OAK RIDGE National Laboratory

Overview of New Quality Control Methods Development at ORNL in FY24

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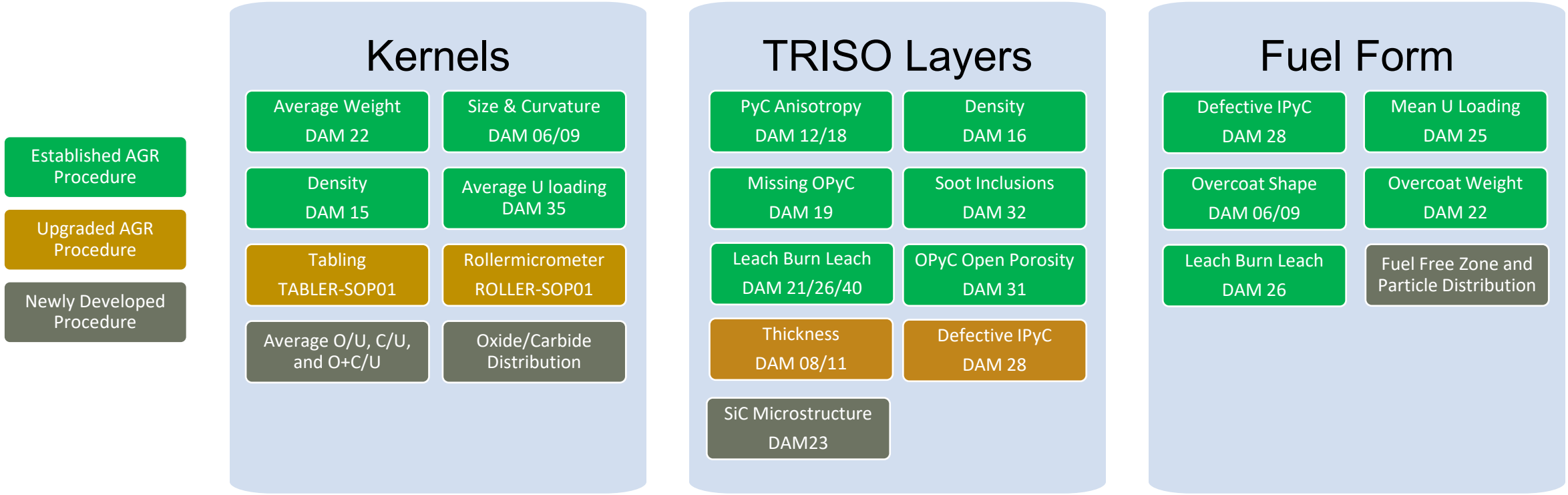
Building on the AGR Characterization Foundation



- TRISO fuel requires a wide range of characterization methods at each stage of fabrication for quality control (QC).
- A foundation of Data Acquisition Methods (DAM's) has been developed at ORNL as part of the AGR Program.
- Further development of new QC characterization methods has continued through various programs.

Better, Faster, Cheaper

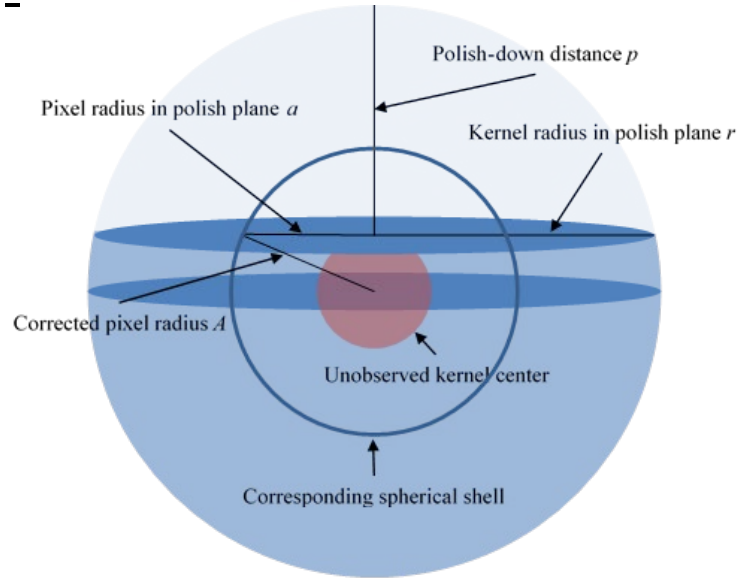
- The primary goal driving TRISO QC characterization methods development is to make new methods better, faster, and cheaper to support economic industrial scale fuel production.
  - Better = more data
    - Qualitative → Quantitative
    - Mean Values → Statistical Distributions
  - Faster = computer automated
  - Cheaper = simpler equipment and less waste
- This talk will focus on four characterization methods investigated at ORNL for use in TRISO QC characterization.

UCO Phase Quantification – Historical Approach

- Prepare and run 3-5 samples of of kernels in a mercury porosimeter to determine average kernel density.
  - Dispose of mixed mercury/LEU waste.
- Prepare and run 3-5 samples of kernels in a LECO CS analyzer to measure average carbon content.
- Prepare and run 3-5 samples of kernels in a LECO ON analyzer to measure average oxygen content.
- Use Davies-Gray titration or mass spectrometry with kernels dissolved in nitric acid to measure average uranium content.
- Calculate mean O/U, C/U, and C+O/U ratios and density.

UCO Phase Quantification – New Approach

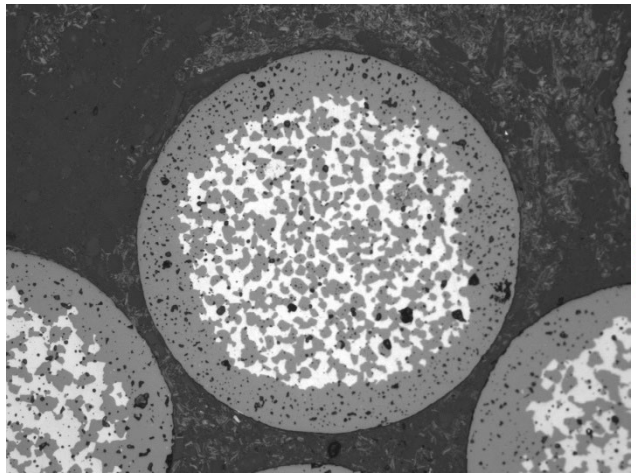
- Prepare a cross-sectional mount of ~100-300 kernels or particles.
- Use an automated imaging script to capture individual optical microscopy images of each kernel.
- Use an automated image processing script to segment phases and calculate phase fractions and density on a per-kernel basis with a polish-down correction.
- Review and confirm image processing results and report statistical distributions for each parameter of interest.

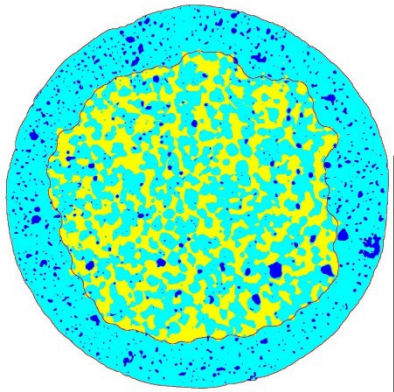


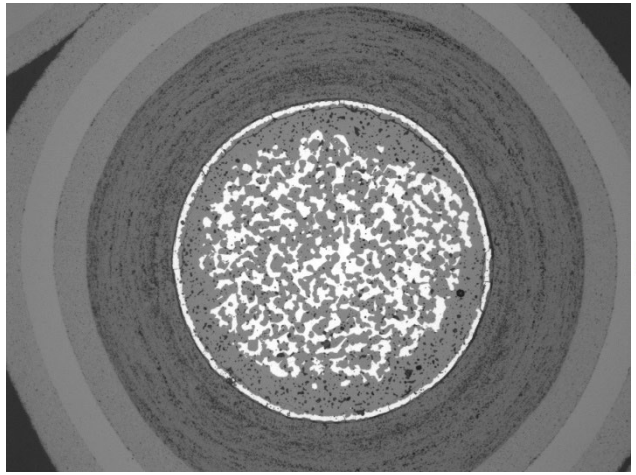
There's always a catch

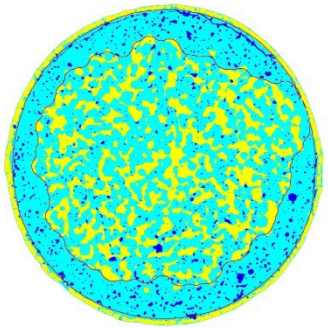
- There are several sources of uncertainty that can affect the results from optical measurement of UCO kernel phases.
  - Heterogenous phase distribution → Use high sampling
  - Features below the resolvable limit → Operator check
  - Impact of UC stoichiometry on density → Minor, but can use XRD
  - Impact of UC stoichiometry on atomic ratios → Shift to phase fractions
- When comparing mean results to bulk chemistry analysis, optical microscopy of UCO kernels holds up well.

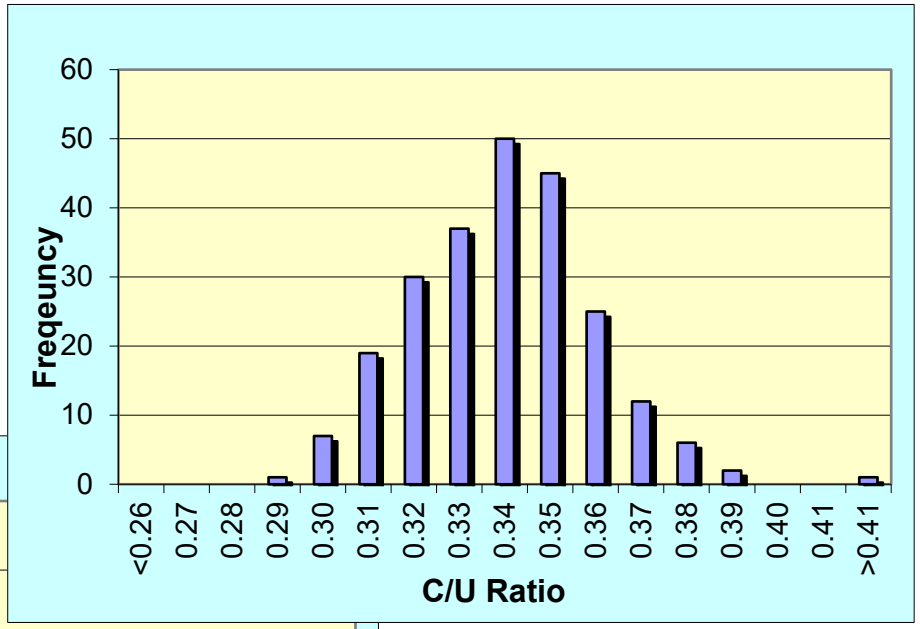
AGR UCO Results

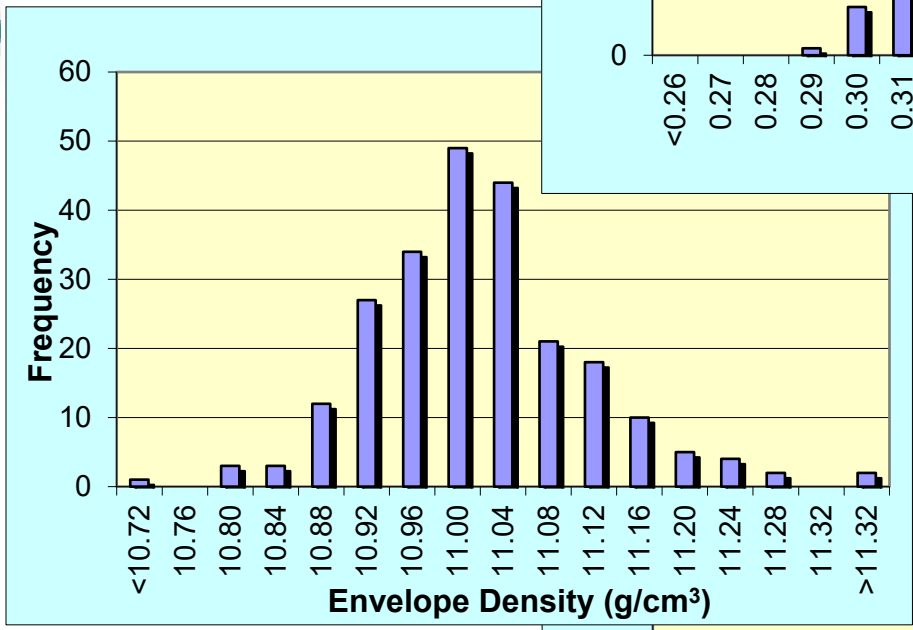


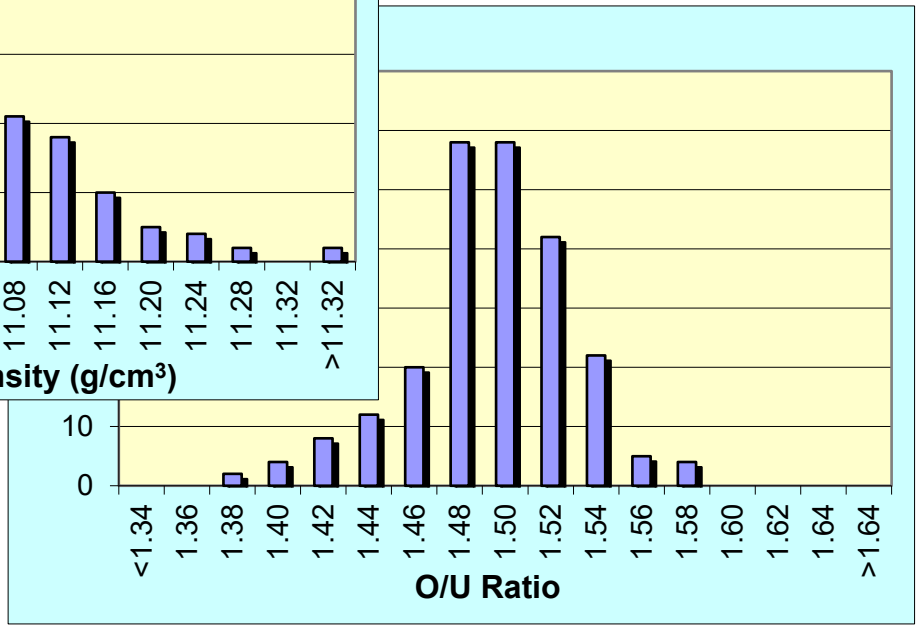












AGR UCO Results

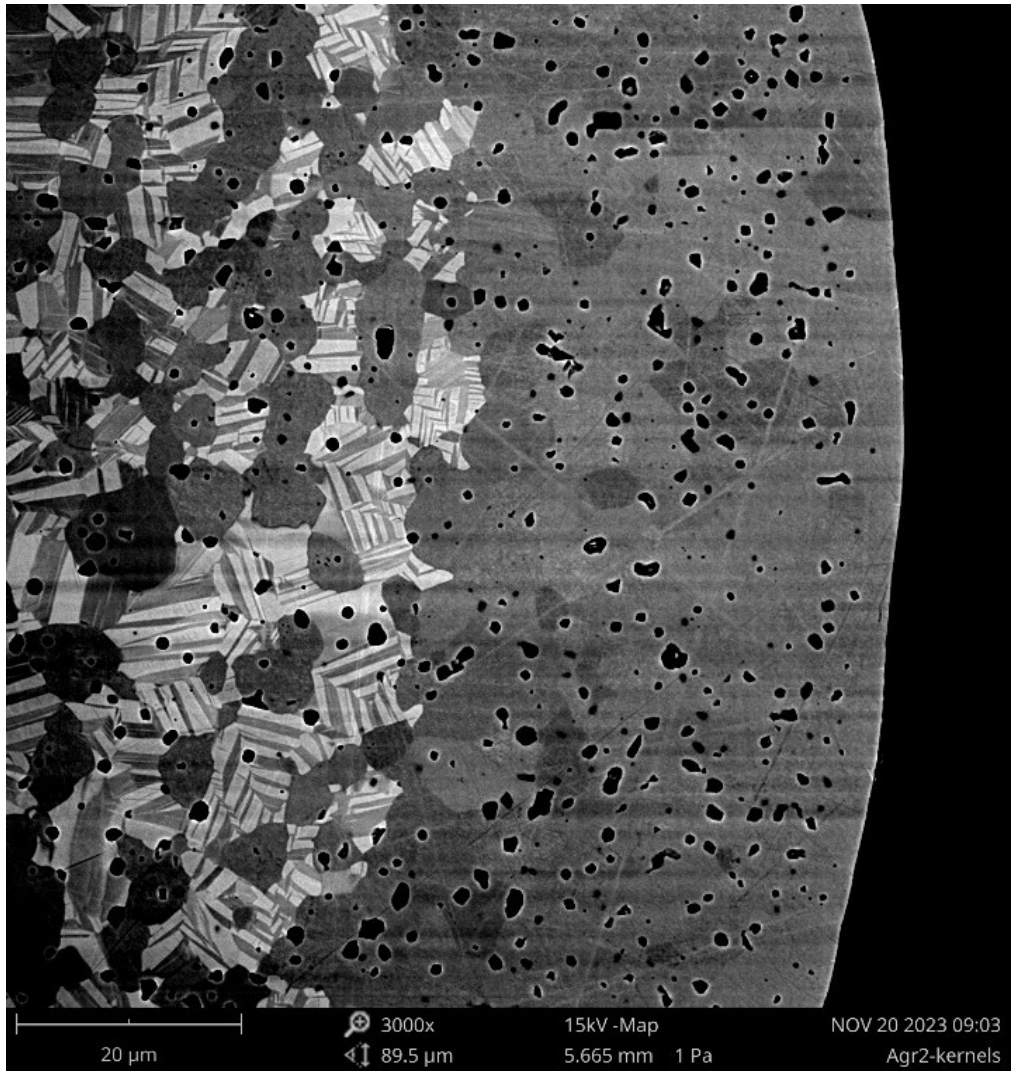
- Samples of kernels and particles from all four AGR irradiation campaigns were analyzed using optical image analysis.
  - No archive of AGR-5/6/7 bare kernels was available at ORNL.
  - XRD was performed for scientific interest and as a confirmatory measurement and is not intended to be applied in this manner for QC.
- Some variation in results was observed between methods.
- The effect of conversion of monocarbide to dicarbide during coating was observed in C/U.
- The loss of friable carbide skin during preparation of particle samples depressed the atom % U in carbide.

Material source	Data source	C/U	O/U	Density (g/cm³)	Atom % U in carbide
AGR-1 kernels	Bulk Analysis	0.325 [0.278–0.385]	1.361 [1.350–1.411]	10.9 (ORNL)	0.319
	XRD	0.345	1.377	—	0.312
	Image analysis	0.298 (0.034)	1.462 (0.061)	10.8 (0.2)	0.269 (0.030)
AGR-1 particles	XRD	0.521	1.360	—	0.320
	Image analysis	0.435 (0.036)	1.466 (0.044)	10.3 (0.3)	0.267 (0.022)
AGR-2 kernels	Bulk Analysis	0.390 [0.39–0.40]	1.430	11.0	0.285
	XRD	0.399	1.417	—	0.292
	Image analysis	0.363 (0.027)	1.447 (0.041)	11.0 (0.1)	0.276 (0.021)
AGR-2 particles	XRD	0.383	1.529	—	0.236
	Image analysis	0.413 (0.022)	1.492 (0.027)	10.7 (0.1)	0.254 (0.013)
AGR-3/4 kernels	Bulk Analysis	0.361	1.430	11.1 (ORNL)	0.285
	XRD	0.388	1.393	—	0.303
	Image analysis	0.335 (0.022)	1.476 (0.034)	11.0 (0.1)	0.262 (0.016)
AGR-3/4 particles	XRD	0.454	1.442	—	0.279
	Image analysis	0.401 (0.026)	1.507 (0.032)	10.8 (0.1)	0.246 (0.016)
AGR-5/6/7 kernels	Bulk Analysis	0.370 [0.33–0.41]	1.441 [1.36–1.48]	11.0	0.280
AGR-5/6/7 particles	XRD	0.511	1.372	—	0.314
	Image analysis	0.416 (0.045)	1.489 (0.055)	10.9 (0.1)	0.255 (0.028)

Note: The associated standard deviation for the measured samples is included in round parentheses for the ORNL image analysis data and represents the kernel-to-kernel variation in the measured composite.
Note: The spread in the BWXT measured average atomic ratios for each kernel batch in the composite is included in square brackets, where available, and represents the extent of batch-to-batch variability in the mean values.

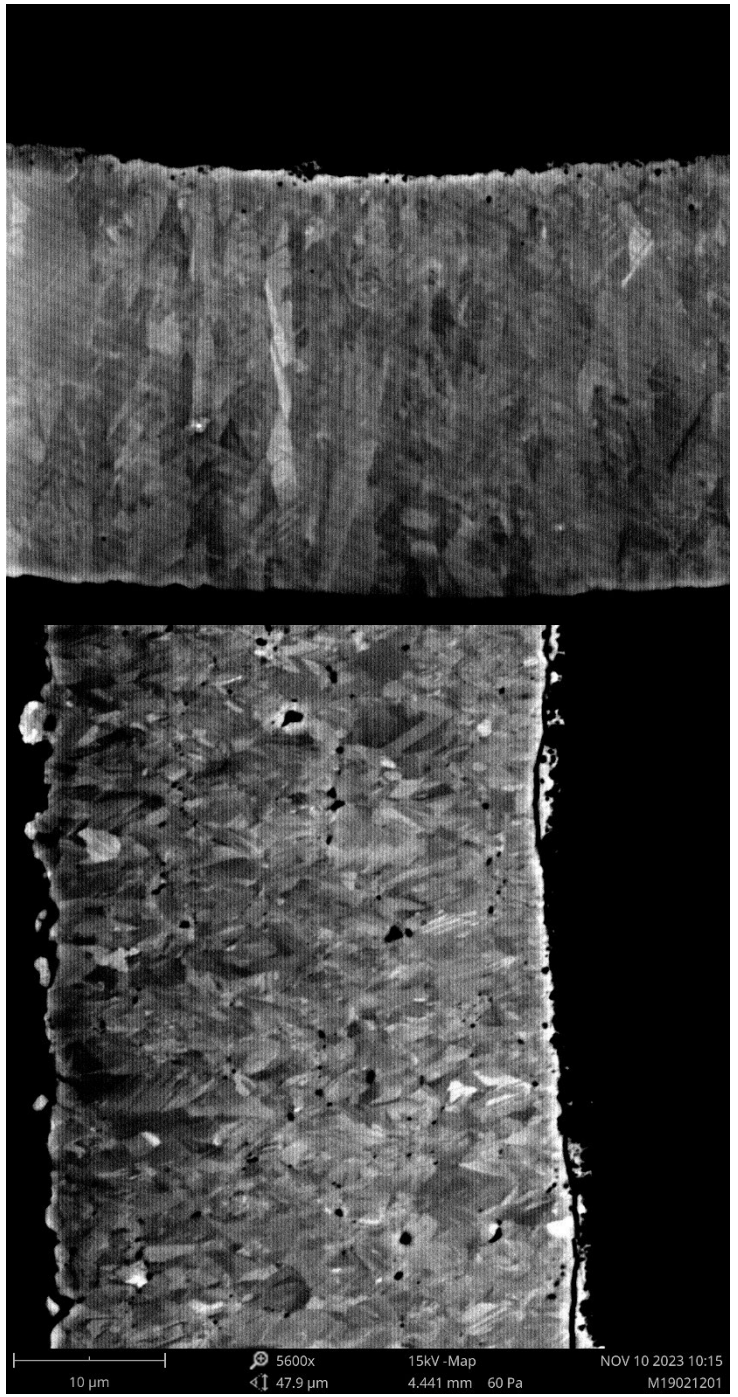
Recommendations for UCO phase fraction measurement

- Optical microscopy analysis of bare UCO kernels is recommended as an industrially appropriate QC method which represents a substantial improvement on existing kernel chemistry measurements.
- Follow-on work may extend this method to image analysis of BSE images acquired using a benchtop system to allow direct differentiation between carbide phases.



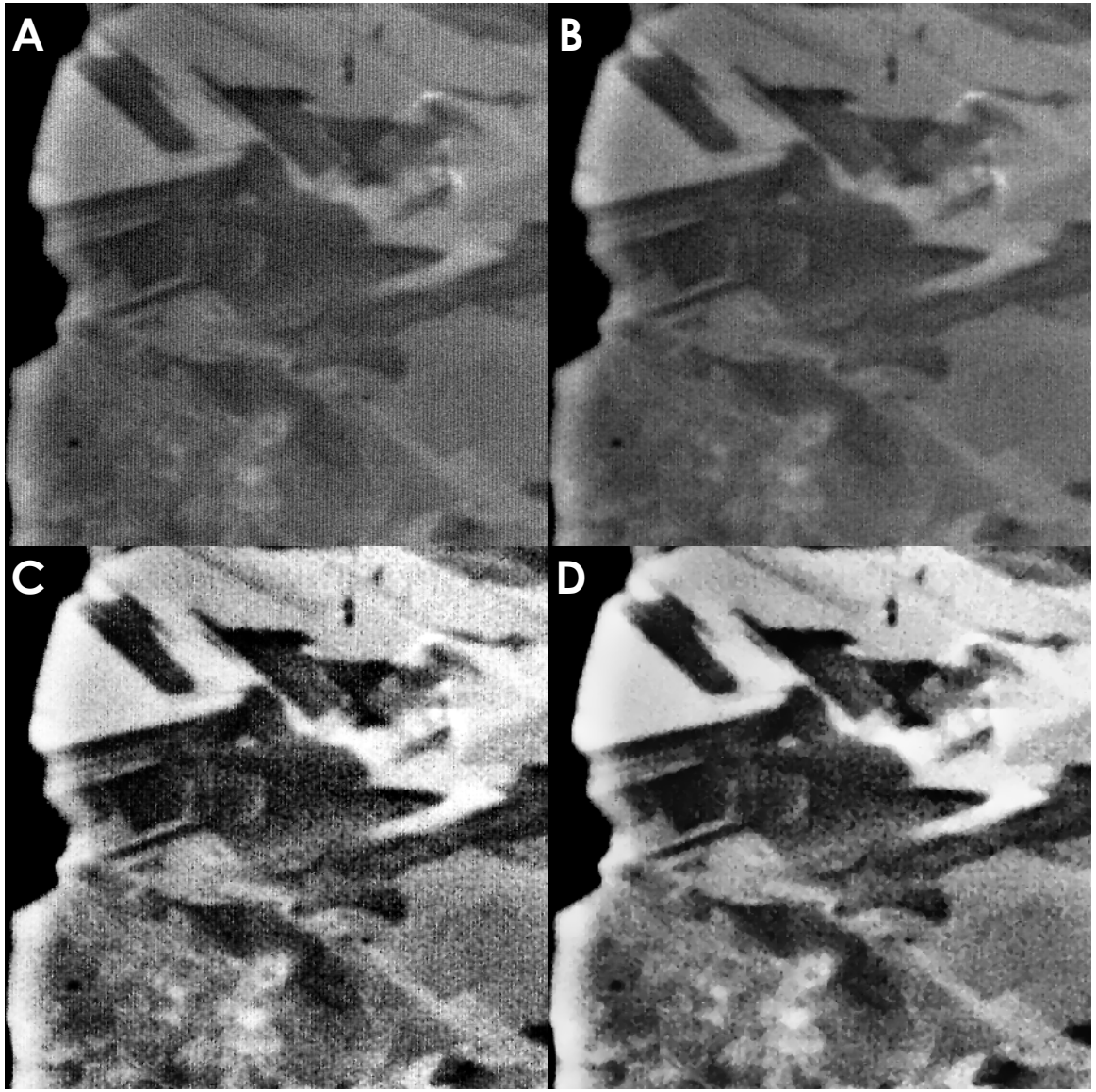
SiC Layer Grain Size

- Existing specification on SiC grain size is based on comparison of BSE images to visual standards showing “good” SiC (bottom) and “columnar” SiC (top).
  - No quantiatative measurement of grain size
  - No lower limit on grain size
- ASTM methods for average grain size determination using automated image analysis have been successfully applied to TRISO SiC layers.



Method: Image filtering

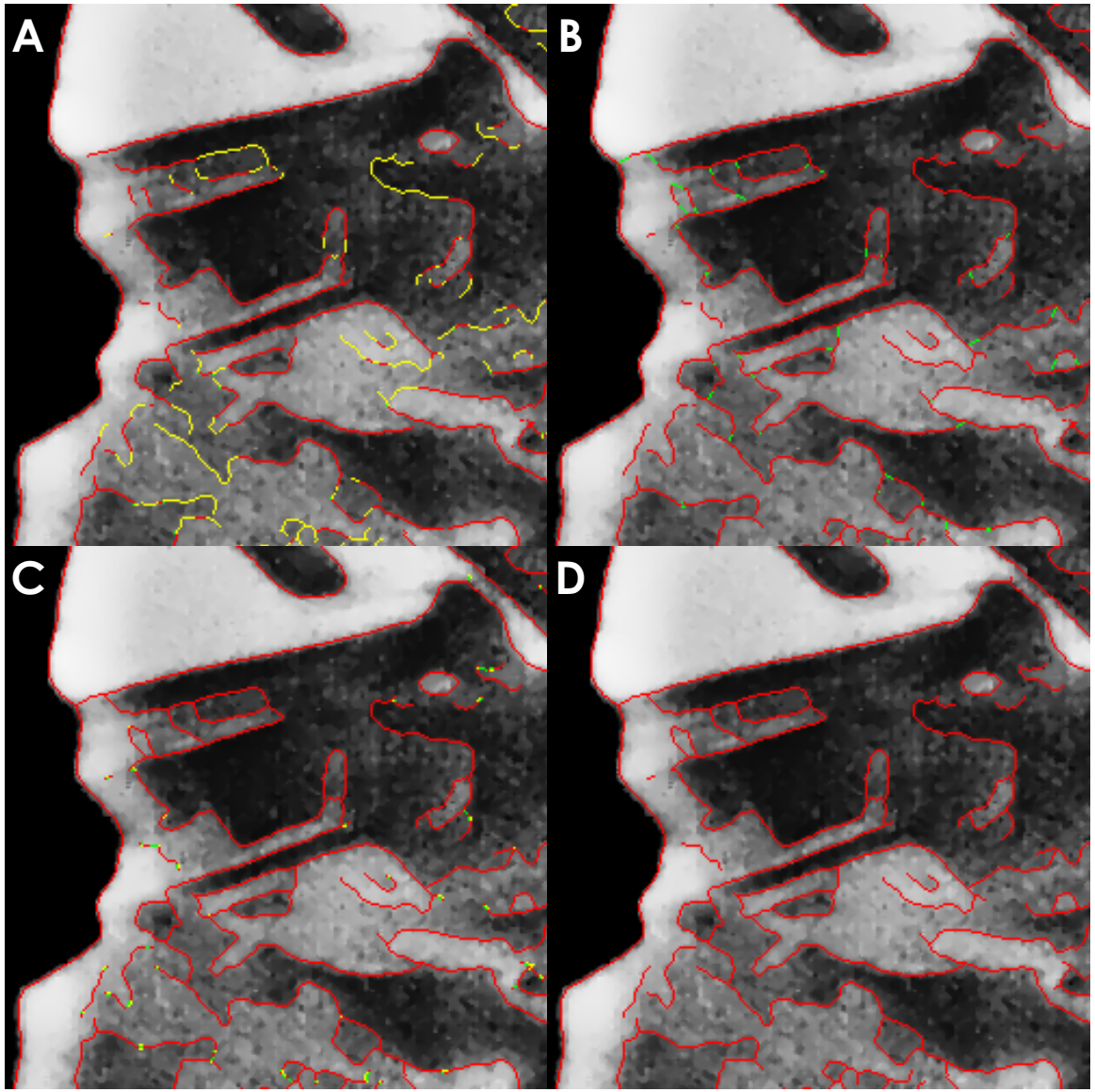
- Several digital filtering steps were applied to optimize grain segmentation.
  - Wiener filter to reduce noise while preserving true edges
  - Contrast-limited adaptive histogram equalization to maximize contrast and normalize image brightness
  - Local-Laplacian filtering to maintain edge contrast while smoothing interior regions
  - Median filtering to smooth final image



Subsection of original image (A), noise-reduced after Wiener filter (B), optimized contrast from adaptive histogram equalization (C), and non-edges smoothed by Local Laplacian filter.

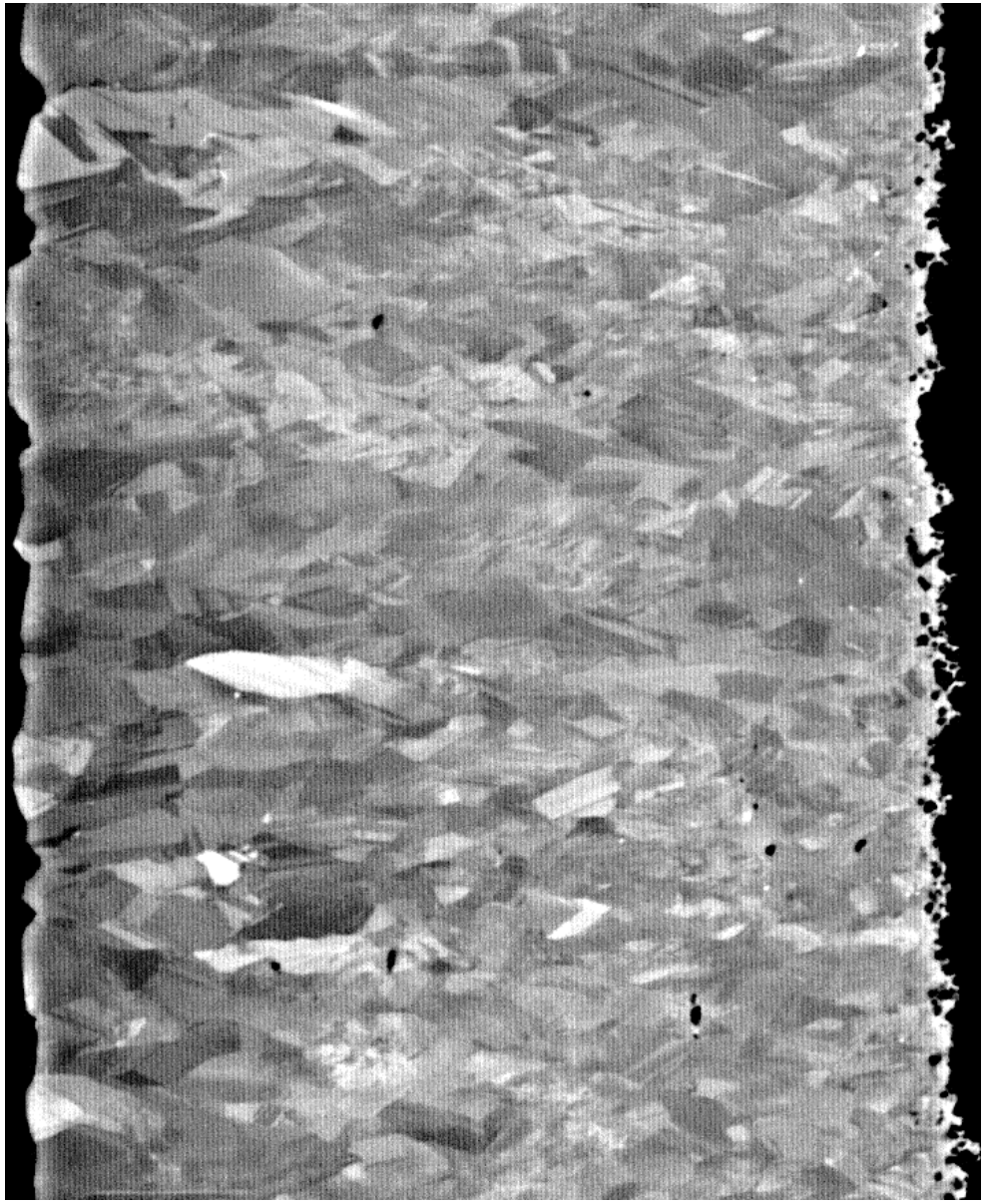
Method: Edge detection

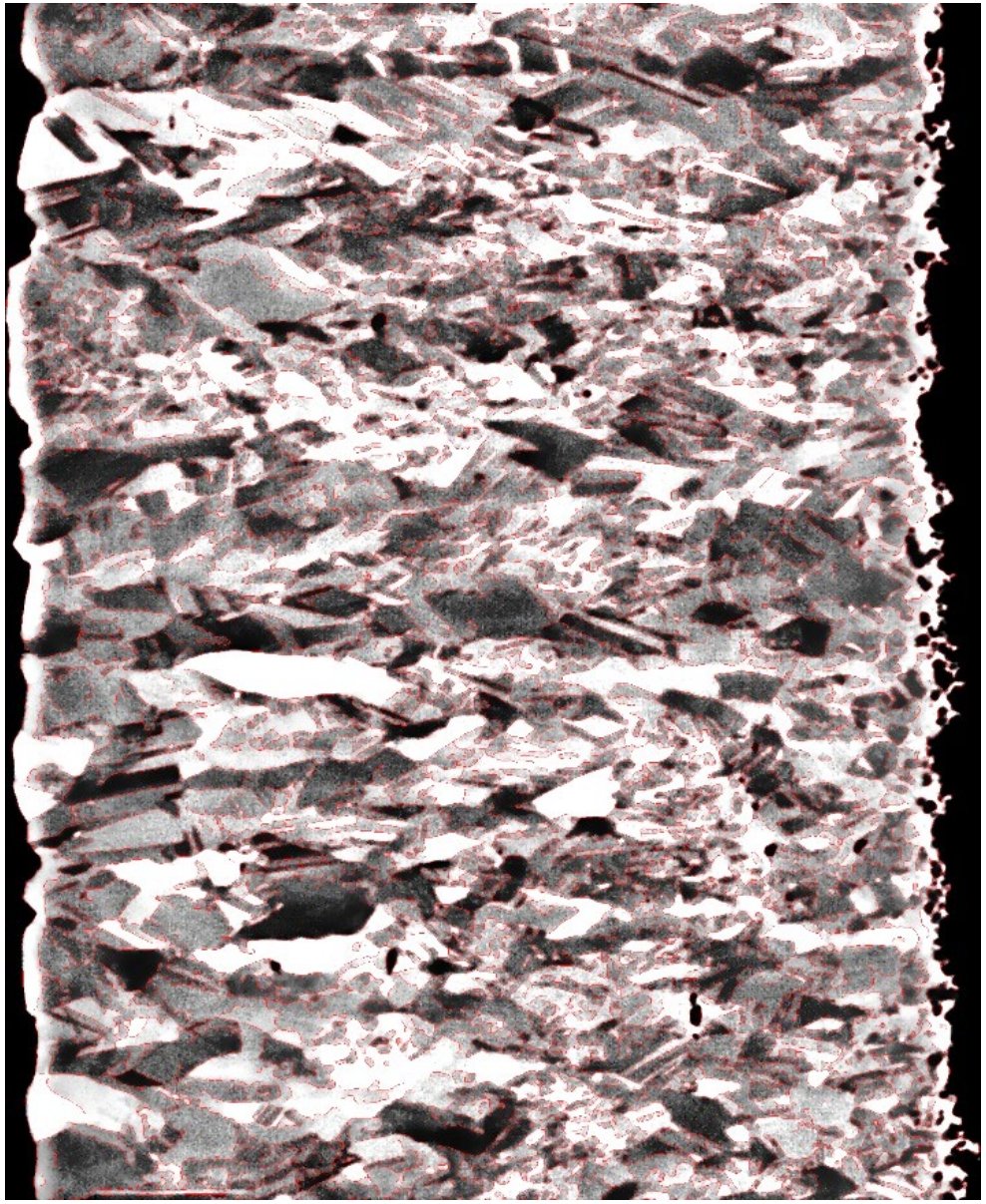
- Initial edge identification by Canny edge detection.
  - Dual-threshold captures strong and connected weak edges
- Momentum linking extends terminal edges.
- Gap-fill linking makes final connections and simplifies over-connected regions.



Strong (red) and weak (yellow) edges found by Canny edge detection (A), edges added by momentum linking (green) (B), edges added (green) or trimmed (yellow) by gap-fill linking (C), and final image (D)

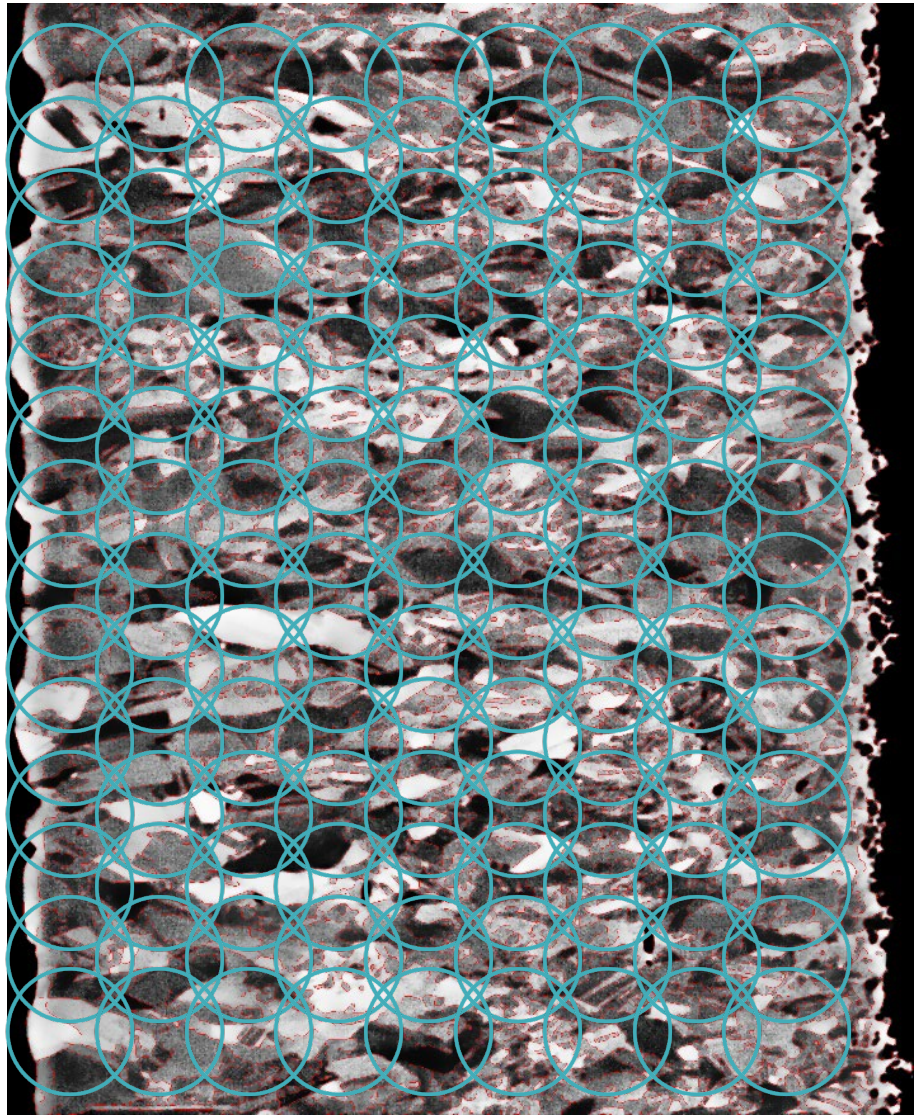
SiC Grain Size Quantification from BSE Images





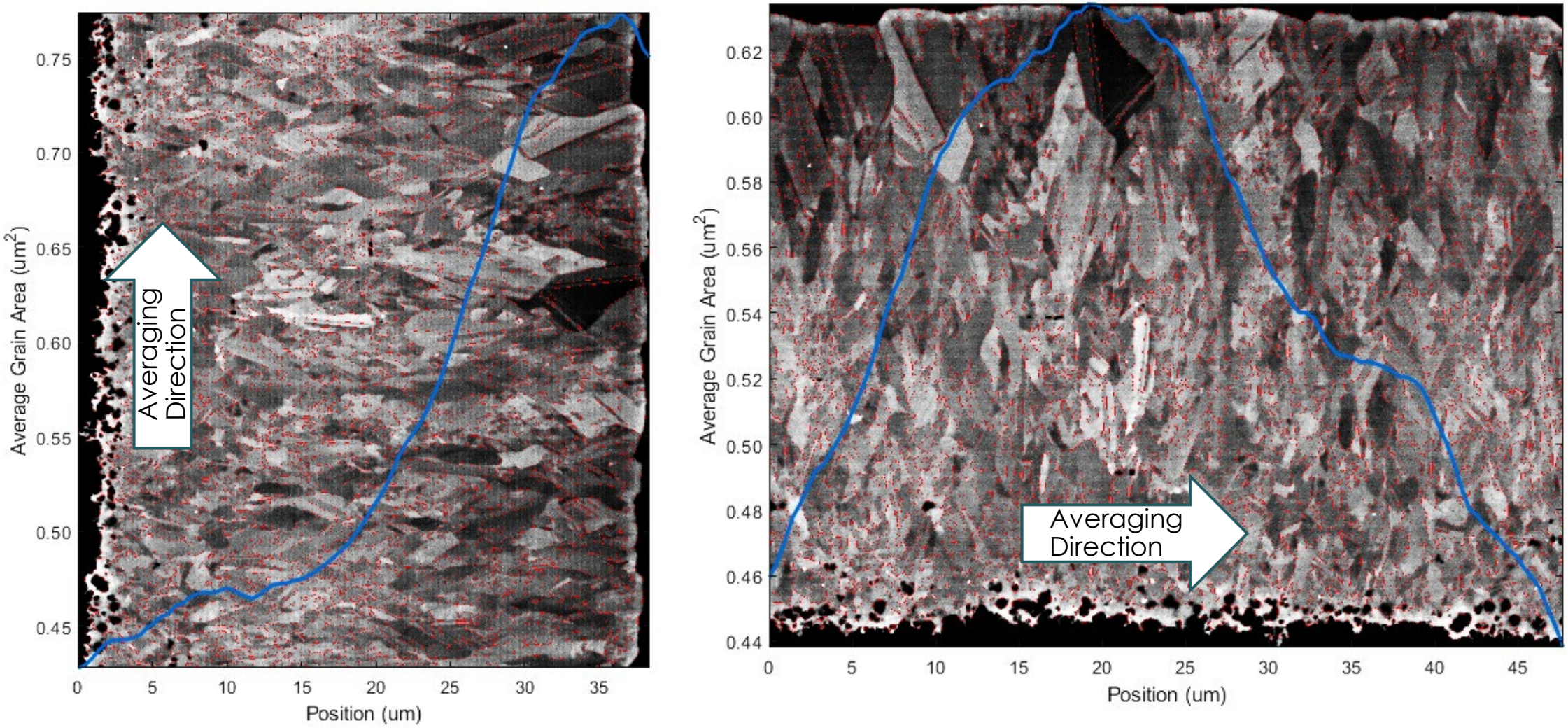
Method: Determination of mean grain area

- Method based on ASTM-E112.
  - Create a circle of area A_i
  - Calculate L_i, the length of all grain boundaries within that circle
  - Calculate grain boundaries per unit area: $P_i = {}^{L_i}/_{A_i}$
  - Calculate average grain area within the circle: $\bar{A}_i = 10^{6.643856 \times Log(P_i) - 0.333}$
- Circular measurement areas reduce bias which line intercept methods encounter for oriented grains.
- Sets of circular intersect results may be averaged to measure grain area trends.



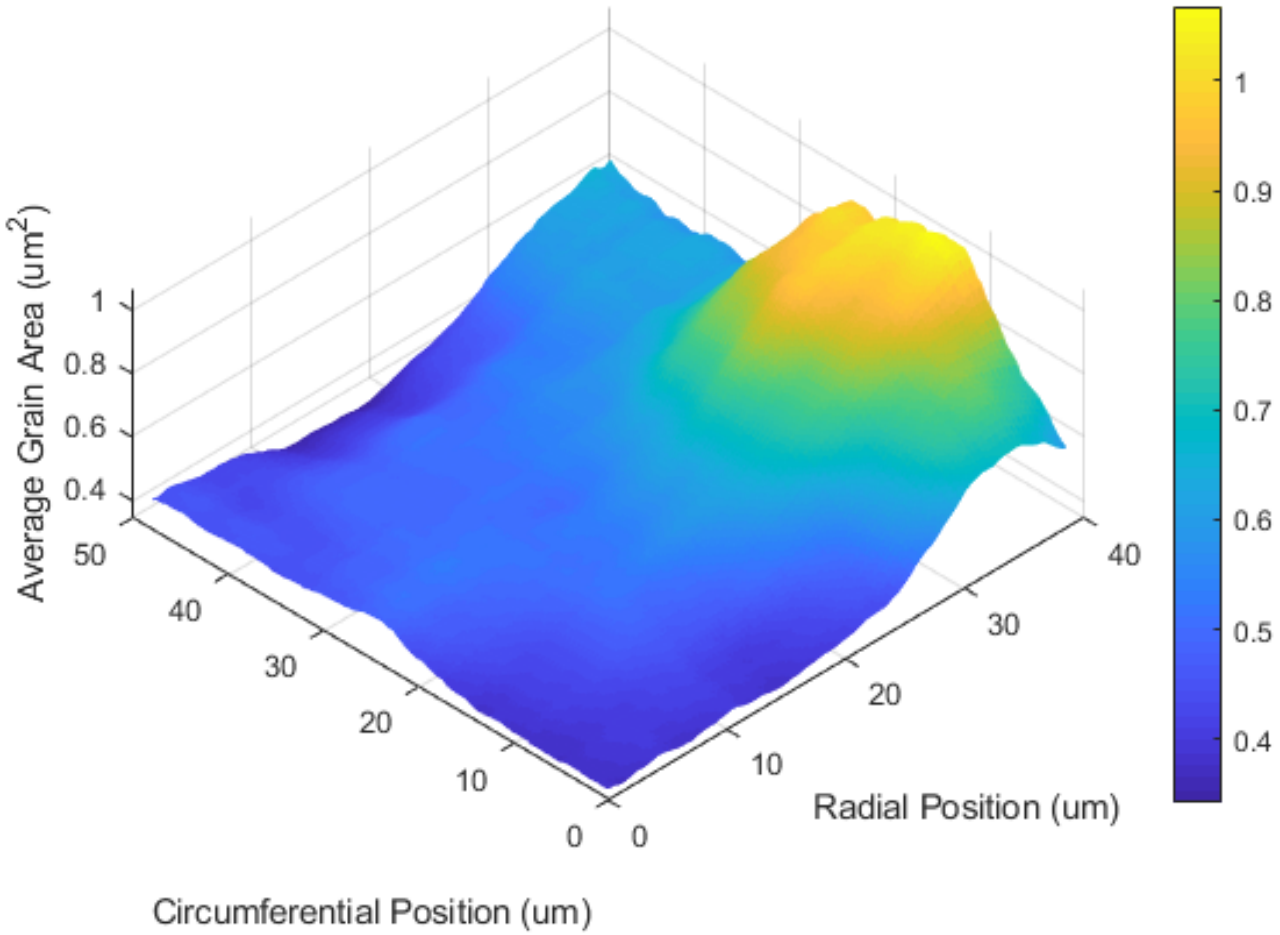
Circular intercept traces overlaying segmented BSE image.

Method: Example results



Local variation in average grain size with averaging in the circumferential direction (left) and with averaging in the radial direction (right).

Method: Example results

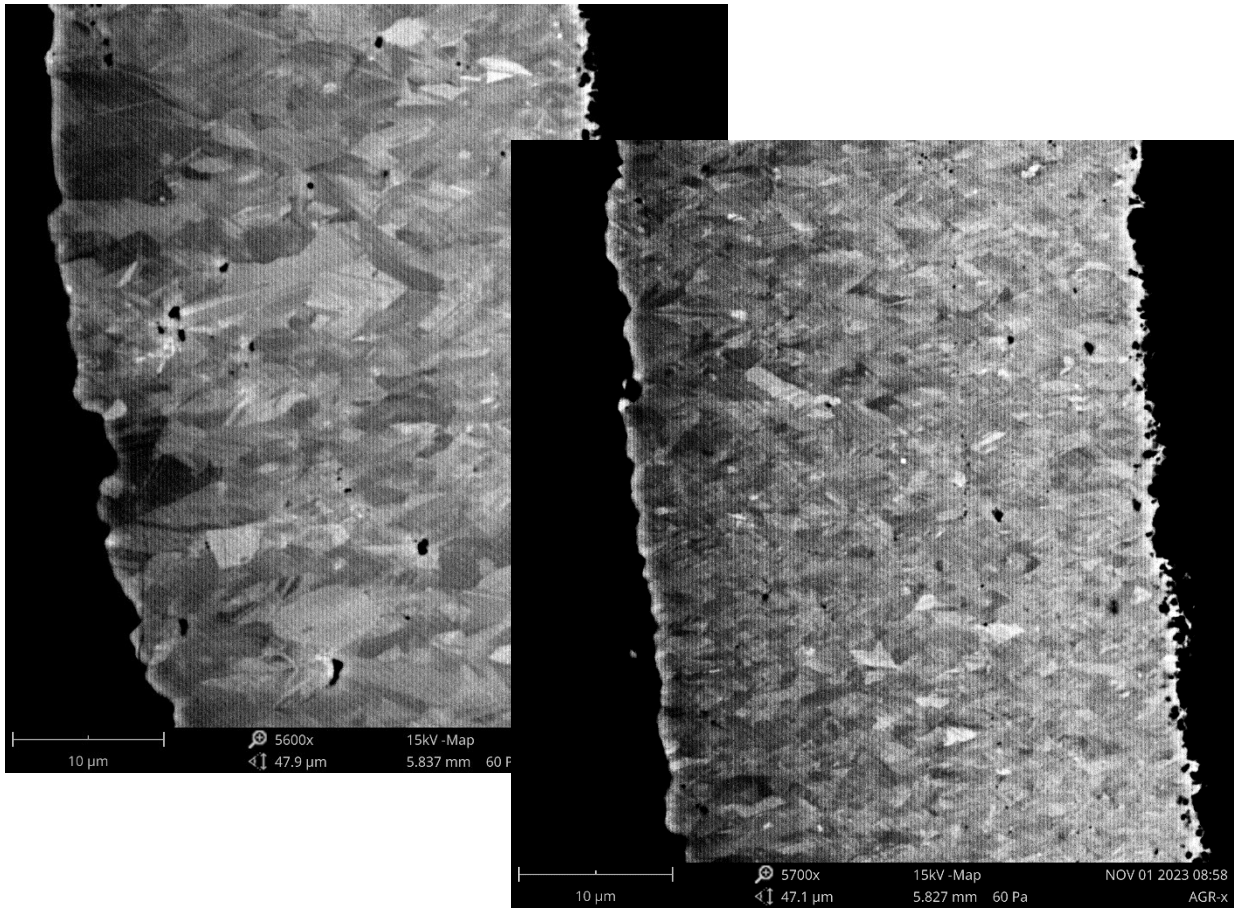


Local variation in average grain size across a SiC layer.

AGR SiC microstructure results

- Initial results for archived material from AGR irradiation campaigns are complete.
  - Note: AGR-3/4 was strongly bimodal, indicating batch-to-batch variation within the composite.
  - Note: AGR-5/6/7 SiC was finer that the reported value as the magnification of the benchtop SEM was not sufficient to resolve all grains.
- Ongoing work using a higher quality SEM and greater particle sampling is being performed now.

Material	Average grain area (μm²)		
	Mean	Standard Deviation	Standard Error
German Reference	0.415	0.056	0.025
AGR-1 Baseline (35T)	0.565	0.042	0.011
AGR-1 Variant 3 (24T)	0.336	0.030	0.008
AGR-2	0.431	0.007	0.003
AGR-3/4	0.581	0.173	0.077
AGR-5/6/7	0.313	0.030	0.013



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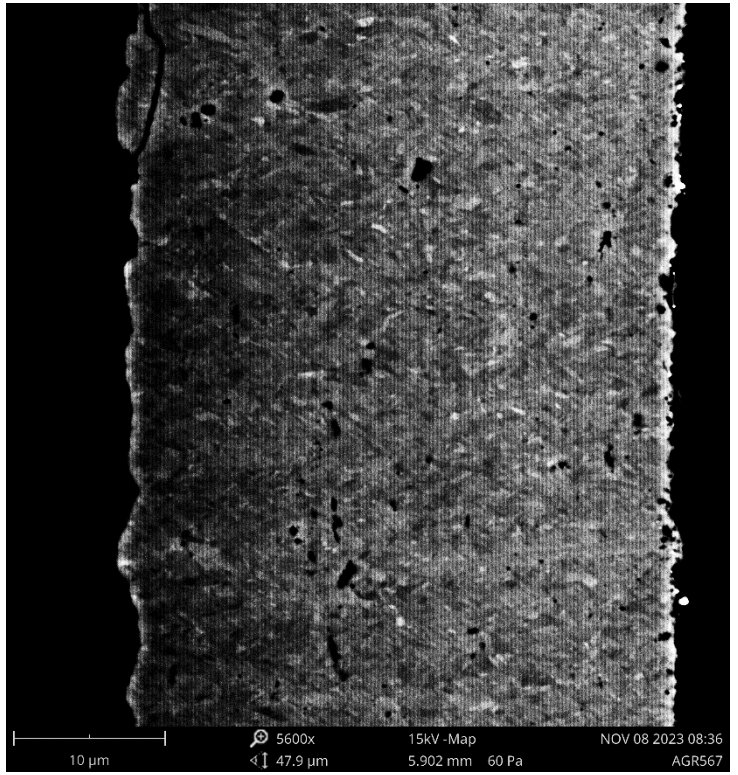


Recommendations for SiC microstructure measurement

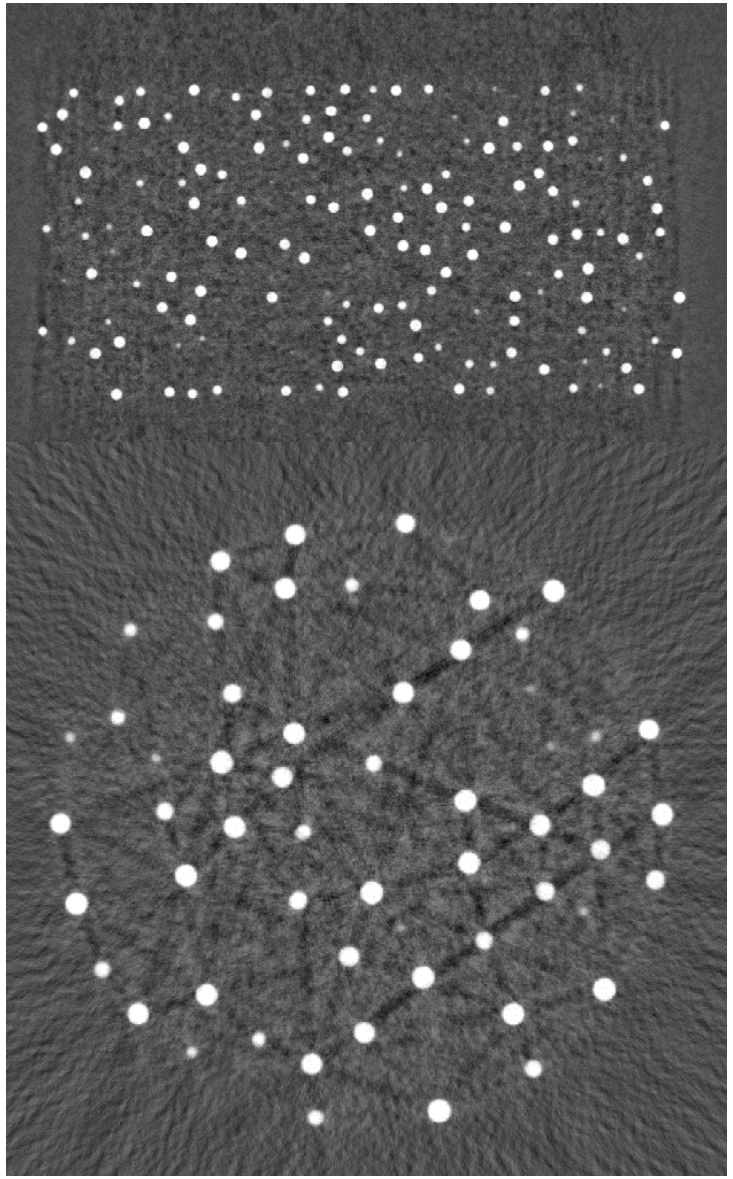
- BSE image analysis for SiC microstructure measurement represents a substantial improvement over the existing qualitative approach.
- Assessment of appropriate upper and potentially lower limits on SiC grain size need to be established, likely based on the range spanned by the AGR irradiations.
- The existing automated grain boundary segmentation method fails to identify twinned grain boundaries common in large SiC grains. Additional development using machine learning should be pursued to remedy this.

Fuel Form Matrix Density

- Matrix density was not specified for the AGR-1 irradiation, but is of interest for its effect on fuel form mechanical and thermal properties.
- Matrix density was added to the AGR-2 fuel specification.
  - The measurement method used overcoated charge weight and mean overcoated particle mass to calculate the number of particles in each compact, then used compact volume, compact mass, and mean particle mass to calculate matrix density.
  - Methanol evaporation between measurement of overcoated charge weight and mean overcoated particle mass resulted in low particle loading relative to the target.

Independent particle counting by rapid XCT

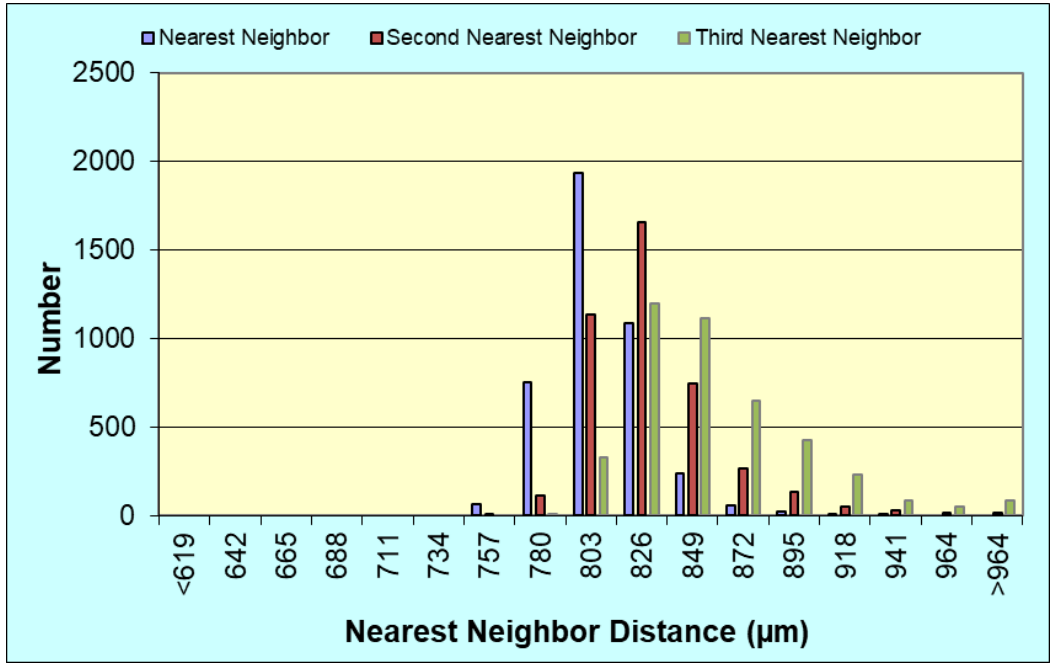
- The use of x-ray computed tomography (XCT) for counting of particles was tested on AGR-1 and AGR-2 compacts.
- Segmentation of the compact for volume measurement was found to be less accurate than caliper measurement.
- Since kernel identification is relatively easy, conditions were optimized for very rapid imaging (~3.5 m per compact)

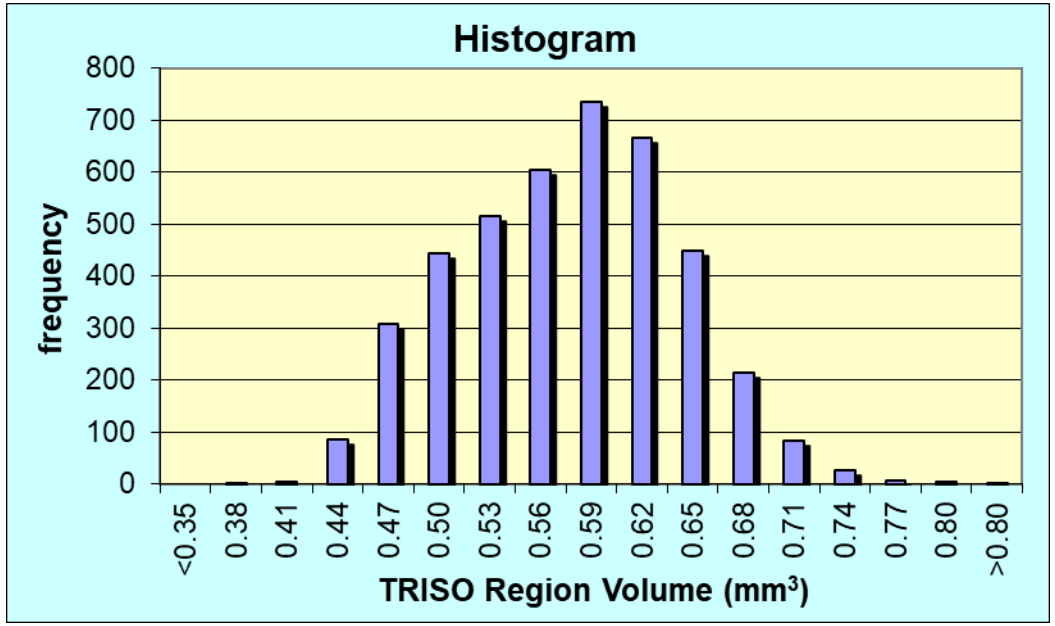


AGR matrix density results

- The impact on calculated matrix density from using the actual number of particles was minor (<0.6%).
- Additional data on nearest neigbor distances and local particle packing were generated for each compact.

Type	Compact ID	Bulk Estimation		XCT Counting	
		# Particles	Matrix Density (g/cm^3)	# Particles	Matrix Density (g/cm^3)
AGR-1	LEU01-46T-Z03	4145	1.290	4155	1.288
AGR-1	LEU01-46T-Z47	4145	1.291	4143	1.291
AGR-1	LEU01-46T-Z68	4145	1.299	4131	1.302
AGR-2 UCO	LEU09-OP2-Z069	3200	1.597	3186	1.600
AGR-2 UCO	LEU09-OP2-Z102	3200	1.590	3167	1.598
AGR-2 UCO	LEU09-OP2-Z139	3200	1.589	3184	1.593
AGR-2 UCO	LEU09-OP2-Z165	3200	1.591	3170	1.599
AGR-2 UO_2	LEU11-OP2-Z045	1566	1.687	1554	1.691
AGR-2 UO_2	LEU11-OP2-Z074	1567	1.678	1537	1.687
AGR-2 UO_2	LEU11-OP2-Z198	1567	1.665	1545	1.672





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AGR-2 UCO	LEU09-OP2-Z102	3200	1.590	3167	1.598
AGR-2 UCO	LEU09-OP2-Z139	3200	1.589	3184	1.593
AGR-2 UCO	LEU09-OP2-Z165	3200	1.591	3170	1.599
AGR-2 UO_2	LEU11-OP2-Z045	1566	1.687	1554	1.691
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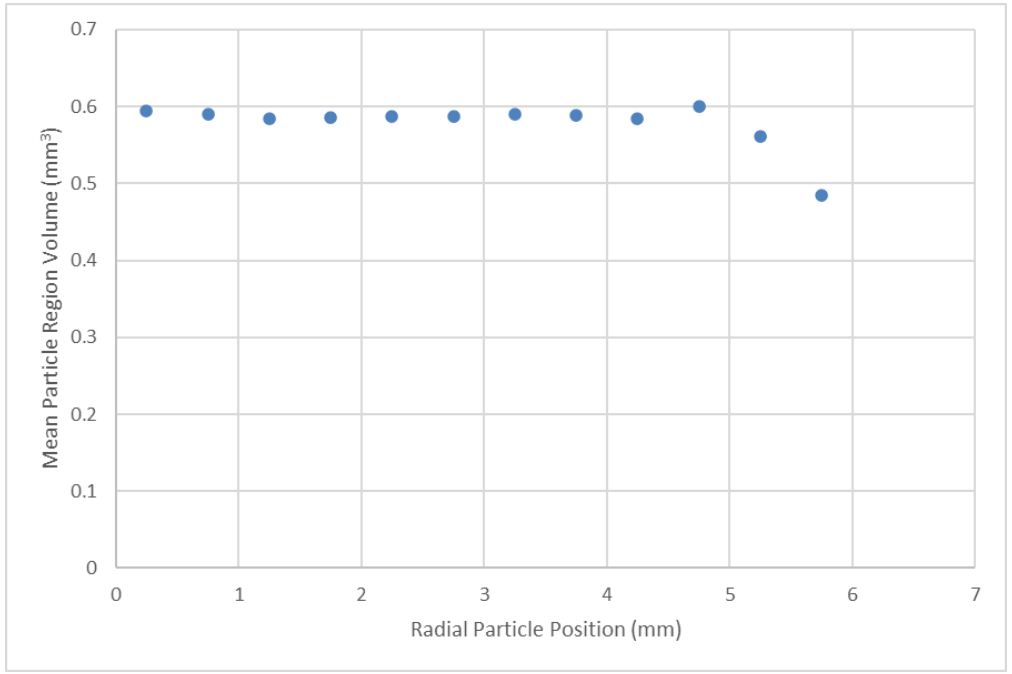
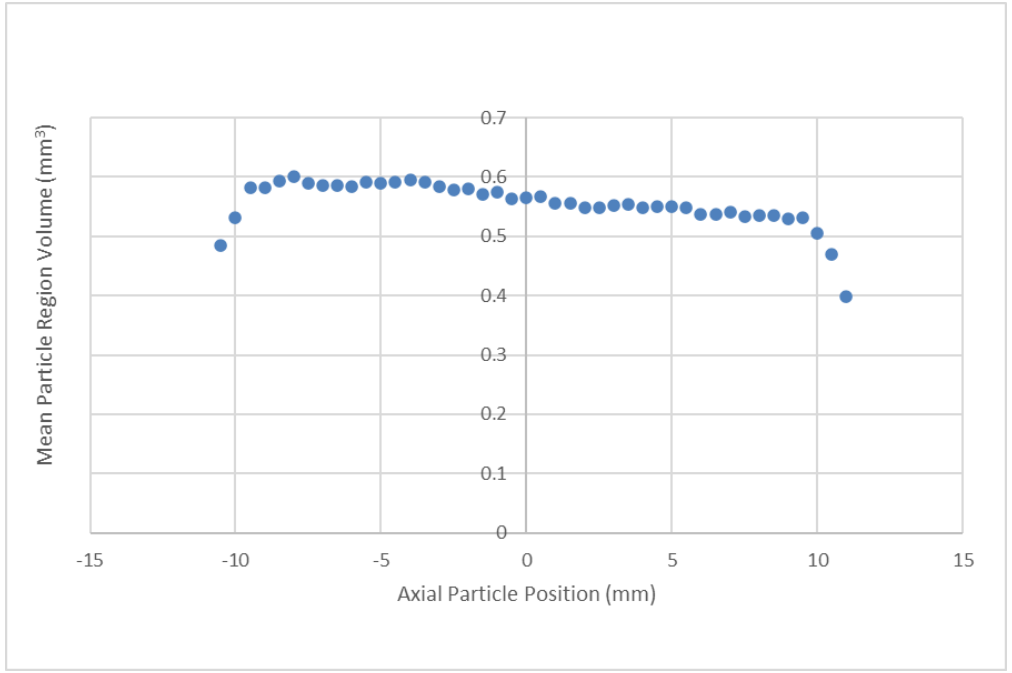




Recommendations for measurement of matrix density

- Counting of particles by XCT may be performed rapidly enough to be applied to individual compacts.
  - Slightly improved accuracy of matrix density calculation.
  - Is independent of fabrication parameters (e.g. overcoated charge weight).
- The Versa 620 system used in this work is overkill for this application. Cheaper benchtop XCT systems are more appropriate for industrial QC use.
- Additional parameters relating to particle packing may be of use as process feedback.

Layer Density Measurement

- The density of TRISO coating layers is an important parameter, but the current methods for measurement have room for improvement.
  - PyC and SiC layer densities are measured using liquid gradient density columns. Highly accurate, but slow and requires interrupted coating runs or hot sampling for IPyC. Also generates halogenated organic waste.
  - Buffer density is measured using mercury porosimetry. Requires interrupted coating runs or hot sampling, only mean density is measured and generates mixed HALEU/mercury waste.
- Buffer density measurement using x-ray radiography was evaluated as a possible new method.

Polychromatic x-ray attenuation

- X-ray attenuation is governed by material and energy dependent coefficients.
  - Transmision I of incident x-ray beam I_0 depends on material thickness t and attenuation coefficient μ.
  - μ is dependent on material composition and density as well as x-ray energy.

$$I = I_0 \times e^{-\mu t}$$

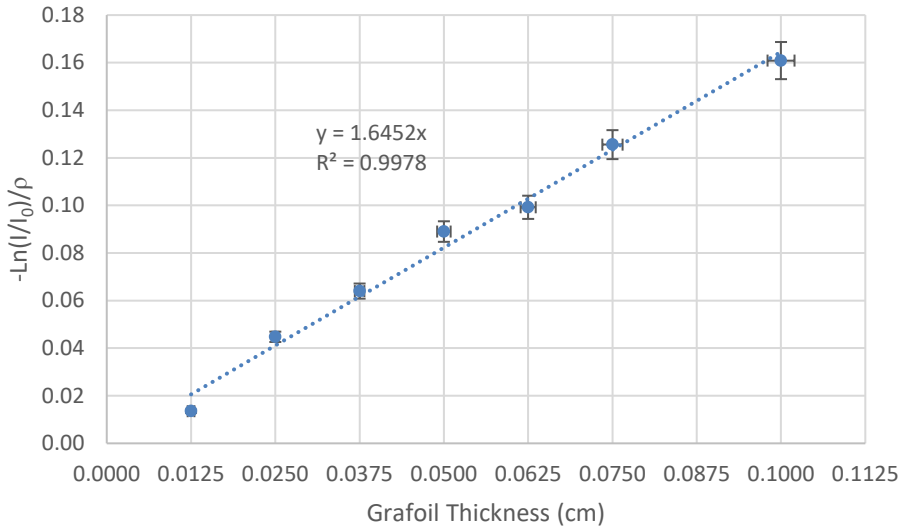
- The most straightforward approach to determining density using a polychromatic x-ray source is to calibrate using samples of the same composition and known density.

$$-ln\left({}^{I}/_{I_0}\right)\Big/_{\rho_s} = {}^{\mu}/_{\rho} \times t_s$$

Polychromatic x-ray attenuation calibration for carbon

- Two thicknesses of grafoil were used with Kapton stickers to generate a series of calibration samples.
  - Grafoil sheet thicknesses were confirmed using calipers
  - The density of each sheet was measured.
  - Kapton stickers were used to mimic the conditions used for particle samples.
- X-ray source conditions were 40kV and 3W with a 60s imaging time.
- The slope of the least-squares fit was taken as the polychromatic

$$-ln\left(I/I_0\right)\Big/_{\rho_s} = \mu/\rho \times t_s$$

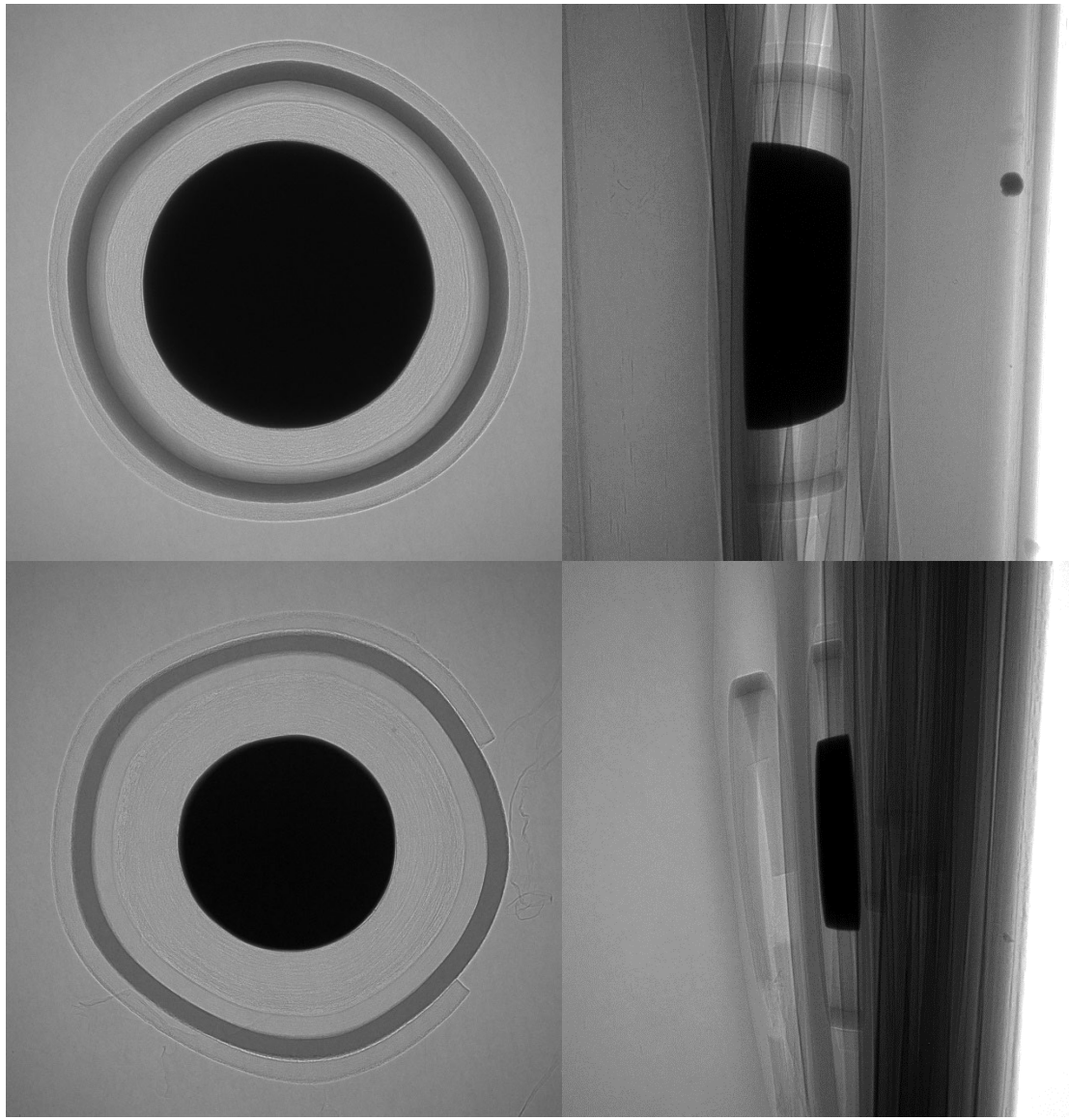


Initial scoping results

- Spherical sections of five surrogate TRISO particles were produced and radiographed.
- Calculated densities were highly variable and generally too high.

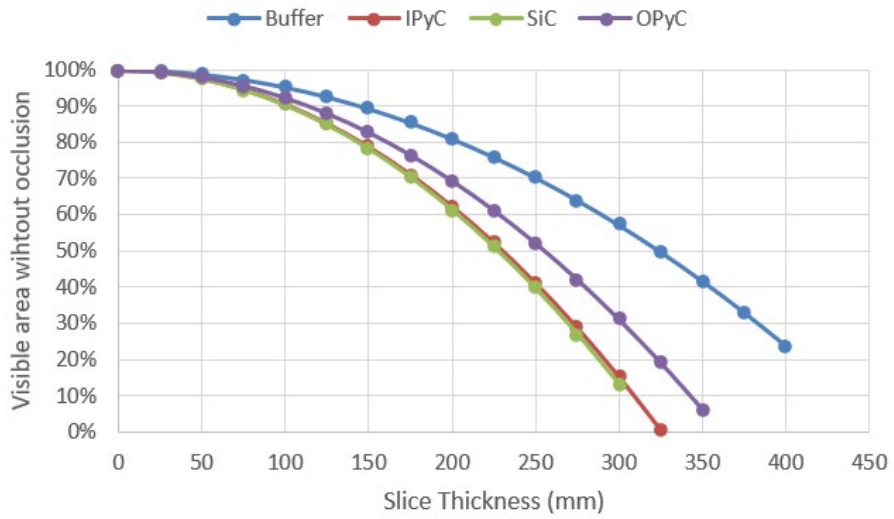
$$\rho_s = \frac{-ln\left(I/I_0\right)}{\mu/\rho \times t}$$

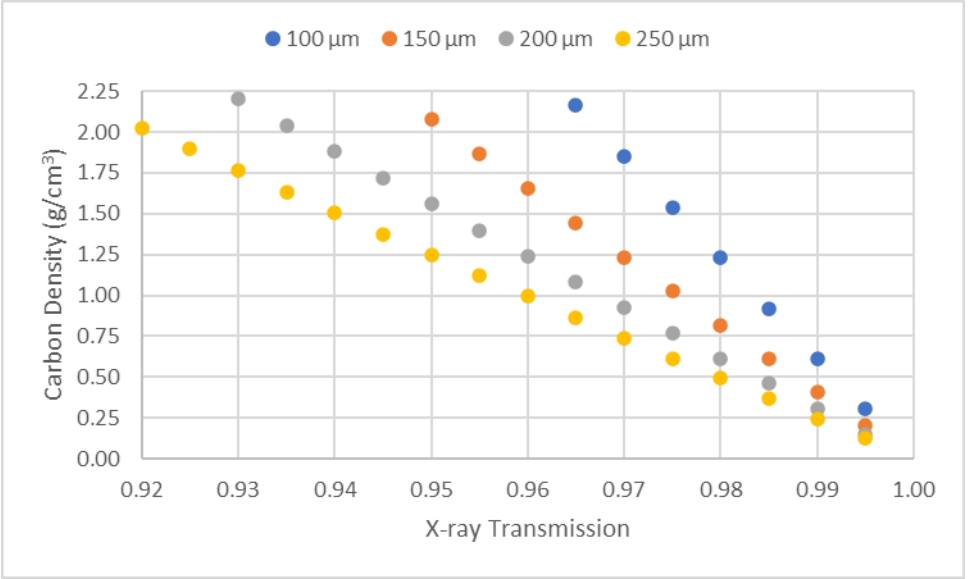
Material	Particle	Thickness (μm)	Transmission	Density (g/cm^3)
ZrX05-33T	P1	178	0.9595	1.411
	P2	179	0.9347	2.295
	P3	176	0.9411	2.093
	P4	179	0.9357	2.259
	P5	177	0.9483	1.825
ZrX05-31T	P1	80	0.9699	2.322
	P2	80	0.9842	1.210
	P3	80	0.9737	2.025
	P4	80	0.9903	0.741
	P5	80	0.9834	1.272



Opportunities for improvement

- Better sample preparation method needed
  - Current approach impregnates buffer with some residual CrystalBond
  - Cutting and grinding a disk from a multiparticle epoxy mount would remove this issue and enable statistical sampling.
- Need to maximize sensitivity of transmission to sample density
  - Alternate x-ray source can provide higher flux of low energy x-rays to interact with buffer.
  - Removal of Kapton tape encapsulation will increase x-rays on target.
  - Optimize sample thickness.
- Better calibration may improve accuracy. Ideally want standards of known density and comparable attenuation to samples. Also may want SiC calibration standards.





Summary and future work

- Optical microscopy of UCO kernels for measurement of composition and density is ready for industrial scale QC implementation.
  - Further development of a BSE image analysis analogue can add separation of UC and UC_2 phases.
- BSE microscopy of SiC layers for grain size measurement is near-ready for industrial scale QC implementation.
  - ML approaches to twin identification should be pursued.
  - Bases for upper and possibly lower limits on SiC grain size should be considered.
  - Assessment of segmentation of very fine-grained SiC using laboratory level SEM is ongoing.
- Rapid XCT for particle counting in support of matrix density calculation is ready for industrial scale QC implementation.
  - Industry will need to decide if improvement in density calculation along with feedback on particle packing and distribution is worth additional equipment and characterization step.
- Scoping of layer density measurement by x-ray radiography completed on buffer layer.
  - Several challenges were identified in the first approach tested, but there are paths forward.
  - Given the potential impact if successful, further work should be performed.