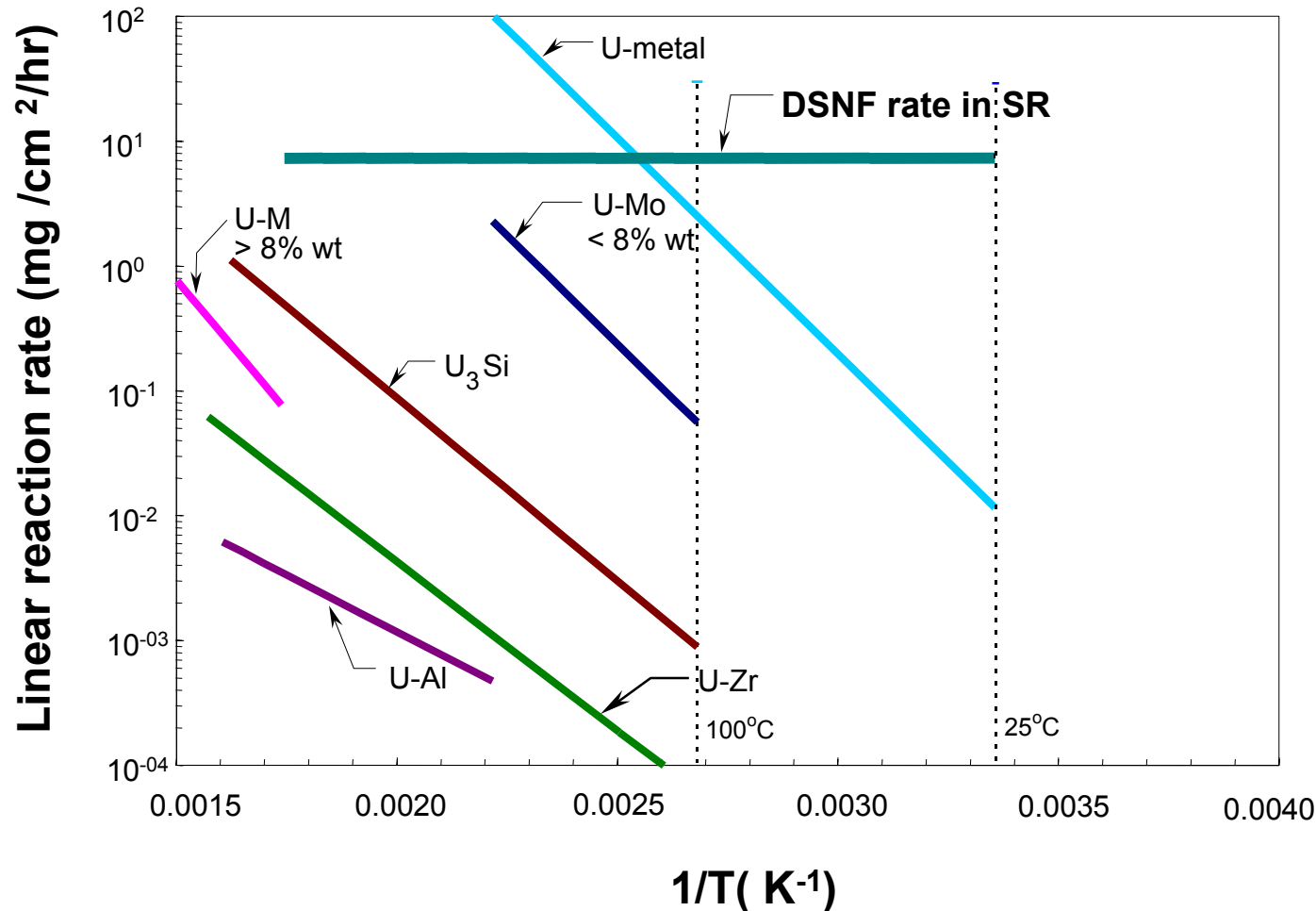


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Figure Modified From
00020DC-WFD-PMR-24-M&O Graphics/LV.ai

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

Constant Degradation Rate Used for DSNF

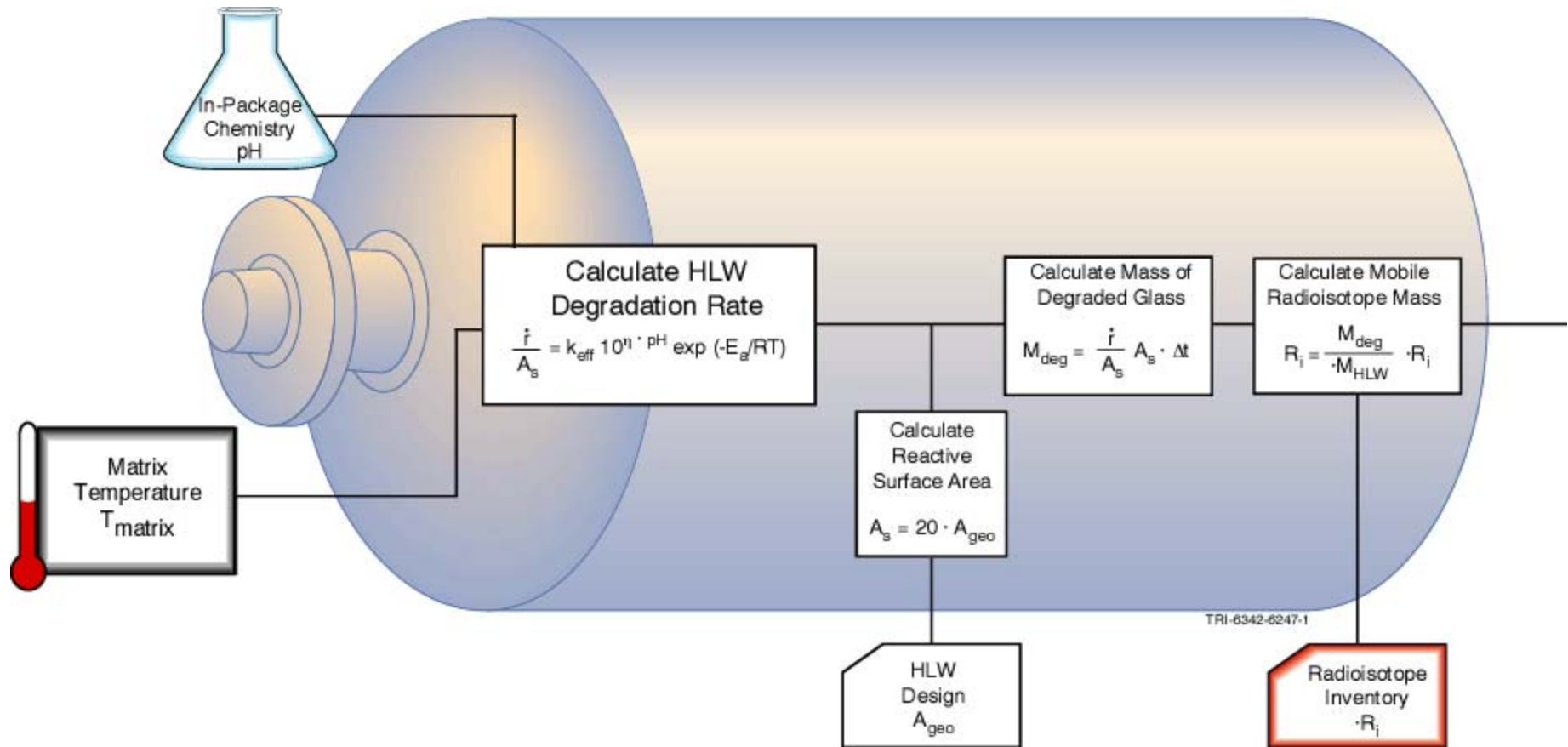
Category Bounds all Measured Degradation Rates



- Addresses TSPAI3/ENG4 issue on RN release

HLW Degradation Component Uses Arrhenius Rate Expression with First Coefficient Dependent on pH

- Addresses CLST4 and TSPAI/ENG4

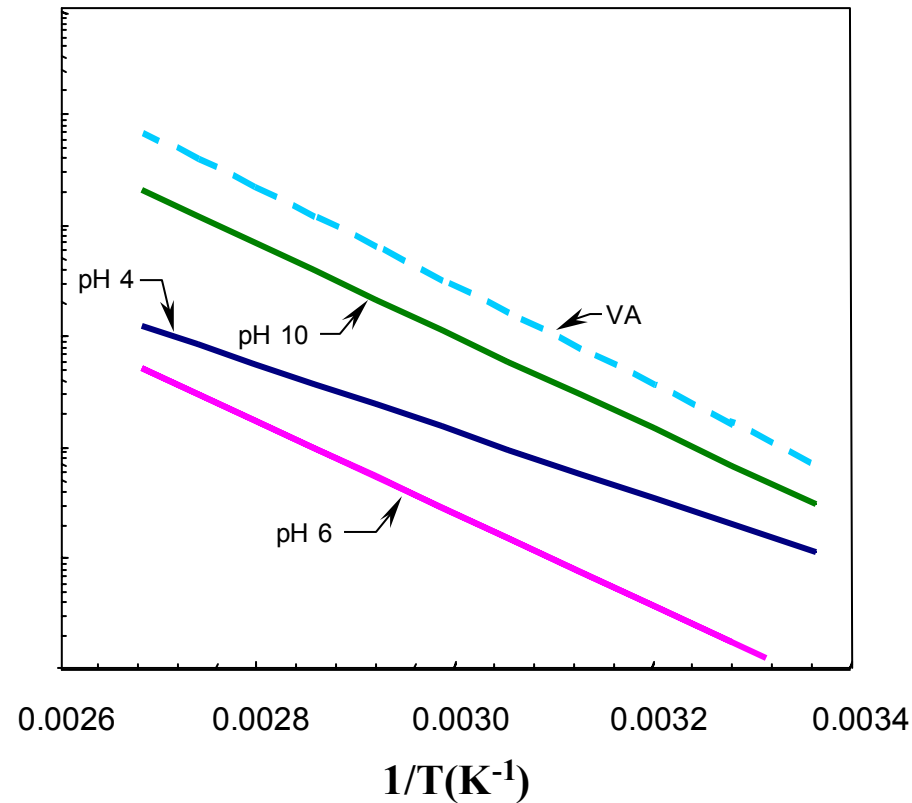
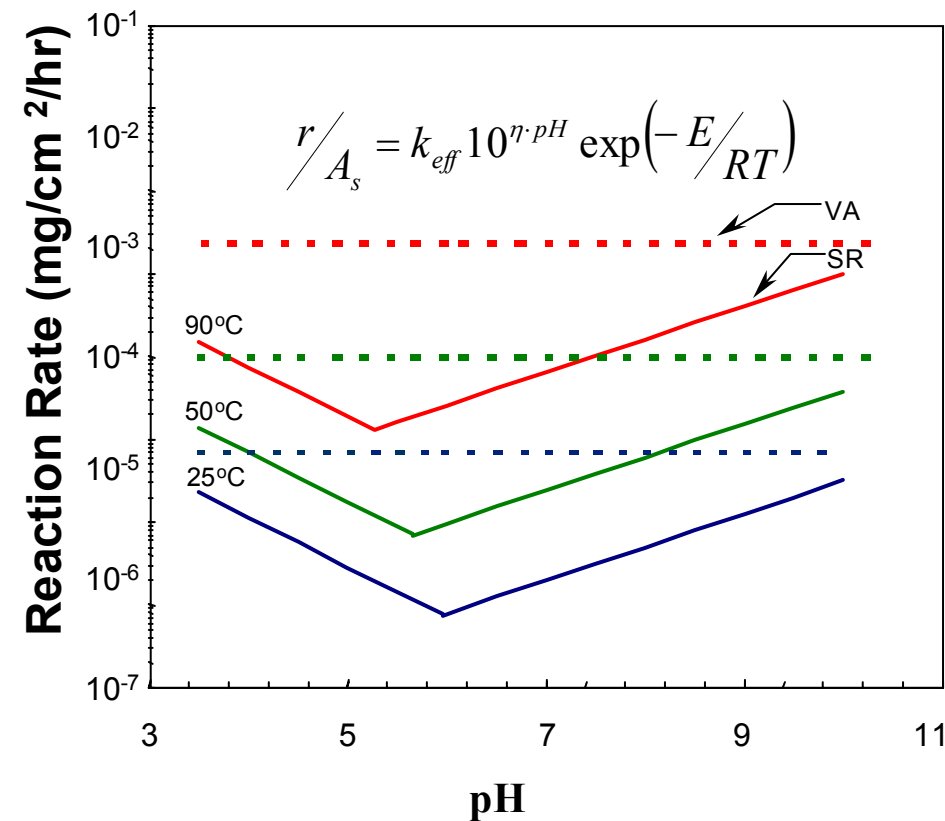


* Discussed in Defense High Level Waste Glass
Degradation-AMR

Figure Modified From:
00020DC-WFD-PMR-26-M&O Graphics/LV.ai
Drawing Not To Scale
HLW Degradation Component.ai

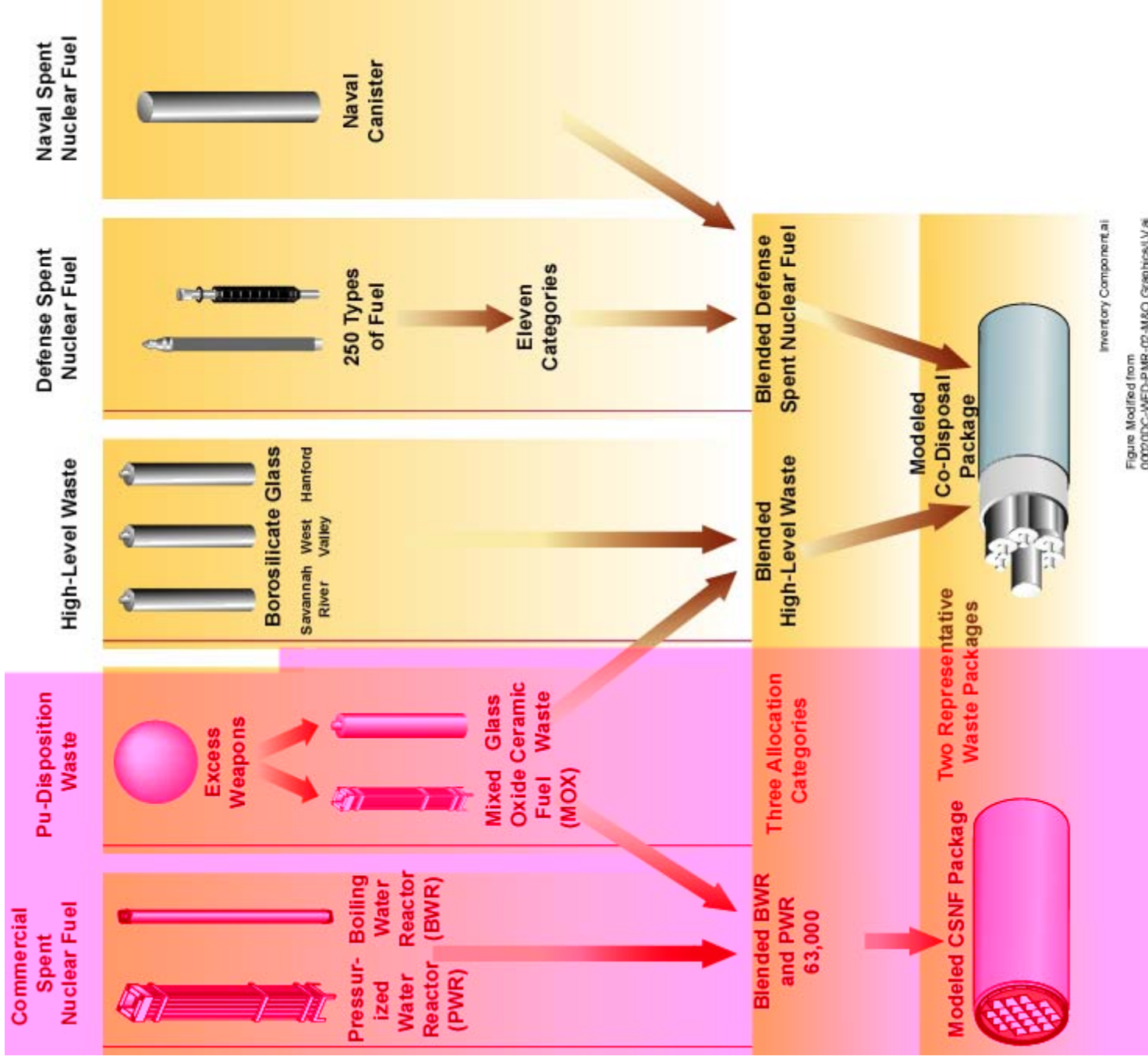
HLW Reaction Rates in SR similar to VA

- Addresses HLW degradation in CLST3 and RN release in TSPA13/ENG4
- HLW rate less than CSNF degradation except at high pH and high temperature
- HLW rate bounds stage I, II and III degradation rates

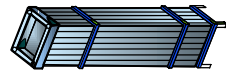


Inventory of Two Modeled Packages Based on Blending Inventory From Five Major Waste Types

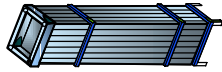
- Addresses request to update and justify inventory in CLST3 and TSPA/ENG4



Radioisotope Activity in a CSNF Waste Package is Based on Average Inventory in 5 Groups



Spent MOX
Fuel Assemblies



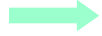
Spent PWR Fuel
Assemblies



Spent BWR
Fuel Assemblies

All of the CSNF Inventory (84,000 MTHM)

Grouping based on thermal load and critically potential (B. Colton CAL-MGR-MD-00001)



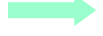
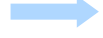
Average inventory
per assembly for
Group 1

Average inventory
per assembly for
Group 2

Average inventory
per assembly for
Group 3

Average inventory
per assembly for
Group 4

Average inventory
per assembly for
Group 5



x 21 assemblies x 21 assemblies x 12 assemblies

x 44 assemblies x 24 assemblies



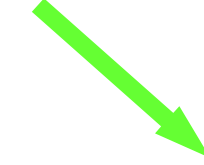
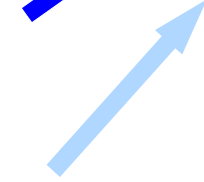
Inventory
for group 1

Inventory for
group 2

Inventory
for group 3

Inventory
for group 4

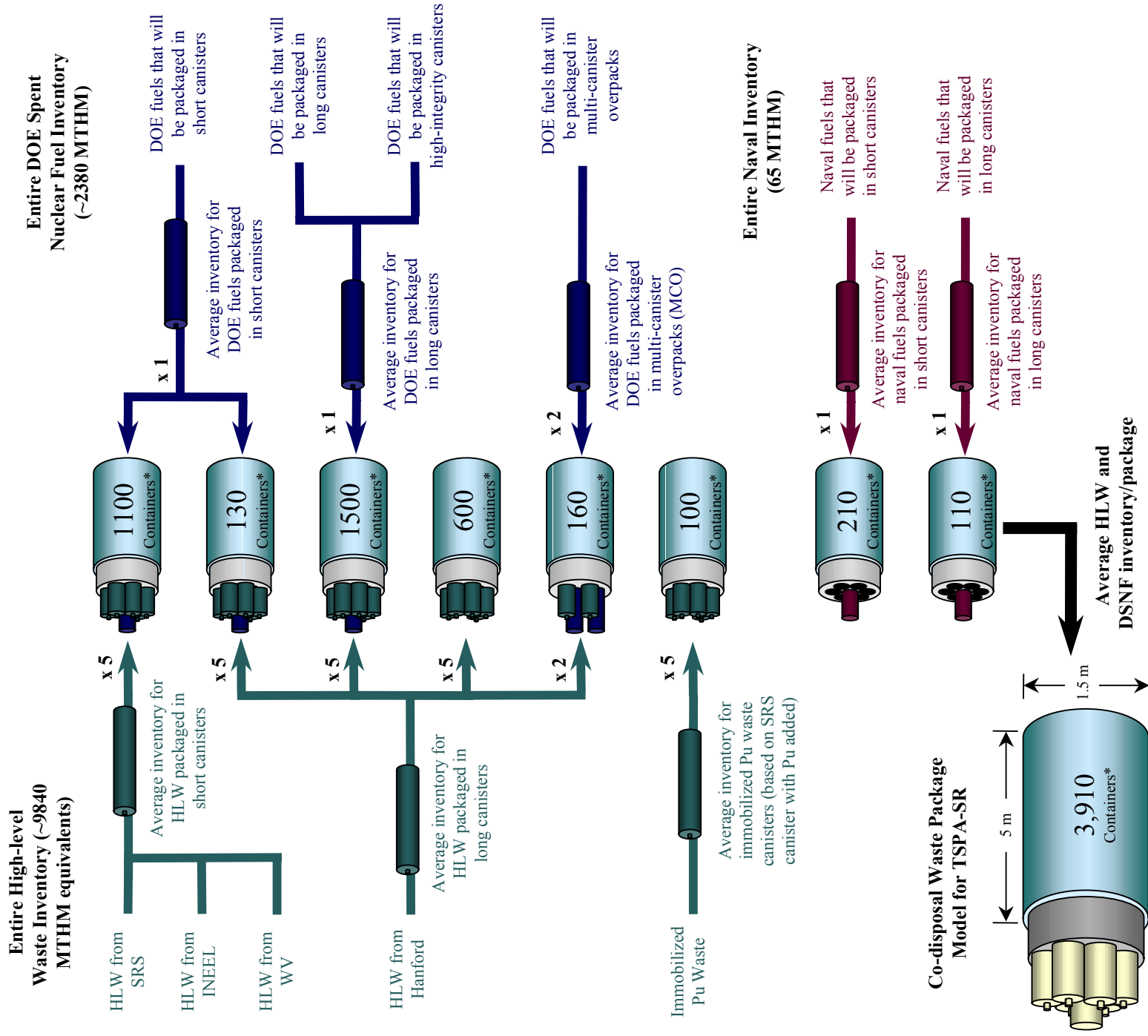
Inventory for
group 5



Average CSNF inventory
per package in TSPA-SR

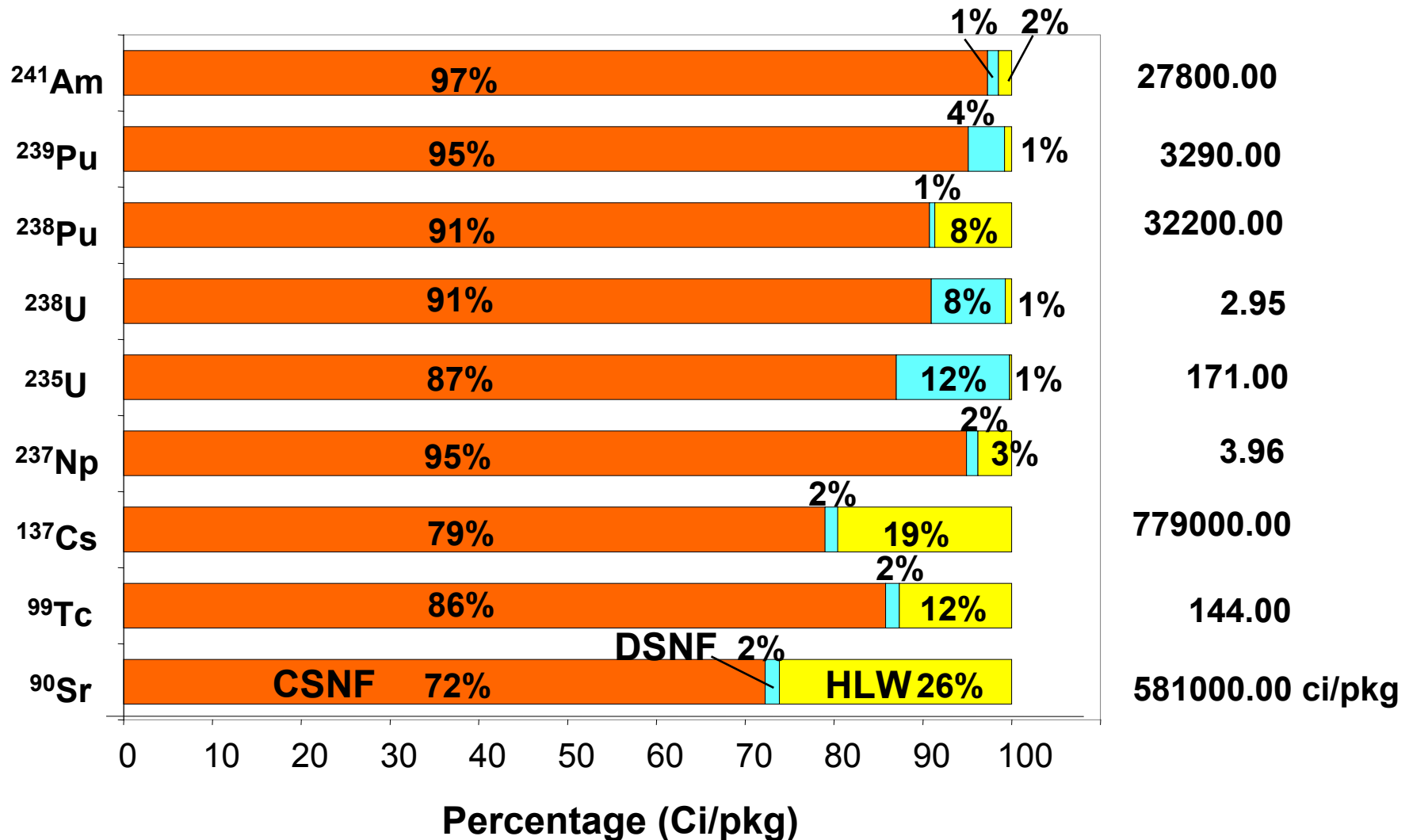
*The number of waste packages for each configuration is dictated by Stroupe 2000

Radioisotope Activities in a Co-disposal Waste Package Based on Average Inventory in Several Packing Groups

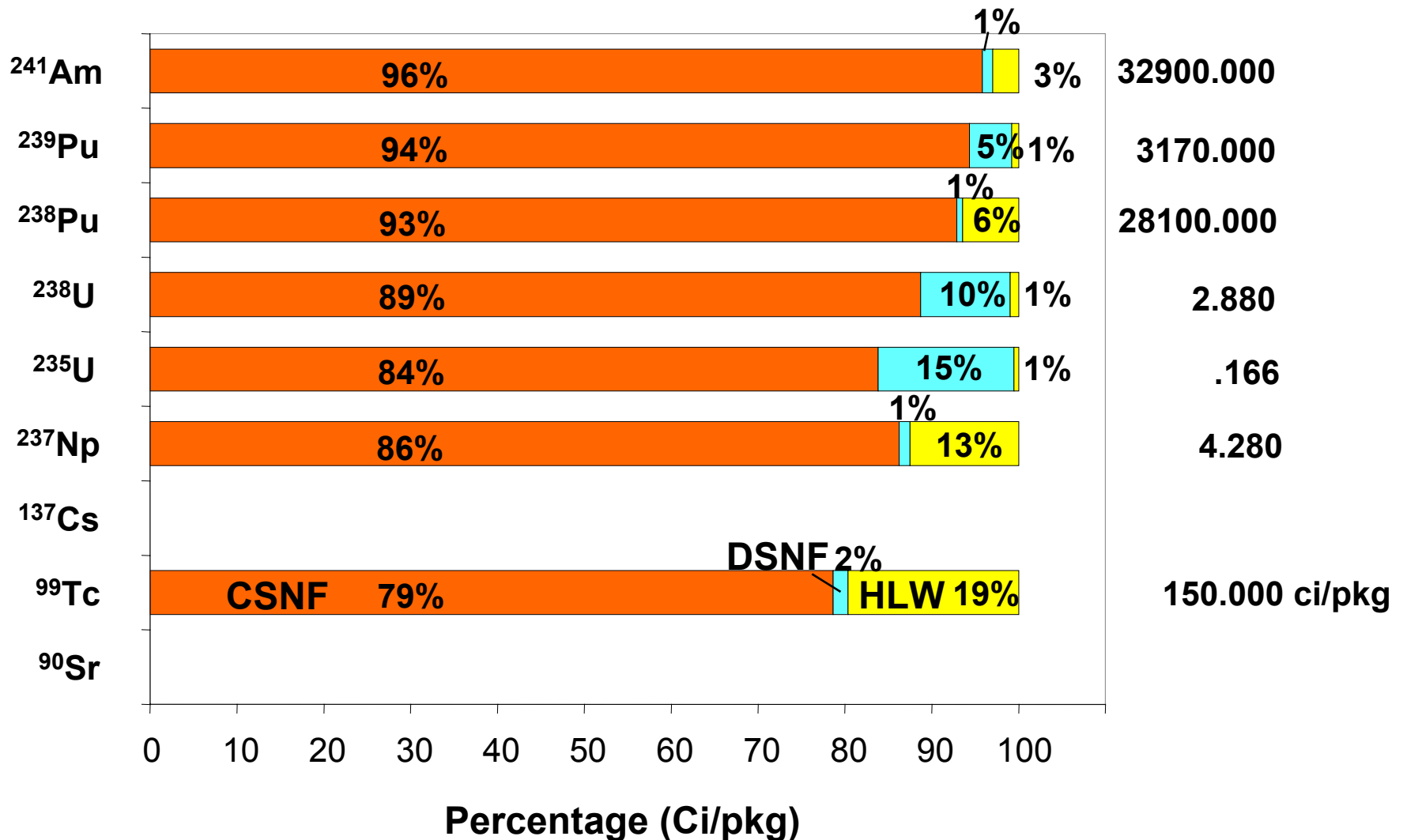


*The number of waste packages for each configuration is dictated by Stroupe 2000

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



Percentage of Radioisotopes in Packages Similar in TSPA-VA



Radioisotope Selection Updated for TSPA-SR As Requested in CLST3 and TSPA3/ENG4

Isotope	TSPA-SR						TSPA-VA	NRC IPA 99	TSPA 93 & 95	Comments
	Nominal		Volcanic		Intrusion					
	10 ⁴	10 ⁶	10 ⁴	10 ⁶	10 ⁴	10 ⁶				
Total	16	21	12	16	18	23	9	20	38	
¹⁴ C	•	•			•	•	•	•	•	Aqueous ¹⁴ C, nonsorbing tracer
⁹⁹ Tc	•	•			•	•	•	•	•	Tracked since 1984
¹²⁹ I	•	•			•	•	•	•	•	Tracked since 1984
²²⁷ Ac	•	•	•	•	•	•			•	
²²⁹ Th	•	•	•	•	•	•			•	
²³⁰ Th		•		•		•		•	•	10 ⁶ yr runs
²³² Th									•	Groundwater protection; originally screened in for SR
²³² U	•	•	•	•	•	•				
²³³ U	•	•	•	•	•	•			•	In DSNF
²³⁴ U	•	•			•	•	•	•	•	
²³⁵ U									•	In DSNF; helps track in growth of ²³¹ Pa; originally in for SR
²³⁶ U	•	•			•	•			•	
²³⁸ U	•	•			•	•		•	•	
²³⁷ Np	•	•			•	•	•	•	•	Tracked since 1984; daughter of ²⁴¹ Am & ²⁴¹ Pu
²³⁸ Pu	•	•	•	•	•	•			•	
²³⁹ Pu	•	•	•	•	•	•	•	•	•	
²⁴⁰ Pu	•	•	•	•	•	•		•	•	
²⁴² Pu		•		•		•	•		•	10 ⁶ yr runs
²⁴¹ Am	•	•	•	•	•	•		•	•	
²⁴³ Am	•	•	•	•	•	•		•	•	
⁹⁰ Sr			•	•	•	•				Volcanic & intrusion scenario
¹³⁷ Cs			•	•	•	•		•		Volcanic & intrusion scenario
²¹⁰ Pb		•		•		•		•	•	10 ⁶ yr runs
²³¹ Pa		•	•	•		•	•		•	
²²⁶ Ra		•		•		•		•	•	Groundwater protection, 10 ⁶ yr runs
²²⁸ Ra									•	Groundwater protection; originally screened in for SR

Radioisotope Selection Updated for TSPA-SR as Requested in CLST3 TSPA13/ENG4

Isotope	TSPA-SR						TSPA-VA	NRC IPA 99	TSPA 93 & 95	Comments
Total							9	20	38	
³⁶ Cl									•	Estimate updated from CDB value-no longer important
⁵⁹ Ni									•	
⁶³ Ni										Originally screened in for SR
⁷⁹ Se							•	•	•	Error in half-life corrected
^{93m} Nb									•	
⁹⁴ Nb								•	•	
⁹⁹ Zr									•	
¹⁰⁷ Pd									•	
¹²⁶ Sn									•	
¹³⁵ Cs								•	•	
¹⁵¹ Sm									•	
²⁴¹ Pu									•	
^{242m} Am									•	
²⁴⁴ Cm									•	
²⁴⁵ Cm								•	•	
²⁴⁶ Cm								•	•	

Dissolved Concentration Component* Determines Aqueous Concentration of Radioisotopes

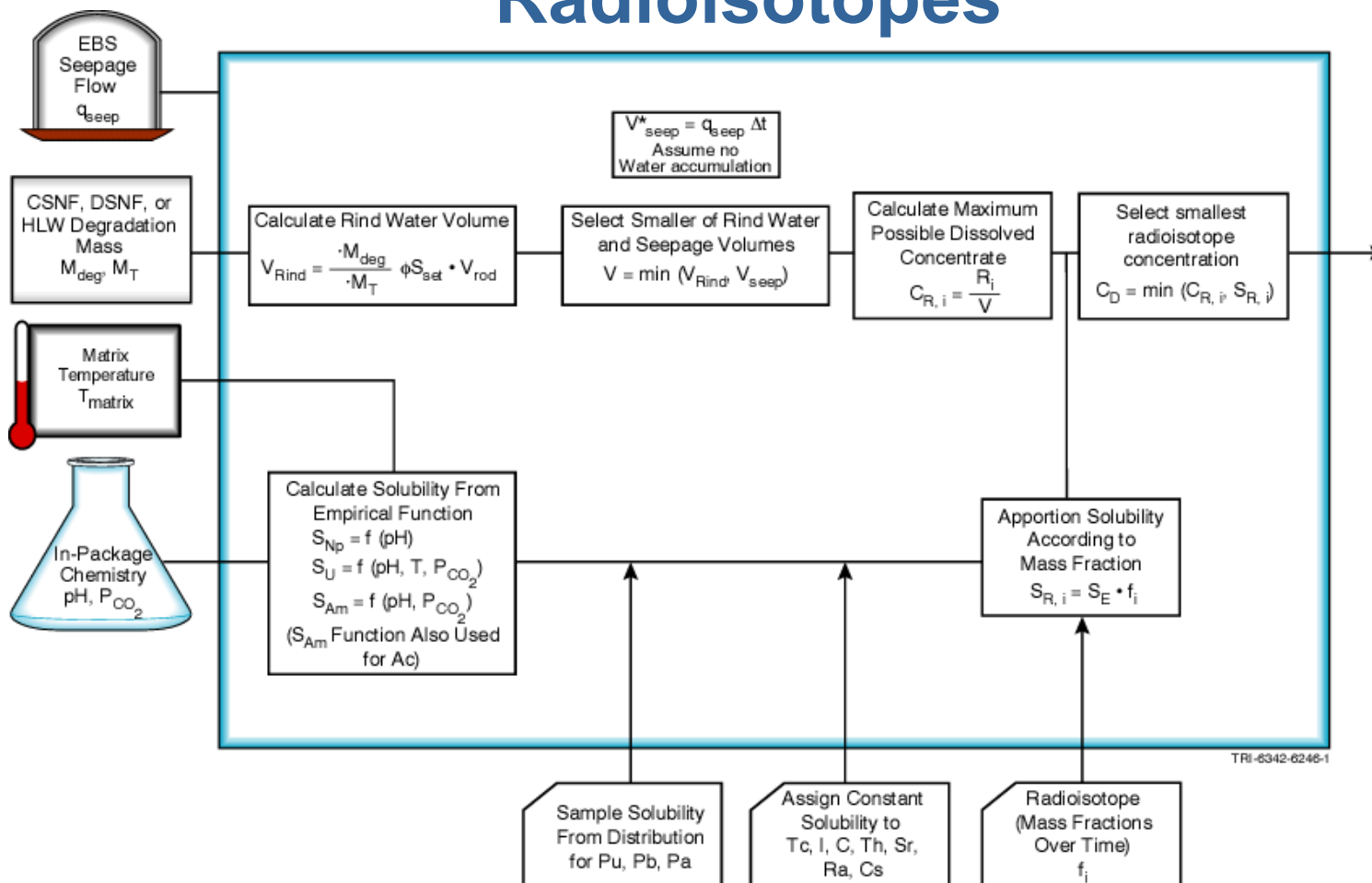


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Drawing Not To Scale
Dissolved Concentration Component.ai

* Discussed in Summary of Dissolved Concentration Limits_AMR

Solubility of Important Radioisotopes updated for TSPA-SR

Empirical functions

- $Np = f(\text{pH})$
- $Am = f(\text{pH}, P_{\text{co}_2})$
- $U = f(\text{pH}, P_{\text{co}_2}, T)$

Distributions

- Pu
- Pa
- Pb

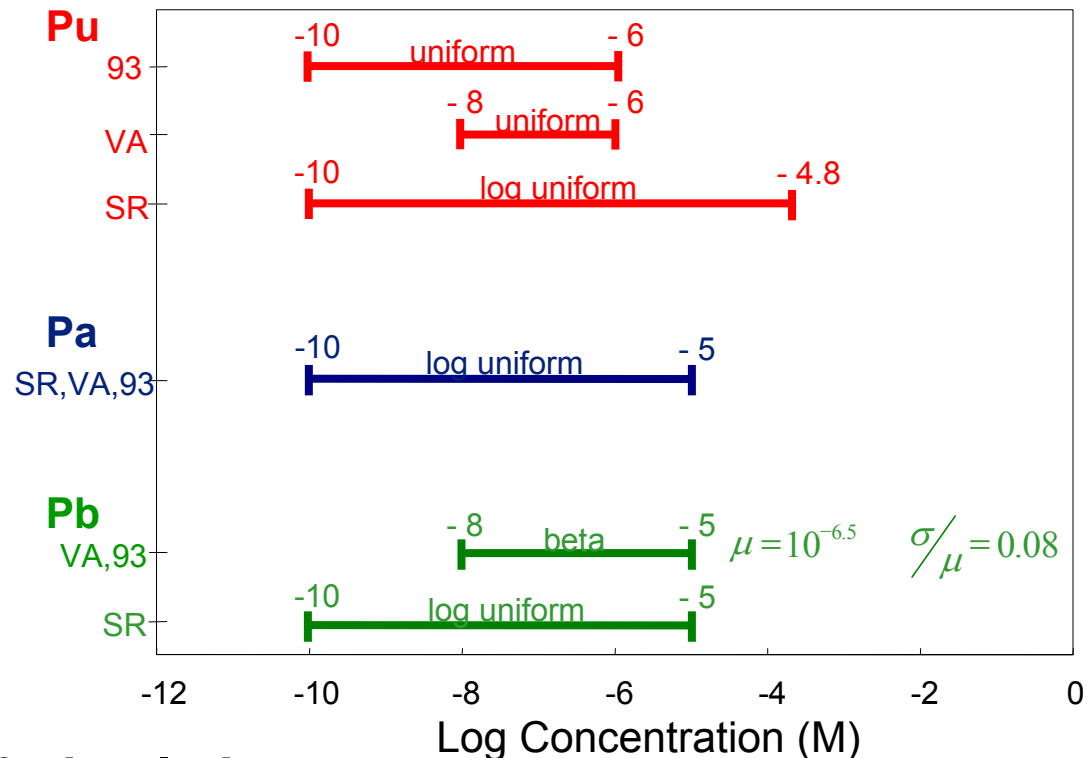
Constants

- $Tc = I = Sr = Cs = C = 1M$

Estimates based on EQ3/6 simulation

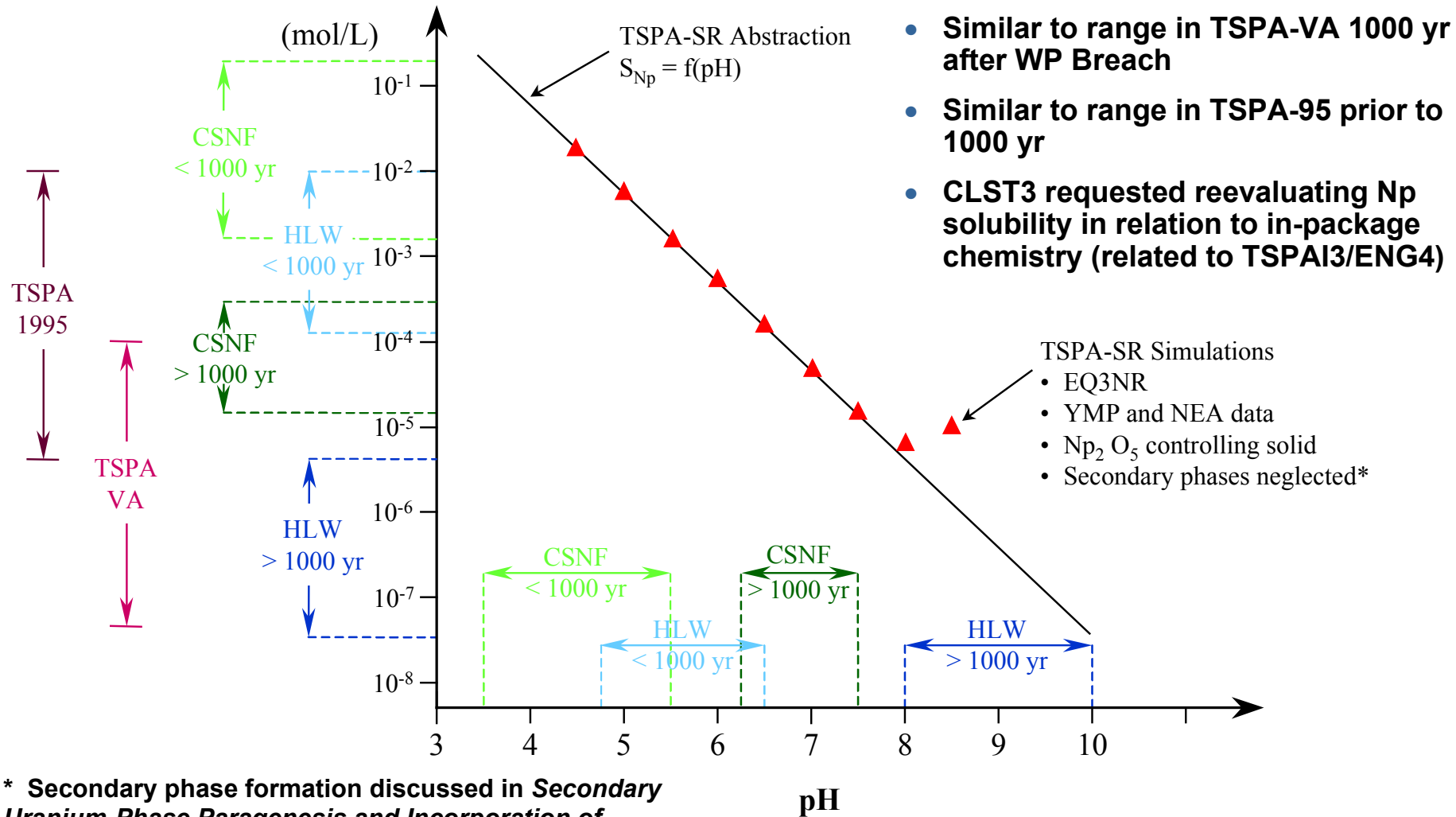
- Thermodynamic data used recent NEA and literature

Reevaluation addresses CLST3 and TSPA13/ENG4 request to update solubilities



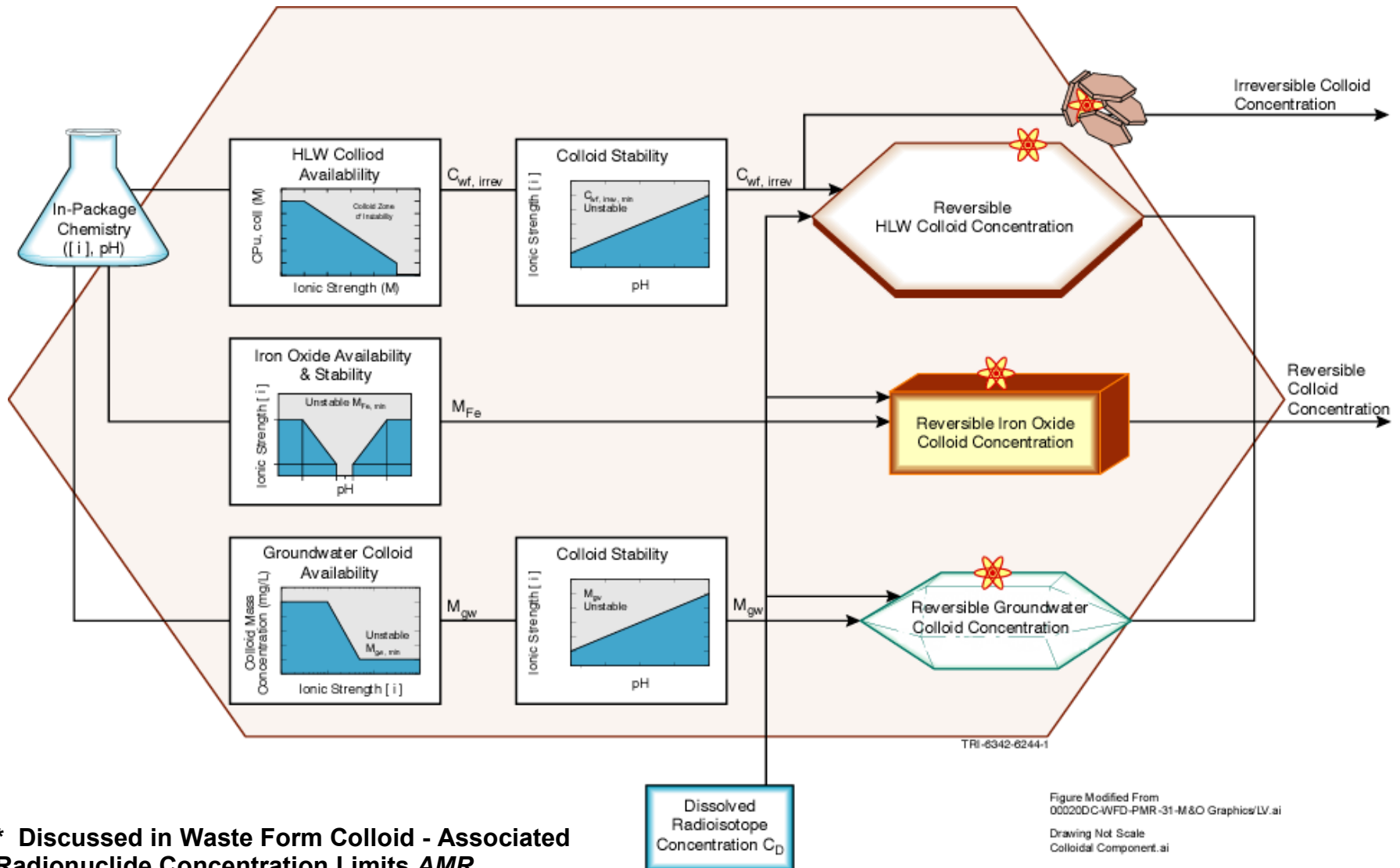
* Discussed in Summary of Dissolved Concentration Limits_AMR

Np Solubility Range in SR Similar to Range in TSPA VA and TSPA-95



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

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

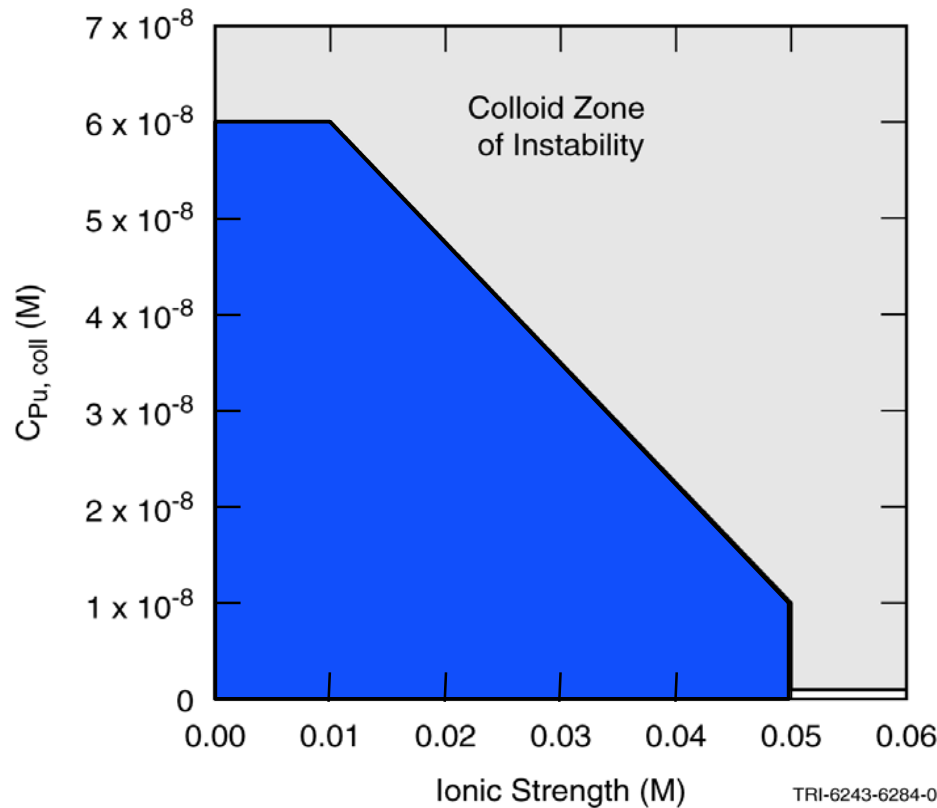


* Discussed in Waste Form Colloid - Associated Radionuclide Concentration Limits_AMR

Figure Modified From
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Colloidal Component.ai

Colloid Component uses Lab Results and Estimated pH and Ionic Strength to Determine Availability and Stability of Colloids

Relationship Between Colloid Concentration and Ionic Strength



- ***Irreversible*** waste colloid concentration function based on HLW degradation data (colloids treated separately from HLW degradation)
- Calculated ionic strength and pH of water used to determine concentration
- Pu and Am transported as ***irreversible*** colloids
- Pu, Am, Th (Ra, Pb), Pa (Ac), Sr, Cs transported as ***reversible*** colloids
- CLST and TSPA I3/ENG4 states colloids of low concern but recommends continued modeling