Large-Scale High-Resolution P-FIB Tomography of Gilsocarbon

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Overall PhD Aim

Focusing on Gilsocarbon, utilise advanced characterisation techniques to quantify the key structural features of nuclear graphite as a means of developing a mechanistic understanding of the material performance, damage, and deformation, at all length scales during reactor operation.

Presentation Aims

1. Present Plasma-Focused Ion Beam (P-FIB) Serial Slicing Tomography (SST) as a means of quantifying, previously unmeasured, microstructural features in 3-Dimensions.

2. Highlight some of the key hurdles when technique applied to nuclear grade graphites.

3. Suggest how the developing technique can fit into a correlative tomography framework.
Background

• Although there is an extensive property database, microstructural information is limited.

PhD objective: Investigate the link between graphite properties and nano- and microstructural features, and how these features evolve during reactor operation.

• To realise true mechanistic understanding of material behaviour, further knowledge is required on the microstructure at the micron/sub-micron scale.

• How can FIB fit into a correlative tomography framework, allowing for quantification of every microstructural feature present in nuclear graphite?

Comparison of 3D microscopy methods by ‘Volume of Material Analysed’ and ‘Spatial Resolution’. Non-destructive methods represented by dashed lines. Adapted from Burnett et al. (2016), and Cantoni & Holzer (2014)
Microstructural Characterisation

• Virgin AGR samples.
  – Work on reactor and MTR irradiated materials coming at a later date.

• Samples extracted from as-fractured faces.
  – Avoiding damage imbued during sample preparation techniques.
  – Ensures samples are representative of bulk material.
  – Chunk extraction possible due to increased milling rates of Plasma-FIBs.

• FIB Serial Sectioning Tomography (SST) technique allows users to accurately resolve smallest pores/cracks in 3-Dimensions.
  – “Slice and View” inherently destructive technique but ‘high-resolution’
  – In-plane resolution limited by electron column and detectors.
  – Slice thicknesses limited by precision of ion-beam.
SST “Slice and View” Example

As-prepared Gilsocarbon sample.
Cross-section of Coke particle.
Volume approximately 10*10*10 µm.
Prepared by Ga PFIB.
Main Hindrances

1. Curtaining.
2. Formation of **Rippling** artefact.
3. **Trench Shadowing**.
4. Sample **preparation induced damage** bands, distorting near surface microstructure.
5. Algorithmically segmentable **Pore-Material contrast** and 3D nature or porosity/microcracks
Imaging Conditions and Segmentation

Aim: Algorithmically separate porosity from material

- Too many features to process manually, therefore need for algorithmic segmentation.
- Algorithmic segmentation simple in principle but complicated by imaging noise, pore topography, and FIB artefacts.
- Optimise imaging conditions for 'best' pore-material segmentation.
- How can pre-segmentation image processing be utilised to aid segmentation?

**Table:**

<table>
<thead>
<tr>
<th>Pore-Material Contrast</th>
<th>Ease of Algorithmic Segmentation</th>
<th>Prominence of Artefacts</th>
</tr>
</thead>
<tbody>
<tr>
<td>Greyscale Value</td>
<td>Frequency</td>
<td>Set Threshold Value</td>
</tr>
<tr>
<td>Porosity</td>
<td>Material</td>
<td></td>
</tr>
<tr>
<td>Idealised</td>
<td>Histogram broadening due to artificial noise</td>
<td>Low kV SE imaged pore</td>
</tr>
</tbody>
</table>

Figure adapted from M. Jordan et al. (2018)
Larger interaction volume of BSE results in a slightly blurrier image.

But, greater pore-material contrast in BSE images.

Cannot rely on edge lightening in SE images as not always present.

BSE image easier to segment, therefore suited to quantifying porosity over a range of sizes.

SE suggested for work on small volumes, which can be manually processed, looking at smallest pores.
Curtaining

- Inherent due to the spatial variation of sputter rate, caused by porosity, and modulation of current density by uneven surfaces.
- Exacerbated by BSE imaging…
- Worsened by rocking mill…
- More prevalent in P-FIBs…

✓ Can be removed by Fourier filtering

Gilsocarbon cross-section imaged in BSE mode using ICE detector at 5 kV, 50 pA: upper) raw, cropped image, lower) same image post Fourier filtering
Rippling Artefact

• Self organization

• Material dependent
  – Carbonaceous materials particularly susceptible.

• Milling ion dependent
  – Larger discrepancy between incident ion and sample, the more likely rippling will be observed.

• Current density dependent

Typical ion rippling of Xe P-FIB milled cross-section on virgin Gilsocarbon, showing the effect of Pt thickness on ‘clean milling’ and presence of artefact.
‘Large’ Volume 3D Dataset Acquisition

1. CHUNK LIFTOUT
- 72 x 34 x 42 μm chunk extracted from as-fractured face
  - Remove damage caused by sample preparation.
  - Can now image perpendicular to face, removing trench-shadowing & perspective distortion.

2. PRE-ASV PREPARATION
- Chunk attached to Si chip edge, thick Pt layer added, & cleaning of imaged face
  - Protective Pt layer prevents rippling artefact from forming.
  - Mounting on edge gives more degrees of freedom.

3. DATA ACQUISITION
- High-resolution Auto Slice & View (ASV) programme adapted to material
  - Low kV BSE imaging, CBS detector.
  - In-plane pixel resolution of 12.20 nm, HFW of 75 μm, 6144 x 4096 pixels.
  - 20 nm slice thickness.

4. IMAGE PROCESSING
- Tailored workflow allows for near-automated segmentation, in AVIZO®
  - Allow for construction of 3D model.
  - Quantitative analysis of individual pores/microcracks (volume, orientation, elongation) or global pore network (connectivity, & volume %).
ASV slice #010 with zoomed insert: imaged at 5kV with CBS detector. Insert rotated 90° anti-clockwise. Micrograph HFW of 75 microns. 6144 x 4096 pixels gives a theoretical spatial resolution of 12.20nm but actual resolution will be lower due to nature of backscattered imaging.
Deterioration of Pt Layer

- ASV process killed after 20 slices due to deterioration of Pt layer, resulting in the emergence of rippling artefact.

- Potential causes:
  - Pt layer too soft (not enough C%)
  - Uneven top of chunk, inherent from fracture face

- An additional sample was prepared with a flattened top face and ‘superior’ protective layer, but sample lost due to equipment issues.
2.5D Dataset

Image processing and analysis conducted in Avizo. Reconstruction volume 75 x 34 x 0.4 microns
Preliminary Results (data extracted)

- Wide range of pore measurements can be extracted, both local and global.
  - Total pore volume %, Volume, Surface Area, Feret Dimensions (X, Y, Z), Elongation, Flatness, Equivalent Diameter of Fitted Sphere, Eigenvectors, Centre of Mass, Pore connectivity...

- Analysis identified 41914 distinguishable ‘features’, most are noise.

- 803 actual, resolvable, features
  - For this dataset, ‘resolvable’ determined as having a minor chord length of 100nm
In-plane resolution limit?

- Montage/Mapping process of stitching together higher magnification images to a single image.
- 20 micrographs, 5 x 4 array, 10.2 micron HFW, 2kV BSE CBS detector, in total 30,720 x 16,384 pixels.
- Smaller the feature the greater the potential for error.
<table>
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<tr>
<th>Region</th>
<th>index</th>
<th>FeretLength (nm)</th>
<th>FeretWidth (nm)</th>
<th>EqDiameter (nm)</th>
<th>Orientation</th>
<th>Perimeter (nm)</th>
<th>Symmetry</th>
<th>Area (nm^2)</th>
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Conclusions

• Resolving porosity, in 3-Dimensions, at all length scales is imperative for better understanding of material behaviour.

• Once rippling issue has been removed, P-FIB serial sectioning tomography is a suitable technique for resolving porosity/microcracks in 3-Dimensions below the resolution limit of x-ray tomography.

• In-plane resolution limited by the need to image in backscatter mode.

• Slice thicknesses limited by precision of ion-beam (approximately 20nm).
  – Depth resolving by Bayesian statistics may offer a future solution but process currently too computer intensive.

• Multi-level FIB characterisation?

• Next step: Irradiated samples.
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- The views presented here do not necessarily reflect those of NNL, EDF Energy, or Loughborough University.
Thank You For Listening!
Any Questions?
SUPPLEMENTARY SLIDES
Sample preparation damage bands
Casino Monte Carlo. 50,000 simulated electrons. 2kV into pure carbon.