STRENGTH AND DIMENSIONAL CHANGE OF NEUTRON-IRRADIATED HIGH-QUALITY 3D CARBON FIBER COMPOSITE - L. L. Snead, T. D. Burchell, and A. L. Qualls, Oak Ridge National Laboratory*

OBJECTIVE

The objective of this work is to gain understanding of the dimensional and strength change in high quality carbon fiber composite under high temperature neutron irradiation.

SUMMARY

The effects of neutron irradiation to ~7 dpa at 500 and 800°C on a high quality, three-dimensional balanced weave composite (FMI-222) is presented. Strength and dimensional stability for this system is compared to earlier work on this material, at lower dose, and contrasted with that of a well studied isotropic graphite (POCO AXF-5Q) irradiated at identical conditions. For both irradiation temperatures the composite strength in bending is substantially increased. Interestingly, while both irradiation temperatures cause contraction along the bend bar axis, the amount of contraction is greater for the higher temperature irradiation. Moreover, for the 500°C specimens nearly isotropic contraction leads to a corresponding decrease in volume, though an apparent large increase in volume occurs for the 800°C irradiated composite due to very anisotropic dimensional change. As the FMI-222 is a balanced-weave, isotropic composite, non-isotropic swelling behavior was unexpected, and is explained in terms of the fiber dimensional stability model previously presented, and the composite nature of this material.

PROGRESS AND STATUS

<u>Introduction</u>

The effect of neutron irradiation on the strength and dimensional stability of graphites has been well studied and are strongly related. [1-4] For isotropic graphite, irradiation causes an initial densification with increased strength and Young's modulus. Densification is attributed to strain relief and closing of internal porosity by the migration of irradiation-induced carbon interstitials. The irradiation-induced increase in strength for graphite can be quite substantial. For example, nuclear graphites such as Graphnol N3N [5], Grade TSX, H451, and others[6] exhibit a peak increase in brittle-ring strength of approximately 100%. The effect of irradiation on composites has received less attention, though similar trends with graphite have been observed,[7-9]. As example, Burchell demonstrated a 64% increase in brittle-ring strength following an intermediate irradiation dose at 600°C for the high quality, balanced weave FMI-222 composite.

In polycrystalline graphites, neutron irradiation initially causes volumetric shrinkage. On the crystallite scale, the irradiation behavior is quite anisotropic with vacancies forming voids, or microcracks at the crystallite boundaries, and new basal planes by interstitial agglomeration. This causes shrinkage in the <a> direction and strain perpendicular to the basal planes (<c> direction.) Initially, the <c> axis strain is absorbed by intrinsic misalignment of the basal planes and porosity. However, at some (irradiation temperature dependent) dose, the ability to accommodate the <c>

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axis strain saturates. The newly forming basal planes then cause <c> swelling and the "turnaround" from macroscopic densification to swelling occurs, leading to multiplication of the strain-induced cracks and severe degradation in the material strength. The point at which the swelling returns to zero is typically taken as the useful lifetime.

It has been speculated that carbon fiber composites, by virtue of their ability to balance the anistropic swelling on a macroscopic scale, may possess superior mechanical and dimensional

properties at high doses and temperatures as compared to graphites. The purpose of this work is to study the dimensional and mechanical performance of a high-quality balanced-weave composite at doses sufficient highly to cause anisotropic swelling in the fiber.

Experimental and Results

Manufacturer supplied thermophysical properties for the two materials of this study are given in Table 1. Bend bars were machined in the as-received condition into 2.3 x 6 x 30 mm and baked at 200°C in air prior to loading into graphite holders. Due to the effect of the relatively large unit cell volume for the composite materials, and the volume constraints associated with irradiation capsules, it was decided not to use the brittle ring geometry typical of previous graphite studies and the previous work with FMI-222. The statistical variability was found to be less with bend bars. The 14J irradiation capsule was irradiated for 8 cycles in the removable beryllium (RB) position of the High Flux Isotope Reactor (HFIR.) The capsule included thermocouples and active sweep gas control for 500 and 800°C temperature regulated zones. Bend testing was carried out at room temperature with cross-head displacement of 0.0085 mm/s. Load and support spans were 6.45 and 19.05 mm, respectively.

Table 1. Thermophysical properties of materials studied

Property	Poco AXF-5Q	FMI-222
Manufacturer	Poco	Fiber Materials Inc.
Architecture	Isotropic Graphite	Balanced 3d weave
Precursor	Pitch based	P-120 Pitch fibers, Pitch Matrix
Grain size/Unit Cell Size (μm)	5	~900
Thermal Conductity (W/m-K)	95	200
Apparent Density (g/cc)	1.78	1.96
Flexure Strength MPa	110	175

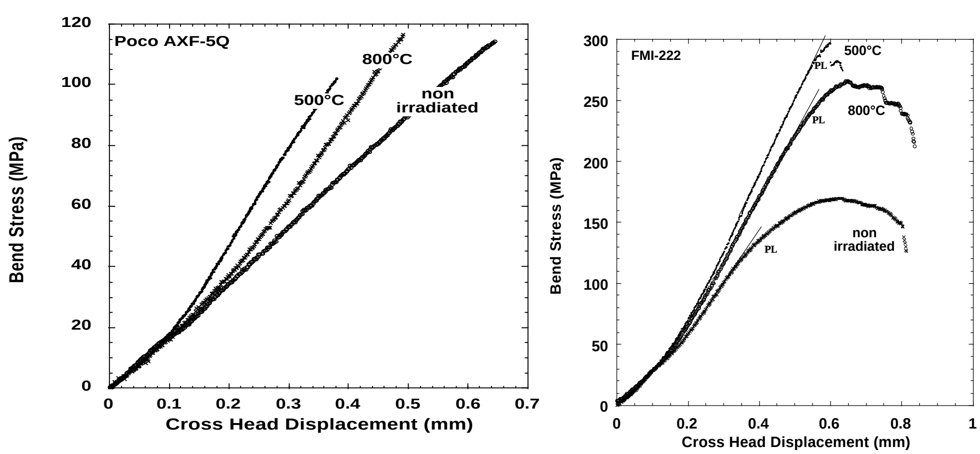
Table 2 gives the dimensional and flexural strength results as well as the fast neutron fluences. While it is commonly understood that graphite strength is best represented using Weibull statistics, the six samples available for the irradiated condition are less than the 15-30 considered adequate for such analysis. For this reason the normal statistical mean ± 1 standard deviation is given. It is seen that there is a slight decrease in flexural strength and density (implied from the length change) for Poco irradiated at 500°C. However, this decrease is within the standard

Table 2. Physical property changes due to irradiation

	Non-Irradiated	500°C 6 x 10^{25} n/m^2 (E>0.1 MeV)	800°C 7.7 x 10^{25}n/m^2 (E>0.1 MeV)
Poco AXF-5Q (#tests)	6	6	5
Ultimate Bend Strength, MPa (% change)	113±9	107±7 (-5%)	98±11 (-13%)
Length Change (%)	-	0.06±0.09	1.11±0.17
FMI-222	6	3	3
Proportional Limit, MPa (% change)	135±16	266±23 (+97%)	205±14 (+52%)
Ultimate Bend Strength, MPa (% change)	176±20	286±25 (+63%)	241±22 (+37%)
Macroscopic Length Change (%)	-	- 1.5	-3.6
Macroscopic Width Change (%)	-	- 1.4	1.4
Macroscopic Thickness Change (%)	-	- 1.4	5.9
Apparent Volume Change (%)	-	- 4.3	3.0

deviation. For the 800°C irradiation, a statistically significant decrease in strength (-13%) and density (-3.3%) occurs in Poco. In table 2, the flexural data for the FMI material is presented both with a proportional limit and ultimate bend strength. As the FMI material is a composite architecture, the flexural curve exhibits a departure from linearity, exhibiting “pseudo-ductility,” as crack propagation is mitigated by the fiber tows. This difference in flexural behavior, and the changes in stiffness and strength for the Poco and FMI materials, is illustrated in Figure 1. The point where the flexure curve departs from linearity is defined as the proportional limit.

Figure 1. Flexural behavior of: a) Poco AXF-5Q and b) FMI-222. (PL is proportional limit)



The as-irradiated behavior of the FMI material contrasts with the Poco. At both irradiation temperatures a large increase proportional limit, flexural strength, and length change occurs in FMI. The increase in the proportional limit for FMI at 500°C (97%) exceeds that of the 800°C (52%), which also holds true for the ultimate bend strength. However, the length change at 500°C is less than half the 3.6% densification observed for the FMI material irradiated at 800°C.

Discussion

As mentioned in the introduction, increased strength with neutron irradiation prior to "turnaround" is well known for graphites and is attributable to: (1) pinning of basal plane dislocations by irradiation-induced defects in the graphite crystallites, and (2) the reduction of internal porosity due to irradiation-induced volume shrinkage (densification.) The transition from strengthening to ultimate disintegration and total loss of load carrying capability is driven by the strains induced by anisotropic dimensional change. The commonly accepted irradiation-induced dimensional change model is for initial densification of isotropic graphite followed by "turnaround" to swelling. This turnaround occurs because the because of pore generation resulting from the mismatch of irradiation-induced crystal strains. Obviously, the removal of carbon atoms from existing basal planes to form new planes leaves behind vacancies causing shrinkage in the <a> direction. Typically, the amount of densification is less, and the point of "turnaround" to swelling behavior occurs at a lower dose, as the irradiation temperature is increased.[4] The contrasting dimensional change behavior for the FMI-222 composite irradiated at 500 and 800°C can be explained using the previously proposed "core-sheath" microstructural model [8] in which the graphite planes are oriented circumferentially on the fiber, and radially in the fiber core. Following this model, there will be continuous axial shrinkage under irradiation with the fiber diameter moving from shrinkage to swelling behavior.

In the previous work on the FMI-222 composite [8], an apparently linear densification of 1.3% occurred per 1 x 10^{25} n/m^2 (E>0.1 MeV) neutron dose. The maximum fluence of that study was ~ 4.7 x 10^{25} n/m^2 (E>0.1 MeV.) A similar densification occurred for a PAN fiber composite, FMI-223, possessing identical matrix and processing as the pitch based fiber composite FMI-222 of this study. The transition to "turnaround" swelling behavior was observed for the FMI-223 material, though not observed in FMI-222. This difference was attributed to the superior radiation stability of the pitch-based fibers. The neutron fluence in this study (7.7 x 10^{25} n/m^2) was chosen to achieve turnaround behavior. Based on the dimensional change results (Table 2,) turnaround was achieved. However, the current results are not easily comparable with the earlier work because of the new finding of non-isotropic swelling for the bend bar. Using the shrinkage in length from table 2, apparent densification of 0.6 and 1.4 % per 1 x 10^{25} n/m^2 occurs at the 500 and 800°C irradiation temperatures, respectively. This qualitatively agrees with the 1.3% per 1 x 10^{25} n/m^2 value of the previous 600°C irradiation. The transition to swelling behavior is evident, however, when taking into account the swelling in the width and thickness of the bend specimens. This gives ~ 4% swelling at 500°C and ~ 5-10% at 800°C.

This anisotropic macroscopic dimensional changes can be explained using the core-sheath model for fiber dimensional changes. Assuming that the macroscopic composite behavior is dominated by the fiber changes, one would expect the fibers to initially shrink in the diametral and axial direction, then begin diametral swelling with continued axial shrinkage. For higher temperatures, turnaround occurs more rapidly due to thermal closure of porosity, yielding greater dimensional changes for equivalent neutron dose. Even though this composite is a balanced weave, and would be expected to have isotropic dimensional change, the fact that the length direction of the bend bar has continuous fiber tows parallel to its axis, the length change is dominated by the behavior of the fibers. However, the macroscopic width and thickness dimensions of the bend bar are dominated by the radial swelling of fiber bundles.

This anisotropic behavior is evident by inspection of SEM micrographs of the top surface of the bend bars (Figure 2.) By comparing the 500 and 800°C images it is clear that at 800°C the fiber bundles have undergone significantly higher shrinkage causing gaps as the bundles have shrunk away from the surface. For the 500°C irradiation, the fiber tows and pitch matrix appear to remain coherent with the tow shrinkage equal to the macroscopic dimensional changes of the composite. Tables 3 and 4 give the dimensional analysis of the fiber bundles within the composite (eg measurement from Figure 2 micrographs.) It is clearly seen from Table 3 that the fiber tow lengths following irradiation decrease, with the least change in the longer, and thereby more constrained, dimensions. Table 4 gives the corresponding increase in width of the fiber bundles caused by the radial swelling of the individual fibers.

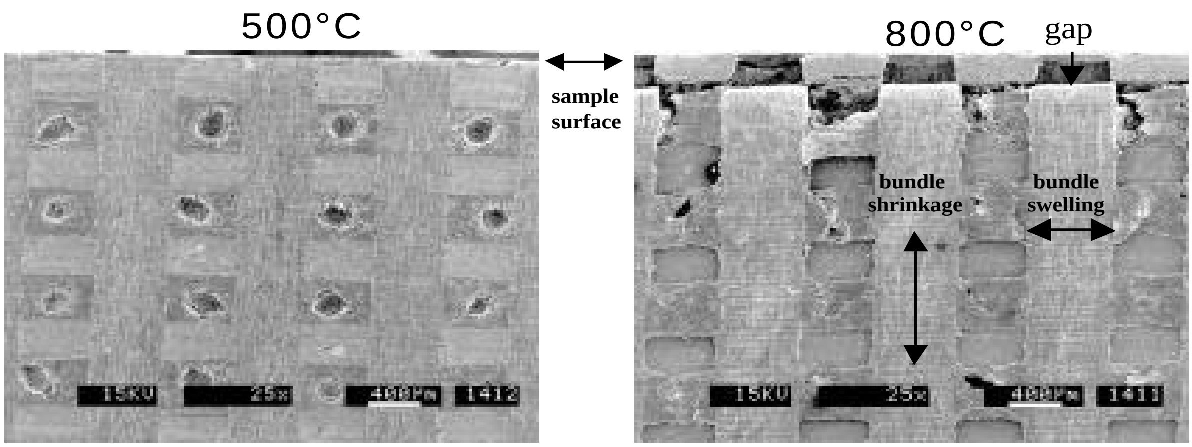


Figure 2. SEM image of surface of 500°C and 800°C irradiated FMI-222 composition.

Table 3. Dimensional change occurring in the fiber bundle lengths upon irradiation.

	Non-Irradiated Dimension (mm)	Change in Length (%) 500°C	800°C
Bundle Length Parallel to Bar Length Axis	30	-1.5	-4.25
Bundle Length Parallel to Bar Width Axis	6	-1.4	-5.5
Bundle Length Parallel to Bar Thickness Axis	2.3	-1.4	-7.2

Table 4. Dimension change occurring in the fiber bundle diameter upon irradiation.

	Bundle Width (μm)	Change in Width (%)
Non-Irradiated	320	-
500°C, Irradiated	331	3.4
800°C, Irradiated	363	13.4

It is important to note that, while this balanced weave, isotropic composite has undergone anisotropic dimensional changes, this behavior is being affected by the geometry, and associated constraints, of the sample. It is likely that larger samples would behave in a manner consistent with the fiber-axis-dominated shrinkage seen along the axis of the bend bars. Referring to Table 2, the positive volume changes given are dominated by the bend bar width and thickness swelling, where it is speculated that were the sample cubic, and large enough for many unit cells, the volume change would be better represented by the cube of the length change. However, as the fluence is increased, the strains associated with the anisotropic swelling must eventually lead to destruction of the composite as the fiber diameter becomes increasingly large and resultant strains cause internal fractures.

Previous brittle-ring strength measurements made on FMI-222 irradiated to a dose of ~ 2.2 x 10^{25}n/m^2 (E>0.1 MeV) at 600°C exhibited a strengthening of about 64% and a corresponding densification change of ~ 3%. In this study, where the fibers are in a regime of gross anisotropic dimensional change (especially at 800°C,) the composite has maintained the radiation enhanced strength.

Specifically, in bending, the fracture strength is 63% higher at 500°C, and 37% higher at 800°C irradiation. This behavior is in contrast to the Poco materials which at identical irradiation and testing conditions underwent a decrease in strength and had entered the isotropic swelling regime.

CONCLUSIONS

This study has shown that, for a very high quality, balanced weave carbon fiber composite, radiation enhanced fracture strength is retained in the anisotropic swelling regime generally associated with severe loss in strength. This has been demonstrated by comparison of the standard isotropic graphite Poco AXF-5Q and the balanced weave pitch-based fiber composite FMI-222. At the highest dose and temperature, 7.7 x 10^{25}n/m^2 and 800°C, the graphite material was seen to undergo swelling with an associated 13% decrease in strength, while the composite material exhibited a 37% higher strength.

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