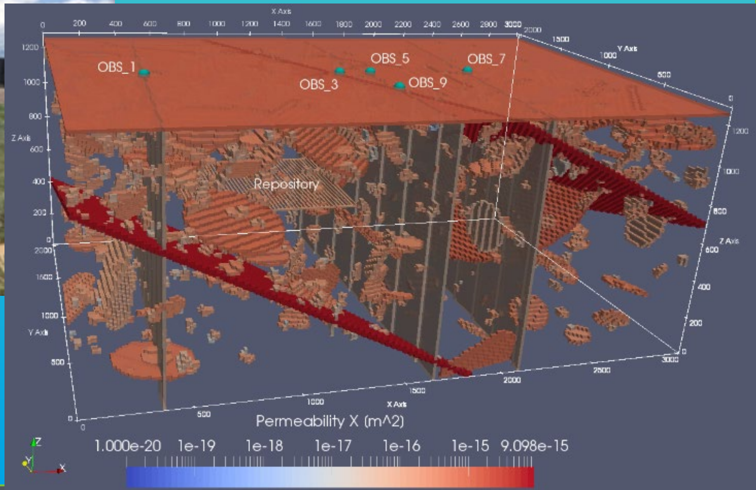
U.S. DEPARTMENT OF ENERGY | Office of NUCLEAR ENERGY
Spent Fuel and Waste Science and Technology (SFWST)
Clean. Reliable. Nuclear.



Ongoing Research and Development: Consequences of Nuclear Criticality in DPCs After Disposal

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

Laura Price, Sandia National Laboratories

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.
Unclassified, unlimited release. SAND2020-7153 PE

Disclaimer

This is a technical presentation that does not take into account 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.

Legal Notice

Acknowledgements

Results discussed below are based on work by:

- Halim Alsaed – Termination of criticality
- Amanda Barela – Inventory
- Pat Brady – In-package chemistry and radionuclide solubilities
- Mike Gross and Fred Gelbard – Thermal analyses
- Scott Painter (ORNL) and Michael Nole – PFLOTRAN calculations
- Jeralyn Prouty – Reference repository diagrams

Objectives

- Develop tools to model the consequences of postclosure criticality
  - Couple neutronics calculations and thermal-hydraulic calculations
  - Build sub-module in PFLOTRAN to account for postclosure critical event
- Further our understanding of the features, events, and processes important to modeling postclosure criticality
- Examine processes leading to permanent termination of critical event
- Identify areas where further work is needed

Approach

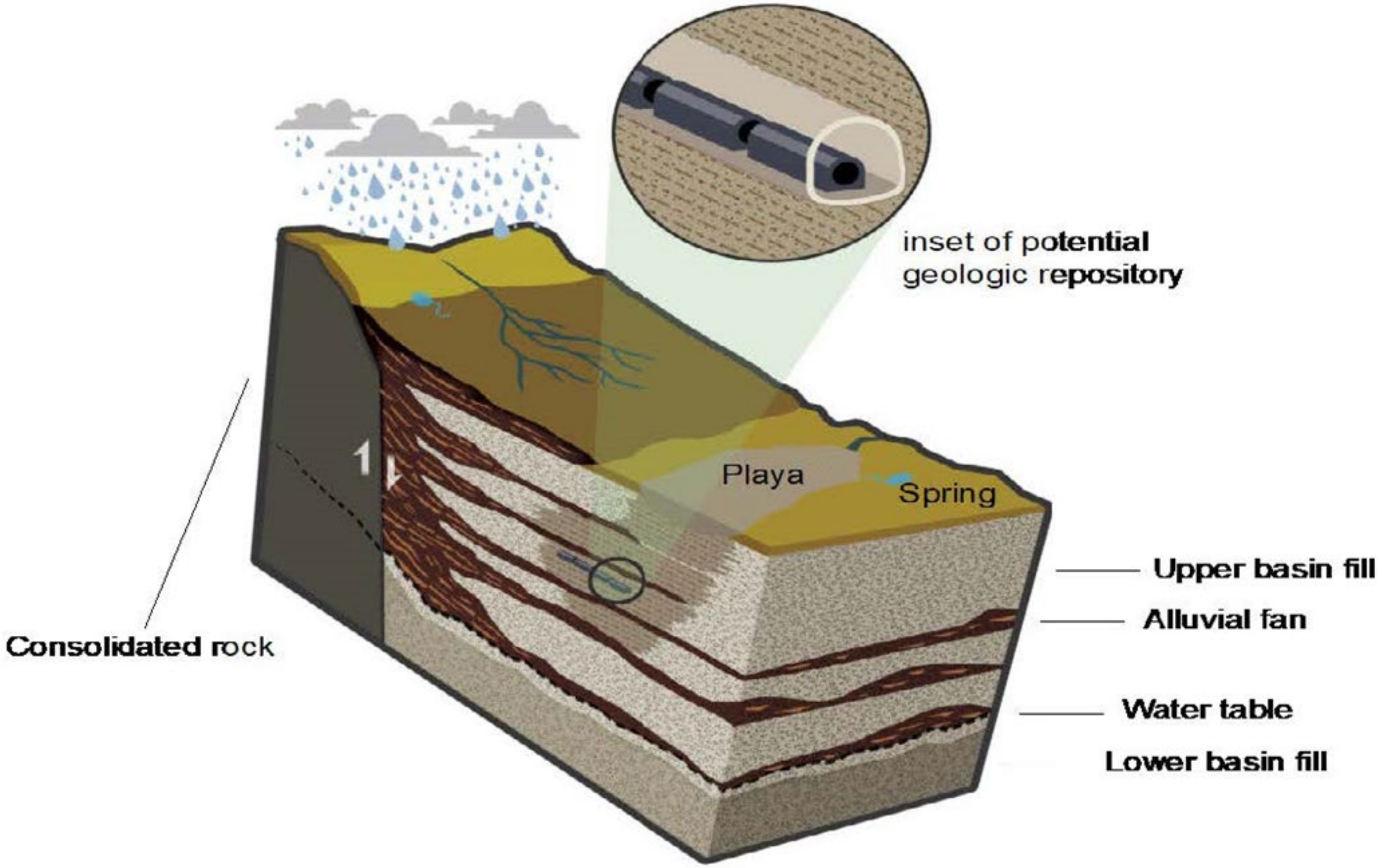
- Two hypothetical repositories considered
  - Saturated repository in shale (Mariner et al. 2017)
    - 500 m depth
    - Backfilled with bentonite
    - Hydrostatic pressure is 50 bars
  - Unsaturated repository in alluvium (Mariner et al. 2018)
    - 250 m depth
    - Backfilled with crushed alluvium
    - Percolation rate up to 10 mm/yr
- Calculate radionuclide concentrations in the host rock with and without the occurrence of a critical event
  - Steady-state criticality (9,000 – 19,000 years postclosure)
  - Transient criticality (9,000 years postclosure)
- Single waste package (37 PWR)

Assumptions

- A waste package is breached; criticality occurs 9,000 years after closure*
- Fuel assembly lattice remains intact (i.e., intact grid spacers) and cladding permits radionuclide release (e.g., through pin holes and cracks)*
- Al-based neutron absorbers are not present
- The steady-state critical event is not cyclic*

* Will be investigated as the research effort moves forward

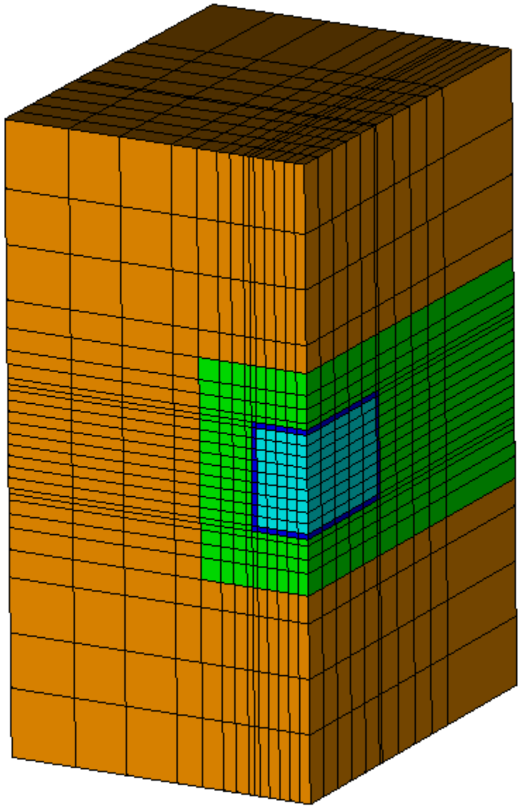
Hypothetical Repository in Alluvium



Mariner et al. 2018

Single DPC Model Setup

- Geometry
  - Consistent with GDSA Unsaturated Alluvium reference case (Sevougian 2019; Hardin and Kalinina 2016)
  - 40 m drift spacing, 40 m center-to-center spacing within drift
  - Square cross-section for drift (4m x 4m) and DPC (1.67 m x 5 m x 1.67 m)
  - 0.1 m overpack/shell
- Properties
  - Permeability 10^{-14} (alluvium) 10^{-13} (backfill)
  - Thermal conductivity = 1 W/m^2-K (dry) and 2 W/m^2-K (wet)
  - Canister internals = hydraulic properties of backfill
- Scenario
  - Postclosure with 37-PWR assembly and backfilled drifts in place
  - Top of DPC shell breached at 9000 years allowing water to enter
  - Initiate criticality event when canister is filled with water
- Cases
  - 10 mm/year and 2 mm/year percolation into waste package
  - Range of power outputs for criticality event



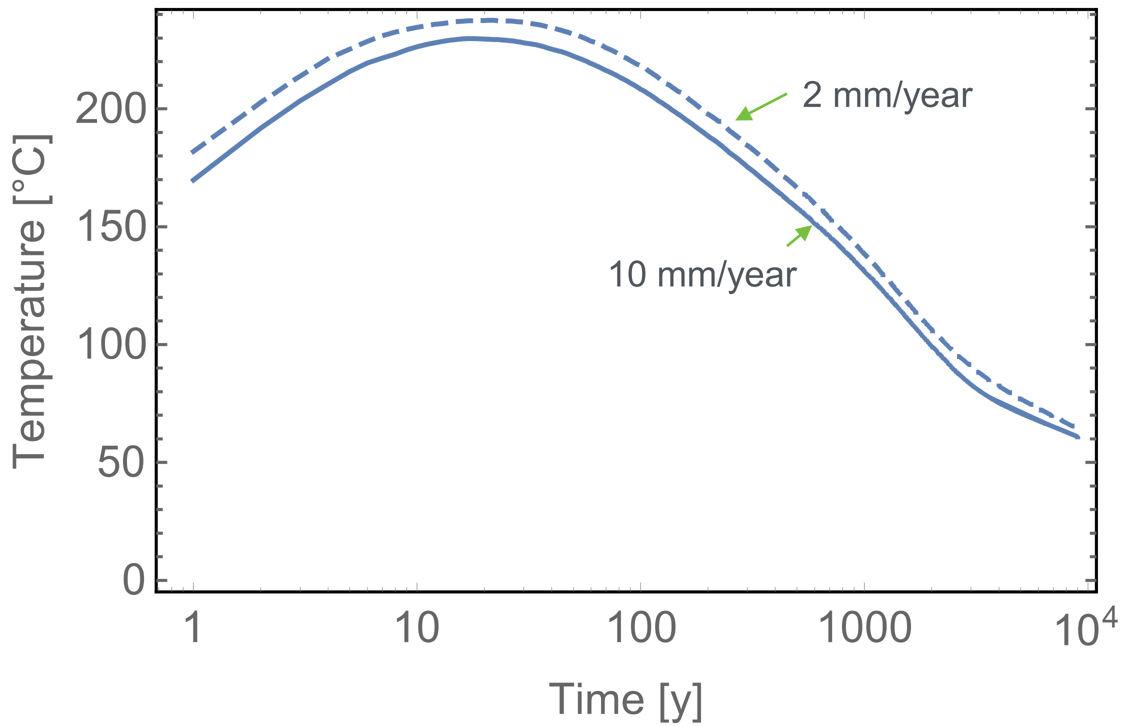
Objective is to estimate (bound) power output that could be sustained before driving water out of the package

PFLOTRAN

- Open source code for thermal hydrology and reactive transport in variably saturated porous geologic media
- Highly parallel by domain decomposition
- “General mode” solves coupled conservation equations over two phases
  - Water as liquid and vapor
  - Air as gas and dissolved in liquid
  - Energy (advection and conduction)
  - Variable switching to accommodate phase disappearance/reappearance
- Lichtner, Hammond et al. www.pflotran.org
  Hammond, Lichtner and Mills 2014 Water Resources Research

Temperature at DPC Center Prior to Critical Event

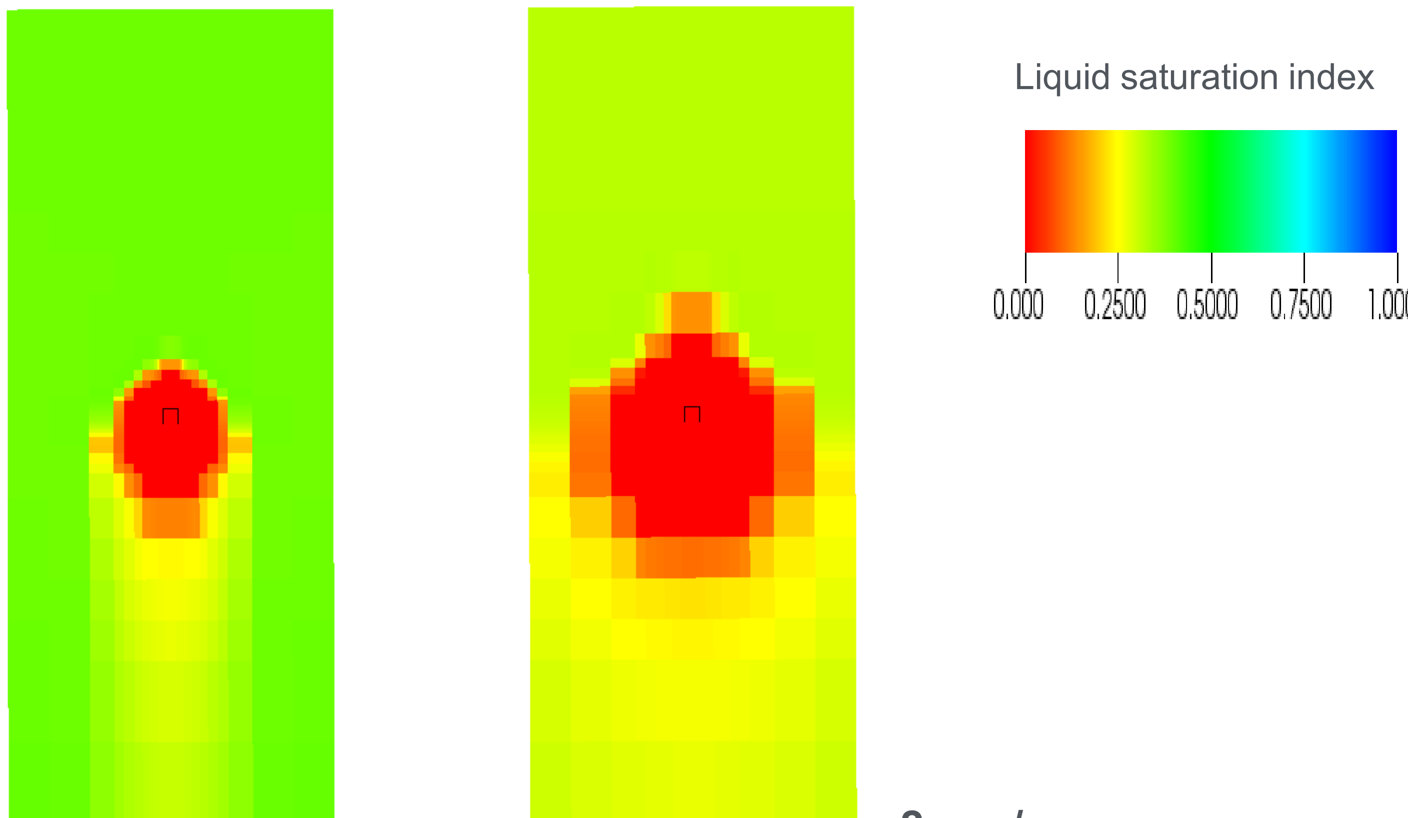
The 2 mm/year case has slightly higher temperatures because of less latent heat of vaporization to overcome and slightly lower thermal conductivity



Liquid Saturation Index at Time of Maximum Dryout

40 m x 80 m
Vertical cross section

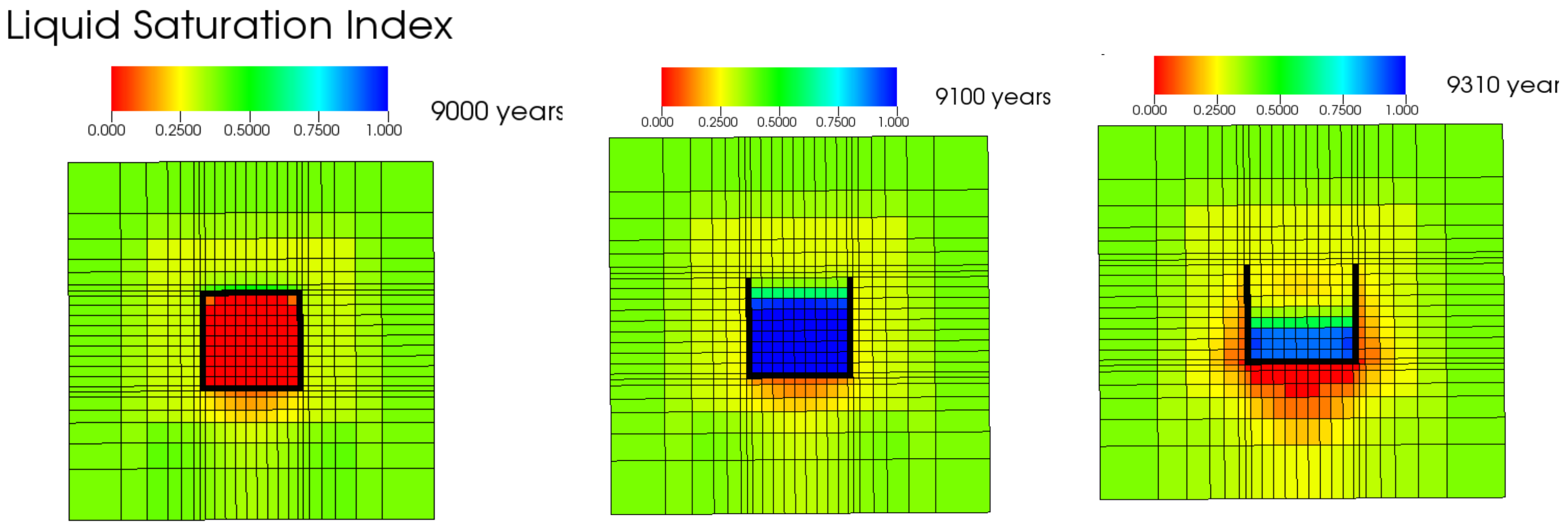
Note that dryout does not extend to the pillar centerline between drifts



10 mm/year
500 years postclosure

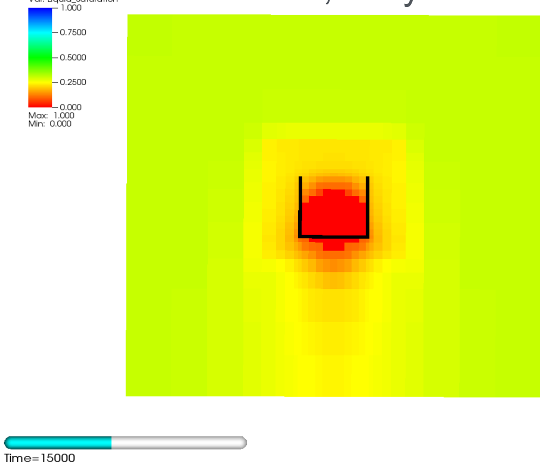
2 mm/year
750 years postclosure

Liquid Saturation Index for 10 mm/yr Case, 400 W Criticality Event

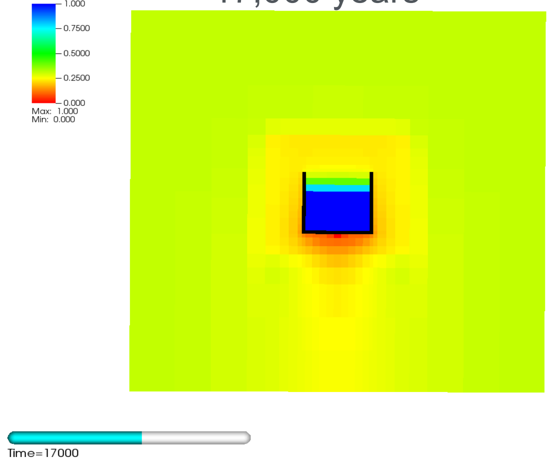


Liquid Saturation Index for 2 mm/yr Case, 100 W Criticality Event

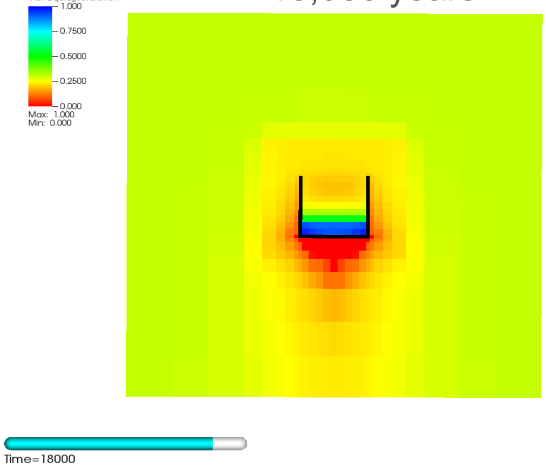
15,000 years



17,000 years



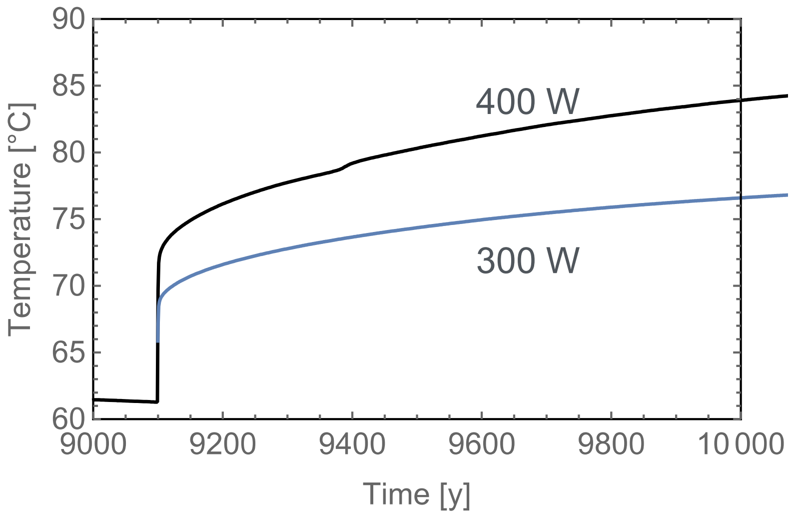
18,000 years



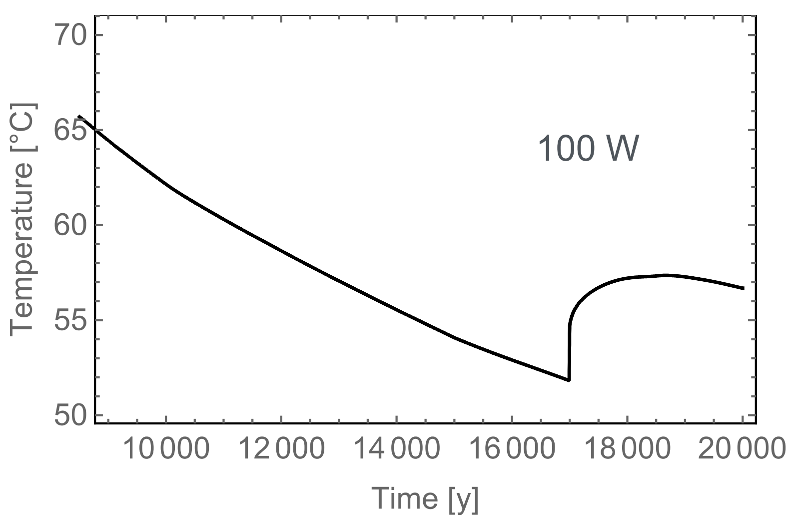
- 100 W event could desaturate the package in about 100 years
- Evaporation without boiling is sufficient to keep the waste package dry in low infiltration unsaturated alluvium

Post-breach Waste Package Temperatures

10 mm/year



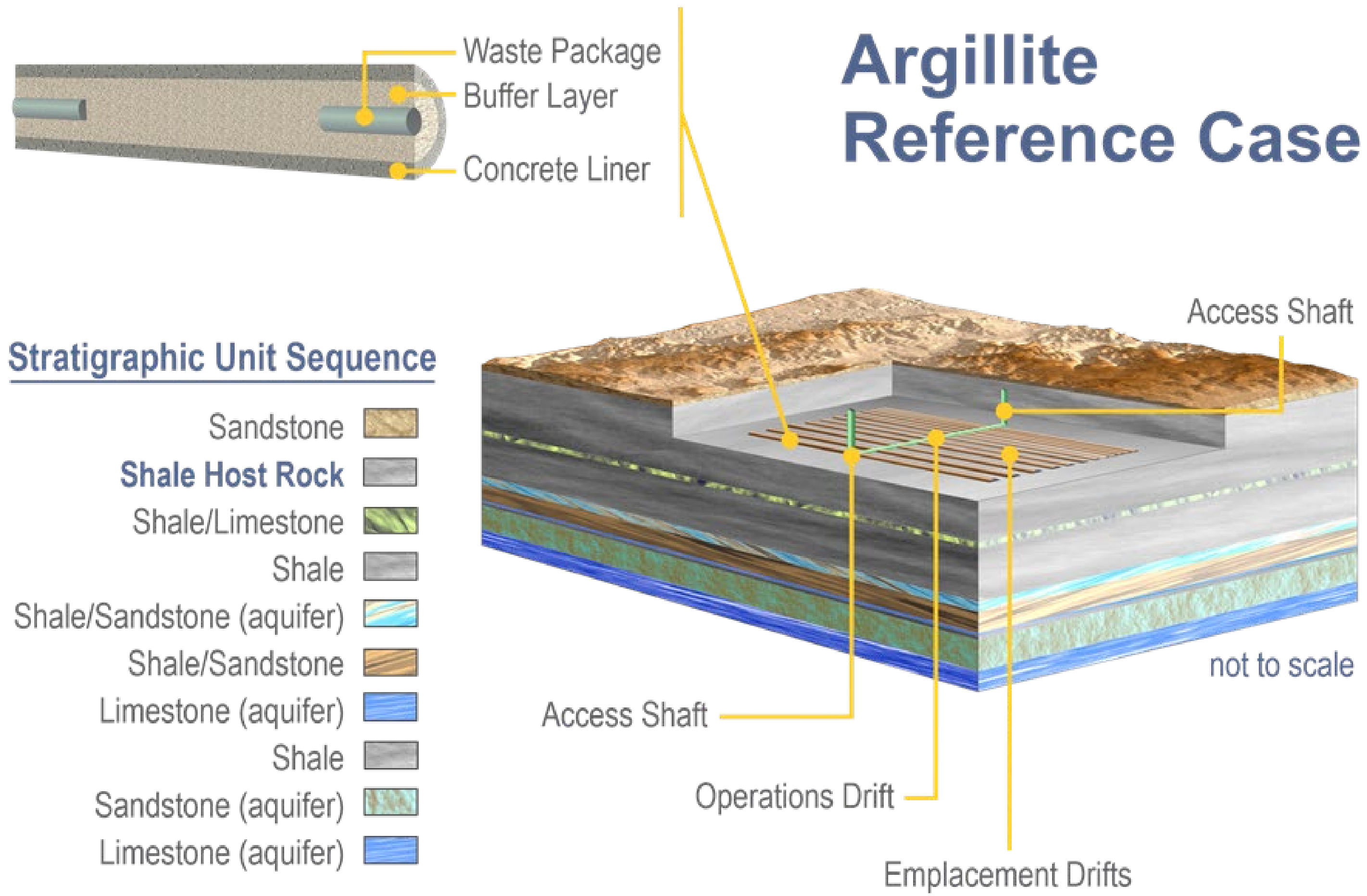
2 mm/year



Conclusions for Unsaturated Case

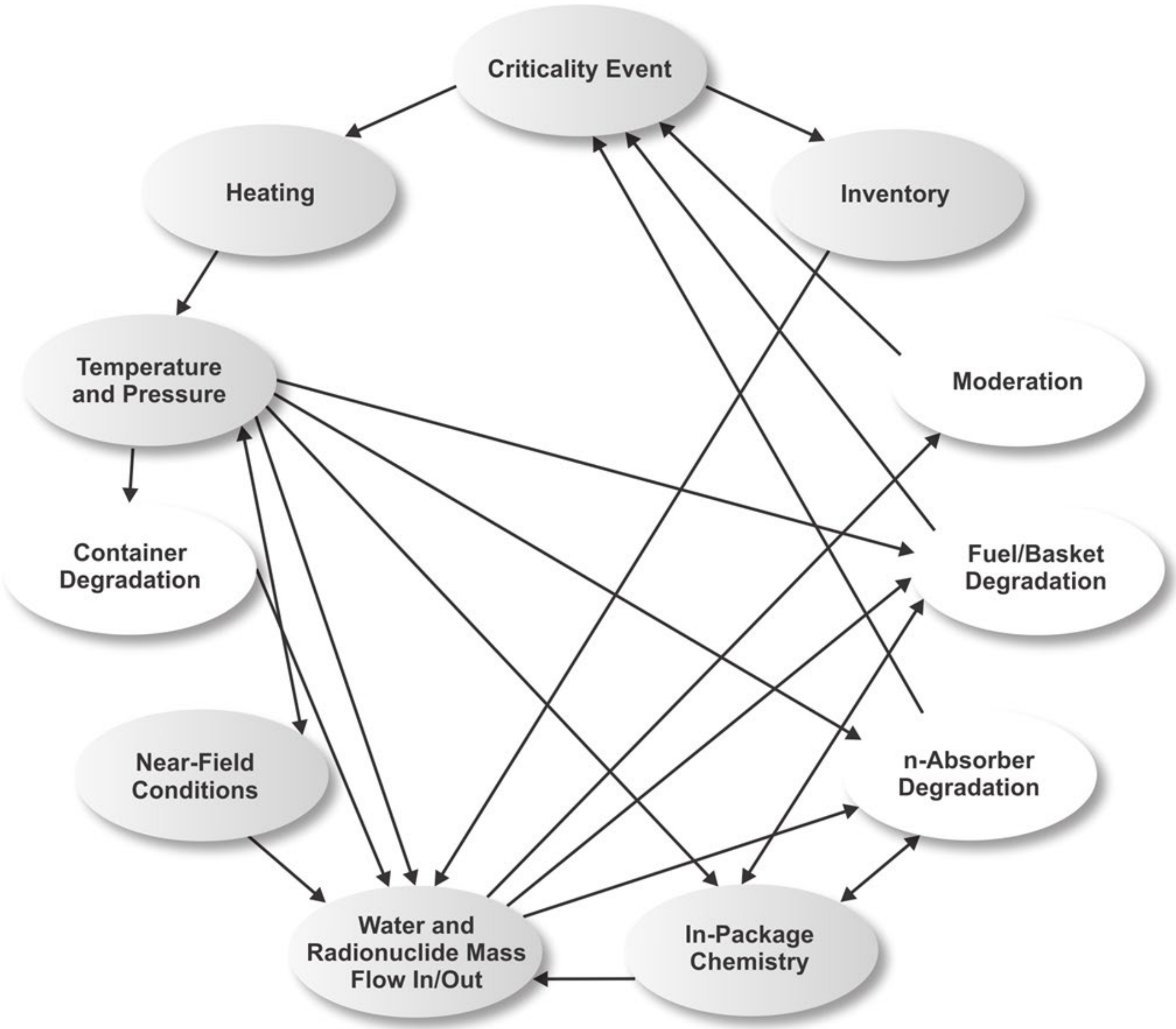
- DPC center temperature peaks around 20 years after closure
  - ~235ºC for 2 mm/year case
  - ~225ºC for 10 mm/year case
- Dryout zones around individual DPCs do not coalesce, allowing for vertical drainage
- Criticality is possible after water returns to the emplacement drifts
  - ~9,100 years postclosure for 10 mm/year case
  - ~17,000 years postclosure for 2 mm/year case
- Long-term average power output from criticality event is limited by thermal hydrology of the unsaturated alluvium
  - <400 W per DPC for 10 mm/year case
  - <100 W per DPC for 2 mm/year case

Hypothetical Argillite Repository



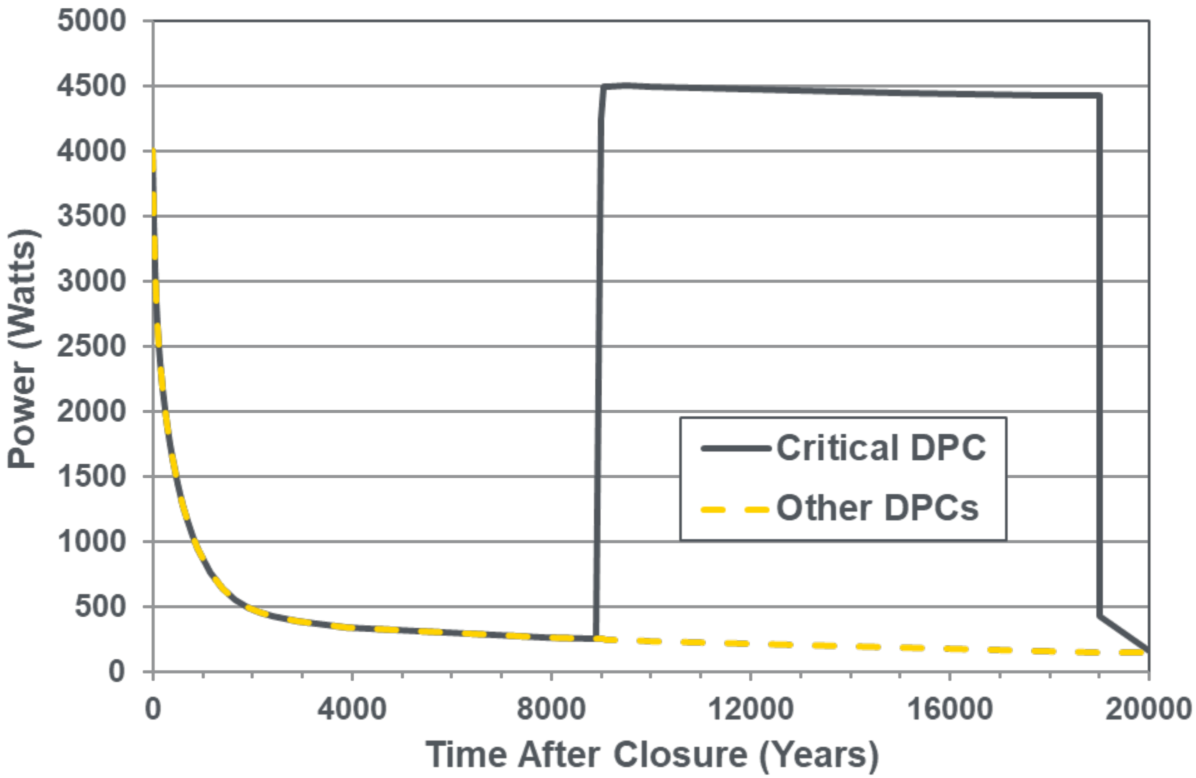
Mariner et al. 2017

Coupling Scheme Between Processes

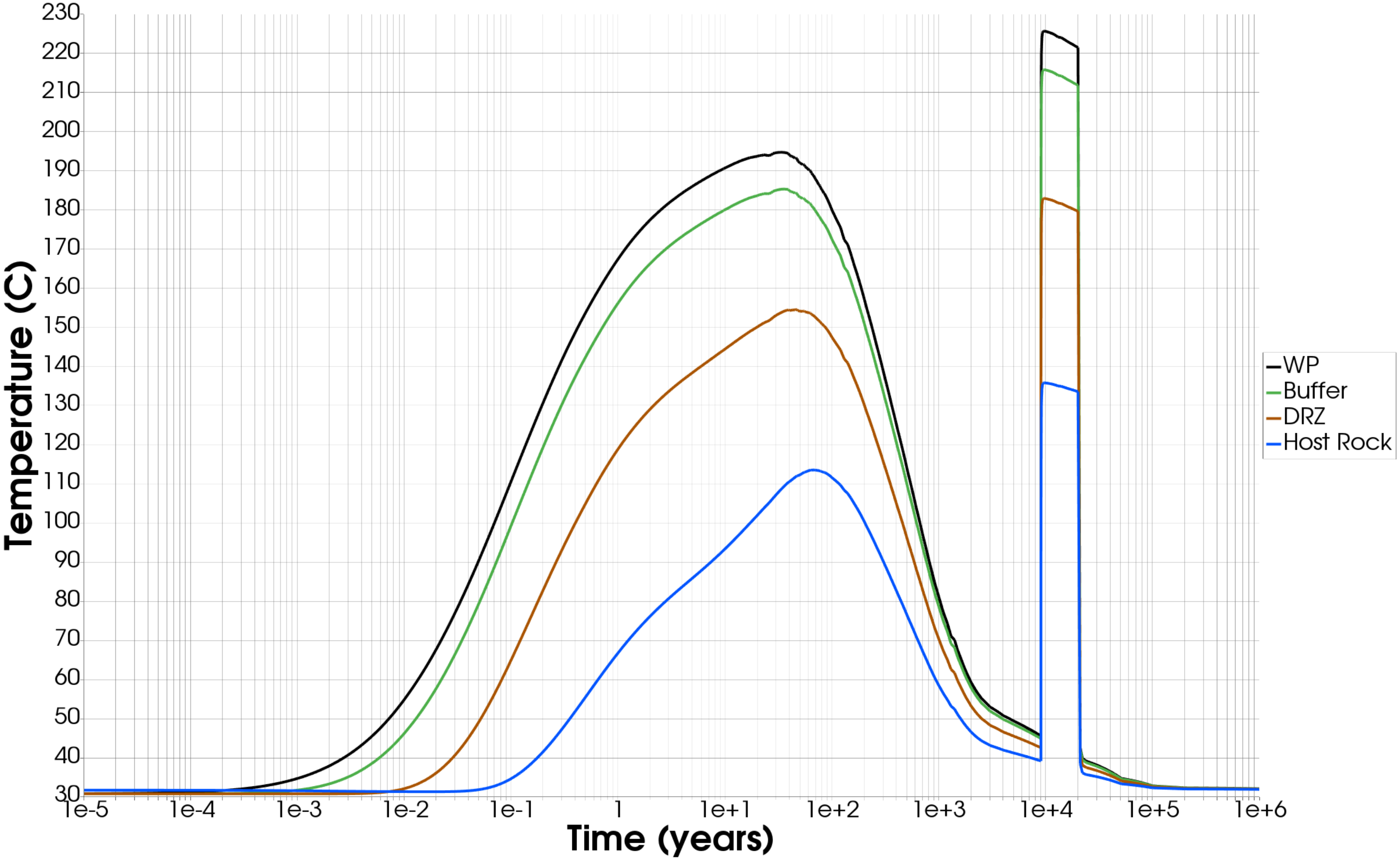


Thermal Analyses – Power Generation

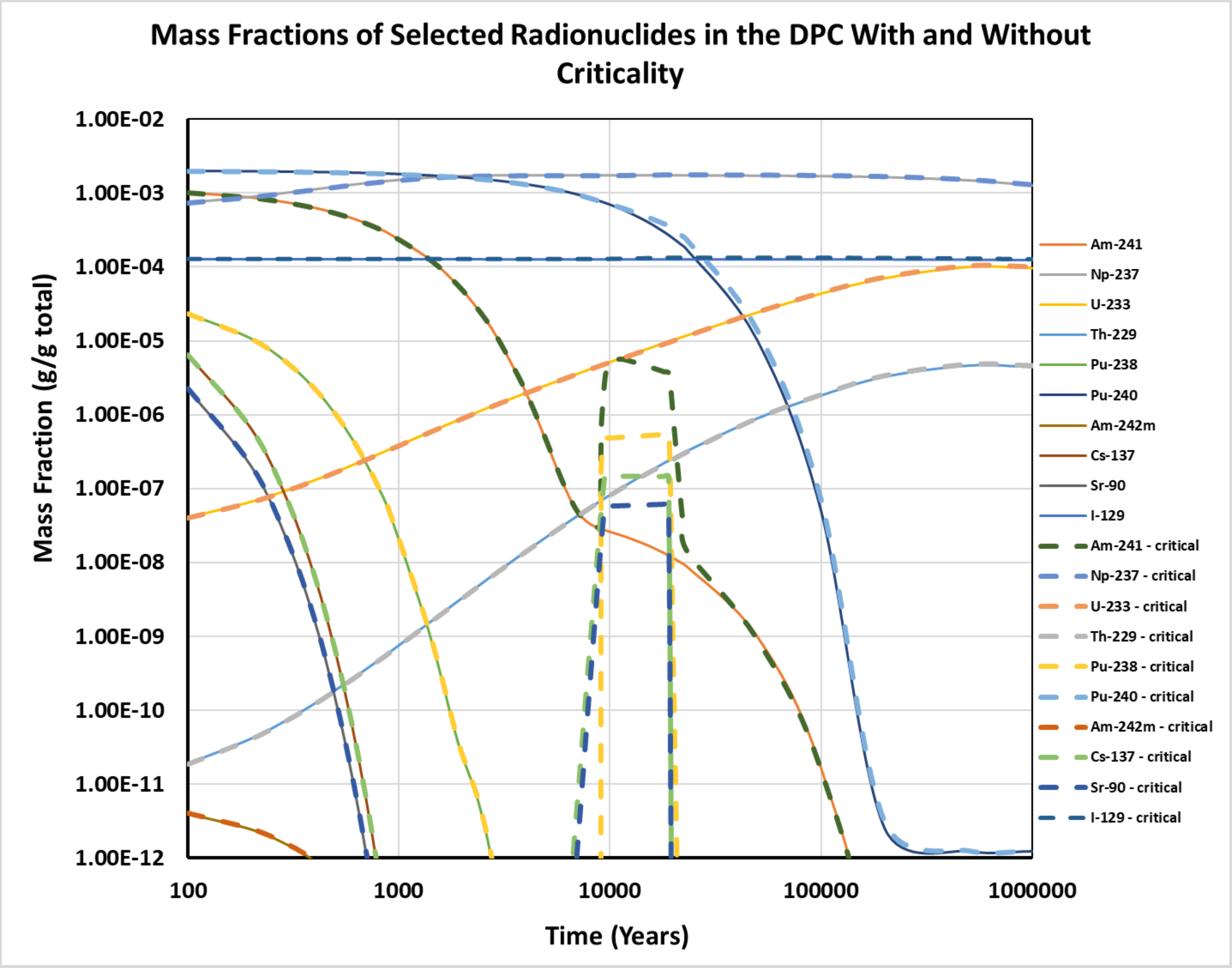
- For saturated repository, maximum power produced by steady-state critical event is assumed to be 4 kW based on scoping calculation
- Boiling point is 264 °C
- Heat transfer is via conduction



Temperature vs. Time



Inventory Changes



Factors Affecting In-Package Chemistry During Steady-State Critical Event

- Chemistry inside waste package affected by
  - New fission products
  - Increased temperature
  - Increased radiolysis
  - Stainless steel corrosion
  - Spent fuel degradation
- Increased temperature accelerates corrosion rates of DPC materials
- Steel corrosion leads to reducing conditions (saturated shale repository) BUT
- Radiolysis produces oxidants (H_2O_2, NO_2 in unsaturated case)
- Coupled calculation of radiolysis, steel degradation, spent fuel degradation needed

Radionuclide Solubilities

- Degradation of SNF produces relatively insoluble actinide oxides containing Pu, U, Am, Np, and Th
- Solubilities of these oxides control actinide release and tend to decrease as temperature increases
- pH affects radionuclide solubilities; in general, actinide solubilities are higher away from neutral pH
- For fission products that are not solubility limited (e.g., I), releases into the host rock depend on SNF degradation rates and uptake by backfill
- As temperature increases, there is a decrease in solubilities of oxides and carbonates of neutron poisons (^{149}Sm, ^{157}Gd, ^{143}Nd)

Engineered Barrier System Degradation

- In the hypothetical repositories assumed in this work, engineered barriers consist of
  - Waste package outer barrier
  - DPC
  - Fuel cladding
  - Backfill (bentonite)
- Waste package is assumed to have failed for critical event to occur – no longer serving as an engineered barrier but is still right circular cylinder
- Cladding is assumed to maintain configuration but have small holes
- Bentonite backfill is assumed to not act as a barrier to radionuclide transport during critical event

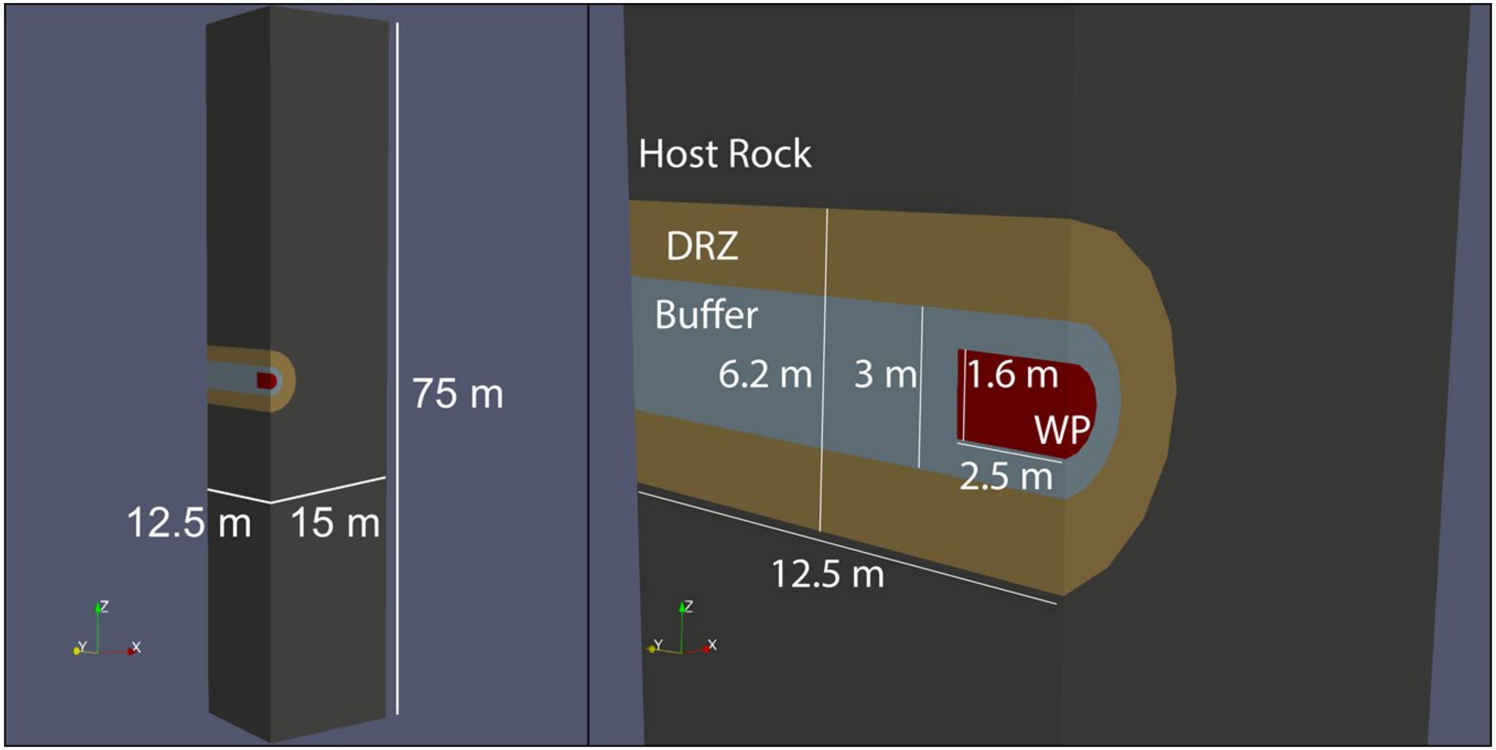
Termination of Criticality

- Radioactive decay provides limited changes in reactivity after ~100,000 years.
- Buildup of ^{233}U from decay of ^{237}Np results in a relatively small reactivity increase over a few million years
- Depletion and production of fissile material from additional burnup from steady-state postclosure criticality occurs very slowly
  - For saturated repository, 4kW for 10,000 years results in additional ~1 GWd/MTU average burnup
- Grid spacer corrosion/collapse resulting in uniform pin pitch reduction of ~3 mm could result in permanent termination of criticality for most DPCs
- Dissolution and transport of neutron-absorbing isotopes could increase reactivity
- Dissolution and transport of ^{239}Pu ($t_{1/2}$ = 24,100 years) prior to about 100,000 years could reduce reactivity
- Dissolution and transport of uranium would likely have a small effect on reactivity because of the large mass of uranium in a DPC

Performance Assessment Calculations

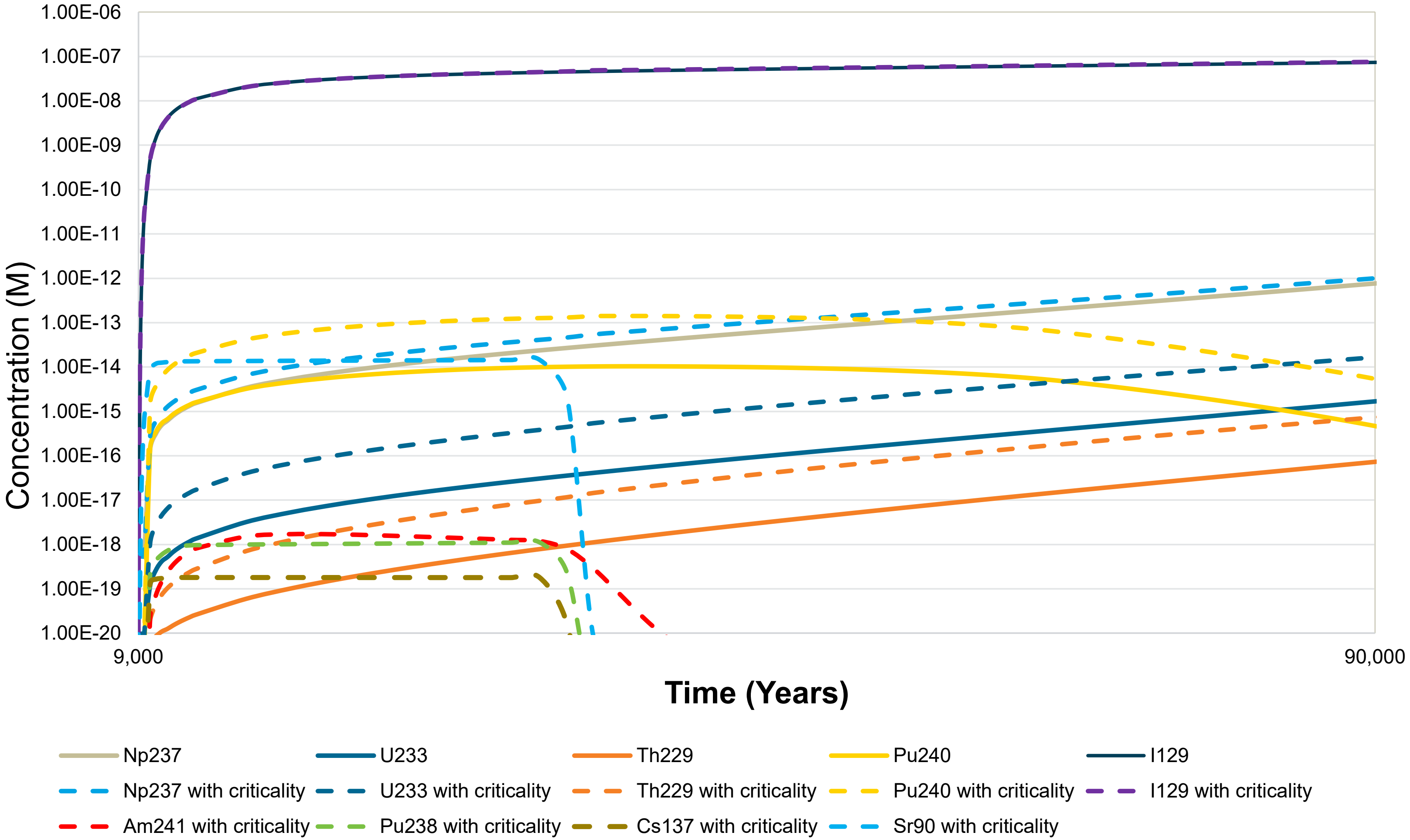
- Developed a criticality sub-module in PFLOTRAN
  - Added capability to specify a steady-state heat from a critical event for a specified period of time
  - Added capability to change radionuclide inventory at a specified time
- Considered case without steady-state critical event and case with steady-state critical event
- Present results for saturated shale case only; unsaturated alluvial case was too dry for chemistry model to run

Performance Assessment Model Setup



Model domain for a 3D, single-drift, single-waste package simulation using quarter symmetry boundaries.

PFLOTRAN Model Results in Shale Next to Drift



Conclusions (1/2)

- Developed new criticality sub-module for PFLOTRAN that accounts for additional heat and additional radionuclides generated by postclosure critical event
- The power generated by a postclosure steady-state critical event in a saturated repository has the potential to be much higher than that in an unsaturated repository
- Qualitative insights into in-package chemistry and radionuclide solubility
  - Acids produced by additional radiolysis can be buffered by stainless steel corrosion products
  - Coupled calculation of radiolysis, steel degradation, spent fuel degradation needed
  - Both actinides and neutron-absorbing radionuclides are less soluble at higher temperatures, but also affected by pH

Conclusions (2/2)

- Behavior of EBS in saturated repository with postclosure critical event not well understood, needs further study
- Insights into permanent criticality termination
  - Fuel can remain reactive for entire postclosure period
  - Identified termination mechanisms for future study
- Insights into repository performance
  - Importance of newly generated radionuclides to dose is dependent on radionuclide travel time from repository to dose receptor
  - Concentration of ^{129}I in the near field increases about 3% in the long term
  - Concentration of ^{237}Np in the near field increases about 50% in the long term
  - Concentrations of ^{240}Pu, ^{229}Th, and ^{233}U in the near field increase about an order of magnitude in the long term
  - ^{241}Am, ^{90}Sr, ^{137}Cs, and ^{238}Pu appear only in the case with criticality because they had decayed to nothing in the case without criticality.

References

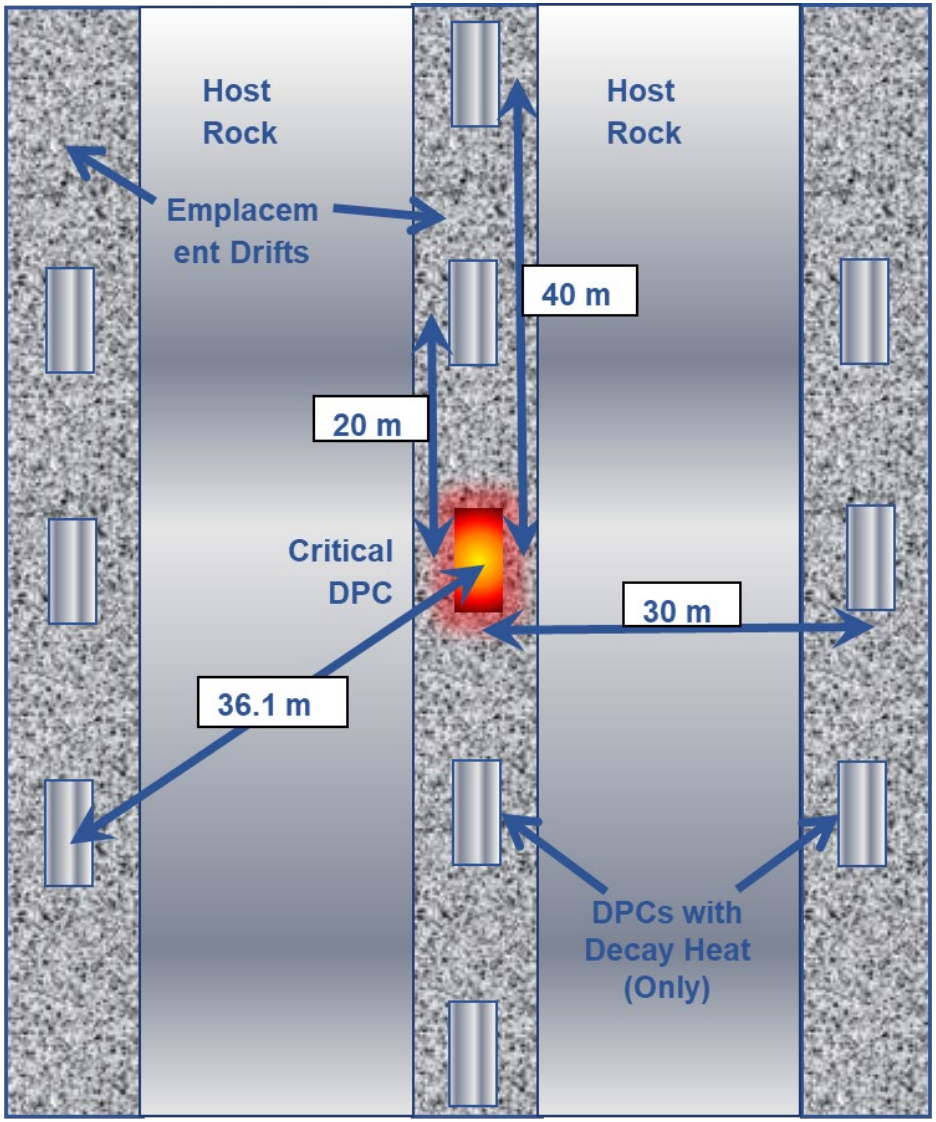
Mariner P.E., E.R. Stein, J.M. Frederick, S.D. Sevougian, and G.E. Hammond 2017. *Advances in Geologic Disposal System Modeling and Shale Reference Case*. SFWD-SFWST-2017-000044; SAND2017-10304R. Albuquerque, NM: Sandia National Laboratories.

Mariner, P.E., E.R. Stein, S.D. Sevougian, L.J. Cunningham, J.M Frederick, G.E. Hammond, T.S. Lowry, S. Jordan, and E. Basurto 2018. *Advances in Geologic Disposal Safety Assessment and an Unsaturated Alluvium Reference Case*. SFWD-SFWST-2018-000509; SAND2018-11858R. Albuquerque, NM: Sandia National Laboratories.

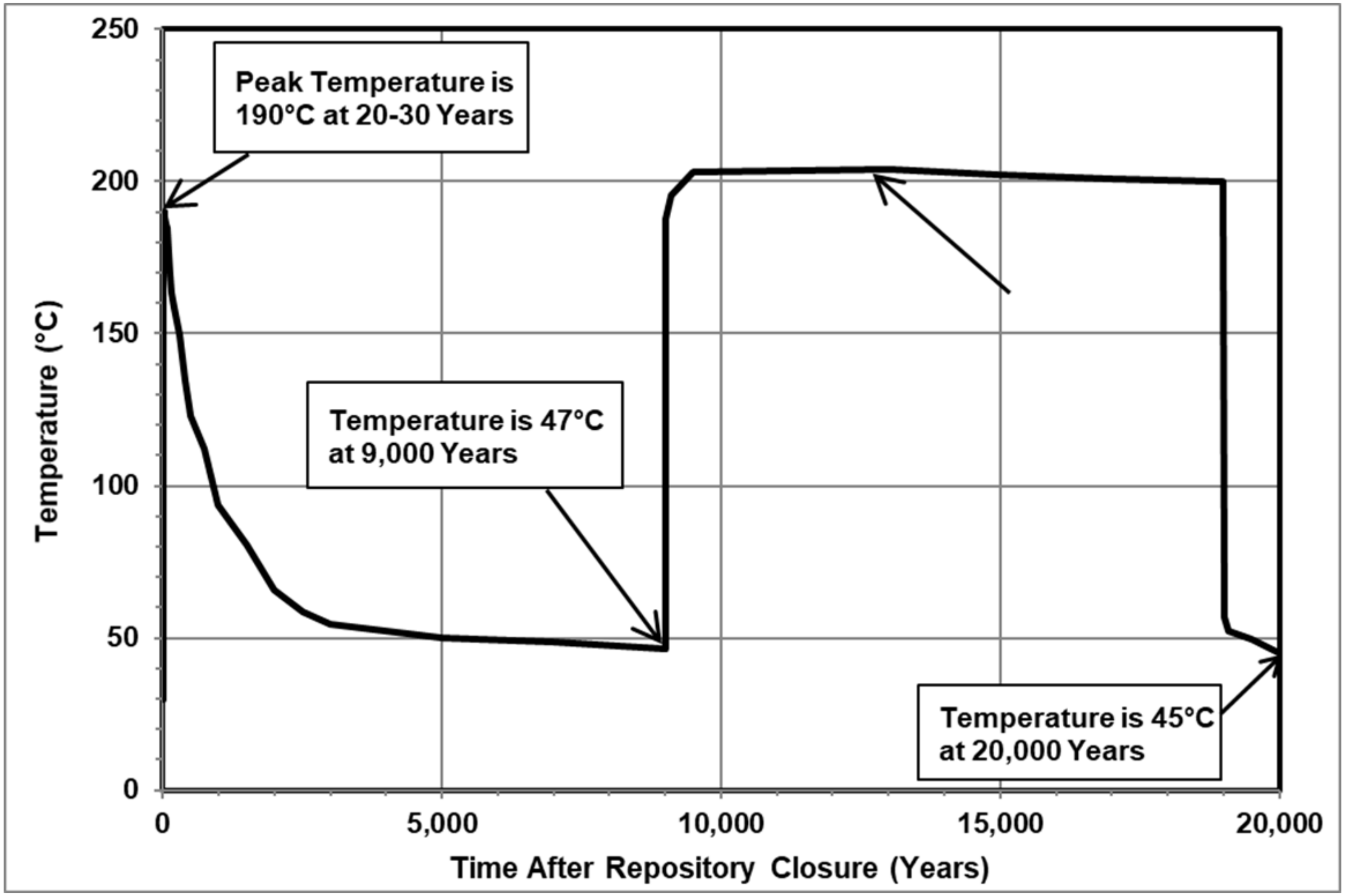
Questions?

Backup Slides

Plan View of Model for Thermal Analyses

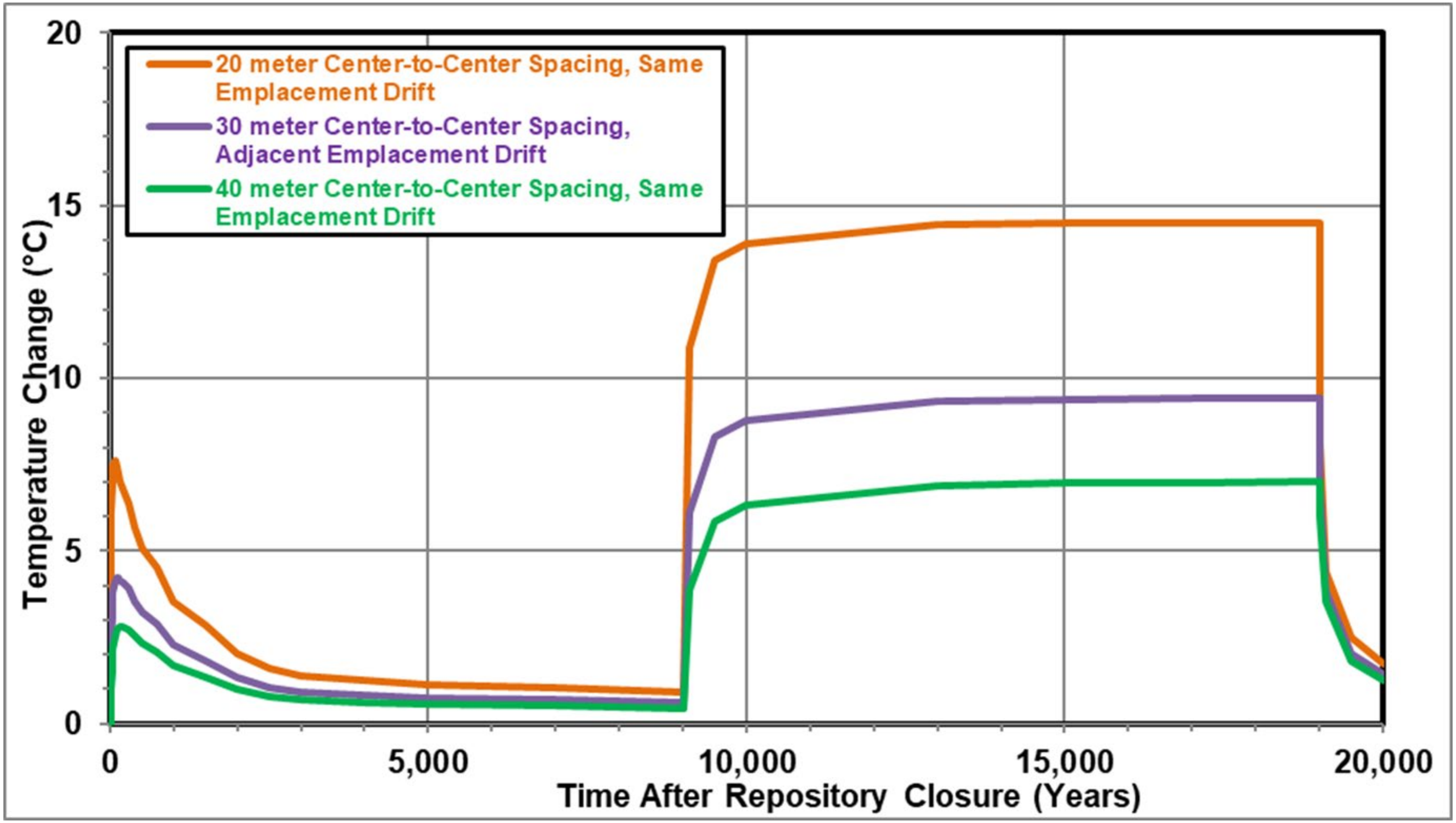


Waste Package Temperature



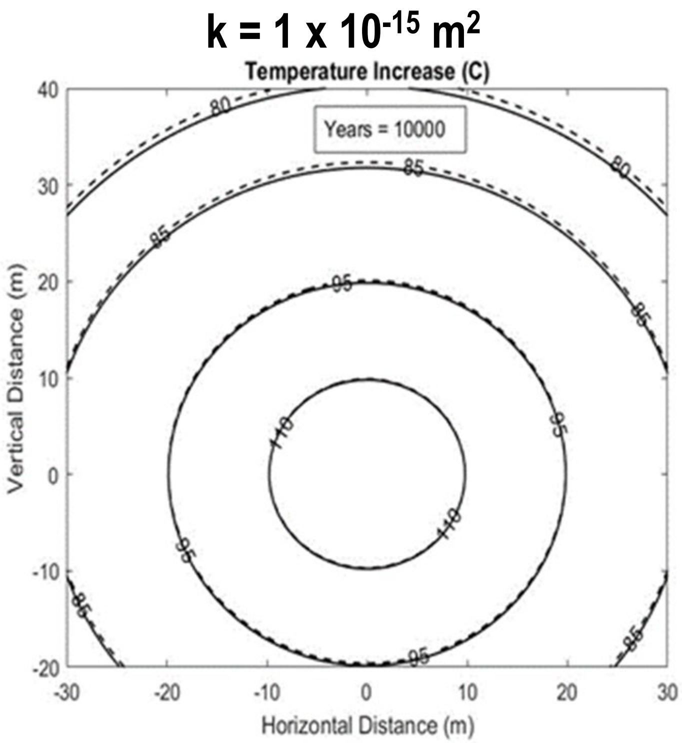
Temperature History for 4 kW Criticality from 9,000 to 19,000 Years with Thermal Properties for Shale Host Rock.

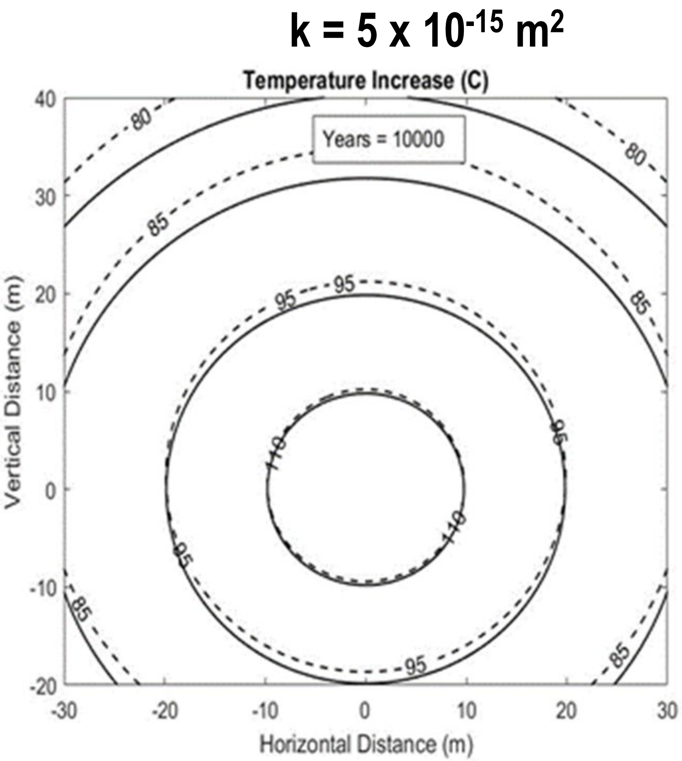
Temperature Change – Adjacent Waste Packages



Temperature Change in Adjacent DPCs Separated by 20, 30, and 40 meters from the Central (Critical) DPC in Shale Host Rock

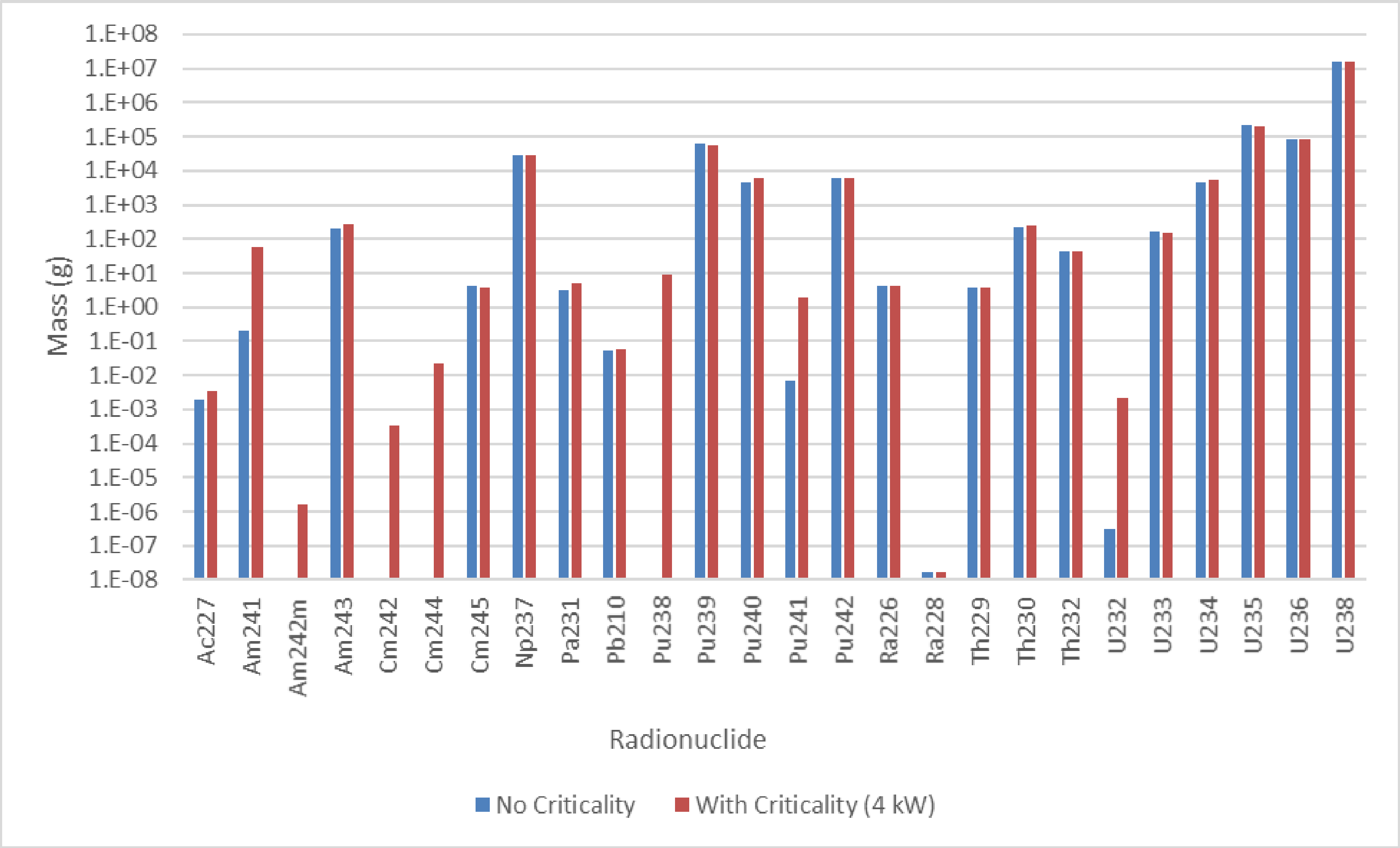
Effects of Convection – 10,000 Years



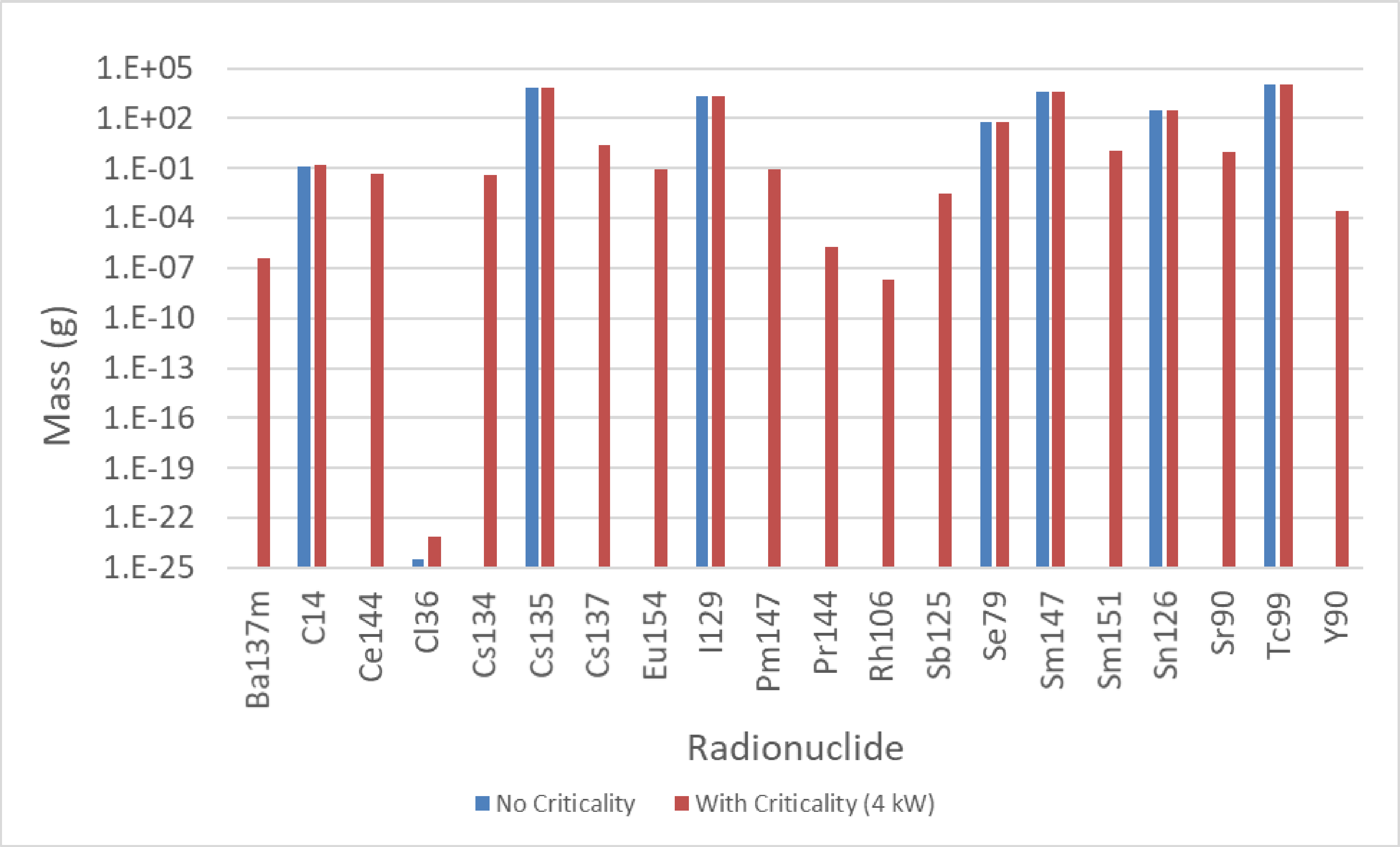


Contours of Increased Temperature Above Ambient at 10,000 Years. Solid Lines are Conduction Only, and Dashed Lines are Conduction and Convection

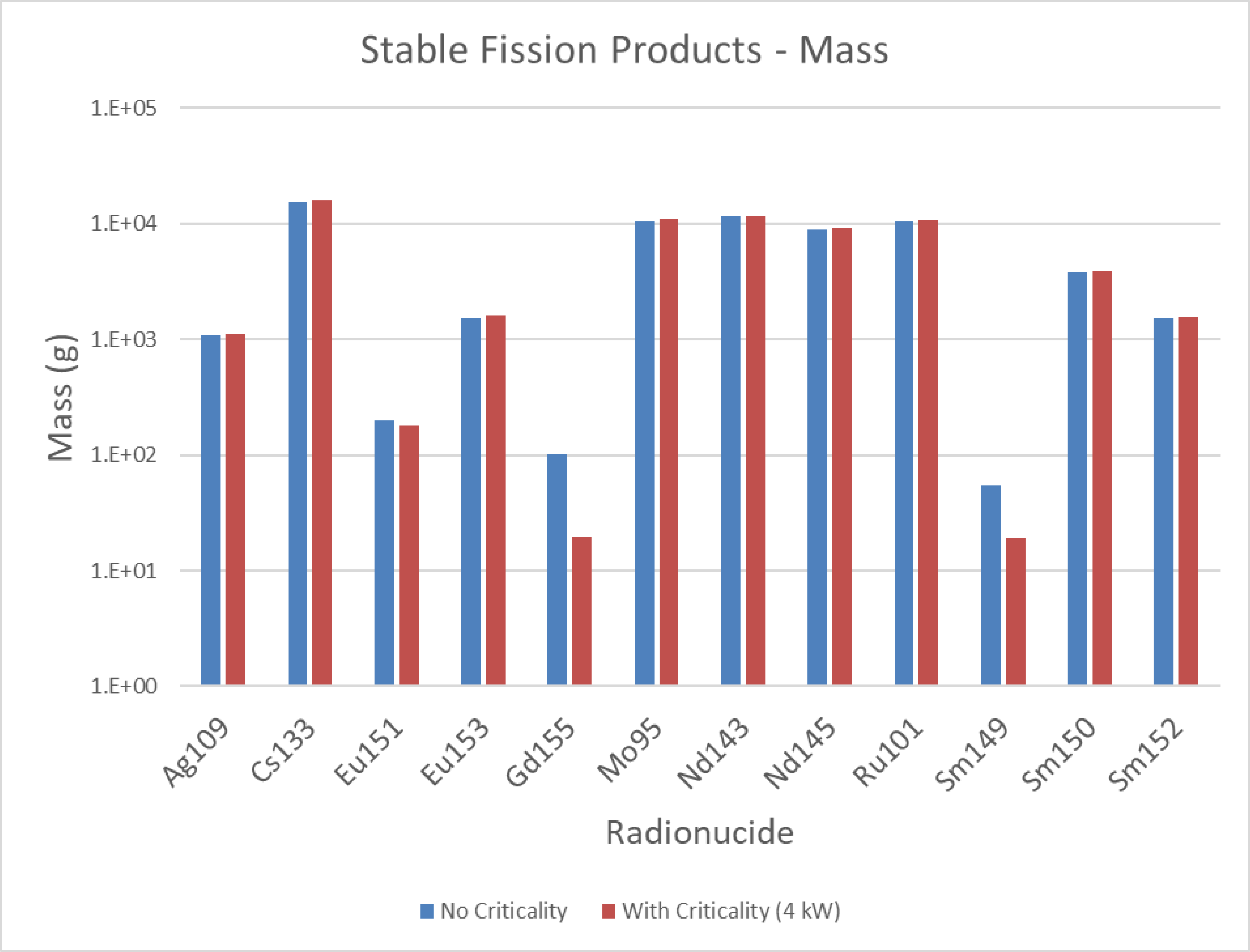
Inventory Changes – Actinides and Their Decay Products



Inventory Changes – Fission and Activation Products



Inventory Changes – Stable Fission Products



Chemistry Inside the Waste Package During Steady-State Critical Event

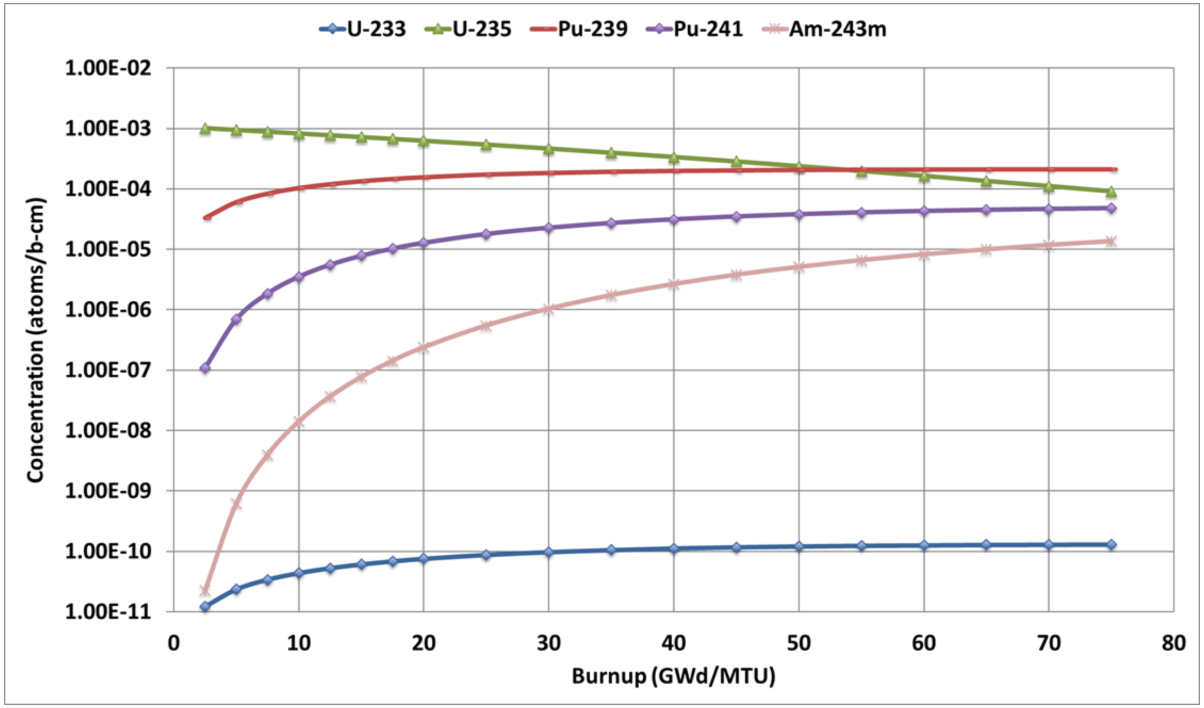
- Arrhenius equation predicts corrosion rates of SS
  - 0.00008 µm/day at 100° C (alluvial repository)
  - 0.002 µm/day at 169° C (shale repository)
- In hypothetical unsaturated alluvium environment, lower SS corrosion rate is not likely to produce enough trevorite to buffer acid produced by radiolysis (assuming “bathtub”)
- In hypothetical saturated shale environment, higher SS corrosion rate is likely to produce enough trevorite to buffer acid produced by radiolysis and inhibit oxidative degradation of SNF (assuming “bathtub”)

Termination of Criticality - Approach

- What, how, and when could SNF or DPC characteristics be affected by disposal events and processes such that the potential for criticality initiation or continuation becomes permanently significantly diminished?
- To begin to answer this question, examined eight typical criticality control parameters
- Determined four parameters were worthy of further examination
  - Radioactive decay
  - Burnup
  - Irreversible geometry changes
  - Compositional changes due to corrosion or dissolution

Reactivity Perturbations Due to Burnup

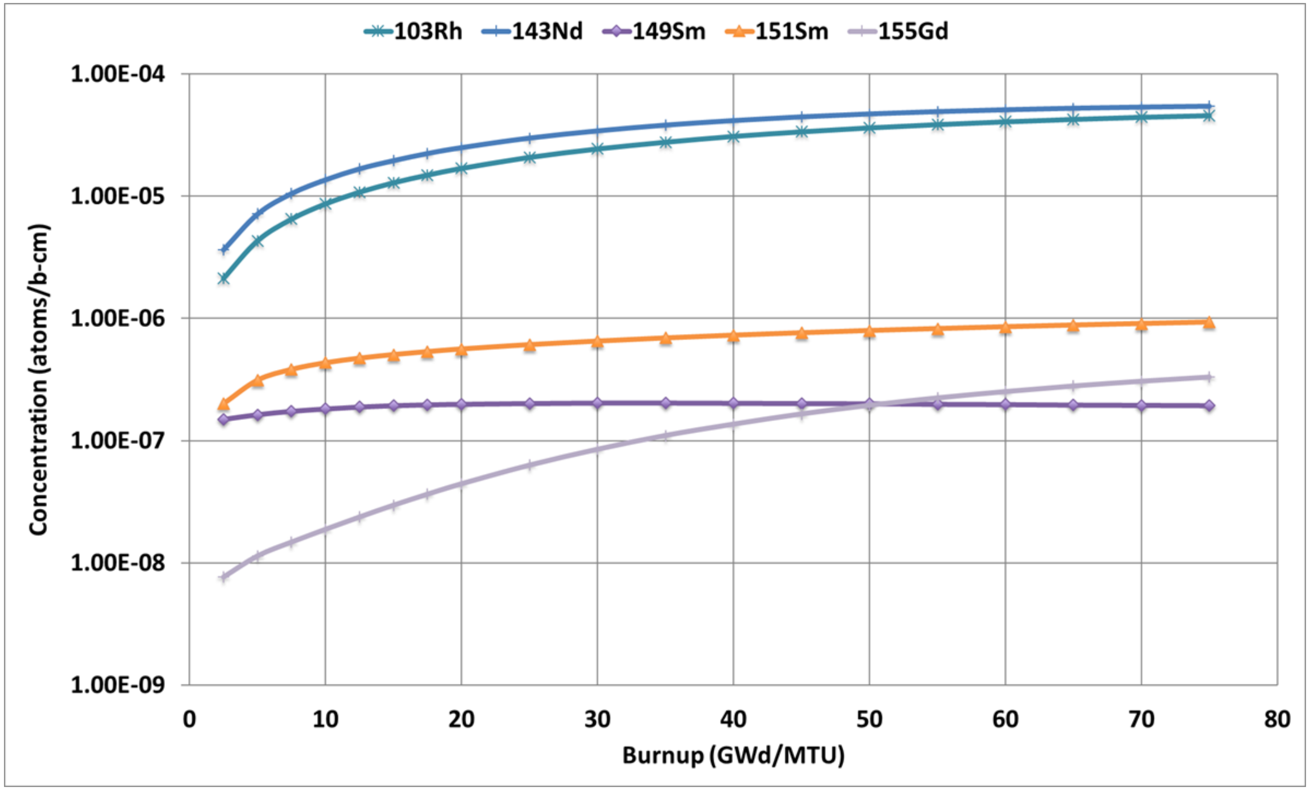
Pu-239 becomes the primary fissile isotope; it reaches an equilibrium concentration at ~30 GWd/MTU



Concentration of Fissile Isotopes as a Function of PWR SNF Burnup

Reactivity Perturbations Due to Burnup (cont'd)

Fission product neutron absorber concentration continues to increase



Concentration of Neutron Absorber Isotopes as a Function of PWR SNF Burnup