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Predictions of inter-granular cracking and dimensional changes of irradiated polycrystalline graphite under plane strain

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ABSTRACT

An original model is proposed that predicts the response of a polycrystalline graphite agglomerate to temperature change and irradiation. The model explicitly considers the graphite crystal anisotropy and the development of cracks along the interfaces between differently oriented grains. Using only graphite single crystal data, the model yields predictions of the macroscopic elastic stiffness, thermal expansion coefficient, irradiation-induced creep and irradiation-induced dimensional change (initial shrinkage followed by swelling at higher fast neutron fluence). The behaviour of the virgin polycrystal is properly predicted using a self-consistent mean-field model. However, predicting the pattern of inter-granular cracks requires a finite element model with cohesive zones. The latter simulations are performed on a periodic model polycrystal in 2D, leading to a qualitative agreement with experimental data.

1. Introduction

In gas cooled nuclear reactors, structural components made of graphite act as the moderator and it is a scientific challenge to predict how their mechanical properties are progressively changed by fast neutron irradiation. Such reactor components have a heterogeneous microstructure made of filler particles and voids as well as a binder with a polycrystalline nature. At the scale of the crystal lattice, graphite is very strongly anisotropic: elastic stiffness, thermal expansion, creep and irradiation-induced dimension changes all differ by orders of magnitude (or in sign) depending on the crystal axis considered [1]. Consequently, both the production process of polycrystalline graphites intended for nuclear reactor application, and the response of these polycrystalline graphites to irradiation, involve strains well beyond the elastic limit of the graphite crystals. Graphite crystals have only two independent slip systems, less than the five independent slip systems that are required to accommodate arbitrary shear deformations [2]. Thus, strains beyond the elastic limit will often cause cracking; indeed cracks form during the initial manufacturing process and strongly influence properties of the manufactured material (for example, strength, elastic modulus, density, thermal expansion coefficient) as pointed out by Mrozowski [3].

At present, there is no theoretical basis on which to predict polycrystalline behaviour from single crystal behaviour; in particular there is no quantitative theoretical description of crack formation and crack closure and the effect they have on the graphite properties. Such a description would be very useful for prediction of the behaviour of graphite components in an operating nuclear reactor. The present contribution begins the construction of a theoretical basis on which to predict polycrystalline response from single crystal response.

There has been much experimental and theoretical work on the properties of single crystals of graphite and highly oriented pyrolytic graphite (which is thought to have similar behaviour), see [1] for references. Based on these earlier experiments, an approximate constitutive law for a single crystal is presented in Section 2. The single crystal constitutive law accounts for anisotropy, thus allowing us to investigate how mismatch in properties of misaligned crystals affects properties of the polycrystalline aggregate. At this stage of development of the model, it is appropriate to use an approximate constitutive law; a description of the anisotropy to a certain accuracy is expected to yield a quantitative description of crack formation and closure of similar accuracy. Thus, for example, in the model, irradiation of graphite crystals is assumed to change their shape while conserving their volume, whereas, in reality, irradiation changes the volume very slightly (as compared to the thermal expansion). In this paper, the words "crystal" and "grain" mean a region of the solid graphite where

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basal planes are approximately aligned, in other words, a region wherein the effects of the anisotropy are the same. A "crystal" is not meant to imply a region that is crystallographically perfect.

The model considers a granular microstructure that is representative of the binder, a major constituent of nuclear graphite. Filler particles and initial porosity are ignored here because their characteristic scale is at least an order of magnitude larger than the average diameter of crystals in the binder. The average response of such a granular polycrystal can be predicted using several scale transition schemes. However, only a few are applicable when internal cracking occurs. The present study is split in two. In the first part, some reference predictions are produced in order to study the build up of internal stresses in an undamaged graphite polycrystal. A model predicting the influence of the pattern of intergranular cracks on the properties of the polycrystalline material is presented in the second part of the paper. In both cases, the problem is simplified by considering a two-dimensional polycrystal section undergoing plane strain.

The homogenized response of an undamaged granular agglomerate can be predicted using Eshelby's solution of the equivalent inclusion problem [4,5]. In self-consistent (SC) schemes, each grain is treated as an isolated inclusion embedded in a uniform matrix, the behaviour of which is iteratively identified and set equal to the average response of all grains [6,7]. The latter solution is exact when constituents are linear elastic and it has been extended in order to develop approximate mean field solutions in the case of non-linear viscoplastic [8] and plastic [9] materials. The SC method has been applied successfully to single phase polycrystals in which strain heterogeneity results from the anisotropy of individual grains. For instance, SC models produce valid prediction of the viscoplasticity of metallic alloys with hexagonal close packed (hcp) crystal symmetry, in which dislocation glide is favored along certain atomic planes so that the crystal lacks the five independent slip systems that allow achieving an arbitrary isochoric deformation [2,10]. An original SC solution is developed here in order to investigate thermal strains in polycrystalline graphite undergoing plane strain. It makes use of the strain concentration tensor computed by Dvorak [11] and then takes advantage of the macroscopic transverse isotropy in order to derive the macroscopic coefficient of thermal expansion.

The most widespread technique in order to model cracking of solid material is the use of cohesive zones [12,13]. The underlying idea is that the crack extends fictitiously beyond its tip, along an interface across which load is carried by springs with non-linear stiffness and decaying strength. Integration of the traction separation law until full opening defines the toughness of the interface, i.e. the energy dissipated in the crack extension. The main drawback of cohesive zone modelling is that potential cracking paths must be known a priori.

Unfortunately, Eshelby's solution cannot be extended easily to the case of an imperfect interface between the inclusion and the surrounding matrix. The mean field approach remains accurate only in the case of spherical inclusions under hydrostatic loading [14–16]. Other cases require a numerical solution of the stress and strain fields throughout the microstructure.

Finite element modelling including cohesive zones has been used extensively to predict crack propagation along the grain boundaries of polycrystalline materials [17–19]. The technique is applied here to irradiated polycrystalline graphite. One of the difficulties, when relying on an implicit finite element solver, is the resolution of convergence issues due to the softening response of the damaged cohesive zones. The most common work-arounds are to introduce a low viscosity in the cohesive law [20] or to force a controlled opening of the internal cracks instead of applying load at the external boundaries of the finite element mesh [21]. The first of these two solutions is applied here.

Finite element modelling of some aspects of the microstructure of nuclear graphite has been attempted previously [22]. This paper states "The underlying assumption of this procedure was that the filler behaviour was the dominant force behind the polycrystalline material changes" and "the binder was assigned unirradiated properties of an isotropic graphite (Gilsocarbon)". Thus changes in binder properties produced by irradiation were assumed to be insignificant and furthermore the binder properties were not predicted from the crystal properties. This approach is not suitable for a model aimed at fundamental understanding. Of course, the filler particles in nuclear graphite may well influence its behaviour, but our results show that the binder behaviour is also important.

2. Mechanical response of individual graphite crystals

For conciseness, the components of symmetric second- and fourth-order tensors are denoted using contracted forms. For instance, the components of the Cauchy stress tensor $\boldsymbol{\sigma}$ are denoted:

$$[\sigma_1,\sigma_2,\sigma_3,\sigma_4,\sigma_5,\sigma_6]^T=[\sigma_{11},\sigma_{22},\sigma_{33},\sigma_{23},\sigma_{13},\sigma_{12}]^T \tag{1}$$

Since graphite crystals have hexagonal symmetry, their mechanical response is transverse-isotropic around the so-called "c-axis", which is normal to the "basal plane" (see Fig. 1). The elastic stiffness operator $\mathcal{C}$ then has only five independent non-zero components:

$$\begin{bmatrix}\sigma_1\\ \sigma_2\\ \sigma_3\\ \sigma_4\\ \sigma_5\\ \sigma_6\end{bmatrix}=\begin{bmatrix}\mathcal{C}_{11} & \mathcal{C}_{12} & \mathcal{C}_{13} & & & \\ \mathcal{C}_{12} & \mathcal{C}_{22} & \mathcal{C}_{23} & & & \\ \mathcal{C}_{13} & \mathcal{C}_{23} & \mathcal{C}_{33} & & & \\ & & & 2\mathcal{C}_{44} & & \\ & & & & 2\mathcal{C}_{55} & \\ & & & & & 2\mathcal{C}_{66}\end{bmatrix}\cdot\begin{bmatrix}\varepsilon_1^{el}\\ \varepsilon_2^{el}\\ \varepsilon_3^{el}\\ \varepsilon_4^{el}\\ \varepsilon_5^{el}\\ \varepsilon_6^{el}\end{bmatrix} \tag{2}$$

If the c-axis corresponds to $\boldsymbol{e}_1$, then $\mathcal{C}_{22}=\mathcal{C}_{33},\mathcal{C}_{12}=\mathcal{C}_{13},\mathcal{C}_{55}=\mathcal{C}_{66}$ and $\mathcal{C}_{44}=(\mathcal{C}_{33}-\mathcal{C}_{23})/2$.

We consider four contributions to the deformation of graphite crystals: elasticity, thermal expansion, irradiation-induced deformation and irradiation-induced creep. Since the total strain remains small, the total strain rate may be additively decomposed into its various parts:

$$\dot{\boldsymbol{\varepsilon}}=\boldsymbol{\mathcal{C}}^{-1}:\dot{\boldsymbol{\sigma}}+\boldsymbol{\alpha}\dot{T}+\boldsymbol{\beta}\frac{\dot{\Phi}}{\Phi_0}+\dot{\boldsymbol{\varepsilon}}^{creep} \tag{3}$$

Here, superscript dots denote time derivatives, $\boldsymbol{\alpha}$ is the thermal expansion tensor, $\boldsymbol{\beta}$ is the irradiation induced dimensional change tensor, and Φ is fast neutron fluence (the total number of fast

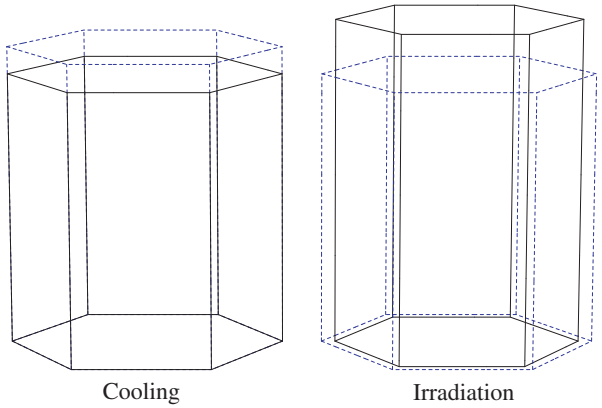

Fig. 1. Illustration of the anisotropy of thermal expansion and irradiation-induced shape change in graphite crystals. The crystal c-axis is vertical and the hexagons lie in the basal planes. Cooling is followed by irradiation. The deformations (only a few percent) are amplified to ease the visualisation.