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Numerical modelling of the effects of porosity changes on the mechanical properties of nuclear graphite

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Abstract

Finite Element (FE) models based on three-dimensional images from X-ray tomography are used to study the relationships between the microstructure and the bulk mechanical properties of nuclear graphite. The increase in porosity due to oxidation is simulated by setting different thresholds to the grey-scale of the images. The numerical predictions seem to follow the Knudsen law and show good agreement with experiments.

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1. Introduction

Graphite is used as a structural material as well as a moderator in nuclear reactors. During operation, graphite suffers from irradiation-induced damage to its microstructures, thus resulting in changes to its mechanical properties. In some gas-cooled reactors, the graphite components are also subjected to radiolytic oxidation which further undermines their structural integrity. The design and safety assessment of nuclear graphite components is often based on empirical rules derived from material irradiation data. With the accumulation of operational experience, such rules have been shown to be inadequate in some cases and a more fundamental understanding of the relationship between changes in microstructures and bulk mechanical properties would be useful.

The rapid development of computer science and image processing technologies in recent years has resulted in new techniques for material characterization. In particular, the use of detailed finite element models, created from images of the microstructures of materials, allows a mechanistic understanding of the relationship between microstructures and bulk mechanical properties. There is thus the possibility of using such a technique to supplement and/or extrapolate existing experimental database. Originally developed in medical science and biomechanics

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in the 1990s [1], the technique makes use of 3D images obtained from micro-tomography scans.

The aim of this work is to explore the potential of the modeling technique described above for analyzing graphite microstructures. The fine grained isotropic graphite Gilsocarbon, used in the UK advanced gas-cooled reactors, forms the subject of the present study. The specific objective is to verify some of the empirical rules which govern the relationship between the mechanical properties and the microstructural changes induced by thermal and/or radiolytic oxidation.

2. Image analysis and mesh generation

The Simpleware [2] commercial package was used to create detailed 3D meshes from high-resolution tomographic images. Since nuclear graphite is a polycrystalline material composed of three main phases (around 20% of porosity, 65% of filler coke particles and a coal tar-pitch binder between the particles), the first step in creating the Finite Element mesh was an image processing step using ScanIP^{TM} [2] to separate the different phases by setting suitable thresholds to the grey-scale of the raw images (Fig. 1(a)).

This was the so-called image segmentation process and had to be treated with care to ensure that the correct amount of porosity, say, was generated. Indeed, certain details could be lost through the use of incorrect thresholds, leading to a wrong evaluation of the actual composition. In this preliminary study, the increasing percentages of porosity induced by oxidation in the graphite were simulated by adjusting the greyscale threshold to different arbitrary values. This technique was used to study the evolution of the mechanical properties with increasing porosity in a sample of isotropic nuclear graphite. For simplicity, no distinction was made between the filler particles and the binder.

By using the ScanFE^{TM} package [2], a voxel-based FE mesh was then created (Fig. 1(b)). This software automatically generated an optimal mesh with either tetrahedral elements only or a combination of tetrahedral and hexahedral elements. The latter was useful in refining the model, especially profiles near the pores. However, this option could lead to a significant increase in the number of elements and, hence, the computing time. The final mesh was then exported to the ABAQUS finite element software [3] for analysis.

3. Determination of mechanical properties

3.1. Young's modulus

The density ρ of a material, the porosity ratio η, and the fractional weight loss x due to oxidation are linked by the following relationships:

$$x = 1 - \frac{\rho}{\rho_0} = 1 - \left(\frac{1-\eta}{1-\eta_0}\right), \tag{1}$$

where the subscript '0' denotes values of virgin graphite.

Knudsen [4] has proposed a relationship between Young's modulus (E) and the porosity fraction of a porous material by an empirical expression known as the Knudsen law:

$$E = E_{(\eta=0)} e^{-b\eta}. \tag{2}$$

In this equation, b is an empirical parameter determined by experiments. Buch [5] has shown this parameter to be dependent on the pore shape ratio a/c:

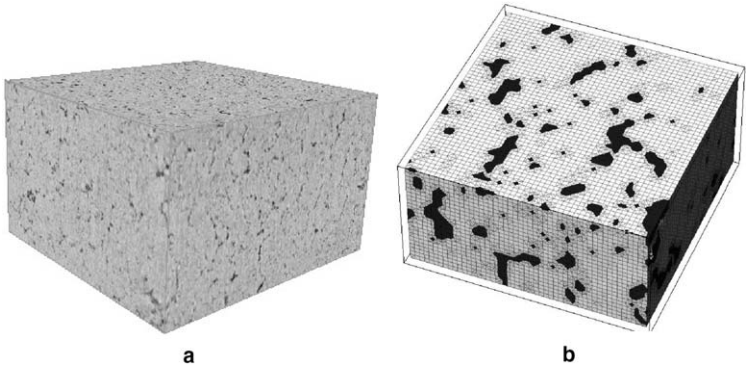

Fig. 1. (a) Reconstructed 3D tomographic image ($14 \times 14 \times 9$ mm^3) and (b) finite element mesh ($2 \times 2 \times 1$ mm^3).

$$b = 1 + 0.594\frac{a}{c}, \quad (3)$$

where a is the dimension in the major axis, and c is the dimension in the minor axis.

Kelly et al. [6] measured properties such as Young's modulus, strength and thermal resistivity of Gilsocarbon graphite subjected to radiolytic oxidation. In relation to the Knudsen law (2), they found a value of $b = 3.6$ for weight losses ranging from 0% to 30%.

Brocklehurst and Adams [7] simulated the effect of radiolytic oxidation by drilling holes into graphite specimens which were then tested to determine their mechanical properties. They found a value of $b = 2.8$ for weight losses ranging from 0% to 50%.

In the present study, Young's modulus was predicted for graphite with different porosity ratios between 0% (perfect crystal) and 45%. The basic input data was obtained from nano-indentation experiments, carried out at the Manchester School of Materials, which provided an average value of $E_0 = 15$ GPa for the 'pore-free' graphite crystal.

A computed tomography (CT) scan of Gilsocarbon was converted to a Finite Element mesh for a cube of 8 mm^3 in volume, which was then extended to 64 mm^3 by imposing symmetric boundary conditions in the x–y, y–z and z–x planes. The mesh consisted of about 100 000 eight-node cubic elements, with each voxel of the original image having a length of 42 μm. Tensile loading was applied in each direction and Young's modulus was calculated using the average stress and strain thus produced. The time of calculation was around 1 h on an Onyx 300 SGI server with 32 processors running at 600 MHz, and 4 GB of memory was required.

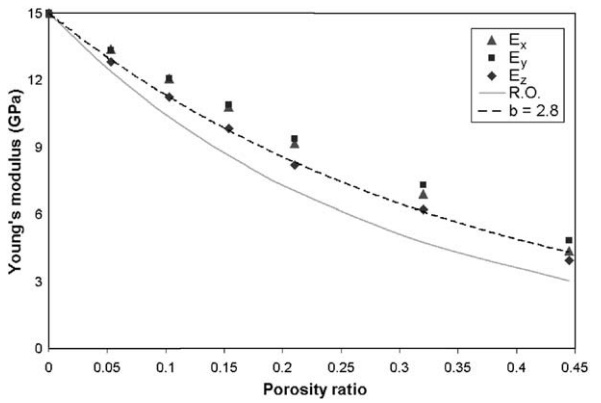

Fig. 2. Evolution of Young's modulus with increasing porosity.

Fig. 2 shows the predicted Young's modulus of graphite in each direction x, y and z for different porosity fractions ranging from 0% to 45%.

It can be seen that the numerical predictions follow closely the Knudsen law with the empirical parameter being based on the hole-drilling experiments carried out by Brocklehurst and Adams [7]. Larger discrepancy exists between the numerical solutions and the measurements made on radiolytically oxidized (R.O.) graphite [6]. It can also be seen that there is a small difference (about 10%) between the predicted Young's moduli in the different directions, due probably to the random distribution of the porosities.

3.2. Tensile strength

In the absence of suitable data, an artificial failure model in the form of a non-linear inelastic stress–strain curve was assumed for pure graphite crystals in order to assess the effects of oxidation on the tensile strength of nuclear graphite. The material was assumed to behave linear-elastically up to a certain yield point. After a very short yield zone, the stress reduces linearly to zero with increasing strain, at which point a crack was assumed to form.

In order to control the geometric (large displacements) and material (stress–strain) non-linearities that appear after the initiation of failure, the non-linear system of equations was solved at each increment by ABAQUS using the Newton–Raphson method. The apparent load was assumed to be applied quasi-statically, so that the incremental algorithm is implicit. On average, 30–40 increments were necessary to obtain a converged solution.

The volume of the model considered was 1 mm^3, with about 13,000 C3D8R elements [3] which use a reduced integration scheme. Fig. 3 shows an example of the stress distribution within the model under load, showing several failure initiation sites. A mesh sensitivity study indicated that the results were strongly mesh-dependent. For a fixed volume, the most stable and most accurate solution was obtained with the use of 20-node cubic elements with reduced integration. However, such a model required significantly longer calculation times than those using 8-node elements.

Fig. 4 shows the stress–strain curves obtained with increasing weight loss, again induced artificially by changing the threshold of the grey-scale. These curves show softening of the material as

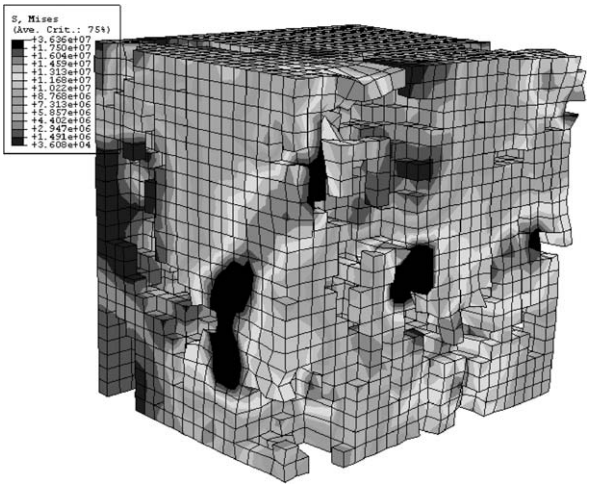

Fig. 3. Cut-out of model ($1 \times 1 \times 1\ mm^3$) showing von-Mises stress distribution after the initiation of failure.

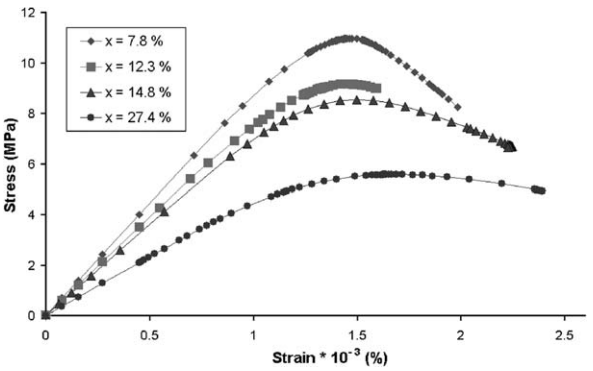

Fig. 4. Stress–strain curves with increasing weight loss.

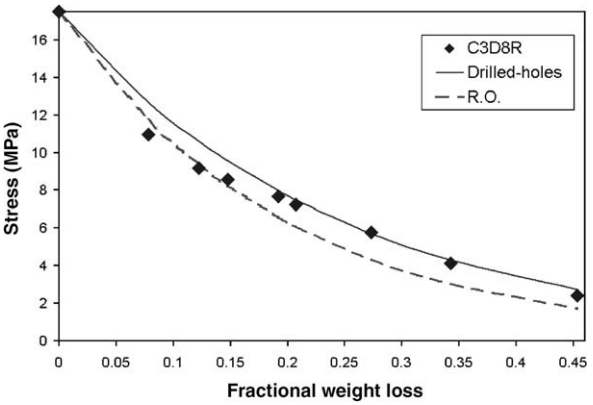

Fig. 5. Evolution of the critical tensile stress with increasing weight loss.

weight loss increases. The critical stress at which failure initiated decreased with increasing weight loss, as shown in Fig. 5.

The obtained numerical values are compared with the hole-drilling and radiolytic oxidation (R.O.) experimental mean curves reported in [6, 7]. Good agreement between numerical predictions and experiments can again be seen in Fig. 5.

4. Discussion

The present study demonstrated the capability of using FE models of microstructures created from 3D tomographic images to evaluate the mechanical properties of nuclear graphite with increasing amount of porosities. Changes in the porosity ratio, or weight loss, due to oxidation were simulated by setting the different thresholds for the greyscale of the images.

The softening behavior predicted for graphite needs experimental verification, and a more robust numerical failure model for predicting damage initiation will have to be developed. In particular, the mesh sensitivity of the results needs to be minimized. This, together with the very different pore geometries involved, means that the apparent good agreement between numerical predictions and experiments for the failure stress is perhaps fortuitous.

The issue of length-scale, i.e., property dependence on the volume of material analyzed, will also need to be explored, but increasing the model size will make the highly non-linear analysis very time-consuming. The volume of material that can be analyzed is limited by the number of elements of the FE model, which in turn is determined by the number of voxels within the 3D image. Too small a volume may cause the results to be unrepresentative. One can increase the volume of material to be analyzed by reducing the mesh density of the model, i.e., reducing the resolution of the image. However, this has the disadvantage of losing important details of the microstructures.

A quantitative study of the effects of pore shape and orientation on the mechanical properties is in progress. Further work will also consist of refining the models by considering the filler coke particles as a different phase from the binder.

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