CHARACTERIZATION OF A 2D-SIC$_F$/SIC COMPOSITE MADE BY ICVI WITH HI-NICALON™ TYPE S FABRIC – G. E. Youngblood and R. H. Jones (Pacific Northwest National Laboratory)*

OBJECTIVE

The objective of this task is to examine SiC fibers and SiC/SiC composites fabricated by various processing methods designed to improve the composite properties. Specifically, it is desired to optimize the thermal conductivity and the mechanical behavior of these composites to meet expected requirements for advanced fusion energy systems.

SUMMARY

In this report, the mechanical and thermal properties of a 2D-SiC$_f$/SiC composite made by the CVI-process with the Hi-Nicalon™ type S fabric are assessed in detail with respect to meeting fusion design requirements. Minimum strength and stiffness structural requirements likely can be met by CVI-processed SiC$_f$/SiC composites when made with advanced SiC fibers. Unfortunately, it appears unlikely that the minimum thermal conduction goals can be met for CVI-processed material. Even for an optimized 2D SiC$_f$/SiC system, the margin of improvement required is just too large for only minor improvements potentially possible through CVI-processing upgrades or other structural or architectural methods.

PROGRESS AND STATUS

Introduction

SiC/SiC composite has been proposed as a structural material for the first wall and blanket in several conceptual designs of a D-T fusion power reactor. The reactor concepts: TAURO in the European Union and the ARIES-AT in the United States utilize Pb-17Li self-cooled blankets, while DREAM in Japan utilizes 10 MPa helium cooling. In these designs, modules built from SiC$_f$/SiC composite would be from 0.5 to several meters high, and would operate under high-energy (14.1 MeV) neutron irradiation in a relatively high temperature range (600°C-1000°C). Important mechanical and thermal design criteria at this time are: elastic modulus > 200 GPa, tensile strength > 200 MPa, and through-thickness thermal conductivity (K_{eff}) at 1000°C > 15 W/mK [1]. With the use of crystalline, near-stoichiometric SiC fibers (e.g., Hi-Nicalon™ type S or Tyranno™ SA fibers), SiC$_f$/SiC composites made by the CVI-process exhibited little degradation of strength and stiffness after a 7.7 dpa irradiation dose at 800°C [2]. Thus, it appears likely that the fusion mechanical design criteria can be met by using SiC$_f$/SiC composites made with advanced SiC fibers. However, for a relatively low dose (typically less than a dpa in SiC) a steady-state concentration of point defects (vacancies and interstituals) will be induced in the SiC lattice during neutron irradiation. Such point defects efficiently scatter phonons, and depending upon the irradiation temperature will degrade the thermal conductivity by 50% or more [3]. To compensate for the inevitable thermal conductivity degradation and still attain the fusion energy K_{eff}-goal during operation, it is estimated that unirradiated SiC/SiC composite should have starting K_{eff}-values of at least 43 and 27 W/mK at 600 and 1000°C, respectively [4]. In this report, the mechanical and thermal properties of a 2D-SiC$_f$/SiC composite made by the CVI-process with the Hi-Nicalon™ type S fabric are assessed in detail with respect to meeting the listed fusion design criteria.

Experimental Procedure

Two SiC$_f$/SiC composite plates (15 x 23 cm^2) were purchased from GE Power Systems Composites

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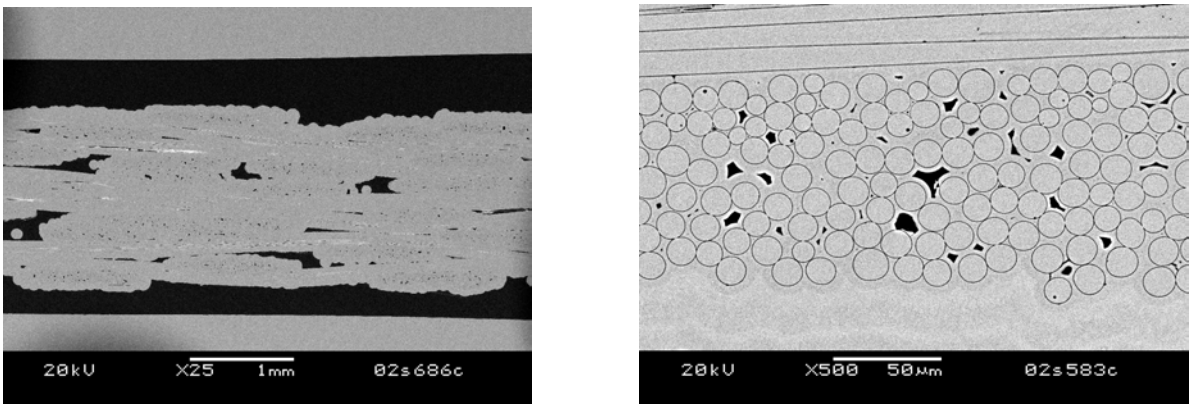
(formerly DuPont Lanxide). One plate was made with eight plies, the other with six plies of 5HS-weave Hi-Nicalon™ type S fabric (18 epi, 500 filaments/yarn, lot number SCS0102, manufactured by Nippon Carbon Co in December, 2001). The properties given by Nippon for the type S fiber were: density 3.0 g/cc, tensile strength 2.8 GPa, tensile modulus 380 GPa, and oxygen content 0.7 wt%. A pyrocarbon (PyC) fiber coating (nominally 150 nm thick) was applied by CVD to each fabric lay-up prior to SiC matrix infiltration by the isothermal CVI-process (ICVI). The nominal bulk density of the plates was 2.69 g/cc after ICVI-SiC infiltration and the nominal overall fiber content was 40%.

Laser flash thermal diffusivity measurements were performed on several representative disc samples (1.79 mm x 9.00 mm dia.) cut from the as-received six-ply plate. Individual diffusivity sample bulk density values ranged from 2.60 to 2.74 g/cc. The samples were mounted four at a time in a sliding sample tray so that diffusivity measurements could be made in air on each sample for 50°C temperature steps from RT to about 400°C. Laser pulses and infrared temperature detector readings were made on the composite sample as-received surfaces. Further experimental details were presented previously [5]. Using previously measured fiber thermal conductivity values (K_f) and measured $K_{eff}(T)$-values, the constituent matrix and fiber coating thermal conductivity values were estimated using the H2L thermal conductivity model developed at PNNL [6].

Four-point bend flexure tests were performed on six ICVI-SiC/SiC composite bars cut from the six-ply plate of the Hi-Nicalon™ type S material (nominal bar dimensions 5.95 x 30.0 x 1.85 mm^3). The outer tensile and compressive bar surfaces were not machined. Bar dimensions were measured at several locations along each bar with a vernier caliper. Side views of the bars by SEM indicated that each surface profile had a roughness variation of about ±0.16 mm. Since the thickness was measured peak-to-peak with the caliper, the average bar thickness was obtained by subtracting an average 0.16-mm profile correction factor from the actual measured thickness values. Bar samples were mounted in a fully articulated inconel fixture in an Instron load frame. The four contact roller pins were made from sintered Hexalloy™ SA SiC and had an outer/inner pin spacing of 20/10 mm. A load deflection rate of 8.5 x 10^{-4} mm/s was used. Three bars each were tested at ambient (RT) and three at 800°C. The bend tests at 800°C were performed in flowing, dry argon. Linear elastic beam theory (LEBT) was used to calculate the maximum stress and strain expected at the outer tensile surface of the bar samples from measured bar midpoint displacement and load data, even though LEBT generally is not valid in the non-linear region of the stress-strain curves for fiber reinforced ceramic composites. The stress-strain data were corrected for machine compliance by testing two CVD-SiC bars of the same dimensions at RT and assuming that the elastic modulus for the monolithic CVD material was 460 GPa. The flexural modulus of the composite samples was then estimated from the corrected slope of the linear portion of the “pseudo-plastic” stress-strain curves. For comparison, similar 4-pt bend tests were performed on several bar samples of SiC_f/SiC composite, also made by ICVI (by DuPont Lanxide) with plain weave (PW) Hi-Nicalon™ fabric and a thin (0.110 μm) PyC fiber coating. Polished cross-sections of representative samples and several fracture surfaces of bars broken in the 4-pt bend tests were examined by SEM.

Results

In Figures 1(a-b), typical low and high magnification SEM micrographs of polished cross-sections of the 2D Hi-Nicalon™ type S composite are presented. From several views of this type, the following structural parameters were determined and used in the analysis of the thermal conductivity with the H2L model. The average fiber coating thickness was 0.188±0.036 μm; the average fabric- and matrix-layer volume fractions were 0.827±0.070 and 0.173±0.070, respectively; the average fiber packing and the microporosity volume fractions within the fabric layers were 0.502±0.060 and 0.065±0.007, respectively; and the interlayer macroporosity eccentricity factor was 0.17±0.05 (the average interlaminar macropore height-to-breadth ratio) [4]. Importantly, for the Hi-Nicalon™ type S composite the bulk density and fabric-layer volume fractions were higher, and the fiber packing and microporosity volume fractions were lower compared to those same values determined for the similarly fabricated 2D Hi-Nicalon™ composite [6].



Figures 1(a-b). SEM micrographs (backscatter electron mode) of polished cross-sections depicting (a) the typical fiber bundle geometry and macroporosity content, shape and orientation, and (b) the excellent matrix infiltration of the individual fiber bundles for the 2D Hi-Nicalon™ type S SiC_f/SiC composite.

In Figure 2, the effective through-thickness thermal conductivity (K_{eff}) determined as a function of temperature for the type S composite is compared to K_{eff}-values similarly determined for the 2D Hi-Nicalon and a 2D Tyranno™ SA composite [4]. The K_{eff}-values for the type S composite decreased continuously from a 28 W/mK maximum at 100°C down to 14 W/mK at 1000°C. Also, the K_{eff}-values for the type S composite were greater than the K_{eff}-values for either the Tyranno™ SA or the Hi-Nicalon™ composites. H2L model analysis of the temperature dependent K_{eff}-curve for the type S composite led to the predictions that (a) the thermal conductivity of the PyC fiber coating constituent increased from 28 W/mK at RT up to a maximum 34 W/mK at 300°C then decreased gradually to 28 W/mK at 1000°C, and (b) the thermal conductivity of the "intrinsic" ICVI-matrix constituent decreased continuously from a maximum value of about 40 W/mK at RT down to a value of 18 W/mK at 1000°C [4].

In Figure 3, the 4-pt bend stress-strain behavior for the 2D Hi-Nicalon™ type S composite bars tested at RT in air and 800°C in argon is illustrated. Also presented are data for similarly fabricated 2D Hi-Nicalon™ composite bars tested at 800°C. For clarity, the strain data has been offset by 0.2% for each group of data. All the stress-strain curves exhibit "graceful" failure, which is characteristic of a toughened

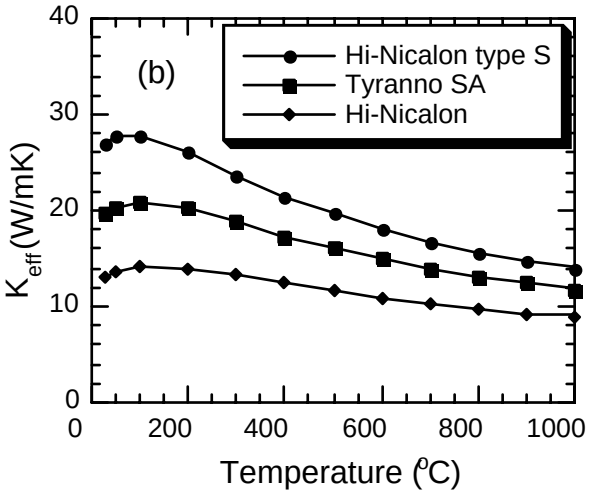


Figure 2. Average K_{eff}-values for 2D SiC_f/SiC composites made by ICVI with either Hi-Nicalon™ type S, Tyranno™ SA or Hi-Nicalon™ woven fabrics [4].

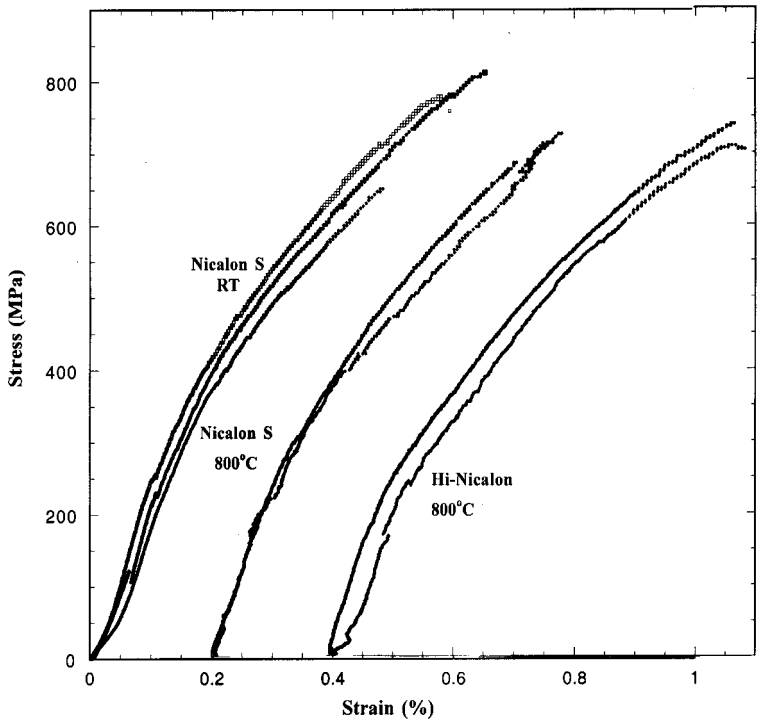


Figure 3. Stress-Strain curves for similarly CVI-fabricated SiC_f/SiC composite made with either Hi-Nicalon™ type S or Hi-Nicalon™ 2D woven fabric.

pseudo-plastic SiC_f/SiC composite. In Table 1, the average values of the ultimate stress and strain, the elastic modulus of the linear portion of the stress-strain curves, and the proportional limit stress determined from the stress-strain curves illustrated in Figure 3 are listed in bold print. For comparison, values for similarly tested 2D SiC_f/SiC composites made by FCVI with either PW type S or PW Hi-Nicalon™ fabric (by ORNL) and reported by Hinoki, et al [2] are given in standard print.

Table 1. Average values of ultimate stress and strain, elastic modulus and proportional limit stress determined from stress-strain curves for Hi-Nicalon™ type S and Hi-Nicalon™ SiC/SiC composites made by the CVI-process and tested in 4-pt bend configuration.

Material	Test Temp (°C)	Ult. Stress (MPa)	Ult. Strain (%)	Modulus (GPa)	Prop. Stress (MPa)
Nic S, 5HS	**RT**	**749 (66)***	**0.55 (0.08)**	**284 (16)**	**240 (20)**
Nic S, 5HS	**800**	**720 (8)**	**0.56 (0.01)**	**243 (12)**	**225 (25)**
Nic S, PW [4]	RT	425	1.42#	32#	280
Hi-Nic, PW	**RT**	**650 (60)**	**0.83 (0.10)**	**239**	**160**
Hi-Nic, PW	**800**	**724 (15)**	**0.66 (0.02)**	**255 (12)**	**180 (20)**
Hi-Nic, PW [4]	RT	450	1.40#	42#	200

* maximum variation from average values given in parentheses.

#Apparently the machine compliance was not correctly applied to this data as the ultimate strain values obviously are too high and the modulus values too low for SiC_f/SiC composite.

At least 6-8 bar samples should be tested for each condition to achieve statistically reliable average strength values. Since only a limited number of samples (6 total) were available at the time of testing, the results are expected to indicate trends, not statistically reproducible values. Also, in Table 1 the proportional limit stress was taken as the first occurrence of non-linearity in the stress-strain curve. For fiber reinforced composites, the PLS has importance primarily as an indication of the first matrix cracking stress, thus can be useful as a fabrication parameter.

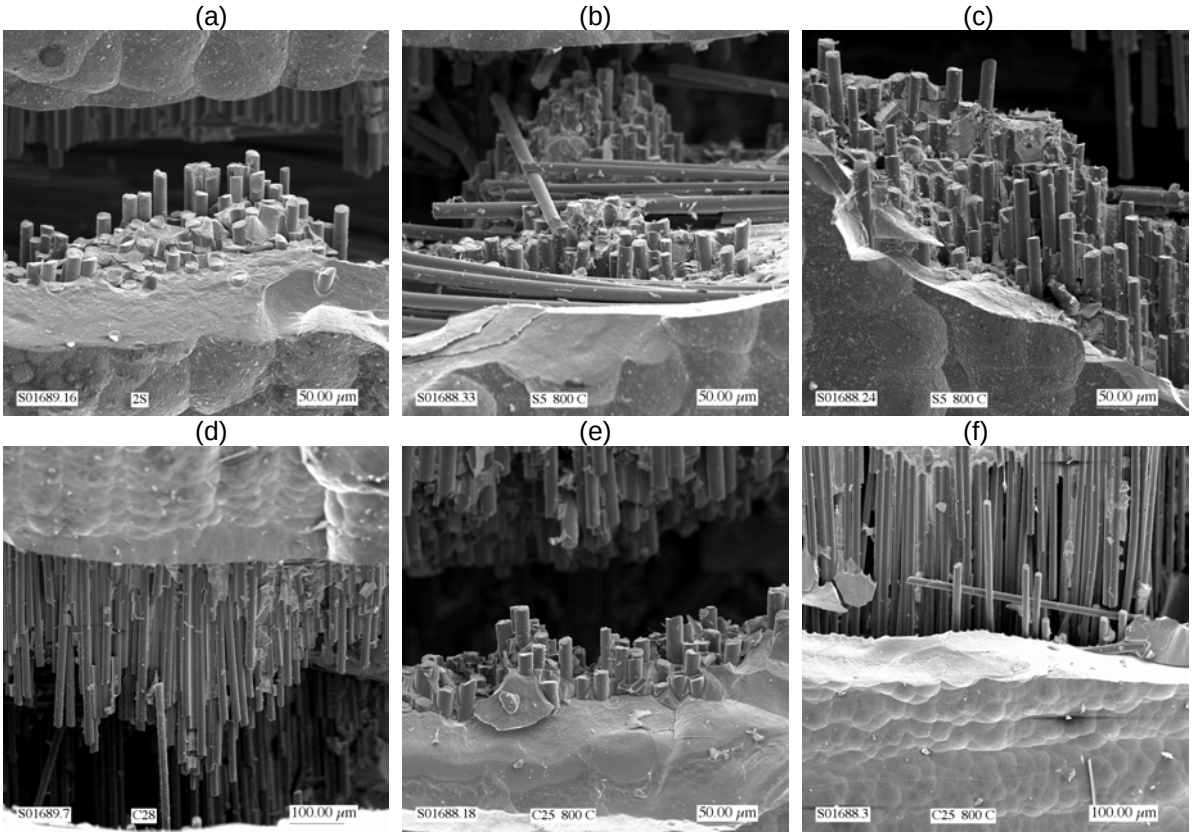
Discussion

The as-received 2D Hi-Nicalon™ type S composite made by ICVI-processing had extremely good fiber bundle infiltration that produced well-bonded fiber/matrix interfaces with a high interfacial conductance of about 3 x 10^4 W/cm^{2}K (estimated using the H2L model) [7]. This material also had a relatively low microporosity within the fiber bundles and low interlaminar macroporosity content and shape factor for CVI-processed material (see Figure 1). For these reasons, the thermal conductivity and bend strength values also were considerably improved over corresponding values for either the Hi-Nicalon™ or the Tyranno™ SA composites similarly fabricated. The approximate factor of two improvement of K_{eff}-values for the type S over that of the Hi-Nicalon™ material was not unexpected since the Nicalon™ type S fiber thermal conductivity (K_f) values are ≈x4 higher than K_f-values for the Hi-Nicalon™ fiber [8]. However, the Tyranno™ SA composite was expected to have the highest K_{eff}-values because of its high K_f-values, larger than a factor x2 that of the Nicalon™ type S fiber [8]. Instead, K_{eff}-values for a similarly fabricated Tyranno™ SA system were 20-30% lower than values for the type S system (see Figure 2). According to the H2L model, the relatively high macroporosity content and shape factor for the SA system offset thermal conductivity gains expected from using the high conductivity SA fiber. Furthermore, the H2L model predicted that the intrinsic matrix constituent thermal conductivity resulting from the ICVI-process was about 25% higher than that resulting from the FCVI-process [4]. The ICVI-matrix was formed over a period of about a month, while the FCVI-matrix infiltration was completed in a few days. Apparently, subtle substructural differences between the slowly formed matrix of the ICVI- and the more rapidly formed FCVI-matrix components were responsible for the H2L-model predicted differences in matrix constituent conductivities, although this explanation needs further examination.

Further evidence for obtaining a “better” composite when using the ICVI- rather than the FCVI-process is revealed by examining the bend strengths for similarly tested type S or Hi-Nicalon™ composites (see Table 1). The ultimate bend strengths for the ICVI-processed type S and Hi-Nicalon™ composites were quite high (749 and 650 MPa at RT and 720 and 724 MPa at 800°C, respectively). The ultimate bend strengths for both the FCVI-processed type S and Hi-Nicalon composites were considerably lower (425 and 450 MPa at RT, respectively).

Interestingly, the RT bend strength of the Hi-Nicalon™ composite was about 75 MPa lower than its bend strength at 800°C, while little difference in strengths at RT and 800°C was observed for the type S composite. Subtle differences in the residual radial clamping stress likely are the reason for this temperature dependence. Since the thermal expansion coefficient for Hi-Nicalon™ fiber is less than that for CVI-SiC matrix, a clamping stress is expected to slightly decrease the strength for the Hi-Nicalon™ material, more so for the RT case. In contrast, the stoichiometric type S SiC fiber likely has similar thermal expansion characteristics as the CVI-SiC matrix constituent, and residual stresses due to thermal expansion mismatch are not expected in the as-received type S composite. Moreover, irradiation induced swelling is expected to be similar in these type S composite constituents, which likely is the major reason little strength degradation was observed for this material by Hinoki, et al when it was irradiated [2].

The bend stress-strain curves for the type S composite tested at either RT or 800°C (see Figure 3) exhibit similar behavior, i.e., besides high bend strengths (>720 MPa), the curves indicate a high ultimate strain (>0.55 %), bend modulus (>243 GPa) and proportional limit stress (>225 MPa). Furthermore, these stress-strain curves indicate a slow transition from linear to pseudo-plastic behavior with a rather small



Figures 4(a-f). SEM micrographs showing characteristic fiber pull-out at tensile surface of bend bars tested at RT or 800°C: type S at (a) RT, (b) 800°C and (c) 800°C, respectively; and Hi-Nicalon at (d) RT, (e) 800°C and (f) 800°C, respectively.

decrease in the non-linear slopes. Several typical SEM views of fracture surfaces for the 2D type S and Hi-Nicalon™ composites taken at the tensile surface of the broken bend bars are compared in Figures 4(a-c) and 4(d-f), respectively. For the type S material tested at either RT or 800°C, Figures 4(a-c) reveal relatively short (10-50 μm), but random fiber pull-out lengths within a fiber bundle. Such observations were typical of views taken at many other locations. For the Hi-Nicalon™ material, some views revealed similar short, random pull-out length distributions (Figure 4e), but others revealed noticeably longer, and a not so random distribution of pull-out lengths (Figures 4d at RT and 4f at 800°C). The short, but random distribution of fiber pull-out lengths exhibited by the type S material are characteristic of a composite with relatively high fiber-matrix interfacial shear stresses as well as with uniformly high individual fiber strengths within the fiber bundles. Such stochastic fracture behavior leads to a desirable, relatively high proportional limit stress and also a gradual transitioning from linear to non-linear stress-strain, as observed. Although the mechanical properties of the Hi-Nicalon™ composite were quite good, the occurrence of some fiber bundles with longer and less randomly distributed fiber pull-out lengths suggests somewhat poorer fiber bundle infiltration for this material. This characteristic then leads to a slightly diminished proportional limit stress (160 and 180 MPa at RT and 800°C, respectively) compared to the type S material and a more abrupt transitioning from linear to non-linear stress-strain, also as observed.

For either 2D composite, the minimum fusion design strength and stiffness requirements can be met by ICVI-processing. Unfortunately, it does not appear likely that minimum fusion K_{eff}-goals can be obtained even for an optimized 2D SiC_f/SiC composite system when using the CVI process for matrix infiltration. The margin of improvement required to meet fusion design K_{eff}-goals is just too large for only minor

improvements potentially possible through CVI-processing upgrades or structural or architectural methods. Other strategies to obtain fusion K_{eff}-goals are recommended.

Future Work

Slow crack growth tests currently are being performed on compact tension samples cut from the eight-ply Hi-Nicalon™ type S plate. Additional bars and discs of the six-ply material are available for testing by others.

Acknowledgments

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