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RESEARCH ARTICLE

Wenjie DANG, Yongming SONG, Qingwen WANG, Weihong WANG

Improvement in compatibility and mechanical properties of modified wood fiber/polypropylene composites

Abstract To improve the interfacial compatibility between wood fibers and polypropylene and the toughness of wood-fiber/polypropylene composites, maleic anhydride grafted polypropylene (PP-g-MAH) and maleic anhydride grafted styrene-ethylene-butadiene-styrene copolymers (SEBS-g-MAH) were used as modifiers. Mechanical properties of wood-fiber/polypropylene (WF/PP) composites were improved when PP-g-MAH or SEBS-g-MAH was added. When either of these copolymers was added, the composites had better interfacial compatibility than the unmodified composite. This was verified by scanning electron microscope (SEM) observations and dynamic mechanical analysis (DMA). The mechanical properties of the composites were significantly improved because of the good interfacial bonding between wood fibers and polypropylene when PP-g-MAH and SEBS-g-MAH were added.

Keywords wood fiber, polypropylene, PP-g-MAH, SEBS-g-MAH

1 Introduction

Wood fiber (flour)/thermoplastic composites have developed rapidly because of their high cost-effectiveness and advantages in recycling (Clemons, 2002; Winandy et al., 2004). However, the dispersion of wood fibers in a plastic matrix and their mechanical properties are not satisfactory because of poor interfacial compatibility between wood fibers and thermoplastics (Oksman and Clemons, 1998). This compatibility of wood with plastic composites is critical. Wood fiber surfaces are usually treated by physical or chemical methods to improve the quality of surfaces, or by adding a coupling agent to improve interfacial compatibility (Raj et al., 1989; Wu et al., 2000). Both maleic anhydride grafted polypropylene (PP-g-MAH) and maleic anhydride grafted styrene-ethylene-butadiene-styrene copolymers (SEBS-g-MAH) are non-polar molecular chains attached to polar groups could be used as coupling agents between polar and non-polar materials. The SEBS segments of SEBS-g-MAH have thermoplastic properties of non-hydrogenated SBS and high flexibility under high temperatures (Gao and She, 2004). SEBS-g-MAH has multi-functions, including being both a hardener and a coupling agent. We studied the performance of WF/PP composites when PP-g-MAH or SEBS-g-MAH was added and verified the interfacial compatibility with scanning electron microscope (SEM) observations and dynamic mechanical analysis (DMA).

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Wenjie DANG (✉)
China Wood Conservation and Substitution Development Center, Beijing 100834, China
E-mail: wjdang@163.com

Yongming SONG, Qingwen WANG, Weihong WANG
Key Laboratory of Bio-based Material Science and Technology of Ministry of Education, Northeast Forestry University, Harbin 150040, China

2 Materials and methods

2.1 Materials

Polypropylene (T30S) was provided by the Daqing Petrochemical Company, China. Its melting index was 3.02 g per 10 min.

Wood fibers from a poplar species (particle size 500–2500 μm) were obtained by screening.

A commercially available maleic anhydride-polypropylene copolymer (PP-g-MAH) was used as a coupling agent to improve the compatibility and bonding strength between the wood fibers and the polypropylene agent. Maleic anhydride grafted styrene-ethylene-butadiene-styrene copolymer (SEBS-g-MAH) was used as a toughener.

2.2 Equipment and methods

The following equipment were used in our tests: an SJSH30/SJ45 dual-band extruder (Najing), a mechanical

testing machine (RGT-20A, Shenzhen), an impact testing machine (XJ-50G, Hebei), a dynamic mechanical analyzer (NeTZSCH Gerätebau GmbH DMA 242, Germany), a high-speed mixer (SHR-10A) and a scanning electron microscope (SEM) (FEI QUNGTA200, USA).

Tensile and flexural tests were performed at a test speed of 2 mm/min given the guidelines of GB/T1040-1992 and GB/T9341-2000 (The State Bureau of Quality and Technical Supervision, 1992, 2000). Impact tests were carried out using five unnotched samples according to GB/T1043-1993 (The State Bureau of Quality and Technical Supervision, 1993).

Viscoelastic performances were investigated with a frequency of 1 Hz using a dynamic mechanical analyzer (NeTZSCH Gerätebau GmbH DMA 242).

The morphology of the wood fiber reinforced PP composites with and without PP-g-MAH or SEBS-g-MAH were investigated using the scanning electron microscope (SEM) (FEI QUNGTA200). Fractured surfaces of flexural test samples were studied with the SEM after being coated with gold.

2.3 Compounding and sample preparation

2.3.1 Fiber screening

Wood fibers, passing through a 40-mesh sieve, were used. Short fibers are beneficial to a uniform distribution.

2.3.2 Fiber dying

Screened wood fibers were oven dried (DHG-9140) for 2 h and moisture content was controlled at about 3%.

2.3.3 Compounding

Wood fibers and polypropylene were mixed in a high-speed mixer (SHR-10A) at pre-determined ratios (Table 1).

Table 1 Formulation of wood fiber/polypropylene composites (unit: %)

experiment No.	wood fiber	polypropylene	PP-g-MAH	SEBS-g-MAH
1	30	70	–	–
2	30	67	3	–
3	30	65	5	–
4	30	62	8	–
5	30	65	–	5
6	30	62	–	8
7	30	60	–	10

2.3.4 Sample preparation

The mixed wood fibers and polypropylene were processed by a SJSH30/SJ45 extruder at temperatures between 155°C and 190°C. This material was ready for use after one day. Table 1 shows the formulation of wood fiber/polypropylene composites.

3 Results and discussion

3.1 Effect of change in PP-g-MAH on mechanical properties of composites

PP-g-MAH, a widely applied grafted copolymer, was introduced as a modifier to enhance the compatibility between fibers and polypropylene. Both tensile and flexural strength of the resultant WF/PP composites with PP-g-MAH are shown in Fig. 1. In general, the wood fiber polypropylene composites showed an increase in the strength of mechanical properties with the addition of increased amounts of PP-g-MAH (Qin, 2002). Tensile strength improved 44.3%, while flexural strength improved 18.4% when 5% PP-g-MAH was added. Mechanical properties of the composites improved slowly when PP-g-MAH content was added at a lower level (Zang et al., 2001). However, mechanical properties decreased when the amount of PP-g-MAH added was greater than 8%, which shows that there is a limit to the amount of PP-g-MAH that can be added (Guo et al., 2002). The addition of excessive amounts is not necessary and in fact counterproductive. Pure PP-g-MAH has a lower rupture strength than pure polypropylene (Zhang et al., 2004). If excessive amounts of PP-g-MAH were added, the composites would break at the weak interface layer where PP-g-MAH would be concentrated and lead to decreased mechanical properties.

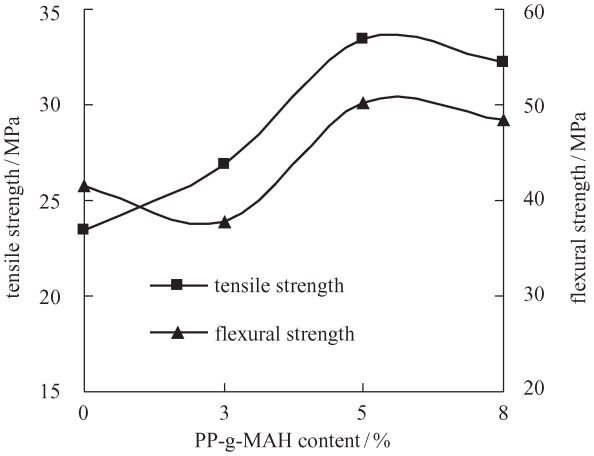


Fig. 1 Tensile and flexural properties of composites with different PP-g-MAH components

Unnotched impact strengths of WF/PP composites with PP-g-MAH are shown in Fig. 2. The figure also illustrates that improved interfacial compatibility with increased unnotched impact strength (nearly 39%) resulted from

the addition of an amount with 5% PP-g-MAH. Similar to tensile strength, unnotched impact strength decreased when PP-g-MAH was added at 8%.

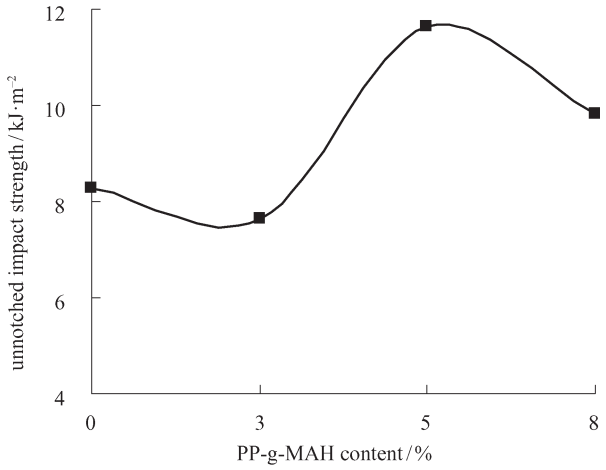


Fig. 2 Unnotched impact strength of composites with different PP-g-MAH components

3.2 Effect of change in SEBS-g-MAH on mechanical properties of composites

Figure 3 shows the variation of tensile and flexural strength of wood fiber-polypropylene composites when SEBS-g-MAH is added. The increase of tensile strength indicates that SEBS-g-MAH enhanced the interfacial compatibility of wood fiber-polypropylene composites up to 5% SEBS-g-MAH. When the amount of SEBS-g-MAH increased, tensile strength was reduced because of the low tensile strength of the copolymer SEBS-g-MAH and stress concentration (Du et al., 2002). It is important to control the amount of SEBS-g-MAH added.

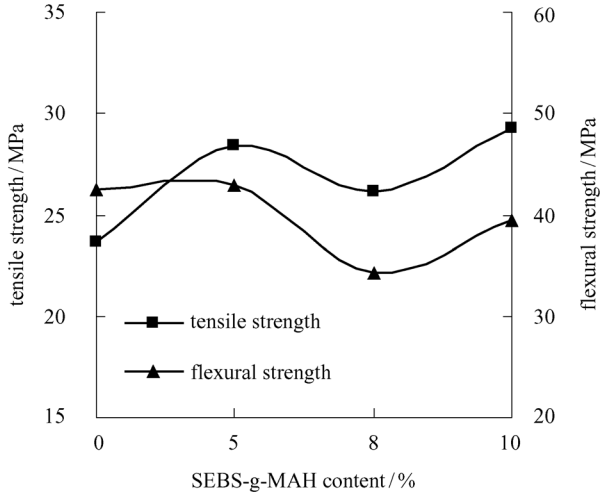


Fig. 3 Tensile and flexural properties of composites with different SEBS-g-MAH contents

Figure 4 shows that the unnotched impact strength of WF/PP composites increased with higher amounts of SEBS-g-MAH. A modest amount added to the flexible interface layer could thus improve impact properties, due to the existence of SEBS-g-MAH, between wood fibers and polypropylene. When the amount of SEBS-g-MAH added was 5%, the unnotched impact strength of WF/PP composites improved nearly 55.7%. With an additional 10% of the copolymer, the impact strength increased by 81.7%.

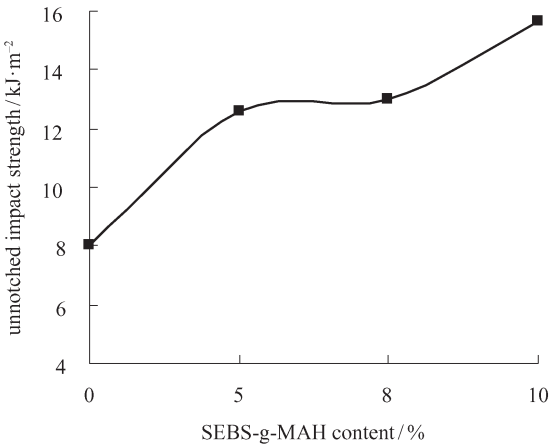


Fig. 4 Unnotched impact strength of composites with different SEBS-g-MAH contents

3.3 Dynamic mechanical analysis of wood fiber-polypropylene composites

A dynamic mechanical analysis can show the actual performance of materials in service compared with a static mechanical analysis (Hristov and Vasileva, 2003). Figure 5 shows the storage modulus (E') of WF/PP composites without a coupling agent, with 5% PP-g-MAH and with 5% SEBS-g-MAH respectively. The storage modulus of these three kinds of composites was reduced when the temperature increased because of molecular movement. The storage modulus was rapidly reduced when the temperature reached the glass transition temperature (T_g) at about $-7°C$.

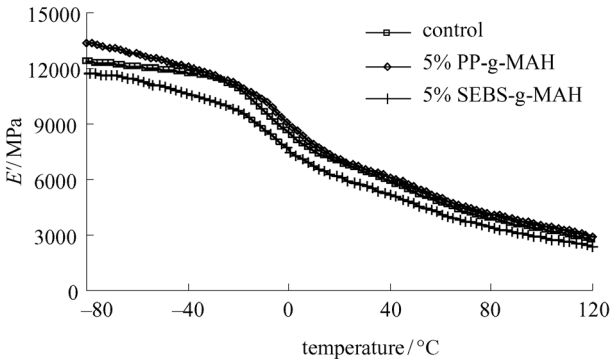


Fig. 5 Temperature spectra of storage modulus of composites with different coupling agents

The storage modulus of the composite with 5% PP-g-MAH was higher than that of the control sample, which indicated that the stiffness of the composite improved at the glass transition temperature of polypropylene. This was attributed to the limited chain movement of polypropylene due to the wood fibers, while the ester formed between wood fibers and polypropylene. The storage modulus of the composite with 5% SEBS-g-MAH was reduced over the temperature range because the SEBS chain of SEBS-g-MAH has a low storage modulus. However, the flexible SEBS of SEBS-g-MAH improved the toughness as indicated in Fig. 4, in which the unnotched impact properties of the composites increased.

Figure 6 shows that the loss factor (tanδ) of WF/PP composites changes with temperature. Over the temperature range analyzed, the first transition is related to unrestricted amorphous PP chains. Compared to the control sample, both 5% MAPP-g-MAH and 5% SEBS-g-MAH reduced the value of tanδ, which suggests that both PP-g-MAH and SEBS-g-MAH could improve the interfacial compatibility of wood fiber with polypropylene, i.e., the friction loss was reduced when the interfacial compatibility of composites improved.

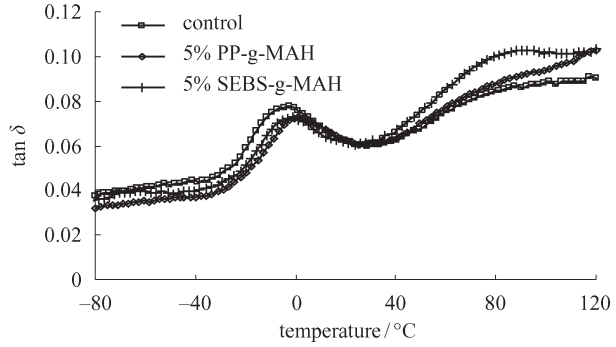


Fig. 6 Temperature spectra of mechanical loss factor of composites with different coupling agents

The composite with 5% SEBS-g-MAH had the highest tanδ value, at 80°C, which indicates it had good damping properties and unnotched impact properties. It is consistent with having an increase of unnotched impact properties in static mechanical analysis.

3.4 Micrographs of wood fiber-polypropylene composites

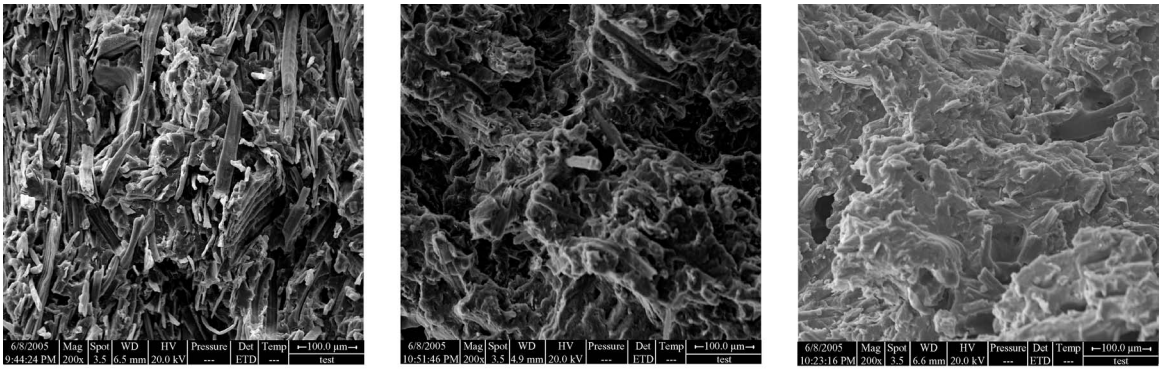
The flexural fractured surfaces of the unmodified control sample and the modified WF/PP composites were examined with the SEM. The results are presented in Fig. 7. Figure 7(a) shows a large number of fibers pulled out, debonding and fibrillation from the unmodified control sample. That more fibers were pulled out in the unmodified WF/PP composites is due to the poor bonding strength between wood fibers and the polymer matrix. Figures 7(b) and 7(c) represent the microstructure of modified wood fiber-polypropylene composites. They show better interaction between the wood fibers and the PP matrix (Qin and Yan, 1999). This indicates that maleic anhydride provides good bonding between wood fibers and polypropylene — a behavior consistent with mechanical properties.

4 Conclusions

The wood fiber-polypropylene composite with 5% MAH-PP shows better tensile and flexural properties, as well as unnotched impact properties compared with those of the control sample. Unnotched impact properties improved by about 55.7% when 5% SEBS-g-MAH was added to the wood fiber-polypropylene composite.

The wood fiber-polypropylene composite with 5% PP-g-MAH had a higher storage modulus and lower tanδ value compared with the control sample, which indicates a good interfacial interaction between wood fibers and polypropylene.

The composites with 5% SEBS-g-MAH had the highest tanδ value at 80°C compared with the control sample,



(a) control (×200) (b) 5% PP-g-MAH (×200) (c) 5% SEBS-g-MAH (×200)

Fig. 7 SEM micrograph of the tensile fractured surface of composites for different coupling agents

which indicates that this kind composite has good damping properties and unnotched impact properties.

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