July 26, 2023

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Statistical modeling of the effect of microstructural heterogeneity on the irradiation behavior of TRISO fuel buffer layer

NEUP: 20-19556

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Funding: $800,000

Period: 10/1/2020 – 9/30/2023 (with NCE to 6/30/2024)

ART
Advanced Reactor Technologies

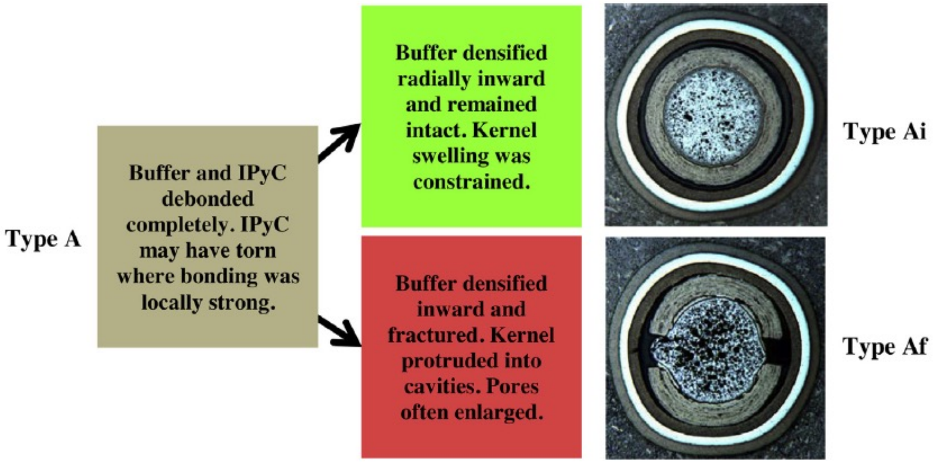
Outline

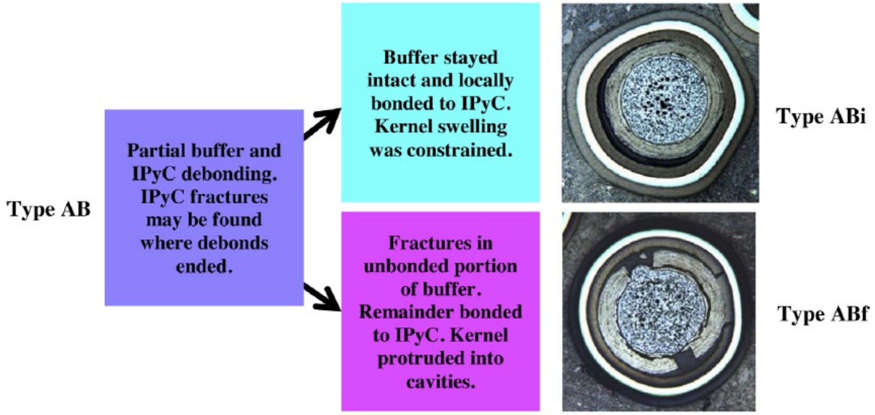
- **Background & objective**
- **Hypothesis**
- **Research design**
- **Accomplishments**
  - ➢ How do we describe buffer? matrix microstructure + porosity
  - ➢ How does buffer porosity affect its fracture behavior?
  - ➢ Irradiation induced changes in porosity and matrix microstructure
  - ➢ Super-elasticity of unirradiated buffer pyrocarbon
- **Outlook**
- **Summary**

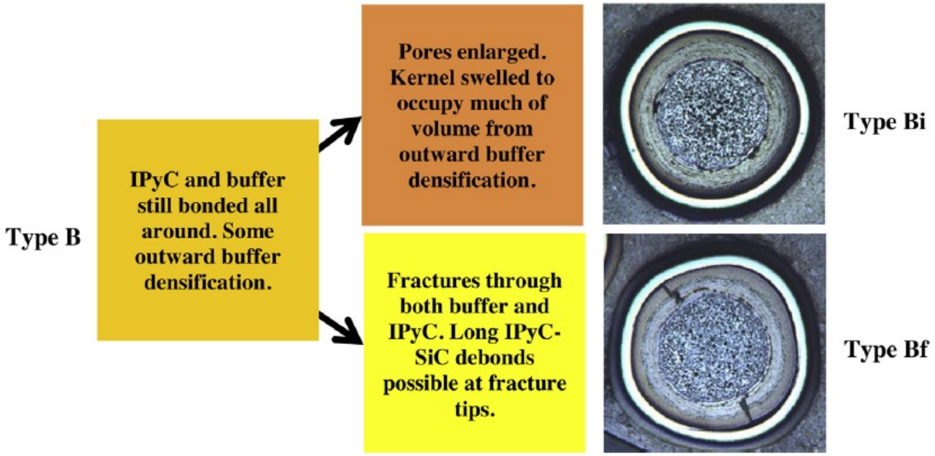
Background & objective

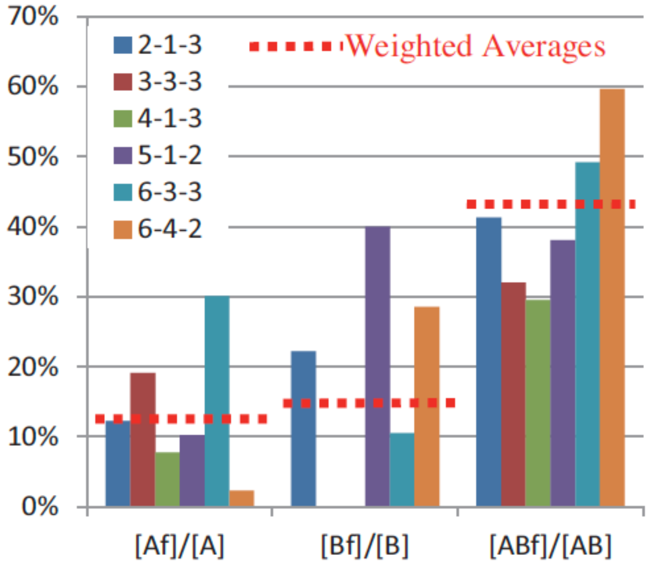
- Buffer fracture is stochastic showing three different modes: Debonding (**A**), radial fracture (**B**), and partial debonding (**AB**), while mode **AB** leads to higher probability of failure.

- **Our objective is to answer:**
  - Why fracture is stochastic?
  - What governs the selection of fracture mode?
  - Can we control the fracture mode?







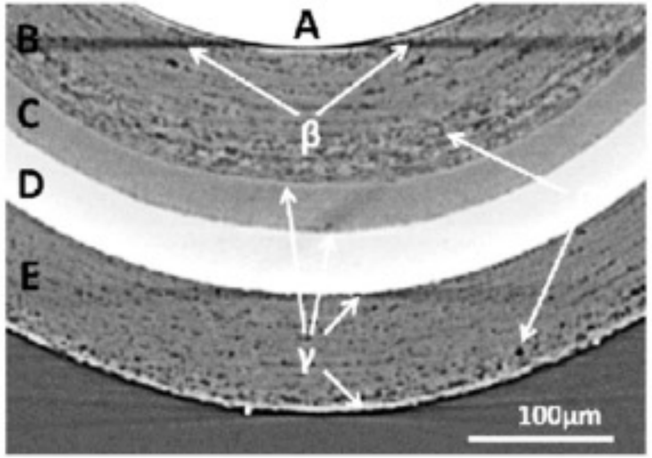


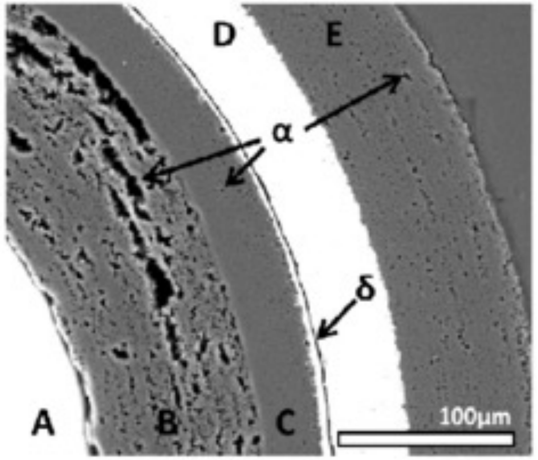
Ploger, S. A., Demkowicz, P. A., Hunn, J. D., & Kehn, J. S. (2014). Nuclear Engineering and Design, 271, 221-230

Hypothesis: The heterogeneous distribution of buffer porosity determines the initiation and propagation of buffer fracture.

Buffer porosity:

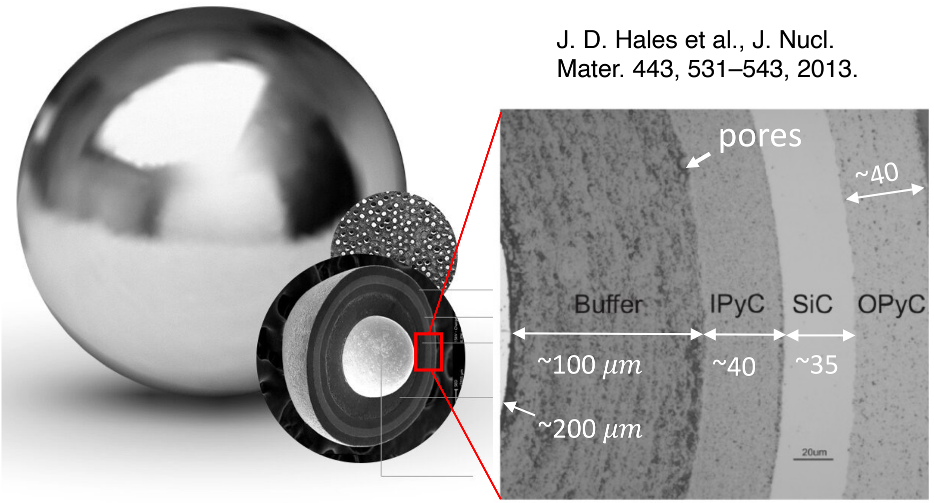
- Random distribution
- Local fluctuation
- Radially increasing
- Connectivity along tangential direction



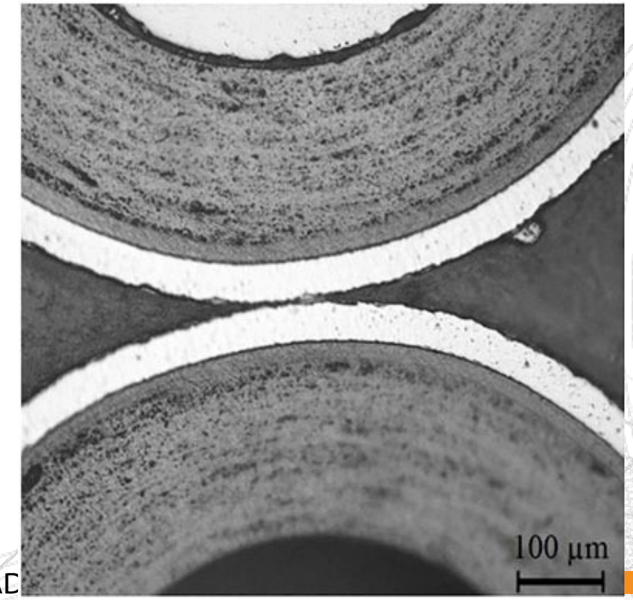


T. Lowe et al. JNM 461, 29-352015

J. D. Hales et al., J. Nucl. Mater. 443, 531–543, 2013.

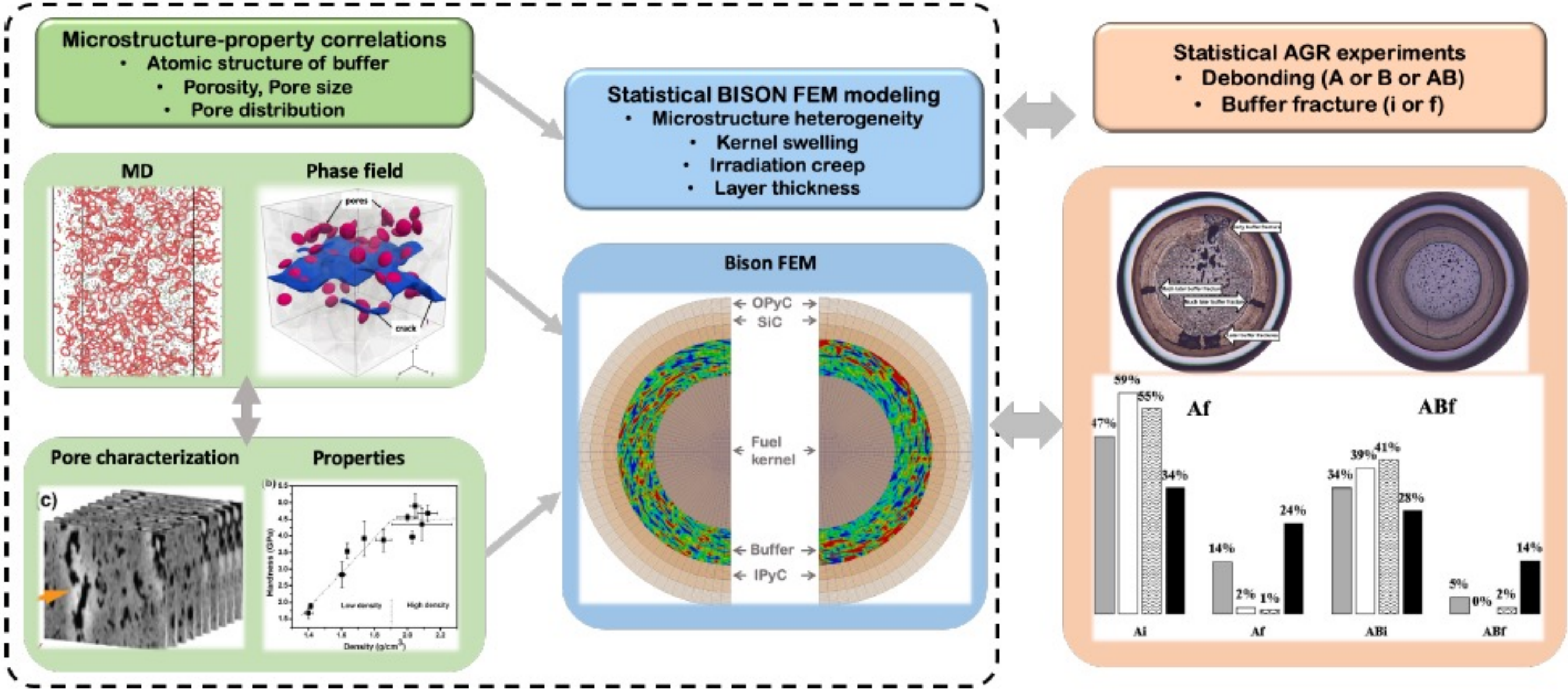


S. Liu et al. Front. Mater., 04 January 2023



Research design

- What are the actual porosity in buffer and its spatial distribution?
- What's the atomic structure of buffer, which is a low density, non-textured pyrocarbon?
- How does porosity correlate with mechanical properties: moduli, toughness, etc.?
- How does porosity distribution affect the buffer fracture (tearing)?



How do we describe buffer?

- ## FIB-SEM characterization of buffer porosity

  - *Griesbach et al. Microstructural heterogeneity of the buffer layer of TRISO nuclear fuel particles, JNM 574 (2022)*

  1st place Winner of FY 2023 Innovations in Nuclear R&D Student Competition: Fuel Cycle Technologies

- ## Atomistic simulations of pyrocarbon matrix

  - *R. David et al. Correlations between atomic structure and elastic properties in low-density non-textured pyrocarbon (in preparation)*

Buffer porosity morphology in unirradiated TRISO buffer (surrogate particles)

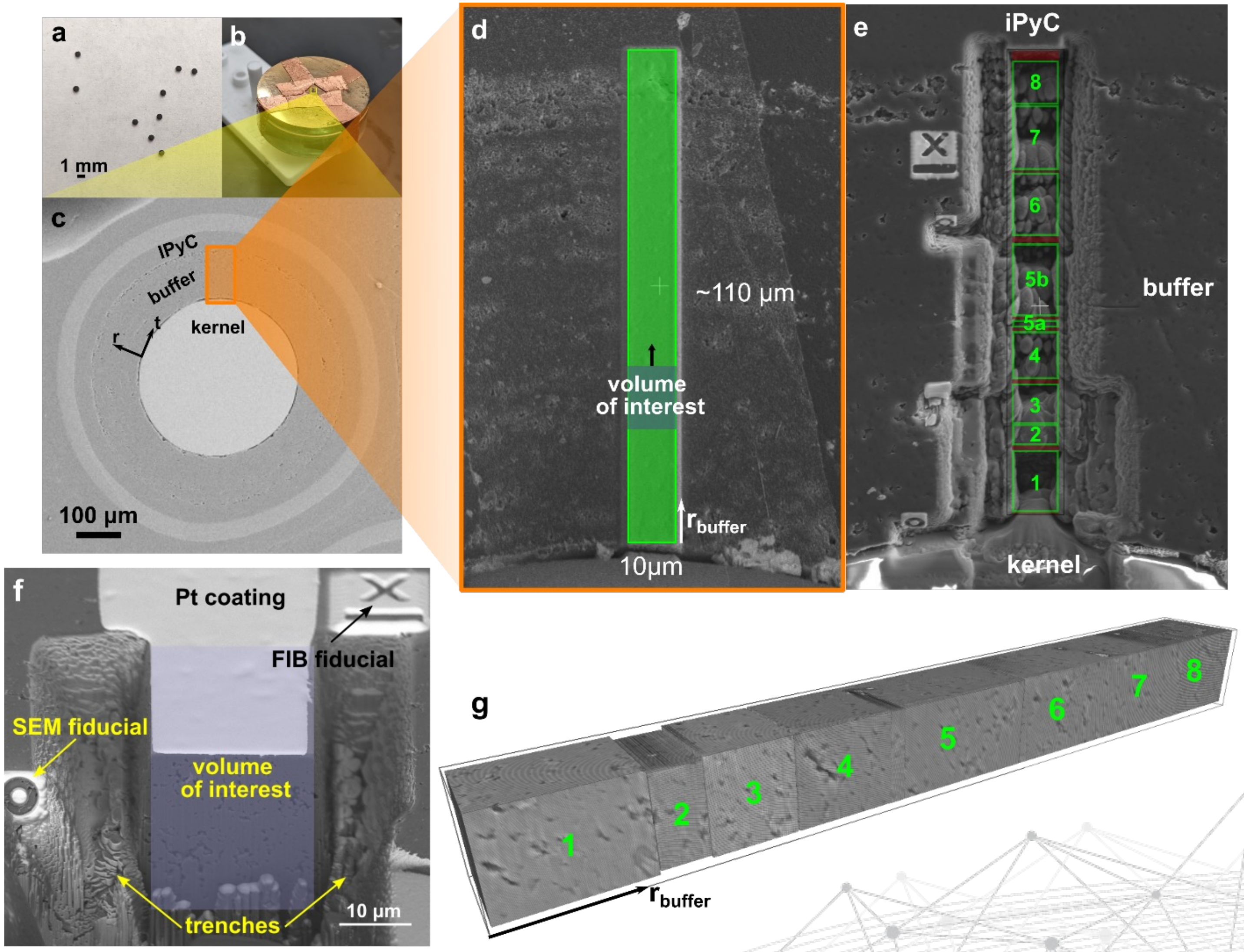
A robust data analysis workflow is established incorporating artificial intelligence assisted pore identification and segmentation for porosity characterization of FIB-SEM image.

FIB-SEM tomography

- FEI Helios PFIB with 30 kV Xe plasma
- Scan volume: 10×10×10 μm^3
- Slice thickness: 50 nm
- Voxel size: 38×49×50 nm^3

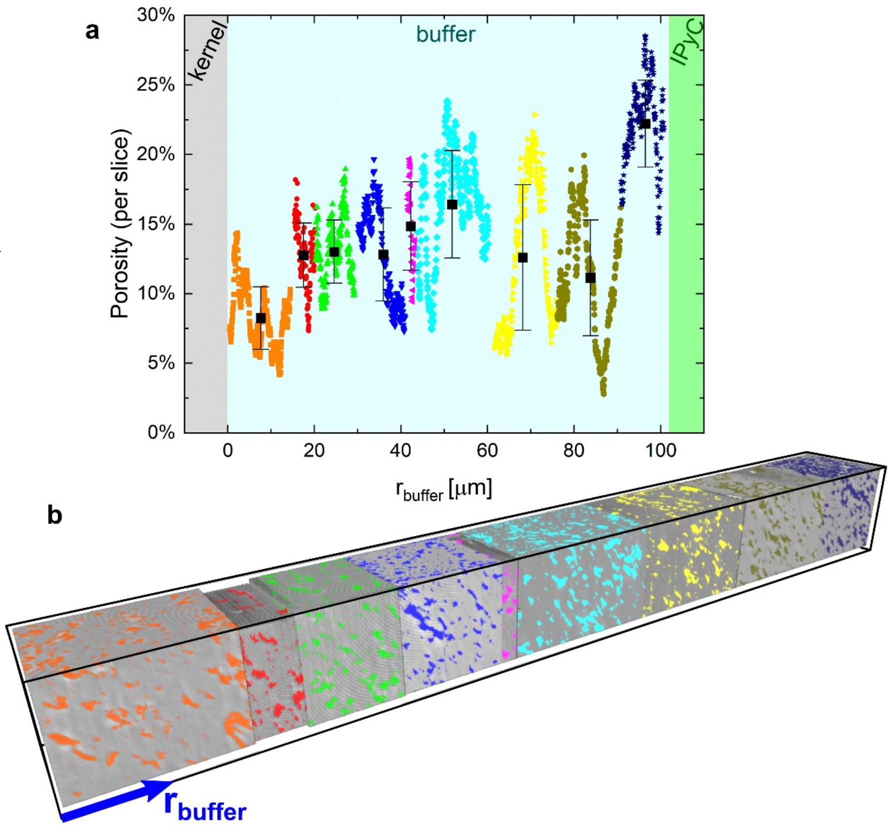
- Image processing and data analyses:
    - Avizo 9.5 and Dragonfly 2021.1
    - Deep learning segmentation

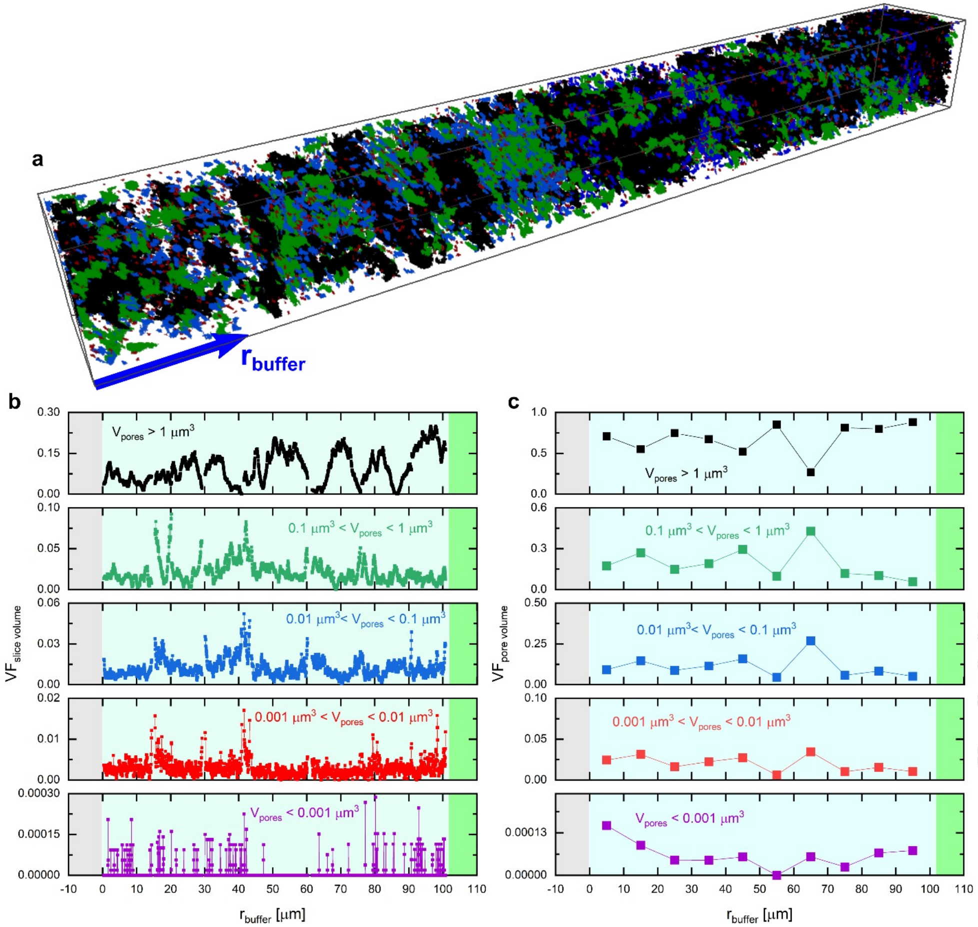
- 3D reconstruction



Buffer: low density pyrocarbon + randomly distributed pores

- **The Average buffer porosity is about 14% in reference to the 50% smeared theoretical density.**
- **Pores distribute randomly.**
- **Porosity increases radially.**
- **Porosity fluctuates locally.**
- Large pores (>1μm^3) dominates the porosity albeit fewer of them.

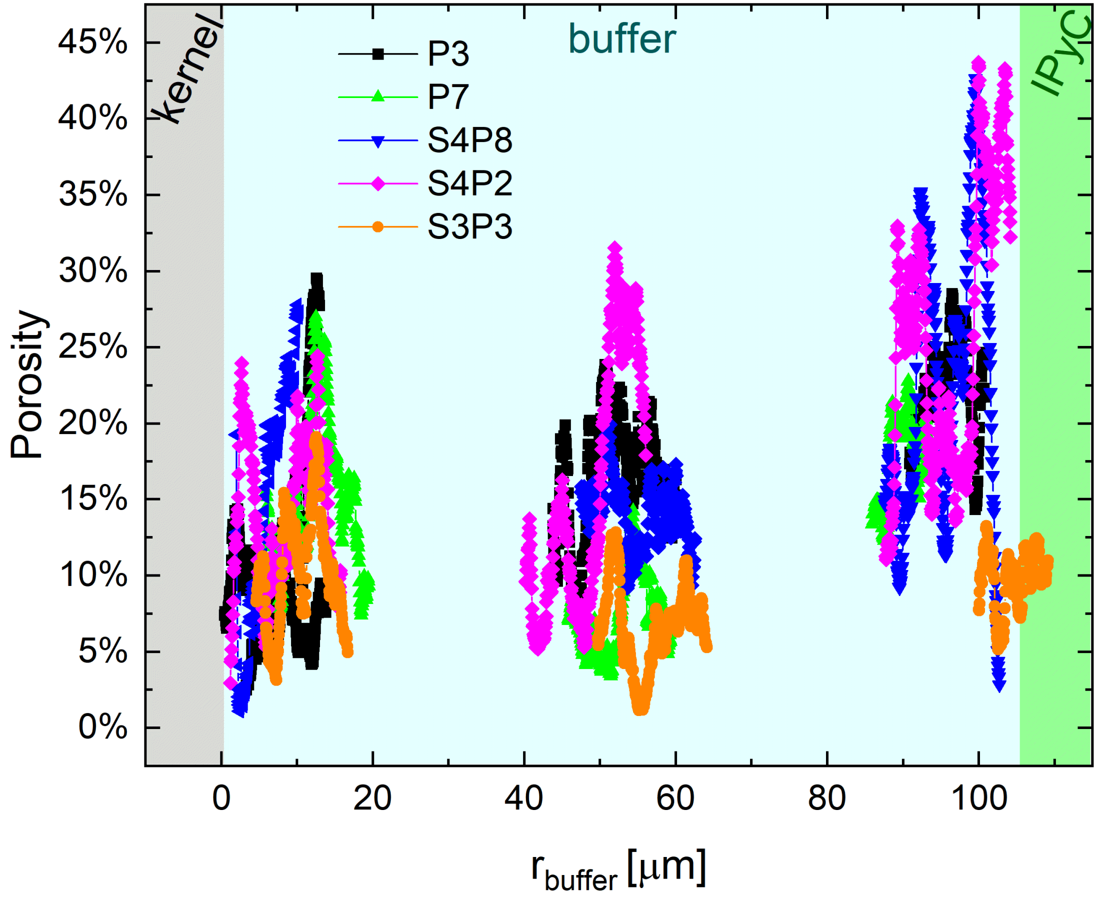


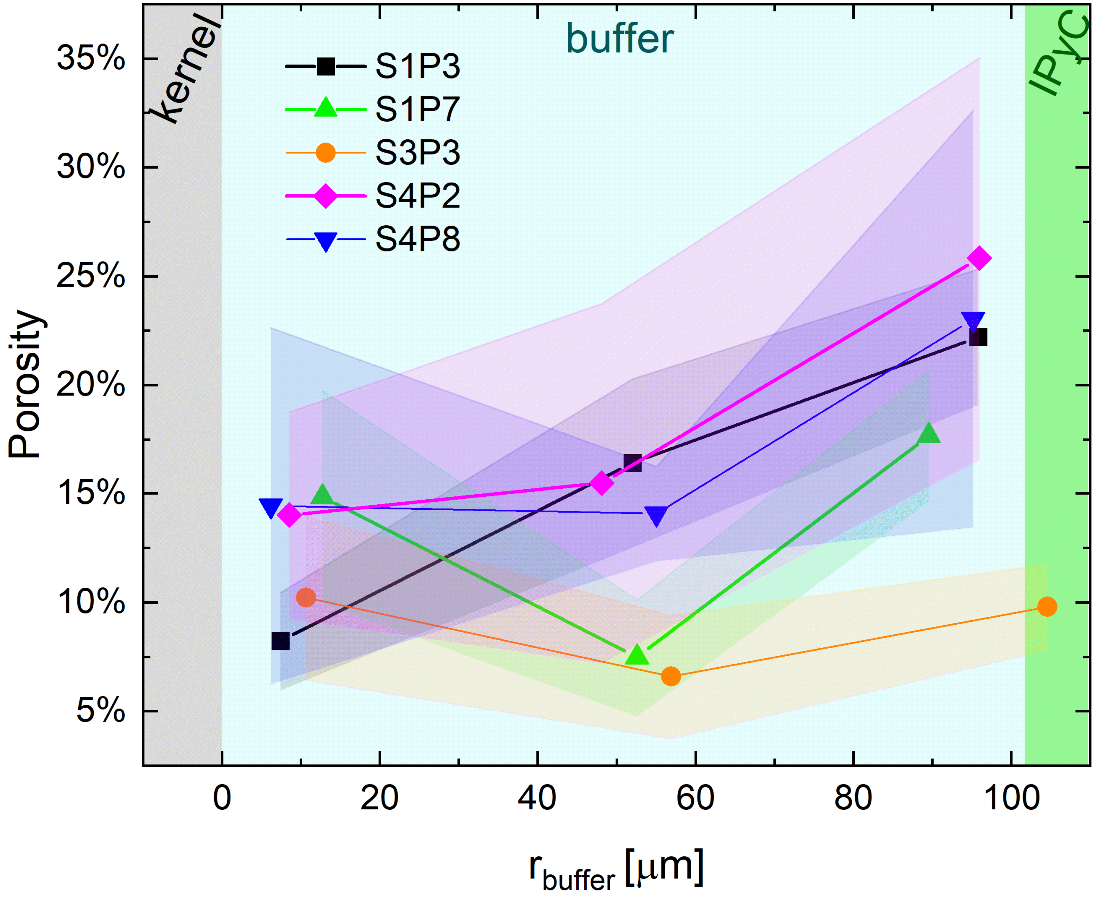


Buffer consists of low density pyrocarbon with 58% theoretical density and randomly distributed pores that give a total porosity of 14%.

Inter-particle (and radial direction) differences

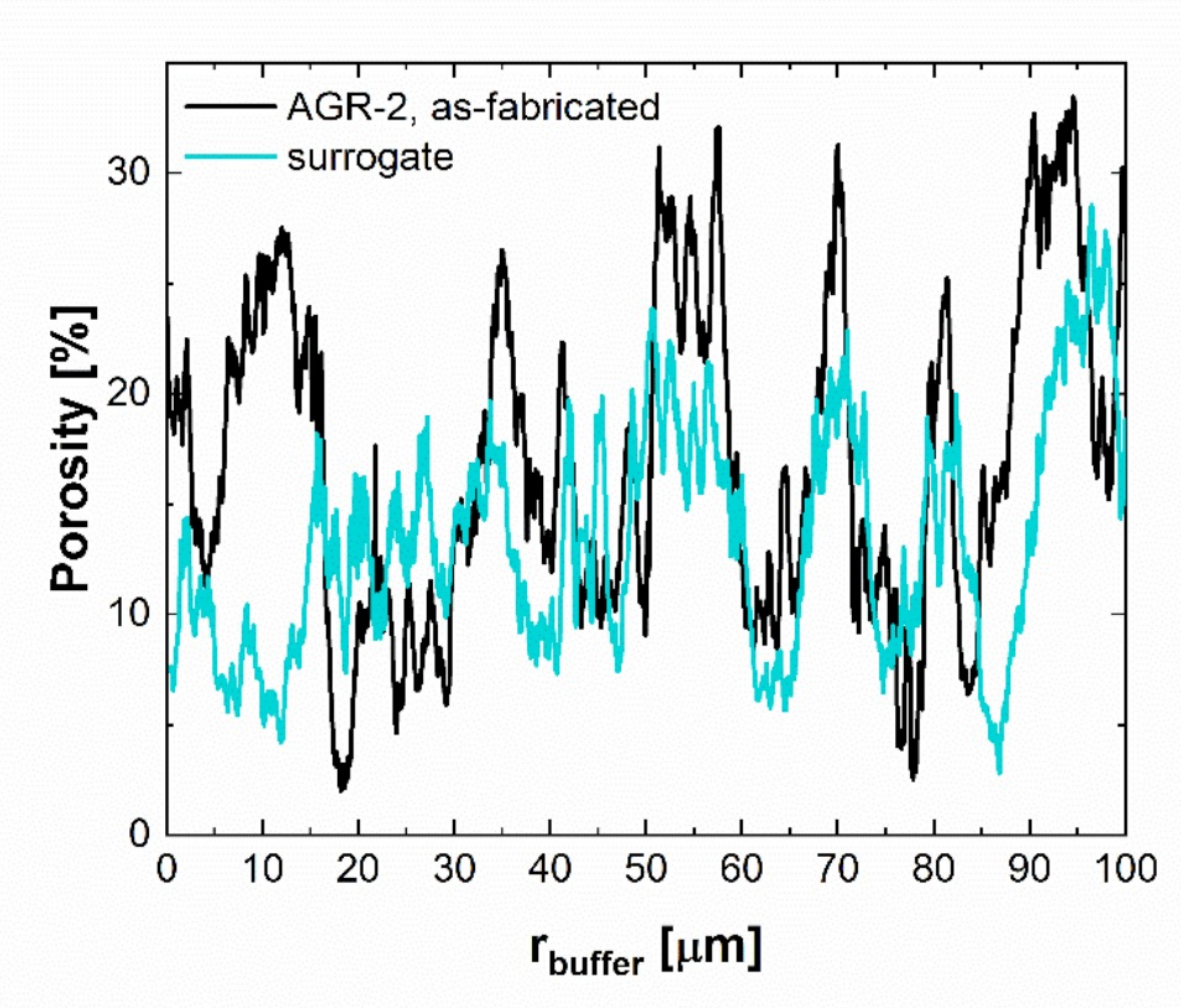
- The radial porosity distribution varies upon the radial direction and from particle to particle
- However, the average porosity, magnitude of local fluctuation and radial gradient are similar.
- The radial increase of porosity is consistent with the fabrication procedure.
- More characterizations (which are expensive) are desired.





Difference between surrogate and AGR-2 TRISO particle

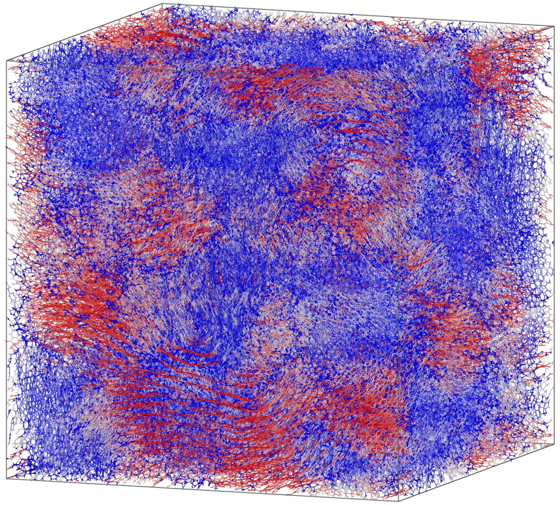
- **Similar average porosity.**
- **Similar radial distribution.**
- **Larger fluctuation in the AGR-2 particle (?)**



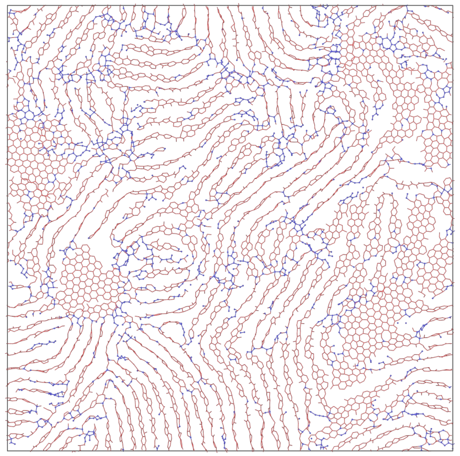
Atomic structure of low density pyrocarbon

- Molecular dynamics simulations show that the low-density pyrocarbon is non-textured, containing randomly oriented graphite crystallites. Three parameters, **density**, and **in-plane** and **out-of-plane crystallite sizes**, are extracted from correlation analysis for describing the matrix microstructure.

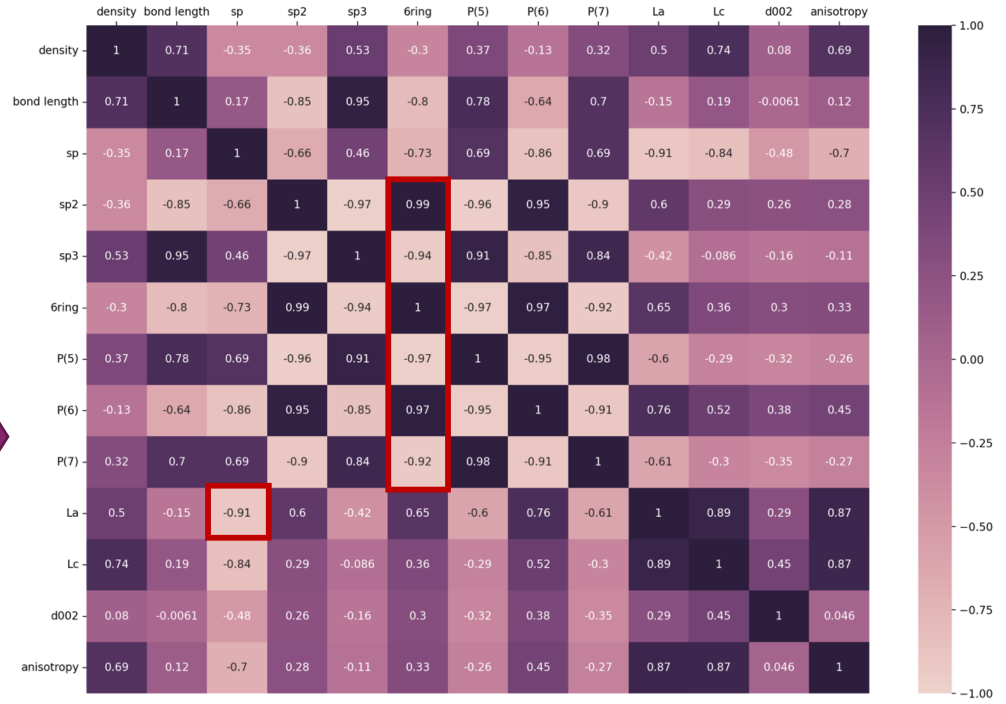
A 3D cell with atoms colored by atomic orientation



A slice in the cell with graphite-like atoms (red, sp2 bonded and in 6-member rings) and defect (others)



Correlations among descriptors

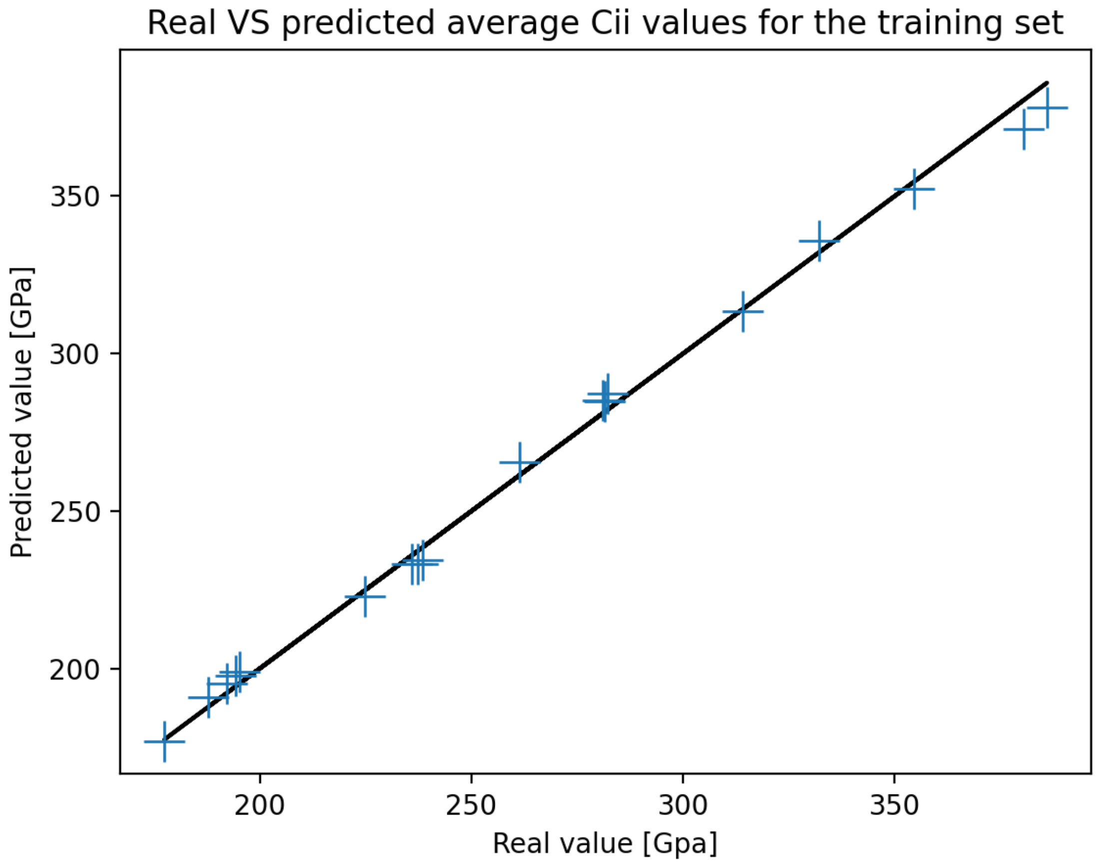


N. A. Marks, PRB 65 (2000), 075411
C. de Tomas et al. / Carbon 109 (2016) 681-693.

C. de Tomas et al. / Carbon 155 (2019) 624e634
http://www.carbonpotentials.org/

Microstructure descriptors of buffer

- Porosity parameters
  - Average porosity
  - Radial gradient
  - Local fluctuation
  - Others?
- Matrix Parameters
  - Density
  - In-plane crystallite size L_a
  - Out-of-plane crystallite size L_c
  - Others?



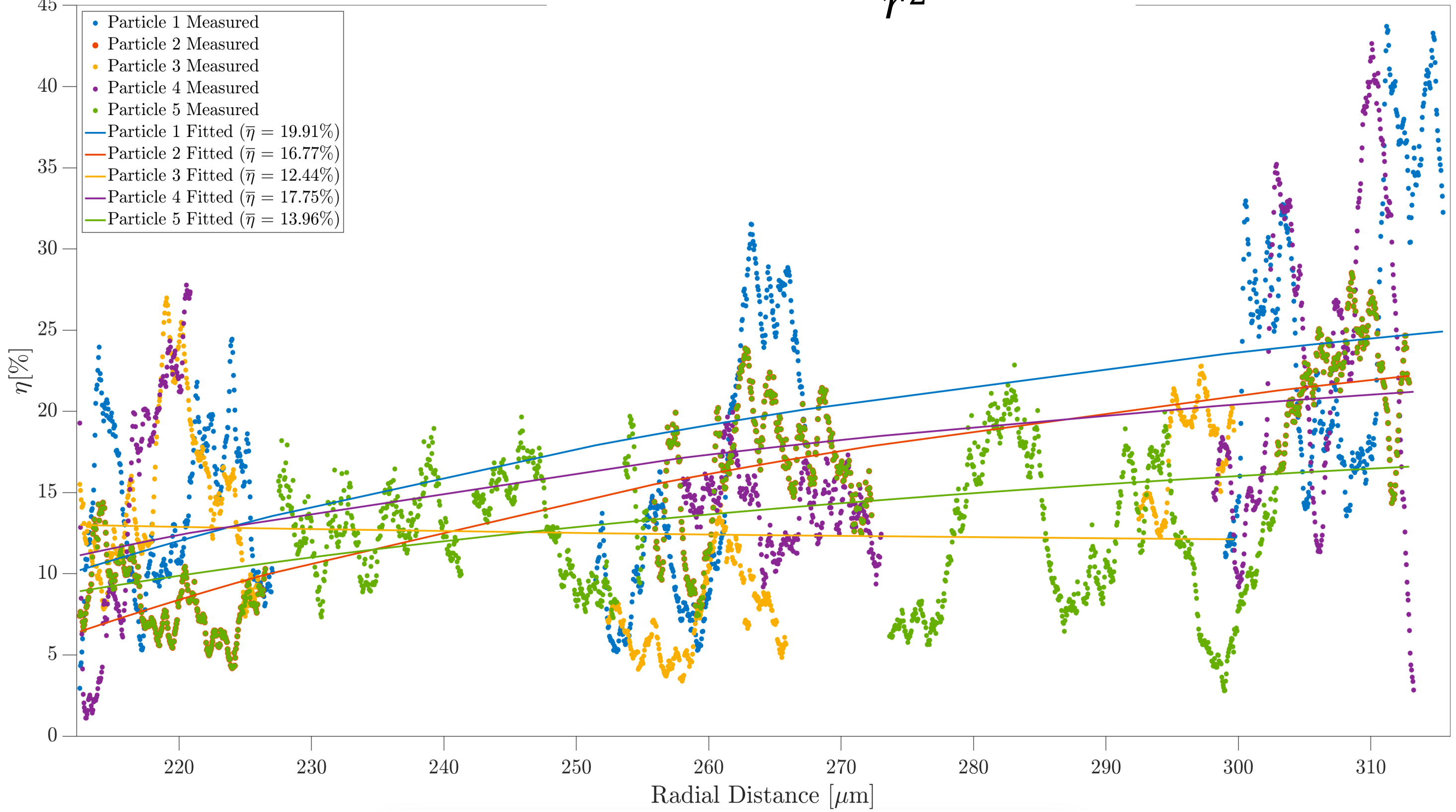
How does buffer porosity affect its fracture behavior?

- **Statistical BISON simulations of buffer fracture initiation and propagation**
  - *Masri et al. The role of heterogeneous porosity distribution on buffer fracture behavior in TRISO fuel particles (in preparation)*

Representation of buffer porosity in BISON

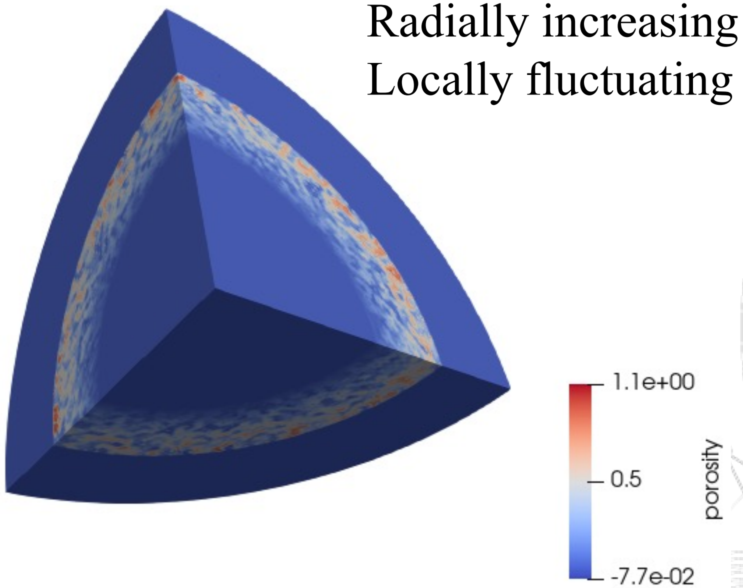
- Buffer porosity is randomly sampled with three parameters: **average porosity (A), radial gradient (B),** and **local fluctuation (Cξ)**, by fitting the experimental results using the below equation.
- The function is selected based on the assumption of constant mass deposition rate per unit radial length.
- Both elastic moduli and fracture stress are made dependent on porosity.

$$\eta(r) = A(1 - B\frac{r_0^2}{r^2})(1 + C\xi)$$



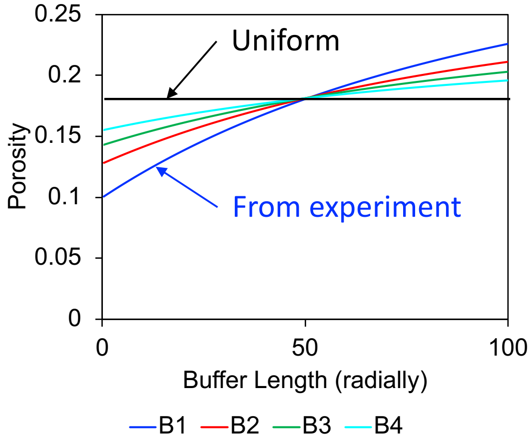
BISON: Williamson, R. L. et al., *J. Nucl. Mater.* 2012, *423* (1–3), 149–163.

Radially increasing
Locally fluctuating

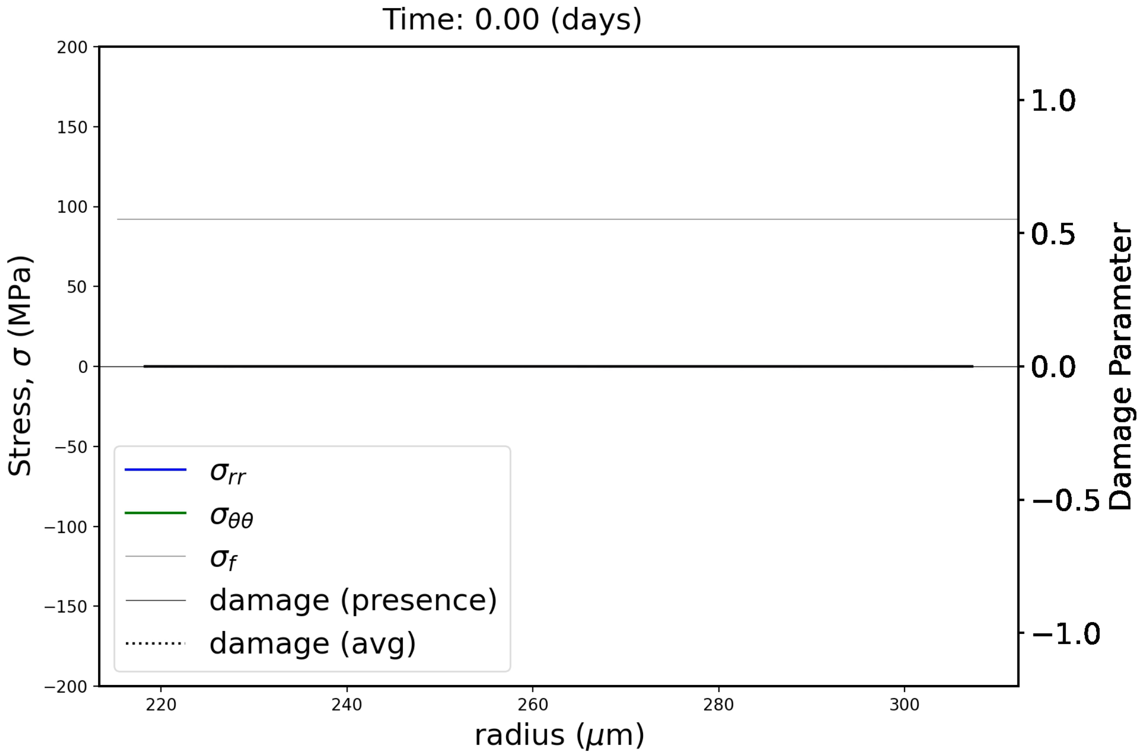


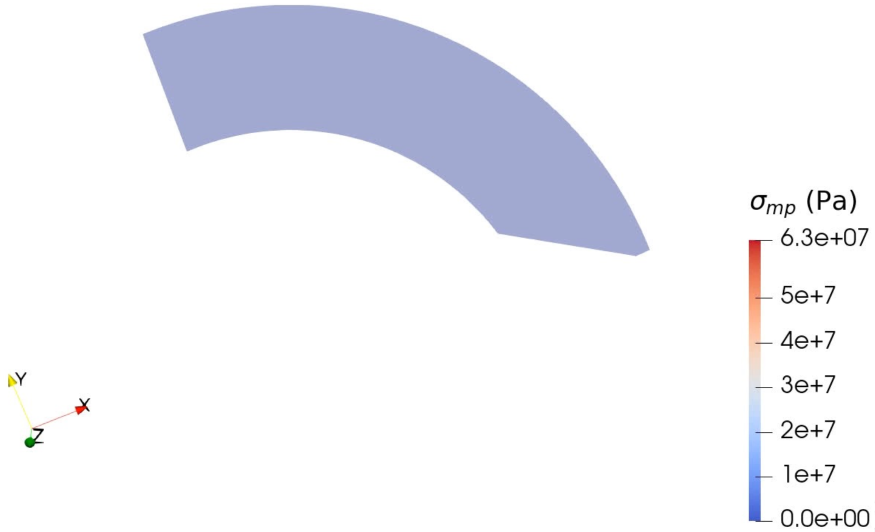
BISON simulations of buffer fraction: uniform porosity

- Both elastic moduli and fracture stress are uniform
- Fracture is caused by **tangential stress**, not radial stress
- Fracture initiates from **kernel/buffer** interface
- Fracture propagates **radially**

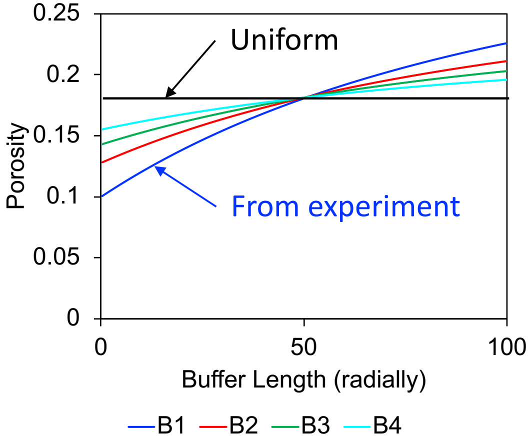


Color: stress
Black: crack

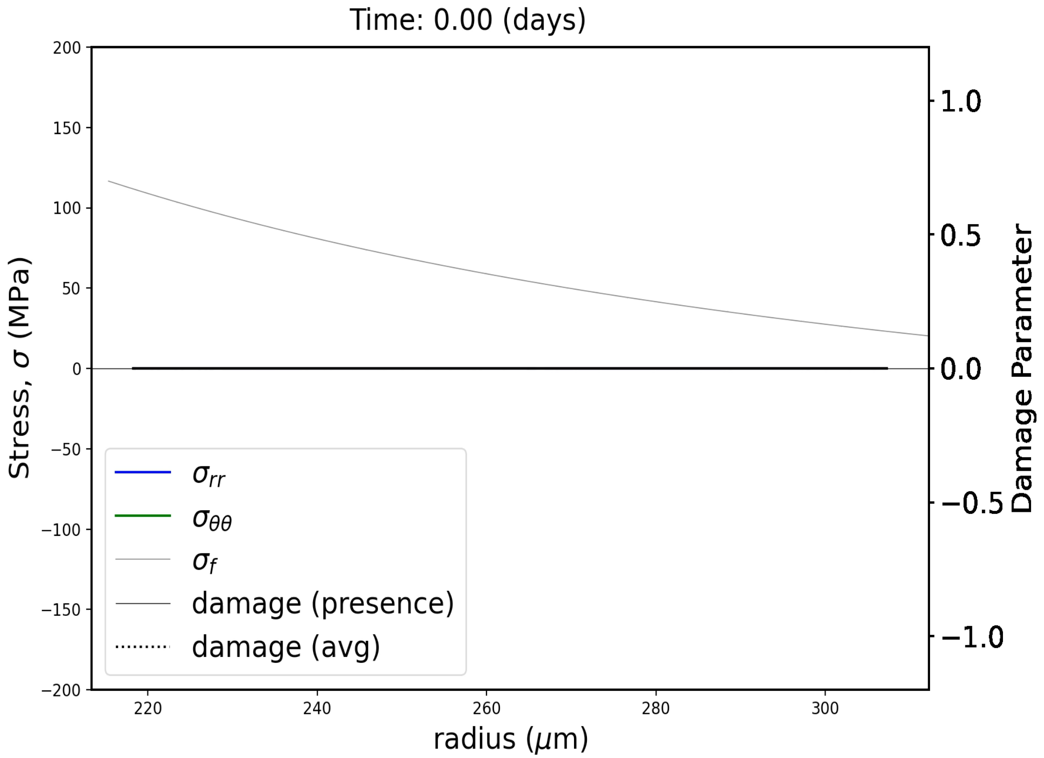


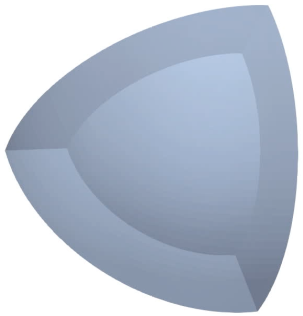


BISON simulations of buffer fraction: radially increasing porosity



- Both elastic moduli and fracture stress decrease radially
- Fracture is caused by **<u>tangential stress</u>**, not radial stress
- Fracture initiates from **<u>buffer/IPyC interface</u>**
- Radial crack connects with each other along tangential direction (**<u>tearing</u>**), instead of propagating radially





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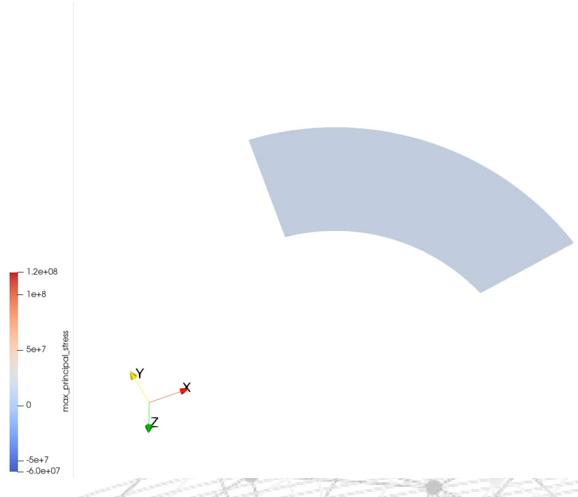
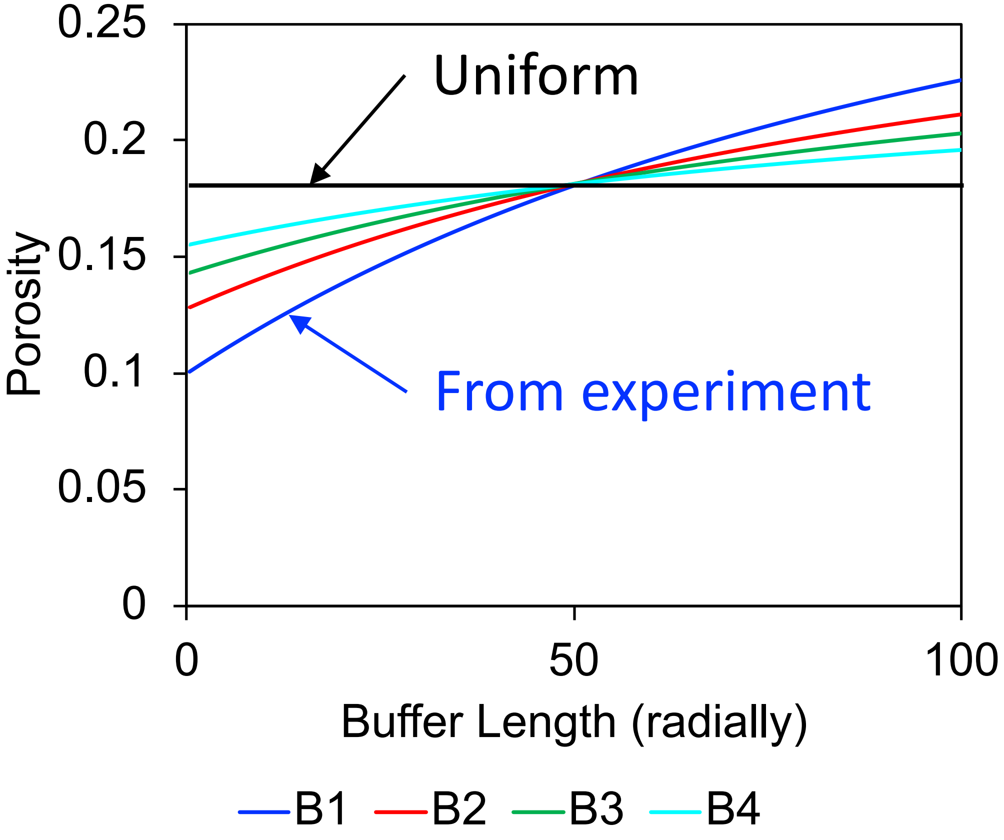
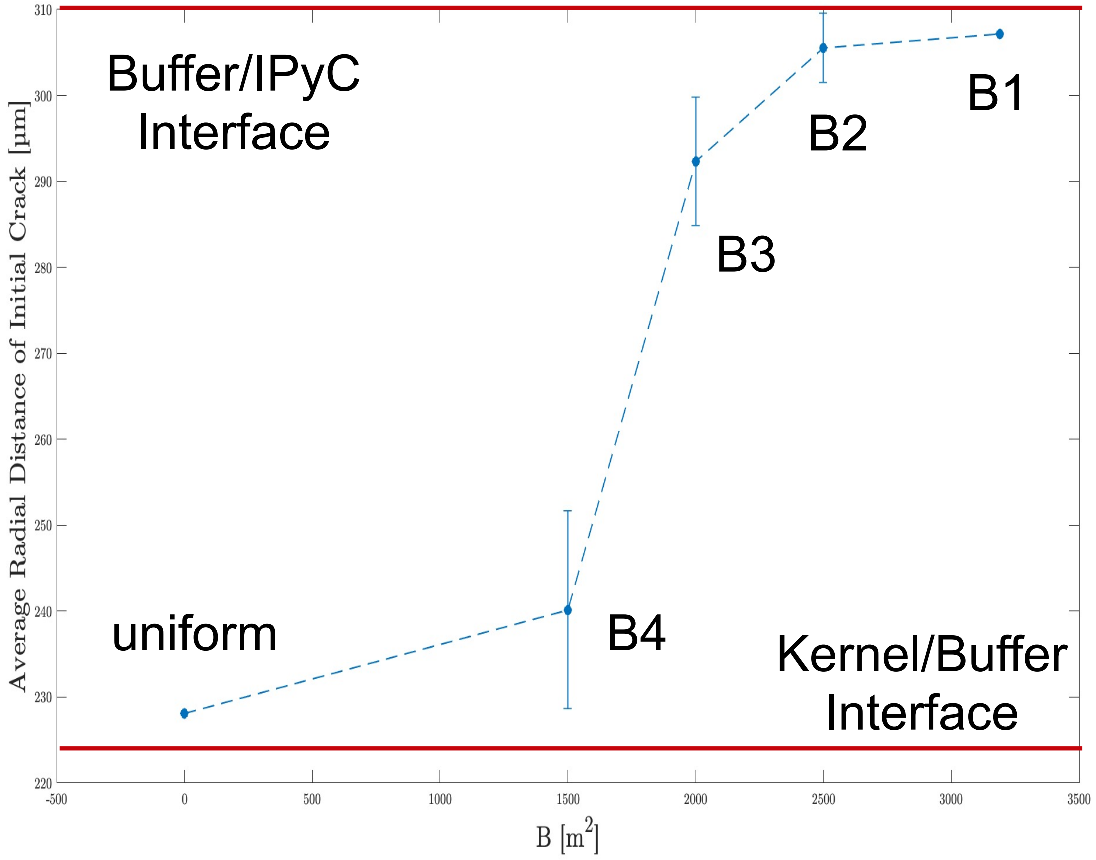


BISON simulations of buffer fracture: fracture initiation

- The fracture initiation point increases radially with the parameter B, which describes the rate of radial porosity increasement, implying the possibility of tailoring fracture by controlling porosity distribution.
- Local fluctuation in porosity induces some fluctuation in fracture initiation point.





Irradiation induced changed in porosity and matrix microstructure

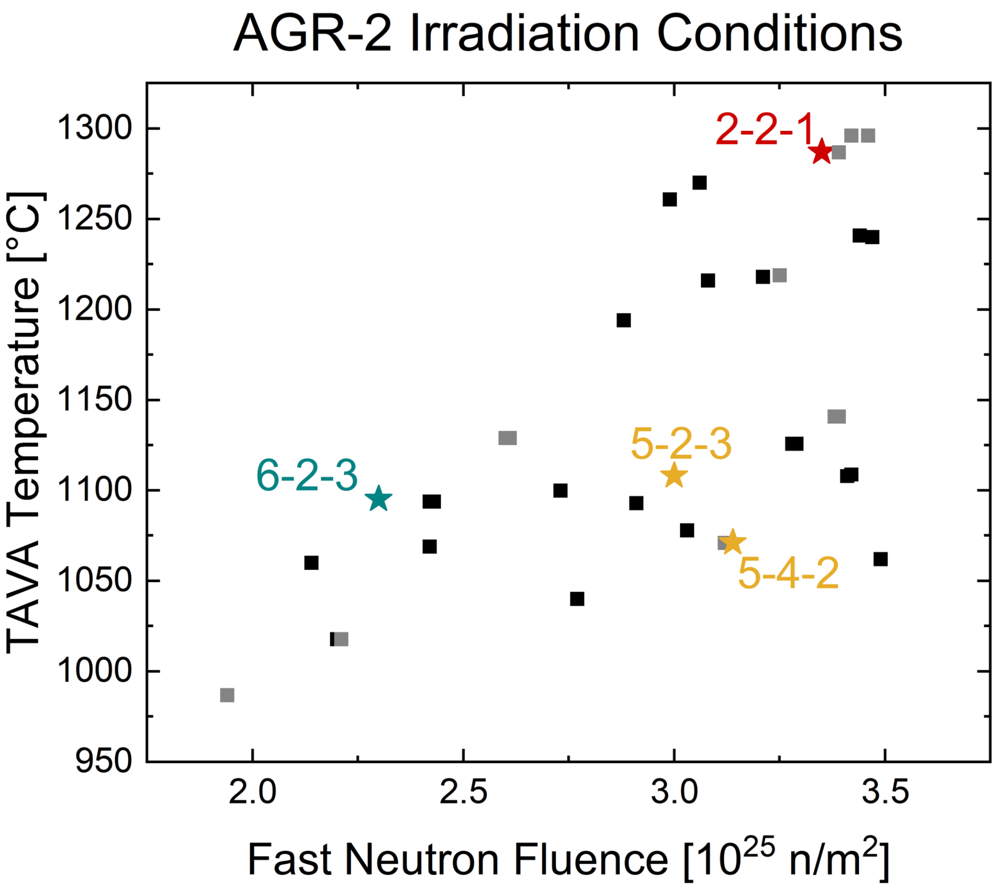
FIB-SEM characterization of buffer porosity

- *C. Griesbach, C. McKinney, Y. Zhang, T. Gerczak, R. Thevamaran, Irradiation-induced changes in the porous buffer microstructure of TRISO nuclear fuel particles (in preparation)*

- *C. Griesbach, J.D. Arregui Mena, E. Lopez, Y. Zhang, T. Gerczak, R. Thevamaran, Irradiation-induced nanostructural changes in porous pyrocarbon (in preparation)*

Samples and irradiation condition

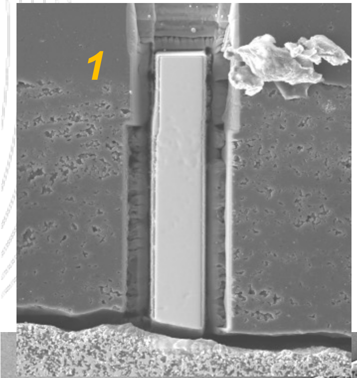
- Various irradiation conditions consider in terms of temperature and neutron fluence.
- **Porosity**, **solid fission products**, **densification**, and **fracture** are characterized.

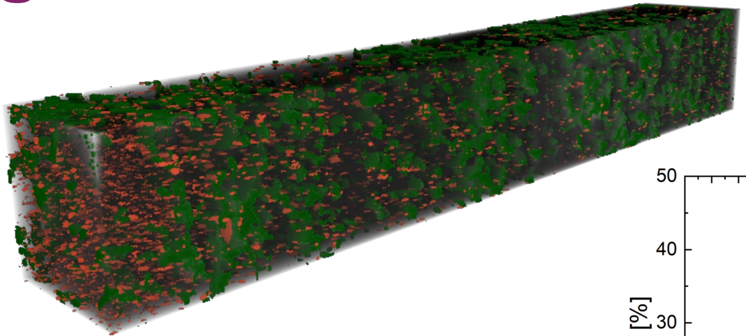


5 – 4 – 2
capsule compact stack

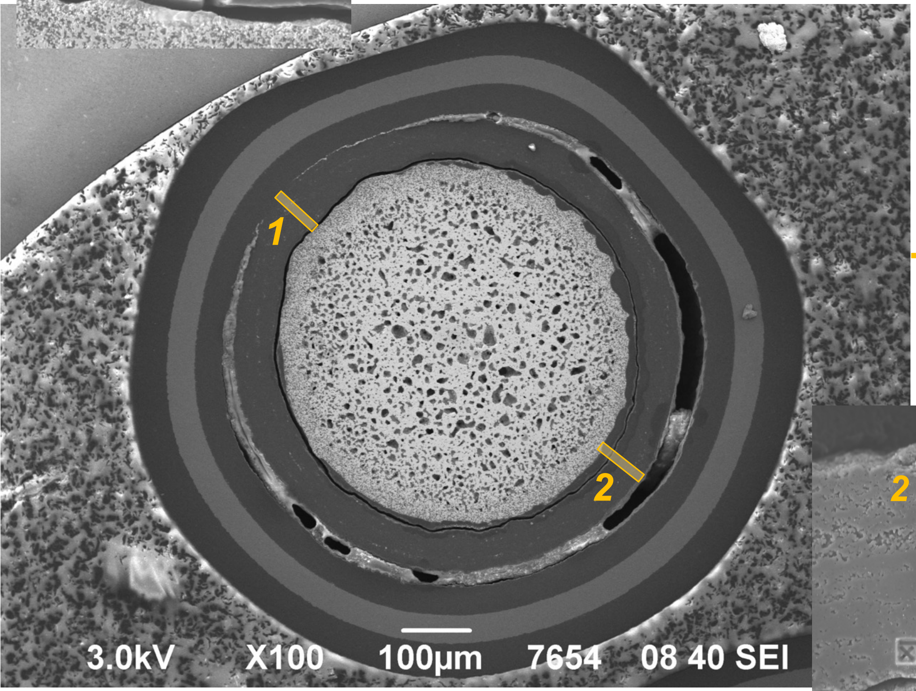
Compact	Mount	Particle	Ag (M_part/M_avg)	Temperature [°C]	Fluence [10^{25} n/m^2]	#full buffer scans	Raman	TEM
221	MM-D67	RS43	1.12	1287	3.35	2	X	X
221	MM-D67	RS46	1.8	1287	3.35	2		X
221	MM-D68	RS39	0.83	1287	3.35	1		
523	MM-D12	RS11	0.48	1108	3.00	1		X
523	MM-D12	RS28	0.43	1108	3.00	1		
542	MM-D55	RS25	1.59	1071	3.14	2	X	
623	MM-D69	RS18	1.87	1095	2.30	2		X
623	MM-D69	RS07	1.72	1095	2.30	1		X
623	MM-D70	RS35	0.81	1095	2.30	1	X	X
As fabricated AGR-2			N/A			1	X	X

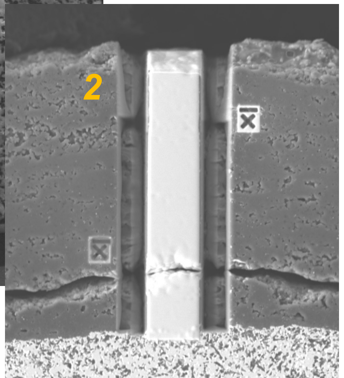
Example: Capsule 2, high temperature and high fluence

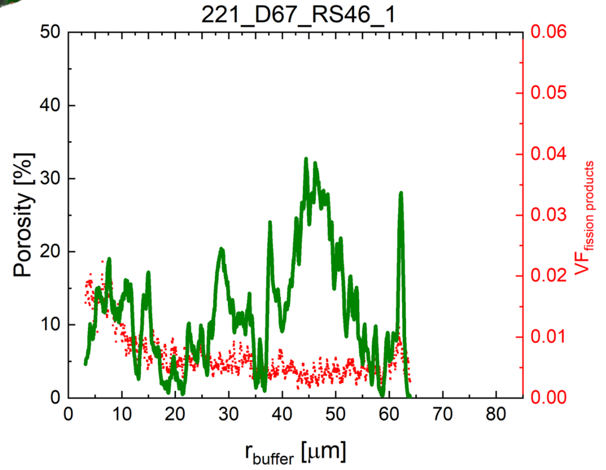


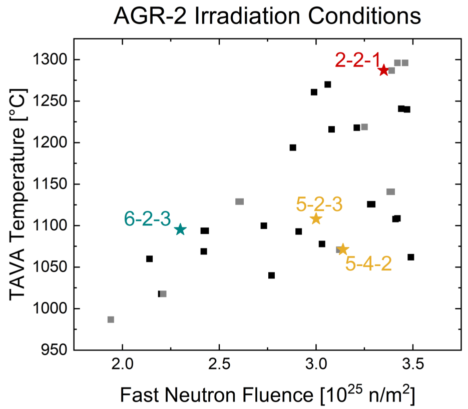


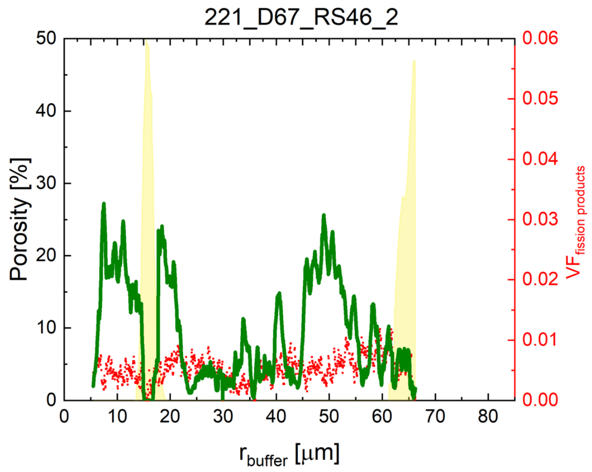
D67-RS46

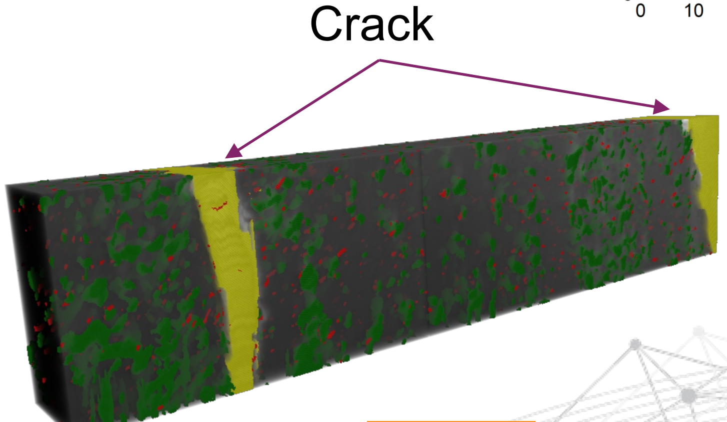






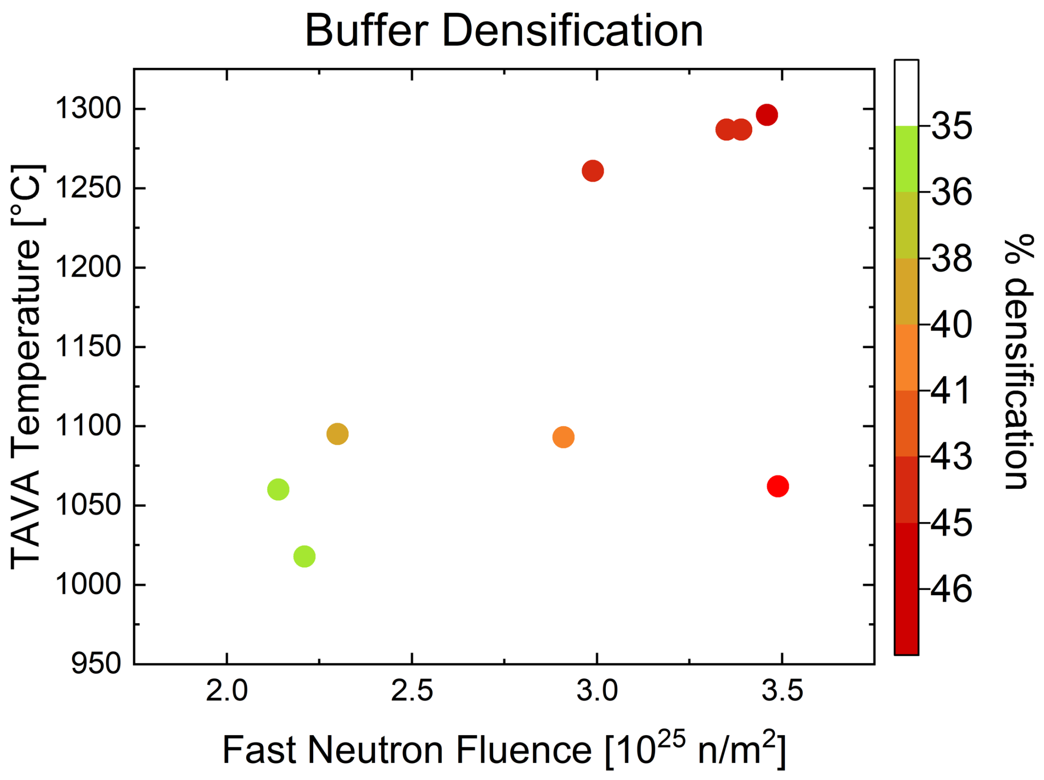


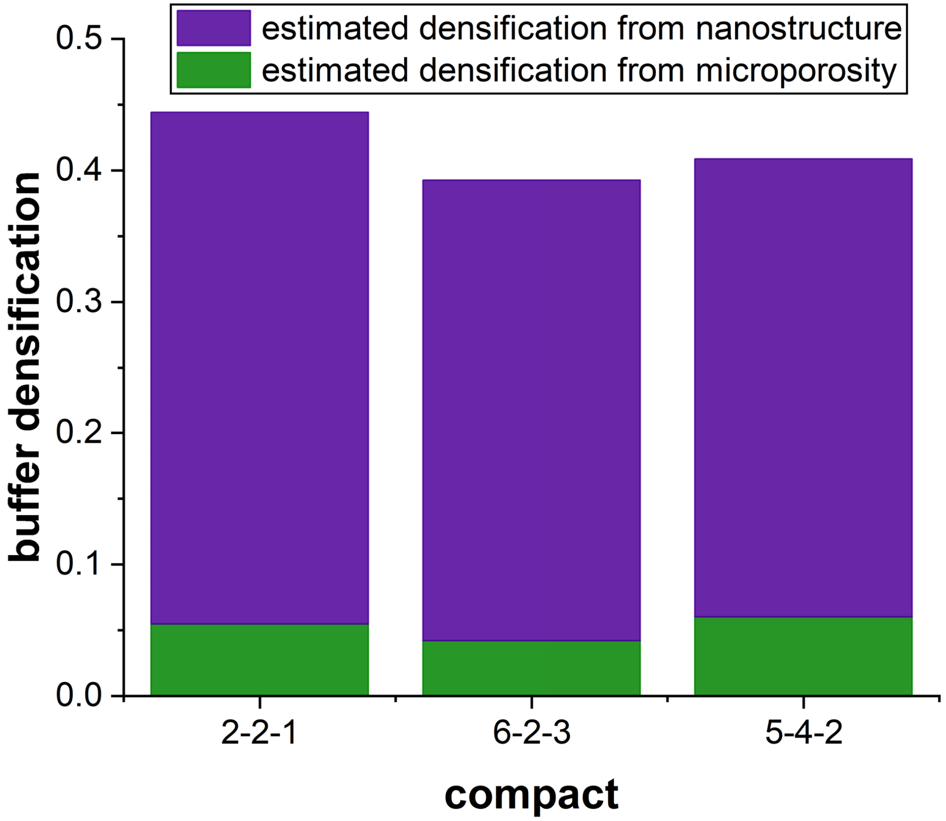




Buffer densification: Change in matrix & porosity

- Substantial densification that correlate positively with fluence and temperature has been observed and found to be dominated by change in matrix microstructure.
- Irradiation changes the average porosity, its local fluctuation and radial distribution.
- The matrix has experienced substantial graphitization as suggested by XRD and Raman analyses.

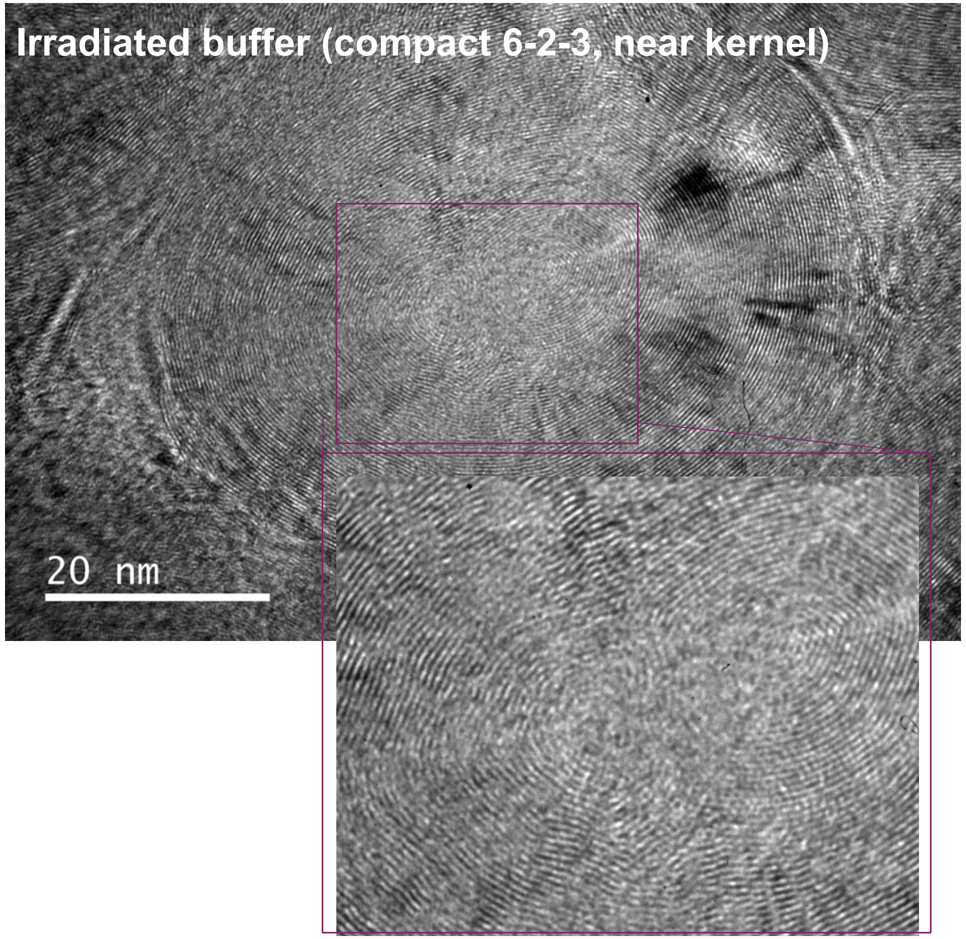




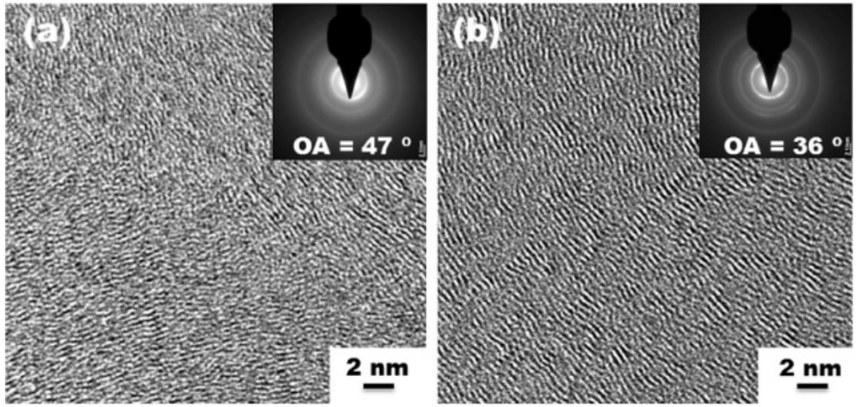
IES

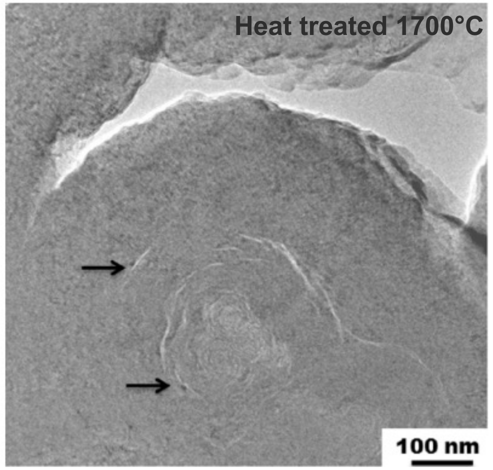
Change in matrix microstructure: formation of unique graphitic onion-like nanofeatures from TEM

- Formation of onion-like nanostructure (left) characterized by TEM. The feature is similar to that has been observed in heat-treated non-textured pyrocarbon (right).



Heat-treated pyrocarbon





Cancino-Trejo et al, Carbon (2016)

Super-elasticity of unirradiated buffer pyrocarbon

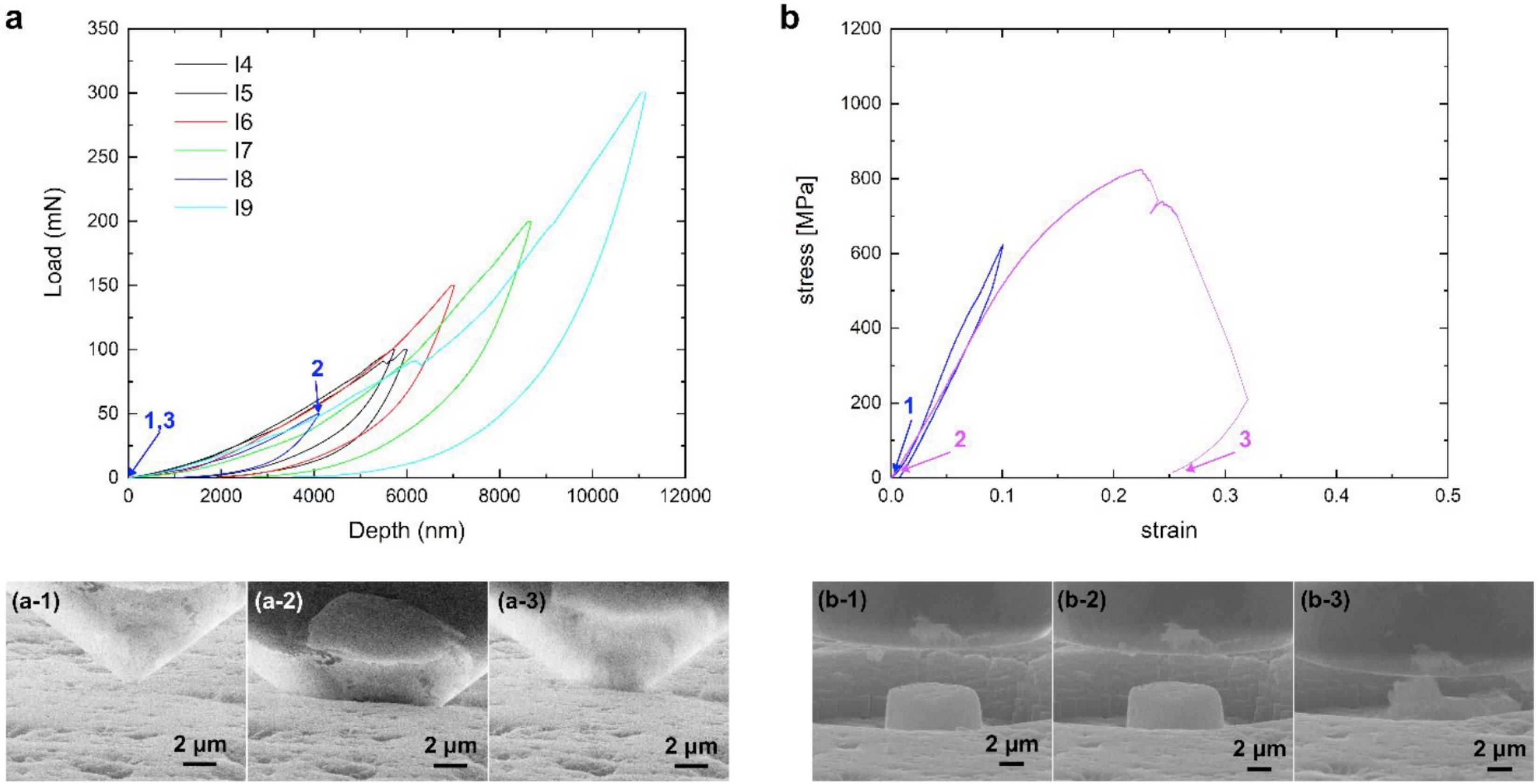
Nanoindentation and micro-compression

- *C. Griesbach, T. Gerczak, Y. Zhang, R. Thevamaran, Super elasticity and anisotropic mechanical response in porous pyrocarbon (in preparation)*

Super-elasticity revealed by nanoindentation and nano-pillar compression

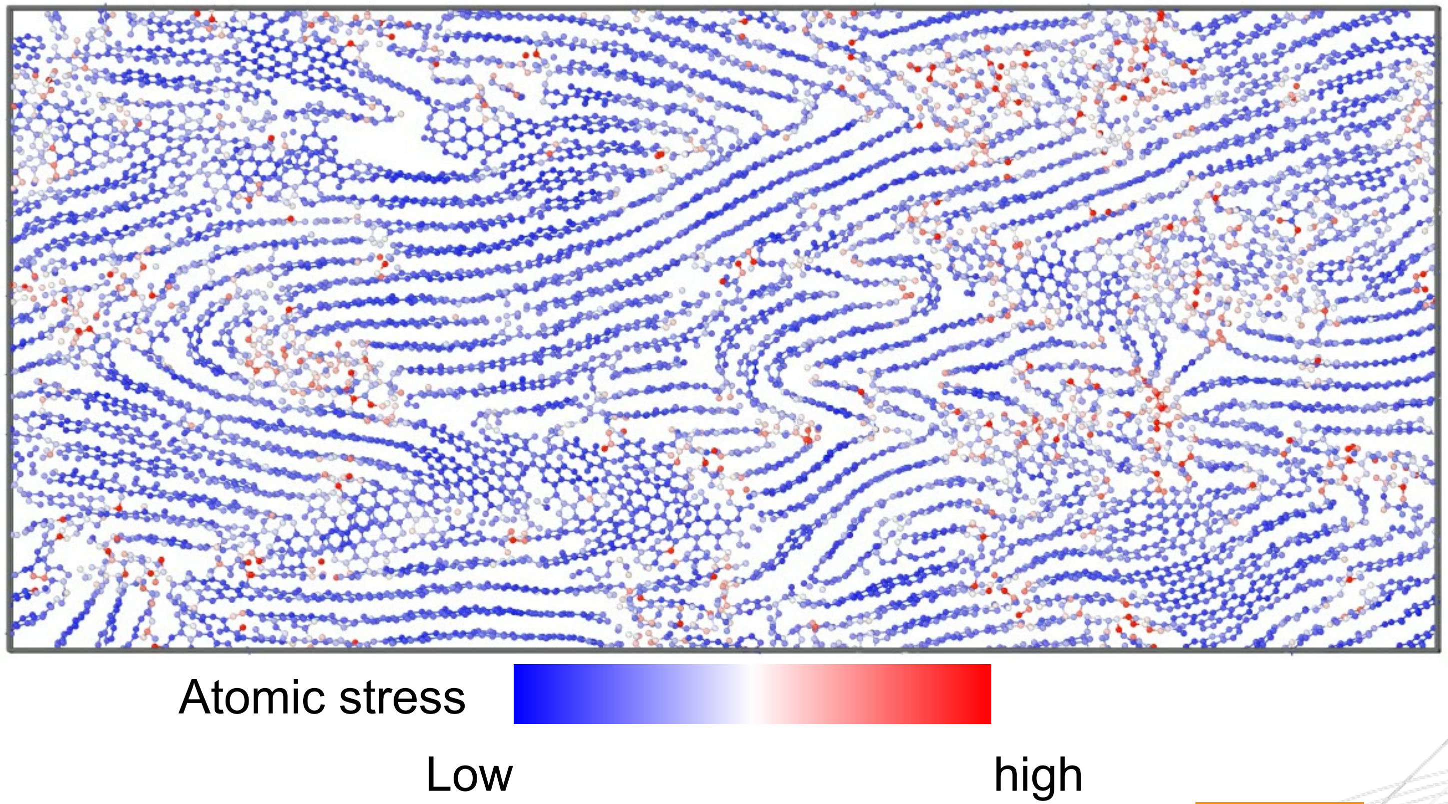
- No failure or plasticity observed during in-situ nanoindentation.
- Brittle failure observed until 30% strain under micro-compression
- Radially compressed micro-pillar show lower yield strength than tangentially compressed.

(a) force-displacement curves from in situ SEM nanoindentation experiments with SEM images at select points during deformation in (a-1) – (a-3) , (b) stress-strain curves from in situ SEM micro-compression experiments on the same sample compressed to ~10% strain without yielding (blue curve) and to ~30% strain exhibiting brittle failure; SEM images at select points during deformation in (b-1) – (b-3).



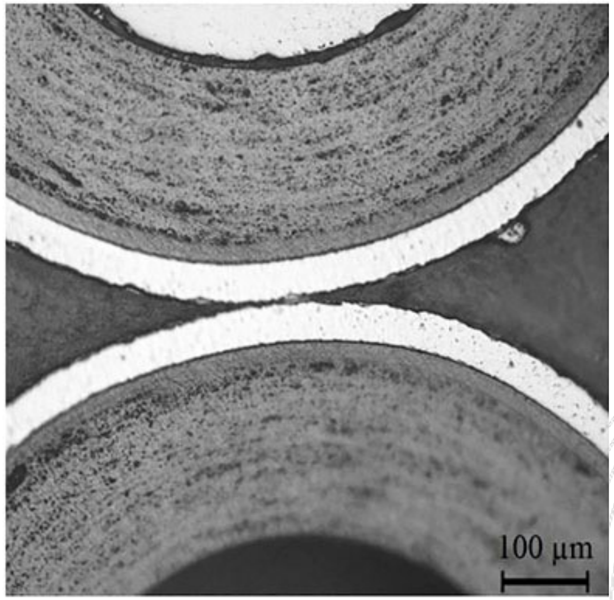
The super-elasticity is related to the unique way of non-textured pyrocarbon for accommodating strain

- Non-textured pyrocarbon can sustain large tensile and compressive strain.
- The stress and plastic deformation (bond breaking and forming) are most accommodated by the disordered regions between ‘crystallites’ (graphite-like regions).
- The crystallites grows and align with each other along the loading direction, enhancing texture.



Outlook

- **Can we optimize the fracture behavior by controlling the initial buffer microstructure and porosity distribution?**
  - ➢ **Anisotropic porosity distribution and mechanical properties**
  - ➢ **Atomic-scale mechanisms responsible for irradiation induced change in matrix microstructure and porosity distribution, which determine the transient mechanical properties**
  - ➢ **Progression of buffer fracture into SiC layer**



- **A Letter Of Intent (LOI) has been submitted for phase-II continuation.**

Summary

- Buffer consists of a low density pyrocarbon matrix (~58% theoretical density) and randomly distributed pores with a total volume of about 14%. The porosity increase radially and fluctuate locally.
- The radial porosity distribution strongly affects the fracture behavior of buffer.
- Irradiation causes significant densification and changes the total porosity, the porosity distribution, and the matrix microstructure. The densification is dominated by the graphitization of the pyrocarbon matrix.
- The unirradiated buffer pyrocarbon exhibits super-elasticity under compression. The strain is accommodated by bending and reorientation of graphite crystallites.
- <u>The findings suggest the possibility of optimizing the fracture behavior by controlling the initial buffer microstructure.</u>

Thank you!

Project information: Statistical modeling of the effect of microstructural heterogeneity on the irradiation behavior of TRISO fuel buffer layer

- PI: Yongfeng Zhang, University of Wisconsin Madison (UW)
- Co-PIs: Ramathasan Thevamaran (UW), Karim Ahmed (TAMU), Tyler Gerczak (ORNL), Wen Jiang (INL)
- Funding: $800,000
- Period: 10/1/2020 – 9/30/2023 (with NCE to 6/30/2024)
- Milestones: (Accomplished in green; M2 in bold)
    1. (M3) TRISO buffer pyrocarbon sample delivery and preparation memo, 9/30/2021
    2. **(M2) Year 1 Annual Progress Report, 9/30/2021**
    3. (M3) Phase field modeling of pyrocarbon fracture with pores, 9/30/2022
    4. (M3) BISON model development for TRISO particle, 9/30/2022
    5. **(M2) Year 2 Annual Progress Report, 9/30/2022**
    6. (M3) Pore structure characterization in TRISO buffer, 12/31/2022
    7. (M3) BISON statistical modeling of buffer tearing, 6/30/2024
    8. (M3) Mechanical property measurement in TRISO buffer, 6/30/2024
    9. **(M2) Final Project Report, 9/30/2024**