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Discontinuous surface-treated submicron-diameter carbon filaments as an interlaminar filler in carbon fiber polymer-matrix composites for vibration reduction

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Abstract—Discontinuous surface-treated submicron-diameter carbon filaments are effective for use as an interlayer between continuous carbon fiber laminae in a nylon-6 matrix composite for enhancing the loss tangent (0.2 and 1.0 Hz) under flexure that involves bending of the fibers, without significant decrease of the flexural storage modulus or the tensile modulus or strength in the fiber direction. The surface treatment is oxidation using ozone. Without treatment, the filaments are not effective. The treated filaments amount to 0.64 vol.% of composite; the interlayer thickness is 77 μm. A viscoelastic interlayer is even more effective than the treated filament interlayer for enhancing the loss tangent, but the accompanying decrease in storage modulus is much more. The loss tangent for composite with viscoelastic interlayer decreases upon heating, so the loss modulus for this composite is less than that of the composite with treated filament interlayer at $\geqslant 50°C$.

Keywords: Polymer–matrix composite; damping; vibration; mechanical; carbon fiber; nylon.

1. INTRODUCTION

Structural vibration control is important for essentially any structure, whether bridges, aerospace structures, turbine blades or skis. Due to the increasingly common use of one-piece composite constructions, vibration damping derived from fastened joints is insufficient and that derived from the composite material itself is needed. A conventional way to increase the damping capacity of a structure is the attaching or embedding of a viscoelastic material on or in the structure [1–7]. For example, in the case of a continuous fiber polymer–matrix composite, which is the most common form of structural composite, a viscoelastic sheet can be placed as an interlayer in the interlaminar region between the continuous fiber layers during composite fabrication. This method is effective for enhancing the damping capacity (described by the loss tangent, or tan δ), but it causes decrease of the stiffness (elastic modulus). Tanimoto reported that the modulus is decreased whenever a viscoelastic interlayer is present, whether an interlayer is present in every interlaminar region of the composite or not [7]. Moreover, Tanimoto reported that the use of a viscoelastic interlayer in every interlaminar region causes the strength to be lower than the corresponding composite without any interlayer, although the use of viscoelastic interlayers in some but not all interlaminar regions of a multidirectional laminate causes the strength to increase due to reduction in interlaminar stress. As stiffness is as important as damping capacity in vibration reduction, both high stiffness and high damping capacity are needed. The objective of this paper is to provide an alternative interlayer material, whereby the damping capacity is enhanced, with essentially no degradation in stiffness.

In this paper, carbon fibers refer to conventional ones of diameter 7 μm, whereas carbon filaments refer to unconventional ones of diameter 0.15 ± 0.05 μm.

Previous work [8] has shown that discontinuous submicron-diameter carbon filaments used as an interlayer material between continuous carbon fiber laminae in an epoxy-matrix composite substantially increase the loss tangent, with only slight decrease in the stiffness. The increase in loss tangent is due to the small diameter of the filaments and the resulting large area of the interface between filaments and matrix. Interfacial slippage is a mechanism of damping. These filaments were made catalytically from carbonaceous gases. They were used as received, i.e. without surface treatment. Recent work [9] has shown that surface treatment of these filaments by ozone gas improves the static mechanical properties (e.g. tensile strength) of cement-matrix composites containing these filaments. Such improvement due to ozone treatment also applies to conventional pitch-based carbon fibers used to reinforce cement [10–12]. Although the effectiveness of this treatment for polymer–matrix composites has not been shown, ozone treatment is an oxidation treatment and surface oxidation of carbon fibers by various other methods (e.g. by using an oxygen plasma, by heating in an oxidizing atmosphere or by acid treatment) has been previously shown to improve the bond between carbon fibers and polymer matrices [13]. Therefore, in this work, a systematic comparison is made among carbon filaments with and without surface treatment and a viscoelastic material with regard to the effectiveness of these three types of interlayer material for vibration reduction. In the work by Hudnut and Chung [8], comparison was not made with respect to a viscoelastic material, but was just made with respect to the case of no interlayer. Because a fair comparison requires testing under the same conditions (loading frequency, displacement amplitude, testing geometry, temperature, etc.), a systematic comparative study is needed. Hudnut and Chung used a thermoset (epoxy) matrix, but this work used a thermoplast (nylon-6) matrix, in order to show the applicability of the filament interlayer to both thermoset and thermoplast matrices.

In this paper, we report that the ozone treatment is highly effective for enhancing both loss tangent and stiffness of the composite containing the filament interlayer. The treated filaments give composites exhibiting lower loss tangent but higher stiffness than the viscoelastic interlayer. The overall effect, as described by the loss modulus, which is the product of loss tangent and storage modulus (stiffness under dynamic loading), is that the treated filaments are less effective than the viscoelastic interlayer at room temperature, but are more effective than the viscoelastic interlayer at 50°C or above. On the other hand, high stiffness is usually a basic requirement of a structural composite. For applications requiring high stiffness, the treated filaments would be more suitable than the viscoelastic material for vibration damping, at whatever temperature.

2. EXPERIMENTAL METHODS

2.1. Materials

Continuous unidirectional carbon fiber (Grafil, Inc., Sacramento, CA, Fiber Type 34-700, tensile strength = 4.5 GPa, modulus = 234 GPa, density = 1.80 g/cm^3, elongation = 1.9%, diameter = 6.9 μm) nylon-6 matrix prepregs supplied by Quadrax Corp. (Portsmouth, RI, Product QNC 4162) were used. This thickness of the prepreg (one ply) was 150 μm. The fiber weight fraction was 62%. The fiber volume fraction was 51%. The fiber areal weight was 150 g/m^2. The density of the nylon-6 matrix was 1.3 g/cm^3. The melting temperature of the matrix was 220°C. The recommended minimum composite processing temperature was 270°C.

Nylon-6 is a polyamide which is characterized by its resistance to oils, greases, solvents, bases, fatigue, repeated impact and abrasion. It exhibits a low coefficient of friction, high tensile strength, toughness, barrier properties, creep resistance and retention of properties over a wide range of temperatures (from − 60 to 110°C).

Discontinuous carbon filaments of diameter 0.15±0.05 μm and exhibiting a naturally bent morphology (resembling cotton wool) were supplied by Applied Sciences Inc. (Cedarville, OH; Product ADNH). Their length was at least 100 μm. Their density was 2.0 g/cm^3. They were made catalytically from carbonaceous gases, in contrast to conventional carbon fibers, which are made from pitch or polymers.

The viscoelastic material used was an acrylic sheet supplied by 3M Vibration Control (St. Paul, MN; Product ISD) for use in damping. It contained no plasticizer. It could withstand temperatures up to 150°C, though the peak damping performance was at 40–100°C. It was tack free at room temperature; heat and pressure were required to bond this material to a substrate. Its thickness was 2 mils (0.002 in, or 0.051 mm).

2.2. Carbon filament surface treatment

The carbon filaments were first cleansed with acetone in order to remove surface contaminants [14]. This cleansing involved washing repeatedly (2 or 3 times) in acetone. Washing was performed by using a blender to stir the slurry of filaments in acetone. After that, the filaments were rinsed with water and then allowed to dry in air at 60°C for about 5 h.

After cleansing, the filaments were surface oxidized by exposure to ozone gas (0.6 vol.% ozone in oxygen) at 160°C for 5 min, using a tube resistance furnace.

2.3. Composite fabrication

Composite materials were fabricated by stacking eight plies of carbon fiber prepreg and consolidating them by compression molding in a steel mold (17 $\times$ 9 cm, platens separated from the prepreg stack by mold release films). The different plies had fibers oriented in the same direction. An interlayer was optionally placed in every interlaminar region of the stack. Hence, there were seven interlayers in each composite material.

Three types of interlayer material were used, namely, a viscoelastic sheet, carbon filaments without surface treatment (i.e. as received), and carbon filaments with surface treatment. The weight of carbon filaments per interlayer was 0.08 g. The volume fractions of filaments, fibers and the nylon-6 matrix, and the thicknesses of composite and interlayer are listed in Table 1. The interlayer thicknesses were determined by optical microscopic observation of polished sections of the composites.

The compression molding was performed at 0.40 and 0.12 MPa respectively for composites without viscoelastic interlayer and that with viscoelastic interlayer. The maximum temperature during compression molding was 270 and 110°C respectively for composites without viscoelastic interlayer and that with viscoelastic interlayer. A lower temperature was used for the composite with viscoelastic interlayer due to the limited temperature tolerance of the viscoelastic material. The maximum temperature was held for no more than 5 min.

Table 1.
Composition of continuous carbon fiber nylon-6 matrix composites with and without interlayers

Interlayer	None	Viscoelastic	As-received carbon filaments	Treated carbon filaments
Interlayer thickness (μm, $\pm$ 3)	0	55	65	77
Composite thickness (mm, $\pm$ 0.01)	0.99	1.20	1.02	1.08
Interlayer volume fraction in composite	0%	4.6%	6.3%	7.1%
Filaments volume fraction in interlayer	0%	0%	10.1%	9.2%
Filaments volume fraction in composite	0%	0%	0.62%	0.64%
Fiber volume fraction in composite	51%	42%	33%	31%
Nylon-6 volume fraction in composite	49%	53%	66%	68%

2.4. Static mechanical testing

Tensile testing up to failure was performed at room temperature to determine the tensile strength, modulus and ductility in the fiber direction of each composite. Each sample was 80 mm long in the stress direction and 10 mm wide. The thickness is shown in Table 1. A resistive strain gage was attached to the center of one of the two opposite large faces of a sample in order to measure the strain during tensile testing. The modulus was taken as the initial slope of the stress–strain curve. Glass fiber reinforced epoxy end tabs were adhered to both sides of each end of a sample to facilitate gripping of the sample for the purpose of applying tension. A hydraulic mechanical testing system (MTS 810) was used at a crosshead speed of 0.1 mm/min. Three samples of each type were tested.

2.5. Dynamic mechanical testing

Dynamic mechanical testing was performed under flexure (three-point bending) at controlled loading frequencies (0.2 or 1.0 Hz) and controlled temperatures (20, 50 or 100°C) to determine the damping capacity ($\tan\delta$), storage modulus and loss modulus (product of $\tan\delta$ and storage modulus).

The testing was performed with the sample in the direction such that the fibers were bent during flexural testing (known as the longitudinal configuration, Fig. 1a) and in the direction such that the fibers were not bent during flexural testing (known as the transverse configuration, Fig. 1b). The longitudinal configuration reflects properties that are governed mainly by the fibers; the transverse configuration reflects properties that are governed mainly by the polymer matrix. The properties of a practical structural composite with fibers in multiple directions are mainly governed by the fibers, so the longitudinal configuration is of more practical importance than the transverse configuration.

The displacement during flexural testing was 8–12 μm. A Perkin-Elmer dynamic mechanical analyzer (DMA 7e) was used. The span in three-point bending was 62 and 15 mm respectively for room temperature testing and controlled temperature

(a)

(b)

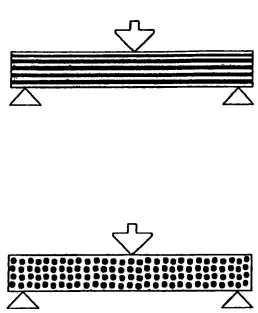

Figure 1. Geometry for dynamic mechanical testing under three-point bending. (a) Longitudinal configuration, in which the fibers (in the plane of the figure) were bent during flexure. (b) Transverse configuration, in which the fibers (perpendicular to the plane of the figure) were not bent during flexure.

Table 2.
Tensile properties in the fiber direction of continuous carbon fiber nylon-6 matrix composites with and without interlayers

Interlayer	None	Viscoelastic	As-received carbon filaments	Treated carbon filaments
Strength (MPa)	1300 ± 5	984 ± 5	1155 ± 5	1231 ± 4
Modulus (GPa)				
Measured	96 ± 2	43 ± 2	63 ± 2	74 ± 1
Calculated*	96	80	62	58
Ductility (%)	1.31 ± 0.05	2.3 ± 0.1	1.8 ± 0.1	1.59 ± 0.06

* Calculated by Rule of Mixtures, relative to the measured value of the composite without interlayer, and assuming that the interlayer makes no contribution to the modulus of the composite.

(20, 50 and 100°C) testing. The sample length in the span direction was 80 and 25 mm respectively for room temperature testing and controlled temperature testing. A smaller span was used for controlled temperature testing due to the sample size limit imposed by the furnace used for controlling the temperature. In controlled temperature testing, the temperature was varied at a constant loading frequency (either 0.2 or 1.0 Hz). Three samples of each type were tested.

2.6. Metallography

The internal structure of composites was examined by observing mechanically polished sections under an optical microscope. Both sections perpendicular and parallel to the fibers were examined. The section perpendicular to the fibers is like Fig. 1b; that parallel to the fibers is like Fig. 1a.

3. RESULTS

3.1. Metallography

Figures 2–5 show optical microscope photographs of composite without interlayer, that with a viscoelastic interlayer, that with as-received carbon filament interlayer, and that with surface treated carbon filament interlayer, respectively. In each figure, (a) shows the section perpendicular to the fibers and (b) shows a section parallel to the fibers. Figure 6 is Fig. 5a at a higher magnification. The laminae in the composite without interlayer essentially could not be distinguished, as is typical of a unidirectional laminate (Fig. 2). For the composites with interlayers, the laminae were observed to be separated by interlayers, such that an interlayer was not uniform in thickness and was not flat (Figs 3–5). The structure within an interlayer could not be observed, even at a relatively high magnification (Fig. 6), due to the small diameter of the carbon filaments in the filament interlayer and the absence of a filler in the viscoelastic interlayer.

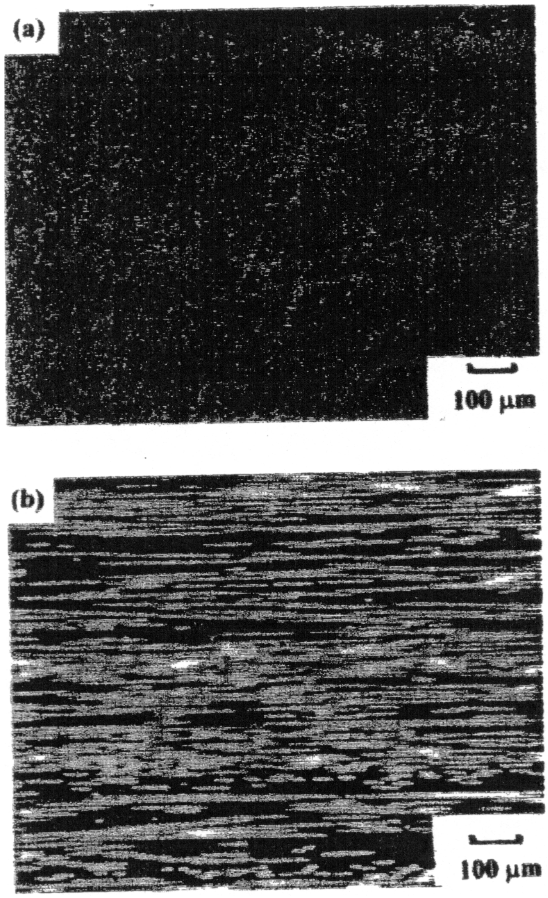

Figure 2. Optical microscope photograph of composite without interlayer. (a) Section perpendicular to fibers. (b) Section parallel to fibers.

3.2. Static mechanical testing

Figure 7 shows tensile stress-strain curves of the four types of composite, i.e. composite without interlayer, composite with viscoelastic interlayer, composite with as-received carbon filament interlayer and composite with surface treated carbon filament interlayer. Table 2 shows the tensile strength, modulus and ductility for each type of composite. The strength and modulus were decreased and the ductility was increased by any type of interlayer, such that the changes were greatest for the case of the viscoelastic interlayer and least for the case of the surface treated carbon filament interlayer. Although the fiber volume fraction was lower for the two cases of filament interlayer than for the case of viscoelastic (Table 1), the strength and modulus were higher for the former (Table 2). Surface treated carbon filaments gave higher strength, higher modulus and lower ductility than as-received

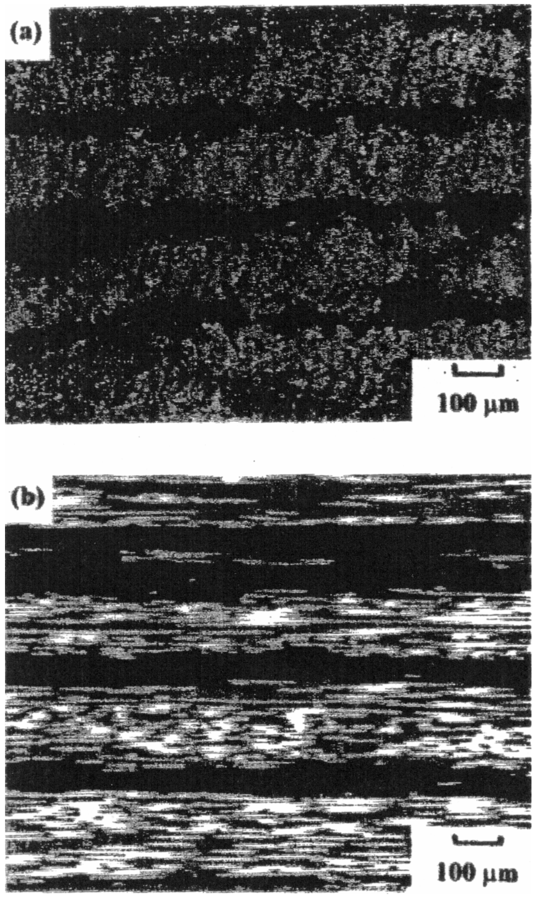

Figure 3. Optical microscope photograph of composite with viscoelastic interlayer. (a) Section perpendicular to fibers. (b) Section parallel to fibers.

carbon filaments, though the fiber volume fraction was a little lower for the former composite.

3.3. Dynamic mechanical testing

Table 3 shows the dynamic mechanical properties at room temperature. The loss tangent for the longitudinal configuration was increased by any of the three types of interlayer; the storage modulus for the longitudinal configuration was decreased by any of the three types of interlayer. For the transverse configuration, the loss tangent was increased by the viscoelastic interlayer, but not much affected by either type of filament interlayer; the storage modulus was decreased by the viscoelastic interlayer and the as-received carbon filament interlayer, but was increased slightly by the treated carbon filament interlayer. The loss tangent for both longitudinal and transverse configurations was increased most significantly by the viscoelastic interlayer, while the storage modulus for both configurations was decreased most

Table 3.
Dynamic flexural properties of continuous carbon fiber nylon-6 matrix composites with and without interlayers

Interlayer	None	Viscoelastic	As-received carbon filaments	Treated carbon filaments
tan δ				
Longitudinal				
0.2 Hz	0.008 ± 0.001	0.43 ± 0.05	0.007 ± 0.001	0.09 ± 0.02
1.0 Hz	< 0.0001	0.36 ± 0.05	0.001 ± 0.001	0.001 ± 0.001
Transverse				
0.2 Hz	0.065 ± 0.005	0.24 ± 0.05	0.060 ± 0.005	0.052 ± 0.005
1.0 Hz	0.080 ± 0.005	0.22 ± 0.06	0.090 ± 0.005	0.073 ± 0.005
Storage modulus (GPa)				
Longitudinal				
0.2 Hz	127 ± 8	37 ± 4	66 ± 5	115 ± 6
1.0 Hz	132 ± 9	67 ± 5	67 ± 3	97 ± 5
Transverse				
0.2 Hz	9.6 ± 0.2	3.8 ± 0.2	6.1 ± 0.2	10.2 ± 0.3
1.0 Hz	9.9 ± 0.3	4.4 ± 0.2	6.3 ± 0.2	10.8 ± 0.3
Loss modulus (GPa)				
Longitudinal				
0.2 Hz	1.0 ± 0.3	16 ± 1	0.35 ± 0.10	9 ± 5
1.0 Hz	< 0.013	23.5 ± 1.5	0.067 ± 0.002	< 0.097
Transverse				
0.2 Hz	0.62 ± 0.03	0.90 ± 0.20	0.067 ± 0.002	0.60 ± 0.05
1.0 Hz	0.79 ± 0.04	0.94 ± 0.20	0.500 ± 0.003	0.78 ± 0.05

significantly by the viscoelastic interlayer. For both configurations, the loss modulus was highest for the case of the viscoelastic interlayer. All effects were much larger for the longitudinal configuration than the transverse configuration.

The storage modulus, particularly for the longitudinal configuration, was much increased upon increasing the frequency from 0.2 to 1.0 Hz, as expected for a viscoelastic material. For the filament interlayer cases, the effect of frequency on the storage modulus was small. For the longitudinal configuration, the loss tangent was much decreased as the frequency was increased from 0.2 to 1.0 Hz; for the transverse configuration, the effect of frequency was small.

Figures 8–10 show the loss tangent, storage modulus and loss modulus respectively as functions of temperature for the four types of composites in the longitudinal configuration at a loading frequency of 0.2 Hz. The loss tangent was highest for the composite with viscoelastic interlayer (Fig. 8), though the value for this composite decreased substantially with increasing temperature, especially from 20 to 50°C. The composite with treated carbon filament interlayer gave higher loss tangent than that with as-received filament interlayer. The values for these filament composites

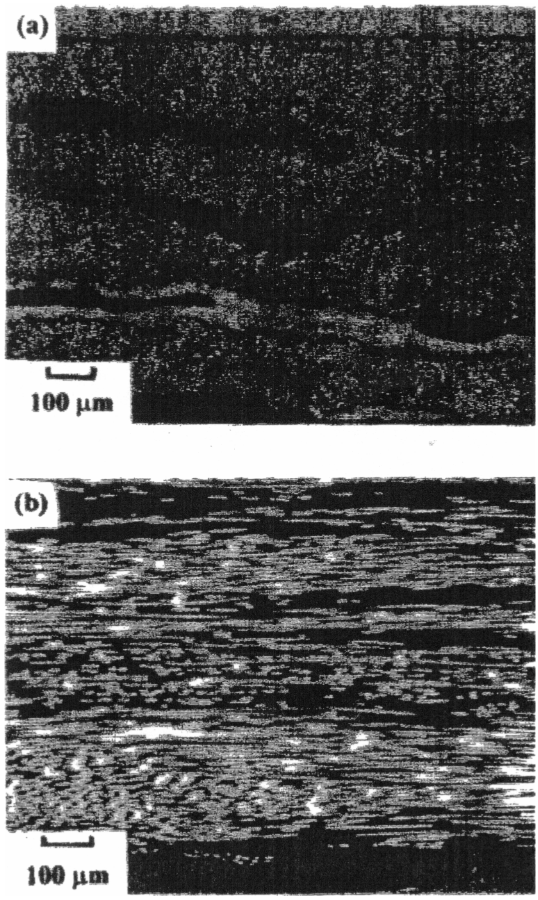

Figure 4. Optical microscope photograph of composite with as-received carbon filament interlayer. (a) Section perpendicular to fibers. (b) Section parallel to fibers.

dropped only slightly with increasing temperature. The storage modulus decreased with increasing temperature for any of the four types of composites (Fig. 9). The loss modulus also decreased with increasing temperature for any of the four types of composites (Fig. 10); the decrease was most significant for the composite with viscoelastic interlayer, particularly from 20 to 50°C. At 20°C, the composite with viscoelastic interlayer exhibited the highest loss modulus; at 50 and 100°C, the composite with treated filaments exhibited the highest loss modulus.

Figures 11–13 show the loss tangent, storage modulus and loss modulus respectively as functions of temperature for the four types of composites in the transverse configuration at a loading frequency of 0.2 Hz. The loss tangent was highest for the composite with viscoelastic interlayer at 20 and 50°C; at 100°C, the loss tangent was highest for the composite with treated filament interlayer (Fig. 11). This is because the loss tangent decreased with increasing temperature for the composite with viscoelastic interlayer, but it increased with increasing temperature for the compos-

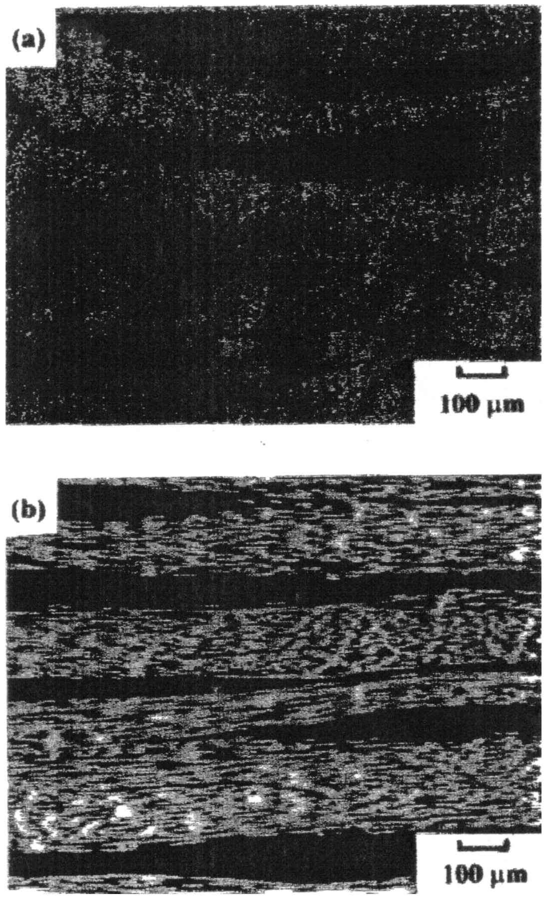

Figure 5. Optical microscope photograph of composite with surface treated carbon filament interlayer. (a) Section perpendicular to fibers. (b) Section parallel to fibers.

ite with treated filament interlayer. For the composite without interlayer and that with as-received filament interlayer, the loss tangent was essentially independent of temperature. The storage modulus decreased with increasing temperature for any of the four types of composites (Fig. 12). The loss modulus decreased with increasing temperature for the composite with viscoelastic interlayer and the composite with as-received filament interlayer, but increased with increasing temperature for the composite with treated filament interlayer (Fig. 13). For the composite without interlayer, the loss modulus increased as the temperature was increased from 20 to 50°C, but decreased slightly as the temperature was further increased to 100°C. At 50°C and 100°C, the loss modulus was highest for the composite with treated filament interlayer and lowest for the composite with as-received filament interlayer and that with viscoelastic interlayer. However, at 20°C, the loss modulus was highest for the composite with viscoelastic interlayer and lowest for the composite without interlayer.

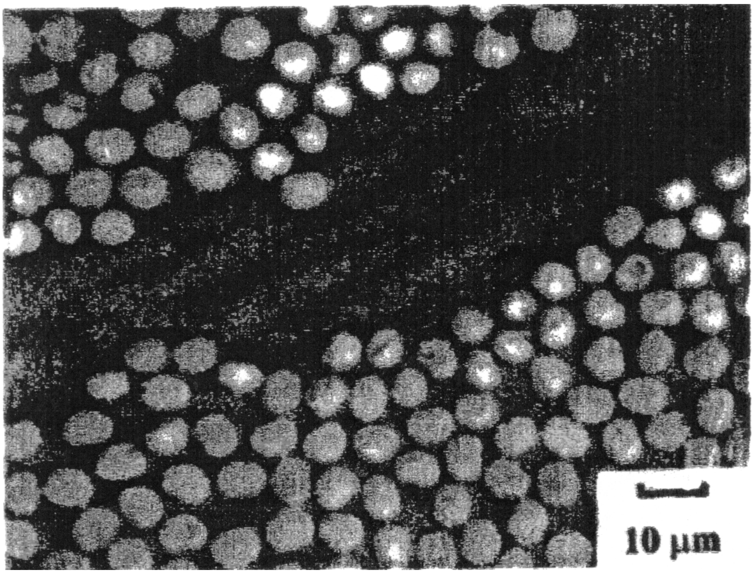

Figure 6. Fig. 5a at a higher magnification.

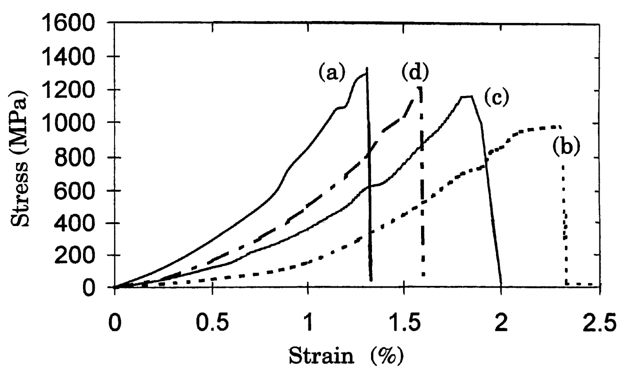

Figure 7. Tensile stress–strain curves of (a) composite without interlayer, (b) composite with viscoelastic interlayer, (c) composite with as-received carbon filament interlayer, and (d) composite with treated carbon filament interlayer.

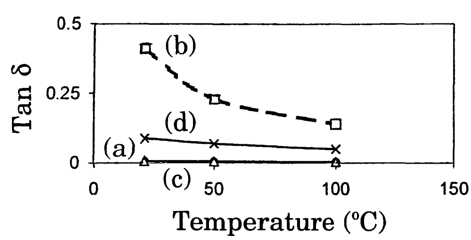

Figure 8. Effect of temperature on the loss tangent for the longitudinal configuration at 0.2 Hz. (a) Composite without interlayer. (b) Composite with viscoelastic interlayer. (c) Composite with as-received carbon filament interlayer. (d) Composite with treated carbon filament interlayer.

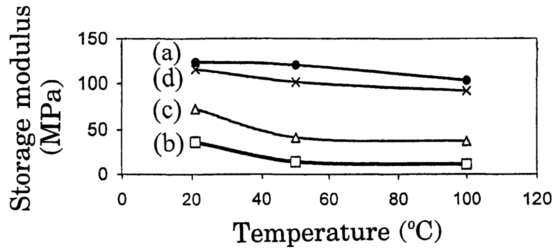

Figure 9. Effect of temperature on the storage modulus for the longitudinal configuration at 0.2 Hz. (a) Composite without interlayer. (b) Composite with viscoelastic interlayer. (c) Composite with as-received carbon filament interlayer. (d) Composite with treated carbon filament interlayer.

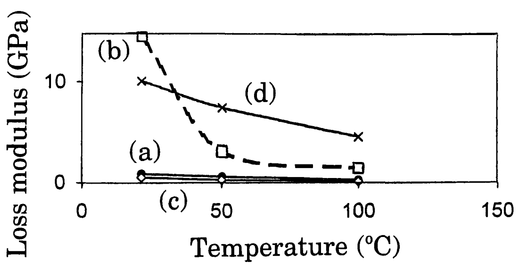

Figure 10. Effect of temperature on the loss modulus for the longitudinal configuration at 0.2 Hz. (a) Composite without interlayer. (b) Composite with viscoelastic interlayer. (c) Composite with as-received carbon filament interlayer. (d) Composite with treated carbon filament interlayer.

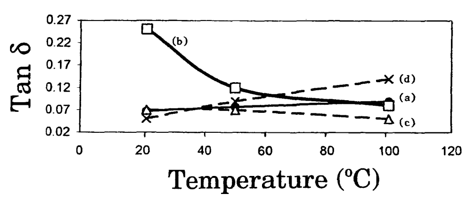

Figure 11. Effect of temperature on the loss tangent for the transverse configuration at 0.2 Hz. (a) Composite without interlayer. (b) Composite with viscoelastic interlayer. (c) Composite with as-received carbon filament interlayer. (d) Composite with treated carbon filament interlayer.

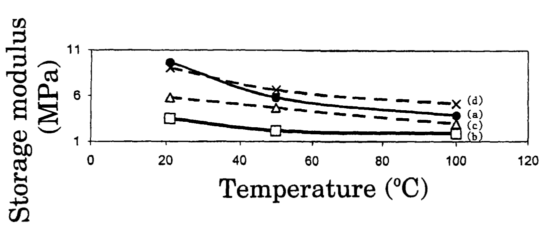

Figure 12. Effect of temperature on the storage modulus for the transverse configuration at 0.2 Hz. (a) Composite without interlayer. (b) Composite with viscoelastic interlayer. (c) Composite with as-received carbon filament interlayer. (d) Composite with treated carbon filament interlayer.

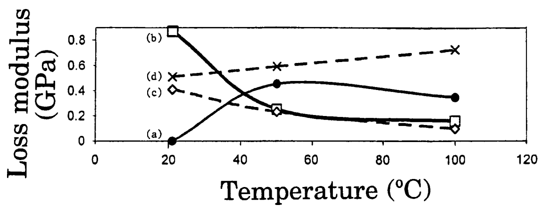

Figure 13. Effect of temperature on the loss modulus for the transverse configuration at 0.2 Hz. (a) Composite without interlayer. (b) Composite with viscoelastic interlayer. (c) Composite with as-received carbon filament interlayer. (d) Composite with treated carbon filament interlayer.

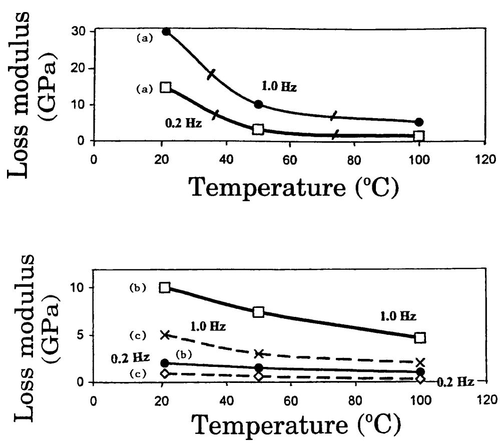

Figure 14. Effect of temperature and frequency on the loss modulus for the longitudinal configuration. (a) Composite with viscoelastic interlayer. (b) Composite without interlayer. (c) Composite with treated carbon filament interlayer.

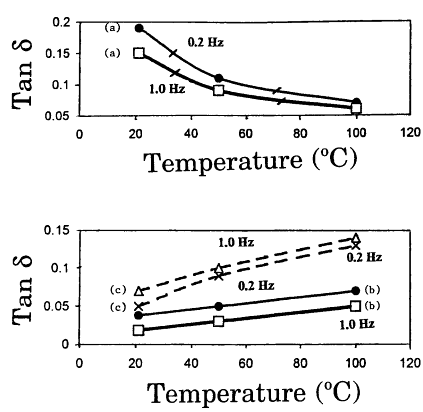

Figure 15. Effect of temperature and frequency on the loss tangent for the transverse configuration. (a) Composite with viscoelastic interlayer. (b) Composite without interlayer. (c) Composite with treated carbon filament interlayer.

For any of the four types of composites in the longitudinal configuration, the loss modulus at any temperature increased as the loading frequency was increased from 0.2 to 1.0 Hz (Fig. 14), mostly because of the increase of the storage modulus with increasing frequency. For the transverse configuration, the loss tangent decreased with increasing frequency for composite with viscoelastic interlayer (Fig. 15a) and that with no interlayer (Fig. 15b), but increased slightly with increasing frequency for composite with treated filament interlayer (Fig. 15b).

4. DISCUSSION

The damping ability of longitudinal composites with filament interlayers is due to the large area of the interface between filaments and matrix and the contribution of interfacial slippage to damping [8]. The increases in static tensile modulus and strength due to the surface treatment of the filaments (Table 2) suggest that bonding between filaments and matrix is enhanced by the surface treatment. This suggestion is consistent with the improved bond strength between pitch-based carbon fibers and cement paste after ozone treatment of the fibers [10–12]. The effect of ozone treatment on the specific surface area is small, as shown for both carbon filaments [15] and pitch-based carbon fibers [10]. The improved bonding is attributed to the increased abundance of oxygen-containing functional groups on the surface of the fibers [10] or filaments [15] after the ozone treatment. Thus, the increase in loss tangent of composite with filament interlayer due to surface treatment of the filaments is not due to an increase in the area of the interface between filaments and matrix, but is due to improved interfacial bonding. When the bonding is too weak, the filaments are less effective for reinforcing as well as damping.

The loss tangent (longitudinal) decreases significantly with increasing temperature for composite with viscoelastic interlayer, but decreases slightly with increasing temperature for composite without interlayer and composites with filament interlayers (Fig. 8). This is due to significant dependence on temperature of the damping ability of the viscoelastic interlayer material. Although the viscoelastic material is rated by its manufacturer to be most effective for damping at 40–100°C, the loss tangent of the composite with viscoelastic interlayer decreases monotonically with increasing temperature from 20 to 100°C. This is probably because the movement of the viscoelastic polymer molecules is affected by both the temperature and the constraint imposed by the laminae adhered to it. Although damping by the viscoelastic mechanism is highly dependent on the temperature, damping by interfacial slippage is only slightly dependent on the temperature. Nevertheless, the damping capacity of composite with viscoelastic interlayer is greater than that of composites with filament (whether treated or not) interlayers at all temperatures from 20 to 100°C.

Table 2 gives the values of the tensile modulus of composites with interlayers, as calculated by the Rule of Mixtures by assuming that the interlayers are zero in the contribution to the modulus of the composite and by using the measured modulus of the composite without interlayer (Table 2) and the volume fractions of fibers in the composites (Table 1). The calculated modulus is lower than the measured modulus for the composites with treated filament interlayers, but is higher than the measured modulus for the composite with viscoelastic interlayer. This means that the treated filament interlayer is positive in modulus contribution, especially if the filaments have been surface treated, whereas the viscoelastic interlayer is negative in modulus contribution. That the composite with viscoelastic interlayer is negative in modulus contribution is probably due to slippage at the interface between viscoelastic interlayer and fiber lamina, as suggested by the high ductility of this composite. The interfacial slippage is possible due to the fiber laminae being not completely in the stress axis and the weak interfacial bond resulting from the low composite fabrication temperature. Therefore, the composite with viscoelastic interlayer has lower values of tensile modulus (Table 2) and longitudinal storage modulus (Table 3 and Fig. 9) than the composites with filament (whether treated or not) interlayers. The longitudinal storage modulus decreases with increasing temperature for all four types of composites (Fig. 9), due to the slight softening of the polymer matrix.

The loss modulus (longitudinal) is lower for composite with treated filament interlayer than that with viscoelastic interlayer at 20°C, but the opposite is true at 50 and 100°C (Fig. 10). This is due to the significant decrease of the loss tangent with increasing temperature for the composite with viscoelastic interlayer (Fig. 8) and the high storage modulus of the composite with treated filament interlayer (Fig. 9).

The loss tangent at room temperature for the transverse configuration is enhanced by the viscoelastic interlayer, but is essentially not affected by the filament (whether treated or not) interlayer (Table 3). This is because viscoelastic damping due to the polymer matrix dominates the damping of composites without viscoelastic interlayer. The addition of viscoelastic interlayer accentuates the viscoelastic damping. As the temperature is increased, the loss tangent of the composite with viscoelastic interlayer is decreased (Fig. 11). This is consistent with the decrease of the same quantity for the longitudinal configuration (Fig. 8). The loss tangent of the composite with treated filament interlayer increases with increasing temperature, although that of the composite with as-received filament interlayer and that of the composite without interlayer are relatively independent of temperature (Fig. 11). As the contribution of the polymer matrix to damping due to the viscoelastic damping mechanism is important for these composites in the transverse configuration, this temperature dependence suggests that the constraint of the polymer matrix due to the filaments (especially if treated) enhances the viscoelastic damping, which is more significant as the temperature increases. On the other hand, the storage modulus for the transverse configuration is greatly decreased by the viscoelastic interlayer (Table 3), probably because the viscoelastic interlayer is softer than the polymer matrix in the fiber laminae. The storage modulus (transverse) is also decreased, though not as greatly, by the as-received filament interlayer, but is slight increased by the treated filament interlayer. This is consistent with the weak bond between as-received filaments and the polymer matrix, the relatively strong bond between treated filaments and the polymer matrix, and the random orientation of the filaments (resembling cotton wool) causing the filaments to stiffen the composite in the transverse direction. In spite of the relatively high storage modulus (transverse) of the composite with treated filament interlayer, the loss modulus (transverse) of this composite at room temperature is lower than that of the composite with viscoelastic interlayer, due to the high loss tangent of the latter. However, due to the temperature dependence of the loss tangent, the loss modulus is higher for the composite with treated filament interlayer than that with viscoelastic interlayer at 50 and 100°C (Fig. 13).

The treated carbon filaments are effective for greatly enhancing the longitudinal loss tangent, though they are not effective for enhancing the transverse loss tangent. Nevertheless, it is the longitudinal configuration that is of practical importance to multidirectional composites. The treated filaments enhance the transverse storage modulus slightly, but decrease the longitudinal storage modulus slightly. The viscoelastic interlayer is much more effective than the treated filament interlayer in enhancing the longitudinal loss tangent; it is also effective for enhancing the transverse loss tangent. However, the viscoelastic interlayer greatly decreases the storage modulus for both longitudinal and transverse configurations. Therefore, for applications that require high stiffness, the viscoelastic interlayer is not suitable.

The significant drop in longitudinal loss tangent of the composite with viscoelastic interlayer upon heating from 20 to 50°C greatly diminishes the attractiveness of the viscoelastic interlayer at 50°C and above. The longitudinal loss modulus of the composite with viscoelastic interlayer is less than that of the composite with treated filament interlayer at 50°C and above (Fig. 10). Hence, the treated filament interlayer is more attractive than the viscoelastic interlayer for vibration reduction at 50°C and above.

The as-received carbon filament interlayer is about as effective for carbon fiber epoxy-matrix composite [8] as the treated filament interlayer is for carbon fiber nylon-6 matrix composite. This is attributed to the strong adhesive ability of epoxy and the resulting good bond between as-received filaments and epoxy.

The filament volume fraction is 0.6% for both epoxy-matrix [8] and nylon-6 matrix composites, but the filament interlayer could not be distinctly observed in the epoxy-matrix composite [8], but could be distinctly observed in nylon-6 matrix composite. The calculated thickness of the filament interlayer is 8.3 μm for epoxy-matrix composite [8], but the observed thickness of the filament interlayer is 65 μm for as-received filaments and 77 μm for treated filaments for nylon-6 matrix composite. This difference between epoxy and nylon-6 matrices is attributed to the higher fluidity of the epoxy resin compared to the nylon-6 polymer during composite fabrication.

Tanimoto used multidirectional laminates [7], whereas this paper uses unidirectional laminates. Due partly to the unidirectional nature of our laminates and partly due to our use of an interlayer in every interlaminar region of a laminate, the positive effect of the viscoelastic interlayer on the strength of laminates having viscoelastic interlayers in some but not all interlaminar regions, as reported in [7], is absent in this work. Nevertheless, our observed decrease in modulus due to the presence of a viscoelastic interlayer is consistent with that reported in [7].

5. CONCLUSION

Discontinuous surface-treated submicron-diameter carbon filaments were found to be effective for use as an interlayer between continuous unidirectional carbon fiber laminae in a nylon-6 matrix composite for enhancing the longitudinal flexural loss tangent (0.2 or 1.0 Hz) without much decrease of the longitudinal flexural storage modulus, tensile modulus or tensile strength. The transverse flexural storage modulus was slightly increased by the interlayer. The surface treatment was oxidation involving exposure to ozone gas. Without the treatment, the filaments were not effective for enhancing the loss tangent and the storage modulus was decreased, both for longitudinal and transverse flexural testing configurations. The treated filaments amounted to 0.64 vol.% of the composite; the interlayer thickness was 77 μm. A viscoelastic interlayer was even more effective than the treated filament interlayer for enhancing the loss tangent, but the accompanying decrease in storage modulus was even more than that caused by the use of as-received filament interlayer. At 50°C and above, the viscoelastic interlayer became less effective than it was at room temperature for enhancing the loss tangent, so the loss modulus (both longitudinal and transverse) was higher for the composite with treated filament interlayer than that with viscoelastic interlayer at 50°C and above.

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