July 13, 2021

Tyler J. Gerczak
Sr. R&D Staff
Particle Fuel Forms Group Leader
Nuclear Energy and Fuel Cycle Division
Oak Ridge National Laboratory

Scanning Electron Microscopy of Irradiated TRISO Fuel Particles

ART
Advanced Reactor Technologies

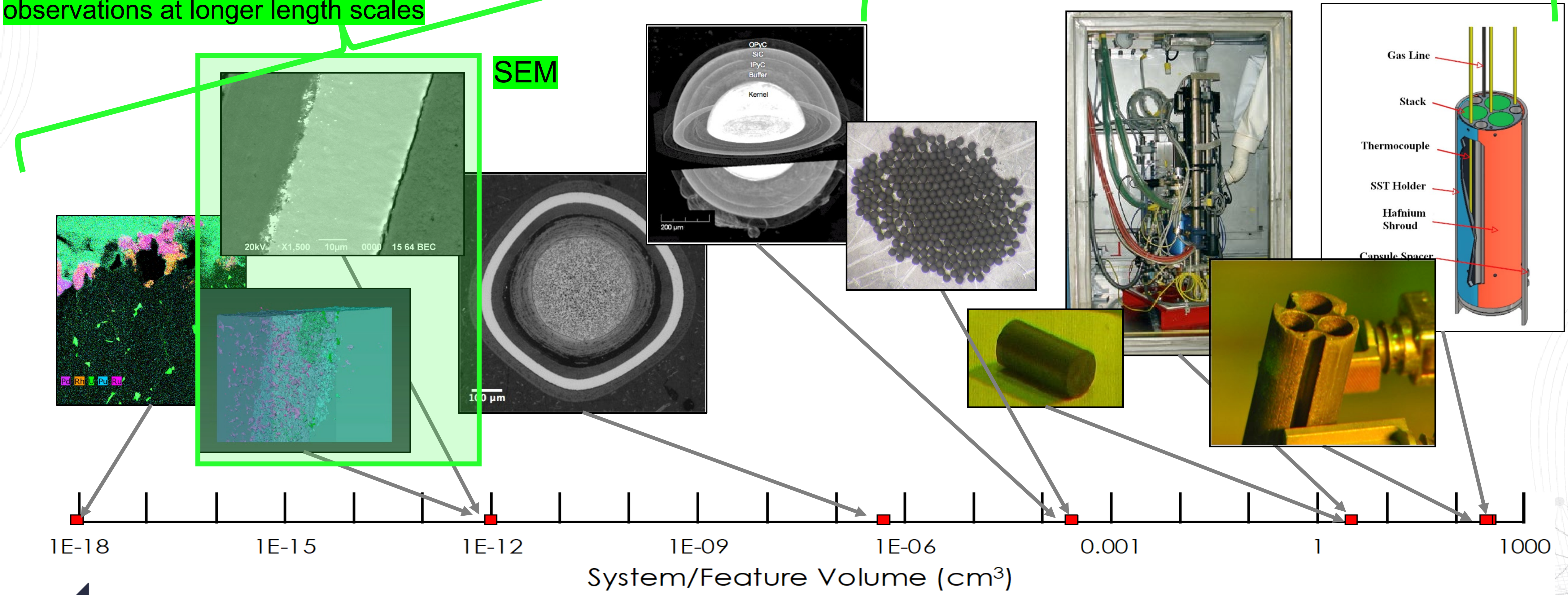
Scanning Electron Microscopy (SEM) Team

- Tyler Gerczak
- Brian Eckhart
- John Hunn
- Fred Montgomery
- Bob Morris
- Rachel Seibert
- Darren Skitt
- Irradiated Fuels Examination Laboratory (IFEL) Staff

Post Irradiation Examination (PIE): Understanding TRISO behavior across multiple length-scales

Mechanistic understanding of FP release/transport to provides interpretation for observations at longer length scales

Overall FP release behavior



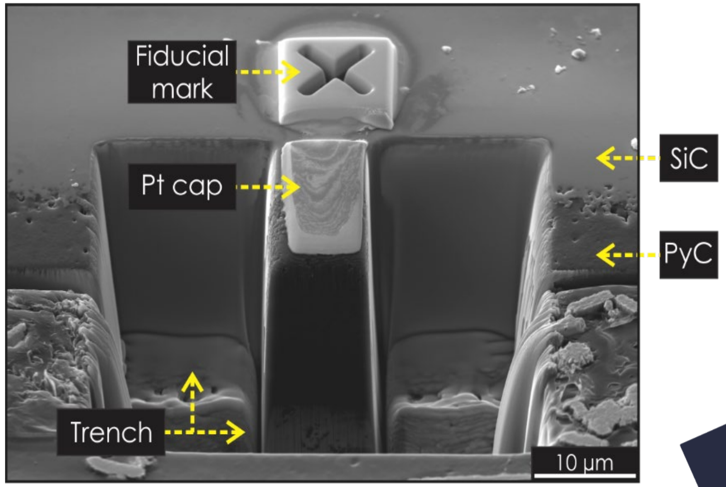
Each PIE effort informs a subsequent stage of analysis

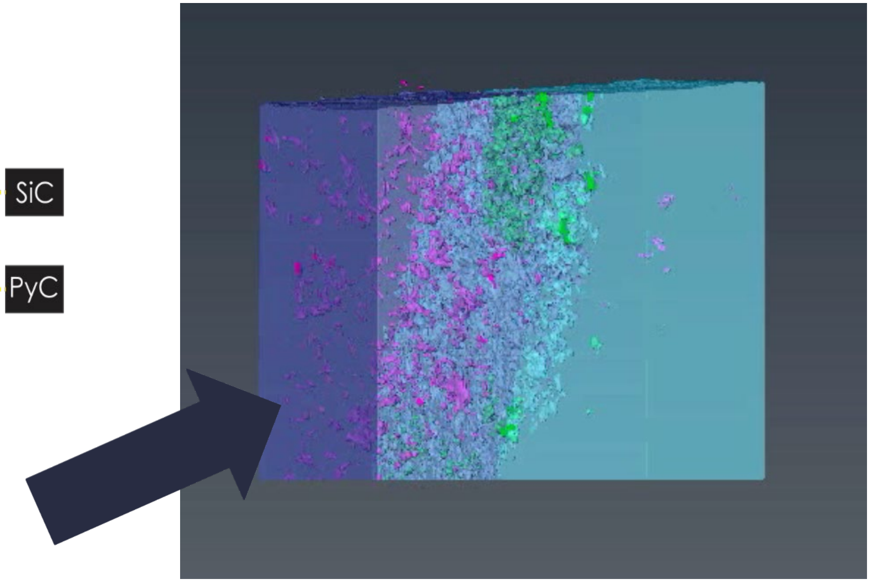
SEM PIE efforts

- SEM with energy dispersive spectroscopy (EDS)
  - Whole particle cross-sectional analysis to observe gross FP distribution and generic feature composition
  - Connect particle behavior with upstream/downstream PIE techniques
- **Focused ion beam/SEM three-dimensional analysis of TRISO layers and interfaces**
  - IPyC/SiC analysis: Determine the role of porosity and interfacial structure on FP accommodation
  - Localized SiC failure analysis: Understand nature of through layer failures
  - Buffer analysis: Quantify the buffer layer porosity and evolution under irradiation

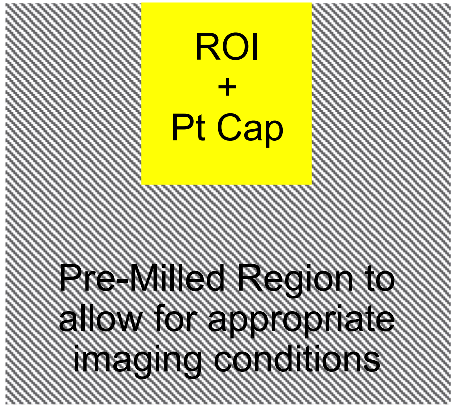
Serial sectioning 3D image analysis of using FIB/SEM

- **A systematic approach to studying the IPyC/SiC interface and buffer in irradiated TRISO fuel (comparison to the as-fabricated case)**
- Advantages:
  - Automation provides large data sets
  - Quantitative interfacial analysis
    - Lots of data to mine
    - "Depth Profiling"
    - Bridges between 2D SEM and STEM analysis length scales
  - Analysis is of a buried surface and interface is not impacted by sample preparation damage
  - Can extend to simultaneous EBSD/EDS Acquisition

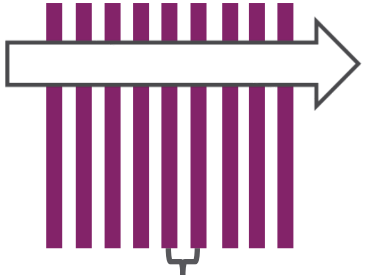




Process: Mill material in slices of defined thickness and image after each milling slice



Top schema



Slice schema

Serial Sectioning Test Matrix from AGR-2

Over 13,500 images collected to date!

Mount ID	Sample ID	Compact	Condition	Silver Content	Region Targeted	# Datasets	Status
-	LEU09-B01	-	Unirradiated	N/A	IPyC/SiC Interface	2	Complete
D51	RS01 RS07	5-4-2	Irradiated	Low	IPyC/SiC Interface	3	Complete
D55	RS25 RS33	5-4-2	Irradiated	High	IPyC/SiC Interface	2	Complete
-	LEU09-B01	-	Unirradiated	N/A	Buffer	1 set of 5, 1 set of 3	Data collected; Segmentation complete
F69	RS29 RS41	5-4-2	Irradiated	Intact & Fractured Buffer	Buffer	*1 – Inserted into FIB 7/2/21*	*Data collection in progress*
D44	RS11	5-4-1	1800ºC, 300 h safety testing	Low	IPyC/SiC Interface	4	Data Collected, Segmentation Refinement
D46	RS26	5-4-1	1800ºC, 300 h safety testing	High	IPyC/SiC Interface	4	Data Collected, Segmentation in Progress
D47	SP02	5-4-1	1800ºC, 300 h safety testing, failed SiC	NA	IPyC/SiC Interface	4	Data Collected, Segmentation in Progress

Serial Sectioning Test Matrix from AGR-2

Over 13,500 images collected to date!

Mount ID	Sample ID	Compact	Condition	Silver Content	Region Targeted	# Datasets	Status
-	LEU09-B01	-	Unirradiated	N/A	IPyC/SiC Interface	2	Complete
D51	RS01 RS07	5-4-2	Irradiated	Low	IPyC/SiC Interface	3	Complete
D55	RS25 RS33	5-4-2	Irradiated	High	IPyC/SiC Interface	2	Complete
-	LEU09-B01	-	Unirradiated	N/A	Buffer	1 set of 5, 1 set of 3	Data collected; Segmentation complete
F69	RS29 RS41	5-4-2	Irradiated	Intact & Fractured Buffer	Buffer	*1 – Inserted into FIB 7/2/21*	*Data collection in progress*
D44	RS11	5-4-1	1800ºC, 300 h safety testing	Low	IPyC/SiC Interface	4	Data Collected, Segmentation Refinement
D46	RS26	5-4-1	1800ºC, 300 h safety testing	High	IPyC/SiC Interface	4	Data Collected, Segmentation in Progress
D47	SP02	5-4-1	1800ºC, 300 h safety testing, failed SiC	NA	IPyC/SiC Interface	4	Data Collected, Segmentation in Progress

First to discuss

Serial Sectioning Test Matrix from AGR-2

Over 13,500 images collected to date!

Mount ID	Sample ID	Compact	Condition	Silver Content	Region Targeted	# Datasets	Status
-	LEU09-B01	-	Unirradiated	N/A	IPyC/SiC Interface	2	Complete
D51	RS01 RS07	5-4-2	Irradiated	Low	IPyC/SiC Interface	3	Complete
D55	RS25 RS33	5-4-2	Irradiated	High	IPyC/SiC Interface	2	Complete
-	LEU09-B01	-	Unirradiated	N/A	Buffer	1 set of 5, 1 set of 3	Data collected; Segmentation complete
F69	RS29 RS41	5-4-2	Irradiated	Intact & Fractured Buffer	Buffer	*1 – Inserted into FIB 7/2/21*	*Data collection in progress*
D44	RS11	5-4-1	1800ºC, 300 h safety testing	Low	IPyC/SiC Interface	4	Data Collected, Segmentation Refinement
D46	RS26	5-4-1	1800ºC, 300 h safety testing	High	IPyC/SiC Interface	4	Data Collected, Segmentation in Progress
D47	SP02	5-4-1	1800ºC, 300 h safety testing, failed SiC	NA	IPyC/SiC Interface	4	Data Collected, Segmentation in Progress

Second to discuss

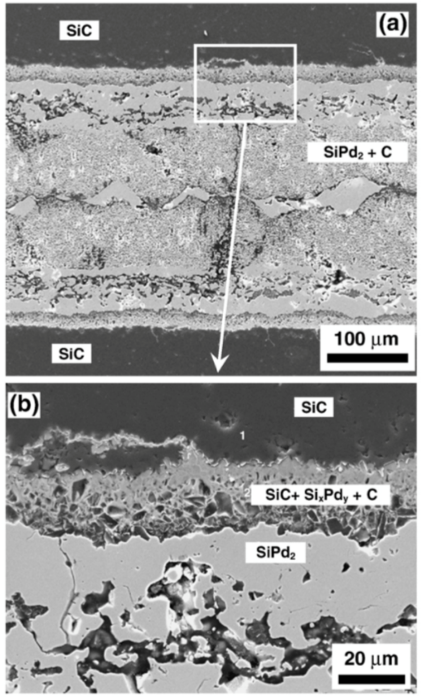
Serial Sectioning Test Matrix from AGR-2

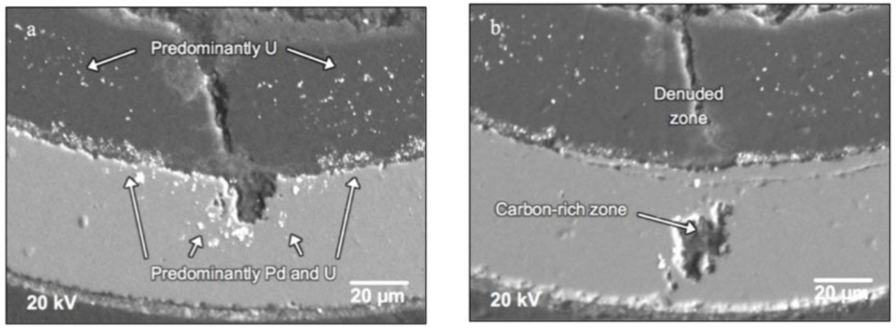
Over 13,500 images collected to date!

Mount ID	Sample ID	Compact	Condition	Silver Content	Region Targeted	# Datasets	Status
-	LEU09-B01	-	Unirradiated	N/A	IPyC/SiC Interface	2	Complete
D51	RS01 RS07	5-4-2	Irradiated	Low	IPyC/SiC Interface	3	Complete
D55	RS25 RS33	5-4-2	Irradiated	High	IPyC/SiC Interface	2	Complete
-	LEU09-B01	-	Unirradiated	N/A	Buffer	1 set of 5, 1 set of 3	Data collected; Segmentation complete
F69	RS29 RS41	5-4-2	Irradiated	Intact & Fractured Buffer	Buffer	*1 – Inserted into FIB 7/2/21*	*Data collection in progress*
D44	RS11	5-4-1	1800ºC, 300 h safety testing	Low	IPyC/SiC Interface	4	Data Collected, Segmentation Refinement
D46	RS26	5-4-1	1800ºC, 300 h safety testing	High	IPyC/SiC Interface	4	Data Collected, Segmentation in Progress
D47	SP02	5-4-1	1800ºC, 300 h safety testing, failed SiC	NA	IPyC/SiC Interface	4	Data Collected, Segmentation in Progress

Third to discuss

Why interface analysis: The nature of the IPyC/SiC interface influences FP interaction with the SiC layer

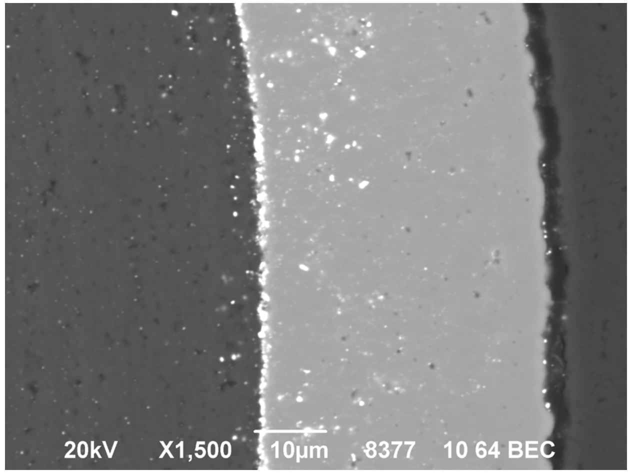




Localized Pd attack at exposed SiC caused a through layer defect with carbon-rich phases remaining from an AGR-1 1700°C safety-tested particle [2].

Exposed SiC surface adjacent to Pd foil heated to 1000°C for 10 h, indicating diffusion-dependent controlled reaction system [1].

212-RS08; ^{110m}Ag M/C ≤ 0.36
1800^{o}C 300 h Safety test



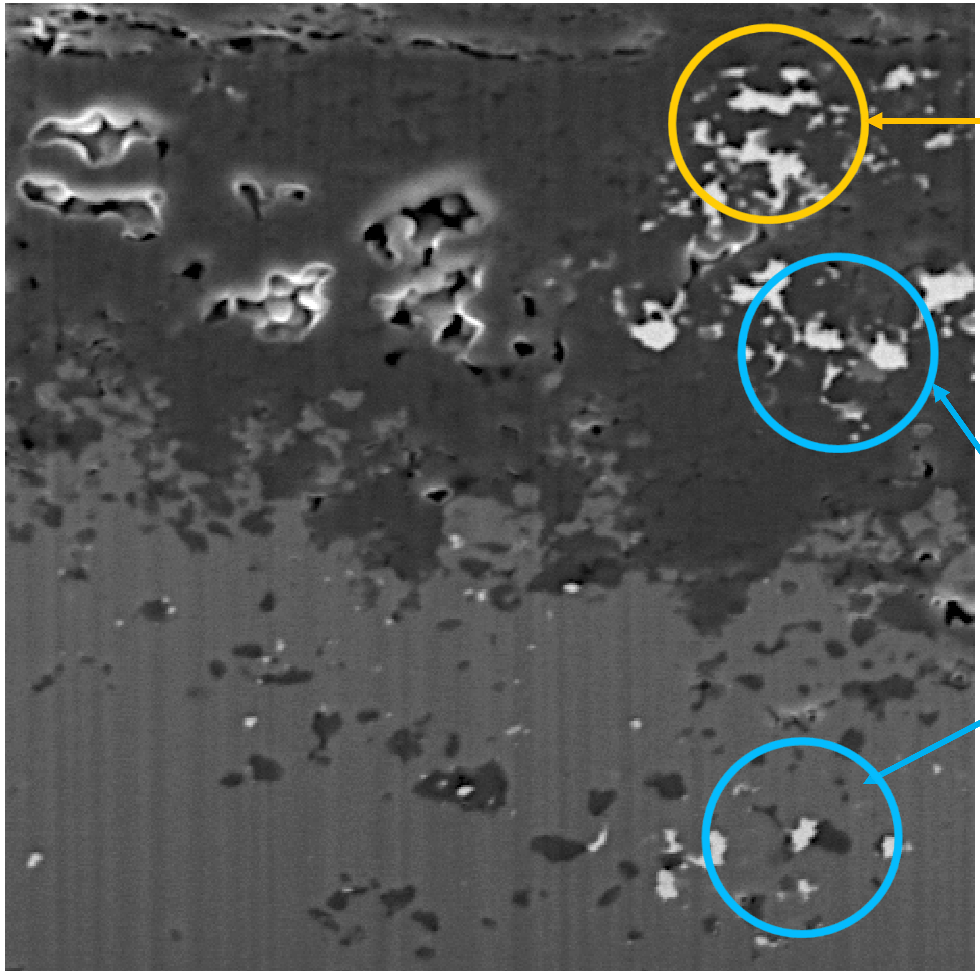
Intact SiC layer after 1800°C, 300 h with significant FP and actinide interaction and pileup

- Pd reacts with SiC at 400ºC [3], the IPyC layer protects the SiC layer during service and influences the nature of the Pd + SiC interaction
- Goal: Define the IPyC/SiC interface and compare unirradiated structure with irradiated structure to understand the features influencing the FP/SiC interaction in TRISO fuel

[1] Demkowicz, P. et al., 2008. High temperature interface reactions of TiC, TiN, and SiC with palladium and rhodium. Solid State Ionics 179 (39), 2313–2321.
[2] 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.
[3] Bhanumurthy, K., Schmid-Fetzer, R., 2001. Interface reactions between silicon carbide and metals (Ni, Cr, Pd, Zr). Compos. A Appl. Sci. Manuf. 32, 6.

Leverage 3D analysis to define interface and assess FP accommodation at IPyC/SiC

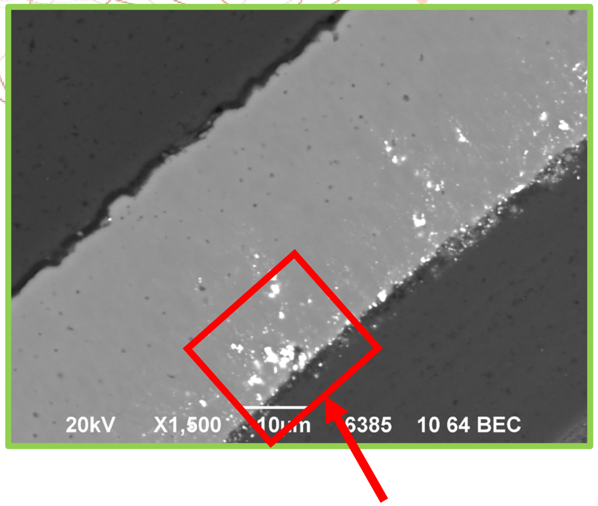
- Step 1: Define the interface and feature distribution in quantifiable metrics
  - “Thickness”
  - Effective surface area (SA) or available SiC surface for reaction
  - Measure total surface area for all permutations
  - SiC/IPyC, SiC/Porosity, SiC/FP, IPyC/FP, etc…
  - Pore/FP size and shape analysis
- Step 2: Compare data sets to understand how the layer structure changes and where the FPs are accommodated
  - Correlate FP location, frequency and shape with initial interface structure



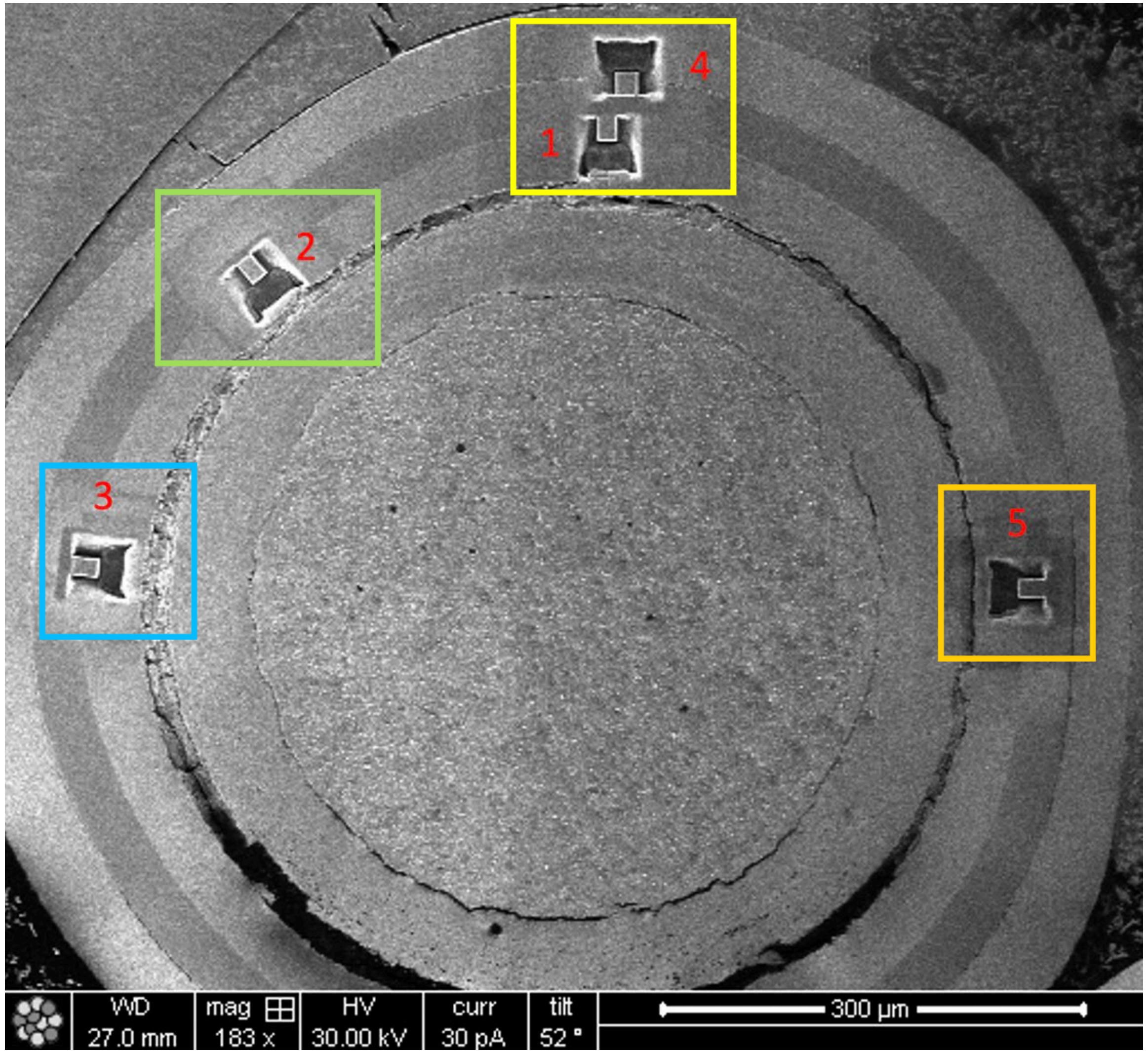
With 2D analysis we assume porosity accommodated FPs

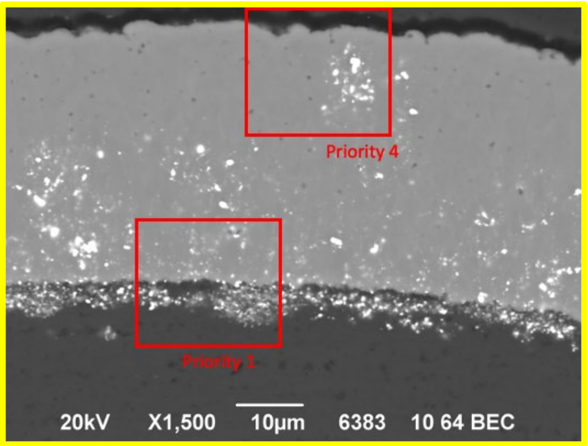
Visible FPs are located adjacent to C-rich regions within the SiC, and adjacent to SiC in the PyC

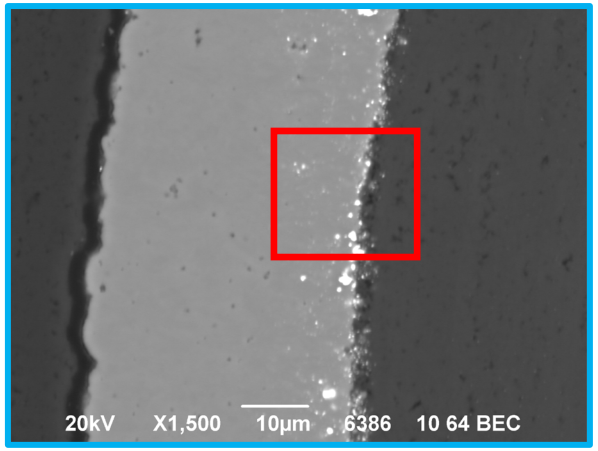
5-4-1 Particle – Low Ag example of variable FP distribution

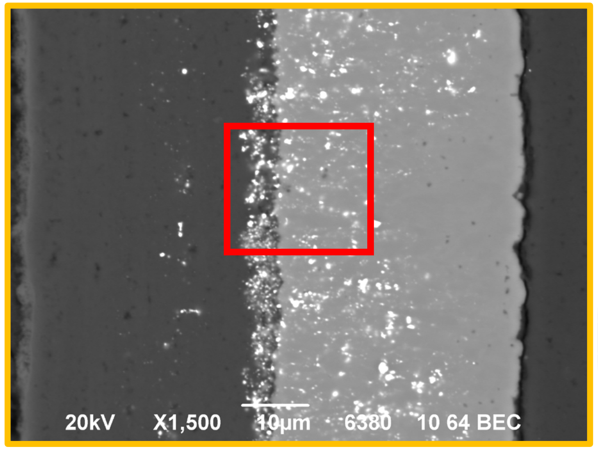


Locally corroded region in the absence of an IPyC crack





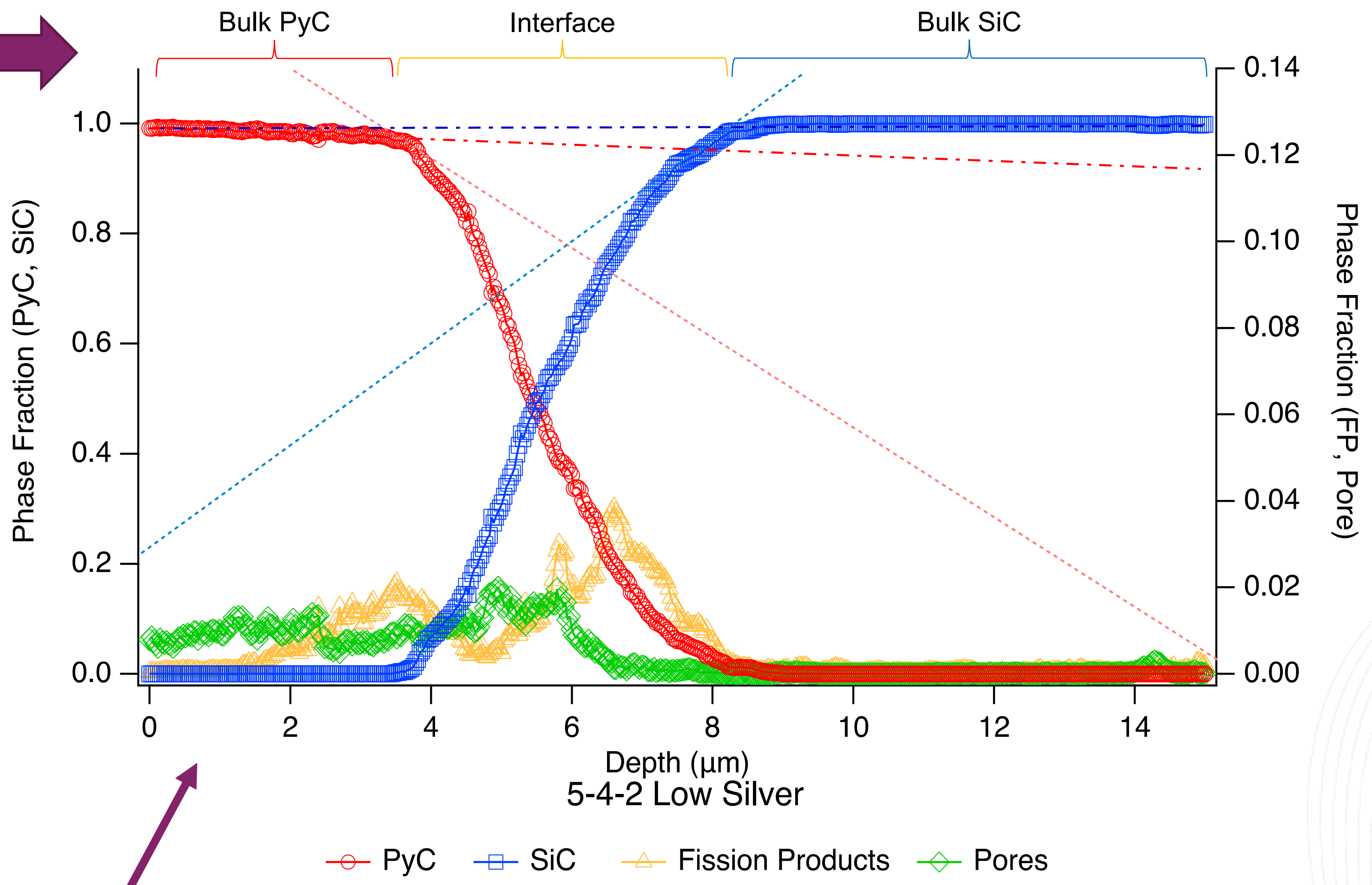


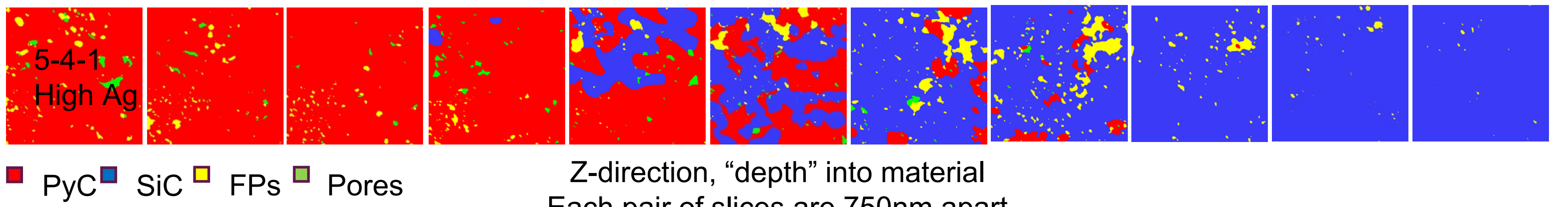


Red Boxes indicate locations of targeted scans

Interface definition for qualitative analysis

- Each slice is integrated to generate an effective depth profile for each phase
- The interface region is defined by the phase analysis, specifically by the deviation of the IPyC and SiC composition from their bulk composition
- Two lines are fit for each bulk phase and the region immediate region which deviates from bulk behavior
  - The intersection of these two points is where the interface is considered to begin and can be defined



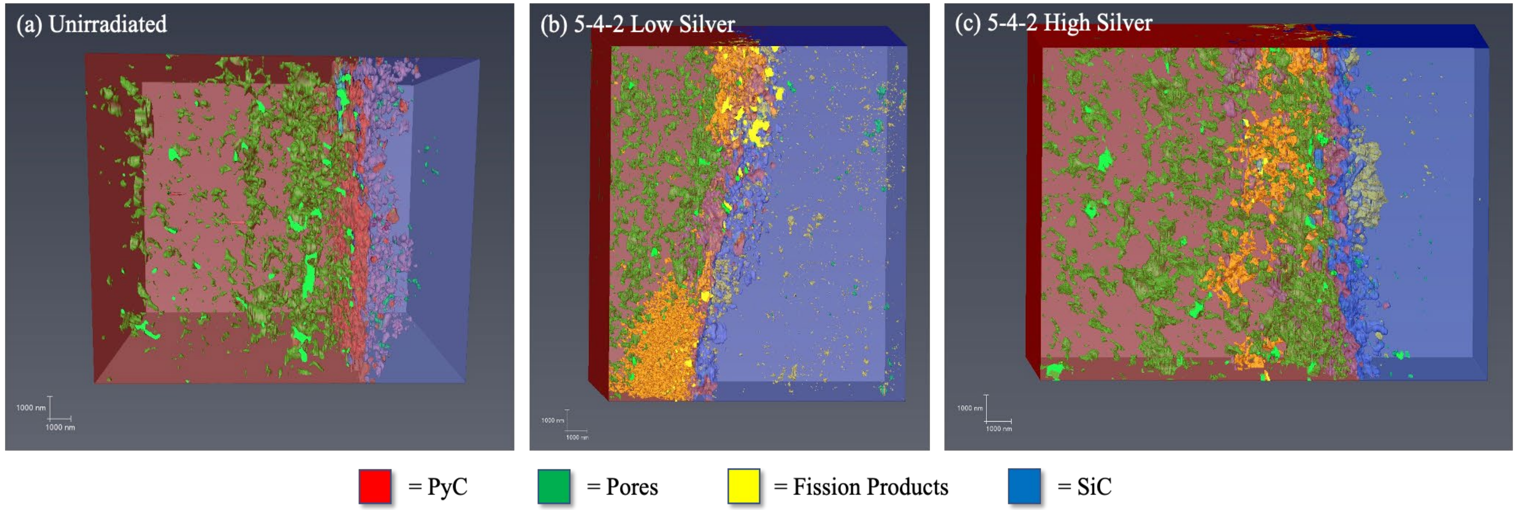


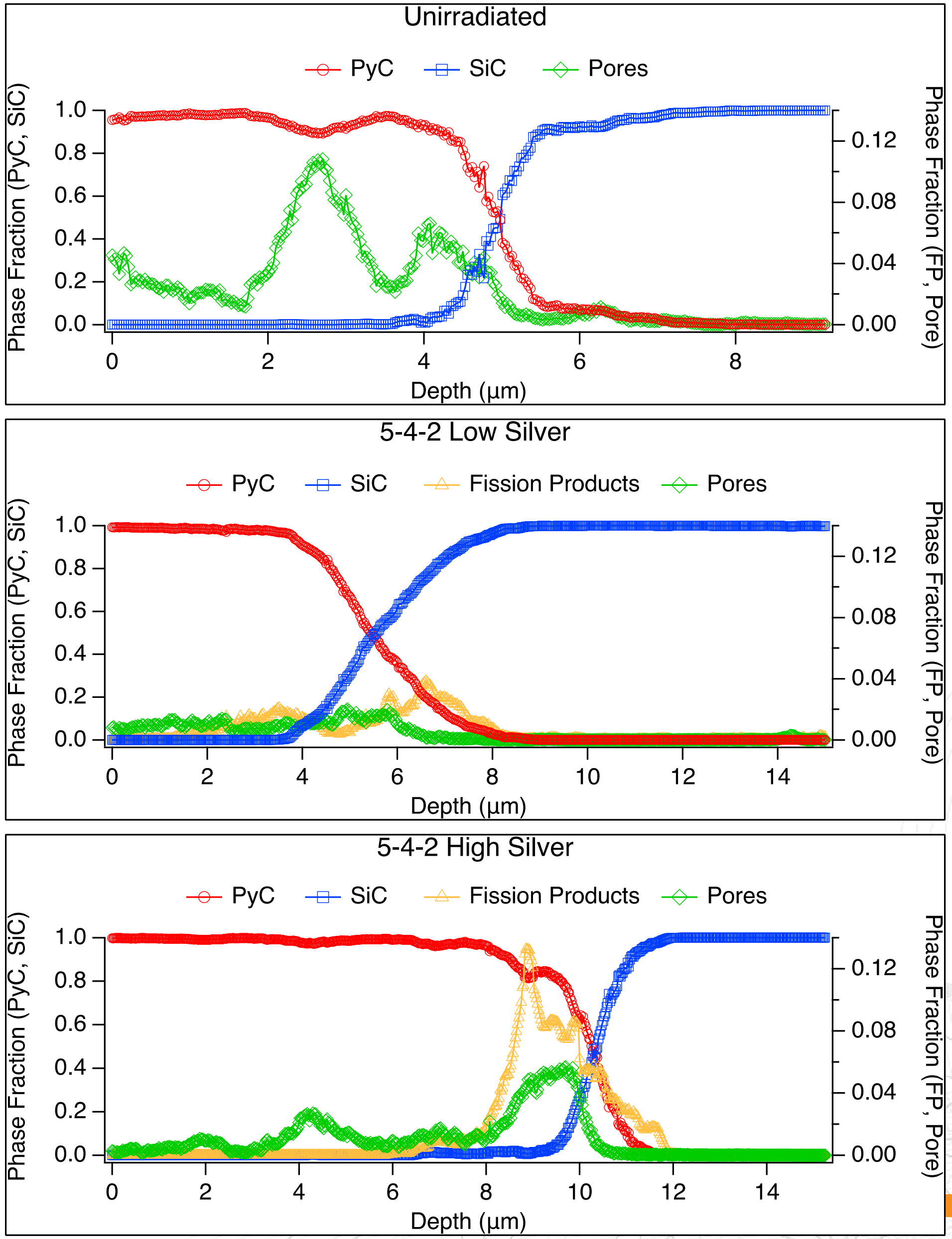
Z-direction, "depth" into material
Each pair of slices are 750nm apart

Interface Structure Detailed Example: 5-4-2

Provides opportunity to back out phase information in each region: Bulk IPyC, Interface, Bulk SiC

	Unirradiated	5-4-2 Low Silver	5-4-2 High Silver
Interface Thickness	3.70 μm	4.68 μm	3.89 μm
Avg. Porosity in Bulk PyC	4.28 ± 0.16%	0.91 ± 0.01%	0.92 ± 0.03%
Peak Porosity in Bulk PyC	10.83%	1.40%	2.71%
Avg. Porosity in Int. Region	2.05 ± 0.16%	0.81 ± 0.05%	2.28 ± 0.16%
Peak Porosity in Int. Region	6.60%	2.00%	5.67%
Avg. Porosity in Bulk SiC	0.09 ± 0.01%	0.02 ± 0.00%	0.01 ± 0.00%
Peak Porosity in Bulk SiC	0.32%	0.29%	0.06%
Avg. FP in Bulk PyC	--	0.67 ± 0.04%	0.14 ± 0.02%
Peak FP in Bulk PyC	--	2.10%	1.13%
Avg. FP in Int. Region	--	1.63 ± 0.07%	5.48 ± 0.26%
Peak FP in Int. Region	--	3.85%	13.25%
Avg. FP in Bulk SiC	--	0.08 ± 0.00%	0.06 ± 0.02%
Peak FP in Bulk SiC	--	0.32%	1.47%





Interface structure analysis

Effective Surface Area ($\mu m^2/\mu m^2$) of SiC in Interface

	Unirradiated	Irradiated (5-4-2)	Safety Tested (5-4-1; 1800°C, 300 h)
Low Silver	10.2	7.8	3.7
High Silver		6.9	6.5

Average FP in Interface

	Unirradiated	Irradiated (5-4-2)	Safety Tested (5-4-1; 1800°C, 300 h)
Low Silver	N/A	1.6 %	5.6 %
High Silver		5.5 %	3.8 %

Average Porosity in Interface

	Unirradiated	Irradiated (5-4-2)	Safety Tested (5-4-1; 1800°C, 300 h)
Low Silver	2.1%	0.8%	0.6%
High Silver		2.3%	1.2%

Average FP in bulk SiC

	Unirradiated	Irradiated (5-4-2)	Safety Tested (5-4-1; 1800°C, 300 h)
Low Silver	N/A	0.1%	0.5%
High Silver		0.1%	1.1 %

Interface structure analysis

Effective Surface Area ($\mu m^2/\mu m^2$) of SiC in Interface

	Unirradiated	Irradiated (5-4-2)	Safety Tested (5-4-1; 1800°C, 300 h)
Low Silver	10.2	7.8	3.7
High Silver		6.9	6.5

Average Porosity in Interface

	Unirradiated	Irradiated (5-4-2)	Safety Tested (5-4-1; 1800°C, 300 h)
Low Silver	2.1%	0.8%	0.6%
High Silver		2.3%	1.2%

High Silver	N/A	0.1%	1.1 %

- Apparent reduction of the SiC surface area (IPyC/SiC, porosity/SiC, FP/SiC) after irradiation and safety testing
- The observation suggests possible preferential reaction of SiC at the interface and/or interface restructuring under irradiation at the interface
- A general trend of increased FP phase fraction and decreasing porosity is observed (with exception of 542 high silver)
- Highlights the need to understand selection bias in targeted locations.

Interface structure analysis

Effective Surface Area ($\mu m^2/\mu m^2$) of SiC in Interface

	Unirradiated	Irradiated (5-4-2)	Safety Tested (5-4-1; 1800°C, 300 h)
Low Silver	10.2	7.8	3.7
High Silver		6.9	6.5

Average Porosity in Interface

	Unirradiated	Irradiated (5-4-2)	Safety Tested (5-4-1; 1800°C, 300 h)
Low Silver	2.1%	0.8%	0.6%
High Silver		2.3%	1.2%

Average FP in Interface

	Unirradiated	Irradiated (5-4-2)	Safety Tested (5-4-1; 1800°C, 300 h)
Low Silver	N/A	1.6 %	5.6 %
High Silver		5.5 %	3.8 %

542	% FP/SiC	%FP/porosity SA	%FP/IPyC SA
Low Silver	75.0%	1.0 %	5.6 %
High Silver	19.5%	0.8 %	79.7 %

- Opportunity to drill deeper and understand how the FPs are positioned in the interface by looking at breakdown of FP surface area distributions in the defined interface region
- Data suggests occupation of some pores and direct segregation at IPyC/SiC interfaces

Interface Structure Analysis

- Even with bias in area selection, an increase in FP phase fraction moving into the SiC is observed in the 541 safety tested particles
- This reflects a FP concentration profile and potential to infer transport kinetics

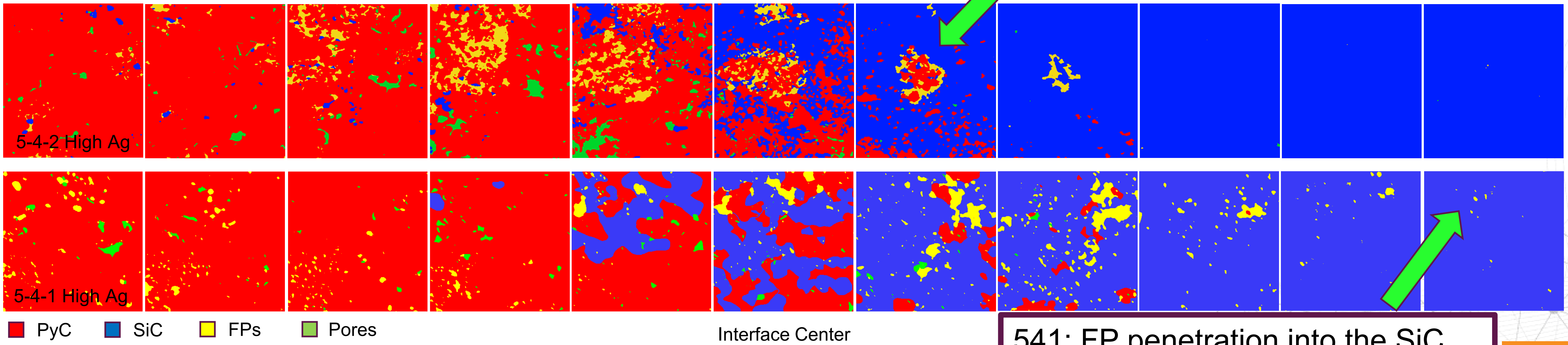
Average Porosity in Interface

	Unirradiated	Irradiated (5-4-2)	Safety Tested (5-4-1; 1800°C, 300 h)
Low Silver	2.1%	0.8%	0.6%
High Silver		2.3%	1.2%

Average FP in bulk SiC

	Unirradiated	Irradiated (5-4-2)	Safety Tested (5-4-1; 1800°C, 300 h)
Low Silver	N/A	0.1%	0.5%
High Silver		0.1%	1.1 %

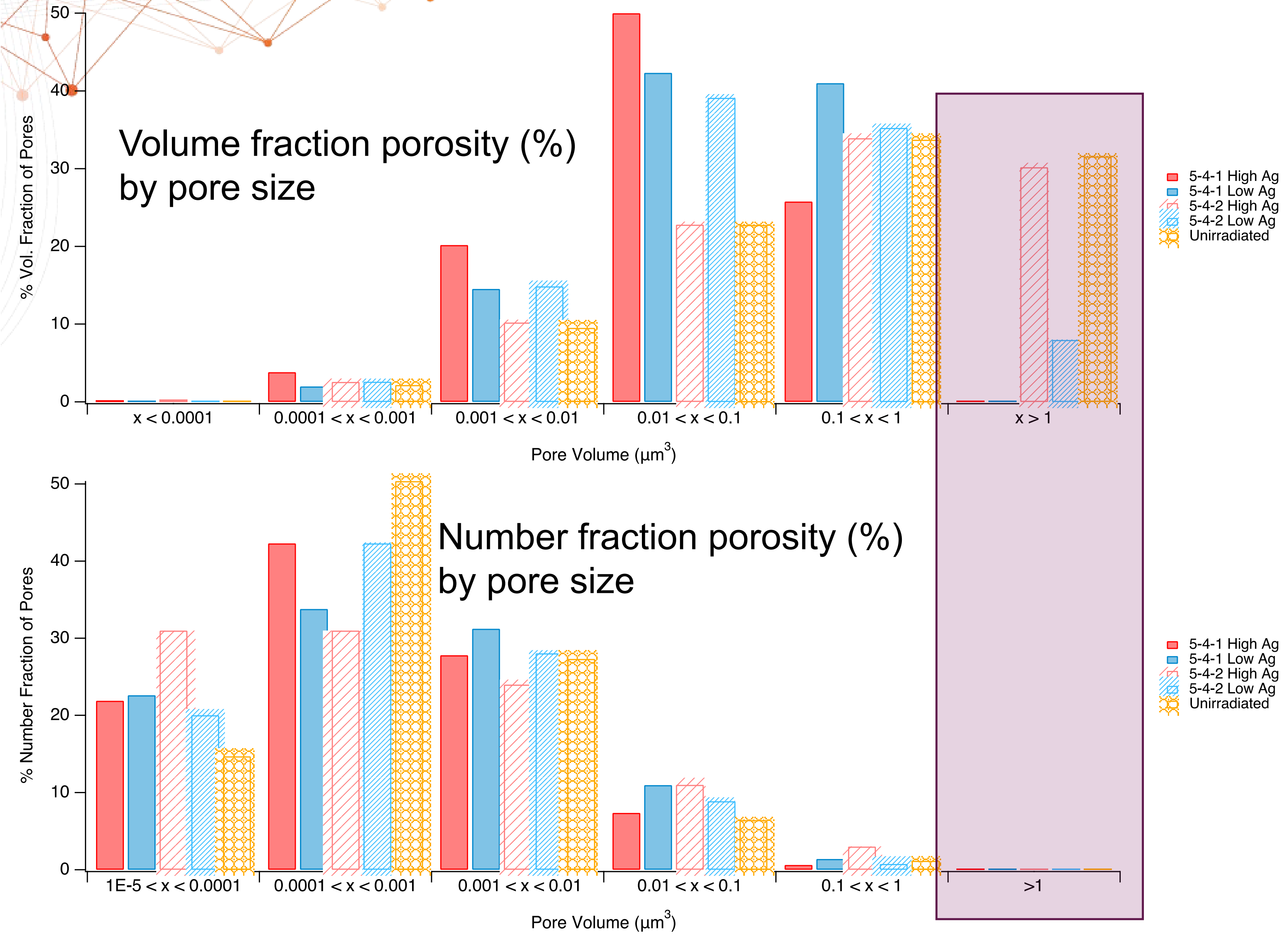
542: FP at SiC interface surface



Interface Center
The distance between two slices is 750 nm

541: FP penetration into the SiC

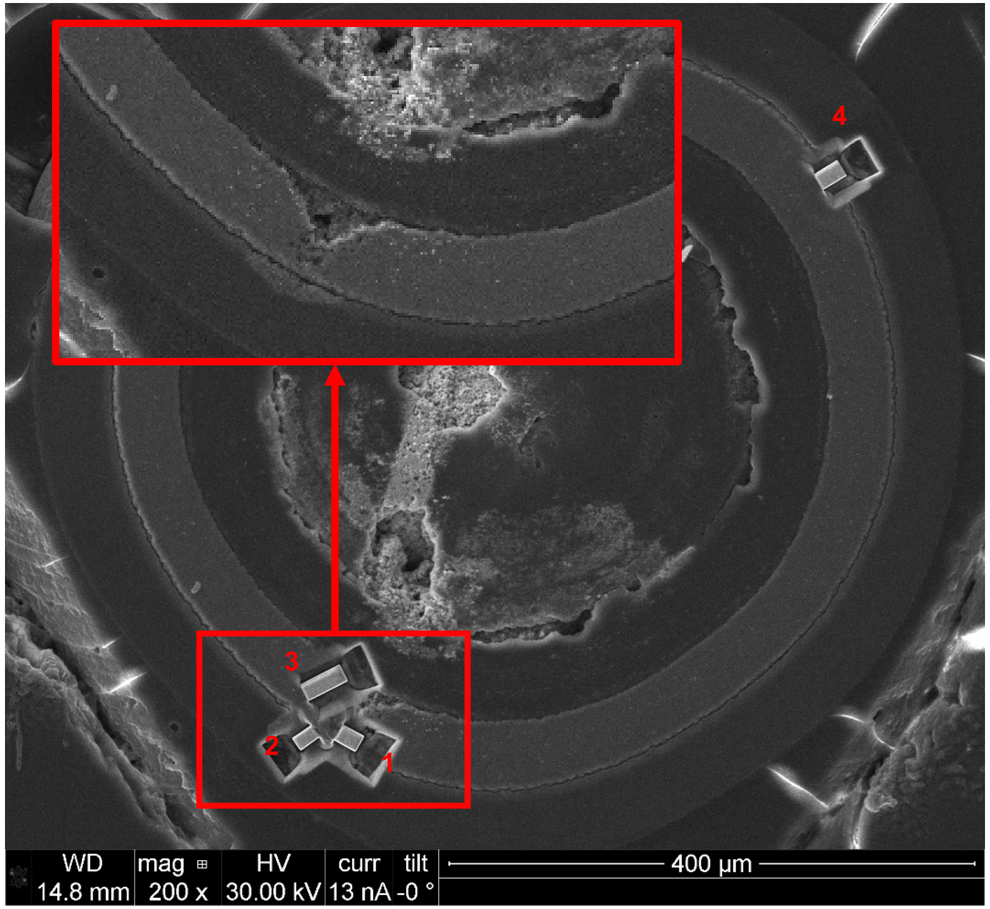
Pore and FP size analysis



- Each individual FP or pore feature can be isolated for shape (not shown)and size analysis
- Comparing number fraction and volume fraction provides opportunity to understand if a few features dominate the statistics
- Primary take away: is 3D Slice and View generates a lot of quantitative data and we need to be smart about how we address analysis bias and draw conclusions: this is underway

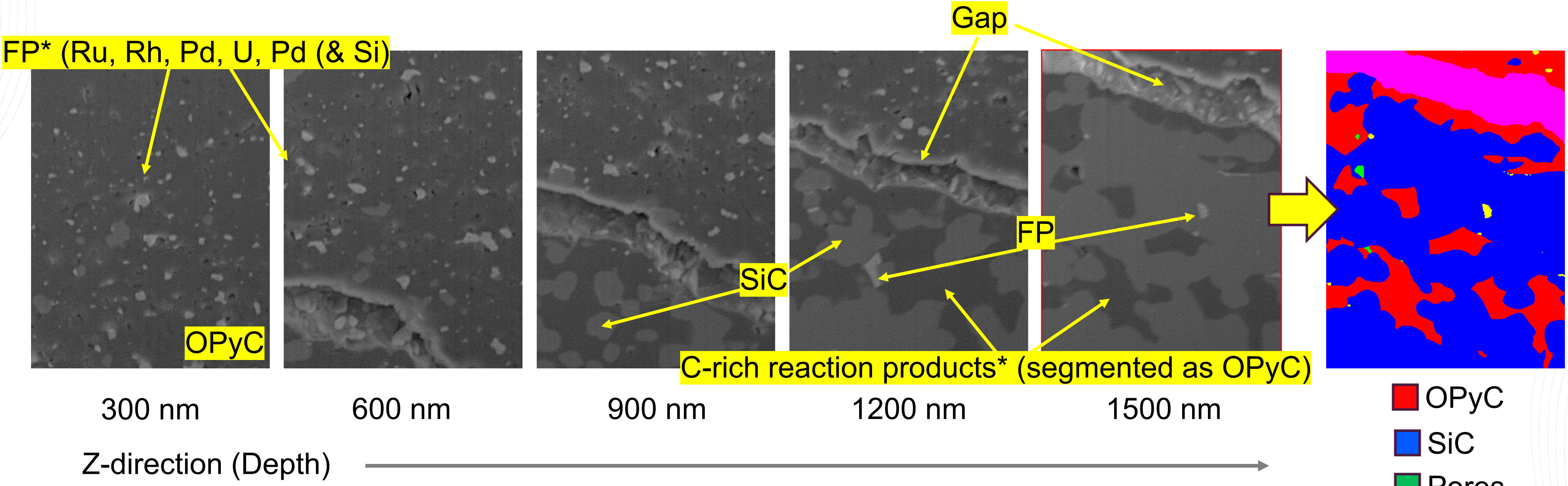
Status 541 Special Particle

- Special Particle chosen due to locally corroded SiC region
  - Three areas of the corrosion were examined, and one area far from/without corrosion (control comparison)
- All regions have been analyzed and segmentation and data interpretation is in progress.



541 Special Particle (cont.)

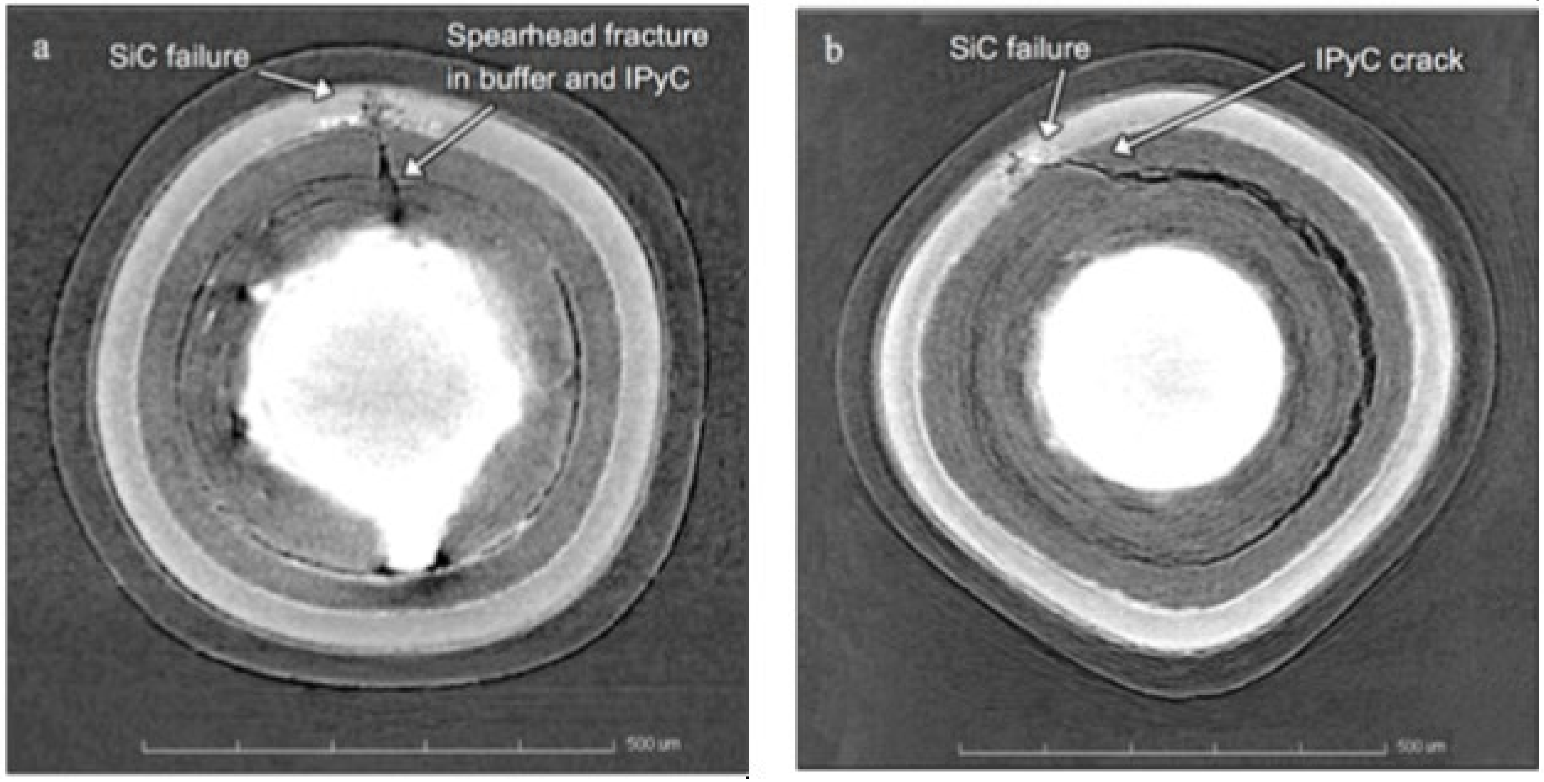
- Example from location 2, which covers both the OPyC and SiC



- Exploring the advanced reaction interface to provide insight into how the reaction proceeds and how typically well retained FPs* move out of the locally corroded region

Why buffer analysis: The predominate TRISO particle failure mechanism is influenced by buffer densification

AGR-1 UCO-TRISO Typical Localized Failures After Safety Testing[1]

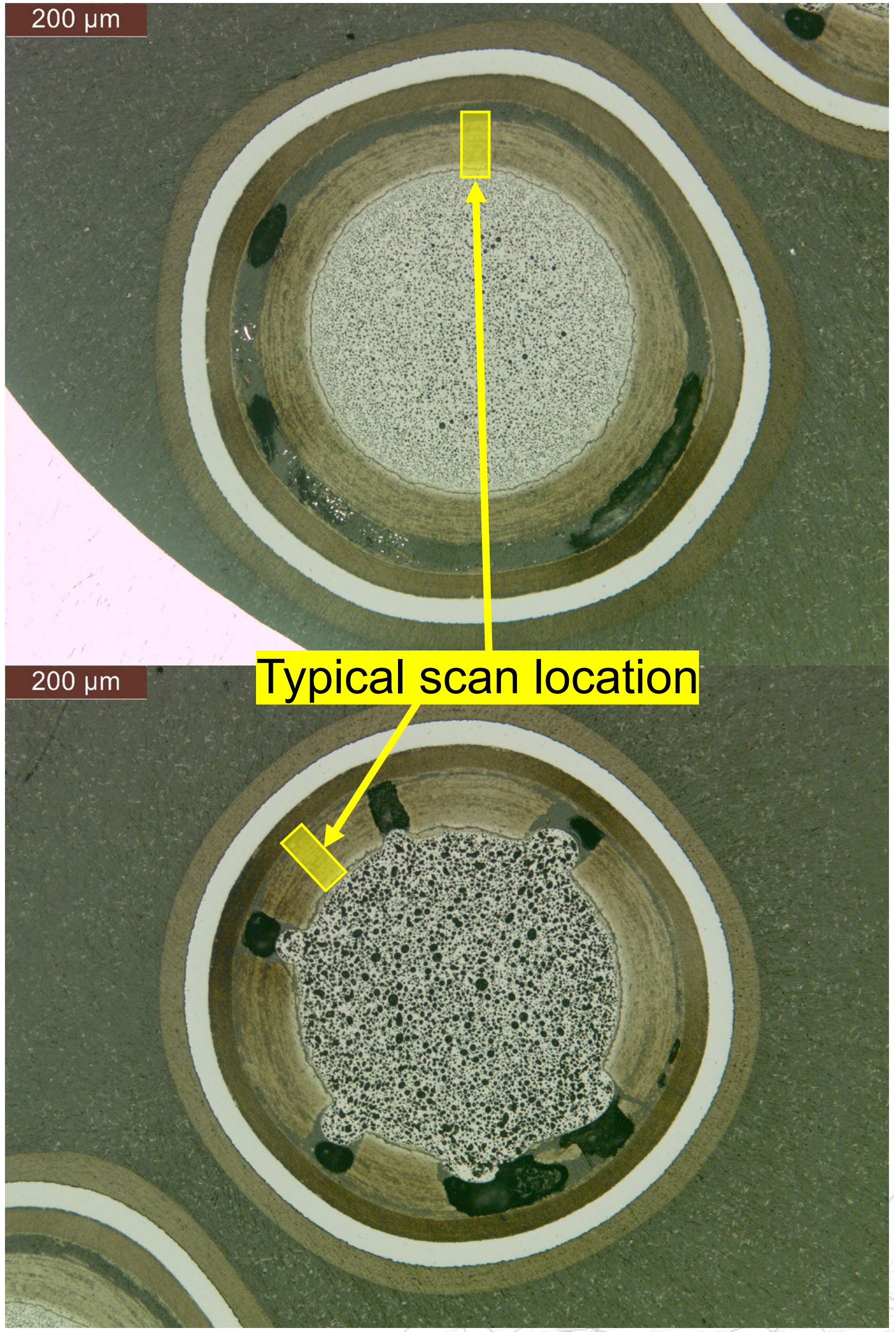


AGR UCO TRISO predominate failure sequence: **incomplete Buffer/IPyC tearing** leading to IPyC crack which exposes the SiC layer and leads to metallic fission product attack (Pd)[1]

[1] J.D. Hunn et al., Nucl. Eng. Des., 306 (2016) 36-46.

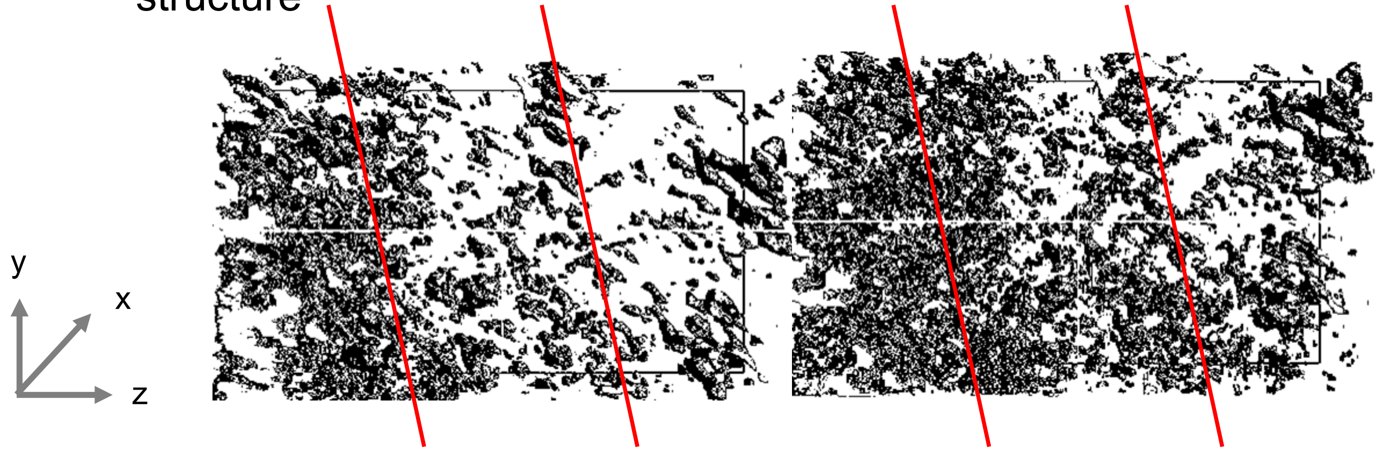
3D Buffer Analysis

- Goal: Develop general understanding of buffer pore structure and its response to irradiation
  - Provide information on relationship between processing and buffer microstructure and densification mechanism
- Approach: compare unirradiated pore structure with intact and fractured buffer
- Exploring intact and fractured analysis will provide insight on densification under different buffer constraints
  - AGR-2 Compact 542: 23% intact, 77% fractured

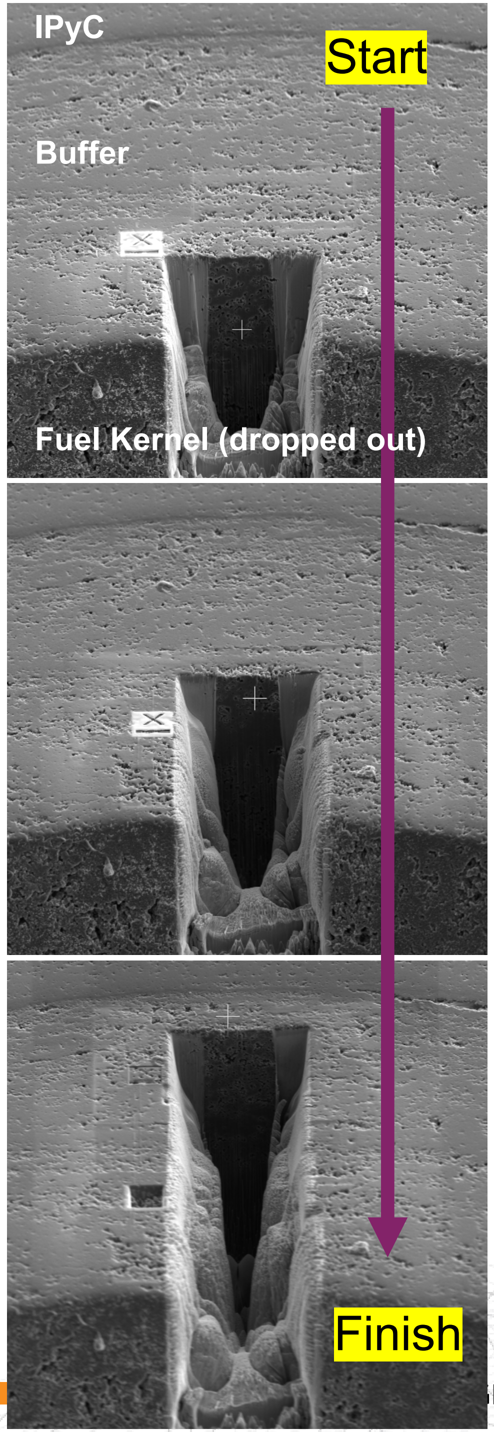


Initial Findings: Unirradiated Buffer Data

- Data is acquired across entire buffer thickness
  - Data under analysis for as-fabricated unirradiated condition; Data acquisition in progress for irradiated samples (inserted into FIB/SEM July 2nd)
- Initial pore reconstruction complete through 40 μm from the kernel
  - Initial analysis suggests variable porosity bands aligned circumferentially (red lines below) and expected to be resultant from variable fluidization process
  - Next step is applying pore shape and size analysis to quantitatively define structure

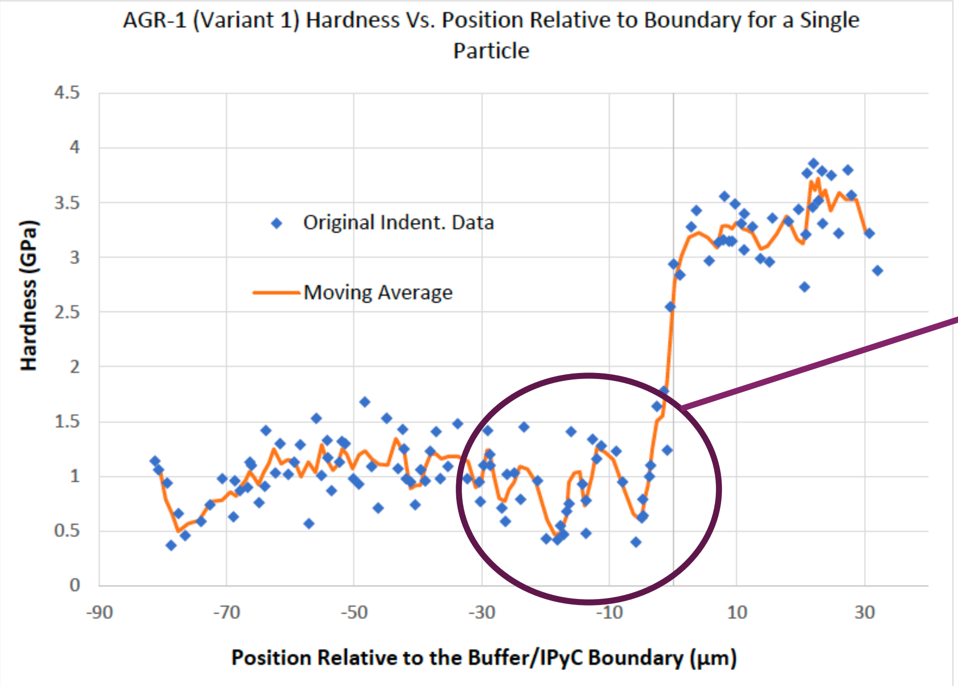


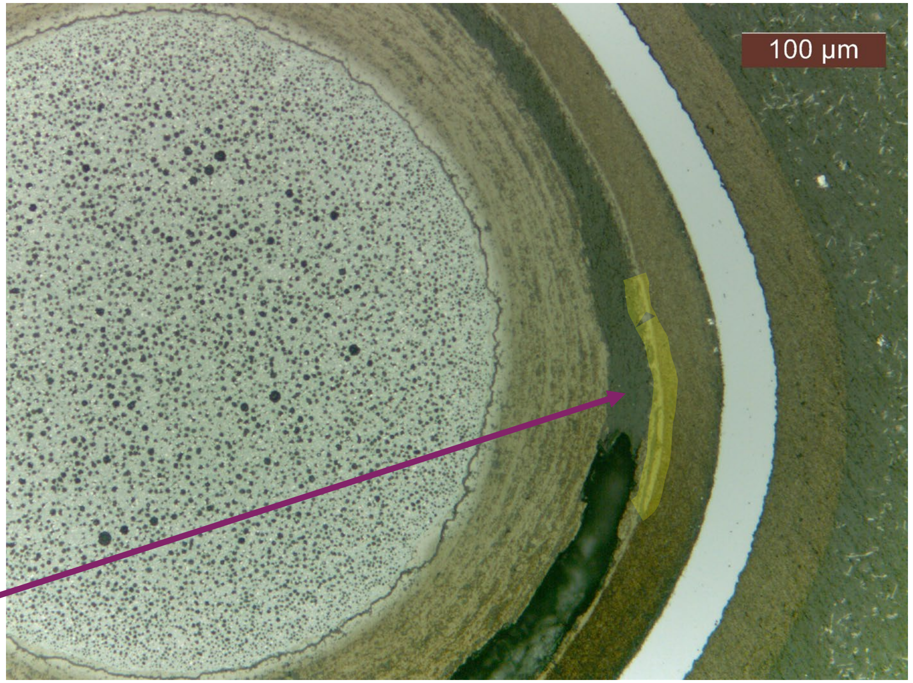
Radial direction is approximately proportional to z-direction (black is porosity)



Striations may have importance regarding internal buffer tearing

- Possible correlation with buffer striations and internal tearing mechanism





Optical cross-section of an AGR-2 Compact 542 selected for slice and view analysis showing residual buffer attached to IPyC layer

Minimum Hardness was ~10 micrometers from discreet Buffer/IPyC interface and hardness appeared to track with the banded buffer striations [1]

[1] A.T. Schumacher, Characterizing and Comparing Tristructural-Isotropic Buffer Properties from AGR-1 and AGR-2 Irradiation Experiments, M.S. Thesis, University of Tennessee, 2019

Summary of 3D analysis efforts

- 3D Slice and View Analysis is a powerful tool for providing quantitative data to describe complex interface and layer properties
- New approaches to quantifying interface structure have been established which is the first step to connect interface structure and properties to TRISO particle performance
  - The IPyC/SiC layer interface governs FP/SiC interactions and 3D Slice and View provides quantitative metrics describing the interface to inform how the interface structure influences the accommodation of FPs
  - The buffer densification play a primary role in the rare instances of particle failure and interface analysis provides opportunity to provide information on the basis for the failure phenomena
- The complexity of the data (+13.5k images to date) and small analysis volume relative to particle size and number of particles in AGR-2 requires caution and proper perspective when drawing conclusions regarding general TRISO behavior (entered with a systematic approach to back out relevant information but now need to take a cautious approach as new information presents itself)
  - Two publication are planned to be submitted in FY21 regarding AGR-2 Compact 542 and AGR-2 Compact 541 IPyC/SiC interface structure and fission product analysis
    - R.L. Seibert, T.J. Gerczak, J.D. Hunn, “AGR-2 Compact 5-4-2 SiC/IPyC Interface Analysis Using FIB-SEM Tomography,” awaiting export control review in Resolution and is to be submitted to Journal of Nuclear Materials July 31st
    - R.L. Seibert, T.J. Gerczak, J.D. Hunn, “AGR-2 Compact 5-4-1 Safety Tested Interface Analysis Using FIB-SEM Tomography,” draft near ready for submission to ORNL’s Resolution system

Questions?

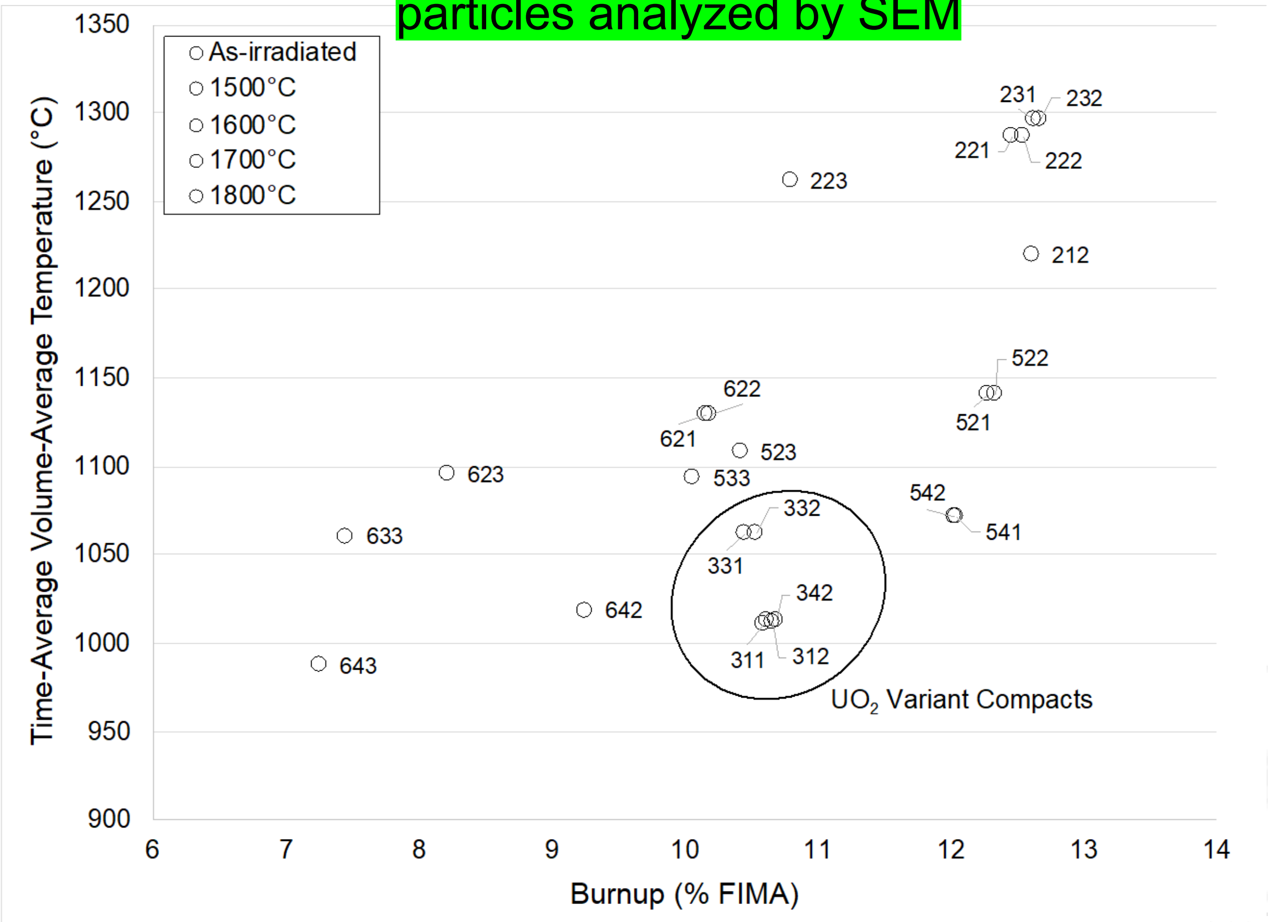
Tyler Gerczak, ORNL
gerczaktj@ornl.gov

- This work was sponsored by the US Department of Energy Office of Nuclear Energy through as part of the Advanced Gas Reactor Fuel Development and Qualification Program.

SEM/EDS analysis provided a range of insight on particle behavior

- **Understand the impact of…**
- a range in irradiation conditions
  - Fast fluence
  - Burnup
  - TAVA Temperature (ºC)
  - Temperature variation (e.g. targeted particle selection based on silver retention)
- the impact of safety testing
- the impact of kernel composition
- the impact of particle internal structure
- **Understand the root cause of infrequent particle failure**
- **Support upstream and downstream PIE efforts**
- **Results have been disseminated in publications, reports, and presentations over the course of the AGR-2 PIE effort**

24 compacts and over 220 individual particles analyzed by SEM

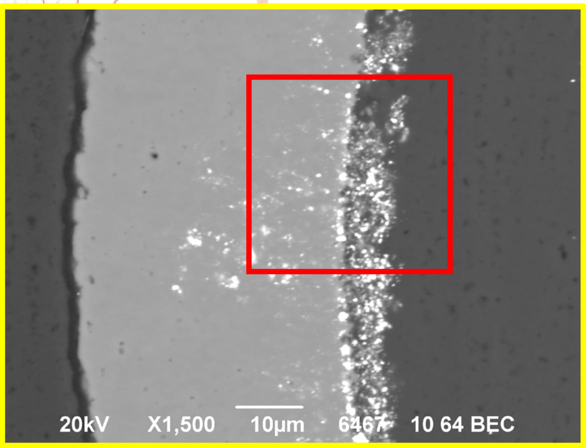


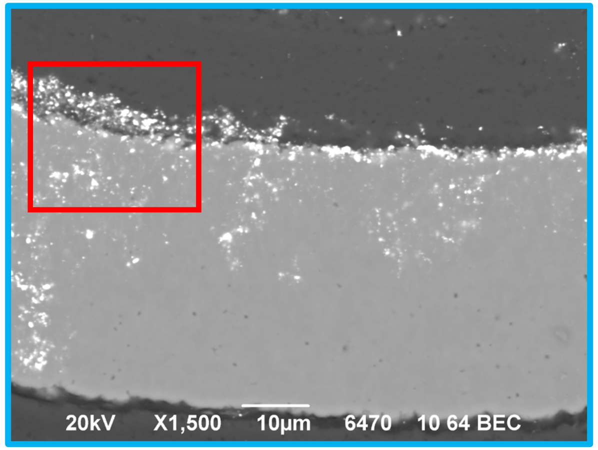
- Temperature; TAVA = time-average, volume-average

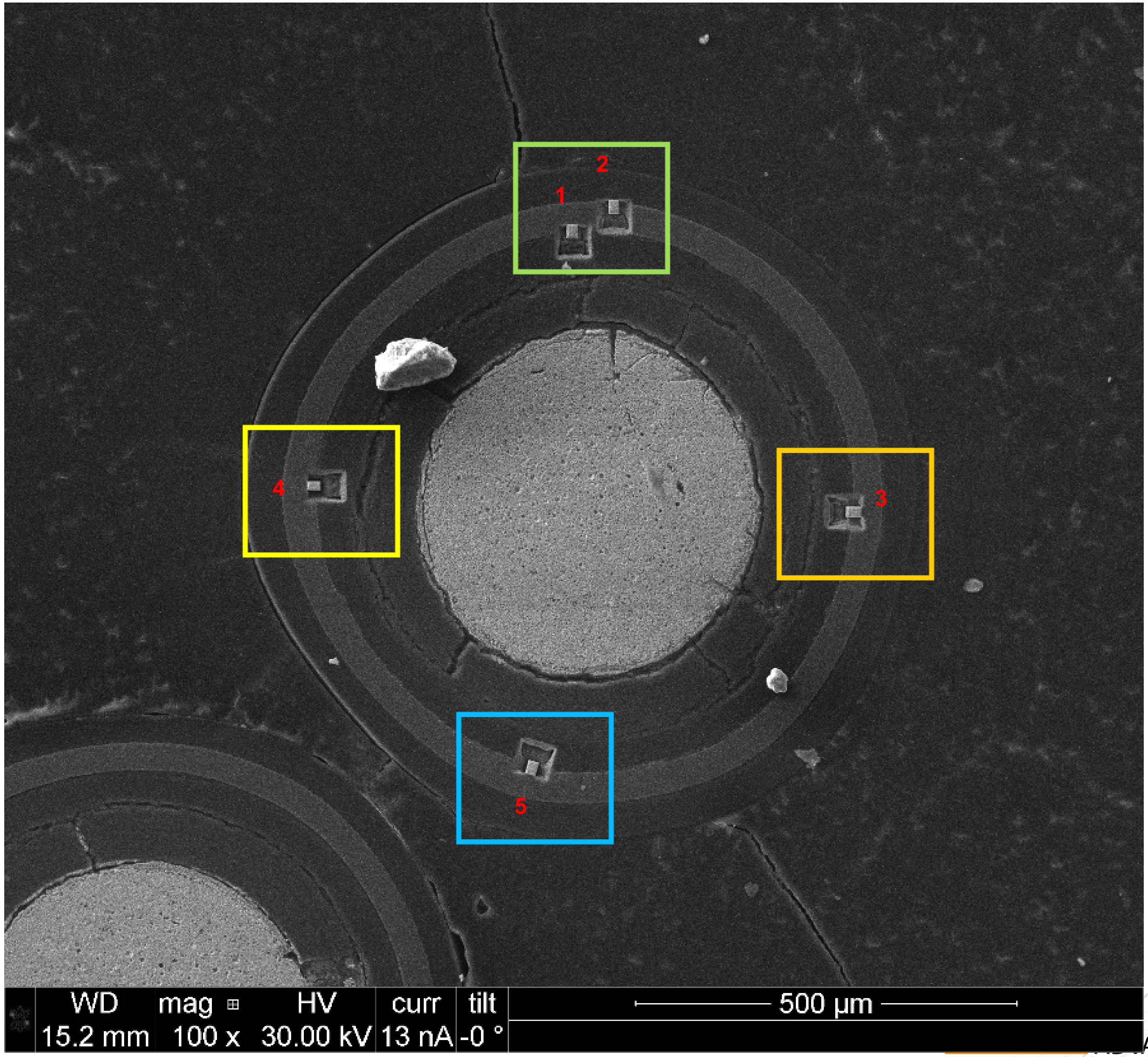
Type of Analysis Completed on AGR-2 Samples

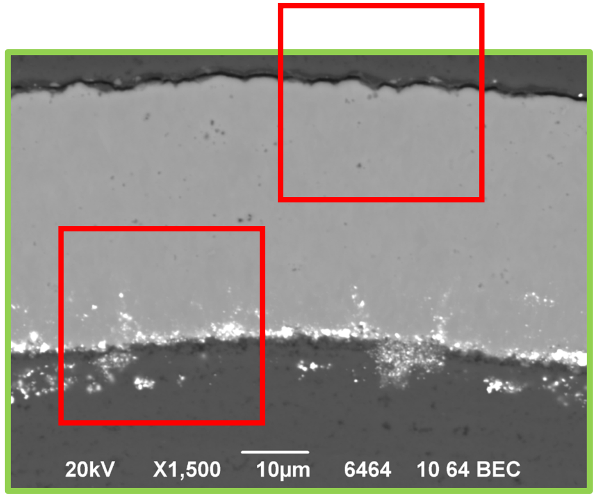
- Interface/Buffer Structure
  - Material composition & spatial information, to determine possible connections between material location and unfavorable behavior in-pile
  - Effective surface areas, to determine if available SiC surface area effects fission product accommodation
  - 3D visual reconstruction, to view the 3D surface as a function of material
- Pore/Fission Product Structure
  - Frequency and size of pores/FPs, to determine if there is preferential formation or types at different locations of interest
  - Shape of pores/FPs, to determine if there is preferential formation at different locations of interest
  - Distribution as function of size and distance to interface

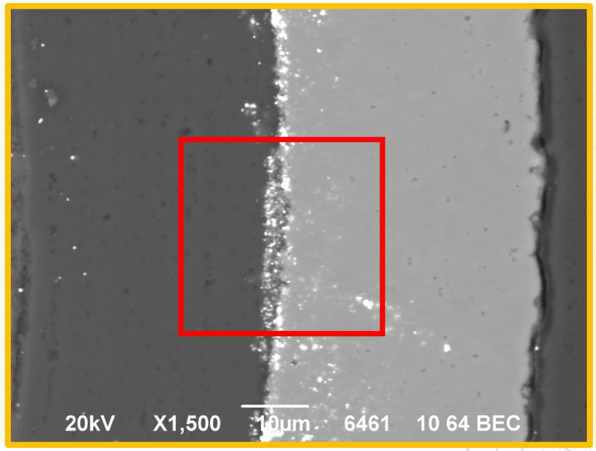
5-4-1 Particles – High Silver – FP distribution is variable analysis may be biased











Pore and FP size analysis