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Consequences of Nuclear Criticality in DPCs after Disposal

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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
- Michael Nole – PFLOTRAN calculations
- Jeralyn Prouty – Reference repository diagrams

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Background

- The US Department of Energy is responsible for disposing of spent nuclear fuel
- DOE's plan to dispose of SNF in canisters designed to prevent postclosure criticality was suspended
- DOE investigating feasibility of disposing of SNF already loaded in dual-purpose canisters
  - Thermal considerations due to higher heat generation rate
  - Handling considerations due to size and weight
  - Postclosure criticality considerations due to performance of neutron absorber and SNF loading
- This presentation focuses on consequences of postclosure criticality, not probability of occurrence

Objectives

- Develop tools to create the ability 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

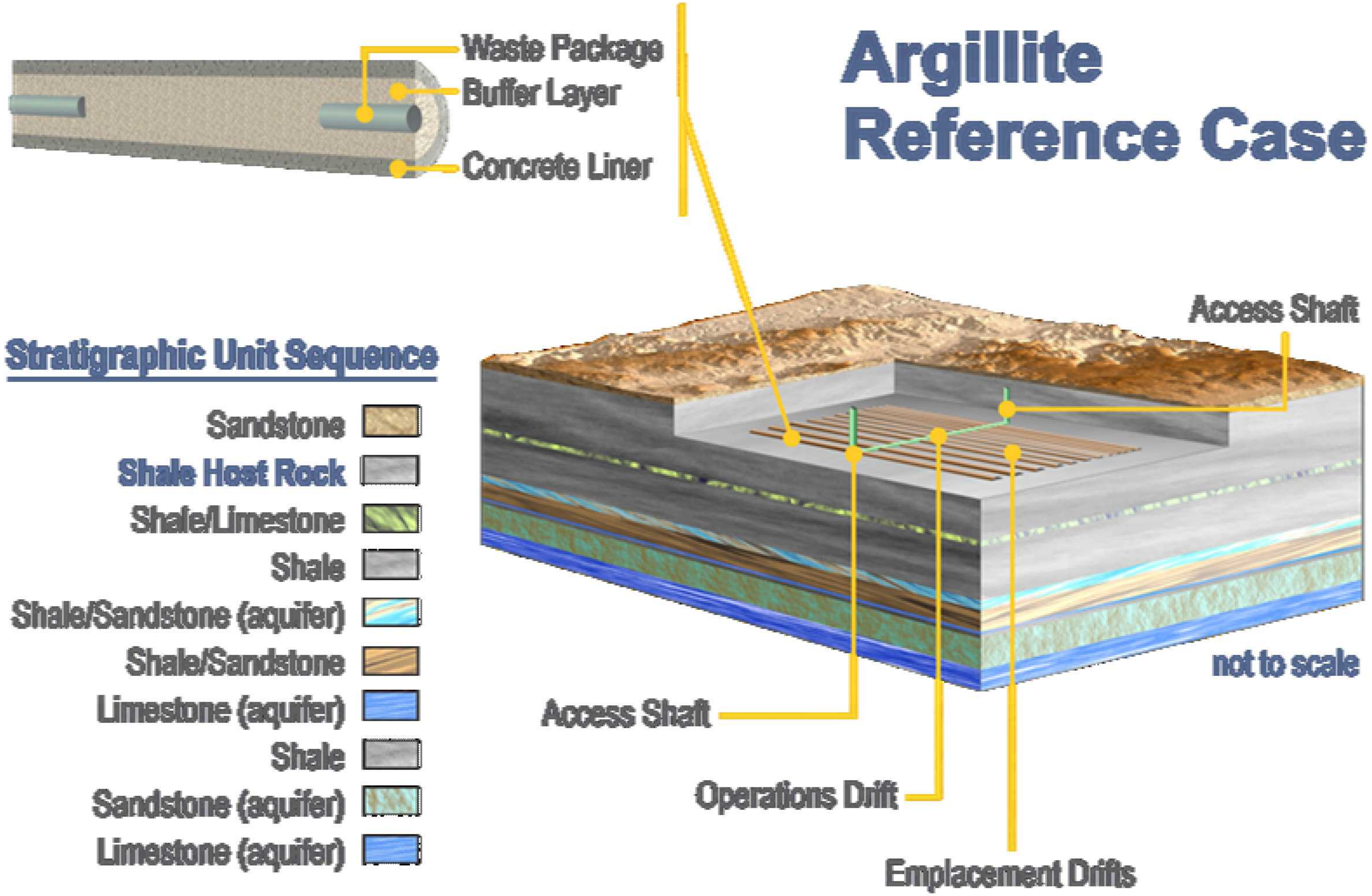
- Two hypothetical repositories considered
  - Saturated repository in shale (Mariner et al. 2017)
    - 500 m depth
    - Backfilled with bentonite
    - Hydrostatic pressure is 50 bars
  - Unsaturated repository in alluvium (Mariner et al. 2018)
    - 250 m depth
    - Backfilled with crushed alluvium
    - 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 Argillite Repository



Mariner et al. 2017

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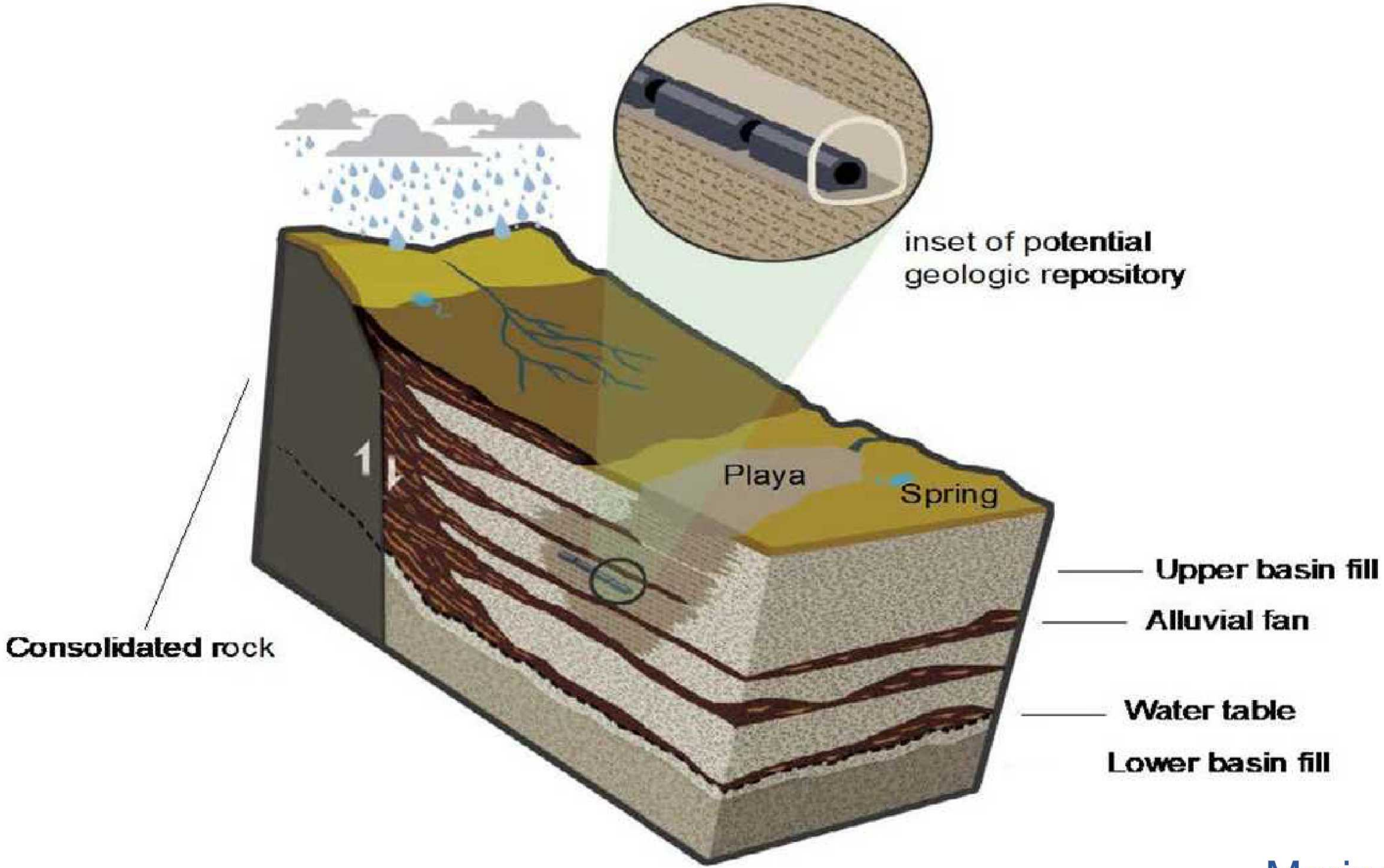
Hypothetical Repository in Alluvium

PLL1

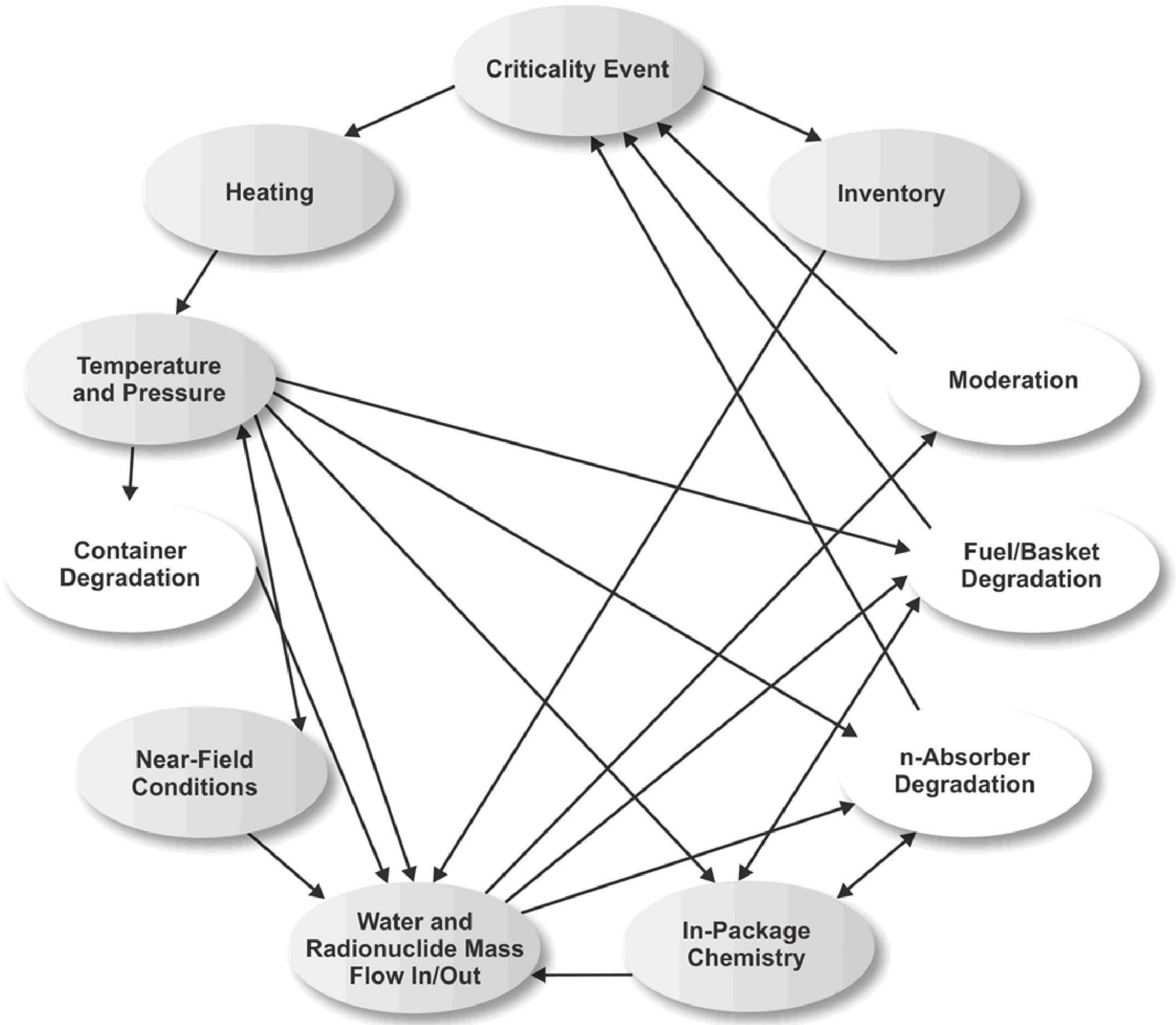


Mariner et al. 2018

Slide 10

PLL1 Price, Laura L, 11/7/2019

Coupling Scheme Between Processes



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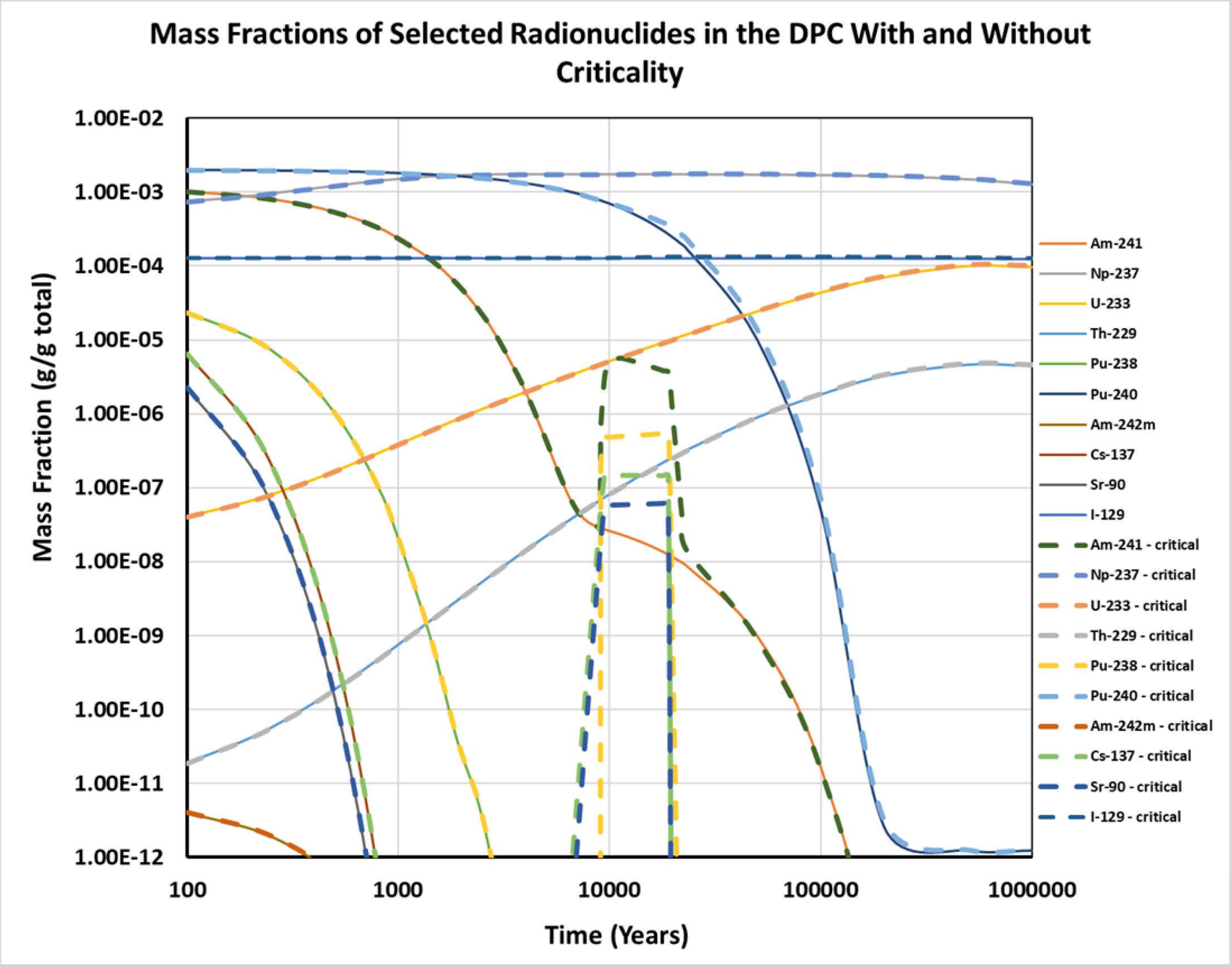
Inventory Changes



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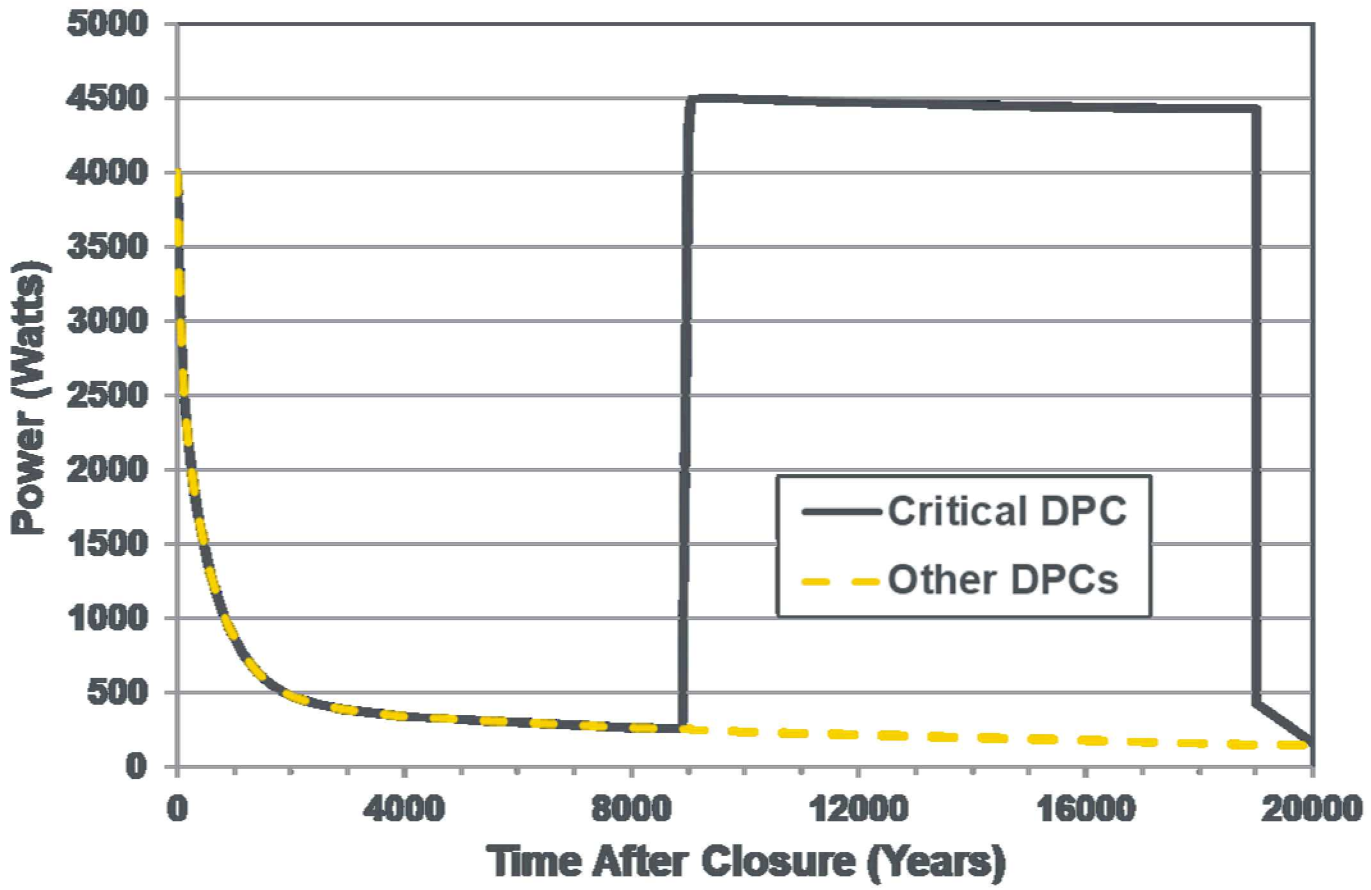
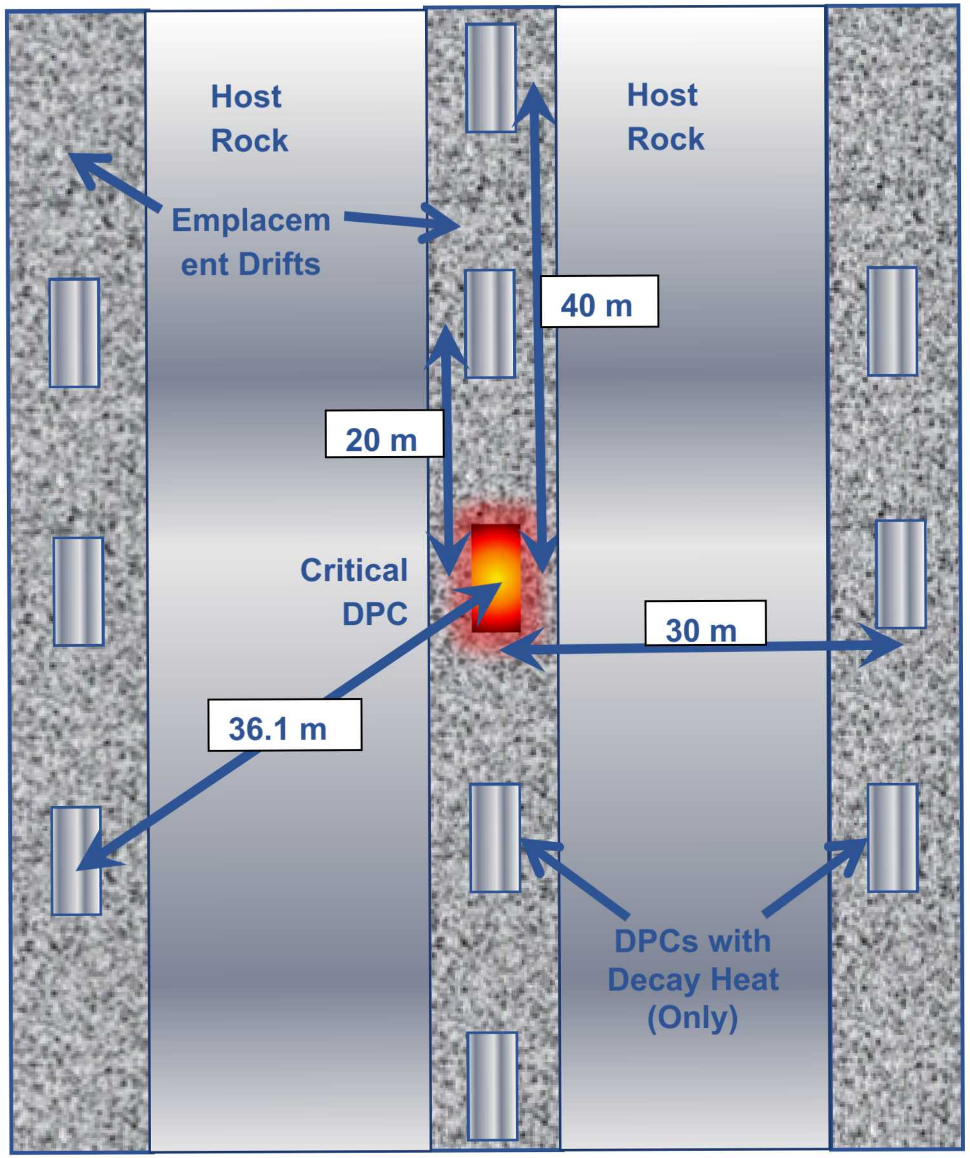
Thermal Analyses – Power Generation

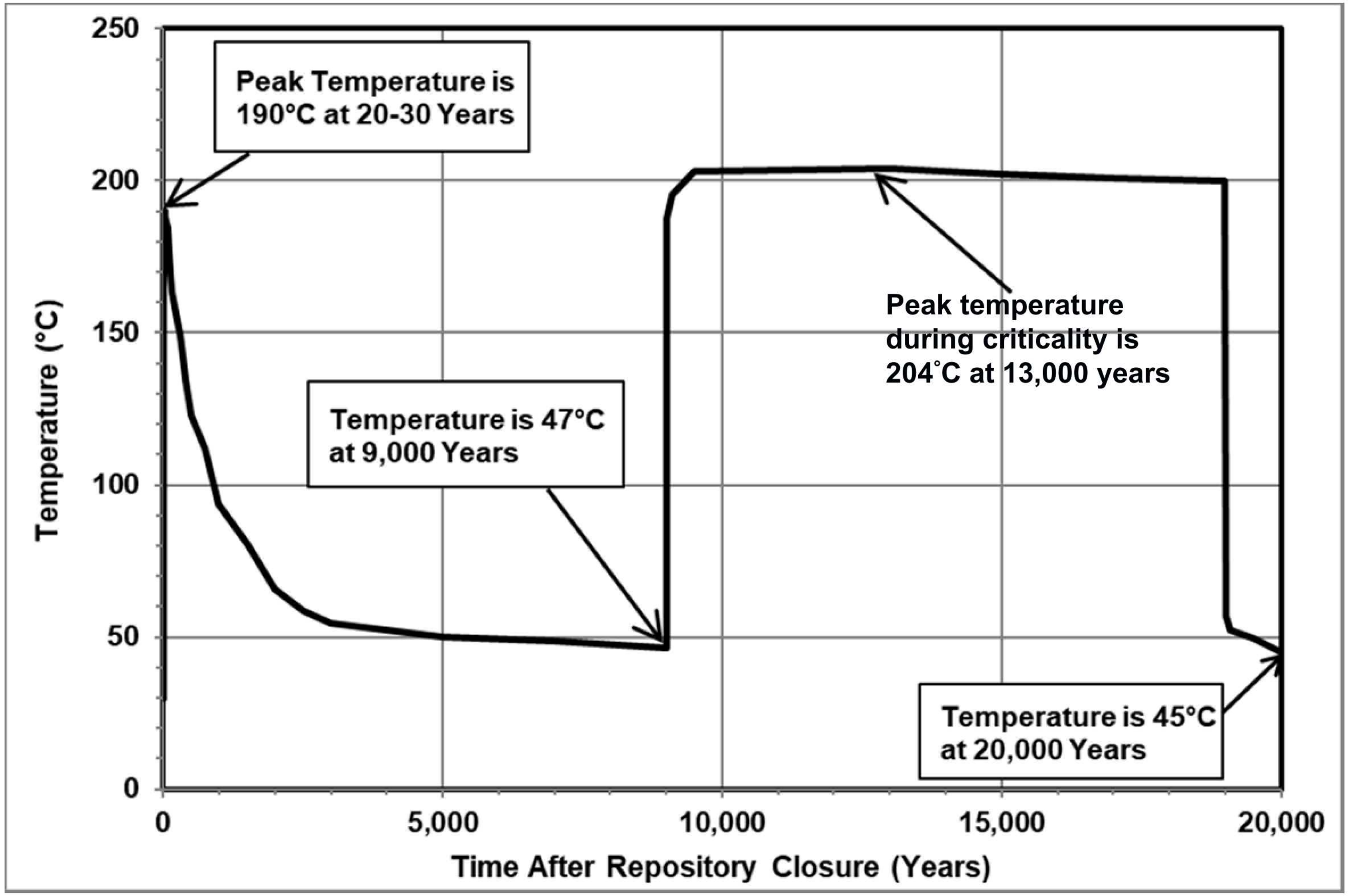
- For saturated repository, maximum power produced by steady-state critical event is assumed to be 4 kW based on scoping calculation
- Boiling point is 264 ^{o}C
- Heat transfer is via conduction



Plan View of Model for Thermal Analyses



Waste Package Temperature – 4kW Steady-State Criticality



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

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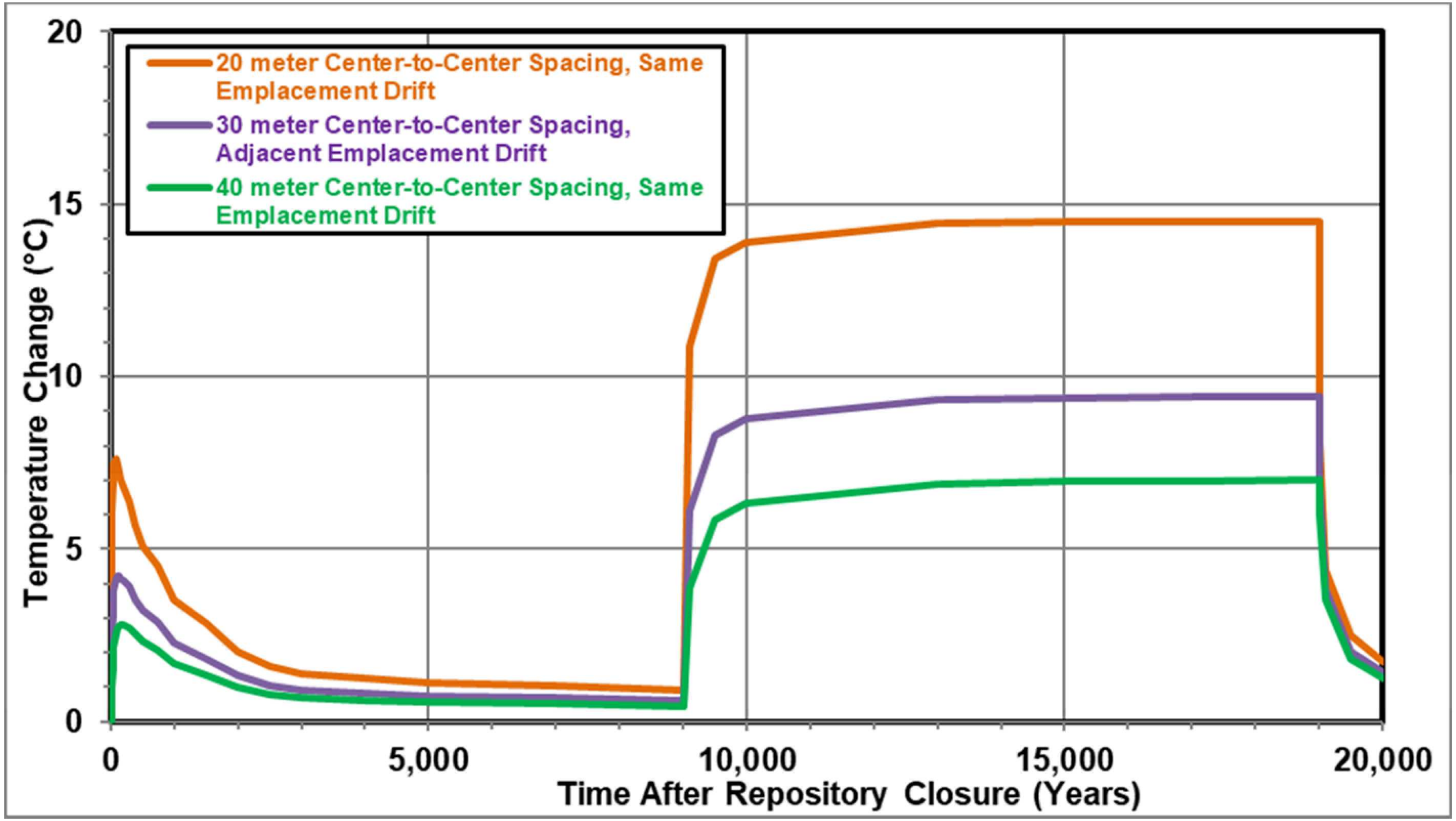
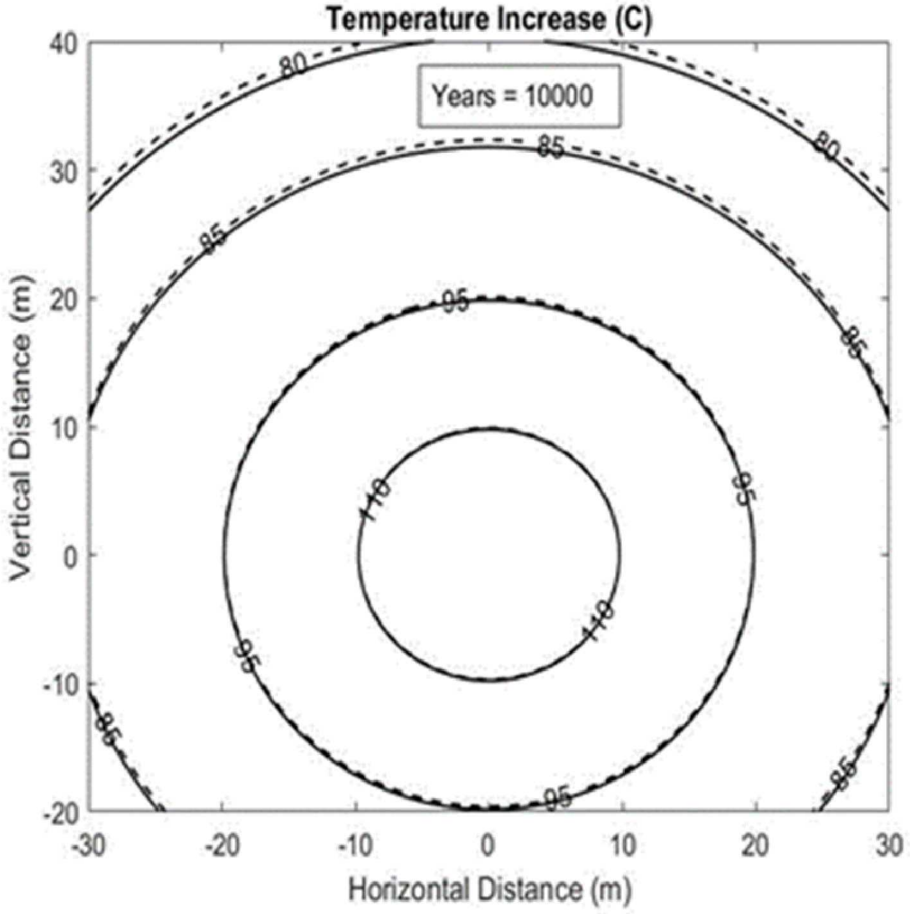
Temperature Change – Adjacent Waste Packages



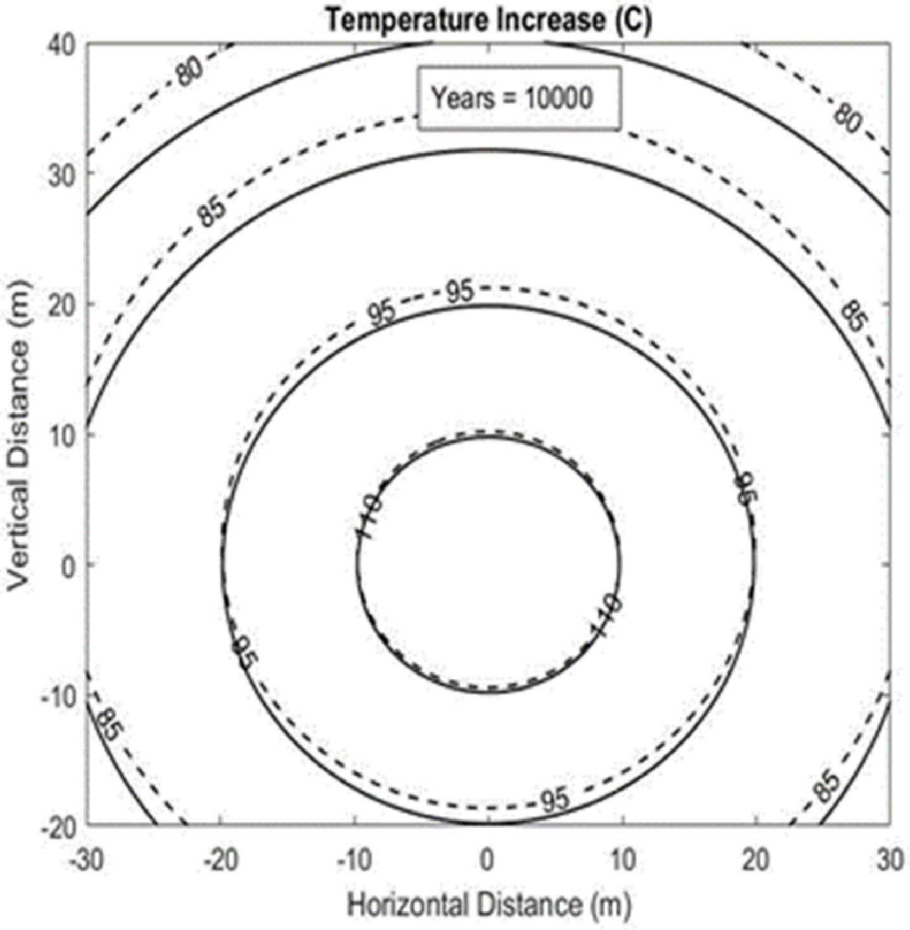
Temperature Change in Adjacent DPCs Separated by 20, 30, and 40 meters from the Central (Critical) DPC in Shale Host Rock

Effects of Convection – 10,000 Years

$k = 1 \times 10^{-15}\ m^2$



$k = 5 \times 10^{-15}\ m^2$



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

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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)

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")
- 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
- These oxides control actinide release, which tends 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)

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Engineered Barrier System Degradation

- In the hypothetical repositories assumed in this work, engineered barriers consist of
  - Waste package outer barrier
  - DPC
  - Fuel cladding
  - Backfill (crushed alluvium, 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 - 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

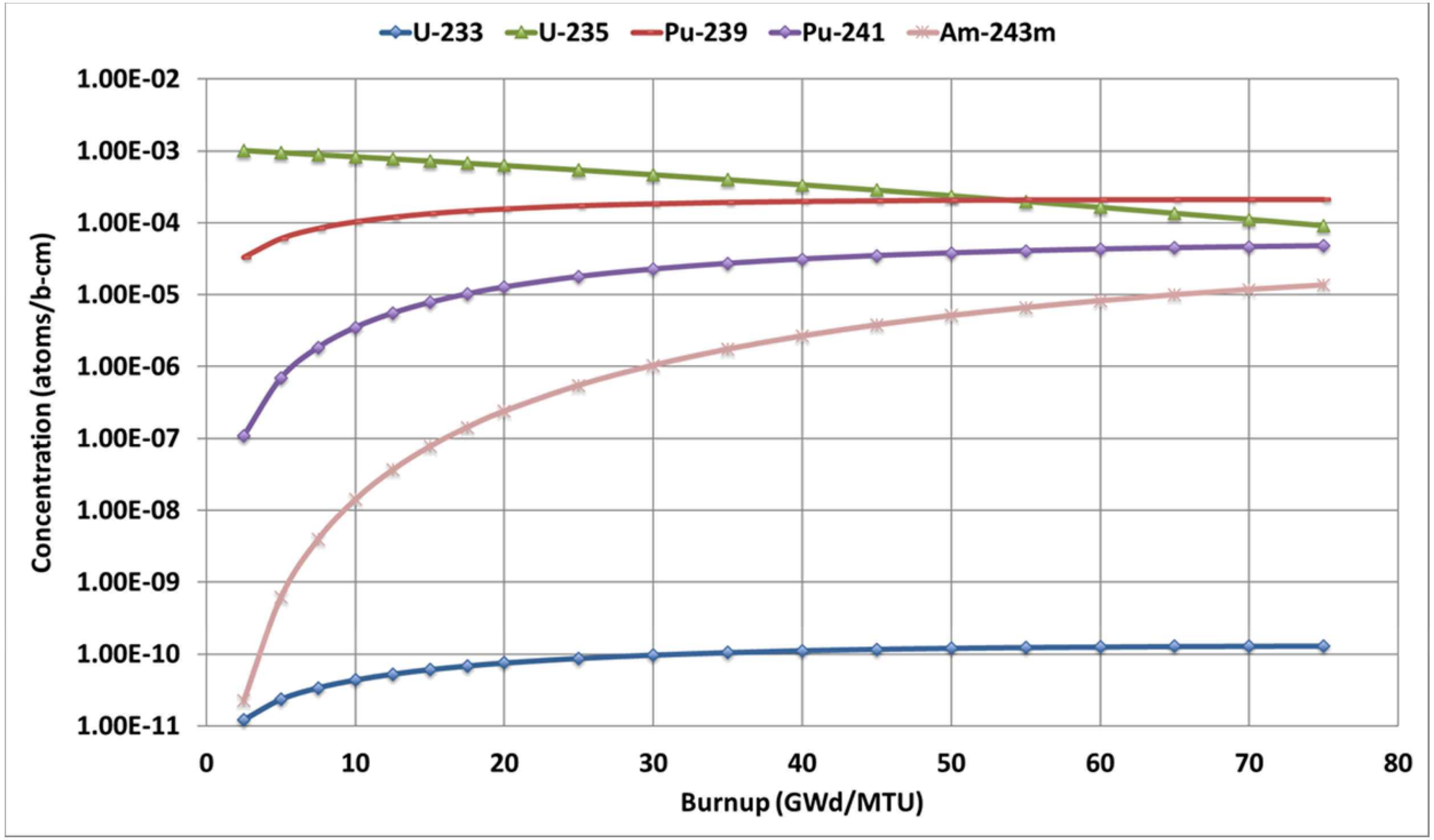
- Determined four parameters were worthy of further examination
  - Radioactive decay
  - Burnup
  - Irreversible geometry changes
  - Compositional changes due to corrosion or dissolution

Termination of Criticality - Conclusions

- 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 unsaturated repository, 400 W for 10,000 years results in additional ~0.1 GWd/MTU average burnup
  - 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

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

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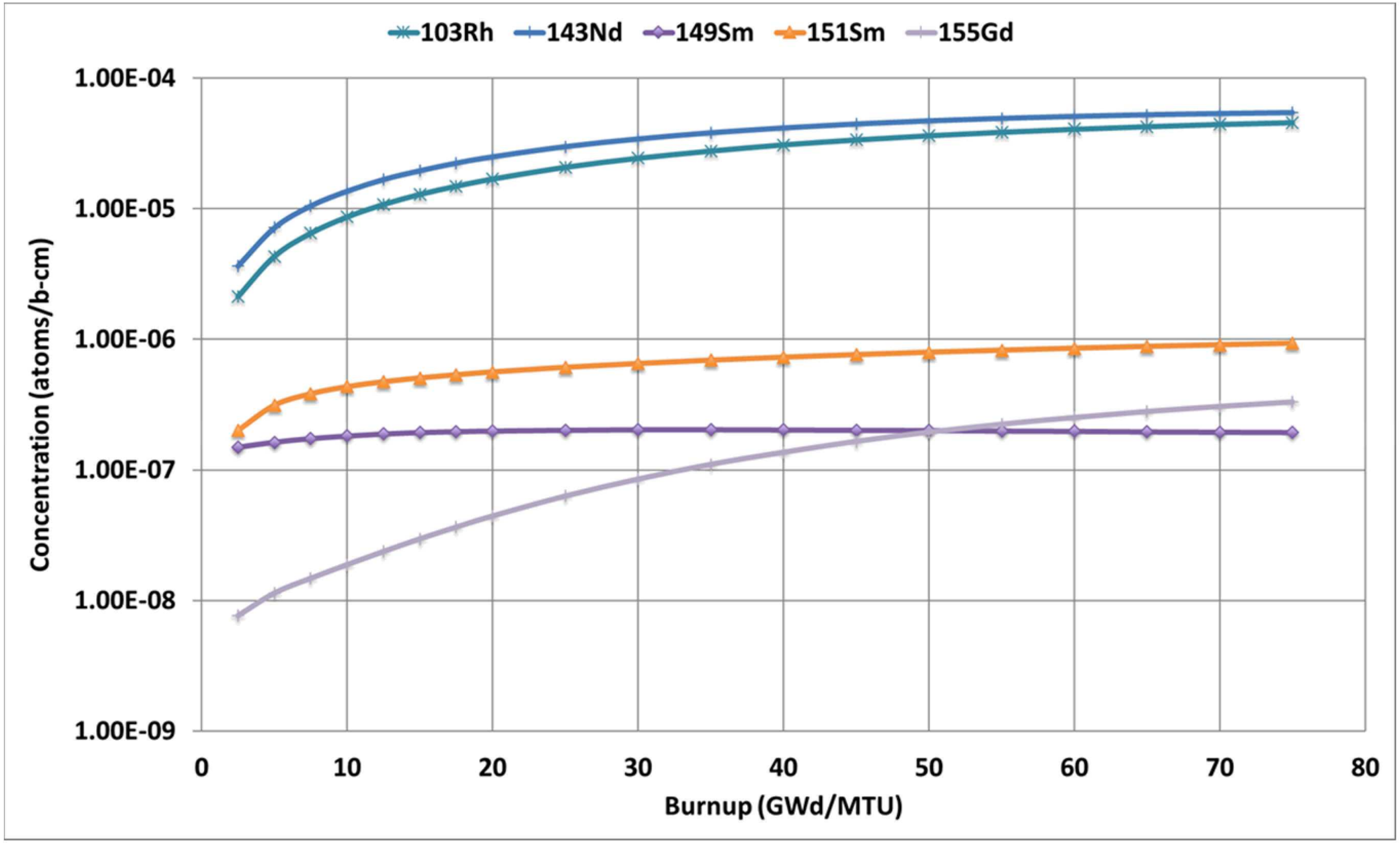
Reactivity Perturbations Due to Burnup (cont'd)

Fission product neutron absorber concentration continues to increase



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

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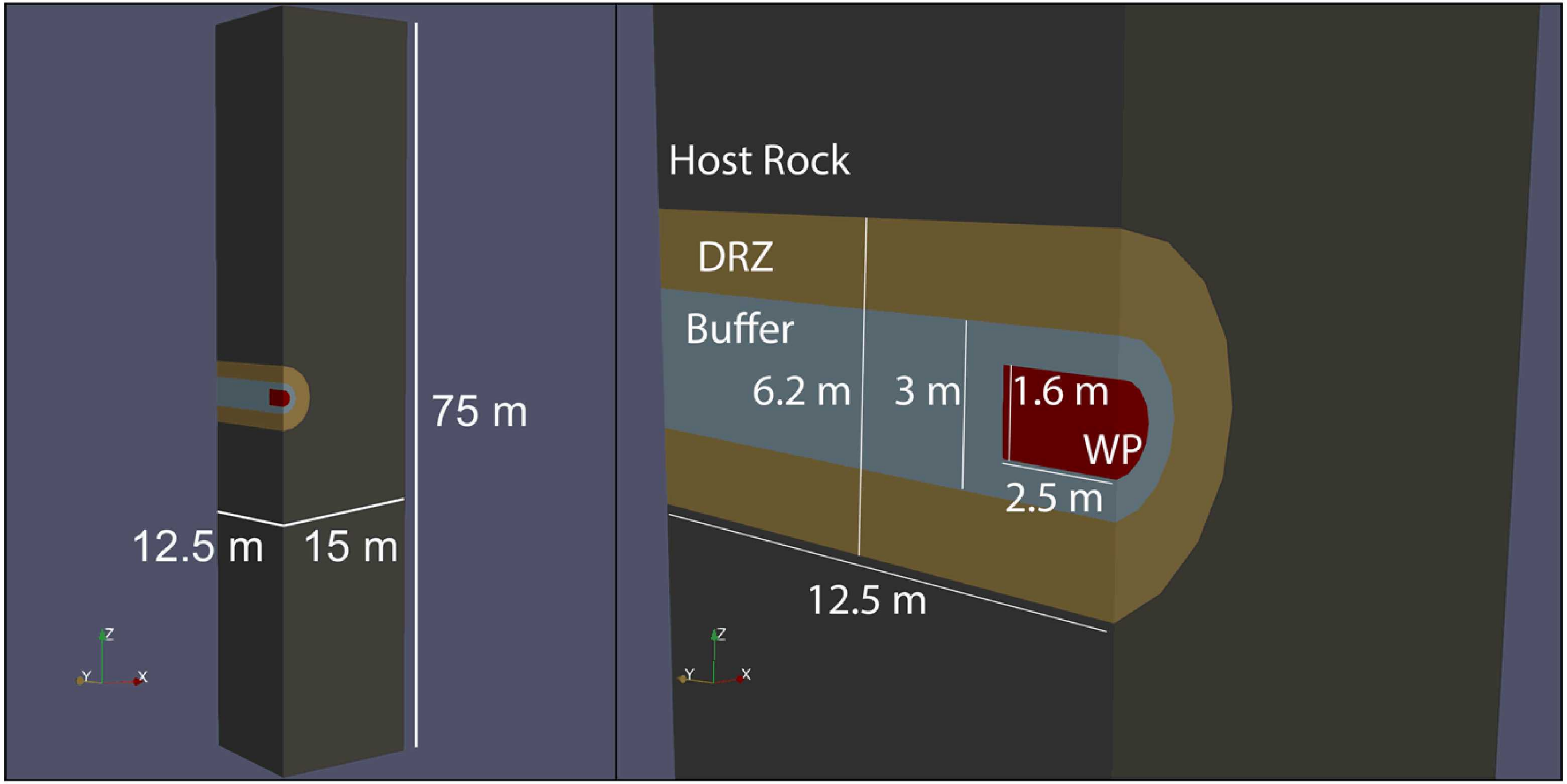
Termination of Criticality – Results (cont'd)

- Further study of corrosion of grid spacers and cladding is warranted
- 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

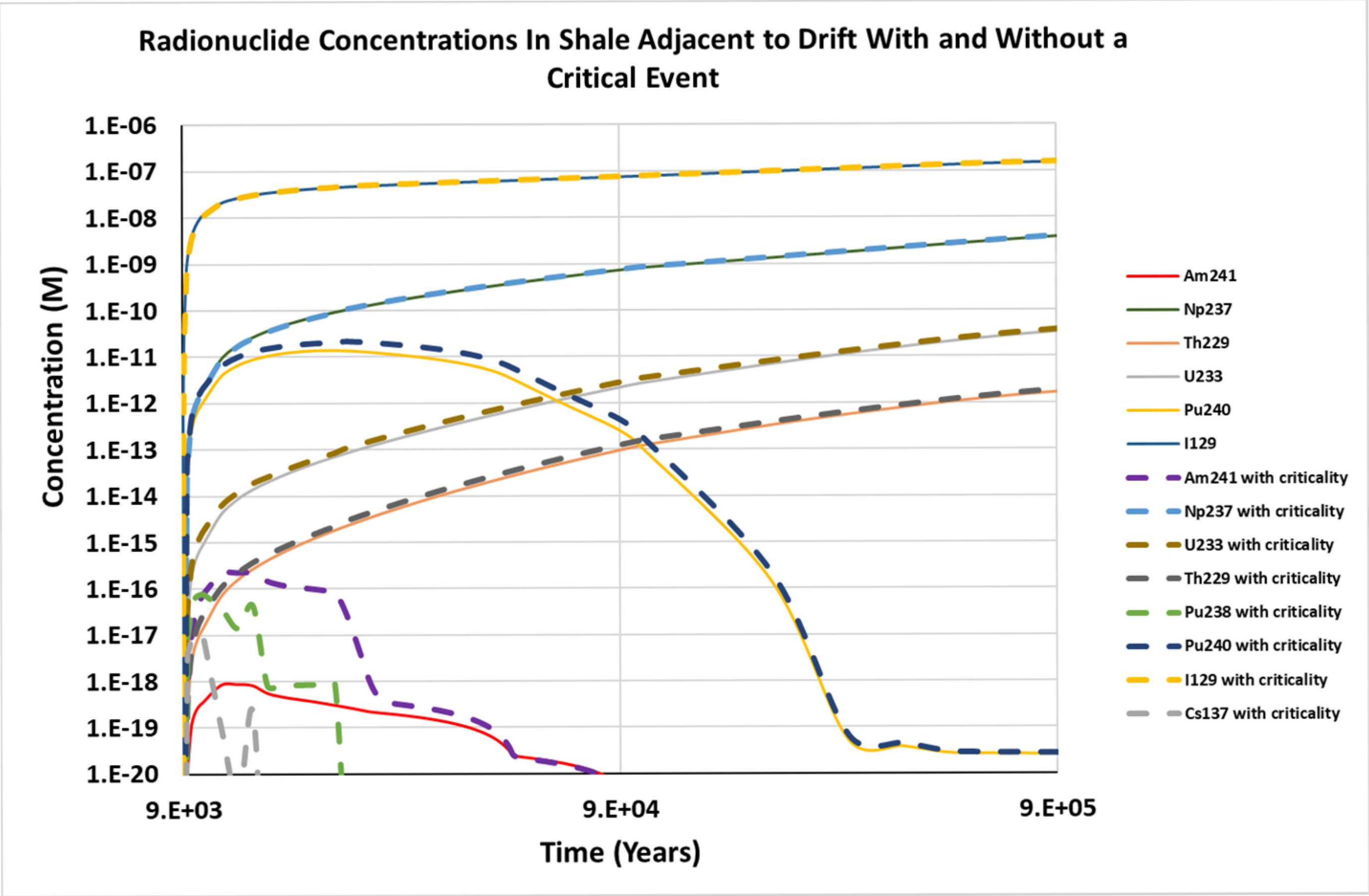
- Used PFLOTRAN
  - Massively parallel subsurface flow and reactive transport simulator
  - Simulated 3D multiphase flow and aqueous radionuclide transport
- Considered case without steady-state critical event and case with steady-state critical event
- Developed a criticality sub-module in PFLOTRAN
  - 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
- 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



Conclusions

- 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
- Insights into thermal processes
  - Temperatures of waste packages adjacent to waste package experiencing a steady-state critical event do not increase significantly (20 m center-to-center spacing)
  - In the low permeability backfill of the hypothetical repository, the effect of convection on heat transfer is negligible

Conclusions (cont'd)

- 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
- 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

Conclusions (cont'd)

- Insights into repository performance
  - Importance of newly generated radionuclides to dose is dependent on radionuclide travel time from repository to dose receptor
  - Concentration of ^{241}Am in the near field increases by about two orders of magnitude for roughly the duration of the critical event
  - Concentrations of ^{129}I and ^{237}Np in the near field increase about 3% in the long term
  - Concentrations of ^{229}Th and ^{233}U in the near field increase about 15% in the long term

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.

Backup Slides

EBS Degradation – Future Work

- Geometry of waste package after failure in saturated environment; external pressure ~20 MPa (hydrostatic + backfill swelling)
- Cladding degradation in the uncontrolled environment inside a disposed-of DPC and effects on criticality
- Behavior of bentonite backfill at pressures and temperatures typical of saturated repository with critical event