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Investigating the effects of stress on the pore structures of nuclear grade graphites

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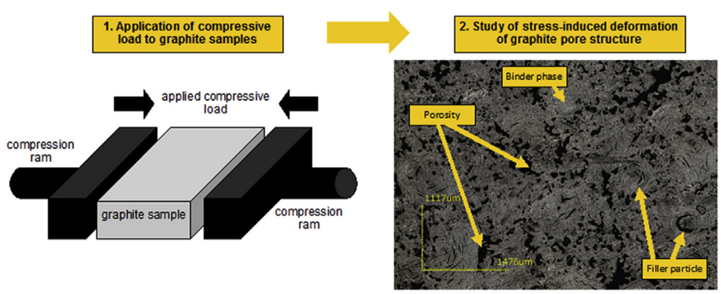
HIGHLIGHTS

- Effects of stress on pore structures of Gilsocarbon and PGA graphites were studied.
- Application of a compressive load was used to generate stresses in graphite.
- Inverse linear relationship between stress and pore area was observed.
- Mean pore eccentricity increased, clockwise pore rotation observed.
- Separation of pores quantified using Voronoi and 'nearest-neighbour' methods.

GRAPHICAL ABSTRACT



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ABSTRACT

Graphite is used as a moderating material and as a structural component in a number of current generation nuclear reactors. During reactor operation stresses develop in the graphite components, causing them to deform. It is important to understand how the microstructure of graphite affects the material's response to these stresses.

A series of experiments were performed to investigate how the pore structures of Pile Grade A and Gilsocarbon graphites respond to loading stresses. A compression rig was used to simulate the build-up of operational stresses in graphite components, and a confocal laser microscope was used to study variation of a number of important pore properties. Values of elastic modulus and Poisson's ratio were calculated and compared to existing literature to confirm the validity of the experimental techniques.

Mean pore areas were observed to decrease linearly with increasing applied load, mean pore eccentricity increased linearly, and a small amount of clockwise pore rotation was observed. The response to build-up of stresses was dependent on the orientation of the pores and basal planes and the shapes of the pores with respect to the loading axis. It was proposed that pore closure and pore reorientation were competing processes. Pore separation was quantified using 'nearest neighbour' and Voronoi techniques, and non-pore regions were found to shrink linearly with increasing applied load.

1. Introduction

Since the construction in 1942 of Chicago Pile 1, the world's first nuclear reactor, graphite has been used for the manufacture of moderating components for a number of types of fission reactor. It

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was adopted for use in British Advanced Gas-Cooled Reactors (AGRs) and Magnox reactors, as well as in Russian RBMK reactors and various designs of High Temperature Reactors (HTRs). Graphite is also a candidate moderating material for next-generation Very High Temperature Reactors (VHTRs). The material is well suited for neutron moderation due to its high moderating efficiency [1], reasonable atomic weight, abundance and relatively low cost.

A wide variety of grades of graphite have been manufactured for use as a moderator in fission reactors. Two graphites of particular interest in the United Kingdom are Pile Grade A (PGA) and Gilsocarbon. PGA graphite has been used in Magnox reactors, while Gilsocarbon is used in AGRs. Both are comprised of a carbonaceous filler material and a binder phase to bond the filler material together [2]. Gilsocarbon is manufactured to be near-isotropic, while PGA has a degree of anisotropy due to the presence of elongated filler particles that align in one direction due to the extrusion process during manufacturing. The alignment in PGA graphite means its physical properties vary in different directions. Herein, samples with the extrusion direction, or grain direction, oriented perpendicular to the orientation being studied are referred to as PGA-AG ('against the grain'), while samples oriented parallel to the grain direction are referred to as PGA-WG ('with the grain').

In an AGR, graphite components are manufactured into keyed bricks of height 80 cm, each with a channel running down the middle into which the fuel assemblies and control rods are inserted. During reactor operation the graphite components are exposed to significant stresses from mechanisms including irradiation, thermal discontinuities between different regions in the material, physical loading and irradiation-induced creep [3]. Such stresses can damage and deform the graphite components, particularly at discontinuities such as keyway roots. If sufficient numbers of graphite components were to fail, for example by cracking, any resulting distortion of the fuel assembly channels, control rod channels or coolant channels could prevent the reactor from cooling the fuel and shutting down, with potential safety implications. Since graphite moderator components cannot be repaired or replaced, deformation of these components is one of the main limitations on the operational lifetime of the reactor.

The response of graphite components to stress is strongly dependent on the microstructure of the graphite. Each individual microstructural property, such as the size, shape and orientation of filler particles and porosity, will affect the response of the graphite components to stress. It is therefore important to gain an understanding of how each of these properties responds to the build-up of stresses, in order to build up a model of the behaviour of the bulk material. This is particularly important for nuclear graphite since its properties vary from brick to brick. Such an understanding can be accomplished through detailed study of specific properties of graphite's microstructure. This paper discusses an investigation of how the pore structure responds to stress, an area where existing research is limited.

Given the UK's investment in graphite moderated reactors, and the country's reliance on nuclear reactors that are aging or have received life extensions [4,5], characterisation of deformation behaviour and the build-up of stresses in reactor grade graphite are particularly important. A detailed understanding of how graphite moderator components respond to stresses is required if we are to be confident of the safe and efficient operation of the reactors, particularly when consideration is being given to prolonging the service of the reactors.

2. Theory

The total strain acting upon a graphite moderator brick in a working reactor is comprised of six main components, as defined by Tsang and Marsden [6]:

$$\varepsilon^{total} = \varepsilon^{e} + \varepsilon^{pc} + \varepsilon^{sc} + \varepsilon^{dc} + \varepsilon^{th} + \varepsilon^{ith} \tag{1}$$

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, and ε^{ith} is the interaction thermal strain [7]. The latter term is not always included in the constitutive equations. In an irradiated graphite brick, the dominant components of this equation are the irradiation-induced stresses, followed by the thermally-induced stresses. The elastic strain term is relatively small.

In the 1960s, Jenkins [8] and Smith [9] investigated the deformation behaviour of nuclear grade graphite, with the intention of explaining the reasons why graphite components fail. Smith [9] studied unirradiated graphite, while Jenkins [8] investigated the behaviour in both irradiated and unirradiated graphites. Two mechanisms were identified that explain the deformation and failure of graphite: microcracking and dislocation movement. Microcracks can form, grow and combine if close enough in proximity, due to the applied stresses. This can result in the formation of large scale fracture paths that may run the length of the graphite brick. These fracture paths reduce the strength of the graphite components. Dislocation movement arises from the effects of irradiation, whereby atoms are forced out of position from the crystal lattice. These atoms can then form semi-stable dislocation loops between lattice planes, and vacancy loops form within the lattice planes where atoms have departed. This can lead to slippage of lattice layers and plastic deformation of the material.

In order to study how stress affects the material and microstructural properties of graphite moderator components, stresses must be generated in the components. There are numerous methods of accomplishing this, such as irradiating the samples or heating the samples to high temperatures. An alternate method would be to simulate the stresses present in a working reactor by applying a compressive load to the samples. The use of axial compressive and tensile loading techniques to simulate development of stresses in nuclear graphite is a well established technique [10,12], and has been used in conjunction with in-situ imaging and quantitative characterisation techniques to study how various nuclear grade graphites respond to stress. This technique has the advantage that the samples themselves need not be irradiated or exposed to high temperatures.

A large amount of research already exists in this field, with two research groups performing particularly relevant studies, and whose research inspired the experimental programme detailed in this paper: the Japanese Atomic Energy Institute [10,12–15] and the Nuclear Graphite Research Group at the University of Manchester [16–20]. Both groups have performed a number of investigations into the effects of stress on bulk graphite using methods such as Young's modulus calculation through ultrasonic time of flight methodology [9,11–13,16] and four point bend testing [17]. These in-situ techniques enabled calculation of both static and dynamic Young's moduli. Imaging studies [16,17] have focussed on the characterisation of various properties of graphite's microstructure, but are limited by these studies being partially qualitative [16] or completely qualitative [17]. Overall, these experiments have yielded significant information on the responses to stress of bulk graphite. However, there is still a lack of understanding of this behaviour at the microlevel. In particular, there is a focus on behaviour of the bulk material in response to stress. Since nuclear graphite is inhomogeneous and its properties can vary between bricks, it is proposed that microstructural studies, such as those by Marrow et al. [21] and Kane et al. [22], will tell us more about why