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Effects of compositions of filler, binder and porosity on elastic and fracture properties of nuclear graphite

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HIGHLIGHTS

- Micromechanics based homogenisation method is used for graphite microstructures.
- Elastic and fracture properties are estimated based on graphite microstructures.
- Fracture behaviours are studied using compact tension simulation.
- Fracture behaviours are studied using four point bending simulation.
- Flexural strengths were obtained and compared to experimental results from literature.

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ABSTRACT

Physical mechanisms at different length scales have to be taken into account while predicting the overall failure of nuclear graphite structures of advanced gas cooled graphite reactors. In this paper, the effect of composition of meso graphite phases and porosity on the aggregate elastic properties is predicted using the Eshelby homogenisation method. Results indicate an overall decrease in elastic modulus with an increase in porosity. Subsequently, the moduli at different porosity levels are used to predict the critical strain energy release rates for crack propagation of graphite, and fracture behaviour is studied using compact tension and four point bending tests. Compared to flexural strength at zero porosity level, significant reduction in strength of up to 80% at 30% porosity level is observed. Evolution of flexural strength due to porosity is also compared against available experimental values of graphite from UK nuclear plants.

1. Introduction

Advanced gas-cooled reactors (AGR) within the UK are near the end of their safety life and currently significant research efforts have been invested to study whether the closure dates can be extended. Since AGRs are graphite-moderated, the damage mechanisms and subsequent fracture behaviour of graphite material need to be understood in order to estimate the safe lifetime of graphite structures.

Polycrystalline graphite used in the nuclear graphite industry is manufactured by mixing and baking a mixture of binder matrix and coke filler particles. Coal tar pitch or petroleum pitch can be used as binder depending on the grade of graphite. The resultant heterogeneous material also has porosity (pore size varies from sub μm to hundreds of μm) and microscopic intra-crystal defects in the forms of microcracks [1]. As this type of graphite undergoes years of operation, the fast neutron damage and oxidation cause dimensional change in the crystals, material properties changes in the phases, and increase in total porosity. These changes can be studied at different scale ranges, from atomistic to mesoscopic scales. Linking these mechanistic multiscale effects to changes in the material properties is necessary. However, the homogenization of random multiscale mechanisms into a macroscopic structural analysis has proved to be challenging.

An attempt to link the mesoscopic and macroscopic scales is presented in this paper. Previous work [2] has shown the use of micromechanics to formulate the properties of aggregates composed of meso phases (binder and filler) and porosity within nuclear graphite. On this basis, a similar micromechanics-based homogenisation technique is applied and a further study of the effects of those phases on the fracture behaviour of nuclear graphite is also carried out. Evolution of flexural strength due to porosity is also compared against available experimental values of graphite from UK nuclear plants from the literature.

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2. Micromechanics based model for effective elastic properties of nuclear graphite

2.1. Eshelby homogenisation method

It is challenging to take account of the effects of microstructures while analysing the failure of engineering components due to large difference in length scales. To avoid the complexity of modelling microstructural graphite phases implicitly using multi-scale models, a homogenisation technique is used to predict the elastic property of heterogeneous nuclear graphite in this paper. The homogenisation scheme applied here is based on Eshelby's method [3] and it is chosen over other continuum micromechanics methods since the method can be applied irrespective of shapes and numbers of phases. The method is briefly discussed here using a material composed of two homogeneous phases without porosity for simplicity, as in [4]. When the inclusion is taken out of the matrix and it is subjected to the eigenstrain (ε^{*}_{kl}) or stress free strain, this eigenstrain would be cancelled by the elastic strain of the inclusion (ε^{el}_{kl}) due to strain compatibility. The stress within the inclusion can be written as Eq. (1) while stress and strain within the matrix are still zero. Once this inclusion is put back into its original place within the matrix, both matrix and inclusion surfaces will experience additional stress (σ^{c}_{ij}) and strain (ε^{c}_{kl}) for the constrained field of Green's function [4]. Due to this additional stress, the stress within the inclusion can now be written as Eq. (2). The term ($\varepsilon^{c}_{kl} - \varepsilon^{*}_{kl}$) represents the elastic strain of the inclusion.

$$\sigma_{ij} = C_{ijkl}\varepsilon^{*}_{kl} = -C_{ijkl}\varepsilon^{el}_{kl} = -\sigma^{*}_{ij} \quad \text{, where } \sigma^{*}_{ij} \text{ is eigenstress} \tag{1}$$

$$\sigma_{ij} = \sigma^{c}_{ij} - \sigma^{*}_{ij} = C_{ijkl}(\varepsilon^{c}_{kl} - \varepsilon^{*}_{kl}) \tag{2}$$

The above method can be extended to inhomogeneous inclusions with an external strain of (ε^{0}_{kl}) by assuming that the stress within the inclusion is identical for both homogenous and inhomogeneous cases. In tensor form, it can be written as Eq. (3). The constrained strain can be linked to the eigenstrain via an Eshelby tensor (S_{ijkl}), which is a fourth order tensor, as shown in Eq. (4). The Eshelby tensor is dependent on the shape of the inclusions and for this study only spherical inclusions are considered for simplicity. For non-spherical inclusions, such as an ellipsoid shape, anisotropic Eshelby tensor is required and the material stiffness will be increased if the loading is applied along the major axis of the ellipsoid or vice versa.

$$C_{ijkl}(\varepsilon^{c}_{kl} + \varepsilon^{c}_{kl} - \varepsilon^{*}_{kl}) = C^{m}_{ijkl}(\varepsilon^{c}_{kl} + \varepsilon^{0}_{kl}),$$
$$\textit{where } C^{m}_{ijkl} \text{ is the stiffness matrix of the inclusion} \tag{3}$$

$$\varepsilon^{c}_{ij} = S_{ijkl}\varepsilon^{*}_{kl} \tag{4}$$

2.2. Formulation of elastic moduli of heterogeneous materials with spherical inclusions

The Eshelby's homogenisation method was modified for the materials with more than one inclusion by Budiansky [5]. The procedure was also applied for heterogeneous coating material by Hermosilla [6] and it will be briefly described here. Assuming that the material is composed of N phases and the sum of volume fractions of those phases (V^{i}_{f}) is one for a volume conservation, the shear modulus (G) and the bulk modulus (K) of the aggregate can be linked to phases properties (G^{i} and K^{i}) as shown in Eq. (5) and (6).

$$\sum_{i=1}^{N} \frac{V^{i}_{f}}{1 + \beta^{t}\left(\frac{G^{i}}{G} - 1\right)} = 1 \tag{5}$$

$$\sum_{i=1}^{N} \frac{V^{i}_{f}}{1 + \beta^{n}\left(\frac{K^{i}}{K} - 1\right)} = 1 \tag{6}$$

β^{t} and β^{n} are linked to the aggregate Poisson's ratio as shown in Eqs. (7) and (8), and aggregate Poisson's ratio can be expressed in terms of the bulk and shear moduli as shown in Eq. (9). For known elastic properties of N phases, Eqs. (5)–(9) can be solved simultaneously to obtain elastic properties of the whole media.

$$\beta^{t} = \frac{2(4 - 5v)}{15(1 - v)} \tag{7}$$

$$\beta^{n} = \frac{(1 + v)}{3(1 - v)} \tag{8}$$

$$v = \frac{3K - 2G}{6K + 2G} \tag{9}$$

3. Application of Eshelby homogenisation method to elastic properties of graphite

To apply the Eshelby homogenisation method to the elastic properties of nuclear graphite, the properties of its constituent phases (filler and binder) are required. The elastic properties of the binder was taken from the nano-indentation test by Berre et al. [7]. The elastic modulus of coke filler was estimated using a zero porosity overall modulus and data from radiolytic oxidation experiments in [2] and the resultant value is taken for this analysis. Due to insufficient data, Poisson's ratio of aggregate graphite from [8] is taken as the value for both filler and binder. The material properties are listed in Table 1. The initial filler volume within quasi-isotropic graphite with spherical coke filler particles is 60% which was taken from X-ray tomography images performed by Berre et al. [2]. This volume is taken as constant for the current analysis and the porosity volume is increased from 0% to 30% with an increment of 5%.

To compare the results from the Eshelby method to other independent results, three sets of data were chosen in which two sets are from simple models (Voigt and Knusden models) and the other set is from the experiments by Kelly et al. [9]. Voigt model uses a weight mean of phase volumetric proportions (V^{i}_{f}) to predict the aggregate properties as in Eq. (10). The model is only applicable to axial loading cases. Knusden model can be expressed in terms of the initial elastic modulus with zero porosity (E_0) and porosity volumetric ratio (P) as shown in Eq. (11) where b is an empirical parameter, which depends on the aspect ratio of the pore (a/c) as shown in Eq. (12).

$$E = \sum_{i=1}^{N} V^{i}_{f}E^{i} \tag{10}$$

$$E = E_{0}e^{(-b \times P)} \tag{11}$$

$$b = 1 + 0.594 \times (a/c) \tag{12}$$

The moduli of graphite based on porosity proportion for all three models are plotted in Fig. 1. For Knusden's model three aspect ratios of porosity, based on different values of b, were also

Table 1
Material properties for binder and filler of graphite.

	Binder	Filler
E (GPa)	15 [7]	41 [2]
v	0.2 [8]	0.2 [8]