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EFTEM Pre- and Post-Irradiation sp^2 to sp^3 R-Ratio Measurements Of SiC/SiC Pyrolytic Carbon Interphases

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This paper presents the results of ongoing investigations on the behavior of a specific component of silicon carbide based ceramic matrix composites (SiC/SiC) under irradiation, namely the pyrolytic carbon (PyC) interphase linking the matrix and fibers together. SiC/SiC composites are currently under heavy investigation because of their potential as a new nuclear fuel cladding to replace the currently used Zirconium based alloys. ENERGY FILTERED TEM (EFTEM) is used to study the hybridization of the carbon atoms forming the aforementioned interface layer by measuring the ratio between sp^2 and sp^3 bonding states (R-ratio), before and after irradiation with ions and neutrons. Indeed, the initial state of the interphase is graphite-like PyC, in which the electrons occupy the π bonding states of the sp^2 orbitals. The ratio between the π and σ occupied levels is different in each carbon allotropes, be it amorphous, graphite or diamond. An effect that radiation damage is known to have is to amorphize and micro-crack nuclear graphite [1], [2]. In cases where the graphitic interphase would become completely amorphous, a potentially impactful change in the composite mechanical and thermal properties would occur.

In this work, TEM lamellae prepared using a focused ion beam (FIB) from both pristine and neutron irradiated SiC/SiC have been examined. These samples are extracted from actual prototype cladding sections produced by General Atomics. In addition to this, the samples prepared from pristine material have been subsequently irradiated in the in-situ TEM JANNuS facility in Orsay, France [3], [4]. Using the unique capabilities of this apparatus, self-ion irradiations with both Si^+ and C+ were performed, up to damage levels approaching one displacement per atom (dpa), whilst observing the effects of said ions at the same time with a TECNAI G^2 20 Twin TEM. The EFTEM measurements are carried out at EPFL, Lausanne, using a Schottky FEG JEOL 2200FS with an in-column omega filter. All of these measurements were carried out as close as technically possible to magic angle conditions[5]. Quantifying the ratio between the peaks corresponding to the π^* and σ^* anti-bonding states is an issue which can be approached with different techniques, two of which are used in the present work and illustrated in Figure 1. First, the TWO WINDOWS METHOD (TWM) [6], whereby the absorption spectrum is numerically integrated over two energy ranges or windows are centered on the aforementioned peaks. Second, spectrum fitting using GAUSSIAN functions [7] is also used to ensure the robustness of the measurement.

The maps displayed in Figure 2 show an overall decrease of the R-ratio after irradiation of the samples, meaning that even sub-dpa damage levels are already inducing an amorphisation of the PyC interlayer. Additionally, the overall layer is homogeneously amorphized, as seen with the very uniform color code of the post irradiation map. Additional data such as local RMS roughness and the overall map standard deviation of the ratio map further corroborates these results.

The authors would like to thank Westinghouse and General Atomics for providing the prototype cladding tubes within the CARAT research program as well as JANNuS-Orsay part of the CSNSM, Orsay, France which is part of the EMIR French accelerators network, where the in-situ experiments were carried out.

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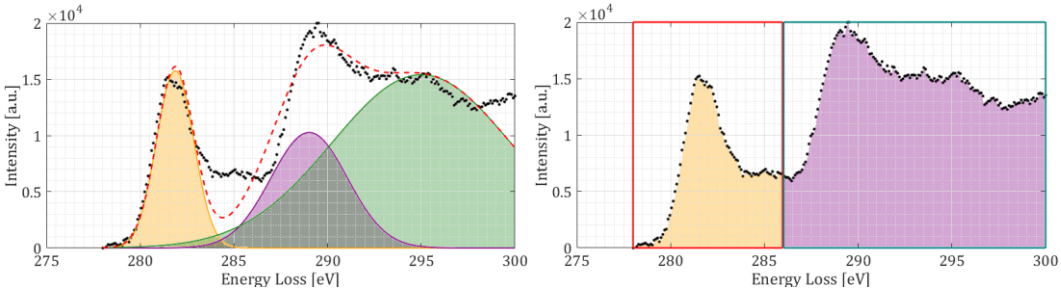

Figure 1: R-ratio quantification of a highly oriented pyrolytic graphite (HOPG) reference via Gaussian fitting (left) and two-windows method (left)

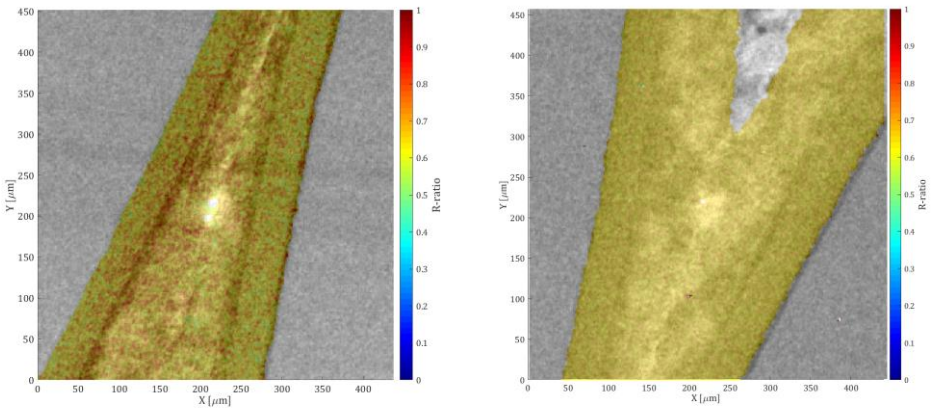

Figure 2: R-ratio maps acquired on a pristine (left) and ion irradiated (right) pyrolytic carbon interphase. Both maps have been recorded at different locations of the same sample

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