



Overview of strength, crack propagation and fracture of nuclear reactor moderator graphite

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HIGHLIGHTS

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ABSTRACT

Nuclear reactor moderator graphite is an aggregate of needle coke filler particles within a matrix of fine coke flour particles mixed with pitch binder. Following extrusion in green condition, impregnation with liquid pitch binder and graphitisation, a polygranular aggregate with orthotropic properties is produced. Its mechanical properties under several different loading conditions and associated cracking behaviour were examined to establish crack initiation and propagation behaviour. Both virgin and radiolytically oxidised material were examined using optical and electron optical microscopy, focused ion beam microscope and digital image correlation. The appearance of force vs. displacement curves varied with type of loading. Mostly linear elastic traces occurred in uniaxial tensile and flexural tests. Large departures from linear elastic behaviour were observed in standard uniaxial and diametral compression testing. Digital image correlation has shown that the initiation of cracking involves formation of a process zone which grows to a critical size of approximately 3–5 mm before a macro-crack is initiated. Cracks straddle a tortuous path which zigzags between the filler particles through the matrix consistent with crack propagation along the filler matrix interface. This paper provides an overview of strength, crack propagation and fracture of nuclear reactor moderator graphite. It reviews the physical processes and mathematical approaches that have been adopted to describe the behaviour of brittle materials and then considers if they apply to reactor core graphites.

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1. Introduction

All commercial nuclear reactors in the U.K., except one, use graphite cores as a moderator which is cooled by carbon dioxide gas. The manufacturing process of graphite, construction and geometry of the core, the operating pressure and temperature are different for Magnox and Advance Gas Cooled reactors (AGR), representing the first and second generation of nuclear plant in the U.K., respectively. The PGA graphite used in Magnox plant is extruded and then graphitised (NGP, 1959) giving the material orthotropic properties related to the extrusion axis. PGA graphite

is a heterogeneous aggregate mixture of needle coke filler particles within a matrix of coke flour and pitch binder. Its microstructure will be described in more detail later. By comparison, the gilsocarbon, used in AGR as moderator, is compacted using spherical filler particles to give isotropic properties. The microstructure is the most obvious difference between these two types of graphite aggregates.

The Magnox reactor cores are constructed of interlocking graphite bricks of pile grade A graphite (PGA) assembled in layers. The arrangement of bricks in the reactor core is shown in Fig. 1. Bricks are either octagonal or square in cross section. Units of eight bricks, with an equal number of each shape, form a repeating pattern in a layer. The core serves a dual role as both the moderator and a receptacle encasing the fuel elements in the channels. These are in the middle of the bricks and are continuous through the core. Each unit of eight bricks has a single interstitial channel at the point

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Fig. 1. Shows arrangement of bricks in reactor core.

where the corners of one set of two octagonal and two square bricks meet. The channels in the middle of the bricks hold fuel elements, and those at the corners of some sets of four bricks serve for insertion of control rods or instrumentation. The properties of graphite are changing during the service due to irradiation hardening and radiolytic oxidation. Neutron irradiation introduces defects at the atomic scale into the graphite crystal lattice causing dimensional change and also increasing the strength as a function of irradiation dose and temperature. Radiolytic oxidation due to energy deposition causes loss of mass which is associated with a reduction of strength (Best et al., 1985; Grover and Metcalfe, 2002). Differential strains between the bore and the outside surfaces of the PGA graphite bricks, mainly due to thermal transients and dimensional changes, generate stresses. These give rise to a concern that bricks may crack in a Magnox reactor core causing leakage of the coolant and a consequential rise of fuel temperature. Structural integrity of the graphite core is assessed throughout the lifetime of the reactor to ensure safe operation (Grover and Metcalfe, 2002; Ellis and Staples, 2005). The assessment makes use of mechanical properties measured on samples either trepanned from the reactor core bricks or those extracted in canisters installed in the core at the time of construction. Most of the experimental work described in this paper was carried out on PGA graphite.

Polycrystalline graphite does not deform plastically and therefore has been classified as a brittle material. The paper reviews the physical and mathematical approaches that have been used to describe the behaviour of brittle materials and then considers whether they apply to graphite. The experimental section describes the deformation, cracking and fracture studies of graphite. This involves a consideration of a range of specimen sizes and testing methods that have been adopted to produce mechanical property data and generated information on fracture characteristics of graphite.

2. Deformation and fracture

Virgin graphite contains porosity, due to shrinkage and cracking during manufacture that is further increased during service as a result of mass loss by radiolytic oxidation (Best et al., 1985; Grover and Metcalfe, 2002). Polycrystalline graphite does not deform plastically in a conventional way by movement of dislocations. Any departure from linear behaviour observed during mechanical testing can be attributed to cracking. Micro-cracking of graphite causes un-recoverable deformation and inelastic behaviour without a significant reduction in load bearing capacity. The behaviour of graphite under tensile loading is elastic and therefore it is considered to be brittle. However, previous studies (Hodgkins et al., 2010, 2011) of graphite led to a conclusion that it is more

appropriate to refer to graphite as a quasi brittle material in common with other aggregate materials such as concrete (Karihaloo, 1995; Schlangen and van Mier, 1992). The main distinction between brittle and quasi brittle materials is that the latter exhibits softening behaviour, post peak load involving a progressive stable decline of stress with increasing strain.

Graphite strength is measured experimentally in a laboratory using smaller specimens than the bricks in the reactor core and the results obtained from these tests need to be scaled for size effect on strength. The form of the relationship for size effect on strength of graphite depends on whether it is regarded as a classical brittle material or whether it is to be treated as a quasi brittle material. The fracture approach for brittle materials is based on Griffith's theory (Griffith, 1920). This involves a thermodynamical condition (Griffith, 1920) that balances surface energy, per unit area, elastic strain energy in a cracked body and work done by displacement of applied forces. This approach predicts that the fracture stress, σ_f , is obtained from:

$$\sigma_f = \sqrt{\frac{2E\gamma}{\pi c}} \quad (1)$$

where E is the Young modulus and c is the crack length. Eq. (1) predicts a decreasing strength with increasing crack length. Two failure models proposed for ferritic steels are consistent with this prediction (Ritchie et al., 1973; Wallin, 1998). Ritchie, Knott and Rice, RKR, proposed (Ritchie et al., 1973) that a critical stress needs to be achieved over a microstructurally significant distance (the characteristic distance) ahead of the crack tip. The second model is the weakest link concept as considered in the Master Curve approach (Wallin, 1998) which is based on a crack initiating by rupture of a large carbide particle, typically in the tail of the probability distribution.

Random variability, x , of fracture toughness or strength, is represented by a probability distribution. Two of these distributions considered further are Weibull and Normal. In a real space, their values vary from 0 to ∞ and $-\infty$ to ∞ , respectively. The Weibull probability density function, $f(x)$, is given by:

$$f(x) = \eta \alpha^{-\eta} x^{\eta-1} \exp \left\{ -\left(\frac{x}{\alpha}\right)^\eta \right\} \quad (2)$$

where η is the Weibull modulus or a shape parameter and α is the scale parameter. Equivalently, in logarithmic space, $\ln(\alpha)$ is the location parameter and $1/\eta$ is the scale parameter. The Normal probability density function is:

$$\frac{1}{\sigma\sqrt{2\pi}} \exp \left(-\frac{(x-\mu)^2}{2\sigma^2} \right) \quad (3)$$

where μ and σ are the mean and standard deviation, respectively. One of the distinctions between the distributions is that the scatter of x is a constant percentage of the median for a Weibull and a constant range about the mean for a normal distribution.

In contrast with the weakest link approach, Knott (2004) arrived to the conclusion that the distribution of strength for bainitic steels with a homogeneous microstructure approximates a Normal distribution. Although the size distribution of brittle particles is not Normal: there being many small, and fewer large particles. However, it is argued that, if a "typically large" (say, 95th percentile) particle is consistently sampled, from test-piece to test-piece, closely similar values of fracture stress, σ_f , will be obtained. Typically for a homogeneous microstructure, both the mean strength and fracture toughness would be single valued (Knott, 2004) and the standard error is due to experimental scatter which is small, typically approximately 2.5% of the mean (Knott, 2004). Substantially larger standard deviation for a Normal distribution or a shape parameter for a Weibull distribution occur in these materials with

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