RADIATION DAMAGE STUDY OF GRAPHITE AND CARBON-CARBON COMPOSITE TARGET MATERIALS*

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Abstract

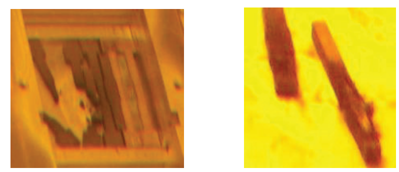
Use of graphite and carbon-carbon composite materials as high intensity proton targets for neutrino production is currently thought to be limited by thermal and structural material properties degraded by exposure to high energy proton beam. Identification of these limits for various irradiation and thermal environments is critical to high intensity targets for future facilities and experiments. To this end, several types of amorphous graphite and one type of carbon-carbon (3D weave) composite were exposed to 180 MeV proton beam at the BNL BLIP facility. Irradiated samples were then thermally, ultra-sonic, and structurally tested and compared to un-irradiated samples. Results show significant changes in material properties even at very low damage levels ($<$ 0.09 DPA) and that significant interstitial annealing of these properties occurs at annealing temperatures only slightly above irradiation temperature. This points the way to optimizing target operating temperature to increase target lifetime. A description of the plan to explore radiation damage in target materials through the new RaDIATE collaboration (Radiation Damage In Accelerator Target Environments) is also presented.

INTRODUCTION

Multi-MW high performance particle production targets are key toward the next generation of accelerator machines for future neutrino and other rare particle beams. One of the future MW accelerators is the LBNE experiment where Fermilab aims to produce a beam of neutrinos with a 2.3 MW proton beam (1.6e14 p/pulse, σ_{rms} = 1.5-3.5 mm, and a pulse length of 9.8 μs) as part of a suite of experiments associated with Project X. These parameters are expected to push many target materials to their limit thus making target design very challenging.

Prior radiation damage studies conducted within the last decade at Brookaven National Laboratory (BNL) using 200 MeV protons at the Brookhaven Linac Isotope Producer (BLIP) target station for LHC and Neutrino Factory revealed structural damage of graphite and carbon composites at proton fluence of about 10^{21} p/cm^2 (Fig. 1) [1]. Moreover, recent performance results from the NuMI experiment indicated that target irradiation damage may be responsible for the gradual degradation of neutrino yield.

Prompted by these experimental and operational observations on targets made of materials desired for the multi-MW LBNE, a comprehensive experimental effort was launched to assess and quantify potential limitations of LBNE target materials using the BNL proton beam. The main objectives were to evaluate irradiation-induced changes in the thermal and mechanical properties of different low-Z target materials, irradiated at varying DPA levels.



(a) IG-43 (b) 2D C/C

Figure 1: Irradiation induced structural damage by 200 MeV protons at BLIP [1].

IRRADIATION EXPERIMENT

The BNL Linac can deliver protons of energies between 66 and 200 MeV, with a pulse length of 525 μs. The BLIP target station, primarily operated to produce medical isotopes, fully arrests the primary proton beam from the Linac operating at 116 MeV. Therefore, to perform the irradiation experiment in tandem with the isotope production, the Linac energy was increased so that beam energy was degraded to the desired beam parameters for optimal isotope production after passing through the target array. As a result, significant fine tuning and multiple sensitivity studies were performed to optimize and configure the final target array.

A comprehensive study, using the MARS15 Monte Carlo code [2], was also launched to confirm whether the irradiation damage experiment at BLIP would be of value and able to answer important target survivability questions associated with the multi-MW LBNE experiment. Table **??** shows a direct comparison of target damage between BLIP and LBNE [3], which demonstrates the significant effect of proton energy on damage (accelerated damage at lower proton energy [4]). It was therefore estimated that for the assumed BLIP beam parameters, one year of accumulated damage of the LBNE target (POCO graphite), operating at 120 GeV/700 kW, can be achieved at BLIP in about 9 weeks of irradiation.

The LBNE BLIP irradiation experiment included various graphite grade specimens (POCO, IG-430, SGL R7650 and C-2020) as well as the 3D C/C composite (orthogonal

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Table 1: MARS15 target damage comparison between 165 MeV BNL BLIP and 120 GeV LBNE protons [3]

	NuMI/LBNE	BLIP
E_p (GeV)	120	0.165
Beam σ (mm)	1.10	4.23
N_p (1/yr)	4.0e20	1.1e22
DPA (1/yr)	0.45	1.50

3D weave). The peak DPA damage in the target specimens was estimated by MARS15 simulations to be about 0.095 [5]. Following irradiation, the target array was allowed to cool down for several months before being transported to the hot cell facility, where post-irradiation experiments were carried out.

RESULTS AND DISCUSSIONS

Thermal Stability and Damage Annealing

Upon irradiation, the lattice structure of graphite changes due to the production of interstitial atoms and vacancies. Interstitial atoms move between layer planes and can be mobile at very low temperatures. On the other hand, vacancies have been assessed to be mobile at temperatures greater than 1000 K [6]. Therefore, to enable partial annealing of the damage (portion attributed to interstitial atoms), temperatures higher than the irradiation temperature should be induced, assuming that interstitial atoms mobile up to that temperature have already been placed back in the lattice, or pinned at grain boundaries. To investigate this annealing process, thermal strain measurements of the specimens were made using a dilatometer.

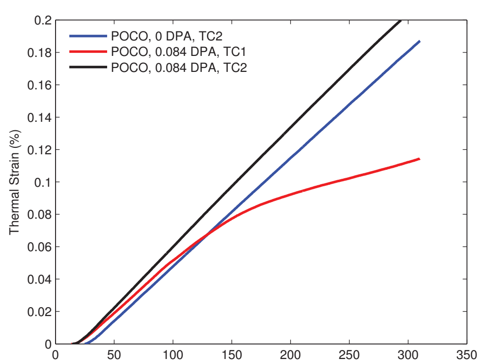

Figure 2: Dimensional changes in un-irradiated and irradiated POCO graphite over two thermal cycles.

Figure 2 shows thermal strain measurements of un-irradiated and irradiated POCO graphite at 0.084 DPA as a function of temperature over two thermal cycles. Upon the first thermal cycle to 300 °C, the irradiated specimen shows significant decrease in thermal expansion. However, in subsequent cycles, the graphite appears to have recovered its un-irradiated expansion characteristics and shows a slightly higher thermal expansion than the control specimen. This trend is qualitatively similar across all the graphite grades [5]. The irradiation temperature, which is about 150 °C, can also be inferred from Fig. 2 (point where irradiated curve diverts from un-irradiated curve). The experimental results give further insight into the interstitial atom annealing and damage reversal process. Similar damage reversal phenomena were observed with the 3D C/C composites, exposed in both water and argon environments during the experiment (Fig. 3). Results shown in Figs. 2 and 3 confirm that significant thermal property changes can occur at low DPA levels while damage reversal is possible at annealing temperatures higher than the irradiation temperature, thus providing practical information on optimal operating temperature of future targets.

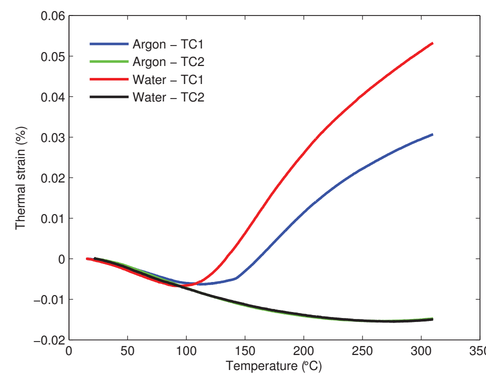

Figure 3: Thermal cycling following irradiation of 3D C/C composite specimens.

Mechanical Testing

Tensile tests were performed on irradiated graphite specimens to investigate the effect of radiation damage on tensile strength and elastic modulus, E. The tensile test results for IG-430 graphites, irradiated at different DPA values, are shown in Fig. 4 and indicate increased elastic modulus and tensile strength than the un-irradiated specimen. However, no distinct dependence on DPA is observed, mainly due to the interrelation of DPA and irradiation temperature. As described in the previous subsection, the irradiation temperature dictates the extent of damage annealing.

Figure 5 shows the damage reversal process of irradiated POCO graphite at 0.07 DPA due to annealing at 350 °C. Both the elastic modulus and the tensile strength revert back to the un-irradiated state. Tensile tests of 3D C/C composites did not provide reliable data because the specimens consistently slipped or failed at the head. Further studies will employ a bending test fixture to evaluate the flexural strength of the 3D C/C composite specimens.

Ultrasonic Tests

Ultrasonic tests were performed to further quantify the change in elastic modulus due to irradiation and post-irradiation annealing [5]. The wave velocity, which is proportional to the elastic modulus, was estimated by mea-

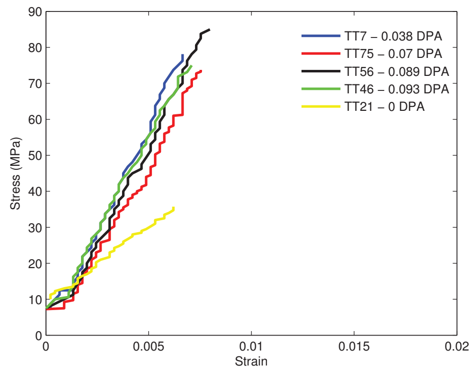

Figure 4: Stress strain curves for irradiated IG-430 at varying DPA levels.

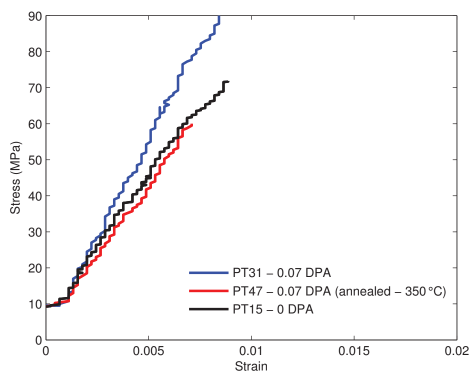

Figure 5: Annealing effect on tensile properties of POCO.

suring the travel time and propagating length of the waves through the specimens.

Figure 6 shows velocity measurements for C-2020 as a function of temperature, with specimen CC12 previously annealed at 310 °C and CC6 unannealed. As seen for temperatures at and below the irradiation temperature (150 °C), the percent velocity change remained constant for both specimens. However, due to interstitial mobilization above the irradiation temperature, a downward trend is exhibited by CC6 after 150 °C while CC12 (previously annealed to 310 °C) remained unchanged until about 310 °C. These results again confirmed changes in elastic modulus due to low DPA irradiation and its annealing-induced restoration.

Ultrasonic tests were difficult to apply to 3D C/C composites because the array and layers of fibers scatter and diffuse the ultrasonic pulse, which has a wavelength smaller than the diameter of each fiber.

FUTURE STUDIES

RaDIATE Collaboration

The Radiation Damage in Accelerator Target Environment collaboration's main objective is to further radiation damage studies in target materials by generating new and useful materials data for application within the accelerator and fission/fusion communities. Current collaborators include scientific and engineering experts from FNAL, PNNL, STFC, BNL and Oxford University with interest in graphite, beryllium and tungsten.

A post-doctoral fellow, based at the Materials for Fusion and Fission Power group at Oxford University, will execute beryllium research over a three year period. Graphite radiation damage studies will be ongoing at BNL while tungsten studies are planned to be carried out at Rutherford Appleton Laboratory (STFC), where prior experience with the ISIS tungsten target stations is at hand. There are opportunities for collaborations with other institutions and more information on the scope and plan of the program is available on the RaDIATE website (http://www-radiate.fnal.gov/index.html).

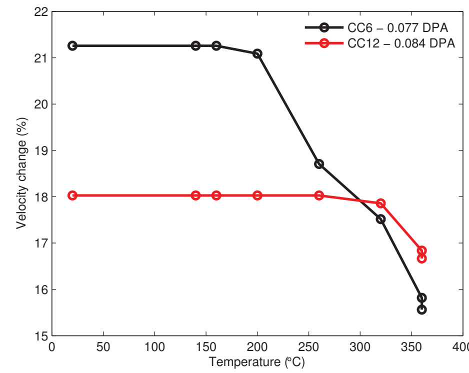

Figure 6: Ultrasonic velocity recovery in irradiated and annealed C-2020 specimens.

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