



AGR-3/4 Post-Irradiation Examination

July 2021

Changing the World's Energy Future

John D Stempien



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**Prepared for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517**

July 13, 2021

John Stempien, Ph.D.
AGR PIE Technical Lead

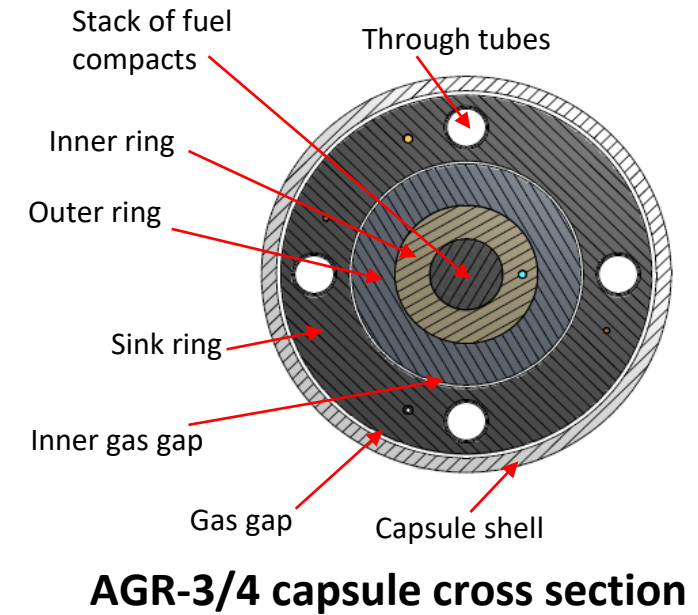
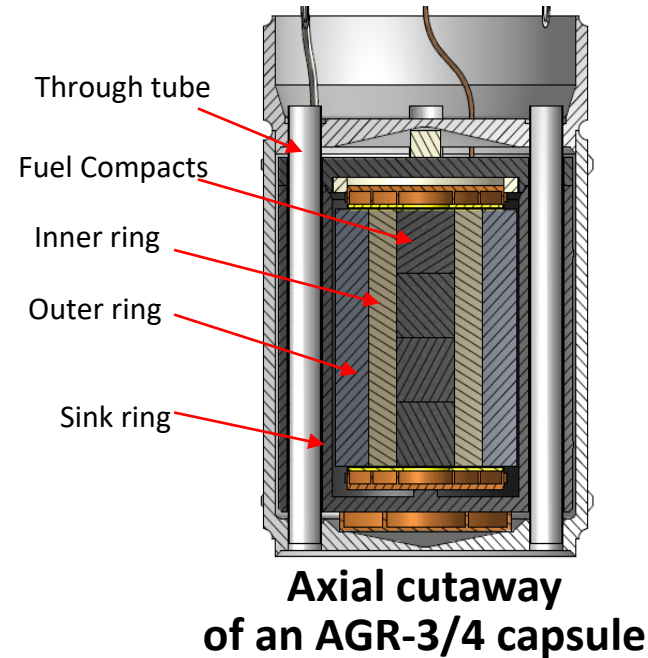
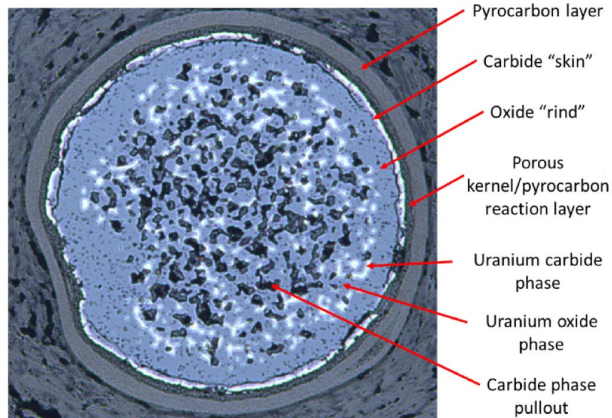
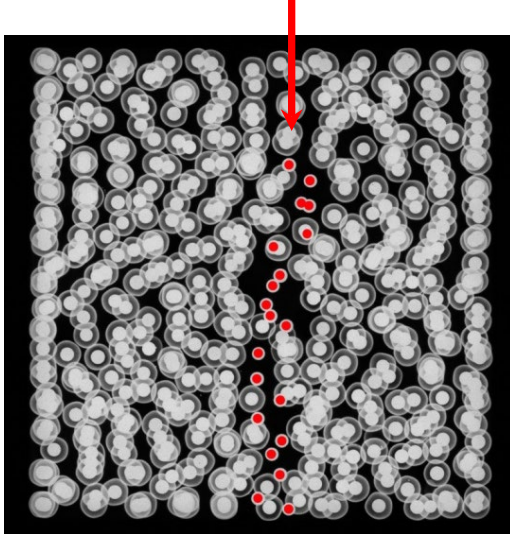
AGR-3/4 Post-Irradiation Examination

AGR-3/4 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
- These goals are different than fuel performance experiments like AGR-1, AGR-2, and AGR-5/6/7.

Besides Fuel Compacts, Carbon Rings are Key Samples

X-ray showing 20 DTF particles in center of compact



Mass Balance of Fission Products Outside of Fuel Compacts was Completed in 2018

Summary of the fission product inventory measured outside of the fuel expressed units of particle equivalents. “Low” includes measured values only. “High” represents measured values plus particle equivalents calculated from MDAs.

Particle Equivalents		Ag-110m	Ce-144	Cs-134	Cs-137	Eu-154	Eu-155	Ru-106	Sr-90 ^b
1	Low	2.23E+1	0.00E+0	5.11E+0	9.53E+0	8.10E+0	1.28E+1	0.00E+0	3.51E-2
	High	9.33E+2	1.55E-1	5.14E+0	9.53E+0	4.53E+1	1.38E+1	8.10E-1	3.51E-2
2 ^a	Low	0.00E+0	0.00E+0	4.18E+0	9.55E+0	0.00E+0	9.91E+0	0.00E+0	5.58E-2
	High	4.90E+1	1.47E-1	4.18E+0	9.55E+0	1.06E+0	1.32E+1	5.70E-1	5.58E-2
3	Low	5.25E+3	5.54E-2	3.59E+1	4.11E+1	3.53E+1	1.26E+1	0.00E+0	2.33E+0
	High	5.26E+3	2.23E-1	3.59E+1	4.11E+1	4.49E+1	1.58E+1	6.55E-1	2.33E+0
4	Low	5.60E+2	0.00E+0	7.21E+1	7.32E+1	0.00E+0	1.88E+1	0.00E+0	7.75E-2
	High	5.67E+2	2.33E-1	7.21E+1	7.32E+1	1.44E+1	1.92E+1	2.08E-1	7.75E-2
5	Low	3.36E+1	8.69E-2	6.34E+1	6.05E+1	0.00E+0	0.00E+0	0.00E+0	3.51E-2
	High	4.63E+1	1.86E-1	6.34E+1	6.05E+1	1.27E+1	3.03E+0	3.25E-1	3.51E-2
6 ^a	Low	1.29E+1	0.00E+0	3.28E+0	4.74E+0	2.33E-3	0.00E+0	0.00E+0	2.57E-2
	High	3.47E+1	1.95E-1	3.29E+0	4.74E+0	6.47E-1	3.41E+0	4.74E-1	2.57E-2
7	Low	7.65E+3	0.00E+0	4.52E+1	4.96E+1	2.33E+2	4.17E+0	7.75E-3	3.33E+0
	High	7.65E+3	1.10E-1	4.52E+1	4.96E+1	2.40E+2	6.52E+0	3.83E-1	3.33E+0
8	Low	6.67E+3	1.69E-2	5.23E+1	6.02E+1	1.11E+1	2.96E+0	2.11E-2	7.09E-1
	High	6.68E+3	1.32E-1	5.23E+1	6.02E+1	1.92E+1	5.87E+0	3.41E-1	7.09E-1
9 ^a	Low	3.14E+1	0.00E+0	2.20E+0	5.07E+0	2.22E+0	1.05E+1	1.43E-2	3.56E-2
	High	6.43E+1	1.83E-1	2.20E+0	5.07E+0	3.78E+0	1.14E+1	9.48E-1	3.56E-2
10	Low	5.52E+3	7.50E-3	4.34E+1	5.03E+1	9.15E+0	4.61E+0	2.44E-2	2.10E+0
	High	5.52E+3	1.32E-1	4.34E+1	5.03E+1	9.96E+0	7.54E+0	4.11E-1	2.10E+0
11 ^a	Low	4.12E+2	8.81E-3	2.65E+1	3.18E+1	1.99E+1	2.28E+1	0.00E+0	1.14E-1
	High	2.17E+3	2.52E-1	2.65E+1	3.18E+1	2.16E+1	2.36E+1	7.59E-1	1.14E-1
12	Low	0.00E+0	0.00E+0	3.25E+0	1.65E+1	1.69E+1	5.61E+0	6.21E-4	1.38E-1
	High	3.07E+3	2.45E-1	4.67E+0	1.65E+1	1.35E+2	1.18E+1	1.41E+0	1.38E-1

a. Capsules 2, 6, 9, and 11 were intact fuel bodies. The inner and outer ring inventories have not been determined. The mass balance is incomplete in these capsules.

b. Does not include inner and outer ring Sr-90 inventory.

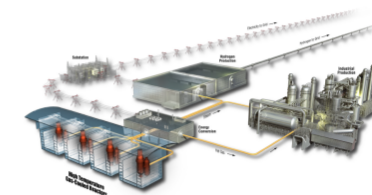
INL/EXT-18-46049
Revision 0

AGR-3/4 Experiment Preliminary Mass Balance

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Paul A. Demkowicz
Jason M. Harp
Philip L. Winston

August 2018

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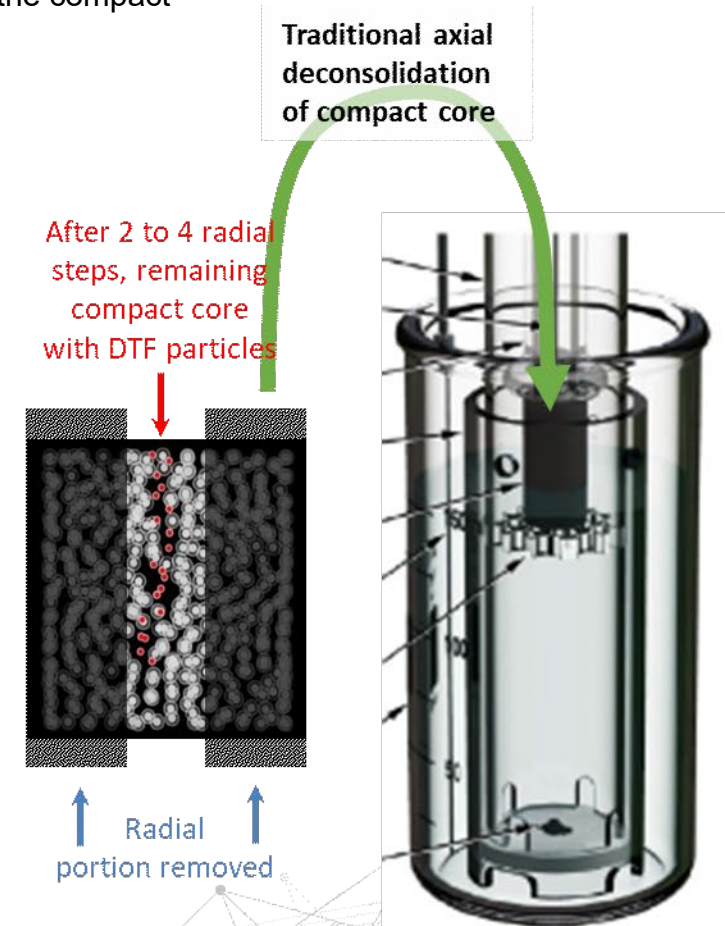
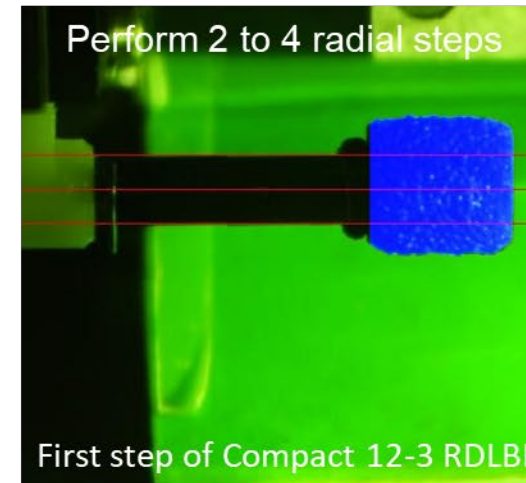
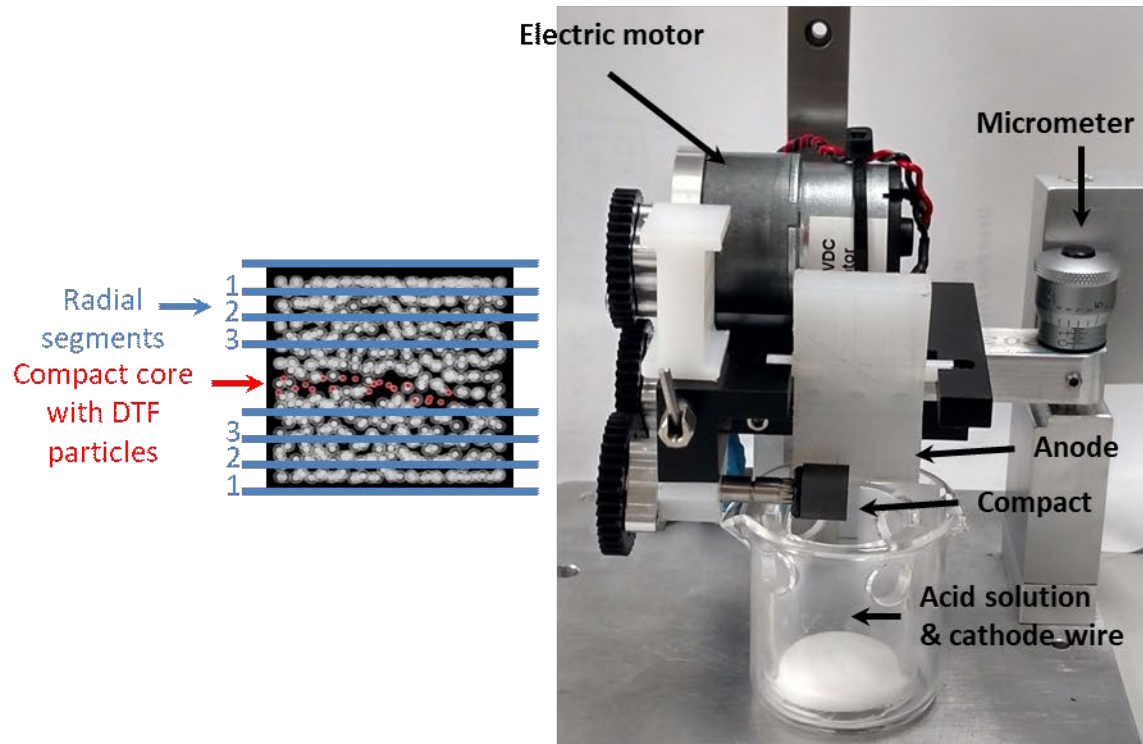




AGR-3/4 Compact Destructive Exams

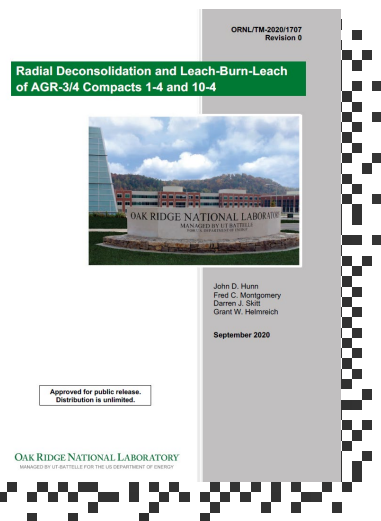
Radial Deconsolidation-Leach-Burn-Leach to Measure Fission Product Radial Concentration Profiles

- 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



As-Irradiated Compacts R-DLBL Status

Compact ID	Burnup (% FIMA)	TAVA Irradiation Temp (°C)	R-DLBL
1-3	6.4	959	In-progress at ORNL in FY21
1-4	6.9	929	Completed ORNL
3-3	12.7	1205	Completed at INL
5-3	14.9	1050	Completed at INL
5-4	15.0	989	Completed at INL
7-3	15.0	1376	Completed at INL
7-4	14.9	1319	Radiochemical Analysis in Progress at ORNL in FY21
8-3	14.5	1213	Completed at INL
8-4	14.4	1169	Radiochemical Analysis in Progress at ORNL in FY21
10-3	11.8	1210	Radiochemical Analysis in Progress at INL in FY21
12-1	5.9	849	Completed at INL
12-3	5.2	864	Completed at INL

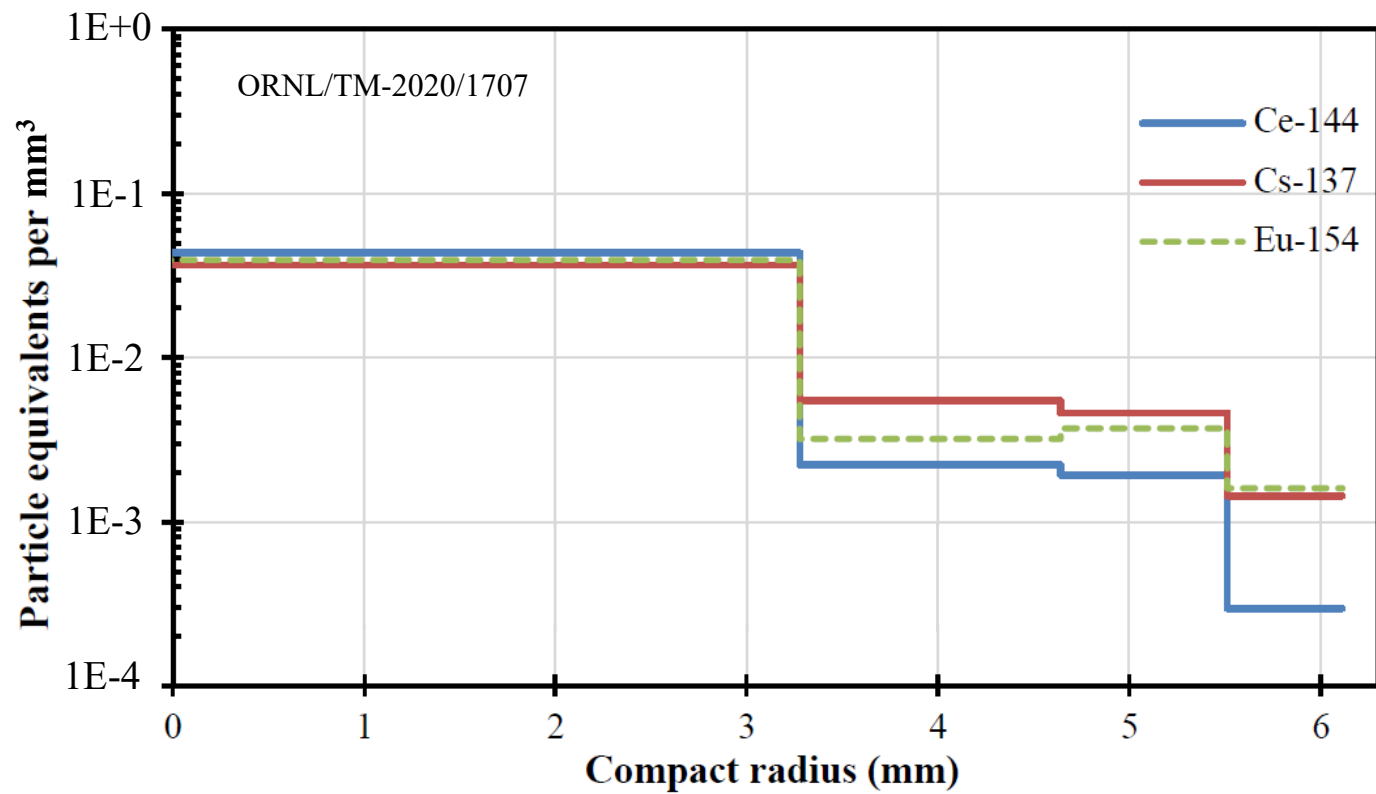


FACS-Tested Compacts R-DLBL Status

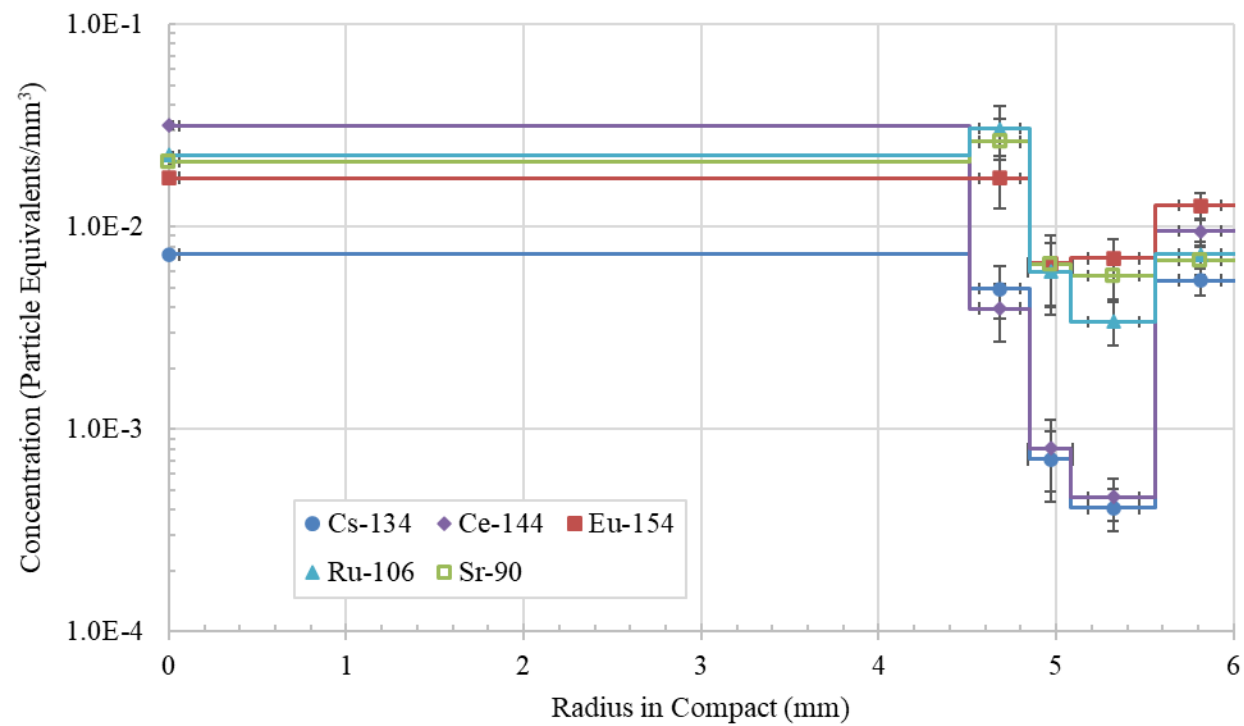
- Expecting to have only four R-DLBLs and radiochemical analysis work remaining to be completed in FY22

Compact	Burnup (% FIMA)	TAVA Irradiation Temp (°C)	FACS Temperature (°C)	Reirradiation?	R-DLBL
1-2	5.9	880	Planned 1400	Planned FY21	Planned for FY22
3-1	12.2	1138	1600	Yes	Planned for FY21 at INL
3-2	12.5	1196	1600	No	Completed at INL
4-3	14.3	1035	Planned 1000	Planned FY21	Planned for FY22
8-1	14.5	1165	1200	Yes	Planned for FY21 at INL
8-2	14.6	1213	1400	No	Completed at INL
10-1	12.1	1172	1400	Yes	Planned for FY22 at ORNL or INL
10-2	12.0	1213	1200	No	Planned at ORNL for FY22
10-4	11.4	1168	1400	No	Completed at ORNL

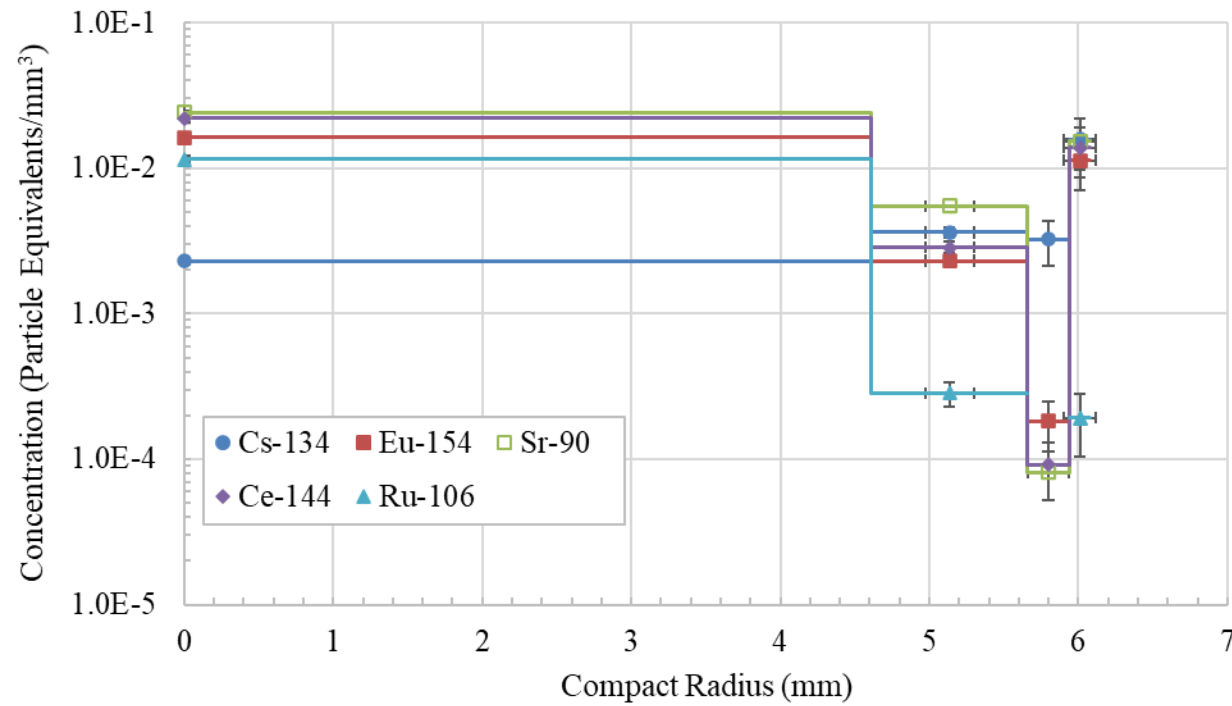
Compact 1-4



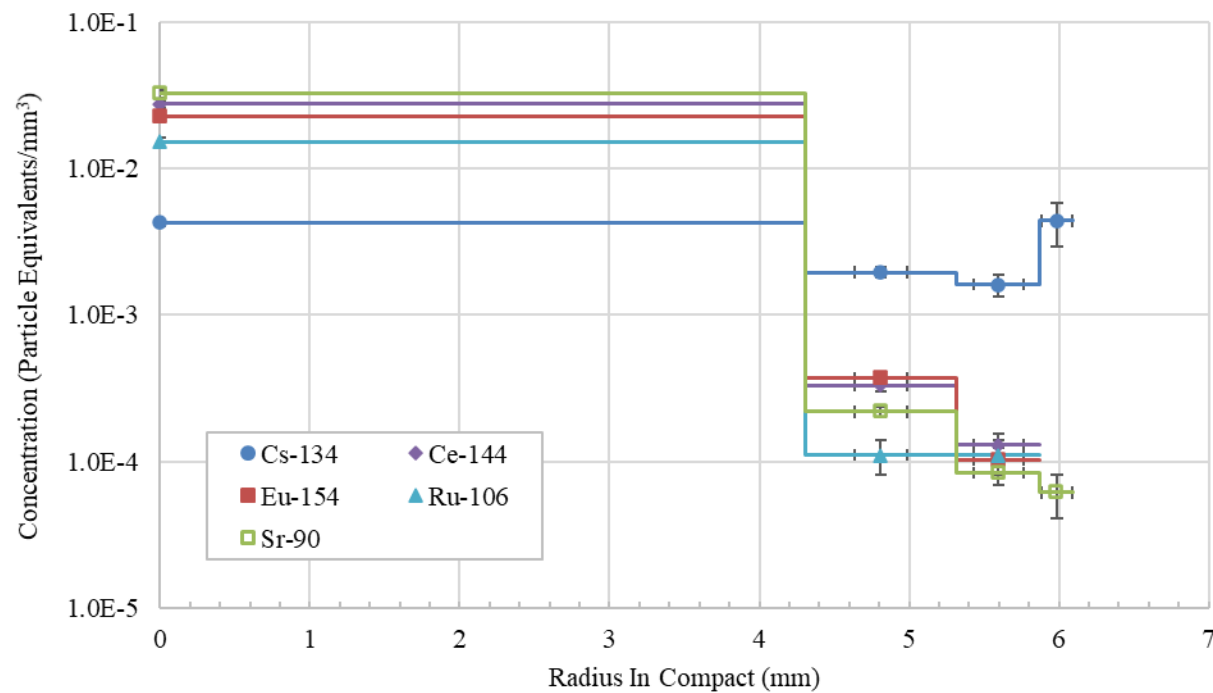
Compact 3-3



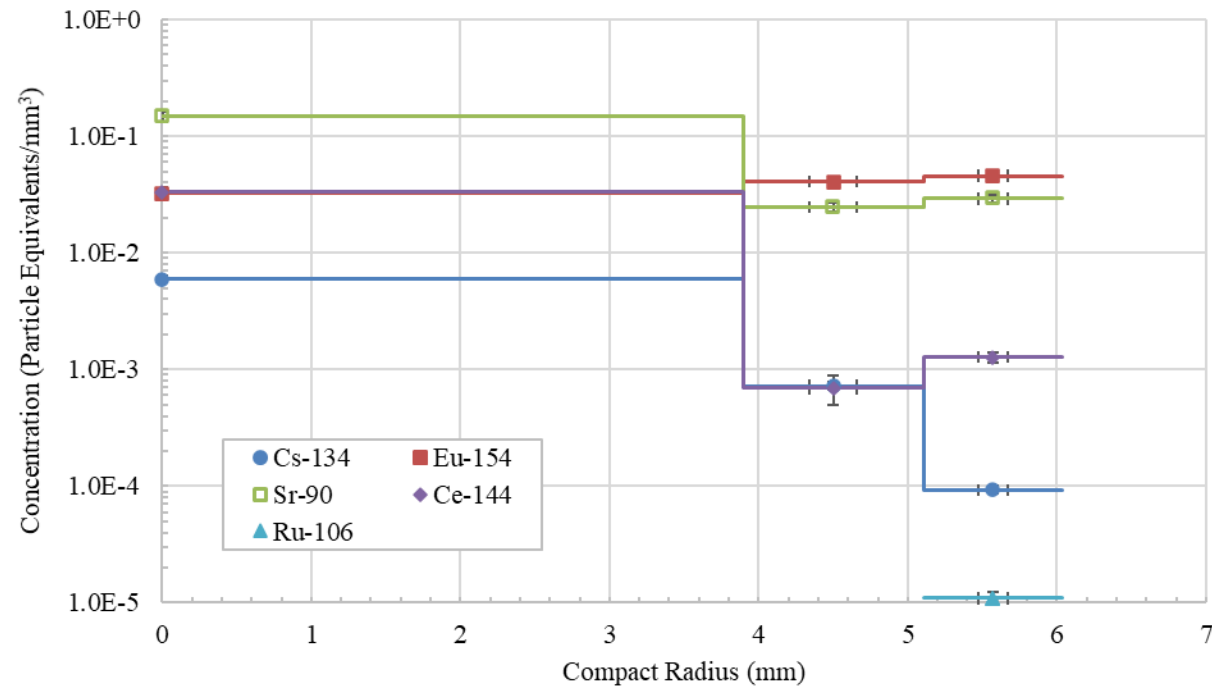
Compact 5-3



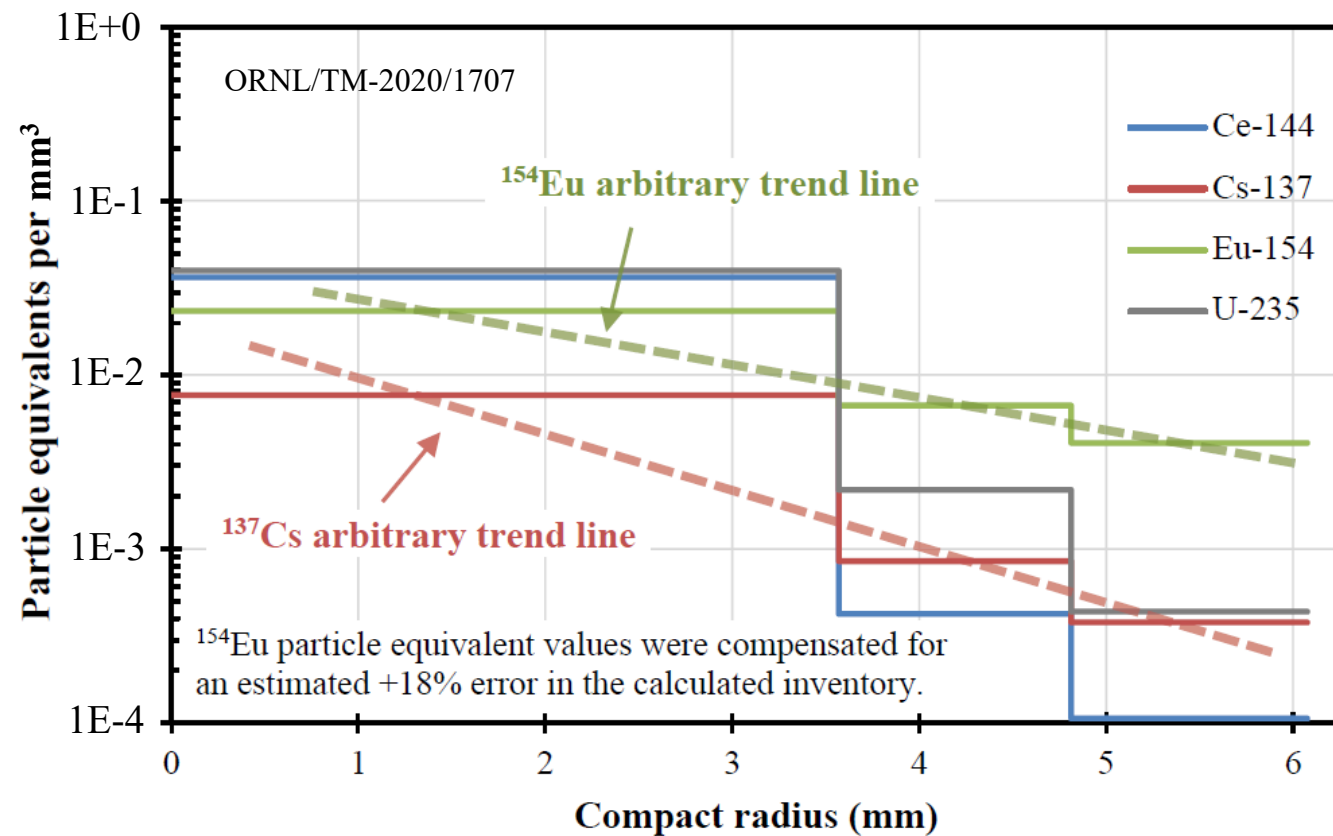
Compact 5-4



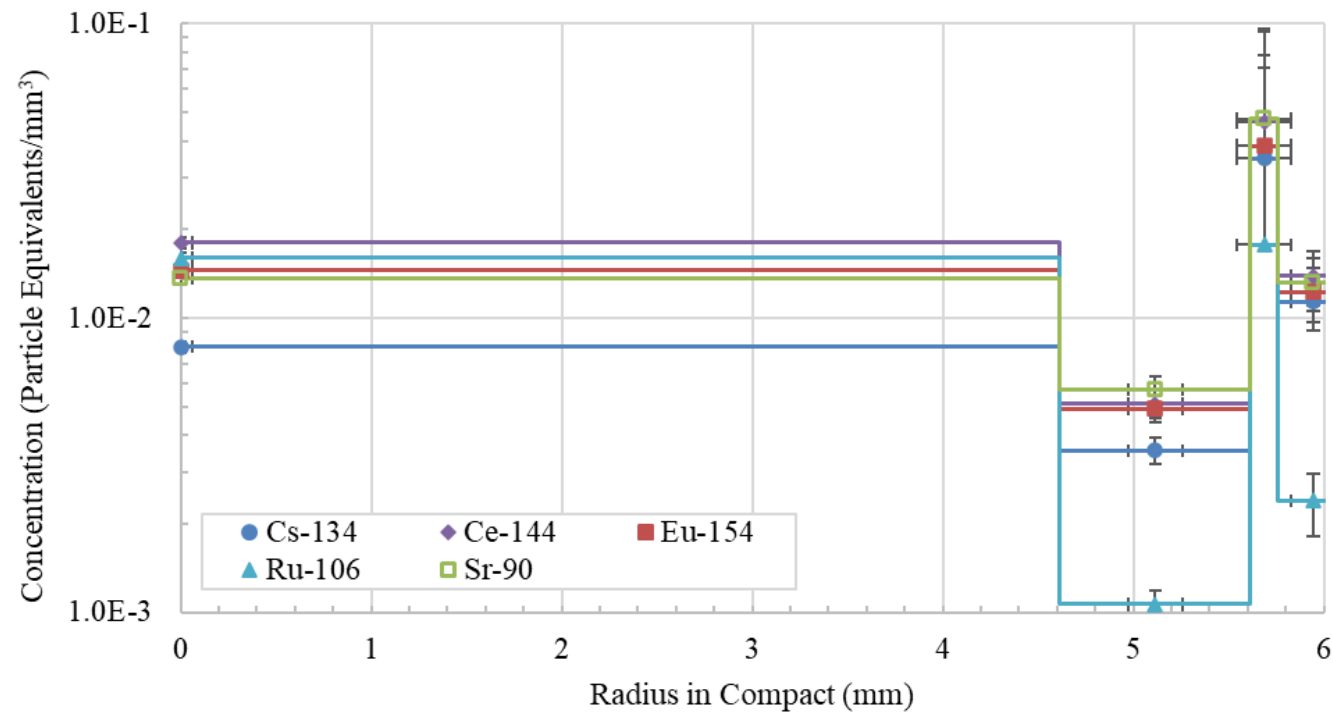
Compact 7-3



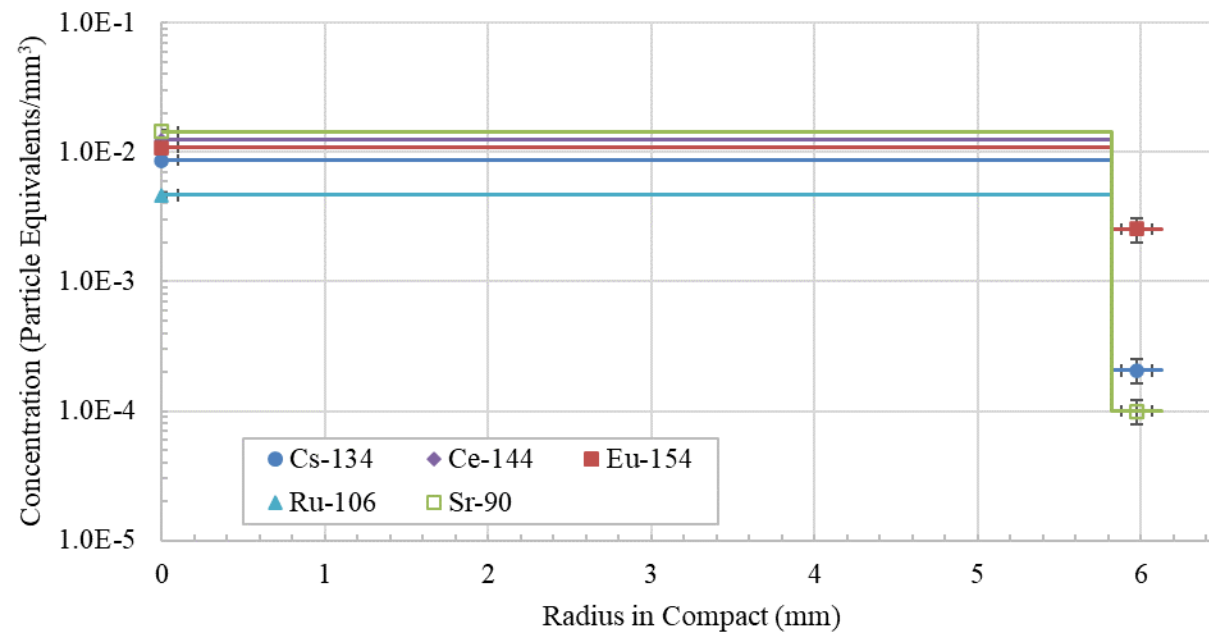
Compact 10-4



Compact 12-1



Compact 12-3





Physical Sampling of Irradiated Rings to Measure Fission Product Concentration Profiles

Completed Construction of Radial Fission Product Profiles in all Carbon Rings in FY2021

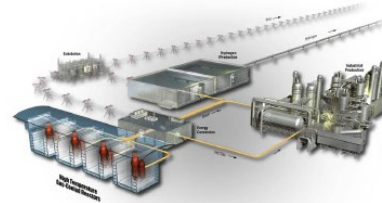
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Measurement of Fission Product Concentration Profiles in AGR-3/4 TRISO Fuel Graphitic Matrix and Nuclear Graphites

June 2021

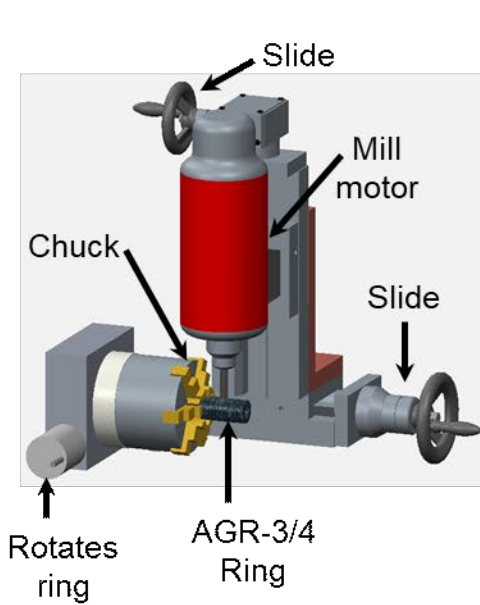
John D. Stempien



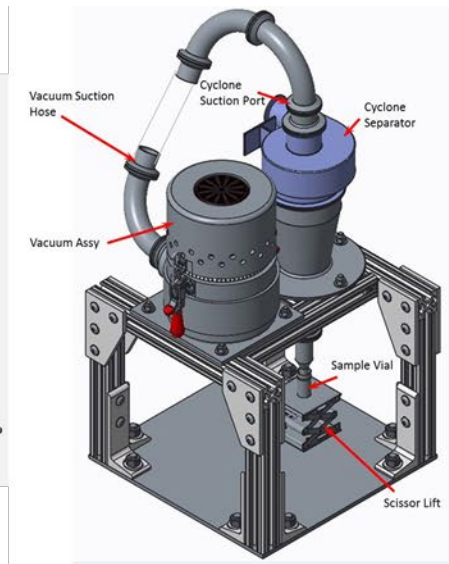
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Physical Sampling for Fission Product Concentration Profiles in Graphite and Graphitic Matrix

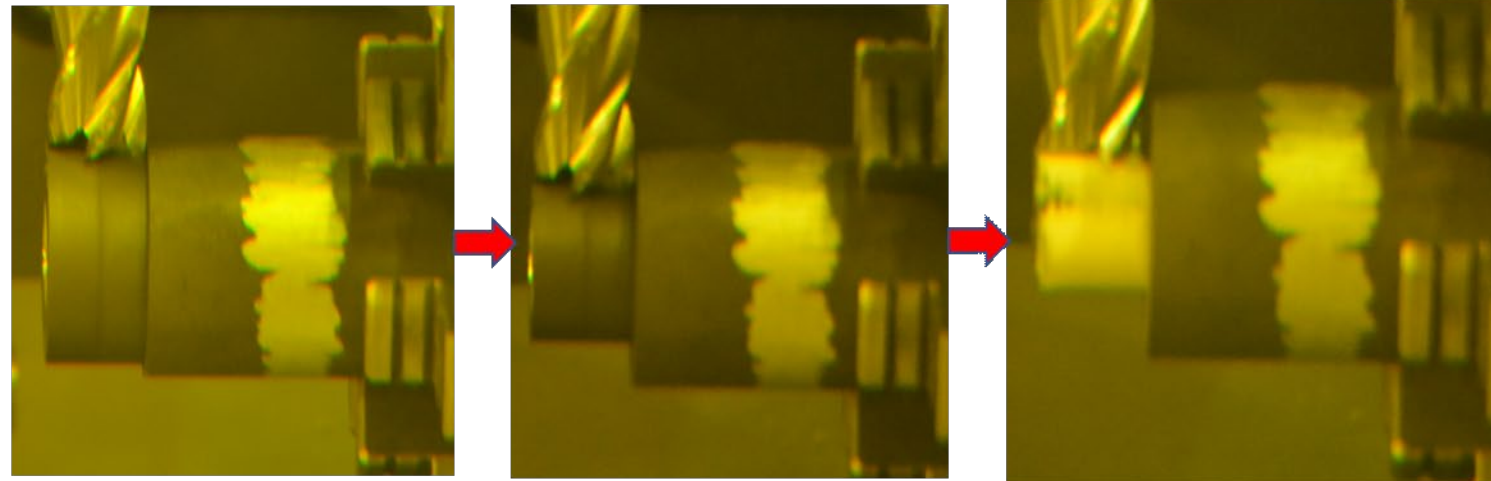
- Machining samples from the graphite rings to measure fission product radial profiles within rings
 - Progressively remove radial segments from rings at one or two axial locations
 - Collected material is gamma scanned and burn-leached for Sr-90 analysis
 - Refine models, compare to PGS, and derive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



Material Removal



Fines Collection



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

Capsule 3 Ring Profiles

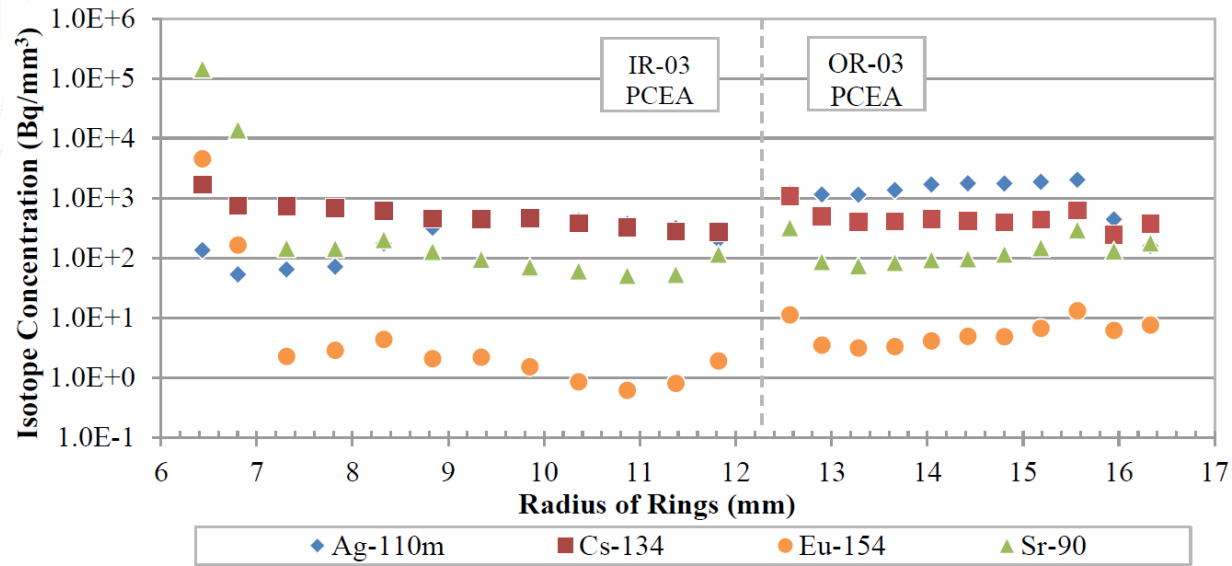


Figure 15. Radial profiles for select fission products at the axial top of the Capsule 3 rings.

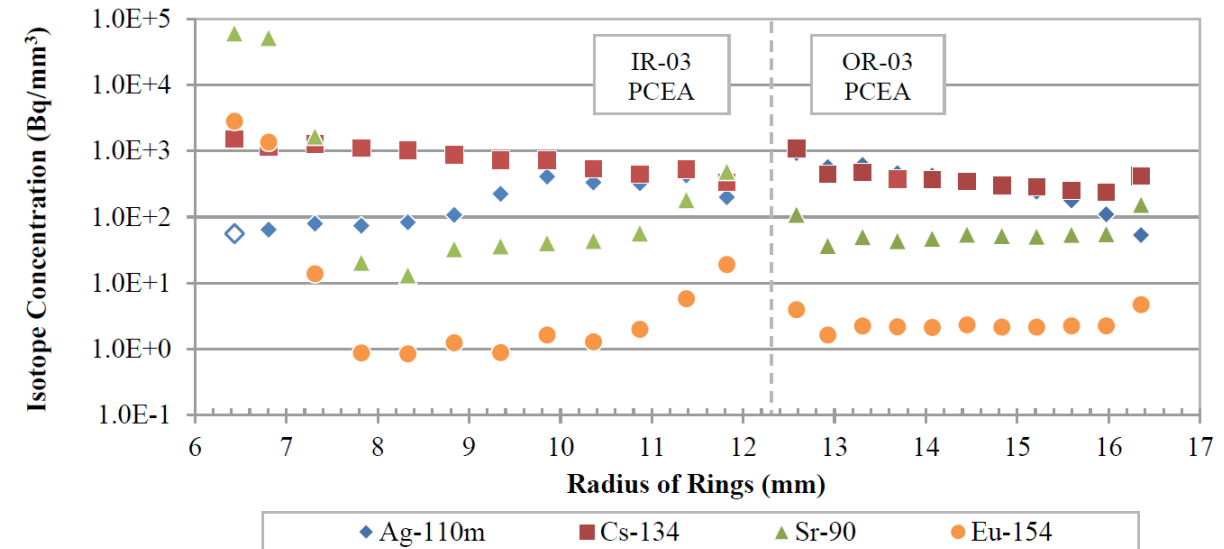


Figure 16. Radial profiles for select fission products at the axial center of the Capsule 3 IRs and ORs. The open symbol for Ag-110m at x = 6.4 mm denotes a value derived from an MDA.

Capsule 5 Ring Profiles

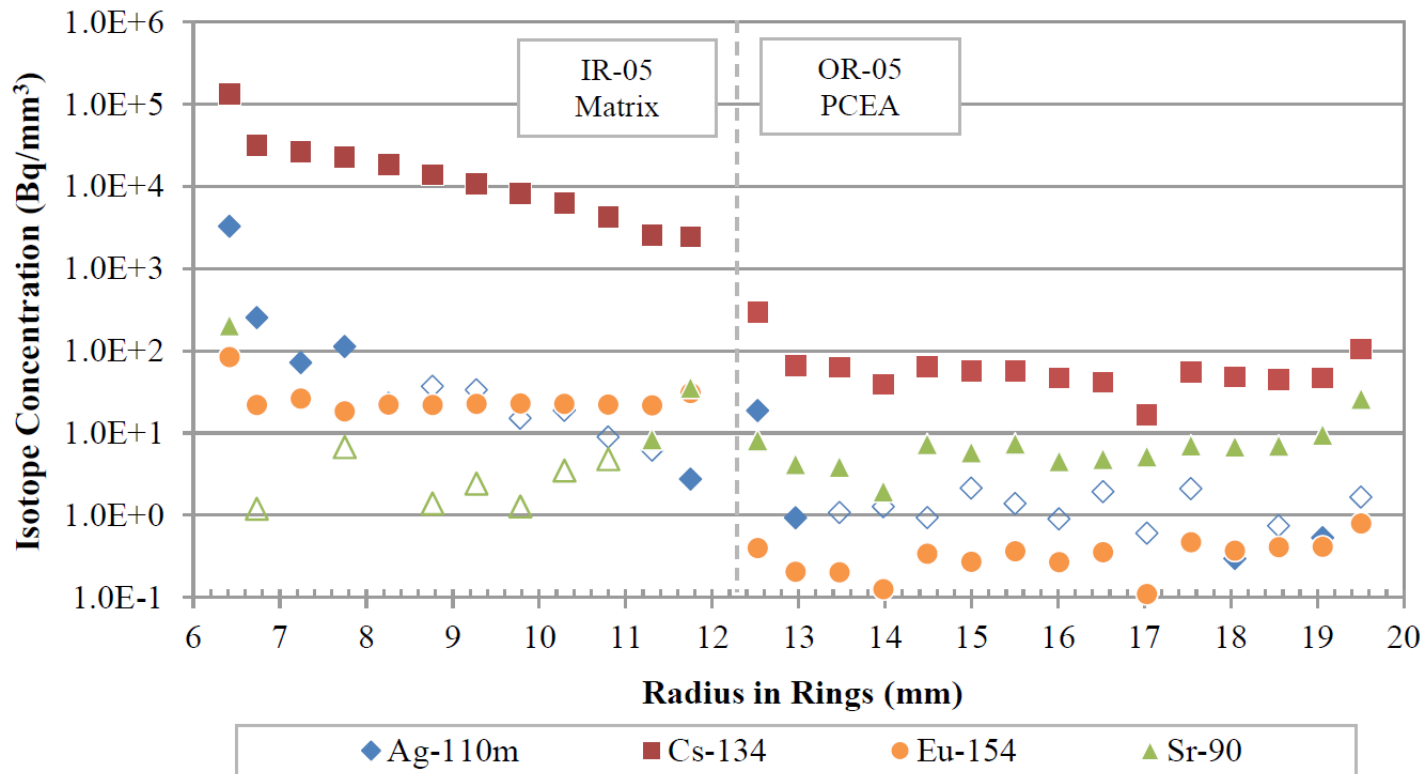


Figure 22. Radial profiles for select fission products at the axial center of the Capsule 5 IR and OR. The open symbols denote values derived from MDAs.

Capsule 7 Ring Profiles

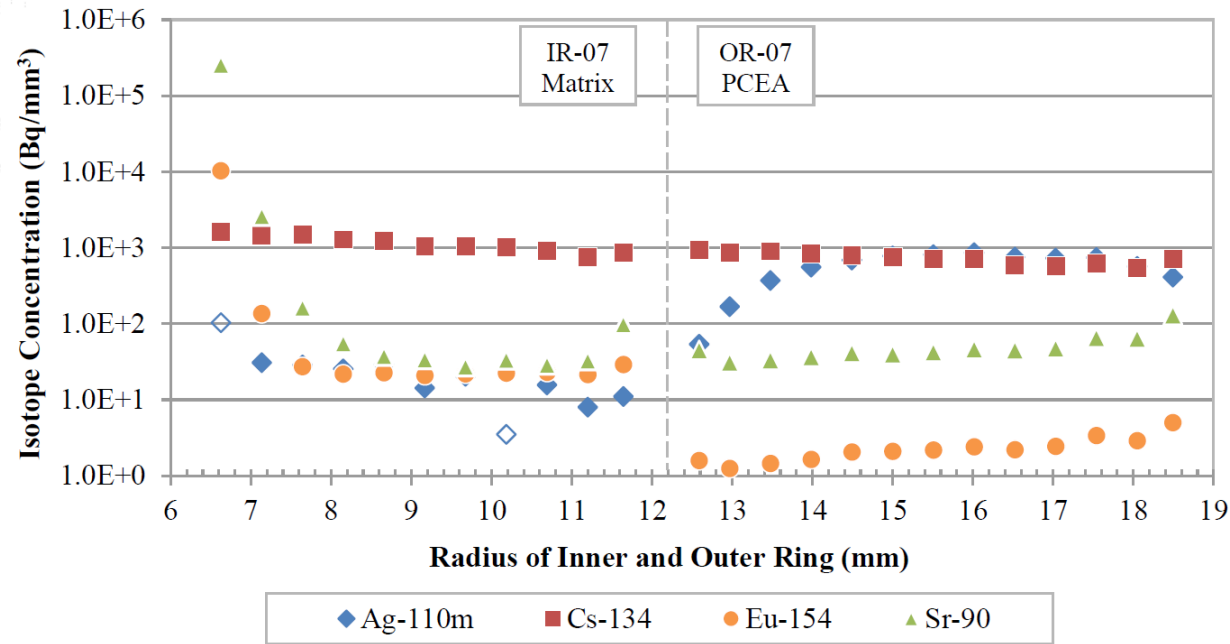


Figure 26. Radial profiles for select fission products at the axial top of the Capsule 7 IR and OR. The open symbols denote values derived from MDAs.

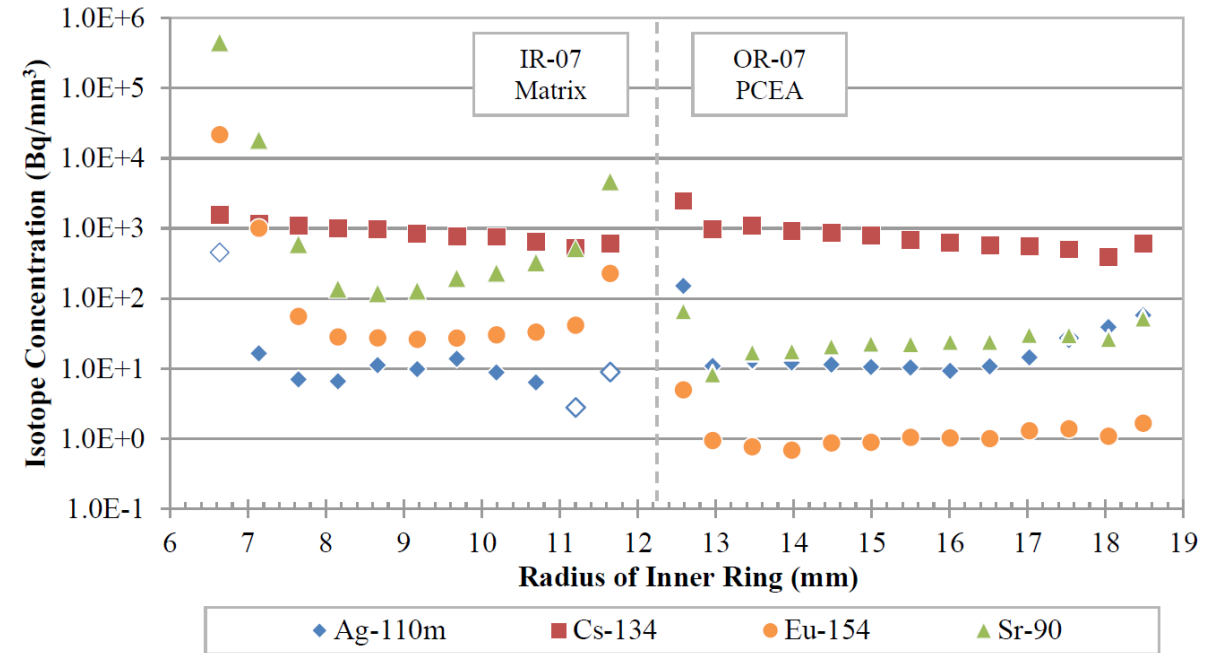


Figure 27. Radial profiles for select fission products at the axial center of the Capsule 7 IR and OR. The open symbols denote values derived from MDAs.

Capsule 8 Ring Profiles

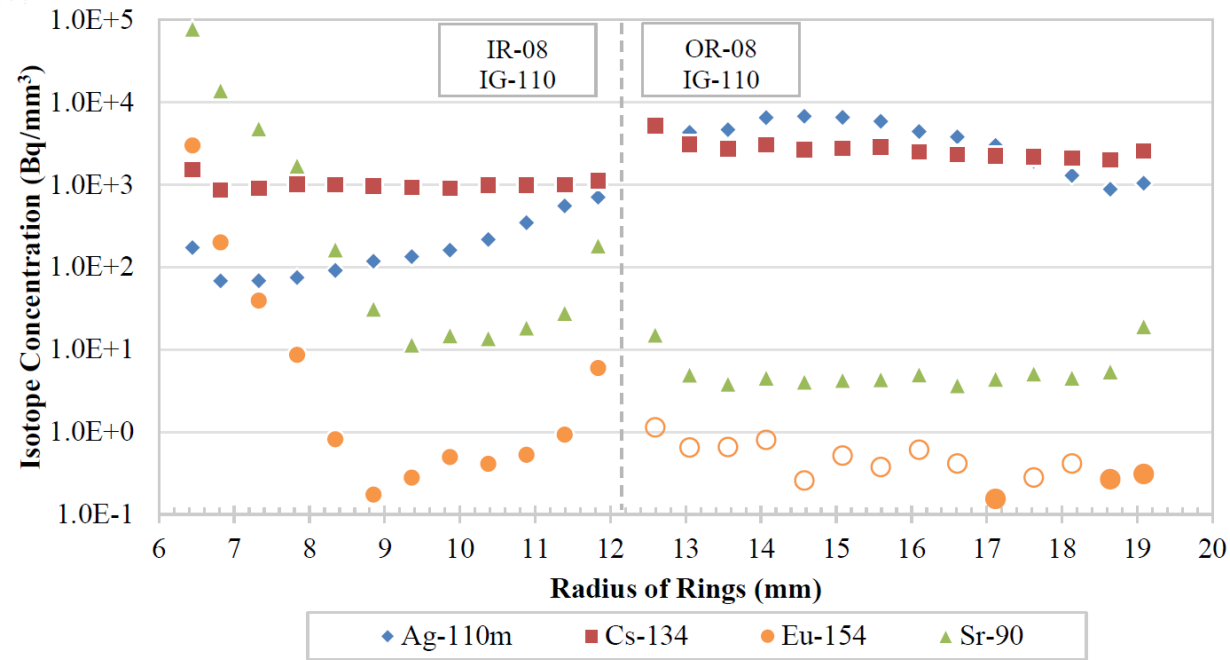


Figure 31. Radial profiles for select fission products at the axial center of the Capsule 8 IR and OR. The open symbols denote values derived from MDAs.

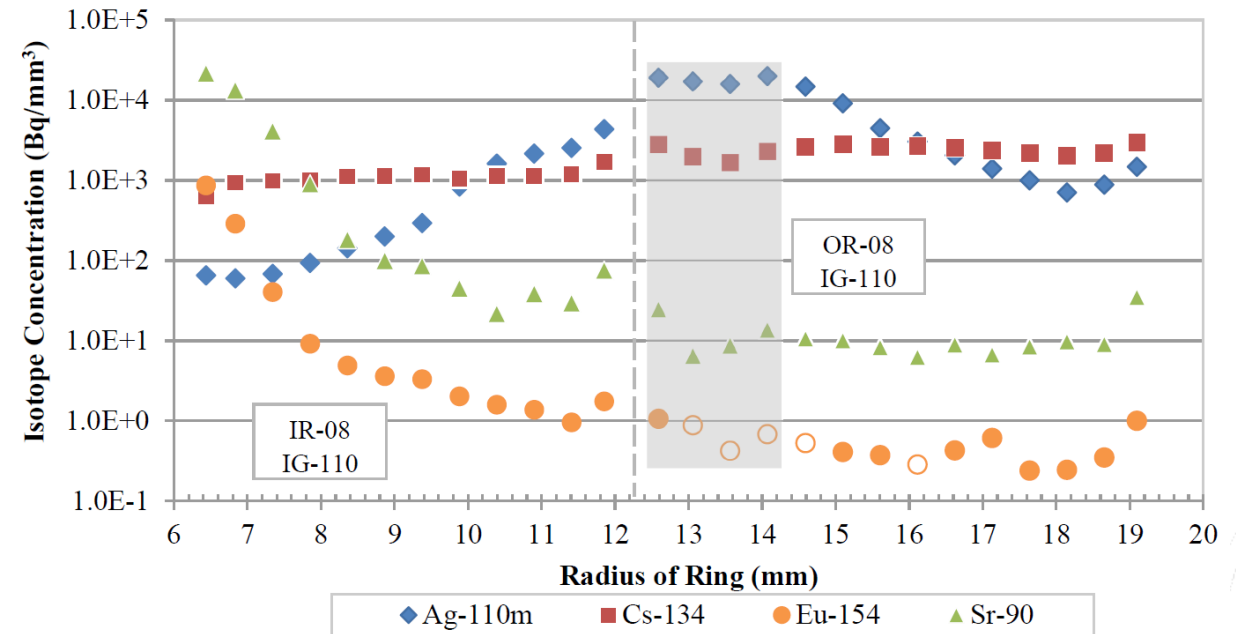


Figure 32. Radial profiles for select fission products at the axial bottom of the Capsule 8 IR and OR. The open symbols denote values derived from MDAs. Gray shading highlights points with greater uncertainty from ring cracking during sampling.

Capsule 10 Ring Profiles

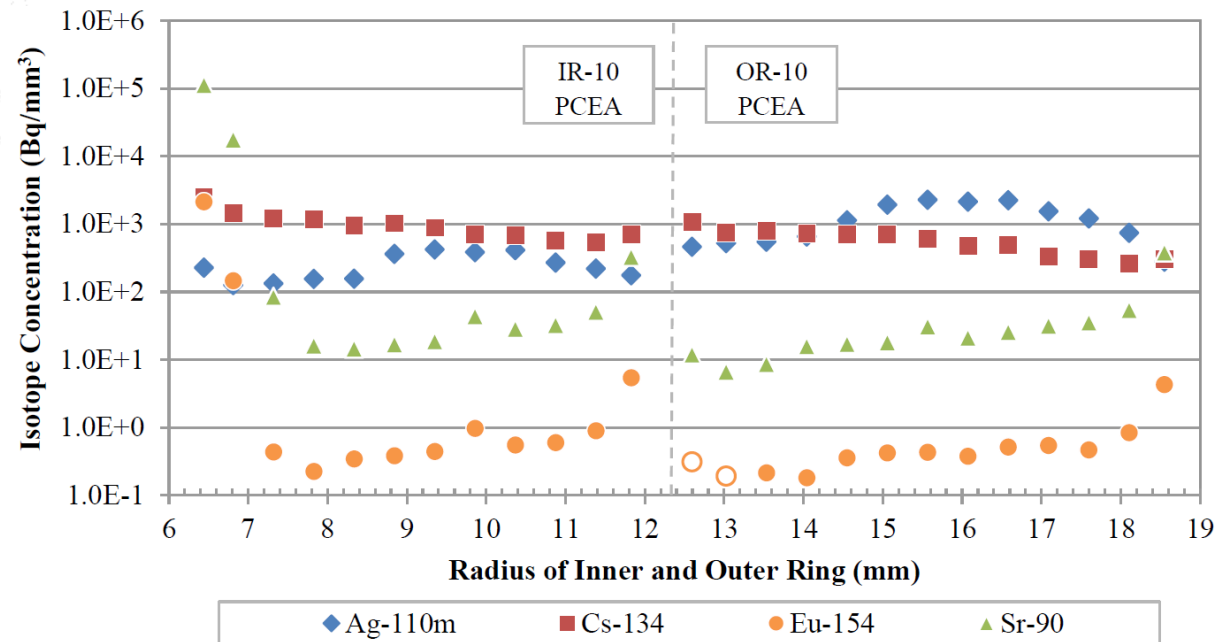


Figure 38. Radial profiles for select fission products at the axial center of the Capsule 10 IR and OR. The open symbols denote values derived from MDAs.

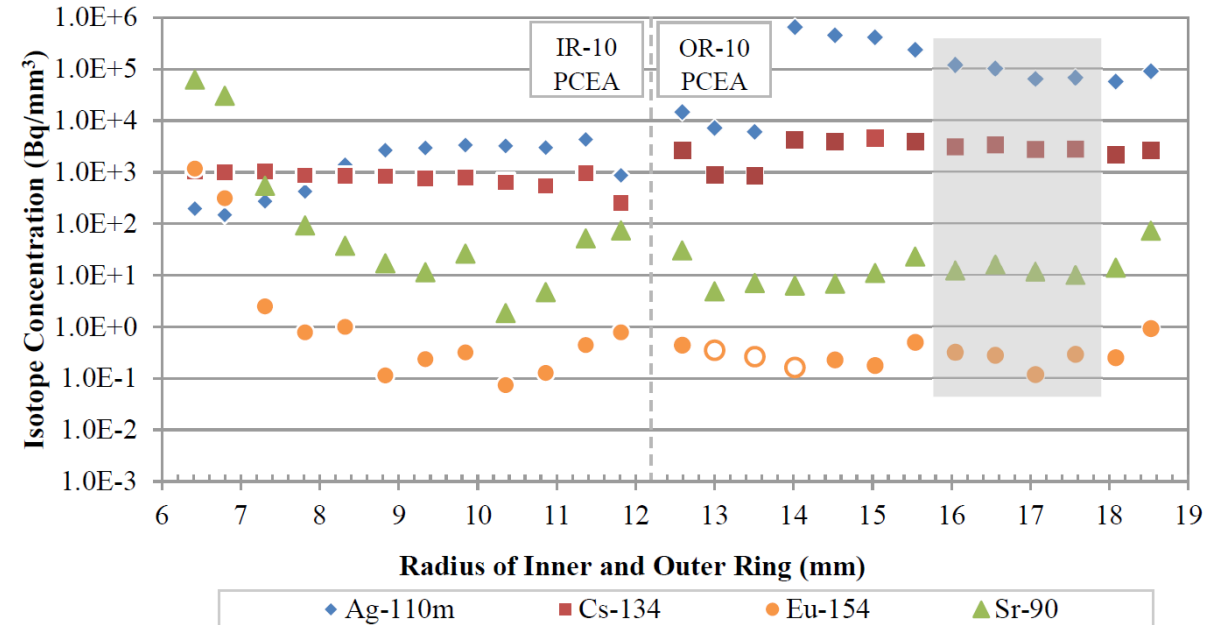


Figure 39. Radial profiles for select fission products at the axial bottom of the Capsule 10 IR and OR. The open symbols denote values derived from MDAs. Gray shading highlights the points with greater uncertainty from ring movement during sampling.

Capsule 12 Ring Profiles

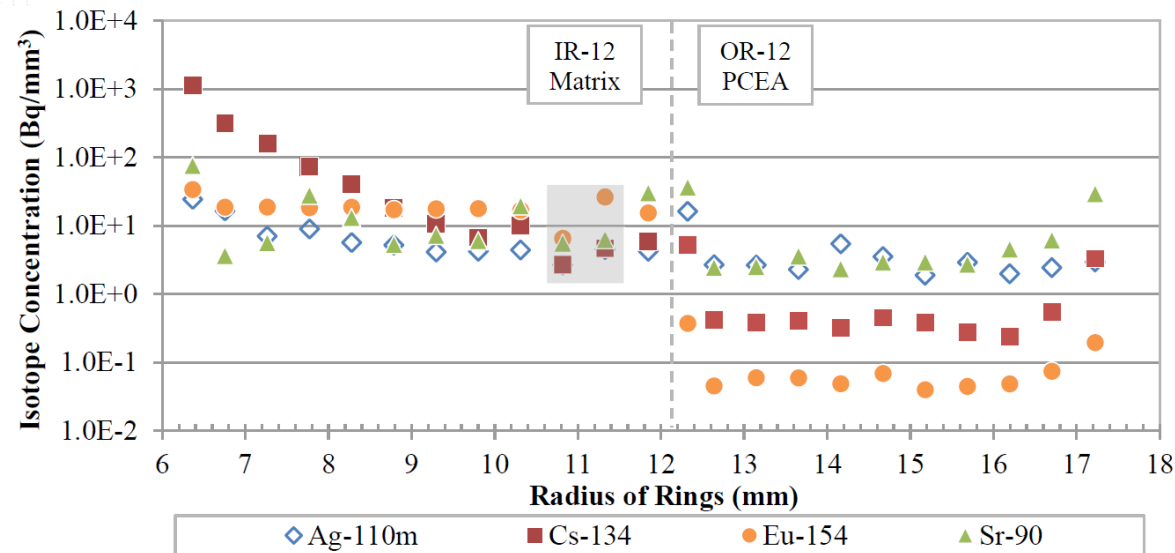


Figure 45. Radial profiles for select fission products at the axial center of the Capsule 12 IR and OR. The open symbols denote values derived from MDAs. Gray shading shows the IR-12 segments where roughly 60% of the third segment (around $x = 10.8$ mm) was collected in the vial for the second segment (around $x = 11.4$ mm).

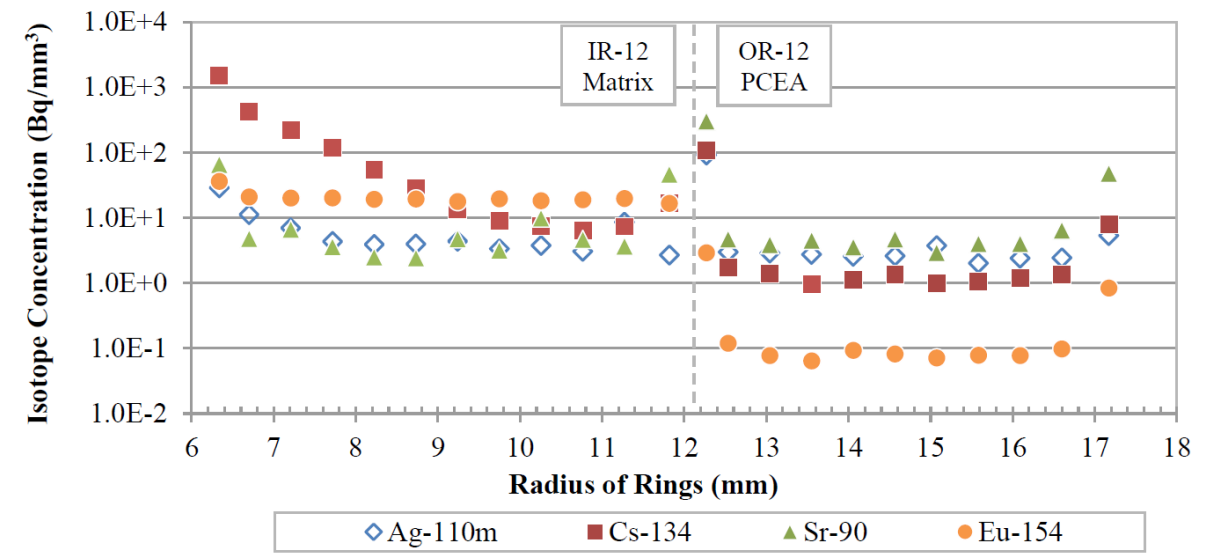


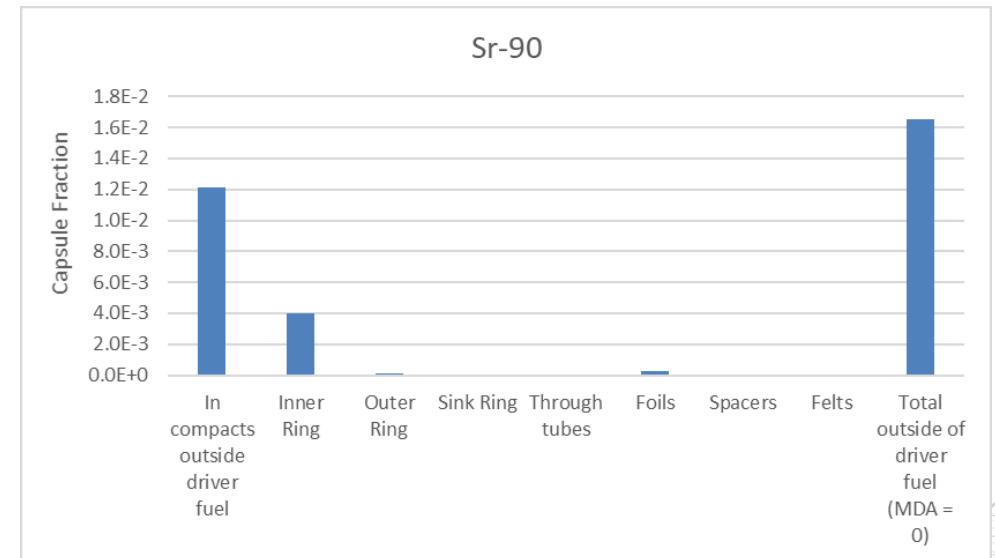
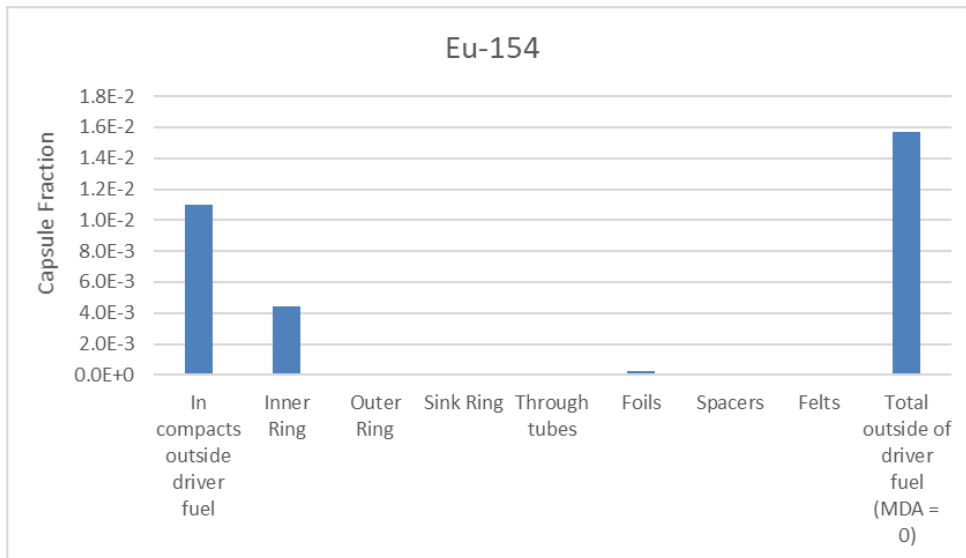
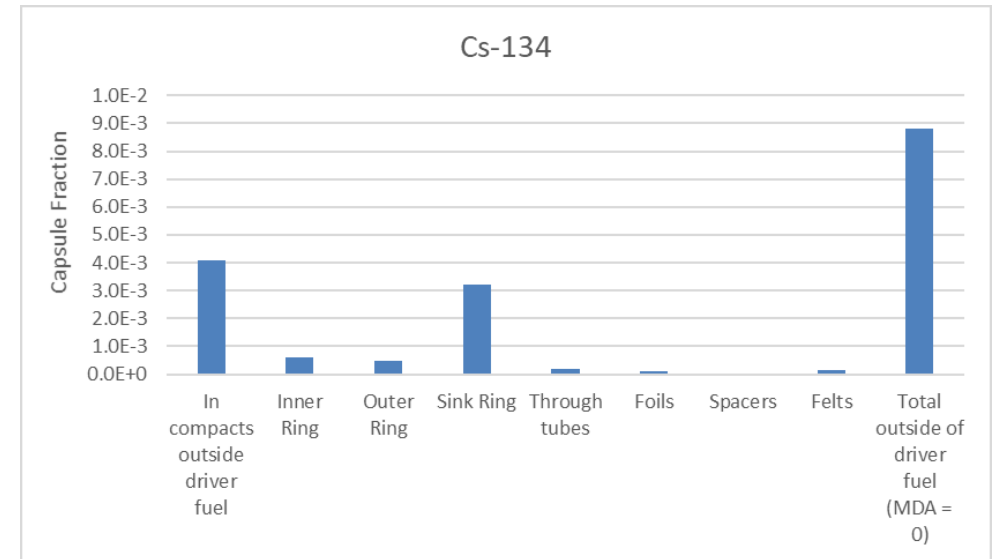
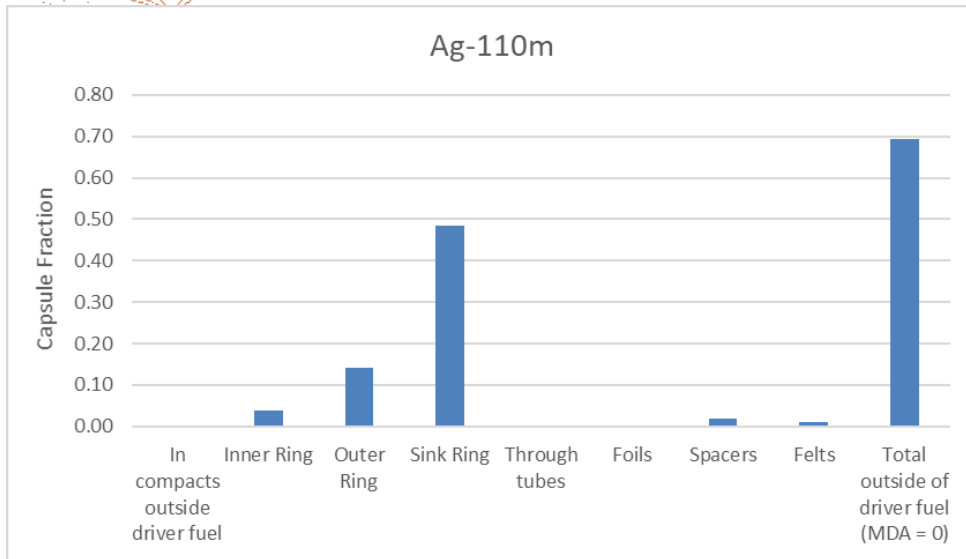
Figure 46. Radial profiles for select fission products at the axial bottom of the Capsule 12 IR and OR. The open symbols denote values derived from MDAs.



AGR-3/4 Mass Balance Outside of Driver Fuel SiC

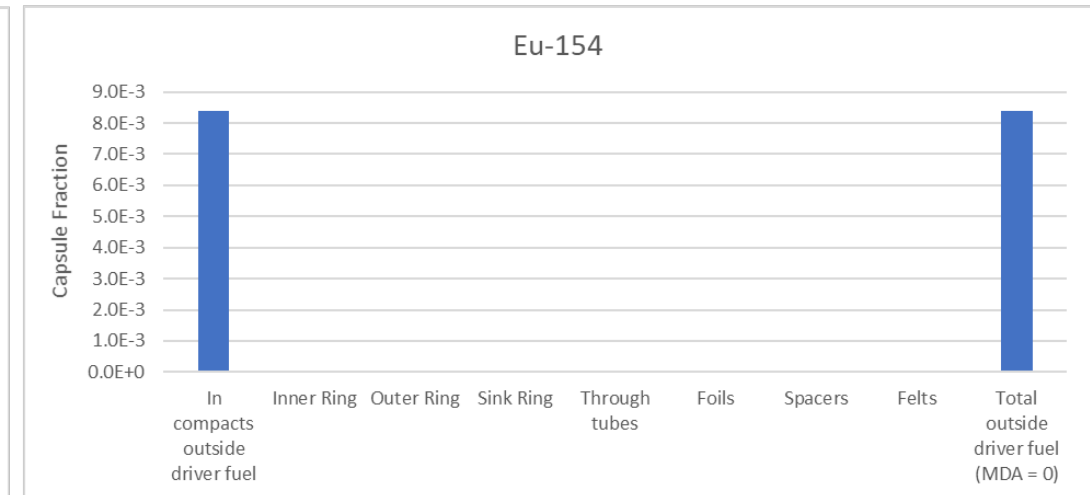
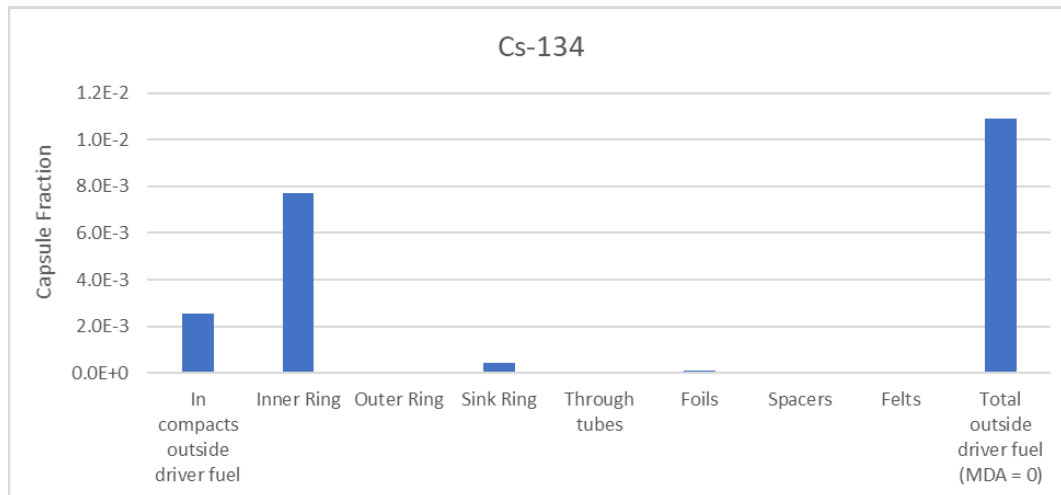
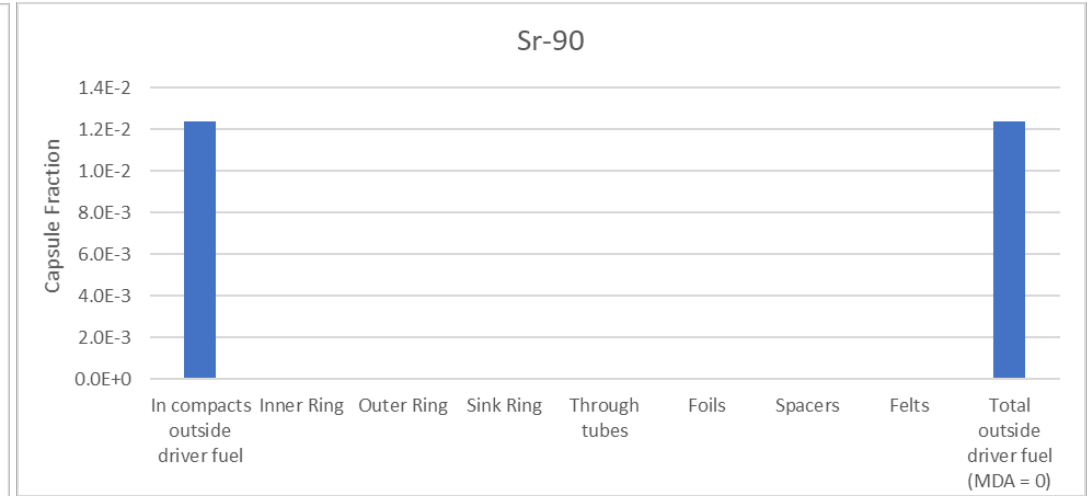
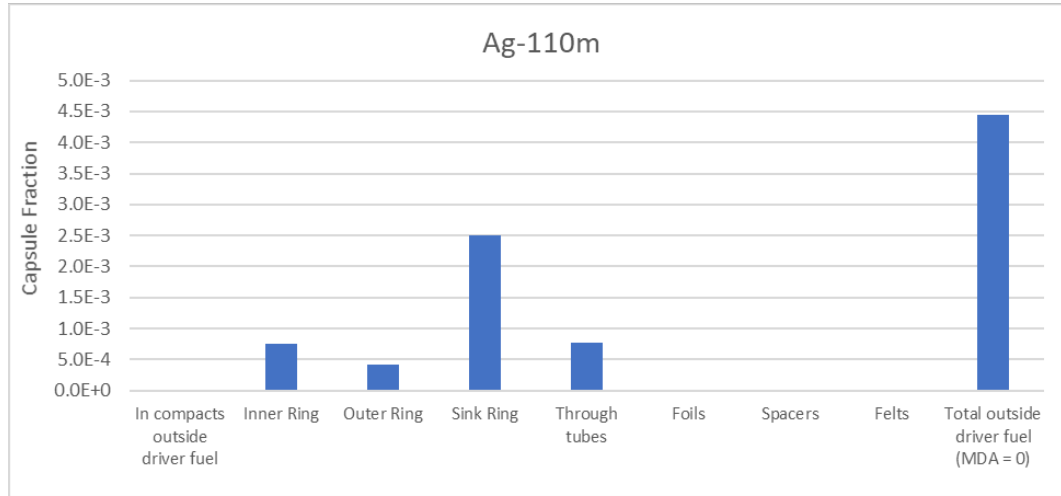
Capsule 3

Use this information to adjust the source from PARFUME used in the AGR-3/4 transport model.



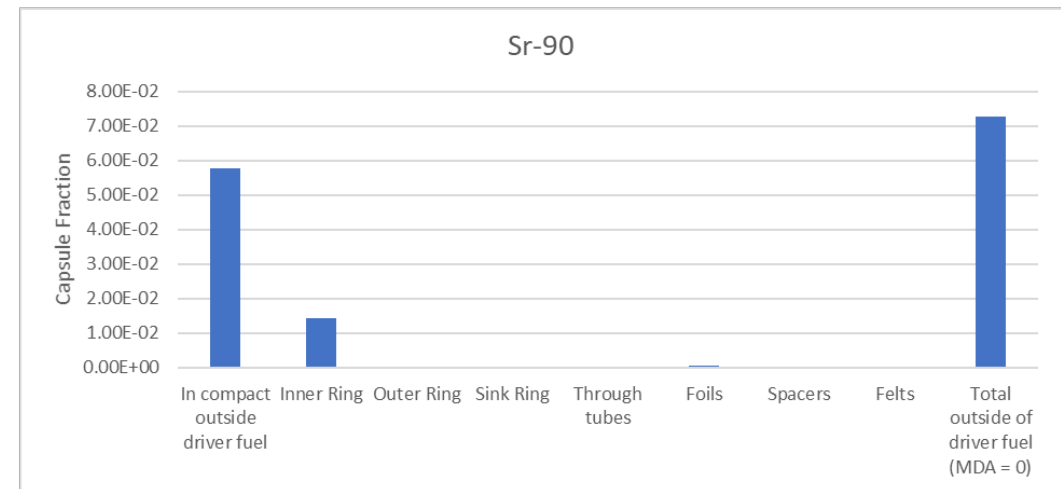
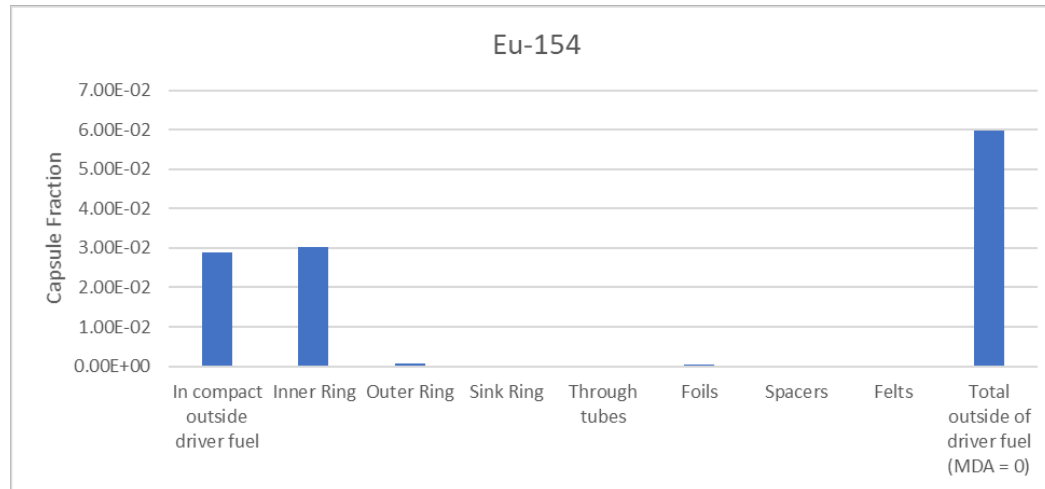
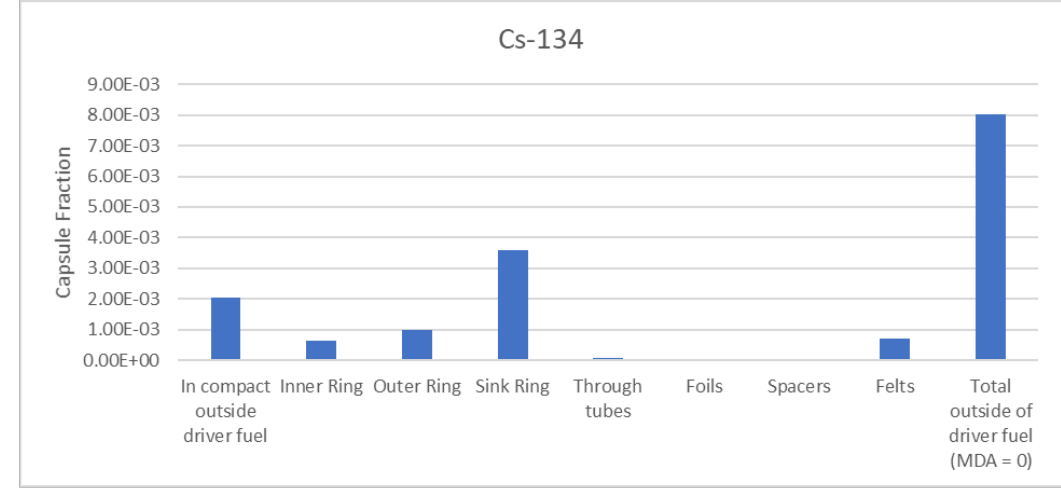
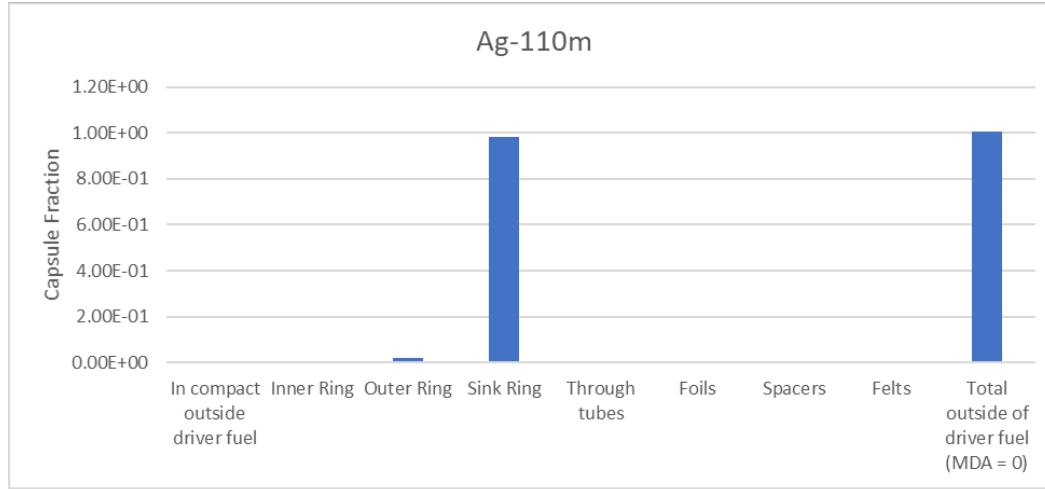
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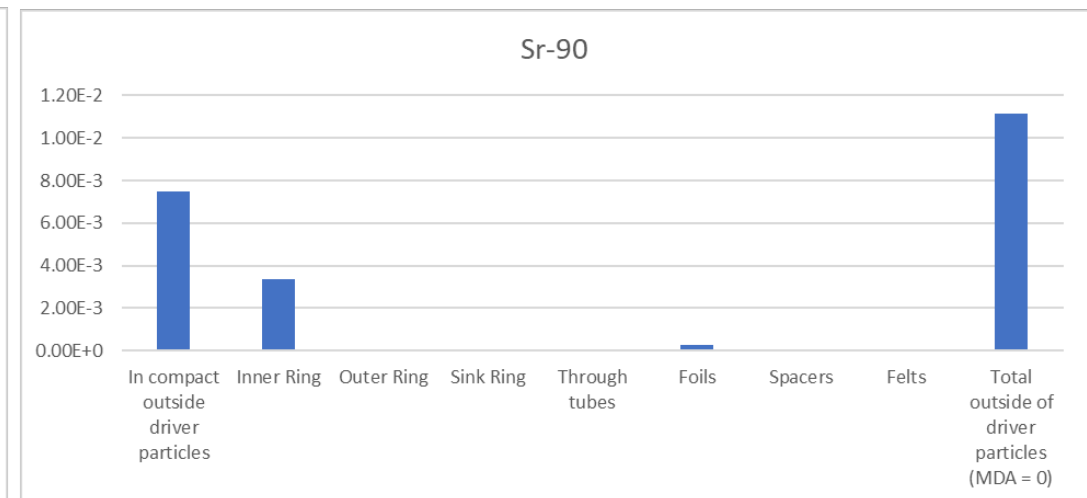
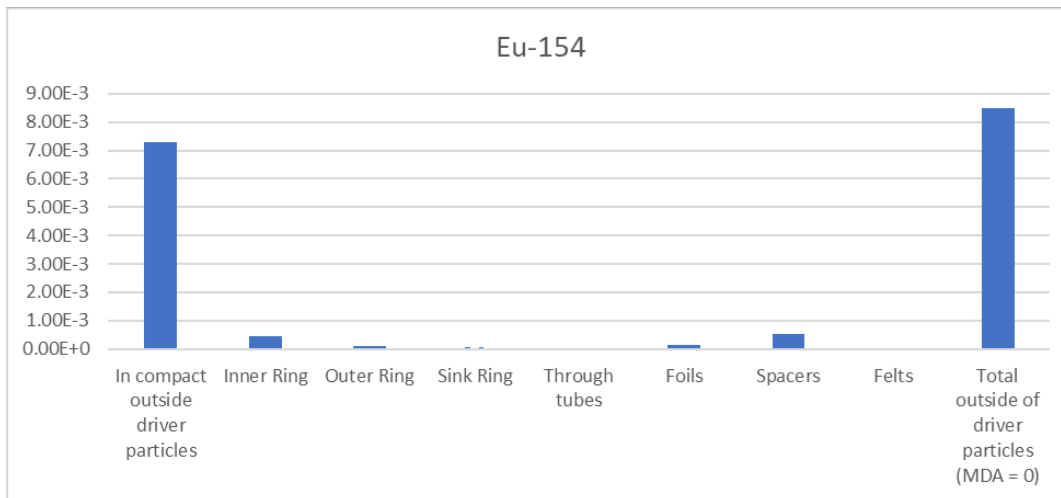
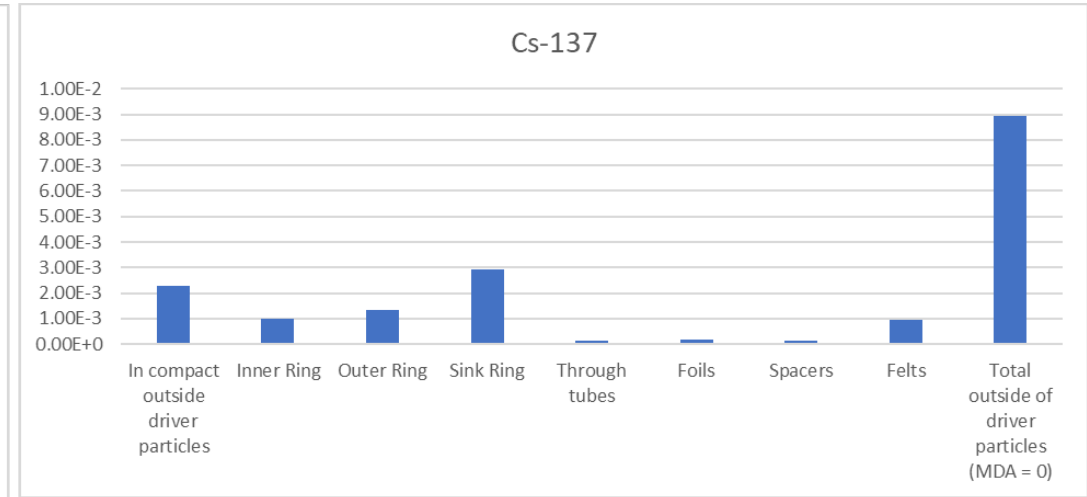
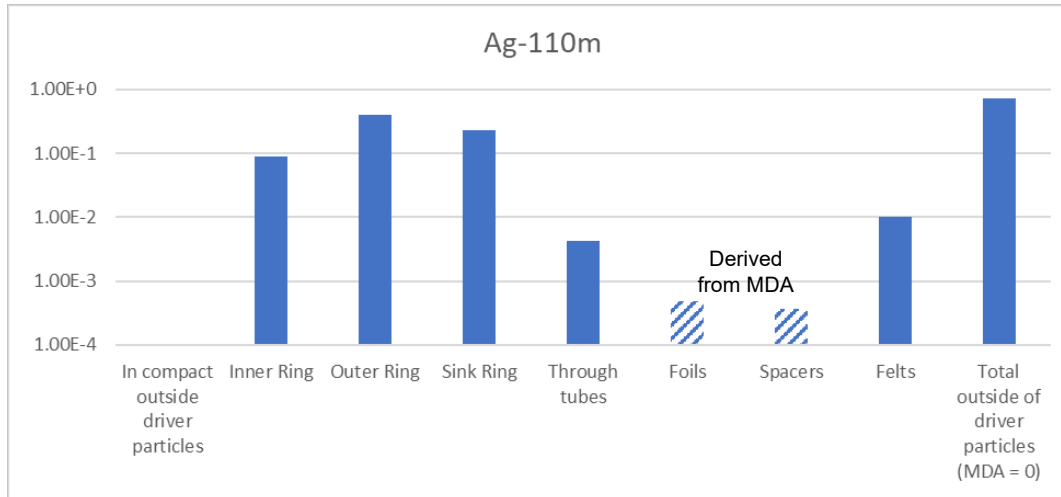
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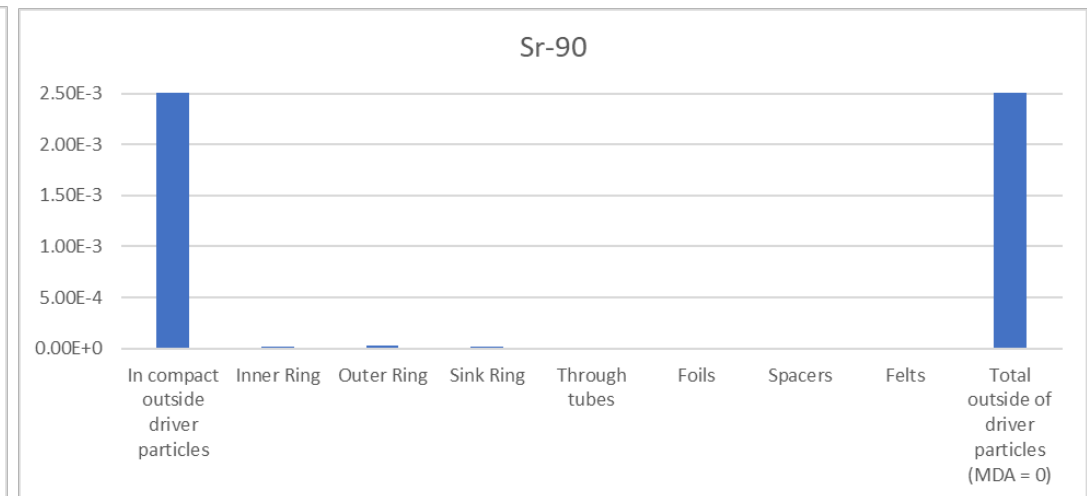
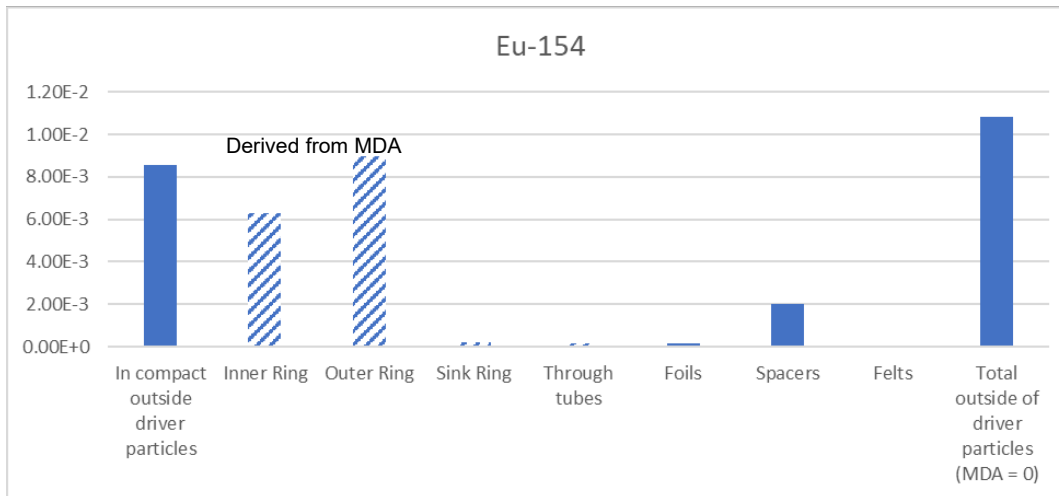
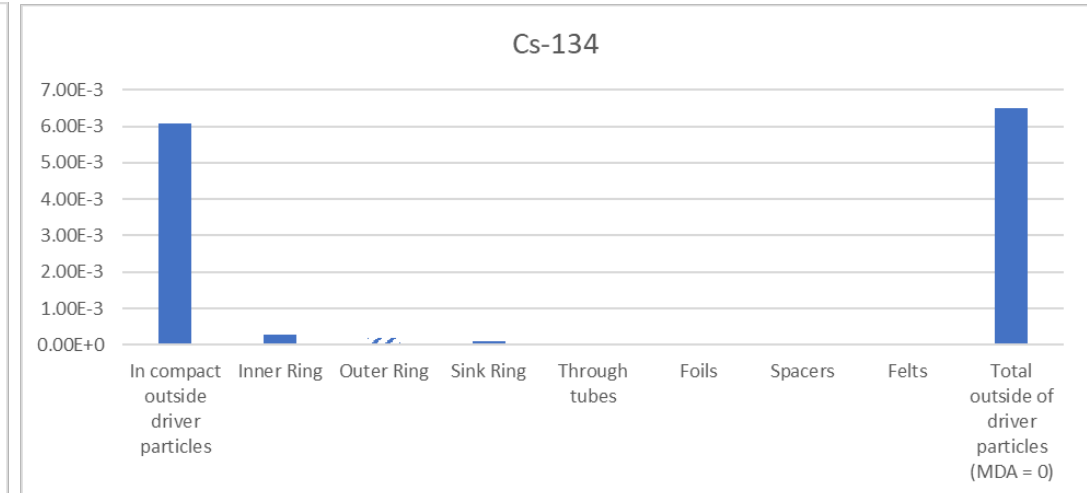
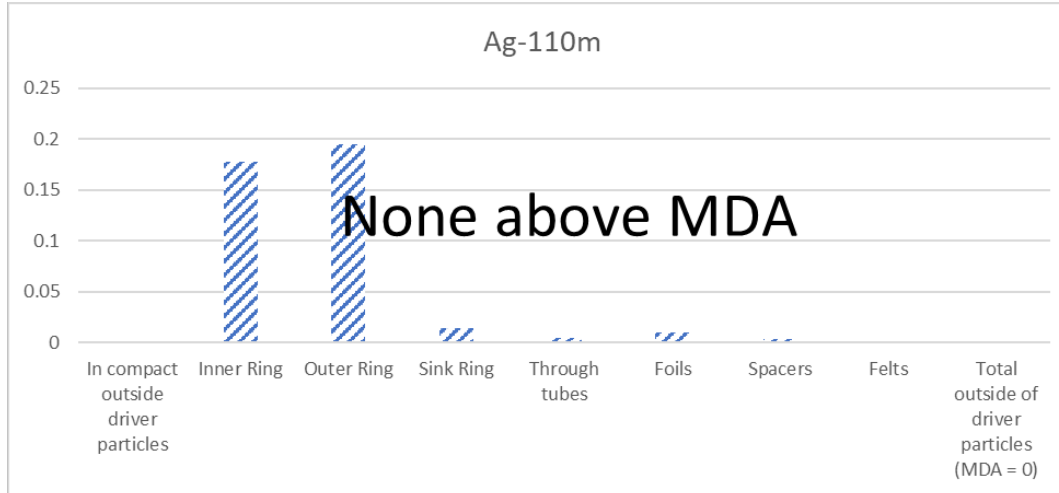
Capsule 10

Use this information to adjust the source from PARFUME used in the AGR-3/4 transport model.



Capsule 12

Use this information to adjust the source from PARFUME used in the AGR-3/4 transport model.



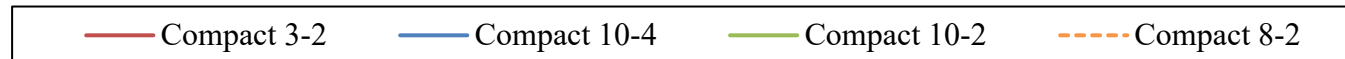
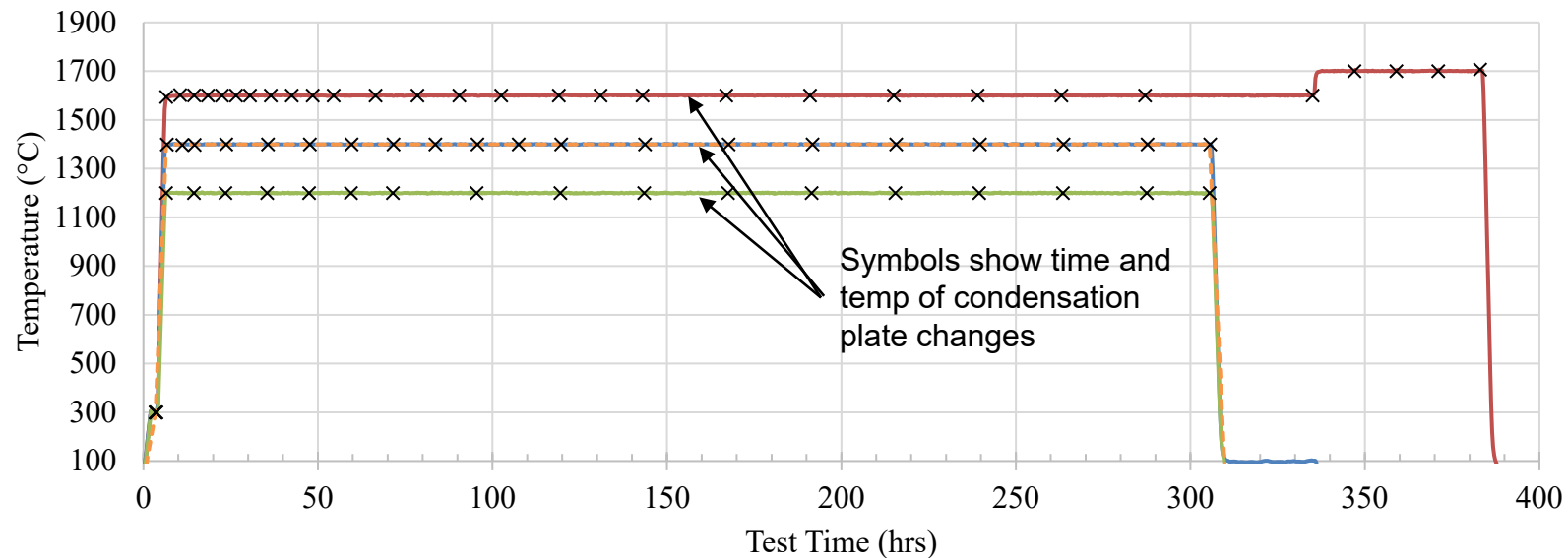


AGR-3/4 Compact Heating Tests

Tests of As-Irradiated Compacts Were Completed in 2018

Compact ID	Burnup (% FIMA)	Fast Fluence (n/m ² , E > 0.18 MeV)	TAVA Irradiation Temp (°C)	Heating Test Temp (°C)
3-2	12.5	4.17E+25	1196	1600/1700
8-2	14.6	5.11E+25	1213	1400
10-2	12.0	4.01E+25	1213	1200
10-4	11.4	3.75E+25	1168	1400

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



Proceedings of HTR 2018
Warsaw, Poland, October 8-10, 2018
Paper HTR 2018-3023

Preliminary results from the first round of post-irradiation heating tests of fuel compacts from the AGR-3/4 irradiation

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Abstract – Three post-irradiation heating tests of fuel compacts from the US AGR-3/4 irradiation experiment were completed. In addition to TRISO-coated driver fuel, each compact contained particles with UCO fuel kernels coated only in pyrocarbon to simulate exposed kernels. Tests at 1600/1700°C, 1400°C, and 1200°C were performed to measure fission product releases as a function of time and temperature. Silver releases were highest in the 1200°C test, supporting the observation that silver release rates are highest in the temperature range of 1100 to 1300°C. Except for Ag-110m (which was released in multiple particle equivalents), releases of Cs-134, Kr-85, Eu-154, and Sr-90 were all less than one particle inventory per compact. This suggests that exposed kernels retain little Kr-85 after irradiation. Compared to tests of AGR-1 compacts with no exposed kernels, the Cs-134 and Kr-85 releases were noticeably higher, and at 1600°C the exposed kernels contributed noticeably more Eu and Sr than in 1600°C tests of AGR-1 fuel. These data can be used to make inferences on retention of fission products in exposed kernels.

1 INTRODUCTION

The US Advanced Gas Reactor (AGR) fuel development and qualification program has fabricated and irradiated tristructural isotropic (TRISO)-coated particle fuels for high-temperature gas-cooled reactors (HTGRs). Three campaigns of fuel fabrication and irradiation in the Advanced Test Reactor (ATR) at the Idaho National Laboratory (INL) have already been completed [1]-[3]. In chronological order, those experiments were titled AGR-1, AGR-2, and AGR-3/4. Destructive and non-destructive post-irradiation examination (PIE) of AGR-1 is complete [4], and PIE of AGR-2 and AGR-3/4 is in progress. The fourth and final AGR fuel irradiation experiment (AGR-5/6/7) began in February 2018.


Whereas AGR-1 and AGR-2 were intended to demonstrate performance of TRISO fuel, the AGR-3/4 irradiation experiment was designed to investigate the release of fission products from exposed kernels and their migration in fuel compact graphite matrix and structural graphite materials. This is an essential area of study needed to refine

fission product transport models and support calculations of fission product releases from the reactor core. The objective was accomplished using “designed-to-fail” (DTF) particles in each AGR-3/4 compact that provided a source of fission products to be released during the irradiation. As part of AGR-3/4 PIE, heating tests of AGR-3/4 fuel compacts are being used to study fission product releases from fuel kernels and fission product transport in the compact matrix under a range of temperatures representative of normal reactor operation and postulated reactor accidents.

II. AGR-3/4 FUEL AND IRRADIATION

II.A. Fuel Description

AGR-3/4 fuel kernels were a heterogeneous mixture of uranium carbide and uranium oxide (UCO) enriched to 19.7 wt % in U-235. Kernels were nominally 350-μm in diameter and were produced at BWX Technologies Nuclear Operations Group (Lynchburg, VA USA).



Three AGR-3/4 Compact Reirradiation Heating Tests Were Completed. Two Remain.

Compact Irradiation Histories Prior to Reirradiation

Compact ID	Burnup (% FIMA)	TAVA Irradiation Temp (°C)	Heating Test Temp (°C)
3-1	12.2	1138	1600
8-1	14.5	1165	1200
10-1	12.1	1172	1400
Planned in FY21: 4-3	14.3	1035	1000
Planned in FY21: 1-2	5.9	941	1400

Proceedings of HTR 2021
Yogyakarta, Indonesia, June 24-26, 2021
Paper HTR 2021-3004

Reirradiation and Heating Testing of AGR-3/4 TRISO Fuels

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Abstract – Three irradiated fuel compacts, each with about 1,898 TRISO-coated particles and 20 designed-to-fail (DTF) particles with kernels coated only in pyrocarbon, were reirradiated and then heated at temperatures from 1200 to 1600°C. Reirradiation enabled measurement of the releases of short-lived fission products I-131 and Xe-133 from the DTF kernels. At a given temperature, the amounts of I-131 and Xe-133 released agreed within about 20%. At 1600°C and 1400°C, the total amounts of I-131 released ranged from 32-46% and the total amount of Xe-133 released ranged from 36-40% of the DTF kernel inventories. I-131 and Xe-133 releases at 1200°C were close to 10 times lower than those at 1600°C and 1400°C. This indicates significant kernel retention of these isotopes, particularly at temperatures below 1400°C. Comparing Cs-134 released from the compacts during heating tests to Cs-134 detected through destructive exams in unirradiated compacts suggests that even with 20 exposed kernels, up to several particles worth of Cs-134 is still retained in the compacts outside of the intact SiC layers after the heating tests.

I. INTRODUCTION

The Advanced Gas Reactor (AGR) Fuel Development and Qualification Program was established to perform research and development on tristructural isotropic (TRISO)-coated particle fuel to support deployment of a high-temperature gas-cooled reactor (HTGR) in the United States. The AGR-1 and AGR-2 experiments focused on fabricating and demonstrating the irradiation performance of mixed uranium carbide/uranium oxide (UCO) TRISO fuel. In contrast, the AGR-3/4 irradiation experiment was designed to investigate the release of fission products from exposed UCO kernels and the subsequent transport of fission products in fuel compact graphite matrix and structural graphite materials. New diffusion coefficients are being derived from results of AGR-3/4 post-irradiation examinations (PIE) [1]. PIE results are also being compared to calculations from predictive fission product transport models of the AGR-3/4 experiment [2].

Comparatively little data are available on the behavior of short-lived fission products in the TRISO fuel system. This is because post-irradiation examination of the fuel typically does not begin until several months after the end of irradiation, and

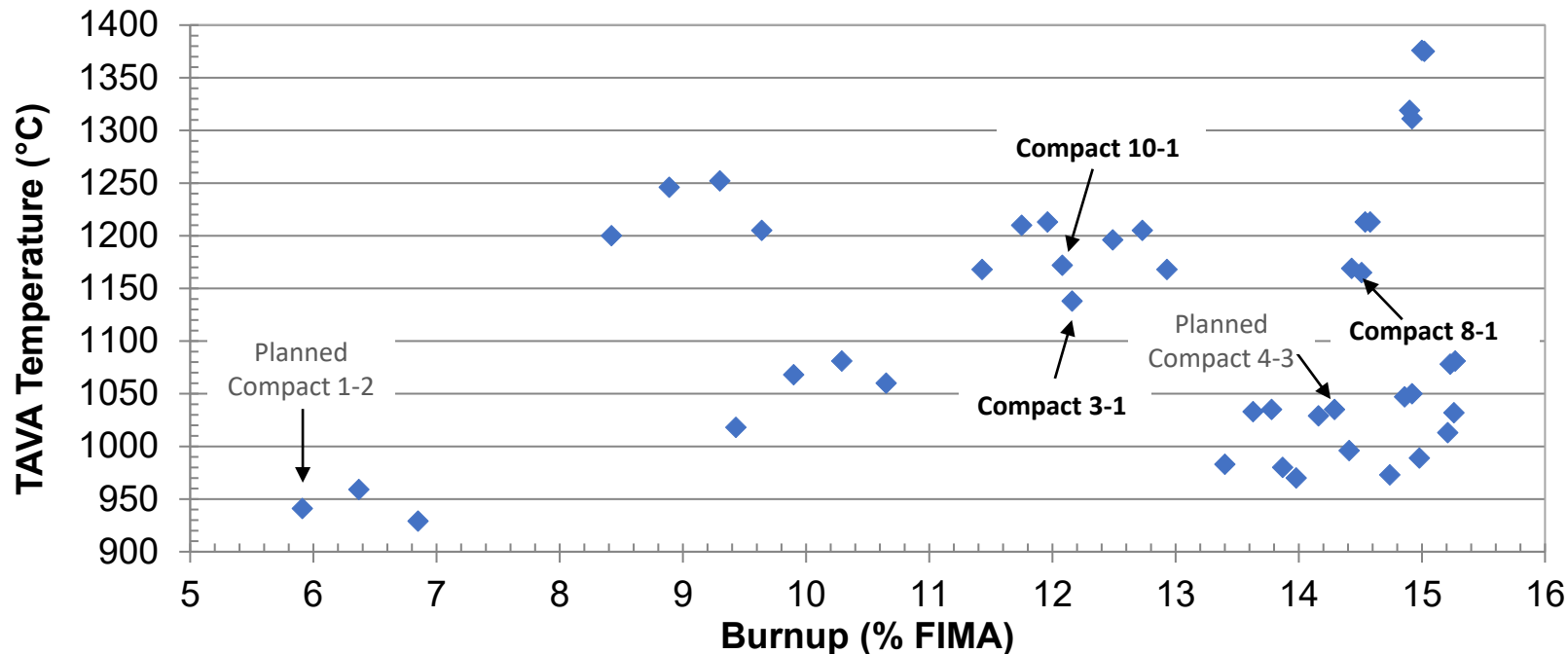
isotopes such as I-131 ($t_{1/2} = 8.02$ d) and Xe-133 ($t_{1/2} = 5.24$ d) have decayed away before PIE can begin.

Understanding the behavior of short-lived I-131, for example, is important because it is a significant contributor to offsite dose during postulated accidents. To study I-131 and Xe-133, the Neutron Radiography (NRAD) Reactor at the Idaho National Laboratory (INL) Hot Fuels Examination Facility (HFEF) was used to reirradiate fuel that was previously irradiated in INL's Advanced Test Reactor (ATR). Reirradiation generates short-lived fission products, and rapidly transferring the fuel from NRAD to the Fuel Accident Condition Simulator (FACS) furnace (also located at HFEF) enables measurement of short-lived fission product releases.

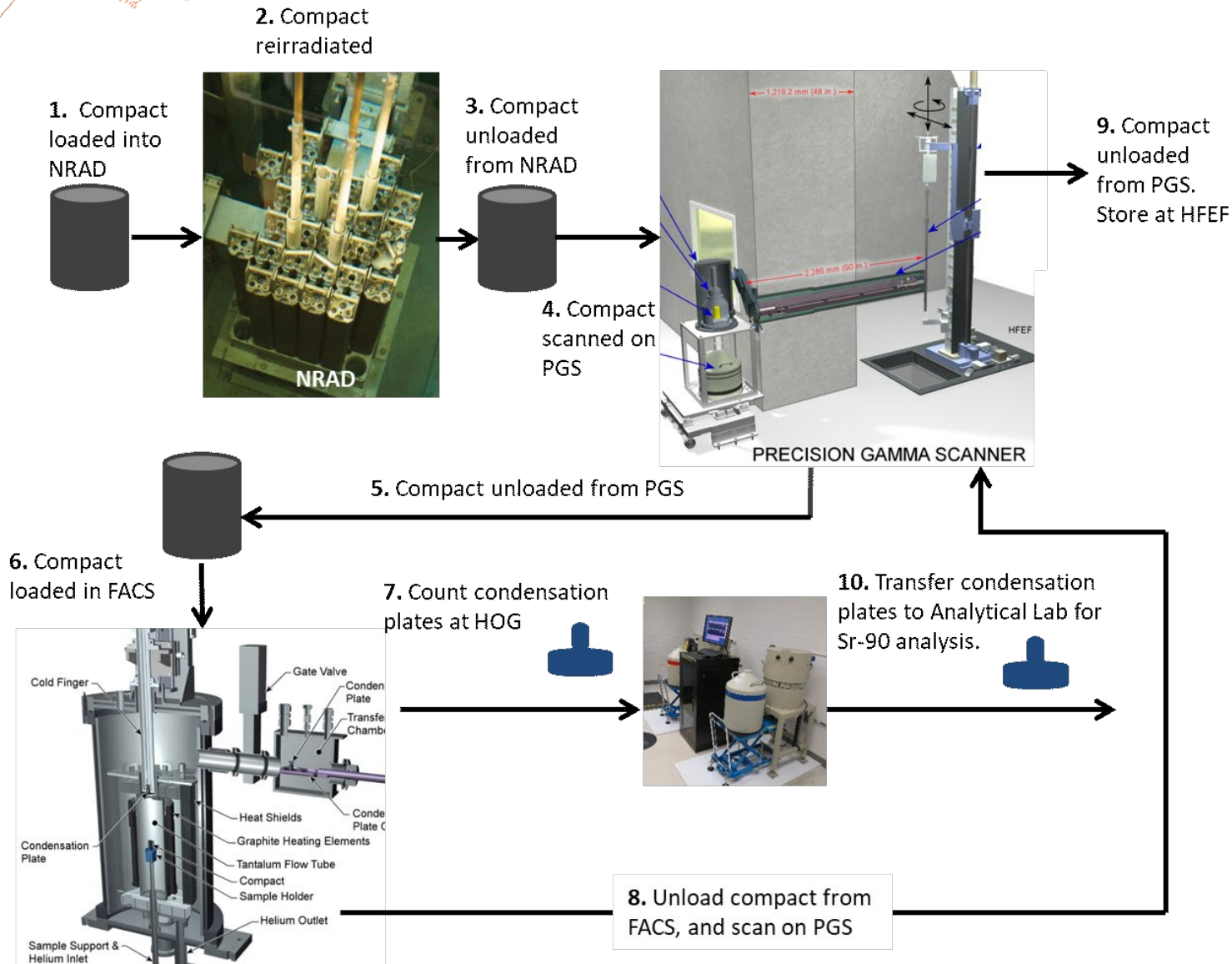
II. AGR-3/4 FUEL AND SAMPLE SELECTION

II.A. Fuel Description

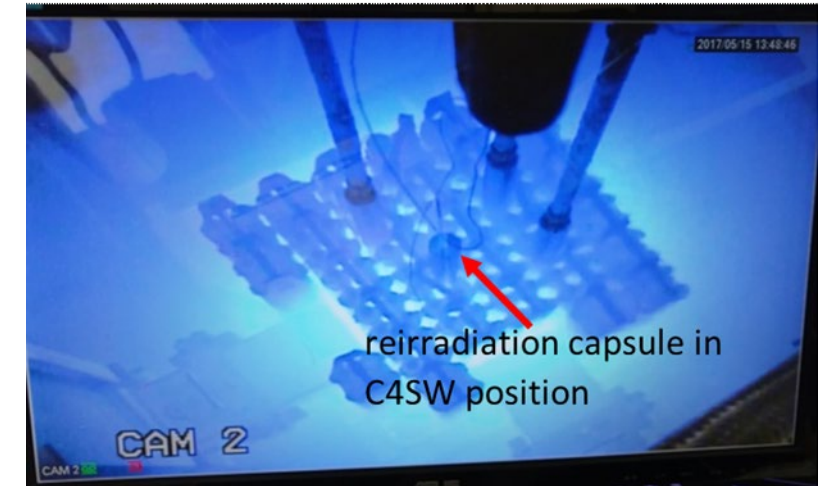
The AGR-3/4 fuel form consisted of cylindrical fuel compacts nominally 1.27 cm in diameter and 1.27 cm in length. Approximately 1,918 fuel particles were in each fuel compact. Of these, exactly 20 particles were designed-to-fail (DTF) particles, coated only with pyrocarbon, and about 1,898 were TRISO-coated particles. Fig. 1 shows an x-



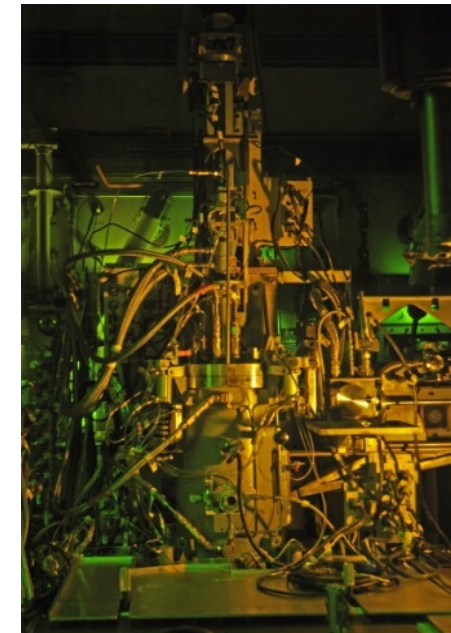
Compact Reirradiation/Heating Tests



Neutron Radiography Reactor (NRAD)



Fuel Accident Condition Simulator (FACS)

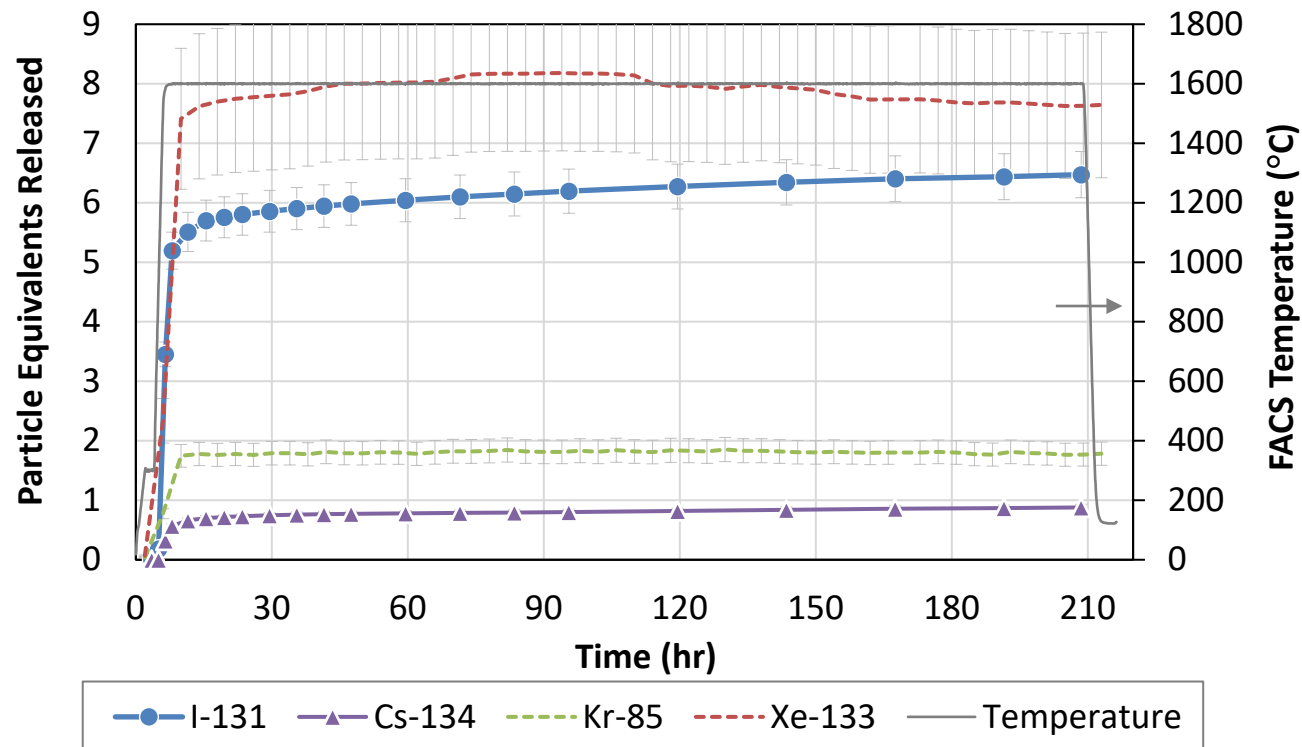


Major Findings from Reirradiation Tests

- Prior irradiation history (e.g., temperature and burnup) affects releases in heating tests.
- I-131 and Xe-133 releases are similar; therefore, Xe-133 is reasonable indicator of I-131 release.
- Kernels may retain some I-131 and Xe-133: 9x less Xe-133 and 5x less I-131 are released at 1200°C than at 1400°C and 1600°C.
- Several DTF particles' worth of Cs-134 likely remains in compacts outside of SiC layers even after heating.
- Two compact reirradiation tests remain to be completed.
- May be possible to determine effective diffusion coefficients from the release data.

Compact 3-1 1600°C Test Summary

- Fission gases and iodine released fastest. I-131 and Xe-133 releases are similar
- Kernels may retain some iodine and xenon
- Compact retained Cs-134 after irradiation that was released in heating test. Based on DLBL and measured releases, up to ~3 DTF particles worth may remain in compact even after heating.
- Short-lived isotope release dominated by inventory in DTF that was generated from reirradiation
- Long-lived isotope release dominated by state of fuel after initial irradiation. Reirradiation does not affect long-lived much

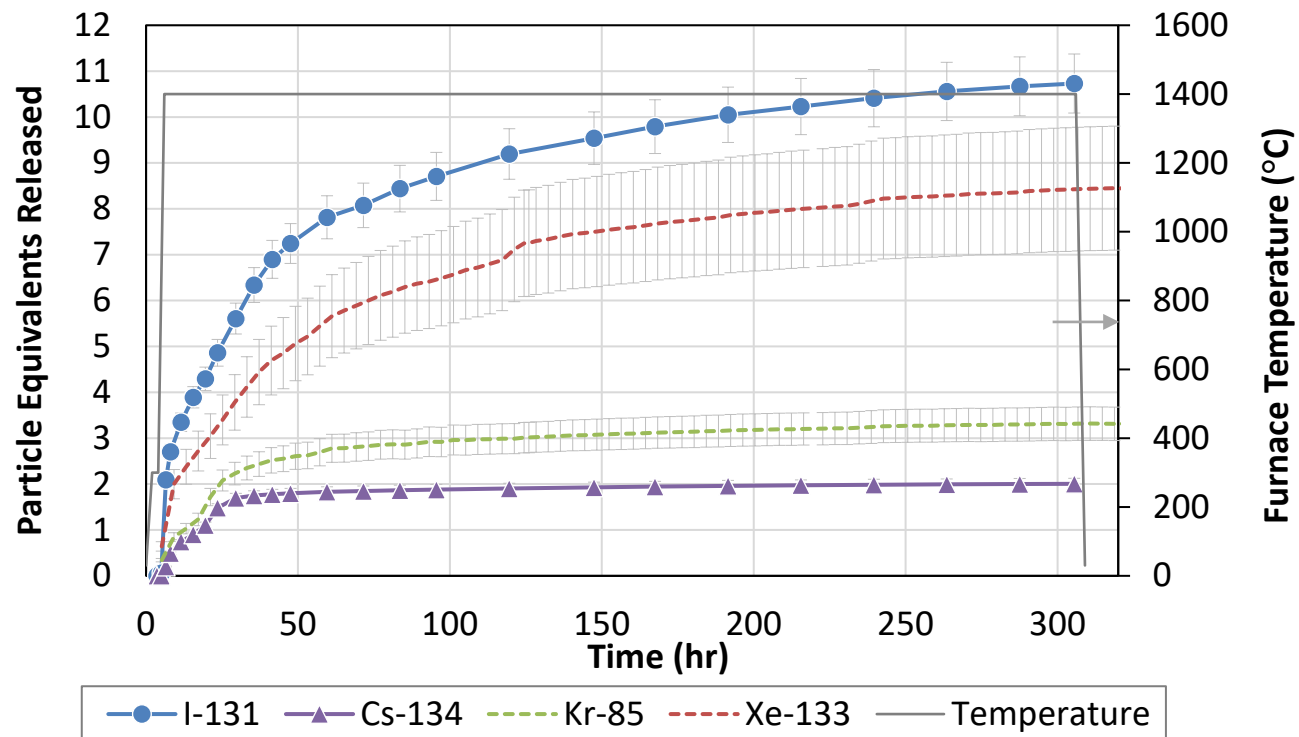


Reirradiation: ~114 hr
Heating test: 1600°C for 202 hr

	DTF Inventory Released (%)
Cs-134	4.4
I-131	32
Kr-85	9.3
Xe-133	41

Compact 10-1 1400°C Test Summary

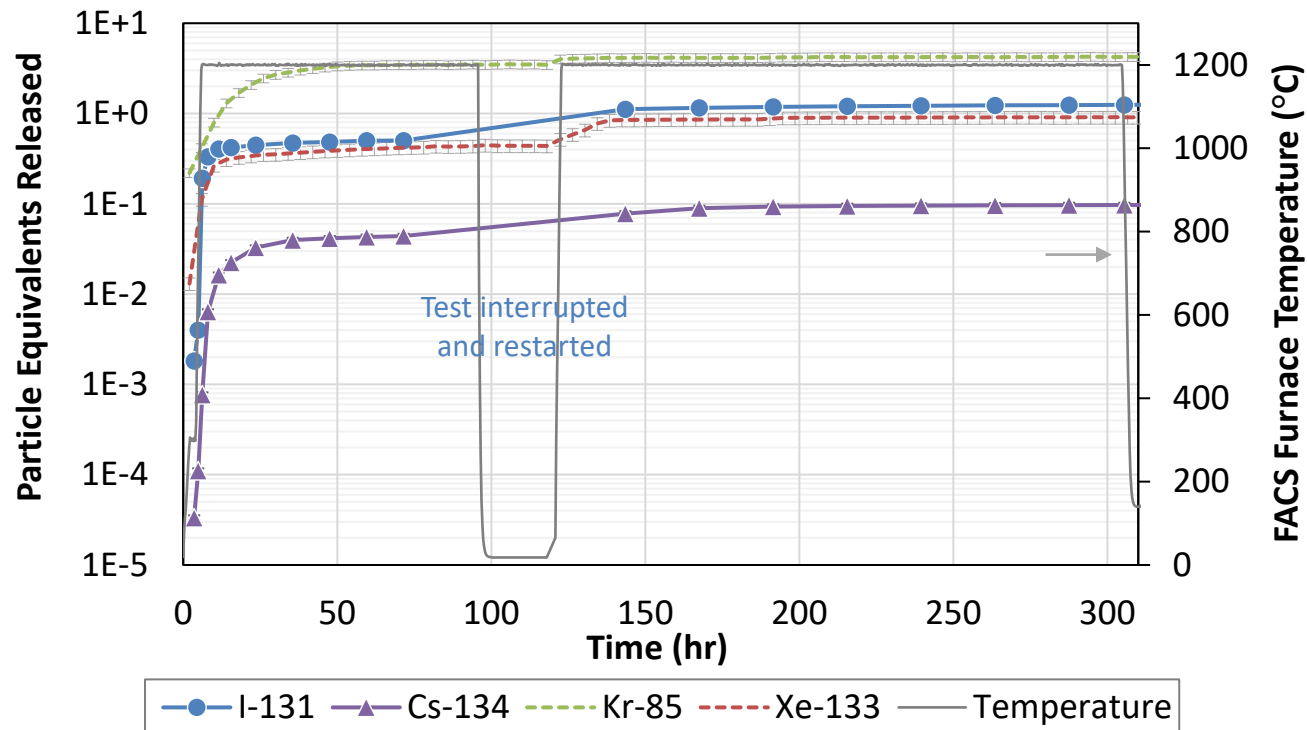
- Compact was reirradiated for ~120 hours, then heated at 1400°C for 300 hours
- Fission gases and iodine released fastest: I-131 and Xe-133 releases are similar
- Kernels seem to retain some iodine and xenon



	DTF Inventory Released (%)
Cs-134	10
I-131	54
Kr-85	17
Xe-133	43

Compact 8-1 1200°C Test Summary

- Compact was reirradiated for ~120 hours, then heated at 1200°C for 272 hours
- Fission gases and iodine released fastest. I-131 and Xe-133 releases are similar
- Temperature cycle from test interruption may have caused a little more release upon reheat phase
- After about 200 hr, 9x less Xe-133 and 5x less I-131 released at 1200°C than at 1600°C



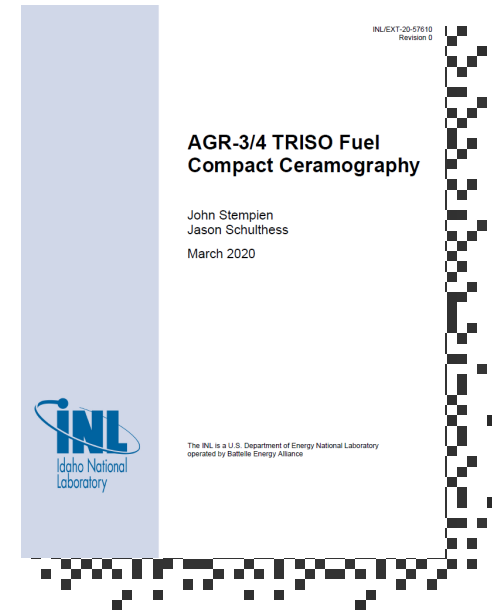
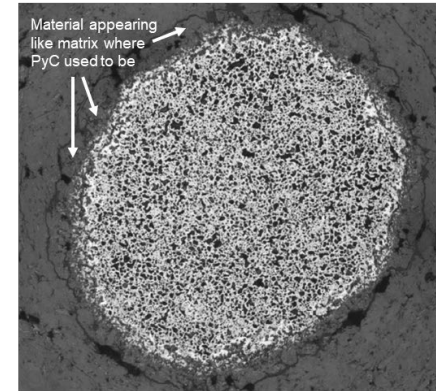
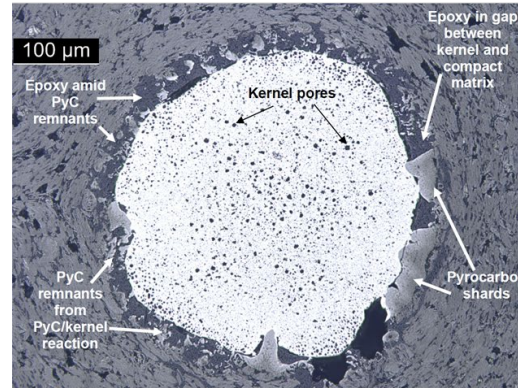
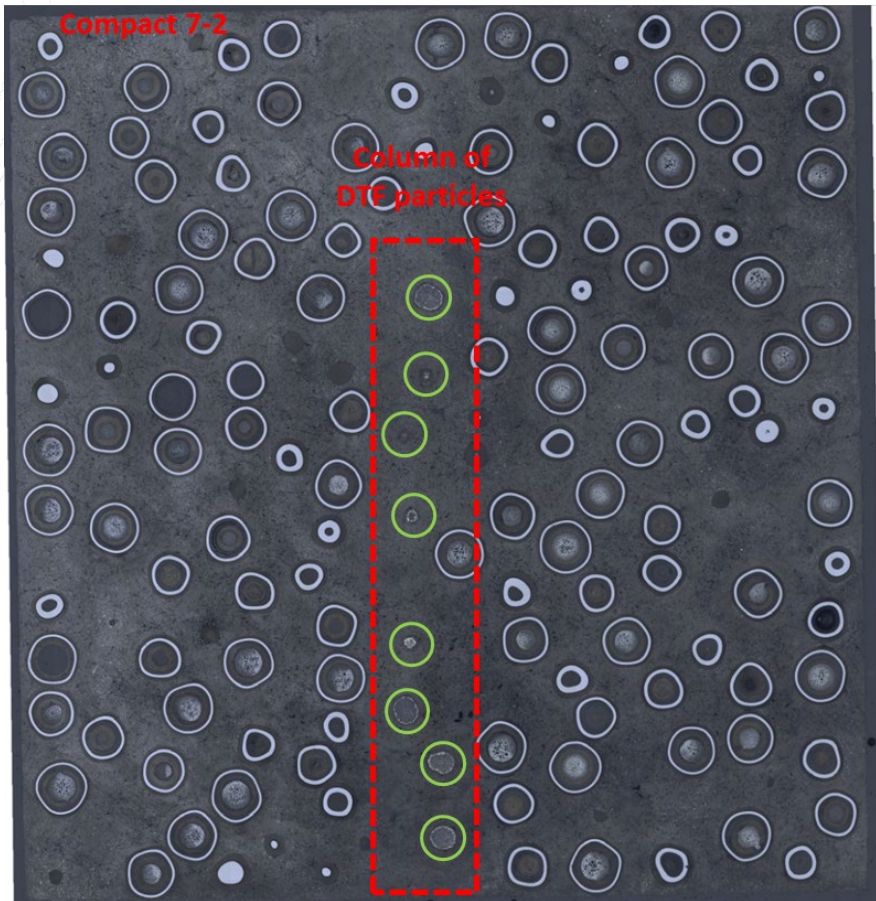
	DTF Inventory Released (%)
Cs-134	0.49
I-131	6.2
Kr-85	21
Xe-133	4.6



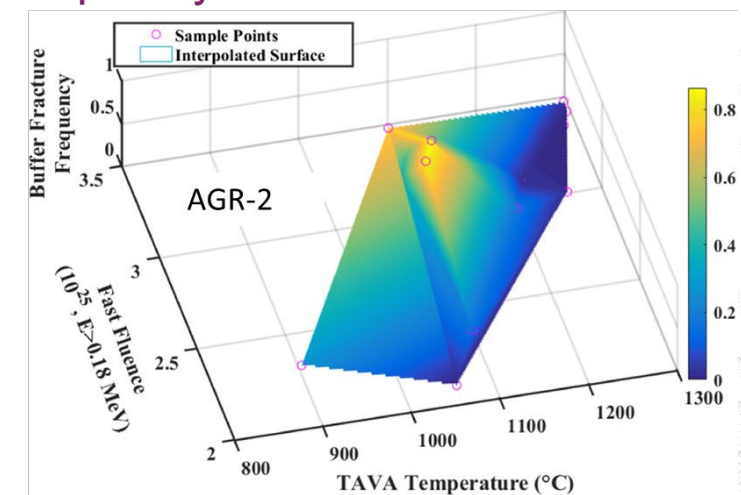
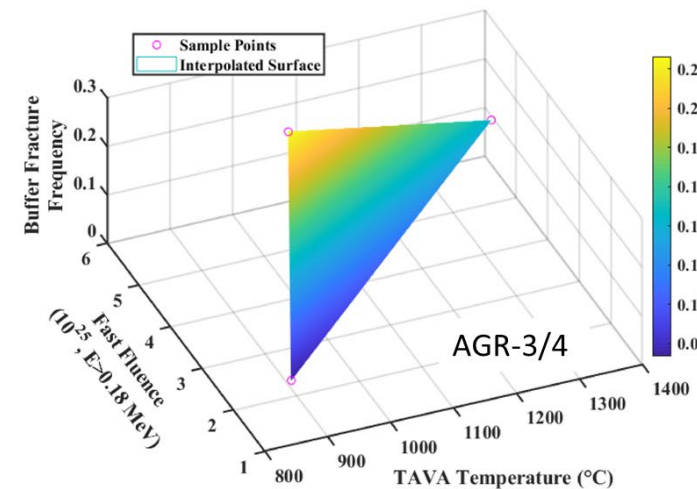
AGR-3/4 Compact Ceramography

Cross-Sectioned Compacts to Observe DTF and TRISO Particle Morphology

All DTF Particles Appeared Failed
Compact 5-2
Compact 7-2



Buffer Fracture Frequency





Conclusions

- Expected remaining experimental work for FY22:
 - R-DLBL of 4 compacts
 - Radiochemical analysis of some samples generated from R-DLBL and NRAD/FACS tests in FY21
- Significant work remains in:
 - Fission product transport model-data comparisons
 - Determinations of transport parameters from AGR-3/4 data

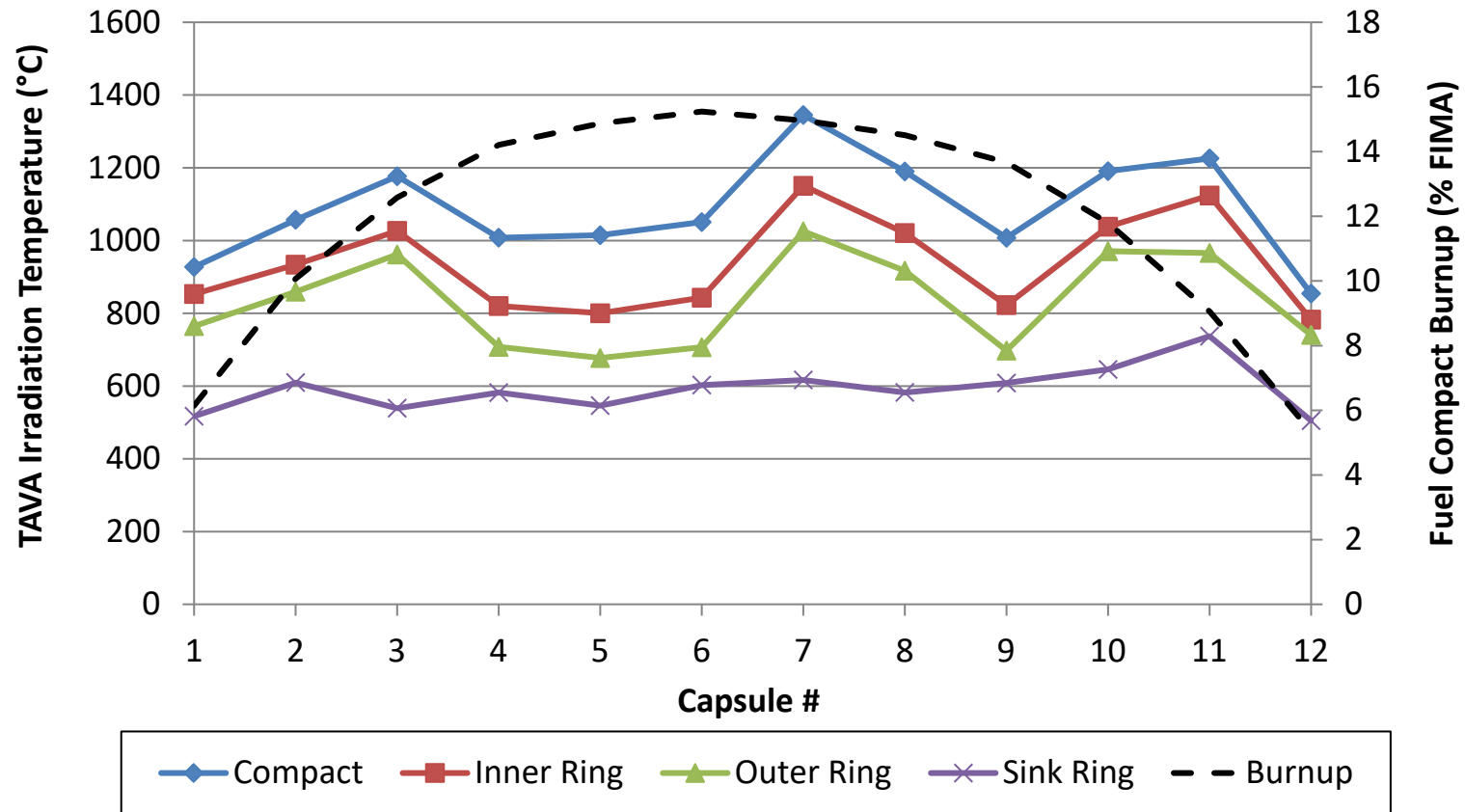


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Irradiation Temperatures and Fuel Burnup



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



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