Effects of Fast Neutron Irradiation on Carbon Fibers

R.J. Price, G.R. Hopkins, and G.B. Engle
GA Technologies Inc., Box. 85608, San Diego, CA 92138

Introduction

Carbon-carbon composites have excellent high temperature strength, good toughness, and low neutron absorption cross section. This combination of properties would make them suitable for use in nuclear reactor cores if they were resistant to radiation damage. However, previous work showed that PAN- and rayon-based carbon fibers shrink excessively at high neutron fluences.[1,2] Crystalline graphite tends to be more radiation stable than poorly crystalline carbon.[3] Thus, more graphitic fibers, such as annealed pitch-based fibers or vapor deposited filaments, offer the possibility of making carbon-carbon composites that are stable enough under irradiation to be useful for reactor core components.

Experimental

A series of commercial PAN-based and mesophase pitch-based carbon fibers were annealed at 2650°C and 3100°C to increase their crystalline order. L_c values measured from the width of the (002) x-ray diffraction peak are shown in Table 1. Vapor deposited carbon filaments, silicon carbide fibers, and highly annealed pyrolytic graphite (Union Carbide HOPG) were included in the experiments. Unannealed and annealed specimens were irradiated in four "piggy back" crucibles in two graphite irradiation capsules in the HFIR reactor at Oak Ridge National Laboratory. The fast neutron fluence ranged from 4.2 to 5.0 x 10^{21} n/cm^2 (E>50 keV) and the irradiation temperature, determined from silicon carbide temperature monitors, ranged from 410 to 620°C. Length changes were measured with a traveling microscope, density changes were measured by suspension in bromoform-methanol mixtures, and changes in Young's modulus and tensile strength were measured on single fibers cemented to paper tabs, using a micro-tensile testing machine.[4]

Results and Discussion

Changes in length and diameter (calculated from the density changes) for specimens irradiated to 5.0 x 10^{21} n/cm^2 at 620°C are shown in Table 1. The fibers all showed longitudinal shrinkage and a smaller diametral expansion. The dimensional changes were very dependent on the fiber type and annealing treatment, with the least graphitic fibers (as received PAN-fiber type 1) shrinking longitudinally more than ten times faster than the most graphitic fibers (pitch-based fiber type 2 annealed at 3100°C). The diametral expansion of the PAN-based fibers was proportionately less than that of the more graphitic pitch-based fibers because the former increased in density during irradiation whereas the latter decreased slightly in density. Based on more limited data, the irradiation stability of both vapor deposited filaments and SiC fibers appeared very good. The longitudinal shrinkage of the fibers is a measure of the a-axis shrinkage of the constituent crystallites, which is lower in more crystalline materials because more interstitials and vacancies recombine before they reach nucleation sites.[3]

The dependence of the longitudinal shrinkage on irradiation temperature is shown in Fig. 1 for three materials: an as-received PAN fiber, a pitch-based fiber annealed at 3100°C, and highly oriented pyrolytic graphite ("HOPG"). The data are normalized to a fluence of 5 x 10^{21} n/cm^2. The two more graphitic materials show a negative temperature dependence, while the less graphitic PAN fibers show a positive temperature dependence. This behavior is consistent with earlier experiments on pyrocarbons and nuclear graphite and can be explained by the hypothesis that the spacing between nucleation sites (such as crystallite boundaries) is less than the mean free path of irradiation-induced defects in poorly graphitized carbons but greater in well graphitized carbons.[5]

Tensile tests were made on the more dimensionally stable fibers after irradiation. Table 1 includes the means of 20 tests on annealed pitch-based fibers and SiC fibers irradiated to 4.2 x 10^{21} n/cm^2 at 440°C. The mean strength of all tested fiber groups increased by 15-45% after irradiation, while Young's modulus of the pitch-based fibers decreased slightly. Young's modulus of the SiC fibers increased by 50%. The strength increases are about the same magnitude as those reported for PAN-based fibers irradiated to much lower fluences.[6]

Conclusions

The results show that annealed pitch-based carbon fibers and SiC fibers have good dimensional stability under fast neutron irradiation and their tensile strength is increased. These fibers have promise as reinforcements in a nuclear reactor environment.

Table 1. Irradiation-induced changes in dimensions and tensile properties of carbon and SiC fibers.

Material	Annealing Temperature (°C)	L_c (Å)	Condition	Length Change (%)	Diameter Change (%)	Mean Tensile Strength (10^3 psi)	Mean Young's Modulus (10^6 psi)
PAN fiber 1	--	17	Unirradiated	--	--	443	32
		--	Irradiated*	-33.8	+17.5	---	--
	2650	61	Unirradiated	--	--	258	45
		--	Irradiated*	- 8.4	+ 3.0	---	--
	3100	77	Unirradiated	--	--	236	46
		--	Irradiated*	- 6.7	+ 0.7	---	--
PAN fiber 2	--	54	Unirradiated	--	--	308	55
		--	Irradiated*	-10.7	+ 3.6	---	--
	2650	67	Unirradiated	--	--	273	48
		--	Irradiated*	- 4.6	+ 1.6	---	--
	3100	101	Unirradiated	--	--	217	46
		---	Irradiated*	- 3.8	+ 0.1	---	--
Pitch fiber 1	--	112	Unirradiated	--	--	286	54
		---	Irradiated*	-13.2	+ 7.3	---	--
	2650	150	Unirradiated	--	--	227	63
		---	Irradiated*	- 5.8	+ 4.2	260	45
	3100	161	Unirradiated	--	--	177	61
		---	Irradiated*	- 4.5	+ 3.4	259	45
Pitch fiber 2	--	168	Unirradiated	--	--	315	135
		---	Irradiated*	- 4.1	+ 3.5	---	--
	2650	193	Unirradiated	--	--	401	111
		---	Irradiated*	- 2.6	+ 1.9	---	--
	3100	336	Unirradiated	--	--	285	85
		---	Irradiated*	- 1.1	+ 1.5	348	82
Vapor deposited filament	3100	1660	Irradiated*	- 0.2	--	---	--
SiC fiber	--	----	Unirradiated	--	--	491	31
		----	Irradiated*	- 1.8	- 0.3	661	48
HOPG	--	----	Irradiated*	- 0.5	+ 1.9	---	--

*Dimensional change data for 5.0×10^{21} n/cm^2 (E > 50 keV) at 620°C. Tensile data for 4.2×10^{21} n/cm^2 (E > 50 keV) at 440°C.

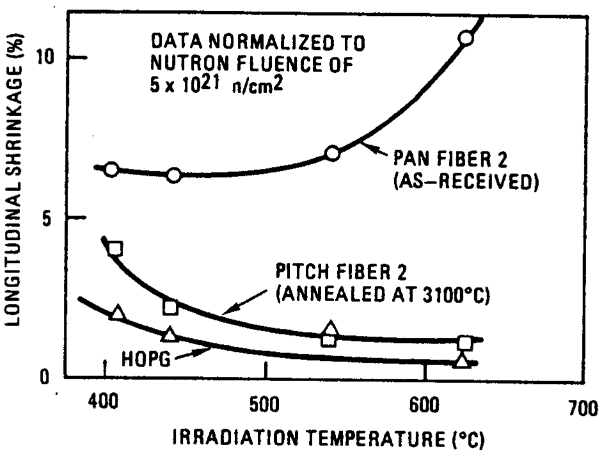

Figure 1. Dependence of longitudinal shrinkage of carbon fibers on irradiation temperature.

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