GAS-COOLED REACTOR
ADVANCED REACTOR TECHNOLOGIES PROGRAM

July 17, 2024

Quantifying the Dynamic and Static Porosity/Microstructure Characteristics of Irradiated Graphite through Multi-technique Experiments and Mesoscale Modeling

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DOE ART GCR Review Meeting

Hybrid Meeting at INL

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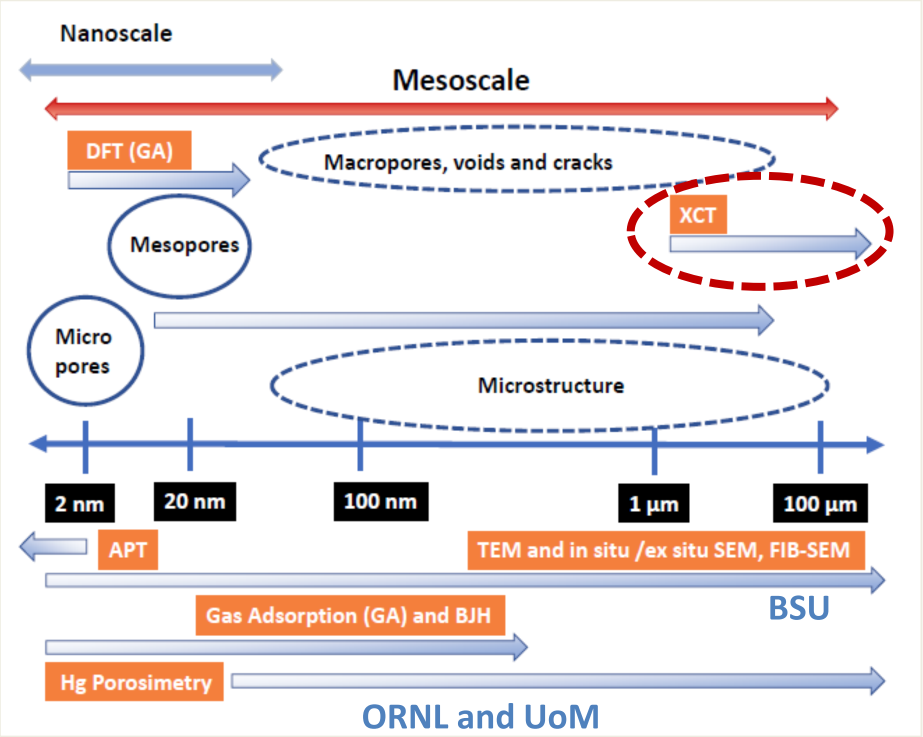
Pores in Nuclear Graphite Grades and Objective of this Work

- All nuclear graphite grades are porous; porosity varies between 14 to 22%.
- Pores are formed during the manufacturing process as well as during in-service (reactor) conditions.
- It is typical to report only a macroscopic measure of porosity, which is defined as the pore volume to the total volume. However, the pore structures are much richer and complex.
- The objective of this work is to quantify the pore structures using **machine learning techniques** to extract metrics such as **distributions** of **pore volume**, **pore shape**, **pore network** and **connecting lengths**, **open** and **percolating porosity**, and **correlations** between the metrics.
- These pore metrics will allow **realistic** modeling and simulation of graphite pores and transport through them.

Grade	Grain Type	Grain Size (µm)	Density (g/cm^3)	Porosity (%)
NBG-18	Large	1600	1.85	14
NBG-17	Medium	800	1.89	14
PCEA	Medium	800	1.84	16
IG-110	Fine	20	1.77	21
G347A	Superfine	11	1.85	-
AXF/ZXF 5Q	Ultrafine	1–5	1.78	20

Pores Size Distribution and Methods



NC State

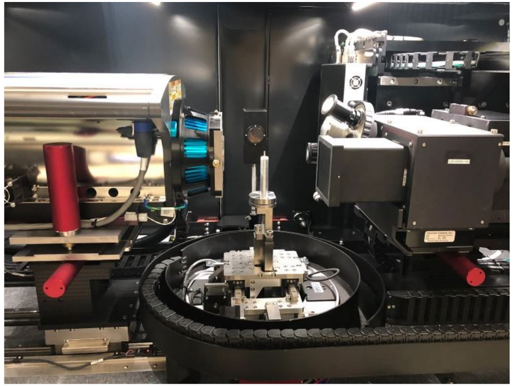
BSU

ORNL and UoM

Characterization Methods

X-ray Computed Tomography

Three cubic samples, each approximately 5 mm^3 in volume, are cut from different sections of bulk graphite from the four grades: **AXF-5Q**, **IG-110**, **NBG-18** and **PCEA**.

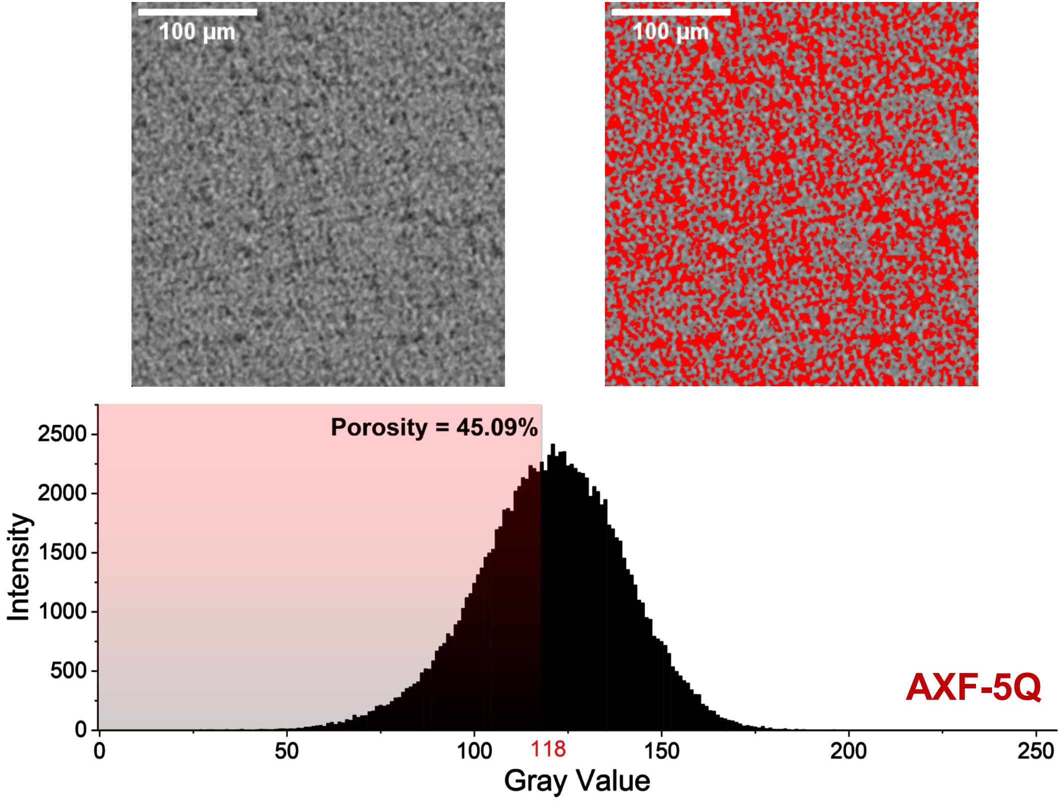


XMReconstructor software is used for image reconstruction.

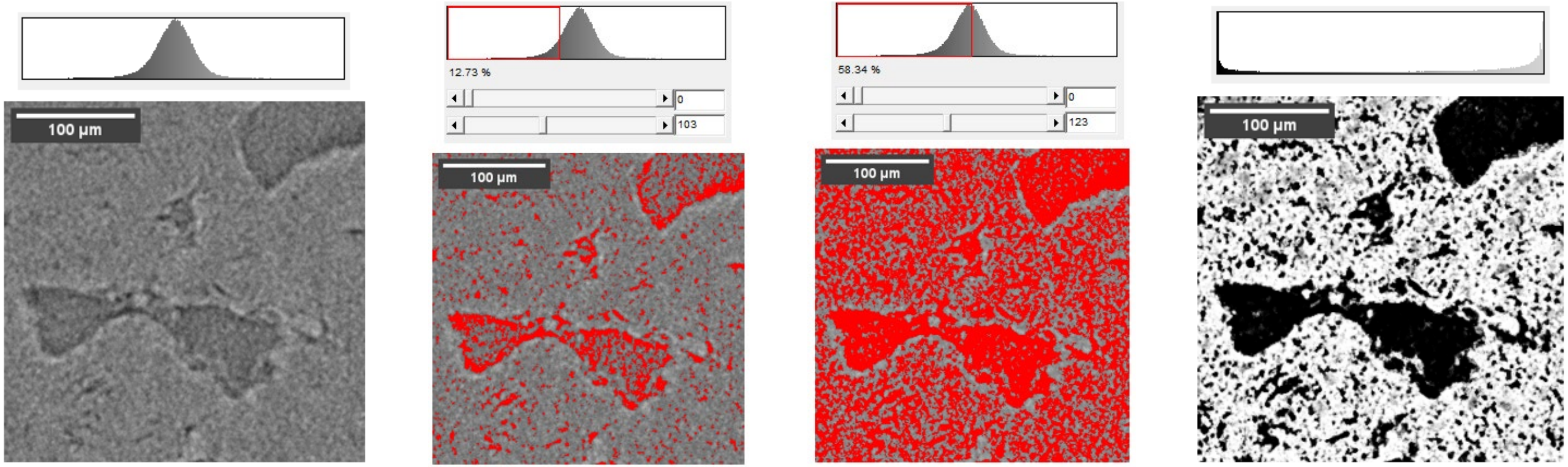
Magnification	20x
Current (mA)	90
Voltage (kV)	100
Exposure time (s)	10
Image size (voxels)	492 x 441 x 478
Resolution (micron/voxel)	0.95
Detector to sample distance (mm)	52.4
Source to sample distance (mm)	29.1

Challenges of Conventional Thresholding in XCT Analysis – Low Contrast

- The low atomic number of graphite results in poor contrast between solid graphite region and and pore regions.
- Conventional thresholding fails due to unimodal histograms lacking clear separation, as illustrated by the single peak in the histogram.
- Standard thresholding methods such as Otsu and IsoData typically overestimates the porosity.



Challenges of Manual Thresholding in XCT Analysis – Complex Pore Structures



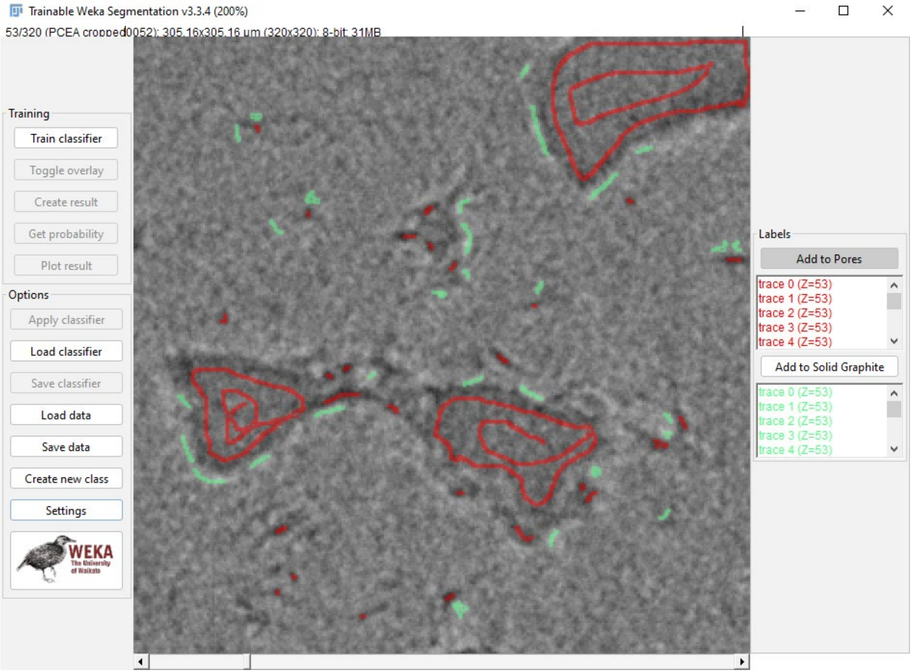
- Manual and conventional methods fail to capture large pore clusters without simultaneously overestimating smaller pores.
- **Machine learning tools** are found to be effective for identifying pore edges. This enhances thresholding accuracy, as demonstrated by the probability map and bimodal histogram (right).

Machine Learning Approach for Pore-Space Segmentation in XCT Images – Overview

- Fast Random Forest (FRF) algorithm is used to classify 3D XCT images into porous and solid graphite regions, reducing human intervention to minimal set of user input (a few confidently identifiable voxels).

- Voxel properties (mean grayscale value, variance, and gradient magnitude) are used to distinguish between porous and dense regions.

- Information Gain and Entropy based on voxel mean and variance are used to find the best node split in decision trees, optimizing the separation of pore and solid phases.

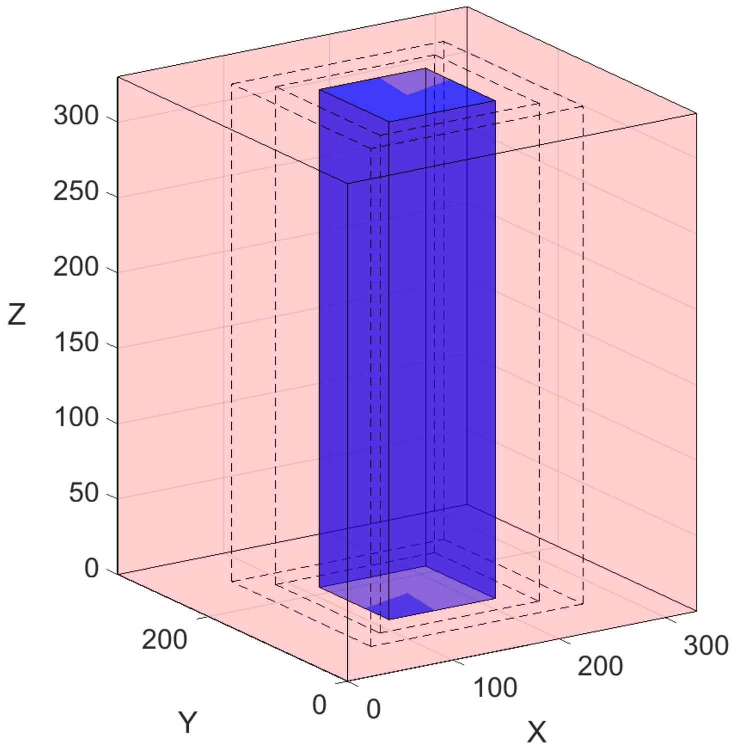


Representative Element Volume Analysis

- REV denotes the smallest volume that captures the main pore characteristics.

- The starting element is a cuboid of the same height as the original volume (320 voxels), and a small cross-sectional area that increases at each step.

- Total porosity is calculated for each volume, and the REV is reached when the relative gradient error of porosity reaches below 0.2 in all directions.

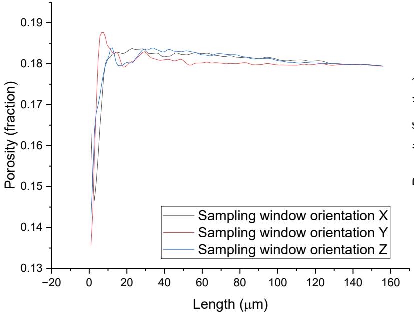


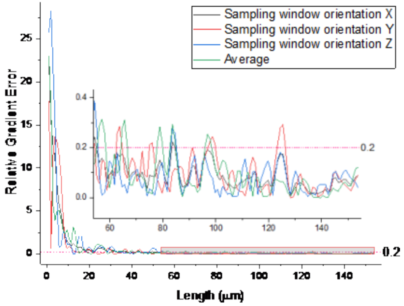
REV Analysis: AXF-5Q

Representative Element Volume Analysis

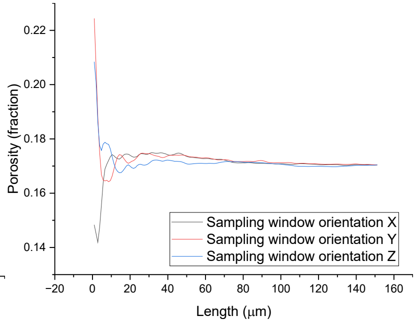
- For the three samples, porosity seems to converge around ~17-18%.
- The relative gradient error, averaging over all directions, seems to fall below 0.2 around 100 microns (105 voxels) which is ~30% of the original volume.

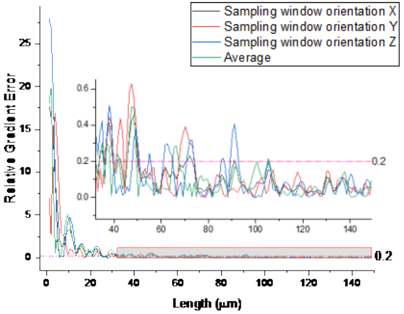
Sample 1



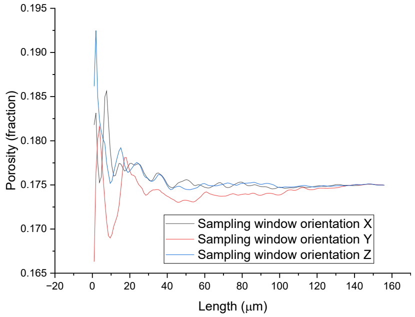


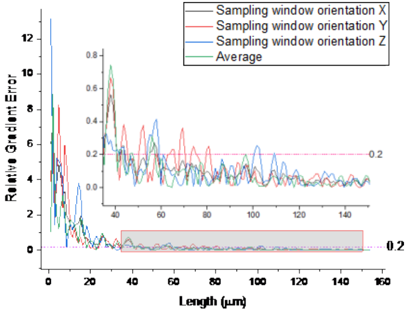
Sample 2





Sample 3

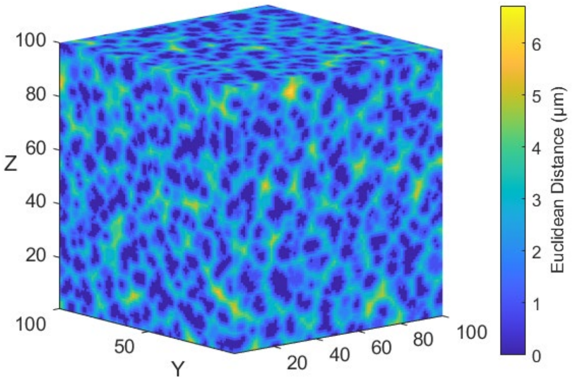




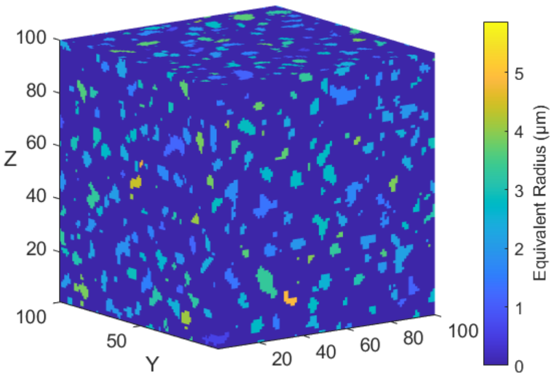
Pore Space Segmentation After Thresholding

- The Euclidean distance transform of the inverse REVs is calculated to represent the distances between centers of neighboring pores.
- The watershed algorithm segments the pore space into individual pores at the narrowest points in the paths between two local maxima over a 3-D 18-connected neighborhood.
- This approach can identify pore regions as well as connections between pores.

Euclidean Distance Transform of Inverse REV



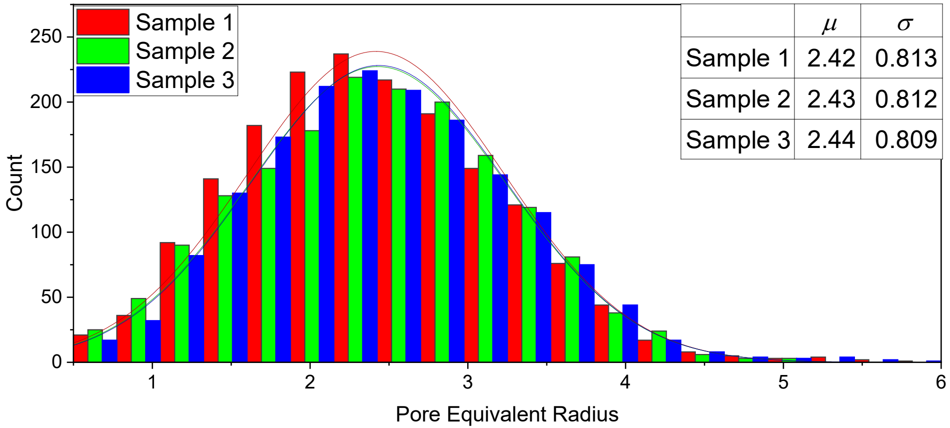
REV Pore Segmentation



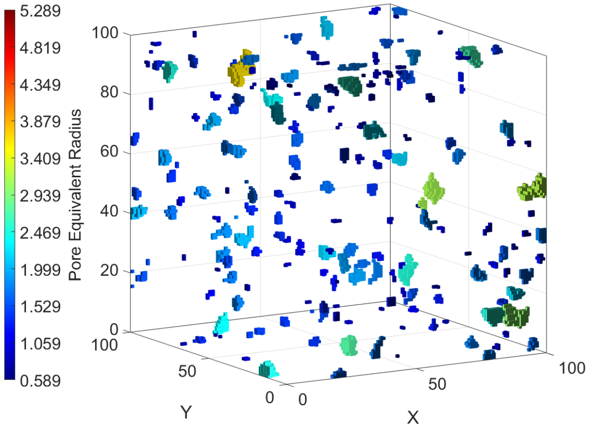
Pore Radius Distribution: AXF-5Q

Pore Radius Distributions

The equivalent radius of each pore is calculated by: $R_{eqv} = \text{Res} \times \left(\frac{3\times\text{volume}}{4\pi}\right)^{\frac{1}{3}}$ where Res is the scan resolution, and volume is the number of voxels per pore.



	μ	σ
Sample 1	2.42	0.813
Sample 2	2.43	0.812
Sample 3	2.44	0.809

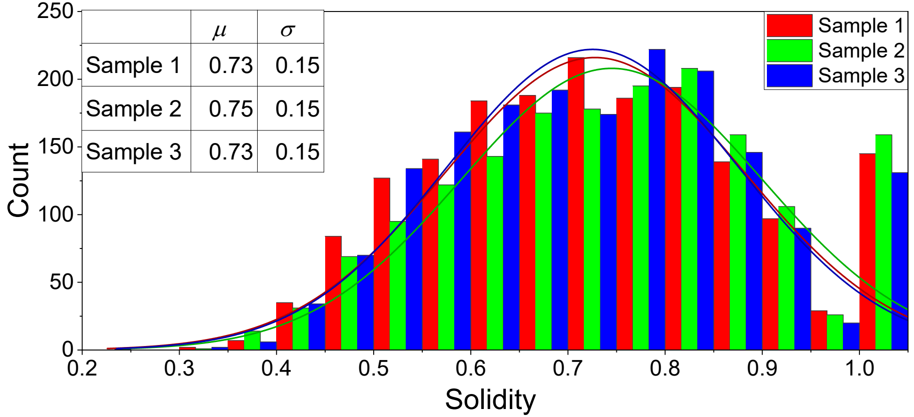


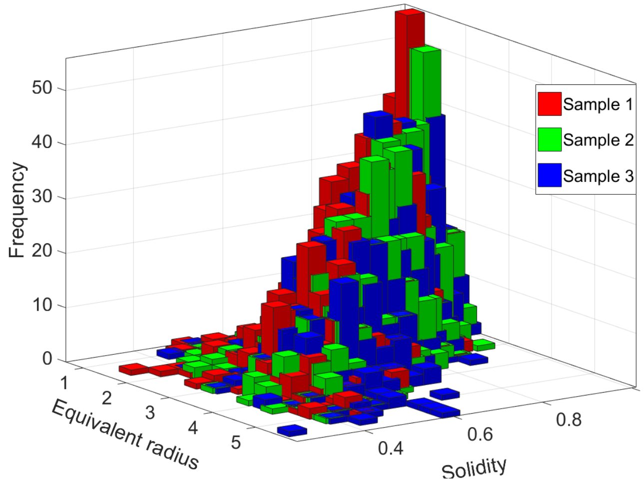
Pores in AXF-5Q have an average radius of ~2.5 microns.

Pore Shape Distribution: AXF-5Q

Pore Shape Distributions

Solidity is the ratio of the true volume to its convex volume, with convex volume being that of the polygon connecting all the corners of that shape. The higher the solidity, the closer the shape is to a perfect sphere.

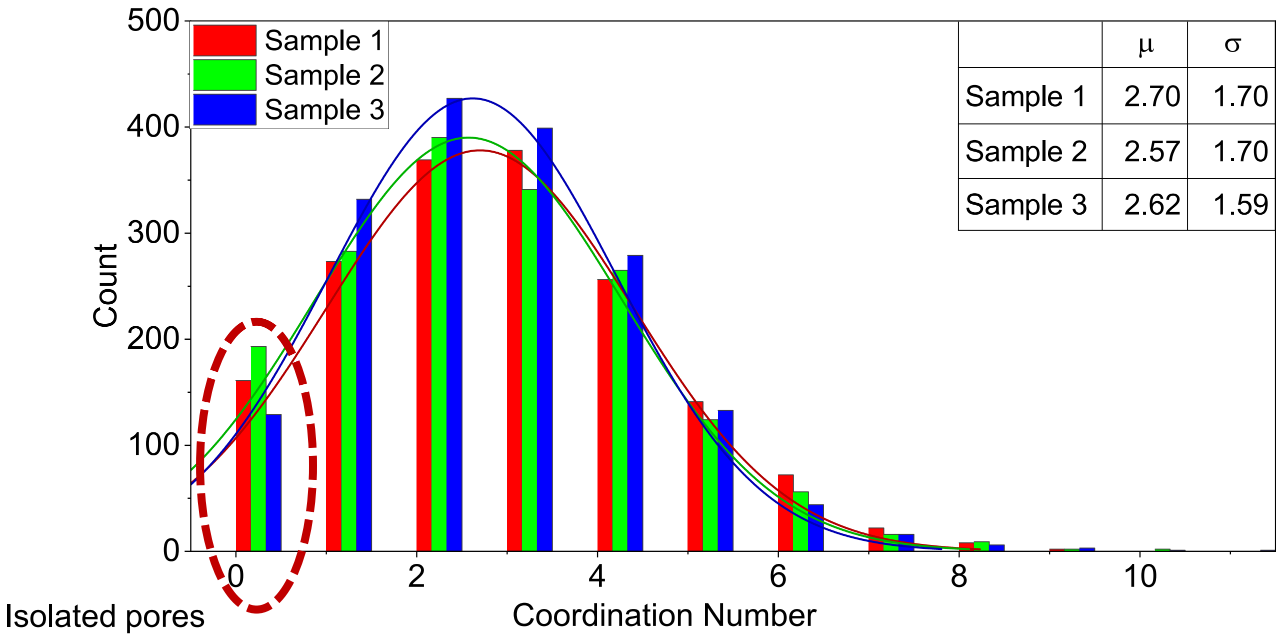




Pores are not perfect spheres! Smaller pores, however, tend to be more spherical.

Porosity Connectivity: AXF-5Q

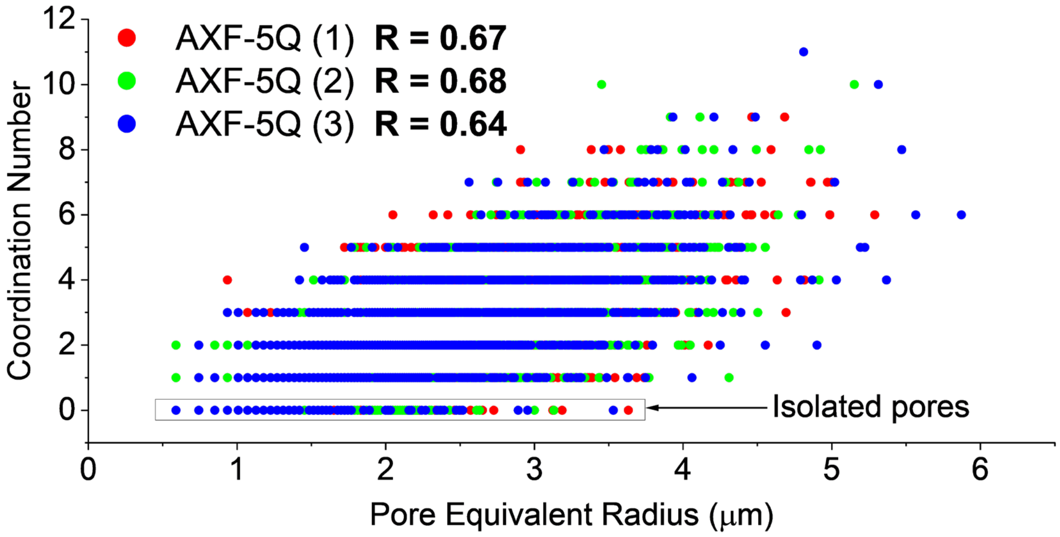
Pore Network Connection – Pore Coordination Number



	μ	σ
Sample 1	2.70	1.70
Sample 2	2.57	1.70
Sample 3	2.62	1.59

On an average, each pore is connected to 2 to 3 pores.

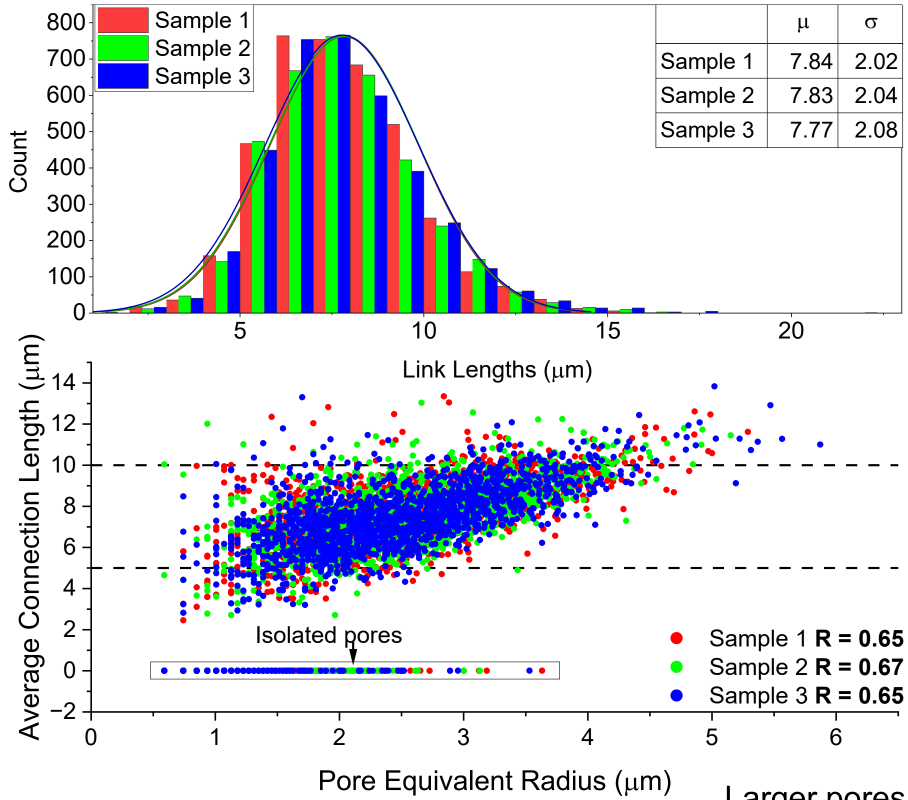
Correlation between Connections and Size



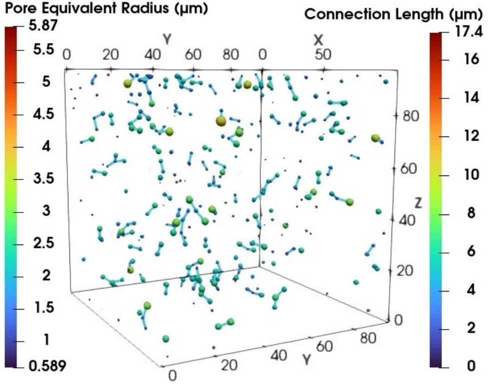
Larger pores tend to have more connections.

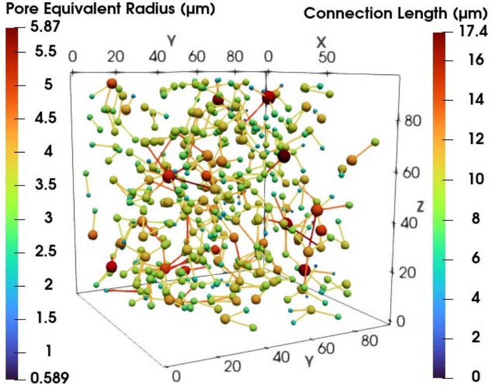
Connection Lengths: AXF-5Q

Pore Network Visualization – Connection Lengths



	μ	σ
Sample 1	7.84	2.02
Sample 2	7.83	2.04
Sample 3	7.77	2.08



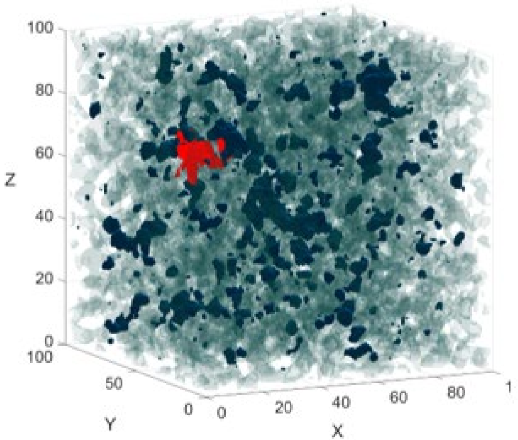


Larger pores tend to be further apart from each other.

Open and Closed Porosity

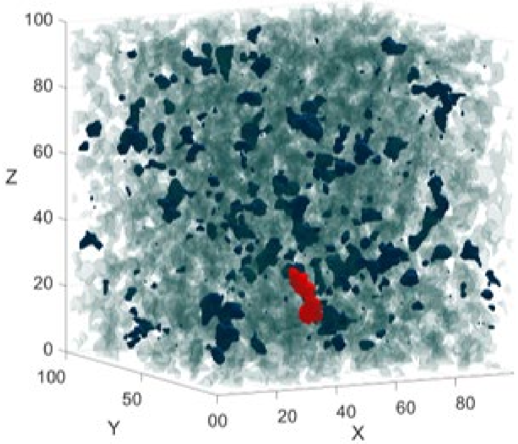
Open pores refer to those that link directly to the external surface, serving as channels for possible diffusion of external agents.

Sample 1



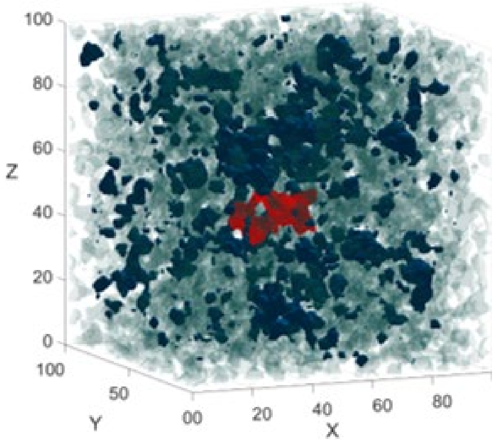
Total Porosity = 18.1%
Open Porosity = 15.3%
(85% of Total Porosity)

Sample 2



Total Porosity = 17%
Open Porosity = 15%
(88% of Total Porosity)

Sample 3



Total Porosity = 17.6%
Open Porosity = 16.2%
(92% of Total Porosity)

AXF-5Q has an open porosity of ~15%

Perecolating Porosity Characterization: : AXF-5Q

Percolating Porosity

Represents the pathways or channels formed by interconnected pores.

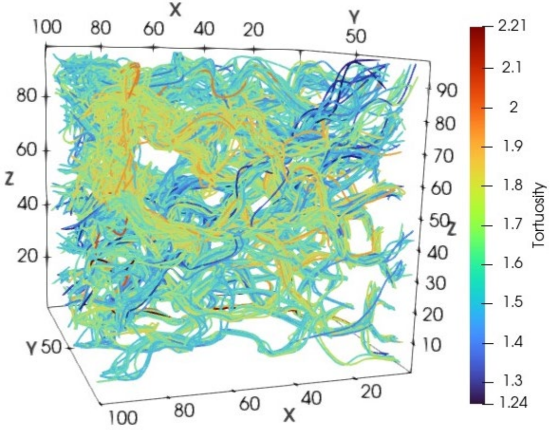
Sample 3

Total porosity = 17.6%

Open porosity = 16.2%

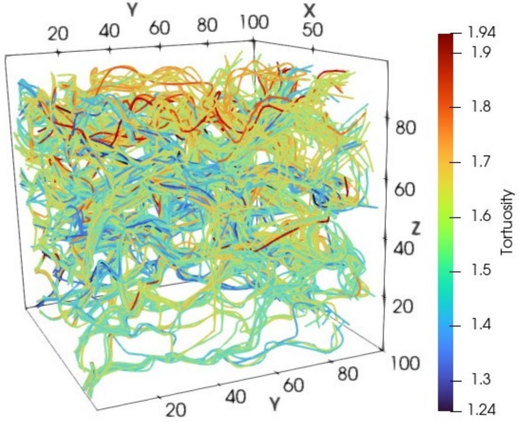
X-direction

Percolating porosity = 10%



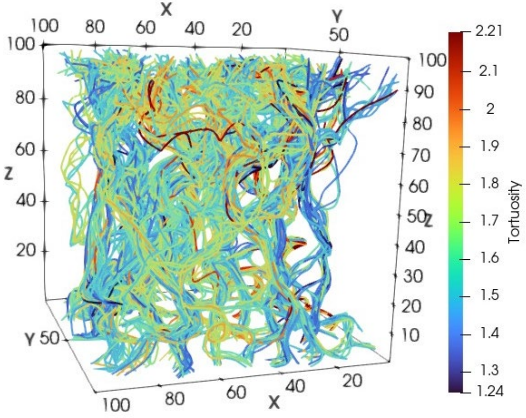
Y-direction

Percolating porosity = 10.6%



Z-direction

Percolating porosity = 9.9%



AXF-5Q has percolating porosity of ~10%

Tortuosity Characterization: AXF-5Q

Tortuosity of The Percolating Paths

A measure of the deviation of the diffusion path through a porous medium from the straight-line, or Euclidean, distance; commonly defined as the ratio of the actual path length to the straight-line distance between the start and end points.

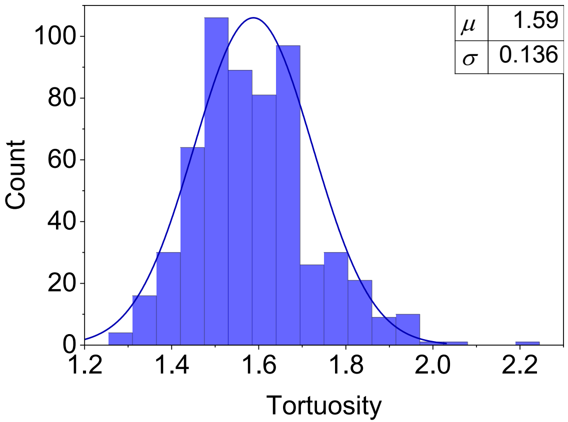
Sample 3

Total porosity = 17.6%

Open porosity = 16.2%

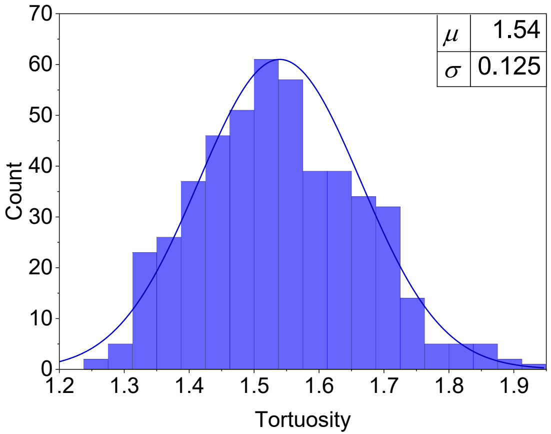
X-direction

Percolating porosity = 10%



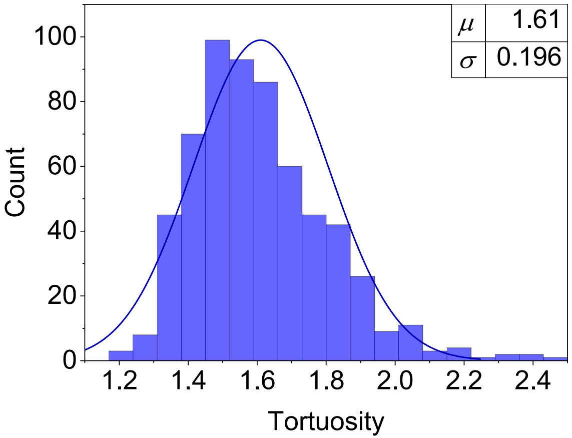
Y-direction

Percolating porosity = 10.6%



Z-direction

Percolating porosity = 9.9%

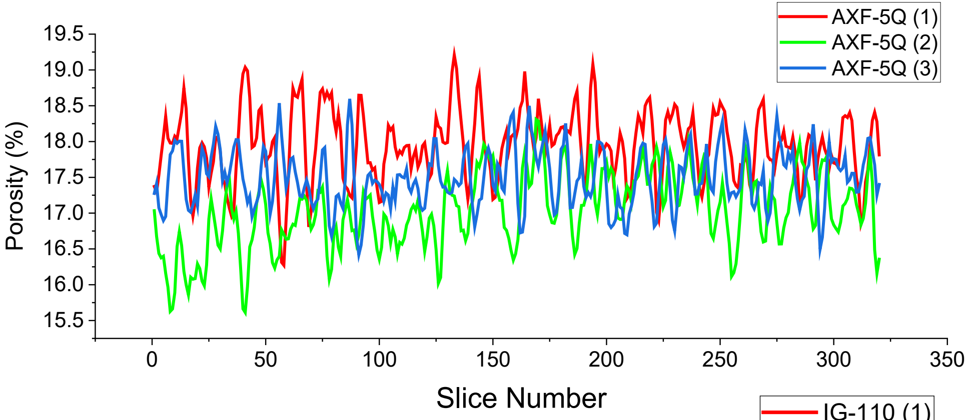


The percolating pores paths are not along straight lines.

Porosity Variation across Different Slices

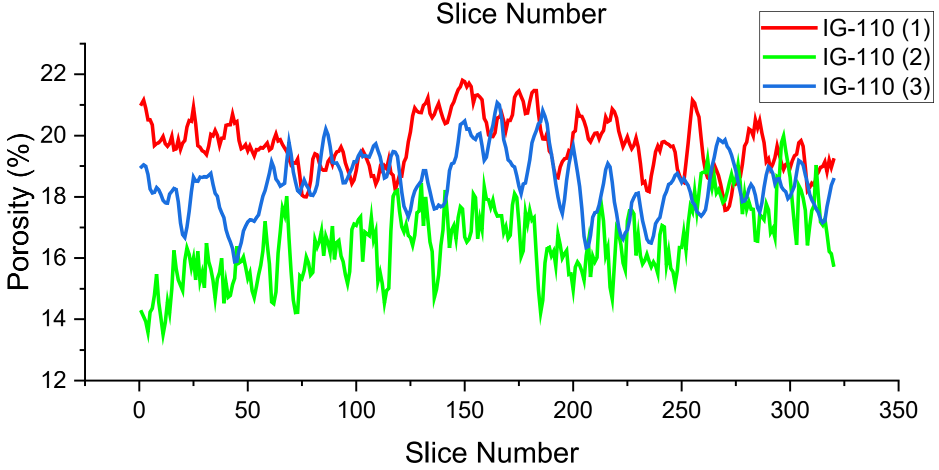
AXF-5Q

320 cubic voxels = $(304\ \mu m)^3$



IG-110

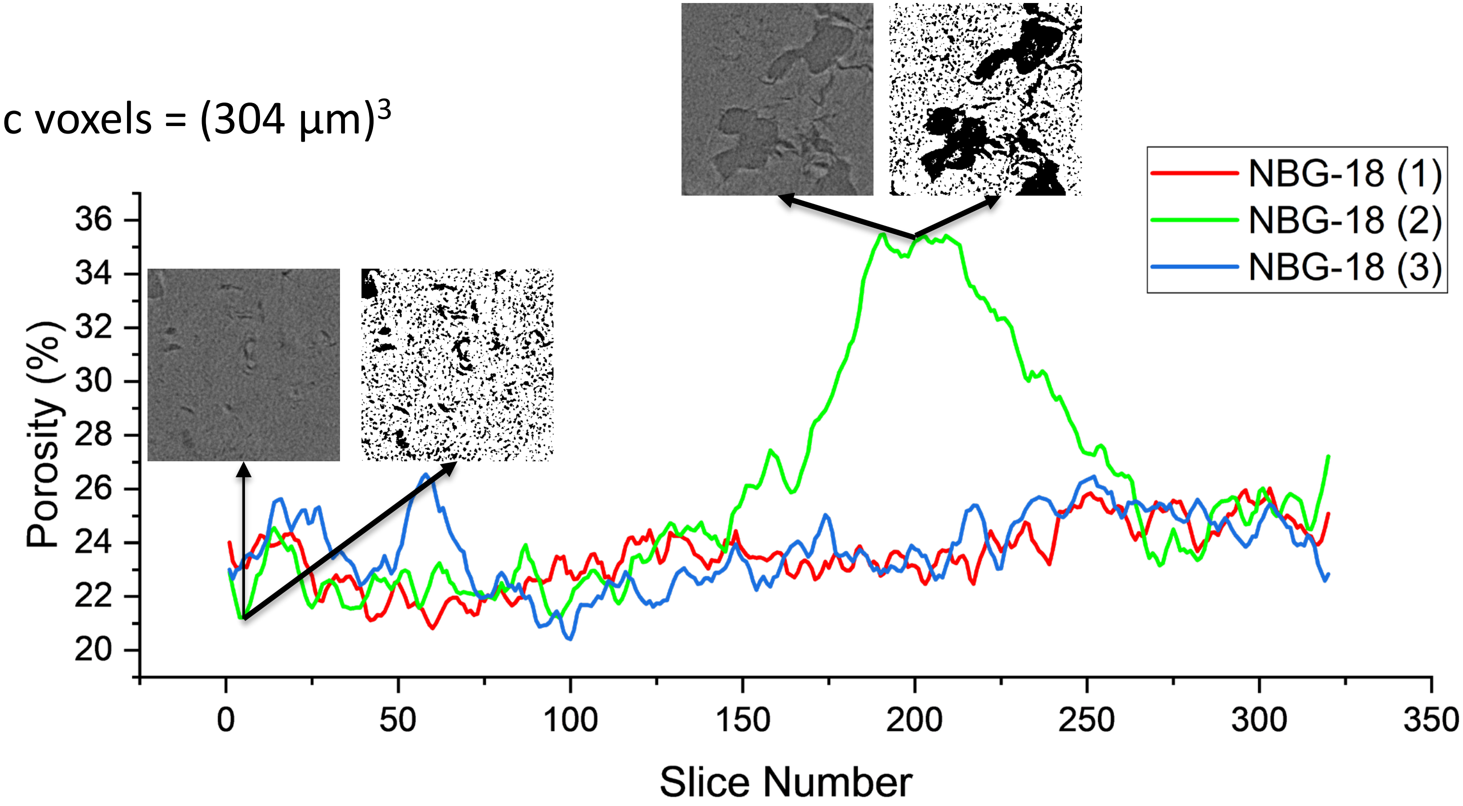
320 cubic voxels = $(304\ \mu m)^3$



Porosity Variation across Different Slices

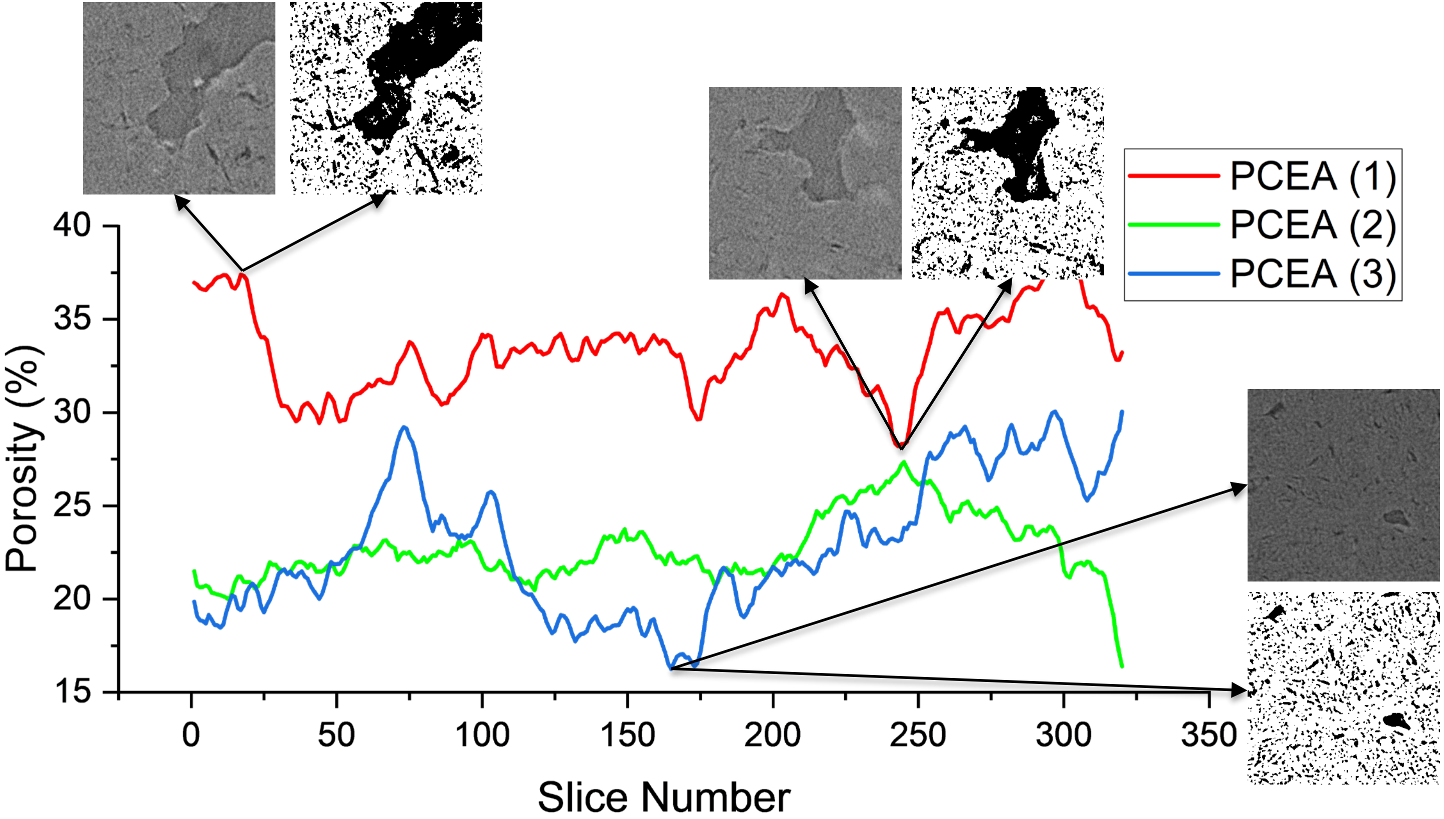
NBG-18

320 cubic voxels = (304 μm)^3



Porosity Variation across Different Slices

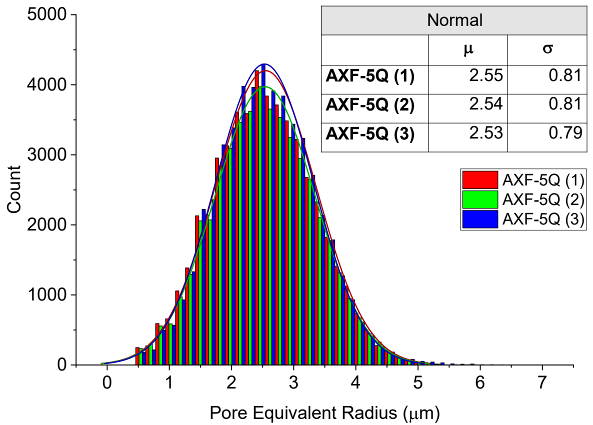
PCEA
320 cubic voxels = $(304\ \mu m)^3$

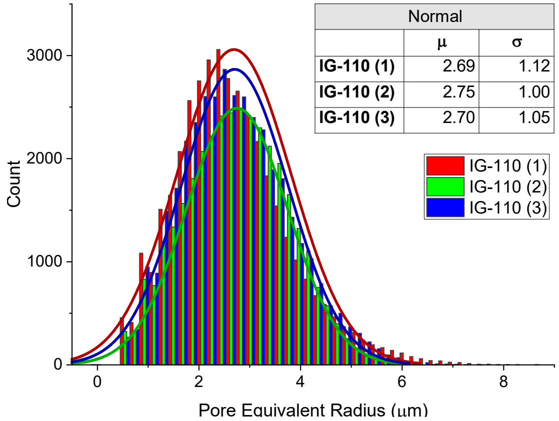


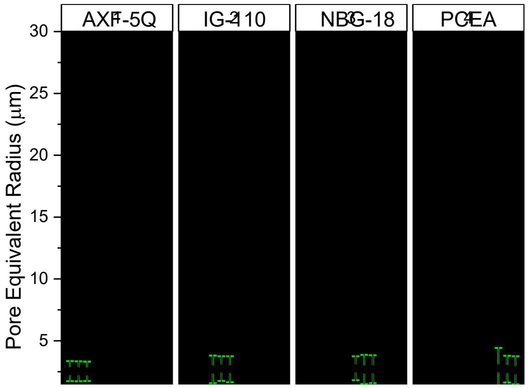
Pore Equivalent Radius

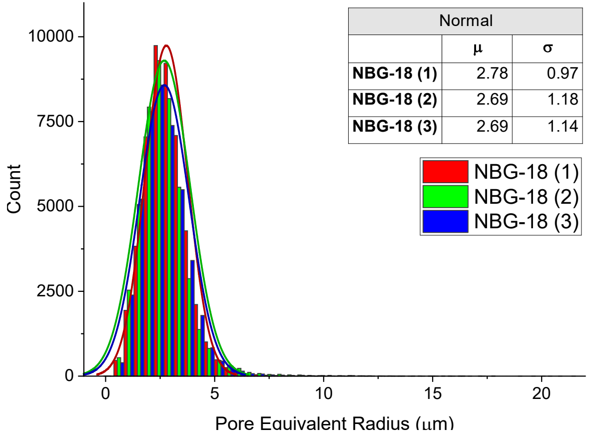
Remarkably, the average pore size is between 2.5 to 2.8 microns across all the grades.

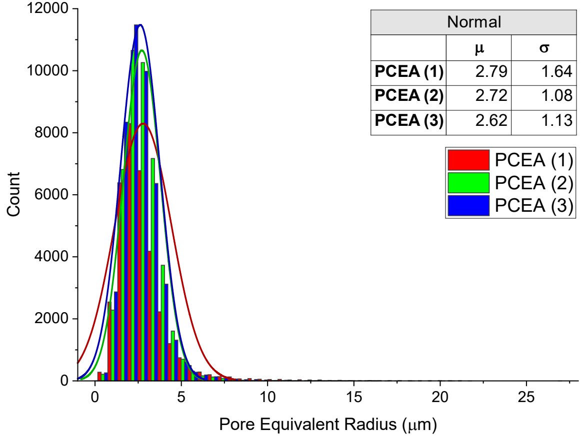
Deviation from the normal distribution for PCEA is notable.







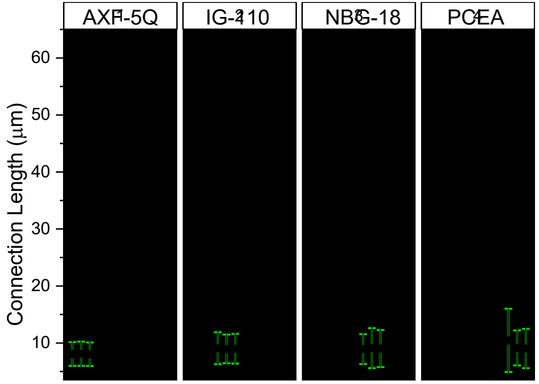


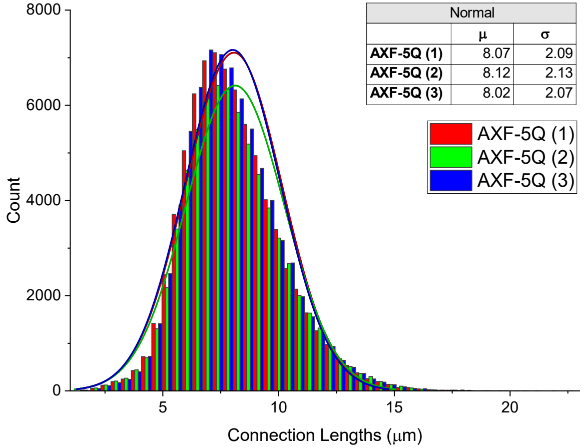


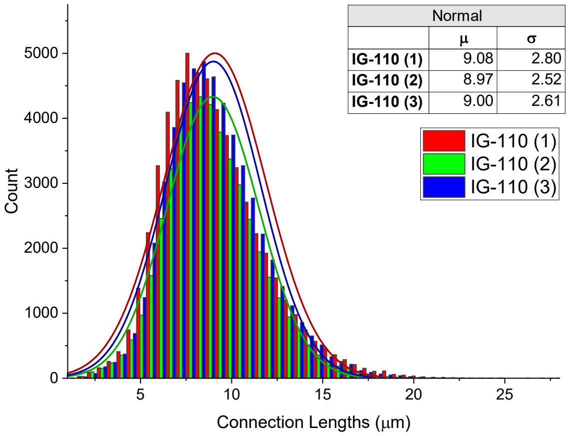
Connection Lengths

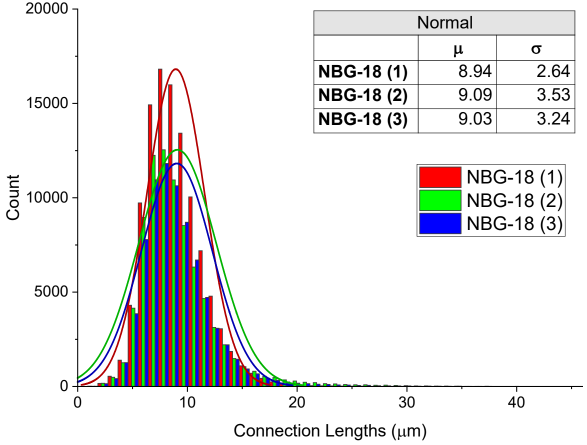
Average connection lengths lie between 8 to 10.5 microns across all the grades.

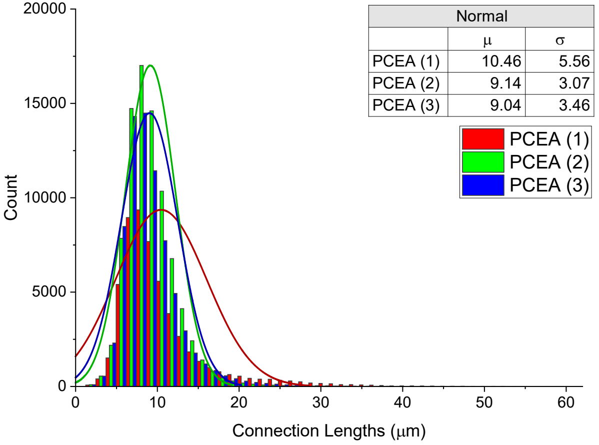
Deviation from the normal distribution for NBG-18 and PCEA is notable.





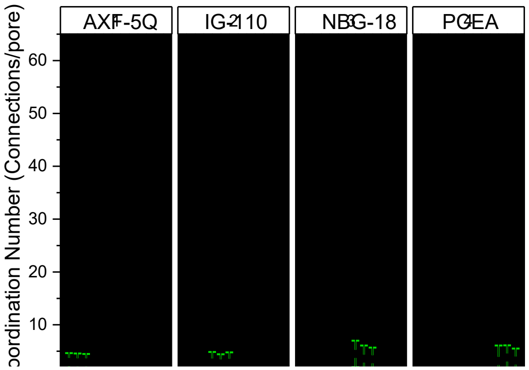


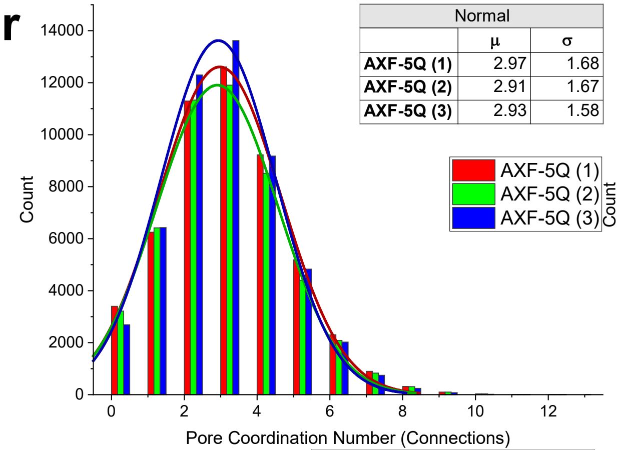




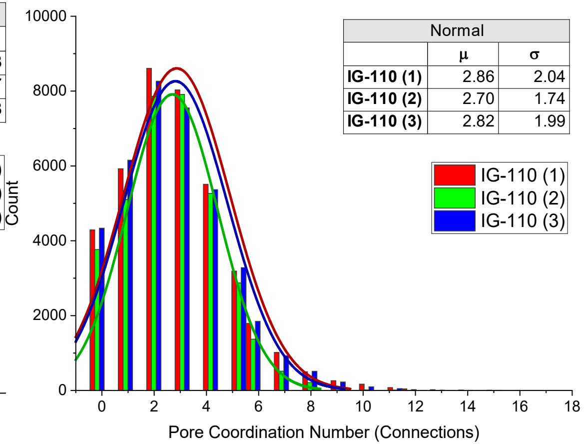
Pore Coordination Number

The average pore coordination numbers range between 2.7 to 4.5 across all grades.

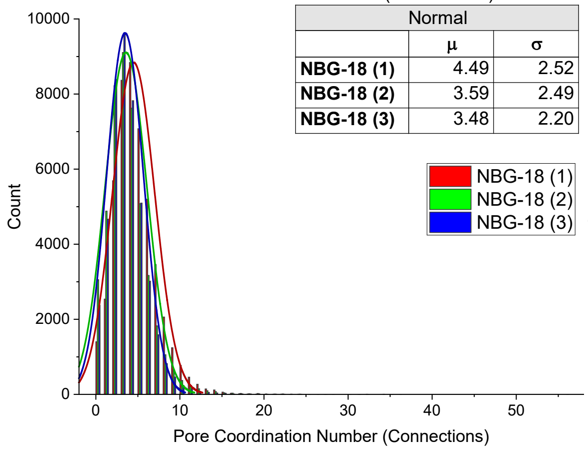




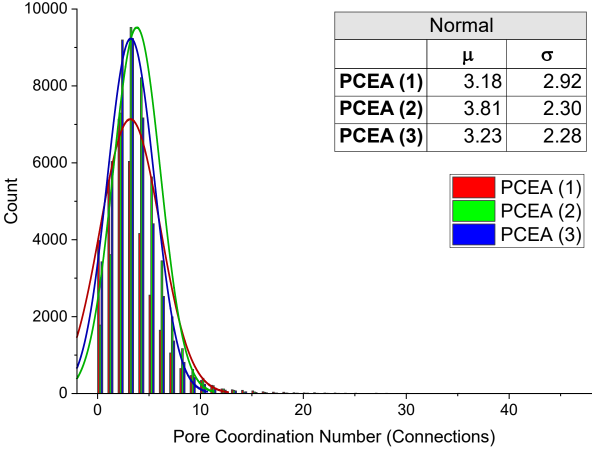
Normal	μ	σ
AXF-5Q (1)	2.97	1.68
AXF-5Q (2)	2.91	1.67
AXF-5Q (3)	2.93	1.58



Normal	μ	σ
IG-110 (1)	2.86	2.04
IG-110 (2)	2.70	1.74
IG-110 (3)	2.82	1.99



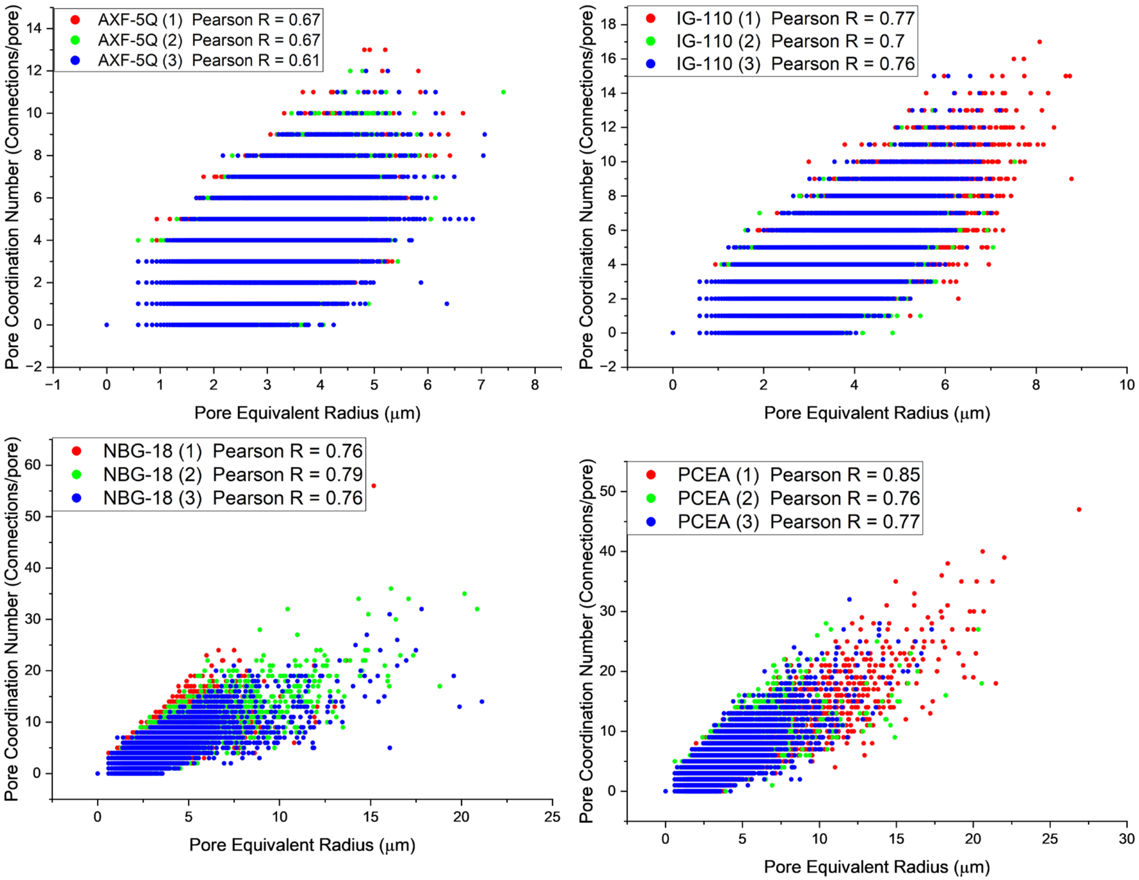
Normal	μ	σ
NBG-18 (1)	4.49	2.52
NBG-18 (2)	3.59	2.49
NBG-18 (3)	3.48	2.20



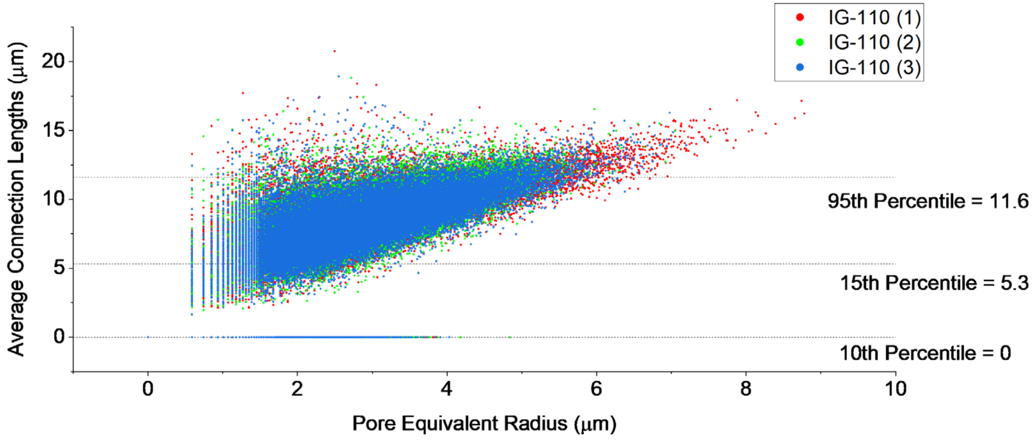
Normal	μ	σ
PCEA (1)	3.18	2.92
PCEA (2)	3.81	2.30
PCEA (3)	3.23	2.28

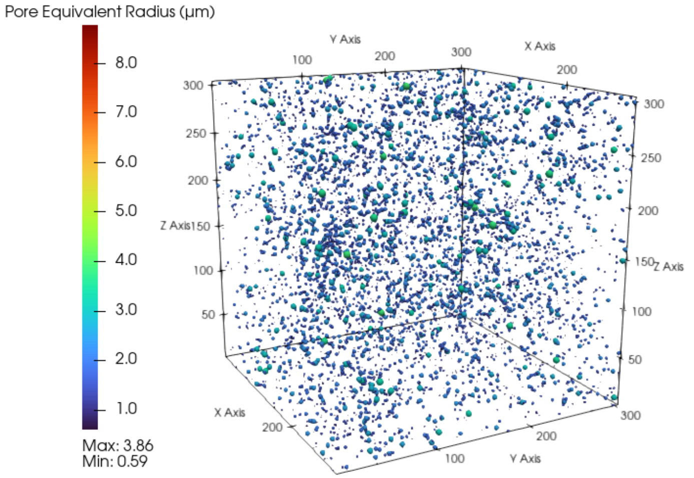
Correlation Between Pore Equivalent Radius and Pore Coordination Number

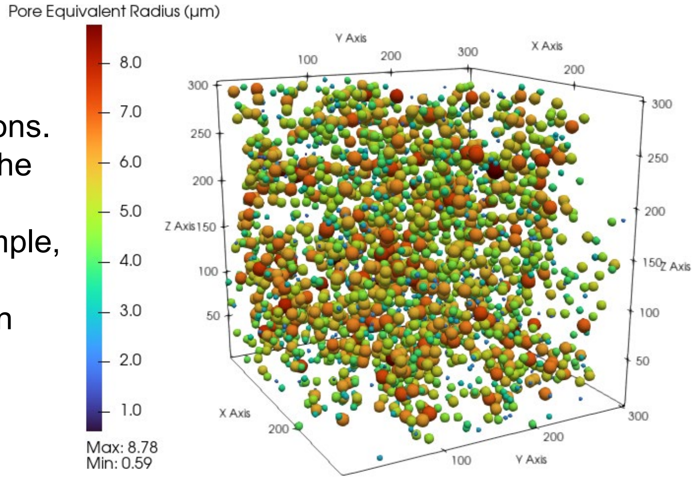
- A moderately strong linear relationship is observed between the size of the pores and their number of connections, irrespective of the grade.
- For both NBG-18 and PCEA grades, the smaller subset of pores that are significantly larger than the majority also appear to conform to the observed correlation pattern.



Correlation Between Pore Equivalent Radius and Average Connection Length Distributions – IG-110

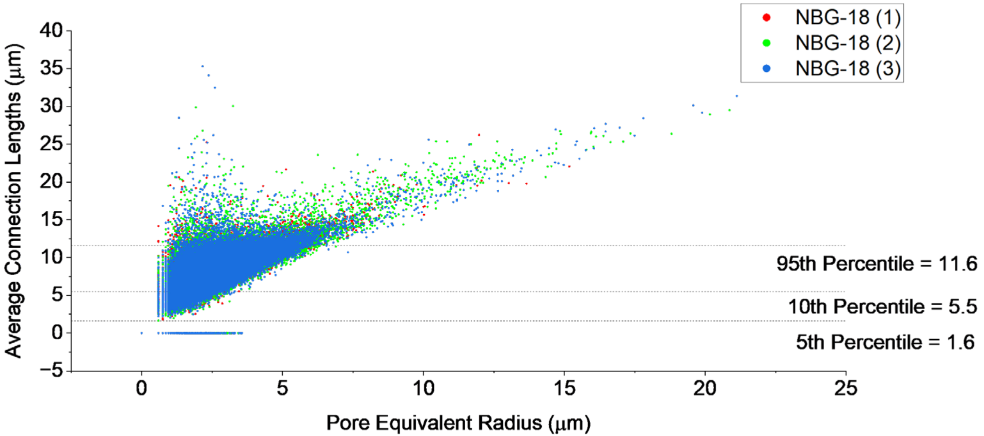


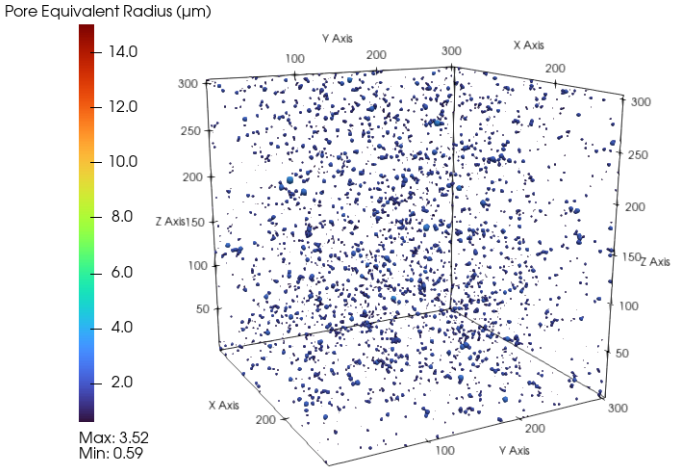


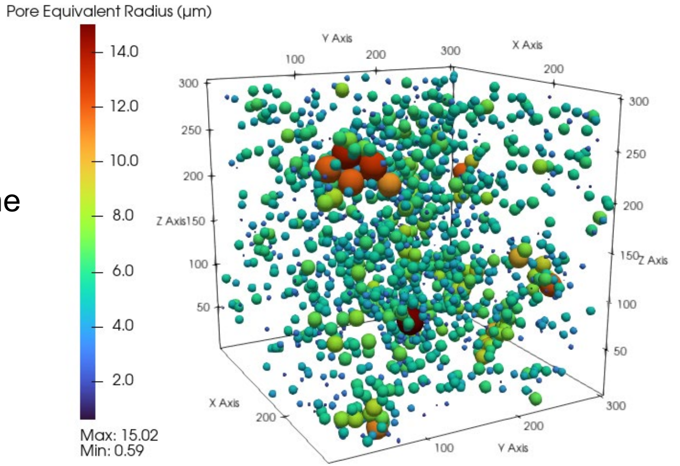


- 10% of all pores are isolated and have a pore equivalent radius <4 microns.
- 5% of all pores have an average connection length >11.6 microns, and the majority of those have an equivalent radius >4.5 microns.
- Both groups of pores are homogeneously distributed throughout the sample, albeit less so than in AXF-5Q.
- The majority of pores (80%) have an average connection length between 5.3 and 11.6 microns.

Correlation Between Pore Equivalent Radius and Average Connection Length Distributions – NBG-18





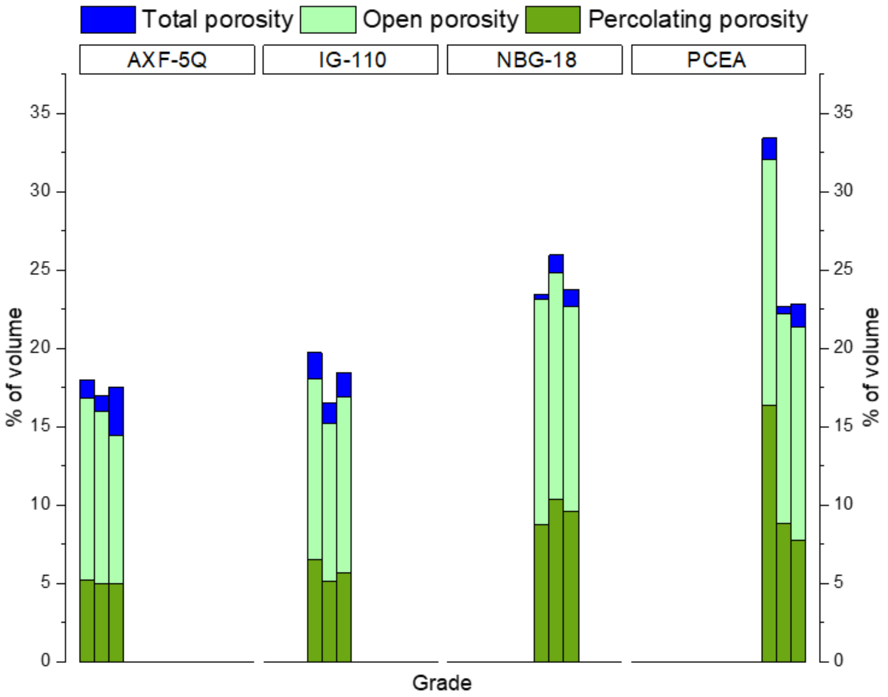


- 5% of all pores are isolated and have an average connection length <1.6 microns with a pore equivalent radius <3.5 microns.
- 5% of all pores have an average connection length >11.6 microns, and the majority of those have an equivalent radius >6 microns.
- Larger pores with longer connections show more clustering compared to previous grades.
- The majority of pores (80%) have an average connection length between 5.5 and 11.6 microns.

Comparative Porosity Characterization

Total, Open, and Percolating Porosities of The Four Grades

- Total porosity increases for larger grain grades.
- Variability between PCEA samples is notable.
- Percolating porosity is slightly less than half of the open porosity in larger-grain grades.



On-going work and Acknowledgement

- A similar analysis is planned for ion-irradiated samples.
- Ion irradiation work has been completed (at the University of Tennessee).

- The work presented here was performed by Dr. Dina Elsayed Elgewaily.

NEUP Nuclear Energy University Program

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Thank you!

1. Introduction

Evaluating Porosity in Nuclear Graphite