Correlating Bulk Properties of Reactor-extracted Gilsocarbon Graphite with Pore Distributions Measured by X-ray Tomography

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Background

• Material properties are controlled by the microstructure

• Pore structures believed to control some material properties
  • Volume, size, shape and connectivity are all important

• Pore structures may be measured by volumetric imaging techniques
  • In 3D many techniques are destructive

• Desire to understand how the pore structure evolution of graphites control their properties
  • Understand current graphites
  • Help to predict the properties of Gen IV+ graphites

Figures: https://tinyurl.com/y453dxa2  https://tinyurl.com/y4xabdzd  https://tinyurl.com/y3zp7lsv  Creative Commons 2.0
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Installed set samples

- AGR installed set cores with weight-loss range 3-8 % (Dose: 18-22 x10^{20} n.cm^{-2} EDN) from Hinkley Point B

- Machined into large beams, fractured and re-machined into 6 x 6 x 19 mm beams
  - Beams selected with highest, median and lowest large beam properties
  - Plain, U-notched and chevron notched

- XCT-imaged in Bruker SkyScan 1273
  - Resolution = 2.97 μm.voxel^{-1}
Blackstone samples

- Ex-AGR samples re-irradiated in Blackstone
  - Mostly Hinkley Point B and Hunterston B material
  - Weight loss: 14-53 %

- Range of geometries
  - Cuboids and cylinders
  - Dimensions in range 6 – 12 mm

- XCT-imaged using the laboratory scanner at NRG, Petten
  - Resolution = 2.50 μm.voxel⁻¹
Aim

To identify and characterise the pore networks from X-ray tomography of graphite, to compare to trends in the physical properties.

The segmentation method should be:

• Robust and work in 3-D
• Have minimal analyst interaction (reducing analyst bias through automation)
• The underlying model should be as simple as possible
• Use open source and well understood tools
• Maximise identification of small pores
• Work for porosities from 0 to over 50 %
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Theory – X-ray tomography

- X-rays absorption is dependent on density and chemical composition

- In X-ray tomography (XCT), transmission maps (radiographs) are captured then reconstructed to 3-D images

- X-ray map is a virtual microstructure, and a pore map if chemistry is constant

- However, a 2-phase material will have a broad range of grey-scale values
Segmentation Theory

- In the tomograph, voxels in bulk material are bright, and pores are dark.

- Want to identify material and pores (binary segmentation).

- Intuitive by eye, but algorithmically difficult to avoid false results.

![Tomographic slice](image)

**Material**

**Pore**

**Binary segmented slice**

Phase 1 (white) = material
Phase 2 (black) = pore

![Area histogram](image)

Frequency: 0 to 255

Greyscale: 0 to 255

Threshold
Segmentation Theory (2)

• Real image is voxelated

• Greyscale distributions are broadened and overlap

• Example broadening effects:
  • X-ray beam hardening
  • Variable density
  • Partial volume
  • Noise

*Simple example – 1 pore, ‘realistically’ imaged*

![Area histogram](image)

- Frequency
- Greyscale
- Decreasing length scale
- Low-pass threshold
Segmentation Rule

- Single-stage thresholding has no suitable value \((t_1)\)

- For multistage process, select \(t_1\) to capture:
  - All pores (no false negatives)
  - Some material as pores (false positives to filter later)

- Can the distribution shape guide the selection?
  - Bi-modal and each \(~\)Gaussian
  - Ratio of the material and pore modes changes with weight loss

Segmentation rule:

\[
t_1 = \text{Material Mode} - k \cdot \Delta
\]

- \(k = 0.30\) (trial and error for a single sample)

Here, an analyst would select \(t_1\) just below distribution mode, but unclear exactly where

\[
\text{Distribution mode} \approx \text{material mode}
\]

Assumption:

Chemistry is constant, so difference in modes \((\Delta)\) is constant, regardless of tomography conditions
$t_1$ determination – real data

Original tomograph  Peak fitting  Initial segmentation (not complete!)
Filtering

After the initial threshold, dataset is filtered using automatic routine, and individual pores characterised

- Coded in FIJI, using MorphoLibJ and 3D IJ Suite
- Basic image tools, applied to obtain 3-D physical results
- Dimension limit to >10 μm

Selecting k (and hence $t_1$) is the main uncertainty in calculating the total pore volume fraction, $f_{TPV}$

- Estimated by considering sensitivity on $f_{TPV}$
- Half the difference over range $f_{TPV}(t_1 - 3) \rightarrow f_{TPV}(t_1 + 3)$
# Segmentation example – installed set

<table>
<thead>
<tr>
<th>Original microstructure</th>
<th>Final segmentation</th>
<th>Positive mask</th>
<th>Negative mask</th>
</tr>
</thead>
</table>

Hinkley Point B Graphite, with weigh loss < 8 % (Porosity < 25 %)
Segmentation example – Blackstone

<table>
<thead>
<tr>
<th>Original microstructure</th>
<th>Final segmentation</th>
<th>Positive mask</th>
<th>Negative mask</th>
</tr>
</thead>
</table>

Segmented with the **same rule** \((k = 0.30)\)

Hinkley Point B Graphite, with 31 % weight loss (43 % porosity)
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Pore properties

- Open pore volume fraction ($f_{OPV}$) is based on the largest detected pore
  - Calculate closed pore volume, ($f_{CPV}$)
    \[ f_{CPV} = f_{TPV} - f_{OPV} \]

- Pores characterised by several measures
  - Only a subset of the closed pores are characterised
  - Measure eg size, surface area, and orientation, sphericity

- Pore components all scale linearly with weight loss
  - Higher weight loss $\Rightarrow$ lower $f_{CPV}$
Property correlation – installed sets

- To examine trends, fracture properties taken as averages of larger beams or cores

- Weight loss range limited compared to uncertainty

- No clear trends with $f_{TPV}$, $f_{OPV}$ or $f_{CPV}$ against any properties
  - Strength and DYM may trend with $f_{OPV}$ component

<table>
<thead>
<tr>
<th>Property</th>
<th>$f_{TPV}$</th>
<th>$f_{OPV}$</th>
<th>$f_{CPV}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Strength</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fracture Toughness</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Crack Initiation</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Dynamic Young's Modulus</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Pore properties - Blackstone

Installed set samples

\[ y = 0.9x + 18.6 \]

\[ y = 1.0x + 16.2 \]

\[ y = -0.0x + 2.4 \]
Property correlation – Blackstone

- DYM [GPa]
- CTE [K\(^{-1}\)]

Graph showing the correlation between Porosity [%] and DYM [GPa], and Porosity [%] and CTE [K\(^{-1}\)].
Limitations

Code is not yet robust for every sample yet and requires post filtering by analyst.
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AGR installed set and Blackstone samples were imaged by XCT prior to fracture

Accompanying PIE measurements were made

Pores segmented by updated semi-automatic method and characterised

Method based on simple statistical rule and applicable to whole series

Lim

Property-porosity correlations examined

Linear trends in $f_{TPV}$, $f_{OPV}$ and $f_{CPV}$ with weight loss

High scatter and few samples makes discerning trends difficult
Next steps

• Improve the high ratio stability of algorithm

• Apply to ex-AGR samples (again)

• Complete enhanced characterisation of available Blackstone samples at NNL

• Understand pore evolution processes

• Correlate further properties
  • Pore size and volume distribution changes
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