1568
doi:10.1017/S1431927619008572
Microsc. Microanal. 25 (Suppl 2), 2019

The Temperature Dependence of Defect Evolution in Irradiated Graphite

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Graphite is commonly used as a moderator in many reactor designs and is a leading candidate core material for the envisioned Generation IV reactor concepts like the very high temperature reactor (VHTR). In reactors graphite is exposed to high-temperature neutron irradiation leading to the creation of lattice defects. These lattice defects cause changes in the mechanical properties of graphite which may adversely affect the performance of reactor components. When initially subjected to neutron irradiation, volumetric shrinkage is observed in nuclear graphite. With prolonged irradiation a turnaround point is reached where the volume then begins to increase. The turnaround point has been shown in literature to be a strong function of temperature [1], which suggests that there exists a temperature dependence to the atomistic nature of accumulated irradiation damage; however, the atomic mechanisms governing this defect evolution are not well understood. This knowledge gap is partly due to a lack of experimental data and difficulties in observing the dynamic nature of the defects.

In situ transmission electron microscopy (TEM) provides a method to monitor the dynamic atomic response of graphite during high-temperature irradiation. In situ electron irradiation and thermal annealing experiments were performed on nuclear graphite IG-110 and natural graphite. IG-110 was prepared by conventional ion-milling techniques for irradiation experiments conducted perpendicular to the *c* axis. In addition, irradiation experiments were conducted on natural graphite parallel to the *c* axis. Graphite specimens were irradiated with a 200 kV electron beam between a temperature range of 25-800°C. Electron energy loss spectroscopy (EELS) was conducted on irradiated areas to analyze bonding character and density. In addition, in situ annealing experiments were conducted at low operating voltages (80 kV) on IG-110 specimens prepared to electron transparency via oxidation-based methods. Oxidized specimens contain no milling-induced artifacts; therefore, experimentally observed defect structures are only the result of irradiation.

Figure 1 shows (a) bright-field and (b) dark-field micrographs of natural graphite irradiated with equivalent electron-irradiation doses at temperatures of 25°C, 400°C, and 800°C. A clear distinction between size, shape and distribution of accumulated irradiation damage is observed at the respective temperatures. Figure 1(c) shows EELS spectra of the respective areas. Assuming a quasi-free electron model, the energy shift of the $\pi+\sigma$ plasmon peak may be correlated to density, in which case a trend of decreasing density as irradiation temperature increases is observed. Figure 2(a) shows an HRTEM image of an oxidized specimen of IG-110 conducted at 800°C with near-zero electron irradiation. At temperatures above 700°C, basal planes often curl and close, which behavior was not observed previously at lower temperatures [2]. Figure 2(b) shows an ion-milled specimen of IG-110 imaged 800°C with near-zero irradiation where the curling and closure of basal planes is also observed. Figure 2(c) shows the same crystallite in (b) post electron-irradiation, where basal planes curl and recombine around a prismatic dislocation resulting in swelling along the *c* axis.

Electron-irradiation studies show clear evidence that the nature and accumulation of defects changes significantly as a function of temperature. In situ TEM results reveal new atomic mechanisms governing dimensional change in irradiated graphite via the formation of newly discovered defect species in high-temperature irradiated graphite. In situ annealing experiments on graphite TEM specimens prepared via oxidation establish a temperature regime where the curling and closure of basal planes due solely to annealing is favourable. In addition, EELS analysis shows qualitative trends in density and bonding changes as a function of irradiating temperature [3].

References:

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[3] The authors acknowledge funding from U. S. Department of Energy's EPSCoR-State/National Laboratory Partnership Program (Award # DE-SC0016427), and that work at the Molecular Foundry was supported by the Office of Science, Office of Basic Energy Sciences, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231. This document was prepared by Steve Johns as a result of the use of facilities of the U.S. Department of Energy (DOE), which are managed by The Regents of the University of California, acting under Contract No.DE-AC02-05CH11231. Neither The Regents of the University of California DOE, the U.S. Government, nor any person acting on their behalf: (a) make any warranty or representation, express or implied, with respect to the information contained in this document; or (b) assume any liabilities with respect to the use of, or damages resulting from the use of any information contained in the document.

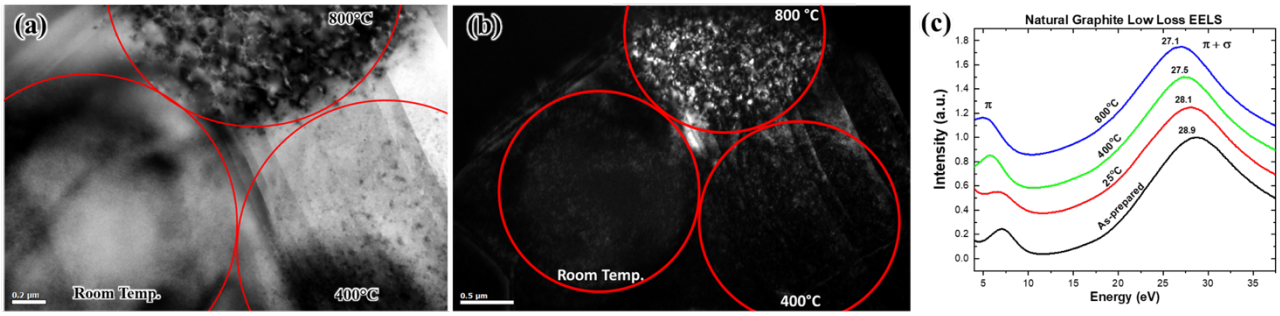

Figure 1. (a) Bright-field and (b) dark-field micrographs of electron-irradiated natural graphite conducted at 25°C, 400°C, and 800°C. (c) Low loss EELS on the respective areas shown in (a) & (b).

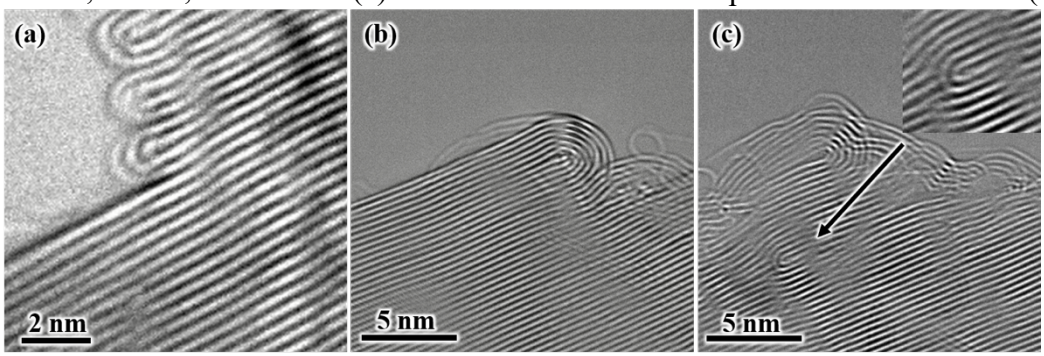

Figure 2. (a) Oxidized and (b) ion-milled specimens of IG-110 imaged at 800°C showing the curling and closure of exposed basal plane edges. (c) Post electron-irradiation of the crystallite in (b) showing basal planes which curl and recombine around a prismatic dislocation.

https://doi.org/10.1017/S1431927619008572 Published online by Cambridge University Press