AGR-3/4 Post-Irradiation Examinations

John D. Stempien, Ph.D.

AGR TRISO Fuels PIE Technical Lead

Presented at the Gas-Cooled Reactor Program Review Meeting

June 18, 2019 at Idaho National Laboratory

ADVANCED REACTOR TECHNOLOGIES

Outline

- Introduction to AGR-3/4 experiment
- AGR-3/4 experiment mass balances
- Update on physical sampling of AGR-3/4 graphite rings for fission product profiles
- Fuel compact radial deconsolidation status
- Summary of completed and planned work

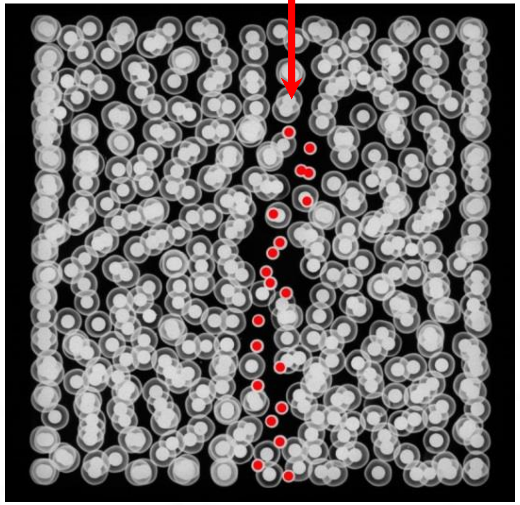
AGR-3/4 Experiment Goals

- Observe metallic fission product (e.g. Ag, Cs, Eu, and Sr) transport within graphitic matrix and nuclear grade graphites (IG-110 and PCEA)
- Measure fission product inventories and spatial distributions within fuel compacts and graphite
- Determine diffusion coefficients of metallic fission products within graphitic materials

AGR-3/4 Compacts

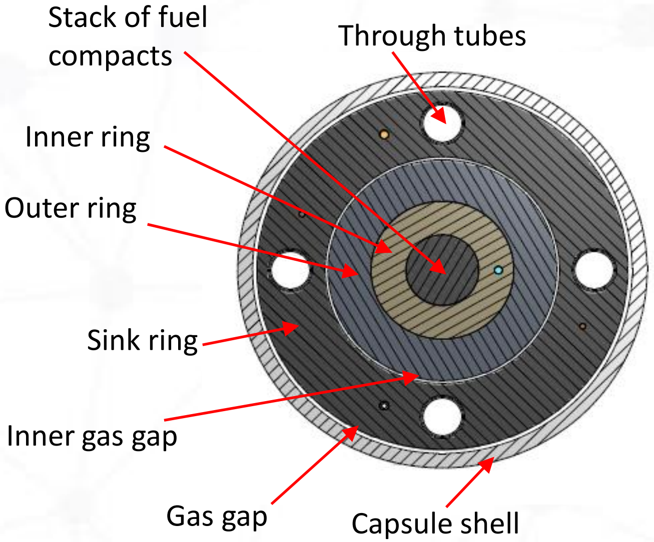
- Each compact is 0.5 in. tall, 0.5 in. diameter
- Approximately 1872 “driver” TRISO particles similar to AGR-1 Baseline fuel
- 20 designed-to-fail (DTF) particles per compact, coated only with 20μm-thick pyrocarbon layer
- DTF provide known source of fission products for transport through compact and rings
- 4 compacts per capsule, 80 DTF particles per irradiation capsule

X-ray showing 20 DTF particles in center of compact

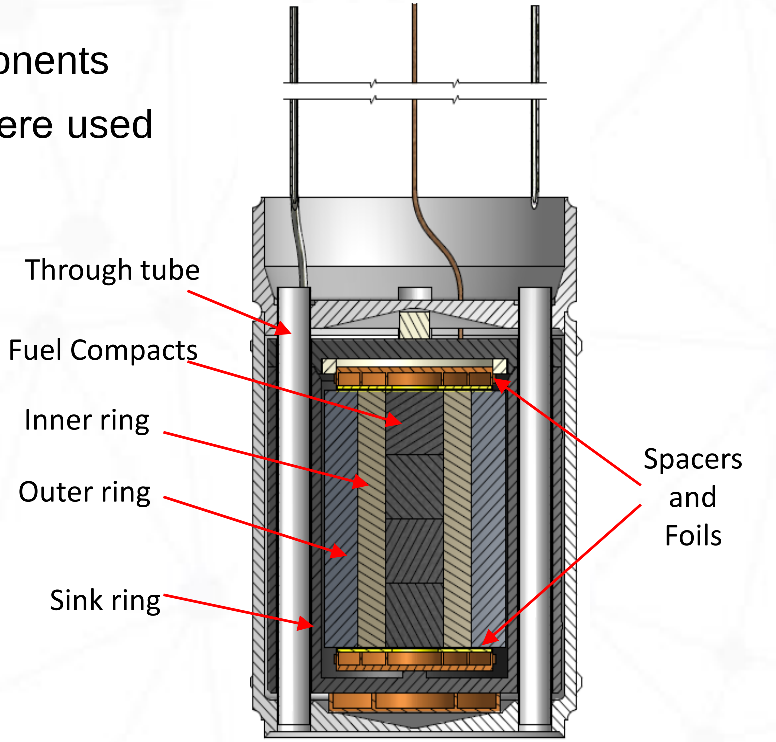


AGR-3/4 Irradiation Capsule and Components

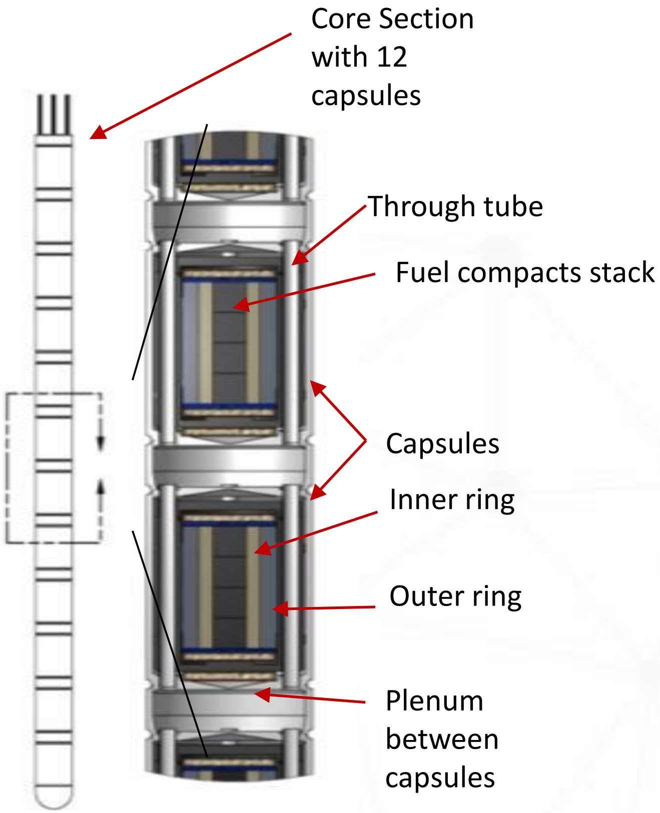
- Four compacts in center of “inner ring” (graphitic matrix, IG-110, or PCEA)
- Inner ring sits within “outer ring” (PCEA or IG-110)
- Outer ring held within cold “sink ring” (PCEA)
- Fission products may transport through components
- Standard capsules and sealed “fuel bodies” were used



AGR-3/4 capsule cross section

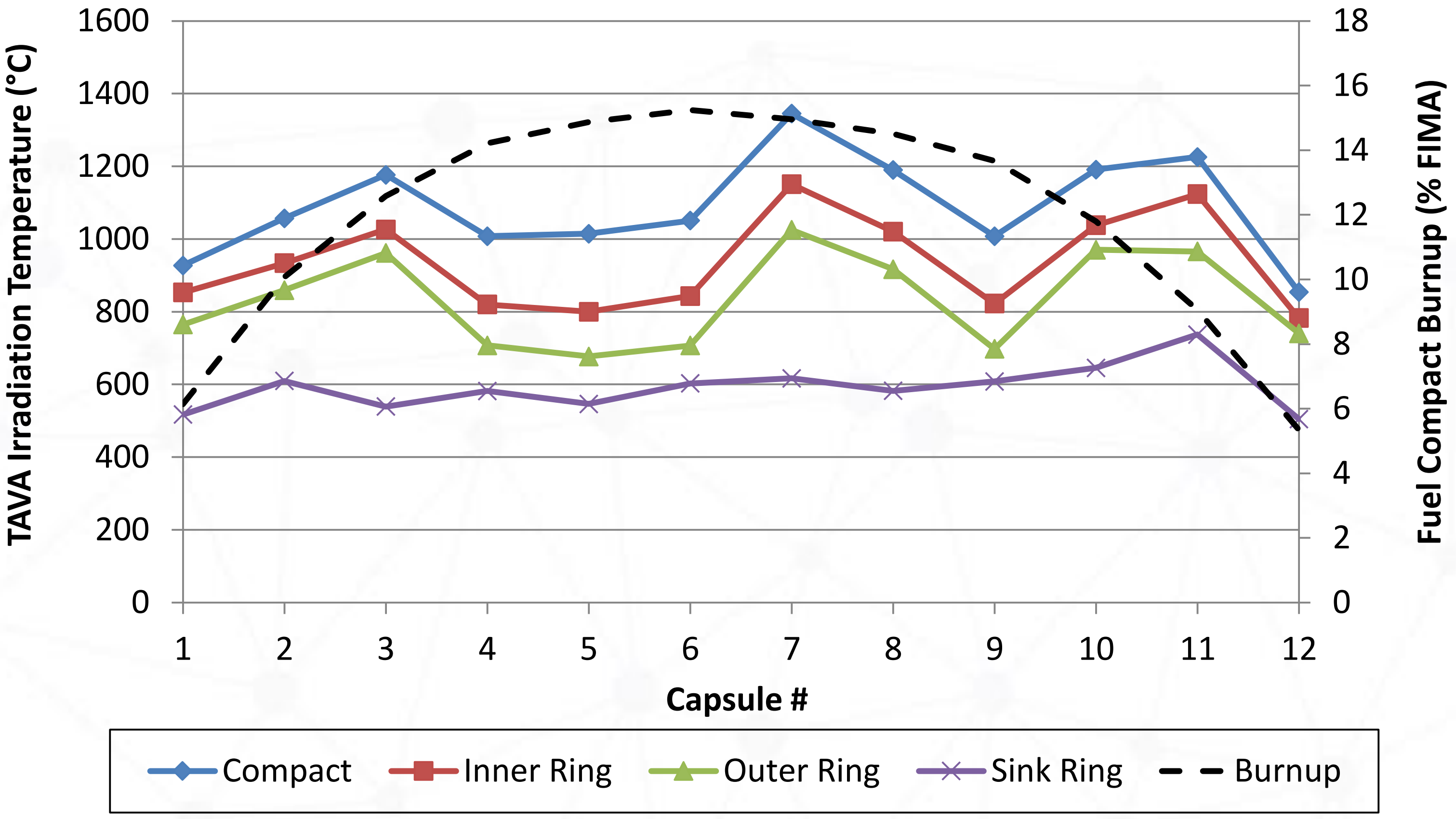


Axial cutaway of an AGR-3/4 capsule



Irradiation test train

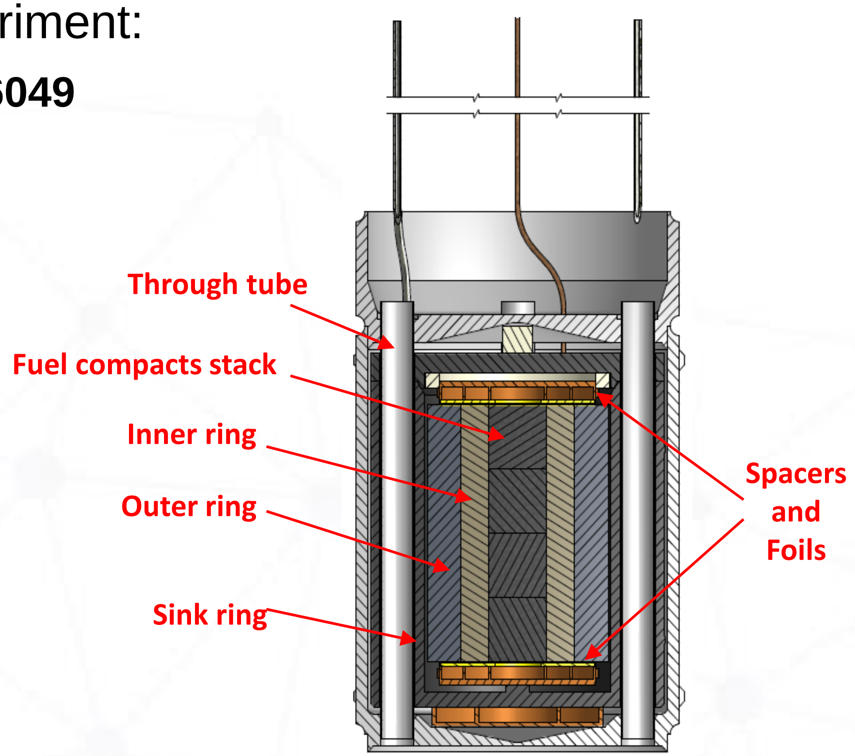
AGR-3/4 Irradiation Temperatures and Fuel Burnup



TAVA = time-averaged, volume-averaged
FIMA = fissions per initial metal atom

AGR-3/4 Components Fission Product Measurements

- Measure total fission product inventories throughout experiment:
  - **Completed AGR-3/4 Mass Balance Report: INL/EXT-18-46049**
    - Through tubes
    - Inner/outer ring
    - Sink ring
    - Spacers/foils
  - Compacts
- Measure fission product radial concentration profiles:
  - Compacts
  - Inner and outer rings

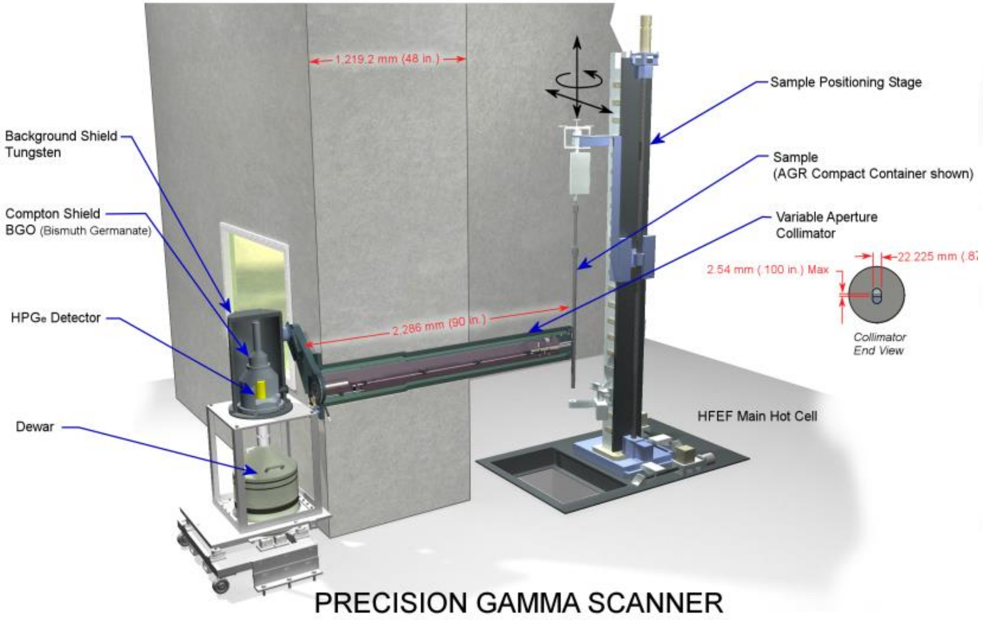


Axial cutaway of an AGR-3/4 capsule

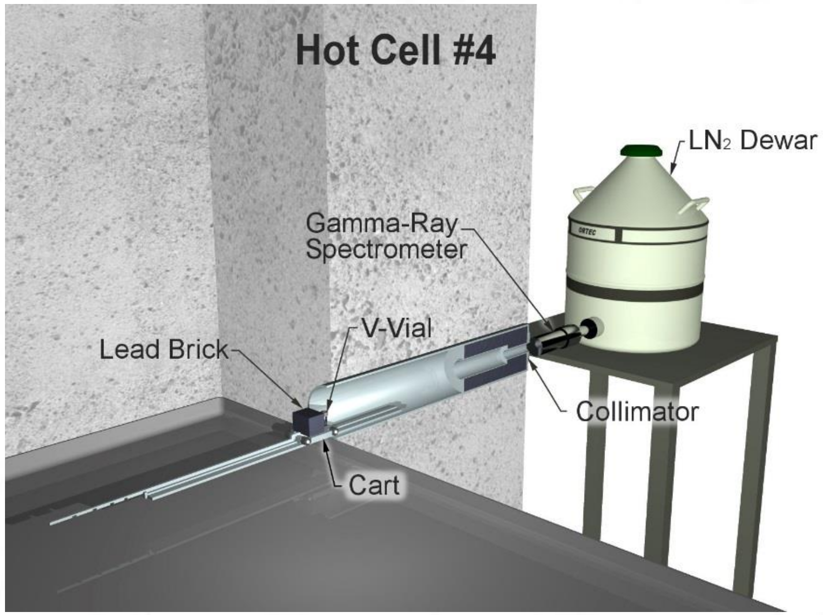
Gamma Counting for FP Inventories

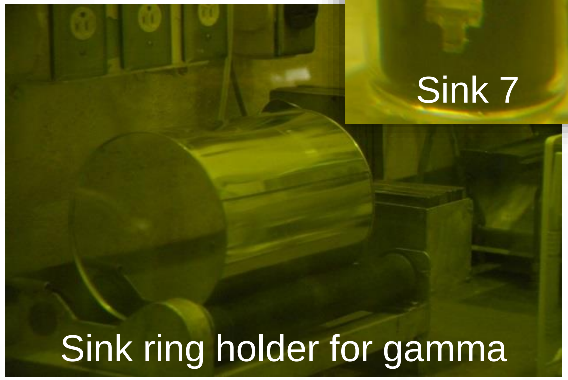
- Counting primarily for Ag-110m, Cs-134/137, and Eu-154/155 (also Sb-125, Ce/Pr-144, Ru/Rh-106)

PGS at HFEF:
compacts and **inner/outer rings** COMPLETE



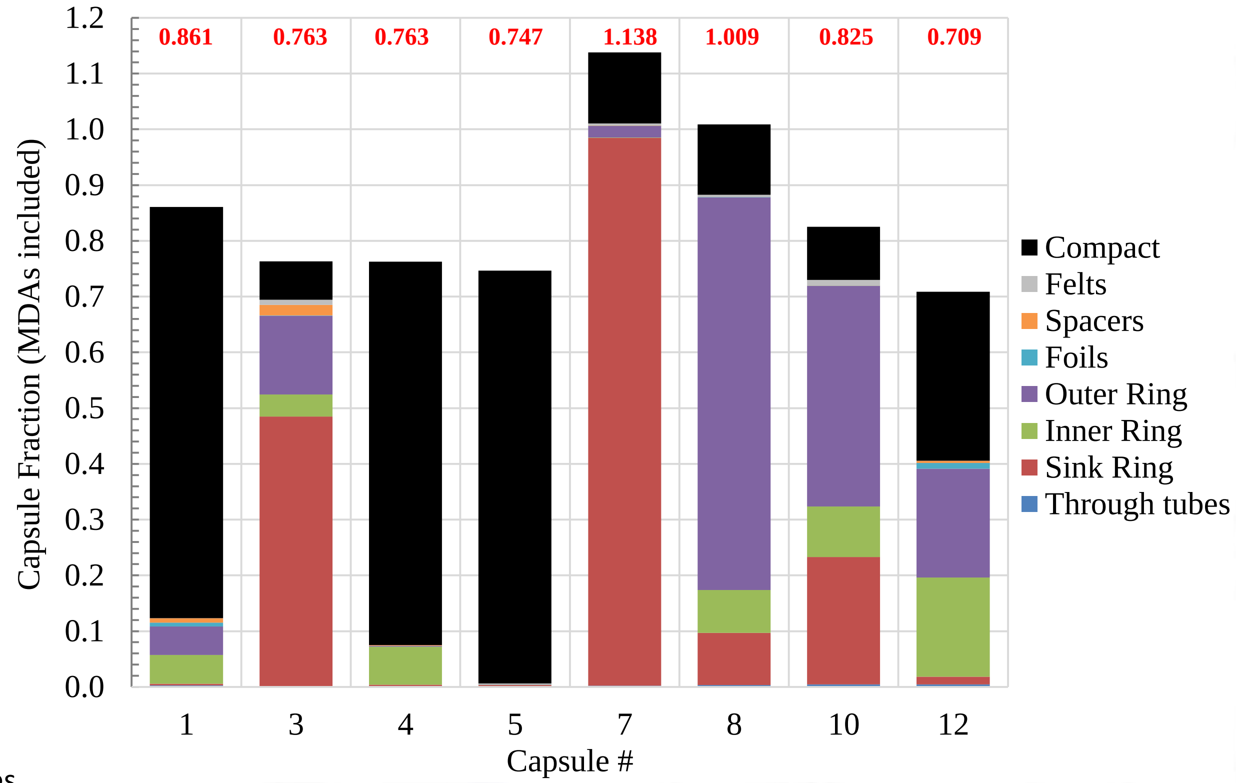
Spectrometer at AL: **foils** and **sink** rings COMPLETE,
second count of **spacers** COMPLETE
(through tubes COMPLETE)





Ag-110m Mass Balance in AGR-3/4 Irradiation Test Train

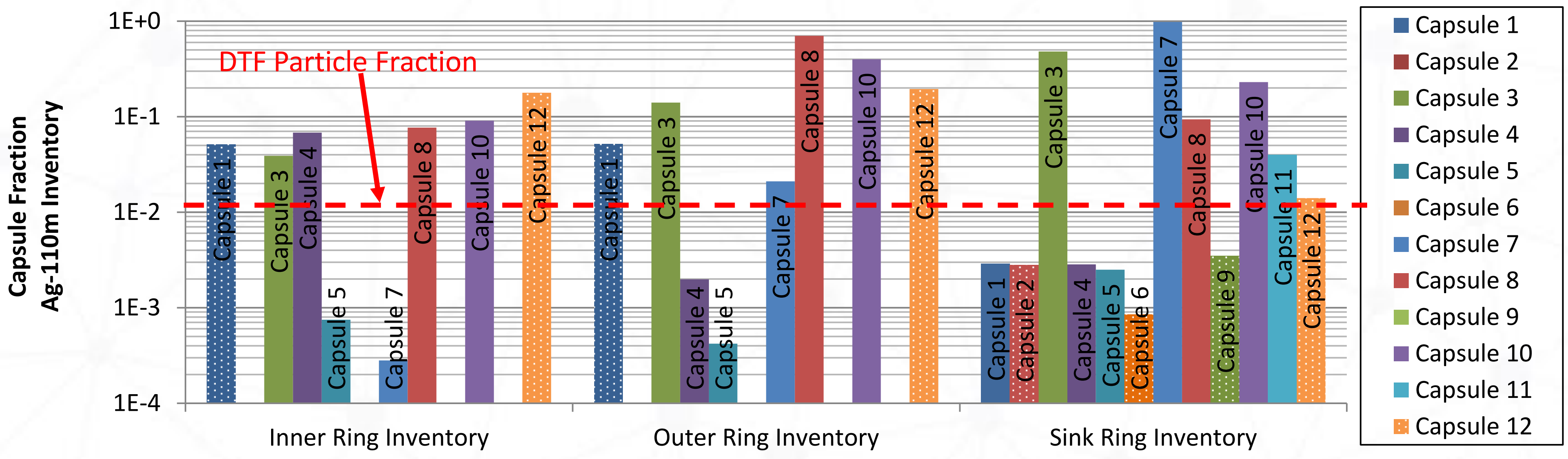
- Significant transport of γ-emitting Ag-110m occurs, leads to some observations not seen for other isotopes
- Measure fission products in each part of the capsule
- Sum all non-fuel components to get total release from fuel compacts and compare to physics calculations
- Agreement of measured with predicted is comparable to AGR-1 and AGR-2



Showing only disassembled capsules, not fuel bodies

Ag-110m Ring Inventories

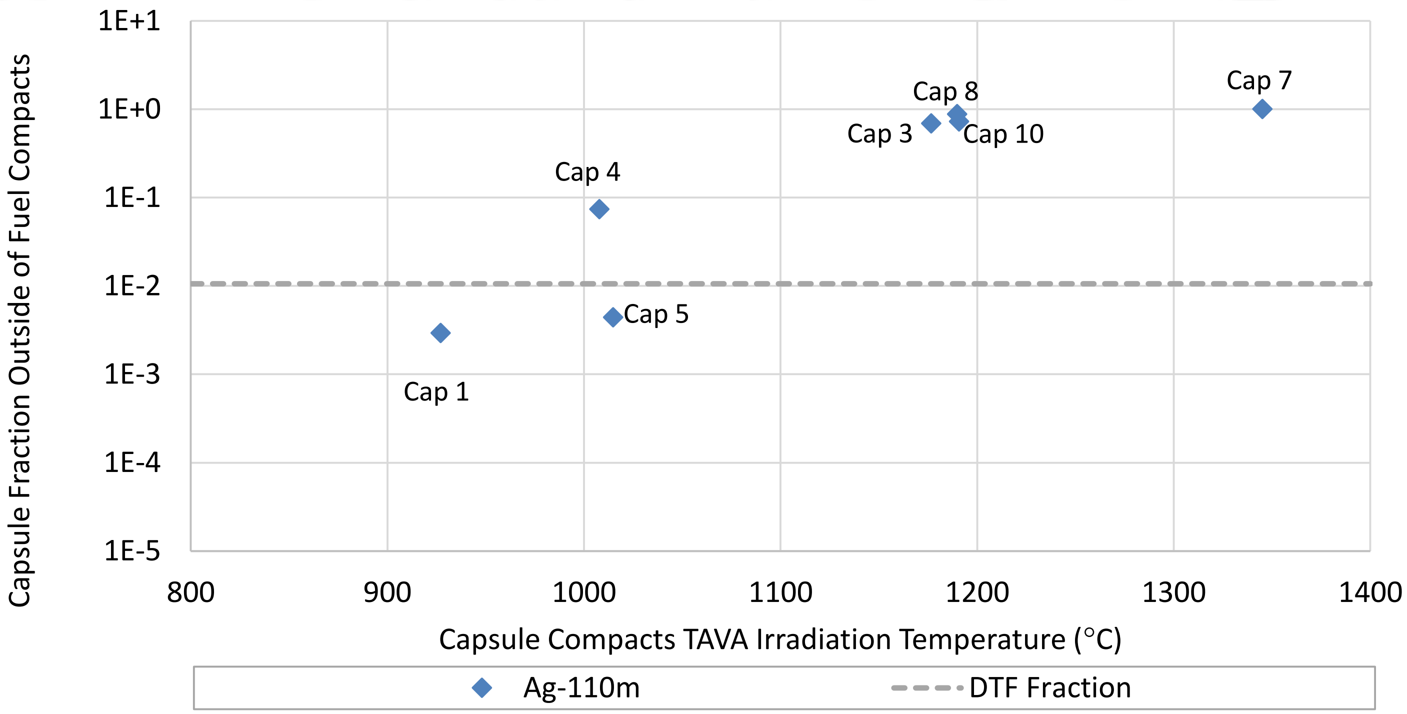
- Ring inventories exceed DTF
- Evidence of release through intact TRISO coatings



Bars with patterned fill indicate values derived from minimum detectable activity (MDA).

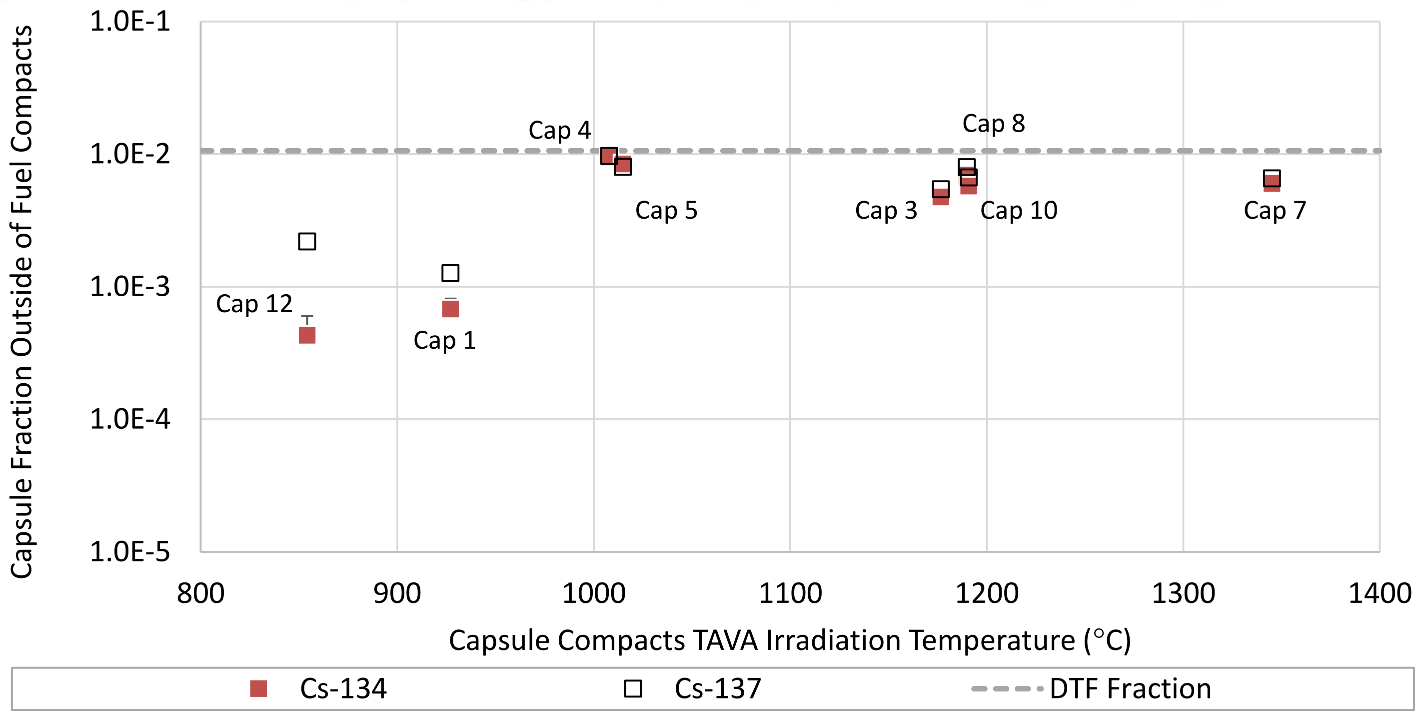
Ag-110m Release Versus Temperature

- Release increases with temp, 70-100% compact release at T ≥1200°C
- Exceeds DTF fraction
- Release through intact TRISO means driver particles contribute to release



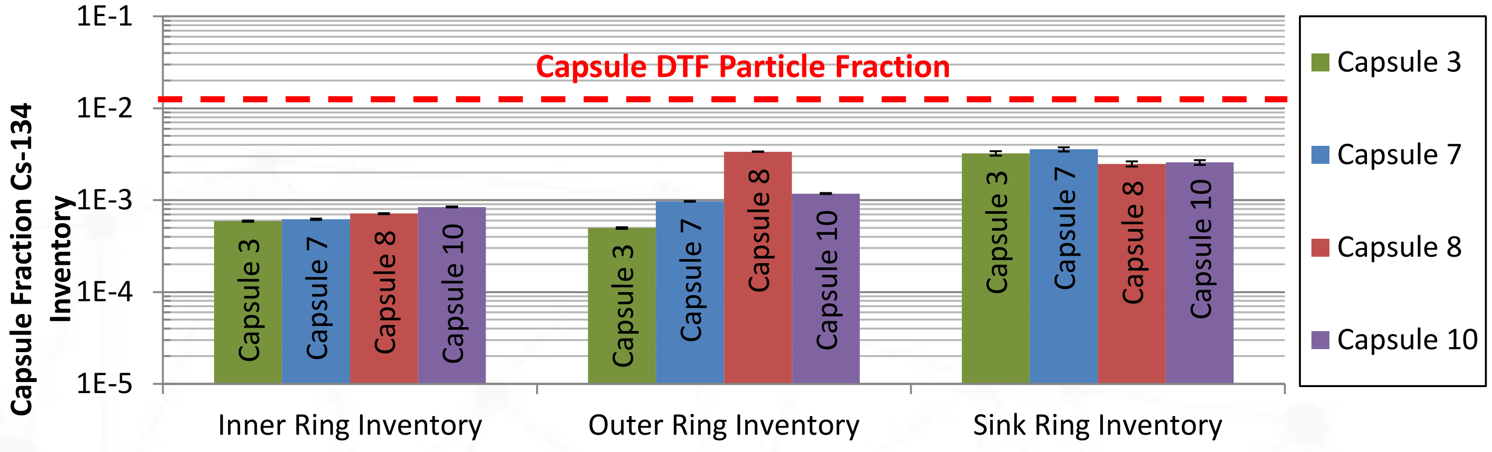
Mass Balance of Cs-134 and Cs-137 Outside of Fuel Compacts

- Cs-134 vs Cs-137: similar (Cs-137 biased high in low burnup Capsules 1 and 12)
- Cs release from compacts is 55-95% of the DTF inventory for T ≥ 1000°C

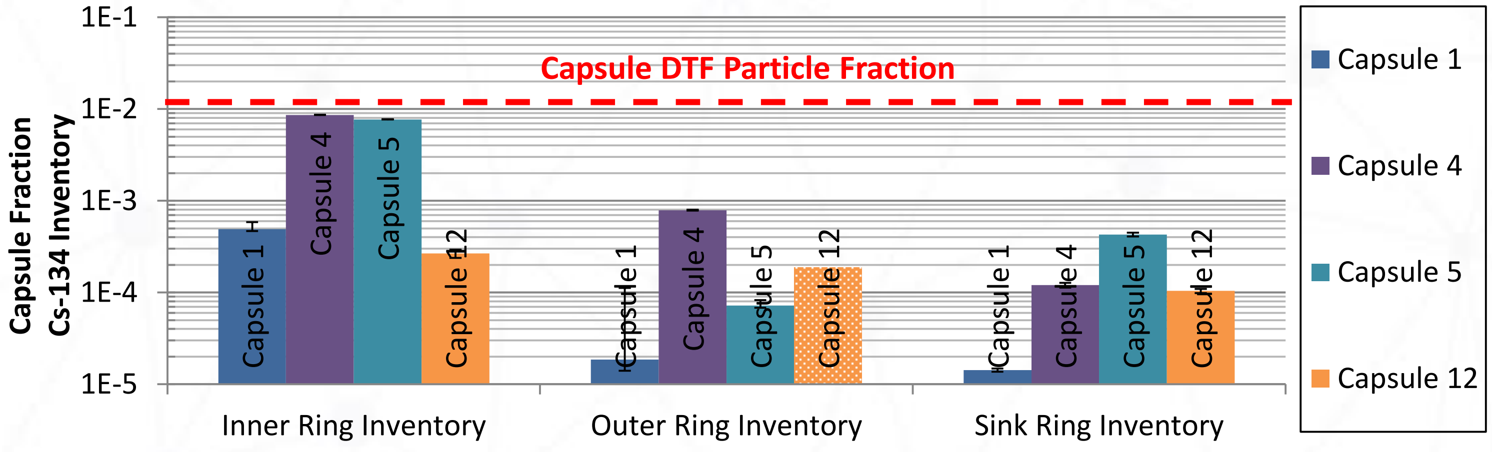


Cs-134 Ring Inventories

- **Hotter capsules (1177-1345°C):** inventories increase from inner ring to sink ring

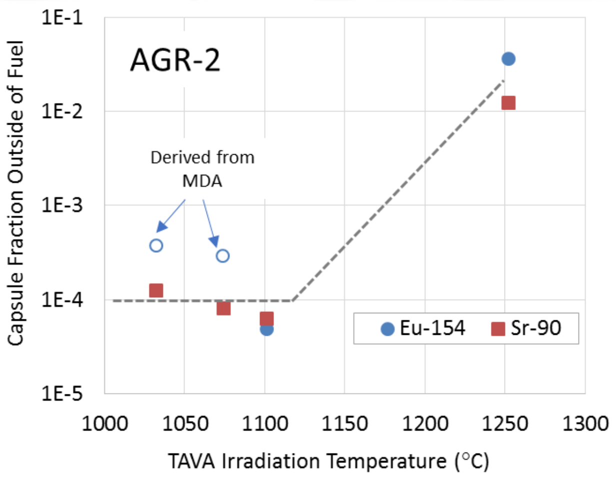


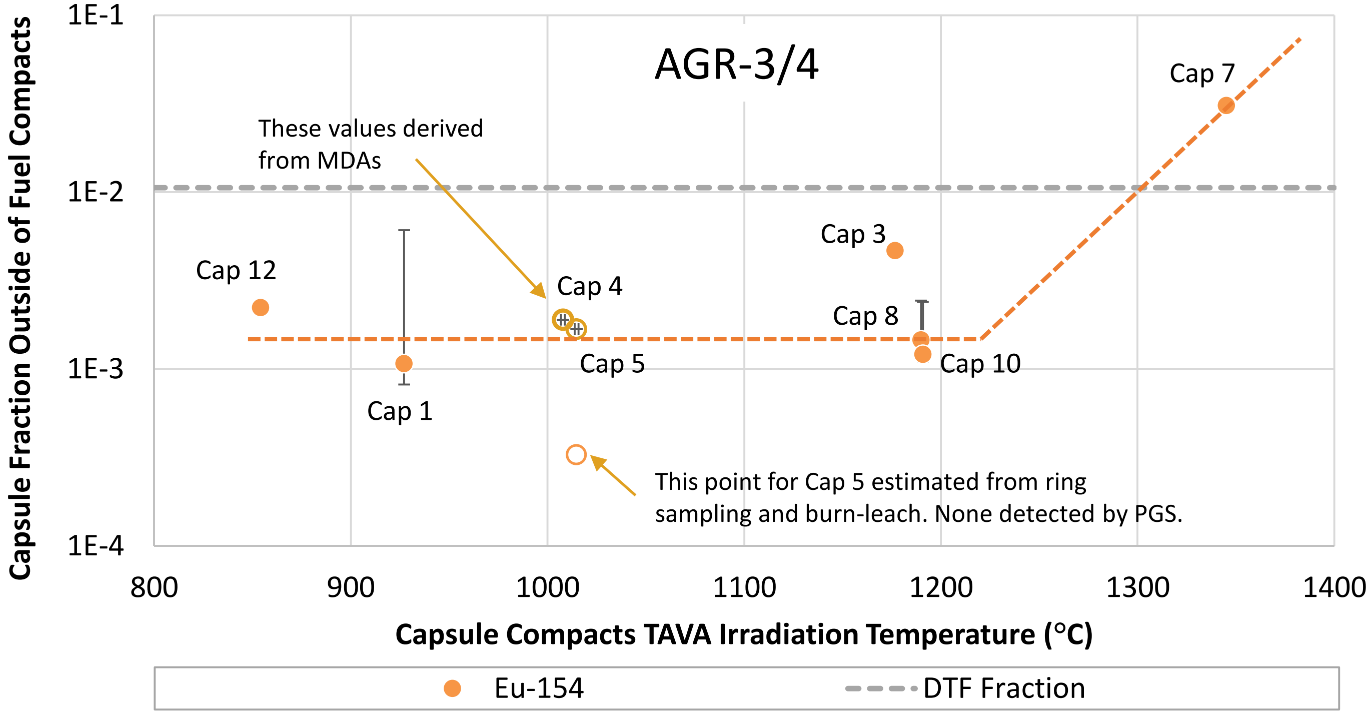
- **Cooler capsules (854-1015°C):** inventories decrease from inner ring to sink ring



Mass Balance of Eu-154 Outside of Fuel Compacts

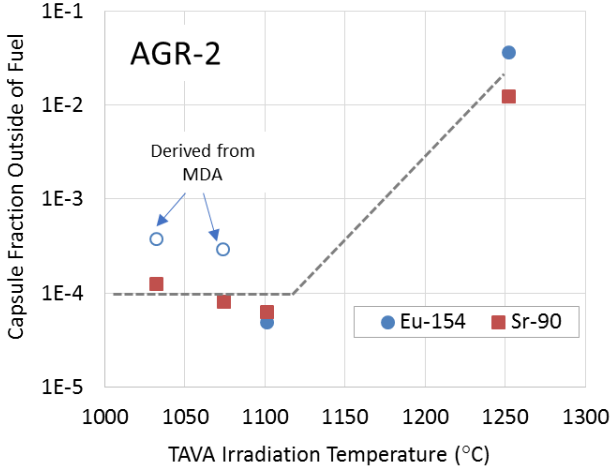
- Capsule 7 (1345°C): release > DTF fraction means release through intact driver particles.
- T < 1200°C: Eu-154 well-retained within fuel.
- Possible accelerated Eu-154 release starting above ~1200°C:
  - T < 1200°C: Caps 1, 8, 10, 12 releases 1E-3 to 2E-3
  - T = 1345°C (Cap 7): 3E-2
  - Similar observation in AGR-2

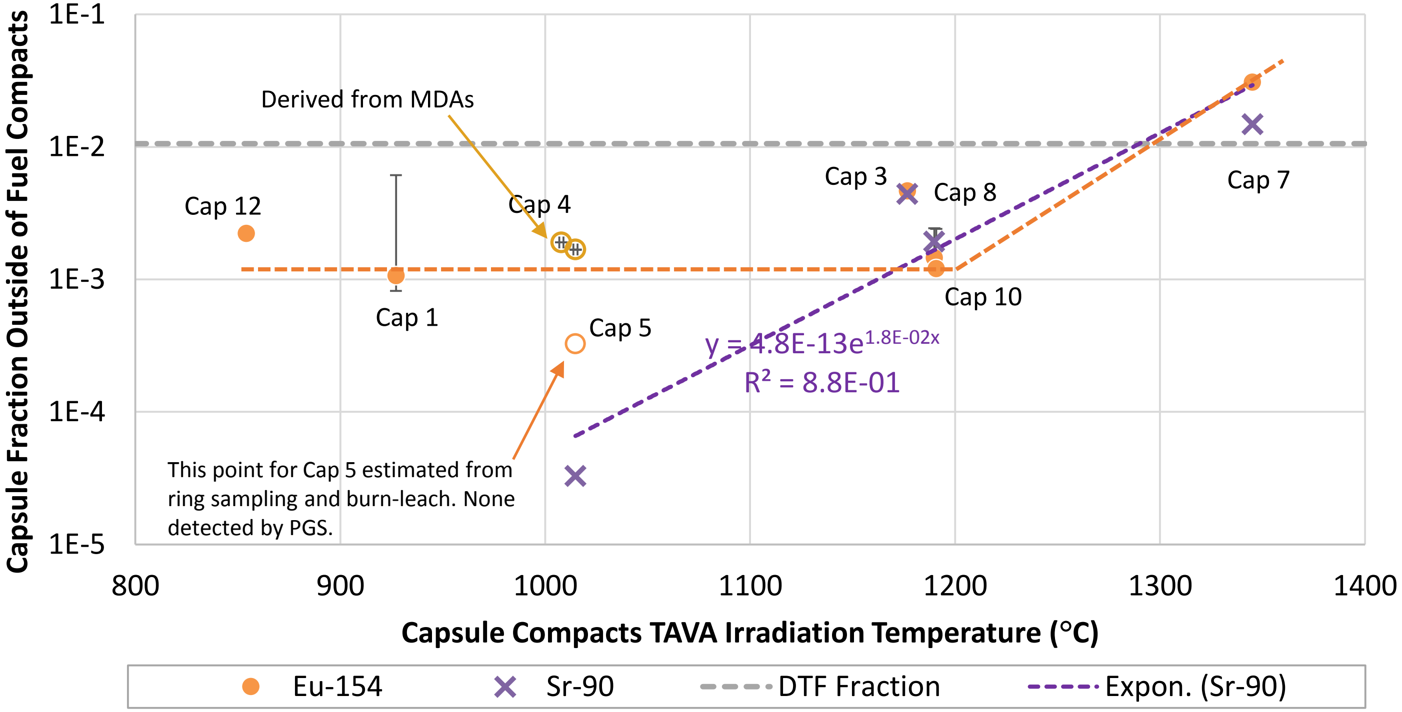




Mass Balance of Sr-90 and Eu-154 Outside of Fuel Compacts

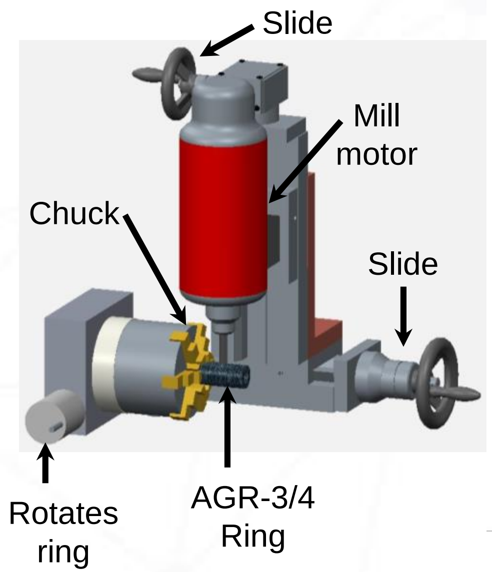
- Completed Sr-90 balances for Capsules 3, 5, 7, and 8 based on ring sampling approximations
- Sr-90 and Eu-154 releases similar in Capsules 3, 8, and 7
- Driver particle release apparent for Capsule 7
- Sr-90 exponential dependence with temperature from ~1000 to 1350°C
- Both Sr-90 and Eu-154 may have similar temp threshold for more rapid release



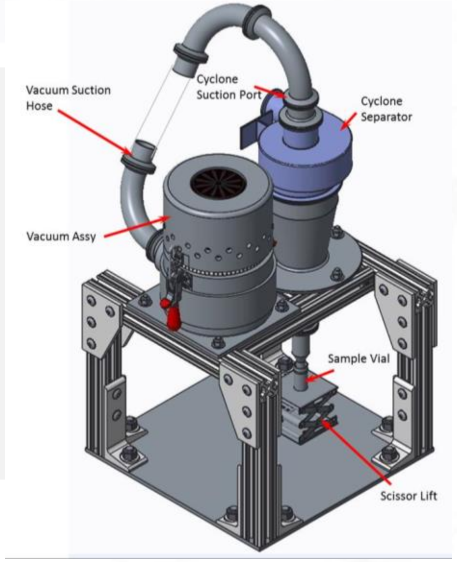


Physical Ring Sampling for FP Concentration Profiles

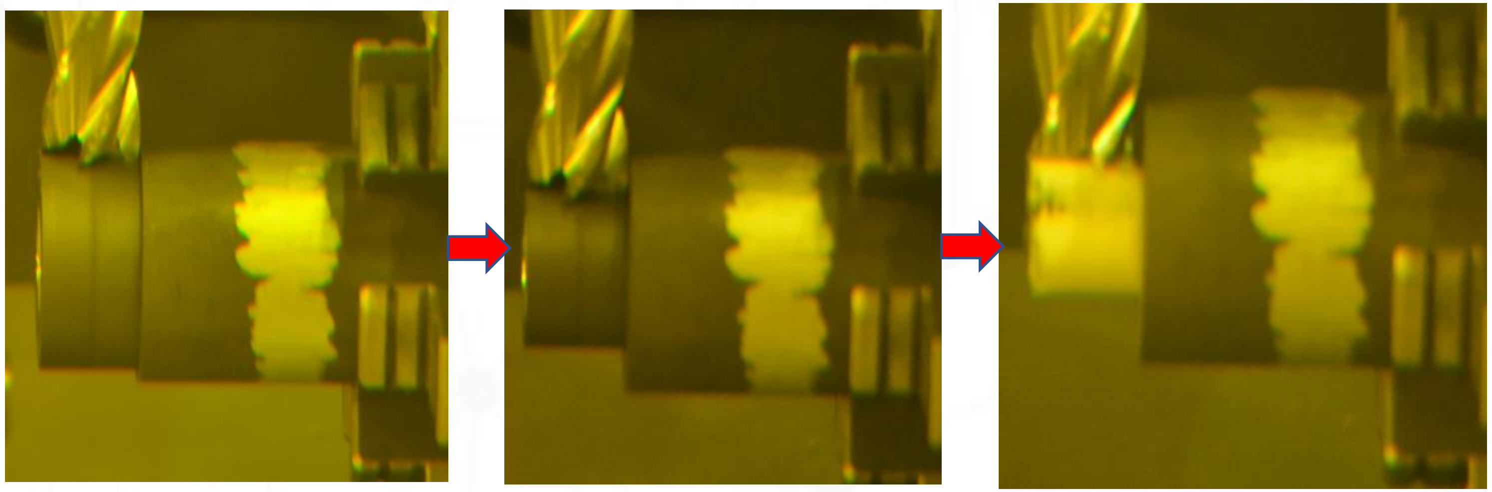
- Machining samples from the graphite rings to measure fission product radial profiles within rings
  - Progressively remove radial segments from rings at one or two axial locations
  - Collected material is gamma scanned and burn-leached for Sr-90 analysis
  - Refine models, compare to PGS, and derive transport parameters (i.e. diffusion coefficients) for FPs in graphite



Material Removal



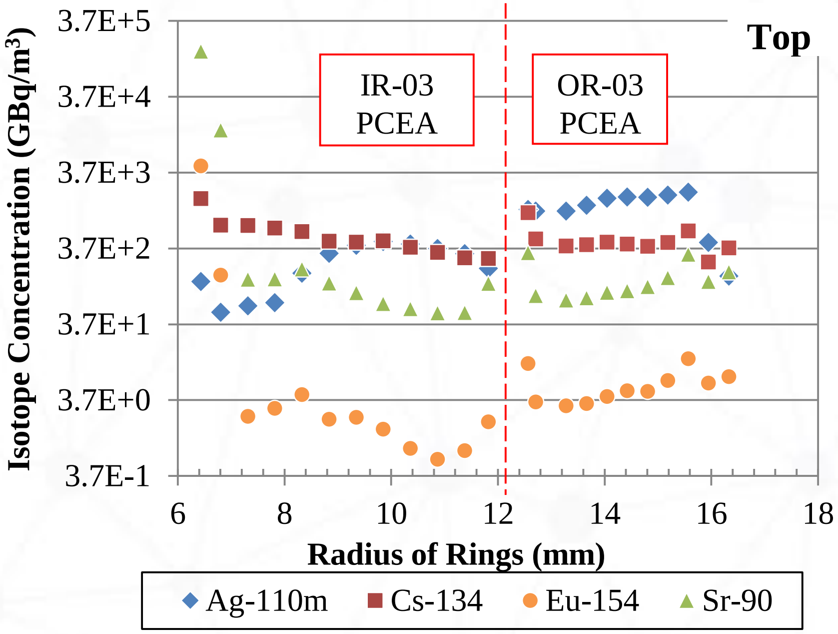
Fines Collection

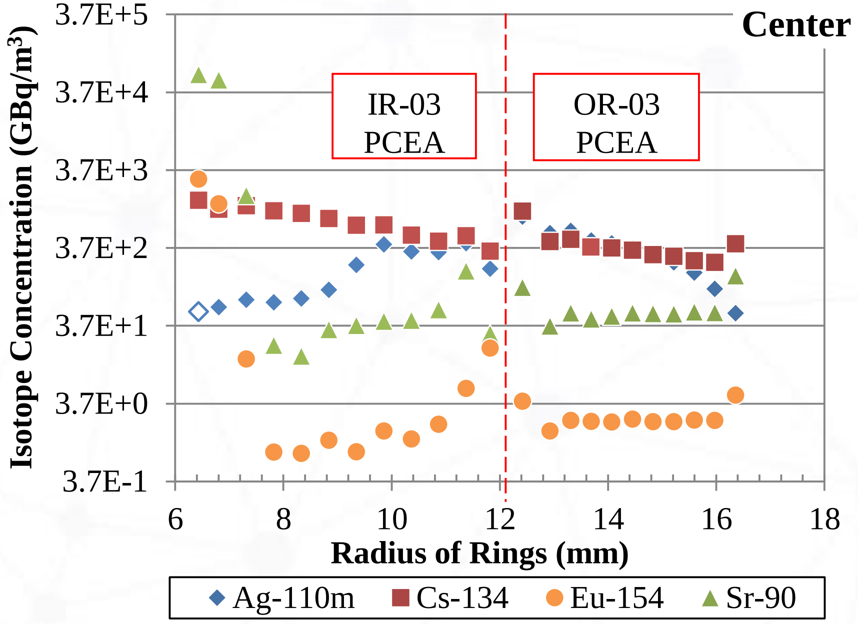


Images of IR-08 at beginning, middle, and end of sampling

Capsule 3 Inner and Outer Ring Concentration Profiles

- Compacts TAVA temp: 1177°C
- Inner ring:
  - PCEA
  - TAVA temp: 1026°C
- Outer ring:
  - PCEA
  - TAVA temp: 962°C

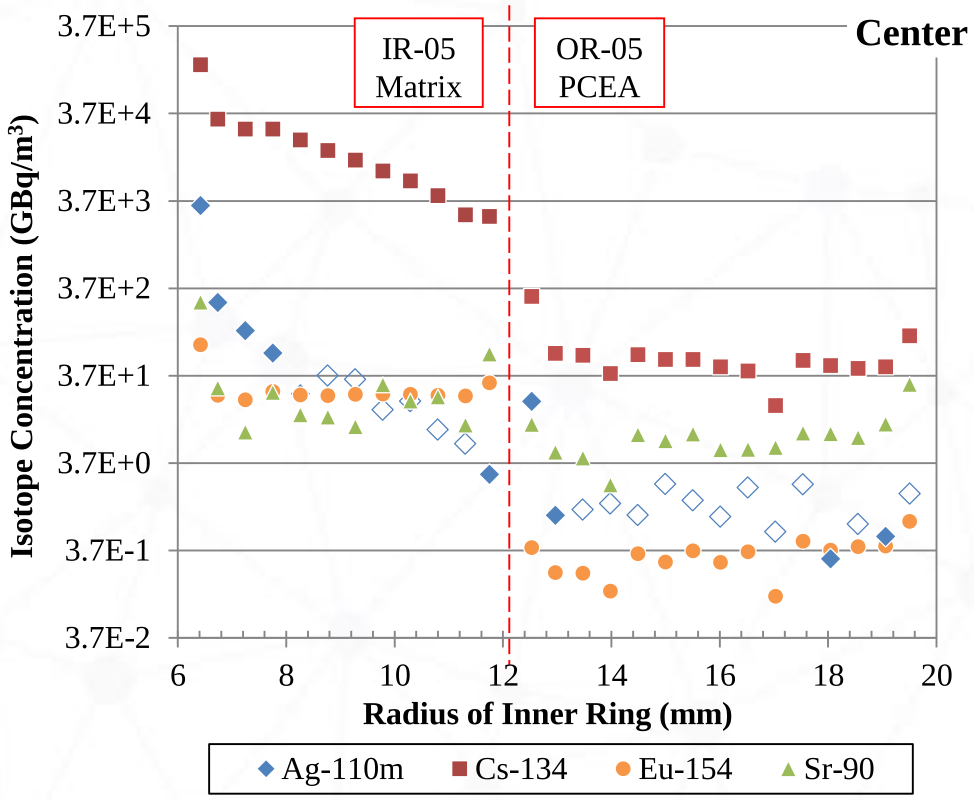




Open symbols indicate value derived from minimum detectable activity.

Capsule 5 Inner and Outer Ring Concentration Profiles

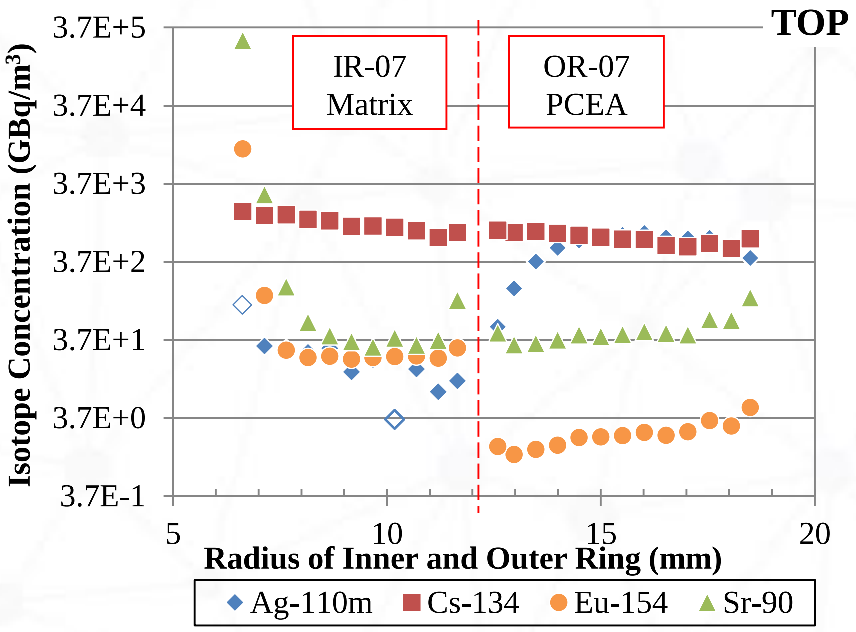
- Compacts TAVA temp: 1015°C
- Inner ring:
  - Matrix A3-27
  - TAVA temp: 800°C
- Outer ring:
  - PCEA
  - TAVA temp: 677°C

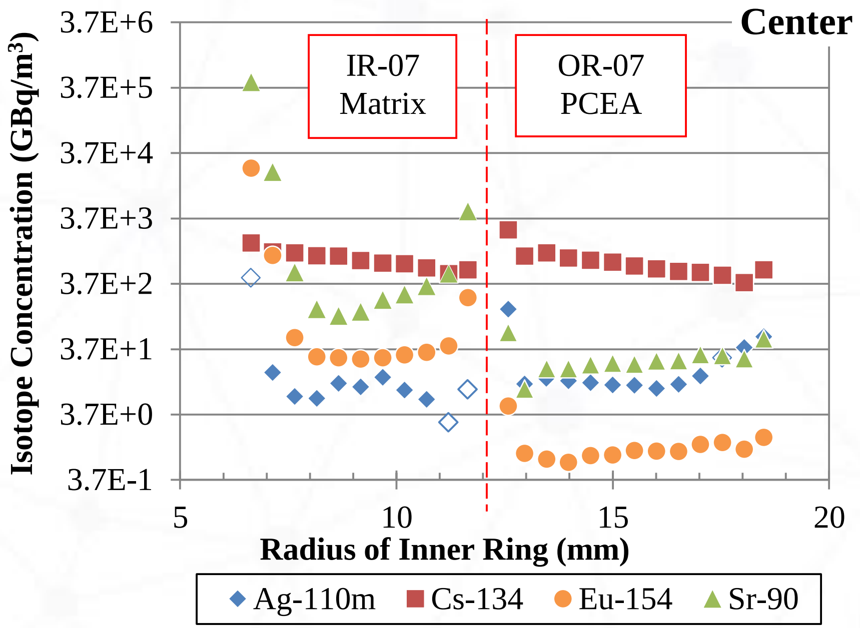


Open symbols indicate value derived from minimum detectable activity.

Capsule 7 Inner and Outer Ring Concentration Profiles

- Compacts TAVA temp: 1345°C
- Inner ring:
  - Matrix A3-27
  - TAVA temp: 1151°C
- Outer ring:
  - PCEA
  - TAVA temp: 1025°C

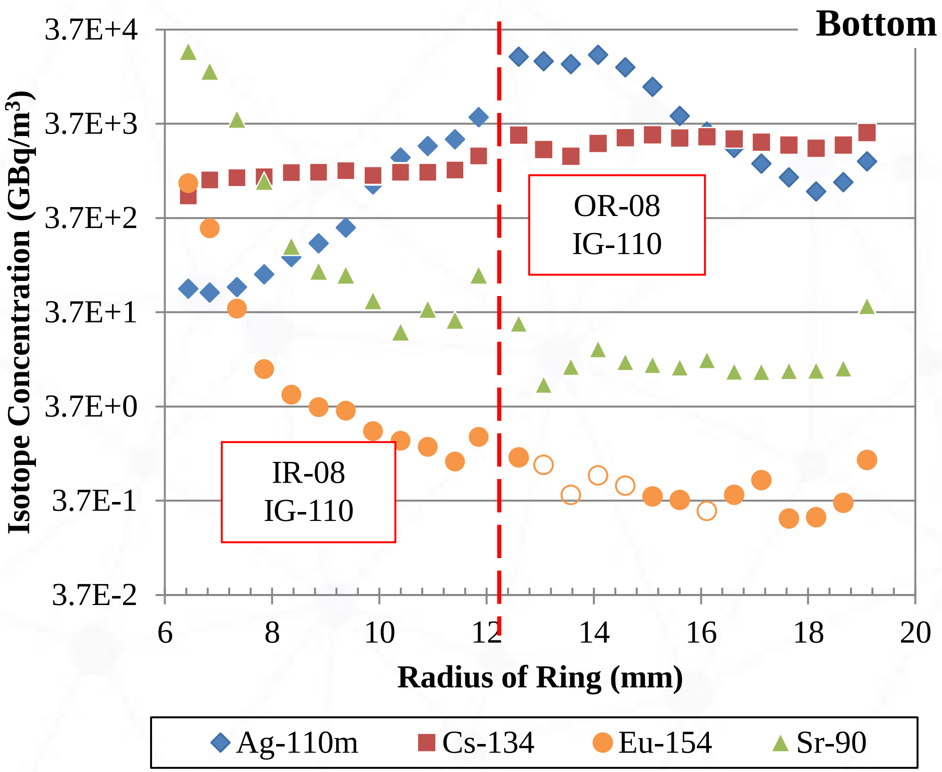




Open symbols indicate value derived from minimum detectable activity.

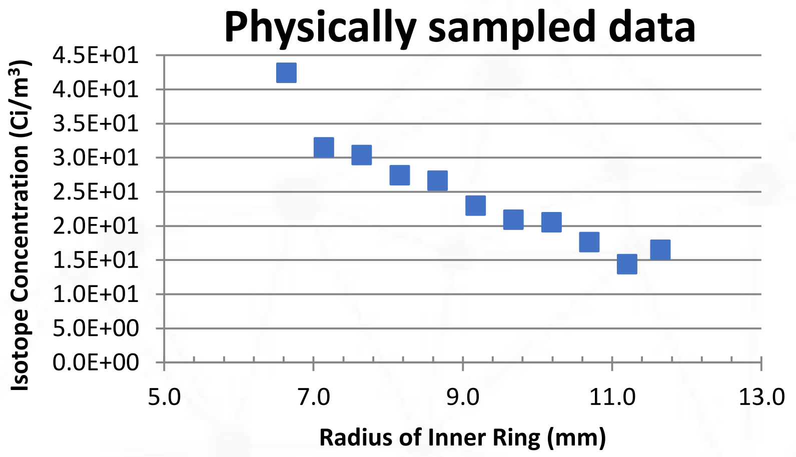
Capsule 8 Inner and Outer Ring Concentration Profiles

- Compacts TAVA temp: 1190°C
- Inner ring:
  - IG-110
  - TAVA temp: 1021°C
- Outer ring:
  - IG-110
  - TAVA temp: 917°C

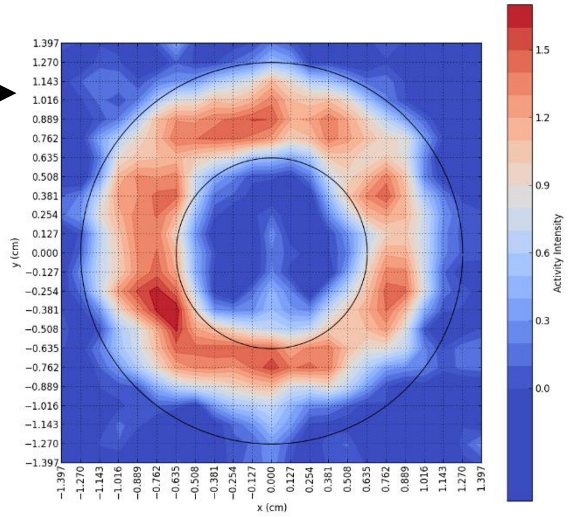


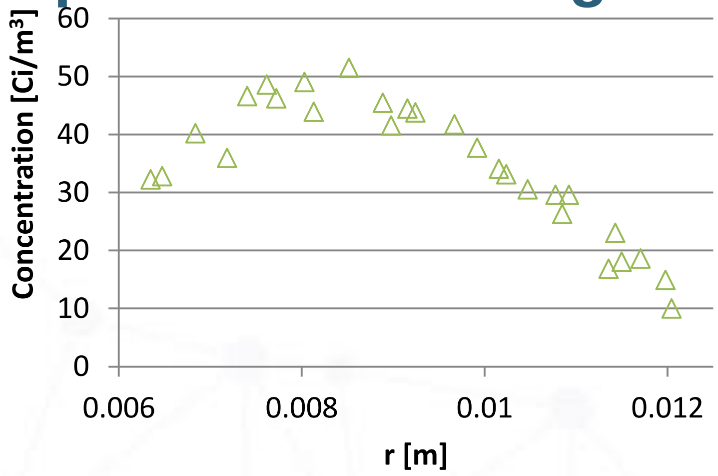
Open symbols indicate value derived from minimum detectable activity.

Comparing PGS and Physical Measurement Cs-134, Capsule 7 Inner Ring

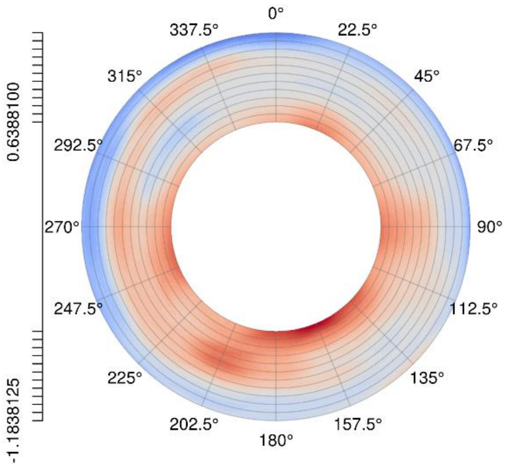


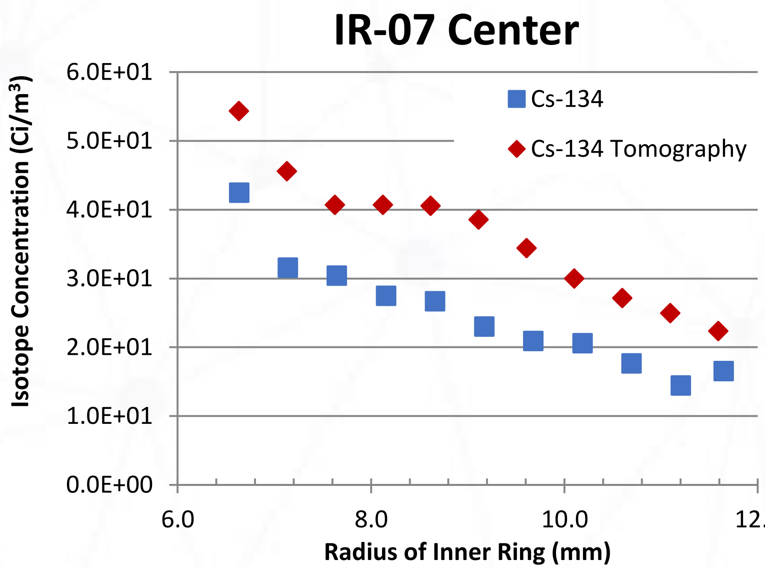
Old Method →





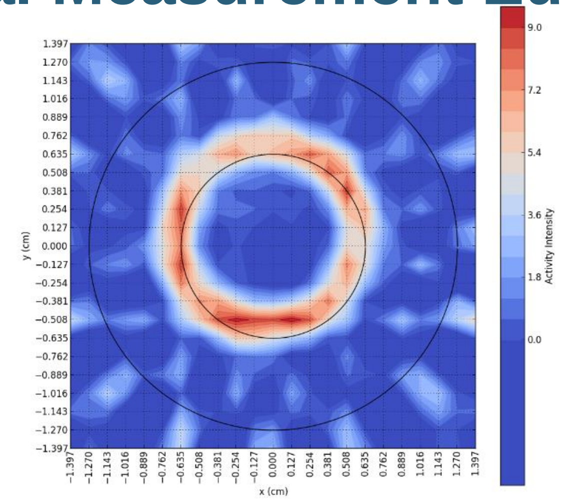
New Method →

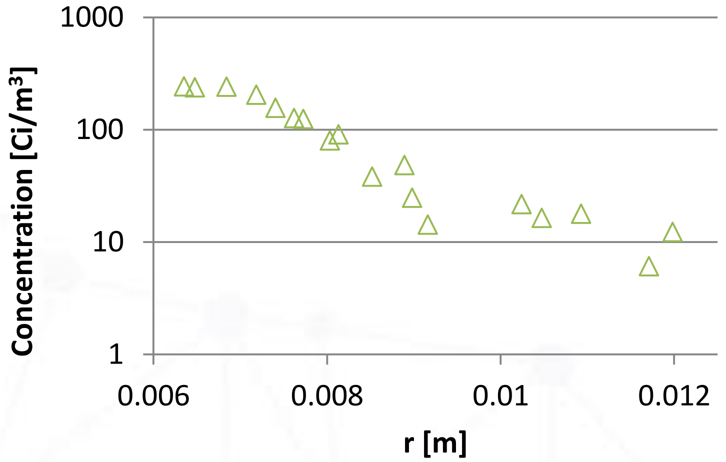


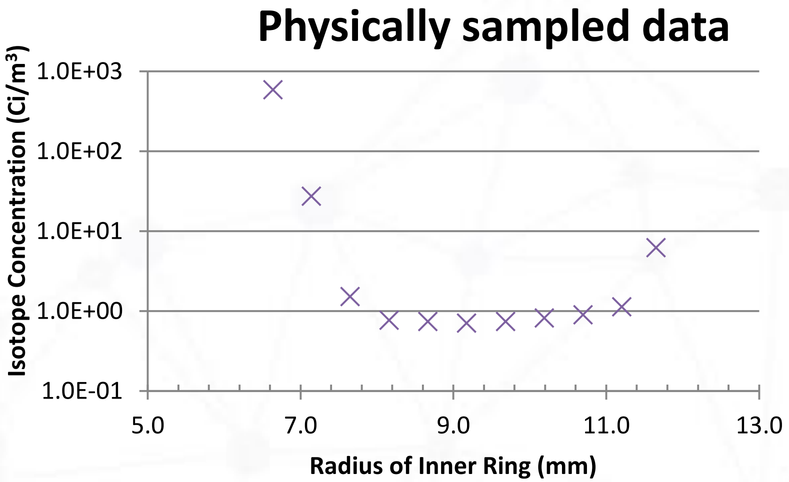


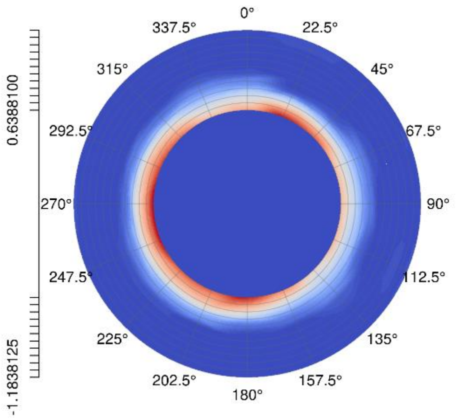
Comparing PGS and Physical Measurement Eu-154, Capsule 7 Inner Ring

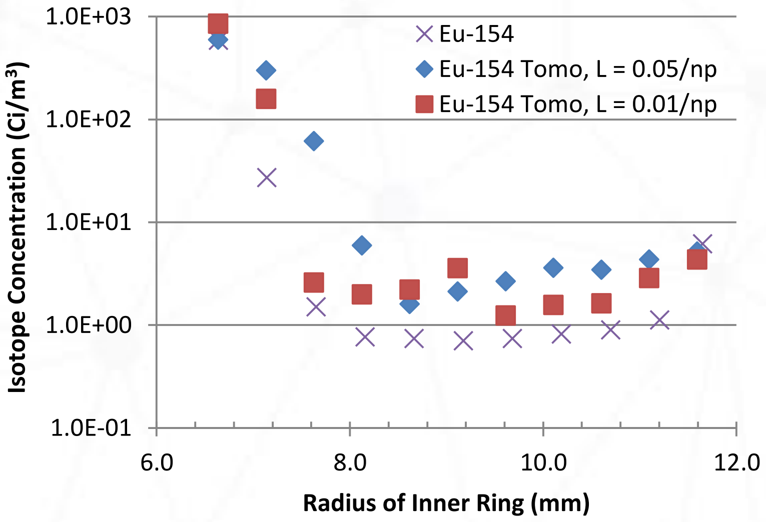
Old Method









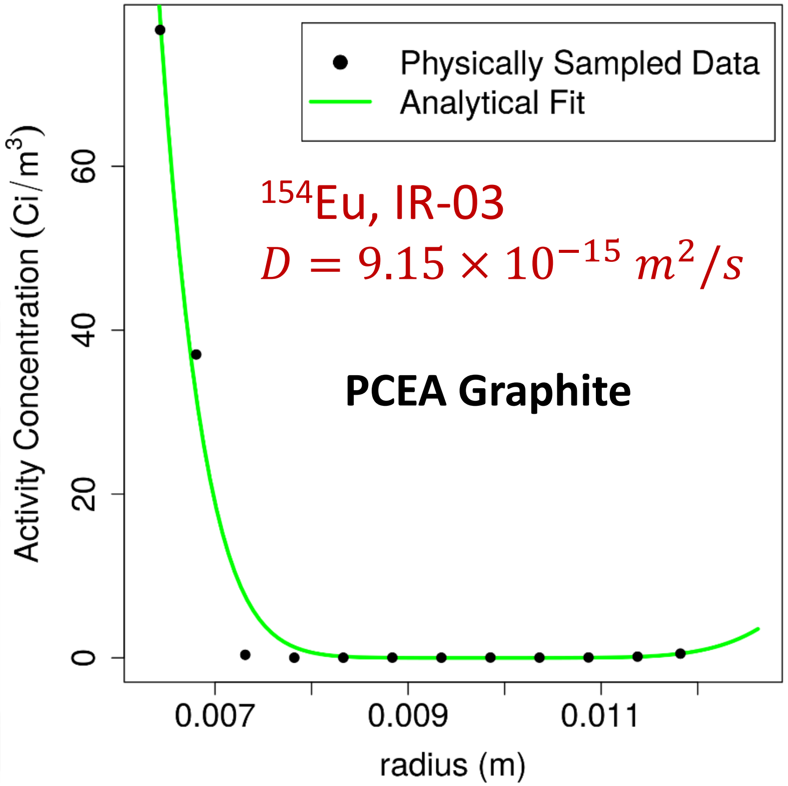


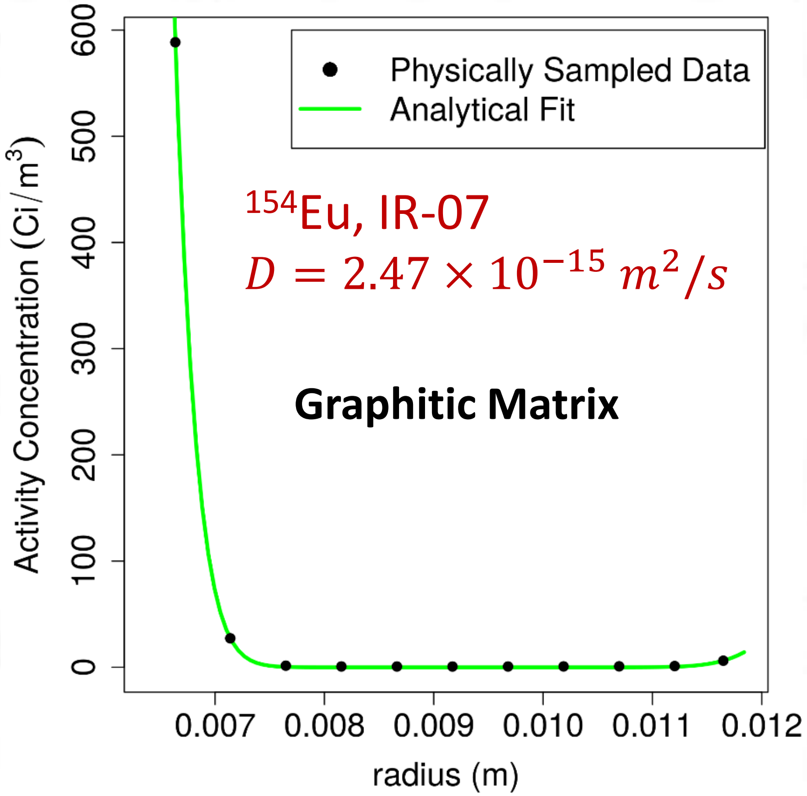
New Method

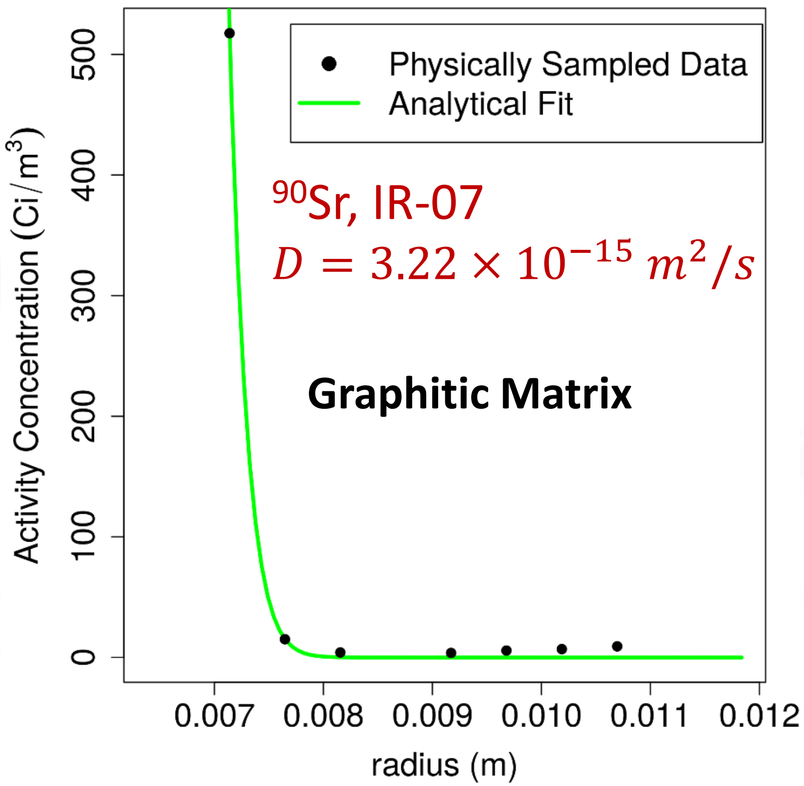
Calculating a New Diffusion Coefficient for Eu-154 & Sr-90

- Direct fits to 1D radial, transient analytical solution, assuming boundary value linearly increasing with time

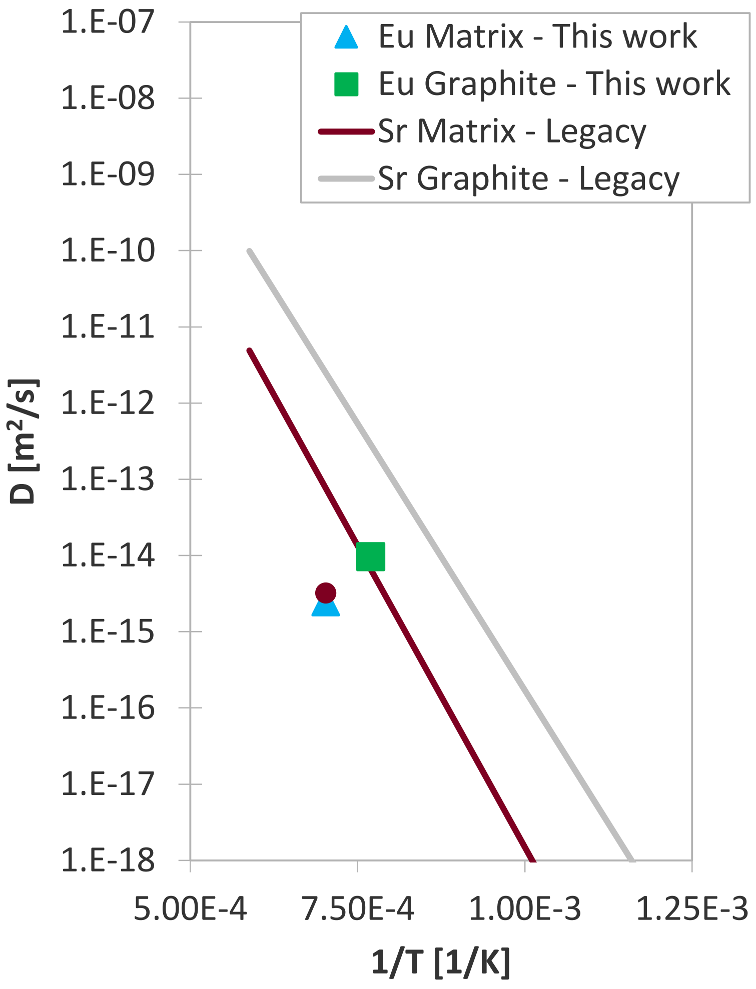
$$C(r,t) = \frac{f(t)\ln\left(\frac{b}{r}\right)+g(t)\ln\left(\frac{r}{a}\right)}{\ln\left(\frac{b}{a}\right)} - \pi\sum_{n=1}^{\infty}\exp(-D\alpha_n^2 t)\frac{J_0(a\alpha_n)U_0(r\alpha_n)}{J_0^2(a\alpha_n)-J_0^2(b\alpha_n)}\left\{J_0(a\alpha_n)\left[g(0)+\int_0^t g'(\tau)\exp(D\alpha_n^2\tau)d\tau\right]-J_0(b\alpha_n)\left[f(0)+\int_0^t f'(\tau)\exp(D\alpha_n^2\tau)d\tau\right]\right\}$$



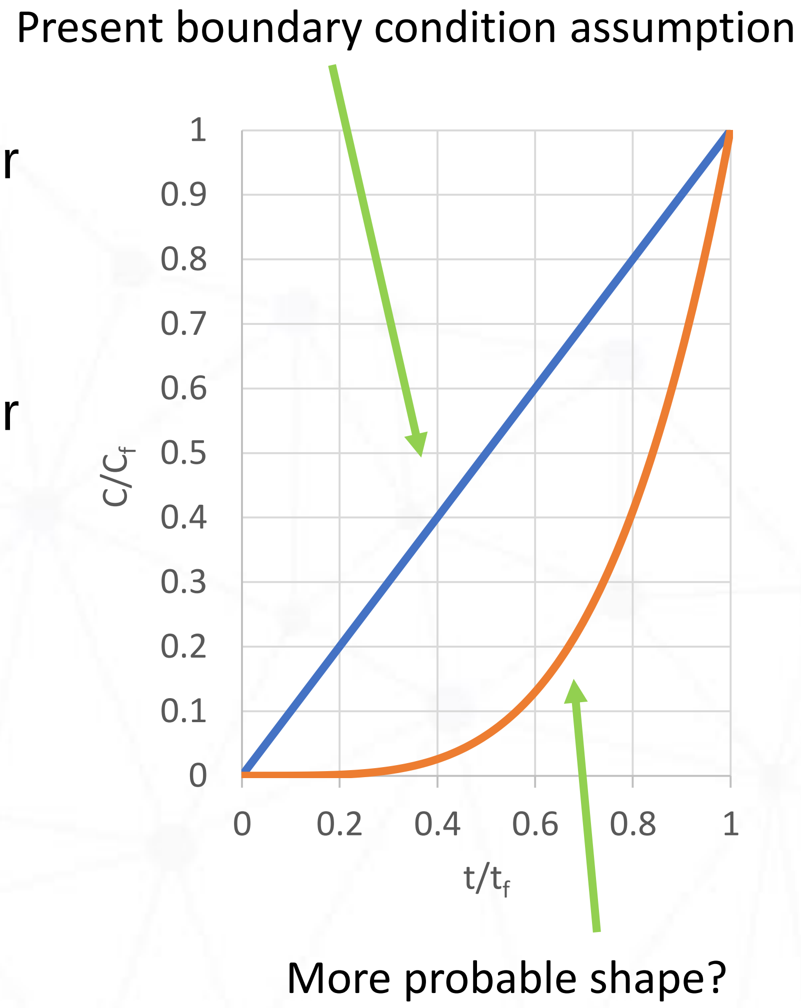




Comparison of AGR-3/4 Diffusion Coefficients to Legacy Data

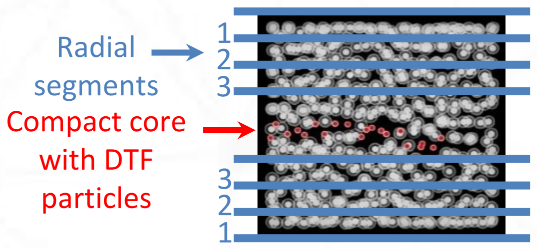


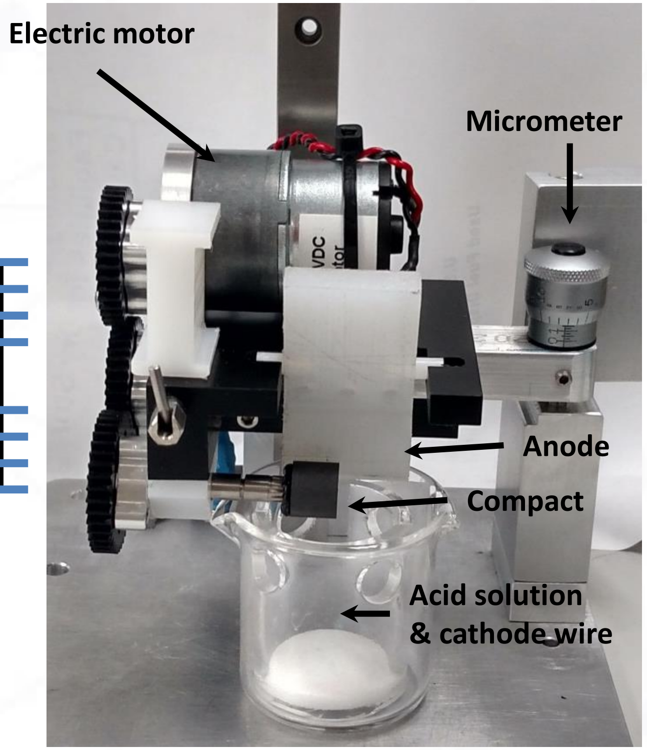
- These estimates are somewhat lower than legacy values
- However, since Eu and Sr are relatively slow diffusers, more of the inventory was probably released later in time (AGR-3/4 gives average)
- If so, these values are likely to be revised upward in the final analysis
- To be further informed by the multiphysics model

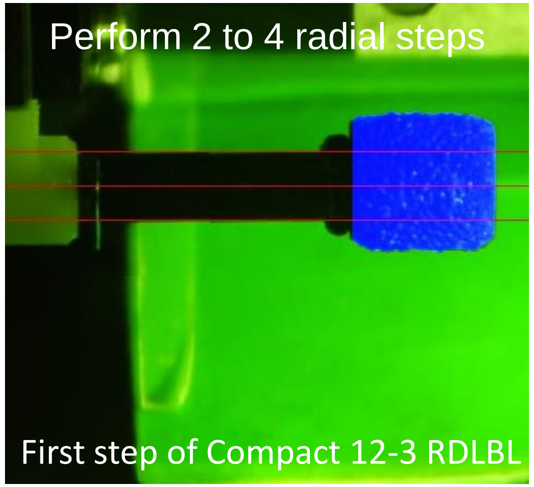


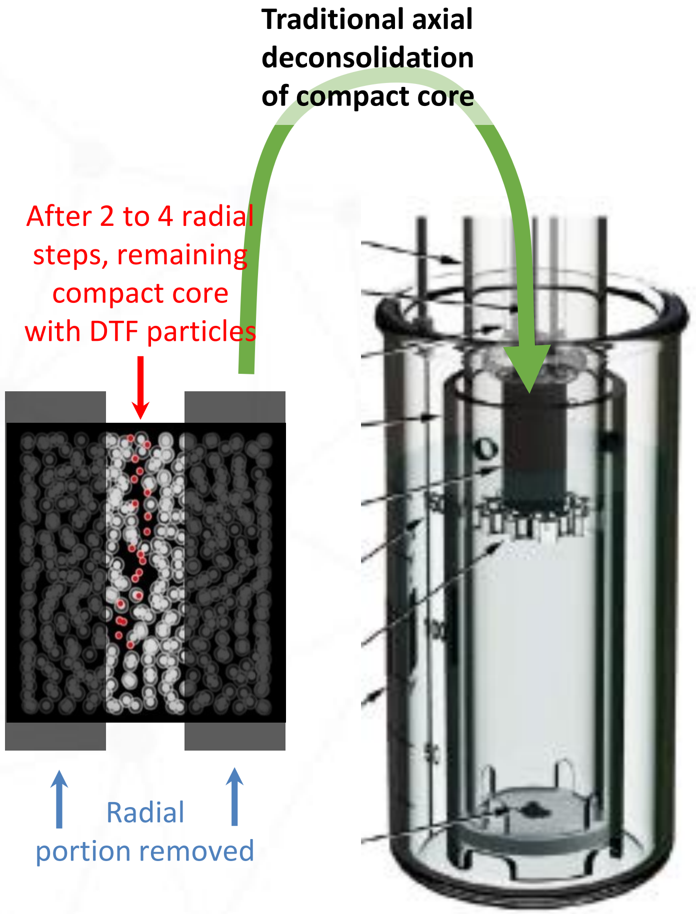
AGR-3/4 Compact Radial Deconsolidation-leach-burn-leach

- Radial deconsolidation-leach-burn-leach (R-DLBL)
  - Measure fission product inventory in compact outside of the SiC layer as a function of the radial position in the compact
  - Collect particles from specific radial portions of the compact for gamma counting and other PIE
  - Avoid deconsolidating DTF particles until the final axial step
  - Compare measured fission product profile with model predictions



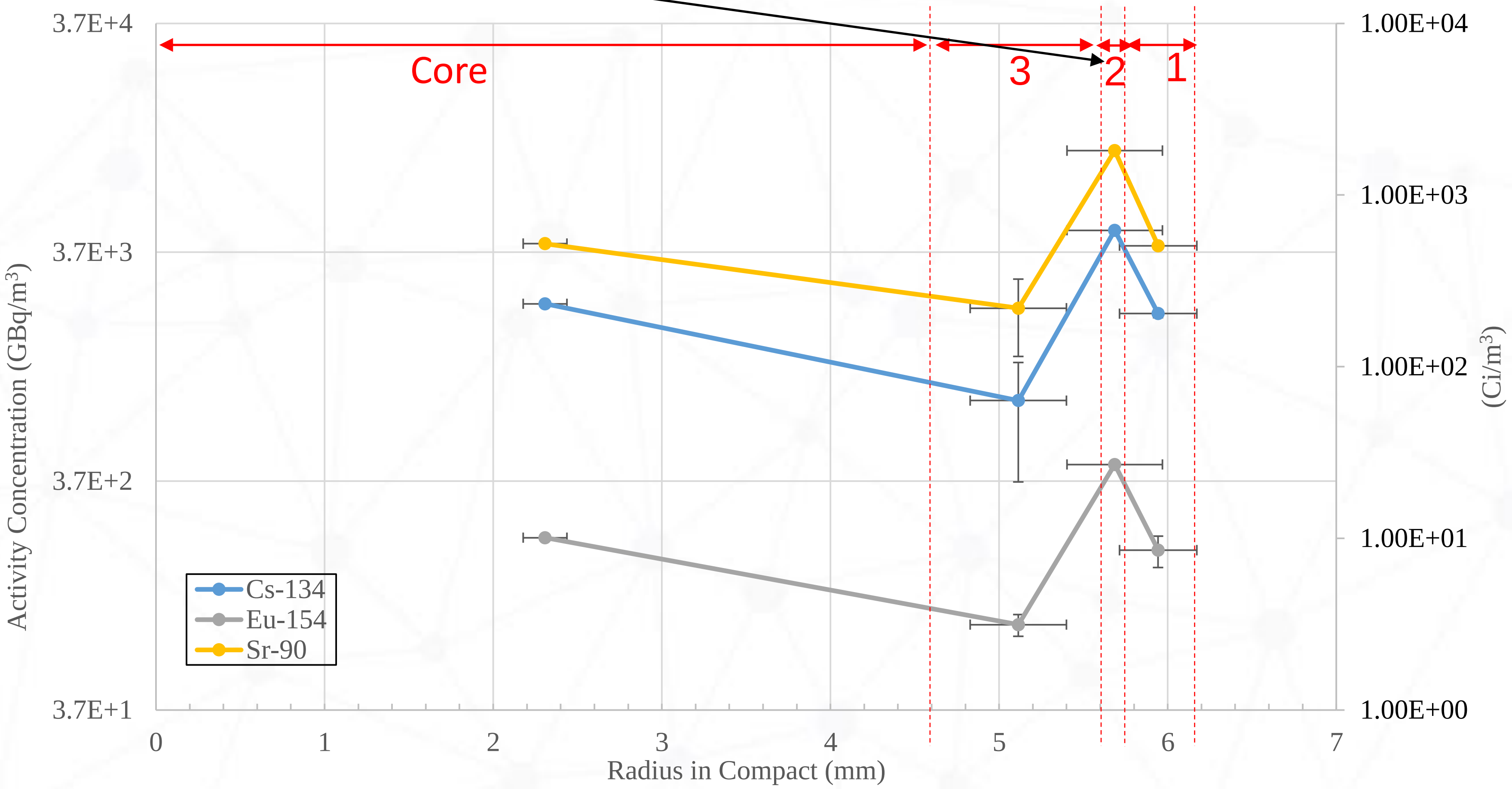






R-DLBL of Compact 12-1 (preliminary concentrations)

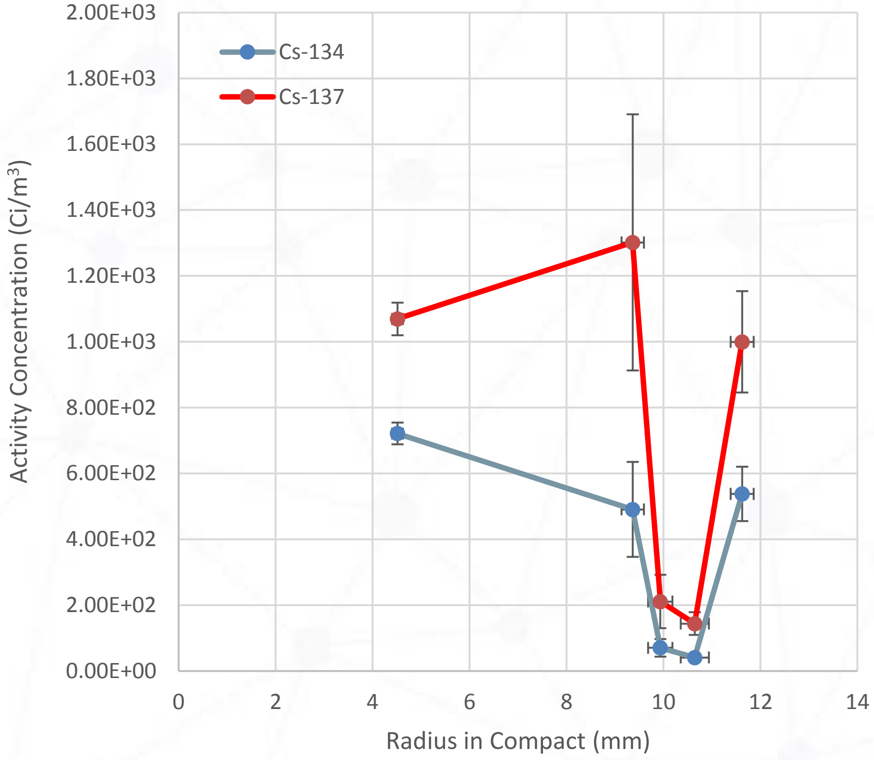
- Concentrations of selected isotopes in 3 radial locations and the central core
- Note: Zone 2 has 102.7% error

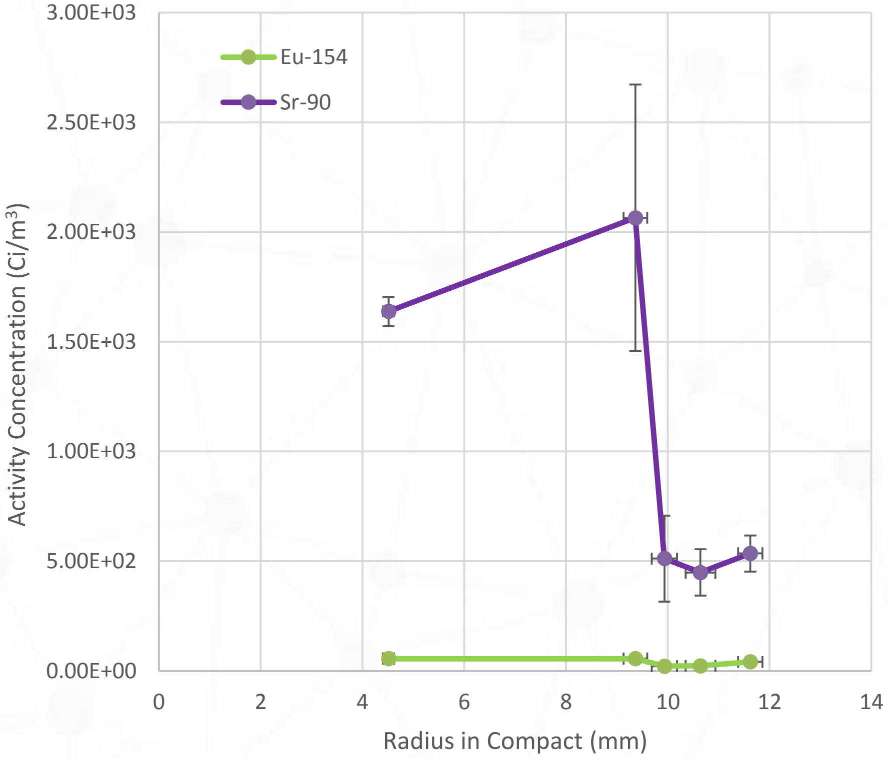


R-DLBL of Compact 3-3 (preliminary concentrations)

- Concentrations of selected isotopes in 4 radial locations and the central core

Totals in Compact	Ce-144	Cs-134	Cs-137	Eu-154	Eu-155	Ru-106	Sr-90
# Particles	27.6	7.7	14.2	20.9	25.6	24.6	23.0
Fraction	1.46E-2	4.09E-3	7.51E-3	1.10E-2	1.35E-2	1.30E-2	1.22E-2

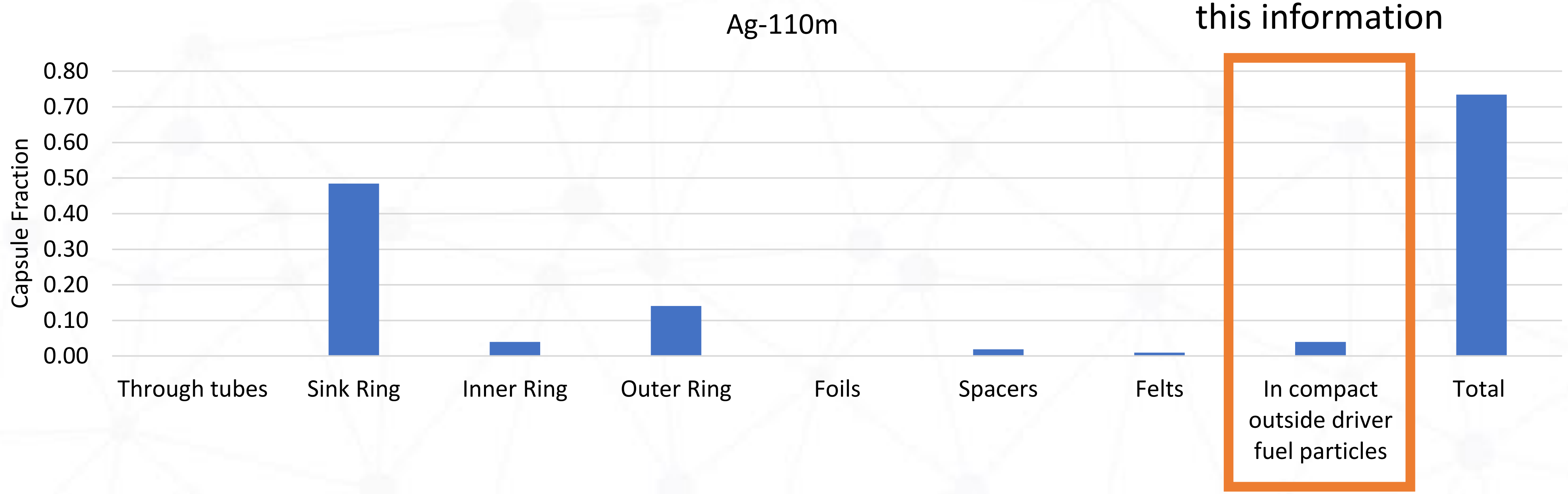




Completed Mass Balance for Capsule 3: Ag-110m

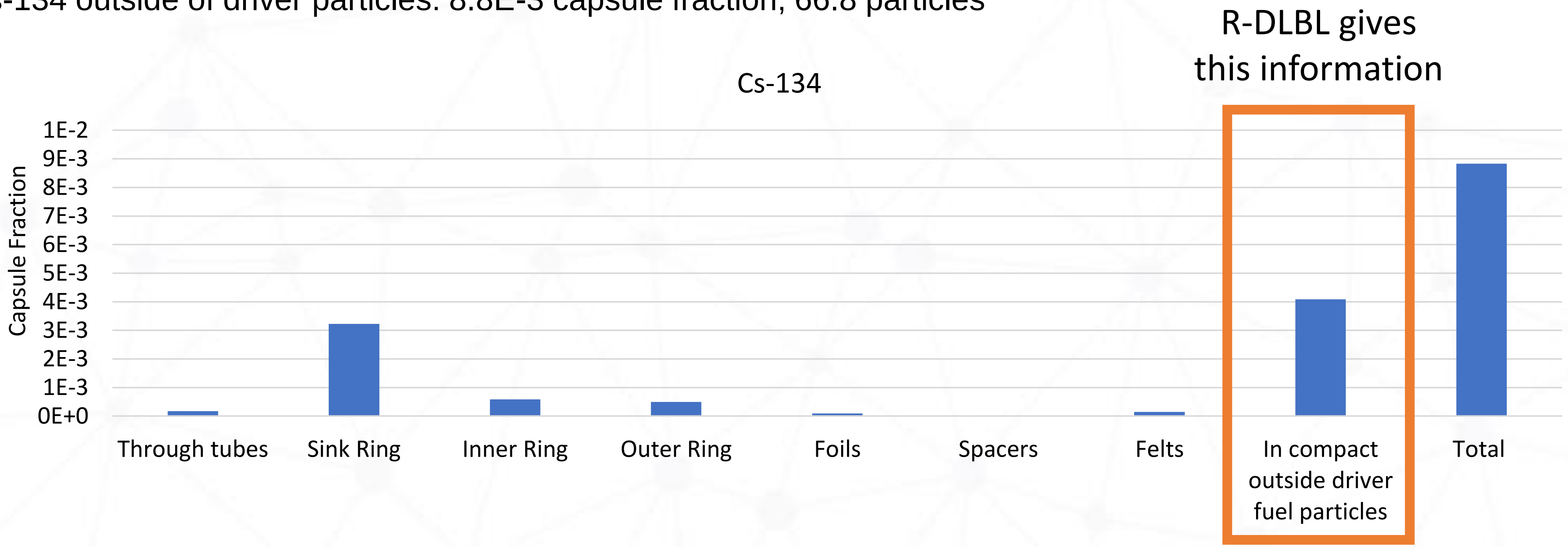
- Capsule 3 fuel, irradiation capsule components, and compacts accounted for
- Compact 3-3 R-DLBL is basis for assuming compact inventory outside of driver fuel

R-DLBL gives this information



Completed Mass Balance for Capsule 3: Cs-134

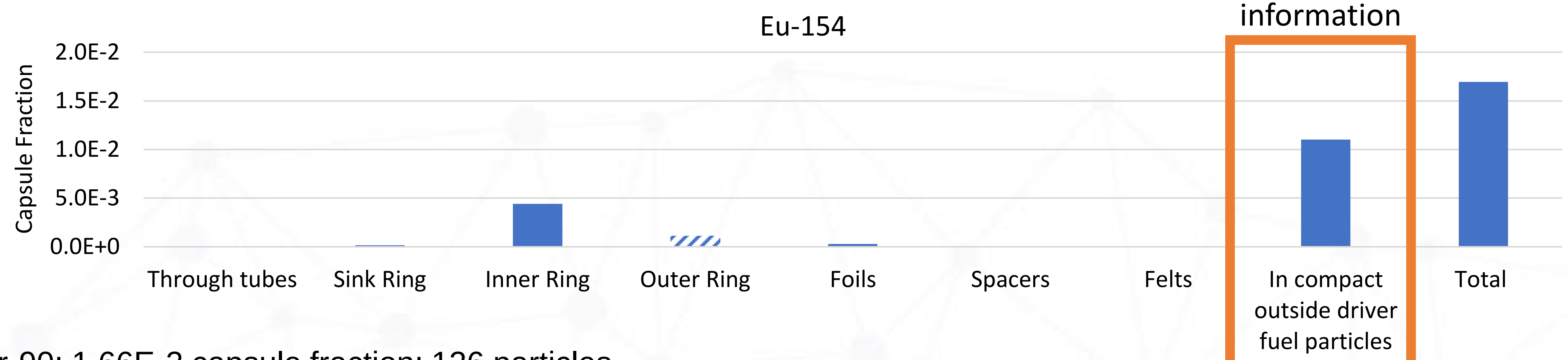
- Capsule 3 fuel, irradiation capsule components, and compacts accounted for
- Compact 3-3 R-DLBL is basis for assuming compact inventory outside of driver fuel
- Total Cs-134 outside of driver particles: 8.8E-3 capsule fraction; 66.8 particles



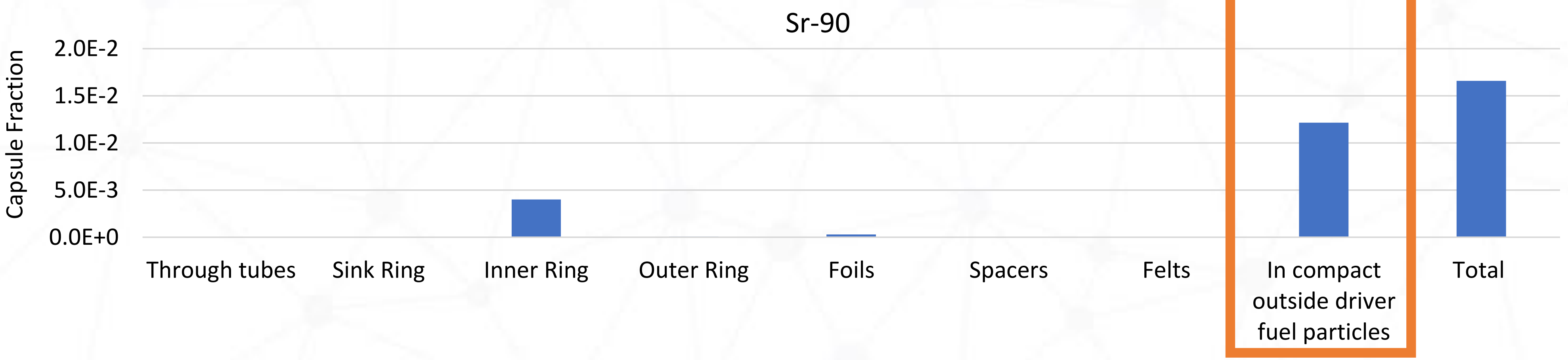
Completed Mass Balance for Capsule 3: Eu-154 & Sr-90

- Total Eu-154: 1.70E-2 capsule fraction; 128 particles

R-DLBL gives this information



- Total Sr-90: 1.66E-2 capsule fraction; 126 particles



By the numbers: Completed

Activity	Number
Capsules disassembled and analyzed for quantitative mass balance	8
Capsules retained intact for possible integral heating testing in He, air, or moisture	4
Compacts counted on PGS	32
Inner/outer rings counted on PGS	16
Inner/outer rings physically sampled (Capsule 10 rings sampling fulfilled FY19 L3 Milestone)	10
Compacts to undergo ceramography	3
As-irradiated compacts to undergo R-DLBL	7
Compacts heated in FACS furnace*	4
Compacts shipped from INL to ORNL for future R-DLBL	6

*See presentation on AGR-2 and AGR-3/4 heating testing after lunch at ~2:15 PM

By the numbers: Planned

Activity	Number
As-irradiated R-DLBL	8
R-DLBL of compacts after FACS heating testing	4
Compact re-irradiation heating tests	4
R-DLBL of compacts after NRAD re-irradiation and FACS heating tests	4

Additional Activities

- Planned:
  - Update to AGR-3/4 fission product transport model/predictions
  - Continue back-calculation of fission product diffusion coefficients
  - Complete radio-chemical analysis and particle counting from compact R-DLBL
  - Reporting
- Possible depending on need:
  - Ring heating followed by PGS and physical sampling – refine diffusion coefficients measurements
  - Intact fuel body capsule heating tests in helium
- Future use of AGR-3/4 samples (not included in AGR-3/4 PIE):
  - Air/moisture atmosphere testing in the Air/Moisture Ingress Experiment (AMIX) furnace
    (see presentation at ~ 4:50 PM today)

INL

Idaho National Laboratory

Questions and Discussion

John Stempien	**AGR PIE Technical Lead**
John.Stempien@inl.gov	Phone (208) 526-8410