

# Wood-Thermoplastic Composites from Wood Flour and High-Density Polyethylene

Cheng Xiong, Rongrong Qi, Yanling Wang

Department of Polymer Science and Engineering, School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University, Shanghai 200240, China

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**ABSTRACT:** High-density polyethylene/wood flour (HDPE/WF) composites were prepared by a twin-screw extruder. The effects of WF, silane coupling agents, polymer compatibilizers, and their content on the comprehensive properties of the WF/HDPE composites have been studied in detail, including the mechanical, thermal, and rheological properties and microstructure. The results showed that both silane coupling agents and polymer compatibilizers could improve the interfacial adhesion between WF and HDPE, and further improve the proper-

ties of WF/HDPE composites, especially with AX8900 as a compatibilizer giving higher impact strength, and with HDPE-g-MAH as a compatibilizer giving the best tensile and flexural properties. The resultant composite has higher strength (tensile strength = 51.03 MPa) and better heat deflection temperature (63.1°C). © 2009 Wiley Periodicals, Inc. *J Appl Polym Sci* 114: 1160–1168, 2009

**Key words:** wood-thermoplastic composites; HDPE; wood flour; silane coupling agent

## INTRODUCTION

Over the past several decades, many types of inorganic fillers, such as calcium carbonate, mica, fiberglass, talc, and clay, have been widely used to improve the property of high-density polyethylene (HDPE).<sup>1–5</sup> Although inorganic fillers currently dominate the thermoplastic industry, organic fillers from renewable resources, such as wood flour (WF) and other cellulose fibers, have aroused great interest because of the increased awareness about recycling and the need for lower cost materials. Wood-plastic composites (WPCs)<sup>3</sup> are advantageous over neat polymers and woods, because of reduced cost variable specific gravity, recyclability, environmental friendliness, and reduced wear on processing equipment. WPCs are mainly used as exterior nonstructural or semistructural building products in the construction industry, such as decking, fencing, siding, window frames, and roof tiles.

However, the poor compatibility between a hydrophilic wood and a hydrophobic plastic is still in presence restricting the application of WPCs. This poor compatibility gives rise to the agglomeration of wood fibers and weak interfacial adhesion between

wood fillers and plastic matrices, which results in poor mechanical properties of the composites. Furthermore, WF is multicomponent, including cellulose, hemicellulose, and keratin, whose low-molecular-weight component may also influence the mechanical properties, such as the tensile strength and Izod impact strength of WPCs. In the last 2 decades, a lot of efforts have been put into improving the interfacial adhesion between the polar wood fiber and nonpolar thermoplastic matrix.<sup>6–10</sup> To improve the compatibility, the WF can be treated with silane coupling agents. But the coupling agent alone has a limited effect on the mechanical properties of WPCs, so polymer compatibilizer is also added to WPCs<sup>1,11–16</sup> to solve the interfacial action between WF and plastic matrix. An ideal compatibilizer can act as a bridge between the WF and the matrix to improve the properties of WPCs, and many researchers are focus on the study of compatibilizers for WPCs. For example, Wang et al.<sup>17</sup> used LLDPE-g-MAH as a compatibilizer in the WPC composite, and Lu et al.<sup>18</sup> investigated the effect of MAPE on the mechanical properties of WF/HDPE system. The benefit from using higher content of WF in WPC includes reduced cost and improved mechanical properties to some extent. However, some concerns still remain such as the agglomeration of fillers<sup>10</sup> and the difficult process because of high viscosity at high filler content.<sup>19,20</sup> Therefore, most of the studies conducted during the last 2 decades were focused on the WPCs with a content of wood fillers less than

Correspondence to: R. Qi (rrqi@sjtu.edu.cn).

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**TABLE I**  
The Structures of Silane Agents

Vinyl silane	$\text{CH}_2=\text{CHSi}(\text{OCH}_3)_3$
Amino silane	$\text{NH}_2(\text{CH}_2)_3\text{Si}(\text{OCH}_2\text{CH}_3)_3$
Epoxy silane	$\begin{array}{c} \text{H}_2\text{C} \quad \text{CH} \quad \text{CH}_2\text{O}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3 \\ \diagdown \quad \diagup \\ \text{O} \end{array}$
Allyl ester silane	$\text{CH}_2=\text{C}(\text{CH}_3)\text{COOCH}_2\text{CH}_2\text{CH}_2\text{Si}(\text{OCH}_3)_3$

50 phr,<sup>21–27</sup> which is helpful to improve interfacial adhesion. Up to now, polyethylene is one of the most commonly used thermoplastics for WPCs, and some polymer compatibilizers should be used to improve the interfacial adhesion between WF and HDPE matrix. However, the choice of an effective coupling agent or compatibilizer for wood-PE composites is limited at present. In this study, the effect of eight types of compatibilizers and four silane coupling agents on the properties of WF/HDPE composites have been thoroughly investigated.

The objectives of this study were (1) to find an effective silane coupling agent to improve the interface adhesion between WFs and HDPE; (2) to investigate the effect of the various polymer compatibilizers on the mechanical properties of the composites; (3) to study the rheology and crystallization behavior of the composites, especially with higher wood filler content. The results reported in this article will promote the understanding of the preparation of high-performance WF/HDPE composites.

## EXPERIMENTAL

### Materials

Wood flour (pine flour, mesh 80, soft, density  $\approx 0.25 \text{ g/cm}^3$ , moisture content, 2.04%) was purchased from Lin'an Mingzhu wood flour Factory (China). HDPE is a product of Yanbet in Saudi Arabia with a melt index of 8 g/10 min (HDPE 80064). Ethanol (water content less than 0.3%, AR) was purchased from Shanghai Lingfeng Chemical Solvent (China). Silane coupling agents [vinyltrimethoxysilane (vinyl silane),  $\gamma$ -aminopropyltriethoxysilane (amino silane),

$\gamma$ -glycidoxypropyltrimethoxysilane (epoxy silane), and  $\gamma$ -ethacryloxypropyltrimethoxysilane (allyl ester silane)] are products of Shanghai Yaohua Chemical Solvent (China), and their molecular formulae are listed in Table I. HDPE-g-MAH, LLDPE-g-MAH, LDPE-g-MAH, and POE-g-MAH made by our group contain about 1.2 wt % maleic anhydride. Grafting reactions were carried out in the molten state with a TSE-35 corotating twin-screw extruder (Ruiya Group, Nanjing, China) at 170°C. HDPE (80064) or LLDPE (2047, Dow Chemical Company, USA) or LDPE (722, Dow Chemical Company, USA) or ethylene-octene copolymer (POE) (Exact 4049, Exxon Mobil Corporation, USA), MAH, initiators, solvent, and comonomers were simultaneously introduced into the twin-screw extruder after dry blending, which was similar to the preparation of ABS-g-MAH.<sup>28</sup> EVA (ethylene/vinyl acetate) and EMA (ethylene methyl acrylate) are the products of DOW Chemical Company (USA). E-MA-GMA (ethylene-methyl acrylate-glycidyl methacrylate terpolymers, with a trade name AX8900 and AX8920, respectively) is kindly supplied by Arkema Company (France). The properties of these compatibilizers are listed in Table II.

### Sample preparation

WF was dried at 105°C for 24 h in a vacuum oven to remove moisture before being dry-blended with HDPE and compatibilizers.<sup>3</sup> The dried WF was treated by silane coupling agents in ethanol solution. The ratio of silane coupling agent to ethanol is 1 : 9, and the silane coupling agent is among 0–10% by the weight of WF, then the treated WF was activated at 120°C for 2 h in a vacuum oven. The mixing of HDPE and WF with various compatibilizers was carried out with a ZE 25A twin-screw corotating extruder (Berstorff GmbH Company, Germany), the screw speed was maintained at 120 rpm, and the barrel temperatures ranged from 140 to 175°C. All composite pellets were dried at 80°C for 24 h to <1% moisture content before injection molding. Test

**TABLE II**  
Properties of Compatibilizers Used

Compatibilizer	MFI (g/10 min, 190°C/2.16 kg)	Melt point (°C)	Tensile strength (MPa)	Degree of grafting (wt %)
HDPE-g-MAH	2.5	137	24	1.2
LLDPE-g-MAH	2.0	121	16	0.9
LDPE-g-MAH	2.0	103	14	0.8
POE-g-MAH	1.9	56	6	0.8
AX8900	6.0	60	4	8
AX8920	6.0	63	7.5	1
EVA	3.5	80	8	11
EMA	3.5	61	6	27–31
Measurement	ASTM 1238	DSC	ASTM D 882	ATO method

**TABLE III**  
**The Formula of the WF/HDPE Composites**

Composites	HDPE (phr)	WF (phr)	Silane coupling agent (phr)	Polymer compatibilizer (phr)
I. Allyl ester silane content series	100.0	50.0	0	0
	100.0	50.0	0.5	0
	100.0	50.0	1.5	0
	100.0	50.0	2.5	0
	100.0	50.0	3.0	0
	100.0	50.0	4.0	0
	100.0	50.0	5.0	0
II. Different silane series	100.0	50.0	2.5 (vinyl silane)	0
	100.0	50.0	2.5 (amino silane)	0
	100.0	50.0	2.5 (epoxy silane)	0
	100.0	50.0	2.5 (allyl ester silane)	0
III. Polymer compatibilizers series (HDPE-g-MAH, LLDPE-g-MAH, LDPE-g-MAH, POE-g-MAH, AX8900, AX8920, EVA, or EMA)	68.7	30.0	0	1.3
	67.4	30.0		2.6
	66.2	30.0		3.8
	64.9	30.0		5.1
	63.7	30.0		6.3
	62.0	30.0		8.0
	58.2	30.0		11.8
IV. Wood flour content series	80.0	10.0	0.5 (epoxy silane)	20.0
	80.0	20.0	1.0 (epoxy silane)	20.0
	80.0	30.0	1.5 (epoxy silane)	20.0
	80.0	40.0	2.0 (epoxy silane)	20.0
	80.0	50.0	2.5 (epoxy silane)	20.0
	80.0	60.0	3.0 (epoxy silane)	20.0
	80.0	70.0	3.5 (epoxy silane)	20.0
	80.0	80.0	4.0 (epoxy silane)	20.0

specimens were molded in a HTBIIOX/1 screw injection molder (Haitian plastic machine company, Ningbo, China). The mold nozzle temperature was 185°C, and the injection pressure reached a peak of 12.5 MPa. The American Society for Testing Materials (ASTM) mold cavity was used. The detail experiment formula is listed in Table III.

### Characterization

#### Mechanical properties

The Izod notched impact strength was conducted on an Izod notched impact machine (Ray-Ran, UK) according to the ASTM D 256 (The samples were cut into 4 mm × 10 mm × 80 mm with a notch deep of 2 mm). The specimens for tensile test were in dumbbell-shape with a dimension of 2 mm × 6 mm × 80 mm, and the testing method used was ASTM D 882 under 50% humidity and 23°C (testing rate: 50 mm/min), which was run on an Instron 4465 (Instron, USA) machine. At least five specimens were tested for each formulation.<sup>29</sup>

#### Rheological behaviors

The rheological properties of the composites were measured with a rotational rheometer (Gemini 200 HR Rheometer, Bohlin Instruments, UK) fitted with a parallel plate geometry. The diameter of the plate

was 25 mm and the gap between two plates was 2.0 mm. The tests were carried out by varying the frequency from 0.01 to 10 rad/s at 170°C under a nitrogen atmosphere to prevent oxidation.

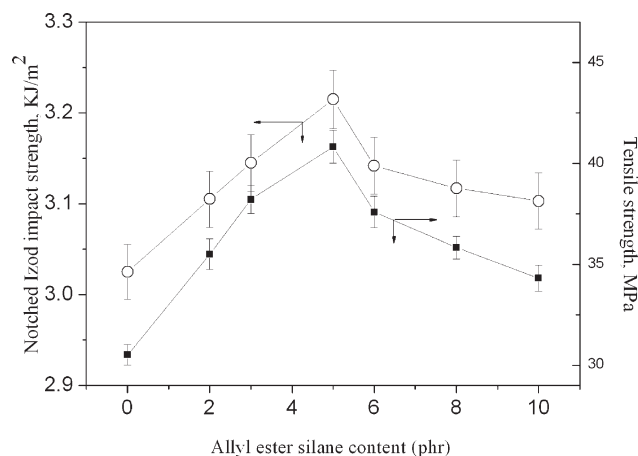
#### Scanning electron microscopy

Scanning electron microscopy (SEM) was used to observe the fractured surface of a sample after cooling in liquid nitrogen. The corresponding cross-sections were examined by using an S-2150 scanning electron microscopy (Hitachi, Japan). The surfaces of sample were dried under vacuum and then gold was evaporated before SEM analysis.

#### Thermal properties

Crystallization behaviors were conducted with a Perkin-Elmer PYRIS-1 differential scanning calorimeter. A sample of 3–6 mg was heated from 20 to 180°C at the heating rate of 50°C/min and then maintained at 180°C for 3 min. After that, the sample was cooled from 180 to 20°C with a cooling rate of 10°C/min; finally, the sample was heated again from 20 to 180°C with a heating rate of 10°C/min. All DSC measurements were performed under a nitrogen atmosphere.

Heat deflection temperature (HDT) was measured as per ASTM-648 with a hot deformation test



**Figure 1** Influence of the allyl ester silane agent content on the impact and tensile properties of WF/HDPE composites.

apparatus (ChengDe COTS Science Measure Col, Chengde, China).

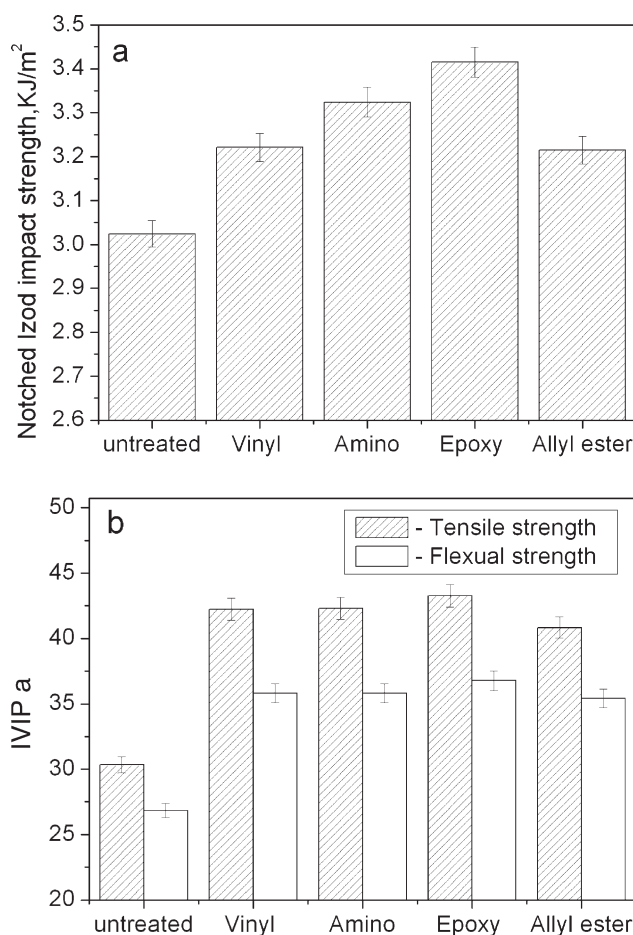
## RESULTS AND DISCUSSION

### Influence of silane coupling agents

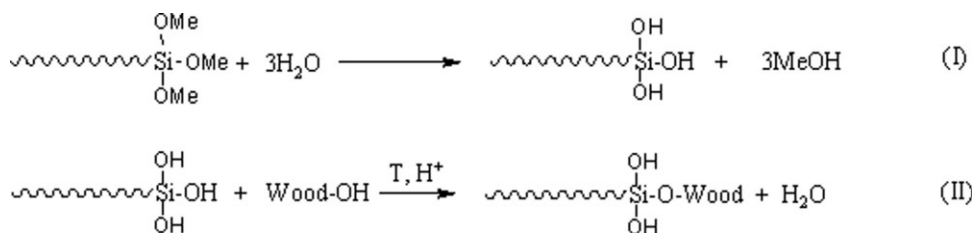
Silane coupling agents were used to improve the interfacial bonding strength between polar WF and nonpolar polyethylene matrix.<sup>17,30</sup> The effect of the loadings of silane coupling agent-allyl ester silane on the mechanical properties of WF/HDPE composites (WF content is fixed at 50 phr in Table III) is shown in Figure 1. From Figure 1, we can see that the mechanical properties of the composites with allyl ester silane are better than those of the composites without silane coupling agent, indicating that silane agent is effective for improving the interfacial binding of WF/HDPE composites. Careful observation reveals that the tensile and impact strength of the resultant composites increase obviously at the low weight percentage (<5.0 phr based on WF) and reach a maximum at 5.0 phr and then gradually decrease with a further increase of silane coupling agent. Although silane coupling agent can improve the interfacial binding of WF/HDPE composites, the silane coupling agent cannot wet the WF completely when its concentration is less than 5.0 phr, which would endow many deformities on the surface of WF and further influence the properties of WF/HDPE composites. However, more allyl ester silane may weaken the link between WF and HDPE and then decrease its mechanical properties because of low molecular weight of allyl ester silane.

The previous work<sup>17</sup> indicated that the chemical structure of silane coupling agents had an important influence on the mechanical properties of the resultant composites, so four different types of silane cou-

pling agent (5 phr based on WF) have been studied (Table III) in this work, and the mechanical properties of the obtained WF/HDPE composites are shown in Figure 2 with the WF fixed at 50 phr. The results show that the modified composite by silane coupling agents had better mechanical properties than that of the untreated composites. The effect of silane coupling agent on the mechanical properties may mainly behave in two aspects. First of all, the active group of silane coupling agent can react with the hydroxyl group of WF (shown in Scheme 1), which may weaken the polarity of WF and further improve its miscibility with the nonpolarity matrix HDPE. The second, the long carbon chain of silane coupling agent can twist with the matrix HDPE. The coeffect makes the bridge role of silane coupling for HDPE and WF in the composites, which may improve the interfacial adhesion between WF and HDPE and then strengthen the mechanical properties of the composites. However, the four silane coupling agents also have differentiated effects on the mechanical properties of the WF/HDPE composites.



**Figure 2** Influence of silane coupling agents on WPCs' mechanical properties. (a) Impact strength and (b) tensile and flexural strength.



**Scheme 1** Mechanism of the reaction between silane and wood flour.

Among these silane coupling agents, epoxy silane is the most effective for the WF/HDPE system, whereas allyl ester silane is the worst. The epoxy silane-treated composite could improve impact strength by 13% [Fig. 2(a)], tensile strength by 42%, and flexible strength by 37% [Fig. 2(b)] compared with the untreated composites. How to explain this phenomenon? This could be related to the structure of silane coupling agent. The structures of four different silane coupling agents are listed in Table I, and many differences can be observed for their active groups, such as double bond, amino group, epoxy group, and ester group. In general, the epoxy group can form more intense chemical bond than other active groups between coupling agent and the matrix (Scheme 2), and that is why epoxy silane is the best effective coupling agent for improving the mechanical properties of WF/HDPE composites. On the other hand, the double bond and ester group in allyl ester silane are less stable at high temperature in the processing, which also influences its mechanical properties.

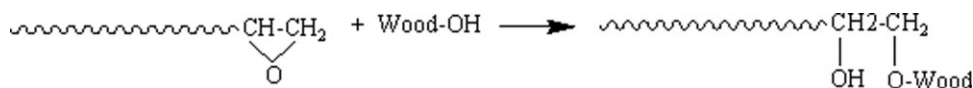
SEM results further indicate that silane coupling agents can improve the interfacial bond between the WF and the matrix. Figure 3 shows the distinct morphology of the fracture surfaces for unmodified [Fig. 3(a)] and silane-modified [Fig. 3(b)] composites, respectively. Besides the relatively smooth surface observed on the WF filler, there are finite gaps near the interfacial region between the PE matrix and WF, indicating the poor adhesion for the unmodified composite [Fig. 3(a)]. Moreover, the unmodified composites also show some fiber pull-out and some clean flake surfaces. However, wood fibers in the composites modified by epoxy silane [Fig. 3(b)] are difficult to differentiate, indicating that the wood fibers are well distributed into the HDPE matrix and the interface between them are improved well. These results also support that suitable silane coupling agent can improve the interfacial bond between the

WF and the matrix and further improve the mechanical properties of wood/HDPE composites.

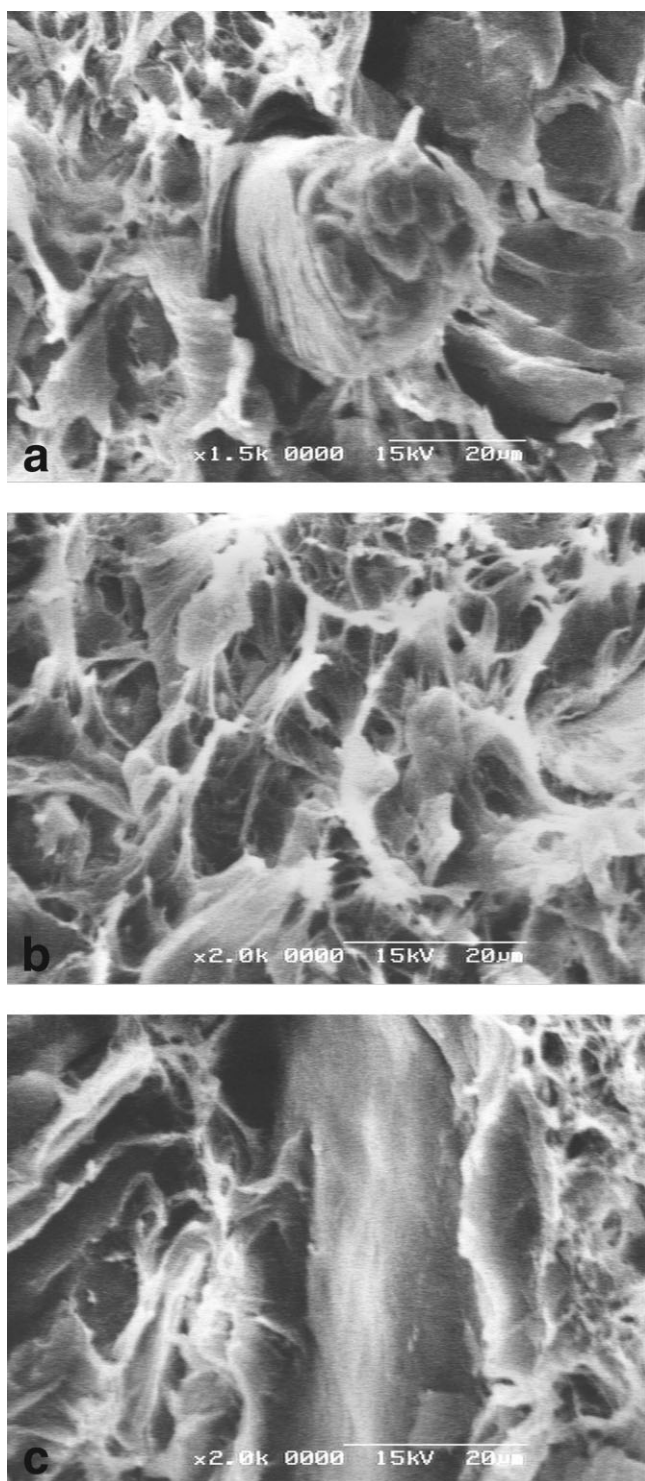
### Effect of polymer compatibilizers

Although silane coupling agents can improve the interfacial bond between the WF and the matrix to some extent, the mechanical properties of the composite are still unsatisfied. To further improve the interfacial adhesion between WF and HDPE matrix, functionalized or polar polymer compatibilizers were also used. The effect of various types of polymer compatibilizers and their contents (Table III) on the mechanical properties of WF/HDPE composites was investigated with WF fixed at 30 phr. As shown in Figure 4, the impact strength of the resultant composites increases with the loading of all kinds of polymer compatibilizer, which indicates that they are effective to improve the interfacial bond between WF and HDPE. Among the compatibilizers, the impact strength of AX8900-treated composites is higher than those with other compatibilizers, especially at the content more than 5 phr. When the content of AX8900 is 11.8 phr, the impact strength of the resultant composite is 4.66 kJ/m<sup>2</sup>, which is increased by 54.2% compared with that of the untreated composites (3.02 kJ/m<sup>2</sup>). The possible mechanism of AX8900 for wood/HDPE composite is shown in Scheme 2.

Contrary to the positive effect on the impact strength, most of the compatibilizers have a negative effect on the tensile strength except HDPE-g-MAH. As shown in Figure 4, the tensile strength of the HDPE-g-MAH-modified composites increases steadily with the increase of HDPE-g-MAH content from 30.31 (untreated composites) to 46.68 MPa (at 11.8 phr), whereas the tensile strength of the other compatibilizer modified composites increases slightly at the low weight percentage of compatibilizer and reaches its maximum at 2.6–3.8 phr compatibilizer



**Scheme 2** Mechanism of the reaction between epoxy group and wood flour.



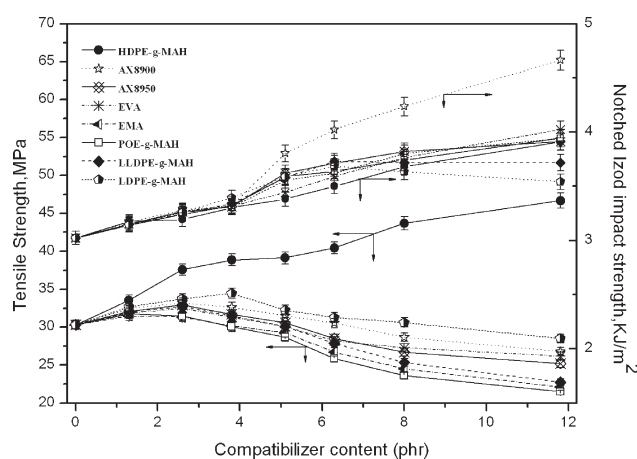
**Figure 3** SEM micrographs of fracture surfaces of WPCs specimens (WF = 30 phr) for (a) an unmodified composite, (b) the composite with 2.5 phr KH560, and (c) the composite with 5.1 phr HDPE-g-MAH.

and then decreases mildly with an increase of compatibilizer content ( $>3.8$  phr). The results could be attributed to the nature and differences of the polymer compatibilizers. First of all, all those compatibilizers can improve the interfacial adhesion, and then

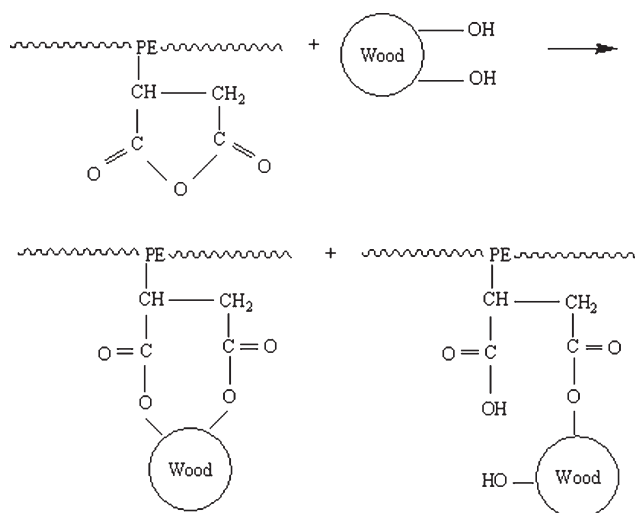
the impact strength and tensile strength could be increased greatly compared with untreated composites. Second, the properties of the resultant composites have a close connection to the nature of compatibilizer. EVA, EMA, AX8900, AX8920, and POE-g-MAH, consisted of many long and soft chemical chains, could decrease the rigid of the composites. Moreover, the tensile strength of LLDPE-g-MAH or LDPE-g-MAH is lower than that of HDPE, so the rigid of the LLDPE-g-MAH or LDPE-g-MAH modified composites decreases at a high weight percentage. However, the nature of HDPE-g-MAH, prepared by HDPE (80064, the same as the HDPE matrix), is similar to the HDPE matrix, so the tensile strength of HDPE-g-MAH modified composites increases with HDPE-g-MAH content increasing because the increasing active groups lead to promote the miscibility between the compatibilizers and the matrix. No clear gaps in the rough interfacial region between the PE matrix and WF also confirm the interface interaction for the HDPE-g-MAH compatibilizer systems [Fig. 3(c)]. The mechanism of MAH group action is shown in Scheme 3.

#### The influence of WF content

Because WF can not only improve the mechanical properties of the composites but also reduce the cost, it is meaningful to study the effect of WF content on the properties of WF/HDPE composites. Furthermore, the aforementioned results indicate that silane coupling agents and functionalized polymer compatibilizers can improve the interfacial bond between WF and HDPE, so epoxy silane and HDPE-g-MAH will be used simultaneously to study the coefficients of them on the mechanical properties of WF/HDPE composites. The effect of WF content on the mechanical properties of modified WPCs is shown in Figure 5. In these experiments, the ratio of

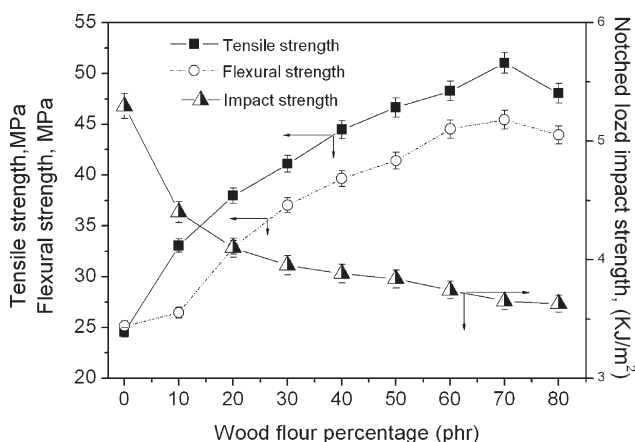


**Figure 4** Mechanical properties of WF/HDPE composites with various types of compatibilizers.



**Scheme 3** Mechanism of the reaction between MAH group and wood flour.

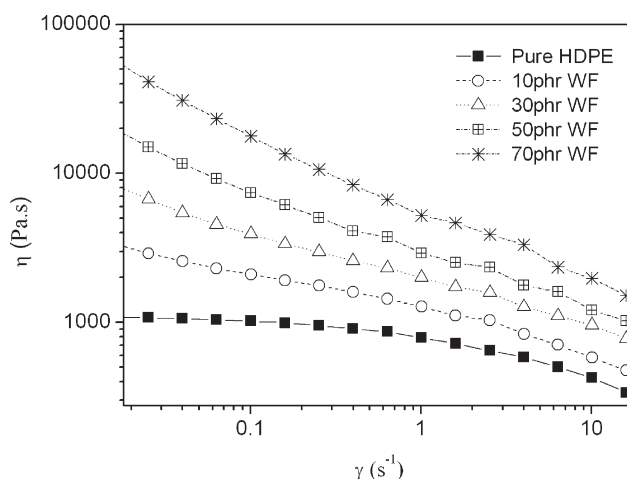
HDPE/HDPE-*g*-MAH is fixed at 5 : 1, and 5 phr epoxy silane is used relative to the WF mass in Table III. It can be seen from Figure 5 that the tensile and flexural strength increase significantly with the increase of WF content, and they are much higher than those of the pure matrix. When the WF percentage is 70 phr, the maximum tensile strength is up to 51.03 MPa, which is increased by 108% compared with pure HDPE matrix (24.68 MPa). The flexural strength also has the same trend as tensile strength. It is well known that the HDPE matrix is a relatively soft plastic with a Young's modulus of 878.4 MPa and tensile strength of 24.68 MPa. Moreover, viewed from the microcosmic aspect, wood fiber in WF has a bunched structure with a high length/diameter ratio and relatively high strength. Therefore, WF could act as a reinforcing filler to strengthen thermoplastics, and both the tensile and flexural strength of the WF/HDPE composites increase with the content increase of WF and reach a maximum at 70 phr,



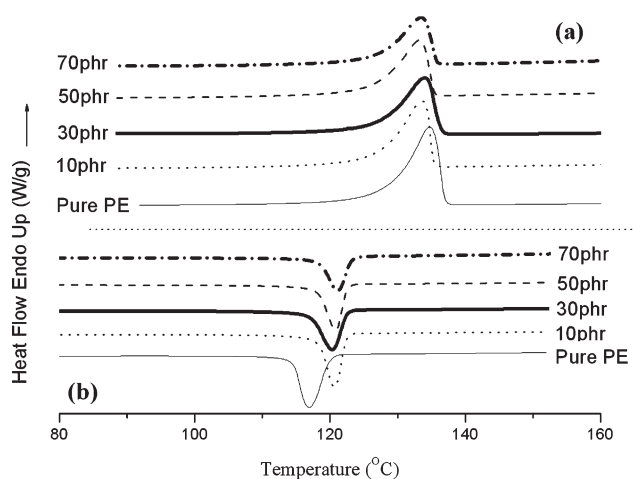
**Figure 5** Effect of wood flour content on the mechanical properties of the resultant composites.

beyond which the strength reduces slightly. On the other hand, the properties of WF/HDPE composites are also relative to the distribution of WF, WF wetting, and HDPE penetration in the processing. Therefore, the viscosity of the composites increases distinctly if the content of WF further increases (more than 70 phr), and this would weaken the interaction between WF and the matrix because of the lower flow of matrix around the WF, and further lead to lower tensile and flexural strengths for the composites. Different from the positive effect on tensile and flexural strength, WF content has negative effect on the impact strength of WPCs. As shown in Figure 5, the impact strength of the resultant composites decreases rapidly with the increase of WF (<30 phr) and then decreases less slightly with further increasing the WF content (>30 phr).

The filler contents of wood plastics are much higher than those for conventional filled plastics that typically are with the filler volume content of ~ 20% or lower. Therefore, the high filler loading results in processing problems, and it is necessary to study the rheology properties of WF/HDPE composites to guide the processing. The effect of WF content on the viscosity of the WF-filled HDPE melts is given in Figure 6. The shear viscosity of all the samples prepared with HDPE-*g*-MAH as a compatibilizer decreases with increasing shear rates and increases with increasing wood content, which is outstanding at low frequencies. In other words, neat HDPE exhibits a Newtonian plateau in the low frequency region and shear thinning at high frequencies, whereas the composites with wood filler exhibit shear thinning at the entire frequency region, and George et al.<sup>30</sup> contributed this phenomenon to fiber orientation. On the other hand, the increased shear thinning in the WF/HDPE composites may probably be due to the high local shear rate in the polymer



**Figure 6** Effect of wood flour content on shear viscosity of WF/HDPE composites.



**Figure 7** DSC result of HDPE with different wood loading levels. (a) Melting curve and (b) crystallization curve.

occupying the space between wood fillers,<sup>31</sup> and the disappearance of Newtonian plateau with the highly filled composites in the entire frequency region may be attributed to yield stress. These phenomena indicate that the viscosity of the system increases sharply with the wood content increasing, and increases the difficulty of processing in return. For the composites with high wood content (>70 phr), there is an obvious sign of extruded surface irregularities and distortions, and the extrudant exhibits typical sharkskin flow instability.

The crystallization and melting behaviors also affect the application and processing of the composites, so melting and crystallization behaviors of pure HDPE and WF/HDPE composites with different WF content are compared in Figure 7, and the results obtained from DSC analyses are summarized in Table IV. Compared to pure HDPE, the melting temperatures ( $T_m$ ) [Fig. 7(a)] of the resultant composites slightly decrease, indicating that the addition of WF does not significantly affect the crystal structure of HDPE in WPC composites. However, as shown in Figure 7(b) and Table IV, the crystallization temperature ( $T_c$ ) of HDPE in the WF/HDPE composites is nearly 4.0°C higher than that of neat HDPE ( $T_c$ , 117.0°C), and then the crystallization temperature increases slightly with the increase of wood content. These results point out that the wood filler presumably acts as a nucleating agent to promote the crys-

tallization of PE, and leads to the composite with a higher number of spherulites of smaller sizes, which is consisted with the reports of Sirisinha.<sup>32</sup> On the other hand, the degree of crystallinity ( $X_c$ ) of HDPE in the composites changes with the content of WF compared with neat HDPE. To compare the crystallinity of only the crystallizable component in each sample, the percentage of crystallinity per unit weight of the crystalline component was calculated using the following relationship:

$$X_c = (\Delta H_f \times M_{\text{composite}}) / (\Delta H_f^i \times M_{\text{HDPE}}) \times 100\% \quad (1)$$

where  $\Delta H_f$  is the observed heat of fusion values and  $\Delta H_f^i$  is the ideal value of 100% crystalline HDPE (270.03 J/g),  $M_{\text{HDPE}}$  is the mass of HDPE include neat HDPE and HDPE-g-MAH in the composite, and  $M_{\text{composites}}$  is the mass of the composites. From Table IV, it can be seen that  $X_c$  increases with the increase of WF content and then decreases if WF content exceeds 70 phr, which further confirms the nucleating role of WF.

The HDT is the temperature at which a polymer or plastic sample deforms under a specified load, and this is a very important property for a given plastic material applied in many aspects of product design, engineering, and manufacture of products using thermoplastic components. So, HDT was also studied in this work and the results are listed in Table IV. The HDT of the composites increases with the increase of WF loading and reaches to 63.1°C when WF content is 70 phr, indicating that WPCs have a better heat resistant performance than that of pure HDPE (37.2°C). This property would endow WF/HDPE composites wide applications.

## CONCLUSIONS

In this article, HDPE/WF composites were prepared by a twin-screw extruder. The effects of silane coupling agents, polymer compatibilizers and their contents, and WF content on the mechanical properties, morphology, rheology, and crystalline behavior of the WF/HDPE composites have been investigated in detail. Some important results were obtained later:

1. Epoxy silane coupling agent could provide the greatest enhancement in the tensile strength, flexural strength, and impact resistance. SEM

**TABLE IV**  
DSC Parameters and HDT of WPC Composites with Different Wood contents

Sample	$T_m$ (°C)	$T_c$ (°C)	$\Delta H_f$ (J/g)	$X_c$ (%)	HDT (°C)
Pure HDPE	134.7 ± 0.1	117.0 ± 0.2	209.3 ± 0.3	77.5 ± 0.1	37.2 ± 0.1
HDPE/HDPE-g-MAH/wood/80/20/10	133.8 ± 0.2	120.8 ± 0.1	198.5 ± 0.2	80.9 ± 0.2	43.8 ± 0.1
HDPE/HDPE-g-MAH/wood/80/20/30	134.2 ± 0.1	120.5 ± 0.3	177.3 ± 0.2	85.4 ± 0.2	54.2 ± 0.2
HDPE/HDPE-g-MAH/wood/80/20/50	133.3 ± 0.3	120.9 ± 0.2	154.6 ± 0.3	86.0 ± 0.2	60.7 ± 0.1
HDPE/HDPE-g-MAH/wood/80/20/70	133.5 ± 0.2	121.0 ± 0.3	125.8 ± 0.1	79.2 ± 0.1	63.1 ± 0.2



analysis indicated that the silane coupling agents could improve the adhesion between wood filler and HDPE matrix.

2. Among eight types of polymer compatibilizers, AX8900 had the best effects on the impact strength and HDPE-g-MAH was more effective in improving the tensile and flexural strength of WF/HDPE composites because of the chemical reaction between epoxy group or maleic anhydride and hydroxyl groups of WF.
3. WF could improve the tensile and flexural properties but had a negative effect on the impact properties. The tensile strength of the composites with 70 phr WF is up to 51.03 MPa twice the strength of pure HDPE (24.68 MPa). And the viscosity of the composites increased with increasing WF loading.
4. Both  $T_c$  and  $X_c$  increased with the increase of WF content, indicating that WF acted as a nucleating agent promoting the crystallization of HDPE.
5. HDT of composites was much improved compared with that of pure HDPE, and it increased with the WF loading increasing. HDT was up to 63.1°C when WF content was 70 phr, which was increased to about 26°C compared with that of pure HDPE.

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