Migration of fission products in graphitic materials used in high-temperature gas-cooled reactors

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Research Reactor
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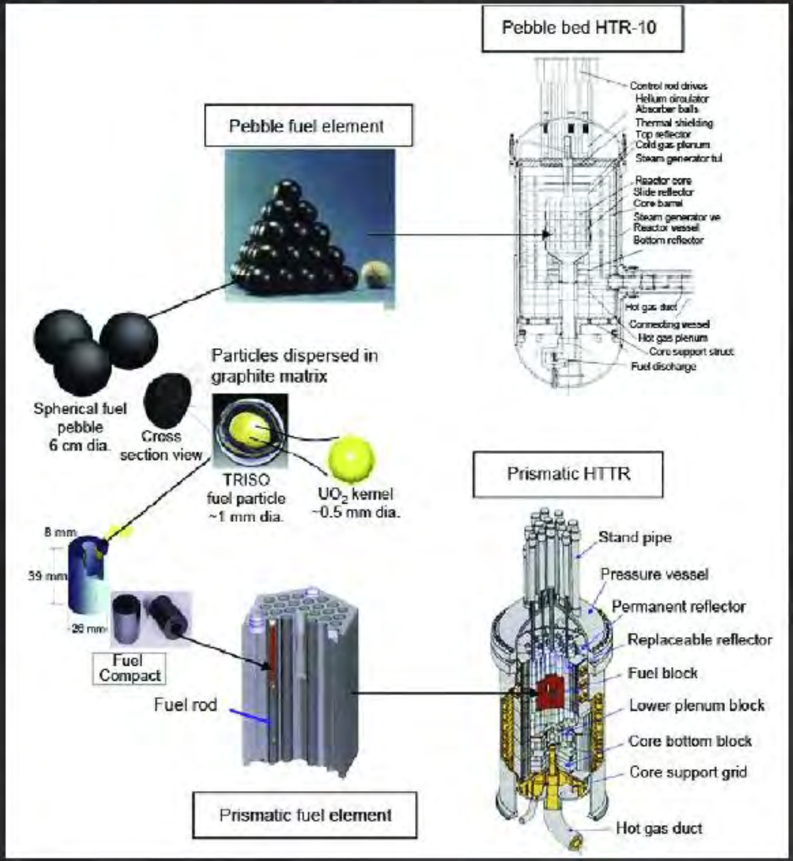
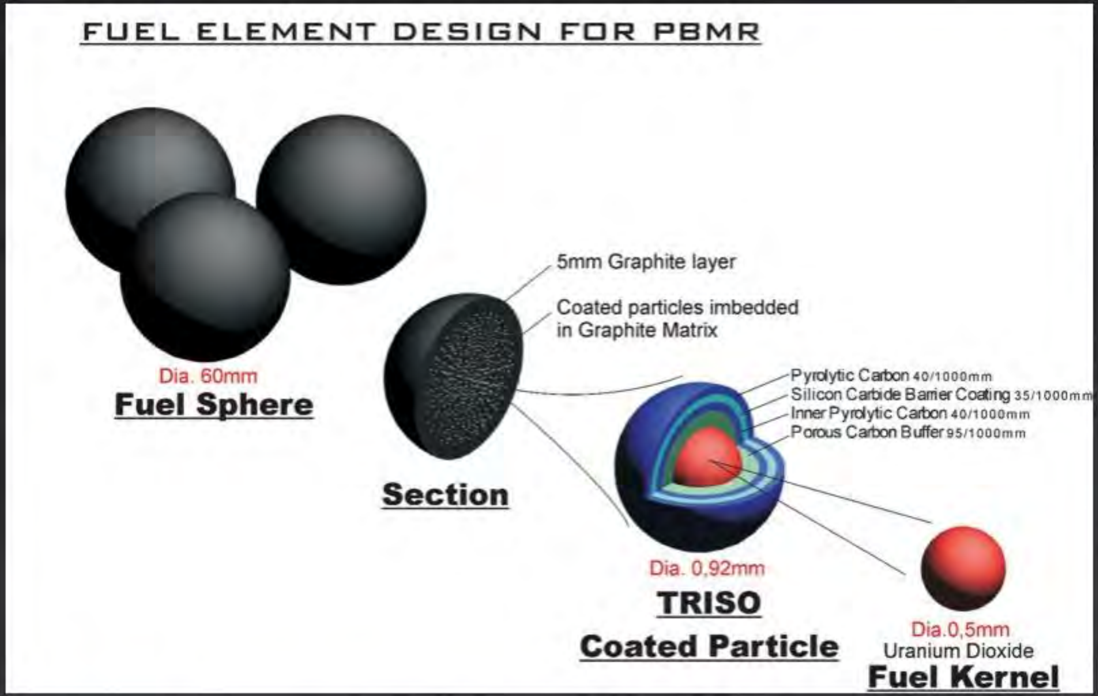
HTGRs and TRISO Fuel





High Temperature Gas Cooled Reactor Fuels and Materials; International Atomic Energy Agency: Vienna, Austria, 2010; p 182.

Photos borrowed :from
https://www.researchgate.net/publication/323778771_Master_Logic_Diagram_An_Approach
tify_Initiating_Events_of_HTGRs/figures?lo=1
http://www.atomic-energy.ru/news/2012/10/07/36571

Fütterer, *Prog. Nucl. Energy*. **2014**, 77, 266-281.

Past, Current, and Future HTGRs

Past

- DRAGON (UK, 1964-1975)
- Peach Bottom (USA, 1966-1974)
- Fort St. Vrain (USA, 1976-1989)
- AVR (Germany, 1967-1988)
- THTR-300 (Germany, 1986-1989)

Current

- HTTR (Japan, 1998)
- HTR-10 (China, 2000)
- HTR-PM (China, 2021)

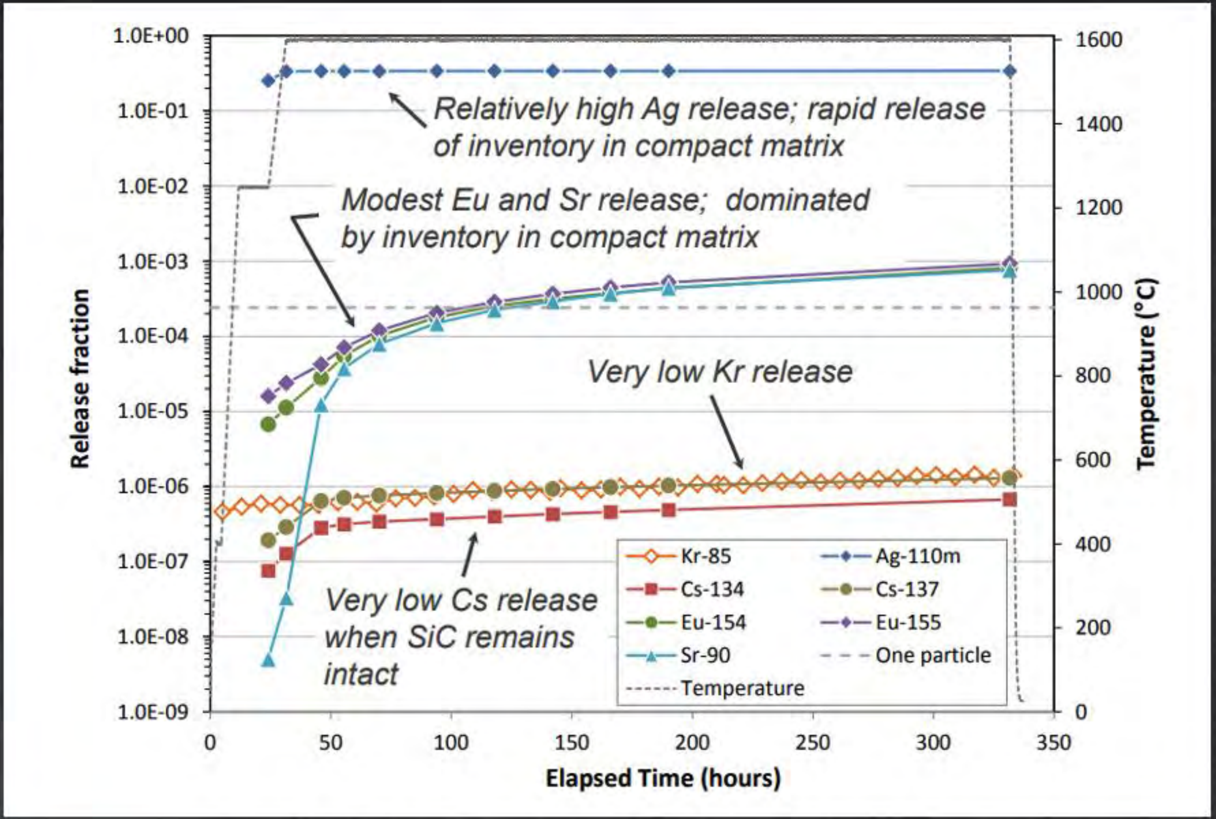
Future

- Xe-100 (X-energy, USA)
- HTR-PM (650 MWe, China)
- EH HTGR (UK, Japan)

Fission Product Diffusion

- TRISO retains 99.99% retention of fission products, but a fraction does escape
- FPs of interest include:
  - **^{134}Cs, ^{137}Cs, ^{110m}Ag, ^{90}Sr, ^{154}Eu, ^{155}Eu**, **^{85}Kr**

A Review of Radionuclide Release From HTGR Cores During Normal Operation, EPRI, Palo Alto, CA: 2003. 1009382.
Demkowicz, *HTR (TRISO) Fuel Safety Research Activities and Needs.*



Relatively high Ag release; rapid release of inventory in compact matrix

Modest Eu and Sr release; dominated by inventory in compact matrix

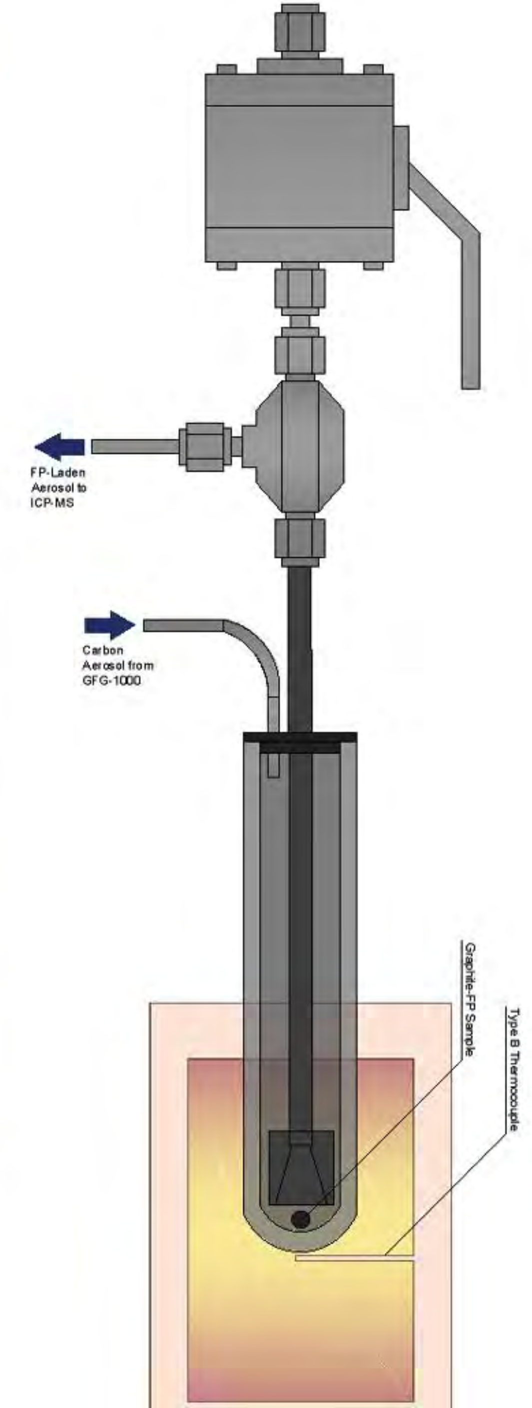
Very low Kr release

Very low Cs release when SiC remains intact

AGR-1 compact safety test results.

Measurement of effective diffusion coefficient of fission products in graphite at high temperatures

- Time-Release experiment
- In this classical approach the term ‘effective’ diffusion coefficient is used to summarize all transport processes, including evaporation, adsorption, diffusion, and trapping, into a single transport process
- Experimentally-derived fractional releases are fit to fractional release rate determined using Fick’s laws
- Results are parameterized using an Arrhenius plot.
- Used by reactor transport models, such as FESCO-II



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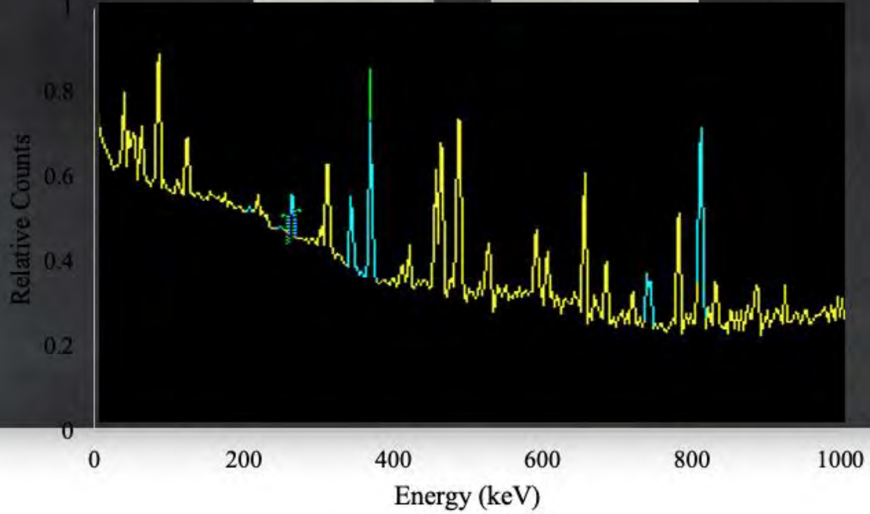
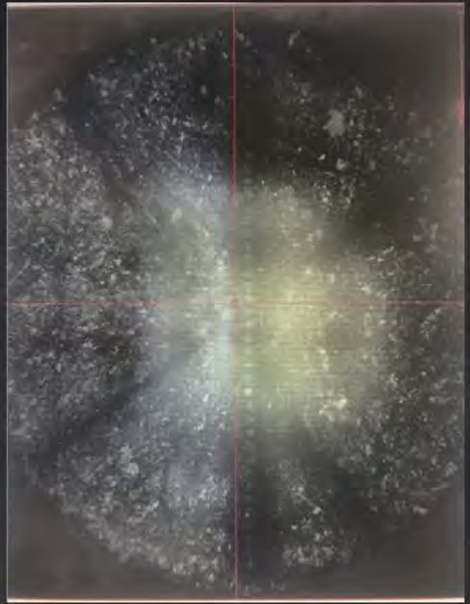
Graphite Loading and Analysis

1. Graphite loaded using pressure vessel and subsequent heating
2. Mass measured using neutron activation analysis
3. Concentration profile measured using laser-ablation ICP-MS

Nitrate standard + water + graphite

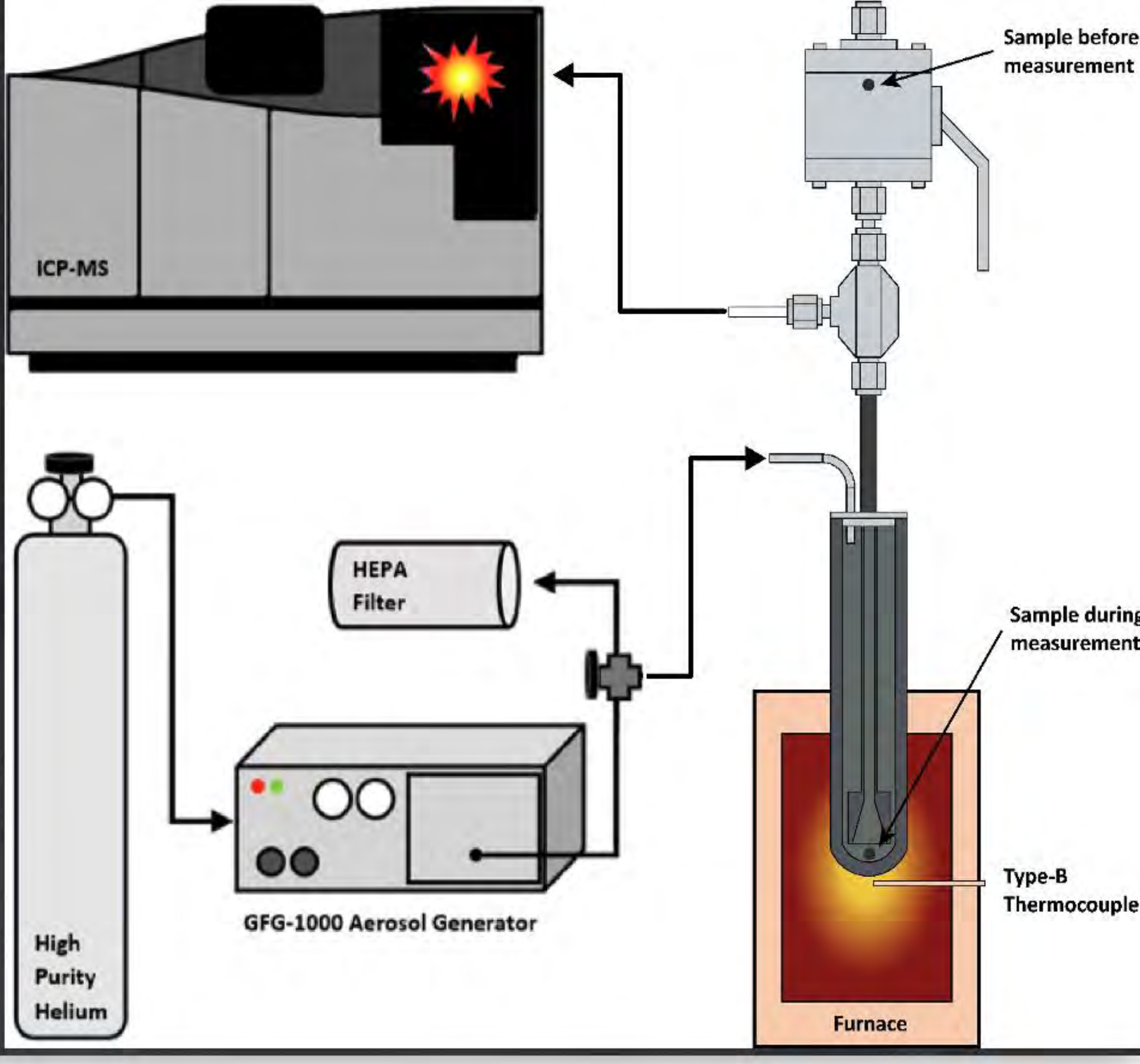




Carter, *J. Nucl. Mater.* **2015**, *466*, 402-408.

Diffusion Experiments

- SiC diffusion cell
- High-temperature box furnace
  - (max temp 1700 °C)
- Carbon aerosol generator + UHP He
- Inductively-coupled plasma mass-spectrometer
- Dual-inlet spray chamber (not pictured)

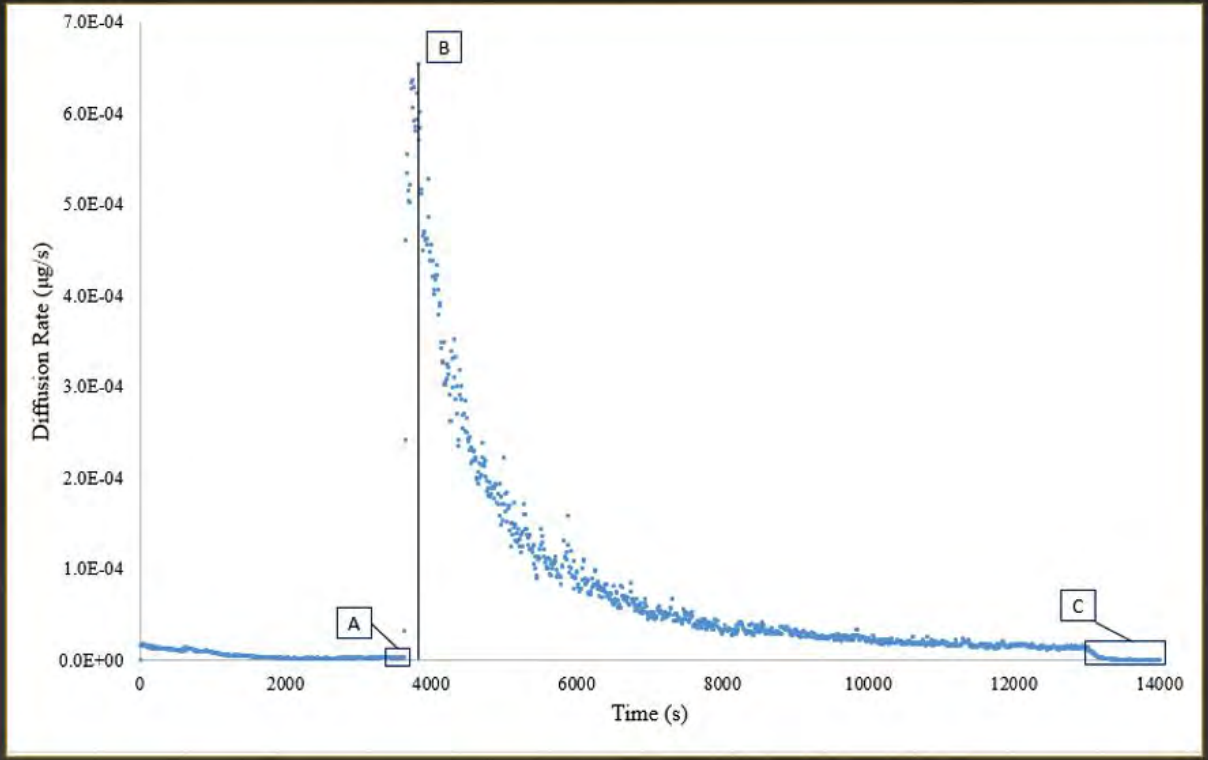


Diffusion Experiments

Weilert, *J. Nucl. Mater.* **2021**, *555*.

Time Release Experiments

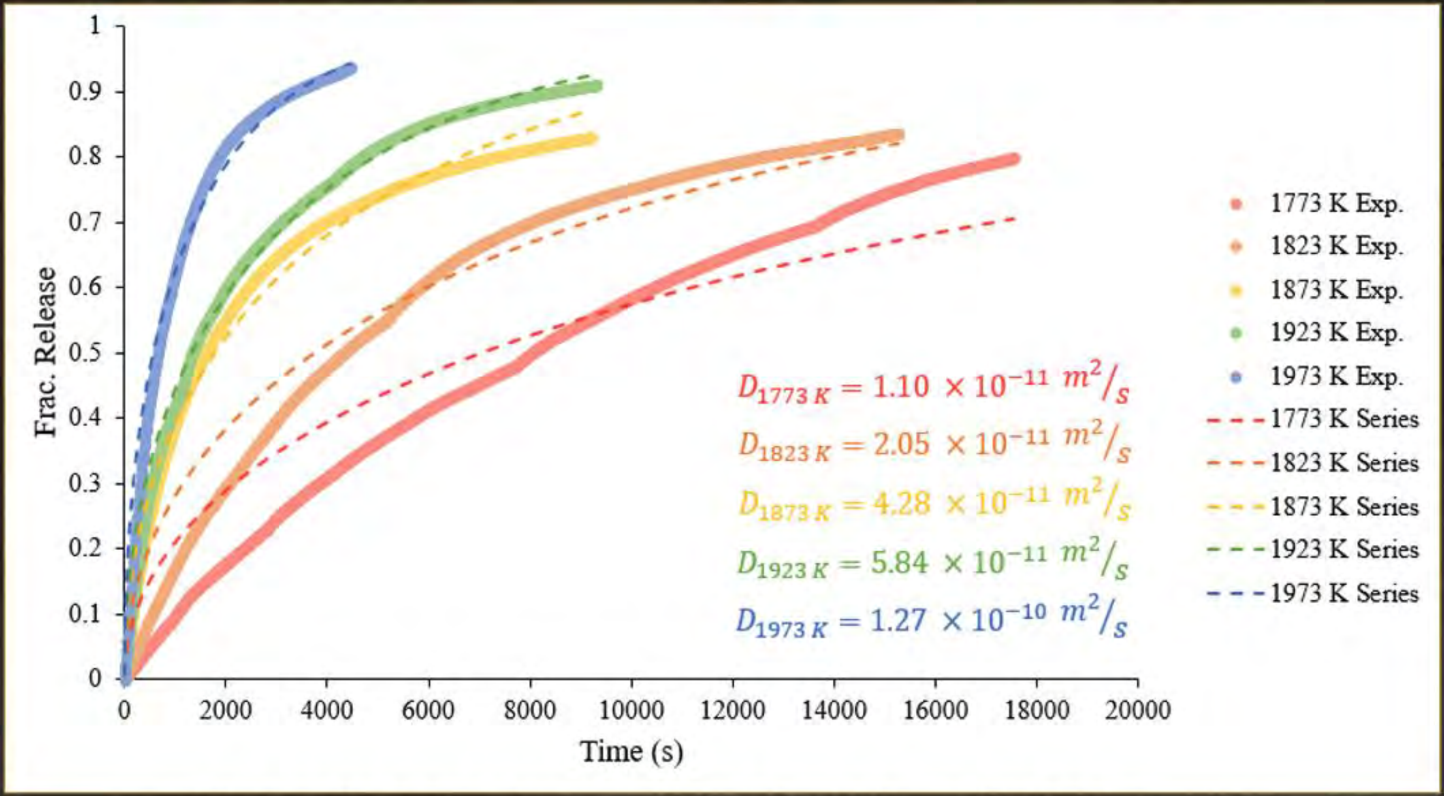
1. Preheat furnace to desired temperature.
2. Start carbon aerosol and data collection.
3. Introduce sample by opening ball valve.
4. Monitor until most of diffusant is believed to have diffused out.
5. Final INAA to determine mass loss.



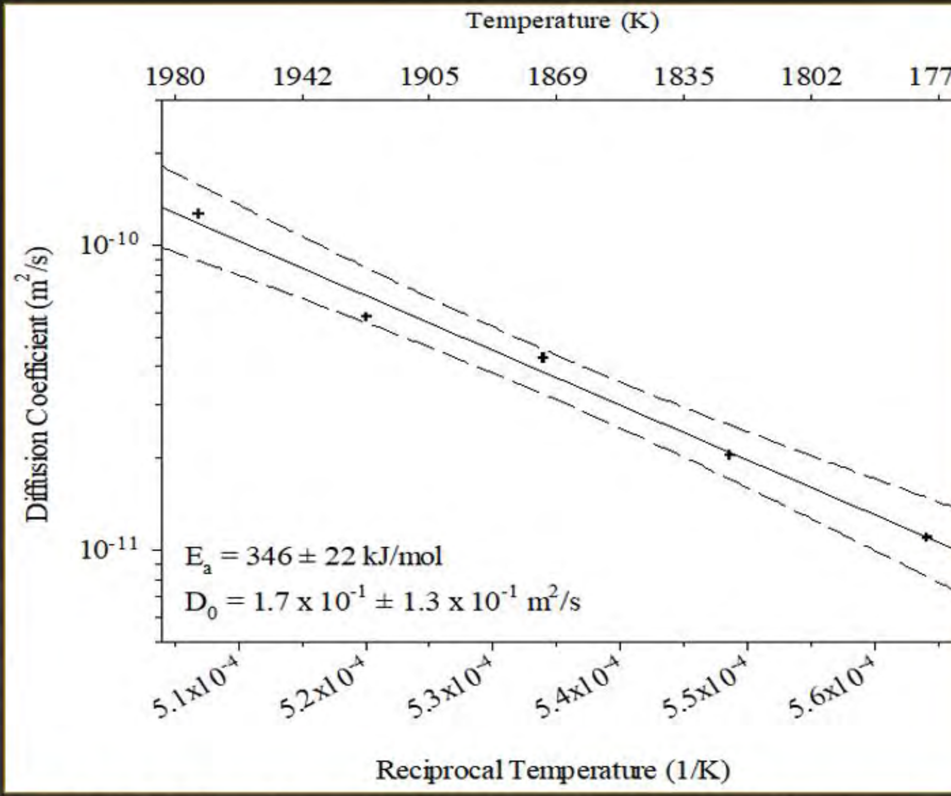
Example: Sr release plot generated at 1873 K.

Weilert, *J. Nucl. Mater.* **2021**, *555*.

Sr Diffusion Experiments



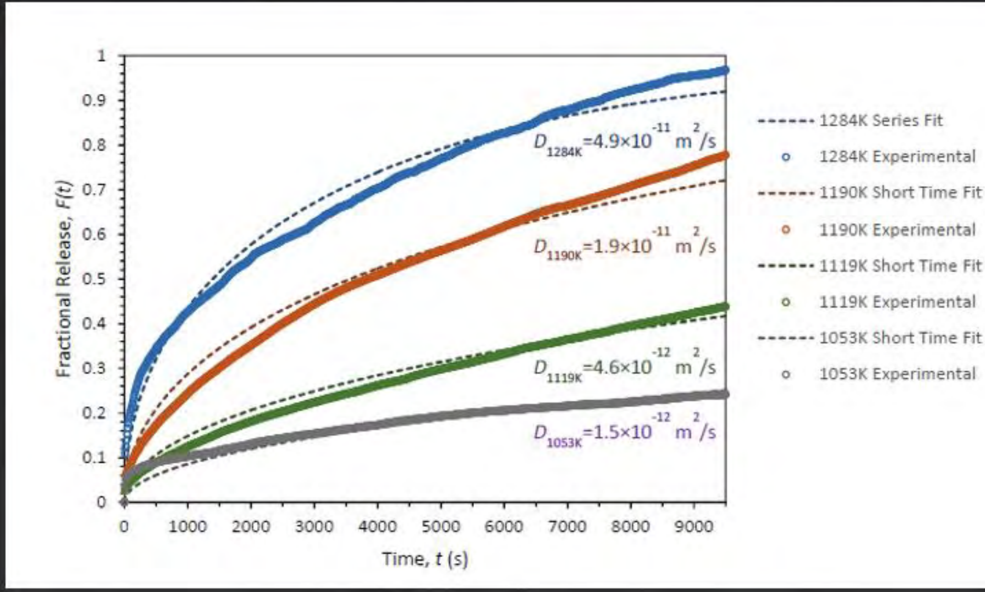
Sr fractional release plots 1773 K – 1973 K



Sr Arrhenius plot 1773 K – 1973 K

Fractional Release of Ag and Pd

Reference	Graphite Type	Temp. Range (K)	E_a (kJ/mol)	D_0 (m^2/s)	D_{1273K} (m^2/s)
Hayashi [35]	IG-110, irradiated	1173-1303	264	6.3×10^{-3}	9.3×10^{-14}
Causey, et.al. [36]	H-451, unirradiated	753-1073	184	$1.7 \times 10^{+1}$	4.8×10^{-7}
Carter, et.al. [23]	IG-110, unirradiated	1048-1284	174 (12)	6.6×10^{-4}	4.8×10^{-11}
This Study	IG-110, unirradiated	1073-1673	224 (4)	2.7×10^{-1}	1.8×10^{-10}
	IG-110, unirradiated + Pd	1223-1673	178 (16)	8.7×10^{-3}	4.2×10^{-10}



Diffusant	Temp. Range (K)	D_0 (m^2/s)	$\pm\Delta D_0$ (m^2/s)	E_a (kJ/mol)	$\pm\Delta E_a$ (kJ/mol)
Sr	1773-1973	1.7×10^{-1}	1.3×10^{-1}	346	22
Ag	1073-1673	2.7×10^{-1}	7.5×10^{-2}	224	4
Ag (+Pd)	1223-1673	8.7×10^{-3}	2.0×10^{-3}	178	13
Pd (+Ag)	1523-1973	6.7×10^{0}	1.2×10^{1}	383	48
Eu	1823-1973	1.5×10^{-3}	3.8×10^{-4}	287	26
Cs	1073-1973	1.9×10^{-6}	4.2×10^{-8}	103	3

Publications

Weilert, T. M., Walton, K. L., Loyalka, S. K., Brockman, J. D*., Diffusion of Cesium in Oxidized and Unoxidized IG-110 Nuclear Graphite. Nuclear Materials, 2022, 570, 153949.

Weilert, T. M., Walton, K. L., Loyalka, S. K., Brockman, J. D*., Europium Diffusion in IG-110 Nuclear Graphite. Nuclear Materials, 2022, 551, 153544.

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Carter, L., Brockman, J.*, Robertson, J., & Loyalka, S. (2016). Diffusion of cesium and iodine in compressed IG-110 graphite compacts. *Journal of Nuclear Materials, 476*, 30-35.

Carter, L., Brockman, J.*, Robertson, J., & Loyalka, S. (2016). ICP-MS measurement of iodine diffusion in IG-110 graphite for HTGR/VHTR. *Journal of Nuclear Materials, 473*, 218-222.

Carter, L. M., Brockman, J. D.*, Loyalka, S. K., & Robertson, J. D. (2016). Calibration of a system for measurements of diffusion coefficients of fission products in HTGR/VHTR core materials. *Journal of Radioanalytical and Nuclear Chemistry, 307*(3), 1771-1775.

Carter, L., Brockman, J.*, Loyalka, S., & Robertson, J. (2015). Measurement of cesium diffusion coefficients in graphite IG-110. *Journal of Nuclear Materials, 460*, 30-36.

Carter, L., Brockman, J.*, Robertson, J., & Loyalka, S. (2015). ICP-MS measurement of diffusion coefficients of Cs in NBG-18 graphite. *Journal of Nuclear Materials, 466*, 402-408.

Future work

- Correlate physical and chemical changes in graphite with the FP effective diffusion coefficient
- Examine realistic FP distributions
- Examine the role of Pd in the migration of Ag in IG-110 graphite
- Examine the effects of irradiation on FP diffusion in IG-110 graphite

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