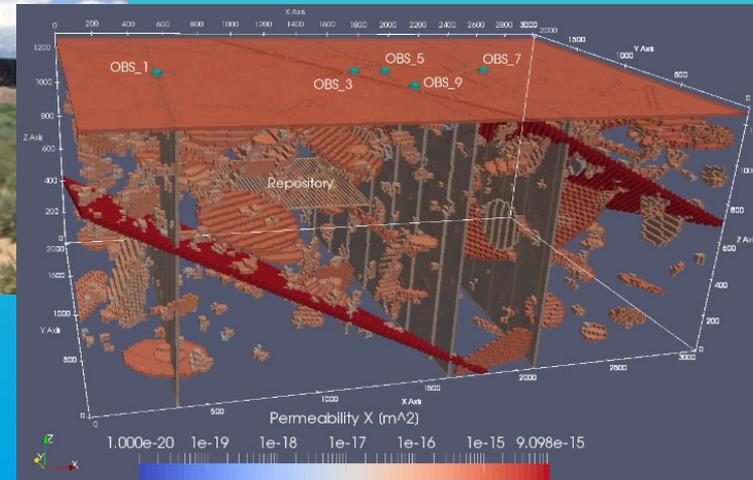


## Spent Fuel and Waste Science and Technology (SFWST)



## Cross-Cutting Research and Development

NWTRB Summer 2020 Board Meeting  
July 27-28, 2020  
Online Virtual Meeting

Geoff Freeze, Sandia National Laboratories  
Robert Howard, Oak Ridge National Laboratory

Sandia National Laboratories is a multi-mission 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.  
SAND2020-7275 PE

# Disclaimer

This is a technical presentation that does not take into account the contractual limitations or obligations under the Standard Contract for Disposal of Spent Nuclear Fuel and/or High-Level Radioactive Waste (Standard Contract) (10 CFR Part 961). For example, under the provisions of the Standard Contract, spent nuclear fuel in multi-assembly canisters is not an acceptable waste form, absent a mutually agreed to contract amendment.

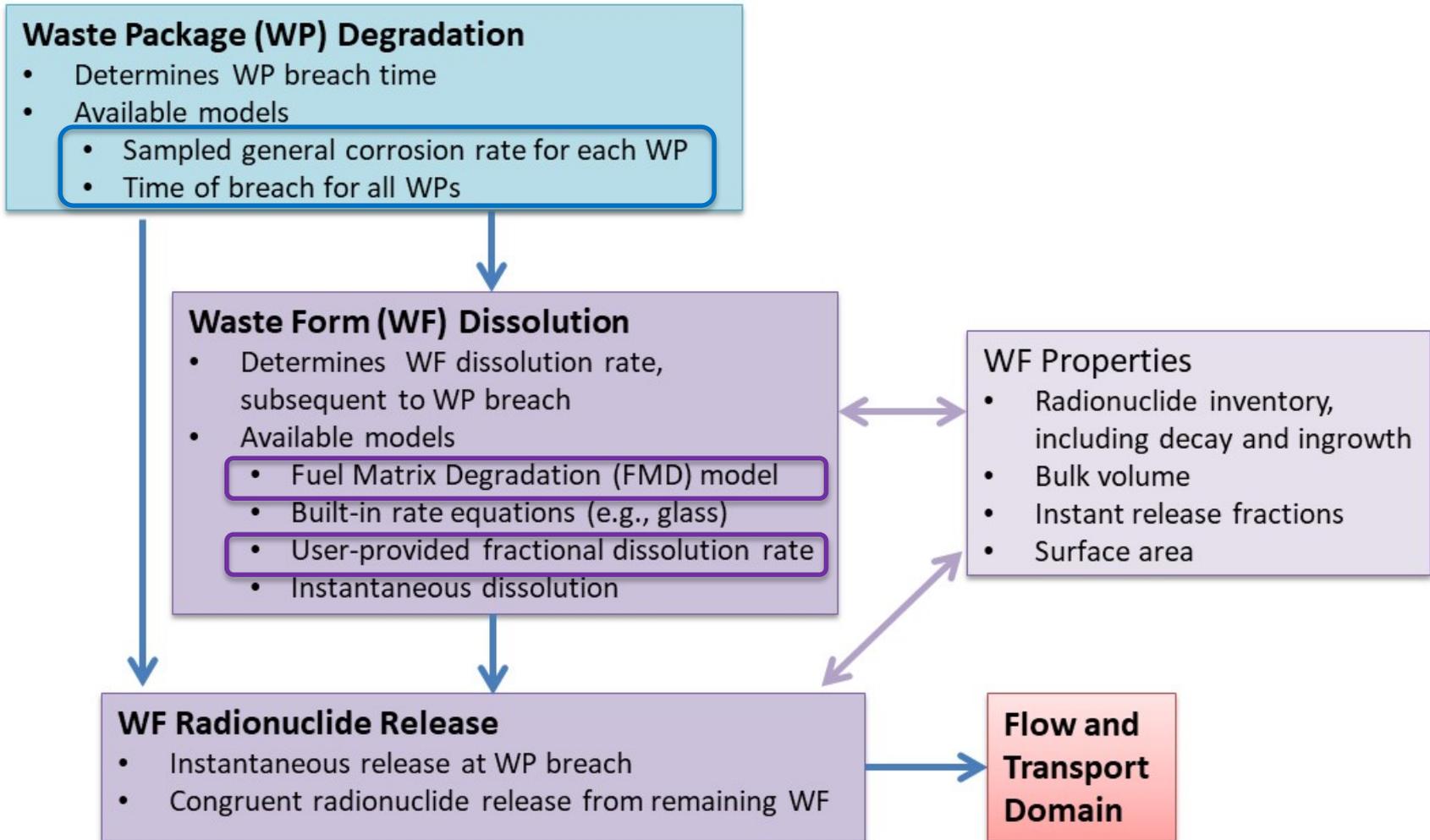
To the extent discussions or recommendations in this presentation conflict with the provisions of the Standard Contract, the Standard Contract governs the obligations of the parties, and this presentation in no manner supersedes, overrides, or amends the Standard Contract.

This presentation reflects technical work which could support future decision making by DOE. No inferences should be drawn from this presentation regarding future actions by DOE, which are limited both by the terms of the Standard Contract and Congressional appropriations for the Department to fulfill its obligations under the Nuclear Waste Policy Act including licensing and construction of a spent nuclear fuel repository.

# Cross-Cutting Research and Development (R&D) Dual Purpose Canister (DPC) Considerations

- Geologic Disposal Safety Assessment (GDSA) reference cases, modeled with the PFLOTRAN code
  - Source Terms – based on large, higher-temperature waste packages
    - Waste package degradation model
    - Waste form degradation model
  - Interactions With Engineered Barriers
    - Effects of different geologies
    - Effect of high-temperature on engineered barriers (e.g., bentonite)
- Thermal and shielding implications for the transportation schedule

# Source Term Processes



Source: adapted from Mariner et al. 2019, Figure 2-4

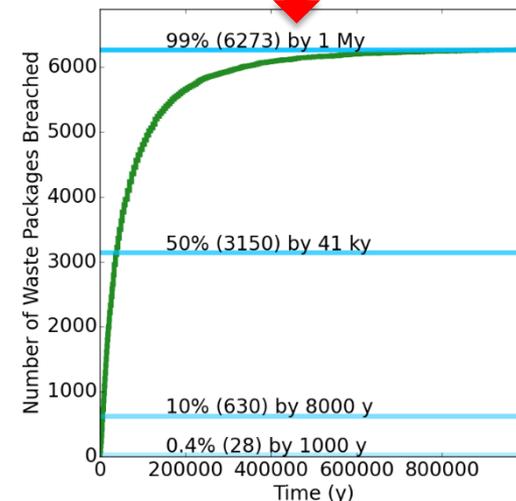
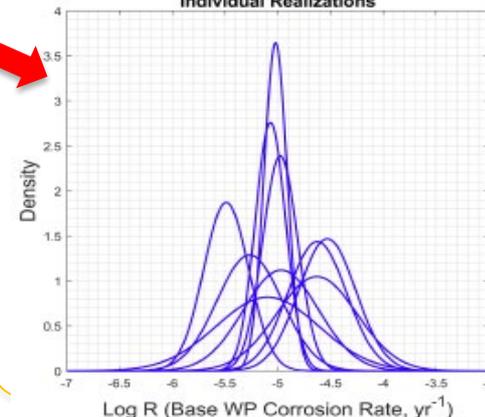
# Waste Package Canister Vitality Model

- **Canister vitality (wall thickness remaining) is a simple probabilistic rate (Mariner et al. 2016)**
  - temperature-dependent general corrosion
  - can also define a breach time (e.g., early failures)
- **Future development (Mariner et al. 2018)**
  - mechanistic corrosion (general, localized)
    - DECOVALEX Task F
  - effects of groundwater chemistry / redox
  - seismic, igneous (site specific)
- **Dual-purpose canister (DPC) considerations**
  - Elevated temperatures
  - Disposal overpack materials (Cu, alloy 22, ... ?)

$R_{\text{eff}}$  = canister degradation rate

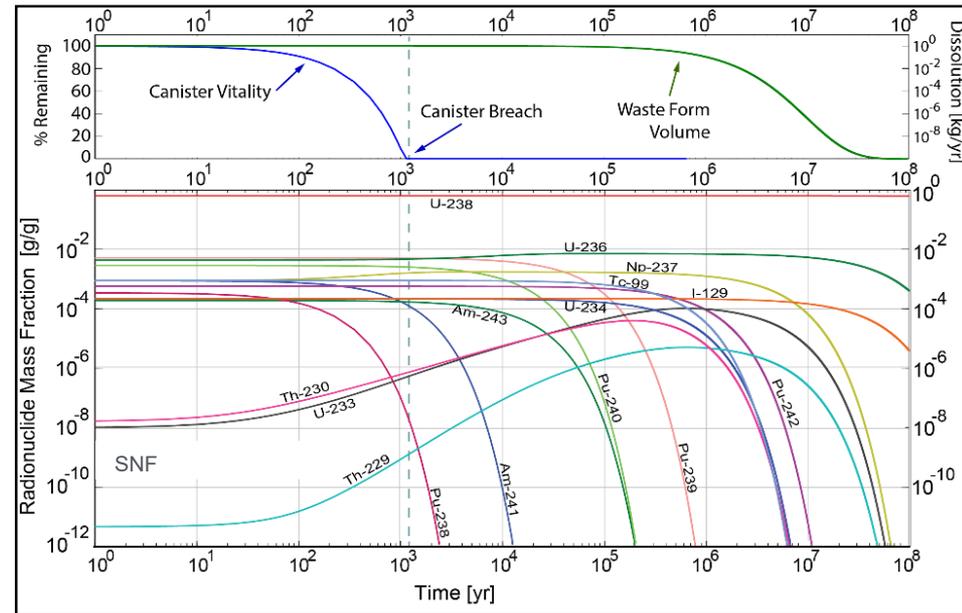
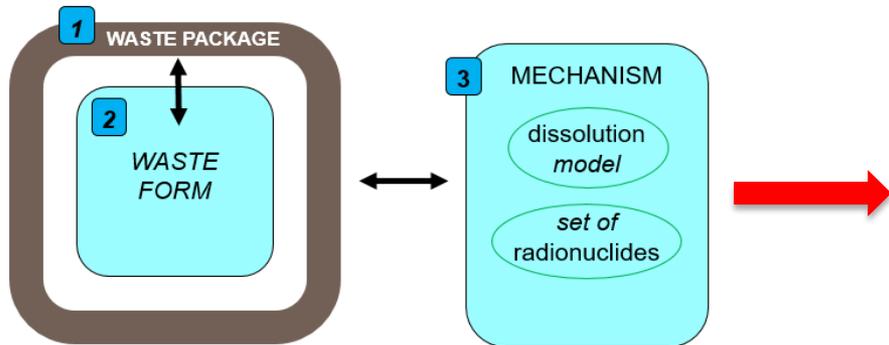
$$R_{\text{eff}} = R \cdot e^{\left[ \frac{1}{60^\circ\text{C}} - \frac{1}{T(x,t)} \right]}$$

Example Log R Distribution among WPs in Individual Realizations



# Waste Form Dissolution Rate Model

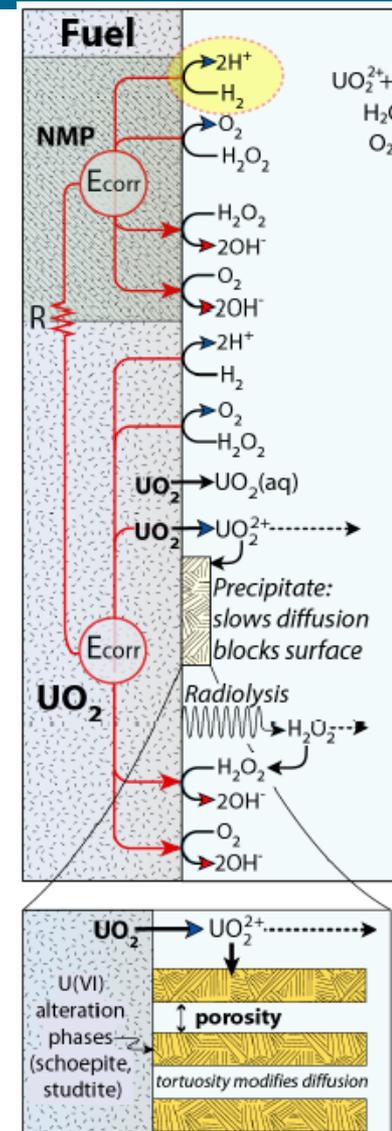
- Spent nuclear fuel (SNF) dissolution rate that begins following waste package failure (Mariner et al. 2016)
  - Instant release fraction (specified radionuclides)
  - Fractional dissolution (e.g.,  $10^{-5}/\text{yr}$ )
- Directly implemented in PFLOTRAN



# Electrochemical Fuel Matrix Degradation (FMD) Model

- 1-D reactive transport model to simulate diffusion of chemical species (Jerden et al. 2017)
- SNF dissolution rate is a function of (Mariner et al. 2018)
  - Radiolysis
  - Growth of alteration layer on  $\text{UO}_2$  surface
  - Diffusion of reactants through the alteration layer
  - Interfacial corrosion potential

Inputs	Outputs
<ul style="list-style-type: none"> <li>Initial concentration profiles across 1D corrosion/water layer (<math>\text{UO}_2(\text{s})</math>, <math>\text{UO}_3(\text{s})</math>, <math>\text{UO}_4(\text{s})</math>, <math>\text{H}_2\text{O}_2</math>, <math>\text{UO}_2^{2+}</math>, <math>\text{UCO}_3^{2-}</math>, <math>\text{UO}_2</math>, <math>\text{CO}_3^{2-}</math>, <math>\text{O}_2</math>, <math>\text{Fe}^{2+}</math>, and <math>\text{H}_2</math>)</li> <li>Initial corrosion layer thickness</li> <li>Dose rate at fuel surface (= f (time, burnup))</li> <li>Temperature</li> <li>Time, time step length</li> <li>Environmental concentrations (<math>\text{CO}_3^{2-}</math>, <math>\text{O}_2</math>, <math>\text{Fe}^{2+}</math>, and <math>\text{H}_2</math>)</li> </ul>	<ul style="list-style-type: none"> <li>Final concentration profiles across 1D corrosion/water layer</li> <li>Final corrosion layer thickness</li> <li>Fuel dissolution rate</li> </ul>



(adapted from Jerden et al. 2017)

- Mechanistic model and emulators coupled to PFLOTRAN (Mariner et al. 2019)

# Waste Form Degradation Model for DPCs

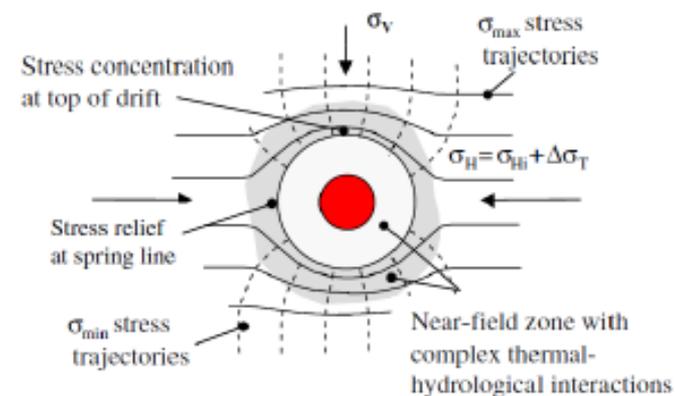
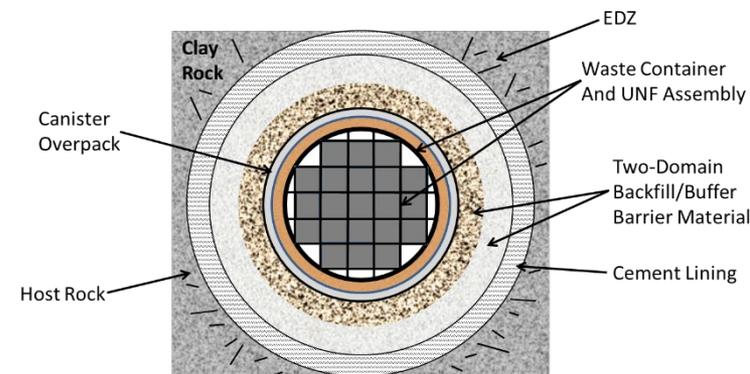
- DPC considerations
  - In-package chemistry and  $\text{UO}_2$  degradation
    - elevated temperature, boiling?
    - reduced instant release fraction for higher burn-up fuels?
    - effects of different geologies (e.g., groundwater chemistry)
    - chemical effects from filler materials
    - criticality event?
      - changes to radionuclide inventory
      - additional radiolytic oxidants from beta and gamma radiation
  - Cladding degradation
    - elevated temperature?
    - criticality event?
      - intact cladding assumed
  - Neutron absorbers
    - degradation of aluminum-based materials
      - e.g., Boral™, Metamic™

# Interactions with Engineered Barriers (Rutqvist 2019)

- For DPC direct disposal, a peak backfill temperature of 200°C is likely to occur, unless the SNF is aged for hundreds of years before backfilling (Hardin et al. 2015)
- For clay-based materials, a peak temperature of 100°C is often adopted to limit thermal-hydrologic-mechanical-chemical effects (e.g., chemical changes, material degradation, clay phase change, smectite swelling)
  - FEBEX: bentonite heated to 100°C in 18-year test at Grimsel Test Site
  - Backfill peak temperature >100°C is currently being evaluated
    - Mont Terri: ongoing in-situ heater test up to 140°C in Opalinus Clay (Rutqvist et al. 2018; 2019)
    - HotBENT: planned heater test at 150°C to 200°C at Grimsel Test Site
  - Bentonite backfill mixtures can be engineered to increase the thermal conductivity by mixing in graphite or graphene oxide
    - Jobmann and Buntebarth 2009; Chen et al. 2018

# Interactions with Engineered Barriers (cont.)

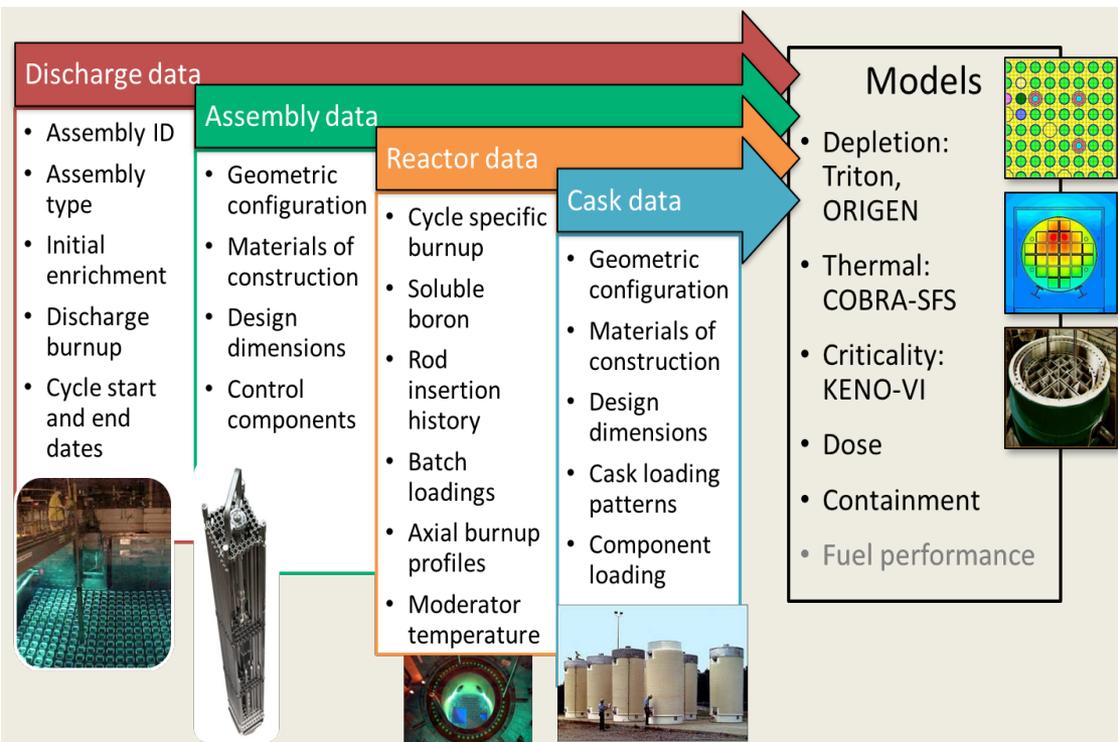
- The thermal-hydrologic-mechanical-chemical (THMC) effects of high-temperature on bentonite and near-field host rock are being examined in multiple SFWST Work Packages
  - Argillite Disposal R&D
  - Engineered Barrier System (EBS) R&D
  - International Collaborations Research
- These effects will be captured in GDSA reference cases
  - DPC disposal in unsaturated alluvium
  - DPC disposal in saturated argillite



(Source: Rutqvist 2019, Figure 1-1)

# Implications for Transportation

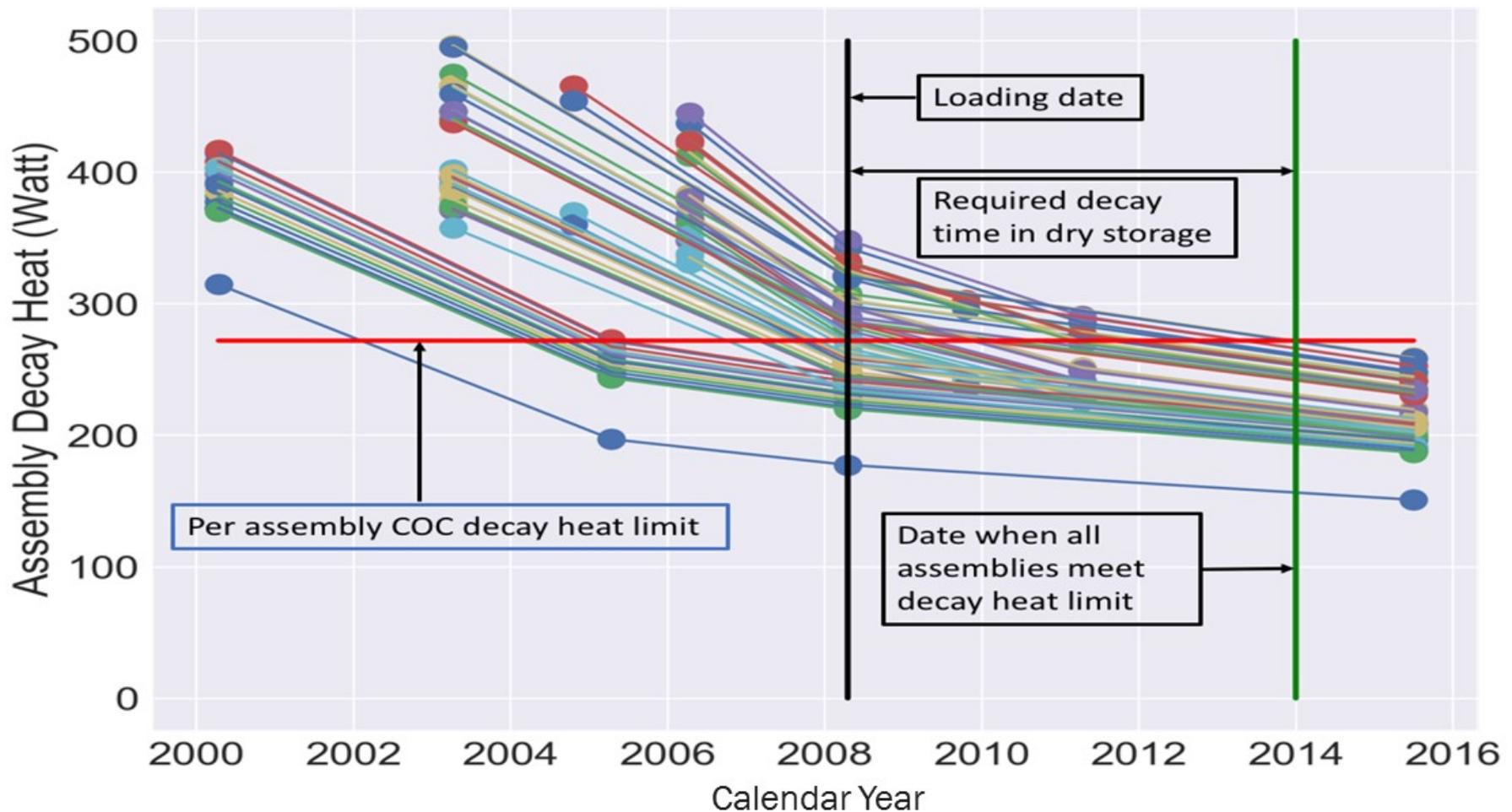
- The same tools and specific data that are used to evaluate criticality margin for the direct disposal of DPCs can also be used to evaluate the thermal and shielding criteria to determine when the DPC is transportable –
  - UNF-ST&DARDS and the Unified Database (UDB)



- Fuel geometry, dimensions, and materials
- Reactor irradiation histories (e.g. reactor cycle length, specific power)
- Cask system data, including Certificate of Compliance (CoC) requirements

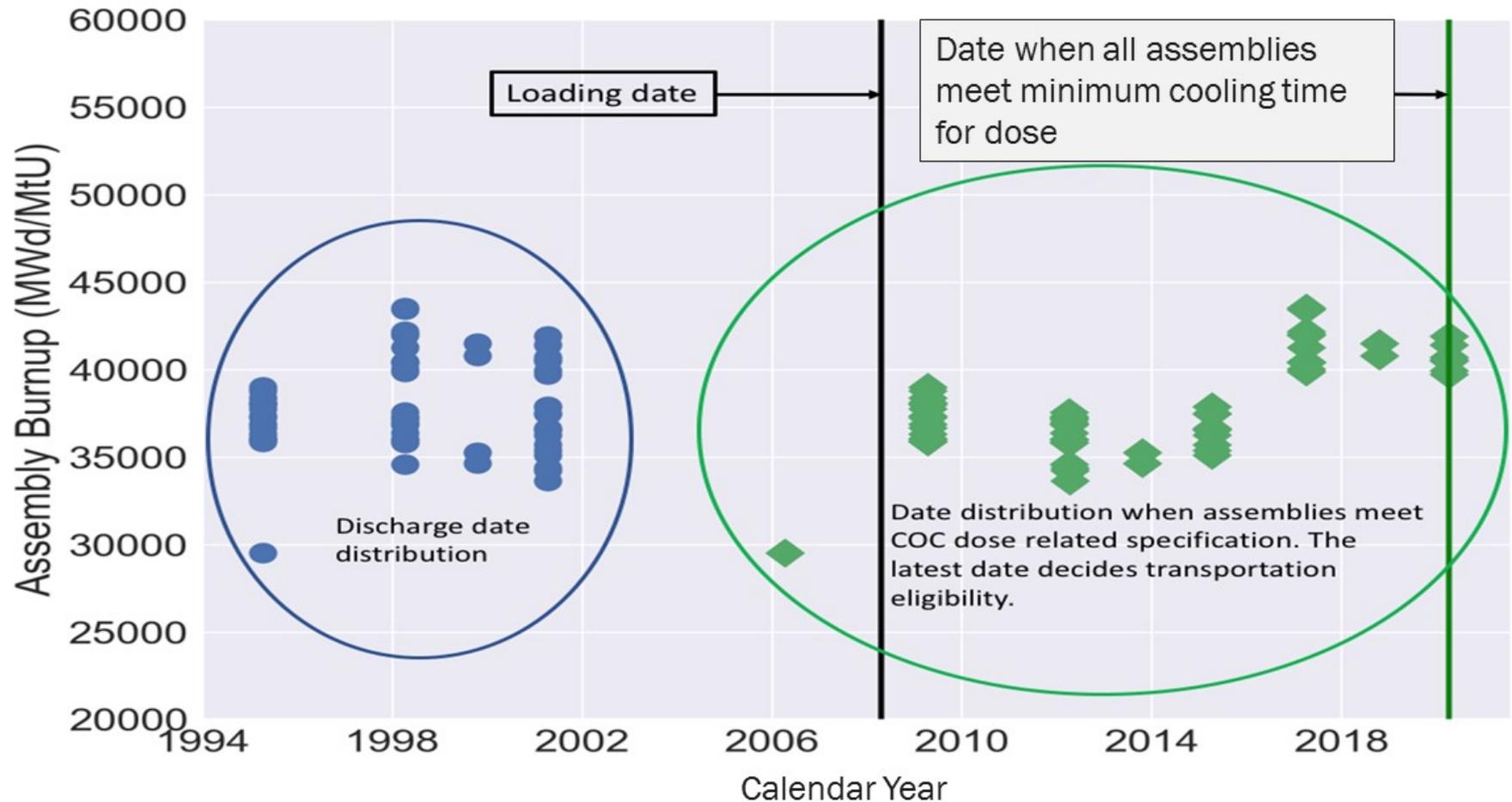
# Unified Database (UDB) checks against transportation Certificate of Compliance (CoC) limits can be used to determine dates when SNF could be shipped

## Assembly Decay Heat Example



# Unified Database (UDB) checks against transportation Certificate of Compliance (CoC) limits can be used to determine dates when SNF could be shipped

## Assembly Minimum Cooling Time Example (Dose Related)



# References

- Chen Y.G., Liu, X.M., Mu, X., Ye, W.M., Cui, Y.J., Chen, B., and Wu, D.B. (2018). Thermal conductivity of compacted GO-GMZ bentonite used as buffer material for a high-level radioactive waste repository. *Advances in Civil Engineering*, 2018, Article ID 9530813, <https://doi.org/10.1155/2018/9530813>.
- Hardin, E., Price, L., Kalinina, E., Hadgu, T., Ilgen, A., Bryan, C., Scaglione, J., Banerjee, K., Clarity, J., Jubin, R., Sobes, V., Howard, R., Carter, J., Severynse, T., Perry, F. (2015) Summary of Investigations on Technical Feasibility of Direct Disposal of Dual-Purpose Canisters. FCRD-UFD-2015-000129 Rev. 0. U.S. Department of Energy, Office of Used Nuclear Fuel Disposition.
- Jerden, J., V.K. Gattu and W. Ebert (2017). *Progress Report on Development of the Spent Fuel Degradation and Waste Package Degradation Models and Model Integration*. SFWD-SFWST-2017-000091, SFWD-SFWST-2017-000095. Argonne National Laboratory.
- Jobmann, M., and Buntebarth, G. (2009). Influence of graphite and quartz addition on the thermo–physical properties of bentonite for sealing heat-generating radioactive waste. *Applied Clay Science* 44, 206–210.
- Mariner, P.E., E.R. Stein, J.M. Frederick, S.D. Sevougian, and G.E. Hammond (2016). *Advances in Geologic Disposal System Modeling and Application to Crystalline Rock*. FCRD-UFD-2016-000440, SAND2016-9610R. Sandia National Laboratories, Albuquerque, NM.
- Mariner, P.E., E.R. Stein, J.M. Frederick, S.D. Sevougian, and G.E. Hammond (2018). *Advances in Geologic Disposal Safety Assessment and an Unsaturated Alluvium Reference Case*. SFWD-SFWST-2018-000509, SAND2018-11858R. Sandia National Laboratories, Albuquerque, NM.
- Mariner, P.E., E.R. Stein, J.M. Frederick, S.D. Sevougian, and G.E. Hammond (2019). *Progress in Deep Geologic Disposal Safety Assessment in the U.S. since 2010*. M2SF-19SN010304041, SAND2019-12001R. Sandia National Laboratories, Albuquerque, NM.
- Rutqvist, J. (2019). *Geotechnical and performance assessment impacts of DPC disposal in various host rock environments*. Prepared for U.S. Department of Energy, Spent Fuel and Waste Disposition, LBNL-2001220, Lawrence Berkeley National Laboratory.
- Rutqvist, J., Kim, K., Xu, H., Guglielmi, Y., and Birkholzer, J. (2018). *Investigation of Coupled Processes in Argillite Rock: FY18 Progress*. Prepared for U.S. Department of Energy, Spent Fuel and Waste Disposition, LBNL-2001168, Lawrence Berkeley National Laboratory.
- Rutqvist, J., Guglielmi, Y., Kim, K., Xu, H., Deng, H., Li, P., Hu, M., Steefel, C., Gilbert, B., Rinaldi, A., Nico, P., Borglin, S., Fox, P., and Birkholzer, J. (2019). *Investigation of coupled processes in argillite rock: FY19 progress*. Prepared for U.S. Department of Energy, Spent Fuel and Waste Disposition, LBNL-2001202, Lawrence Berkeley National Laboratory.

Questions?