In situ X-ray tomography imaging of crack initation and propagation in nuclear graphite at 1000°C

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Abstract
Nuclear-grade graphite is a critically important high-temperature structural material for current and potentially next generation of fission reactors worldwide. It is imperative to understand its damage-tolerant behaviour and to discern the mechanisms of damage evolution under in-service conditions. Here we perform in situ mechanical testing with synchrotron X-ray computed micro-tomography at temperatures between ambient and 1,000 °C on a nuclear-grade Gilsocarbon graphite. We find that both the strength and fracture toughness of this graphite are improved at elevated temperature. Whereas this behaviour is consistent with observations of the closure of microcracks formed parallel to the covalent-sp2-bonded graphene layers at higher temperatures, which accommodate the more than tenfold larger thermal expansion perpendicular to these layers, we attribute the elevation in strength and toughness primarily to changes in the residual stress state at 800–1,000 °C, specifically to the reduction in significant levels of residual tensile stresses in the graphite that are 'frozen-in' following processing. A range of other nuclear grade graphite materials were tested and compared with Gilsocarbon graphite.

1. Introduction
Nuclear-grade graphites are used as a 'neutron moderator' in roughly one-fifth of the world's currently operating nuclear reactors inlcuidng the UK Advanced Gas-Cooled Reactors (AGR). The UK AGRs are designed to operate at temperatures of 420–650 °C, but future Gen-IV designs, such as very high temperature reactors, will potentially operate as high as 1,000 °C with a graphite core. Despite graphite's use in such safety-critical applications, the nano- and micro-structure of nuclear graphite has an extremely high defect population that spans multiple length scales, leading to physical properties that are not completely understood. As the mechanical behaviour of materials is dominated by the presence of defects, we seek here to fully characterize the strength and toughness of this material at typical in-service temperatures between ambient and 1,000 °C.

In particular, Gilsocarbon graphite is currently used in the operating UK AGRs and is a representative material for future nuclear graphites designed for Gen-IV reactors. Gilsocarbon graphite is a moulded, medium-grained, near-isotropic (anisotropy ratio of 1:1.1) GCMB grade polygranular nuclear graphite with the bulk elastic modulus in the range of 10–11 GPa at ambient temperatures. Its structure comprises spherical filler particles of Gilsonite (a naturally occurring solid hydrocarbon bitumen), about 500 μm in diameter, in a binder matrix made from the finer fractions of the coke flour and coal tar pitch. The filler particles, which originate from the coarse-milled Gilsonite coke grains and make up some 70–80% of the total weight, consist of small contiguous crystallites that are misaligned to form a circumferential pattern that contributes substantially to the near-isotropic properties of this grade of graphite.

To investigate how such multiscale defect populations affect mechanical properties at elevated temperatures, here we characterize the (macro-scale) strength and fracture toughness of this Gilsocarbon graphite using three-point bend samples at a temperature range between 20 °C and 1,000 °C. These experiments are performed at a synchrotron X-ray beamline with the samples mounted in a high-temperature test cell (hot cell, see Supplementary Fig. 3). This setup allows real-time three-dimensional (3D) computed micro-tomography, coupled with post-mortem digital volume correlation (DVC), to image

and quantify the micro-scale damage and fracture processes. We find that, contrary to the behaviour observed in the vast majority of materials, both the strength and fracture toughness of nuclear graphite are improved at elevated temperature. We attribute this elevation in strength and toughness to the change of residual stresses accompanied by the closure of nano-scale cracks at temperature.

2. Results

Our experiments afforded two principal findings with regard to Gilsocarbon graphite's mechanical properties. First, with macro-scale tests, the maximum flexural strength of the graphite was found to be about 30% higher at 1,000 °C than at ambient temperature, specifically rising from ~25 MPa at 20 °C to ~32 MPa at 1,000 °C (Fig. 1a). Second, using in situ tomography to quantify the 3D crack geometry, full nonlinear-elastic fracture-mechanics based JR(Δa) crack-resistance curves (R-curves) were derived at 20, 650 and 1,000 °C, as shown in Fig. 1b in terms of J as a function of crack extension Δa; this revealed a corresponding increase in the fracture toughness of the nuclear graphite with temperature. Specifically, mean KIc crack-initiation toughness values increase approximately twofold from ~1–1.5 MPa✓m at 20 °C to ~2–3 MPa✓m at 1,000 °C, when back-calculated in terms of stress intensities. The steepness of the R-curves, representative of the crack-growth toughness, was also observed to increase at the higher temperatures. The full R-curves, re-plotted in terms of stress-intensity factors. Analysis using high-temperature Raman spectroscopy mapping at elevated temperatures showed there are relaxation of tensile residual stresses and this correlated very well with the neutron diffraction data.

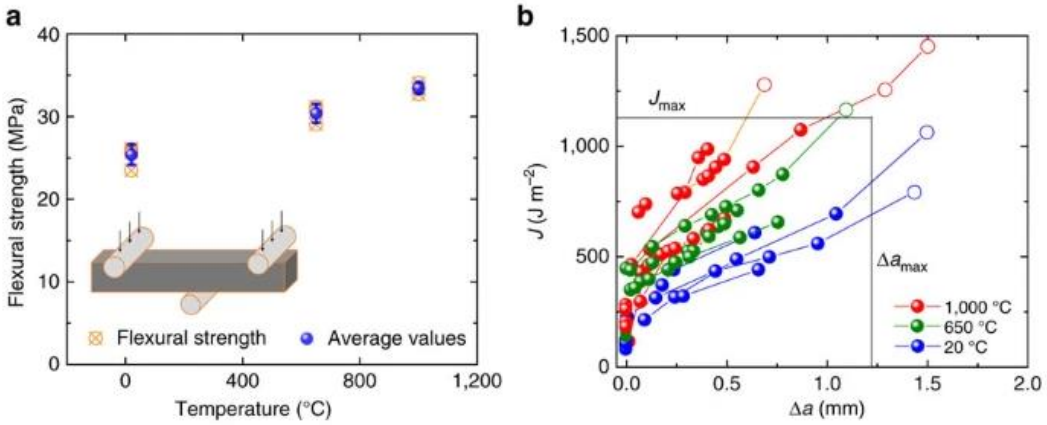

Fig.1. (a) The macro-scale flexural strength (measured on 3–4 mm-sized bend specimens) increases with temperature from ~25 MPa at 20 °C to ~32 MPa at 1,000 °C and (b) the corresponding fracture toughness also increases, in terms of JR(Δa) crack-resistance curves. (It is noteworthy that the open data points on the R-curves in b are outside the maximum J and Δa limits for the size of our specimens, as prescribed by the ASTM E1820 standard41; these data points are not included in the analysis

3. Conclusions

In situ mechanical testing at high temperatures was undertaken, coupled with real-time computed X-ray micro-tomography, to examine the damage evolution, strength and toughness of the nuclear Gilsocarbon graphite, which was observed to display the unusual behaviour of an increase in flexural strength and toughness with increase in temperature. Using additional Raman spectroscopy and neutron diffraction studies, we conclude that the underlying mechanisms responsible for such behaviour are the high-temperature relaxation of tensile stresses 'frozen-in' at ambient temperatures from the high defect concentration that results from cooling from >2,700 °C during manufacture, which aids the closure of nano-cracks at elevated temperatures from the anisotropic thermal expansion in graphite.