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### Modelling fracture of aged graphite bricks under radiation and temperature

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#### ABSTRACT

The graphite bricks of the UK carbon dioxide gas cooled nuclear reactors are subjected to neutron irradiation and radiolytic oxidation during operation which will affect thermal and mechanical material properties and may lead to structural failure. In this paper, an empirical equation is obtained and used to represent the reduction in the thermal conductivity as a result of temperature and neutron dose. A 2D finite element thermal analysis was carried out using Abaqus to obtain temperature distribution across the graphite brick. Although thermal conductivity could be reduced by up to 75% under certain conditions of dose and temperature, analysis has shown that it has no significant effect on the temperature distribution. It was found that the temperature distribution within the graphite brick is non-radial, different from the steady state temperature distribution used in the previous studies [1,2]. To investigate the significance of this non-radial temperature distribution on the failure of graphite bricks, a subsequent mechanical analysis was also carried out with the nodal temperature information obtained from the thermal analysis. To predict the formation of cracks within the brick and the subsequent propagation, a linear traction-separation cohesive model in conjunction with the extended finite element method (XFEM) is used. Compared to the analysis with steady state radial temperature distribution, the crack initiation time for the model with non-radial temperature distribution is delayed by almost one year in service, and the maximum crack length is also shorter by around 20%. © 2017 The Authors. Published by Elsevier Ltd.

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#### 1. Introduction

Nowadays, in the UK, the gas-cooled graphite moderated type dominates over 90% of the nuclear reactor [3]. Graphite is also considered to be used within the design of the new reactor generation (such as the Helium gas cooled very high temperature reactor or VHTR) as structural support and as moderator because of its extreme purity, ability to withstand extremely high temperatures [4]. The core structure of the reactor is formed from joining graphite bricks, with keys and keyways. During operation, graphite is exposed to neutron radiation and temperature gradients that could cause irradiation and radiolytic oxidation. These occurrences will affect material properties such as weight loss, porosity changes and thermal conductivity which can potentially lead to material and structural failure [5].

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Future reactors such as VHTR are expected to operate at higher temperatures than the current reactors and its core outlet temperature (COT) will be about 1000 °C [5-8]. In order to enhance thermal efficiency of future nuclear reactors, extensive research has been carried out, aiming to provide an improved design for future reactors [6-10]. As the reactor is exposed to neutron dose and temperature, many material properties are changed by irradiation and radiolytic oxidation if air or carbon dioxide is used as a coolant. Some of the properties of nuclear graphite which change under service conditions include elastic properties and thermal expansion coefficient, irradiation creep, irradiation induced dimensional changes. The prediction of the nuclear graphite lifetime and its structural integrity are significant issues for the safety and reliability of reactor operation that could be affected by changing properties.

In previous studies [2], a constitutive model has been developed for the effects of mechanical properties changes on stress states within the graphite brick. Resultant crack development from the stress state within the brick was also studied in [1]. For previous studies, steady state temperature distribution was used to calcu-

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late thermal stress assuming that thermal conductivity of graphite brick remains constant across the brick during the operation. It was also assumed that the temperature distribution is linear across the graphite brick. However, studies [11,12] have shown that thermal conductivity of nuclear graphite changes with both temperature and fast neutron irradiation. Hence, a new thermal analysis is required to calculate the temperature gradient across the brick by considering the changes in thermal conductivity due to dose and temperature.

In this paper, an empirical equation for the evolution of thermal conductivity is derived from available experimental data by Marsden et al. [12]. Using this relationship with relevant thermal properties, a thermal analysis is carried out. Resultant nodal temperatures are then implemented into mechanical finite element (FE) model with a constitutive model developed in [2] and fracture mechanics based damage model developed in [1]. Stress analyses and crack development are then carried out. Comparisons are also made to models with the linear temperature distribution across the brick.

## 2. Empirical relationship of thermal conductivity of graphite with neutron dose and irradiated temperature

The effects of temperature gradients and irradiation can induce damage to the graphite bricks and this leads to changes in its thermal conductivity. The common approach is based on relating thermal conductivity (K) to summation of thermal resistances (1/K) as a result of scattering hurdles [13,14]

$$K(\mathbf{x},T) = \frac{\alpha(\mathbf{x})}{\left(\frac{1}{K_{CB}} + \frac{1}{K_{u}} + \frac{1}{K_{RD}}\right)}$$
(1)

where the term  $\alpha(x)$  is a coefficient, which is function of orientation terms (basal plane, x),  $\frac{1}{K_{CB}}$  is the thermal resistance due to grain boundary scattering,  $\frac{1}{K_u}$  is the thermal resistance due to Umklapp scattering or the so-called phonon-phonon scattering [14], and  $\frac{1}{K_{RD}}$  is the thermal resistance due to lattice defect scattering.  $K_{GB}$  depends on the graphite perfection and is taken into account only for lower temperatures, as it is insignificant at intermediate and high temperatures.  $K_u$  can be scaled as a quadratic function of temperature [14] and it controls thermal conductivity at higher temperatures. Finally,  $K_{RD}$  depends on the neutron irradiation, as the latter plays a significant role in producing various types of defects, which are more effective in scattering phonons. Therefore, many studies [5,13,15] emphasise that the change in conductivity is only caused by the phonon scattering in lattice defects. This phonon scattering increases with temperature. Hence it is necessary to consider both irradiation dose and temperature to estimate the change in thermal conductivity.

For significant nuclear irradiated graphite grades e.g. Gilsocarbon, Kelly [11] proposed a formula as shown in Eq (2), to calculate the thermal conductivity. It includes the terms, f and  $S_k$ , obtained empirically as a function of dose and temperature [12].

$$\frac{1}{K(T)} = \frac{1}{K_0(30)} \left[ \frac{K_0(30)}{K_0(T)} + f.\delta(T) \right] S_k$$
(2)

where *f* is the initial irradiation induced change equal to  $\frac{K_0(30)}{K_1(30)} = \frac{K_0(30)}{K(30)} - 1$  and  $S_k$  is a structural term which allows decrease in thermal conductivity at higher dose. Marsden et al. [12] used Eq (2) on irradiated and unirradiated graphite samples. The thermal conductivity is shown to be directly proportional to the operation temperature (T) for irradiated graphite but the opposite is true for unirradiated graphite.

Without using an atomistic physical approach for calculating of the variation of thermal conductivity for the graphite such as



Fig. 1. Thermal resistivity variation with fast neutron dose as a function of dose and temperature [12].

the one proposed in [16], the thermal conductivity dependence on dose and temperature can be deducted from experimental data curves or include empirical terms obtained from the experiments using Material Test Reactor (MTR). In the current paper, thermal conductivity variation results of MTR from Marsden et al. [12] are used to generate the empirical equation. The variation in thermal resistivity with temperature and dose is illustrated in Fig. 1. The data was obtained from the nuclear experiments with equivalent DIDO nickel dose (EDND). The neutron dose is expressed in terms of equivalent DIDO nickel dose (EDND) which is calculated so that the damage rate experienced by the graphite at a given location is equivalent to that of graphite test samples irradiated in the DIDO reactor [17]. 1 n/cm<sup>-2</sup> EDND is equal to  $1.313 \times 10^{-21}$  displacement per atom (dpa) [12].

Using the data of thermal resistivity from Fig. 1, an empirical equation shown in Eq (3) for the dependence of the resistivity on dose (D) and temperature (T) is fitted.

$$K = K_0 \cdot \left[ \left( (a.D + b) \times \exp\left(\frac{c}{T}\right) \right) + 1 \right]^{-1}$$
(3)

where *K* is the thermal conductivity of the nuclear graphite, D is dose  $\times 10^{20}$  n/cm<sup>-2</sup> EDND, *T* is the irradiation temperature in Kelvin and  $K_o$  is the initial thermal conductivity of unirradiated graphite at 30 °C (about 130 W/m/K), and *a*, *b* and *c* are constants equal to 0.004185, 0.9716 and 209.4, respectively.

Please note that the data for 25 °C was excluded from the fitting process to reduce the error sum of squares (SSE) from 3.742 to 1.858, with coefficients of 95% confidence bounds. The resultant Eq (3) can be used to calculate transient change in thermal conductivity for the thermal FE model for which changes in dose and temperature are expected. Finally, to illustrate the accuracy of the calculated thermal resistivity using Eq (5), the values are plotted against experimental results as shown in Fig. 2 at 100°C, 300°C and 600°C.

#### 3. Graphite brick model

#### 3.1. Geometry and mesh of finite element model of graphite brick

In this paper, failure of the graphite brick of Gilsocarbon under irradiation and temperature is estimated. In previous studies [1,2], an assumed profile of temperature distribution across the graphite brick was applied to the model as one of the field variables to model the reactor conditions. The temperature is assumed to vary in the radial direction of the brick while it is kept constant in the circumferential direction and along the depth direction of the brick. Here, a separate thermal FE model is used to assess the effect of conductivity change on the temperature distribution

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