EXPERIMENTAL STUDY OF RADIATION DAMAGE IN CARBON COMPOSITES AND GRAPHITE CONSIDERED AS TARGETS IN THE NEUTRINO SUPER BEAM *

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Abstract

The unique properties of fiber reinforced carbon composites as well as of newly developed grades of graphite have prompted the interest of high power accelerators such as the multi-MW neutrino superbeam as the material for their performance target. Properties of primary interest are the shock and radiation damage resistance of these materials. To assess their behavior under proton irradiation shock and radiation damage experiments have been performed at Brookhaven National Laboratory. This paper presents results of the prolonged radiation exposure of these materials and assesses their suitability as high performance accelerator targets.

INTRODUCTION

Carbon composites have been of primary interest as materials of choice for a multi-MW neutrino superbeam which desires low-Z pion production target. Figure 1 schematically depicts a concept of high power target that relies on the performance of carbon-carbon composite at the MW power level. Beam on target experiments conducted at BNL [1, 2, 3] made the case stronger in their favor, as compared to graphite, by demonstrating their excellent shock resistance which is directly linked with their extremely low thermal expansion. In addition, the superior thermal conductivity characterizing these composites, a property required to quickly discharge energy deposited in the target to the heat sink, made these composites more attractive to graphite. However, since target survivability also depends on resistance to prolonged radiation, a series of irradiation damage studies on carbon composites and graphite were launched. This particular effort was undertaken to assess the effect of proton irradiation on this materials and potentially arrive at some correlation with neutron-based experience data that have been available for these materials from reactor operating experience. Past studies [2] on the response of carbon-carbon composites with different structure (1-D, 2-D and 3-D fiber weaving) under neutron irradiation up to several displacement-per-atom (dpa) damage revealed that these composites experience serious swelling and shrinkage depending on the fiber orientation. In particular, the anisotropic behavior under neutron irradiation manifested itself rapid shrinkage along the fiber direction and swelling normal to the fibers. This anisotropic behavior may have serious implications in the high power target scenario. Figure 2 displays neutron irradiation effects on the dimensional changes of 2-D and 3-D carbon composites [2]. Studies have also focused on the degradation of thermal conductivity as a result of prolonged exposure to neutrons. Figure 3 depicts the dramatic reduction in conductivity observed [3] that takes place in both graphite and carbon composites even at modest irradiation levels.

Using the BNL accelerator complex and in particular the BNL Isotope Production Facility which receives 200 or 117 MeV, 90 μA proton beam from the Linac, a series of irradiation phases were conducted. The effects of the proton irradiation on different graphite grades and carbon composites were studied through extensive post-irradiation analyses that focused on physical and mechanical property degradation. It was observed that while carbon composites at moderate doses exhibited interesting behavior of damage reversal through thermal annealing, at higher dose levels of peak proton fluences $>5x10^{20}$ protons/cm^2 they exhibited serious structural degradation. Also, the experimental study showed that graphite suffered similar structural damage when subjected to the same proton fluence. The latter was a surprise given that reactor experience on graphite indicates that graphite has exhibited survivability under high neutron fluence and to estimated radiation damage of several dpa. It appears that the effects of neutrons and protons on the structure of the material are very different and therefore attention needs to be paid in establishing the right correlation so the wealth of data from reactor, operations can be utilized in the accelerator field.

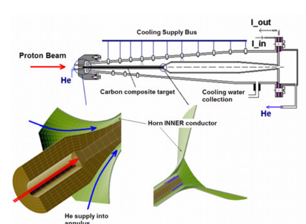

Figure 1: Neutrino super-beam target schematic

EXPERIMENTAL RADIATION DAMAGE

Using the BNL Isotope Production Facility for irradiation and the BNL Hot Cell Lab for post-irradiation analysis, a series of irradiation phases were conducted. During the first phase which led to moderate dpa levels for these low-Z materials (~0.02 dpa) the 3-D carbon composite and the IG-43 graphite were irradiated. The

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post-irradiation analysis addressed the changes that occur in the stress-strain relations as well as the thermal expansion of these materials. The interesting damage reversal behavior achieved through annealing that was exhibited by the carbon composite prompted a second irradiation phase with primary candidate the 2-D structured carbon composite. The higher dose achieved during the second phase revealed that while the 2-D composite exhibits similar annealing behavior as the 3-D counterpart, it suffers structural degradation above a fluence threshold. This prompted a third investigation which addressed the effects of higher dose on the 2-D and 3-D carbon composite as well as the IG-43 and isotropic IG-430 graphite grades when exposed to same conditions. The results and observations of the post-irradiation analysis of the various phases are presented below.

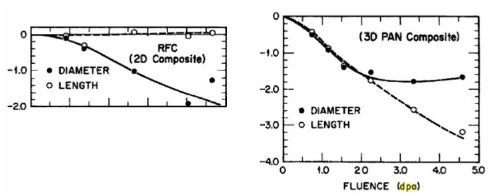

Figure 2: Neutron irradiation dimensional change effects on carbon composites [2]

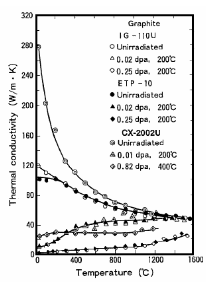

Figure 3: Irradiation effects on thermal conductivity of graphite and carbon composites [3]

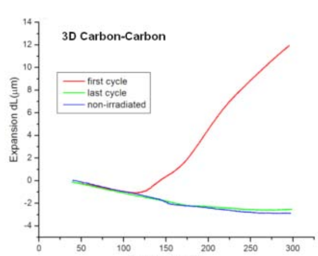

Figure 4: Thermal annealing of irradiated 3D carbon using the BNL 200 MeV proton beam

The post-irradiation analysis of the 3-D composite under modest irradiation damage (0.02 dpa) revealed that the composite is capable of reversing the damage which is in the form of swelling or shrinking through annealing. Shown in Figure 4 is the interesting behavior of the composite. As also observed under neutron irradiation, the material experiences shrinking along the fiber direction. The first thermal cycle, as shown in Figure 4, restores the material behavior in terms of its thermal expansion which has a small but negative value below 800 C. Figure 5 depicts the restoration of the thermal expansion coefficient along the fibers and the 45^{o} plane.

During the second irradiation phase the 2-D carbon composite was exposed to a peak fluence of ~0.5x10^{21} p/cm^{2}. At fluence levels below the peak value the 2-D composite exhibited strikingly similar annealing behavior to the 3-D counterpart. Figure 6 depicts the restoration of the irradiated material along the fiber orientation of two independent irradiated specimens. As was observed in the 3-D case, the material underwent shrinkage along the fiber plane during irradiation. The finding confirms what was also observed for this carbon structure in the neutron irradiation of [2].

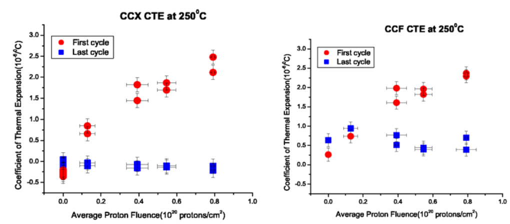

Figure 5: Thermal annealing of irradiated 2D carbon

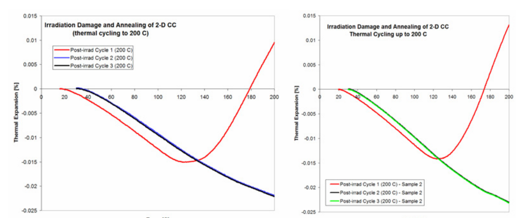

Figure 6: Annealing in 2D carbon exposed to low dose

In the direction normal to the fiber plane the material experiences swelling. Thermal annealing restores the material along that direction also as shown in Figure 7 where the dose effect is captured. Clearly restoration in both directions takes place through marked changes in the thermal expansion coefficient. Important information is depicted in Figure 8 where the annealing cycles to different temperatures are successively applied on the irradiated material. Progressive restoration of the irradiated material is achieved as the annealing temperature increases eventually restoring the material for the entire temperature range. This is a very important finding that points to higher operating temperatures as more appropriate for these composites.

The effect of proton irradiation on the physio-mechanical properties of graphite grades was also explored during the three irradiation phases. Figure 9 depicts the irradiation effect on the thermal expansion coefficient of the IG-43 graphite. The radiation effect on the strength and the stress-strain behavior of IG-43 is shown in Figure 10. The clear effect is the strengthening of graphite with irradiation, a feature exhibited by most materials after irradiation. Shown in Figure 11 is the thermal expansion behavior of the isotropic IG-430 graphite. As shown, the material is stable under irradiation.

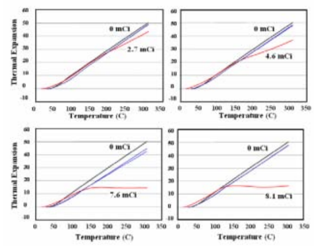

Figure 7: Annealing of 2D carbon in the “weak” direction

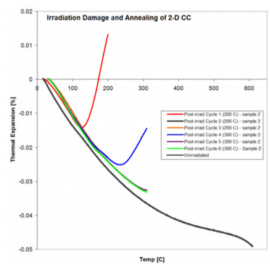

Figure 8: Temperature-dependant annealing of 2-D carbon

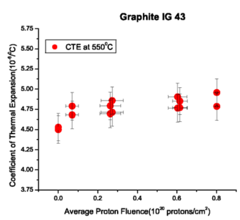

Figure 9: Irradiation effect on the IG-43 graphite CTE

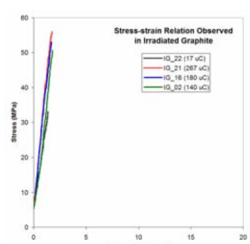

Figure 10: Irradiation effect on the IG-43 graphite stress-strain relationship

While the low dose exposure revealed very interesting annealing behavior of the carbon composites, the higher proton fluence led to serious structural degradation. This was first observed during the irradiation of the 2-D composite. To assess whether this behavior is unique to only the 2-D structure, the experiment was repeated to same fluence level while exposing the 2-D and 3-D composites along with the IG-43 and the IG-430 graphite grades. In addition a special bond between titanium and IG-43 graphite was also included. As shown in Figures 12 and 13 the carbon composites and the IG-43 graphite all suffer structural degradation at fluence > 0.5×10^{21} protons/cm^2. It appears that there is a clear threshold that governs the behavior. Obviously this effect is different than the neutron irradiation effect on these materials and further studies are needed to explain the damage mechanism. The IG-430 did not quite reach the threshold fluence but it shoed no degradation signs. Its resistance to high fluence needs to be further explored.

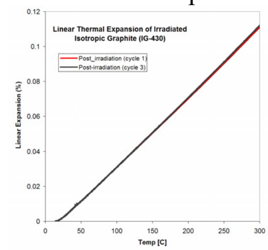

Figure 11: Irradiation effect on the thermal expansion of the isotropic IG-430 graphite

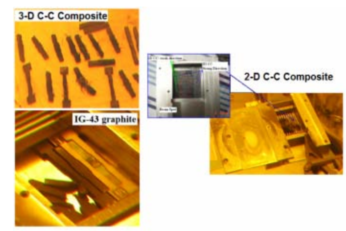

Figure 12: Special graphite and titanium bond

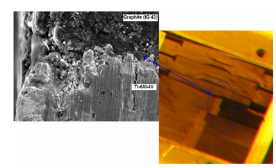

Figure 11: Special graphite and titanium bond

SUMMARY

The extensive array of irradiation experiments on the behavior of carbon composites and graphite grades revealed that while the composites exhibit interesting damage reversal behavior at low proton fluences, they, along with typical graphite grades, experience structural degradation at levels expected during their life time as proton target in high power accelerators. Their behavior under proton exposure appears to be very different than the behavior observed under neutron radiation at much higher dose levels. Further investigations are needed to understand the damage mechanism. The study also confirmed the dramatic loss of conductivity in both the graphite and the carbon composites.

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