Journal of Nuclear Materials 487 (2017) 50–67

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Time of flight measurements of unirradiated and irradiated nuclear graphite under cyclic compressive load

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ARTICLE INFO

Article history:
Received 27 July 2016
Accepted 27 December 2016
Available online 4 February 2017

ABSTRACT

The time-of-flight technique has been used to investigate the stiffness of nuclear graphite with respect to the grade and grain direction. A loading rig was developed to collect time-of-flight measurements during cycled compressive loading up to 80% of the material's compressive strength and subsequent unloading of specimens along the axis of the applied stress. The transmission velocity (related to Young's modulus), decreased with increasing applied stress; and depending on the graphite grade and orientation, the modulus then increased, decreased or remained constant upon unloading. These tests were repeated while observing the microstructure during the load/unload cycles. Initial decreases in transmission velocity with compressive load are attributed to microcrack formation within filler and binder phases. Three distinct types of behaviour occur on unloading, depending on the grade, irradiation, and loading direction. These different behaviours can be explained in terms of the material microstructure observed from the microscopy performed during loading.

1. Introduction

Graphite is and has been used extensively as a moderator within nuclear reactors ever since the first artificial nuclear reactor in Chicago achieved criticality in 1942, through to prospective Generation IV reactor designs. In the United Kingdom, two graphite grades, with very different microstructures, have been used in this moderator role: Pile Grade A (PGA), used in the first generation Magnox reactors; and Gilsocarbon, used in the succeeding Advanced Gas-cooled Reactors (AGRs). Samples of these two grades were used in this work which studied the influence of microstructure on stiffness, however the findings are applicable to newer graphite grades being developed for use in Generation IV High Temperature Reactors (HTRs) and Molten Salt Reactors (MSRs).

Examples of the microstructures of PGA and Gilsocarbon are shown in Fig. 1 and Fig. 2 respectively. The microstructures of these graphite grades can be considered as consisting of three separate phases: binder, filler and porosity [1]; and the most significant difference between the two grades is the difference between the filler material. PGA was manufactured using “needle-shaped” coke particles as the filler material; elongated grist particles which were by-products of the cracking process during petroleum manufacture [2]. Due to the method of billet manufacture, the “needle-shaped” coke preferentially aligns during the extrusion process prior to baking, yielding an anisotropic final product. The anisotropic microstructure of PGA results in strongly anisotropic material properties [3], which are generally quoted as “with grain” (measured along the axis of extrusion), or “against grain” (measured along one of the axes perpendicular to the axis of extrusion); a convention which is used throughout this paper, with grain directions of specimens logged prior to testing. In contrast, Gilsocarbon utilised spherical coke particles which were derived from Gilsonite, a naturally occurring asphalt after which the graphitised material is named [4]. In addition, prior to baking, Gilsocarbon was pressed into shape rather than extruded which, when combined with the spherical particles, resulted in a near-isotropic material.

The other two phases to consider when examining graphite microstructures are the binder and porosity. The binder is a pitch-flour-particle mix which is blended with sized coke particles after their calcination and prior to baking. Porosity in nuclear-grade graphite takes three forms. Firstly, the calcination of coke particles prior to mixing leads to the formation of narrow lenticular shaped cracks within the particles, which are still clearly

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http://dx.doi.org/10.1016/j.jnucmat.2016.12.044

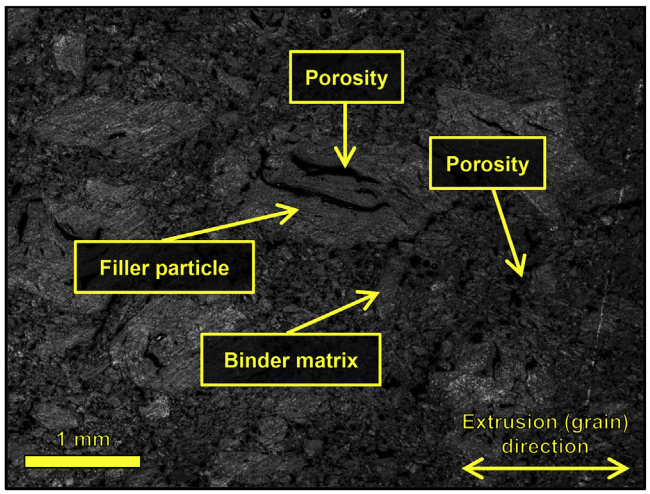

Fig. 1. Microstructures of PGA graphite indicating common features. The extrusion direction is shown; left to right is considered "with grain" while up and down is referred to as "against grain".

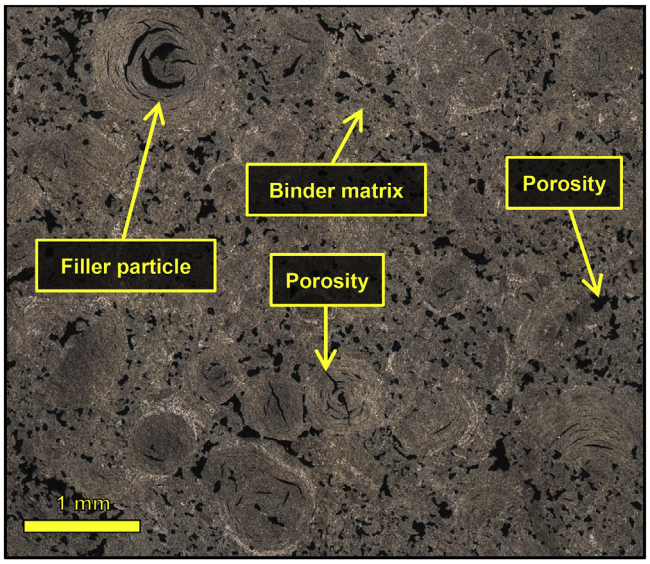

Fig. 2. Microstructure of Gilsocarbon with equivalent features. Note the main difference between this and the PGA microstructure is the shape of the filler particles.

observable in the final graphitised microstructure. These cracks run circumferentially in Gilsocarbon coke filler particles, and along the long axis of PGA grist filler particles. Secondly, the baking process results in the evolution of volatiles from the pitch-particle mix, resulting in large, globular gas-evolution porosity existing within the binder and around coke particles. Lastly, while the graphitisation process leads to a far more ordered material at a crystal level than exists in prior stages in the manufacturing process, the final product is far from a perfect graphite crystal; the difference in density between an ideal graphite material (~2.27 g cm^{-3}), and the skeletal density of the polycrystalline product used in nuclear applications is considered microporosity. One of the sources of this closed microporosity results from cracks which are formed during the thermal contraction of cooling after the graphitisation process, known as Mrozowski cracks [5]. The skeletal density of Gilsocarbon is ~2.00 g cm^{-3}, with bulk density around 1.82 g cm^{-3}; a result of the open porosity within the material [6]. The heterogeneous nature of artificial graphite results in differences in measured values across samples. The differences in material properties between the grades are documented [7].

There is a reduction in Young's modulus when test specimens of polycrystalline graphite are placed under tensile or compressive stress [8]. It is also well observed that the non-linear stress-strain behaviour becomes progressively more linear as the test material becomes irradiated, which is attributed to pinning of dislocations in basal planes [9]. This work follows the evolution in transmission velocity (i.e. the variable used to calculate dynamic Young's modulus) of virgin Gilsocarbon and virgin and irradiated PGA when placed under compressive load, and examines the changes to the microstructure in order to better understand the relationship between microstructure and unloading behaviour.

2. Experimental

2.1. Materials

Compressive specimens of unirradiated Gilsocarbon and PGA were prepared as cubes of dimensions (40 × 40 × 40) mm^3 to allow for testing across all three axes. The PGA samples used for this work were cut from an unused moderator brick prepared for use within the Oldbury Magnox reactor. Gilsocarbon samples were prepared from an unmachined billet of Gilsocarbon provided by EDF; though the designated reactor for this batch of material is unknown, this work is aimed at understanding loading behaviour rather than obtaining specific engineering data for individual reactors. Irradiated PGA specimens originate from spacer material taken from a non-thermocoupled installed set assembly. The reactor mean core irradiation during their discharge was 29,835 MW d/te. No dosimetry calculations specific to the spacer were available, however using data for the adjacent position an irradiation temperature of 325 °C and a dose of 3.5 dpa can be estimated from MCBEND calculations. These samples were irradiated with exposure to carbon dioxide coolant and therefore their densities are reduced because of radiolytic oxidation [10]. The original size of the source material limited the dimensions of the irradiated specimens to cubes of (18 × 18 × 18) mm^3.

Small samples of each material of dimensions (15 × 9.5 × 6) mm^3 where the span of the sample is 15 mm were also prepared for loading in a microtester. The top surfaces of the samples were polished to allow for images to be captured clearly during loading. Because of the strong anisotropy of the properties of PGA, the grain directions of each PGA specimen were noted during machining to enable measurements to be carried out with grain and against grain where appropriate.

2.2. Apparatus

A Panametrics Model 5800 pulser/receiver was used in conjunction with two Olympus 1 MHz V103-RM right angled microdot/BNC contact transducers to generate and receive longitudinal ultrasonic pulses. The pulser/receiver was controlled by a LeCroy WaveRunner 64Xi 600 MHz digital oscilloscope, with Sonatest Sonagel W couplant gel used to provide appropriate signal propagation. Samples were compressed in a servo-hydraulic Mayes 500 universal testing machine with a self-aligning spherical platen using the apparatus shown in Fig. 3 to prevent loading to the transducers during testing. Transducers were attached to the rig using a triggered G-clamp. Strains were measured through the use of Tokyo Sokki Kenyujo 120 Ω FLA 6–11 strain gauges (gauge factor 2.1) attached to the test specimen using M-Bond 200 strain gauge adhesive. The lack of extended experimental durations, humidity or elevated temperatures during these experiments permits the use of