GAS-COOLED REACTOR
ADVANCED REACTOR TECHNOLOGIES PROGRAM

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Supplemental PIE Activities Supporting Mechanistic Understanding of TRISO Performance

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DOE ART GCR Review Meeting

Hybrid Meeting at INL

July 16–18, 2024

It takes a village…

- **Will Cureton**
- **Grant Helmreich**
- Martino Hooghkirk
- **John Hunn**
- Jay Jellison
- Andy Kercher
- Stephen Trewhitt
- Jesse Werden

- ORNL Irradiated Fuels Examination Laboratory Staff (Ops and RCTs)
- INL AGR Team

- Chuck Baldwin
- Bob Morris
- Darren Skitt (now at WEC)

Supplemental activities establish new tools for that can be used to improve our overall understanding of TRISO fuel behavior and establish basis for engineering scale observations

- Modern microscopy of localized failures and anomalous particles – ”Why”
- Three-dimensional analysis of particles and fuel forms
  - FIB/SEM tomography for porosity and fission product distributions in TRISO layers and interfaces (example – Yongfeng’s talk)
  - X-ray Computed Tomography (XCT)
    - Segmentation development for near term challenges in quantify volume change in fractured buffer particles
- Property changes important to TRISO performance
  - **Anisotropy changes of IPyC/OPyC as a function of irradiation conditions**
- Furnace for Irradiated TRISO Testing (FITT)
  - Time dependent release of fission products (^{110m}Ag and ^{154}Eu)
  - **Oxidation response of particles (SiC recession)**

Effective use of M&S tools requires accurate data (inputs), validated and verified models, and modeling of relevant phenomena

AGR-2 Compact Failure Probability[1]

Method	Compact	IPyC cracking	SiC failure due to cracking
BISON	5-1-3	2.23×10^{-1}	9.75×10^{-5}
	5-3-3	1.25×10^{-1}	4.53×10^{-5}
	6-4-2	3.59×10^{-1}	1.16×10^{-4}
	6-4-3	3.15×10^{-1}	1.12×10^{-4}
PIE	2-1-3, 2-4-3, 3-2-3, 5-1-3	3.5×10^{-2} (includes partial tears)[2]	**Not Observed** (~5.25×10^{-5} from Pd attack)

~1 order of magnitude difference between BISON and PIE data

- We use modeling and simulation (M&S) tools to interpret post-irradiation examination (PIE) observations and predict fuel response
  - As M&S tools make greater advances they **will play a larger role in fuel performance safety analysis, fuel qualification, and eventual reactor licensing**
- Fidelity of simulations is related to the ability to capture appropriate mechanisms and the quality/relevancy of data available – primary failure modes associated with IPyC cracking
  - Current Pd related failure modes assume a diffusion mechanism based on Pd penetration into the SiC layer [3] and not the local Pd attack at exposed SiC due to IPyC fracture
- Opportunities exist to **leverage existing data and materials (AGR) to obtain representative data inputs** and support V&V of M&S tools to better understand internal layer restructuring leading to IPyC cracking and SiC failure

[1] W. Jiang et al., TRISO particle fuel performance and failure analysis with BISON, Journal of Nuclear Materials 548 (2021) 152795

[2] F. Rice et al., Ceramography of irradiated TRISO fuel from the AGR-2 experiment, Nuclear Engineering and Design 329 (2018) 73-81

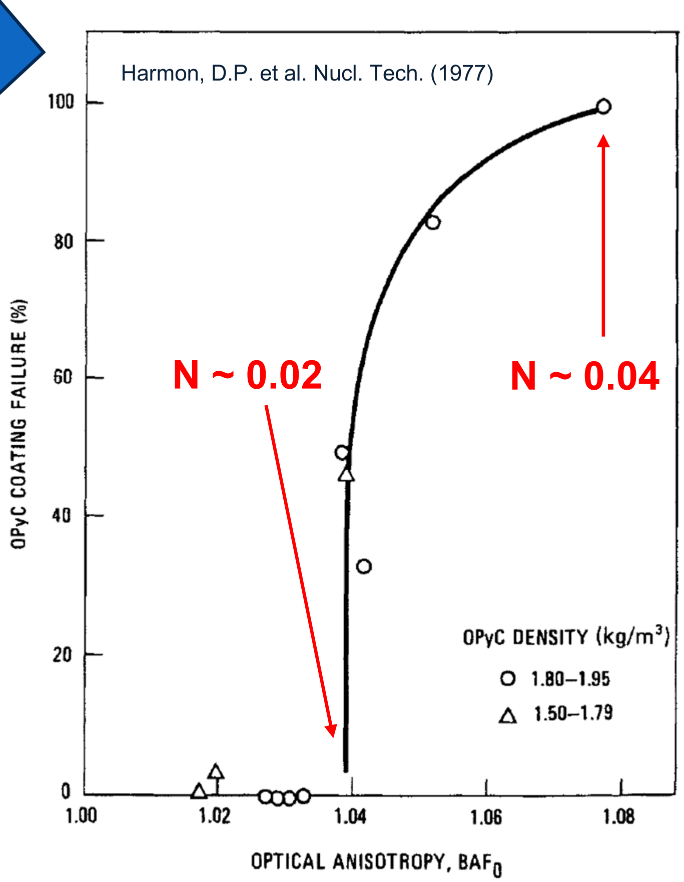
[3] J. Maki et al., et al., The challenges associated with high burnup, high temperature and accelerated irradiation for TRISO-coated particle fuel Journal of Nuclear Materials 371 (2007) 270-280

TRISO fuel specifications require limitations on as-fabricated PyC layer anisotropy

- Anisotropy is a key parameter influencing PyC fracture
- Anisotropy of PyC layers must be controlled and quantified to ensure irradiation performance
- PyC anisotropy can be measured by multiple methods and reported in by different metrics which are related
  - Diattenuation (N), Bacon Anisotropy Factor (BAF), Optical Anisotropy Factor (OPTAF)
- Conservative limits have been determined using modern fabrication/characterization techniques

AGR Specification [1]	Average	Critical Limit
IPyC Diattenuation (N)	≤ 0.0170	≥ 0.0242
OPyC Diattenuation (N)	≤ 0.0122	≥ 0.0242

[1] D. Marshall, AGR-5/6/7 Fuel Specification, INL SPC-1352, Rev. 8


Harmon, D.P. et al. Nucl. Tech. (1977)

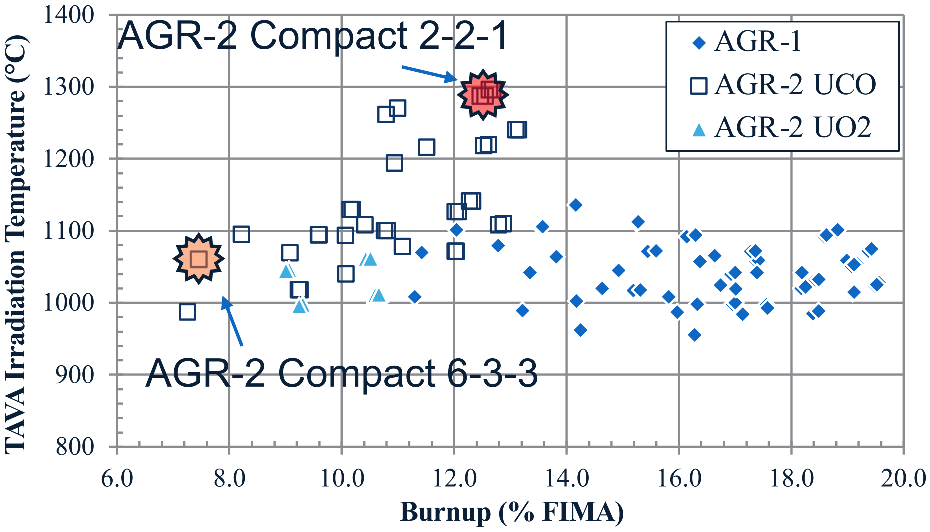
N ~ 0.02

N ~ 0.04

Leveraging Two-Modulated Generalized Ellipsometry Microscope (2-MGEM) for anisotropy analysis of irradiated TRISO

- Same system used for anisotropy measurements of fresh fuel for fabrication quality assurance
- Modified sample preparation to obtain samples with appropriate “flatness” in hot cell
- Conducted comparison on deconsolidated fresh fuel from AGR-2 and bounding irradiation conditions
- Direct measurement of irradiated PyC anisotropy in representative particles

FIMA = Fissions per initial metal atoms
TAVA = Time-averaged volume-averaged



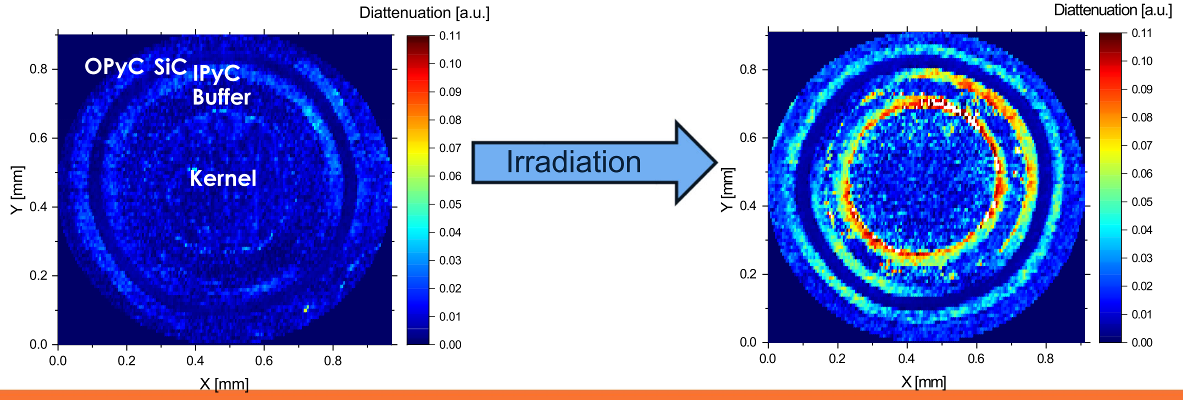
- Measure and report diattenuation (N)
  - $N = \frac{R_{max} - R_{min}}{R_{max} + R_{min}}$
    - N → 0 (isotropic)
    - N → ∞ (anisotropic)
  - Can be used to calculate Bacon anisotropy factor or other metrics

∆ in AGR-2 PyC anisotropy after irradiation

- Initial results show increase in N after irradiation (expected)
  - Particles remain intact despite increase in anisotropy
    - Expected >50% fracture based on measured N from historic data
  - Data used in M&S [1] predicts a smaller change in diattenuation (N) for AGR-2 Compact 221
  - 221 and 633 comparison indicates burnup/fluence alone does not dictate the overall response
- Anisotropy of PyC layers influences the thermomechanical response of the particles (e.g., probability of cracking the layers!)
  - How does this **60–85% measured difference from predicted** impact fuel performance models?

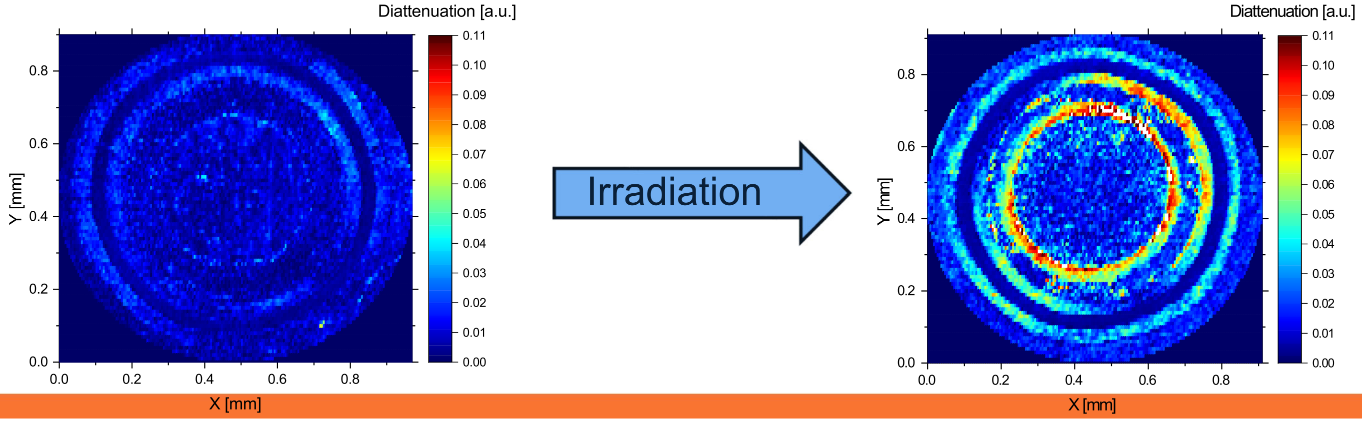
*Preliminary data	**Diattenuation (N) (St. Dev.)**	
Sample	IPyC	OPyC
AGR-2-LEU09 Unirradiated/compacted	**0.0166** (0.009)	**0.0136** (0.007)
AGR-2-221 12.5% FIMA/1287°C TAVA	**0.0378** (0.019)	**0.0382** (0.011)
AGR-2-633 7.5% FIMA/1095°C TAVA	**0.0456** (0.015)	**0.0243** (0.010)
Relative Change in N	**0.0212**	**0.0246**
Predicted Change for AGR-2-221 in N [1]	**0.0133 (~60–85% smaller than measured)**	

Diattenuation values representative of average from measurement of 5 particles



Summary of Anisotropy Effort

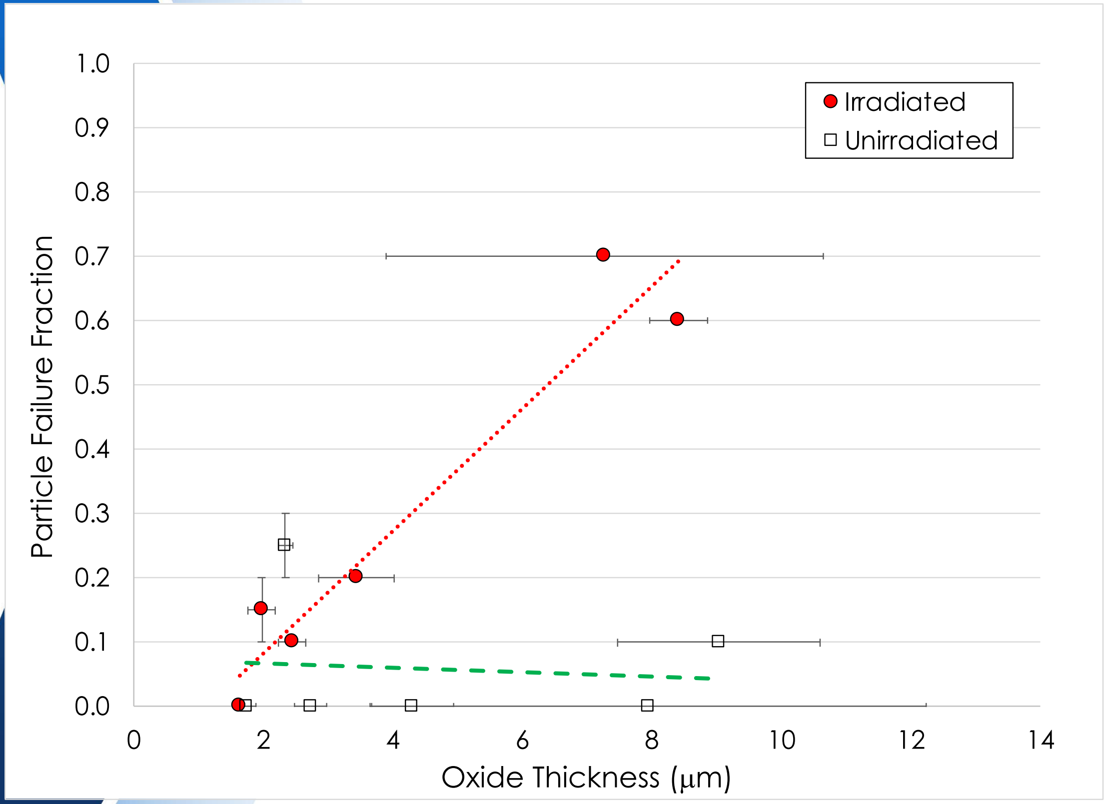
- Irradiation impacts the anisotropy in both IPyC and OPyC layers
- Particles remain intact despite significant increase in anisotropy due to irradiation
- Future work aims to map the anisotropy as a function of irradiation conditions (e.g., temperature, burnup, fluence moving to AGR-5/6/7 next)
- Connect with modelling and simulation activities



Understanding the impact of irradiation on TRISO particle oxidation and failure in AGR-5/6/7

- Approach
  - Simultaneously expose irradiated and unirradiated particles to oxidizing environments
  - Measure particle failure fractions
  - Measure resultant ~~oxide layer thickness and structure~~ SiC layer recession
- Outcome
  - Gain understanding of differences in failure response for different test environments and irradiation conditions
  - Criteria for failure based on SiC thickness
  - Direct insights on differences in oxidation response between fresh and irradiated fuel
  - *Bigger picture* → results will support text matrix development for compact oxidation studies in the Air/Moisture-Ingress Experiment (AMIX) system at INL and potentially feed into modeling efforts to predict fuel performance during off-normal scenarios

AGR-2 showed correlation of oxide thickness with failure fraction



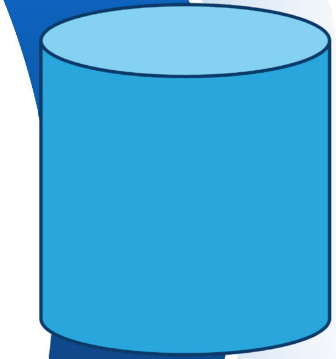
- Why no failure in unirradiated?
  - Change in mechanical properties?
  - Internal layer restructuring and gas pressure?
  - Need M&S to support observations
- Assumption is that thickness is proportional to SiC recession (in passive regime) but not equivalent
- Models predict thin SiC is more susceptible to failure [1] so we need a direct measure of recession

[1] W.F. Skerjanc, J.T. Maki, B.P. Collin, D.A. Petti, Evaluation of design parameters for TRISO-coated fuel particles to establish manufacturing critical limits using PARFUME, Journal of Nuclear Materials 469 (2016) 99–105.

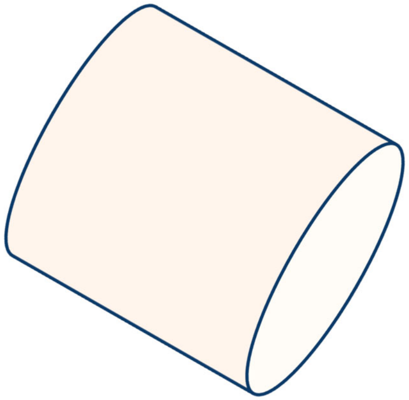
Revisiting FITT Oxidation effort to improve understanding of variables impacting particle failure

1. **Establish methodology to measure SiC recession after oxidation**
   - **Demonstration of use of XCT to track particles before and after testing (complete)**
     - **Blind test with set of 8 particles**
   - **Determine degree of SiC recession post-test (complete)**
2. Execute oxidation testing focusing on failure fraction as a function of exposure time and SiC recession
   - **Measure pre-test particles (in progress)**
   - Complete FITT testing

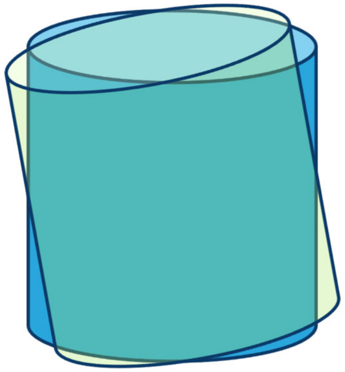
3D analysis provides lots of data, we can use it to increase confidence in particle matching



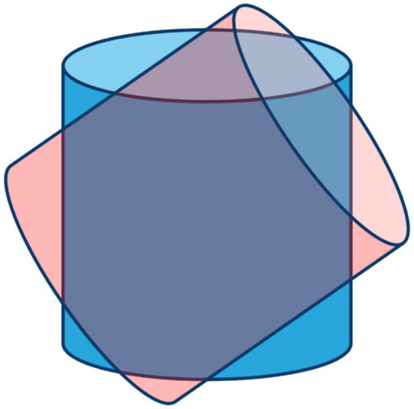
Particle orientation measured pre-test



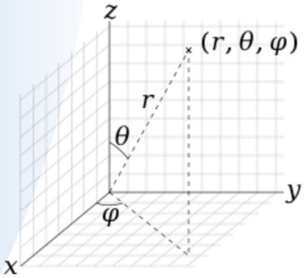
Same particle orientation measured post-test



“Low” error



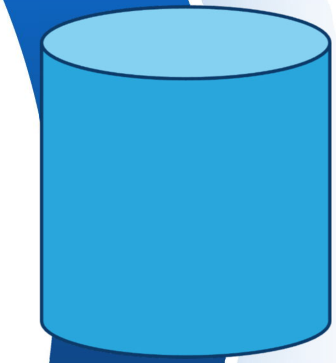
“High” error



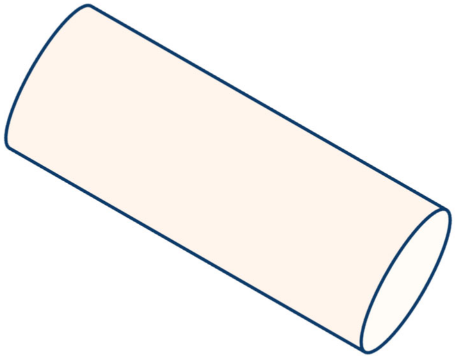
Rotate around 5000 unique θ,ϕ pairs in spherical coordinates, determine difference in radius at each rotation for 5000 radii, and calculate error in radius for each layer

3D analysis provides lots of data, we can use it to increase confidence in particle matching

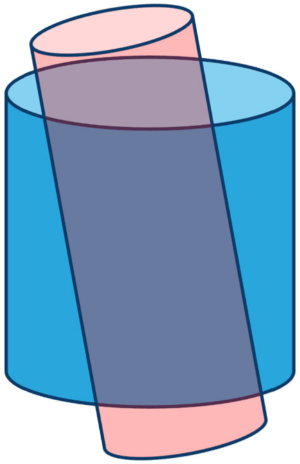
Particle orientation
measured pre-test



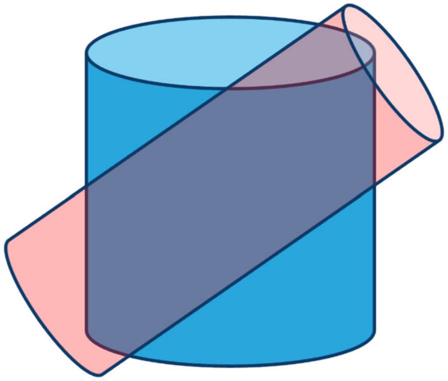
“Different” particle orientation
measured post-test

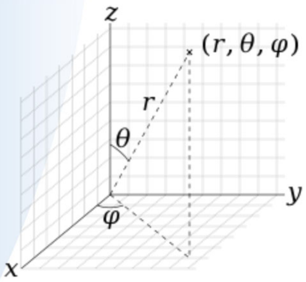


“High” error



“High” error

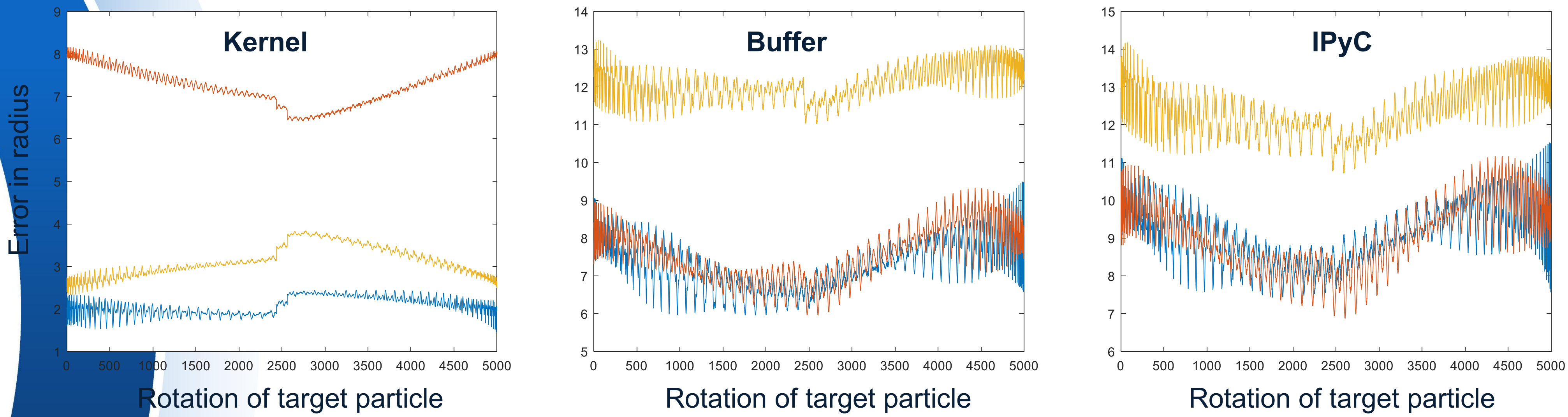




Rotate around 5000 unique θ,ϕ pairs in spherical coordinates, determine difference in radius at each rotation for 5000 radii, and calculate error in radius for each layer

- Minimum in error should reflect the best fit for each particle
- Local minima should reflect best θ,ϕ pair to match orientation

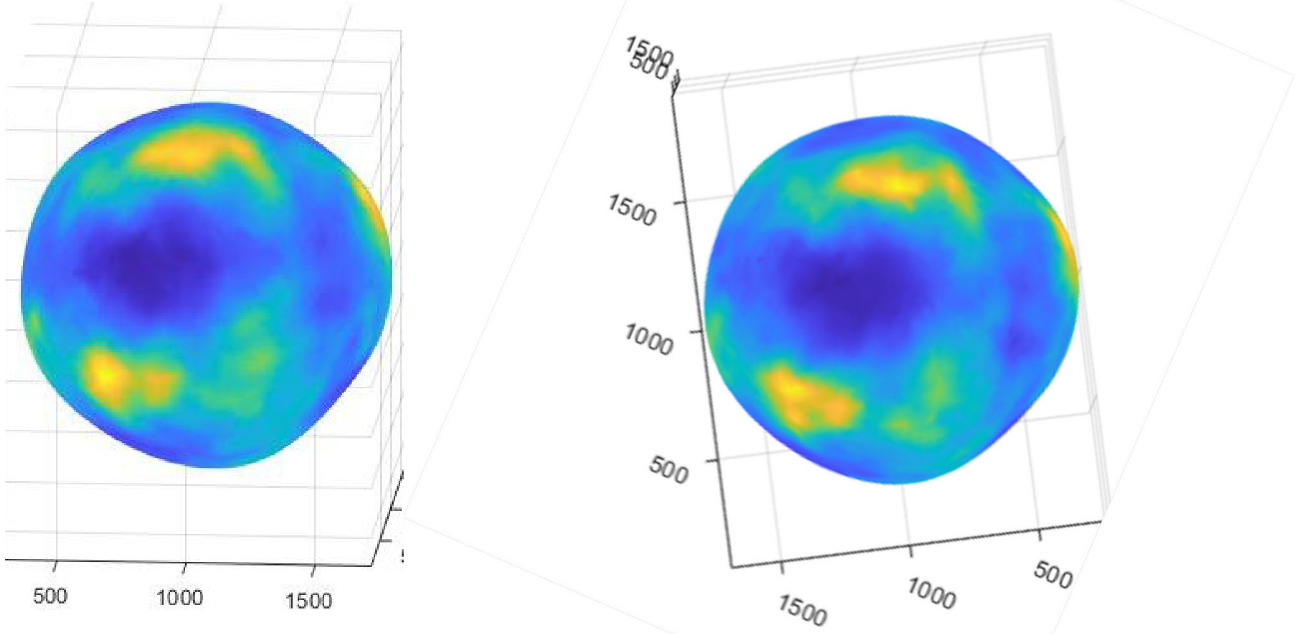
3D segmentation provides lots of data, we can use it to increase confidence in particle matching



Error in outer radius of kernel, buffer, and IPyC as a function of rotation of target particle for three particles (1 – blue; 2 – red; 3 – yellow)

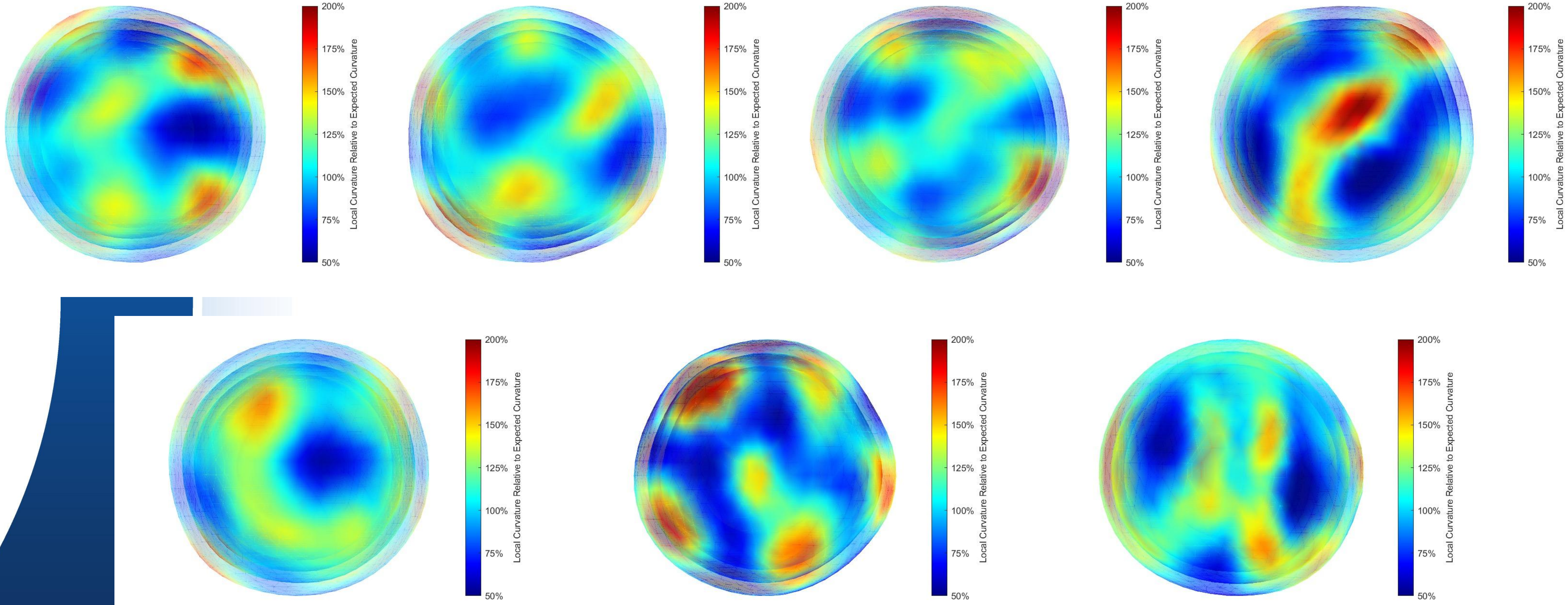
- Particle 1 shows good agreement for all radii explored

Local surface curvature plots provides a sanity check on particle identification and orientation



- Features line up; Particle 1 is confirmed
- The identified orientation can be used to determine new SiC thickness and degree of recession

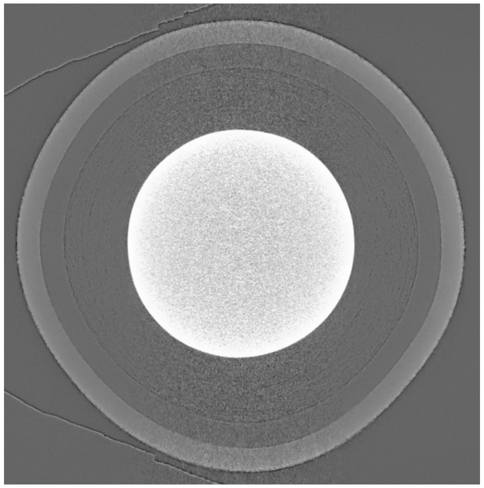
It works because like snowflakes, every particle is unique…

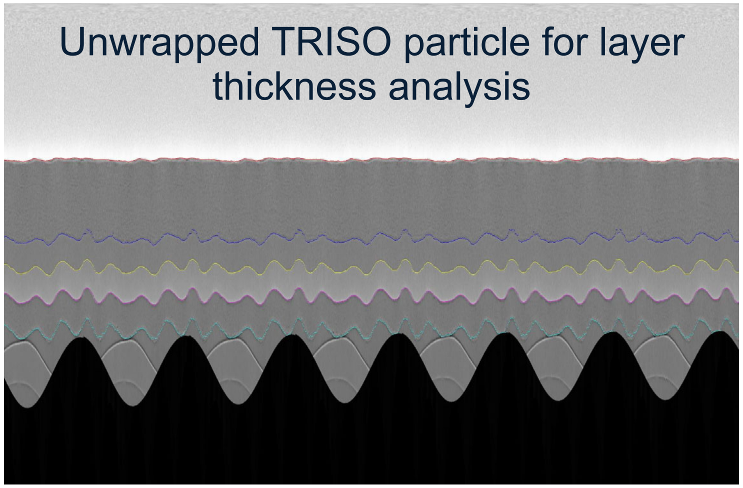


Basis for the surface curvature check

Determination of recession after exposure

XCT of oxidized TRISO particle





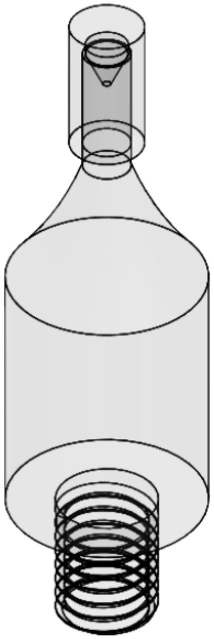
Unwrapped TRISO particle for layer thickness analysis

- Particles were exposed to 1200°C for 200 h, then 1400°C, 200 h in flowing air to ensure an oxide layer was present
  - Three particles imaged after testing while optimizing imaging conditions to best support SiC and SiO_2 segmentation
- Initial efforts used the established layer thickness analysis from Helmreich et al. [1], however, the need existed **to add a new subroutine** for layer thickness analysis to allow segmentation of SiO_2 from SiC layer
- Recession analysis targeting 25,000 angular points (θ,ϕ pairs) compared to 5,000 angular points to improve fidelity of rotational matching and accuracy in determination of local SiC thinning
- 3D analysis of individual exposed particle indicates 1.44 μm of layer growth from unexposed SiC to SiC + Oxide
  - Simplified 2D analysis indicates an average oxide thickness of ~7.5 μm and SiC recession of ~6.0 μm
  - Proof of concept demonstrated, but more importantly that oxide layer thickness is not equivalent to SiC recession

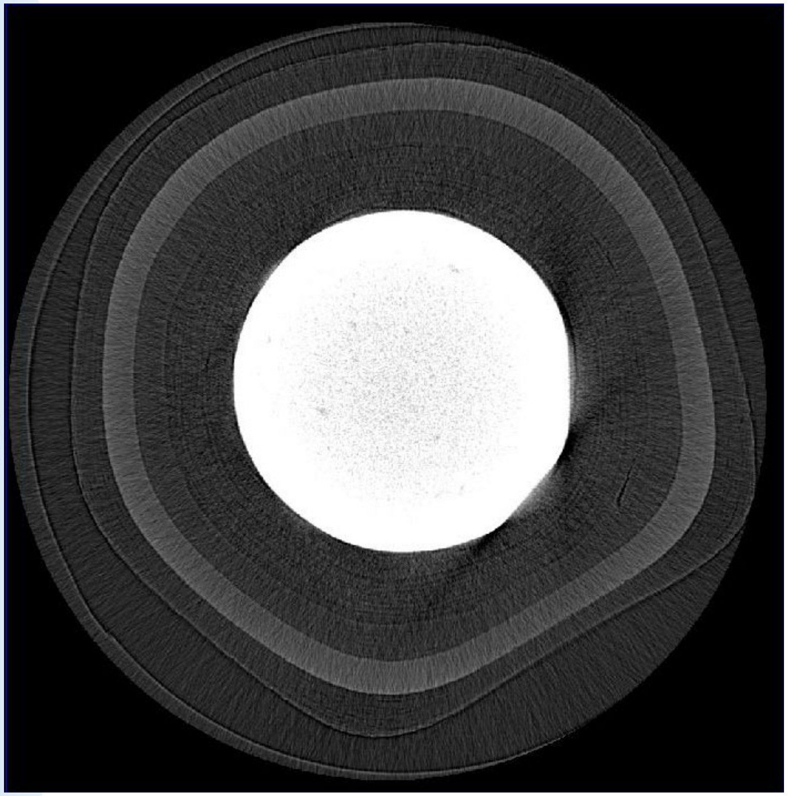
[1] Helmreich et al., JNM, V 539, October 2020, 152255

Particle holder modification for FITT testing

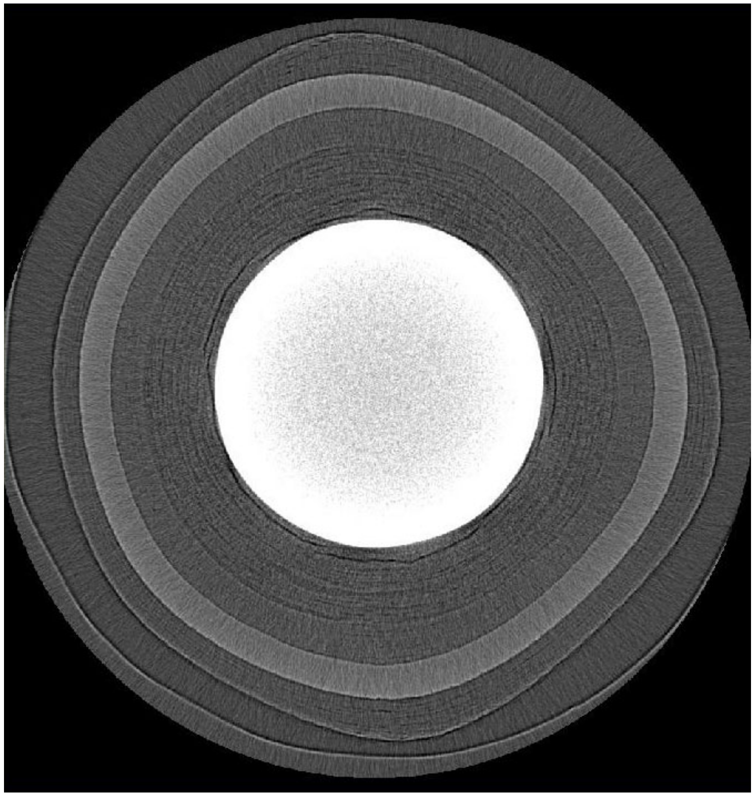
- Initial XCT demonstration was performed on surrogate particles, but **uranium-bearing and irradiated particles require special sample holders**
  - Must securely seal particle for transit to and from hot cells
  - Particle must be removable for heat treatment (no epoxy)
  - Attenuation of low energy x-rays must be minimized for image quality
- Currently used sample holders for irradiated particle XCT use epoxy to permanently secure particle in place
- Initial imaging with screw-top particle holders was found to excessively degrade image quality for low-Z layers
- A new slip-fit sample holder has been developed and tested and found to be secure and acceptably attenuating



Holder modification for FITT testing has led to better signal to noise to improve segmentation



Tomogram acquired using old holder. Low signal to noise ratio in low Z layers.



Tomogram acquired using new holder. Good signal to noise ratio in low Z layers.

In progress for FITT oxidation testing

- AGR-5/6/7 Compact 363 and 221
  - Different conditions:
    - 363 → 1363^{o}C TAVA Temp., 5.47x10^{25} n/m^{2}, 14.77% FIMA
    - 221 → 845^{o}C TAVA Temp., 4.72x10^{25} n/m^{2}, 14.03% FIMA
- Added benefit of 363
  - Ability to determine swelling of kernels via statistical population analysis (30× as-fab)
  - Statistical assessment of kernel mobility/migration (re: Cureton's earlier talk) 30×irradiated

No recession analysis, ***completed***

Temp. (°C)	Compact	Atmosphere	Exposure Times (h)
1200	221	21% O_2 (balance N_2)	400
1400	221	21% O_2 (balance N_2)	10, 100, 400
1400	221	2% O_2 (balance He)	10

Planned with recession analysis

Temp. (°C)	Compact	Atmosphere	Exposure Times (h)
1400	221	21% O_2 (balance N_2)	*400*
	363	21% O_2 (balance N_2)	**100, 200, 400**
1400	221	2% O_2 (balance He)	*400*
	363	2% O_2 (balance He)	*400*

Bold = Priority 1, *italic* = Priority 2

FITT oxidation testing summary

- Recession analysis provide a direct measurement of the key parameter expected to influence particle failure
- New method established and verified to conduct experiment
  - Redesigned holder with limited artifacts
- Extent of experiment provides additional potential benefits regarding statistical XCT analysis

Summary of supporting PIE

Opportunity to answer questions and more

- Supplemental PIE allows us to determine the bases for many critical phenomena and defend the engineering scale observations
  - Communicate why failures or other deleterious behaviors occur and that we have confidence that operation within the prescribed envelope will not lead to unaccounted for behavior
- Intentional approaches may positively impact M&S efforts by providing relevant data on representative systems to support model development and inputs – even more impact when we can coordinate efforts with M&S directly

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Thank you all for your attention!

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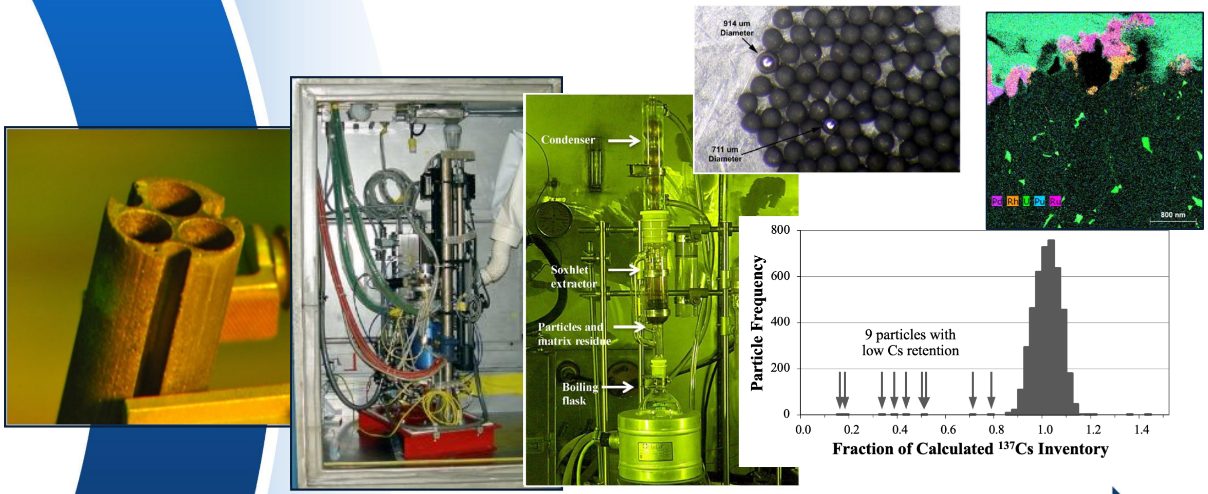
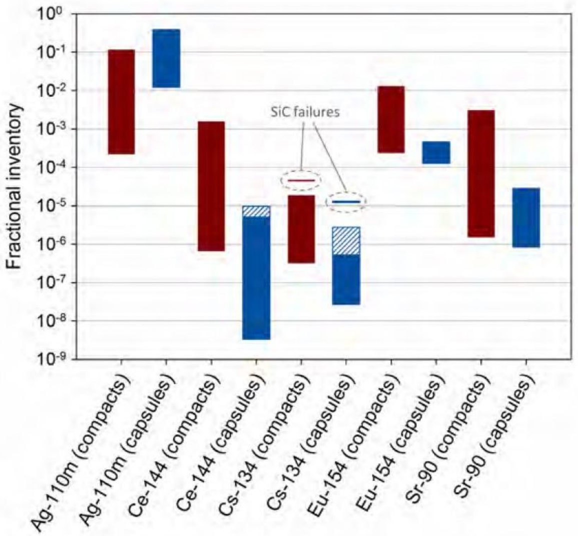
Supplemental PIE across multiple length scales provides opportunity to establish basis for engineering scale observations



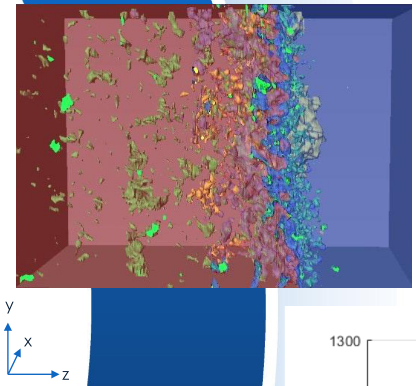


Range of inventory fractions found retained in irradiated compacts outside of the SiC layer (red columns) and on the capsule components (blue columns). (Demkowicz et al. 2015)[1]

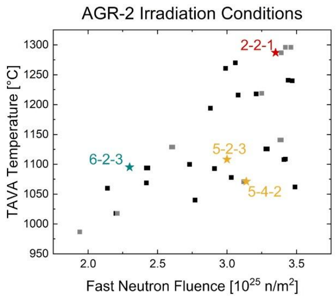
- Multiscale provides both engineering scale data (overall release behavior & failure rate) and a mechanistic understanding of release & failure from lower length scale analysis (the “why” or basis for observation)
- Particle failure deemed to be primary source of radionuclide release in AGR-1 and AGR-2 [1,2]

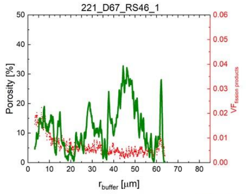
[1] P.A. Demkowicz, J.D. Hunn, R.N. Morris, I. van Rooyen, T. Gerczak, J.M. Harp, S.A. Ploger, *AGR-1 post irradiation examination final report*, INL/EXT-15-36407 Rev. 0, Idaho Falls, ID, Idaho National Laboratory (2015).
[2] J.D. Stempien, J.D. Hunn, R.N. Morris, T.J. Gerczak, P.A. Demkowicz, *AGR-2 TRISO fuel post-irradiation examination final report*, INL/EXT-21-64279 Rev. 0, Idaho Falls, ID, Idaho National Laboratory (2021).

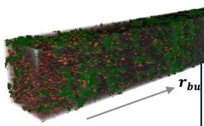
Example: Coupling multiple techniques to expand our understanding of TRISO fuel behavior

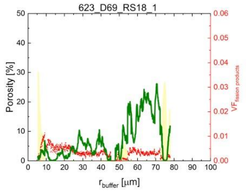


3D FIB/SEM tomography to established ability to conduct analysis of irradiated particles and insights on IPyC/SiC interface structure on Pd accommodation (Seibert et al., 2023)

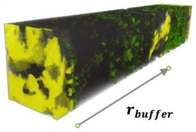




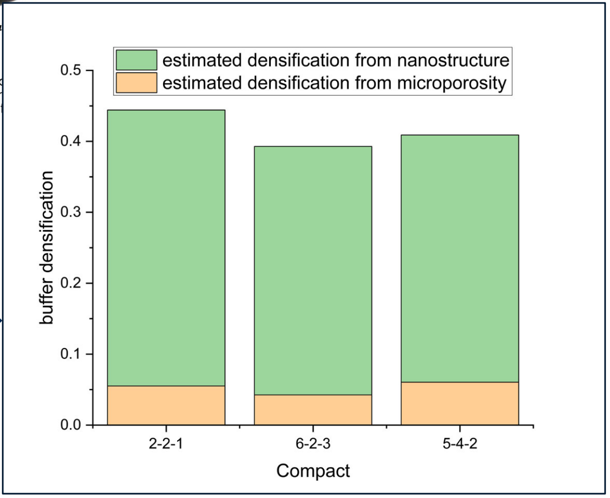


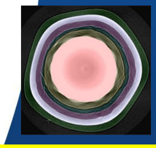


Red = fission products/
Green
Yellow =

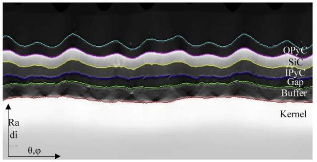


Coupling XCT data and FIB/SEM tomography allows for a determination of the role of microstructure on densification (Yongfeng's talk: Joint work with UW-Madison, Griesbach et al., 2023)

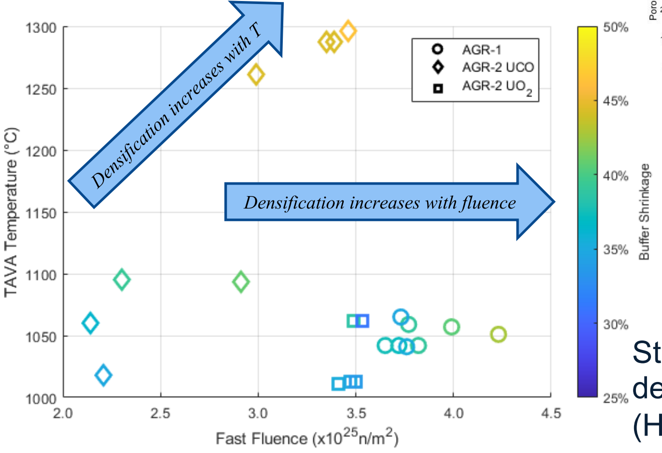




Layer thickness analysis of irradiated TRISO particle



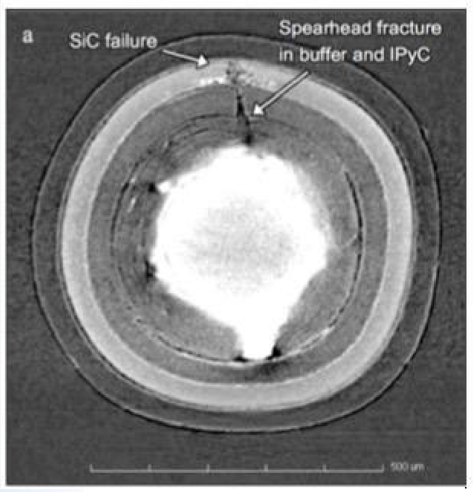
Helmreich et al., JNM, V 539, October 2020, 152255

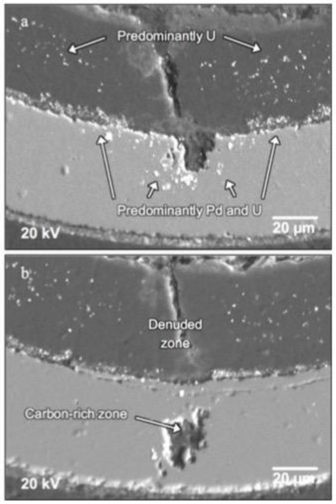


Standard XCT can be used to determine bulk buffer densification (Helmreich et al., 2022)

[1] G.W. Helmreich, A.K. Kercher, T.J. Gerczak, D. Richardson, F.C. Montgomery, D.J. Skitt, J.D. Hunn, Microstructure of irradiated AGR TRISO particle buffer layers as measured by X-ray computed tomography, Journal of Nuclear Materials, Volume 572 (2022) 154061.
[2] C. Griesbach, T. Gerczak, Y. Zhang, R. Thevamaran, Microstructural heterogeneity of the buffer layer of TRISO nuclear fuel particles, Journal of Nuclear Materials, Volume 574 (2023) 154219.
[3] R. Seibert, T. Gerczak, G. Helmreich, J. Hunn, AGR-2 irradiated TRISO particle IPyC/SiC interface analysis using FIB-SEM tomography, Journal of Nuclear Materials, Volume 573 (2023) 154104.

Multiscale analysis ensures arguments for understanding "why" failures occur

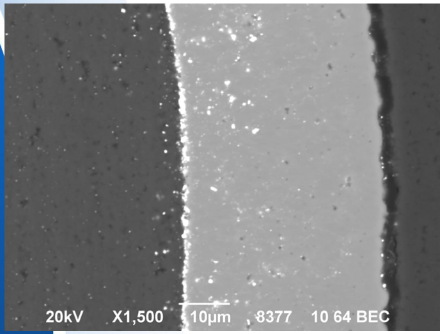




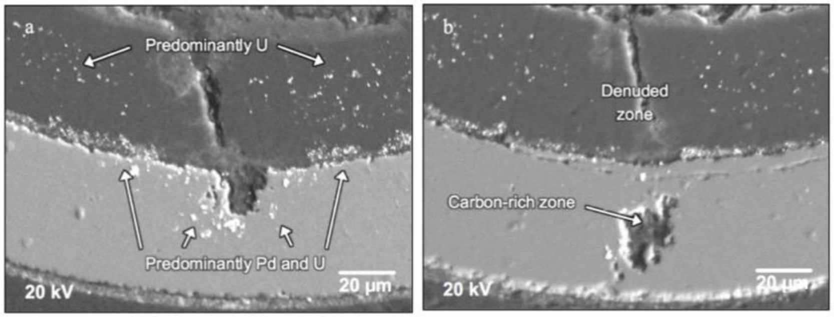
- Buffer densification under irradiation is a primary part of the particle failure sequence observed in rare in pile failures [1]
- Fracture or incomplete buffer tearing transmits a crack through IPyC to the SiC which exposes the layer and becomes susceptible to local Pd corrosion
  - Highlights a need to holistically understand the buffer densification mechanism to predict tearing response

[1] Hunn, J.D., Baldwin, C.A., Gerczak, T.J., Montgomery, F.C., Morris, R.N., Silva, C.M., Demkowicz, P.A., Harp, J.M., Ploger, S.A., 2014. Detection and analysis of particles with failed SiC in AGR-1 fuel compacts. Nucl. Eng. Des.

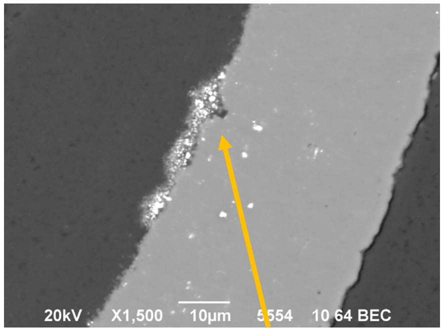
Supplemental PIE can answer questions such as how to facilitate longer operation at temperature?



AGR-1 Intact SiC layer after 1800°C, 300 h with **significant FP and actinide interaction and pileup**



Localized Pd attack at exposed SiC caused a through layer defect with carbon-rich phases remaining from an AGR-1 1700°C safety-tested particle [1].



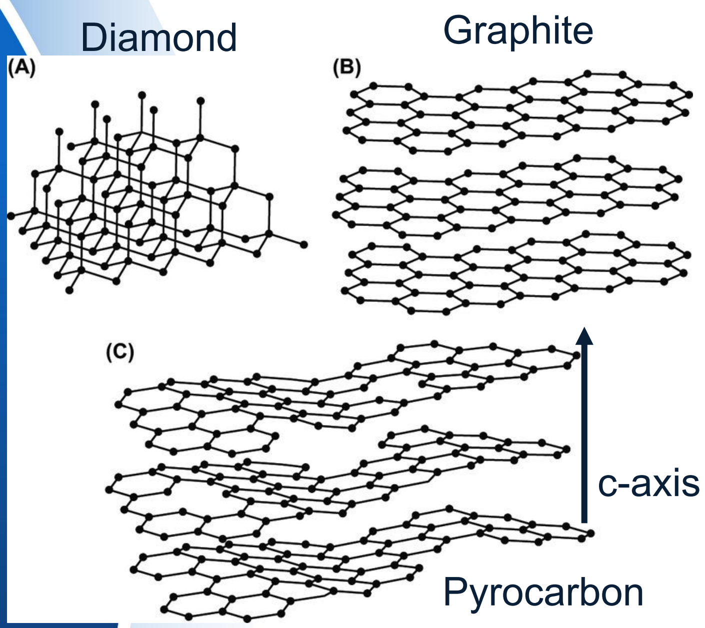
As irradiated AGR-2 localized SiC corrosion by Pd absent exposed SiC

- Multiple microreactor concepts expect to keep fuel in operation >8× longer than AGR irradiation experiments, Pd attack is expected to be temperature dependent and coupled to exposed SiC layer arising from buffer/IPyC interactions
- **Previously failure was assumed when Pd penetration has reached 50% of SiC thickness** based on empirical Pd attack rates (Maki et al. [2])
  - PIE data shows this is not the relevant failure mechanism
- To have confidence at more aggressive operation conditions we need to **understand the mechanism leading to exposed SiC,** Pd transport and interaction with SiC, and nature of observed local corrosion absent exposed SiC (e.g., how IPyC protects SiC)

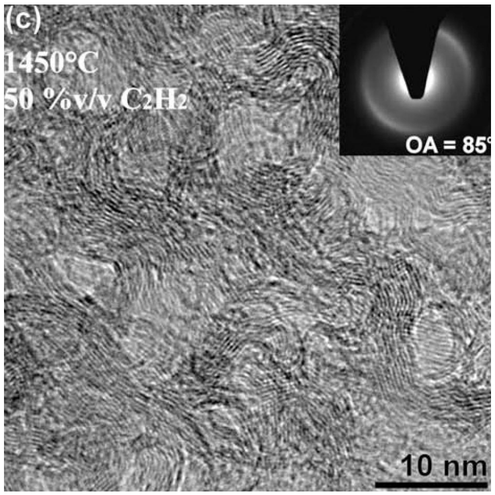
[1] Hunn, J.D., Baldwin, C.A., Gerczak, T.J., Montgomery, F.C., Morris, R.N., Silva, C.M., Demkowicz, P.A., Harp, J.M., Ploger, S.A., 2014. Detection and analysis of particles with failed SiC in AGR-1 fuel compacts. Nucl. Eng. Des.

[2] Maki et al. The challenges associated with high burnup, high temperature and accelerated irradiation for TRISO-coated particle fuel, Journal of Nuclear Materials 371 (2007) 270–280.

Pyrocarbon is a Complex Material

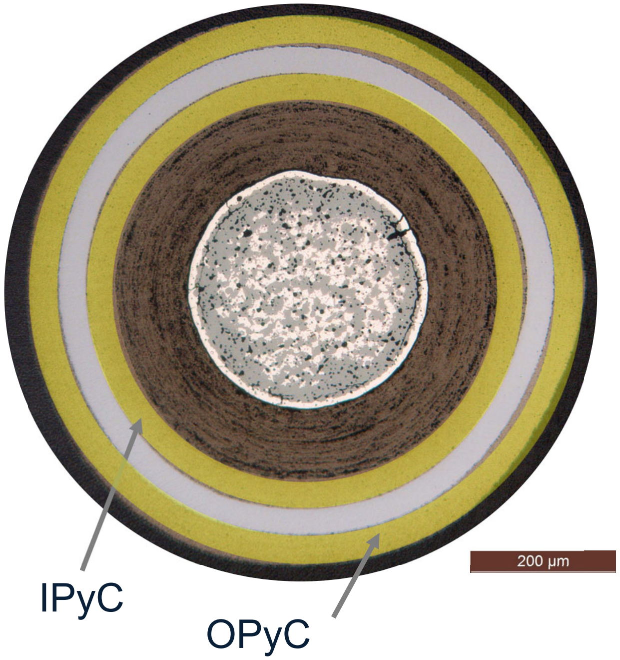


R. More et al. Biomaterials Science (2013)



E. Lopez-Honorato et al. Carbon (2009)

Typical TRISO Particle



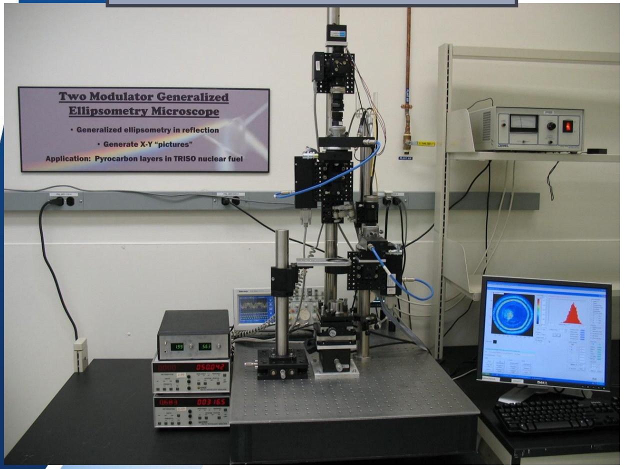
- Pyrocarbon: Amorphous carbon + pores + crystallites
  - Crystallites similar but distinct from graphite
  - Kinked and defective
- Since crystallographic anisotropy is intrinsic, bulk anisotropy must be intentionally avoided.
  - Under irradiation anisotropic dimensional changes, stress could cause the integrity of PyC layers to become compromised

- Diattenuation (N)
- $N = \frac{R_{max} - R_{min}}{R_{max} + R_{min}}$
  - N → 0 (isotropic)
  - N → ∞ (anisotropic)

Two-Modulated Generalized Ellipsometry Microscope (2-MGEM) for anisotropy analysis of irradiated TRISO

2-MGEM developed in early 2000s at ORNL to quantify anisotropy in as-fabricated pyrocarbon layers with higher fidelity than previously used techniques

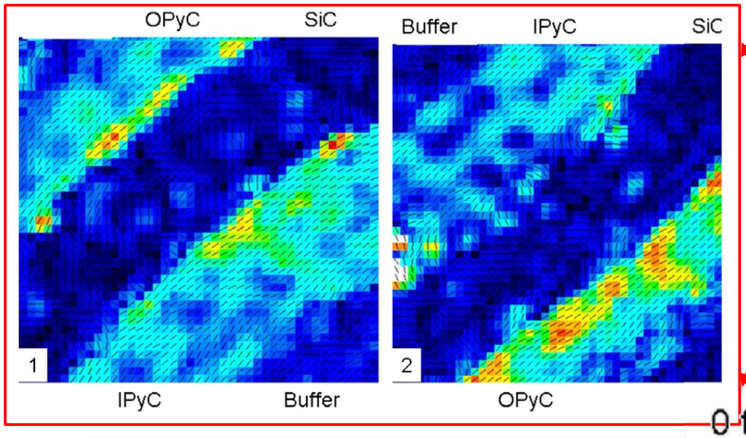
2-MGEM Instrument

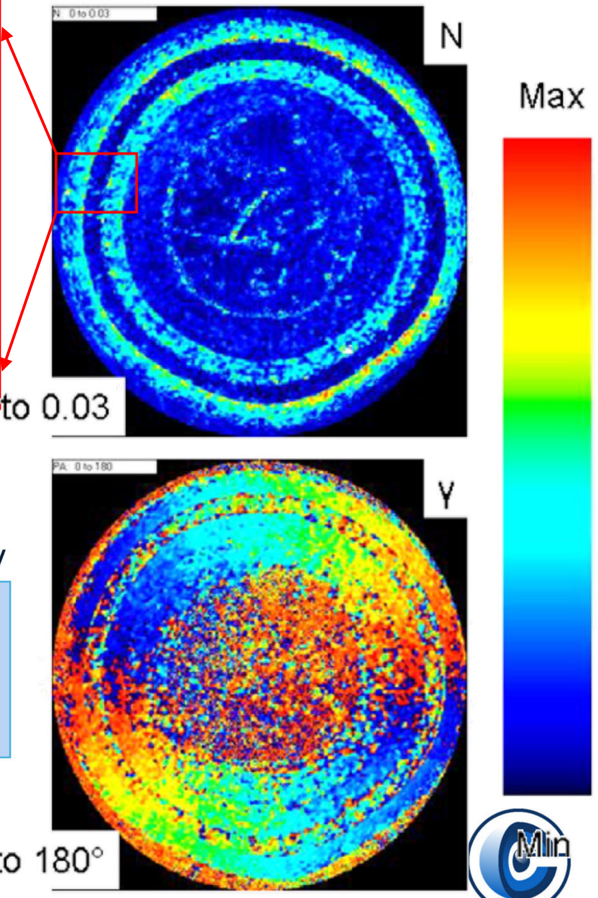


Two Modulator Generalized Ellipsometry Microscope

- Generalized ellipsometry in reflection
- Generate X-Y "pictures"

Application: Pyrocarbon layers in TRISO nuclear fuel





Up to 2 µm spatial resolution imaging of pyrocarbon anisotropy

Ellipsometry compared to traditional Polarimetry

More accurate: measures the entire Mueller matrix

More sensitive: 10 times higher resolution of diattenuation

More robust: Non-specular reflection doesn't affect accuracy

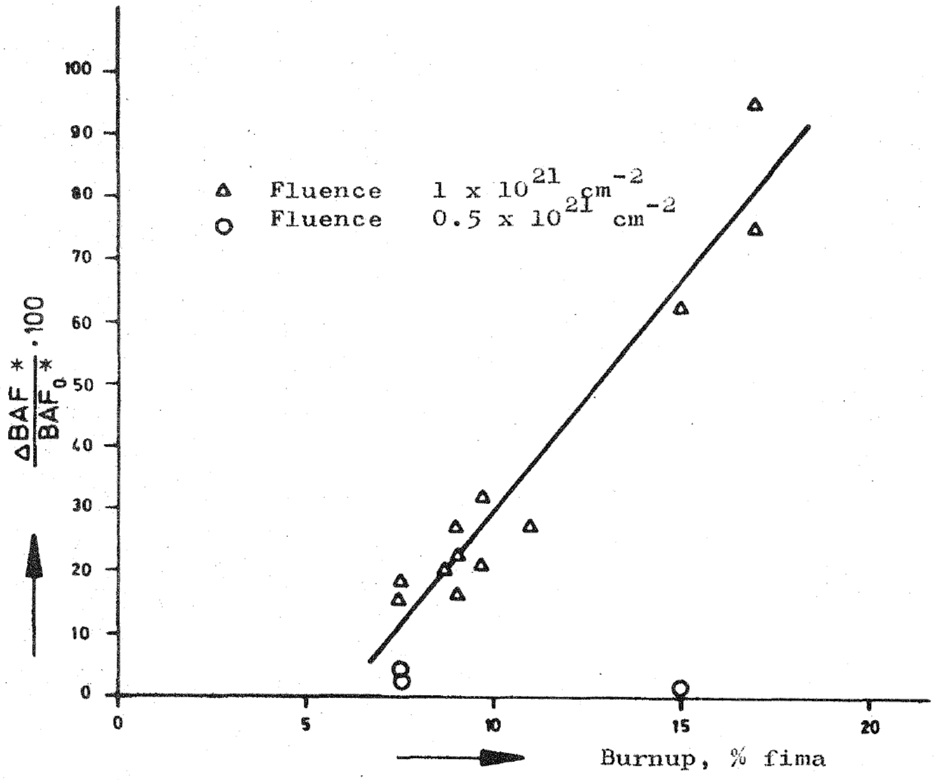
More complete: measures 10,000+ points across the surface

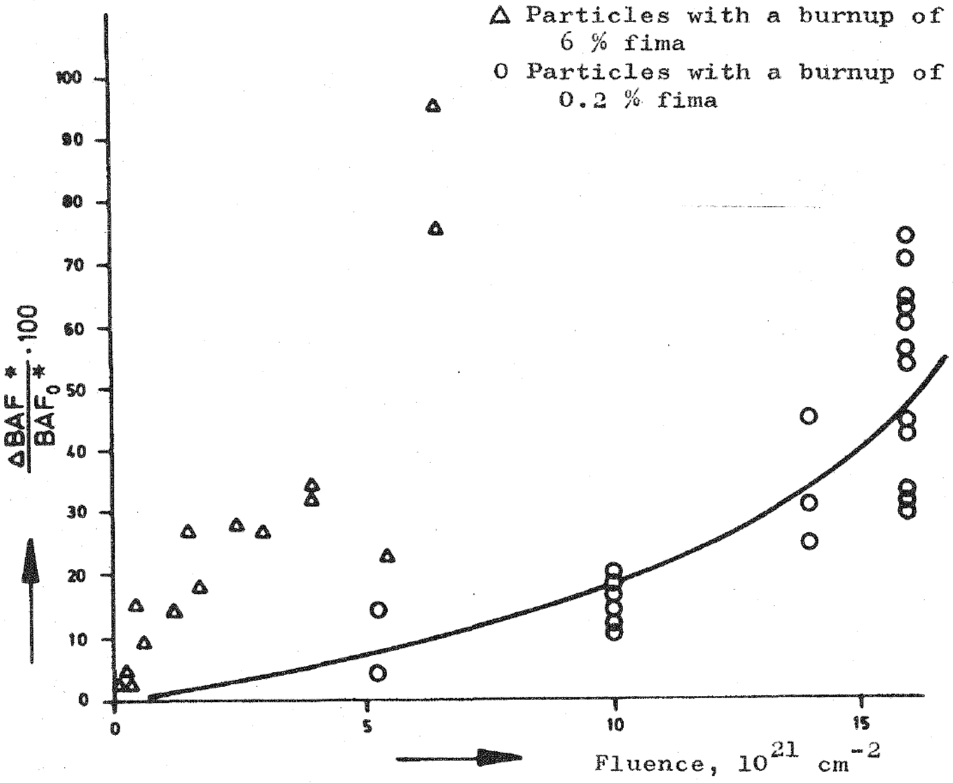
- Measure and report diattenuation (N)
  - $N = \frac{R_{max}-R_{min}}{R_{max}+R_{min}}$
  - N → 0 (isotropic)
  - N → ∞ (anisotropic)

G.E. Jellison Jr., J.D. Hunn, J. Nucl. Mats. (2008)

The Anisotropy in Irradiated PyC Evolves

K. Koizlik, GERHTR-48 (1972)





- Previous work identifies change in anisotropy for coated particle PyC as a function of burn up
- Data is not necessarily from representative samples or similar in pile conditions and could be a source of uncertainty

Particle tracking using XCT

- A set of eight randomly selected particles were measured using XCT
- Particles were then remeasured after initial oxidation testing at 1200^{o}C, 100 h with one particle's identity tracked
- The radius of layer interlayers were measured post test to attempt to match particles
- Just using radius is a good indicator but leaves uncertainty

	Mean Radius (μm)			
Particle ID	Kernel	Buffer	IPyC	SiC
1	204.40	315.22	352.87	390.11
2	197.03	312.14	349.42	386.52
3	206.22	305.61	343.97	376.24
5	201.14	308.86	349.34	386.87
6	204.27	307.83	345.98	384.22
7	214.61	315.66	352.83	389.86
8	192.22	297.75	336.62	376.24
Blind Test Particle	204.61	315.41	353.11	389.55

Likely Particle 1, but…

"Good" agreement highlighted in yellow