Investigating the Effects of Stress on the Material Properties of Graphite Moderators using Confocal Laser Microscopy

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INVESTIGATING THE EFFECTS OF STRESS ON THE MATERIAL PROPERTIES OF GRAPHITE MODERATORS USING CONFOCAL LASER MICROSCOPY

Joshua E. L. Taylor, Graham N. Hall and Paul M. Mummery

A detailed understanding of the properties of graphite is required to ensure its safe and continued use as a moderator, and as a structural component, in British nuclear reactors. Considerable stresses are generated in graphite components during reactor operation, which can affect the reactor's ability to cool the fuel and shut down. Therefore, graphite's response to such stresses must be understood. The behaviour of the pore structure and Young's modulus are of particular interest, due to their importance to the strength and integrity of the material. A confocal microscope was used to image samples while they were axially stressed, allowing three-dimensional surface profiles to be produced. By taking images at varying levels of stress, changes to the microstructure of the material were observed. Digital image correlation was performed on the micrographs to produce strain maps. These observations formed the basis of an explanation of the behaviour of graphite bricks in nuclear reactors, in response to loading stresses.

INTRODUCTION

The UK currently has sixteen nuclear reactors in operation, of which fourteen are Advanced Gas-Cooled Reactors, a second generation reactor type that uses graphite for its moderator. The AGR design is largely unique to the UK, and the use of graphite as a moderating material has led to research into its material properties and its behaviour when exposed to the conditions of a working nuclear reactor. Although many of its properties and responses to reactor conditions are well documented – otherwise the material would not have been cleared for use – there are still behaviours and characteristics that are not fully understood. Through further study it may be possible to gain a greater understanding of graphite as a moderating material and allow a rethink

Nuclear Graphite Research Group, School of Mechanical, Aerospace and Civil Engineering, The University of Manchester, Manchester, UK, M13 9PL

into how it is utilised in current and future reactor designs.

Of particular interest is the response of the material properties of graphite to stress. Graphite components are exposed to considerable stresses in working reactors, generated from a wide variety of mechanisms such as dead weight, temperature discontinuities, irradiation, and irradiation creep. A reactor can also be exposed to externally generated stresses such as an earthquake or an accident during operation, as discussed by Yoda et al. [1]. Such stresses may damage or deform the graphite, reducing its efficiency as a moderator and as a structural component. In severe cases the safety of the reactor itself may be compromised, for example by graphite components deforming and blocking the channels that the control rods travel down when controlling the rate of fission. Therefore, it is desirable to have as detailed an understanding as possible about the stress-induced behaviour of graphite to ensure the safe and efficient operation of the reactor.

The most significant types of stress that affect the behaviour of graphite are thermal stresses, arising from temperature discontinuities between different regions of the moderator; and irradiation induced stresses. Both of these mechanisms have been the subject of numerous studies, and the effects of these stresses are reasonably well understood. By comparison, there has been relatively little research into the effects of loading induced stresses on graphite moderators. While the effects of such stresses are not as significant as the aforementioned temperature and irradiation stresses, it is still desirable to understand how graphite components are affected by loading stresses.

To address this, a series of experiments to investigate the damage and deformation behaviour to the microstructure of various reactor grade graphites by physical loading were performed. A number of reactor grade graphites were axially compressed to varying degrees during imaging with a confocal laser scanning microscope. By taking a series of images at progressively higher stresses, the deformation of the surface microstructure was observed and quantified. Changes to the open pore structure were studied in three dimensions and digital image correlation was performed to produce deformation and strain maps for the samples.

STRESS IN GRAPHITE MODERATOR COMPONENTS

The total strain acting upon a graphite moderator is comprised of seven main components, as defined by Tsang and Marsden [2]:

$$\epsilon^{total} = \epsilon^{e} + \epsilon^{pc} + \epsilon^{sc} + \epsilon^{dc} + \epsilon^{th} + \epsilon^{ith} + \epsilon^{idc} \quad (1)$$

Where ϵ^e is the elastic strain, ϵ^{pc} is the primary creep strain, ϵ^{sc} is the secondary creep strain, ϵ^{dc} is the dimensional change strain, ϵ^{th} is the thermal strain, ϵ^{ith} is the interaction thermal strain and ϵ^{idc} is the interaction dimensional change. The interaction dimensional change term is not always included in constitutive strain equations, as discussed by Tanner et al. [3]. For investigations into how loading stresses affect graphite, the elastic strain term ϵ^e is important.

In graphite, two main mechanisms have been identified by Jenkins et al. [4], Smith [5] and Sutton and Howard [6] that underpin the deformation behaviour and ultimately the failure of graphite as a result of stresses: microcracking and movement of dislocations. Microcracks can form in graphite components, or combine together if close enough in proximity and alignment, to form large scale fracture paths which lower the strength of the material. Dislocations can build up as a result of atoms being forced out of position from the crystal lattice and can allow parts of the lattice to slip, resulting in plastic deformation. Both of these behaviours contribute to the deformation of the microstructure of the graphite components.

Exposing the graphite to radiation causes further growth of defects within the crystal structure. High energy neutrons bombard the graphite components and, if enough energy is transferred, can knock carbon atoms out of the crystal lattice. Many of these atoms will fall back into the lattice, but Thrower [7] identified two inter-lattice structures which may form instead. Semi-stable clusters of atoms may form between crystal planes, known as ‘interstitial loops’; or groups of empty atom sites may collapse parallel to the planes, producing an empty ‘vacancy loop’. These dislocations allow slip to occur at lower stresses than in a perfect crystal lattice and reduce the strength of the material.

The stresses present in nuclear reactors damage the graphite components causing degradation of the material. This is of particular concern late in the reactor’s lifespan when components have suffered more damage and are more susceptible to failure. Therefore it is important to understand the response of graphite to reactor stresses in order to ensure that existing reactors are not operating in a potentially dangerous manner, and to optimize the long term quality and efficiency of graphite reactor components in the future. This issue is particularly relevant for the UK’s nuclear industry, given the large investment in graphite moderated reactors and the significant numbers of reactors operating beyond their original design lifetimes.

IN-SITU CONFOCAL LASER MICROSCOPY ON GRAPHITE

Confocal laser microscopy is a method of acquiring three-dimensional surface profiles of topologically complex materials. It is the ideal imaging method for this investigation since graphite is a highly porous material and the laser can penetrate the open pores - pores that reach the surface of the sample - to a reasonable depth. This technique is known as optical sectioning. The microscope can take into account changes to the sample that occur outside of the original surface plane, for example the sample bending as it is stressed. It also allows three-dimensional analysis of changes to the material that would not be possible with traditional microscopes, such as the study of variation of pore volumes.

To optimise the samples for imaging, the samples were polished. Polishing provides a flat, defect-free surface that reflects a higher proportion of light rather than scattering it. By smoothing the samples with progressively finer abrasives, up to a grit of 4000, small imperfections were removed from the surface of the sample.

Each sample was prepared for imaging by hand polishing, using progressively finer grades of polishing paper, to a grit of 4000. Further polishing was performed using felt polishing pads and diamond compound, removing even finer surface imperfections. A highly smooth surface was obtained, with care taken during polishing to ensure no preferential directionality was introduced to the sample's surface. Samples were cleaned using deionised water, followed by 20 minutes in an ultrasonic bath containing acetone to remove any remaining diamond paste or other impurities from the deep pores in the samples.

A sample of PGA graphite was placed into a Deben Microtester rig capable of applying loads of up to 5 kN. The extrusion direction of the anisotropic graphite was oriented perpendicular to the direction from which the force was applied. The sample was subjected to three loading and unloading cycles, reaching 45%, 75% and 95% of the expected failure load. The sample contraction was measured in real time and periodically the loading or unloading was paused to allow imaging of the sample by the confocal microscope. A graph showing applied force against contraction length for all three cycles is shown in Figure 1.

In the first loading cycle, sample contraction was found to occur linearly after an initial 'bedding-in' period of around 0 – 200 N. When the load was removed, the sample did not fully recover and permanent sample contraction of around 0.1 mm was observed. Therefore, some of the deformation at these relatively low stresses had permanently damaged the microstructure of the graphite. However, the subsequent loading to

significantly greater loads does not cause any permanent contraction, and the sample deformation appears to be fully recoverable. This suggests that there are two distinct types of sample contraction occurring in the PGA sample:

- Fully recoverable deformation that, once any compressive forces have been removed, the components exhibit no permanent changes in the direction that the force is applied. This was observed at high levels of compression.
- Non-recoverable deformation, where compression permanently deforms certain components in the sample; and removing the force is insufficient for fully recovering the damage. This was found to occur only at forces below 1 kN. It is not clear whether components affected by this form of deformation are partially recovering or not recovering at all.

During the loading and unloading process, micrographs were recorded using the confocal microscope to show the effects of the applied stress upon the surface of the samples. Figure 2 shows a subsection of the graphite sample at four different applied loads, magnified by a factor of 10. Regions where there is visible damage being caused to the surface of the material are highlighted with circles. At this magnification, the majority of the visible damage occurs in the open pore structure at high stresses. The data in Figure 1 suggest that this damage is recoverable.

The majority of the microstructural changes are, however, only visible at significantly higher magnifications. Therefore the stress-induced changes are better analysed using quantitative rather than qualitative methods.

QUANTITATIVE ANALYSIS OF CONFOCAL MICROSCOPY DATA

By thresholding the images using appropriate software, it is possible to quantitatively study the behaviour of the individual components of the graphite. For example, changes to the sizes and volumes of the open pores can be calculated by segmenting the pore regions.

Figure 3 shows a frequency plot of pore area at the surface of a number of samples of PGA graphite. Bin sizes were 2 μm^2. Table 1 shows how the pore sizes varied by percentage, starting from the smallest pore.

TABLE 1- Data on pore sizes in PGA graphite. 13630 separate pores were identified. Cumulative pore percentage values start from the smallest pore observed and increase in size.

Cumulative Pore Percentage	No. of Pores	Size [μm^2]
10	1363	< 1.3
20	2726	< 2.1
30	4089	< 3.1
40	5452	< 4.2
50	6815	< 6.5
60	8178	< 9.7
70	9541	< 13.8
80	10904	< 47.0
90	12267	< 206.4
100	13630	< 29400

As the pore size increased, the frequency in which they were observed on the surface of the sample was found to decrease almost asymptotically. Hence, for ease of analysis, the data was plotted on logarithmic axes, as shown in Figure 3. Equation 2 shows the power law that was found to fit the data reasonably well up to pore areas greater than 1000 μm^2, and this region represents 98.4% of the data. t is the total number of pores in a bin of size 2 μm^2 centred on a.

$$t = 5640\, a^{-1.146} \quad (2)$$

This process was repeated as different loads were applied to quantitatively study the changes observed in Figure 2.

The same analysis can be performed using three dimensional data, with the caveat that greater magnifications are required to image the pores accurately, so the area that can be scanned each time is smaller. The confocal microscope's imaging software is capable of calculating volumes and surface areas of three dimensional structures, such as open pores. This is accomplished by defining a region of interest around a pore at the surface level and a horizontal plane representing the top of the pore, and calculating the

visible pore volume within these limits. This work was ongoing at the time of publication.

STRAIN MAPPING OF DEFORMED GRAPHITE

Digital image correlation is a technique that allows the tracking of two dimensional changes between pairs of images. The technique can be used to produce 'strain maps', a visual representation of the deformation of the microstructure that occurred as a result of the application of stress to the samples.

A cross-correlation function is defined which quantifies the degree of similarity between two equally sized areas. By selecting an area in the image of lower/zero stress and running the cross correlation function on the higher stress image, the equivalent section of the image can be located and the displacement of the region can be defined by a vector. By repeating the process across the whole image, a full deformation map can be produced for the pair of images. This will take the form of a vector field. Once the deformation of a specific subsection of an image has been calculated, it is also possible to calculate the strain acting upon that part of the sample.

The Young's modulus of a material is a measure of stiffness and is a useful parameter for material characterisation. It is defined as the ratio of stress to strain along an axis, and values of the localised Young's modulus can be calculated using the strain maps and the total applied stress. Strains can be quantified for specific regions of the strain map, so by dividing the applied stress by the strain present in a region, a value of Young's modulus for this local region can be calculated.

CONCLUSIONS

Confocal laser microscopy is particularly well suited for imaging materials during the application of stresses due to its ability to record high resolution three dimensional images. The technique was applied successfully to the study of graphite's response to loading stresses, and enabled both a qualitative study through observation of micrographs, and a quantitative study through calculation of pore sizes, volumes and displacement fields.

Samples of PGA graphite were scanned by a confocal microscope during application of axial stresses. An understanding of the samples' responses to stress in terms of sample contraction, recoverable and non-recoverable damage and pore structure deformation has

been gained, and with experimental data still being collected - and new types of analyses still being performed - this understanding is likely to continue to increase into the future.

Through analysis of samples of different grades of graphite, including those machined in different orientations with respect to the extrusion direction for anisotropic materials, it is hoped that a full model of the response of bulk graphite moderating components to loading stresses can be produced. This will enable better decisions to be made regarding end-of-life extension of existing fission reactors and the design of novel Generation IV and fusion reactors.

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FIGURES

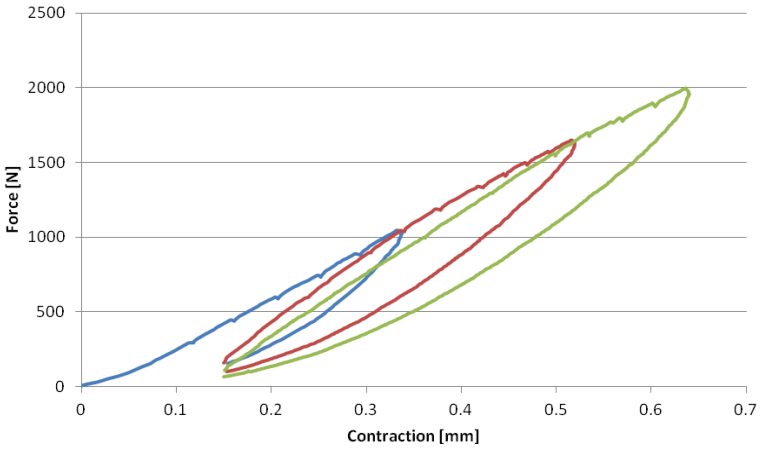

FIGURE: 1 - A graph showing the applied force against the absolute sample contraction for three loading-unloading cycles. During unloading, the force was reduced to a token non-zero force (as opposed to fully removing the force) to prevent the sample from moving freely.

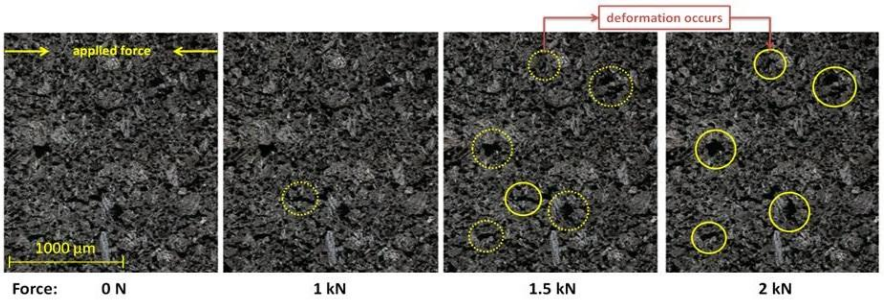

FIGURE: 2 - Images showing a subsection of the surface of the graphite sample at four levels of load. From left to right: 0 N, 1 kN, 1.5 kN and 2 kN. The load was applied from left to right. Regions where the visible deformation has occurred are highlighted with circles.

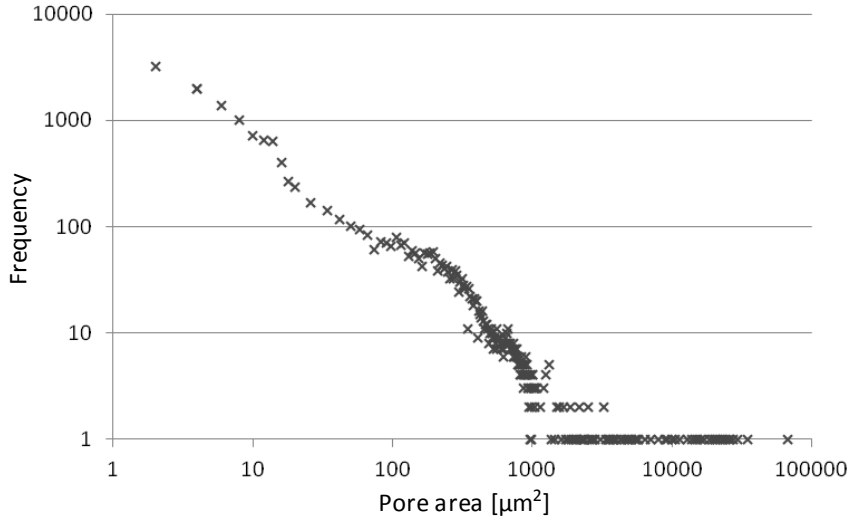

FIGURE: 3 – Frequency plot of pore area at the surface of a number of PGA samples. 13630 pores were identified across all samples. Data were grouped into bin sizes of 2 μm^2.