

Performance improvement of carbon fiber/polyethylene fiber hybrid composites

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Carbon fiber/polyethylene (PE) fiber hybrid composites were fabricated by open leaky mold method. The positional effect of PE fiber was investigated concerning the mechanical performance improvement of carbon fiber/PE fiber hybrid composites. The influence of adhesion level of PE fiber on the hybrid properties was studied using oxygen plasma, γ -MPS, and γ -MPS-modified polybutadiene (PB/ γ -MPS) as surface modifiers. In case of carbon fiber/vinylester composite, γ -MPS and PB/ γ -MPS acted as the bridge to bond carbon fiber to vinylester resin through the chemical bonding. In case of PE fiber/vinylester composite, plasma treatment of PE fiber introduced the etching and micro-pitting rather than the functional group on fiber surface. Therefore, the plasma treated composite exhibited a large increase in flexural strength compared with untreated composite. In the case of carbon fiber/PE fiber hybrid composites, the mechanical properties of hybrid composite strongly depended on the reinforcing fiber position. When carbon fiber was at the outermost layer, the hybrid composite exhibited the highest flexural strength among other hybrid composites. This was attributed to the fact that the compressive and tensile stress had maximum magnitudes in the outermost layer. The surface treatment of PE fiber at outermost layer had a significant effect on the flexural strength of hybrid composite.

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1. Introduction

The term hybrid composite is used to denote the incorporation of two different types of fiber material in a common resin matrix [1–3]. The purpose of hybridization is to construct a new material that will retain the advantages of its constituents but not their disadvantages [4–6]. Recently some studies on hybrid composites have been directed to the carbon fiber/polyethylene fiber system [7–9]. Carbon fibers are well known for their high specific strength and stiffness both in tension and in compression. Composite materials based on these fibers have been increasingly used in primary and secondary structures during the last decade. However, the advantages of these materials are significantly reduced because of their susceptibility to impact damage. Various reports have been presented concerning impact improvement of carbon fiber-reinforced composite by hybridizing with glass, aramid, and polyethylene fibers [10–12]. A relatively new and tough reinforcing fiber is polyethylene (PE) fiber possessing unique mechanical properties in terms of high specific strength and stiffness. Moreover these PE fibers possess a high elongation at break leading to high values of work to break compared with other reinforcing fibers. Due to these properties, PE fibers have a high potential for use in composite structures requiring good impact properties.

However, PE fiber has poor adhesion to polymer matrix and the chemical inertness of it is disadvantageous for structural applications.

It is a well-known fact that the mechanical properties of polymer composite depend on the properties of each primary component, the nature of the interface, and the locus of fiber-matrix interaction. The hybrid composites also obey this principle. Because the PE fiber is extremely inert chemically, the surface treatment is required to produce an adequate bond between fiber and matrix [13–17]. By treating the surface of fiber to control the fiber-matrix adhesion of PE fiber composites, it is possible to control the mechanical and impact property of carbon/PE hybrid composites. Some studies have demonstrated that the amount of performance improvement of carbon/PE hybrid composites was strongly dependent on the adhesion level of the PE fibers [7, 18]. Apart from the adhesion level, the position of PE fiber plies in carbon/PE hybrid structure is also an important parameter for structural applications.

In this study, we describe the effect of the position of PE fiber on the mechanical properties of carbon fiber/PE fiber hybrid composites. We also evaluate the influence of improved adhesion of PE fiber on the hybrid properties through various surface treatments of PE fiber. In addition, we examine the relationship

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TABLE I Physical properties of carbon fiber, PE fiber, and vinylester resin

Physical properties	Carbon fiber	Spectra-900	Vinylester
Density (kg/m ³)	1760	970	1150
Tensile modulus (GPa)	235	117	3.71
Tensile strength (MPa)	2760	2500	63.30
Maximum strain (%)	0.75	3.50	6.30

between the surface treatment of PE fiber and the extent of performance improvement of each hybrid composite.

2. Experimentals

2.1. Materials

The carbon fabric used in this work was T300 plain weave type from Torayca, and the polyethylene (PE) fabric was Spectra-900 plain weave type from Allied Signal Co. Both fabrics were desized with methylene chloride for 3 days, then dried at 60 °C for 48 h in drying oven. The matrix resin was styrene-based XSR-10 vinylester resin supplied by National Synthesis Co. (Korea). Benzoyl peroxide (BPO) was added to the matrix resin as an initiator. The physical properties of carbon fiber, PE fiber, and vinylester resin are given in Table I. The tensile tests of each material were conducted at strain rate of 1 min⁻¹.

2.2. Fiber surface treatment

Low temperature oxygen plasma treatment was used to modify the chemical functionality and the topology on fabrics. The plasma output power was 100 W and the carrier gas flow rate was 10⁻⁵ m³/min. The plasma treatment time was fixed at 1 min. The plasma treated fabrics were treated with silane coupling agent and polybutadiene-modified silane. γ -Methacryloxypropyltrimethoxysilane (γ -MPS) from Petrach System was used as a silane coupling agent for surface treatment. Polybutadiene-modified γ -MPS (PB/ γ -MPS) was synthesized using azobisisobutyronitrile (AIBN) as initiator under benzene solvent. The mole ratio of polybutadiene to γ -MPS was 1 to 9 and the reaction was performed at 65 °C for 18 h under nitrogen atmosphere. γ -MPS and PB/ γ -MPS were prehydrolysed for 1 h in distilled water adjusted to pH 3.5 with acetic acid. The silane concentration was varied from 0 to 0.7 wt % and the plasma-treated fabrics were impregnated in the prehydrolysed silane solution for 10 min. The silane-treated fabrics were dried for 2 days in a hood at room temperature.

2.3. Prepreg preparation

The prepreg of hybrid composite was prepared using vinylester resin with 2 wt % BPO. Each fabric was well impregnated with a solution of this mixture in acetone by hand roller. The resin impregnated fabrics were aged for two days in a hood at room temperature for thickening of the resin.

2.4. Composite manufacturing

The composites were made using open leaky mold method. The eight ply composites were then cured in

a hot press for 20 min at 43 °C and 50 min at 90 °C at a pressure of 1000 psi. The thickness of hybrid composites was 2.0 mm approximately. The laminates were hybridized by interleaving plies of each fabric with a change of the stacking sequence. For interply hybrid laminates, 4 plies of carbon fabric and PE fabric were used respectively.

2.5. FT-IR analysis

PB/ γ -MPS was analysed by transmission technique in a Bomem MB-100 instrument with DTGS (Deuterated Triglycinsulfate) detector. The total 32 scans were coadded and a dry nitrogen purge was used to prevent the interference of water and CO₂.

2.6. Flexural properties

The flexural strength and modulus of the hybrid composites were measured using three point bending test according to the ASTM standard method D 790. The composite specimens of 50 × 20 mm were tested using a support span of 32 mm at a crosshead speed of 2 mm/min. The flexural strength and modulus of each sample was determined from data on 5 test specimens.

2.7. Interlaminar shear strength (ILSS)

The interfacial adhesion between fiber and matrix resin was measured by the short beam three point bending test (ASTM D2344). The width of the test specimen was 10 mm and the length was 14 mm. The span length was 10 mm and the crosshead speed for the test was 2 mm/min. The ILSS of each specimen was averaged with 5 values.

2.8. Scanning electron microscopy (SEM)

Scanning electron microscopy (SEM) was used to observe the fracture surface of hybrid composites. The instrument used in this study was Jeol JSM-35 and all specimens were coated with a thin layer of gold to eliminate charging effects.

3. Results and discussion

3.1. Carbon fiber/vinylester composite

Fig. 1 shows FT-IR transmission spectra of γ -MPS and PB/ γ -MPS. Fig. 1A is the spectrum of γ -MPS showing the characteristic peak at 1719 and 939 cm⁻¹. The peak at 1719 cm⁻¹ is assigned to carbonyl peak of methacrylate group which is conjugated with the adjacent double bond. In addition, the band at 939 cm⁻¹ is associated with the wagging mode of double bond within methacrylate group. Fig. 1B shows the change of peaks of PB/ γ -MPS compared with γ -MPS. The peak at 1728 cm⁻¹ originates from carbonyl group of PB/ γ -MPS and is shifted to higher wavenumber compared with γ -MPS. This is attributed to the fact that the double bond adjacent to carbonyl group disappears due to the reaction of PB with γ -MPS. The disappearance of conjugation enhances the electron density of carbonyl group and leads to higher frequency. In contrast with spectrum (A), the peak at 939 cm⁻¹ is not shown in spectrum (B). This indicates that the double bond of γ -MPS effectively reacts with that of PB. In

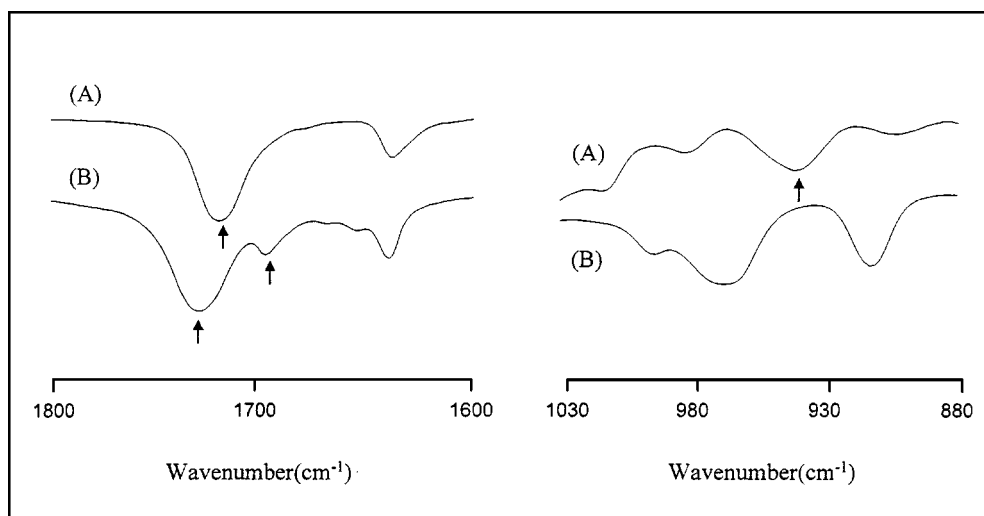


Figure 1 FT-IR transmission spectra of γ -MPS and PB/ γ -MPS: (A) γ -MPS, (B) PB/ γ -MPS.

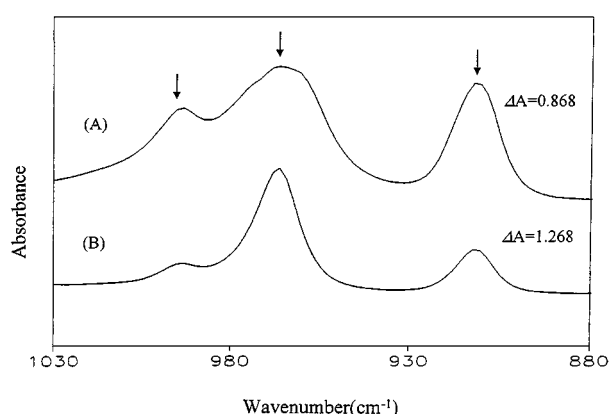


Figure 2 FT-IR absorbance spectra of PB and PB/ γ -MPS: (A) PB, (B) PB/ γ -MPS.

addition, the peak at 1696 cm^{-1} is due to the hydrogen bonding with water in air during measurement.

FT-IR absorbance spectra of PB and PB/ γ -MPS are shown in Fig. 2. Fig. 2A is the spectrum of PB and exhibits the characteristic peaks at 910 , 960 , and 990 cm^{-1} . The peak at 910 and 990 cm^{-1} is associated with CH_2 wagging and trans CH wagging of the vinyl group, respectively. In addition, the peak at 960 cm^{-1} is due to CH wagging of trans group. Fig. 2B is the spectrum of PB/ γ -MPS and shows the same peak position as spectrum (A). However, the intensity of each peak is different in two spectra. The peak intensity at 910 and 990 cm^{-1} decreases compared with spectrum (A). This suggests that the vinyl group of PB primarily reacts with the double bond of γ -MPS to form PB/ γ -MPS.

Fig. 3 shows the variation of flexural strength of carbon fiber/vinylester composite according to the concentration of γ -MPS and PB/ γ -MPS. The flexural strength shows a maximum value and then gradually decreases with further increase in the concentration. γ -MPS and PB/ γ -MPS act as the bridge to bond carbon fiber to the vinylester resin with the chain of primary bond. The difference in the two cases is the concentration at which the maximum flexural strength is shown. After the maximum value, the increase of the concentration leads to the formation of physisorbed layer and

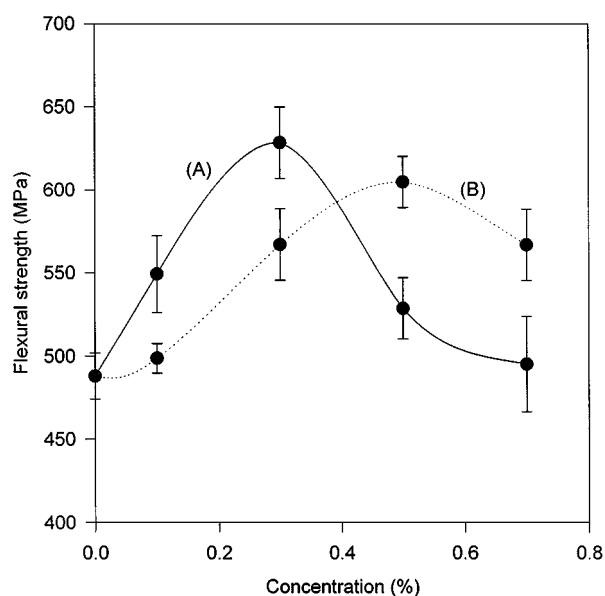


Figure 3 The variation of flexural strength of carbon fiber/vinylester composite according to the concentration of γ -MPS and PB/ γ -MPS: (A) γ -MPS, (B) PB/ γ -MPS.

the composite fails in this region through the slippage of physisorbed layer. Compared with γ -MPS, the long and flexible chains of PB/ γ -MPS lead to the formation of IPN (interpenetrating polymer network) with matrix even at the higher concentration. These results in the maximum value at the higher concentration in PB/ γ -MPS treated composites. In addition, the higher flexural strength is obtained in γ -MPS treated composite at lower concentration, but in PB/ γ -MPS treated composite at higher concentration. At the lower concentration, γ -MPS forms the covalent bonding with carbon fiber and the siloxane bonding occurs between γ -MPS chains. However, at the higher concentration γ -MPS treated composite fails easily due to lubricant action of physisorbed layer, whereas PB/ γ -MPS forms the entanglement between chains and IPN with matrix.

The flexural strength and ILSS of untreated and surface treated carbon fiber/vinylester composite are shown in Fig. 4. For plasma treated composite, the flexural strength and ILSS increase a little compared with

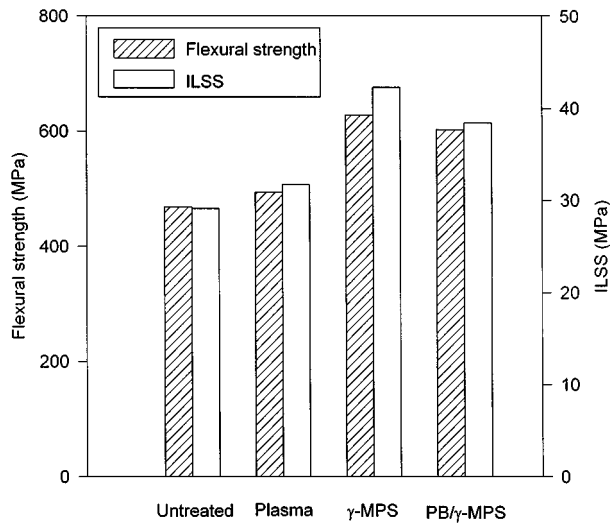


Figure 4 The flexural strength and ILSS of untreated and surface treated carbon fiber/vinylester composite.

untreated composite. This is attributed to the fact that although the functional groups such as hydroxyl and carbonyl groups are created on the surface of carbon fiber by plasma treatment, they cannot effectively form the chemical bonding with double bond of vinylester. On the other hand, γ -MPS and PB/ γ -MPS act as the bridge to bond carbon fiber to vinylester resin because they contain both hydroxyl groups and double bonds. In addition, the increment of ILSS is larger than that of flexural strength. ILSS is sensitive to the change of fiber-matrix interfacial property because it primarily depends on fiber-matrix interfacial strength rather than fiber and matrix property.

3.2. PE fiber/vinylester composite

Fig. 5 shows the variation of flexural strength of PE fiber/vinylester composite according to the concentration of γ -MPS and PB/ γ -MPS. In contrast with carbon fiber, γ -MPS treated composite exhibits higher flexural strength than PB/ γ -MPS treated composite at all

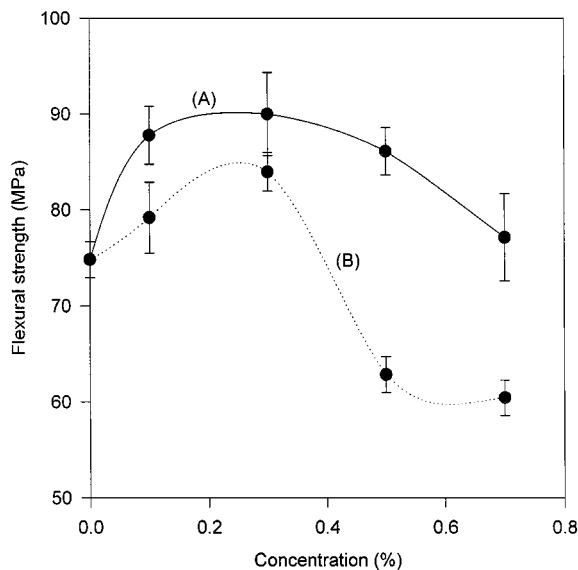


Figure 5 The variation of flexural strength of PE fiber/vinylester composite according to the concentration of γ -MPS and PB/ γ -MPS: (A) γ -MPS, (B) PB/ γ -MPS.

concentrations. At the plasma treatment of 1 min the functional group on PE fiber surface does not be introduced effectively, therefore γ -MPS and PB/ γ -MPS form physical bonding rather than chemical bonding with PE fiber [17]. Therefore, the polarity of coupling agent plays a role in the flexural property of the composites. PB/ γ -MPS has more hydrophobic characteristics than γ -MPS because the chain contains more double bond than silanol group. The chains are excluded from hydrophilic fiber surface, and PB/ γ -MPS treated composite forms the weak interface between fiber and matrix. In addition, at the higher concentration the composite exhibits much lower flexural strength than the untreated composite.

A representative comparison of untreated PE fiber with plasma treated fiber is shown in Fig. 6. The surface

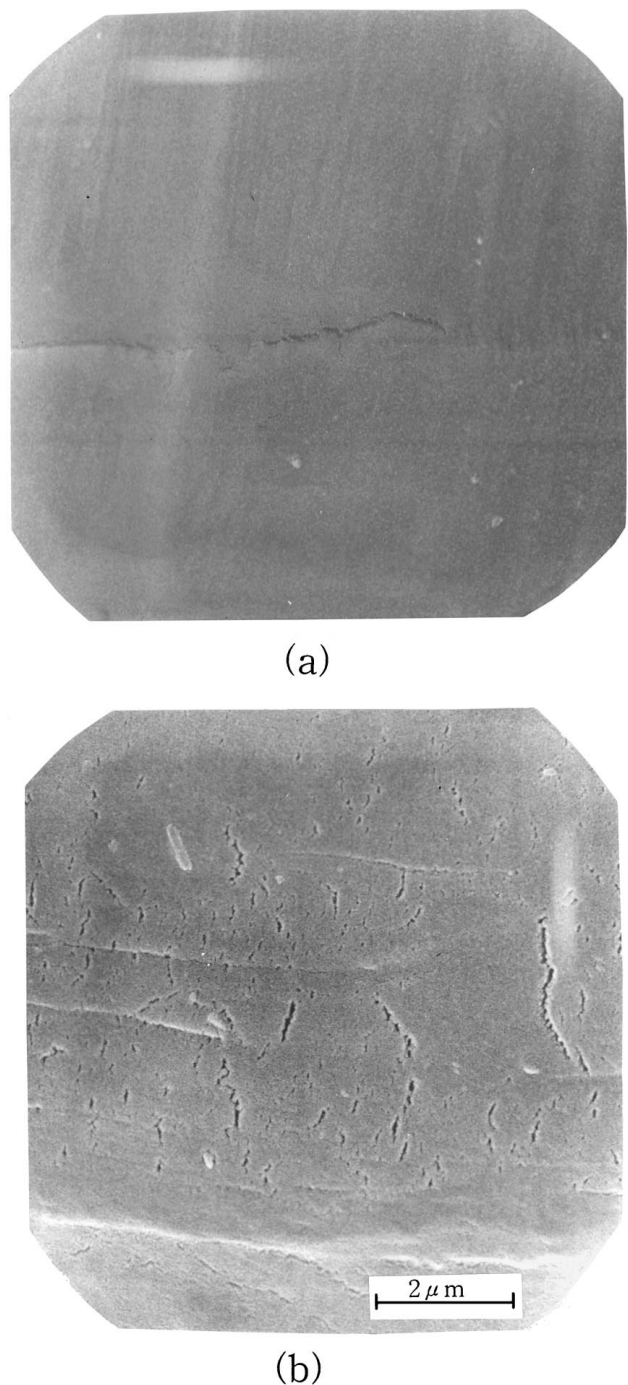


Figure 6 A representative comparison of untreated PE fiber with plasma treated PE fiber: (a) untreated, (b) plasma treated.

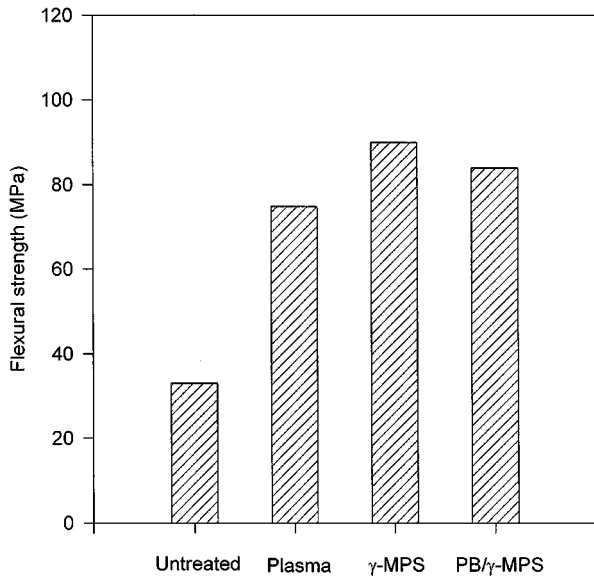


Figure 7 The flexural strength of untreated and surface treated PE fiber/vinylester composite.

of control monofilament reveals it to be quite smooth as shown in Fig. 6a. On the other hand, the oxygen plasma introduces micro-pitting on the fiber surface by etching, and this micro-pitting improves the adhesion between the PE fiber and coupling agent by mechanical interlocking.

Fig. 7 shows the flexural strength of untreated and surface treated PE fiber/vinylester composite. For plasma treated composite, the flexural strength increases considerably compared with untreated composite. However, in the case of γ -MPS and PB/ γ -MPS treated composite, the increment of flexural strength is low compared with plasma treated composite. This result indicates that the plasma treatment introduces etching and micro-pitting rather than functional group formation on the fiber surface. Therefore, plasma treatment followed by coupling agent leads to a little increment in flexural strength. If the plasma treatment primarily leads to the introduction of functional groups on fiber surface, the treatment with coupling agent results in higher flexural strength.

3.3. Carbon fiber/PE fiber hybrid composites

Table II summarizes the flexural strength and modulus of untreated carbon/PE interply hybrid composites with different stacking sequence. All laminates were loaded from the left side. In the flexural strength and modulus of hybrid composites, carbon fiber-only laminate exhibits the highest value, but PE fiber-only laminate shows the lowest value. Interply hybrid composites show a value between those for the two laminates. In the flexural strength of hybrid composites, when carbon fiber is at the outermost layer, the laminate (B1) shows the highest flexural strength. However, the composite (B7) with PE fiber at the skin region exhibits very low flexural strength. This is attributed to the fact that the compressive and tensile stress have maximum magnitudes in the outermost layer. Therefore, in the laminate B1, carbon fiber in the outermost layer bears most of applied load and this leads to higher flexural strength.

TABLE II The flexural strength and modulus of untreated carbon fiber/PE fiber interply hybrid composites with the different stacking sequence

Notation	Lamination sequence	Flexural strength (MPa)	Flexural modulus (GPa)
A	■ ■ ■ ■ ■ ■ ■ ■	467.9	31.2
B1	■ ■ □ □ □ □ ■ ■	205.1	7.0
B2	■ ■ ■ ■ □ □ □ □	188.3	6.6
B3	□ □ □ □ ■ ■ ■ ■	141.7	4.6
B4	■ ■ □ □ ■ ■ □ □	136.8	6.3
B5	■ □ ■ □ ■ □ ■ □	133.2	5.9
B6	□ □ ■ ■ □ □ ■ ■	131.2	6.1
B7	□ □ ■ ■ ■ ■ □ □	126.0	5.8
B8	□ ■ □ ■ □ ■ □ ■	124.1	4.2
C	□ □ □ □ □ □ □ □	33.04	1.2

■ Carbon fiber layer.
□ PE fiber layer.

In addition, carbon/vinylester and PE/vinylester composites are weaker in compression than in tension, and the failure is always in the compression mode. When carbon fiber is at the compressive side, the laminate (B2, B4, B5) exhibits higher flexural strength. The dispersion extent of fibers also has an effect on the flexural strength of interply hybrid composites. The dispersion extent of fibers means that how many same fabrics are neighbored or dispersed. The composites with large dispersion extent (B5, B6, B8) produce much residual stress between different fabric layers, and easy failure of the composite occurs in this layer. The flexural modulus also exhibits similar behavior to the flexural strength. However, the flexural modulus relates to the initial slope in the load-displacement curve on flexural loading and is controlled by the property of the compressive side. Therefore, the laminates with carbon fiber at the compressive side (B1, B2, B4) show the higher flexural modulus, whereas the composites with PE fiber at compressive side (B3, B6, B7) exhibit the lower value.

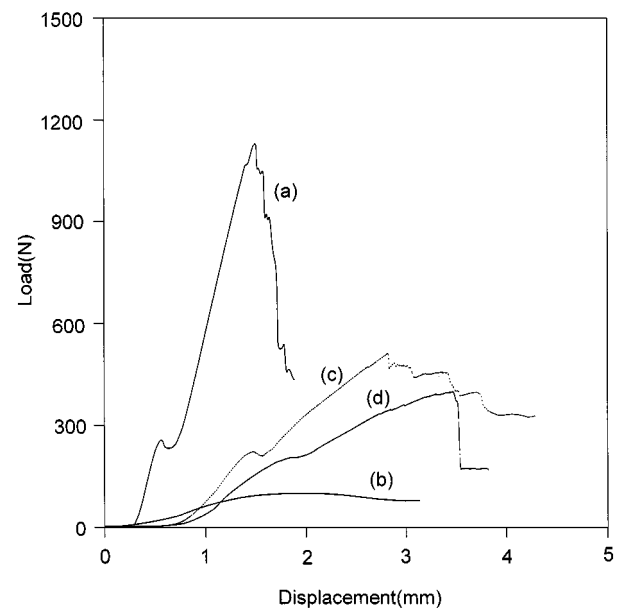


Figure 8 The load-displacement curves of carbon fiber-only, PE fiber-only, and interply hybrid composites: (a) carbon fiber-only, (b) PE fiber-only, (c) laminate B2, (d) laminate B3.

Fig. 8 shows the load-displacement curves of carbon fiber-only, PE fiber-only, and interply hybrid composites. The carbon fiber-only composite shows a rapid load rise, the highest maximum load, and catastrophic failure, indicating that this composite fails in a brittle manner. The PE fiber-only composite shows slow load rise, high yield displacement, and the lowest maximum load. These behaviors suggest that the composite fails in a ductile manner because of the high elongation property of PE fiber. The interply hybrid composites B2 and B3 follow the behavior of both carbon fiber-only and PE fiber-only composite. The initial slope of laminate

B2 is higher than that of laminate B3. This suggests that the early part of the flexural response is controlled by the property of the compressive side. After the maximum load, the load drop of composite B2 is gradual but that of laminate B3 is steep. This result indicates that the latter part of flexural response mainly depends on the property of the tension side.

The flexural strength of carbon/PE interply hybrid composites with surface treated PE fiber is represented in Table III. The plasma treatment time was 1 min and the treatment concentration of silane coupling agents was fixed at 0.3 wt %. The surface treated composites

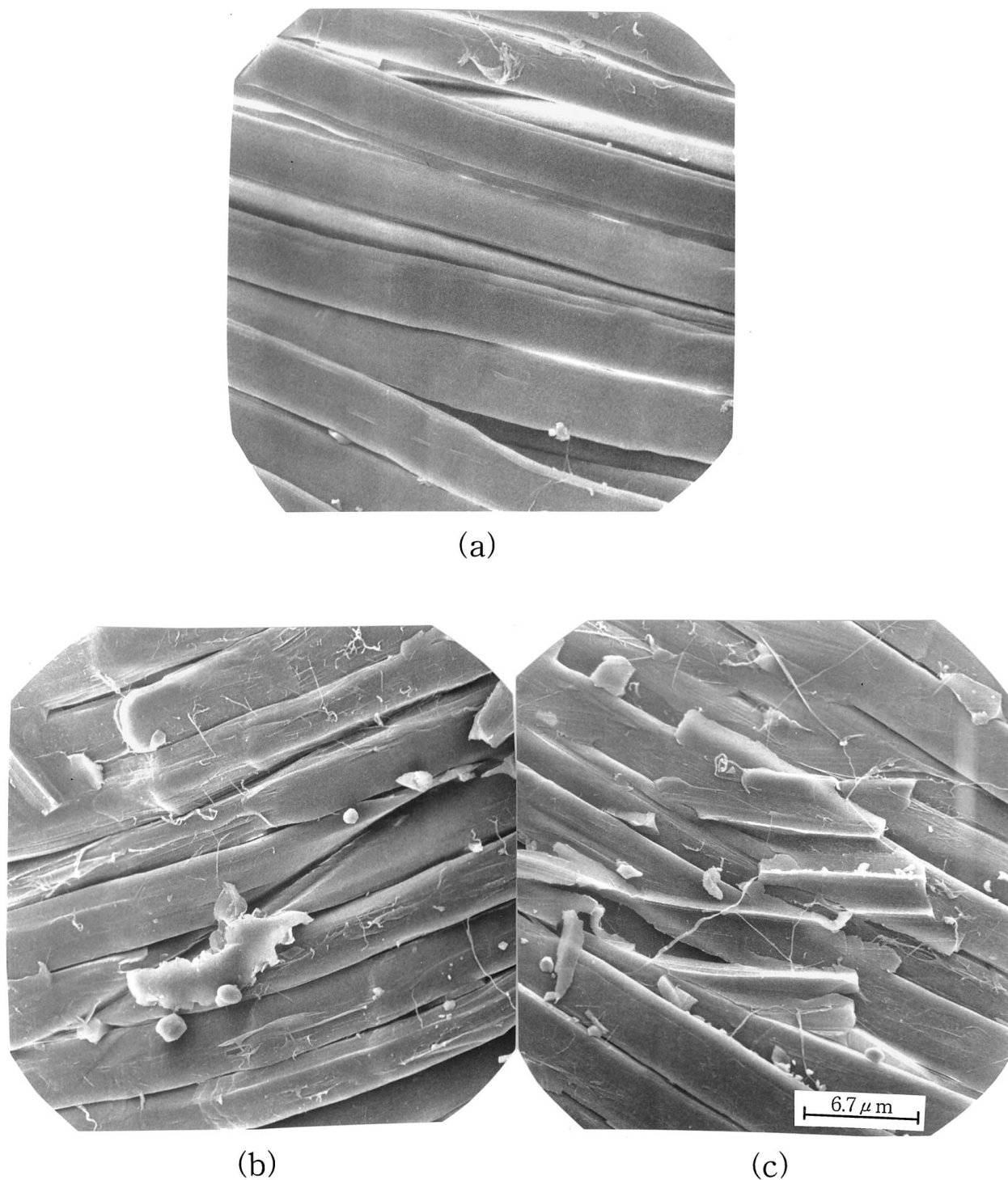


Figure 9 The fracture surface in the compressive side of hybrid composite B7 with the surface treatment of PE fiber: (a) untreated, (b) γ -MPS treated, (c) PB/ γ -MPS treated.

TABLE III The flexural strength of carbon fiber/PE fiber interply hybrid composites with surface treatment of PE fiber

Notation	Lamination sequence	Flexural strength (MPa)		
		Untreated	γ -MPS	PB/ γ -MPS
A	■ ■ ■ ■ ■ ■ ■ ■	467.9	628.4	602.9
B1	■ ■ □ □ □ □ ■ ■	205.1	217.3	207.8
B2	■ ■ ■ ■ □ □ □ □	188.3	247.9	192.5
B3	□ □ □ □ ■ ■ ■ ■	141.7	189.5	163.4
B7	□ □ ■ ■ ■ ■ □ □	126.0	202.4	168.2
C	□ □ □ □ □ □ □ □	33.04	90.0	83.9

■ Carbon fiber layer.
□ PE fiber layer.

show higher flexural strength than untreated composites and γ -MPS treated composite exhibits a higher value than PB/ γ -MPS treated composite. However, each laminate shows much difference in extent of increment of flexural strength. In the composite with carbon fiber at the outermost layer (B1), the effect of surface treatment on the flexural strength of hybrids is negligible. This is due to the dependence of flexural strength on the property of the outermost layer. Therefore, the surface treatment of PE fiber at the core region has little effect on the flexural strength of the composite. However, the laminate with PE fiber at the outermost layer (B7) shows a large increase in the flexural strength. Surface treatment of PE fiber leads to an increase of stress at the compressive and tensile layers and allows the composite to bear a higher load.

Fig. 9 shows the fracture surface in the compressive side of hybrid composite B7 with surface treated PE fiber. The fracture surface of untreated composite shows clean fibre/matrix separation with little fibre and matrix damage, indicating that the composite fails adhesively at the fiber-matrix interface. The fracture surface of γ -MPS treated composite shows some fiber fibrillations with a little matrix damage. Fiber fibrillation and

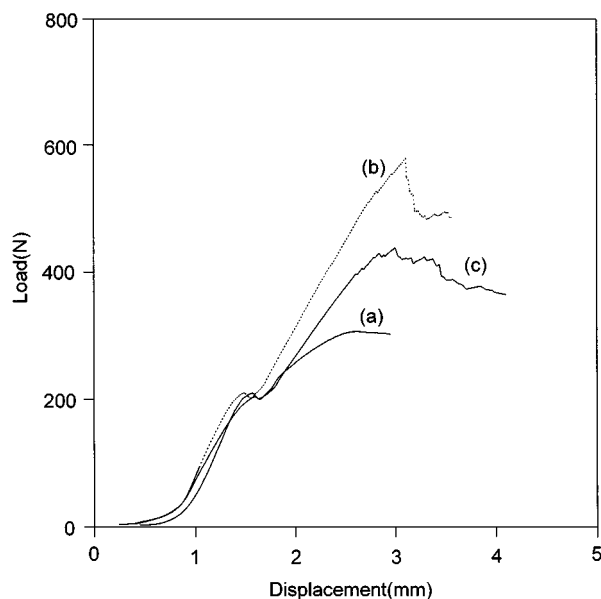


Figure 10 The load-displacement curves of hybrid composite B7 with the surface treatment of PE fiber: (a) untreated, (b) γ -MPS treated, (c) PB/ γ -MPS treated.

splitting result from cohesive failure within fiber bulk region, which contributes to the increase of the flexural strength. For PB/ γ -MPS treated composite, the fracture surface shows the reduced fiber fibrils and clean matrix region. This result indicates that the composite fails more or less adhesively at the PE fibre/matrix interface.

The load-displacement curves of hybrid composite (B7) with surface treated PE fiber are shown in Fig. 10. Untreated and surface treated composites show a similar initial slope of curve but different maximum loads. The surface treatment of PE fiber improves the load-bearing capability of the composite at the compressive and tension side. In addition, after the maximum load, the load drops more or less steeply due to improved adhesion between fiber and matrix.

4. Conclusions

The effect of the position of PE fiber and the adhesion level of PE fiber on the mechanical properties of carbon fiber/PE fiber hybrid composites has been examined. In the case of carbon fiber/vinylester composites, plasma treatment showed a small increment in flexural strength and ILSS compared with the untreated case. This was due to the absence of chemical bonding of the functional group of carbon fiber with matrix resin. γ -MPS and PB/ γ -MPS acted as the bridge to bond carbon fiber to vinylester resin through the chemical bonding. In the case of PE fiber/vinylester composites, plasma treated composite exhibited a large increase in flexural strength compared with the untreated composite. This was attributed to the fact that the plasma treatment of PE fiber introduced etching and micro-pitting rather than producing functional groups on the fiber surface. In the case of carbon fiber/PE fiber hybrid composites, the mechanical properties of the hybrid composite strongly depended on the reinforcing fiber position. When carbon fiber was at the outermost layer, the composite showed the highest flexural strength. The flexural modulus related to the initial slope in the load-displacement curve on flexural loading and was controlled by the property of the compressive side. The surface treatment of PE fiber lead to considerable difference in extent of increment of flexural strength in each laminate. The surface treatment of PE fiber at the outermost layer had a major effect on the flexural strength of the composite.

Acknowledgements

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