Utilizing Big Data Approaches to Unveil the Structure of Disordered Carbon Materials

Boejesen, E.[1], Liu, A.C.Y.[1], Petersen, T.C.[2] and Etheridge, J.[3]

[1] Monash Centre for Electron Microscopy, Monash University, VIC 3800, Australia, Australia, [2] School of Physics and Astronomy, Monash University, VIC 3800, Australia, Australia, [3] Monash Centre for Electron Microscopy, Monash University, VIC 3800, Australia & Department of Materials Science and Engineering, Monash University, VIC 3800, Australia, Australia

Carbon solids are used in many industrial applications due to their useful properties, abundance, and low toxicity. The structural make-up of the well-ordered allotropes is relatively well understood. A similarly reliable structural elucidation of disordered carbon compounds, despite their industrial importance, still remains elusive. In crystals the structure is correlated over large length scales; each atom is a well-defined distance from its neighbors in an array, and the pattern of scattered radiation from a macroscopic specimen is sufficient to solve the structure of the material. In disordered materials, e.g. activated carbons, the correlation length is much smaller. Broad-beam scattering measurements average over too many different local structural environments to specify a unique structure solution for such materials.

In a previous multi-technique study we were able to document changes in density and average bonding along the activation pathway of a range of activated disordered carbons.[1] However, a more subtle understanding of the structural changes along the activation pathway remained elusive. Here we apply a novel characterization method to the same nano-porous carbons; enabling a more detailed understanding of the highly disordered structure.

In modern scanning transmission electron microscopes, electrons may be focused to form a probe with dimensions matching the correlation lengths found in highly disordered materials. The spatial reduction in probe size breaks-up the otherwise isotropic and diffuse amorphous rings into patterns with discrete diffracted intensities. Angular correlations in these patterns are related to the local structure and symmetry. The angular correlations in such scanning nanobeam electron diffraction (SEND) patterns can be quantified by calculating a set of Fourier coefficients for the periodic intensity around a specified diffraction ring. These Fourier coefficients probe three-body correlations and are more discerning of short range order than the isotropic pair distribution function. Subsequently the Fourier coefficients can be mapped to reveal extended structure within the sample. They may also be compared to theoretical calculations of ideal nearest neighbour clusters with a well- defined point symmetry.[2-5]

Here we present an exciting new approach to SEND data analysis. It has been tested upon large datasets consisting of 10,000 diffraction patterns collected from 25 x 25nm areas of differently activated carbons, using a Titan 80-300 aberration corrected electron microscope operating at 80kV, equipped with a fast-readout diffraction camera. The use of principal component analysis (PCA) and non-negative matrix

factorization (NMF) enables the extraction of subtle features from these large datasets, otherwise hidden upon cursory inspection. The extracted components and loadings are examined to deduce valuable information on the dimensions and orientation of *regions of correlated structure*[6] Our analysis shows we are able to correlate changes in local structure to the physical properties of the different carbons. Finally, additional information on pore volume and layer buckling may possibly be extracted using this newly developed method. Thus, here we demonstrate how big data analysis may enable a deeper understanding of highly disordered solids.

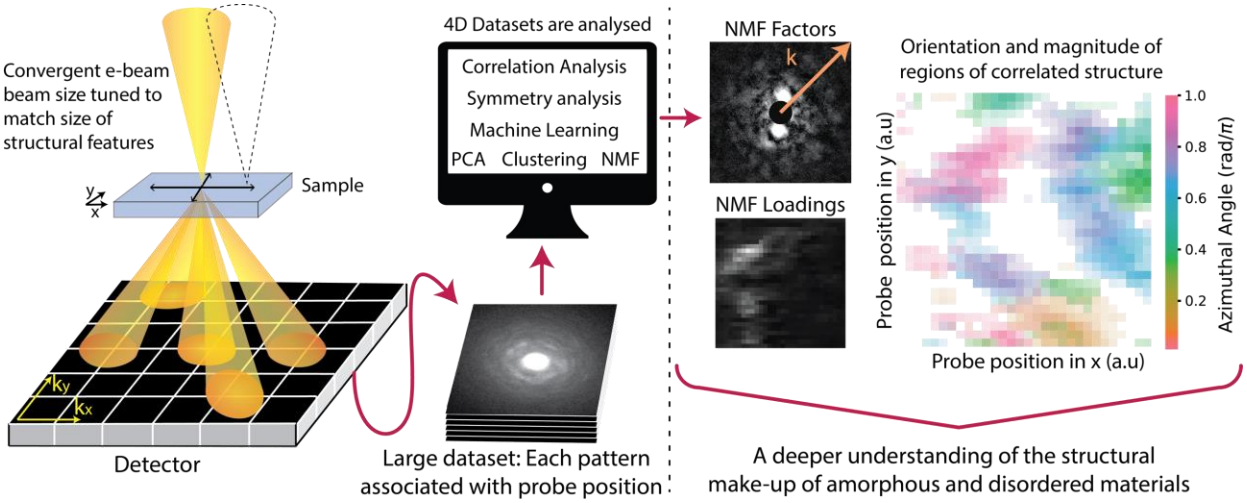

Figure 1 *Large 4D SEND datasets are collected and analysed using a broad range of methods. Big data tools such as NMF and PCA decomposition allow for extraction of otherwise latent features. Regions of correlated structures are revealed and compared to known structural carbon motifs.*

Acknowledgements: This research used equipment funded by Australian Research Council grant ARC Funding (LE0454166). E.D. Boejesen would like to acknowledge funding (VKR023371) from VILLUM FONDEN. "The authors acknowledge use of the facilities and the assistance of Dr. Matthew Weyland at the Monash Centre for Electron Microscopy.

References:

1: Hu, C., Liu, A. C. Y. *et al, Carbon,* **2015**, 85, 119 - 134.

2: Liu, A. C. Y., *et al.*, *Acta Crystallogr Sect Found Adv,* **2015**, 71, 473 - 482.

3: Liu, A. C. Y., *et al., Proc National Acad Sci*, **2017**, 114, 10344 - 10349.

4: Liu , A. C. Y., *et al., Phys Rev Lett,* **2013**, 110, 205505.

5: Liu, A. C. Y., *et al., J Statistical Mech Theory Exp,* **2016**, 054046.

6: Cowley, J. M. , *Ultramicroscopy,* **2002**, 90, 197 - 206.