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EXPERIMENTALLY INFORMED MULTI-SCALE MODELLING OF SIZE EFFECT AND FRACTURE IN POROUS GRAPHITE

Branko Šavija[1], Gillian E. Smith[2], Dong Liu[2], Erik Schlangen[1], Peter E.J. Flewitt[2]

[1] Faculty of Civil Engineering and Geosciences, Delft University of Technology, Delft, Netherlands
[2] HH Wills Physics Laboratories, School of Physics, University of Bristol, Bristol, UK

ABSTRACT

Advanced Gas Cooled reactors operated in the UK are cooled by carbon dioxide gas and contain graphite bricks to moderate the fast neutrons and fulfil structural requirements. These graphite moderator bricks lose mass and become more porous during service due to radiolytic oxidation . Mechanical behaviour of these graphites can be considered as quasi-brittle. As their microstructure is complex and changes over time, computer models can help in understanding the experimental observations. A multi-scale model for porous graphite is presented and the results are compared with PG25 filter graphite, which has a relatively simple microstructure containing 52 vol.% porosity. The mechanical model is based on a microstructural model and micro-mechanical properties of the material were fed directly as input parameters. Specifically, a representative microstructure is created on the scale at which small scale laboratory experiments can be performed (several mm). Three-point bending simulations are performed at different scales to investigate the influence of testing-scale on the measured mechanical response. Modelling results have shown that the average mechanical properties decrease monotonically with the increase in specimen size, and the scatter of the data reduces for larger specimens consistent with experimental observations. In addition, the strain fields and crack patterns are modelled and they fit with experimental observations. The model can, therefore, be regarded as a useful tool in studying the influence of different microstructural parameters on fracture of various nuclear graphites.

INTRODUCTION

Gas cooled nuclear reactors in the UK use graphite bricks which moderate neutrons and provide structural load bearing elements. Brick units are connected by locks and keys, creating a structure which enables movement of control rods and fuel loading/unloading operations (Tsang and Marsden (2006)). Apart from their own weight, these bricks are subjected to various loads occurring as a result of the environment, high temperature, and neutron irradiation. As a result of the interaction with the environment, the microstructure of nuclear graphite evolves as a function of time and exposure. This is primarily related to its strength, porosity, and pore size distribution. The microstructure of nuclear graphite bricks, therefore, changes during the operation as a consequence of neutron irradiation (which increases the strength as a function of the dose of irradiation) and radiolytic oxidation by carbon dioxide (which decreases the material density and alters the pore size and connectivity, thereby reducing the global strength). Mechanical behaviour of porous graphites used in nuclear reactors can be described as quasi-brittle (Moskovic et al. (2013), Moskovic et al. (2014)).,These materials can be considered as having microstructural features which resemble other aggregate containing materials , such as concrete (van Mier (2012)). Similarly to concrete, nuclear graphite does not to exhibit plasticity as it deforms, and any non-linearity in the load-displacement response can be attributed to microcracking. Therefore, techniques developed for simulating concrete fracture can potentially be modified and applied to porous graphite.

In this work, a multi-scale lattice based model was employed to simulate mechanical response and cracking in PG25 filter graphite. This type of graphite is suitable due to its relatively simple microstructure compared to, for example, Gilsocarbon graphite. The microstructure of PG25 graphite

comprises only a graphitized matrix and porosity (52%), without any filler particles common to nuclear grade graphites. As a consequence this material provides a simple basis for comparing model predictions with experimental data . The mechanical model is based on a microstructural model and uses measured micro-mechanical properties directly as input parameters. A representative microstructure is created on the scale at which small scale laboratory experiments can be performed (several mm). Three-point bending simulations are performed at different scales to investigate the influence of testing-scale on the measured mechanical response.

EXPERIMENTAL MEASUREMENTS

Mechanical properties are a necessary input for numerical simulations. These include elastic modulus and fracture strength. To obtain true material properties, measurements need to be performed at the appropriate length-scale. For models used in this work, this is the micrometre-scale. Micromechanical properties of porous nuclear graphites can be obtained by using a novel technique for in-situ testing of micro-cantilever specimens. In this approach, small cantilever specimens of the representative material are prepared and tested in a dual-beam work station (FEI Helios NanoLab 600i) which uses a force measurement system within it. The cantilevers are milled from a region of the PG 25 graphite that have cross- sections in the range of a few μm (2 to 6 μm), with the beam length of 10 to 44 μm, depending on the cross - section size (aspect ratio 5: 1). More details about the technique can be found in Liu et al. (2014).
Measurements on this scale were used as input for the multi-scale model. On the larger scale, a three-point bending experiment was simulated, according to figure 1.

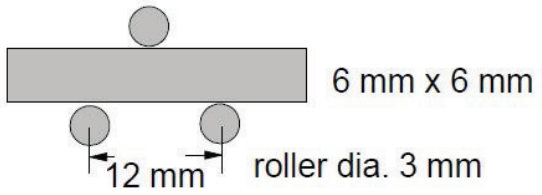


Figure 1. Geometry of the full-scale beam which was used in the simulations (total specimen length is 18mm)

MICROSTRUCTURAL MODEL

As previously stated, filter graphite is simpler than other nuclear graphite materials, with the microstructure consisting of pores and graphitized matrix only. The size of individual pores is relatively small compared to the size of the simulated beam (figure 1), and has been measured to be around 200 μm in diameter. In reality, however, these pores are not spherical. In the microstructural model, the complex porosity was created in the following way: first the number of spherical pores of the average measured size needed to give the target 52% porosity was calculated. These were placed randomly within the model This is shown in figure 2.

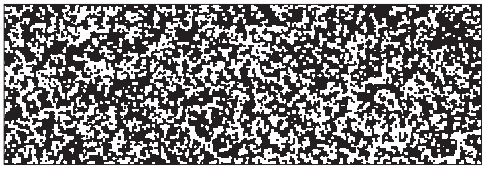


Figure 2. A 2D cross-section of a 3D model filter graphite microstructure, with 52% porosity

In order to employ the multi-scale modeling scheme (as described later), the beam was divided into cubic blocks of 1 x 1 x 1mm in size (in total, 6 x 6 x 18 = 648 such blocks). Due to the random microstructure created, these blocks are not uniform, and have different levels of porosity, depending on their location in the beam specimen. The variation of porosity in small cubes is shown in figure 3.

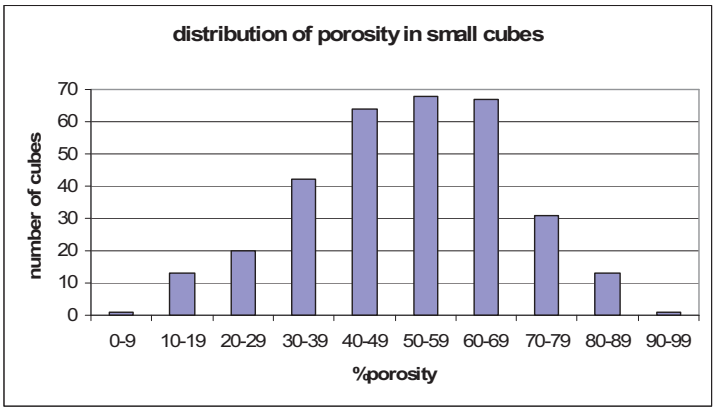


Figure 3. Distribution of porosity across 1mm cubes, "cut" from the beam specimen

MULTI-SCALE MODELLING

Description of the model

In this paper, a lattice-type model was used for the deformation and fracture analysis (Schlangen and Qian (2009), Smith et al. (2013)). In the model, the material is discretised as a set of small beam elements. A regular cubical grid of beam elements, with equal lengths, was used. A set of linear analyses is then performed by calculating the response of the lattice mesh for a particular external displacement. The beam with the highest stress to strength ratio is identified and removed from the lattice network. In each of the analysis steps, a single lattice beam is removed from the mesh, representing the creation of a small crack, and causing a change in the specimen compliance.The analysis is then repeated, with an updated mesh.
In order to capture the microstructural details of porous graphite, the computational effort necessary for a single-scale approach is too high. Instead, in this study, a multi-scale modelling scheme, initially proposed in Smith et al. (2013), was used. The approach is schematically shown in fig. 4.

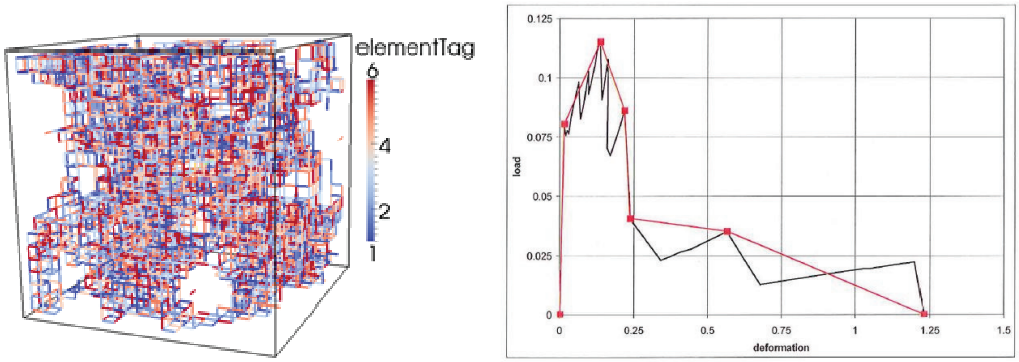


Figure 4. (Left) A small cube, 20x20x20 voxels. The colours represent the type of element (see table 1); (Right) Black line is the load-displacement curve for a small cube (the outcome of the simulation). The red line is a a multi-linear curve with six segments , which represents black line. (adapted Smith et al. (2013)).

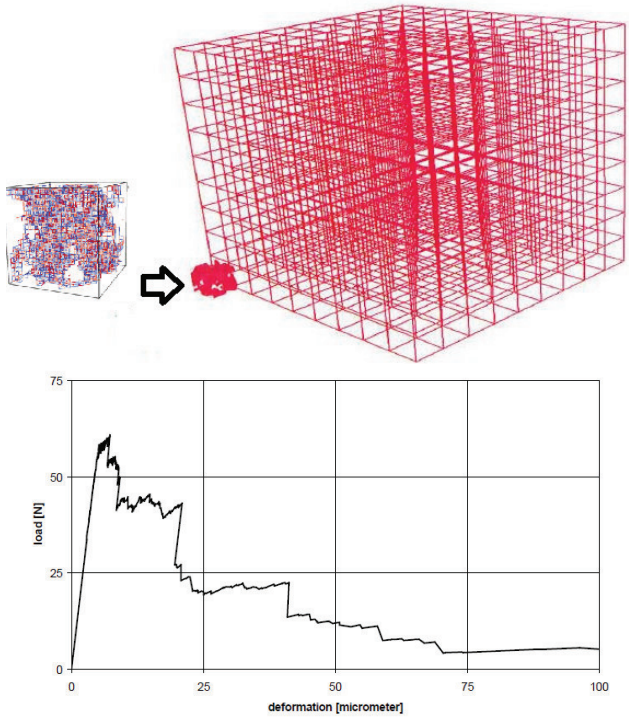


Figure 5. (Top) The detailed mesh on the left represents the small cube (20 x 20 x 20) at its location in the large cube; (Bottom) An example of a load-displacement curve obtained from a simulation on a large cube. (adapted from Smith et al. (2013)).

The multi-scale modelling procedure can be summarized as follows: first, the beam microstructure is divided into a number of small cubes (in this case, each cube was 1x1x1mm, with the total of 648 cubes); then, a direct tension test is simulated on each of these small cubes (in these simulations, beam elements were assigned brittle behaviour), resulting in load - displacement curves, which are then schematised as multi-linear (figure 4). These multi-linear curves for small cubes are used as constitutive relations for elements in the larger specimen (figure 5), which is then tested, resulting in a load-displacement response and cracking patterns of the full-sized specimen (an example is shown in figure 5 right).
In the simulations on the larger scale, the local behaviour of each beam element was, therefore, not brittle. In fact, each element was assigned a multi-linear constitutive relation which resulted by simulating the "local" microstructure on the smaller scale. Consequently, on this scale, the beam elements were not removed in each loading step, but for "damaged" elements, the stiffness and strength would be adjusted according to their respective constitutive relation. This is schematically shown in figure 4 right.

Modelling of porous graphite on different scales

To study the effect of the specimen size on the mechanical response of PG25 filter graphite, the multi-scale modelling approach is used. Mechanical properties obtained from micro-cantilever tests and the microstructure model (see figure 6), both described before, were used as input for numerical simulations.

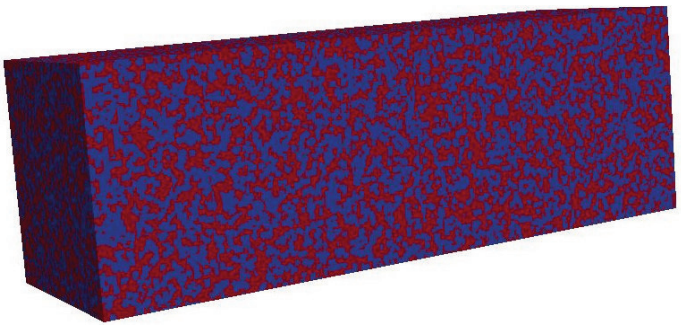


Figure 6. Microstructure of filter graphite with 52% porosity. Pores are displayed as blue, while the graphitized matrix is shown in red. The size of the microstructure is 6x6x18 mm.

The microstructure shown in figure 6 is sliced into small cubes (1x1x1 mm) , which are then tested in a virtual direct-tension test. Their tensile response is then passed on to the higher scale. To take the variability of micro-scale tests into account, properties of one of the six beam types (see table 1) to each of the beams in the small scale simulation, as shown in figure 4.

Table 1: Mechanical properties of beams used in the small scale simulation, according to the micro-scale tests

Beam type	Tensile strength (MPa)	Elastic modulus (GPa)
1	192	10
2	201	21
3	261	16
4	281	11
5	514	25
6	604	35

Due to the different porosity and random distribution of micromechanical properties in the matrix in each of these small cubes, their uniaxial tensile strength varies substantially, as shown in figure 7. This variability underlines the effect of sample size on “measured” mechanical properties of porous graphite.

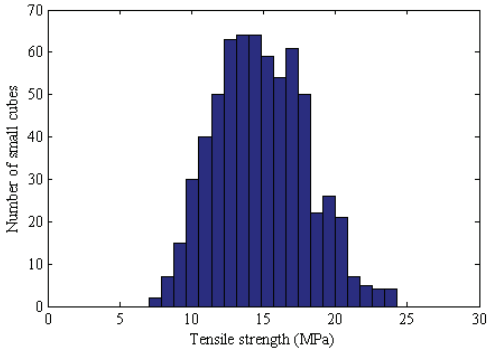


Figure 7. Variability of uniaxial tensile strengths between cubes used in the small-scale simulation

Numerical specimens were created for simulating the size-effects in porous graphite. Specimens of different sizes were created by (randomly) slicing from the full-scale microstructure, which is shown in

figure 6. Specimens of different sizes were subjected to simulated three-point bending tests. In total, five different specimen sizes were tested:

- 0.3mm x 0.3mm x 0.3 mm (6 x 6 x18 voxels from the whole specimen)
- 0.6mm x 0.6 mm x 0.6 mm (12 x 12 x 36 voxels from the whole specimen)
- 1.2mm x 1.2 mm x 3.6 mm (24 x 24 x 72 voxels from the whole specimen)
- 3 mm x 3 mm x 9 mm (60 x 60 x 180 voxels from the whole specimen)
- 6 mm x 6 mm x 18 mm (120 x 120 x 360 voxels), the full-sized beam as shown in figure 6

For the three smallest specimen sizes, a single-scale approach was used (i.e. without the upscaling of properties), which was possible due to their relatively small size. For the last two scales, the previously described multi-scale modelling scheme was used. For each scale, a number of simulations were performed, to assess the variability between individual simulations. The ratio between the support span and the specimen size was kept constant across the scales. The hypothesis was that the specimen-to-specimen variability in terms of mechanical properties should decrease with the increase in specimen size. For all simulations on different scales, fracture energy (G_f) was calculated using the methodology proposed by RILEM (1985):

$$W_f = \int_0^{u_1} P du \tag{1}$$

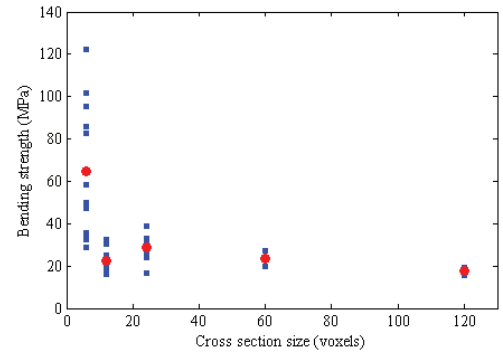
$$G_f = \frac{W_f}{bd} \tag{2}$$

where W_f is the work of force P, b and d are dimensions of the cross section, and G_f the effective (average) fracture energy. The work of force P does not include the linear elastic part of the load-displacement curve so that this represents the energy to form (pre-peak micro-cracking) and propagate (post –peak) the macro-crack .

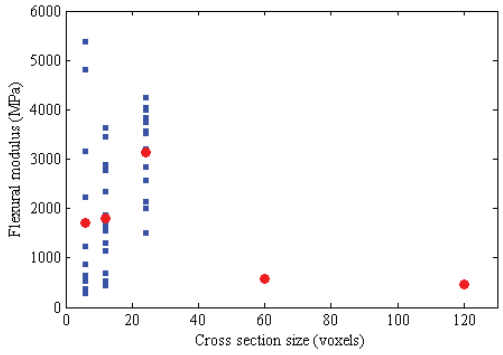
RESULTS AND DISCUSSION

Simulations performed on different scales have revealed a pronounced length -scale effect in terms of bending strength, flexural modulus, and fracture energy (figures 8-10). All of these parameters decrease with increasing test specimen size. It is also interesting that the scatter in simulation results between simulations at the same scale also decrease with the increasing sample size. A similar trend has been reported for experiments. The size effect in terms of fracture energy for porous graphite has not been reported in the literature previously, according to the authors' knowledge. However, it was observed in concrete (Man and van Mier (2008)). For numerical concrete, an increase in fracture energy was observed due to aggregate particles of different sizes sampled on different scales. In filter graphite, the opposite trend is expected, as the size of pores relative to the specimen size changes with scale, instead of the size of aggregate particles. This is exactly what was found in presented simulations (figure 10). Experimental verification is, however, still needed.

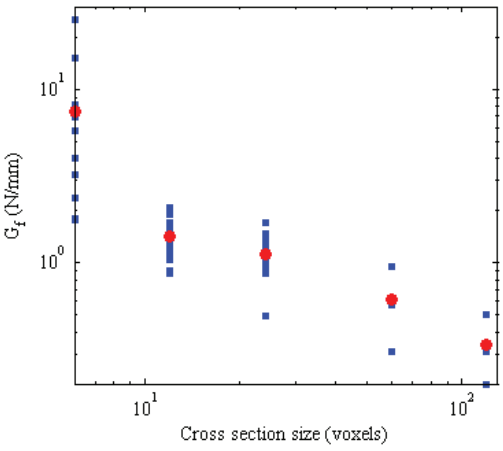
As stated, the variation between individual experiments on the same scale decrease as the scale of observation increases (figures8 a to c). This is important when selecting the specimen size for experimental measurements. This means that, in essence, if a tested specimen is too small, it may result in a gross overestimation of mechanical properties of the bulk material. Specimen size of the graphite material for testing is the main cause of that, as some smaller specimens may have a significantly lower porosity compared to the bulk.



(a)



(b)



(c)

Figure 8. Effect of the size of specimen on (a) the simulated bending strength (blue – all performed simulations; red – average for a scale) (b) the simulated flexural modulus (blue – all performed simulations; red – average for a scale) (c) the simulated fracture energy on a logarithmic scale (blue – all performed simulations; red – average for a scale)

Cracks developing in one of the full-scale simulations are shown in figure 9. On the left-hand side of figure 10, relative local strains in each of the elements in the large specimen are displayed. Cracks at peak (top) and at failure (bottom) are shown. Here the final crack pattern resembles closely the crack pattern

observed experimentally (see figure 12). In some of the full-scale simulations, it was also revealed that the position where initial microcracking devlops in the pre-peak regime is not necessarily the position of the final failure (figure 11 left). A similar phenomenon was previously observed experimentally using the digital image (DIC) technique by Nakhodchi et al. (2013). Clearly, this is a consequence of the pronounced heterogeneity of the porous graphite material.

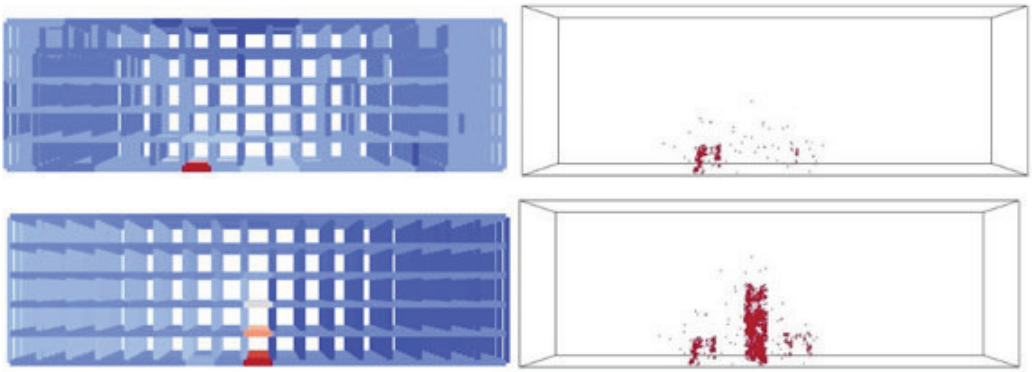


Figure 9. Relative strains in lattice elements (left) and crack patterns (right) in a full-scale three-point bending simulation. Top – at peak load; bottom – at failure.

Figure 10. Experimental crack pattern obtained using a three-point bending test on a filter graphite specimen

Figure 11 displays the stress/strain curves for four simulations performed on the full-scale specimens. The failure mechanism is brittle, with little post-peak deformation in all simulated cases. On this length- scale, differences between individual simulations in terms of load-displacement response are minimal. This results in very small differences of simulated mechanical properties on this scale (see figures 8 a to c). The simulated mechanical properties are of the same order of magnitude as the experimental data. The obtained results show a bending strength of about four times that obtained by experiments. This can in part be attributed to the simplifying assumptions made in creating the material microstructure. The starting point were the spherical pores, which coalesce, creating intricate porosity. In reality, pores are probably more elongated and sharp, resulting in stress concentrations, which reduce the strength.

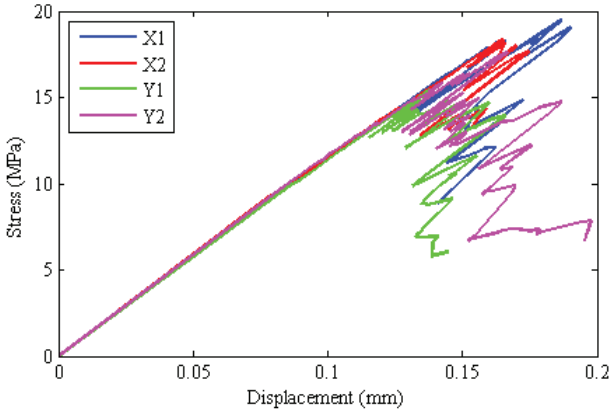


Figure 11. Stress-displacement curves for four full-scale three point bending simulations

CONCLUSION

From the presented results, the following conclusion can be drawn:

- The proposed modelling scheme is innovative in a sense that it uses micro-scale experimental results (i.e. mechanical properties) as input, while the larger scale experiments can be used for validation. No assumptions are made on "real" mechanical properties of the solid phase. This is an important improvement compared to models used in the literature.
- There is a significant scale effect of mechanical and fracture properties. This is in line with experimental data.
- The scatter for both flexural modulus and strength decreases as the specimen size increases. This is also in line with available experimental data.
- The model can predict realistic crack patterns for a filter graphite eg PG25.
- The model showed that, at peak load, microcracking and even some localised cracking sometimes occurs outside the zone of final failure. This corresponds to experiments performed using digital image correlation.

As the obtained mechanical properties are of the same order of magnitude, it is reasonable to conclude that the model can be successfully used in studying deformation and fracture in porous graphite. Future work will focus on more complex graphite types, and effects of irradiation on graphite mechanical properties.

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