GAS-COOLED REACTOR
ADVANCED REACTOR TECHNOLOGIES PROGRAM

7-16-2024

Thermophysical Property Characterization of Irradiated TRISO Compacts

---

Ethan Hisle

INL Research Staff | Materials & Fuels Complex | Characterization| Thermophysical Properties Group

DOE ART GCR Review Meeting

Hybrid Meeting at INL

July 16–18, 2024

Research Group & Contributors

Ethan J. Hisle[1], Tsvetoslav R. Pavlov[1], Amey Khanolkar[2], Zilong Hua[2], Narayan Poudel[1], Chuting Tsai[1],Isaac Cutler[1], Matthew Goodson[1], Scott T. Anderson[3],Tanner Mauseth[3], Cameron Howard[3], Mario D. Matos III[3],Riley S. Moeykens[1],Swapnil K. Morankar[4], William C. Chuirazzi[4], Subhashish Meher[5], Lu Cai[6], John D. Stempien[6]

[1]Thermophysical Property Group – Characterization & PIE - INL

[2]Center for Thermal Energy Transport Under Irradiation – Condensed Matter & Materials Physics – INL

[3]Correlative Microscopy Group – Characterization & PIE – INL

[4]Diffraction and Imaging Group – X-Ray and Neutron Sciences - INL

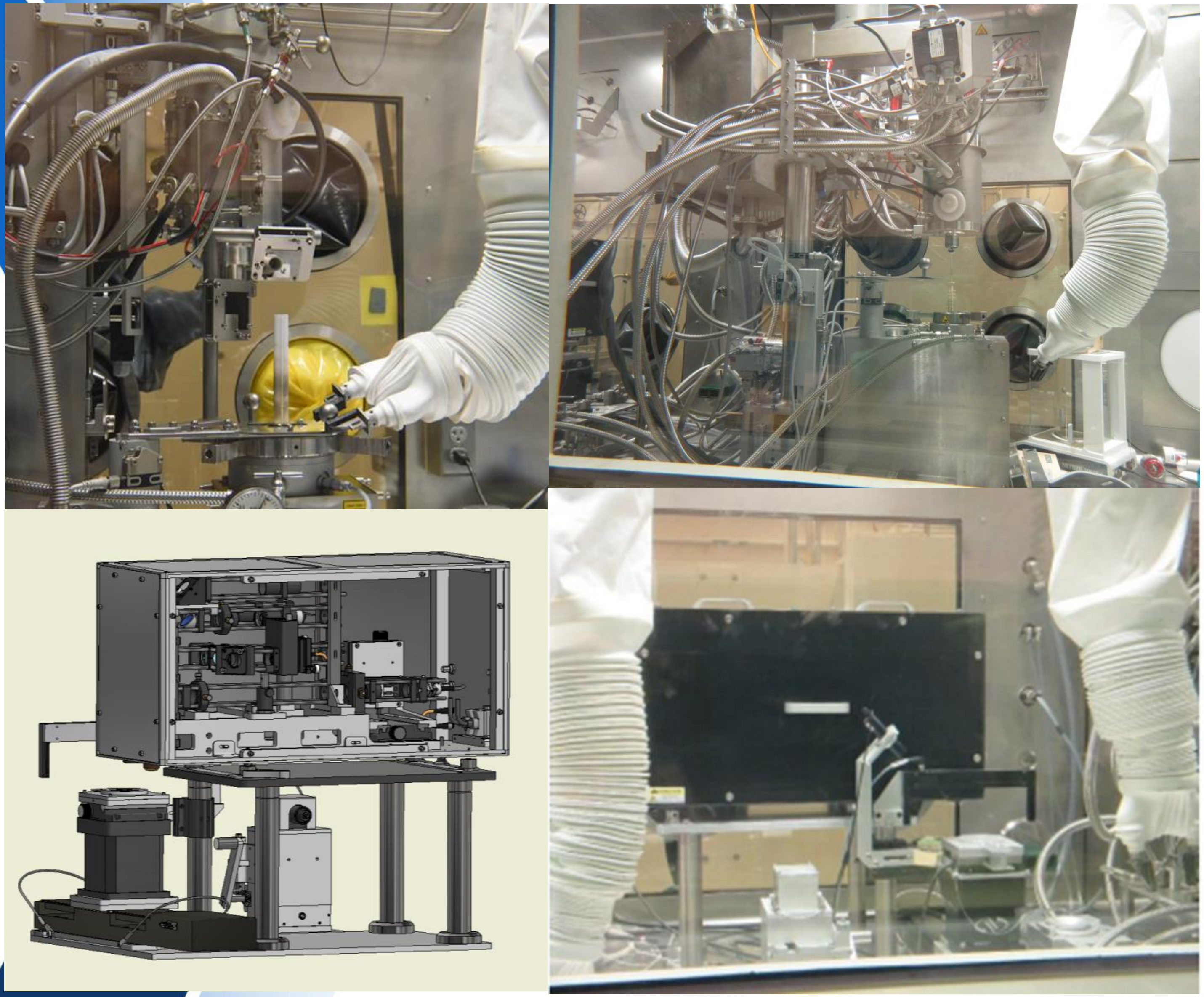
[5]Energy and Environment Directorate - PNNL

[6]Fuel development, Performance & Qualification Group - INL

Special thanks to IMCL operations staff for their support

Overview

- Motivation
- Experiment Background
- TCM Experimental Method
- TCM Results
- Summary and Conclusions
- Future/Ongoing Work



Top Left: LFA at IMCL, Top Right, STA at IMCL
Bottom Left: TCM diagram, Bottom Right, TCM at IMCL

Motivation

- Measurements of thermophysical properties are used in thermomechanical fuel performance code to model the performance of reactors in normal and abnormal conditions
- The temperature across the fuel specimen (governed by thermophysical properties) in a nuclear reactor will affect:
  - mechanical properties
  - Fission product/gas transport and release
  - Fuel compact pressurization (stress states of the fuel, layers, and matrix)
- This work provides first-of-a-kind PIE on mesoscale thermal transport data for TRISO particles
  - A novel thermophysical properties source for fuel performance codes
  - Will contribute to an understanding of the microstructural evolution of TRISO materials thermal transport properties from pre- to post-irradiation

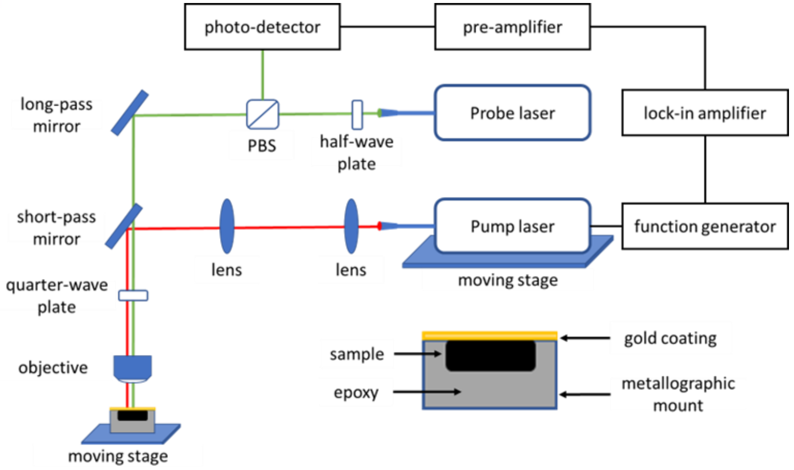
AGR-2 Compacts and Characteristics

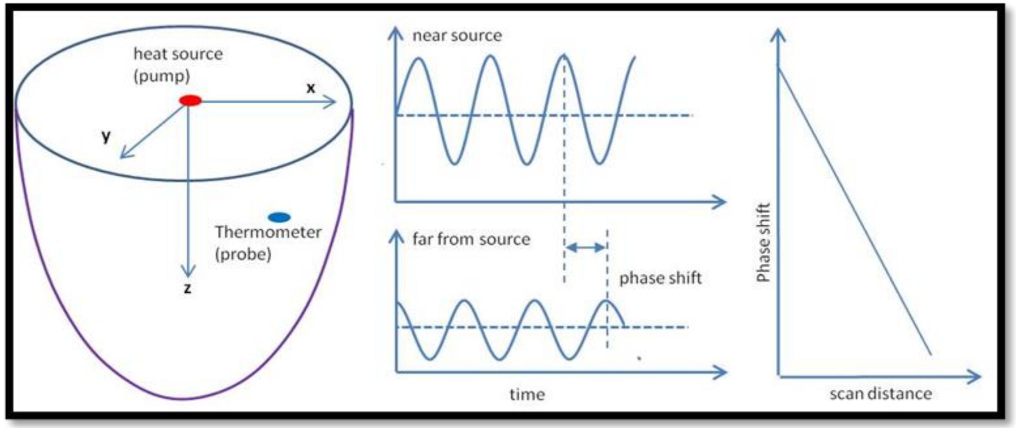
Compact	Sample ID	TAVA Irradiation Temperature (°C)	Burnup (% FIMA)	Fuel Type	Fast Fluence (10^{25} n/m^2 E>0.18 MeV)	Enrichment (weight % U-235)	Mean Particle Diameter (um)
5-1-3	MNT-64X	1078	11.09	UCO	3.03	14	873
2-4-3	MNT-58X	1216	11.52	UCO	3.08	14	873
2-1-3	MNT-67X	1194	10.95	UCO	2.88	14	873
LEU09-OP2-z	LEU09-F52	NA	NA	UCO	0	14.3	873

- Three samples are from compacts were sourced from the AGR-2 experiment [1]
- One sample consisting of 3 as-fabricated TRISO particles in a surrogate matrix as a baseline [2]

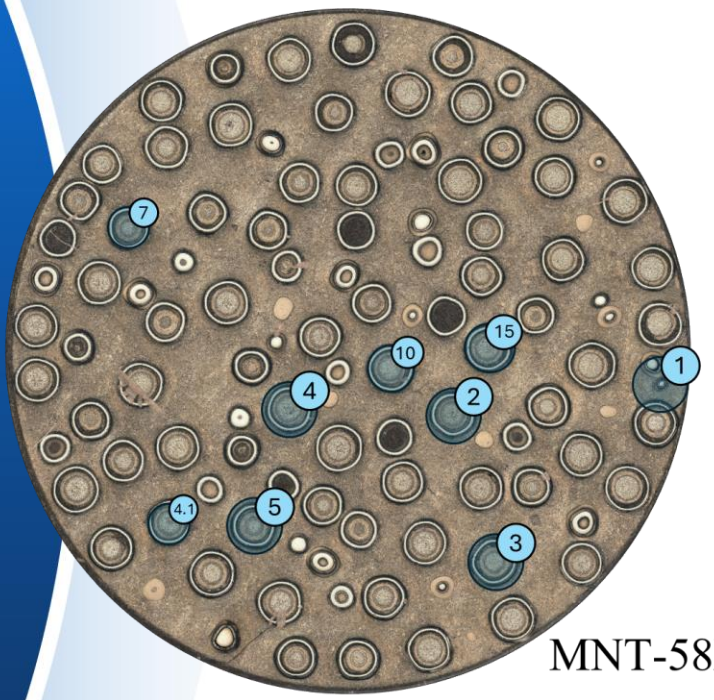
Thermal Conductivity Microscope (TCM) Methods

- The TCM is a state-of-the-art instrument built by INL scientists in collaboration with NSUF to deliver a mesoscale thermal transport measurement capability [3].
- The TCM is the only instrument in the world capable of mesoscale thermal transport PIE
- It uses a hybrid thermoreflectance technique to measure thermal conductivity and thermal diffusivity on the microscale (~12 – 50 um)
  - Amplitude modulated (1-100 kHz) red heating pump laser generates thermal waves in the substrate
  - A green probe laser, spatially separated from pump laser, measures the effects of changes in optical reflectivity of substrate, mathematically related with heat transport properties of substrate [4].

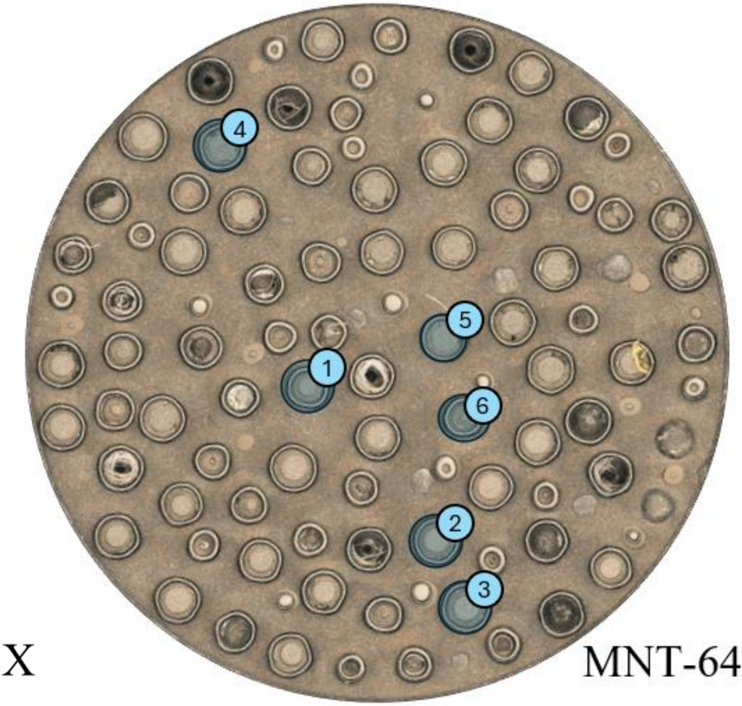




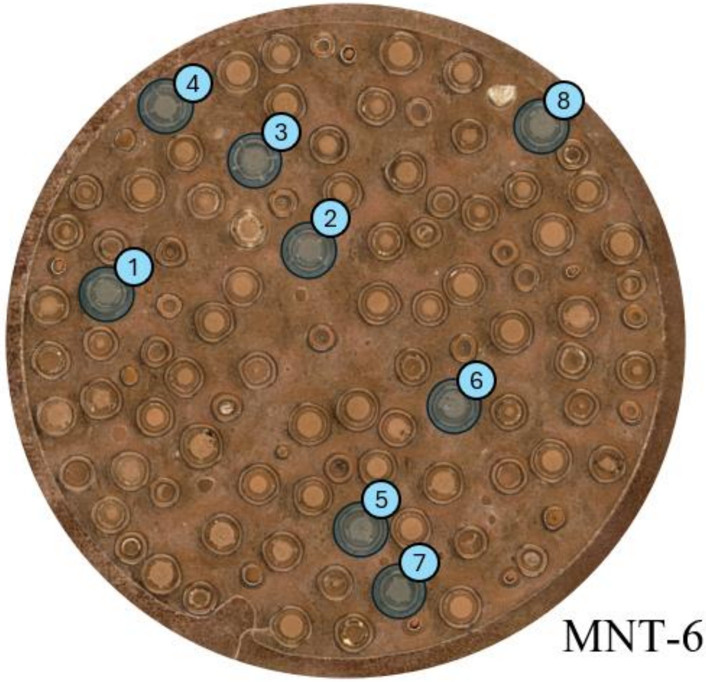
TCM Particle Selection



MNT-58X



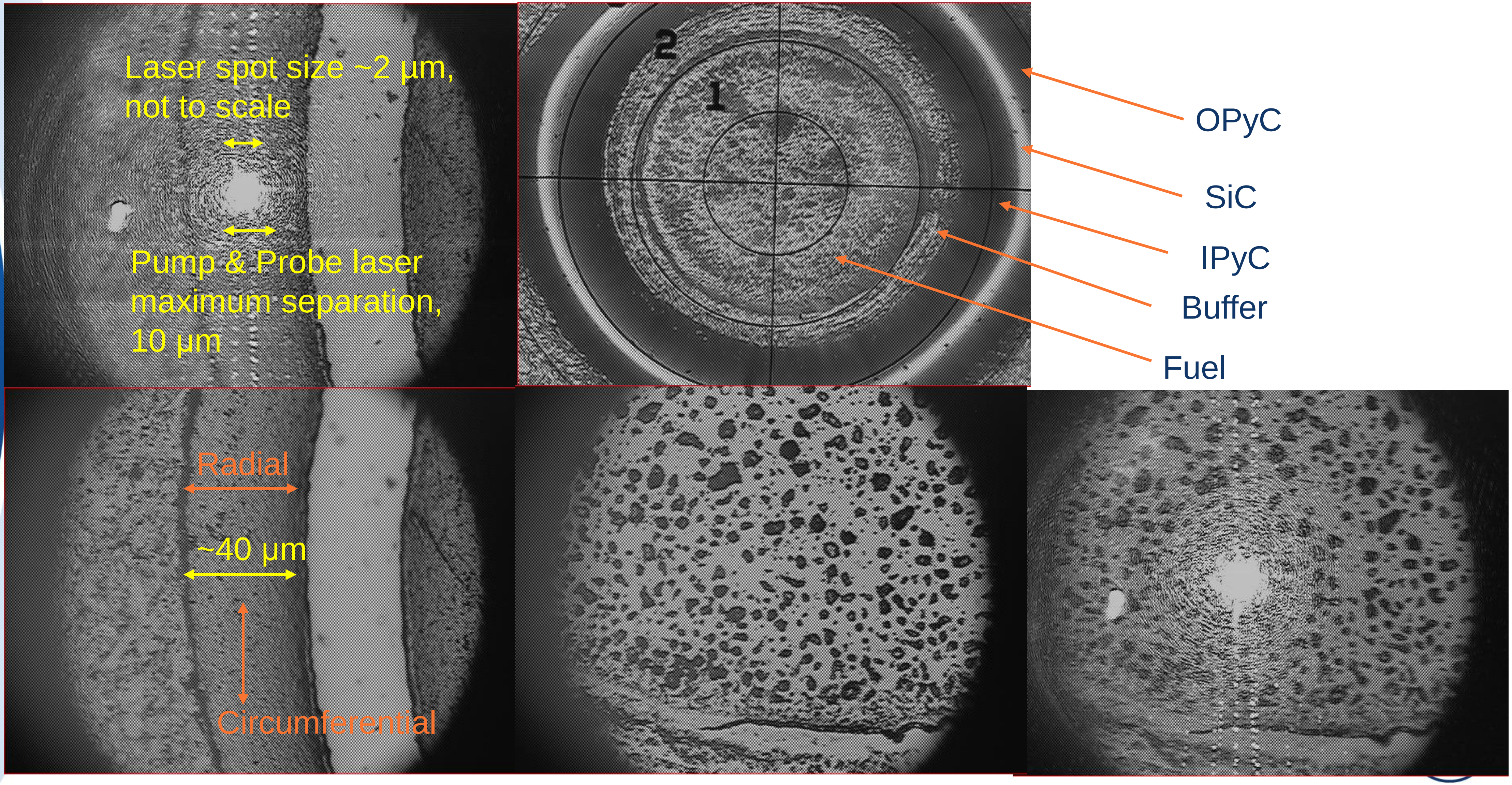
MNT-64X



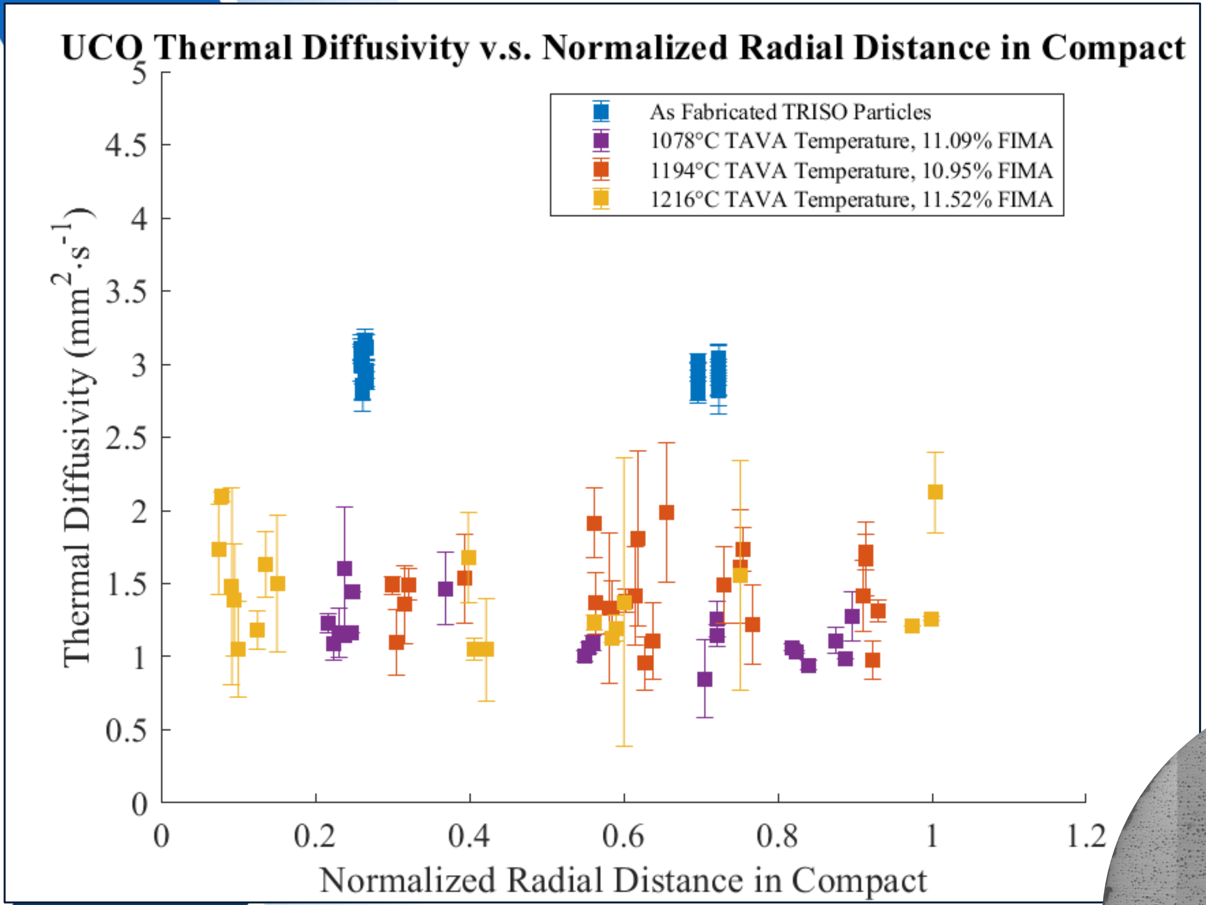
MNT-67X

- Intact, whole particles were chosen for SEM high resolution microscopy and TCM measurements
- Particles were selected based on evaluating effects of radial distance from the center of the compact

TCM Measurement Examples

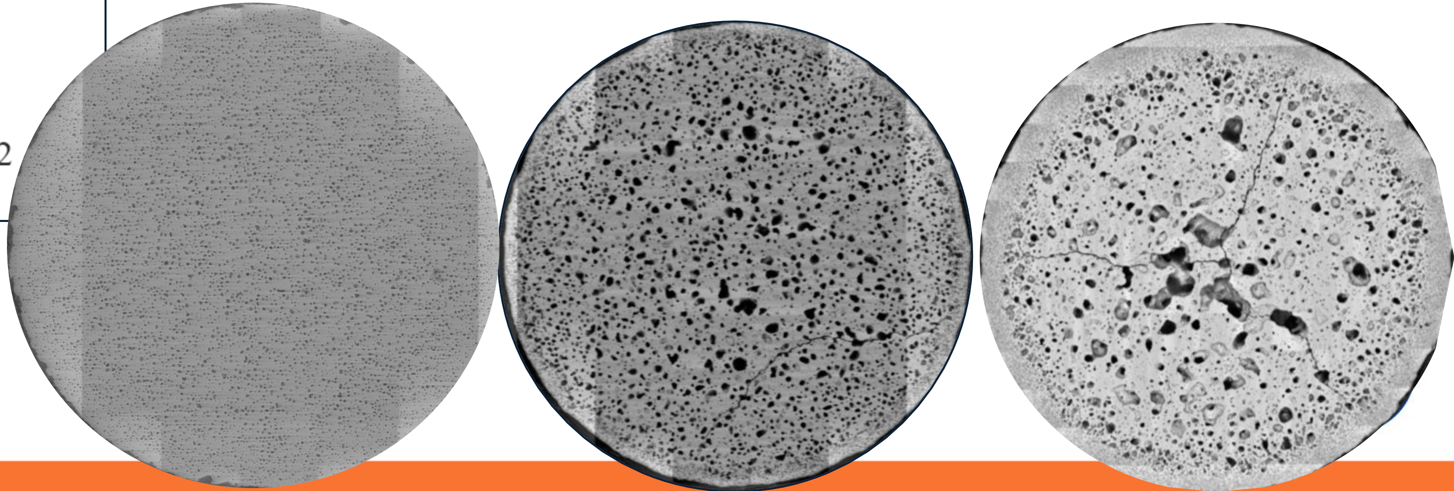


UCO Kernel Pre- to Post-Irradiation

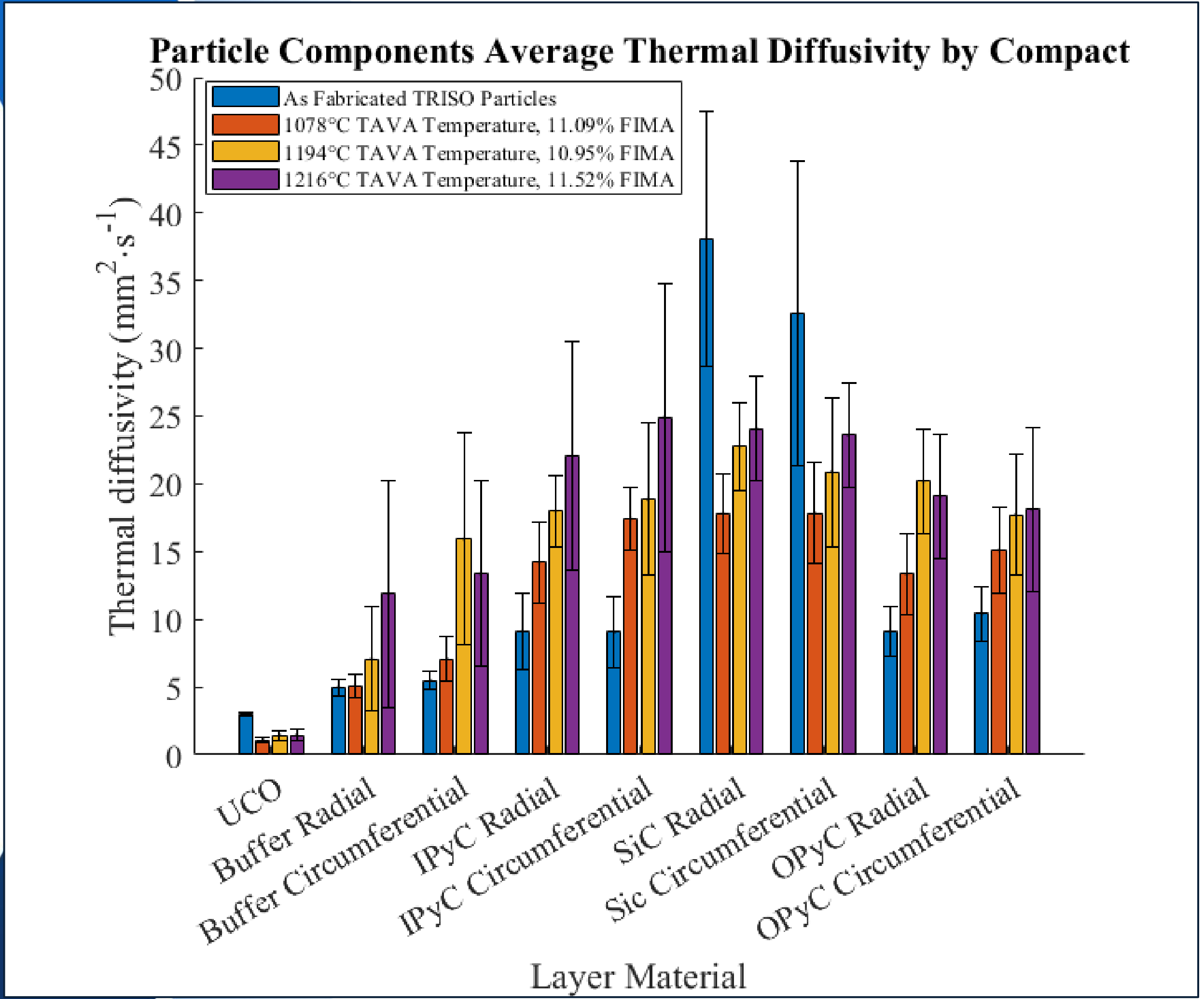


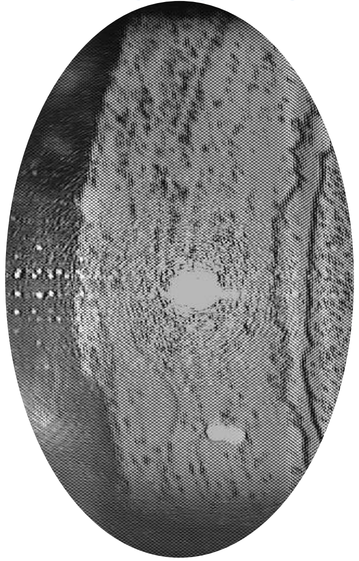
- Compared to as fabricated UCO kernels:
  - Average thermal diffusivity in the higher TAVA temperature compacts is decreased by ~50%
  - Average thermal diffusivity in the lower TAVA temperature compacts is decreased by ~64%
- At higher TAVA temperature UCO kernel thermal diffusivity may recover slightly
- Porosity profiles will be needed to better understand degradation of thermal diffusivity and conductivity [5]

TCM Measured fuel particle SEM images by TAVA temperature
Left to Right: 1078°C, 1194°C,1216°C



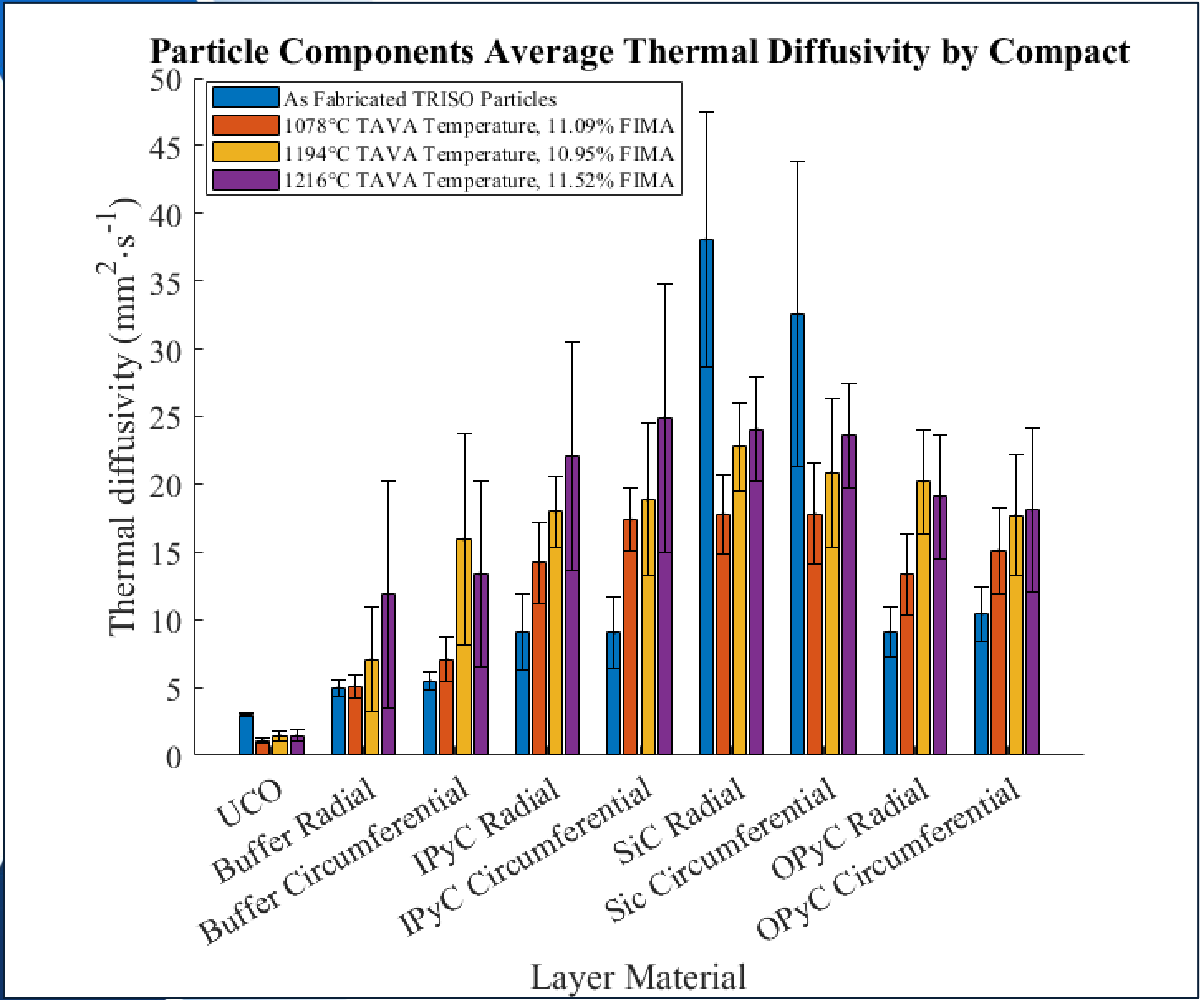
Buffer Layer Pre- to Post-Irradiation

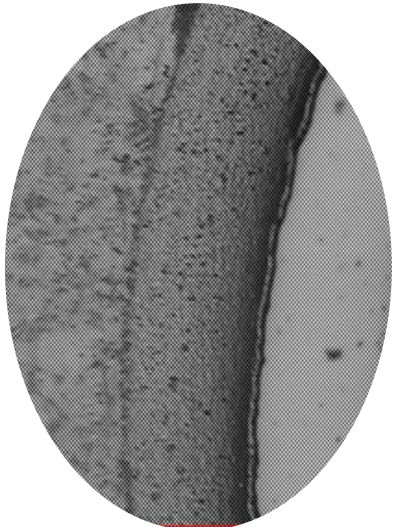




- Thermal diffusivity tends to increase post-irradiation with increases correlated with TAVA temperature
- Likely driven by:
  - Densification of the layer
  - Simultaneous point defect annealing at higher temperatures
- Significant anisotropy is difficult to detect given high deviation from particle to particle
  - Deviation from particle to particle may be due to localized annealing
  - Fabrication defects could also cause variation [6].

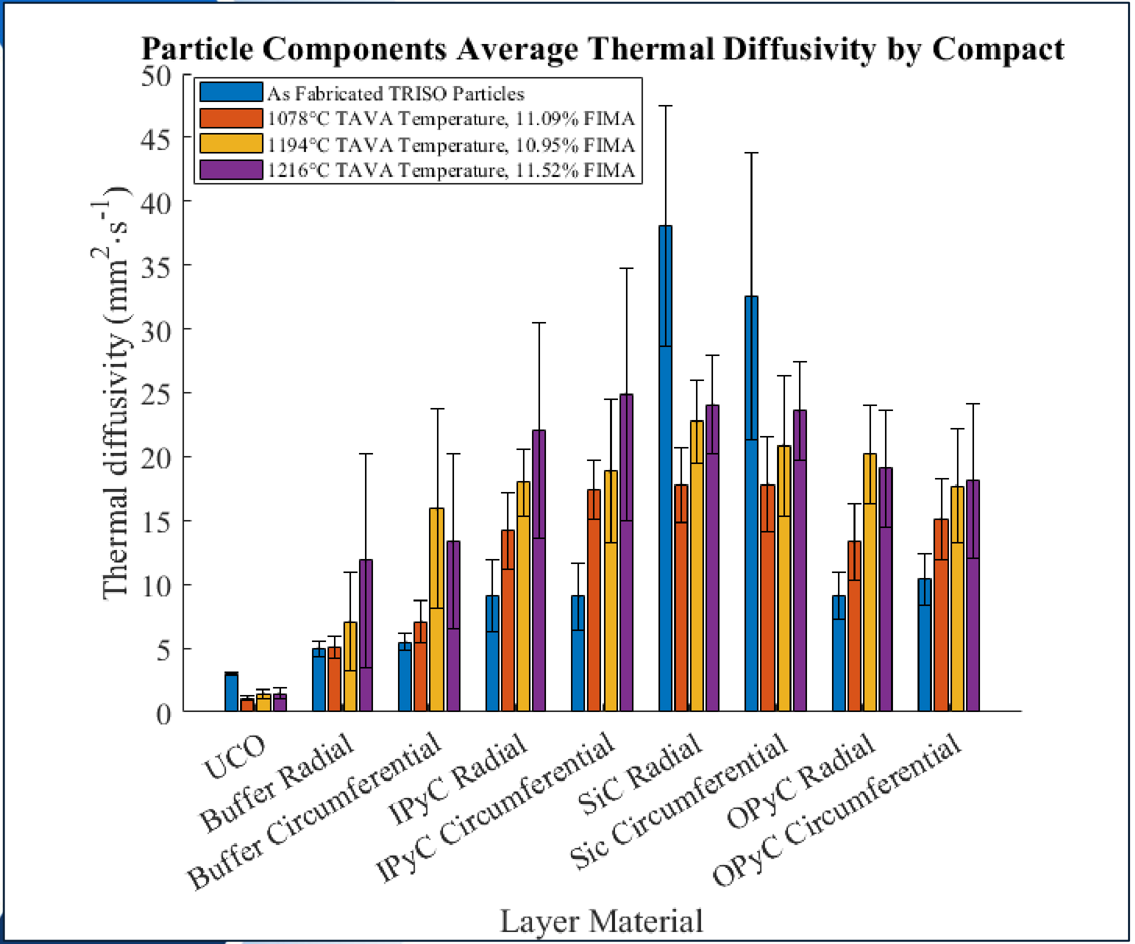
Pyrolytic Carbon Layers Pre- to Post-Irradiation

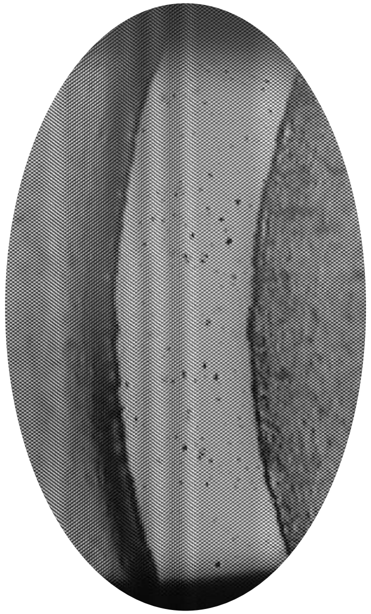




- Thermal diffusivity increases post-irradiation
  - Potentially due to annealing of fabrication induced defects
- Higher TAVA temperature leads to a higher thermal diffusivity due to annealing of irradiation induced defects
- Difficult to detect any significant anisotropy

Silicon Carbide Layer Pre- to Post-Irradiation





- Strong degradation from pre- to post-irradiation in thermal diffusivity
  - Irradiation leads to point defects which reduce thermal diffusivity
- Higher TAVA temperatures lead to higher thermal diffusivity values due to annealing of irradiation induced defects
- Potentially other effects contributing to degradation include:
  - Nanovoids [7]
  - Lower thermal conductivity precipitates (e.g. silicides)

TCM Measurements Conclusions

- Compared to as fabricated material:
    - Fuel and Silicon Carbide thermal diffusivity degrades significantly (50 – 60% and 37 – 53% respectively)
        - Some recovery in thermal diffusivity likely due to annealing at higher TAVA temperatures
    - Pyrolytic Carbons increase thermal diffusivity (ranging from 46 – 175%)
        - Increase correlates to increasing TAVA
- No significant anisotropy found
- Measurements of matrix graphite were attempted with some success, however the surface was non-ideal for TCM measurements
- More than 800 measurements across the 3 compacts

TCM Measurement Discussion & Takeaways

- Thermal diffusivity values measured by the TCM at RT suggest that PARFUME may be underestimating layer thermal transport (PARFUME's approach is conservative)
- PARFUME values used from 600 to 1300ºC [7]:
  - PyC thermal conductivity is assumed to be **invariant** at 4 W/m·K
    - TCM values at RT are between 212% and 712% higher
  - Buffer thermal conductivity is assumed to be **invariant** at 0.5 W/m·K
    - TCM values at RT are between 900% and 1900% higher
  - SiC thermal conductivity is assumed to decrease from 20 to 13 W/m·K
    - TCM values at RT are between 87% and 150% higher
- Annealing of defects in the layers with increasing TAVA temperature may contribute to a higher effective thermal conductivity than currently modeled [8][9].

Ongoing and Future work at IMCL

- Laser Flash Analysis – high accuracy bulk measurements
  - Custom geometric FEA and EM for heat transfer
  - 1 material property can be extracted using TCM as source for other materials, or whole compact can be compared against TCM
- Simultaneous Thermal Analysis – Differential Scanning Calorimetry
  - Bulk heat capacity
  - Annealing effects
  - Fission gas release
- X-ray Computed Tomography – Search for voids in compacts, establish geometric mesh for EM/FEA modeling for LFA and STA
- Porosity profiles for each TCM measurement for correlation with thermal diffusivity
- Calculation of thermal conductivities using experimental density and heat capacity*

Research Group & Contributors

Ethan J. Hisle[1], Tsvetoslav R. Pavlov[1], Amey Khanolkar[2], Zilong Hua[2], Narayan Poudel[1], Chuting Tsai[1],Isaac Cutler[1], Matthew Goodson[1], Scott T. Anderson[3],Tanner Mauseth[3], Cameron Howard[3], Mario D. Matos III[3],Riley S. Moeykens[1],Swapnil K. Morankar[4], William C. Chuirazzi[4], Subhashish Meher[5], Lu Cai[6], John D. Stempien[6]

[1]Thermophysical Property Group – Characterization & PIE - INL

[2]Center for Thermal Energy Transport Under Irradiation – Condensed Matter & Materials Physics – INL

[3]Correlative Microscopy Group – Characterization & PIE – INL

[4]Diffraction and Imaging Group – X-Ray and Neutron Sciences - INL

[5]Energy and Environment Directorate - PNNL

[6]Fuel development, Performance & Qualification Group - INL

Special thanks to IMCL operations staff for their support

Questions?

- Additional questions can be sent to:
  - Ethan.hisle@inl.gov
  - Tsvetoslav.pavlov@inl.gov
  - John.Stempien@inl.gov

References

1. F.J. Rice, J.D. Stempien, P.A. Demkowicz, *Ceramography of irradiated TRISO fuel from the AGR-2 experiment*,Nuclear Engineering and Design,Volume 329,2018,Pages 73-81,ISSN 0029-5493, https://doi.org/10.1016/j.nucengdes.2017.10.010
2. Hunn, John D., Montgomery, Fred C., and Pappano, Peter J. *Data Compilation for AGR-2 UCO Variant Compact Lot LEU09-OP2-Z*. United States: N. p., 2010. Web. doi:10.2172/1630497.
3. Hurley, David H, Hua, Zilong, and Schley, Robert S. *Closeout Phase II Qualification of the Thermal Conductivity Microscope for IMCL*. United States: N. p., 2018. Web.
4. David H. Hurley, Robert S. Schley, Marat Khafizov, Brycen L. Wendt; Local measurement of thermal conductivity and diffusivity. *Rev. Sci. Instrum.* 1 December 2015; 86 (12): 123901. https://doi.org/10.1063/1.4936213
5. Stempien, John D., Plummer, Mitchell A., Schulthess, Jason L., and Demkowicz, Paul A. *Measurement of kernel swelling and buffer densification in irradiated UCO and UO2 TRISO fuel particles from AGR-2*. United States: N. p., 2019. Web. doi:10.2172/1599772.
6. V. Barabash, I. Mazul, R. Latypov, A. Pokrovsky, C.H. Wu,The effect of low temperature neutron irradiation and annealing on the thermal conductivity of advanced carbon-based materials,Journal of Nuclear Materials,Volumes 307–311, Part 2,2002,Pages 1300-1304,ISSN 0022-3115,https://doi.org/10.1016/S0022-3115(02)00961-3.
7. Meher, S., van Rooyen, I.J. & Lillo, T.M. A Novel Dual-Step Nucleation Pathway in Crystalline Solids under Neutron Irradiation. Sci Rep 8, 98 (2018). https://doi.org/10.1038/s41598-017-18548-8
8. Skerjanc, William F, & Collin, Blaise P. Assessment of Material Properties for TRISO Fuel Particles in PARFUME. United States. https://doi.org/10.2172/1874979
9. G.E Youngblood, D.J Senor, R.H Jones,Effects of irradiation and post-irradiation annealing on the thermal conductivity/diffusivity of monolithic SiC and f-SiC/SiC composites,Journal of Nuclear Materials,Volumes 329–333, Part A,2004,Pages 507-512,ISSN 0022-3115,https://doi.org/10.1016/j.jnucmat.2004.04.111.
10. Charles Folsom, Changhu Xing, Colby Jensen, Heng Ban, Douglas W. Marshall, *Experimental measurement and numerical modeling of the effective thermal conductivity of TRISO fuel compacts*,Journal of Nuclear Materials,Volume 458,2015,Pages 198-205,ISSN 0022-3115,https://doi.org/10.1016/j.jnucmat.2014.12.042.
11. Stempien, John D., and Schulthess, Jason L. AGR-3/4 TRISO Fuel Compact Ceramography. United States: N. p., 2020. Web. doi:10.2172/1843925.

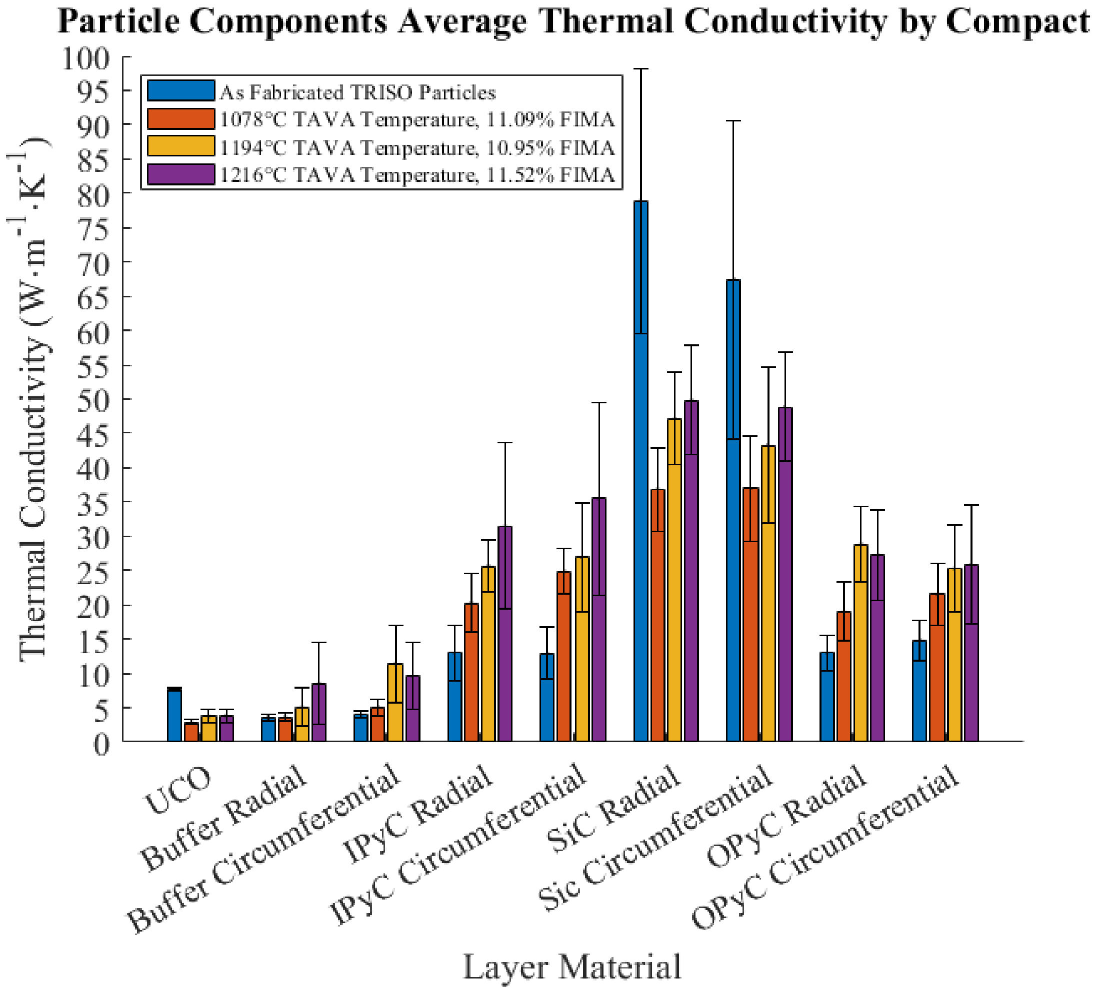
GAS-COOLED REACTOR

ADVANCED REACTOR TECHNOLOGIES PROGRAM

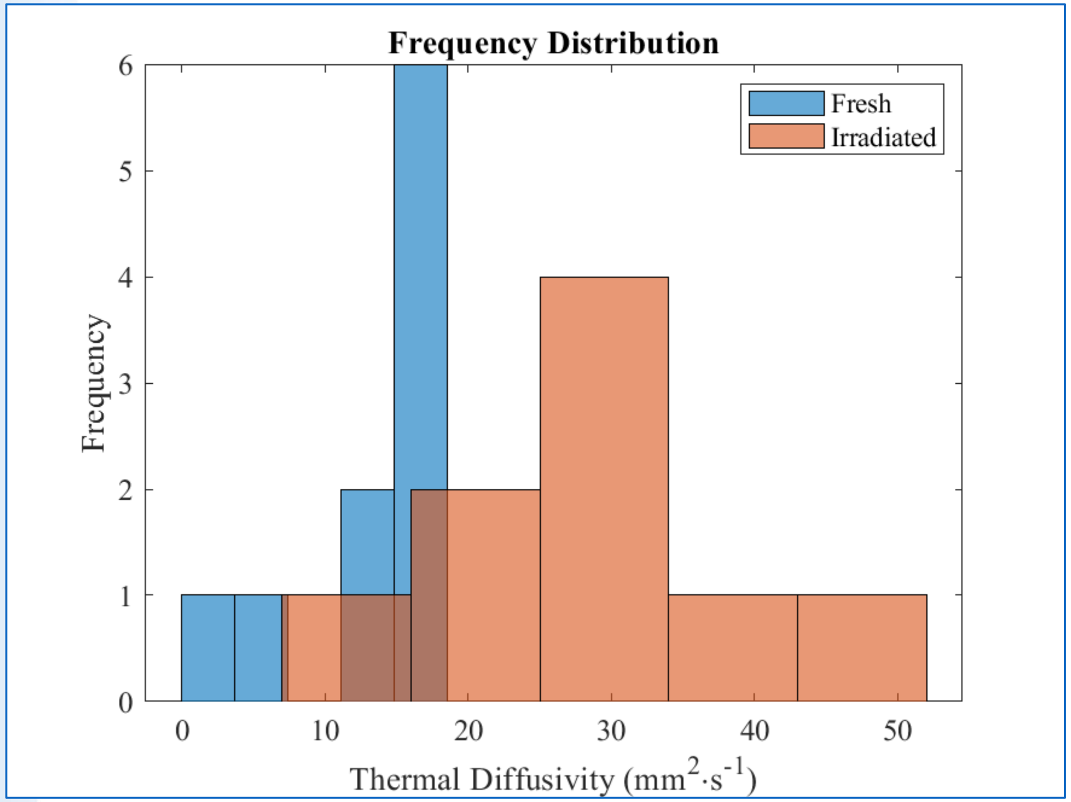
U.S. DEPARTMENT OF ENERGY

Supporting Slides

Thermal Conductivity Calculations

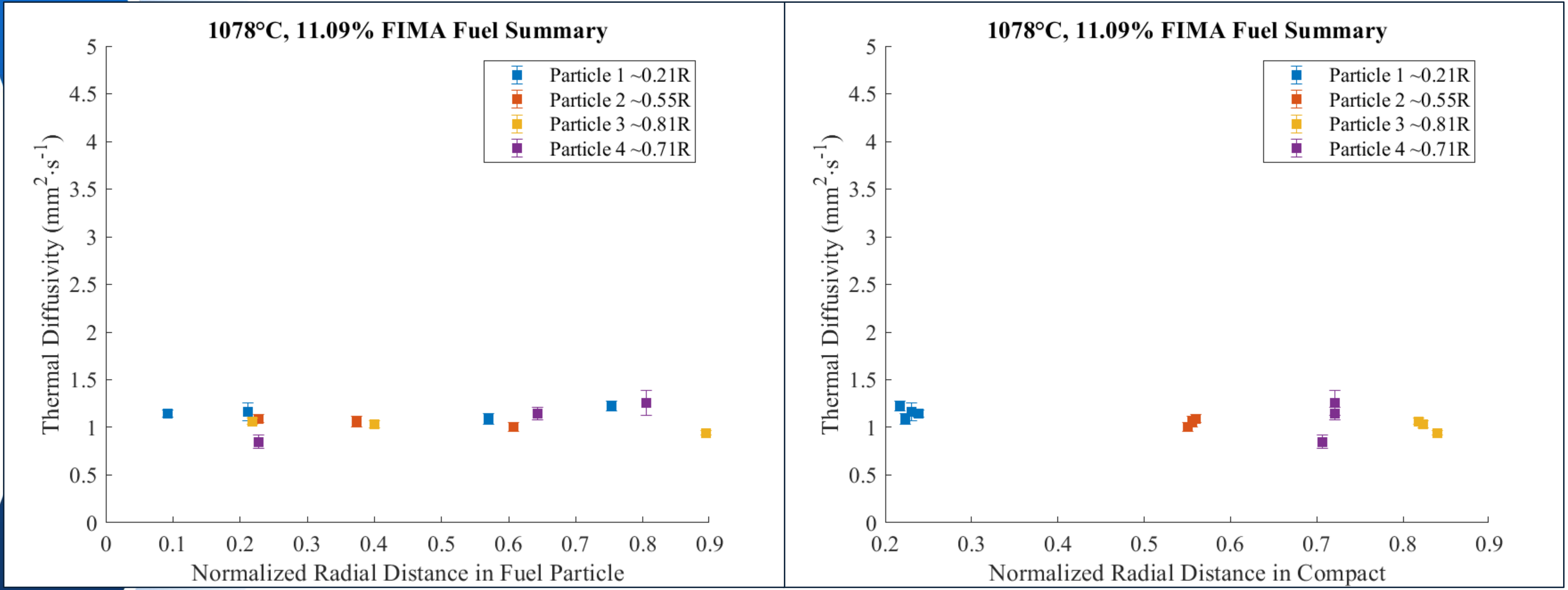


Graphite Initial Results

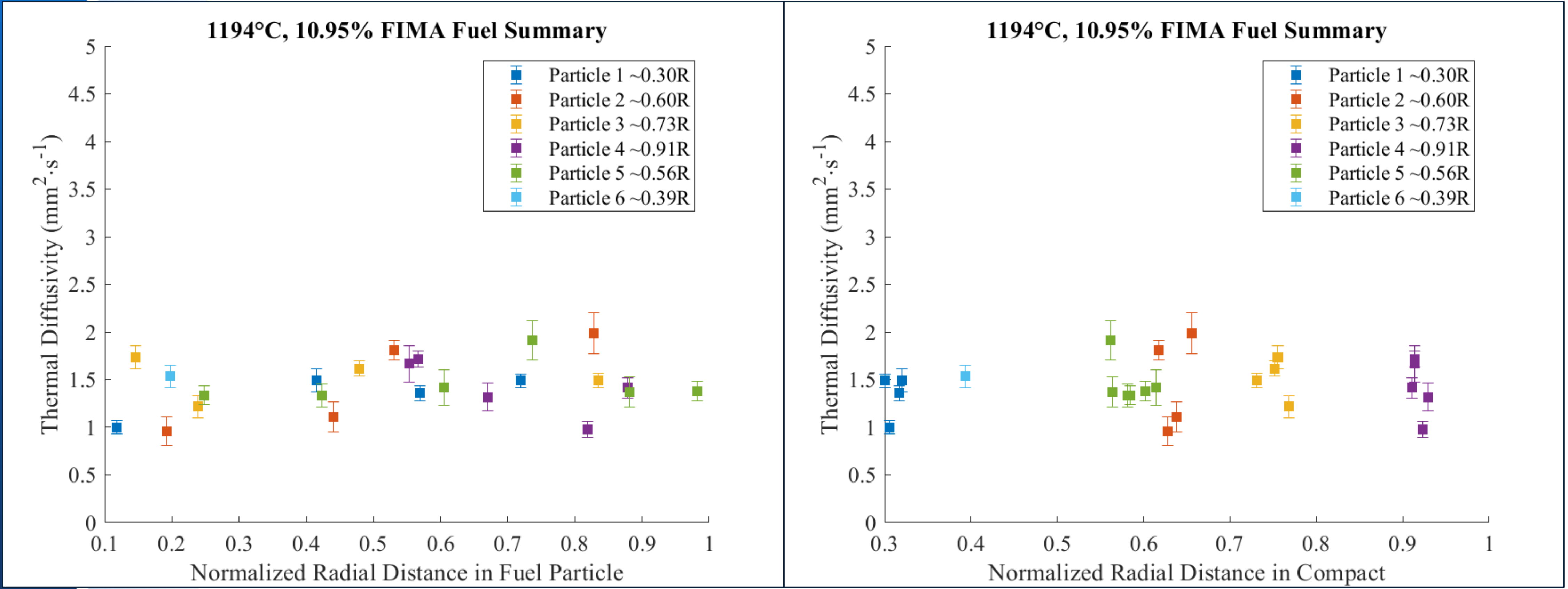


Fresh		Irradiated (1216ºC)	
Thermal Diffusivity (mm^2/s)	Standard Deviation (mm^2/s)	Thermal Diffusivity (mm^2/s)	Standard Deviation (mm^2/s)
13.89	5.38	28.6	9.92

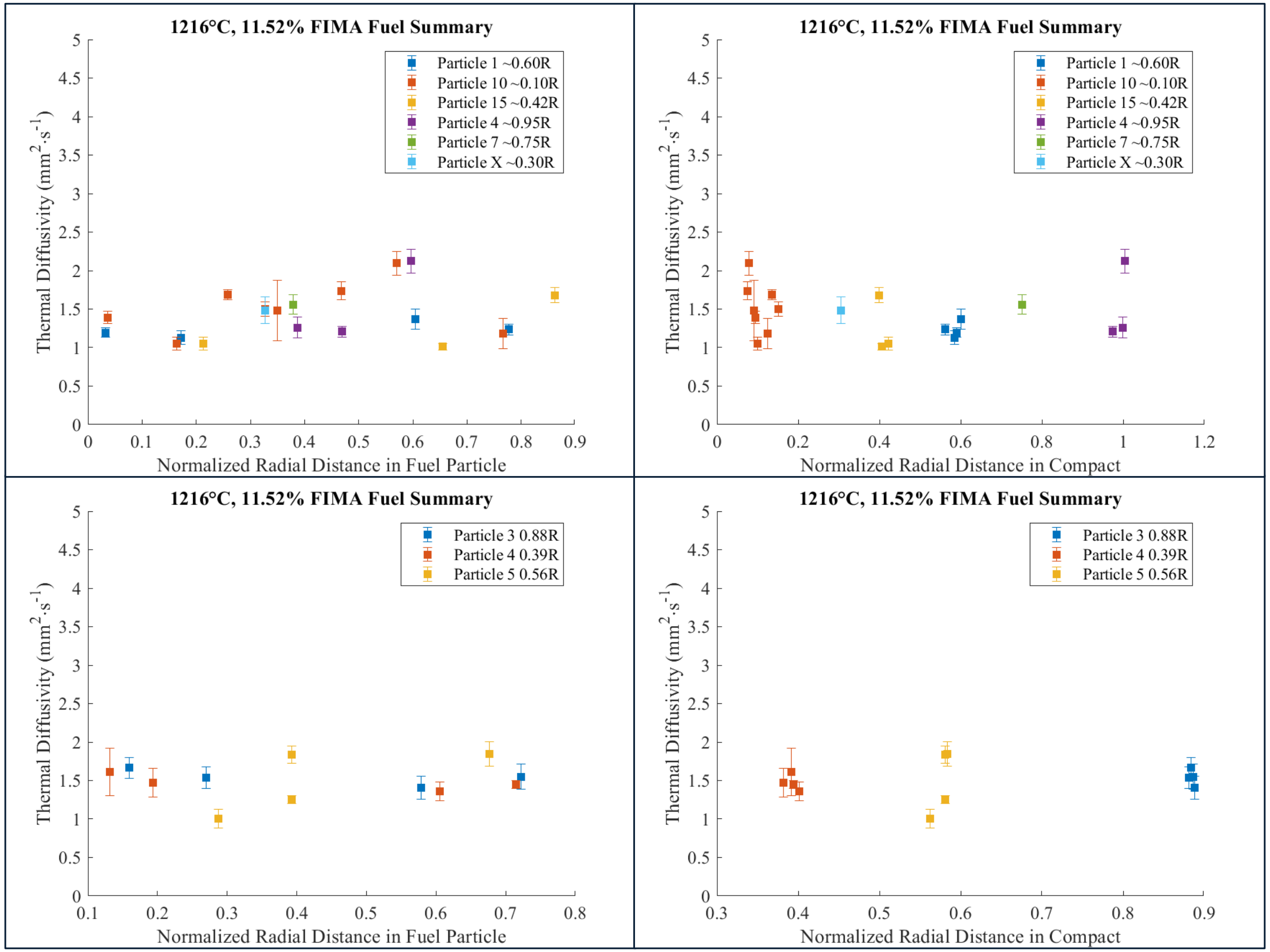
MNT-64X Fuel Summary



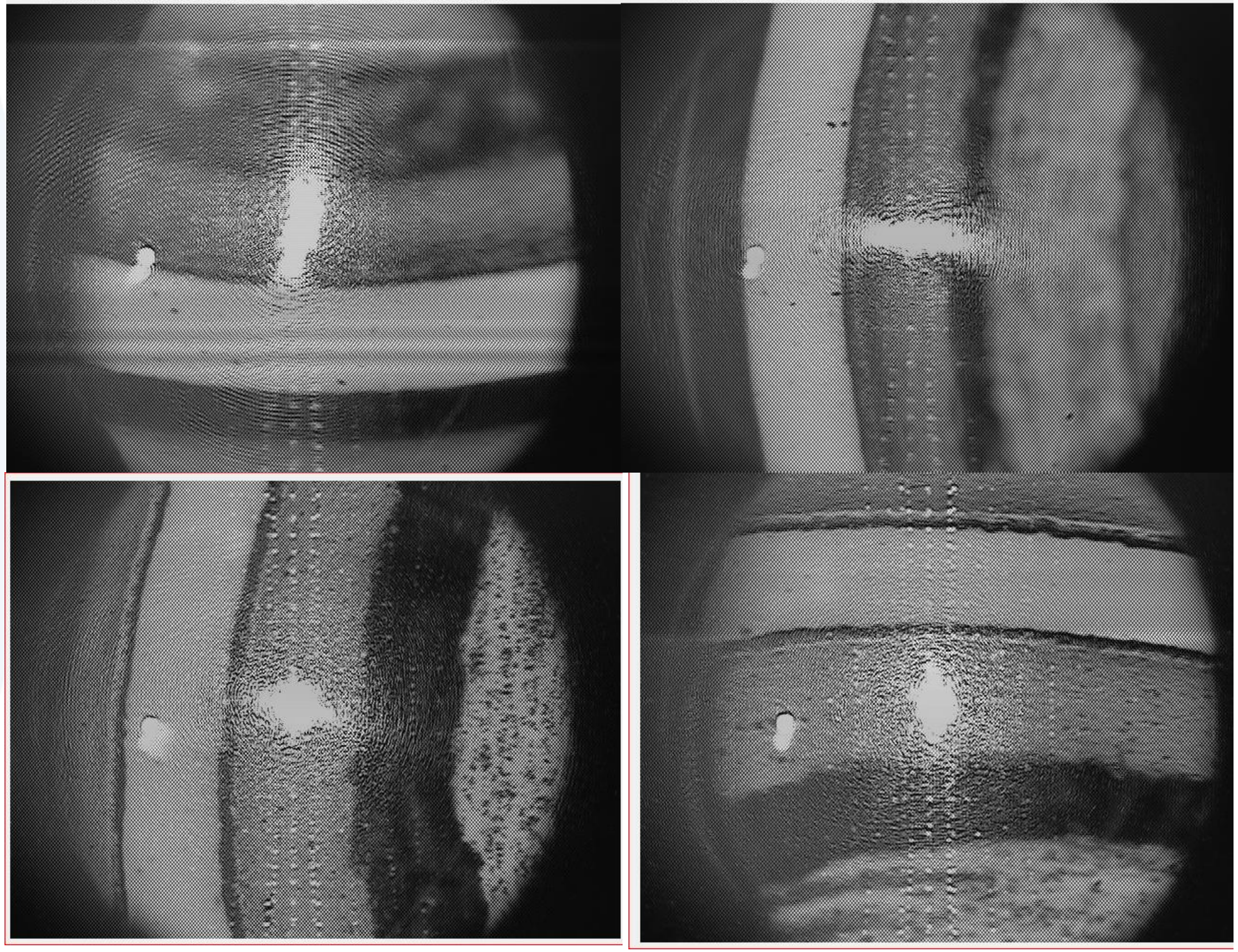
MNT-67X Fuel Summary



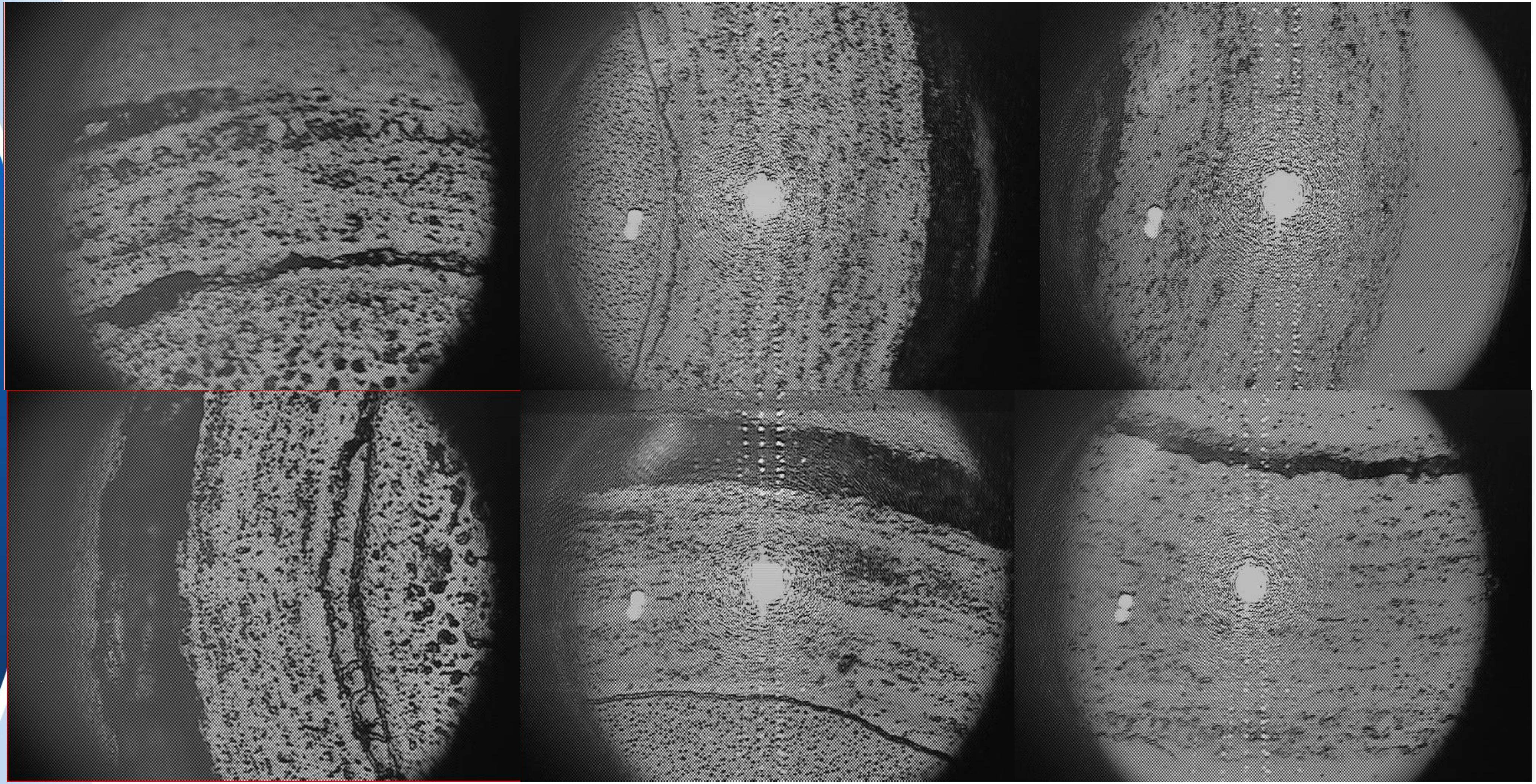
MNT-58X Fuel Summary



Substrate surface was not always ideal



Buffer Layering By Compact



From left to right, 1216ºC TAVA, 1078ºC TAVA, and as fabricated fresh particles

EMT of TRISO Compacts using PARFUME by Folsome et al. [9]

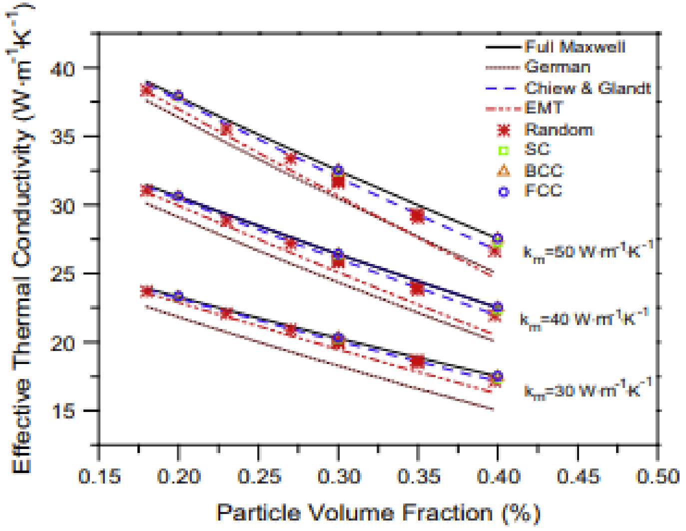


Fig. 5. FEA results for the ETC of the fuel compact for multiple matrix thermal conductivities (k_m) compared to analytical ETC models as a function of particle-volume fraction. Particle thermal conductivity is taken as 4.13 W m^{-1} K^{-1}.

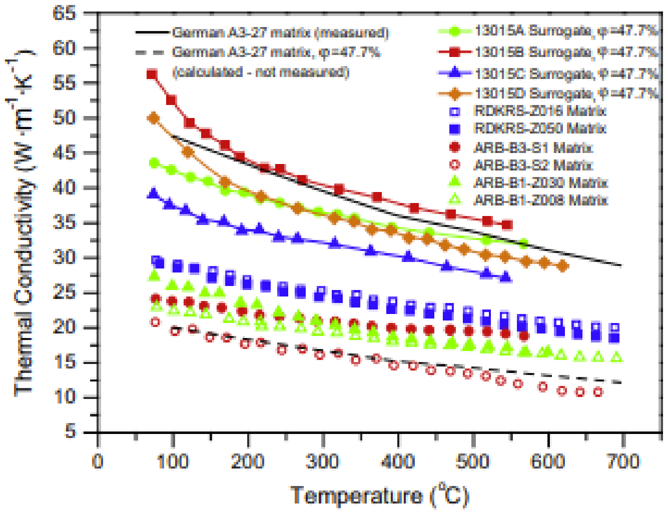


Fig. 6. Measured thermal conductivity results of matrix-only and surrogate samples plotted with legacy German data. Sample information can be found in Section 3.2.

- Developed an EMT of the TRISO compacts based on values from PARFUME
- Predicted that addition of relatively insulating particles (~4 W/m·K) into the matrix would reduce the ETC of the bulk
- Experimentation proved the opposite. The addition of the particles lead to a higher thermal conductivity of the bulk compact. Possible causes:
  - The disordered matrix becomes more ordered with the addition of particles, leading to a higher thermal conductivity
  - The particle layers are treated as very insulating, and the particle thermal conductivity would need to be (~100 W/m·K) to explain experimental results
- Likely some combination of local reordering of the matrix around the particles, and higher layer thermal conductivity

Stempien et al. findings on UCO fuel

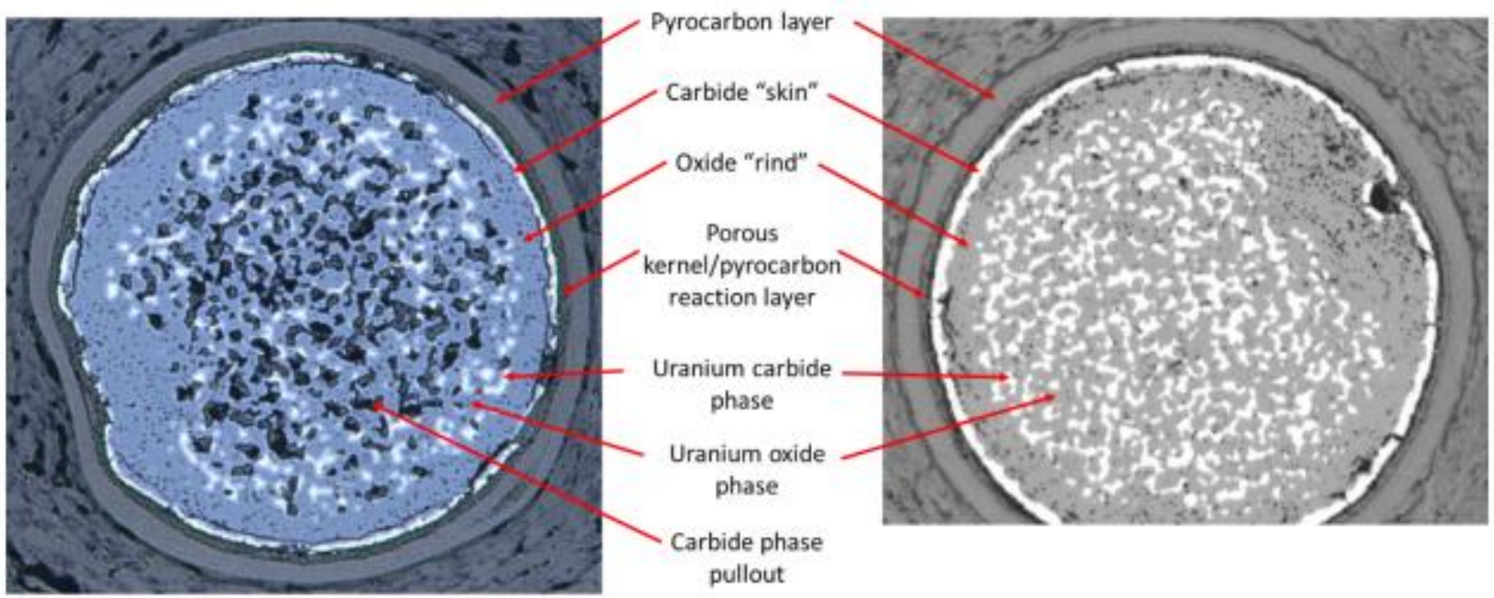


Figure 9. Cross-section of unirradiated DTF particle “1” from MNT78A (see Figure 8) in this work (left) and from work on a different, unirradiated compact in Hunn et al. 2011 Figure 1-13 (right).

- Fuel kernel forms a distinct uranium carbide phase and a uranium dioxide phase
- Edge of fuel kernel forms an oxide rich fuel region
- Layer of carbide forms between buffer and fuel kernel