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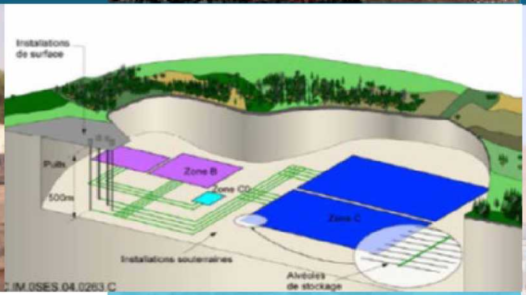
SAND2019-4107C

A Stochastic Model for Degradation Behavior of Tristructural-Isotropic Coated Particle Spent Fuels



PRESENTED BY

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International High-Level Radioactive Waste Management Conference

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U.S. DEPARTMENT OF ENERGY NNSA

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Outline

Introduction

- U.S. Program Wastes
- Tristructural Isotropic (TRISO) Particle Fuels

Considerations for TRISO Spent Fuels

- Features, Events, and Processes (FEP)
- Degradation Model Development
  - Coupled SiC Layer General Corrosion and Radionuclide Diffusion
  - Consideration of Localized Corrosion of SiC Layer
  - Stochastic Failure of TRISO Particles in Graphite Compact
  - Porous Media Diffusion through Graphite Compact

Summary and Conclusions

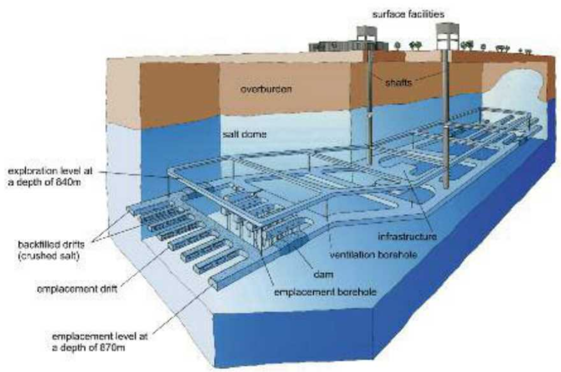
Spent Nuclear Fuel and High-Level Radioactive Waste Disposal: The Goal

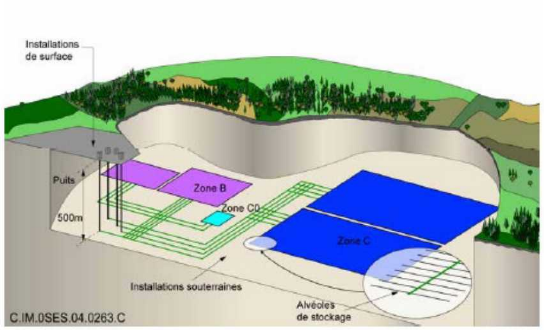
"There has been, for decades, a worldwide consensus in the nuclear technical community for disposal through geological isolation of high-level waste (HLW), including spent nuclear fuel (SNF)."

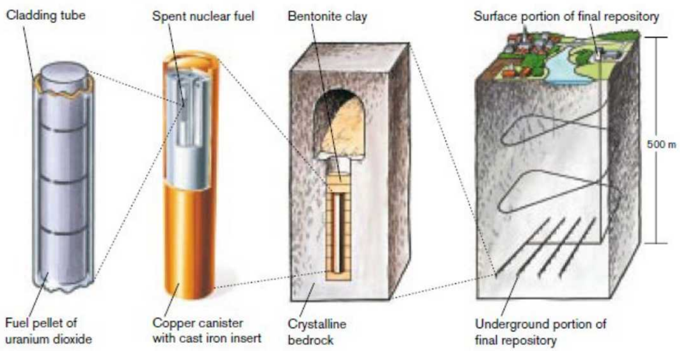
"Geological disposal remains the only long-term solution available."

National Research Council, 2001

Deep geologic disposal has been planned since the 1950s (SNL, 2014 provides recent analysis of disposal options)

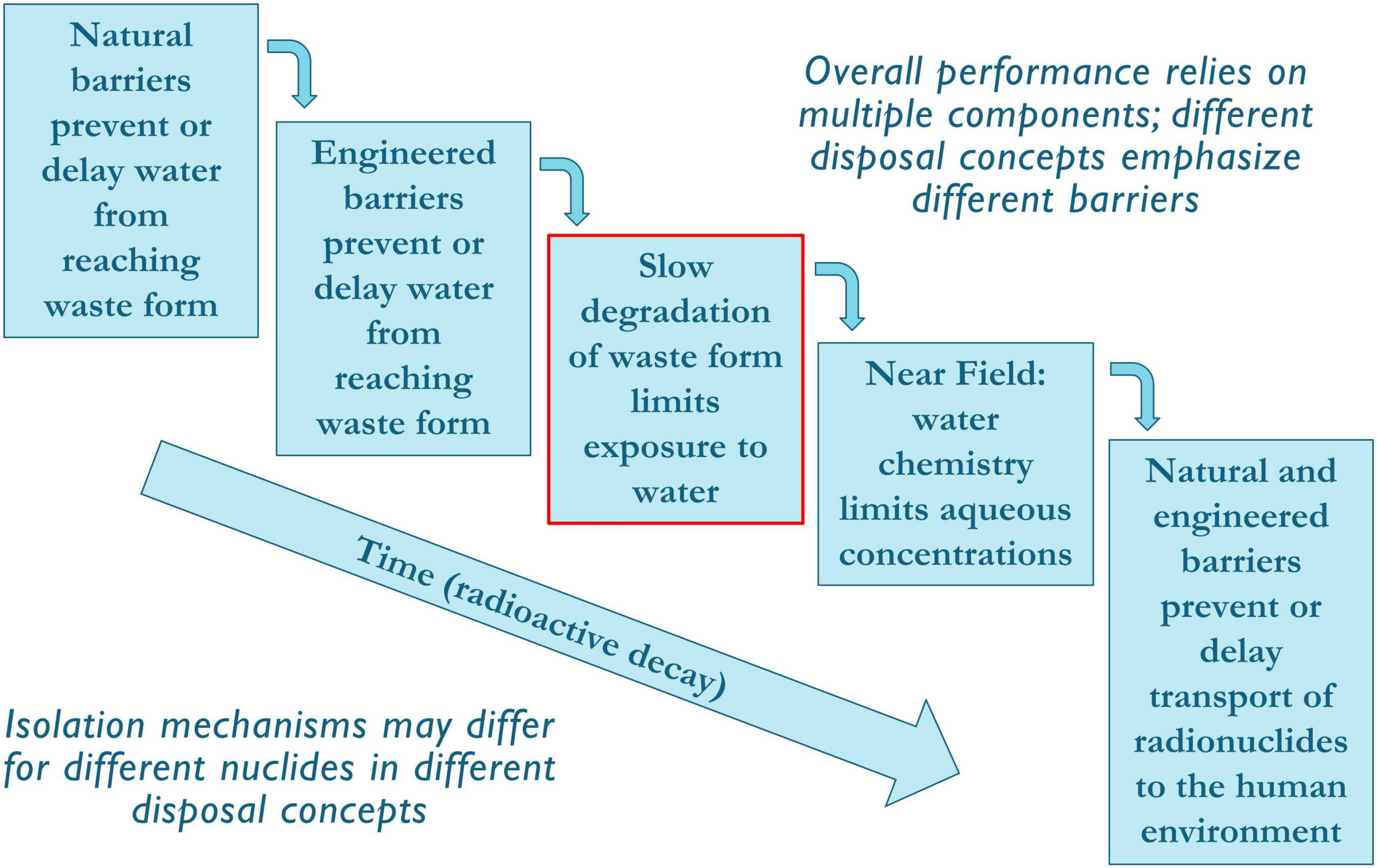






How Repositories Work

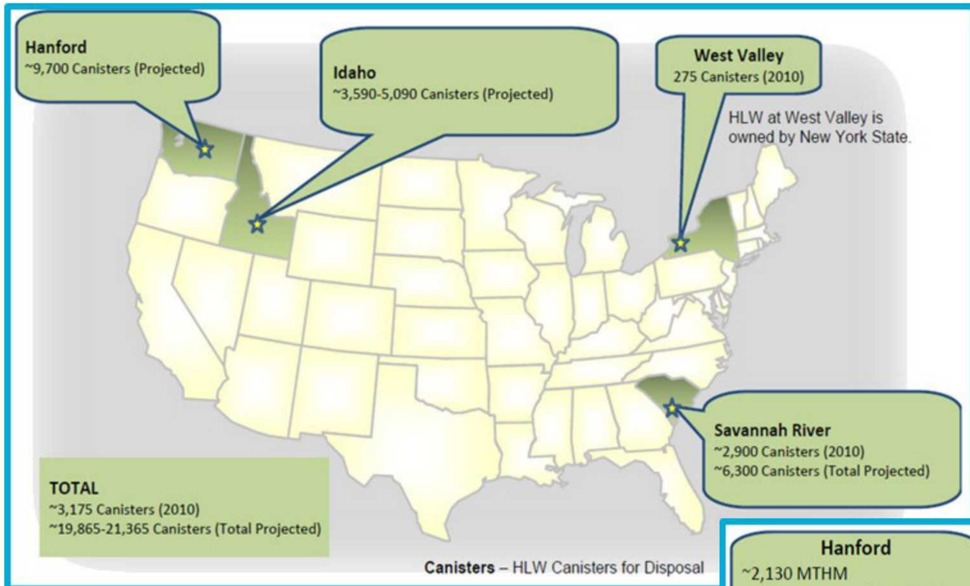
Overall performance relies on multiple components; different disposal concepts emphasize different barriers



Isolation mechanisms may differ for different nuclides in different disposal concepts

Geologic Disposal in the US: The Reality

DOE-managed SNF and HLW is in Temporary Storage at 5 Sites in 5 States



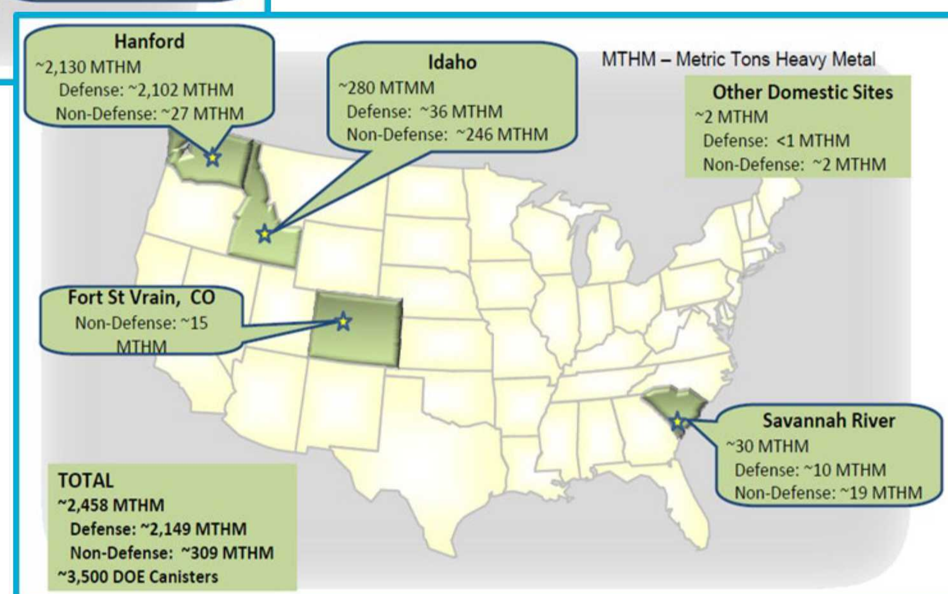
DOE-Managed HLW

~20,000 total canisters (projected)

DOE-Managed SNF

~2,458 Metric Tons

FSVR TRISO (~23.5 MTHM)



Source: Marcinowski, F., "Overview of DOE's Spent Nuclear Fuel and High-Level Waste," presentation to the Blue Ribbon Commission on America's Nuclear Future, March, 25, 2010, Washington, DC.

TRISO Particle Fuels Overview

(from van den Akker and Ahn, 2013)

Characteristics of TRISO fuel with a UO_X fuel kernel of radius 250 - 300 μm

Layer	Nominal Thickness (μm)	Purpose/function
porous pyrolytic carbon buffer	60 - 95	- allows kernel to swell - stops recoiling fission products from reaching SiC layer - provides void volume for gases
inner dense pyrolytic carbon (IPyC)	30 - 40	- barrier to gaseous fission products - slows down metallic fission product transport
Silicon Carbide (SiC)	25 - 35	- main fission product barrier - structural support to contain gas pressure
Outer dense pyrolytic carbon (OPyC)	40 - 45	- protects SiC layer from chemical and mechanical damage - adds to support to contain gas pressure

Sources: Minato et al., (1994); Moormann, et al., (2001); Nabielek et al., (2010); Fachinger (2006).

- Previous work by van den Akker and Ahn (2013) evaluated releases in repository setting
  - Relies mainly on graphite matrix chemical corrosion longevity (oxidation)
    - Fuel element graphite
    - Individual graphite compacts

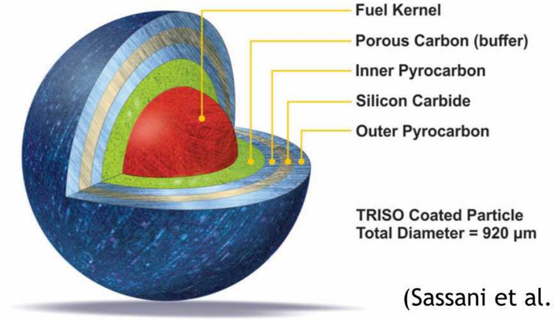
Degradation and Release Mechanisms for TRISO Particles

Corrosion of SiC and PyC Layers

- Using Fachinger et al. (2006) data for various fluids and temperatures

Models of van den Akker and Ahn (2013) to Assess SiC Layer Rupture

- Corrosion rates;
- Helium internal pressure buildup
- Statistical variability of SiC strength
- Possible protection via outer PyC

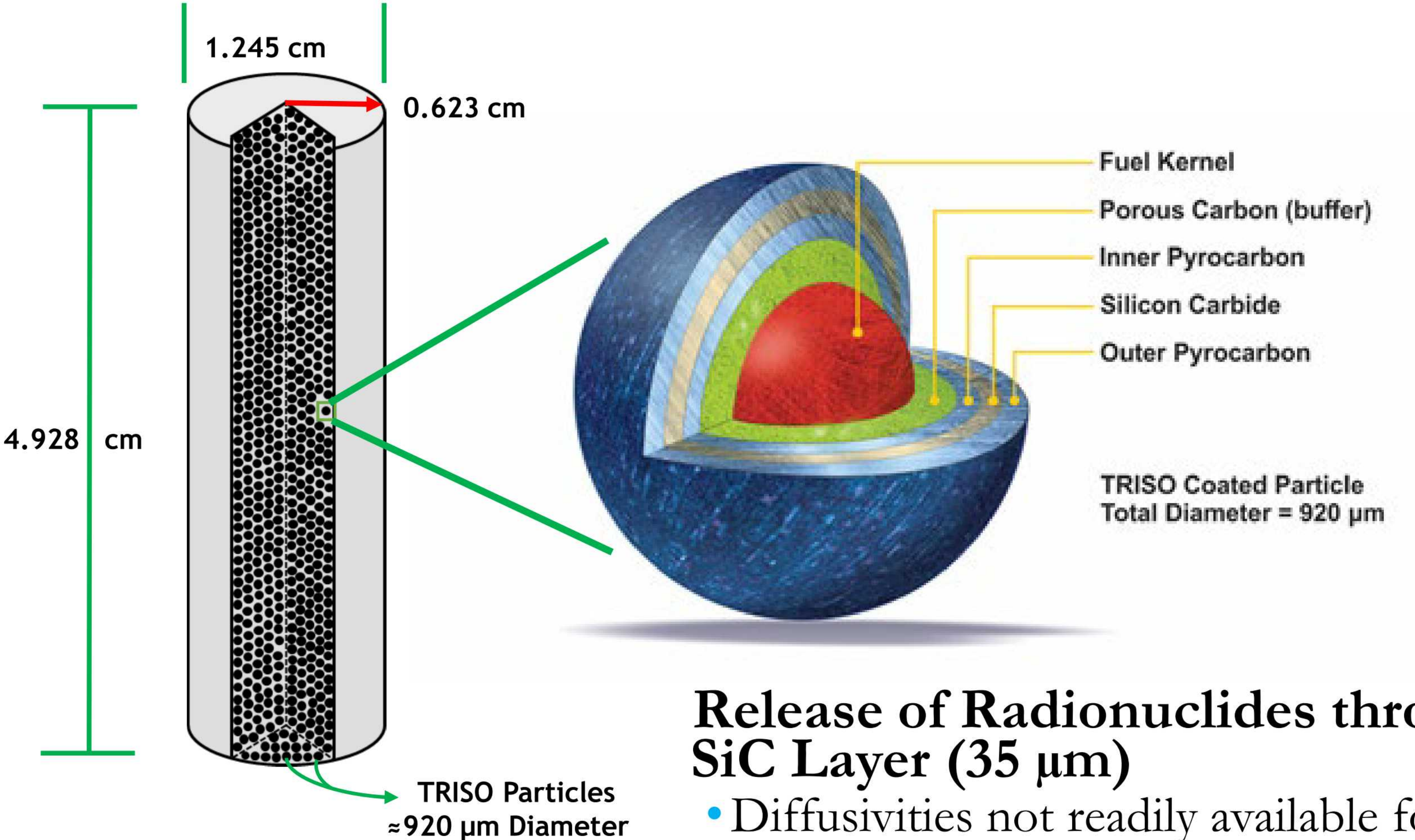


(Sassani et al., 2018)

Additional FEP Considerations

- Degradation mechanisms for graphite matrix (elements and compacts)
  - Seismic disruption: (compacts likely more durable than larger fuel elements)
  - Porosity/permeability evolution over time – advective pathways likely for element (channels)
- Diffusive release through graphite matrix
  - Likely fast compared to graphite corrosion lifetimes (10^6 – 10^8 yrs.)
  - Graphite compact likely diffusive pathway (advective potentially beyond that)
- Condition of particles (e.g., radionuclide distribution; Demkowicz et al., 2017)

Focus on TRISO Particles in Compacts



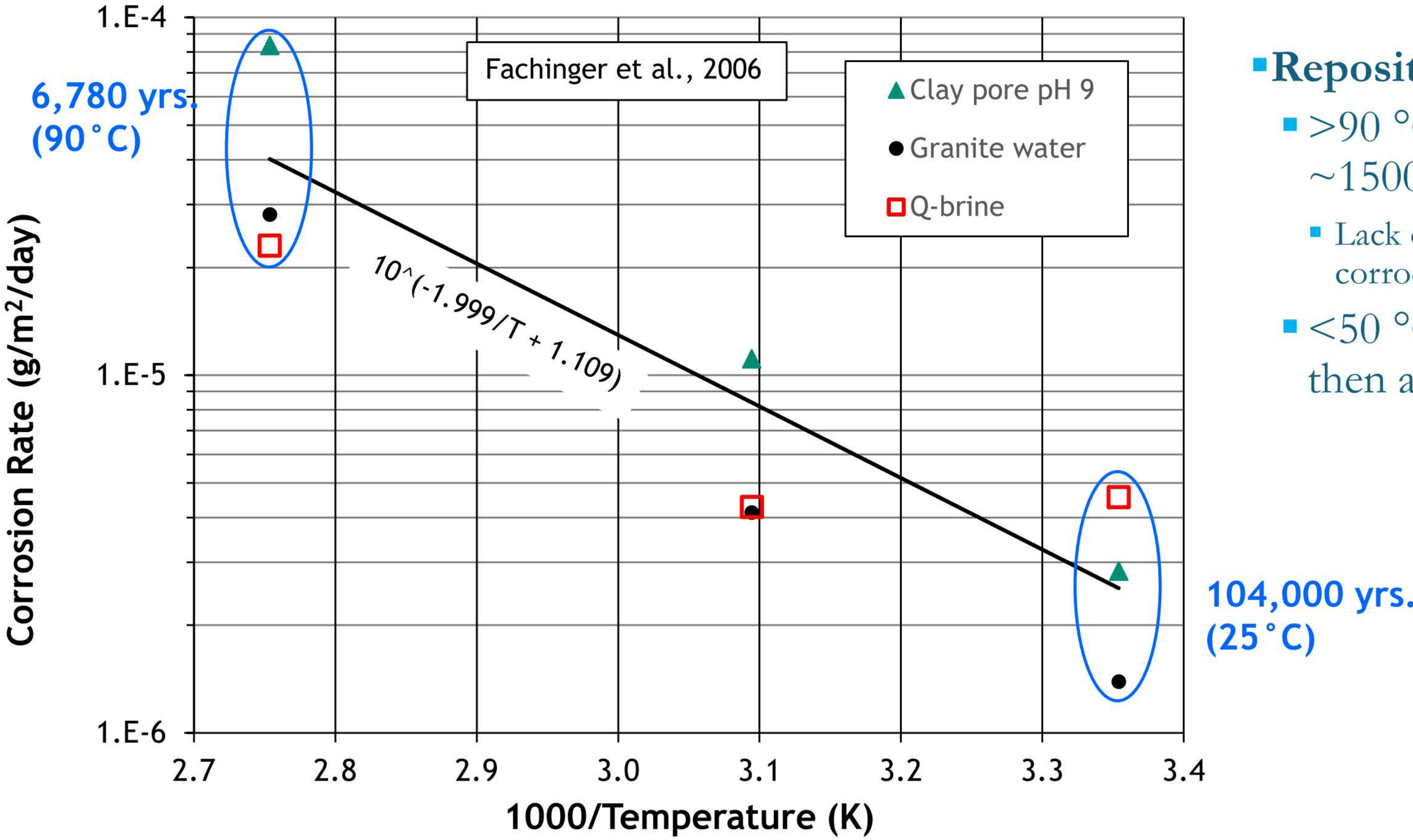
Graphite Compact

Release of Radionuclides through SiC Layer (35 µm)

- Diffusivities not readily available for SiC at repository conditions
- Sensitivity study of diffusion compared to SiC layer corrosion
  - Coupled diffusion through, and corrosion of, SiC layer
  - Assess relative importance at various conditions

SASSANI ET AL. IHLRWMC 2019

SiC Layer Corrosion Lifetime for Particle

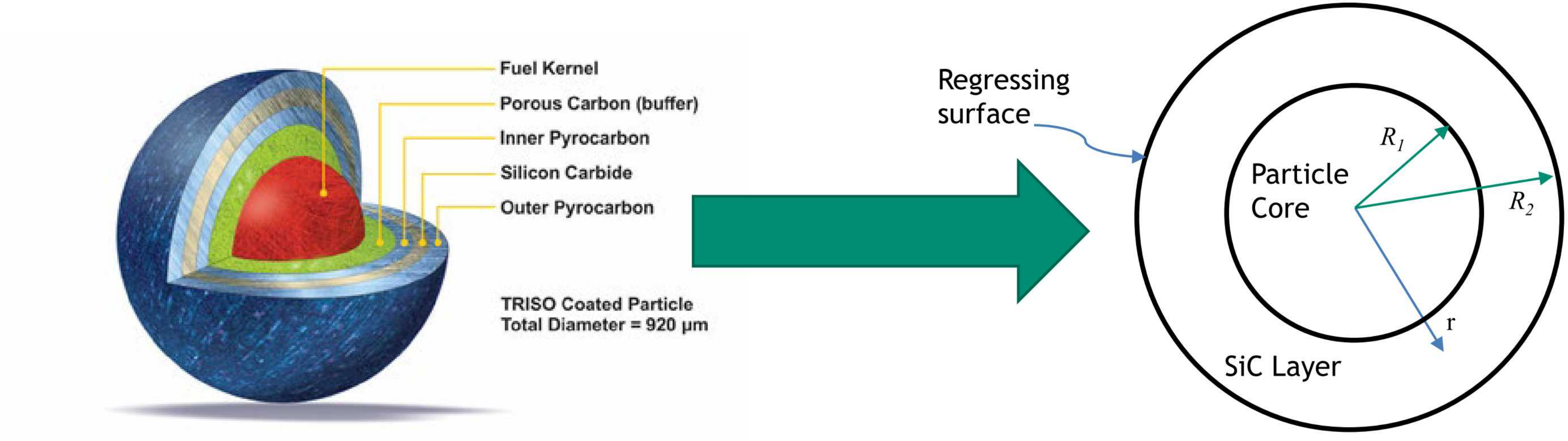


- **Repositories**:
  - >90 °C for 100's to ~1500 yrs
    - Lack of water in WP to corrode SiC
  - <50 °C ~10^3 to 10^4 yrs, then ambient

- SiC layer corrosion data for different groundwater indicate (assuming uniform/general corrosion):
  - At 90 °C, the 35 μm SiC layer would last ~7000 years (average at constant T)
  - At 25 °C, the SiC layer would last ~100,000 years (average at constant T)
- Estimated layer lifetime will depend on thermal evolution
  - Uncertainties: corrosion rate, thermal history, hydrologic condition (~10^4 to 10^5 years)
  - Slow corrosion of OPyC layer may protect and add to lifetime (~10^6 years; van den Akker and Ahn, 2013)

Simplifications for Simultaneous SiC Layer Corrosion and Radionuclide Diffusion

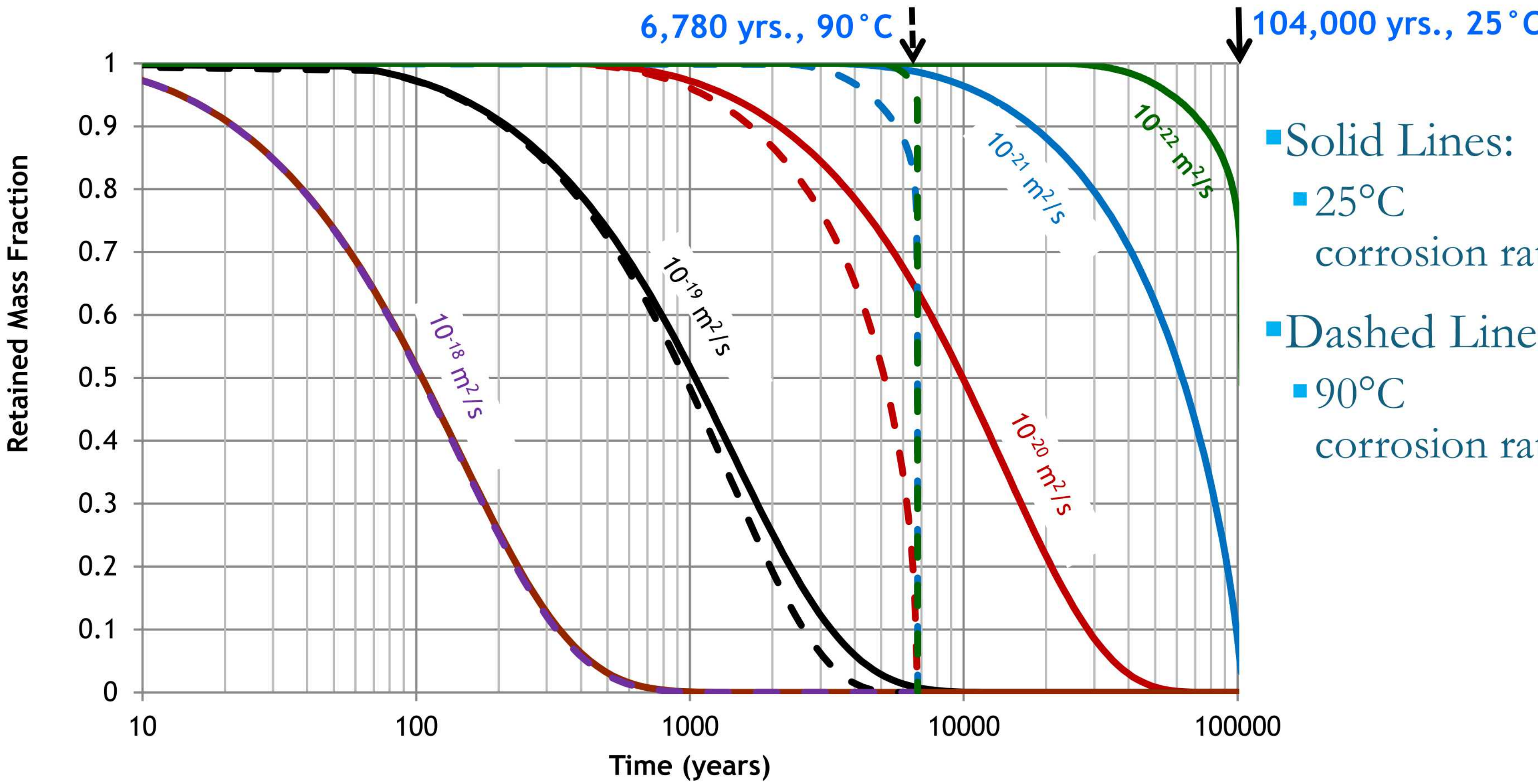
- Radionuclides distributed uniformly within particle core
  - Particle core = fuel kernel, and porous carbon and IPyC layers
    - Distribution of radionuclides during reactor operation
    - Diffusion likely faster within inner carbon layers than for denser SiC



Evaluate range of Diffusivities through SiC, while Layer Corrodes

- Compare releases for 25 °C and 90 °C corrosion rates
  - Coupled model analyzes retention of radionuclides within the regressing SiC surface (Gelbard and Sassani, 2018)

Simultaneous radionuclide diffusion through, and corrosion of, the SiC layer



- Solid Lines:
  - 25°C corrosion rate
- Dashed Lines
  - 90°C corrosion rates

- If diffusivity through SiC is about 10^{-18} m^2/s
  - Diffusive release would dominate over corrosion release
- If diffusivity through SiC is about 10^{-21} m^2/s
  - Diffusive release may only contribute at low T, with slow corrosion

Properties of SiC Layer – Considerations for Porous Medium (Aqueous) Diffusion

High Temperature Diffusivities in SiC for both Ag and Cs (Malherbe, 2013)

- Extrapolate to well below 10^{-21} m^2/s at repository temperatures (orders of magnitude lower)
  - Mechanisms not relevant to release through SiC in repository
    - Solid state and grain boundary diffusion processes very slow at lower T

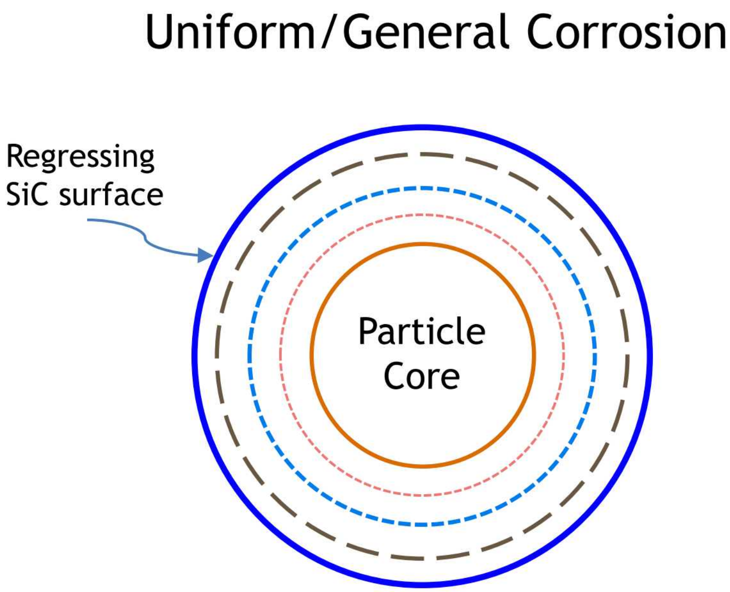
Porous Media Diffusion Processes much Faster

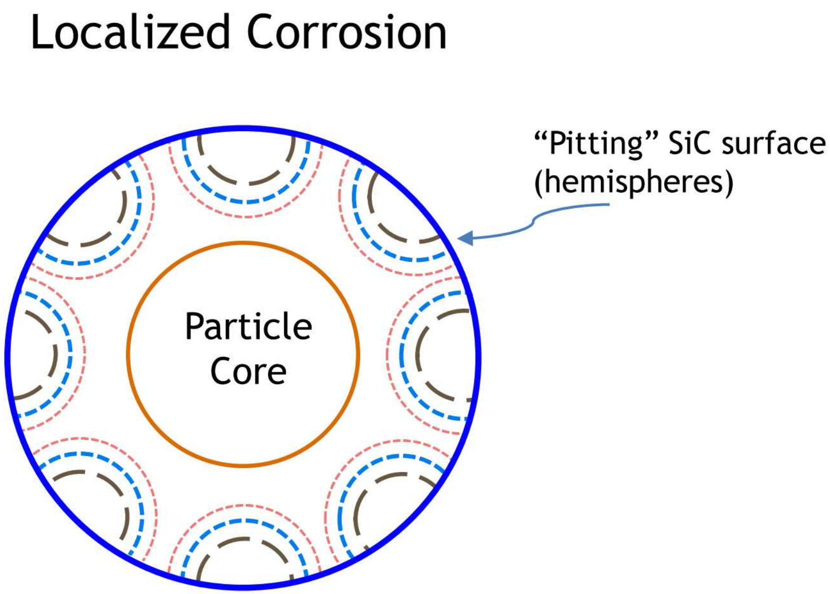
- Diffusivities of ^{36}Cl, ^{90}Sr, and ^{134}Cs through graphite in different brines (Fachinger et al., 2006)
  - Range from 1.2×10^{-13} m^2/s to 6.3×10^{-13} m^2/s
    - At least 10^8 times that of high-T SiC diffusion mechanisms values ($\leq 10^{-21}$ m^2/s)

Consider SiC Porosity as Porous Media (?)

- Porosity is small (~3-5%; Slavin and Quinn, 1986)
  - Pore structure of SiC does not appear to be connected porosity
  - Corrosion products may be porous media

Evaluation of SiC Localized Corrosion

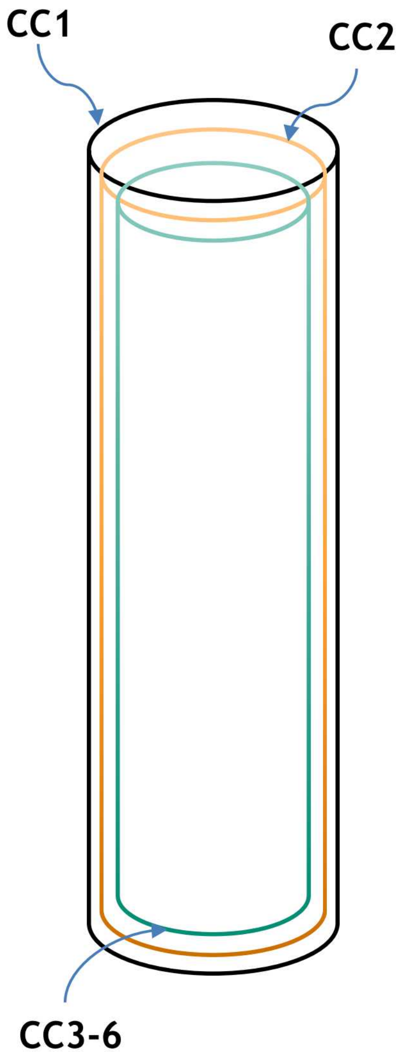


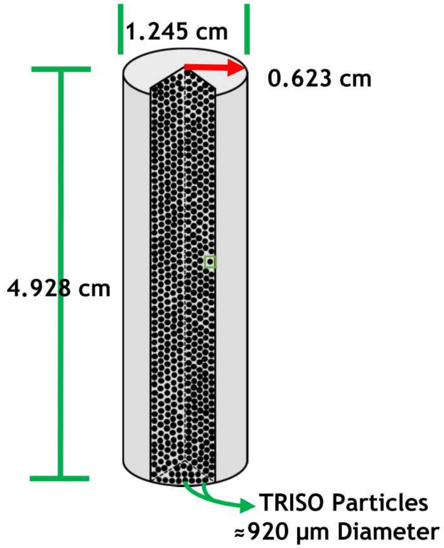


Geometric Considerations for Corrosion Data

- General corrosion: ~7000 to 100,000 years
  - If mechanism is localized, penetration may be faster
- Simple geometric scaling interpretation of measured rates
  - For hemispherical pitting: SiC lifetime ~4000 to 60,000 years
  - For conical pitting: SiC lifetime ~2000 to 30,000 years
- Formation of SiO_2 surface layer during corrosion may decrease corrosion rates (e.g., Hagan and Opila, 2015)

Failure of TRISO Particles Failures and Diffusion in Graphite Compacts





Failure of TRISO Particles (~5580)

- SiC layer failure times assigned randomly
  - General corrosion lifetime distributions
  - Localized corrosion lifetime distributions
- Uniform particle distribution in compact
  - Particle distributions for 6 concentric cylinders
  - CC1 – CC6 each ~0.104 cm thickness
  - ~33% particles in outermost CC1 (~59% in CC1 + CC2)
- Assume all radionuclides are at particle edge
  - Average diffusion path length ~0.06 to 0.58 cm (CC1 – CC6)
    - SiC + OPyC thickness negligible (~80 μm thick)
    - Could use graphite diffusivity for corroded SiC

Transport of Radionuclides through Compact Graphite Porous Medium

- Diffusivities from Fachinger et al. (2006)
  - 1.2×10^{-13} m^2/s to 6.3×10^{-13} m^2/s
- Assumed to be 1-D via average path (CC1 – CC6)
- Diffusion time frame is about years, at most

Diffusion through Graphite very Fast compared to Graphite Corrosion

- Can simplify representation of compact
- Radionuclide release effectively instant at SiC failure
  - Failure time distributions of particles

Summary and Conclusions

SiC Layer Corrosion Lifetimes

- About 100,000 to 7,000 years from SiC average
  - General corrosion depending on thermal history and fluid composition
  - If OPyC is protective, may be longer (~10^6 yr; van den Akker and Ahn, 2013)
- Shorter SiC penetration time if localized corrosion
  - About 60% to 30% of general corrosion lifetime
- Secondary phase (SiO_2) may provide protective layer to extend lifetime

Simultaneous SiC Corrosion and Radionuclide Diffusion

- Simplified moving boundary problem with diffusion
- Diffusion through SiC contributes to release for
  - Long corrosion lifetimes ~10^5 years with SiC diffusivity $\geq 10^{-21}$ m^2/s
  - SiC diffusivity ~10^{-18} m^2/s
  - Data for Ag and Cs for high-T diffusion mechanisms suggest those would be negligible at repository conditions
- SiC layer does not appear to be porous medium (small, non-connected porosity)

Porous Media Diffusion in Graphite Compact (& Pyrolitic Carbon)

- Higher diffusivities result in short release time (years at most)
- Release from compact would be driven by SiC layer failure times
  - Compact could be represented with a simple model of particle failure distributions
- The SiC layer appears to be the primary barrier to radionuclide release – further refinement of corrosion behavior and potential protection mechanisms

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