3D analysis of buffer porosity before and after irradiation

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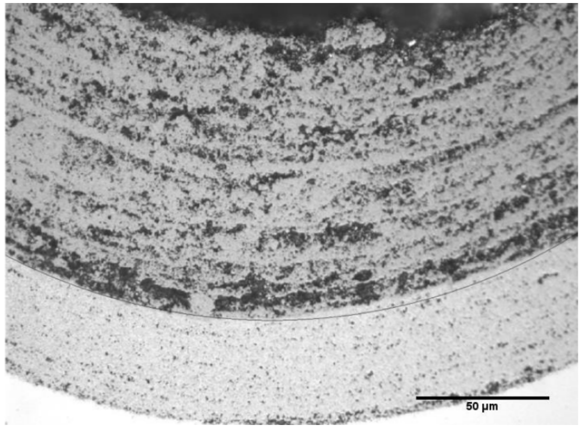
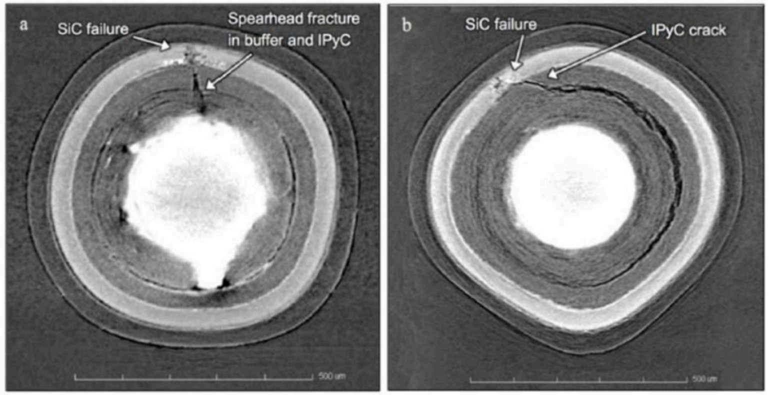
Nuclear Fuel Development Section

Oak Ridge National Laboratory

Background

- Densification of buffer under irradiation generally causes partial or full tearing of the buffer from the IPyC.
- There is some indication from PIE that fractures beginning in the buffer may propagate into the IPyC and expose the SiC layer, which may lead to concentrated attack of fission products in UCO TRISO.
- The nature of the buffer and the transition region into the IPyC possibly influence the fracture propagation into IPyC.
- Work has been done already to study the buffer, but serial sectioning presents a higher resolution method for examining pore structure.
  - XCT has been used to study pores in the buffer (Bari et. al.), but has lower resolution than serial sectioning
  - Schumacher showed that samples with a greater porosity were able to tolerate only a lower fraction load and flexure strength, hardness value, and modulus, meaning it took a lower load to fracture.





Images from top to bottom:

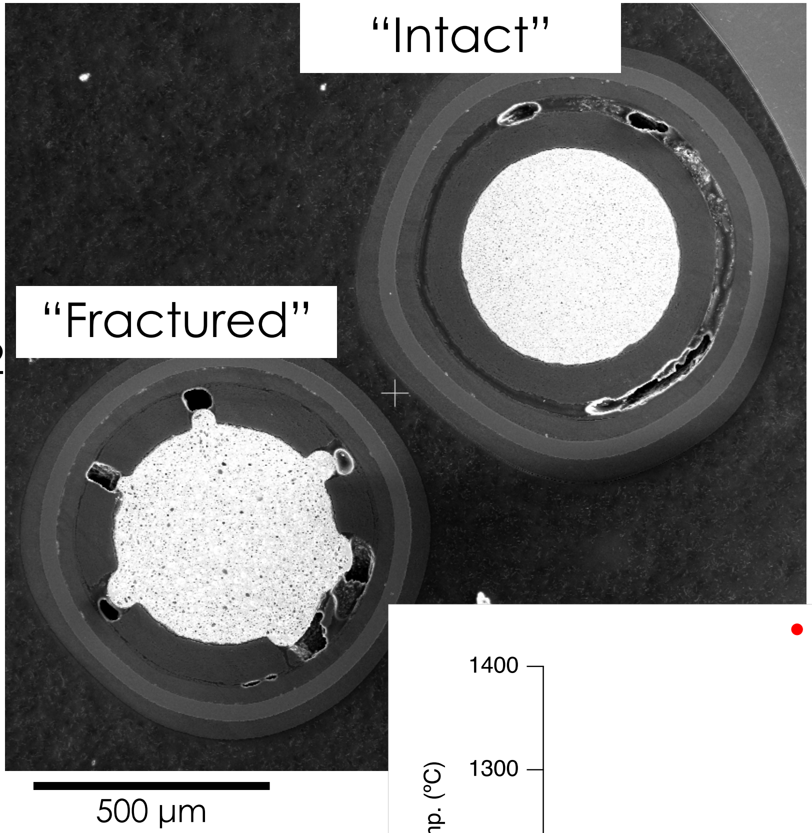
Schumacher, Austin, Master's Thesis, University of Tennessee, 2019. https://trace.tennessee.edu/utk_gradthes/5667

Bari, K., C. Osarinmwian, E. López-Honorato and T. J. Abram (2013). Nucl. Eng. Des. 265: 668-674.

Hunn, J. D., C. A. Baldwin, T. J. Gerczak, F. C. Montgomery, R. N. Morris, C. M. Silva, P. A. Demkowicz, J. M. Harp and S. A. Ploger (2016). Nucl. Eng. Des. 306: 36-46.

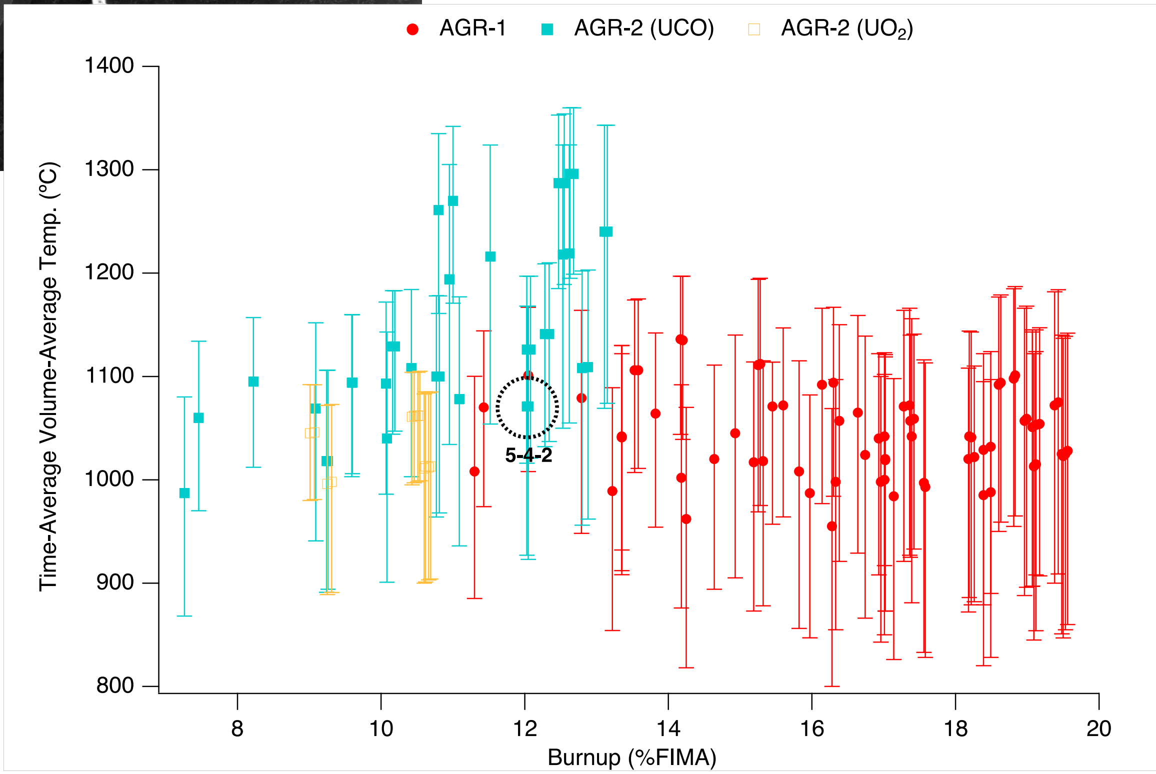
Sample Pedigree

- Unirradiated
  - Deconsolidated particle from an AGR-2 compact from the same AGR-2 compact lot used in the irradiation experiment
- Irradiated (AGR-2 Compact 5-4-2)
  - "Intact" Buffer
  - "Fractured" Buffer
  - Allows exploration of densification for different buffer responses
- 5-4-2 experienced midrange irradiation conditions
  - 12.03% FIMA
  - 3.14 x 10^{25} n/m^2 Neutron Fast Fluence
  - 1071 °C Time-Average Volume-Averaged Temperature



Data for plot from:
B.P. Collin, "AGR-2 Irradiation Test Final As-Run Report," INL/EXT-14-32277
B.P. Collin, "AGR-1 Irradiation Test Final As-Run Report," INL/EXT-10-18097

Error bars represent the TAVA minimum and maximum for each compact.



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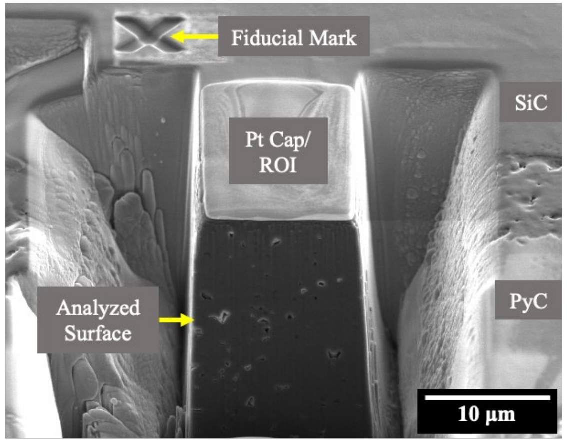
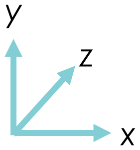
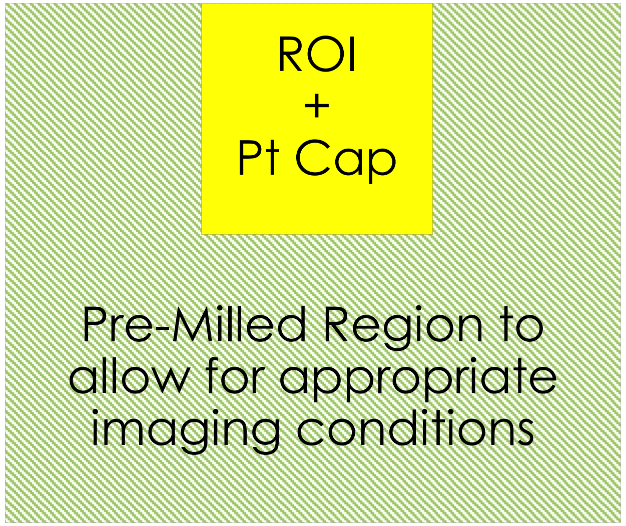
3D reconstruction analysis provides a quantitative method to describe the buffer

Sample Preparation for sectioning

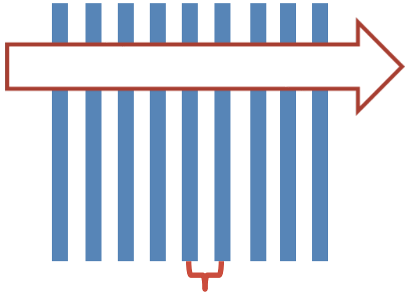




Top schema

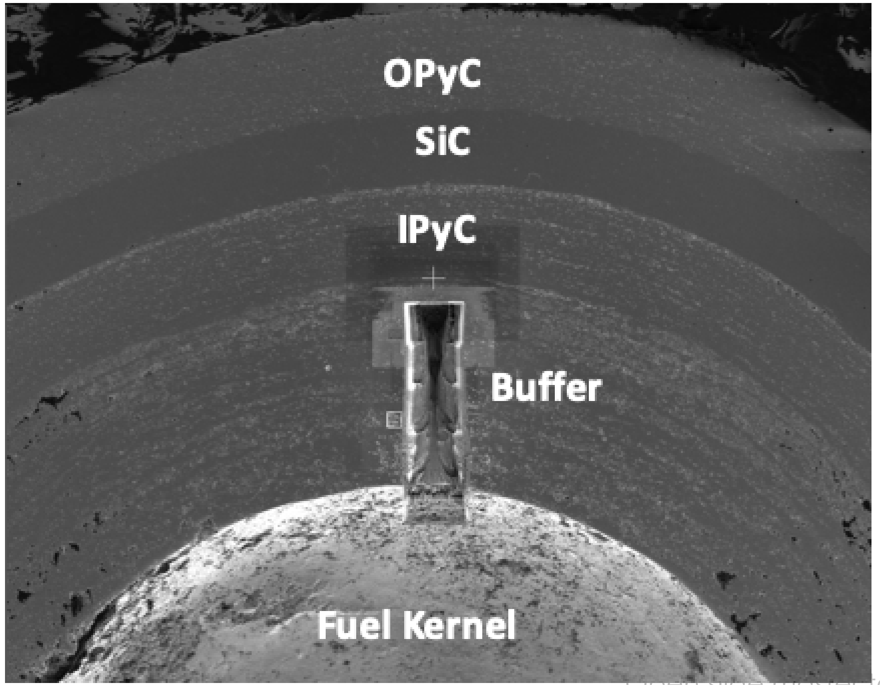


Slice schema



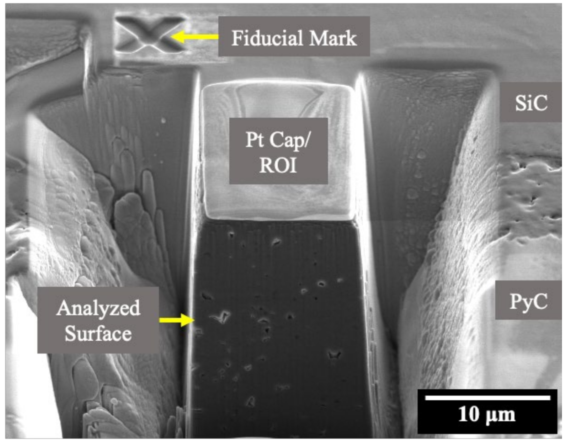
Spacing of 25-30 nm between each slice

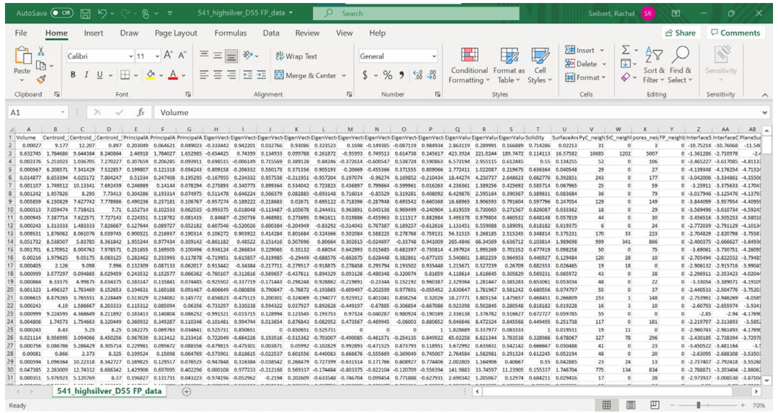
- Use of a focused ion beam to mill material of x thickness
- Image after each milling "slice" using scanning electron microscopy
- Possibility to couple with analytical techniques such as energy dispersive x-ray spectroscopy (EDX) or electron backscatter diffraction (EBSD)



Data Analysis

- Data collected contains image stacks of 300-600 images
- Images are segmented into regions (Pores, Buffer) using a combination of thresholding, manual segmentation, and some machine learning
  - Avizo Amira (Thermo Fisher), Dragonfly (The Objects – work initialized by Claire Griesbach, U. Wisconsin)
- After segmentation, data mining takes place to gather information
  - Each feature is isolated and identified, and unique statistics are defined
  - Spreadsheets produced through materials statistics and analysis via Avizo Amira (Thermo Fisher)
- There is a LOT of data to mine, and identifying what is crucial is an important (and difficult) process



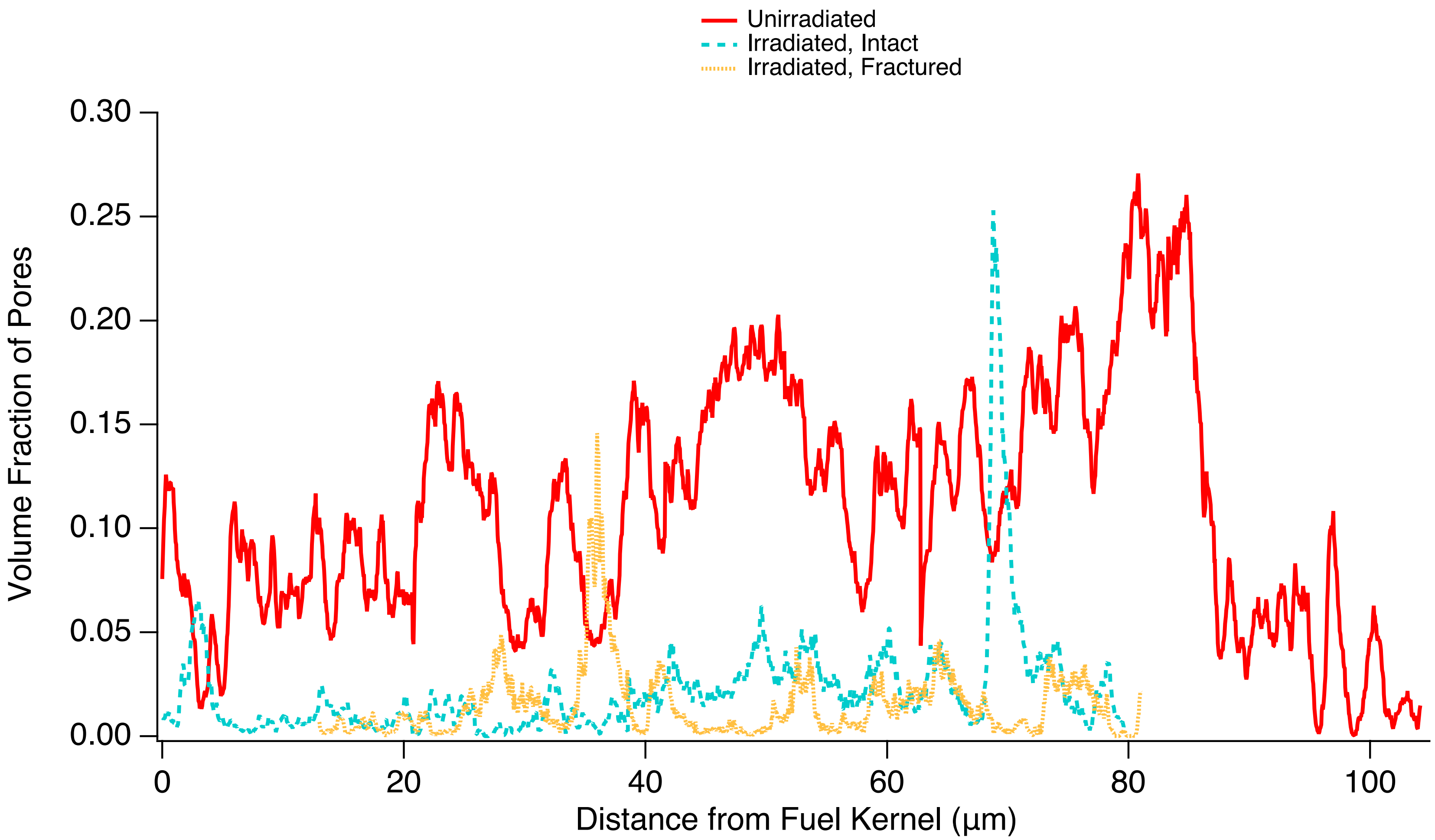


Volume
Centroid position (x,y,z)
Centroid position from interface
Feature shape and size
Solidity
Total surface area and volume of features
Surface area associated with each phase
Etc.

Pore density as a function of distance from the fuel kernel

Key Findings:

- Irradiation condenses the buffer radius
- Average pore fraction decreases from an average of 11% in the unirradiated to 1% in the fractured buffer and 2% in the intact buffer
- Despite shrinkage, irradiated samples still maintain apparent buffer “striations”



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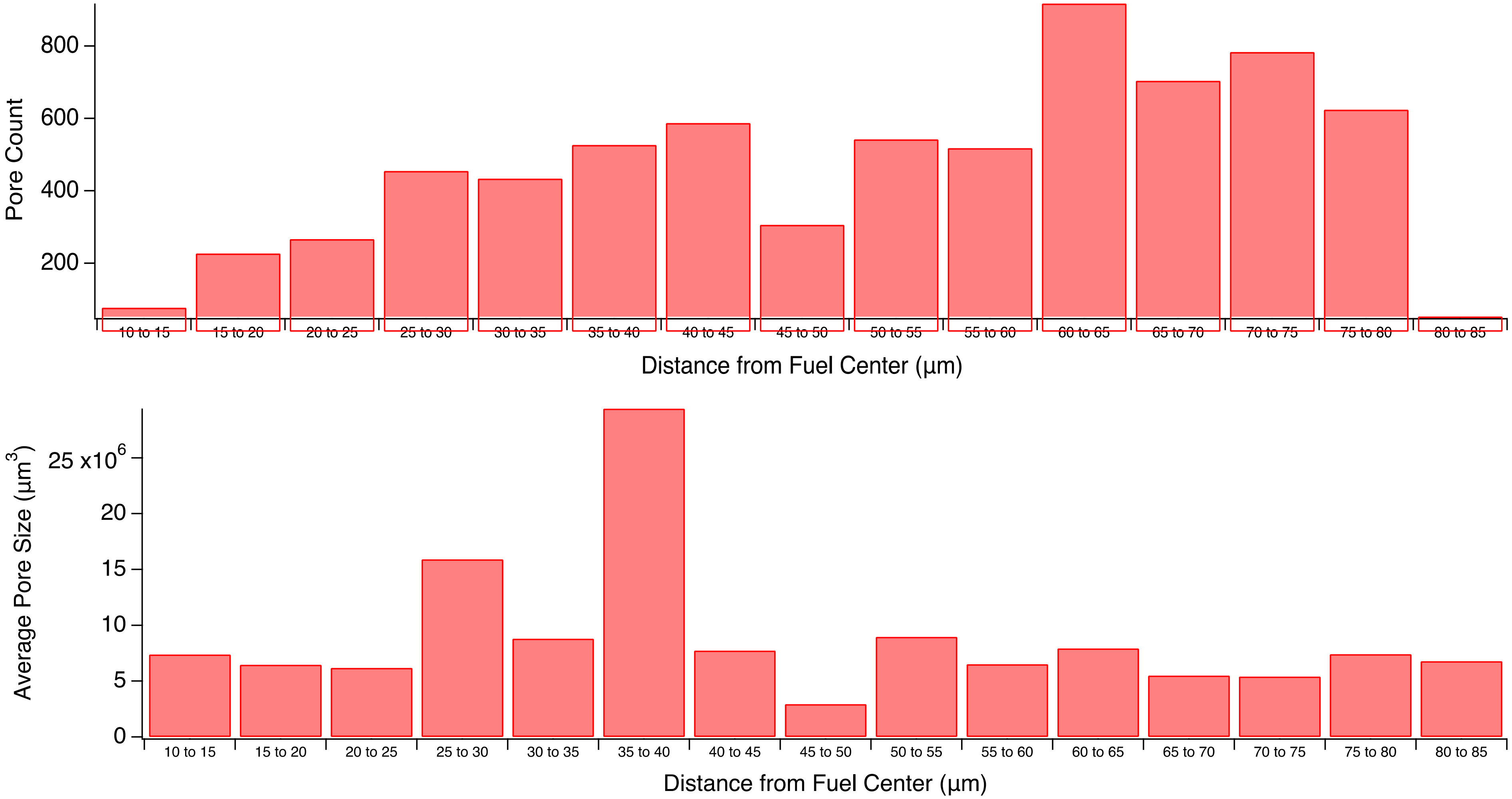
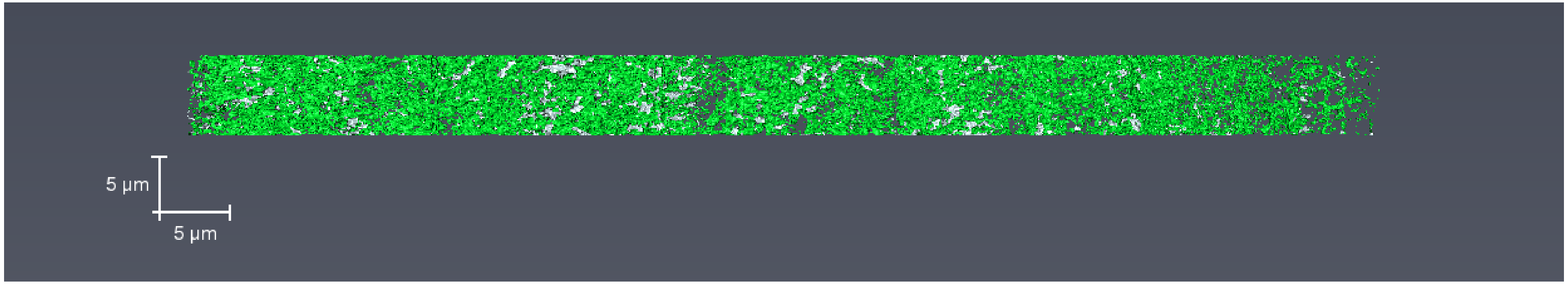
Pore Count and Average Size as a function of Distance



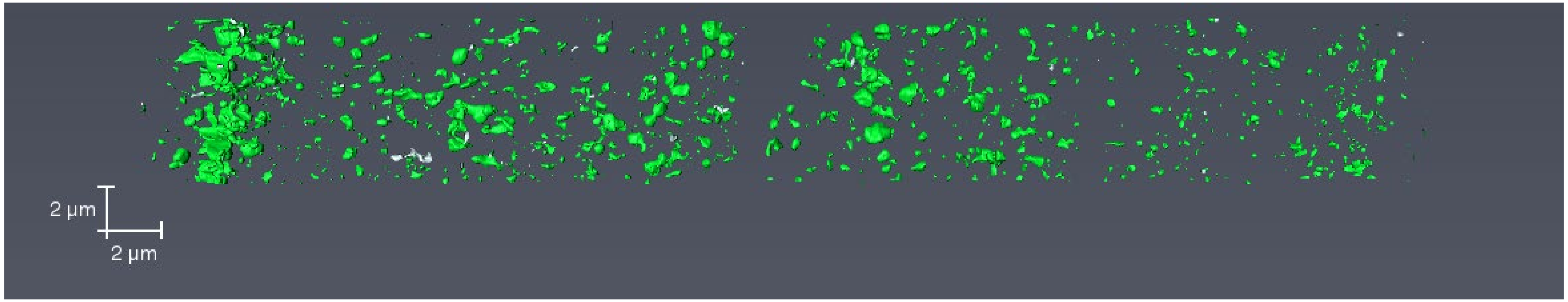
In all samples, the pore count was increased near the IPyC rather than at the Fuel/Buffer interface, which is not unexpected due to the buffer deposition methods. This held true in all samples, an example of which is shown here from the fractured sample

Pore Visualization

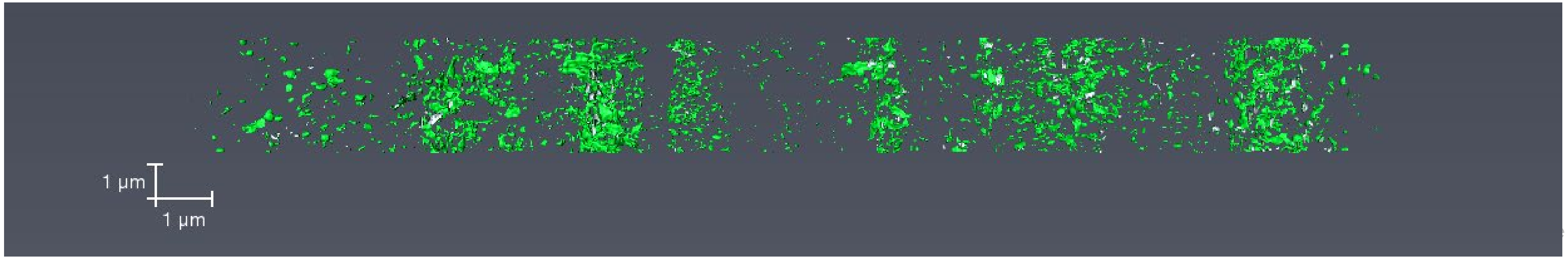
Unirradiated



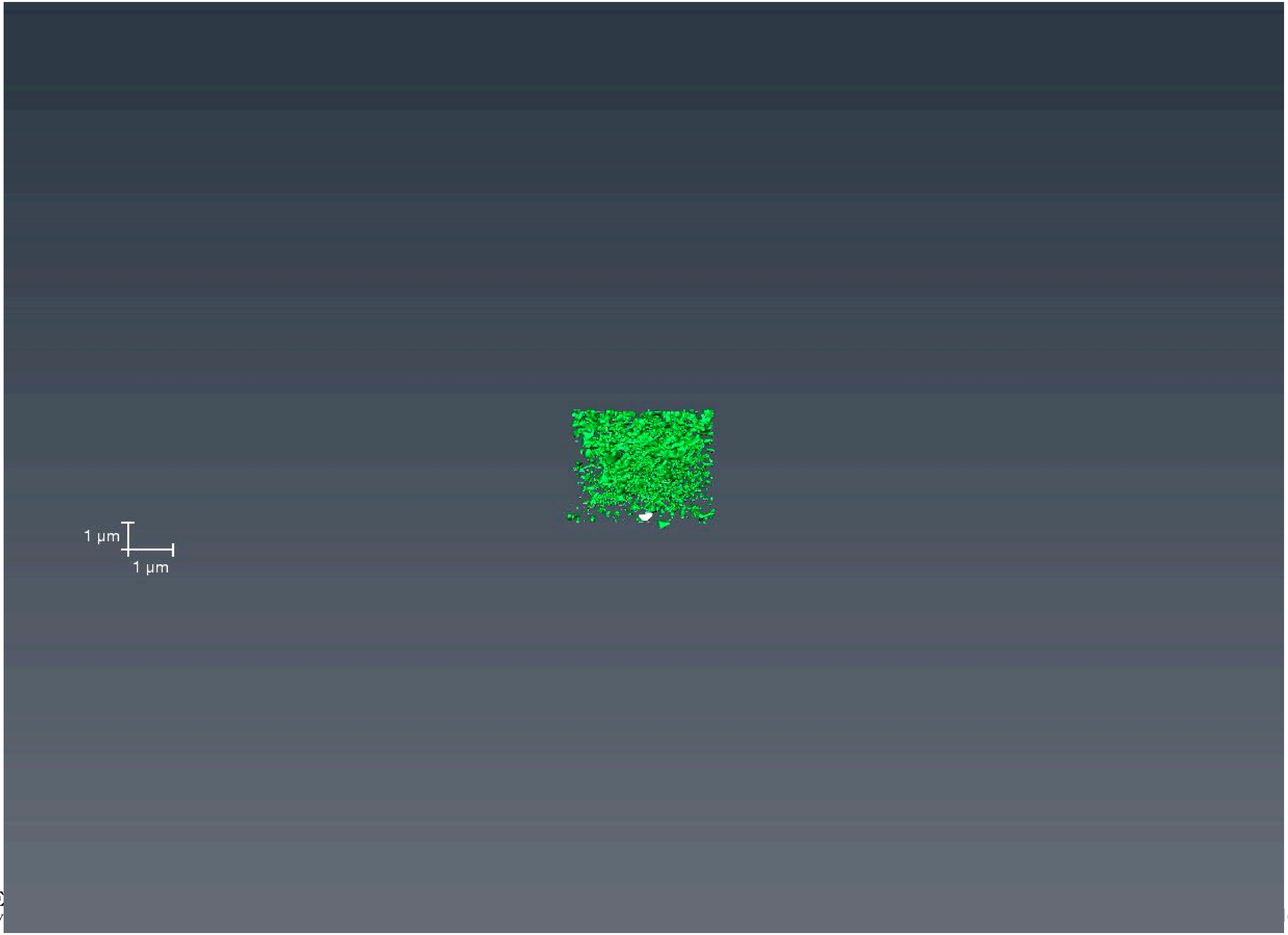
Irradiated - Intact



Irradiated - Fractured

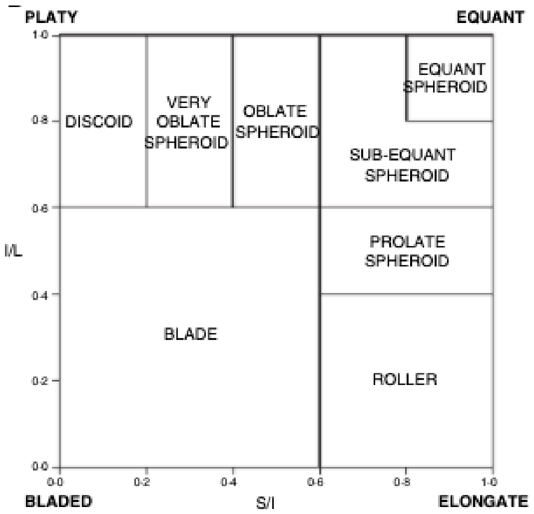


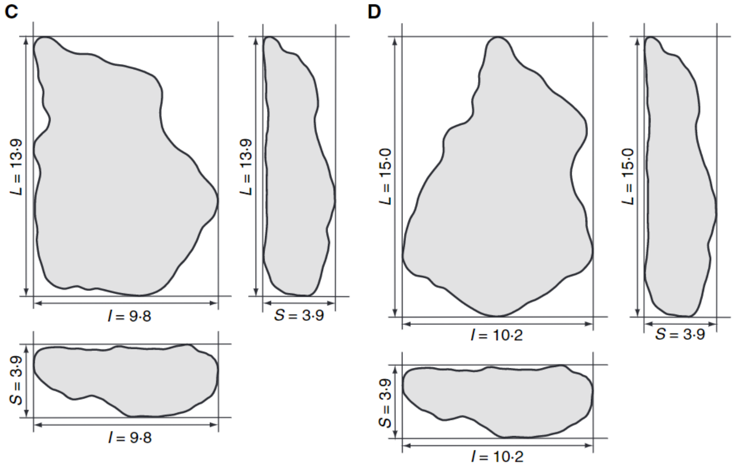
Fractured example



Pore Analysis – Shape and Size

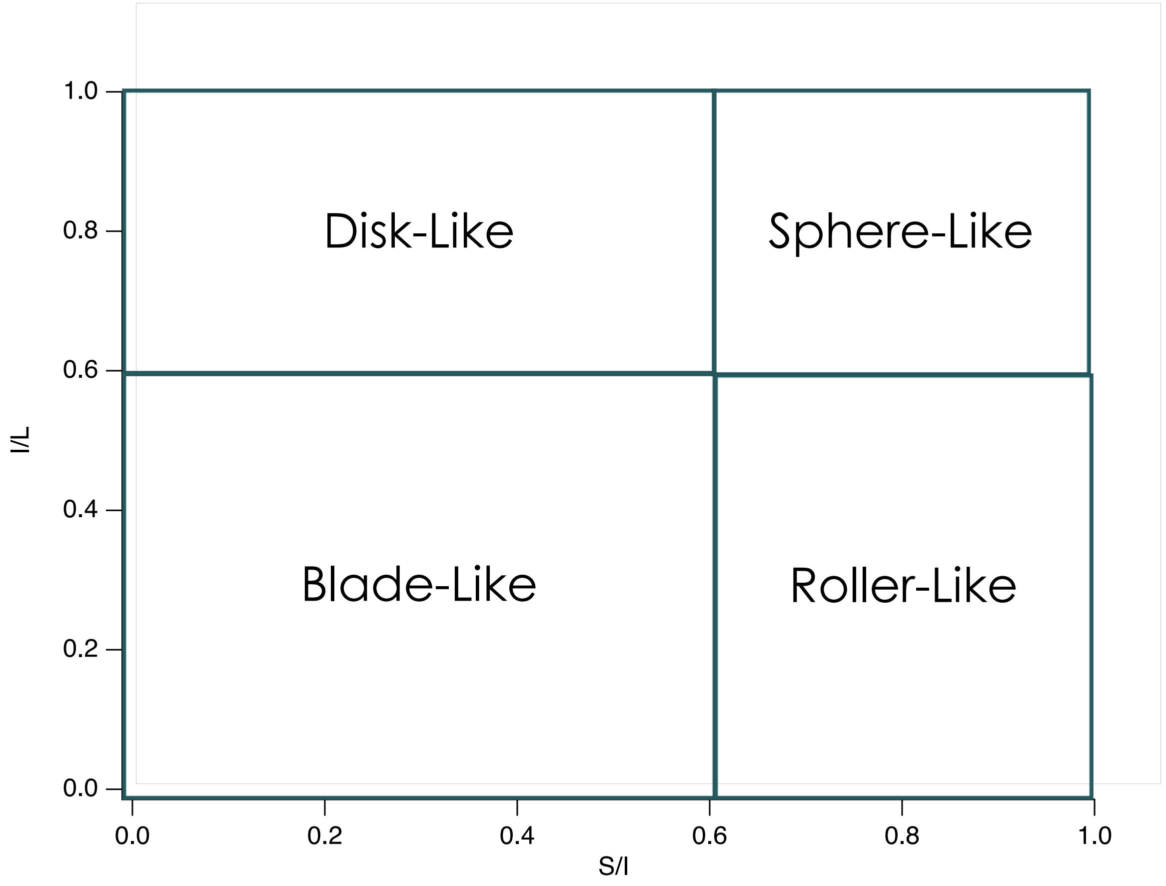
- Builds off shared surface area analysis
- Pore shape analyzed using methodology proposed for sediment by Blott and Pye [1], with Sneed and Folk 1958 notation.
  - Use ratios of L (longest dimension), I (longest dimension perpendicular to L), and S (perpendicular to both L and I)

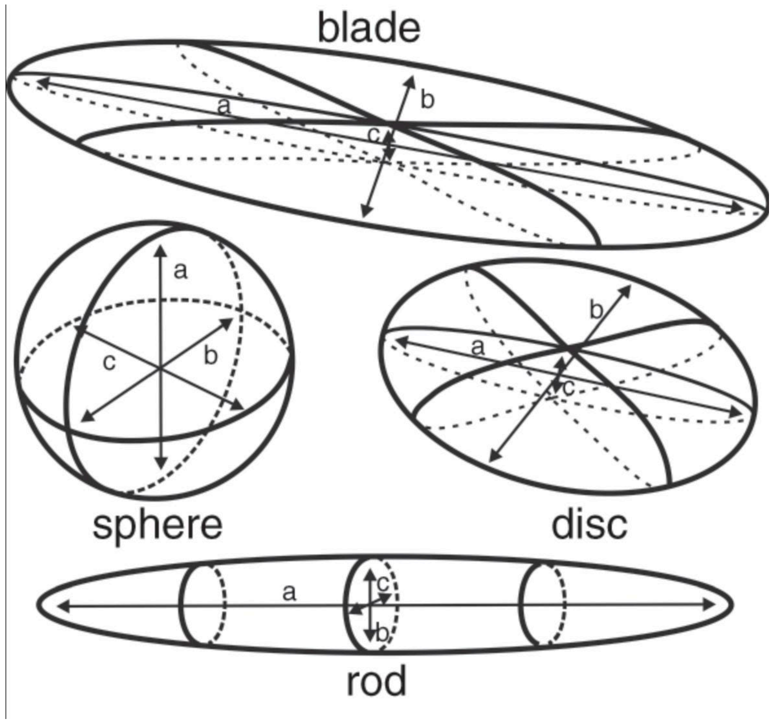




[1] S.J. Blott and K. Pye, “Particle shape: a review and new methods of characterization and classification,” Sedimentology 55(1), p. 31-63, 2007. Figures from [1].

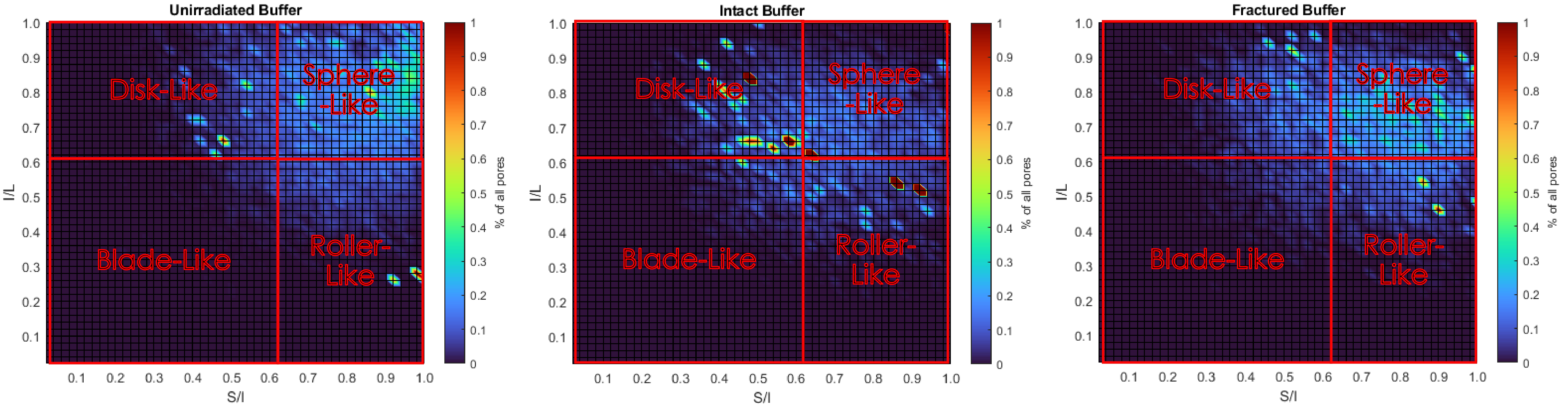
Pore Shape: Pores elongate with irradiation





Rendering from B. Rhoads River Dynamics Appendix B – Characterization of Fluvial Sediment, Cambridge University Press (2020)

Pore Shape: Pores elongate with irradiation



There was a slight shift observed between the unirradiated and the irradiated samples where sphere-like pores were elongated and squished into more rod or disk-like shapes. This is to be expected during shrinkage, as material is moving into the void space of pores

Implications and Conclusions

- **The data showed a difference before and after irradiation in pore size, shape, and frequency, but thus far has not alluded to significant insight in structural changes between the fractured and intact irradiated samples, with the exception being that the average volume fraction is marginally lower in the fractured samples.**
- **Densification of the buffer is not completely accounted for by porosity change. There are at least two influencing mechanisms here, including porosity structure change and a change in structure in the dense material.**
- 3D Slice and View Analysis is a powerful tool for providing quantitative data to describe complex interface and layer properties
  - Approaches to quantifying interface structure have been established which is the first step to connecting interface structure and properties to TRISO particle performance

Acknowledgements

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