Preliminary Fission Product Transport Modeling Results

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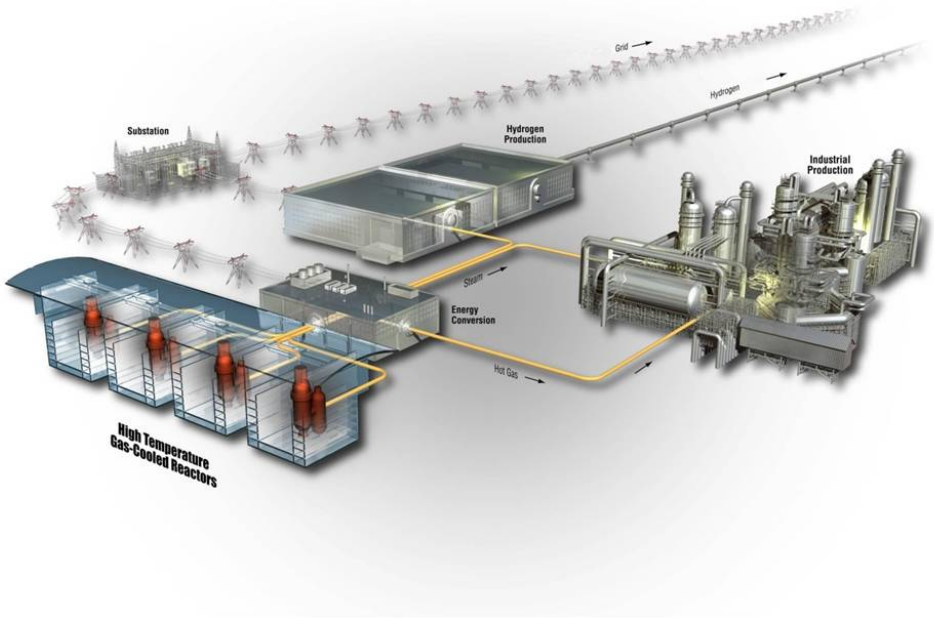

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Objectives

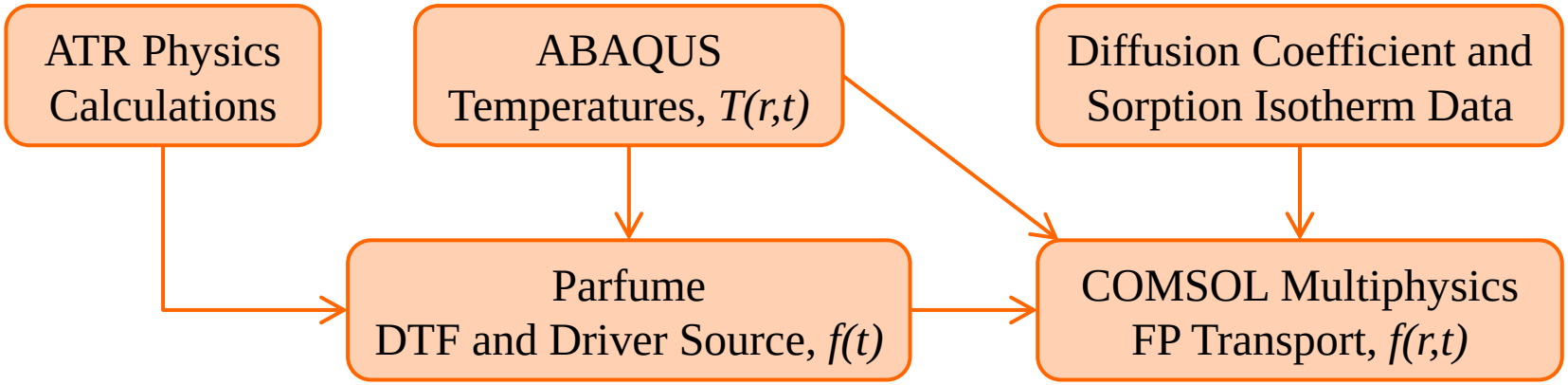
- Using our current best estimates of diffusion coefficients and sorption isotherms, develop a model of AGR-3/4 to perform fission product transport calculations to:
  - Guide PIE plans and outline measurement objectives/expectations
  - Identify appropriate parameters for heating tests
  - Provide an integral check on measured/calculated diffusion coefficients
- Identify a suitable method of determining temperature-dependent diffusion coefficients of key fission products from data measured in PIE

Model Description

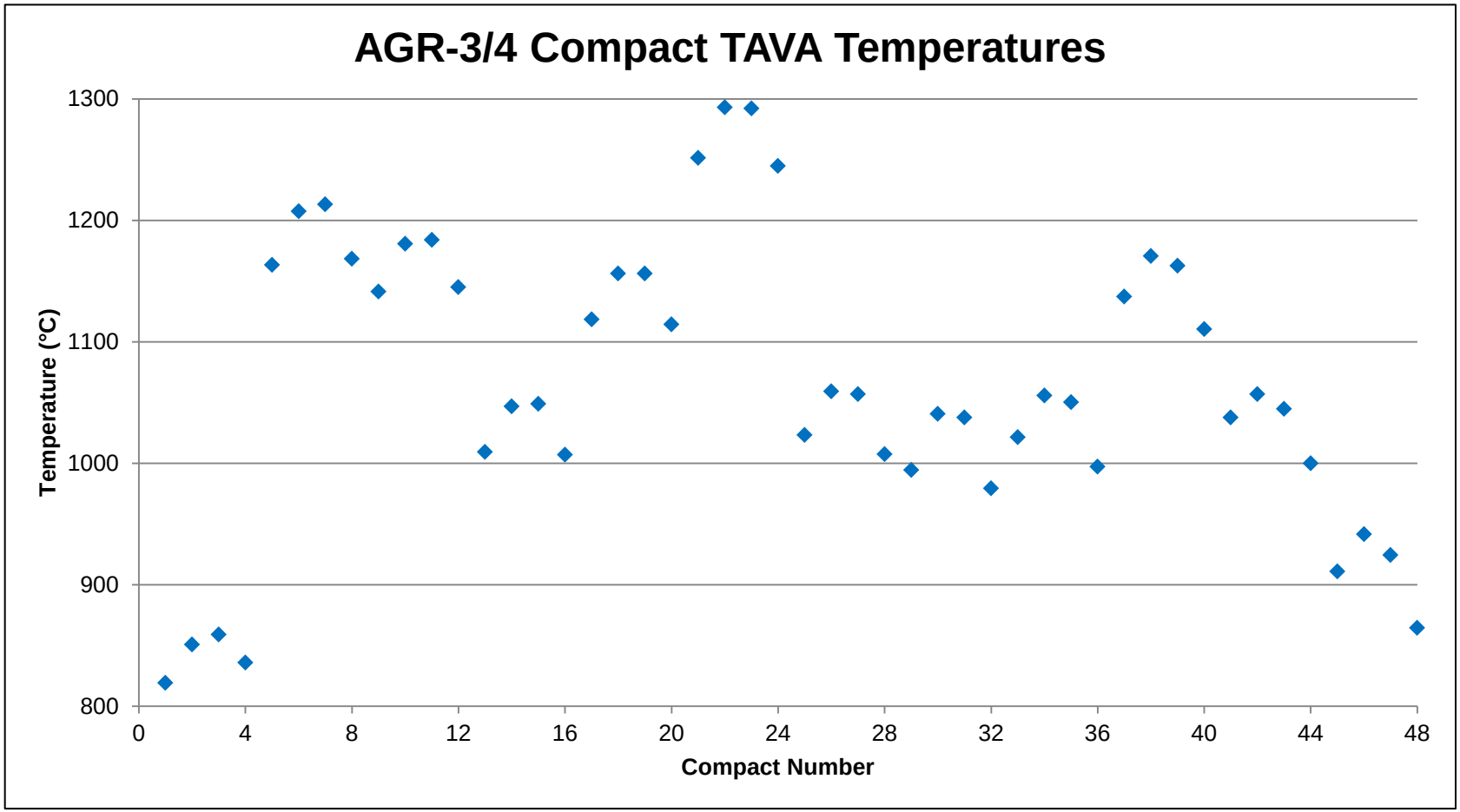
- 1D radial diffusion equations for Ag, Cs, and Sr) are solved in COMSOL Multiphysics

$$\frac{\partial C}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(rD\frac{\partial C}{\partial r}\right)$$

- Concentrations are discontinuous at the gaps between rings; partition coefficients are given by temperature dependent sorption isotherms
- Time and space-dependent temperature profiles are based on ABAQUS calculations
- Time-dependent source terms for each fission product (from both DTF and driver fuel) are calculated with PARFUME



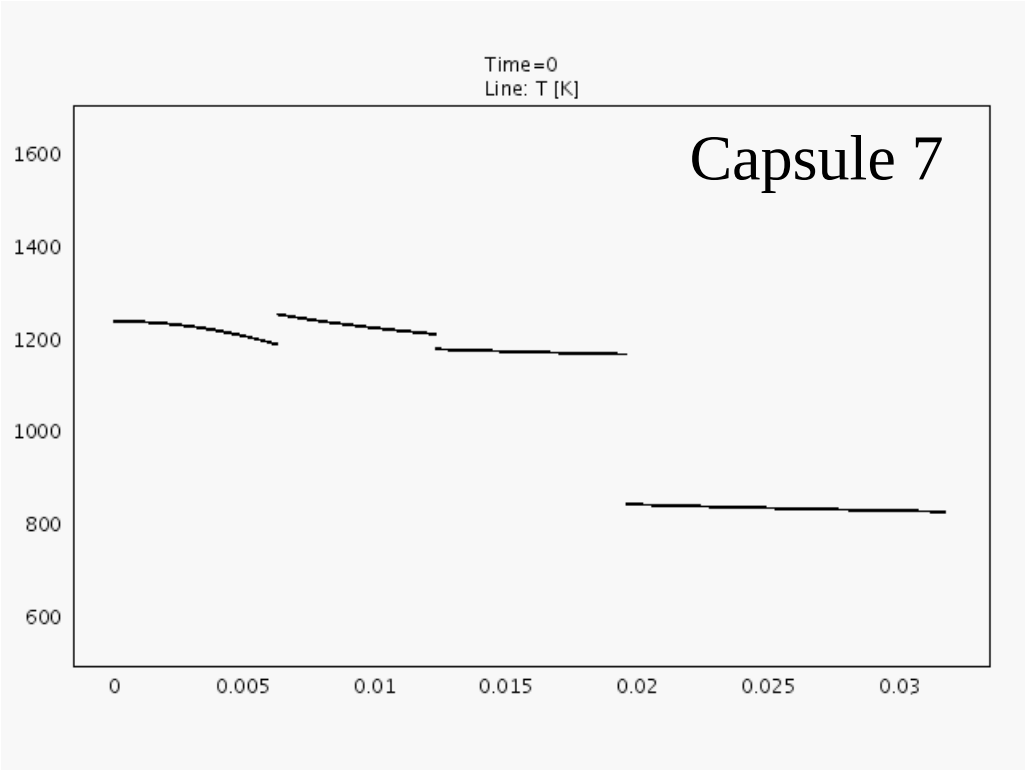
AGR-3/4 Compact TAVA Temperatures



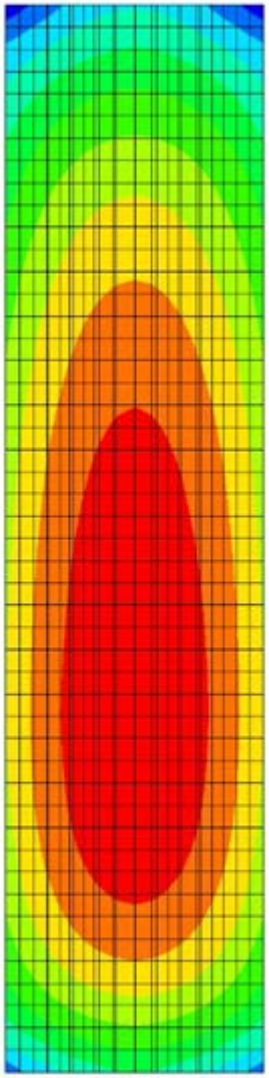
Compacts 12-4 (1) to 1-1 (48)

Temperature Distribution

- Time varying temperature profile at compact center from ABAQUS analysis
- Temperatures fixed at boundaries, solved in COMSOL
  - Heat generation in fuel region backed out from analytical solution



Capsule 12

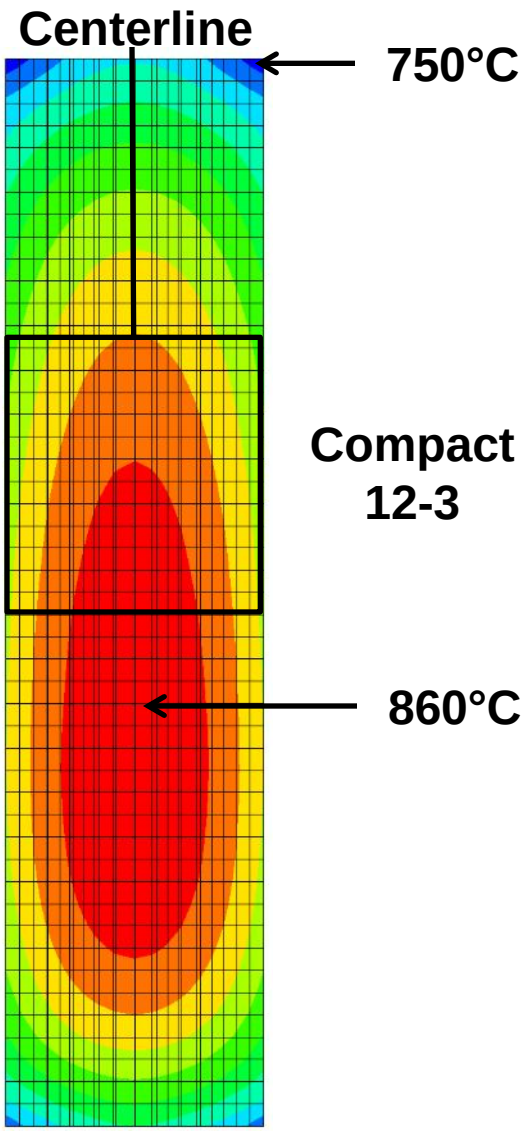


PARFUME Compact Source Terms

- 3 capsules
  - Capsule 12 → coldest (TAVA 841°C)
  - Capsule 7 → hottest (TAVA 1270°C)
  - Capsule 2 → intermediate (TAVA 1035°C)

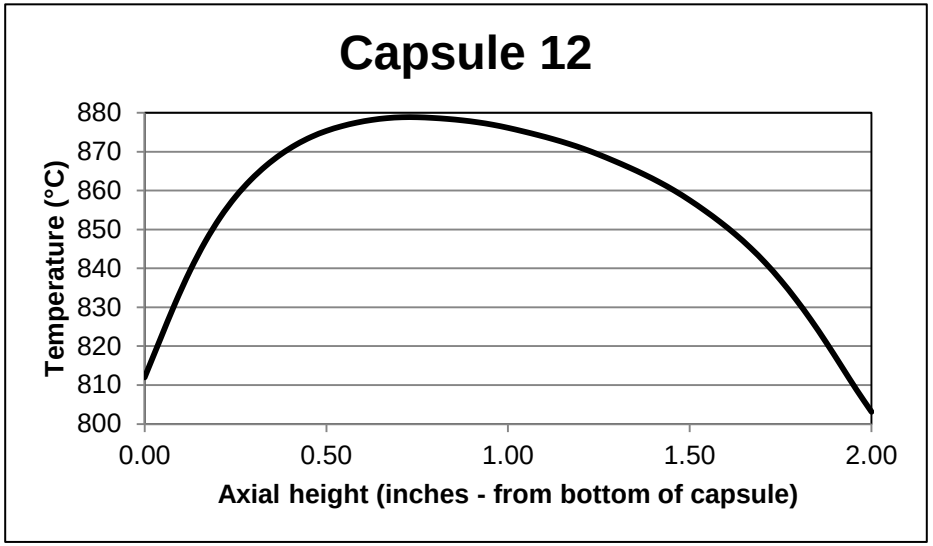
- 18 sources per compact
  - 3 FPs → elemental Ag/Cs/Sr
  - 3 "Driver Fuel" (1872 p/c) + 3 "DTF" (20 p/c)

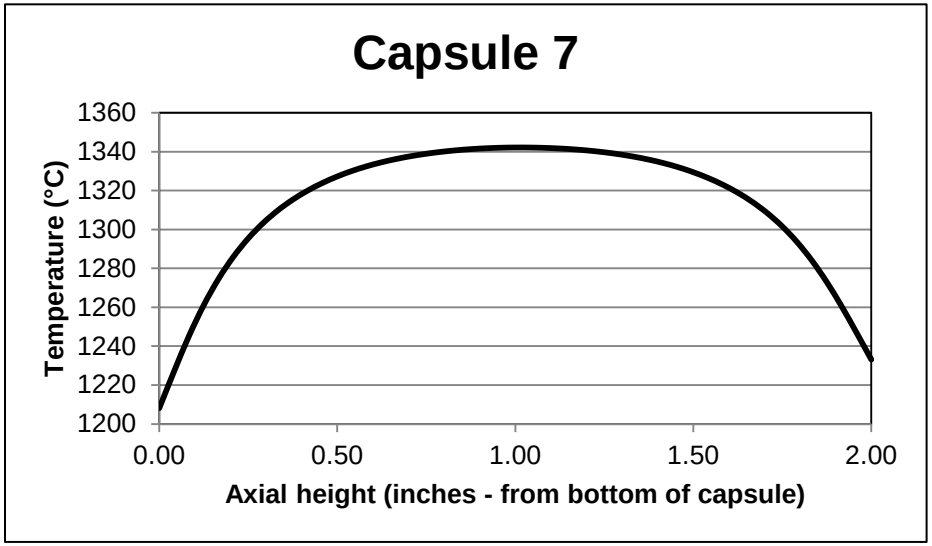
- Sources calculated by PARFUME
  - AGR-3/4 fuel characteristics
  - Compact irradiation conditions
    - As-run burnup and fast fluence
    - As-run daily temperatures (min / avg / max)
      - Driver Fuel → compact
      - DTF → centerline
    - DTF fails at "t = 0"

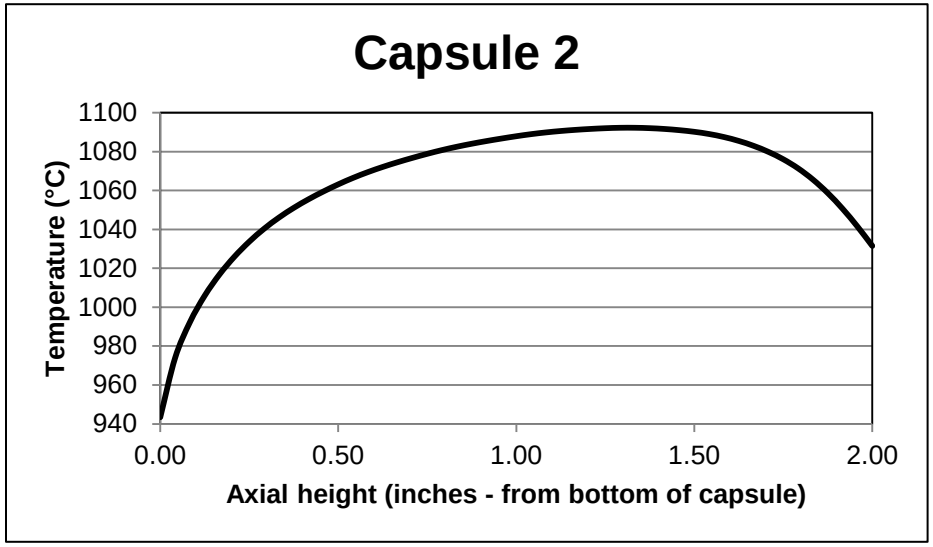


Capsule 12 - Cycle 154B – 246 EFPD

Axial Temperature Profiles







PARFUME Source Term – Capsule 12

Compact	Fuel	Ag			Cs			Sr		
		Min	Avg	Max	Min	Avg	Max	Min	Avg	Max
12-4	Driver	2.5×10^{12}	2.5×10^{12}	2.5×10^{12}	1.6×10^{14}	2.2×10^{14}	2.5×10^{14}	2.0×10^{14}	2.0×10^{14}	2.0×10^{14}
	DTF	3.6×10^{14}	4.5×10^{14}	5.1×10^{14}	1.4×10^{16}	2.0×10^{16}	2.5×10^{16}	3.3×10^{10}	3.3×10^{10}	3.4×10^{10}
12-3	Driver	2.7×10^{12}	2.8×10^{12}	2.8×10^{12}	2.4×10^{14}	2.6×10^{14}	2.7×10^{14}	2.2×10^{14}	2.2×10^{14}	2.2×10^{14}
	DTF	5.6×10^{14}	5.9×10^{14}	6.1×10^{14}	2.6×10^{16}	2.9×10^{16}	3.1×10^{16}	3.6×10^{10}	3.7×10^{10}	3.8×10^{10}
12-2	Driver	3.0×10^{12}	3.0×10^{12}	3.1×10^{12}	2.7×10^{14}	2.8×10^{14}	2.9×10^{14}	2.3×10^{14}	2.3×10^{14}	2.3×10^{14}
	DTF	6.7×10^{14}	6.8×10^{14}	6.8×10^{14}	3.3×10^{16}	3.4×10^{16}	3.4×10^{16}	4.0×10^{10}	4.0×10^{10}	4.0×10^{10}
12-1	Driver	3.3×10^{12}	3.3×10^{12}	3.3×10^{12}	2.2×10^{14}	2.8×10^{14}	3.1×10^{14}	2.4×10^{14}	2.4×10^{14}	2.4×10^{14}
	DTF	5.1×10^{14}	6.6×10^{14}	7.4×10^{14}	1.9×10^{16}	2.9×10^{16}	3.5×10^{16}	3.9×10^{10}	4.0×10^{10}	4.2×10^{10}
Total	Driver	1.2×10^{13}	1.2×10^{13}	1.2×10^{13}	8.9×10^{14}	1.0×10^{15}	1.1×10^{15}	8.9×10^{14}	8.9×10^{14}	8.9×10^{14}
	DTF	2.1×10^{15}	2.4×10^{15}	2.5×10^{15}	9.2×10^{16}	1.1×10^{17}	1.3×10^{17}	1.5×10^{11}	1.5×10^{11}	1.5×10^{11}

Uranium contamination: 3.5×10^{-5} (Driver Fuel) / 5.25×10^{-7} (DTF)

PARFUME Source Term – Capsule 7

Compact	Fuel	Ag			Cs			Sr		
		Min	Avg	Max	Min	Avg	Max	Min	Avg	Max
7-4	Driver	8.7×10^{16}	1.8×10^{17}	2.5×10^{17}	1.9×10^{16}	1.4×10^{17}	4.0×10^{17}	1.7×10^{16}	2.3×10^{17}	8.6×10^{17}
	DTF	4.2×10^{15}	4.2×10^{15}	4.2×10^{15}	2.9×10^{17}	2.9×10^{17}	2.9×10^{17}	1.1×10^{15}	3.3×10^{15}	5.8×10^{15}
7-3	Driver	1.7×10^{17}	2.2×10^{17}	2.6×10^{17}	1.3×10^{17}	2.6×10^{17}	4.9×10^{17}	2.2×10^{17}	5.3×10^{17}	1.1×10^{18}
	DTF	4.2×10^{15}	4.2×10^{15}	4.2×10^{15}	2.9×10^{17}	2.9×10^{17}	2.9×10^{17}	6.0×10^{15}	6.8×10^{15}	7.2×10^{15}
7-2	Driver	1.7×10^{17}	2.2×10^{17}	2.6×10^{17}	1.3×10^{17}	2.6×10^{17}	4.9×10^{17}	2.1×10^{17}	5.2×10^{17}	1.1×10^{18}
	DTF	4.2×10^{15}	4.2×10^{15}	4.2×10^{15}	2.9×10^{17}	2.9×10^{17}	2.9×10^{17}	5.7×10^{15}	6.7×10^{15}	7.2×10^{15}
7-1	Driver	8.1×10^{16}	1.7×10^{17}	2.5×10^{17}	1.3×10^{16}	1.2×10^{17}	4.0×10^{17}	1.3×10^{16}	1.9×10^{17}	8.3×10^{17}
	DTF	4.2×10^{15}	4.2×10^{15}	4.2×10^{15}	2.9×10^{17}	2.9×10^{17}	2.9×10^{17}	6.4×10^{14}	2.8×10^{15}	6.9×10^{15}
Total	Driver	5.1×10^{17}	7.9×10^{17}	1.0×10^{18}	2.9×10^{17}	7.8×10^{17}	1.8×10^{18}	4.6×10^{17}	1.5×10^{18}	3.9×10^{18}
	DTF	1.7×10^{16}	1.7×10^{16}	1.7×10^{16}	1.2×10^{18}	1.2×10^{18}	1.2×10^{18}	1.3×10^{16}	2.0×10^{16}	2.7×10^{16}

Uranium contamination: 3.5×10^{-5} (Driver Fuel) / 5.25×10^{-7} (DTF)

100% release

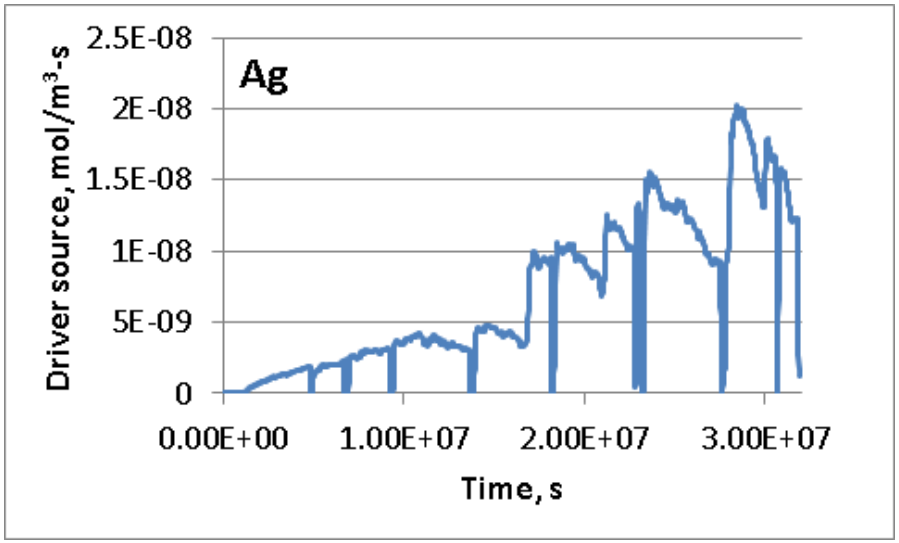
PARFUME Source Term – Capsule 2

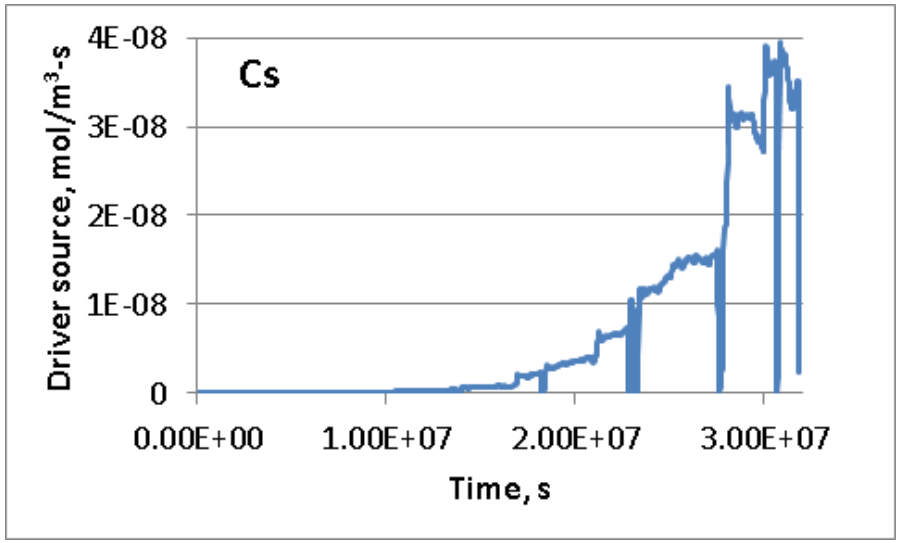
Compact	Fuel	Ag			Cs			Sr		
		Min	Avg	Max	Min	Avg	Max	Min	Avg	Max
2-4	Driver	1.7×10^{14}	2.6×10^{15}	9.7×10^{15}	6.4×10^{14}	6.6×10^{14}	7.2×10^{14}	4.0×10^{14}	4.1×10^{14}	5.1×10^{14}
	DTF	2.4×10^{15}	2.5×10^{15}	2.5×10^{15}	1.6×10^{17}	1.8×10^{17}	1.9×10^{17}	1.5×10^{12}	5.3×10^{12}	1.0×10^{13}
2-3	Driver	1.9×10^{15}	4.4×10^{15}	9.8×10^{15}	6.4×10^{14}	6.5×10^{14}	7.0×10^{14}	3.9×10^{14}	4.1×10^{14}	5.0×10^{14}
	DTF	2.3×10^{15}	2.3×10^{15}	2.4×10^{15}	1.8×10^{17}	1.8×10^{17}	1.8×10^{17}	9.1×10^{12}	1.0×10^{13}	1.0×10^{13}
2-2	Driver	8.3×10^{14}	2.9×10^{15}	8.3×10^{15}	6.1×10^{14}	6.2×10^{14}	6.6×10^{14}	3.8×10^{14}	3.9×10^{14}	4.5×10^{14}
	DTF	2.2×10^{15}	2.2×10^{15}	2.2×10^{15}	1.7×10^{17}	1.7×10^{17}	1.7×10^{17}	4.2×10^{12}	6.4×10^{12}	8.8×10^{12}
2-1	Driver	1.4×10^{13}	4.9×10^{14}	4.1×10^{15}	5.5×10^{14}	5.8×10^{14}	5.9×10^{14}	3.6×10^{14}	3.6×10^{14}	3.8×10^{14}
	DTF	1.8×10^{15}	2.0×10^{15}	2.0×10^{15}	8.9×10^{16}	1.4×10^{17}	1.6×10^{17}	1.1×10^{11}	1.1×10^{12}	4.0×10^{12}
Total	Driver	2.9×10^{15}	1.0×10^{16}	3.2×10^{16}	2.4×10^{15}	2.5×10^{15}	2.7×10^{15}	1.5×10^{15}	1.6×10^{15}	1.8×10^{15}
	DTF	8.7×10^{15}	9.0×10^{15}	9.1×10^{15}	6.0×10^{17}	6.7×10^{17}	7.0×10^{17}	1.5×10^{13}	2.3×10^{13}	3.3×10^{13}

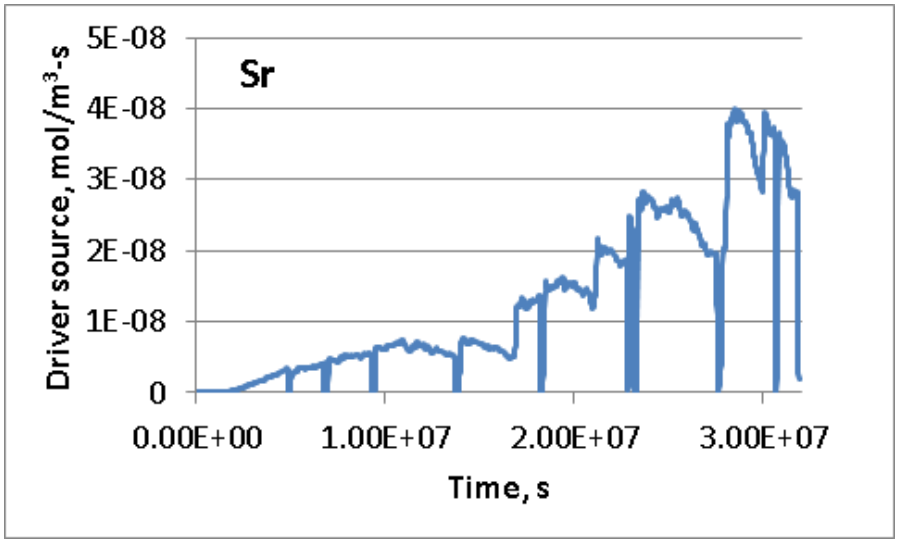
Uranium contamination: 3.5×10^{-5} (Driver Fuel) / 5.25×10^{-7} (DTF)

100% release

Driver Sources – Capsule 7





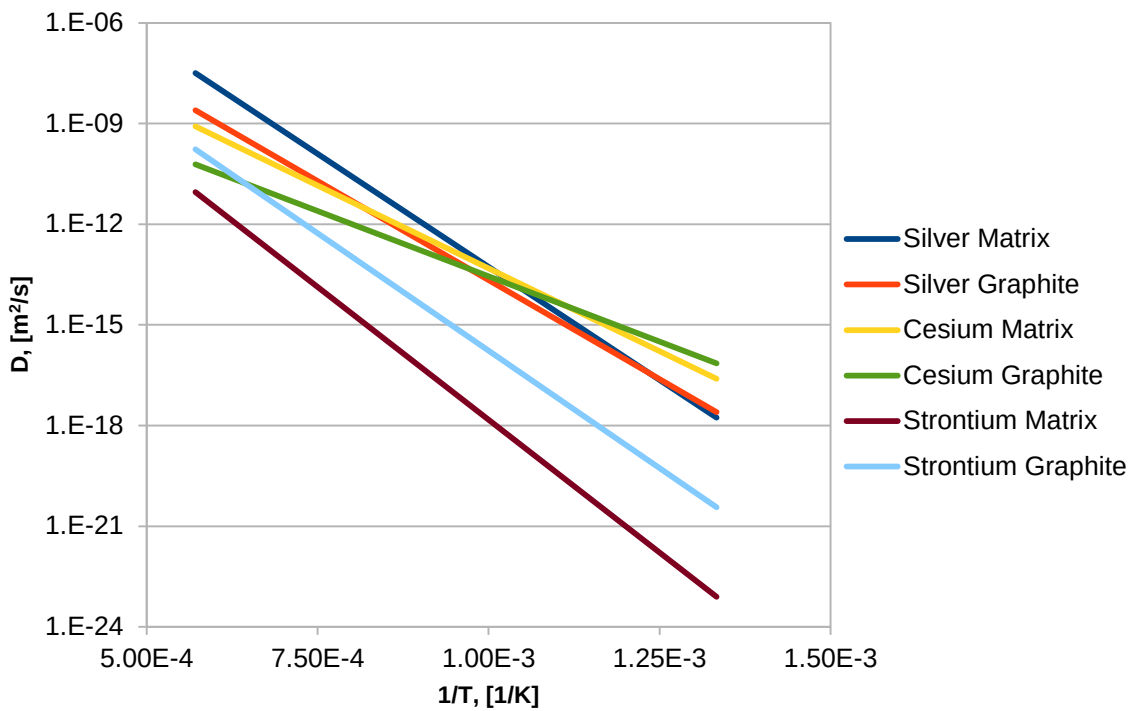


Diffusion Coefficients

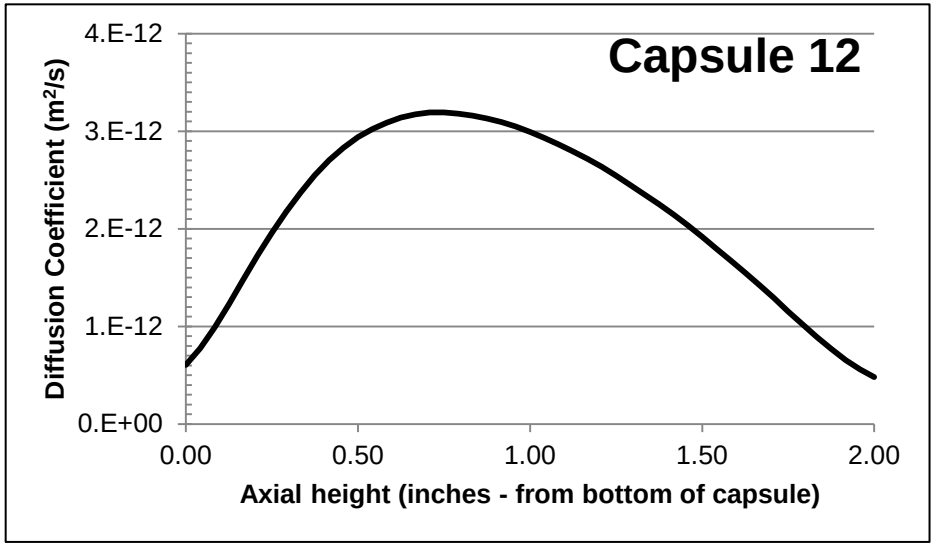
- From TECDOC-978, GA report 911200
- Arrhenius law with temperature:

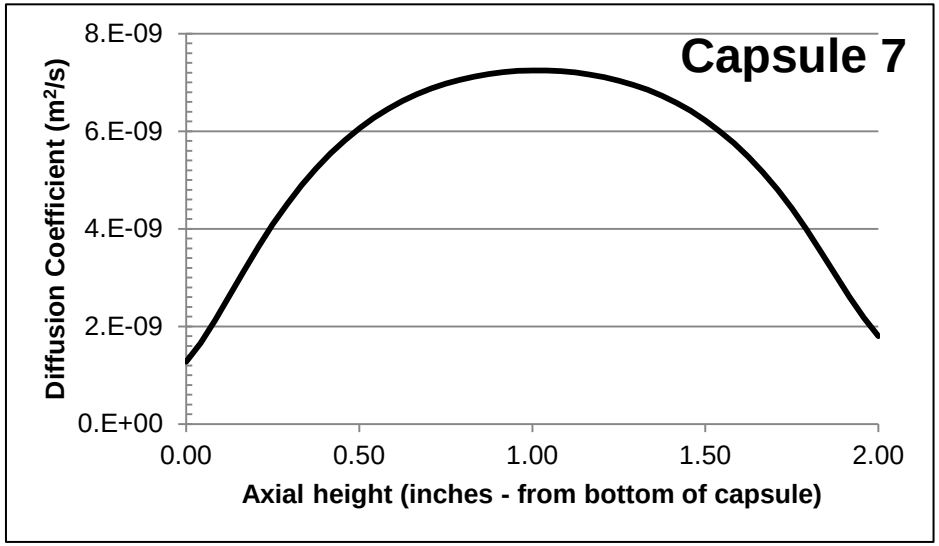
$$D = D_0 \exp\left(-Q/RT\right)$$

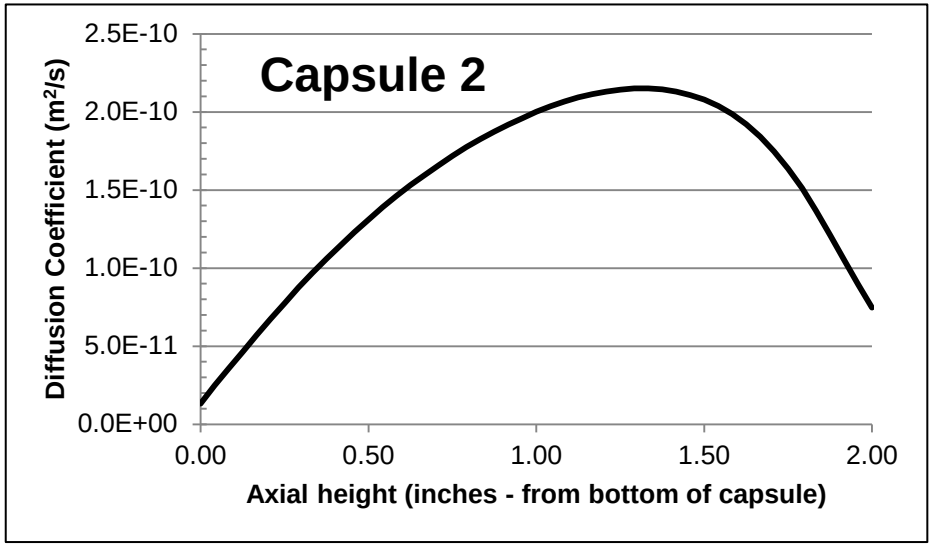
Material	Silver		Cesium		Strontium	
	D_0 [m^2/s]	Q [kJ/mol]	D_0 [m^2/s]	Q [kJ/mol]	D_0 [m^2/s]	Q [kJ/mol]
Matrix	1.6	258	3.60×10^{-4}	189	1.00×10^{-2}	303
Graphite	1.38×10^{-2}	226	1.70×10^{-6}	149	1.70×10^{-2}	268



Diffusion Coefficient Axial Profiles – Ag in Matrix

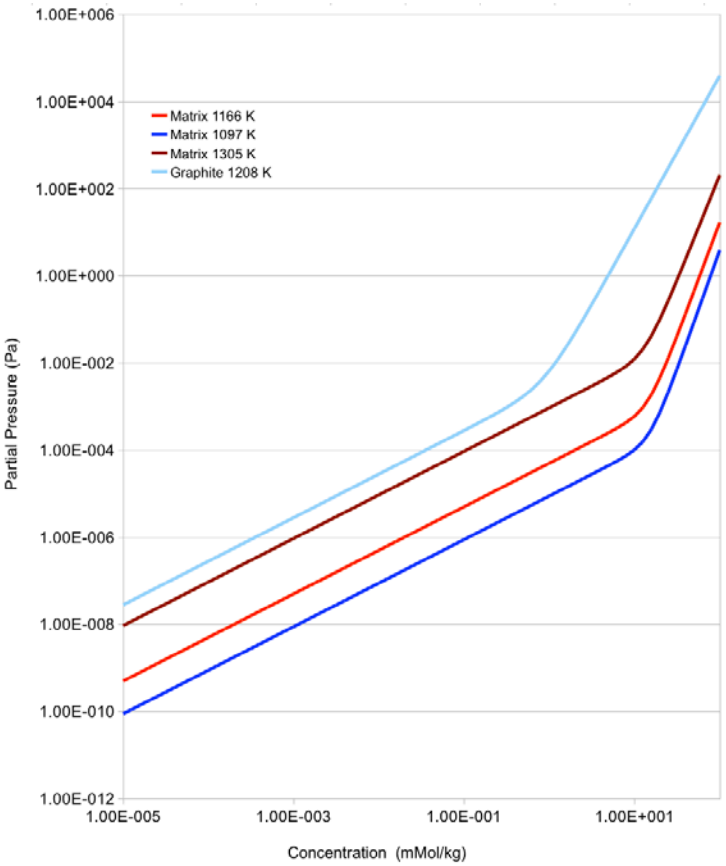






Boundary Conditions - Sorption Isotherms

- Gaps between regions result in abrupt changes in concentration
  - Sorption isotherms relate partial pressure in gap to surface concentrations
  - Temperature drops give different concentrations even for the same material
  - Matrix/graphite interface has different isotherms on each side of the gap



	Matrix						Graphite					
	A	B	D	E	d_1	d_2	A	B	D	E	d_1	d_2
Cs	19.3	-47300	1.51	4340	3.4	6.15×10^{-4}	24	-35700	-1.56	6120	$2.04^{\dagger}$	1.79×10^{-3}
Sr	54.3	-149000	-8.52	28500	3.13	0	19.4	-40100	-0.32	4090	-2.12	0

Sorption Isotherms

- Sum of Henrian (linear) and Freundlich (power law) contributions

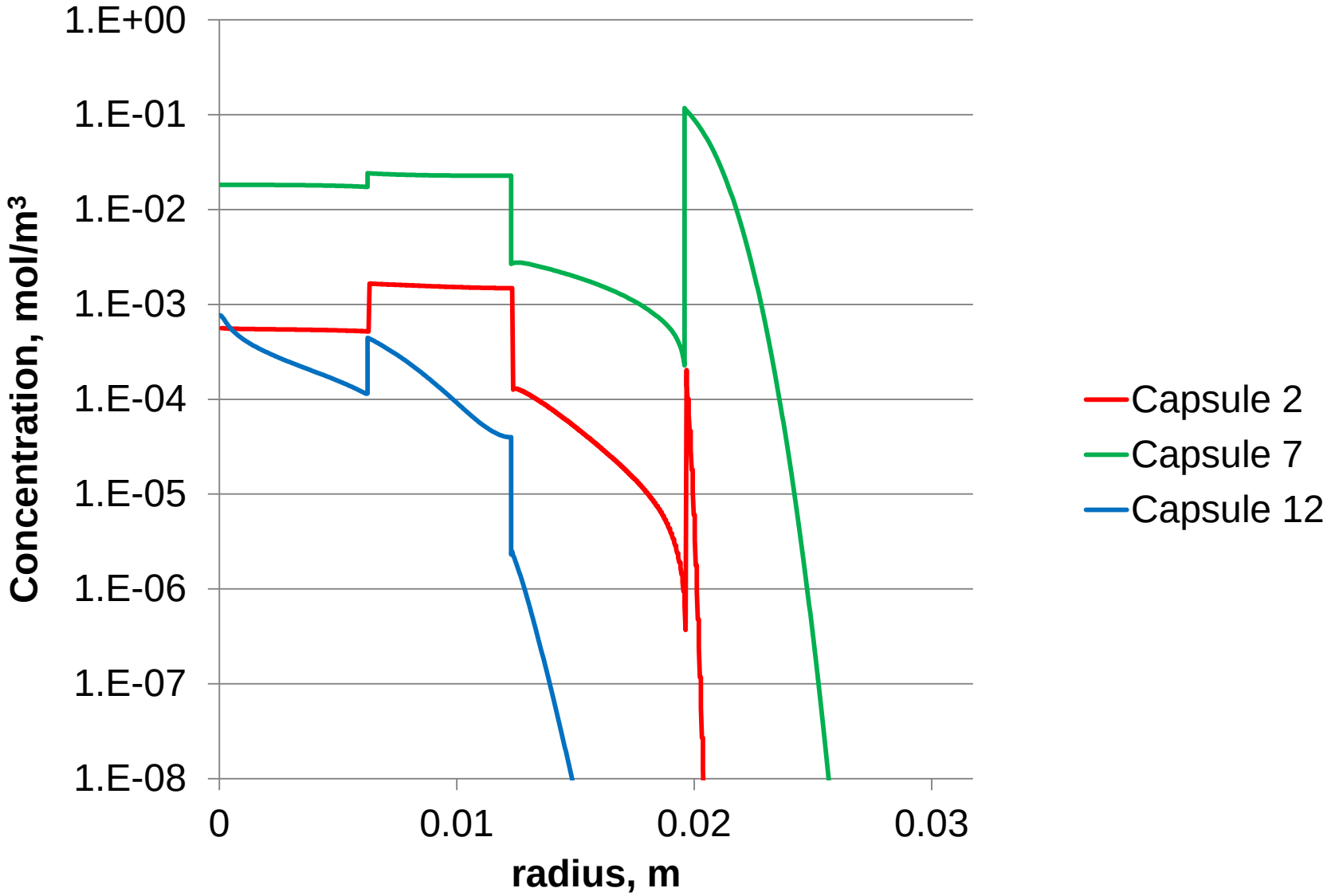
$$P = \exp\left(A + \frac{B}{T}\right) C^{\left(D + \frac{E}{T}\right)} + \exp\left[\left(A + \frac{B}{T}\right) + \left(D - 1 + \frac{E}{T}\right)\left(d_1 - d_2 T\right)\right] C$$

- It is assumed that both surfaces (different materials and temperatures) are in equilibrium with this pressure
- Presently only the linear portion is implemented; this allows for the definition of a partition coefficient:

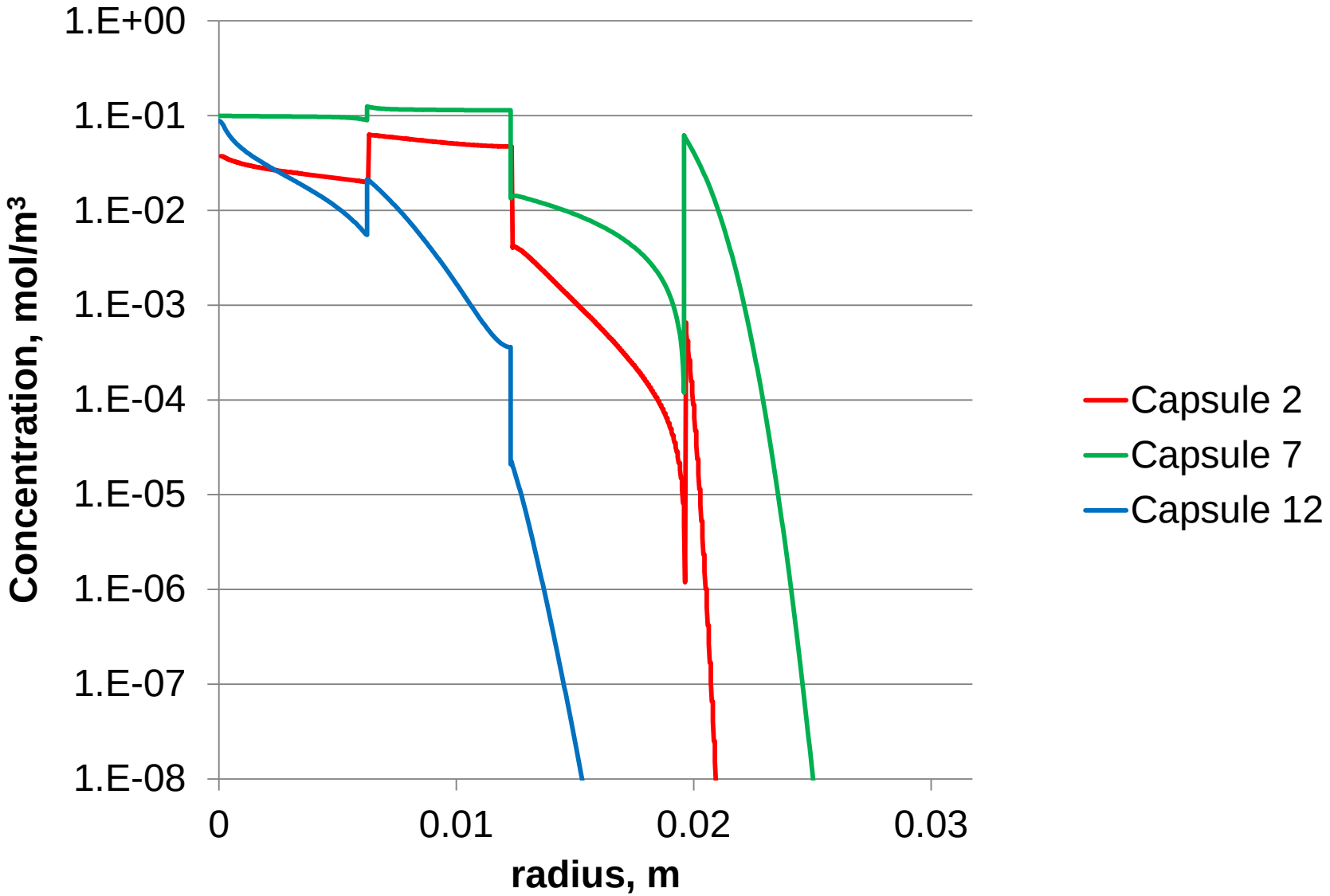
$$\frac{C_0}{C_1} = \frac{\exp\left[\left(A_1 + \frac{B_1}{T_1}\right) + \left(D_1 - 1 + \frac{E_1}{T_1}\right)\left(d_{1_1} - d_{2_1} T_1\right)\right]}{\exp\left[\left(A_0 + \frac{B_0}{T_0}\right) + \left(D_0 - 1 + \frac{E_0}{T_0}\right)\left(d_{1_0} - d_{2_0} T_0\right)\right]}$$

- Flux balance to conserve mass: $D_{0_0} \exp\left(\frac{-Q_0}{RT_0}\right) \frac{\partial C}{\partial r}\bigg|_0 = D_{0_1} \exp\left(\frac{-Q_1}{RT_1}\right) \frac{\partial C}{\partial r}\bigg|_1$

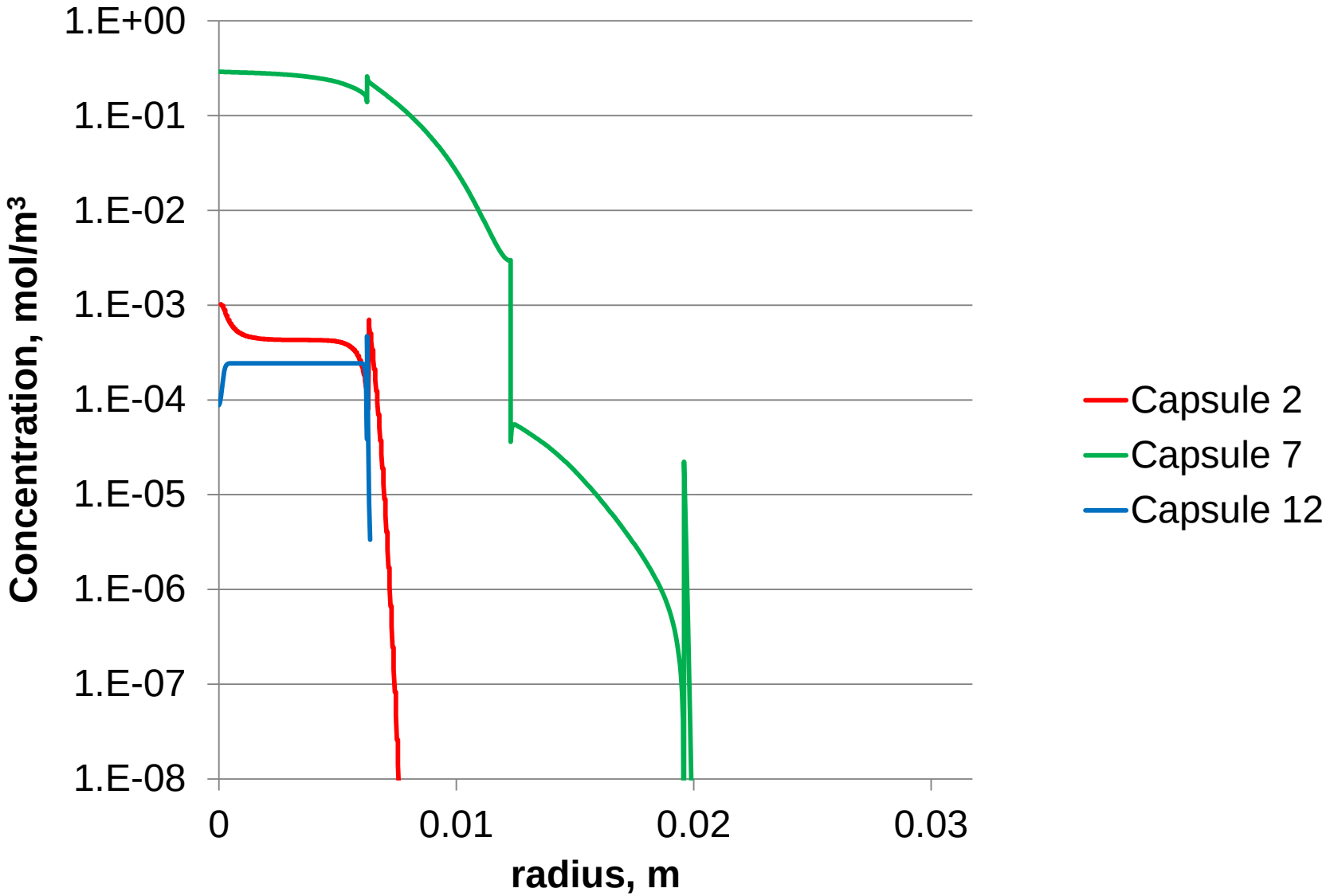
Silver profiles in capsules 2, 7, and 12



Cesium profiles in capsules 2, 7, and 12



Strontium profiles in capsules 2, 7, and 12



Determining Diffusion Coefficients: Method #1

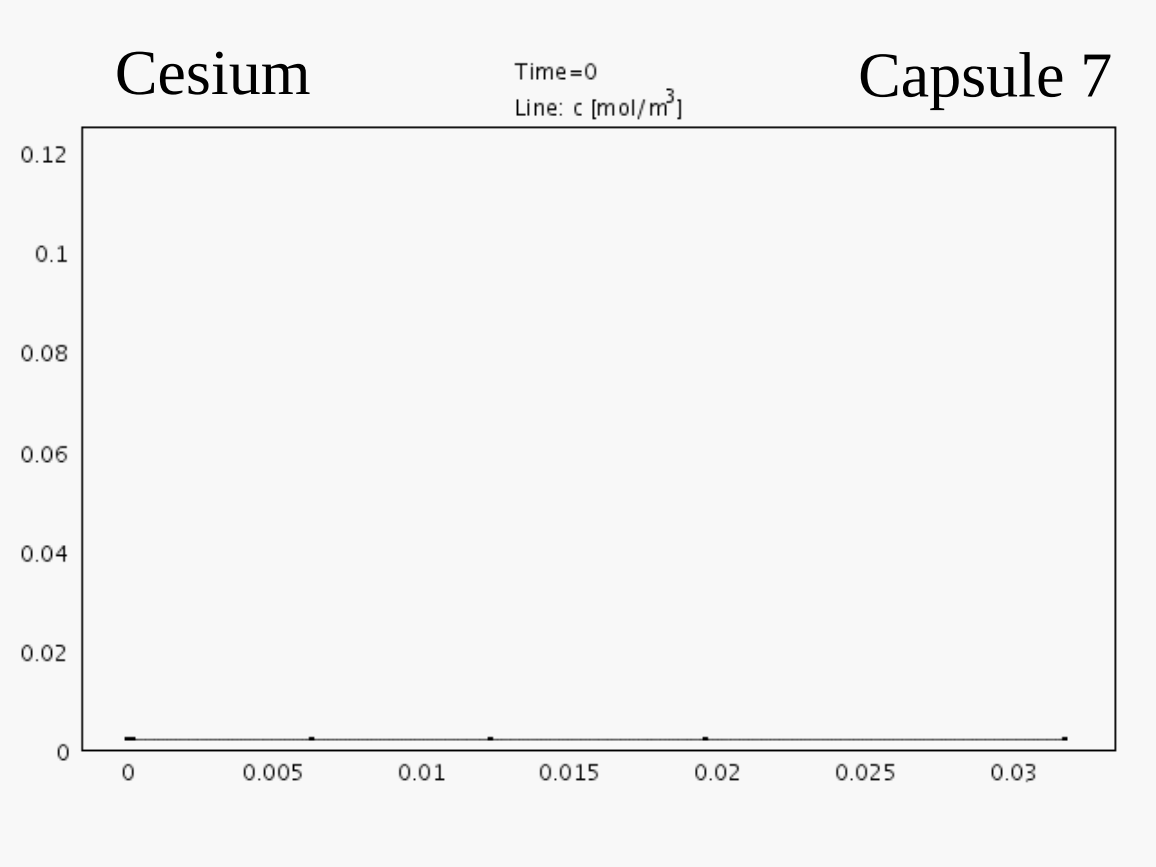
- If:
  - 1) diffusion is steady state (*dC(r)/dt = 0*), and
  - 2) The concentrations C_1 and C_2 at each boundary are constant,
- The profile across a ring is given by

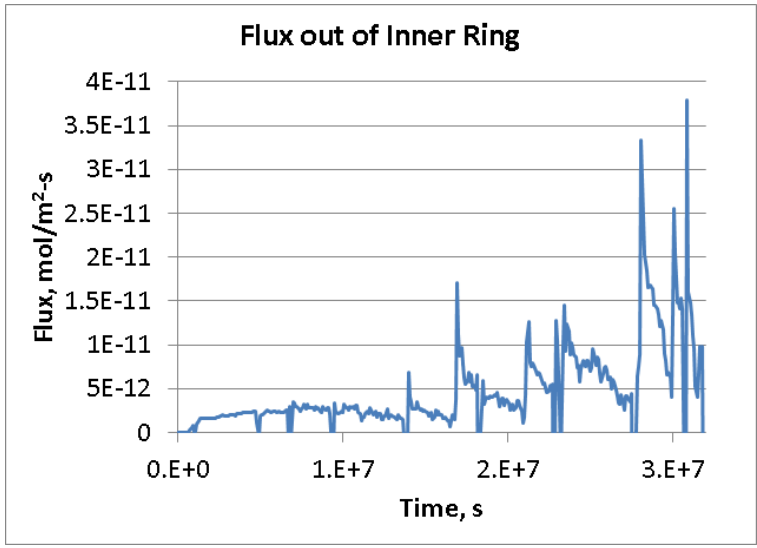
$$C(r) = C_1 + \frac{(C_2 - C_1)\ln\left(r/r_1\right)}{\ln\left(r_2/r_1\right)}$$

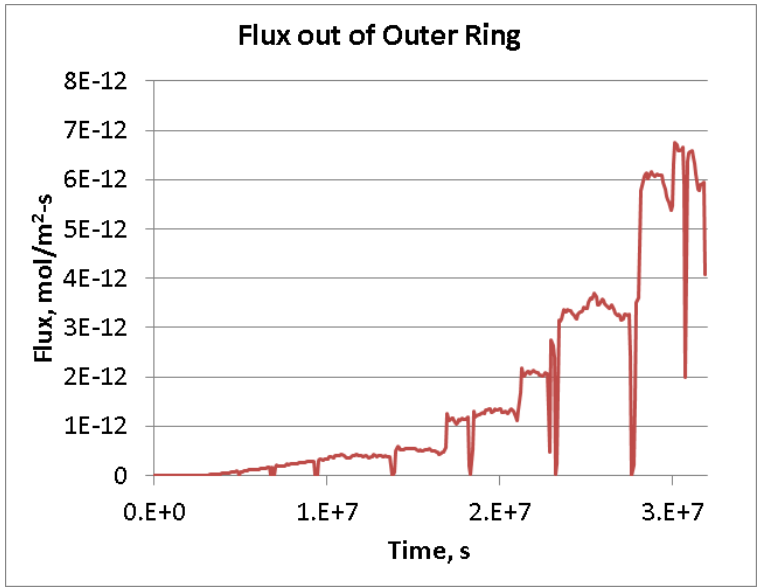
- The (constant) flux through the ring is given by $J = \dfrac{D(C_1 - C_2)}{r_2 \ln\left(r_2/r_1\right)}$
- and the diffusion coefficient is given by $D = \dfrac{M \ln\left(r_2/r_1\right)}{2\pi L t (C_1 - C_2)}$
- where *M* is the total inventory outside the ring (which we can measure)

The Problem with Method #1

- The cold "sink" really acts as a diffusion *barrier*, which prevents any of the rings (in any of the capsules) from approaching a steady state







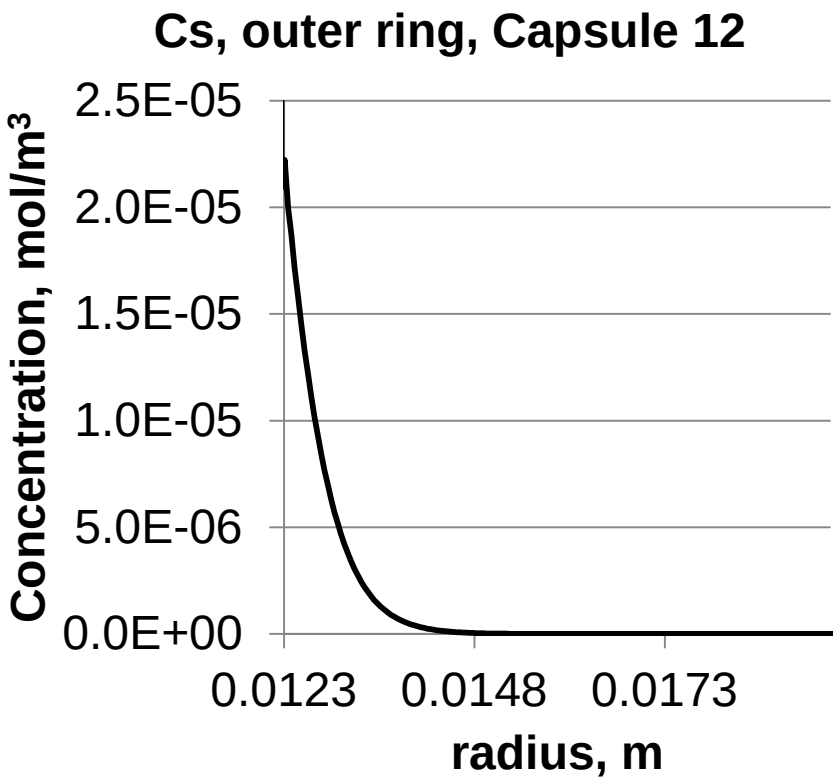
Determining Diffusion Coefficients: Method #2

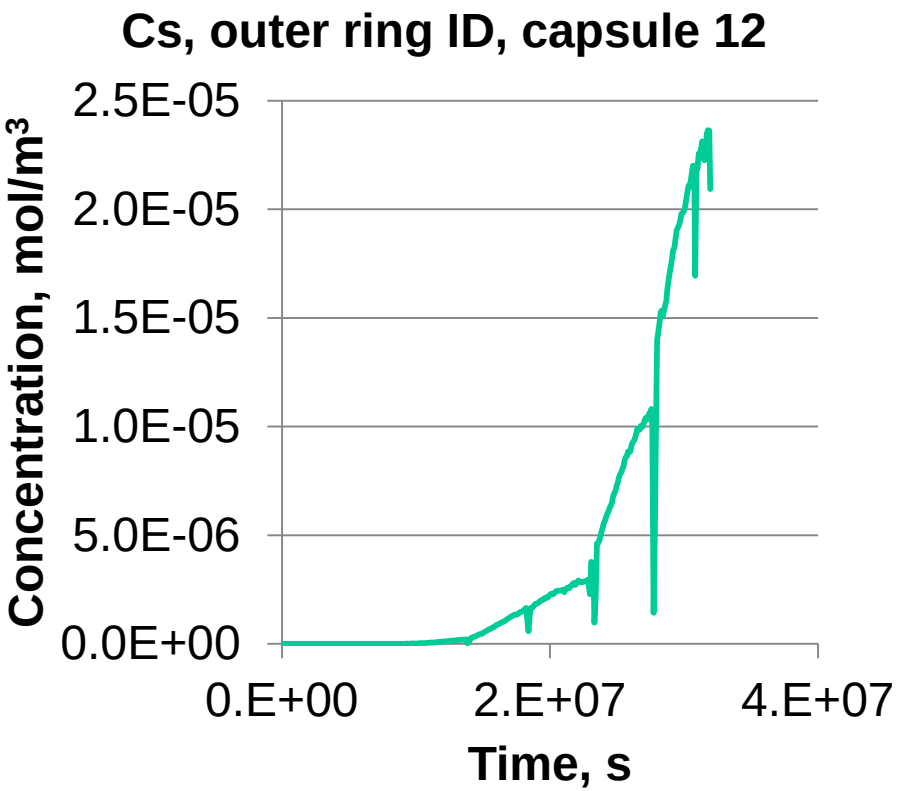
- If:
  - 1) we can approximate the cylindrical geometry as cartesian,
  - 2) a ring is effectively infinite, i.e. its thickness is large relative to a diffusion distance (~$(Dt)^{1/2}$), or the concentration at the outer diameter is ~0, and
  - 3) the concentration C_1 at the inner diameter is constant (i.e. it reaches a steady state), then
- The profile across a ring is given by $C(x,t) = C_1\left[1 - \mathrm{erf}\left(\frac{x}{2\sqrt{Dt}}\right)\right]$
- Since we can measure the concentration C_2 at some position x (and we know the time t) the diffusion coefficient can be calculated from

$$D = \frac{x^2}{4t\left(\mathrm{erf}^{-1}\left(1 - \frac{C_2}{C_1}\right)\right)^2}$$

Problems with Method #2

- Most of the rings are not effectively infinite, i.e. there is a non-zero concentration (and flux) at their outer diameter
- Those that are effectively infinite do not have constant concentrations at their inner diameter:





The General Problem

- AGR-3/4 is complicated:
  - There are multiple, coupled diffusion problems (both spherical particle layers and cylindrical rings)
  - Both temperatures and source terms vary in space and time
- It does not appear to be analytically tractable
- There are 16 material parameters related to diffusion and sorption in the rings (not including the fuel particles) that are required to model a capsule
  - Hand-tuning all of these does not appear to be a credible method of “fitting” these to measured data
- Suggestion (open for discussion): we can use heating tests to do controlled experiments to measure diffusion as a function of temperature

Measuring diffusion coefficients in heating tests

- Diffusion coefficients can be determined with heating tests in the following way:
  - 1) Separate a ring from the compact and other rings
  - 2) Measure the FP profile/inventory in the ring
  - 3) Heat to a constant temperature for a specified time
    - The boundary conditions must be controlled, e.g. fixed at zero
  - 4) Measure the new FP concentration profile/inventory, and/or the release from the ring during heating
- There is an analytical solution for this problem in 1D:

$$C(r,t)=\frac{\pi^2}{2}\sum_{n=1}^{\infty}\frac{\alpha_n^2 J_0^2(r_1\alpha_n)}{J_0^2(r_1\alpha_n)-J_0^2(r_2\alpha_n)}\exp\left(-D\alpha_n^2 t\right)U_0(r\alpha_n)\int_{r_1}^{r_2} rC(r,0)U_0(r\alpha_n)dr$$

- 2D analyses are feasible if necessary since there is only one parameter (the diffusion coefficient *D*) to vary

Summary

- A finite element model for fission product transport in AGR-3/4 has been developed in support of PIE planning
- The fission product concentration profiles that can be expected are complex, resulting from time and space-varying temperatures and mass sources, and coupling between multiple spherical and cylindrical layers
- As a result, there does not appear to be an appropriate analytical method for extracting diffusion or sorption parameters from direct measurements
- It is proposed that heating tests be used with some rings to measure diffusion coefficients in a more controlled way
- Measurements of FP concentration profiles from other capsules can still provide valuable *integral* data for comparison

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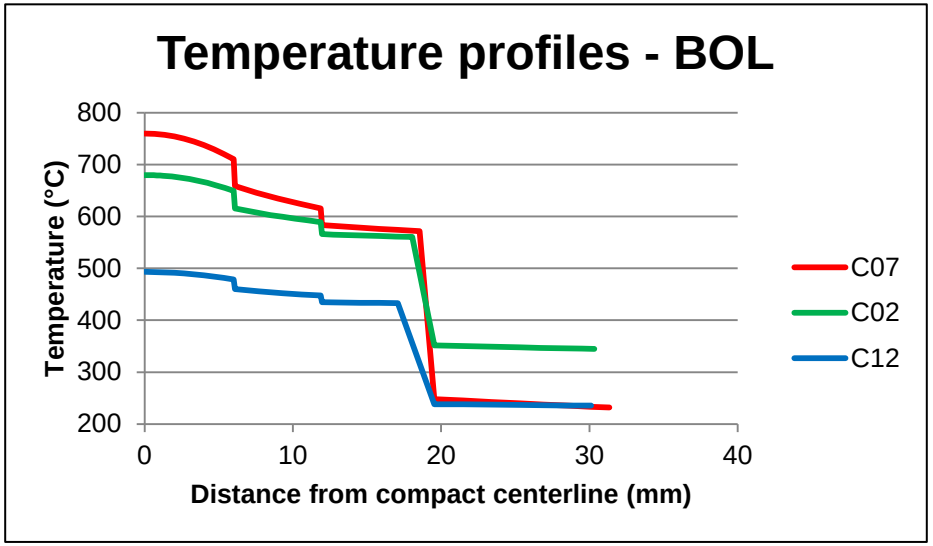
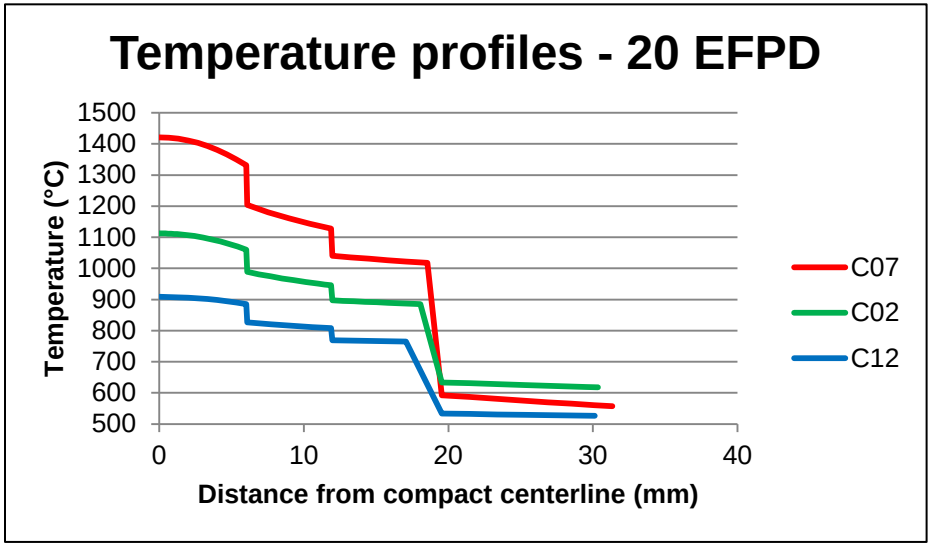
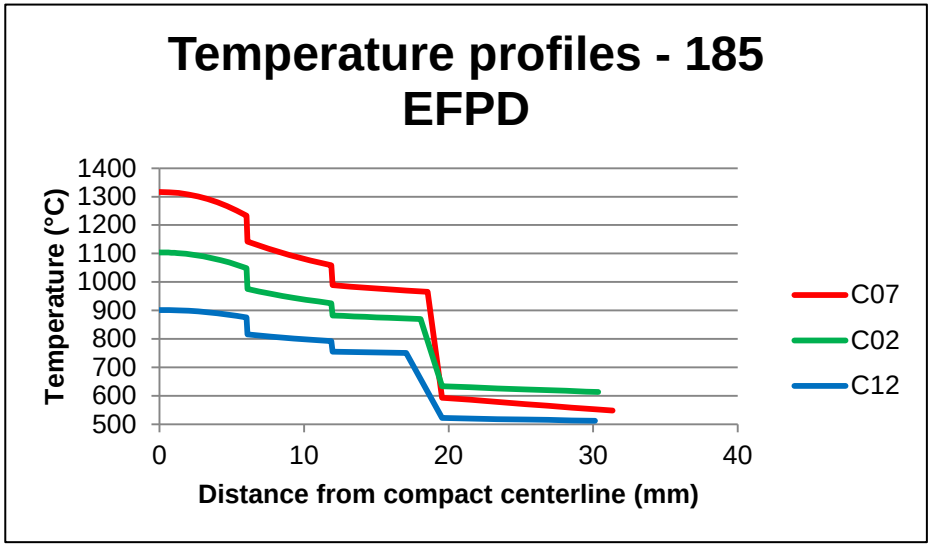
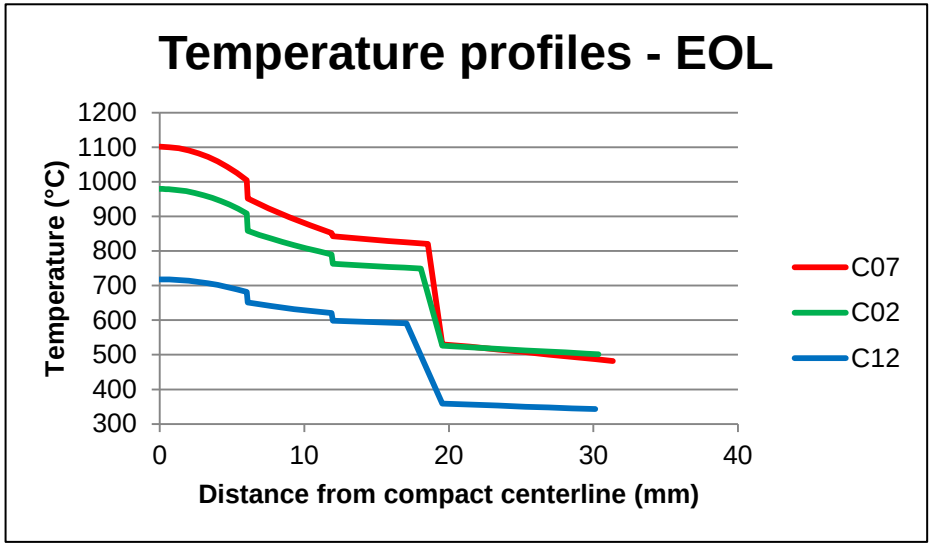
Radial Temperature Profiles









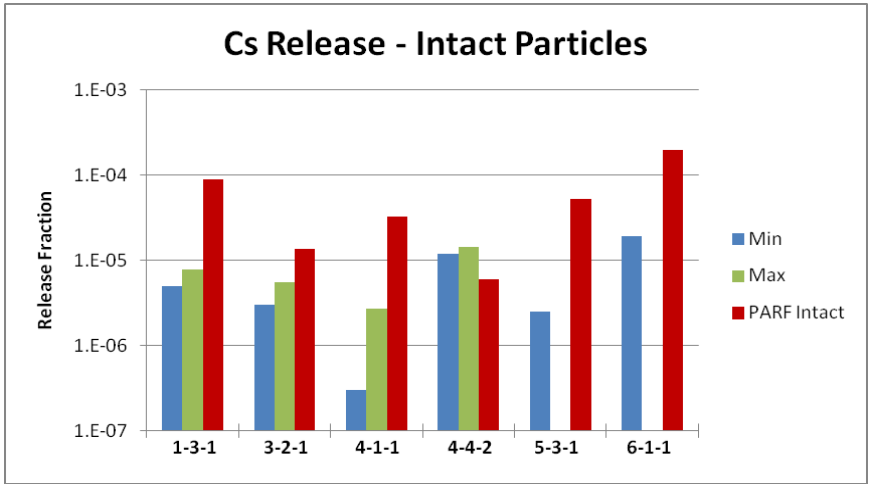
PARFUME Source Term - Summary

Caps	Level	Ag		Cs		Sr	
		Driver	DTF	Driver	DTF	Driver	DTF
12	4	2.5E+12	4.5E+14	2.2E+14	2.0E+16	2.0E+14	3.3E+10
12	3	2.8E+12	5.9E+14	2.6E+14	2.9E+16	2.2E+14	3.7E+10
12	2	3.0E+12	6.8E+14	2.8E+14	3.4E+16	2.3E+14	4.0E+10
12	1	3.3E+12	6.6E+14	2.8E+14	2.9E+16	2.4E+14	4.0E+10
11	4	3.1E+16	1.7E+15	2.0E+15	1.6E+17	3.1E+15	1.9E+14
11	3	5.5E+16	1.9E+15	8.2E+15	1.7E+17	1.6E+16	5.9E+14
11	2	6.2E+16	2.0E+15	1.1E+16	1.8E+17	2.2E+16	7.0E+14
11	1	4.0E+16	2.1E+15	3.3E+15	1.8E+17	5.0E+15	2.0E+14
10	4	3.8E+16	2.8E+15	2.8E+15	2.2E+17	3.7E+15	1.8E+14
10	3	6.4E+16	2.9E+15	1.0E+16	2.2E+17	1.7E+16	5.6E+14
10	2	6.8E+16	3.0E+15	1.2E+16	2.3E+17	2.0E+16	6.2E+14
10	1	4.4E+16	3.0E+15	4.0E+15	2.3E+17	4.9E+15	2.0E+14
9	4	2.1E+15	3.5E+15	8.3E+14	2.3E+17	4.9E+14	6.5E+12
9	3	7.0E+15	3.6E+15	8.8E+14	2.5E+17	5.4E+14	2.1E+13
9	2	7.5E+15	3.7E+15	8.9E+14	2.5E+17	5.5E+14	2.3E+13
9	1	2.0E+15	3.7E+15	8.6E+14	2.3E+17	5.1E+14	5.3E+12
8	4	7.4E+16	4.0E+15	7.7E+15	2.8E+17	1.5E+16	5.9E+14
8	3	1.1E+17	4.0E+15	2.4E+16	2.8E+17	5.1E+16	1.4E+15
8	2	1.1E+17	4.0E+15	2.5E+16	2.8E+17	5.2E+16	1.4E+15
8	1	7.1E+16	4.0E+15	7.4E+15	2.8E+17	1.4E+16	5.2E+14
7	4	1.8E+17	4.2E+15	1.4E+17	2.9E+17	2.3E+17	3.3E+15
7	3	2.2E+17	4.2E+15	2.6E+17	2.9E+17	5.3E+17	6.8E+15
7	2	2.2E+17	4.2E+15	2.6E+17	2.9E+17	5.2E+17	6.7E+15
7	1	1.7E+17	4.2E+15	1.2E+17	2.9E+17	1.9E+17	2.8E+15

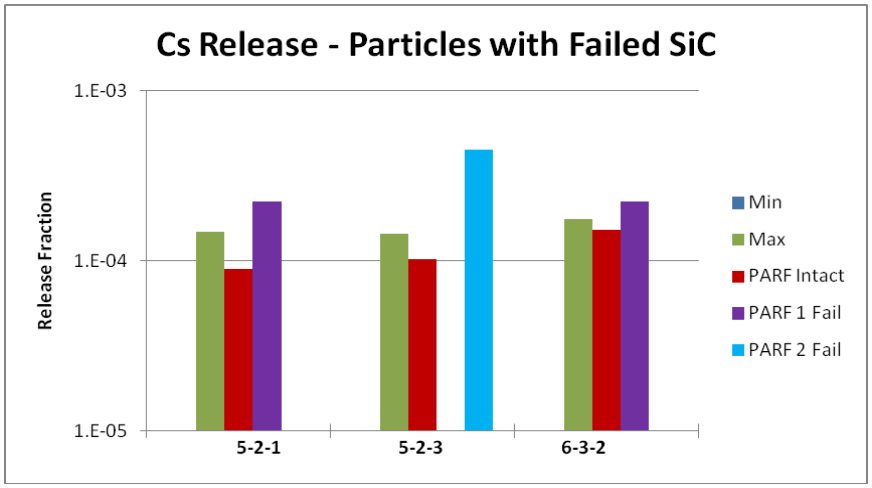
Caps	Level	Ag		Cs		Sr	
		Driver	DTF	Driver	DTF	Driver	DTF
6	4	4.9E+15	4.3E+15	9.6E+14	2.8E+17	5.6E+14	1.5E+13
6	3	1.3E+16	4.3E+15	1.1E+15	2.9E+17	6.5E+14	4.0E+13
6	2	1.2E+16	4.3E+15	1.1E+15	2.8E+17	6.4E+14	3.8E+13
6	1	2.9E+15	4.3E+15	9.5E+14	2.6E+17	5.5E+14	7.6E+12
5	4	8.6E+14	4.1E+15	9.1E+14	2.3E+17	5.4E+14	3.1E+12
5	3	4.7E+15	4.1E+15	9.4E+14	2.6E+17	5.7E+14	1.5E+13
5	2	4.3E+15	4.1E+15	9.3E+14	2.6E+17	5.7E+14	1.4E+13
5	1	4.0E+14	3.9E+15	8.9E+14	2.2E+17	5.3E+14	1.6E+12
4	4	3.5E+15	3.9E+15	9.0E+14	2.5E+17	5.3E+14	1.2E+13
4	3	9.9E+15	3.9E+15	9.7E+14	2.6E+17	6.0E+14	3.3E+13
4	2	8.5E+15	3.8E+15	9.3E+14	2.6E+17	5.7E+14	2.8E+13
4	1	1.3E+15	3.9E+15	8.6E+14	2.3E+17	5.1E+14	4.1E+12
3	4	3.2E+16	3.3E+15	2.5E+15	2.4E+17	2.6E+15	1.2E+14
3	3	5.0E+16	3.3E+15	6.4E+15	2.4E+17	8.4E+15	3.2E+14
3	2	4.3E+16	3.2E+15	4.7E+15	2.4E+17	5.9E+15	2.5E+14
3	1	1.9E+16	3.0E+15	1.1E+15	2.2E+17	9.7E+14	4.8E+13
2	4	2.6E+15	2.5E+15	6.6E+14	1.8E+17	4.1E+14	5.3E+12
2	3	4.4E+15	2.3E+15	6.5E+14	1.8E+17	4.1E+14	1.0E+13
2	2	2.9E+15	2.2E+15	6.2E+14	1.7E+17	3.9E+14	6.4E+12
2	1	4.9E+14	2.0E+15	5.8E+14	1.4E+17	3.6E+14	1.1E+12
1	4	4.8E+12	1.1E+15	3.9E+14	6.4E+16	2.8E+14	8.5E+10
1	3	1.3E+13	1.1E+15	3.8E+14	7.4E+16	2.6E+14	1.8E+11
1	2	5.3E+12	9.3E+14	3.4E+14	6.1E+16	2.4E+14	1.0E+11
1	1	3.0E+12	6.8E+14	2.9E+14	3.4E+16	2.3E+14	4.1E+10

Per AGR-1, Cs from Driver Fuel is over-predicted by up to 100 & Sr by up to 400

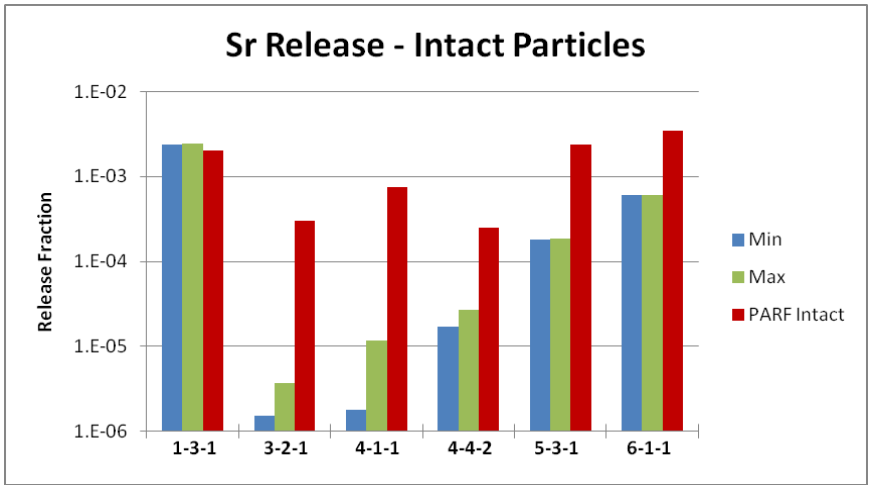
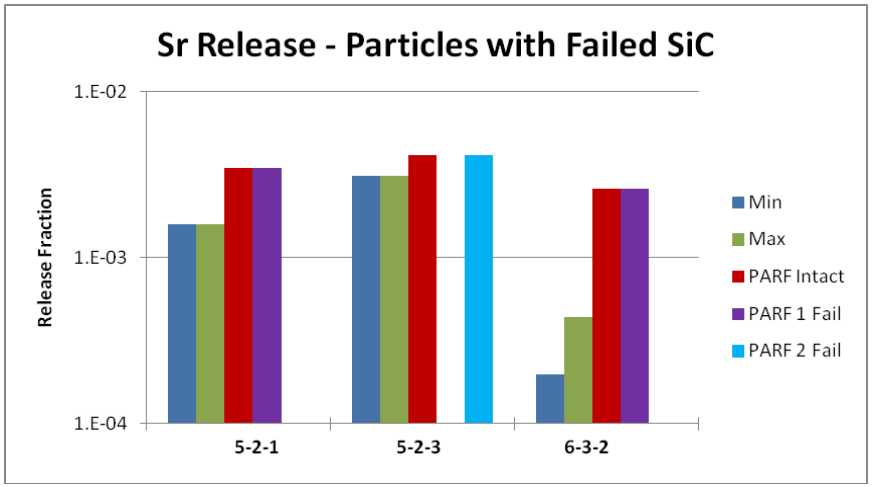
PARFUME AGR-1 over-predictions - Cs



Intact x2 to x100
Failed x1.3 to x3



PARFUME AGR-1 over-predictions - Sr



Up to x400