THE RADIATE COLLABORATION R&D PROGRAM: STATUS AND UPDATE*

K. Ammigan†, P. Hurh, B. Hartsell, N. Mokhov, FNAL, Batavia, IL 60510, USA
N. Simos, BNL, Upton, NY 11973, USA

Abstract

With the increasing power of future proton accelerator sources impacting target facilities mostly, there is an immediate need to further the understanding of the structural radiation response of window and target materials. The RaDIATE (Radiation Damage In Accelerator Target Environments) collaboration, currently including various US national laboratories and UK institutions, draws on existing expertise in the fission and fusion power materials field to formulate a research program that applies a unique combination of facilities and expertise of the participating institutions to a broad range of high power accelerator projects of interest. The research program includes activities to develop a better understanding of radiation damage mechanisms and the related thermal and mechanical properties response for materials of interest to future high power proton beam target facilities. The window/target materials currently identified as being of particular interest include graphite, beryllium and tungsten. Graphite radiation damage (proton-irradiated) research is already ongoing at Brookhaven National Laboratory as part of the RaDIATE program. This work along with the status and future plans of the RaDIATE collaboration R&D program will be presented.

INTRODUCTION

The increasing power of future proton accelerator particle sources and target facilities calls for the need to better understand and predict the radiation response of structural window, target and collimator materials. In addition, next generation fission reactors as well as the demonstration of fusion reactors, will require components that endure more extreme temperature and irradiation environments than current reactors. Consequently, the RaDIATE (Radiation Damage In Accelerator Target Environments) collaboration [1] aims to draw on existing expertise in the related fields of fission and fusion research to formulate and implement a research program which will generate new and useful materials data for application within both accelerator and fission/fusion communities. This involves recruiting and developing new scientific and engineering experts to initiate and coordinate a continuing synergy between research in these currently disparate communities, with eventual benefit to both proton accelerator in science and industry and carbon-free energy technologies.

The initial scope of the program involves radiation damage study of materials which are of particular interest for targets and beam windows. The materials include graphite and carbon-carbon composites (for existing and future neutrino facility targets), beryllium (for beam windows and future neutrino facility targets), and tungsten (for spallation neutron source targets and muon production targets). Graphite has a long history of use in nuclear fission applications backed by a large body of research on irradiation effects mostly due to low-energy neutrons. For beryllium and tungsten, there is only limited and recent work directed at fast neutron environments, such as the Joint European Torus (JET) fusion reactor at Culham Center for Fusion Energy (CCFE), which is equipped with a tungsten/beryllium wall system. However, the irradiation environment in proton accelerators is somewhat different to reactors due to the high energy and intensity of the pulsed proton beam, which lead to relatively higher gas production rates, higher DPA dose rates, and larger temperature and DPA dose concentrations/gradients in the irradiated material (Table 1). As a result, further research and understanding of radiation damage response in proton-irradiated materials is critical.

Table 1: Comparison of irradiation parameters [2].

Source	**DPA rate (DPA/s)**	**He gas prod. (appm/DPA)**	**Irradiation temp. (°C)**
Fission reactor	3×10^{-7}	0.1	200-600
Fusion reactor	1×10^{-6}	10	400-1000
HE proton beam	6×10^{-3}	100	100-800

The ultimate goal of the collaboration is to eventually predict operating lifetimes for the materials of interest in terms of integrated proton fluence for the high energy proton accelerator parameter space (temperature, dose rate, duty factor and dynamic stresses), while accepting that such predictions are inherently challenging. The initial RaDIATE collaborators include Fermi National Accelerator Laboratory (FNAL), Brookhaven National Laboratory (BNL), Pacific Northwest National Laboratory (PNNL), Science and Technology Facilities Research Council (STFC)/Rutherford Appleton Laboratory (RAL) and Oxford University. Materials that are not included in the initial program could be addressed in future

* Work supported by Fermi Research Alliance, LLC, under Contract No. DE-AC02-07CH11359 with the U.S. Department of Energy.
† ammikav@fnal.gov

or parallel collaborative activities, with titanium alloys and superconductors being considered.

R&D PROGRAM PLAN

The RaDIATE R&D program was developed to be carried out in three possibly overlapping stages.

1. Exploratory/development stage: An initial six-month study was launched to develop the specific research activities required to achieve the goals of the program. This included an extensive radiation damage literature review, including past target facility experiences, to synergize current radiation damage research and identify possible correlation of radiation damage data of low energy fast neutron and heavy ion to high energy proton accelerator irradiation conditions. The parameter space of future proton accelerator facilities was also established [1], which included the anticipated proton beam parameters, operating temperatures, dose rate and total DPA. The testing capabilities for post-irradiation examinations of each of the collaborators' facilities were determined, such as the Radiochemical Processing Laboratory (RPL) at PNNL, Brookhaven Linear Isotope Production (BLIP) facility at BNL and the Materials for Fusion and Fission Power (MFFP) Laboratory at Oxford University. Lastly, the process to recruit a post-doctoral researcher dedicated to beryllium radiation damage study was initiated. The post-doc will be based in the MFFP group at Oxford University, with potential work at collaborating institutions.

2. Research activities stage: This stage will involve the coordinated research activities performed at the different participating institutions, including the post-doctoral research work of beryllium at Oxford University and the ongoing graphite work at BNL BLIP. The identification, collection and preparation of irradiated materials for different post-irradiation examination techniques will be carried out at pertinent institutions. Finally, after data analysis and computations, models will be developed to help understand material response to high energy proton radiation damage.

3. Results reporting stage: The final stage is dedicated to results reporting. This will comprise of periodical reports on research progress as well as continual presentations of results and findings at various conferences relevant to the particle accelerator community. Results will be published in scholarly journals while future research avenues will also be identified.

GRAPHITE STUDIES AT BNL

As part of ongoing work of the RaDIATE collaboration, graphite and 3D carbon-carbon composite specimens are being tested at BNL BLIP. A target array, containing graphite specimens, was placed upstream of the BLIP facility beam line and irradiated with 200 MeV protons, while operating in tandem with isotope production. A comprehensive study, using MARS15 Monte Carlo code [3], estimated that for the assumed BLIP beam parameters, one year of accumulated damage of the LBNE (Fermilab's proposed Long Baseline Neutrino Experiment) POCO graphite target, operating at 120 GeV/700 kW, would be achieved at BLIP in about 9 weeks of irradiation [4]. The irradiation experiment included various grades of graphite specimens (POCO, IG-430, SGL R7650 and C-2020) as well as 3D C/C composite (orthogonal 3D weave) specimens. The peak DPA damage in the target specimens was estimated by MARS15 simulations to be about 0.095 [5]. After several months of cool down period, the specimens were transported to the hot cell facility, where post-irradiation experiments were carried out. The following subsections provide some initial radiation damage results stemming from thermal and mechanical tests of the irradiated specimens.

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 only mobile at temperatures greater than 1000 K [6]. The peak irradiation temperature of the BLIP specimens was estimated to be around 430 K. Therefore, to enable partial annealing of the damage (portion attributed to interstitial atoms), temperatures higher than the irradiation temperature should be induced. This is under the assumption that interstitial atoms mobile up to that temperature have been placed back in the lattice, or pinned at grain boundaries. To investigate and confirm the annealing and potential damage reversal process, thermal strain measurements of the specimens were made using a dilatometer. Figure 1 shows thermal strain measure-

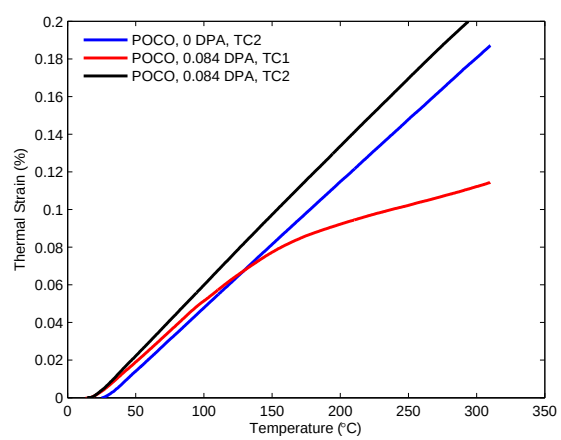

Figure 1: Thermal expansion measurements of unirradiated and irradiated POCO graphite.

ments of POCO graphite, comparing an unirradiated control specimen to an irradiated specimen at 0.084 DPA, with the latter performed over two thermal cycles. During the first thermal cycle to 310 °C, the irradiated specimen deviates significantly from its initial linear behavior to show a decrease in thermal expansion. However, in the subsequent cycle, the specimen recovers most of its unirradiated expansion characteristics with its coefficient of thermal expansion about 10% higher than the unirradiated one. This trend is qualitatively similar across all the graphite grades [5] and confirms the interstitial atom annealing and damage reversal process. No correlation was observed in the change of coefficient of thermal expansion as a function of DPA, during the thermal cycles. This can be explained due to the fact that higher dose rate specimens also experienced higher irradiation temperatures and therefore, exhibited less damage due to the annealing effect. The irradiation temperature, which is estimated to be about 150 °C for this particular POCO specimen, can be inferred from the plot (Figure 1), the point at which the irradiated curve diverted from the unirradiated curve.

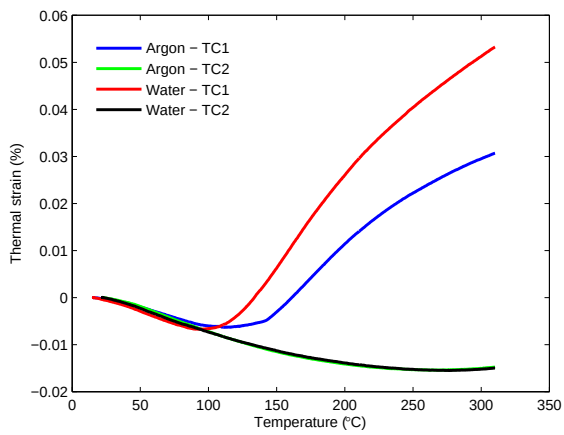

Figure 2: Thermal expansion measurements of irradiated 3D C/C composite specimens.

Similar damage reversal phenomena, shown in Figure 2 were observed with the 3D C/C composites, which were exposed to both water and argon environments during irradiation. The negative thermal expansion coefficient characteristic of unirradiated 3D C/C composite was regained after the irradiated specimen underwent a second thermal cycle. Results from both Figures 1 and 2 confirm that significant property changes can occur at low DPA levels in graphite, while damage reversal is possible at annealing temperatures higher than the irradiation temperature. This can provide optimal operating temperatures for future accelerator targets or windows.

Mechanical testing

To quantify the effect of radiation damage on the tensile strength and elastic modulus of the specimens, tensile tests were carried out. Figure 3 shows an increase in both the elastic modulus and ultimate tensile strength of irradiated IG-430 specimens when compared to the control specimen. However, no distinct dependence on DPA is observed, primarily due to the interrelation of DPA and irradiation temperature. As explained in the previous section, higher DPA specimens are irradiated at higher temperatures and therefore show less damage. Figure 4 confirms the damage reversal process of an irradiated POCO specimen after annealing at 350 °C.

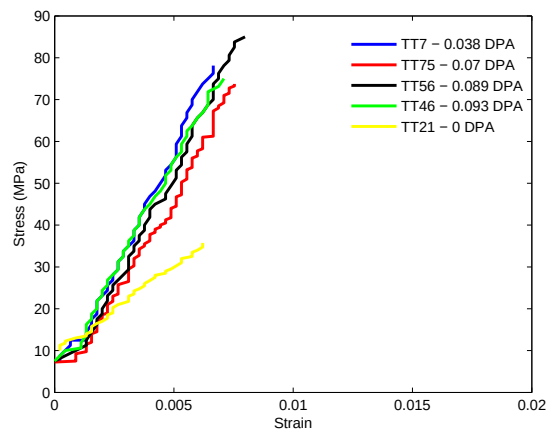

Figure 3: Stress strain curves for irradiated IG-430 at varying DPA levels.

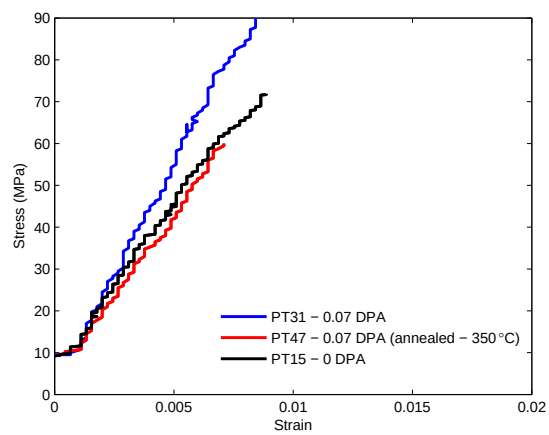

Figure 4: Damage reversal annealing of irradiated POCO.

Tensile tests of 3D C/C composite specimens did not provide reliable experimental data due to constant slipping of the grips and failure at the specimen head. As a result, the specimens were recently tested in a three-point bending test to obtain consistent failure in the middle of the gage. Figure 5 shows flexural strength data of a control specimen compared to irradiated specimens (0.05 DPA) in both argon and water environments. Clearly, the irradiation environment plays an important role in radiation damage, as

indicated by the contrasting strength and stiffness change between the argon and water specimens relative to the control specimen. The irradiated specimen in argon shows a significant increase in both flexural modulus and strength. Further flexural tests of 3D C/C will be carried out to study the damage annealing effects and allow for DPA comparisons.

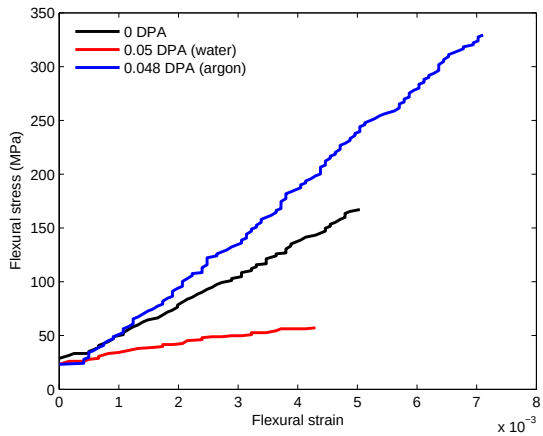

Figure 5: Flexural strength of 3D C/C composite specimens irradiated in both water and argon.

Ultrasonic tests

Ultrasonic tests were also performed to quantify the change in the elastic modulus of the specimens due to irradiation and annealing [5]. The longitudinal wave velocity was first estimated by measuring the travel time and propagation length of the waves through the specimens, after which the elastic modulus was evaluated using Equation 1, where V_L is the longitudinal velocity, ρ the density, and ν the Poisson's ratio.

$$E = \frac{{V_L}^2 \rho (1+\nu)(1-2\nu)}{(1-\nu)} \quad (1)$$

Figure 6 shows the elastic modulus of irradiated IG-430 specimens as a function of DPA, evaluated using the ultrasonic test method. The irradiated specimens show a significant increase in the elastic modulus, about a factor of two, when compared to the control specimen. It should be noted that the higher DPA specimen (0.095 DPA) reveals a smaller increase in elastic modulus than the 0.054 DPA one, which is consistent with the previously stated notion that higher DPA specimens are irradiated at higher temperatures and therefore incur less damage due to the annealing process. After annealing of the irradiated specimens to 310 °C, the elastic modulus decreases to about 12 GPa, which is around 70 % higher than that of the control specimen, regardless of DPA levels. This trend is qualitatively similar to results from the tensile tests.

Table 2 compares the percentage change of elastic modulus with respect to the control specimen, of irradiated

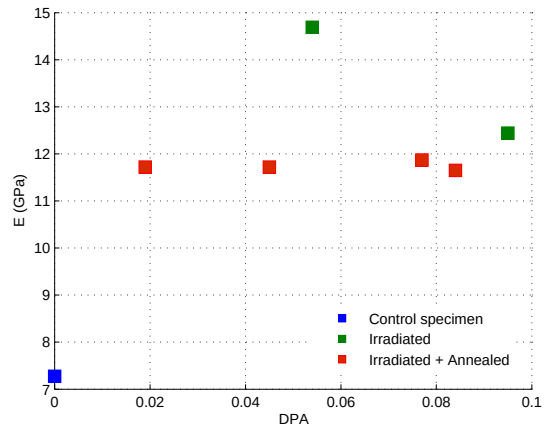

Figure 6: Elastic modulus of IG-430 graphite specimens (ultrasonic method).

and annealed IG-430 specimens obtained via the ultrasonic and tensile test methods. Clearly, the tensile tests yielded higher percentage changes than the ultrasonic tests, when ideally both methods should provide similar results. One possibility for the discrepancy could be the assumed constant density and Poisson's ratio values in evaluating the elastic modulus with Equation 1. Other possible causes are being investigated further and more specimens will be tested to provide better statistics on the current results.

Table 2: Comparison of percentage change in elastic modulus between tensile and ultrasonic tests (IG-430).

DPA	Ultrasonic	Tensile
$\sim$ 0.095 (unannealed)	72%	126%
$\sim$ 0.070 (annealed)	64%	89%
$\sim$ 0.045 (annealed)	61%	87%

Ultrasonic tests were unsuccessful with 3D C/C composites because the array/layer of fibers scattered and diffused the ultrasonic pulses, which had a wavelength smaller than the diameter of each fiber. Further mechanical and thermal tests of the proton-irradiated specimens are ongoing at BNL and will yield more results to allow for comparisons of the various graphite grades. This radiation damage work ties in well with the main objectives of the RaDIATE collaboration and will serve as a platform for radiation damage studies of other materials.

IRRADIATED MATERIALS AND TESTING

Part of the initial activities of the collaboration was to identify already irradiated components at various institutions, which may be suitable for post-irradiation examination. These components include previously failed or retired

targets/windows, made of graphite, beryllium or tungsten. An extensive list of the components as well as the estimated irradiation parameters were drawn up [1] and efforts are currently under way to recover the components that are of most interest.

Furthermore, to avoid long and costly high energy proton irradiation runs (eg. BNL BLIP), low energy ion irradiation tests are also being considered. Ion irradiation provides good control on irradiation temperature, dose and dose rate, with high doses easily achievable in a short time. There is also the advantage of low specimen activation, which facilitates post-irradiation examination. However, one main issue is the shallow depth of penetration (a few micrometers), which induces radiation damage only in the proximity of the specimen's surface. But with recent advancements in micro-mechanic testing, material properties from irradiated miniature specimens may be extracted. Figure 7 shows a micro-mechanics test cantilever produced at Oxford, which can be mechanically tested using nano-indenters. Eventually, the micro-mechanics technique will require validation for each material in order to confirm whether micro properties are representative of bulk material properties.

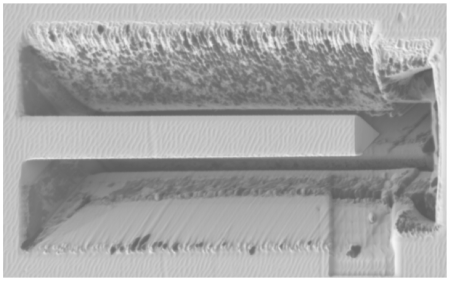

Figure 7: Micro-cantilever (17μm $\times$ 3μm) machined in tungsten (image courtesy of D. Armstrong, Oxford).

Other important issues to address with ion irradiation tests are the absence of transmutation effects and ion implantation into the specimens. A method to generate controlled gas production (He and H) during irradiation is necessary so as to replicate high energy proton irradiation conditions (Table 1). And finally, validations must also be performed to confirm that ion implantation does not induce composition changes and interstitials into the test specimen.

SUMMARY

Stage 1 of the RaDIATE program plan is complete and provides a clear vision for coordinated radiation damage research among the participating institutions. Stage 2 will follow soon, with results for graphite, beryllium, and tungsten expected in 2015-2016. As briefly presented in earlier sections, important findings have already been obtained from graphite studies at BNL BLIP which will benefit both the nuclear fission/fusion and particle accelerator communities. Lastly, the RaDIATE program is open to other collaborators and is also considering other materials of interest for radiation damage studies, such as titanium alloys and superconductors.

REFERENCES

[1] RaDIATE website, www-radiate.fnal.gov

[2] P. Hurh, K. Ammigan, B. Hartsell, R. Tschirhart, 'Targetry challenges at megawatt proton accelerator facilities', Proc. of the 4th International Particle Accelerator Conference, Shanghai, China (2013).

[3] N.V. Mokhov, 'The MARS code system user's guide', Fermilab-FN-628 (1995).

[4] N.V. Mokhov et al., 'Simulation and verification of DPA in materials', Proc. of the Workshop on Applications of High Intensity Proton Accelerators, Fermilab, World Scientific (2010) 128.

[5] N. Simos et al., 'Long Baseline Neutrino Experiment (LBNE) target material radiation damage from energetic protons of the Brookhaven Linear Isotope Production (BLIP) facility', Final Report, Fermilab (2013).

[6] B.T. Kelly, 'The behavior of graphite under neutron irradiation', J. Vac. Sci. Technol. A4 (1986) 1171.