Investigation on the Uniformity of High-Density Polyethylene/Wood Fiber Composites in a Twin-Screw Extruder

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ABSTRACT: Uniform dispersion of wood fiber in high-density polyethylene can improve the mechanical properties and surface finish of the wood plastic composites. However, it is difficult to achieve uniform dispersion when the wood content is high, due to its low thermal stability, incompatibility with polymer, and affinity for agglomeration. This work was undertaken to improve the uniformity of high-density polyethylene/wood composites by designing screw configurations, optimizing screw speed, and altering material compositions. The rheological properties and scanning electron microscope micrograph were used to characterize the uniformity. The results showed that the medium dispersive and distributive mixing, medium screw speed, and lubricant were all beneficial in improving uniformity. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 113: 2081–2089, 2009

Key words: wood fiber; uniform dispersion; screw configuration; screw speed; lubricant

INTRODUCTION

Wood–plastic composites (WPC) have gained rapid growth in recent years due to advantages such as better weather/environment tolerance, improved strength, and lower maintenance requirements compared to natural wood.[1–4] In North America, large amounts of WPC are being produced to replace wood in the decking and railing industry. The industry trend is to produce WPC with high wood content, such as 50 wt % or higher, to increase their economic competitiveness.

The wood fibers (WF) are hydrophilic, whereas most polymers are hydrophobic, which results in poor compatibility between them. Moreover, due to their hydrophilic nature, the WF tend to agglomerate, particularly at higher concentrations.[5–7] Also, WF easily degrades during the processing because of low thermal stability. Therefore, uniform dispersion of high wood contents in a plastic matrix is quite challenging.

The uniformity of wood fiber dispersion in the plastic matrix is important for improving the mechanical properties of WPC.[8,9] Thus it is critical to prepare raw material pellets with uniform wood dispersion. The mixing of polymer and WF is strongly influenced by material components, processing conditions, and machine characteristics. The material components refer to the type and content of polymer, wood, and additives. The processing conditions include screw speed, barrel temperature, and feed rate. The machine characteristics refer to the type of the processing machine and screw configuration. A number of works deal with adding a coupling agent or lubricant to improve wood dispersion in the polymer.[10–12] The processing conditions have also been studied by several researchers.[13,14] However, limited studies demonstrate the effect of machine characteristics on mixing of WPC.

Among the processing equipment of WPC, both the counter-rotating and the co-rotating twin-screw extruder (TSE) are widely used, especially the intermeshing, co-rotating type. Fully intermeshing twin screws provide a narrow residence time distribution, therefore, provide uniform heat history to most wood fibers, which prevents wood degradation. Co-rotating screws are effective in alternating the direction of applied stresses through the use of different mixing elements, thus producing different mixing effects. Two types of mixing are widely used (i.e.,

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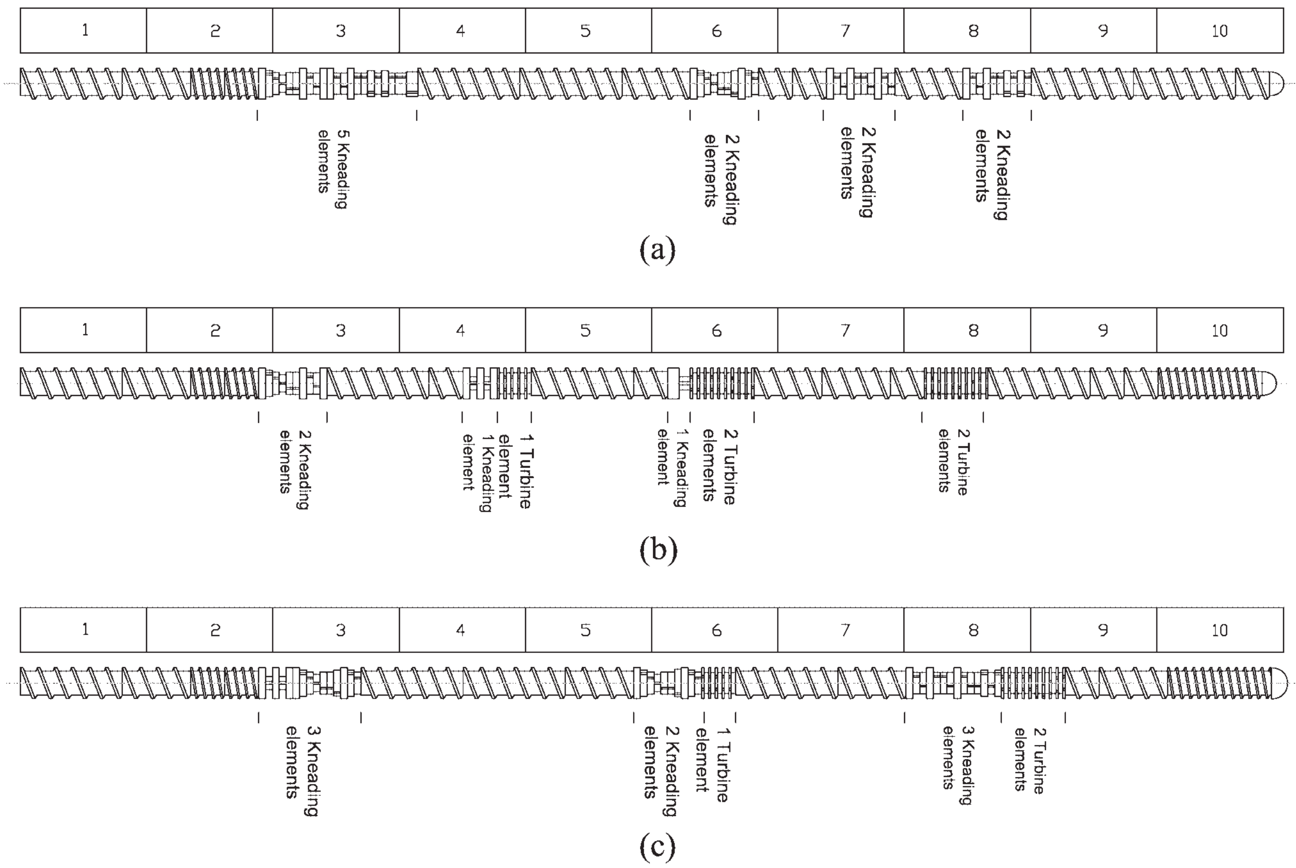


Figure 1 Schematic diagrams of three screw configurations: (a) screw I, (b) screw II, and (c) screw III.

the dispersive mixing and the distributive mixing).[15] Dispersive mixing is the breaking up of clumps or agglomerates of particles into the ultimate particulate size. Distributive mixing is the distribution of particles without decreasing their size. However, the effects of these two types of mixing on the preparation of WPC pellets are not clear. To study these effects, we first have to determine a suitable method for characterize uniformity. The scanning electron microscope (SEM) micrograph,[16–18] mechanical properties,[8,9] and rheological properties[5–7] are the most common methods to investigate the wood dispersion in polymer. SEM micrographs can directly reflect the dispersion state of WF in WPC. Dynamic rheological data can reflect the dispersion state of the filler in the polymer because the test condition is indestructible to the internal structure of the WPC[19]; thus the repeatability of the dynamic rheological data can be used to indicate the uniformity of WPC. The nonuniform WPC will show poor repeatability due to the sensitivity of its rheological response to the state of filler dispersion.[20] Therefore, these two methods were used to characterize the uniformity of WPC in this study.

Among the thermoplastics, polyethylene resin is reported to be the dominant resin in North America, accounting for 83% of the 685,000 tons of plastics consumed annually.[21] Therefore, in the present work, high-density polyethylene (HDPE)/wood composites were used to examine the effects of machine characteristics on WPC dispersion uniformity. Three screw configurations were designed to obtain different types of mixing. A dynamic frequency sweep test was carried out on each WPC formulation to examine changes in its uniformity after processing. The influence of screw speeds and lubricants was also studied.

Designing the screw configurations of TSE

The TSE manufactured by Leistritz has a screw diameter of 27 mm, and a length-to-diameter ratio of 40. The kneading element and turbine element are the most commonly used elements in TSE for mixing. The kneading element can provide dispersive and distributive mixing, depending on the kneading disc width and staggering angle. The turbine element can split the material stream to generate new interfacial surfaces and enhance the chances of material exchange, which, therefore, provide high distributive mixing. In this study we looked at the mixing effectiveness of three screw configurations.

The schematic diagrams of these screw configurations are shown in Figure 1. The idea of designing these configurations is melting the composites first,

TABLE I
The Number of Kneading and Turbine Elements in Three Screw Configurations

Screw configuration	The number of kneading elements	The number of turbine elements	Mixing type
Screw I	11	0	High dispersive mixing, low distributive mixing
Screw II	4	5	Low dispersive mixing, high distributive mixing
Screw III	8	3	Medium dispersive mixing, medium distributive mixing

mixing them with kneading or turbine elements, relaxing and conveying them with conveying elements, then repeat mixing and relaxing them again. The first kneading zone in the three screw configurations have the same function in common: melt the polymer and then wet WF with polymer. However, the three screw configurations differ in the type of mixing they impart. In screw configuration I, eleven kneading elements formed four kneading zones with screw elements between them. By using kneading elements with wide discs and large staggering angles, a high dispersive mixing was achieved. In screw configuration II, there were five turbine elements and four kneading elements with a small staggering angle, which provided high distributive mixing. In screw configuration III, there were three turbine elements and eight kneading elements, which provided both medium dispersive and distributive mixing. In the second mixing zone, the kneading elements provided dispersive mixing followed by turbine element, which provided distributive mixing. Subsequently the dispersive and distributive mixings were applied again in the third mixing zone. The number of kneading and turbine elements in the three screw configurations was summarized in Table I.

MATERIALS AND METHODS

Materials

HDPE 2710, with a melt flow index of 17 g/10 min, was supplied by Nova Chemicals (Calgary/Canada). The WF was standard softwood (pine) with a grade of 12020, provided by American Wood Fibers (Columbia, Maryland). Fifty weight percent of these fibers passed through the sieve of 120 mesh size (125 μm) and were retained on the 140 mesh (106 μm) sieve. Maleic anhydride-*g*-HDPE (Fusabond MB-100D, MFI = 2.0 g/10 min) was used as coupling agent, supplied by DuPont Canada (Mississauga, Canada). Lubricant (TPW 113) was supplied by Struktol America (Stow, Ohio).

Sample preparation

Before mixing, the WF was oven-dried at 100°C for 12 h to remove moisture from the wood. All the materials were dry-blended and then were fed in the main hopper of the TSE. The coupling agent content was maintained at 3% of the total WPC. The barrel temperature was set at 150°C, and the die temperature was set at 160°C. The extruded strands were cooled in air and pelletized. Eleven WPC were compounded in different screw configurations, screw speeds, and material components as described in Table II.

For each WPC formulation, seven batches of pellets were taken randomly to test the repeatability of rheological data. Test samples from each of these seven batches were denoted as Samples 1–7, respectively. Each batch of pellets was dried at 100°C for 12 h and then compression molded into a disc with a size of ϕ 25 mm × 1.4 mm at 170°C and 10 MPa for 20 min, which was then used for the rheological measurements.

Rheological measurements

The rheological measurements were performed in a dynamic stress rheometer (TA, Mississauga/Canada) by using parallel plates 25 mm in diameter. The discs were placed between the plates, which were heated to 170°C. After allowing enough time for the sample to reach the set temperature, the distance between the plates was adjusted to 1 mm before starting the tests. A dynamic frequency sweep test was then performed to obtain the complex viscosity

TABLE II
Material Components and Compounded Conditions of HDPE/Wood Composite

ID code	Material component HDPE : WF : coupling agent : lubricant (phr)	Screw configuration	Screw speed (rpm)
WPC30I	70 : 30 : 3 : 0	Screw I	100
WPC30II	70 : 30 : 3 : 0	Screw II	100
WPC30III	70 : 30 : 3 : 0	Screw III	100
WPC50I	50 : 50 : 3 : 0	Screw I	100
WPC50II	50 : 50 : 3 : 0	Screw II	100
WPC50III	50 : 50 : 3 : 0	Screw III	100
WPC50rpm	50 : 50 : 3 : 0	Screw III	50
WPC150rpm	50 : 50 : 3 : 0	Screw III	150
WPC200rpm	50 : 50 : 3 : 0	Screw III	200
WPC50II/lub	50 : 50 : 3 : 3	Screw II	100
WPC50III/lub	50 : 50 : 3 : 3	Screw III	100

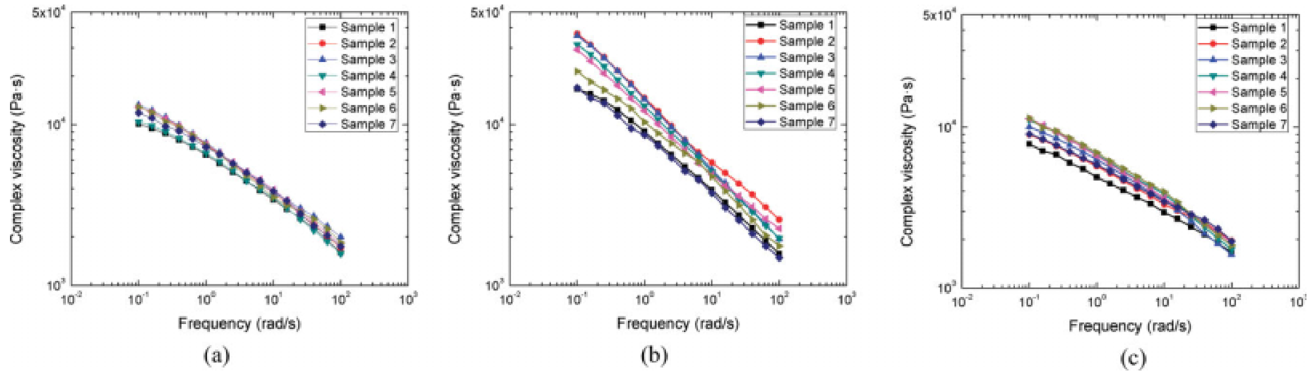


Figure 2 Complex viscosity of (a) WPC30I, (b) WPC30II, and (c) WPC30III. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

over a frequency range of 0.1–100 rad/s at 170°C. Seven samples of each material formulation were tested.

Scanning electron microscope

A sample from the extruded strands of each WPC formulation was dipped in liquid nitrogen and then fractured. The fractured surface was then gold-coated by using a sputter coater (E 50000C PS3), and the microstructure was examined by using a SEM (Hitachi 510).

RESULTS AND DISCUSSION

Effects of screw configuration

The effect of screw configuration on uniformity of WF dispersion in WPC melt was first investigated for 30 wt % wood content. Figure 2 shows the complex viscosity of the WPC material prepared by using the screw speed of 100 rpm, but on different screw configurations. It can be seen that the complex viscosity curves of WPC30I and WPC30III show better repeatability than WPC30II. Furthermore, it also shows that the overall magnitude of the complex viscosity of WPC30II is higher in the low-frequency regions than that of WPC30I and WPC30III. As mentioned above, both screw I and screw III can provide greater dispersive mixing, which forces the WPC to pass through higher shear zones generated between narrow clearances. Therefore, it is likely that the WF aspect ratio is decreased under this high shear. These shortened fibers will have less flow resistance; therefore, they can flow smoothly in the polymer. This would indicate that the WF is uniformly dispersed in the polymer. Screw II provides distributive mixing, generating low shear rates, which cannot break down the WF. Therefore, the fibers with longer aspect ratio, which were in the original raw material, still exist in WPC melt. Because the turbine elements in this configuration do not wipe the entire barrel wall, they provide a low-velocity zone for material stagnation and the long WF may be trapped in the turbine element. Thus, the WF most likely are not distributed uniformly into the polymer matrix. Also, the presence of long WF will increase the complex viscosity. This results in nonuniform WPC with high complex viscosity.

Examination of the microstructure of WPC gave information about how the screw configurations affect the WF length. Figure 3 shows the SEM micrographs of original WF, WPC30I, WPC30II, and WPC30III. The original WF shows the long fiber shape, the length of which is about 106–125 μm. This long WF is not evident in the WPC30I and WPC30III, the length of which is about 15 μm and could be due to the breakage of WF. In Figure 3(c), the long WFs are clearly visible in the WPC30II, the length of which is about 60 μm. This supports our hypothesis that the effect of screw II on breaking down the WF is not very severe.

When the wood content is high, such as 50 wt %, more WF may be bound together to form agglomerates due to its hydrophilic nature. This increase of agglomeration will enhance the difficulty of uniform mixing of the WPC melt. The effects on complex viscosity indicating the uniformity of WPC melt, with 50 wt % wood content and prepared by using the same three screw configurations and the screw speed of 100 rpm, were also observed and are shown in Figure 4. It can be seen that the complex viscosity curves of WPC50III show the best repeatability. It seems that by only applying dispersive mixing or distributive mixing, uniform WPC with high wood content cannot be prepared. When only using dispersive mixing to prepare high wood content composites, the agglomeration breaks down to small particles by the applied shear force, but this is different from the low wood content WPC, as there

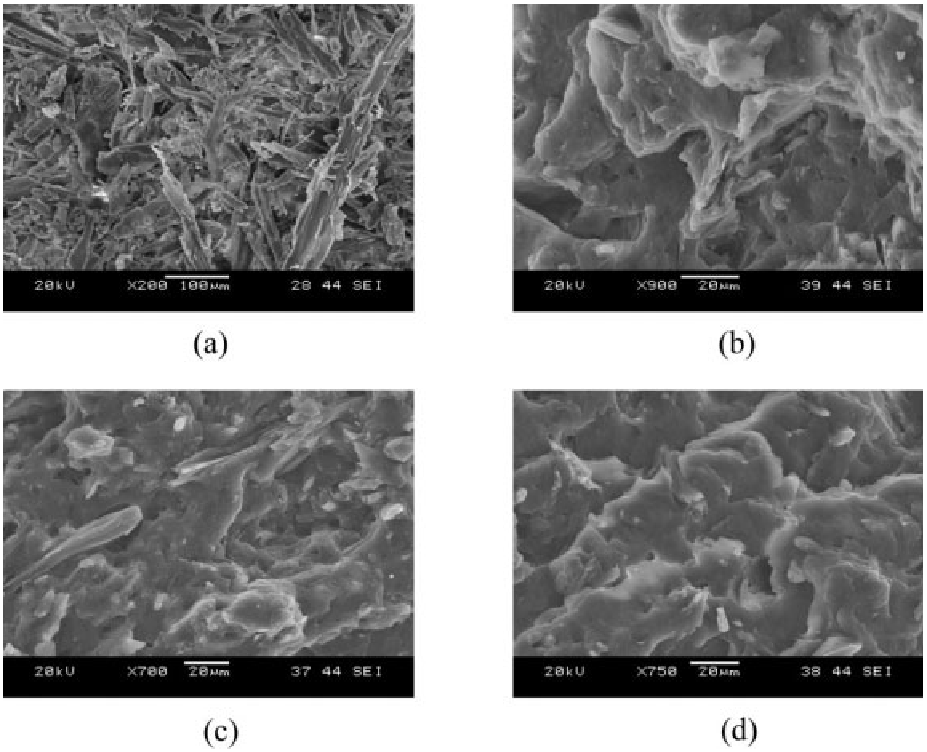


Figure 3 SEM micrographs of (a) original WF, (b) WPC30I, (c) WPC30II, and (d) WPC30III.

are a large amount of small particles in the polymer. These particles may stay in some localized regions and are not transported uniformly to other locations due to the low distributive mixing. Therefore, the final composite is not uniform. The SEM micrographs of these WPC were also examined to understand the effect of screw configuration and are shown in Figure 5. From Figure 5(a), it can be seen that several short WFs stay together in the WPC50I. This illustrates the ineffectiveness of screw I on uniformly distributing WF in polymer. When only using distributive mixing to prepare high wood content composites, the long WF and the agglomeration cannot be broken down due to the low shear generated by distributive mixing, which makes the material stagnation in the turbine element much more severe. The existence of agglomeration and long WFs are clearly seen in the SEM micrograph in Figure 5(b). The length of the WF is about 70 μm. As a result, the composite is not uniform.

By repeatedly applying dispersive and distributive mixing, the uniformity of the composite with high wood content that was prepared with screw III was then improved. The agglomeration was broken

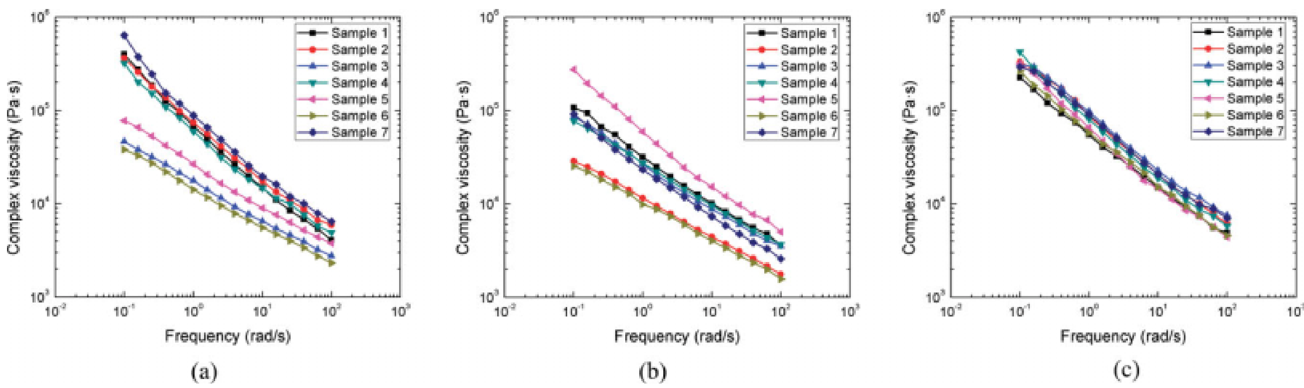


Figure 4 Complex viscosity of (a) WPC50I, (b) WPC50II, and (c) WPC50III. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

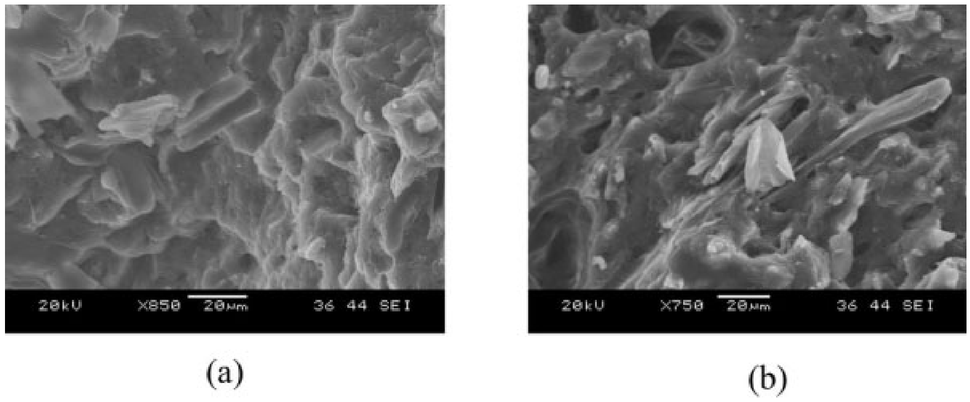


Figure 5 SEM micrographs of (a) WPC50I and (b) WPC50II.

down into its smallest physical particles by dispersive mixing, and then the position of these particles was randomized in the polymer by distributive mixing, thus causing repeated stream splitting and recombination, which then prepared for uniform WPC.

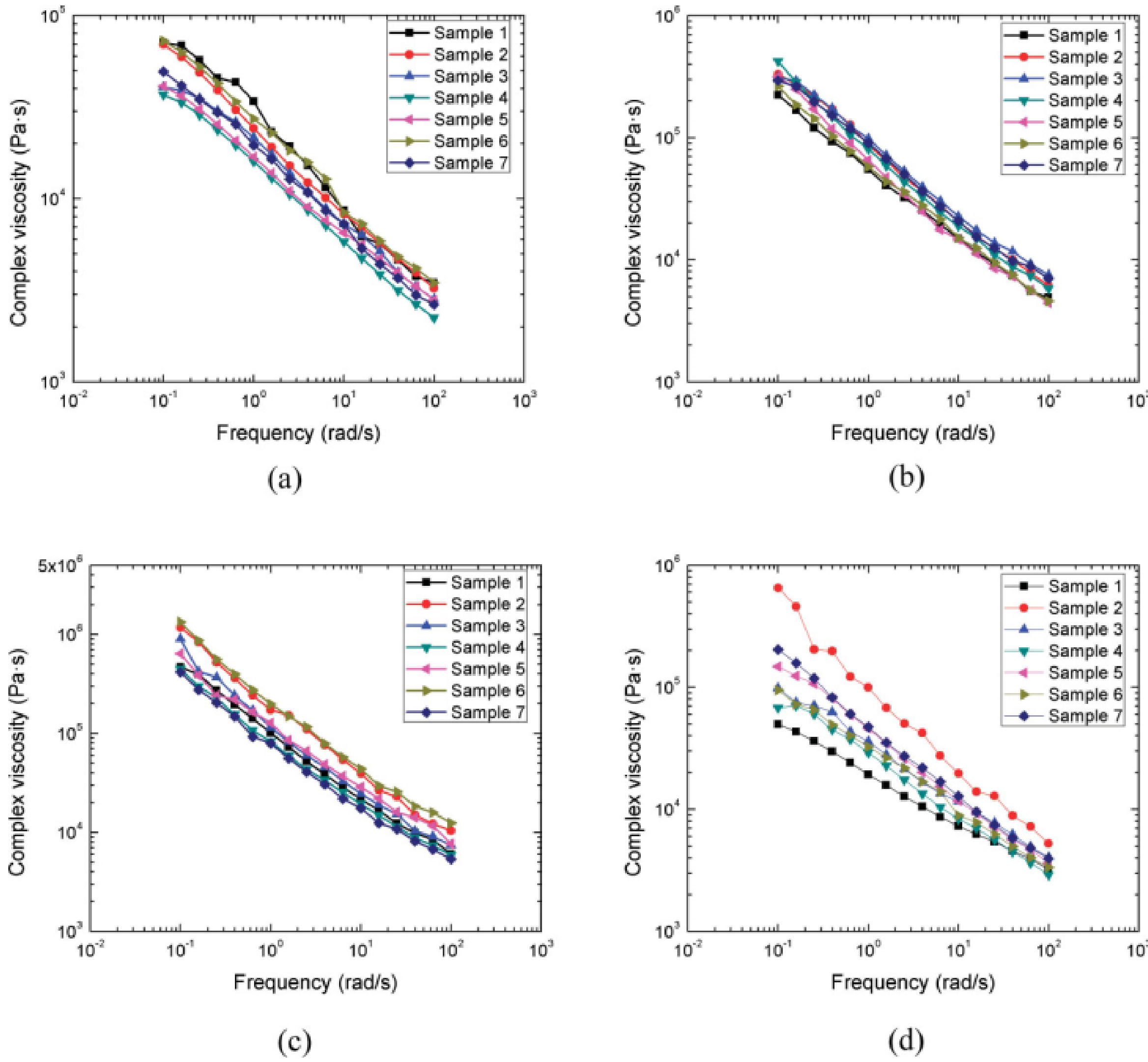


Figure 6 Complex viscosity of (a) WPC50rpm, (b) WPC100rpm, (c) WPC150rpm, and (d) WPC200 rpm. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

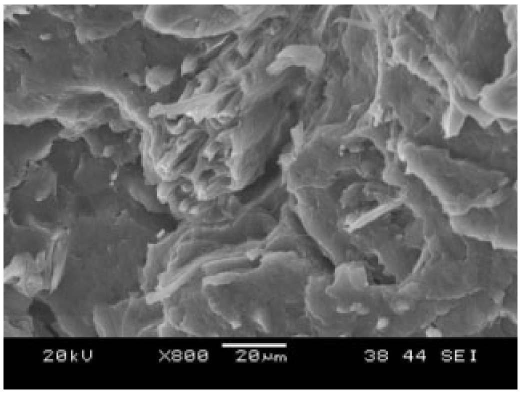


Figure 7 SEM micrograph of WPC50rpm.

Effects of screw speed

The screw speed is one of the important parameters that influences the mixing ability. Four screw speeds were used to study the effects of screw speeds on the uniformity of composites with 50 wt % wood content prepared with screw III. The results, shown in Figure 6, indicate that the uniformity of WPC had increased when the screw speed increased from 50 to 100 rpm. The SEM micrograph of WPC50rpm was investigated, as shown in Figure 7. The existence of WF agglomeration is obvious in the SEM micrograph. This indicates the screw speed of 50 rpm is ineffective in breaking down the WF agglomeration. Increasing the screw speed increases the average shear rate, which enhances the dispersive mixing and, thereby, helps in the uniform dispersion of WF in the WPC melt. However, when the screw speed was further increased to 200 rpm, the uniformity deteriorated. The reason is that the increase of screw speed also increases the shear heating, resulting in higher melt temperatures. Because WF is a temperature-sensitive material, some WFs may have degraded due to the higher temperatures generated when the screw speed increased to 200 rpm. The dark appearance of WPC200rpm was observed during the experiment. The degraded WF will block the flow of WPC, reducing the chance of material exchange, thus deteriorating the uniformity of the WPC.

Another possible reason for the nonuniform WPC at high screw speed is the short residence time of material in the extruder. Because residence time determines the mixing time of the melt, shorter resident times will result in lesser effectiveness of mixing.

Effects of lubricant

Besides the coupling agent, lubricants are another additive that is commonly used in WPC. The lubricants are expected to have the three following effects: melt homogenization, viscosity modification, and surface smoothness improvement. These effects are achieved by both internal lubrication that affects interactions between the components of the melt and external lubrication that affects interactions between the melt and process equipment. In this study, the effects of one lubricant on the complex viscosity of WPC with WF content of 50 wt %, prepared with screw II and screw III, were examined. All the samples were mixed at the screw speed of 100 rpm. The results are shown in Figures 8 and 9. From both the figures, it is obvious that the repeatability of complex viscosity curves is improved by the addition of the lubricant, indicating improved uniformity of WPC. This is because of the better distributive

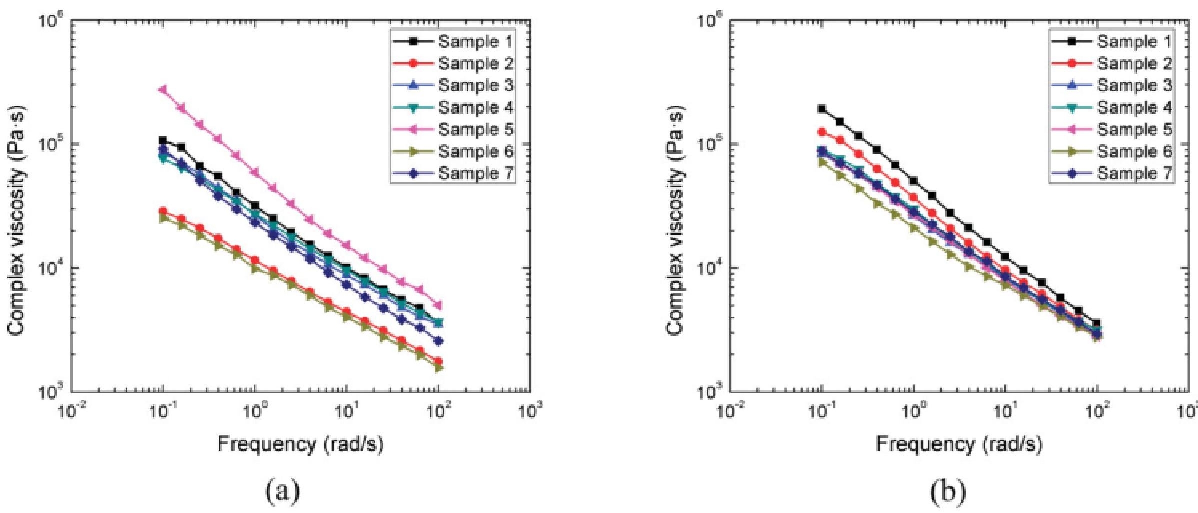
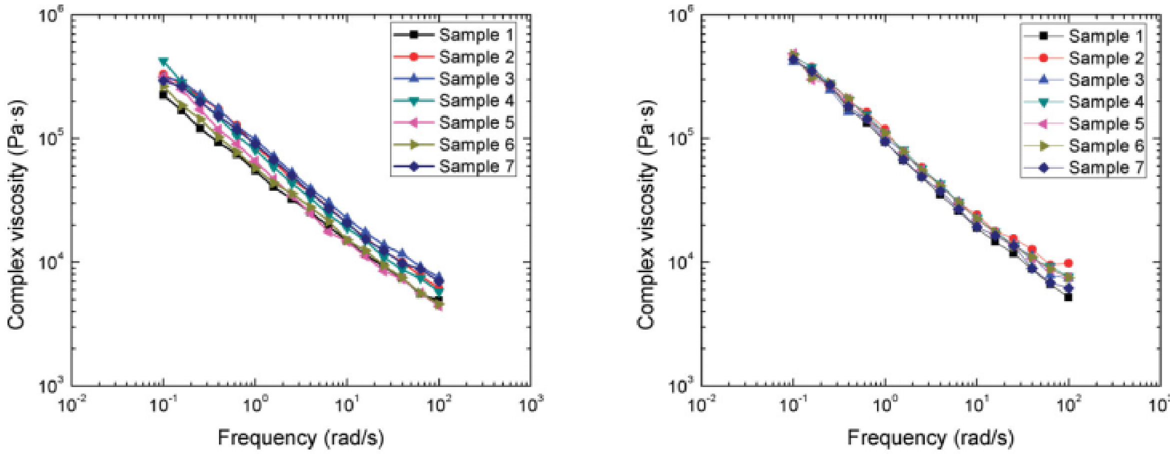


Figure 8 Complex viscosity of (a) WPC50II and (b) WPC50II/lub. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]



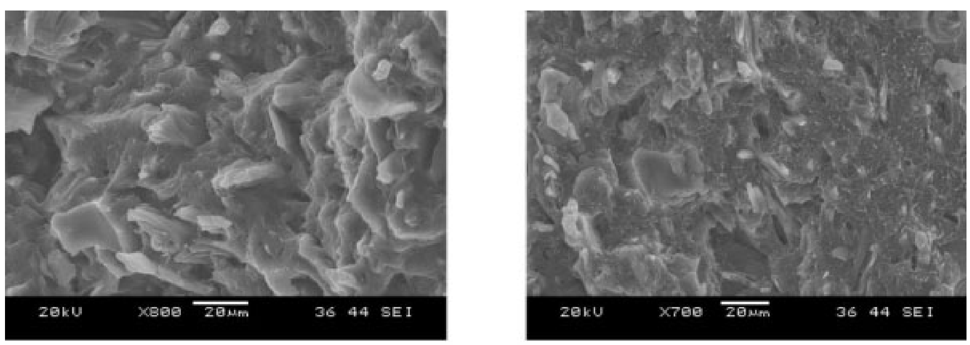
(a) (b)

Figure 9 Complex viscosity of (a) WPC50III and (b)WPC50III/lub. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

mixing of WF due to the decreased friction between WF and polymer by lubricant.

In the case of the WPC50III/lub, all of the complex viscosity curves were nearly coincidental, which shows very good uniformity in the composite. However, the WPC50II/lub does not show as good uniformity as that of WPC50III/lub. As mentioned above, the WF is long and may be easily trapped in the turbine element of screw II. The addition of lubricant enhances the flow of the long WF, resulting in improved uniformity. However, the addition of lubricant is not enough to make very uniform composites without the suitable effect of screw. By applying medium dispersive and distributive mixing and adding a lubricant that increases the WF mobility in the melt flow, very uniform WPC can be achieved.

It is interesting to note that the complex viscosity of the WPC50III/lub is a little higher than that of WPC50III in the low frequency. This is surprising as the complex viscosity was expected to decrease due to the lubricating effects. A possible reason may be due to the lubricant, which reduces the friction between the kneading element and the melt, thus reducing the shear effects and causing the WF breakage to be reduced. The SEM micrographs of WPC50III and WPC50III/lub were investigated to identify the effect of lubricant, as shown in Figure 10. It can be seen that the WF length is about 20 μm, indicating some WFs in WPC50III [Fig. 10(a)] are broken down severely. While in WPC50III/lub [Fig. 10(b)], the WF length is about 40 μm, indicating the WF breakage is less and most of the WF are embedded in polymer. Therefore, with the lubricant, two different effects on the complex viscosity were observed: one was a decrease in the complex viscosity due to the lubrication effects and the other was an increase in the complex viscosity due to lesser breakage of the WF. The combined effect is a small increase in the complex viscosity of the WPC containing lubricant, at low frequencies.



(a) (b)

Figure 10 SEM micrographs of (a) WPC50III and (b) WPC50III/lub.

CONCLUSIONS

In this article, the uniformity of HDPE/wood composites with high wood content was studied by using different screw configurations, screw speeds, and lubricants. Two methods, rheological measurement and SEM micrograph observations, were used to evaluate uniformity. The results showed that by designing screw configurations to provide medium dispersive and distributive mixing cyclically, the uniformity of highly filled HDPE/wood composites was improved. The screw speed should be controlled to achieve efficient mixing without thermal degradation of wood. The use of lubricant enhanced the WF flow due to the reduction of friction among the WF and the molten plastic. The HDPE/wood composites, containing the lubricant and processed by medium dispersive and distributive mixing, showed the best uniformity.

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