

## Preparation of Soft Wood–Plastic Composites

Jiaqi Lu,<sup>1</sup> Rongrong Qi,<sup>1</sup> Xinli Hu,<sup>1</sup> Yu Luo,<sup>1</sup> Jieyu Jin,<sup>1</sup> Pingkai Jiang<sup>2</sup>

<sup>1</sup>School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University, Shanghai 200240, China

<sup>2</sup>Shanghai Key Lab of Electrical Insulation and Thermal Aging, Department of Polymer Science and Engineering, Shanghai Jiao Tong University, Shanghai 200240, China

Correspondence to: R. Qi (E-mail: rraqi@sjtu.edu.cn)

**ABSTRACT:** To extend the applications of wood–plastic composites (WPCs), soft WPCs were prepared with ethylene vinyl acetate (EVA) and wood flour (WF) as major components via a two-step process involving two-roll mixing and compression molding. The effect of the various factors, such as WF, unsaturated fatty acid (UFA), and dicumyl peroxide contents, on the mechanical properties, processability, and morphology of the WPCs was investigated in detail. The addition of UFA could effectively improve the processability and flexibility of the composites. The *in situ* grafting reaction between UFA and EVA considerably improved the adhesion of the WF and resin matrix. Consequently, soft WPCs with good performances in flexibility and processability were successfully obtained. © 2013 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 130: 39–46, 2013

**KEYWORDS:** composites; fibers; thermoplastics

Received 13 September 2012; accepted 7 December 2012; published online 8 March 2013

**DOI:** 10.1002/app.38916

### INTRODUCTION

Wood–plastic composites (WPCs) are usually formulated from a blend of natural fiber, for example, wood flour (WF), fruit hull powder, bamboo powder, or crop straw stalk powder, and thermoplastic polymers, such as polypropylene (PP) and polyesters,<sup>1</sup> and they have gained considerable attention in recent years because of their environmental benefits and low cost. On the other hand, their properties of low moisture absorption, resistance to biological attack, dimensional stability, high specific stiffness and strength, less abrasion during processing, and low density<sup>2,3</sup> make them have wide applications in windows and door frames, interior panels in cars, railings, decking, cladding and fences among others.<sup>4</sup> However, the hydrophilicity of wood flour and the hydrophobicity of plastic make the dispersion and interfacial adhesion between them difficult; this leads further to rather poor mechanical properties of the composites.<sup>5,6</sup> Thus, many studies have been done to overcome these drawbacks. For example, Renner et al.<sup>7</sup> used maleic anhydride modified polypropylene (MAPP) as a compatibilizer to improve the interfacial adhesion between PP and Bengtsson et al.<sup>8</sup> used vinyl trimethoxysilane to improve the adhesion between WF and high-density polyethylene (PE). Ashori and Nourbakhsh<sup>9,10</sup> studied the reinforcing effects of nanoparticles on rice straw/PP composites and WF/PP composites. They found that the physicochemical properties of WPCs could be significantly improved with an appropriate combination of coupling agent content and nanoclay loading in the com-

posites. We also improved the compatibility between WF and high-density PE composites through the grafting of silane by an *in situ* reaction extrusion process.<sup>11</sup> Although coupling agents can effectively improve the interaction between the WF and the matrix, the agglomeration of fillers in the obtained composites<sup>12</sup> and difficult processing due to the high viscosity at high filler contents<sup>13,14</sup> are still the main problems to be solved.

In general, thermoplastics are used as matrices in the manufacturing of WPCs; these include PE,<sup>15,16</sup> PP,<sup>17,18</sup> polystyrene,<sup>19</sup> and poly(vinyl chloride),<sup>20,21</sup> and the obtained WPCs usually have properties of high strength and hardness and low toughness. However, soft WPCs with a certain of softness and flexibility will have important applications in some special fields, such as indoor floors and automotive interiors.<sup>22</sup> To the best of our knowledge, few studies have been reported on these WPCs up to this point.

Ethylene vinyl acetate (EVA) is a kind of polymer with outstanding low-temperature flexibility, toughness, and good adhesion to a variety of materials, including plastics and wood,<sup>23</sup> and their properties are influenced by their composition; for example, an increased content of ethylene promotes mechanical strength and block resistance, whereas an increased content of vinyl acetate (VA) provides higher flexibility and adhesion and better low-temperature performance.<sup>24</sup> The works of Dikobe and Luyt<sup>25</sup> indicated that the interfacial adhesion between WF and EVA was better than those of other traditional WPCs

because there might have been hydrogen bonding between the —OH groups on the cellulose and the —C=O groups in the VA. Malunka et al.<sup>26</sup> prepared EVA–sisal fiber composites with better physical properties, and they observed that dicumyl peroxide (DCP) was effective in the grafting of EVA to sisal fiber during the initiated crosslinking process.

Unsaturated fatty acid (UFA) is a kind of low-molecular-weight molecule with rich carboxyl groups and unsaturated double bonds; it can soak WF and graft onto the EVA backbone through the initiation of DCP and further improve the interfacial action between the WF and resin matrix. In this study, UFA was used as a plasticizer, and soft WPCs with good flexibility were prepared with EVA as the matrix and WF as the filler. The effects of the various factors, such as the contents of WF, UFA, and DCP, on the morphological, mechanical, rheological, and hardness properties of the EVA/WF composites were also investigated in detail.

## EXPERIMENTAL

### Materials

EVA with a 26.5% VA content was obtained from BASF-YPC Co., Ltd. (Yangzhou, China). It had a melt flow index of about 6.0 g/10 min and a specific gravity of 0.9 g/cm<sup>3</sup>. Pine WF (nominal 100 mesh, moisture content <8%, specific gravity ≈0.24 g/cm<sup>3</sup>) was supplied by Lin Pai WF Co., Ltd. (Huzhou, China); its composition included cellulose (42 ± 2%), hemicellulose (27 ± 2%), lignin (28 ± 2%), and volatile matter (3 ± 1%). DCP as an initiator was obtained from Shanghai Chemical Reagent Co. (Shanghai, China). UFA was derived from soybean oil (supplied by Golden Dragon Fish Co., Ltd., Shanghai, China).

### Composite Preparation

WF was dried in a vacuum oven at 80 ± 2°C for 24 h to remove water and low-molecular-weight organic materials before the preparation of the samples.<sup>27,28</sup> The components of the samples are shown in Table I. All of the samples in Table I were prepared by the mixture of EVA, WF, and other additives on a SK-160 two-roll mill (Shanghai Sinan Rubber Machinery Works, Shanghai, China) at 80°C for 15 min; the mixture was then pressed on a compression-molding machine (Shunli Rubber Machinery Co., Ltd., Huzhou, China). The preprepared mixtures were poured into four steel square molds with dimensions of 80 × 80 × 1 mm<sup>3</sup> preheated at a temperature of 140°C for 10 min before they were compression-molded under a pressure of 7.5 MPa at a mold temperature of 140°C for 6 min; they were then cooled for 5 min to obtain the EVA/WF composite sheets.

### Characterization

**Mechanical Properties.** The tensile properties were determined according to ISO 527-2 on an Instron testing machine (Instron 4465, Instron Corp., Boston, MA). The tests were performed at a crosshead speed of 10 mm/min. All samples were dumbbell-shaped with dimensions of 1 × 4 × 80 mm<sup>3</sup> and were preconditioned at 23°C and 50% humidity for 24 h. At least five replicate specimens were tested for each formulation.

**Hardness Properties.** The hardness of the samples was measured according to ISO 868 on a LX-D durometer (Aotuo

**Table I.** Formulas of the EVA/WF Composites

| EVA (phr) | WF (phr) | UFA (phr) | DCP (phr) |
|-----------|----------|-----------|-----------|
| 100       | 0        | 0         | 0         |
| 100       | 40       | 0         | 0         |
| 100       | 60       | 0         | 0         |
| 100       | 80       | 0         | 0         |
| 100       | 100      | 0         | 0         |
| 100       | 120      | 0         | 0         |
| 100       | 80       | 2         | 0         |
| 100       | 80       | 5         | 0         |
| 100       | 80       | 10        | 0         |
| 100       | 80       | 15        | 0         |
| 100       | 80       | 20        | 0         |
| 100       | 80       | 20        | 0.05      |
| 100       | 80       | 20        | 0.1       |
| 100       | 80       | 20        | 0.5       |
| 100       | 80       | 20        | 1.0       |

Instruments, Wuxi, China) and is expressed as shore D hardness.

**Rheological Measurement.** The rheological characteristics of the EVA/WF composites were measured on an AR-G2 rheometer (TA Instruments, New Castle, PA). The diameter of the plate was 25 mm, and the gap between the two plates was 1.0 mm. The studies were carried out at 160°C with scanning frequencies from 0.01 to 100 Hz.

**Fourier Transform Infrared (FTIR) Spectra.** The FTIR spectra of the EVA, UFA, and UFA/EVA composites were recorded on an FTIR spectrophotometer (Spectrum 100, PerkinElmer, Inc., Waltham, MA). Both the sample and KBr (at a ratio of ~1 : 20) were homogeneously ground in a mortar, and then, 4–10 mg of the mixture was taken and pressed in a hydraulic press to make the pellets.

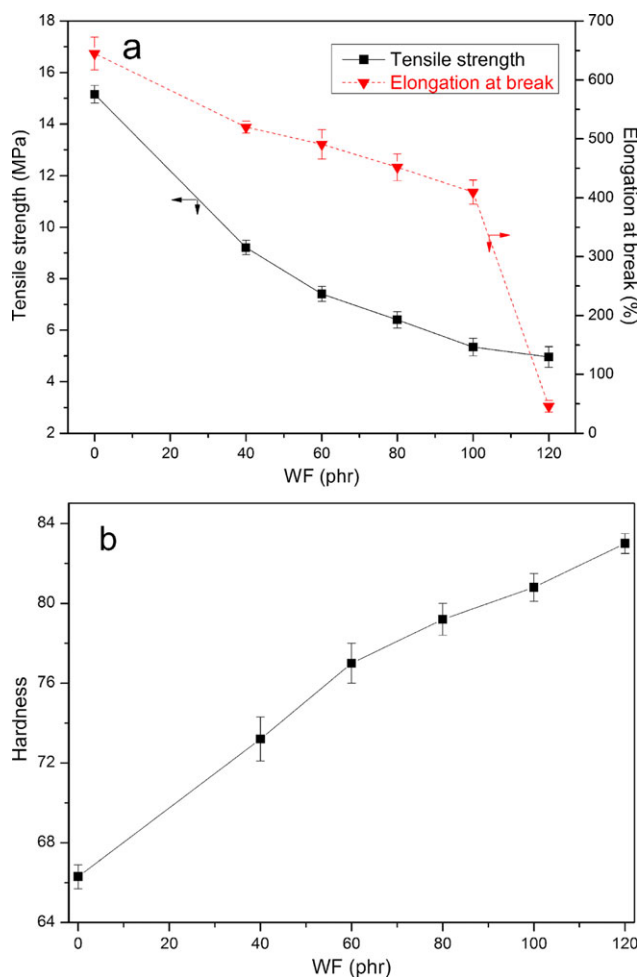
**Scanning Electron Microscopy (SEM).** SEM of the composites was conducted on a Hitachi S-2150 SEM instrument (Hitachi High-Technologies Corp., Tokyo, Japan) with an accelerated voltage of 15 kV. The samples were broken in liquid nitrogen to prevent any deformation of the samples and were then sputter-coated with a thin layer of gold.

## RESULTS AND DISCUSSION

### Influence of the WF Content

Because of the low density, biodegradability, renewability, and relatively low cost of WF, we examined a high content of WF in the composites. However, the poor interfacial adhesion between the WF and the matrix and the difficulty in processing because of the high viscosity at the high filler content, hindered production of very high filler content composites. The effect of the WF content on the properties of the composites was studied first.

Our experiments revealed that the processing was smooth when the content of WF was less than 120 phr, whereas a higher content of WF led to processing difficulties. So the influence of the



**Figure 1.** (a) Tensile properties and (b) hardness of the EVA/WF composites as a function of the WF content.

WF content (0–120 phr) on the tensile properties was investigated. From Figure 1(a), one can see that the tensile strength of the composites decreased obviously with increasing WF content, especially at low contents (40–80 phr), and then, there was no significant change in strength with further increases in the WF content. However, the elongation at break of the composites decreased marginally with increasing WF content at low content, whereas it nearly vertically dropped above 100 phr (from 400% at 100 phr to 50% at 120 phr). The decrease in the tensile strength and elongation at break of the composites was attributed to WF agglomeration and the formation of voids,<sup>29</sup> and this resulted in a stress concentration region and the subsequent coalescence of the voids into large voids, which lead to brittle fracture at a low stress level. The elongation at break of the composites maintained a high level at low WF content may have been due to fact that the EVA interacted well with WF, so that the WF particles could move with the polymer matrix under large deformation, and the flexible chain could convert stress through stress relaxation. On the other hand, the hardness of the EVA/WF composites increased with increasing WF [Figure 1(b)] because of the higher hardness of WF, and the reinforcement of WF also restricted the migration of the

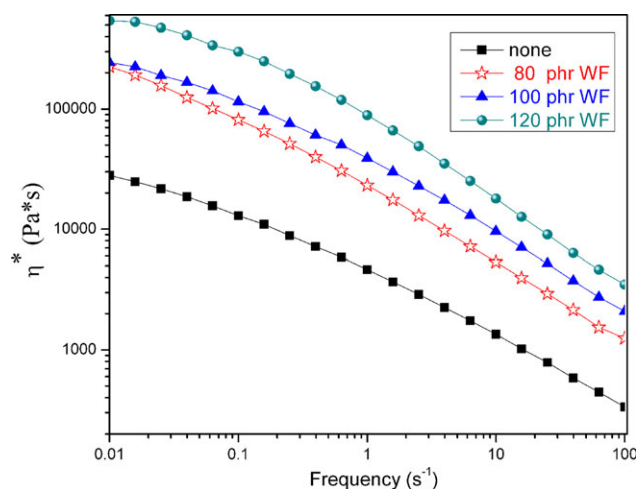
polymer chain and led to a decrease in the flexibility of the composites.

In this study, the filler contents of the WPCs were much higher than those of conventional filled plastics, which typically have filler volume contents of about 30% or lower.<sup>30</sup> Therefore, the high filler loading resulted in processing problems, and it was necessary to study the rheological properties of the composites. As shown in Figure 2, the complex viscosity ( $\eta^*$ ) of the composites increased as the loading of WF increased because WF did not melt and its surface roughness resisted flow deformation; similar results were derived from previous studies.<sup>31</sup> On the other hand, the  $\eta^*$  values of all of the blends decreased significantly as the shear rate increased; this helped us to elucidate a shear-thinning or pseudoplastic flow of the EVA/WF composites in all of the frequency ranges measured. In general, the shear-thinning behavior of composites is related to the viscoelastic behavior of the matrix and the linkage between polymer and particles/clusters.<sup>32</sup> At high frequencies, the polymer chains did not have enough time to recover the original filler particle distribution, and this resulted in WF orientation and a decrease in the viscosity.

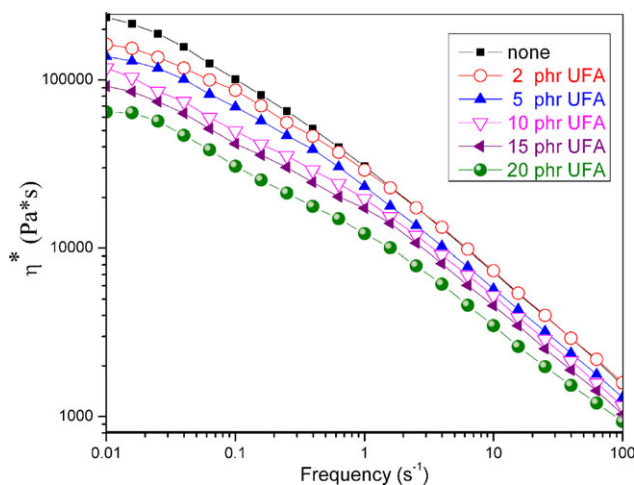
Taking into account the mechanical and rheological properties, 100/80 EVA/WF was chosen as appropriate formula for the following studies.

#### Influence of UFA

Compared to the neat EVA resin, the composites with added experienced declines in their flexibility and processing ability. In the past few years, some plasticizers, such as dioctyl phthalate and liquid paraffin,<sup>33</sup> have been used to improve composites. In this study, UFA was used. The rich carboxyl groups of UFA caused it to interact well with the WF particles through hydrogen bonding with the hydroxyl radical groups on the WF; this was beneficial to the dispersion of WF particles and the processing of composites.<sup>34</sup> On the other hand, UFA is a kind of low-molecular molecule with a long hydrophobic carbon chain, which fits easily into polymer chains and twists with the EVA matrix.<sup>35</sup> The previous synchronous effects of UFA improve the processing of composites. Furthermore, the low molecular



**Figure 2.**  $\eta^*$  of the EVA/WF composites as a function of the frequency.



**Figure 3.**  $\eta^*$  of the EVA/WF (100/80) composites as a function of the frequency.

weight of UFA also weakened the attractive forces between the EVA chains, increase the free volume and segmental motions, and result in a lower hardness. The effect of the UFA content on the properties of the EVA/WF composites was studied (Table I).

The effect of the UFA content on the  $\eta^*$  values of WPCs was investigated by rotational rheometry. One can see that all of the composites showed shear-thinning behavior (Figure 3), and a decrease in  $\eta^*$  took place with increasing UFA content in the WPCs from 2 to 20 phr; this implied that the addition of UFA could effectively improve the processing behavior of the composites.

Furthermore, we also found that the hardness of the obtained EVA/WF composites decreased with increasing UFA concentration [Figure 4(a)]. This was because the stiffness of materials was due to weak bonds between the composites, for example, Van der Waals forces, hydrogen bonding, and crystalline structure, and the addition of UFA weakened these interactions and spaced them apart. This allowed the polymer molecules to move more freely and made it softer. On the other hand, low-molecular-weight UFA also acted as a lubricant and weakened the interfacial friction between the polymer chain and the WF particle; this was also beneficial for decreasing the viscosity of the composites and softening them.

The influence of the UFA content on the tensile properties of the WPC blends is shown in Figure 4(b), and the addition of UFA led to a slight decrease in the tensile strength of the composites and had a feeble influence on the elongation at break. This phenomenon was in accordance with the hardness discussed before, in which the effective plasticization of UFA resulted in a decrease in the tensile strength of the composites.

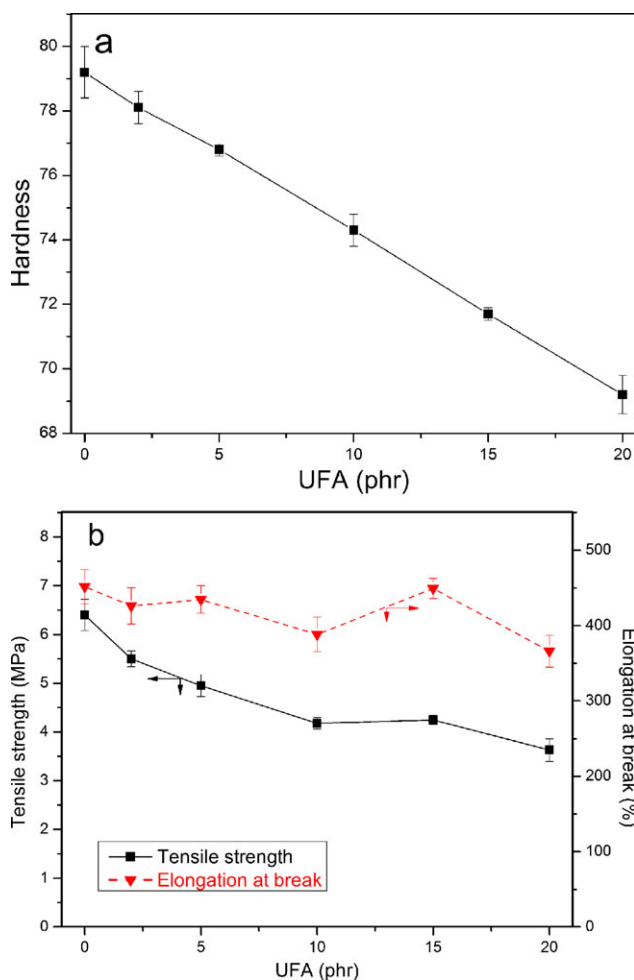
#### Influence of the DCP Concentration

The previous works revealed that the addition of UFA could effectively improve the processability of the composites and soften them because of its low molecular weight. Nevertheless, the addition of UFA harmed the tensile strength of the compo-

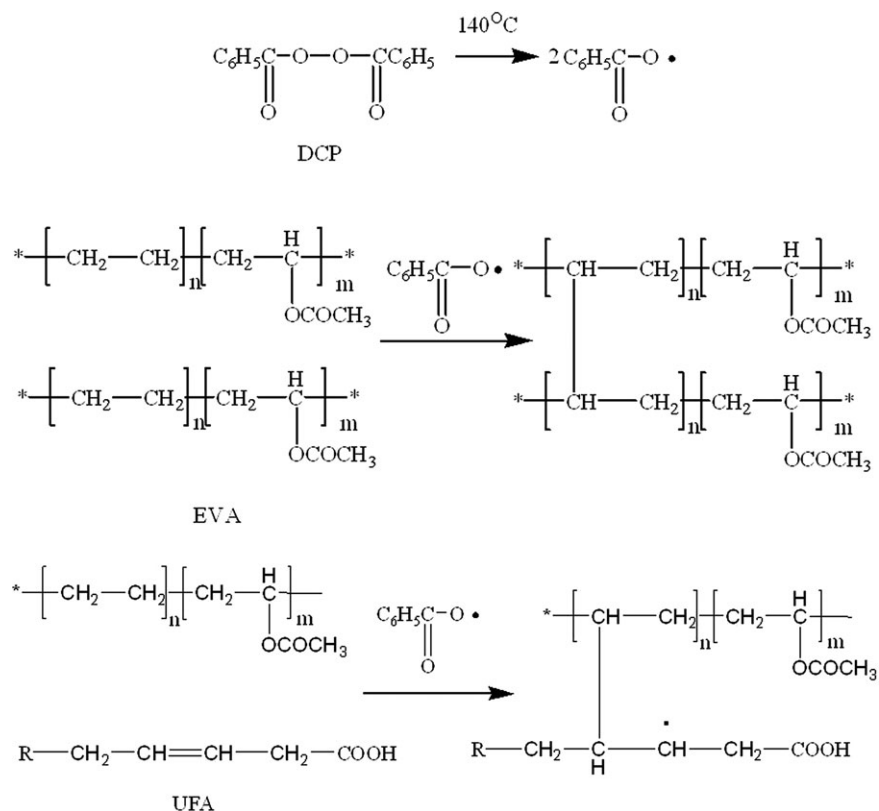
sites to some degree. On the other hand, UFA was rich in unsaturated double bonds; in addition, the carboxyl group on UFA could form hydrogen bonds with the hydroxyl radical groups on WF. The unsaturated double bonds in UFA could be grafted onto the EVA backbone through the initiation of DCP, which also improved the interfacial action between the WF and EVA matrix and further improved the mechanical properties of the WPCs.

The grafting reactions between EVA and UFA initiated by DCP are described in Scheme 1. The initiator DCP decomposed to oxy-free radicals at a certain temperature, and then, the newly formed oxy radicals attacked EVA to release hydrogen from the EVA chains and caused a crosslinking reaction. Meanwhile, the newly formed oxy radicals also attacked the double bonds of UFA and converted them into free radicals. Then, UFA could be grafted onto the EVA backbone. This reaction was verified by the FTIR spectra.

The FTIR spectra of EVA, UFA, and UFA-grafted EVA (the samples were extracted in boiling acetone for 48 h to remove the unreacted UFA) are shown in Figure 5. In the spectrum of EVA [Figure 5(A-a)], the absorption peaks at 2930, 2856, and 1468



**Figure 4.** (a) Hardness and (b) tensile properties of the EVA/WF composites as a function of the UFA content.



Scheme 1. Grafting reactions between EVA and UFA initiated by DCP.

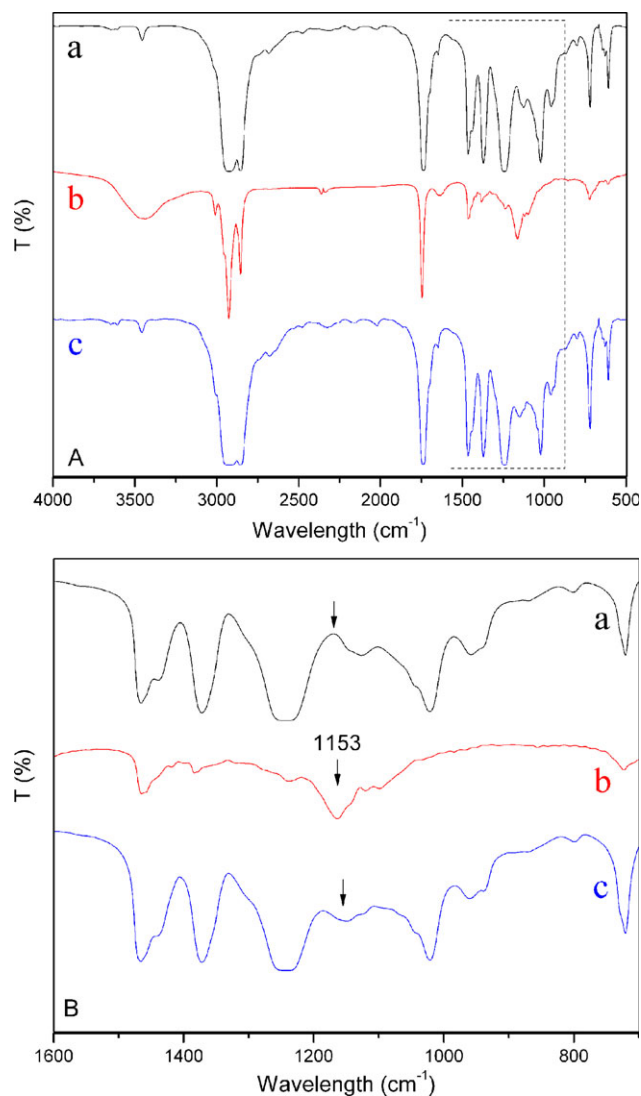
$\text{cm}^{-1}$  were assigned to the asymmetric, symmetric, and scissor modes of  $-\text{CH}_2$  stretching in the methylene chains, respectively.<sup>36</sup> The absorbance at  $1743 \text{ cm}^{-1}$  was attributed to  $\text{C}=\text{O}$  stretching, and those at  $1238$ ,  $1025$ , and  $608 \text{ cm}^{-1}$  were due to the symmetric and asymmetric  $\text{C}-\text{O}-\text{C}$  stretching of ester groups in the VA regions.<sup>37</sup> In the FTIR spectrum of UFA [Figure 5(A-b)], there were bands at  $3453 \text{ cm}^{-1}$  for  $-\text{OH}$  stretching;  $3004 \text{ cm}^{-1}$  for the stretching vibrations of  $=\text{CH}$ ;  $2930$ ,  $2856$ , and  $1468 \text{ cm}^{-1}$  for the asymmetric, symmetric, and scissor modes of  $-\text{CH}_2$  stretching;  $1749 \text{ cm}^{-1}$  for  $\text{C}=\text{O}$  stretching; and  $1166 \text{ cm}^{-1}$  for the  $\text{C}-\text{O}-\text{C}$  stretching of ester groups in unsaturated triglyceride. The UFA-grafted EVA [Figure 5(A-c)] exhibited peaks at  $2930$ ,  $2856$ , and  $1468 \text{ cm}^{-1}$  for  $-\text{CH}_2$  stretching;  $1743 \text{ cm}^{-1}$  for  $\text{C}=\text{O}$  stretching; and  $1238$ ,  $1025$ , and  $608 \text{ cm}^{-1}$  for the  $\text{C}-\text{O}-\text{C}$  stretching of ester groups in VA regions that were identical to those in the EVA sample. To more clearly illustrate the differences between the spectra of EVA and the UFA-grafted EVA, a partial enlarged drawing of Figure 5(A) (from  $700$  to  $1600 \text{ cm}^{-1}$ ) is presented in Figure 5(B). The appearance of peak at  $1153 \text{ cm}^{-1}$  was associated with the  $\text{C}-\text{O}-\text{C}$  stretching of ester groups in UFA; this proved that UFA was grafted onto the EVA backbone successfully.

The influence of the DCP concentration on the mechanical properties is shown in Figure 6. One can see that the tensile strength of the EVA/WF composites increased slightly at a low DCP concentration and then sharply reached a maximum value of  $8.5 \text{ MPa}$  at  $0.5\%$  DCP; the elongation of the composites decreased radically when only  $0.1 \text{ phr}$  of DCP was added and

then maintained no changes. These results imply that the grafting reaction between UFA and EVA occurred; this improved the interaction between the WF and EVA matrix and further improved the tensile strength of the composites. On the other hand, the crosslinking reaction of the composites<sup>38</sup> also had a great influence on them. Only slight changes in the tensile properties were observed for the crosslinked samples at low DCP concentrations because there were a large number of uncrosslinked regions existing within the crosslinked polymers, whereas the elongation at break significantly declined because the formed crosslink network restricted the movement of the polymer chains. With increasing DCP concentration, the tensile strength of the composites increased because the density of the crosslink network increased, and this further restricted the movement and flexibility of the polymer chains.

However, the DCP content had little influence on the hardness of the WPCs [Figure 6(b)], and the hardness of the composites increased slightly with increasing DCP concentration. The improvement was due to the decrease in the mobility of the polymer chains caused by the crossing reaction and the increase in interaction between the WF and polymer caused by the grafting reaction initiated by DCP, as explained earlier. Nevertheless, the composites still had good flexibility and could be easily curled.

Figure 7 shows the morphology of the composites. From Figure 7(a), one can see that the interfacial boundary between EVA and WF was indistinct, even though without any UFA, fewer



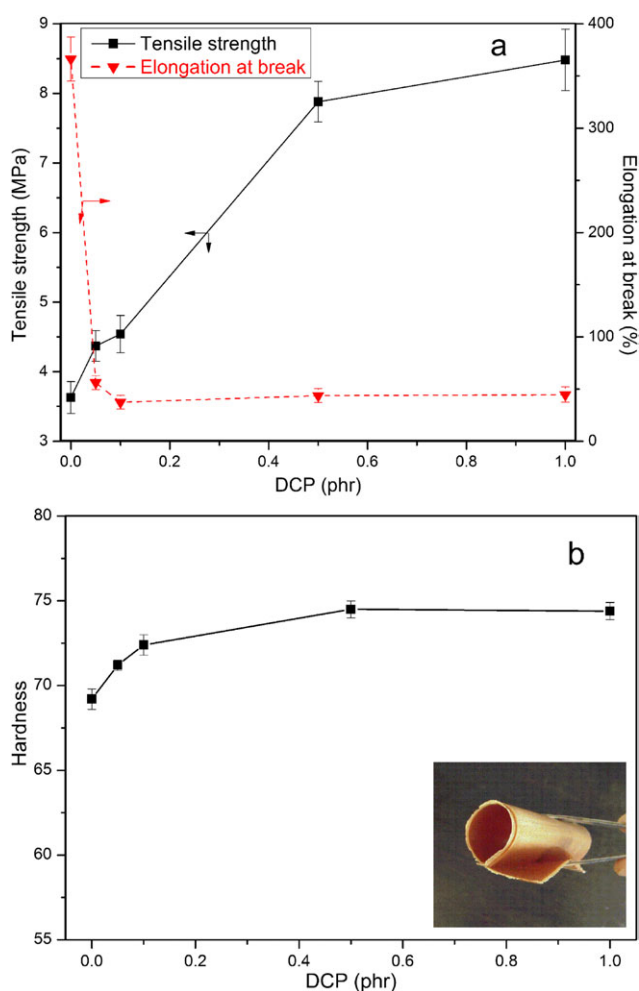
**Figure 5.** (A) FTIR spectra of (a) EVA, (b) UFA, and (c) UFA grafting EVA. (B) Partial enlarged drawing of part A (from 700 to 1600  $\text{cm}^{-1}$ ).

voids and cavities could be observed on the fracture surface; this implied better interfacial adhesion between WF and EVA because of the hydrogen bonding existing between the  $-\text{OH}$  groups on WF and the  $-\text{C}=\text{O}$  groups in VA. Although the agglomerates of WF particles were visible on the fracture surface, this was also consistent with the poor elongation at break and tensile strength in the EVA/WF composites. When UFA was added, an indistinct interfacial boundary between EVA and WF was still observed [Figure 7(b)], but the distribution of WF became more homogeneous. This also implied that UFA improved the compatibility between WF and the matrix effectively. From the fracture surface of the composites with UFA and DCP [Figure 7(c)], we observed that WF was embedded within EVA matrix; this implied that a grafting reaction existed between EVA and UFA, and it was effective in improving the wettability and interfacial interaction between the WF and EVA matrix. This was in accord with the improvement in the tensile strength.

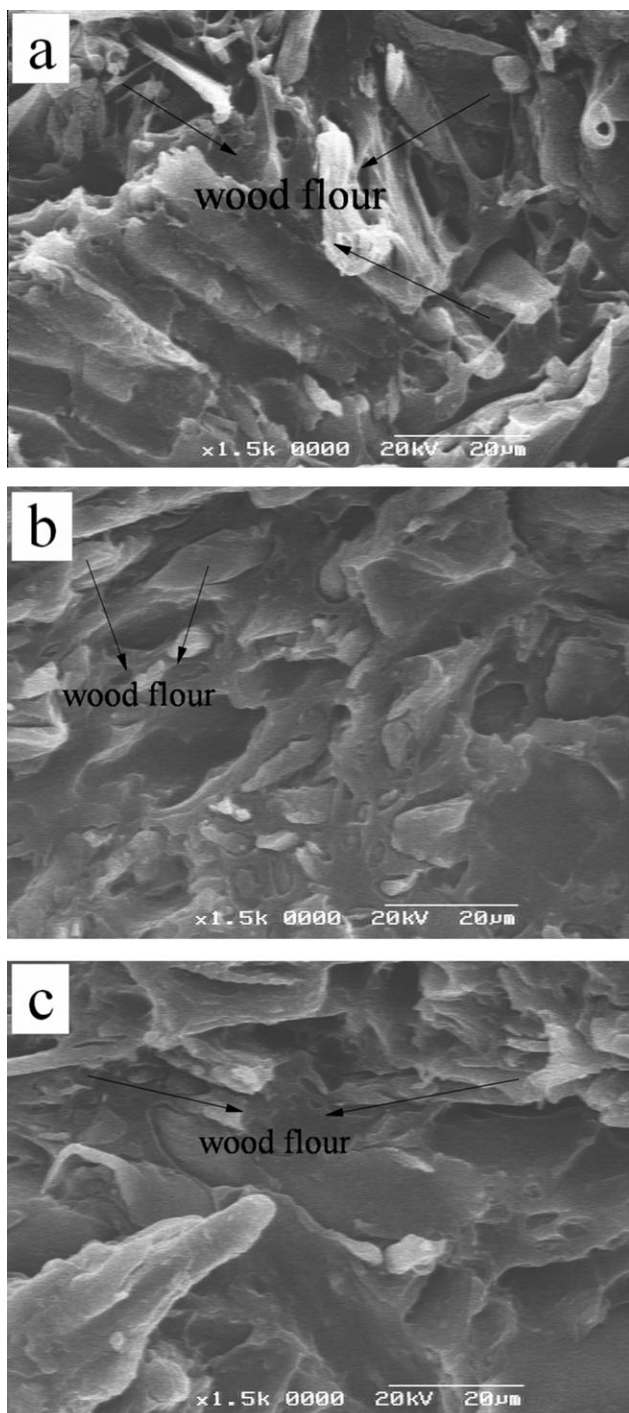
## CONCLUSIONS

A kind of soft wood–plastic composite was successfully prepared through the mixture of EVA and WF by a two-roll mill and a vulcanizing press. The effects of various factors on the properties of the EVA/WF composites were investigated in detail:

1. EVA/WF composites prepared without any interfacial modifiers showed a slight decline in the tensile properties at low WF contents. Nevertheless, highly filled WPCs presented difficulty in processing and poor properties.
2. UFA improved the interfacial interactions between the WF and EVA matrix effectively, and the addition of UFA reduced the hardness and viscosity of the EVA/WF composites; this was beneficial in the preparation of soft and flexible WPCs and improved the processability of the composites.
3. The grafting reaction between UFA and EVA could be initiated by DCP, and the mechanical properties of the EVA/WF composites were effectively improved, whereas the obtained composites still had good flexibility and could be easily curled.



**Figure 6.** (a) Tensile properties and (b) hardness of the EVA/WF (100/80) composites as a function of the DCP content.



**Figure 7.** SEM micrographs of the fracture surfaces of the EVA/WF (100/80) composites for (a) none, (b) 20 phr UFA, and (c) 20 phr UFA and 0.5 phr DCP.

#### ACKNOWLEDGMENTS

The work described in this article was supported by the National Science Foundation of China (contract grant numbers 21173145 and 51133003) and Shanghai Leading Academic Discipline Project (contract grant number B202).

#### REFERENCES

- Malkapuram, R.; Kumar, V.; Negi, Y. S. *J. Reinf. Plast. Compos.* **2009**, *28*, 1169.
- La Mantia, F. P.; Morreale, M. *Compos. A* **2011**, *42*, 579.
- Kalia, S.; Kaith, B. S.; Kaur, I. *Polym. Eng. Sci.* **2009**, *49*, 1253.
- Valente, M.; Sarasini, F.; Marra, F.; Tirillò, J.; Pulci, G. *Compos. A* **2011**, *42*, 649.
- Xie, Y. J.; Hill, C. A. S.; Xiao, Z. F.; Militz, H.; Mai, C. *Compos. A* **2010**, *41*, 806.
- Cui, J. Q.; Mei, C. T.; Jia, C.; Zhou, Z. B.; Zhang, Y. *Polym. Bull.* **2011**, *67*, 375.
- Renner, K.; Kenyó, C.; Móczó, J.; Pukánszky, B. *Compos. A* **2010**, *41*, 1653.
- Bengtsson, M.; Gatenholm, P.; Oksman, K. *Compos. Sci. Technol.* **2005**, *65*, 1468.
- Ashori, A.; Nourbakhsh, A. *Eur. J. Wood Prod.* **2011**, *69*, 663.
- Ashori, A. *J. Compos. Mater.* **2013**, *47*, 149.
- Xiong, C.; Qi, R. R.; Gong, W. J. *Polym. Adv. Technol.* **2009**, *20*, 273.
- Morreale, M.; Scaffaro, R.; Maio, A.; La Mantia, F. P. *Compos. A* **2008**, *39*, 503.
- Li, T. Q.; Wolcott, M. P. *Polym. Eng. Sci.* **2005**, *45*, 549.
- Li, T. Q.; Wolcott, M. P. *Polym. Eng. Sci.* **2006**, *46*, 114.
- Xiong, C.; Qi, R. R.; Wang, Y. L. *J. Appl. Polym. Sci.* **2009**, *114*, 1160.
- Grubbströmn, G.; Holmgren, A.; Oksman, K. *Compos. A* **2010**, *41*, 678.
- Ashori, A.; Nourbakhsh, A. *Bioresour. Technol.* **2010**, *101*, 2515.
- Kim, T. W.; Lee, S. Y.; Chun, S. J. *J. Compos. Mater.* **2011**, *15*, 1595.
- Mansour, S. H.; El-Nashar, D. E.; Abd-El-Messieh, S. L. *J. Appl. Polym. Sci.* **2006**, *102*, 5861.
- Marathe, D. S.; Joshi, P. S. *J. Appl. Polym. Sci.* **2009**, *114*, 90.
- Chaochanchaikul, K.; Sombatsompop, N. *Polym. Eng. Sci.* **2011**, *51*, 1354.
- Ashori, A. *Bioresour. Technol.* **2008**, *99*, 4661.
- Chen, X. M.; Zhong, H.; Jia, L. Q. *Int. J. Adhes. Adhes.* **2001**, *21*, 221.
- Tlili, R.; Cecen, V.; Krupa, I. *Polym. Compos.* **2011**, *32*, 727.
- Dikobe, D. G.; Luyt, A. S. *J. Appl. Polym. Sci.* **2007**, *104*, 3206.
- Malunka, M. E.; Luyt, A. S.; Krump, H. *J. Appl. Polym. Sci.* **2006**, *100*, 1607.
- Ou, R. X.; Zhao, H.; Sui, S. J.; Song, Y. M.; Wang, Q. W. *Compos. A* **2010**, *41*, 1272.
- Pilla, S.; Gong, S. Q.; O'Neill, E.; Yang, L. Q.; Rowell, R. M. *J. Appl. Polym. Sci.* **2009**, *111*, 37.

29. Huang, H. X.; Zhang, J. J. *J. Appl. Polym. Sci.* **2009**, *111*, 2806.
30. Clemons, C. M.; Sabo, R. C.; Hirth, K. C. *J. Appl. Polym. Sci.* **2011**, *120*, 2292.
31. Wang, Y. L.; Qi, R. R.; Xiong, C.; Huang, M. *Iran. Polym. J.* **2011**, *20*, 281.
32. Ares, A.; Bouza, R.; Pardo, S. G.; Abad, M. J.; Barral, L. *J. Polym. Environ.* **2010**, *18*, 318.
33. Feldman, D.; Banu, D.; Manley, R. S. J.; Zhu, H. *J. Appl. Polym. Sci.* **2003**, *89*, 2000.
34. Adhikary, K. B.; Park, C. B.; Islam, M. R.; Rizvi, G. M. *J. Thermoplast. Compos.* **2011**, *24*, 155.
35. Turhan, K. N.; Sancak, Z. O. E.; Ayana, B.; Erdogdu, F. J. *Food Process. Eng.* **2007**, *30*, 485.
36. Li, H.; Tripp, C. P. *Langmuir.* **2002**, *18*, 9441.
37. Barrueso-Martinez, M. L.; Ferrandiz-Gomez, T. D.; Romero-Sanchez, M. D.; Martin-Martinez, J. M. *J. Adhes.* **2003**, *79*, 805.
38. Sirisinha, K.; Meksawat, D. *Polym. Int.* **2005**, *54*, 1014.