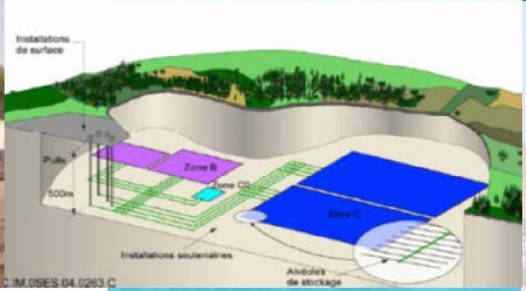
Sandia National Laboratories

SAND2018-8909C

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



PRESENTED BY

David Sassani, Distinguished Member Tech Staff

Fred Gelbard, Principal Member Technical Staff

Goldschmidt 2018

August 15, 2018, Boston, MA

U.S. DEPARTMENT OF ENERGY NNSA

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & 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.
SAND2018-XXXX PE

Outline

- Introduction
  - U.S. Program Wastes
  - Tristructural Isotropic (TRISO) Particle Fuels
- Considerations for TRISO Spent Fuels
  - Features, Events, and Processes (FEP)
  - Coupled SiC Layer Corrosion and Radionuclide Diffusion (coauthor Fred Gelbard, SNL)
- Summary and Conclusions

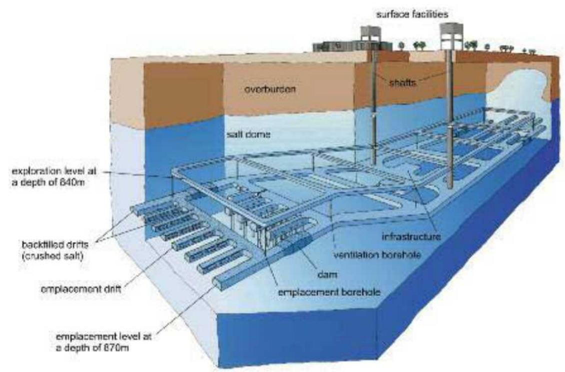
Spent Nuclear Fuel and High-Level Radioactive Waste Disposal: The Goal

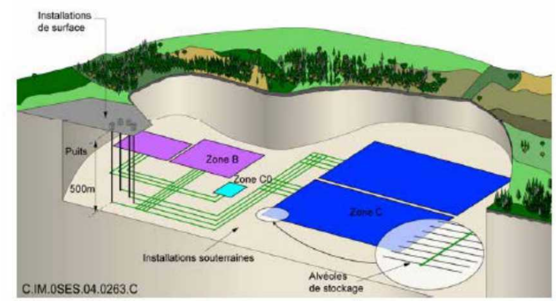
"There has been, for decades, a worldwide consensus in the nuclear technical community for disposal through geological isolation of high-level waste (HLW), including spent nuclear fuel (SNF)."

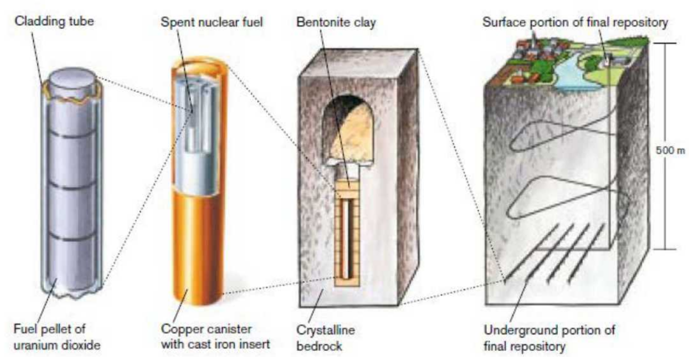
"Geological disposal remains the only long-term solution available."

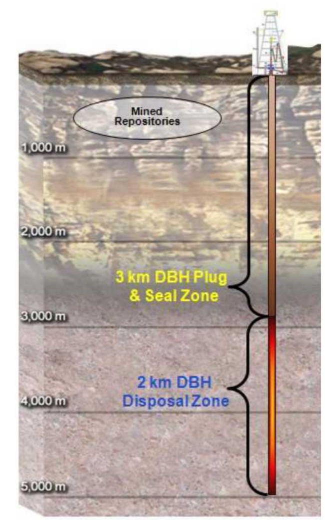
National Research Council, 2001

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









How Repositories Work

Natural barriers prevent or delay water from reaching waste form

Engineered barriers prevent or delay water from reaching waste form

Slow degradation of waste form limits exposure to water

Near Field: water chemistry limits aqueous concentrations

Natural and engineered barriers prevent or delay transport of radionuclides to the human environment

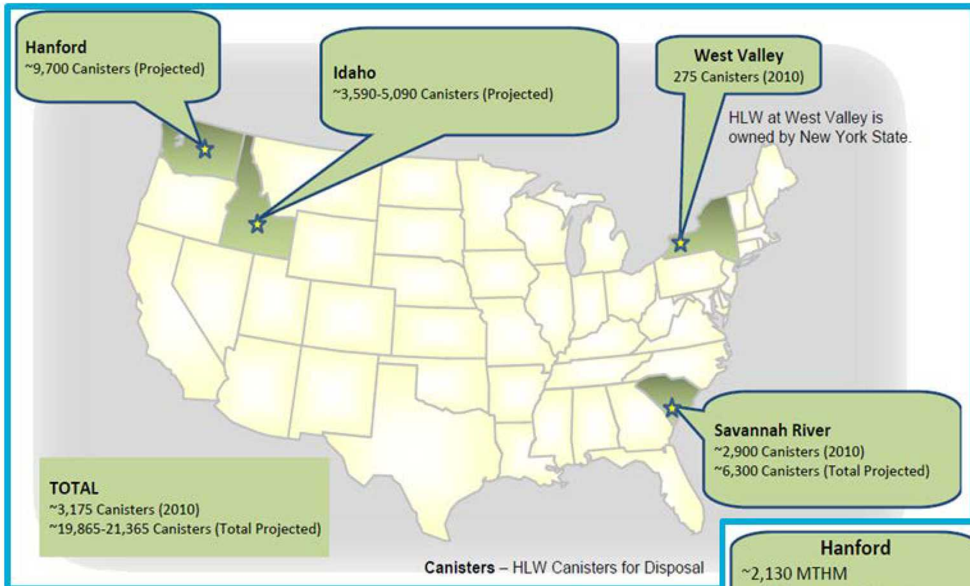
Time (radioactive decay)

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



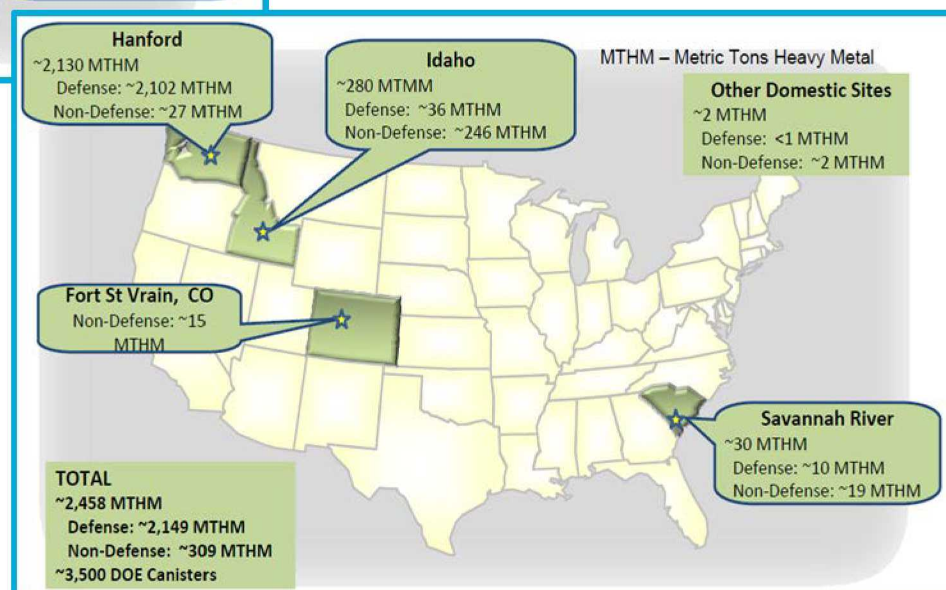
DOE-Managed HLW

~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 core of radius 250 - 300 μm

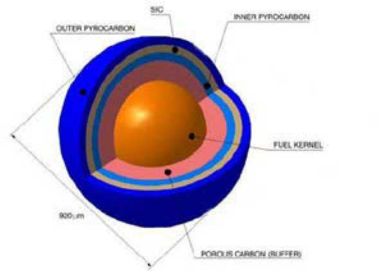
Layer	Nominal Thickness (μm)	Purpose/function
porous pyrolytic carbon buffer	60 - 95	- allows kernel to swell - stops recoiling fission products from reaching SiC layer - provides void volume for gases
inner dense pyrolytic carbon (IPyC)	30 - 40	- barrier to gaseous fission products - slows down metallic 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 longevity (oxidation)
    - Fuel element graphite
    - Individual graphite compacts

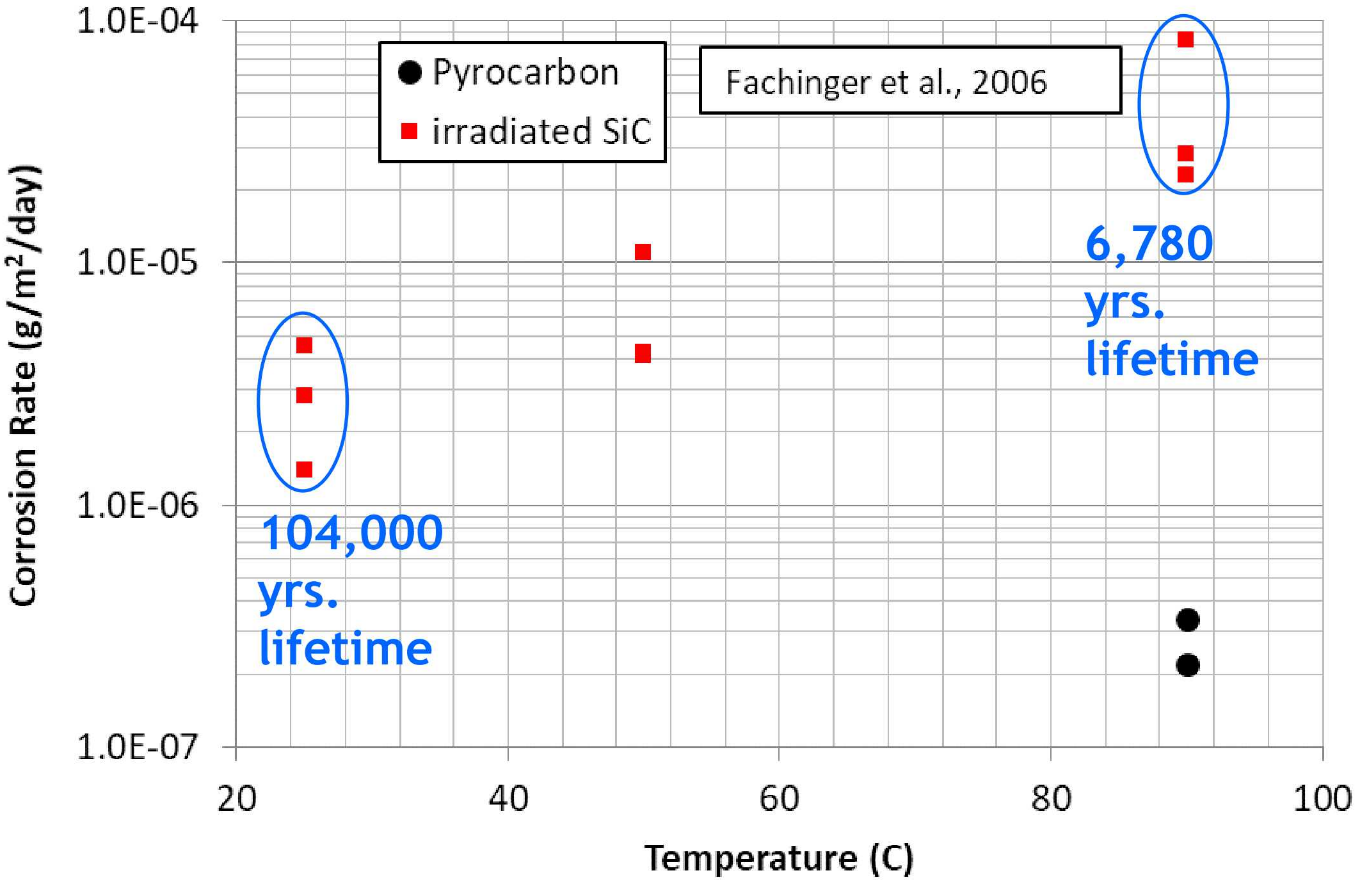
Degradation and Release Mechanisms for TRISO Particles

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



- Additional FEP Considerations
  - Degradation mechanisms for graphite matrix (elements and compacts)
    - Seismic disruption: (compacts likely more durable than elements)
      - Porosity/permeability evolution over time – advective pathways
  - Diffusive release through graphite matrix
    - Compare to lifetimes 10^6 – 10^8 yrs
  - Condition of particles (e.g., location of radionuclides; Demkowicz et al., 2017)
- Diffusion of radionuclides through particles (Gelbard, 2002)
  - Diffusivities not readily available for SiC
  - Sensitivity study of diffusion compared to SiC layer corrosion
  - Coupled diffusion and corrosion of SiC layer
  - Kinetic models used to estimate/assess magnitude of diffusivities

SiC layer corrosion lifetimes for TRISO fuel



- **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

- SiC layer corrosion data for different brines indicate:
  - At 90 °C the 35 μm layer would last ~7000 years (average at constant T)
  - At 25 °C the would layer 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)
  - Pyrolitc carbon layer protection may add to lifetime (~10^6 years; van den Akker and Ahn, 2013)

Example radionuclides of interest for geological repositories

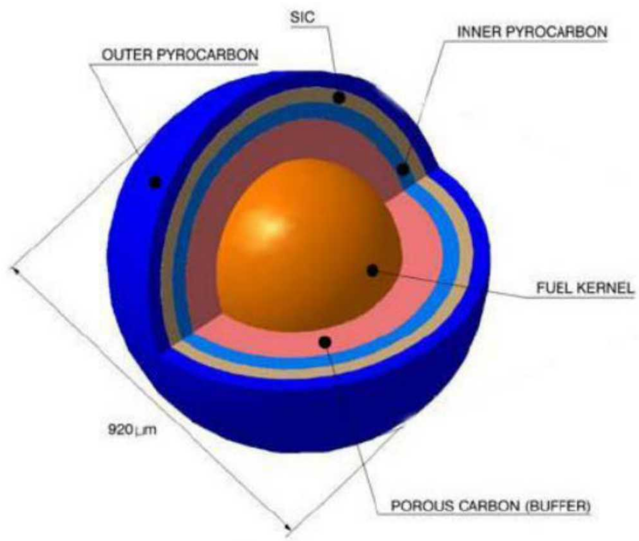
Radionuclide	Half-life (years)	Decay Product
^{129}I	1.57×10^7	^{129}Xe
^{36}Cl	3.01×10^5	^{36}Ar
^{226}Ra	1.60×10^3	$^{222}Rn \rightarrow {}^{218}Po \rightarrow {}^{214}Pb \rightarrow {}^{214}Bi \rightarrow {}^{214}Po \rightarrow {}^{210}Pb \rightarrow {}^{210}Bi \rightarrow {}^{210}Po \rightarrow {}^{206}Pb$
^{79}Se	2.95×10^5	^{79}Br
^{99}Tc	2.13×10^5	^{99}Ru

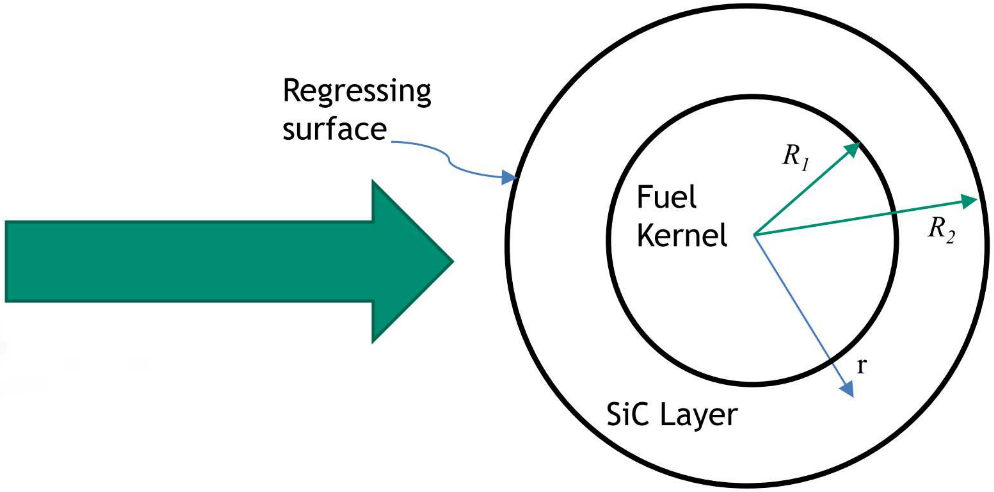
Actinides

- Data for diffusivities through SiC
  - Not readily available in general, and specifically at repository temperatures
- Conduct sensitivity analyses for coupled radionuclide diffusion and SiC layer corrosion to identify threshold diffusivities

Simplifications for simultaneous SiC layer corrosion and radionuclide diffusion

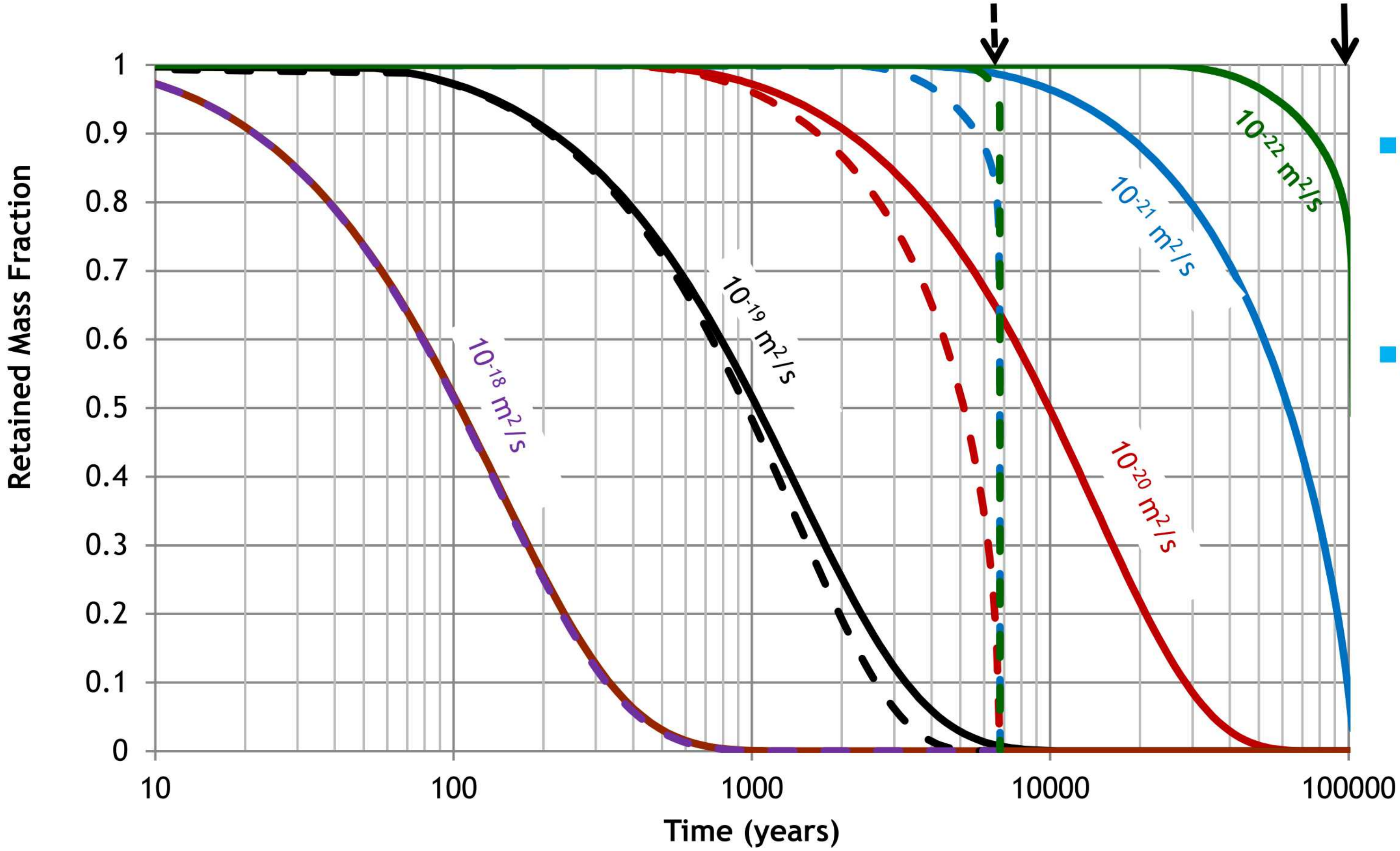
- Diffusivities of ^{36}Cl, ^{90}Sr, and ^{134}Cs through graphite in different brines range from 1.2×10^{-13} m^2/s to 6.3×10^{-13} m^2/s (Fachinger et al., 2006)
  - Likely faster than for denser SiC, so assume instant transport for carbon layers





- Evaluate range of diffusivities through SiC, while layer corrodes
  - Compare releases for 25 °C and 90 °C corrosion rates.

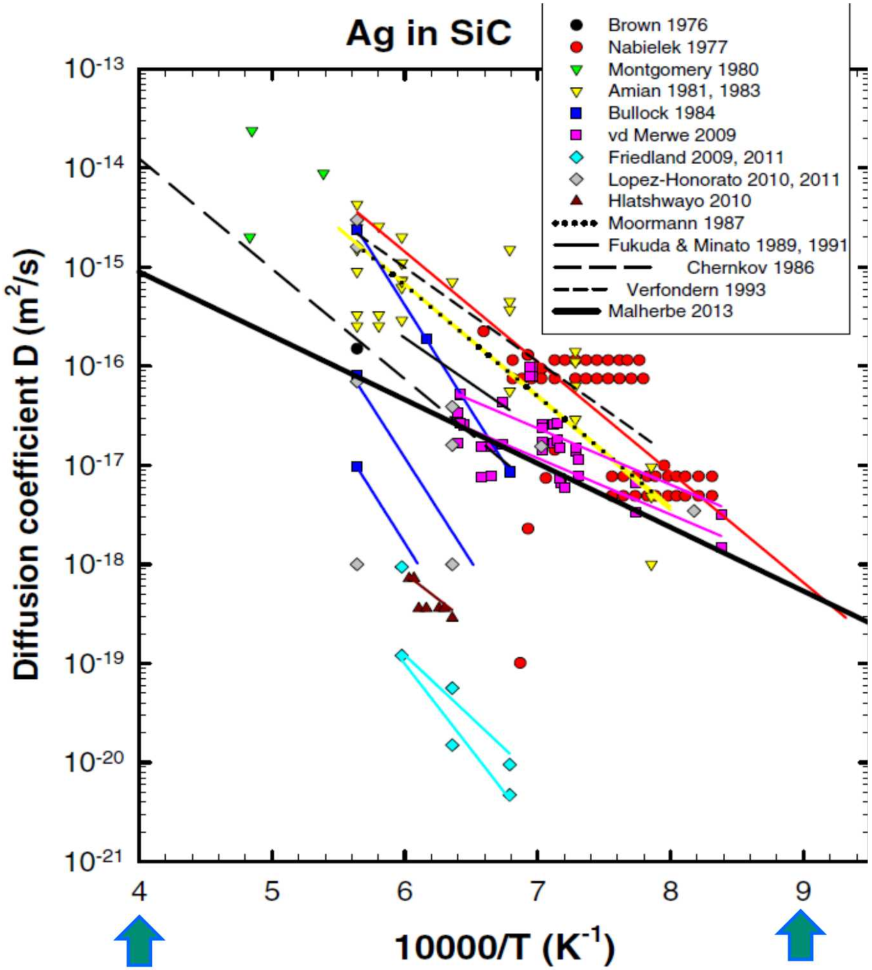
Simultaneous radionuclide diffusion through, and corrosion of, the SiC layer

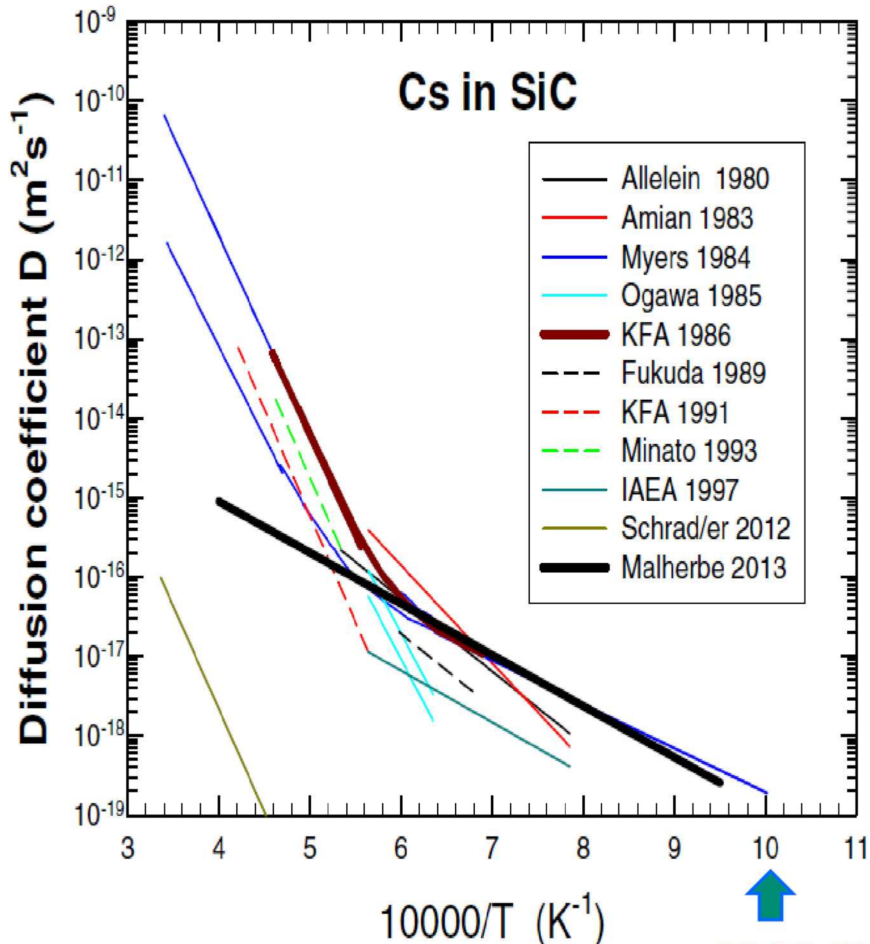


- Solid Lines:
  - 25°C corrosion rate
- Dashed Lines
  - 90°C corrosion rates

- If diffusivity through SiC is 10^{-18} m^2/s => diffusive release dominates
- If diffusivity through SiC is 10^{-21} m^2/s => diffusive release contributes only for lower T's
  - This is 10^{-8} times than that for the carbon/graphite materials (~10^{-13} m^2/s)

Diffusivity data analyzed by Malherbe (2013) are at higher temperatures than disposal conditions





2500 K **1111 K** **1000 K**

- Extrapolate to T where D=>10^{-21} m^2/s
  - Diffusivities for both Ag and Cs hit this at ~750 K (~480 °C)

Summary and Conclusions

- SiC Layer Lifetimes
  - About 100,000 to 7,000 years from SiC average corrosion rates
    - Fluid composition and thermal history
    - If OPyC is protective, may be longer (~10^6 yr; van den Akker and Ahn, 2013)
- Simultaneous SiC Corrosion and Radionuclide Diffusion
  - Simplified moving boundary problem with diffusion
  - If diffusivity through SiC is 10^{-21} m^2/s
    - Diffusive release contributes to release only for lower temperatures
    - High T data for Ag and Cs suggest values would be well below this value
    - Diffusion in graphite/pyrolitic carbon may be much higher
- Next Steps
  - Develop model further, coupling stochastics
    - Added layers, including diffusion through compact graphite matrix
    - Evolving temperatures for repository
  - Assess major uncertainties
    - Diffusivities, especially in SiC
    - Mechanical behavior/evolution of graphite matrix (elements and compacts)

References

- ANDRA (Agence nationale pour la gestion des déchets radioactifs), 2005. Dossier 2005: Argile. Tome: Safety Evaluation of a Geological Repository (English translation: original documentation written in French remains ultimately the reference documentation).
- Bonano, E., Kalinina, E., and Swift, P., 2018, The Need for Integrating the Back End of the Nuclear Fuel Cycle in the United States of America. MRS Advances, 1-13. doi:10.1557/adv.2018.231
- Demkowicz, P., Hunn, J., Petti, D., and Morris, R., 2017. Key results from irradiation and post-irradiation examination of AGR-1 UCO, Nucl. Eng. and Design, (j.nucengdes.2017.09.005).
- Fachinger, J., M. den Exter, B. Grambow, S. Holgersson, C. Landeman, M. Titov, and T. Podruhzina, 2006. Behavior of spent HTR fuel elements in aquatic phases of repository host rock formation, Nuclear Engineering and Design 236, 543-554, 2006.
- Faybishenko, B., Birkholzer, J., Sassani, D., and Swift, P., 2016. International Approaches for Deep Geological Disposal of Nuclear Waste: Geological Challenges in Radioactive Waste Isolation, Fifth Worldwide Review, LBNL-1006984, Lawrence Berkeley National Laboratory.
- Gelbard, F., 2002. Analytical Modeling of Fission Product Releases by Diffusion from Multicoated Fuel Particles, SAND2002-3966, Sandia National Laboratories, Albuquerque, New Mexico, 2003
- Maheras, S.J., R.E. Best, S.B. Ross, K.A. Buxton, J.L. England, P.E. McConnell, L.M. Massaro, P.J. Jensen, 2017, Preliminary Evaluation of Removing Used Nuclear Fuel from Shutdown Sites (U.S. Department of Energy Nuclear Fuels Storage and Transportation Planning Project SFWD-IWM-2017-000024, PNNL-22676 Rev. 10, 2017) 452 p.
- Malherbe, J. B., 2013. Diffusion of fission products and radiation damage in SiC, Journal of Physics D: Applied Physics 46 (47) 473001, 2013.
- NAGRA (Nationale Genossenschaft für die Lagerung Radioactiver Abfälle [National Cooperative for the Disposal of Radioactive Waste]), 2002, *Project Opalinus Clay Safety Report: Demonstration of disposal feasibility for spent fuel, vitrified high-level waste and long-lived intermediate-level waste (Entsorgungsnachweis),* Technical Report 02-05.
- National Research Council / National Academies, 2001. Disposition of High-Level Waste and Spent Nuclear Fuel: The Continuing Societal and Technical Challenges, Washington, DC, National Academy Press.
- SNL (Sandia National Laboratories) 2014. Evaluation of Options for Permanent Geologic Disposal of Used Nuclear Fuel and High-Level Radioactive Waste Inventory in Support of a Comprehensive National Nuclear Fuel Cycle Strategy. FCRD-UFD-2013-000371. SAND2014-0187P; SAND2014-0189P. Revision 1. Albuquerque, New Mexico: Sandia National Laboratories.
- SKB (Svensk Kämbränslehantering AB [Swedish Nuclear Fuel and Waste Management Co.]), 2011. Long-Term Safety for the Final Repository for Spent Nuclear Fuel at Forsmark: Main Report of the SR-Site Project, Technical Report TR-11-01.
- Swift, P.N., and Bonano, E.J., 2016, “”Geological Disposal of Nuclear Waste in Tuff,” Elements vol. 12, pp. 263-268.
- US DOE (United States Department of Energy) 2008. Yucca Mountain Repository License Application, DOE/RW-0573, Rev. 1.
- Van den Akker, B. P. and J. Ahn, 2013. Performance assessment for geological disposal of graphite waste containing TRISO particles, Nuclear Technology 181 (3) 408-426, 2013.
- Wagner, J.C., J.L. Peterson, D.E. Mueller, J.C. Gehin, A. Worrall, T. Taiwo, M. Nutt, M.A. Williamson, M. Todosow, R. Wigeland, W. Halsey, R. Omberg, P.N. Swift, and J.T. Carter, 2012, Categorization of Used Nuclear Fuel Inventory in Support of a Comprehensive National Nuclear Fuel Cycle Strategy, ORNL/TM-2012/308, FCRD-FCT-2012-000232, Oak Ridge National Laboratory, Oak Ridge, TN.