Effect of Fiber Pretreatment Condition on the Interfacial Strength and Mechanical Properties of Wood Fiber/PP Composites

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ABSTRACT: The effect of fiber surface pretreatment on the interfacial strength and mechanical properties of wood fiber/polypropylene (WF/PP) composites are investigated. The results demonstrate that fiber surface conditions significantly influence the fiber–matrix interfacial bond, which, in turn, determines the mechanical properties of the composites. The WF/PP composite containing fibers pretreated with an acid–silane aqueous solution exhibits the highest tensile properties among the materials studied. This observation is a direct result of the strong interfacial bond caused by the acid/water condition used in the fiber pretreatment. Evidence from coupling chemistry, rheological and electron microscopic studies support the above conclusion. When SEBS-g-MA copolymer is used, a synergistic toughening effect between the wood fiber and the copolymer is observed. The V-notch Charpy impact strength of the WF/PP/SEBS-g-MA composite is substantially higher than that of the WF/PP composite. The synergistic toughening mechanisms are discussed with respect to the interfacial bond strength, fiber-matrix debonding, and matrix plastic deformation. © 2000 John Wiley & Sons, Inc. J Appl Polym Sci 76: 1000–1010, 2000

Key words: wood fiber/polypropylene composite; interfacial adhesion; impact strength

INTRODUCTION

In recent years, significant efforts have been made to manufacture thermoplastic composites using such natural fibers as wood sawdust, wheat straw, nut shell fiber, and jute fiber.[1–4] The rationale behind these efforts is that the use of natural fibers offers several benefits, including low cost, high specific properties, renewable nature, and biodegradability. Wood fiber is one of the most abundant, low-cost materials among the natural fibers; therefore, it has attracted much research interest. Attempts have been made with varied success to use wood fiber as reinforcement for

thermoplastic polymers; for example, polyethylene, polypropylene, polystyrene, and poly(vinyl chloride). Possible relationships between the structure and properties of the wood fiber/thermoplastic composites were proposed.[5–8] Many major issues have been identified in the processing of the composites.[9,10] These include low compatibility between the wood fiber and polymer matrix; instability of the fiber at temperatures above 200°C; high moisture intake, and poor dispersion of the fiber in polymer melt. In particular, the low compatibility between the hydrophilic wood fiber and hydrophobic polymer matrix is one of the major reasons for limited use of wood fiber as reinforcement. In many cases, poor mechanical and physical properties of a wood fiber composite can be attributed to the weak fiber–matrix interfacial bond caused by the low compatibility. Obviously, how to improve the compatibility between the two components is a key to success in the area.

To increase the fiber–matrix compatibility, both matrix resin modification and fiber surface treatment have been considered. For example, maleic anhydride modified polyolefins and such copolymers as styrene butadiene styrene butadiene styrene (SBS) were employed in the preparation of wood fiber composites. It was reported that the fiber–matrix interfacial adhesion increased, and the mechanical properties of the composites improved after matrix modification.[11,12] On the other hand, pretreatment of wood fibers to alter the chemical nature of the fiber surface, instead of matrix modification, has been considered to be a more promising means of compatibilization.[13] Coutinho and co-authors attempted to coat wood fibers with polypropylene through *in situ* propylene polymerization and reported that the adhesion between wood fiber and polypropylene matrix was improved.[14] In addition to coating with polymer, pretreatment of wood fibers using low molecular weight coupling agents, such as silane coupling agents, has been regarded as a more convenient and efficient way of enhancing the fiber–matrix adhesion. Several different types of silane coupling agents have been employed in previous studies, focusing on the effect of silane content on the physical and mechanical properties of wood fiber/polypropylene composites.[14–16]

According to the principles of interface coupling, the hydrophilic group of a silane coupling agent is expected to react chemically with the functional groups on the wood fiber surface, and

the hydrophobic group should react or have relatively high compatibility with the polymer matrix. The combined effects of these interactions will effectively improve the compatibility of the fibers and matrix. However, to ensure successful coupling, the chemical reaction should occur under controlled conditions. The extent and rate of the chemical reaction may greatly affect the compatibility; hence, the study of these factors is warranted.

In the present study, a silane coupling agent, vinyl-trimethoxy silane, was employed in the pretreatment of wood fiber to improve the compatibility of the wood fiber and PP matrix. Four different pretreatment methods were used to determine the optimized conditions for the fiber pretreatment. The effects of fiber pretreatment conditions on the fiber–matrix adhesion were studied based upon rheological measurements and electron microscopy. A styrene-ethylene-butylene-styrene copolymer grafted with maleic anhydride (SEBS-g-MA) was also employed as a compatibilizer in the composite preparation. The mechanical properties and the rheological behavior of the composites were investigated to determine the relationships among the fiber pretreatment condition, interfacial characteristics, and mechanical properties of wood fiber reinforced polymer materials.

EXPERIMENTAL

Materials and Fiber Treatment

Materials

The polymer matrix used in this study was a commercially available polypropylene (PP), Cosmoplene PP Y101. It has an MFI of 12 g/10 min and a density of 0.9 g/cm^3. Wood fibers (WF) in the form of sawdust were obtained from an indoor workshop. The sawdust was ground into wood fiber and surface treated under different conditions before it was compounded with the PP matrix. Details of the pretreatment condition are described in the following section. Two modifiers were employed to improve the interfacial adhesion between the WF and the PP matrix. One was a vinyl-trimethoxy silane coupling agent (supplied by Union Carbide) and the other was a maleic anhydride (MA) grafted styrene-ethylene-butylene-styrene (SEBS) triblock copolymer (Kraton FG1901X obtained from Shell Chemical Co.). Acetic acid was used in the preparation of the aque-

Table I   Physical Properties of the Wood Fibers

Fiber Code	Moisture Content (% by Weight)	Particle Distribution (% by Weight)			
		<150 μm	150–250 μm	250–300 μm	>300 μm
WF-N	3.80	12.1	31.2	56.4	0.3
WF-D	3.81	12.9	31.3	55.6	0.2
WF-G	3.92	12.6	31.4	55.8	0.2
WF-A	3.93	12.1	31.2	56.4	0.3

ous silane solution to obtain a desired pH value, which was required for chemical reactions among WF, PP, and silane.

Wood Fiber Pretreatment

The sawdust from our workshop was first sieved with a 500-μm sieve. Wood particles smaller than 500 μm were collected and dried at 60°C for 48 h in a vacuum oven. The dried wood particles were subsequently ground into wood fibers using a high-speed grinder for about 1 h and 40 min. The wood fibers were then treated with the silane agent; that is, vinyl-trimethoxy silane, under different conditions. In total, four kinds of wood fibers were obtained after the pretreatment. The designations and pretreatment conditions are listed below.

1. WF-N is the plain wood fiber obtained after grinding without silane coupling agent treatment. It was used as the control in the present study.
2. WF-D is the wood fiber obtained by physically mixing the dry WF-N fibers with 2 wt % vinyl-trimethoxy silane using a mixer. This is the traditional method for surface treatment of wood fibers.
3. WF-G is the wood fiber obtained by grinding the sieved dry sawdust particles and 2 wt % vinyl-trimethoxy silane simultaneously using a high-speed grinder for 1 h and 40 min. It was hoped that the new fiber surface generated during grinding would be able to contact and react with the silane coupling agent *in situ,* resulting in a higher coupling efficiency.
4. WF-A is the wood fiber obtained by mixing 100 parts WF-N fibers with 20 parts coupling agent solution. The coupling agent solution consisted of 10% vinyl-trimethoxy silane and 90% distilled water by weight.

The acidity of the coupling solution was adjusted using acetic acid. The pH value of the solution was around 4 to 5.

After the surface treatment, all wood fibers were dried in an oven at 60°C for 48 h before being blended with PP. The moisture content and size distribution of the wood fibers are shown in Table I.

Sample Preparation

The wood fibers and PP were compounded using a WP ZSK-30 co-rotating intermeshing twin-screw extruder with kneading blocks. The temperatures of the five processing zones were 180°C, 185°C, 190°C, 190°C and 190°C, respectively. The extrusion speed was 22 to 25 rpm. The resident time of the wood fibers and PP inside the extruder was approximately 2 to 3 min. No thermal degradation of the wood fiber was observed within this residence time at the above processing temperatures. The WF/PP extrudates were quenched in a cold water bath and then pelletized into granules. After the granules were dried in an oven for 2 h at 80°C, they were injection molded into standard specimens for mechanical tests. All samples were conditioned at room temperature for 48 h before testing. Because the major interest of the present study was to investigate the effect of fiber surface conditions on the properties of the composites, the content of fibers was kept constant at 10 wt %.

In the preparation of the WF/PP/SEBS-g-MA composite, wood fibers, PP, and SEBS-g-MA copolymer pellets at 10 : 10 : 80 weight ratio were mechanically mixed using a mixer at room temperature. The mixture of the dry materials was then compounded using the same extruder under the same processing condition as described above. The resultant composites are designated as WF-x/PP/SEBS-g-MA, where x is N, D, G, or A, representing the fiber pretreatment conditions applied.
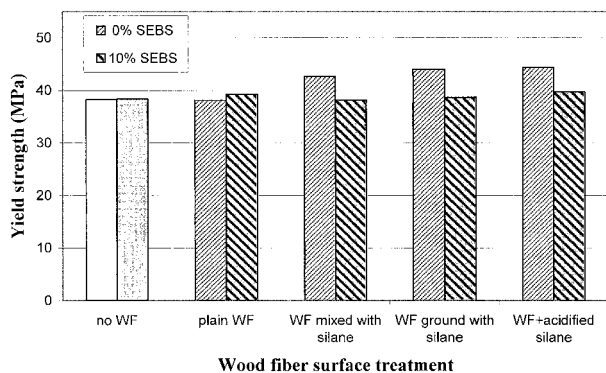
Figure 1 Variation of yield strength against wood fiber surface treatment conditions for the WF/PP and WF/PP/SEBS composites.

Mechanical Property Test

The tensile properties of the composites were measured using a Sintech D/10 Universal Tensile Machine at a crosshead speed of 50 mm/min according to the standard ASTM D638. The impact strength was measured on a Charpy impact test machine following the standard ASTM D 256. The data reported in the present paper are the mean values of at least seven measurements. The rheological properties of the composites were measured using an RDS-II rheometer with a parallel-plate fixture at 190°C. The diameter of the plates was 25 mm, and the dynamic oscillatory mode was adopted. Complex viscosity (η^*) was measured as a function of frequency at a constant strain of 5%.

The fracture surfaces of the composites obtained from mechanical tests were examined using a JEOL 6300 scanning electron microscope (SEM). A thin layer of gold approximately 100 Å was coated on the fractured surfaces for good conduction before SEM examination.

RESULTS AND DISCUSSION

Wood Fiber/PP Composites

Mechanical Properties

The tensile properties of the WF/PP composites are shown in Figures 1 to 3 by the light striped columns. In general, the WF/PP composites had a higher tensile modulus, yield stress, and tensile strength than the pure PP (the empty column) or the PP/SEBS-g-MA blends (the solid column). The effect of fiber pretreatment condition on the ten-
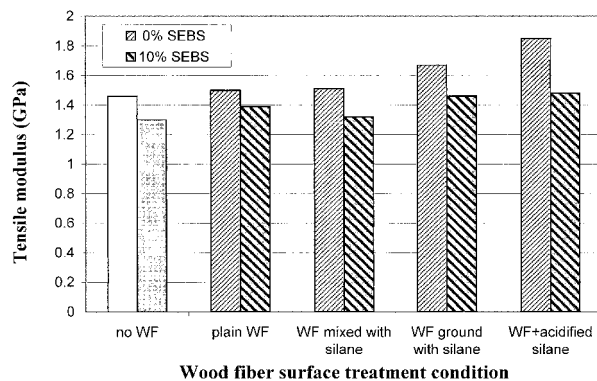

Figure 2 Variation of tensile modulus against wood fiber surface treatment conditions for the WF/PP and WF/PP/SEBS composites.

sile properties of the composites is apparent. Among the four different composites without SEBS-g-MA, the one containing the acid–silane solution pretreated fibers; that is, the WF-A/PP composite, had the highest modulus (1.85 GPa), yield stress (44.4 MPa) and tensile strength (40.7 MPa). It is followed by the WF-G/PP composite and then the WF-D/PP composite. The tensile properties of the composite containing plain wood fibers (i.e., WF-N/PP composite) were the lowest and very close to those of the pure PP. These results suggest that the tensile properties of the WF/PP composites are closely related to the fiber pretreatment conditions. The strengthening effect of the wood fiber can only be realized when they are treated by the coupling agent under proper pretreatment conditions.

Coupling Chemistry of Silane Agent

According to the interfacial adhesion mechanisms reported for fiber/polymer composites,[17–21] silane
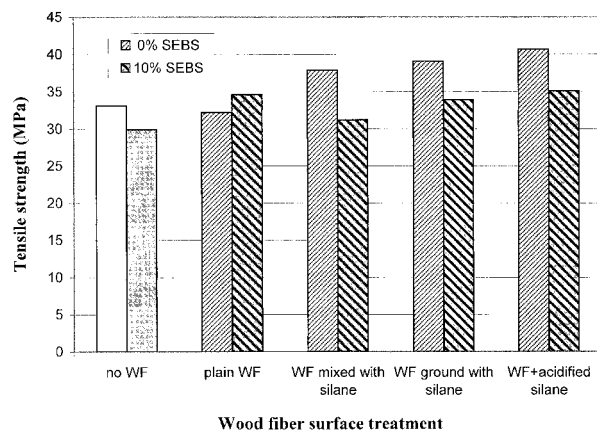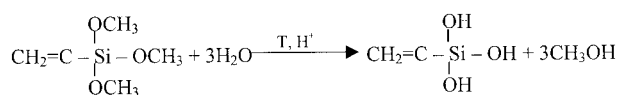

Figure 3 Variation of tensile strength against wood fiber surface treatment conditions for the WF/PP and WF/PP/SEBS composites.

coupling agents improve the adhesion by forming a thin interfacial layer that bridges the two components. The bridging effect of the interfacial layer is achieved through chemical and physical interactions among the coupling layer, fiber, and polymer matrix. In the present study, the vinyl-trimethoxy silane was used to establish such a coupling layer between the wood fibers and PP matrix. Based on the molecular structure of the silane coupling agent, it is known that the methoxyl group (CH_3O) of the silane can be easily turned into a hydroxyl group under proper conditions by the following hydrolysis reaction (Scheme 1):

$$CH_2{=}C\underset{\underset{OCH_3}{|}}{\overset{\overset{OCH_3}{|}}{-}}Si-OCH_3 + 3H_2O \xrightarrow{T,\ H^+} CH_2{=}C\underset{\underset{OH}{|}}{\overset{\overset{OH}{|}}{-}}Si-OH + 3CH_3OH$$

Scheme 1

The resultant hydroxyl group is hydrophilic in nature, therefore, the fiber surface is easily wetted by the hydrolyzed silane. Moreover, the hydrolyzed silane is able to react with the hydroxyl groups on the wood fiber surface to form strong chemical bonds through the following dehydration process (Scheme 2):
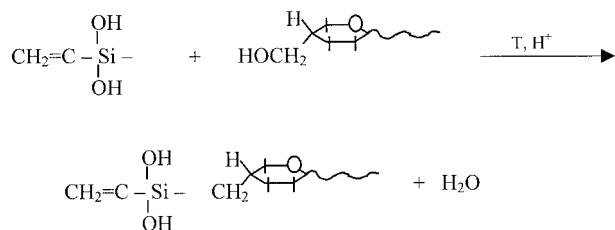
Scheme 2

Strong chemical bonding between the wood fiber and the silane layer will be established by these reactions.

At the other end of the silane agent, the hydrophobic vinyl group ($CH_2{=}C$) has a relatively high miscibility with the hydrophobic PP matrix. Moreover, because of the double bond in the vinyl group, some covalent bonds between the silane and the PP may form during extrusion. This is because the mechanical scissoring force and the high temperature in the extrusion process may break certain PP chains and create some polymeric free radicals. The polymeric radicals are capable of opening the double bond in the vinyl group of the silane to form a strong covalent bond with the silane. If this process occurs, strong

chemical bonding between the coupling layer and the PP matrix will be established.

However, it must be emphasized here that the hydrolysis of the methoxyl group must be initiated and completed in the presence of water. In addition, both the hydrolysis and dehydration reactions should be carried out in an acid environment, because the speed and proportion of these two chemical reactions are low under a neutral condition.[22] Because the wood fibers used in the present study had been thoroughly dried before being mixed with the silane, the hydrolysis of the methoxyl group should not occur during the dry mixing. Thus, the fiber–matrix interface bond of the WF-D/PP and WF-G/PP composites was most likely to be weak.

For the same reasons, the fiber–matrix interfacial bonds of the WF-A/PP composite was strong. When preparing the WF-A fibers, the silane coupling agent was first dissolved in water with a suitable amount of acid, as described previously. Thus, the hydrolysis of the methoxyl group of the silane occurred under an acidic condition and was catalyzed by the acid. When the fiber was treated with the hydrolyzed silane agent, wetting was easy, uniform, and complete. Dehydration between the wood fiber and the silane was likely to take place after the solution treatment. A covalent bond between the WF-A fiber and PP could be established during extrusion leading to a strong fiber–matrix interfacial bonding. The good tensile properties of the WF-A/PP composite are a direct reflection of the strong interfacial bonding produced by the acid-silane pretreatment.

Rheology and Microscopy

The above conclusion is supported by the results from the rheological and microscopic studies. As demonstrated in Figure 4, the complex viscosity of the WF-A/PP composite is the highest among all WF/PP composites in the entire frequency range. Because the complex viscosity represents the viscoelastic resistance of the composite melts during flow, a high viscosity implies a strong fiber–matrix interaction.[23,24] Thus, the observed high complex viscosity of the WF-A/PP composite suggests that the WF-A fibers have a strong interfacial bonding with the PP matrix.

Meanwhile, the complex viscosity of the WF-D/PP or WF-G/PP composites, which was lower than that of the WF-N/PP composite in the low frequency range (<10 rad/s), implied that the in-
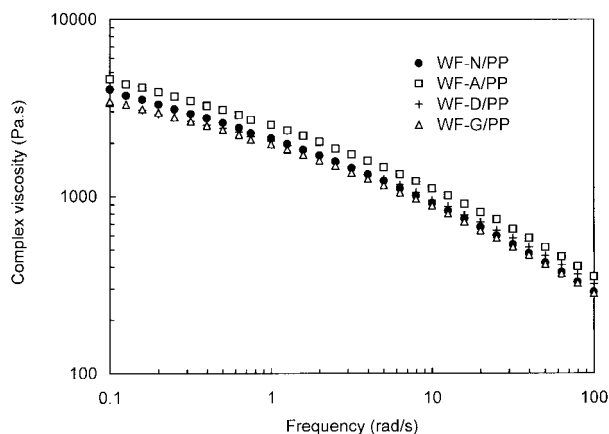
Figure 4 Variation of complex viscosity against frequency for the WF/PP composites with different fiber surface treatment.

teractions between the silane coating and the WF-D and WF-G fibers were weak. It is well known that during the shear flow of composite melt, wood fibers tend to move and align in the direction of the applied shear force. Among many factors affecting the viscosity of the melt, the friction caused by the relative movement of the fibers and matrix during the fiber alignment makes a significant contribution to the viscosity. Because the fibers in the WF-N/PP composite received no pretreatment, the fiber surface was rough. When being mixed with the polymer, the rough fiber surface would have a relatively strong mechanical interaction with the matrix, resulting in a high complex viscosity. It is proposed here that when the wood fibers were treated with the silane coupling agent, the silane coating had mutually counteracting effects on the complex viscosity. If the silane coating formed strong bonds with both the fiber and matrix, as in the WF-A/PP composite, it would increase the viscosity. In contrast, however, if the bonding between the silane and fiber or PP was weak, the silane coating could act as a lubricant reducing the friction between the fiber and matrix, leading to a relatively low viscosity. Based on this argument, the observed low viscosity of WF-D/PP and WF-G/PP can be inferred as a weak interaction between the silane coating and wood fiber. This indicates that the anticipated interfacial modification cannot be achieved by simply mixing the silane coupling agent with wood fibers.

In the higher frequency range (10–1000 rad/s), there was no significant difference in the complex viscosity among the WF-N/PP, WF-D/PP, and

WF-G/PP composites. This is attributed to the improved alignment of the PP chains and formation of a thin "dead-layer" of PP on the WF-N fiber surface at a high shear rate.[25,26] It appears that the relative movement of WF-N fiber and matrix occurred at the interface between the "dead-layer" of PP and matrix at a high shear rate. The "dead-layer" acted as a lubricant and reduced the friction and the complex viscosity of the WF-N/PP composite.

The effect of fiber surface treatment on interfacial adhesion was further studied using SEM. Figure 5 indicates PP particles adhering onto the wood fibers in the WF-A/PP composite, implying strong interfacial bonding and cohesive failure within the PP matrix. However, because the interfacial bond was very strong, the matrix tended to fail in a rather brittle mode. The mechanisms of this interface-related brittle failure are discussed in detail below. On the other hand, the fractured surface of the WF-D/PP composite, as shown in Figure 6a, shows that some wood fibers were pulled out. The magnified view illustrated in Figure 6b shows that the pull-out process involves little plastic deformation of the surrounding matrix. As indicated by the arrows in Figures 6b and c, there are, in fact, gaps between the wood fibers and matrix, suggesting that the fiber–matrix interface is, indeed, poor.

Wood Fiber/PP/SEBS Composites

Mechanical Properties

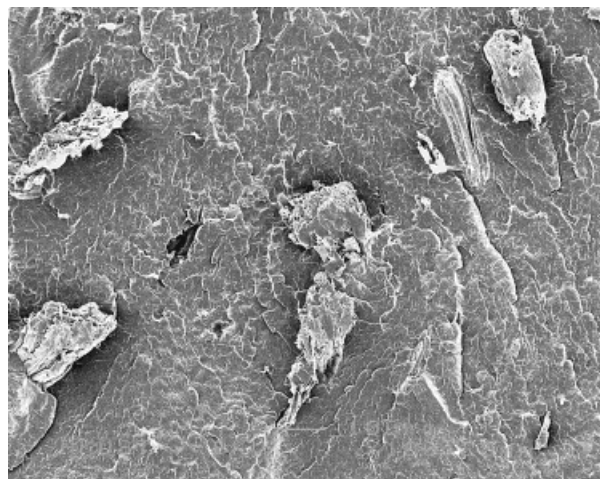When SEBS-g-MA copolymer was employed, the tensile properties of the PP/SEBS-g-MA blend
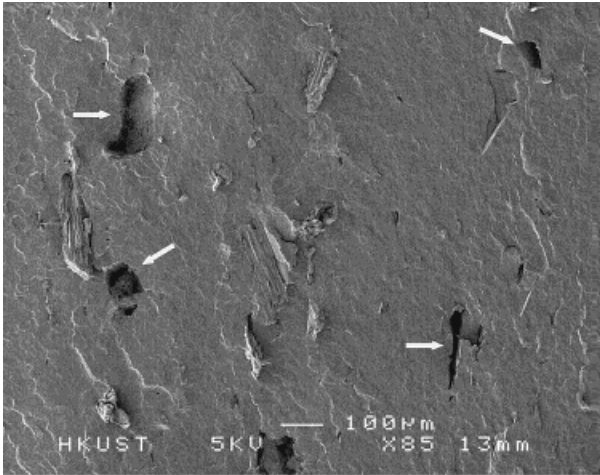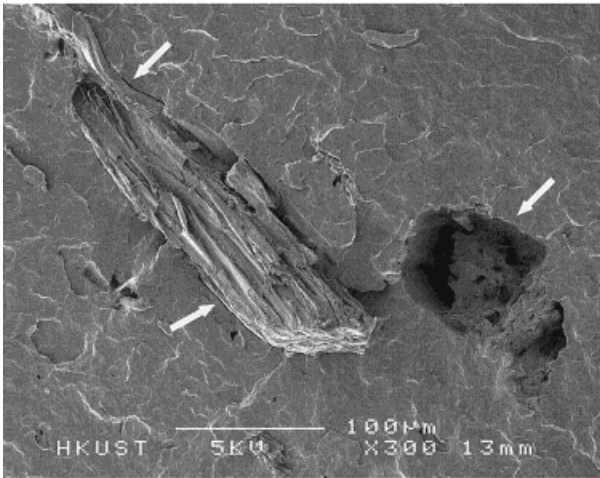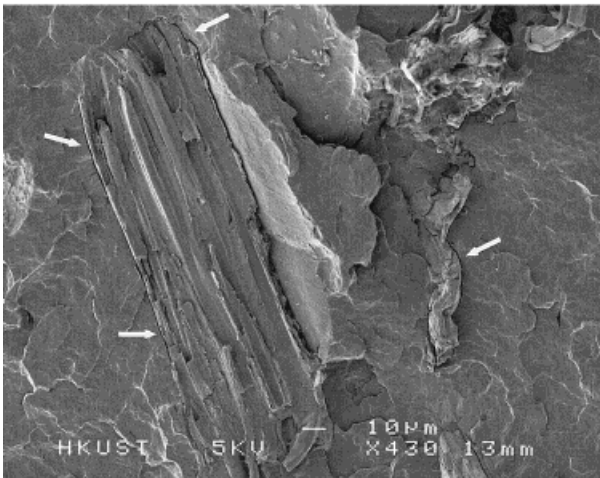

Figure 5 SEM of a fractured surface of the WF-A/PP composite.

(a)

(b)

(c)

Figure 6 SEM of a fractured surface of the WF-D/PP composite.
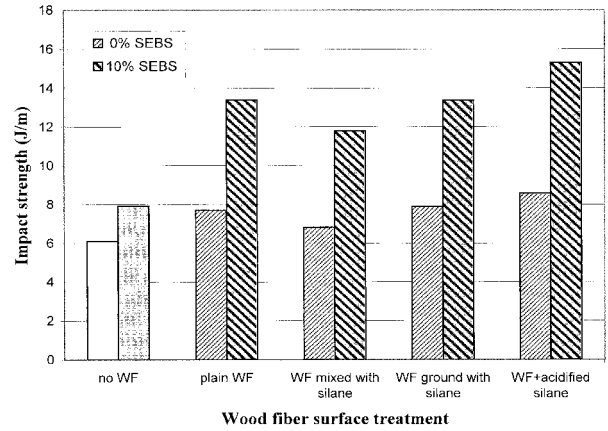

Figure 7 Variation of impact strength against wood fiber surface treatment conditions for the WF/PP and WF/PP/SEBS-g-MA composites. Synergistic toughening effect between wood fiber and SEBS-g-MA copolymer is clearly seen.

were lower than those of the pure PP. Addition of wood fibers into the PP/SEBS-g-MA blend improved the tensile properties only slightly. The strengthening effect of the wood fiber merely compensates the softening effect of the SEBS-g-MA copolymer. The tensile properties of the WF/PP/SEBS-g-MA composites are essentially the same as that of the pure PP, as shown by the dark striped columns in Figures 1 to 3.

The major benefit of adding SEBS-g-MA in the WF/PP composites was the substantial improvement in the V-notch Charpy impact strength of the WF/PP/SEBS-g-MA composites. The use of SEBS-g-MA alone to modify the PP improved its impact strength only marginally from 6.1 J/m to 7.93 J/m, refer to the dark solid column in Figure 7. Similarly, mixing wood fibers with different surface treatments alone with the PP matrix provided slight improvement in its impact strength. As shown by the light striped columns, the impact strength of the WF/PP composites fluctuated between 6.1 J/m and 8.5 J/m, depending upon the fiber surface treatment method used. However, when both the wood fiber and SEBS-g-MA copolymer were added into the composites, a synergistic toughening effect attributable to the presence of both constituents was achieved. For a given fiber pretreatment condition, the WF/PP/SEBS-g-MA composite had a much higher impact strength than the WF/PP counterpart (refer to the dark striped columns in Fig. 7). For example, the impact strength of the WF-N/PP/SEBS-g-MA composite is 13.39 J/m; whereas that of the WF-

N/PP is only 7.7 J/m. Again, the acid-silane solution treated fiber composite (WF-A/PP/SEBS-g-MA) has the highest impact strength ($\sim$ 15.31 J/m) among all variations studied. This value is significantly higher than the impact strength of pure PP (6.1 J/m), PP/SEBS-g-MA blend (7.93 J/m) and WF-A/PP composite (8.57 J/m). Considering that the tensile properties of the WF/PP/SEBS-g-MA composites are higher than that of the PP/SEBS-g-MA blend and comparable to that of the pure PP, the mechanical integrity of the WF/PP/SEBS-g-MA composites is, therefore, superior to PP, WF/PP, or PP/SEBS-g-MA. Although a similar synergistic toughening effect attributable to SEBS-g-MA copolymer and glass beads in a PP-based composite was reported recently,[27] the synergistic toughening attributable to wood fiber and SEBS-g-MA has never been reported, to our knowledge. Therefore, further discussion on the possible mechanisms behind the phenomenon is made as follows.

Toughening Mechanisms in WF/PP/SEBS Composites

It is now well established that the fracture toughness of polymer composites arises from various sources of energy dissipation mechanisms, including fiber-matrix debonding, fiber pull-out, matrix shear yielding, and fracture of matrix and fibers.[28,29] Some of the energy dissipating events is fiber and interface related and the others are matrix related. However, major sources of toughness for the composites consisting of ductile matrix and short fiber or particle reinforcements are the matrix-related ones, including energy dissipation attributable to shear deformation of matrix. Hence, the matrix-related mechanisms should be promoted further. In fact, as indicated by Karger-Kocsis,[30] the fiber- or interface-related toughening mechanisms may augment the toughness contributions attributable to the matrix-related ones under favorable circumstances. An example is that the fiber pull-out process may induce matrix shear yielding, which may not only terminate the unwanted growth of crazes, but also may absorb a large amount of fracture energy. On the other hand, incorporation of rigid fibers with a ductile polymer matrix very often reduces the fracture toughness markedly. The causes for the reduction are diverse and complicated. The most common ones are the decreased matrix ductility caused by the plastic constraint imposed by the rigid fibers; the high stress concentration at fiber ends or crossings, and poor interfacial adhesion because of low compatibility between the fibers and matrix. In many cases, whether the presence of fibers will bring in positive or negative effect on the toughening of polymer composites depends largely upon the fiber–matrix interfacial bonding. From the micromechanics point of view, neither very strong nor very weak interfacial bond is desired for high fracture toughness.[31]

Adding SEBS-g-MA copolymer into the WF/PP composite can introduce a ductile interface between the wood fiber and PP matrix in the WF/PP/SEBS-g-MA composites based on the following mechanisms. Because the MA segment of the copolymer can react with the hydroxyl groups (OH) on the wood fiber surface,[11] the copolymer tends to form a thin encapsulating layer on the fiber surface. Direct evidence of this SEBS encapsulation was obtained in a glass bead/PP/SEBS-g-MA system by Karger-Kocsis.[27] In addition, the SEBS has a good interfacial adhesion with PP phase, because the SEBS is able to diffuse into the PP phase under micelle formation.[32] Combination of the interactions among the PP, wood fiber, and the SEBS-g-MA will lock the copolymer at the WF-PP interface, forming a polymeric and rubbery coupling layer. A schematic of this process is given in Figure 8.

The SEBS interface has many advantages over the low molecular weight silane interfaces when fracture toughness is a major concern. For example, because of its macromolecular nature, the SEBS interface is highly ductile. When subjected to a plane strain condition, the rubbery interface has the ability to relax laterally in response to the triaxial tension. At a high triaxial stress level, the polymeric interface may undergo debonding cavitation through disentanglement of polymer chains. Both processes are extremely important in toughening polymer-based composites, because the relaxation of the interface prevents the fiber–matrix interface, as well as the matrix, from premature brittle failure at the early stage of loading. The debonding cavitation can release the plastic constraint imposed by the rigid fibers before the triaxial stresses reach the fracture stress of the matrix to cause brittle failure. Obviously, for a given composite system, there exists a critical interfacial bond strength: when the fiber–matrix adhesion is stronger than the critical value, brittle matrix failure is likely to occur before the fiber–matrix debonding cavitation; if the interfacial bond is lower than the critical value, debonding cavitation at interface is preferably to

take place. In an ideal case, the cavitation strength of the polymeric interface should be designed slightly lower than the critical interfacial bond strength. The composites with the optimized interfacial bond strength will gain the highest fracture toughness without scarifying other important mechanical properties, including the tensile strength and modulus. It seems that the interfacial bond strength of the WF-A/PP/SEBS-g-MA composite is closer to the critical value than the other composites, because the impact strength, modulus, yield, and tensile strengths of the former composite were the highest among the all WF/PP/SEBS-g-MA composites. As demonstrated in Figures 9a and b, the SEM taken from the fractured surfaces of the WF-A/PP/SEBS-g-MA composites, large-scale plastic deformation occurred in PP matrix. Wood fibers are coated with thick polymer residuals, reflecting that the bonding between the fibers and polymer was good, and the debonding process took place, most probably, in the SEBS interface. The large space between the fibers and the surrounding matrix

(a) Molecular structure of cellulose of wood fiber:
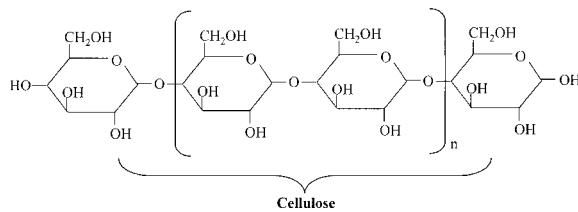
(b) Chemical reaction between hydroxyl group of cellulose and MA of SEBS:
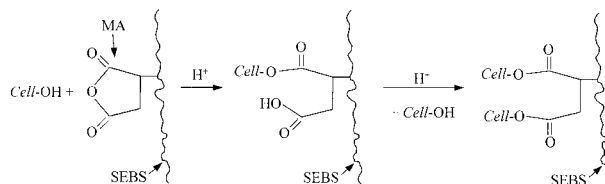
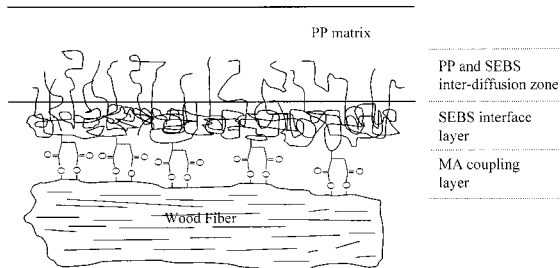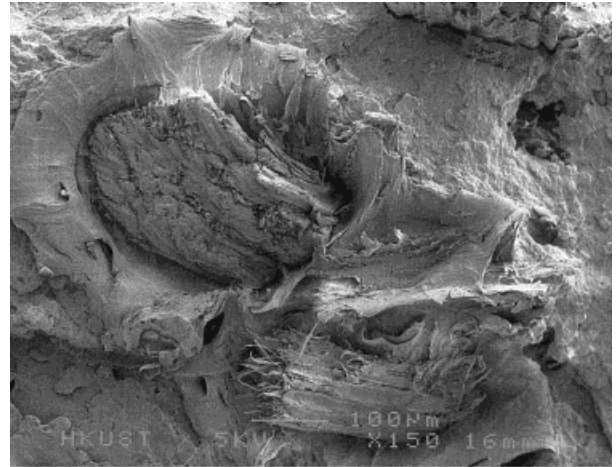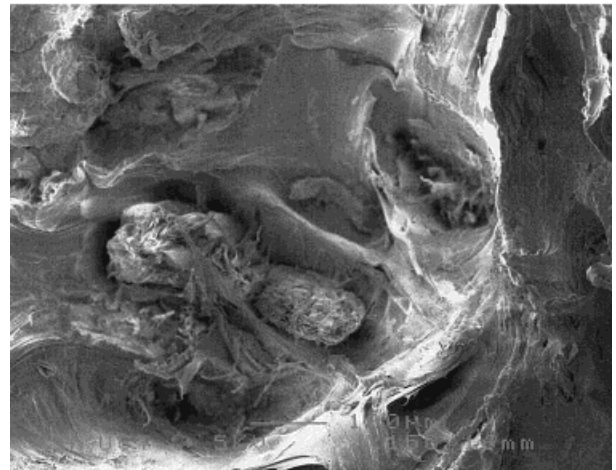(c) Micro-structure of SEBS-g-MA interface layer:

Figure 8 Schematic of the formation of the SEBS-g-MA interfacial layer between the wood fiber and PP matrix.

Figure 9 SEM of a fractured surface of the WF-A/PP/SEBS-g-MA composite.

was obviously caused by volume expansion and deformation of the matrix under triaxial tension, indicating that there was a high triaxial stress field that was released later by means of debonding cavitation of the polymeric interface.

Toughening Mechanisms in WF/PP Composites

The results for other composites without SEBS-g-MA copolymers support the above toughening theories. In fact, the observed low-impact strength of the WF-N/PP, WF-D/PP, and WF-G/PP composites reflects the poor interfacial strength caused by improper conditions used in the fiber pretreatment. As seen in Figures 6a–c, the fracture of the WF-D/PP composite occurred

within the PP matrix without noticeable plastic deformation. Fibers were pulled out of the PP matrix with smooth and clean surfaces because of the poor interfacial adhesion.

The fracture of the WF-A/PP composite is, however, a typical example of brittle failure caused by very strong interfacial bond strength. As mentioned in the previous section, the fiber–matrix interface in the WF-A/PP composite was the strongest among all WF/PP composites studied. Moreover, the low molecular weight of silane coupling agent often makes a rigid coupling layer. Therefore, the rigid fibers in the WF-A/PP composite were strongly bonded to the PP matrix by means of a rigid coupling layer. When the composite was subjected to impact loading, plastic deformation of the PP matrix was suppressed by the high strain rate as well as the constraint imposed by the rigid fibers. Materials in front of the crack tip in this circumstance were subjected to plane strain condition. The crack would propagate through the PP matrix with little plastic deformation. Also, because of the strong interfacial bonding, some wood fibers on the crack path were split into two pieces rather than being debonded and pulled out from the matrix, as shown in Figures 10a and b. It is seen that the fracture toughness of this composite was mainly caused by the fracture energy of wood fiber and matrix, which is relatively low.

CONCLUSIONS

The effect of fiber surface pretreatment on the interfacial bond strength and mechanical properties of wood fiber/polypropylene (WF/PP) composites was studied. The interfacial bond strength played a critical role in determining the tensile and impact strength of the composites. The wood fibers pretreated with the acid–silane aqueous solution gave rise to a high interfacial bond strength with the PP matrix, resulting in improved mechanical properties of the composite. Both rheological and microscopic studies were carried out to investigate the interfacial strength and toughening mechanisms.

A synergistic toughening effect between the wood fiber and SEBS-g-MA copolymer was observed in the impact tests of the WF/PP/SEBS-g-MA composites. To our knowledge, the same has never been reported in the literature. The impact strength of the WF/PP/SEBS-g-MA composite was found to be much higher than that of
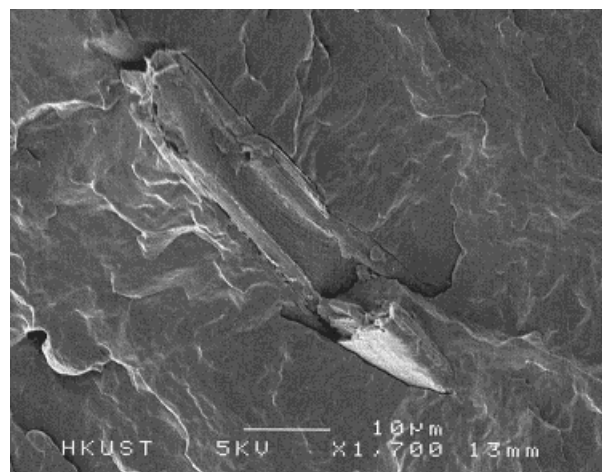
(a)
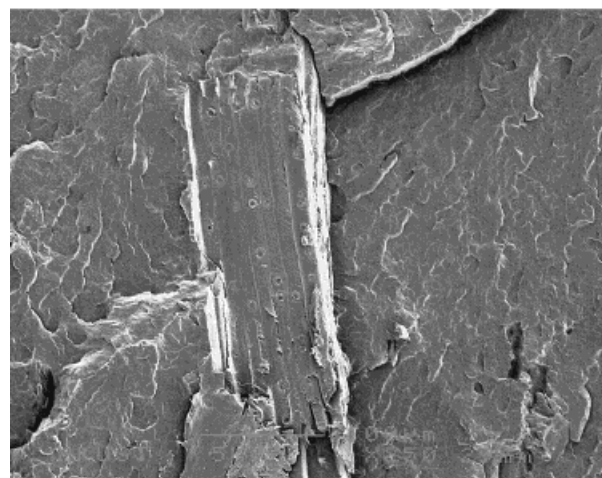
(b)

Figure 10   SEM of a fractured surface of the WF-A/PP composite.

the WF/PP composite or PP/SEBS-g-MA blend. The synergistic toughening was attributed to the ductile SEBS interface layer formed between the wood fibers and PP matrix, because the rubbery nature of SEBS prevented both the fiber and PP matrix from premature brittle fracture at the early stage of impact. The SEBS interfacial layer was able to cavitate under triaxial tension, which, in turn, effectively released the plastic constraint from the rigid fibers and induced massive plastic deformation in the PP matrix.

Materials Characterization and Preparation Facility (MCPF) at the Hong Kong University of Science & Technology for use of their equipment.

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