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Transmission Electron Microscopy Study of Graphite under *in situ* Ion Irradiation

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Abstract. Graphite is employed as a moderator and structural component in 18 of the United Kingdom's fleet of Magnox and Advanced Gas-cooled Reactors (AGRs). During the operational lifetime of a reactor, graphite undergoes complex physical and mechanical property changes including dimensional modification, owing to the effects of temperature, oxidation and irradiation-induced atomic displacements. In order to safely extend the lifetime of the current fleet of AGRs, and also to develop materials for GenIV concepts such as the Very-High-Temperature Reactor (VHTR), it is important to gain a better understanding of the fundamental atomic processes which underpin the behaviour of graphite under current and future operational conditions.

This study has focused on the effects of temperature and displacing radiation on the evolution of Mrozowski cracks in highly-orientated pyrolytic graphite (HOPG) using the new Microscope and Ion Accelerator for Materials Investigations (MIAMI) facility. This instrument allows transmission electron microscopy to be performed *in situ* whilst simultaneously ion irradiating to radiation damage levels typically reached in a reactor. By using this technique, it is possible to explore the development of radiation damage under a range of different conditions continuously from start-to-finish rather than just observing the end-states accessible in *ex situ* studies.

1. Introduction

Graphite is used as both a moderator and structural component in Magnox and Advanced Gas-cooled Reactors (AGRs). The combined effects of the neutron flux, temperature and oxidation which the graphite experiences cause it to undergo significant physical modification including dimensional change [1–3]. These effects are not fully understood in terms of the underlying fundamental atomistic processes or the implications for continued operation of these reactors. In order to safely extend the operating lifetime of the existing fleet of AGRs and Magnox reactors – as well as develop materials for GenIV concepts such as the Very-High-Temperature Reactor (VHTR) – it is important to gain a better understanding into the behaviour of graphite under these conditions.

One of the features of nuclear graphite which is known to change as a consequence of both displacing radiation and temperature are Mrozowski cracks – the opening and closing of these cracks are two of the phenomena behind dimensional change as well as the modification of other physical properties [4]. In this study, the behaviour of these cracks has been monitored under irradiation with 30 kV He ions and subsequent annealing.

Transmission electron microscopy (TEM) with *in situ* ion irradiation allows the dynamic effects of radiation damage on the structure of materials to be explored at the nanoscale whilst controlling experimental parameters such as temperature. These effects are the result of a complex competition between multiple processes and so it is important to observe them as they occur rather than simply examining the end-states accessible in *ex situ* studies.

2. The MIAMI Facility

The Microscope and Ion Accelerator for Materials Investigations (MIAMI) facility was originally constructed at the University of Salford and has recently moved to the University of Huddersfield [5]. It consists of a JEOL JEM-2000FX 200 kV TEM interfaced to a 100 kV ion accelerator. A summary of the key specifications of the MIAMI facility is given in table 1.

Table 1. Specifications of the MIAMI facility.

Ion accelerating voltage	0.5–100 kV	Resolution (lattice)	0.14 nm
Ion species	H to Xe	Resolution (point)	0.31 nm
Typical flux (10 kV He)	3×10^{13} ions.cm ⁻² .s ⁻¹	Sample tilt	±20°
Ion/electron beam angle ^a	30°	Digital cameras	Gatan ES500W 1.4 MP
Electron beam voltage	80–200 kV		Gatan ORIUS 4 MP
Magnification range	50–800 k	Sample temperature	100–373 K or RT to 1273 K

^a Previously 25°.

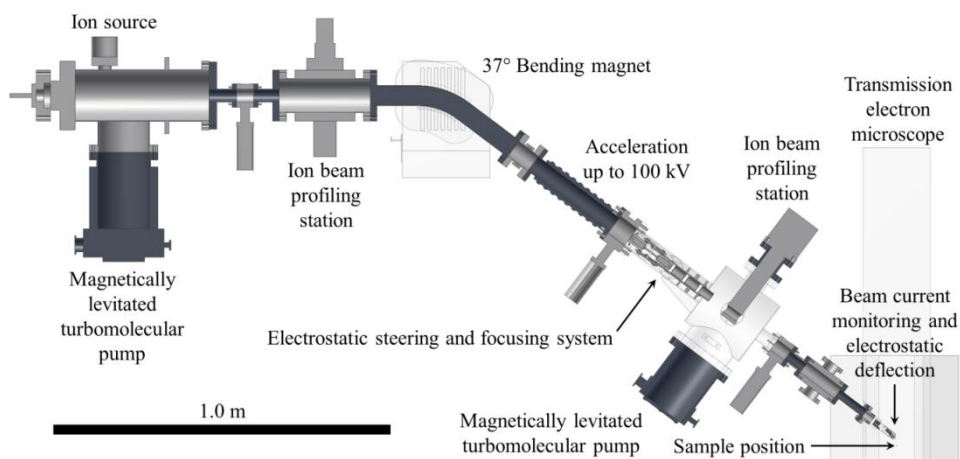


Figure 1. Key components of the MIAMI facility.

A Colutron G-2 is used as the ion source and can generate ion beams from either a gas supply or solid charge with an accelerating voltage of up to 10 kV. Whilst this is adequate for many experiments including the implantation of light atoms such as H or He into most samples, the source can also be floated up to 100 kV with post acceleration performed after the bending magnet – see figure 1. The ion beam then enters the TEM via a high take-off angle x-ray port at an angle of 53° to the vertical. Electrostatic deflection is then performed inside the microscope column in order to make the ion beam incident upon the sample at angle of 30° to the vertical.

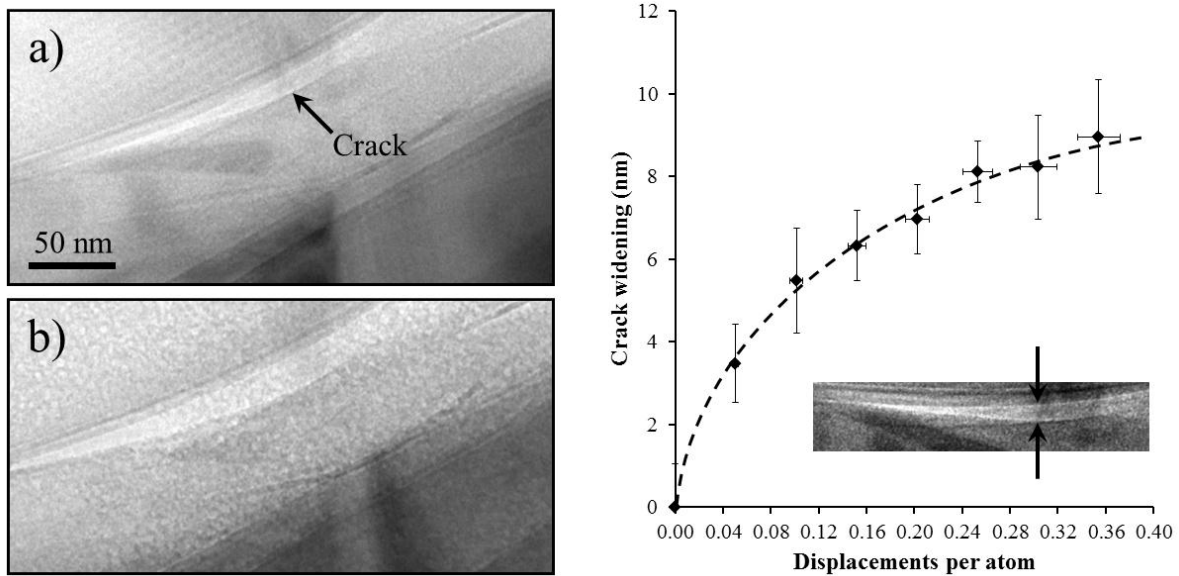


Figure 2. Bright-field transmission electron micrographs (underfocus $\approx 1 \mu\text{m}$) showing the widening of a Mrozowski crack in HOPG under 30 keV He irradiation at room temperature: a) virgin; and b) after a fluence of $4.5 \times 10^{16} \text{ ions.cm}^{-2}$ (0.35 dpa). Scale marker applies to both micrographs. Trendline provided as guide to the eye and is not fitted to data.

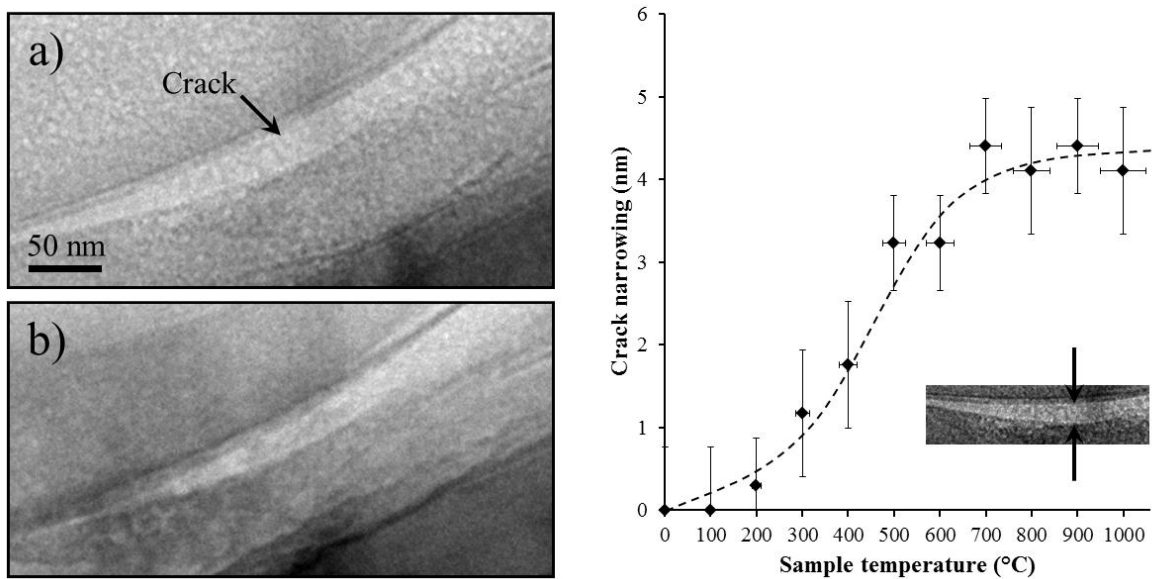


Figure 3. Bright-field transmission electron micrographs (underfocus $\approx 1 \mu\text{m}$) showing the narrowing of an irradiation-widened Mrozowski crack in HOPG during temperature-ramp to 1000°C: a) room temperature; and b) after ramp to 1000°C. Scale marker applies to both micrographs. Trendline provided as guide to the eye and is not fitted to data.

3. Experimental Method

Electron-transparent samples of graphite for TEM were produced by exfoliation of bulk highly-orientated pyrolytic graphite (HOPG) using adhesive tape. The adhesive was dissolved using acetone and the thin layers of graphite were captured in copper mesh oyster-grids.

TEM of *in situ* ion irradiation and annealing was performed at room temperature using the MIAMI facility [5] using an electron beam accelerating voltage of 200 kV. Irradiation was performed using 30

keV He ions with a flux of 2×10^{13} ions.cm⁻².s⁻¹. Using this ion species and energy, 1 displacement per atom (dpa) can be achieved in graphite at a fluence of approximately 5×10^{16} ions.cm⁻² whilst introducing only 0.1% additional atoms to a 100 nm thick sample according to calculations performed using the Stopping and Range of Ions in Matter (SRIM) 2008 computer program [6]. The electron beam was kept on during both irradiation and annealing to allow video to be captured. Still images and video were both recorded using a Gatan ORIUS SC200 CCD digital camera and sample annealing was performed using a Gatan Model 652 double-tilt heating holder with a ramp rate of 1°C.s⁻¹.

4. Results

Mrozowski cracks were observed to widen under irradiation by 30 keV He at room temperature (see figure 2) and to narrow during annealing to 1000°C (see figure 3). The crack shown in figures 2 and 3 was observed to widen significantly to 0.2 dpa and to widen more slowly at higher doses; during annealing it was observed to narrow during annealing to 800°C. No significant effects were observed as a result of electron beam irradiation (without ion beam) and the sample did not undergo any detectable tilting which could otherwise account for the observed changes to crack width

5. Discussion and Conclusions

The widening of Mrozowski cracks under displacing irradiation could be caused by either the rearrangement of the material inside the cracks (possibly under strain applied by the adjoining crystal) or by the transport of displaced carbon atoms from the surrounding bulk. Assuming the structure factor of the material inside the crack was relatively constant during the irradiation, the mass-thickness contrast within the crack in figure 2 appears to remain constant suggesting that the latter is the more likely mechanism.

As the narrowing of the crack was observed to proceed rapidly above 400°C and to be complete by 800°C, it would appear that the mechanism(s) responsible were activated in this temperature range. Previous work by Wen *et al* concluded that crack closure in HOPG during annealing to 800°C was caused by dimensional changes in the surrounding crystals due to thermal expansion [7].

The crack followed in this study was parallel to the edge of the sample and far from the support grid which was in good contact with, but not physically attached to, the HOPG. Therefore, it is not expected that differing coefficients of thermal expansion between the grid and sample could have contributed to the narrowing of the crack.

It should be noted that the crack widened by approximately 9 nm under irradiation but narrowed by only a little more than 4 nm under annealing to 1000°C. It is possible that further narrowing may occur at temperatures higher than those available in the MIAMI facility. Further exploration of both phenomena is required to ascertain the mechanisms and activation energies for crack growth and shrinkage and will be the subject of further study.

Acknowledgments

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