RHEOLOGICAL BEHAVIOR OF HDPE-WOOD FLOUR COMPOSITES WITH DIFFERENT WOOD CONTENTS*

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Abstract

Rheological behavior of high-density polyethylene (HDPE)-wood flour composites with different wood contents was examined using steady shear sweep, oscillatory frequency sweep, creep test, and stress relaxation test. The samples were prepared using a melt compounding method in a twin-screw extruder. The results showed that the yield stress did not appear for the composites prepared under the processing conditions used in this work. Oscillatory frequency sweep showed that both storage and loss moduli of the composites increased with increasing wood content. However, they exhibited a plateau for the composites with wood contents higher than 30% at low frequency. The creep test showed that the composites with high wood content were difficult to deform. The relaxation modulus of the composites obtained from stress relaxation test increased with increasing the wood content.

Keywords: HDPE-wood flour composites; rheological behavior; melt compounding

Introduction

Wood-plastic composites (WPC) have been studied extensively in recent years, due to the attractive benefits they offer, such as increased durability, dimensional stability, and low maintenance [1–4]. An important reason of these composites being widely used in decking and building industry is the economical competitiveness of wood flour. Wood flour has relatively low density compared to inorganic filler, which provides a capacity for manufacturing composites with higher content of wood at lower costs.

However, high wood content results in processability problem of WPC. Since the rheological properties have been widely used to assess the processibility of filled polymers, this study investigates the rheological behavior of WPC.

Due to different flow behavior of filler and polymer and the complex interaction between two phases, the rheological behavior of the composites is complex. Several researchers [5–8] have investigated the rheological behavior of WPC. Li et al. [5] investigated the nonlinear feature of HDPE-wood composites through both oscillatory shear and step shear tests. The oscillatory tests showed nonlinear storage moduli at the strain levels far below the values for neat matrix polymer. The step shear tests revealed the time-strain factorizability for the shear relaxation modulus. Xiao et al. [6] found the melt elasticity of the composites decreased with increasing wood content from the creep tests. The strain response showed a sudden decrease at 50% wood content, indicating the composites was very inelastic. Maiti et al. [7] used the first normal stress difference as an indicator of melt elasticity. It was found that the elasticity initially decreased up to a wood volume content value of 9.2 % at a low shear rate, then increased with wood content. At a higher shear rate, the melt elasticity increased by 6–32%.

In this study, the rheological behavior of the HDPE-wood composites with different wood

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contents was investigated using steady shear sweep, oscillatory frequency sweep, creep test, and stress relaxation test.

Experimental

Materials

The plastics matrix used was HDPE (grade 5300B, Sinopec Group Daqing Co.) with a melt index of 0.41 g/10min (190℃ and 2.16 kg). The wood flour had a 100-mesh size. A commercially available titanate was used as a coupling agent.

Sample Preparation

The wood flour was dried at 80 ℃ in a vacuum oven for 12 h. Then titanate with a content of 5 wt% of the wood flour was added to the wood flour to improve the compatibility and adhesion between the wood flour and the HDPE matrix. The HDPE and wood flour were compounding using a TSE-20 co-rotating twin-screw extruder (Ruiya Company, China) with a screw diameter of 20 mm and a length to screw diameter ratio of 42. The barrel temperature was set at 80-120-150-160 -170-170-170℃ (toward the die). The screw speed was 200 rpm. The feed rate was set at 1 kg/h. The extruded strands were cooled in air and palletized. The pellets were dried at 80℃ for 12 h, and then were compression molded into a disk with a size of $\phi 25\times 1$ mm at 170℃ and 10 MPa for 20 min.

Rheological Measurement

The above-prepared disk was used to measure the rheological properties of the composites. The measurement, including steady shear sweep, oscillatory frequency sweep, creep test, and stress relaxation test, was conducted using Bohlin Gemini 200 Rheometer System with parallel plate of 25 mm diameter at 170℃. Among them, steady shear sweep was performed at a controlled strain mode from 0.001 to 0.1 s^{-1}. To measure the storage modulus (G'), loss modulus (G''), and damping factor ($\tan\delta$), oscillatory frequency sweep was performed from 100 to 0.01 s^{-1} at a strain level of 1% which ensured the measurement was carried out in linear region. Then creep and stress relaxation tests were carried out to further examine the viscoelastic properties of the composites. In the creep test, a constant stress of 1000 Pa was applied to the samples and held for 150 s, then the stress was removed and the strain was monitored over time. In the stress relaxation test, the sample was subjected to a rapidly applied strain of 10% which was then held for the remainder time of the test. The relaxation behavior was then studied by monitoring the steadily decreasing value of shear stress.

Results and Discussion

Shear Stress-Shear Rate Curve

Figure 1 shows the steady shear sweep data of the composites with different wood contents at a controlled strain mode. It is evident that the stress increases with increasing wood content. In order to examine whether the yield stress exists for the HDPE-wood composites prepared in this work, the shear stress-shear rate curves shown in Figure 1 were fitted using the Herschel-Bulkley equation (1).

$$\sigma = \sigma_y + K\dot{\gamma}^n \qquad (1)$$

where σ_y is the yield stress, K and n are material parameters. The fitted results show the values of σ_y for all the composites are negative, indicating the yield stress do not appear for the composites prepared under the processing conditions used in this work. The result is different from that by Xiao et al. [6] who observed the yield stress for the composite with a 50% wood content. The result in this work demonstrates that the HDPE-wood composites bahavior as a pseudo-plastic flow.

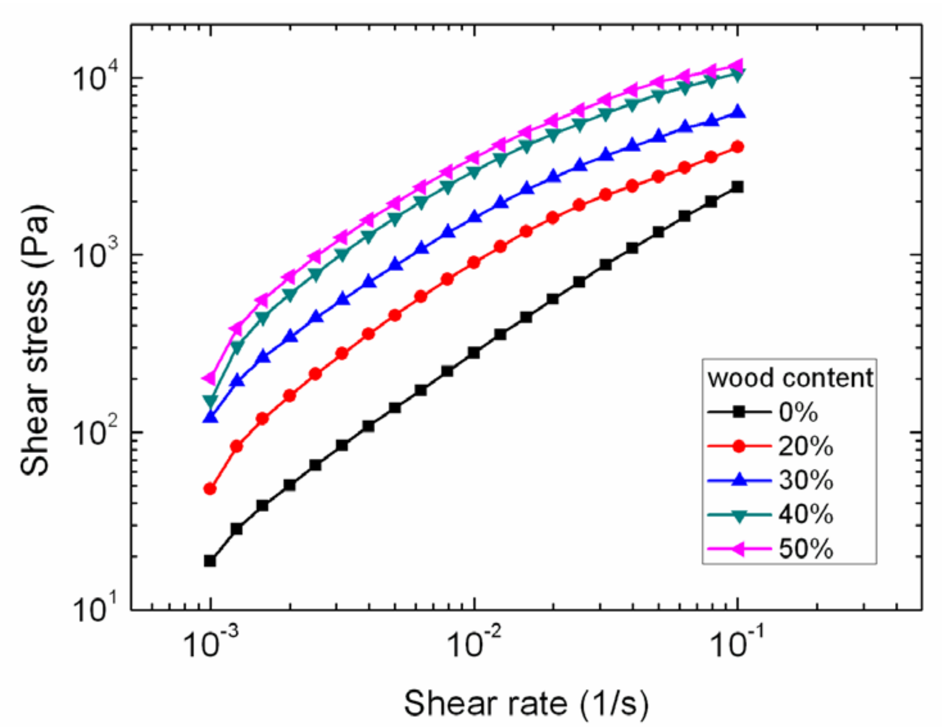
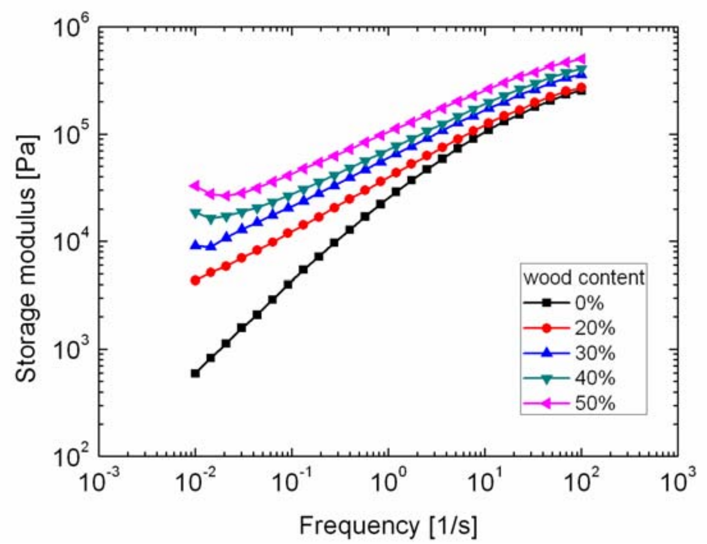


Figure 1. Steady Shear Sweep Data of HDPE-Wood Composites.

Storage Modulus, Loss Modulus, and Damping Factor

Figure 2 shows the oscillatory frequency sweep results of different wood content composites. As can be seen, both storage modulus (G') and loss modulus (G'') increase with increasing wood content and frequency. It is interesting to note that the storage and loss moduli of the composites with wood contents higher than 30% exhibit a plateau at the low frequency. The results agree with the report of Li et al. [5] who also observed a low frequency plateau for composite with a wood content of 60%. The $\tan\delta$ value for neat HDPE decreases dramatically with increasing frequency, but it is less dependent on the frequency for the composites. Moreover, increasing the wood content leads to the decrease of the $\tan\delta$. It is well known that $\tan\delta$ is defined as the ratio of G'' to G'. So the decrease of the $\tan\delta$ means that the storage modulus increases more than loss modulus with increasing wood content.



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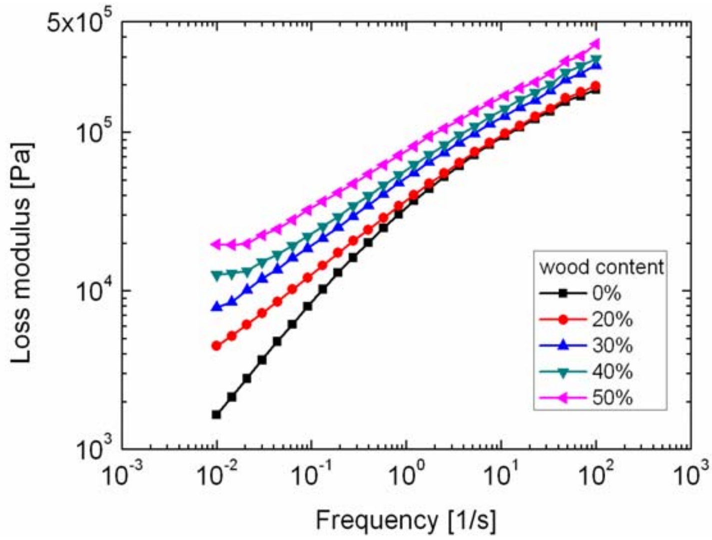

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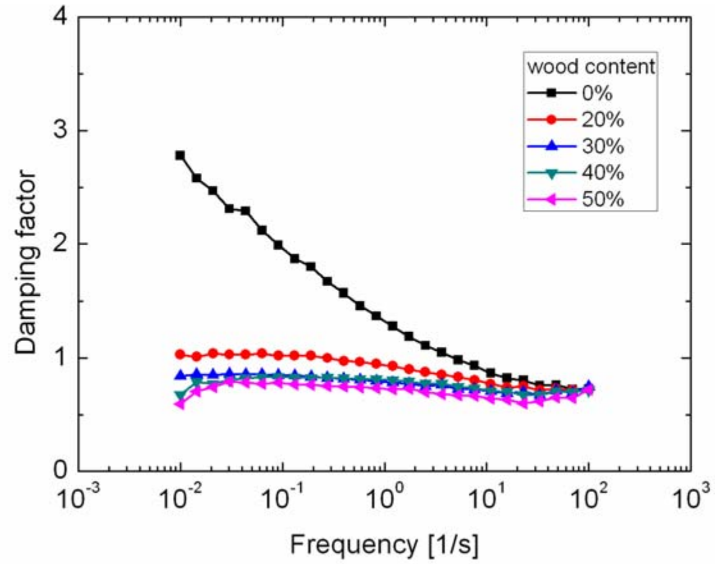

c

Figure 2. Storage Modulus (a), Loss Modulus (b) and Damping Factor (c) of HDPE-Wood Composites.

Strain-Time Curve

To further examine the effect of wood content on the viscoelastic properties of the composites, creep test was performed. The obtained strain versus time curves are plotted in Figure 4. As can be seen, the strain response for the composites shows a sudden drop, indicating the wood has a profound effect on the viscoelastic behavior of the polymer.

It can be seen from Figure 4 that the slope of initial strain curves decreases with increasing the wood content. This may be attributed to the fact that strain behavior is mainly influenced by the elastic character of the composites at initial time when the stress is applied, whilst the elastic character of the composites increases with increasing the wood content. After 150 s when the stress is removed, the strain of the composites decreases slightly with increasing time due to the gradual relaxation of the molecular chain of the polymer. After 500 s, the strain of the composites exhibits a plateau. This may be attributed to the fact that the elastic character of the composites has been consumed and the strain behavior is mainly influenced by the viscous character of the composites.

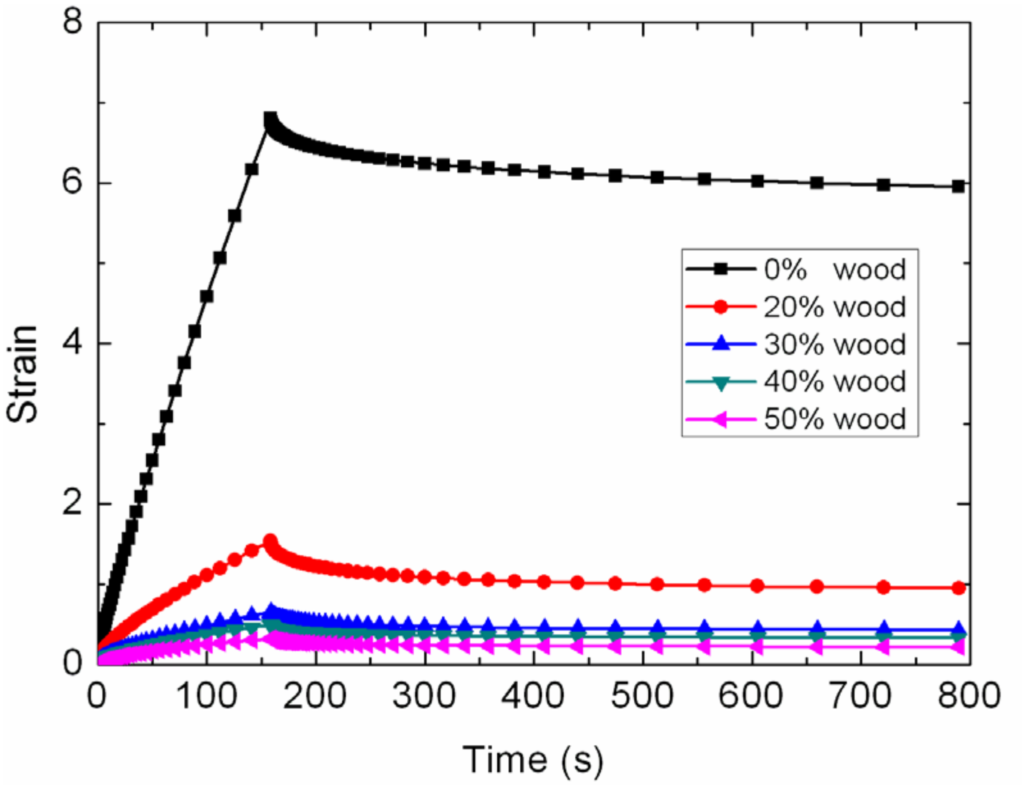

Figure 4. Strain Response of HDPE-Wood Composites.

Relaxation Modulus-Time Curve

Stress relaxation test was performed at a strain of 10%. The relaxation modulus $G(\gamma,t)$ is defined as

$$G(t) = \frac{\sigma}{\gamma} \qquad (2)$$

where σ is the measured stress, and γ is the strain applied to the sample. The measured relaxation modulus versus time curves are plotted in Figure 5. It is observed that $G(\gamma,t)$ increases with increasing the wood content, indicating a larger stress is needed to obtain the same strain when increasing the wood content.

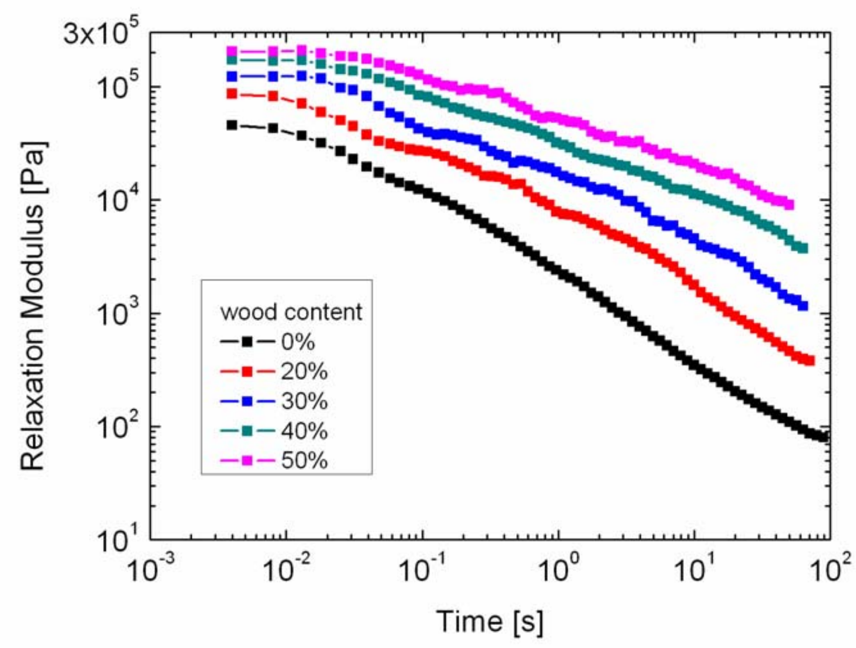

Figure 5. Relaxation Modulus of HDPE-Wood Composites.

Conclusion

The rheological properties of HDPE-wood composites with wood contents of 20~50% were studied through steady shear sweep, oscillatory frequency sweep, creep test, and stress relaxation test. The following conclusions can be drawn from the experimental results:

The yield stress did not appear for the composites prepared under the processing conditions used in this work. Oscillatory frequency sweep showed that both storage and loss moduli of the composites increased with increasing wood content. However, they exhibited a plateau for the composites with wood contents higher than 30% at low frequency. The creep test showed that the deformation of the composites with high wood content was hard. The relaxation modulus of the composites measured from stress relaxation test increased with increasing the wood content.

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