Fabrication and Mechanical Properties of Glass Fiber-Reinforced Wood Plastic Hybrid Composites

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ABSTRACT: To fully utilize the resource in the municipal solid waste (MSW) and improve the strength and toughness of wood plastic composites, glass fiber (GF)-reinforced wood plastic hybrid composites (GWPCs) were prepared through compounding of recycled high-density polyethylene (HDPE) from MSW, waste wood fibers, and chopped GF. Mechanical tests of GWPCs specimens with varying amounts of GF content were carried out and the impact fractured surface of GWPCs was observed through scanning electron microscope (SEM). The tensile strength of GWPCs and the efficiency coefficient values were predicted by Kelly-Tyson method. The results indicated that the tensile strength and impact strength of GWPCs could be improved simultaneously by adding type L chopped GF (L-GF), and would be dropped down when type S chopped GF (S-GF) was included. The tensile strength of GWPCs was well accordant with the experimental result. The efficiency coefficient values of S-GF and L-GF are −0.19 and 0.63, respectively. Inspection of SEM micrographs indicated that L-GF had achieved full adhesion with the plastic matrix through addition of maleic anhydride-*g*-polyethylene. The main fracture modes of GWPCs included pullout of GF, broken of matrix, and interfacial debonding. Because of the synergistic effects between hybrid components in GF/wood fiber/HDPE hybrid system, a special 3D network microstructure was formed, which was the main contribution to the significant improvement in the tensile strength and impact strength of L-GF-reinforced hybrid composites. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 112: 1250–1257, 2009

Key words: wood plastic composite; glass fiber; hybrid; synergistic effects; recycled; high-density polyethylene

INTRODUCTION

Wood fibers reinforced recycled plastics composites (WRPCs) offer interesting combination of properties. They are cheaper than competitive materials, especially those made of natural fibers and virgin resin. By using recycled plastics rather than virgin resin, these WRPCs provide an additional market for recycled plastics, thereby helping to reduce the burden of waste disposal in landfills. WRPCs can also be fabricated using recycled wood fibers, such as wood flour from sawmills, providing an additional usage for recovered wood fibers and thus further reducing waste in landfills.[1–3]

The WRPCs offer many advantages over natural wood. They do not warp, expand, or shrink; they are weather-, fungus-, and termite-resistant; and they require very little maintenance. Moreover, not only can they be recycled but also they can be made entirely from recycled materials. The usage of WRPCs is rapidly growing because of their many advantages. Many applications for WRPCs products include decking, window and door profiles, automobile paneling, panel inserts, and flower pots which were classified as decorative materials.[4–6]

However, these composites suffer from the disadvantage of being heavier in weight and lower in toughness as compared with wood, which makes them unsuitable for many applications. Physical, chemical, and hybridization methods have been used to enhance the mechanical properties of natural fiber-reinforced composites. Physical methods, such as stretching, calendering, and thermal treatment, can be used to change the structural and surface properties of the fiber, but do not affect its chemical composition. Chemical methods (e.g., oxidation, acetylation, mercerization, or using coupling agents) can also change the surface structure of the natural fiber. The main functions of the chemicals used are to react with the hydroxyl group on the fiber surface, making the surface wettable by the polymer, and promote covalent bonding with the matrix.[7] Chemical coupling is by far the most effective method of obtaining good interfacial adhesion at the fiber/matrix interface. For example, the treatment of cellulose fibers with maleic anhydride-*graft*-polypropylene (MAPP) can result in the formation of covalent bonds across the interface.

However, it is difficult to significantly improve the mechanical properties of WRPCs through

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interfacial modification. Existing literatures have reported a number of works[8,9] attempting to improve the structural or mechanical properties, and environmental behavior of the WRPCs products in various means, such as use of synthetic fiber, metal inserts, and glass fiber (GF). Among them, incorporation of GF reinforcements into polymer matrix is the most effective method to improve the mechanical properties of plastics. Using a hybrid composite that contains two or more types of different fibers, the advantages of one type of fiber could complement deficiencies in the other. As a consequence, a balance in performance and cost could be achieved via appropriate material design.

Rozman et al.[10] made a preliminary study on the use of glass and coconut fiber as reinforcements in polypropylene hybrid composites. They found that the incorporation of both fibers into the PP matrix has resulted in the reduction of flexural, tensile, and impact strengths and elongation at break. The reduction has been attributed to the increased incompatibility between the fibers and the PP matrix.

Tungjitpornkull et al.[11] studied E-chopped strand GF-reinforced wood-polyvinyl chloride (WPVC) composites. The results suggest that the tensile and flexural modulus and strengths of the wood/PVC composites increase with increasing GF contents. The tensile and flexural modulus and strengths of the WPVC composites at 10–20 phr GF loadings are more dependent on carbonyl (C=O) content on the fiber surface, but those at 30 phr GF loadings are influenced by the average final length of GF.

Mishra et al.[8] studied the mechanical performance of biofiber/glass-reinforced polyester hybrid composites. They found that the addition of small amount of GF in the pineapple leaf fiber and sisal fiber-reinforced polyester showed positive hybrid effect, which improved the mechanical properties of these composites.

Such variables were confirmed by Fu and Lauke[12] who studied the tensile strength of short fiber-reinforced polymers (SFRP) by considering the effect of fiber length and fiber orientation distributions, and found that the tensile strength of SFRP increased rapidly with increasing mean fiber length for the cases of small mean fiber length (as value close to critical fiber length, L_c) but was unchangeable at large mean fiber length ($>5\ L_c$). They also suggested that the composite strength increased with the decrease of critical fiber length. In aspect of fiber orientation distributions, they found that the maximum and minimum tensile strengths were shown for fiber oriented parallel and transverse to test direction, respectively.

Jiang et al.[13] studied PVC-based WRPCs with L and S type GF and found that impact strength improved upon adding 5% of type L GF but not upon adding type S.

Kitano et al.[14] also studied the effects of long and short GF, along with other fibers, on the properties of high-density polyethylene (HDPE)-based composites containing 20 vol % of fibers. They found that tensile strength decreased upon increasing the fiber content when long fibers were used, whereas the short fibers did not exhibit this pattern. They did not use any coupling agent.

Rozman et al.[15,16] studied the properties of a hybrid of GF and empty fruit bunch in a PP matrix and found significant increases in properties when suitable coupling agents were used and when the fibers were subjected to an oil extraction preprocess before actual molding.

Arbelaiz et al.[17] studied flax-fiber/GF hybrid composites in a PP matrix, varied the GF ratios from 0% of fibers to 100%, and found improvements in properties when MAPP was used as a coupling agent. In all these studies, the amount of GF used was usually quite high.

Thwe and Liao[9] looked at hybrid bamboo-GF composites in a PP matrix and found MAPP to be an effective coupling agent. The GF content ranged from 0 to 20%. They investigated the mechanical properties of bamboo-GF-reinforced polypropylene matrix hybrid composites (BGRP) before and after environmental aging. They found that before environmental aging, both modulus and strength in tension and flexural tests increased with incorporation of GF, while the mechanical properties reduced after environmental aging. They strongly suggested that hybridization of GF could improve durability of bamboo fiber-reinforced polypropylene (BFRP). They also showed that the fiber length, orientation, and distribution in the composites were important variables on the mechanical properties of the composites. They also reported that the moisture sorption and strength reduction are further suppressed by using maleic anhydride polypropylene (MAPP) as a coupling agent in both types of composite system.[18,19]

In this study, mechanical tests of WRPC specimens with and without chopped GF were characterized. The impact fractured surface of glass fiber-reinforced wood plastic hybrid composites (GWPCs) was observed through scanning electron microscope (SEM). The tensile strength of GWPCs and the efficiency coefficient values were predicted by Kelly-Tyson method.

EXPERIMENTAL WORK

Raw materials

The wood fibers collected from wood sawdust were separated by sifting into different mesh particle sizes. For the wood fibers of 48–100 mesh, the average length of which was 225 μm, they were then dried in

an oven at 105°C for 24 h to ensure the moisture content of final wood fiber was less than 3%. A combination of the alkaline and silane methods (ASM) was used to treat wood fiber. The experimental procedure for the ASM treatment of the wood fibers was already detailed in our previous work.[20]

Postconsumer HDPE containers were cleaned, washed, and dried. The dried plastic containers were compressed and heated at 180°C for 15 min with the pressure gradually increased to 2.5 MPa. The temperature and pressure were kept at 180°C and 2.5 MPa, respectively, for 5 min. The plastic sheet was then cooled down to room temperature and was cut into strips with widths from 25 to 35 mm, which were then processed in a Collin Teach-Line® E 20T single screw extruder to make recycled HDPE (RPE) pellets. The temperatures of the initial, middle, and final chambers were 185, 192, and 187°C, respectively. The temperature of the die was 177°C. The turning speed of the screw was 55 rpm, and the extrusion pressure was 13 MPa. The melt flow index of the recycled HDPE is 8.4 g/10 min (170°C).

The coupling agent used for improving the adhesion between the hydrophobic HDPE and the hydrophilic wood fiber was the maleic anhydride-*g*-PE (MAPE, Epolene G-2608 from Eastman Company, ring and ball softening point of 122°C, molecular weight of 65,000, UK), and in all compositions it was 10 wt % of wood fiber.

The GF used was E-glass chopped fiber, 2 mm (S) and 5 mm (L) in length with 13 μm in diameter, provided by Sinoma, China. The L/day ratios of which were 143 and 357, respectively.

Preparation of GWPCs pellets

The weight fraction of wood fiber inside RPE was 35%. The content of E-GF to be introduced in the wood plastic composites varied from 5 to 15 phr of compound. The powder was dry-blended with various necessary additives as listed in Table I. Wood fiber, RPE pellets, and chopped GF were blended for 2 min to produce a homogeneous mixture in an M-10 L high-intensity laboratory mixer, which is a kind of mechanically drived corotating mixing device.

TABLE I
The Ingredients for GWPCs

Ingredients	Concentration (phr)
Wood fiber	35.0
RPE	65.0
MAPE	3.5
Calcium stearate	2.0
Calcium carbonate	8.0
GF	Varied/0, 5, 10, 15

The mixture was processed with the Collin Teach-Line® ZK 25T double screw extruder to make GWPCs pellets. The temperatures of cylinder were set to 185, 192, and 187°C, respectively. The temperature of die was 177°C. The screws speed was 45 rpm, and the extrusion pressure was 11 MPa.

Compression molding of GWPCs panels

The above pellets were compression-molded into sheets by using an oil-heated press (TMP Moore 50-Ton) with a nominal maximum pressure of 5.5 MPa, which was used for the molding. The temperature of the press platens was maintained at 180°C. After forming, the sheets were cooled under pressure to ambient temperature and cut into dog-bone-shaped samples conforming to ASTM D638 Type-V specifications.

Five GWPCs panels were produced for each type of ingredients and prescription. The panels were air conditioned for at least 48 h at 23°C ± 2°C and 50% ± 5% humidity to eliminate the processing residual stresses.

Mechanical property testing

The dog-bone-shaped samples were cut from the compression-molded panels. Tensile test was performed on a SANS tensile tester (Shenzhen, China) at a crosshead speed of 5 mm/min. The tensile test procedure followed the ASTM D638 (1990) Specimen Type I. For each composition, 10 samples were tested, and the average values were used for interpreting the results.

Izod impact tests, following ASTM D256 (1990), were performed on a XJU-5.5 impact tester (Chengde, China) with the notched side facing the pendulum.

Scanning electron microscope characterization of fracture surface

The samples were cut near the fracture surface, and SEM micrographs of fracture surfaces were taken by using a Fei Quanta 200 SEM. For this operation, each sample was first gold-coated by using a sputter coater, and then the microstructure was examined.

RESULTS AND DISCUSSION

Tensile strength

Figure 1 showed the tensile strength variations of GWPCs with different weight fractions and types of chopped GF. As can be seen, the reinforcing effect of GF on WRPCs was decided by the fiber length, where L chopped GF (L-GF) showed significant improvement in tensile strength of GWPCs, the

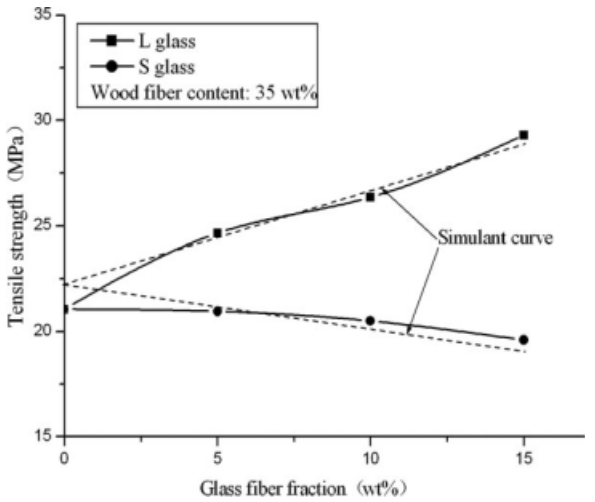

Figure 1 The relationship between tensile strength of GWPCs and GF fraction.

strong GF did contribute to increasing the load-bearing ability of the WRPCs. Compared with WRPCs, the tensile strength of GWPCs was increased by 38.7%, from 21.09 to 29.25 MPa, when 15 wt % of L-GF was introduced. One advantage of using the MAPE as a coupling agent is that it promotes adhesion between plastic and both wood fiber and GF, so that only one coupling agent can be used for both materials. This finding in Figure 1 also indicated that the maleic-anhydride coupling agent successfully promoted adhesion between GF and the plastic matrix.

However, there will be about 6.5% of decrease in tensile strength for S chopped GF (S-GF) under the same conditions. The explanation is that there is a critical length, l_{cr}, for this kind of short fiber-reinforced composites, which may be computed according to eq. (1).

$$\frac{l_{cr}}{d} = \frac{\sigma_f}{2\tau_s} \tag{1}$$

where d is the fiber length, σ_f is the tensile strength of fiber, τ_s is the interfacial shear strength.

As can be seen that only the GF length is higher than the critical length, it can show reinforcing effect. If the GF is too short or less than l_{cr}, it can only serve as filler, which will destroy the continuity of matrix. On account of this, the load cannot transfer from the matrix to GF, which will instead decrease the tensile strength of WRPCs.

In this research work, the tensile strength and the diameter of GF is 2100 MPa and 14 μm, respectively. So the interfacial shear strength between GF and WRPCs is about 2.94–7.34 MPa, which is far-forth lower than that of the same kind of fiber-reinforced thermoplastic composites (28 MPa).[21]

The tensile strength of GWPCs can be given by Kelly-Tyson method,[22] in eq. (2).

$$\sigma_c = k\eta_0\eta_L V_f\sigma_f + (1 - V_f)\sigma_{um} \tag{2}$$

where k is efficiency coefficient, which refers to the contribution of GF to the tensile strength of WRPCs. η_0 is the orientation coefficient of GF. Thomason et al.[22] gave its value for GF, which is 0.206. η_L, V_f, σ_f is the efficiency coefficient of GF length, the volume fraction of GF, and the tensile strength of GF, respectively. σ_{um} is the strength of GF when GWPCs is fractured, which may be computed based on eq. (3).

$$\sigma_{um} = \frac{E_m}{E_f}\cdot\sigma_f \tag{3}$$

where E_m is the tensile modulus of WRPCs.

For GWPCs with 30 wt % of wood fiber, E_m is 1.2 GPa, E_f is the tensile modulus of GF, the general value is around 75 GPa. So the tensile strength of GWPCs when GF fails is 22.4 MPa based on eq. (3).

According to Kelly-Tyson theory, η_L may be given based on eq. (4).

$$\eta_L = \frac{1}{V_f}\left[\sum\frac{L_i V_i}{2L_c} + \sum V_j\left(1 - \frac{L_c}{2L_j}\right)\right] \tag{4}$$

where V_i, L_j is the volume fraction of fiber with length of L_i, L_j, respectively, L_c is the critical fiber length.

The corresponding values of η_L for the two types of GF may be computed according to eqs. (5) and (6).

$$\eta_L = \frac{L_f}{2L_c} \tag{5}$$

$$\eta_L = \left(1 - \frac{L_c}{2L_f}\right) \tag{6}$$

where L_f is the GF length.

If choose the average length (3.5 mm) of the two types of GF as the critical length for this system, the η_L values of the two fibers are 0.286, 0.65, respectively, and the tensile strength of GWPCs can be predicted according to eqs. (7) and (8).

$$\sigma_c = 22.4 + (82.48k - 22.4)V_f \tag{7}$$

$$\sigma_c = 22.4 + (187.46k - 22.4)V_f \tag{8}$$

$$V_f = \frac{\frac{W_f}{\rho_f}}{\frac{W_f}{\rho_f} + \frac{1-W_f}{\rho_m}} \tag{9}$$

where W_f is the weight fraction of fiber, ρ_f and ρ_m are the density of fiber and matrix (2.54 g/cm^3, 1.10 g/cm^3), respectively.

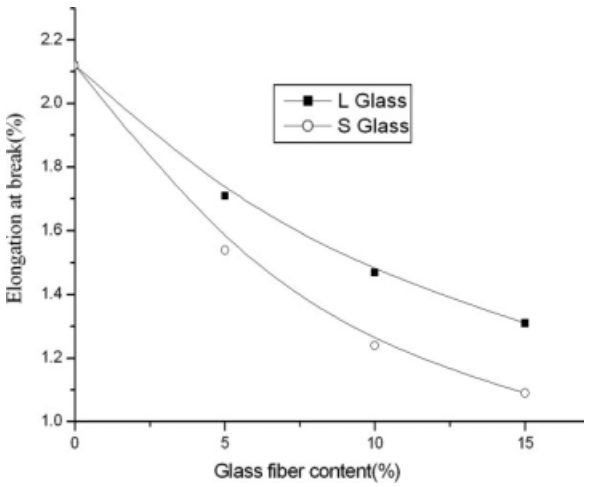

Figure 2 Elongation at break of GWPCs containing different types of GF.

According to the results in Figure 1, and the conversion relationship between weight fraction and volume fraction of GWPCs, the k values of S-GF and L-GF are −0.19 and 0.63, respectively, by curve simulation.

Elongation at break

Figure 2 showed the elongation at break of GWPCs with different weight fractions and types of chopped GF. As can be seen, the addition of GF decreases the elongation at break because of the increased stiffness provided by these fibers. Increasing the GF content also reduces the elongation at break owing to reduction in the amount of ductile plastic matrix in the composite. But the decrease in the elongation at break of GWPCs containing L-GF is less than that of S-GF. The reason for this result is the same as the tensile strength. L-GF can carry and transfer more external load during failure which increases the fracture strain of GWPCs.

Impact property

The incorporation of GF into WRPCs will significantly affect its impact property. The impact strength of WRPCs may be distinctly improved for L-GF, in Figure 3. When 15 wt % of L-GF was introduced, the impact strength of GWPCs was increased by 40.6%, from 5.61 to 7.89 kJ/m^2. The impact strength value of composite materials indicates their energy absorption capability during fracture, which represents the interfacial shear strength and combination of composite materials.

Because the diameter of GF is less than that of wood fiber (around one tenth), a lot of interface will be formed when it is compounded with GWPCs matrix. On the other hand, a major issue in achieving true reinforcement for wood plastic composites is the inherent incompatibility between the hydrophilic fibers and the hydrophobic polymers, which results in poor adhesion and therefore in poor ability to transfer stress from the matrix to the reinforcing fibers.[23] A number of investigators have explored the capability of additives to enhance adhesion and thereby improve properties, such as the tensile and flexural strengths of these composite materials.[24–26] The most commonly used additives is coupling agent, which may form chemical bonds between the cellulose chains in the fiber and the polymer matrix.[27] There has been a lot of research over the past decades on different types of coupling agents to improve the adhesion between the wood and the plastic. The most commonly used coupling agents are maleated polyolefins.[28]

In this research work, MAPE served as coupling agent which may also improve adhesion between the plastic and GF, because there is also a lot of hydroxyl on the surface of GF. The strong interfacial combination between wood fiber and plastic, GF and plastic was formed, so the introduction of GF could theoretically enhance the impact resistant properties. The interface inside this kind of hybrid GWPCs may absorb more energy when the crack in the matrix spreads to the interface.

Compared with L-GF, there were more "rigid rods" in the GWPCs reinforced with S-GF under the same weight fraction, then the probability of formation of defects will increase. Because these short "rigid rods" cannot carry external load, the cracks in the matrix will link together when they extend to the interfacial area, which lead to earlier failure and lower impact strength.

Although there are still some demerits such as large density, high cost, and abrasion of equipment,[29] GF is a good reinforcement for GWPCs, especially for those

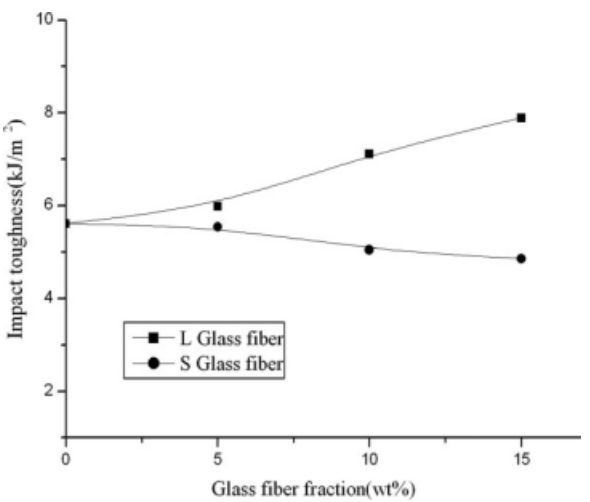

Figure 3 The relationship between impact strength of GWPCs and GF fraction.

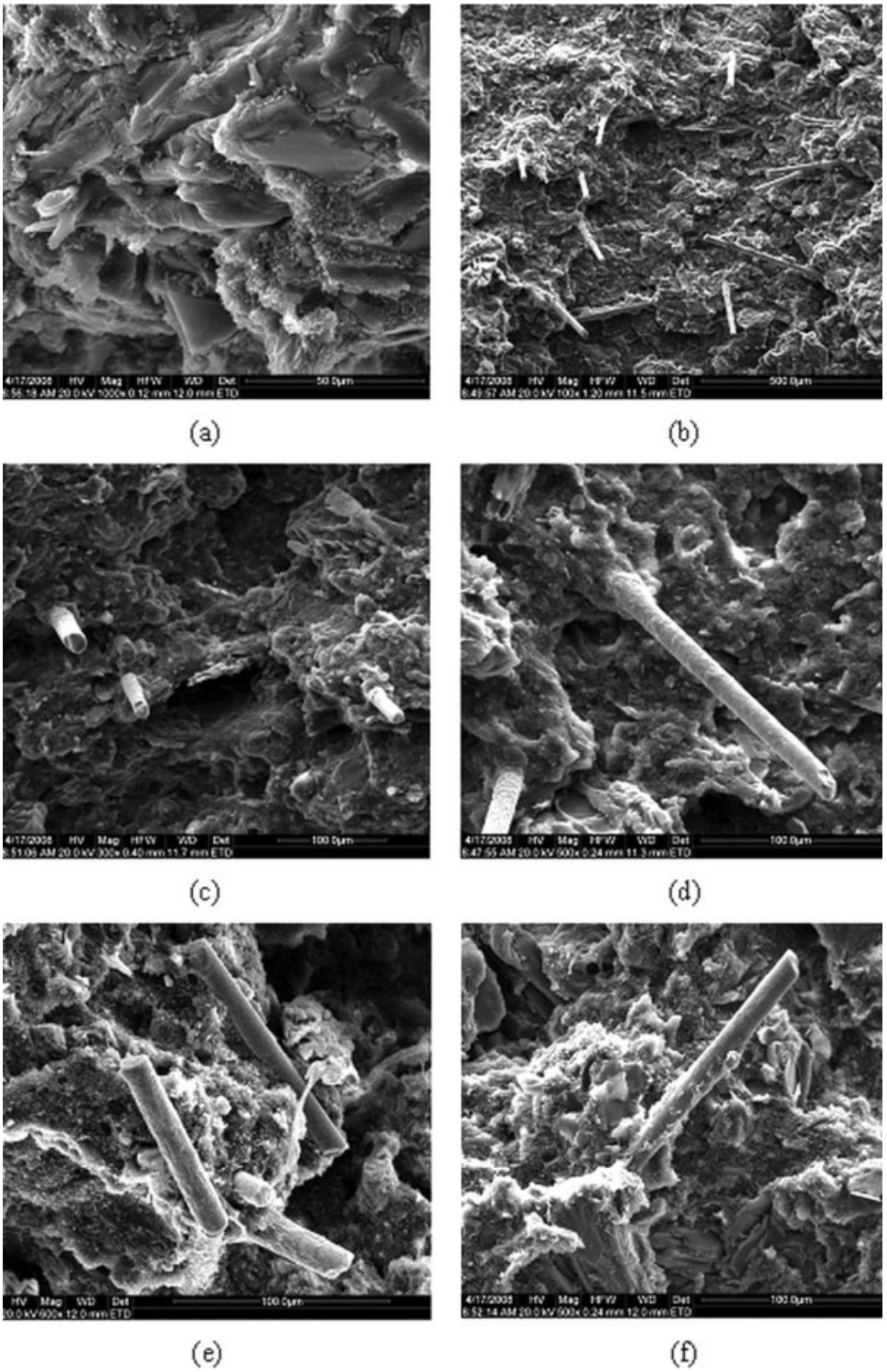

Figure 4 Microstructure of fracture surface of GWPCs specimen containing different types of GF.

applications where high tensile strength and impact strength are required.

Microstructure of the fracture surfaces

The interfacial microstructure will significantly affect the mechanical properties of GWPCs, because the interface plays a quite important role in the transferring of stress and spread of crack between fiber and matrix.

Figure 4(a) showed the microstructure of fracture surface of WRPCs, where very good interfacial adhesion occurred when wood fibers were treated by ASM and MAPE was used as coupling agent.

Figure 4(b–f) showed the microstructure of fracture surface of GWPCs specimen containing different types of GF. As can be seen, fiber pull-out is the main failure mode of GWPCs reinforced by L-GF, in Figure 4(b). Other failure modes include interfacial debond, matrix fracture, Figure 4(c).

In Figure 4(d), the GF can be seen clearly, and apparently the coupling agent has fully acted on the glass surface, as adhesion is observed. However, the wood fiber is not distinguishable, thus indicating that the amount of coupling agent used is sufficient for fully compatibilizing the wood fiber and the GF, which is different from the results of Rizvi and Semeralul.[30]

Interfacial debond is the main fracture mode of S-GF-reinforced GWPCs, where few GF pull-out happened, as seen in Figure 4(e,f). Another noticeable information is that no fractured GF was found on the fractured surface, which is accordant with the results of tensile strength and impact strength (Figs. 1 and 3). This implied that GF did not effectively carry and transfer stress during fracture process, exert its reinforcing effect and improve the stress level of plastic matrix. The crack in the matrix would quickly spread along the interface between the GF and the matrix under the action of impact load. During the extrusion process, GF will be broken into shorter ones. This can be seen in Figure 4(e) where the dimensions of the fibers have been reduced in the longitudinal directions.

These broken fibers show little attachment to HDPE matrix. Only a small amount of matrix is left on the surface of the fractured GWPCs, which is shown in Figure 4(f), where also indicated that the interface of GWPCs containing S-GF was poor compared with that of L-GF.

Synergetic reinforcing effect

When two or more kinds of reinforcements were used to fabricate hybrid composite materials with only one matrix, there may be some physical or chemical synergetic reinforcing effects between the reinforcing components. This kind of synergetic reinforcing effect in wood fiber/PP/SEBS-*g*-MA hybrid composite system has been reported by Rozman et al.[15] A lot of wood microfibril was formed under the friction between GF rod and wood fiber after they were mixed under high speed and compounded in molten state. Then a special kind of three-dimensional hollow network structure came into being between wood fiber, L-GF, and matrix, which is the main reason why the impact properties of GWPCs were improved, in Figure 5. The wood fiber and the polymeric molecule can fill in the cavity in this kind of 3D microstructure. Thus, very strong framework was generated in the matrix during compression molding which effectively restricted the spread of crack in the matrix.

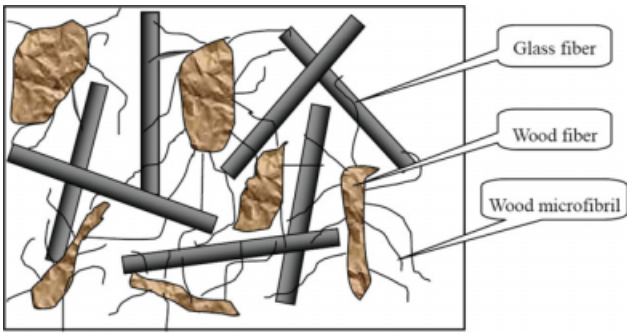

Figure 5 Three-dimensional network structure in GWPCs. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

However, this 3D microstructure is seldom observed in the GWPCs reinforced with S-GF. Under this circumstance, the hybrid components were only mechanically and randomly mixed together.

It is obvious that the reinforcing effect of 3D network lied on the length or length/diameter ratio of GF and the volume fraction of matrix. Because when the weight fraction of reinforcement is too high, the cavities in the 3D network structure can not be entirely filled by resin or matrix, which even generate defects like inanition.

SUMMARY

GF-reinforced wood plastic hybrid composites were prepared through compounding of RPE from municipal solid waste, waste wood fibers, and chopped GF. Mechanical tests of GWPCs specimens with varying amounts of GF content were carried out and the impact fractured surface of GWPCs was observed through SEM.

The tensile strength and the impact strength of GWPCs could be improved simultaneously by adding L-GF and would be dropped down when S-GF was included. Compared with GWPCs, the tensile strength of GWPCs was increased by 38.7%, from 21.09 to 29.25 MPa, when 15 wt % of L-GF was introduced the impact strength of GWPCs increased by 40.6%, from 5.61 to 7.89 kJ/m^2. The tensile strength of GWPCs was predicted by Kelly-Tyson method. The efficiency coefficient values of S-GF and L-GF are −0.19 and 0.63, respectively. Inspection of SEM micrographs indicated that the L-GF had achieved full adhesion with the plastic matrix through the addition of maleic anhydride-*g*-polyethylene. The main fracture modes of GWPCs included pullout of GF, broken of matrix, and interfacial debonding. Because of the synergistic effects between hybrid components in GF/wood fiber/HDPE hybrid system, a special 3D network

microstructure was formed, which was the main contribution to the significant improvement in the tensile strength and impact strength of L-GF hybrid composites.

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