

# Wood-Fiber/High-Density-Polyethylene Composites: Compounding Process

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**ABSTRACT:** The compounding process directly influenced the compounding quality of wood-polymer blends and finally affected the interfacial bonding strength and flexural modulus of the resultant composites. With 50 wt % wood fiber, the optimum compounding parameters for the wood-fiber/high-density-polyethylene blends at 60 rpm were a temperature of 180°C and a mixing time of 10 min for the one-step process with a rotor mixer. The optimum compounding conditions at 90 rpm were a temperature of 165°C and a mixing time of 10 min. Therefore, a short compounding time, appropriate mixing temperatures, and a moderate rotation speed improved the compounding quality of the modified blends and the dynamic mechanical properties of

the resultant composites. The melt torque and blend temperature followed a polynomial relationship with the loading ratio of the wood fiber. The highest melt torque and blend temperature were obtained with 50% wood fiber. The coupling treatment was effective for improving the compatibility and adhesion at the interface. The two-step process was better than the one-step process because the coupling agents were more evenly distributed at the interface with the two-step process. © 2004 Wiley Periodicals, Inc. *J Appl Polym Sci* 93: 2570–2578, 2004

**Key words:** blends; compounding; fibers; thermoplastics

## INTRODUCTION

The compounding process directly influences the compounding quality of wood-polymer blends and the coupling agent performance in the resultant composites.<sup>1–3</sup> For wood-fiber/polymer composites, compounding is normally divided into one-step and two-step processes.<sup>4</sup> The former is appropriate for continuous mixers (e.g., extruder and injection molding), whereas the latter is preferred for batch mixers (e.g., roll mills and rotor mixers). Krzysik and Younquist<sup>5</sup> reported that the two-step process was better for the manufacturing of air-formed wood-fiber/polymer composites.

For refiner ground pulp and polypropylene composites, the optimum mixing conditions are a period of 10 min, a mixing temperature of 180°C, and a rotation speed of 50 rpm.<sup>2</sup> Takase and Shiraishi<sup>2</sup> reported that rotation speeds have an influence on coupling effectiveness similar to that of the mixing time, and moderate mixing speeds are preferred for better fiber length distribution and coupling effectiveness. Usually, the mixing temperature is controlled to be less than 200°C for most coupling treatments to avoid the decomposition and degradation of wood fiber and some thermoplastic matrices.<sup>2,6,7</sup>

Maldas and Kokta<sup>3</sup> reported that with a two-roll mill system, the maximum improvement in the mechanical properties of chemithermomechanical pulp and polystyrene composites was achieved when the mixing time was 15 min and the temperature was 175°C. The blends were remixed 5 and 10 times (ca. 6–8 min) during compounding. Hence, the two-step process resulted in a better distribution of the coupling agents at the interface, providing a uniform mixture of the coupling agents with the polymer and wood fiber.<sup>7</sup>

Coutinho et al.<sup>8</sup> investigated the effects of the treatment and mixing conditions on the mechanical properties of wood-fiber/polypropylene composites with silane coupling agents. They reported that the optimal mixing conditions for wood-fiber/polypropylene composites were a mixture temperature of 180°C and a mixing time of 10 min at a rotation speed of 60 rpm. Through the pretreatment of the wood fiber with silane A172, the mechanical properties of the resultant composites were optimized under the aforementioned compounding conditions.

Rozman et al.<sup>9</sup> compared the effects of two different blending systems (an internal mixer and a single-screw extruder) on the mechanical properties of oil-palm-empty-fruit-bunch/polypropylene composites. For the internal mixer, mixing was conducted at 180°C at a rotation speed of 25 rpm. The single-screw extruder was operated at a rotation speed of 20 rpm with

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increasing temperature zones of 160, 170, and 180°C. They reported that the composites produced by the internal mixer had higher tensile strength, tensile modulus, and impact strength than those produced by extrusion because the internal mixer produced better filler dispersion and improved the wetting of the filler surface.

Blending the wood fiber with a polymer is the key step of composite production because the compounding process helps to uniformly distribute the wood fiber and coupling and modifying agents in the thermoplastic matrix, decrease the pore ratio, and stabilize the filler/matrix interaction.<sup>10</sup> However, there has been no criterion for determining the optimum compounding conditions. Usually, the compounding conditions vary with the mixing machine type, compounding steps, weight ratio of the wood fiber and polymeric matrix, moisture content of the wood fiber, and species of the thermoplastics and wood fiber. Sometimes, it may be difficult to isolate these factors. In this study, a statistical method was used to explore the relationship of the rotation speed, mixing temperature, and compounding time for wood-polymer blends in an inner mixer and their influence on the mechanical properties of the resultant wood-polymer composites.

The objectives of this study were (1) to investigate the influences of the compounding conditions on the mechanical properties of the resultant composites, (2) to investigate the relationship of the rotation speed, mixing temperature, and compounding time with statistical analysis, (3) to determine the best compounding conditions for compounding quality and the mechanical properties of the resultant composites, and (4) to compare one-step and two-step processes.

## EXPERIMENTAL

### Materials

Mixed thermomechanical pulp (TMP) fiber was obtained from Temple-Inland Co. (Diboll, TX). Before the compounding process, the TMP fiber was dried in an oven at 100°C for 24 h, and its moisture content was controlled within 2–3%.

High-density-polyethylene (HDPE) pellets (PE10462N, Dow Chemical, Midland, MI) were obtained from a local plastic company. The density of HDPE was 962.5 kg/m<sup>3</sup>. Its melting temperature and melt index were 134°C and 10 g/10 min, respectively. The tensile strength and modulus of PE10462N were 12 MPa and 0.75 GPa, respectively. It had an ultimate elongation of 1200%.

Epolene E20, an oxidized polyethylene, was obtained from Eastman Chemical Company (Longview, TX). E20 is a good textile lubricant and softener. It is also extensively used as an extrusion lubricant for rigid poly(vinyl chloride) and HDPE. In this study,

**TABLE I**  
Experimental Design for Compounding Conditions in a One-Step Process

Experiment	Rotation speed (rpm)	Temperature (°C)	Time (min)
S1Mix 1	60	150	10
S1Mix 2	60	150	15
S1Mix 3	60	150	20
S1Mix 4	60	165	10
S1Mix 5	60	165	15
S1Mix 6	60	165	20
S1Mix 7	60	180	10
S1Mix 8	60	180	15
S1Mix 9	60	180	20
S1Mix 10	90	150	10
S1Mix 11	90	150	15
S1Mix 12	90	150	20
S1Mix 13	90	165	10
S1Mix 14	90	165	15
S1Mix 15	90	165	20
S1Mix 16	90	180	10
S1Mix 17	90	180	15
S1Mix 18	90	180	20

The weight ratio of the wood fiber to HDPE was 50:50. The concentration of E20 was 5 wt% of the oven-dried wood fiber.

E20 was used as a coupling agent for wood-fiber/HDPE composites. E20 has a weight-average molecular weight of 7000, and its acid number is 17. The concentration level of E20 was 5% with respect to the weight of the oven-dried wood fiber.

### Compounding processes for the wood-fiber/HDPE blends

The melt-molding processes used for manufacturing the wood-fiber/polymer composites were one-step and two-step.<sup>4</sup> The weight ratio of the wood fiber to HDPE was 50:50. For the one-step process, the wood fiber, thermoplastics, and coupling agent were sequentially fed into a Haake Rheomix 600 blender (Dreieich, Germany). Both the blending torque and compounding temperature were measured during the compounding. After the mixing, the melts were removed from the blender and cooled to room temperature. The melts were ground with a Thomas-Wiley 3383L10 (Swedesboro, NJ) miller to pass through a 20-mesh screen. For the one-step process, a 3 × 3 × 2 completely randomized design factorial experiment was conducted to determine the optimum blending conditions for the wood-fiber/HDPE composites (Table I).

For the two-step process, a small amount of HDPE (10% with respect to all the thermoplastics) was first kneaded with all the E20 pellets and 10% wood fiber and ground into a powder (20-mesh). Then, the pre-treated blends were further mixed with the remaining

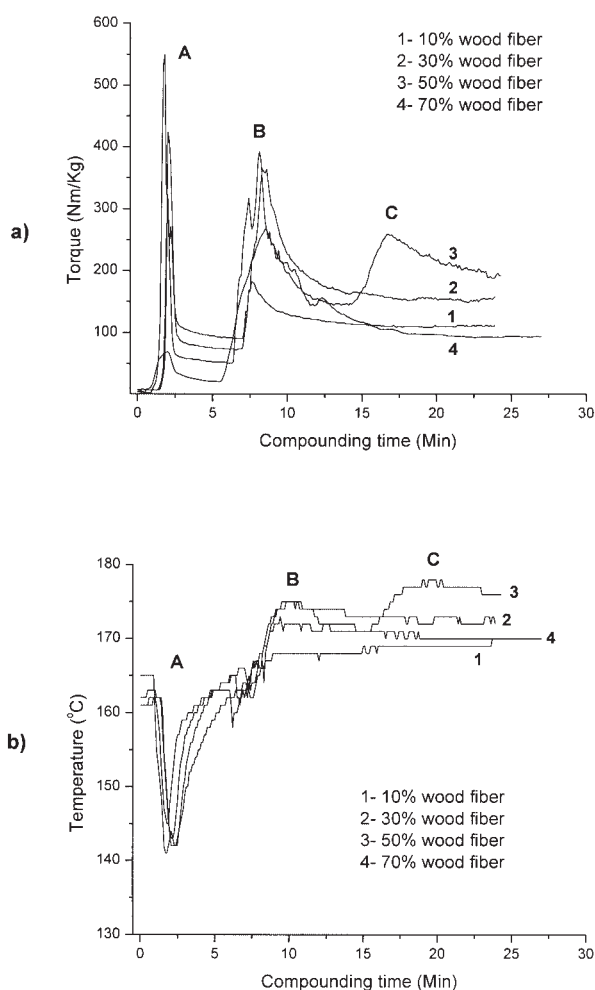
wood fiber and thermoplastics. After the compounding, all the resulting blends were ground again into a powder (20-mesh).

### Manufacture of the wood-fiber/HDPE composites

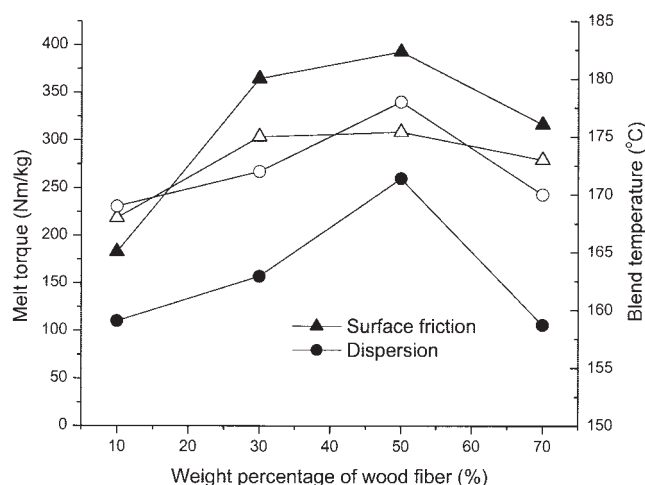
A ground powder of the required weight was placed in a two-piece aluminum molding set. The mold was pressed with a miniature hot press at 168°C for 3 min and cooled to room temperature at the same pressure for 1 min. The pressure for heating and cooling was controlled to be 0.16 MPa. Specimens were made with this molding set for dynamic mechanical analysis. The densities of all the specimens were controlled to be  $1000 \pm 50 \text{ kg/m}^3$ .

### Measurement of the dynamic mechanical properties of the resultant composites

The dynamic mechanical properties of the resultant composites were analyzed with a Seiko DMS 110 dy-



**Figure 1** Effect of the wood loading for the wood-fiber/HDPE blends on (a) the melt torque and (b) the blend temperature. The blends were mixed at 165°C and 60 rpm for 15 min.



**Figure 2** Torque and temperature changes for the wood-polymer blends. The solid symbols represent the melt torque, whereas the empty ones indicate the blend temperatures.

amic mechanical analysis system (Chiba, Japan). The sample size was 60 mm × 10 mm. At room temperature, all the samples were measured in a three-point bending mode at a frequency of 100 Hz.

### Data analysis

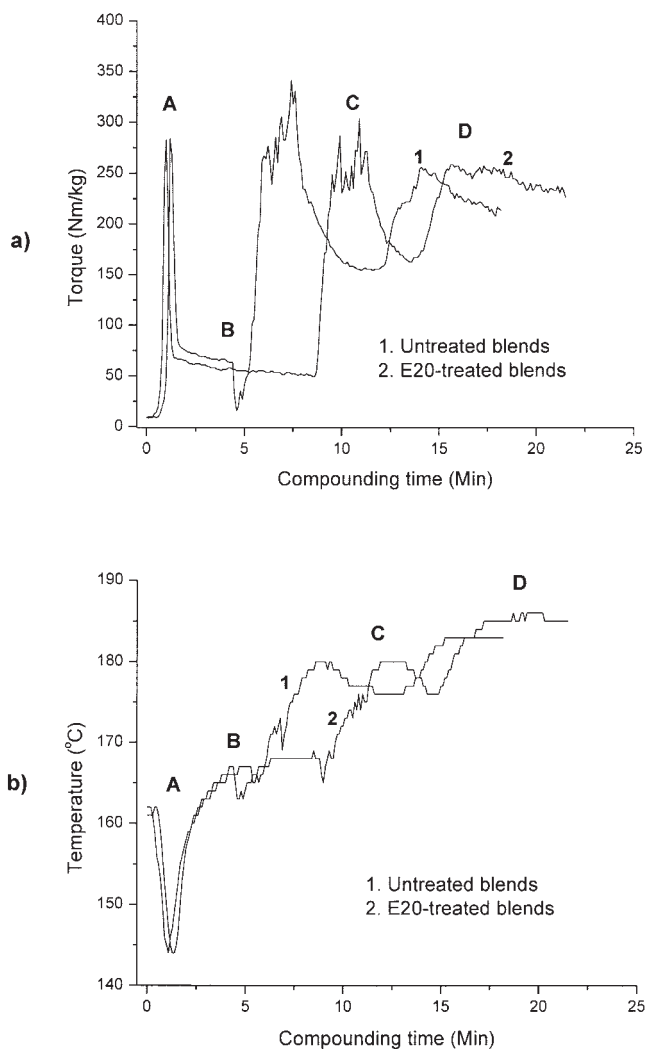
For the one-step blending process, a three-way analysis of variance (ANOVA) was conducted to investigate the main effects and interaction effects of the mixing temperature, rotation speed, and compounding time on the flexural moduli of the resulting composites. The differences in the levels of these three factors were compared with Duncan's multiple-range test.

## RESULTS AND DISCUSSION

### Characterization of the wood-fiber/HDPE blending process

During the compounding, the wood-fiber/HDPE blends mainly underwent stages A and B in the one-step process (Fig. 1). During stage A, the blend torque sharply increased as HDPE pellets were added. At the same time, the blend temperature (or chamber temperature) sharply decreased because the inner temperature dropped with the feeding of cold thermoplastics into the mixing chamber. After HDPE was melted, the melt torque decreased to around 50–100 N m/kg, and the blend temperature decreased back to the setting point. The actual torque values varied with the amount of HDPE, as expected. The more HDPE pellets were added, the more torque was needed.

After the addition of wood fibers during stage B, there were rapid increases in the machine torque and blend temperature because of the surface friction be-



**Figure 3** Compounding characterization of the modified wood-fiber/HDPE blends (50:50): (a) the melt torque and (b) the compounding temperature. Untreated and treated blends were mixed at 165°C and 90 rpm for 10 min. The concentration of E20 was 5 wt % of the oven-dried wood fiber.

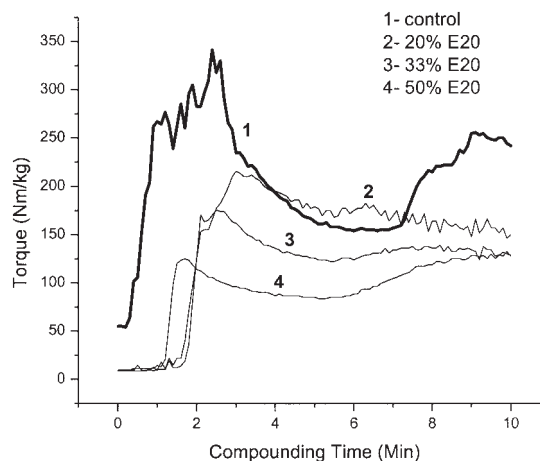
tween the wood fiber and HDPE (Fig. 1). With continuous compounding, the melt torque of the wood-HDPE blends gradually decreased, and the blend temperature was stabilized to around 170°C. The melt torque by surface friction and the blend temperature both had a polynomial relationship with the weight percentage of the wood fiber (Fig. 2). They had their biggest jump at 50% wood fiber.

Under further compounding, the fillers were dispersed into the matrix by shear stress, which was significantly influenced by the loading ratio of the fillers.<sup>11</sup> During stage C, the melt torque in the dispersion increased with an increase in the weight percentage of the wood fibers at low loading ratios (Figs. 1 and 2). At 10% wood fiber, the melt torque was around 100 N m/kg. For the wood-HDPE blends with

a weight ratio of 50:50, there was an increase in the melt torque and blend temperature during stage C. The torque dramatically increased to around 260 N m/kg when the loading ratio of the wood fiber was as high as 50%. However, the torque decreased back to 100 N m/kg at 70% wood fiber (Fig. 2). The increase in the melt torque and blend temperature was due to the fact that the total surface contact area of the individual fibers with the matrix was close to the maximum value around 50% wood fiber, and so the surface friction and dispersion resistance reached the maximum value. At 50% wood fiber, the largest torque was built up by shear stress [Fig. 1(a)]. The shear stress also increased the blend temperature by about 8°C (Fig. 2).

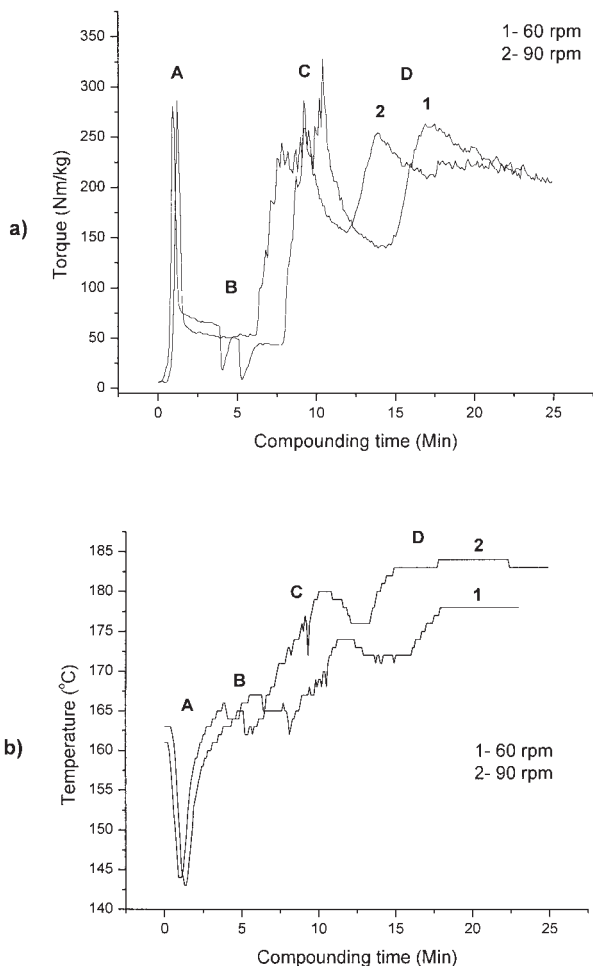
The modified wood-fiber/HDPE blends presented compounding characterization similar to that of untreated blends. There were four different compounding stages for the modified wood-fiber/HDPE blends (50:50; Fig. 3). After the addition of a coupling agent during stage B, the torque slightly dropped (ca. 50 N m/kg) and then went back to 50 N m/kg. During stage C [Fig. 3(a)], the torque experienced a large jump after the addition of wood fibers because surface friction was produced at the interface. This friction also caused a large increase in the blend temperature [Fig. 3(b)]. However, the torque value of the E20-treated blends decreased by 50 N m/kg with respect to that of the untreated blends. This implied that adding a coupling agent improved the affinity between the wood fiber and the thermoplastic matrix.

Like the untreated blends, the E20-treated blends also experienced large jumps in the torque and temperature during stage D because of dispersion (Fig. 3). For the modified blends, the melt torque gradually decreased during stage D, but its decreasing slope was smaller than that of untreated blends. There was a



**Figure 4** Effect of the coupling agent concentration on the melt torque of the wood-HDPE blends (50:50). The blends were mixed with no coupling agent (control), 20% E20, 33% E20, or 50% E20 at 165°C and 90 rpm for 10 min.





**Figure 5** Influence of the rotation speed on (a) the melt torque and (b) the compounding temperature for the wood–HDPE blends with 5% E20. The mixing temperature and compounding time were 165°C and 15 min, respectively.

small temperature increase during stage D, which was perhaps related to the coupling reaction at the interface. The torque was finally stable around 220 N m/kg for the modified blends (50:50). Therefore, stage D presented a difference between the modified blends and unmodified blends. For the unmodified blends, the temperatures were stable and independent of the compounding time, whereas there was a temperature increase for the modified blends because of the coupling reaction at the interface.

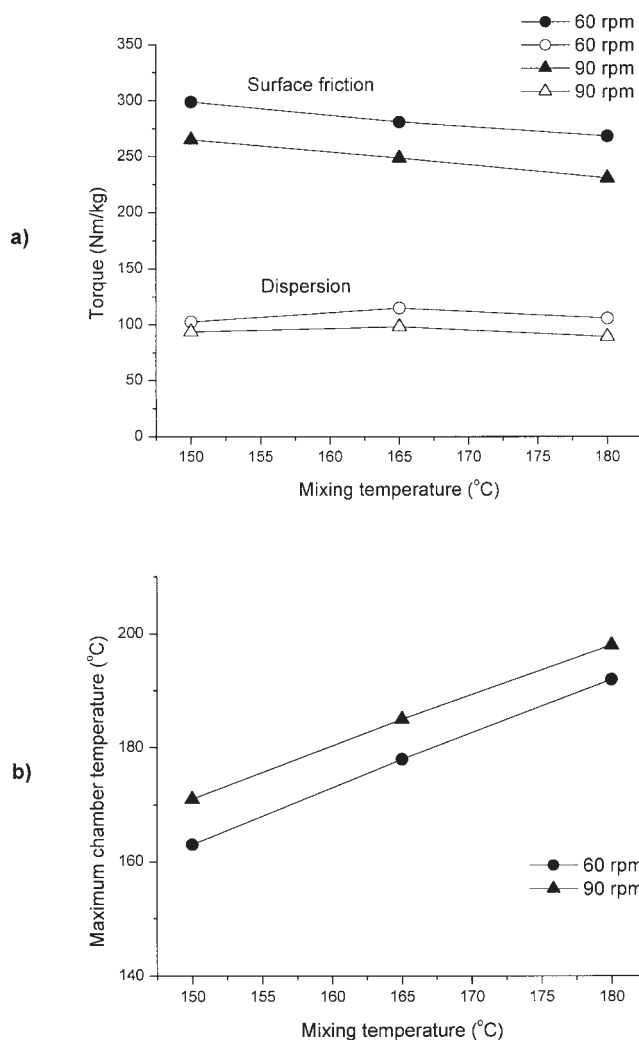
**Effect of the coupling agent concentration**

For the untreated wood-fiber/HDPE blends (50:50), the melt torque by surface friction and that by dispersion were around 330 and 250 N m/kg, respectively (Fig. 4). The melt torque by surface friction decreased by 50 N m/kg when the E20 concentration was 5%. This implied that there was little interaction between the wood fiber and thermoplastics for untreated

wood–polymer blends. The surface friction and dispersion stress significantly changed when the coupling agent concentration was greater than 10%. The melt torque decreased to around 220 N m/kg when the coupling agent concentration was 20%. It was equal to 130 N m/kg, around one-third of that of the untreated blends, when the concentration of E20 was as high as 50%. During dispersion, the melt torque for all the modified blends converged to about 130 N m/kg, which was one-half that of the untreated blends (Fig. 4). Consequently, the coupling treatment effectively improved the compatibility between the wood fiber and thermoplastics.

**Effect of the compounding conditions on the melt torque and blend temperature**

As shown in Figures 5 and 6, the rotation speed influenced the melt torque of the modified blends and



**Figure 6** Effect of the different compounding conditions on (a) the melt torque of the wood–polymer blends and (b) the maximum chamber temperature.

TABLE II  
Melt-Torque (Q) and Blend-Temperature Changes Under Different Compounding Conditions for the One-Step Process

Rotation speed (rpm)	Temperature (°C)	Time (min)	Q (N m/kg)			$\Delta Q$ (N m/kg)		$\Delta T$ (°C)	
			Adding HDPE	Adding E20	Adding fiber	Friction	Dispersion	Friction	Dispersion
60	150	10	5.56	55.09	46.62	312.58	100.40	13	6
	150	15	7.15	59.33	47.94	285.81	101.72	12	6
	150	20	7.15	50.85	38.14	298.50	106.21	14	7
	165	10	7.15	52.45	47.95	264.63	120.26	12	7
	165	15	5.56	50.86	43.71	288.47	124.50	12	7
	165	20	2.91	45.30	38.14	289.87	100.39	14	7
	180	10	2.91	41.06	36.82	278.52	117.61	13	6
	180	15	2.91	41.06	36.82	285.84	104.90	12	7
	180	20	2.91	35.50	35.50	240.16	94.57	10	6
	150	10	5.56	63.57	53.77	250.57	90.26	15	9
90	150	15	5.56	66.49	49.54	273.13	96.16	15	9
	150	20	4.24	58.01	41.06	271.53	94.57	18	9
	165	10	8.48	63.57	49.54	254.30	95.89	15	10
	165	15	7.15	63.57	53.77	236.02	100.39	16	8
	165	20	4.24	55.10	45.30	255.90	98.81	16	8
	180	10	8.48	56.69	46.62	218.01	97.48	13	7
	180	15	7.15	53.77	45.30	253.24	85.03	14	5
	180	20	4.24	52.45	45.30	220.66	85.03	15	6

The weight ratio of the wood fiber to HDPE was 50:50. The concentration of E20 was 5 wt% of the oven-dried wood fiber.

blend temperature. The melt torque by surface friction and dispersion at 60 rpm was larger than that at 90 rpm [Fig. 5(a)], but the maximum blend temperature at 90 rpm was larger than that at 60 rpm because the high rotation speed (i.e., 90 rpm) usually generated more heat per minute. During stages C and D, the blend temperature at 90 rpm increased by 10–11°C in comparison with that at 60 rpm [Fig. 5(b)]. At the same mixing temperature, the melt torque by surface friction at 60 rpm was larger than that at 90 rpm [Fig. 6(a)]. The melt torque by dispersion, however, was not as sensitive to the rotation speed. The melt torque by dispersion at 60 rpm was a little higher than that at 90 rpm [Fig. 6(a)]. Usually, a high rotation speed resulted in a high chamber temperature at the same mixing temperature [Fig. 6(b)].

The mixing temperature also influenced the melt torque and chamber temperature. At the same rotation speed, the melt torque by surface friction decreased with an increase in the mixing temperature [Fig. 6(a)]. However, the chamber temperature increased with an increase in the mixing temperature [Fig. 6(b)]. At high mixing temperatures (e.g., 180°C), the melt torque by friction decreased at both rotation speeds because the thermoplastic matrix had better flowability under high temperatures. Similarly, the melt torque was related to the shear stress and was not as sensitive to the mixing temperature [Fig. 6(a)]. For both rotation speeds, the melt torque at 165°C was a little higher than at other mixing temperatures [Fig. 6(a)]. The melt torque decreased as the compounding time increased

(Table II) because a longer compounding time caused degradation of the wood fiber and some polymer matrices at high temperatures. Therefore, a longer compounding time weakened the quality of the wood-polymer blends.

The dispersion of the wood fiber in the matrix was more related to the mixing temperature and rotation speed. As shown in Figure 7, a high mixing temperature or high rotation speed caused a left shift in a dispersion peak with a small change in the torque.

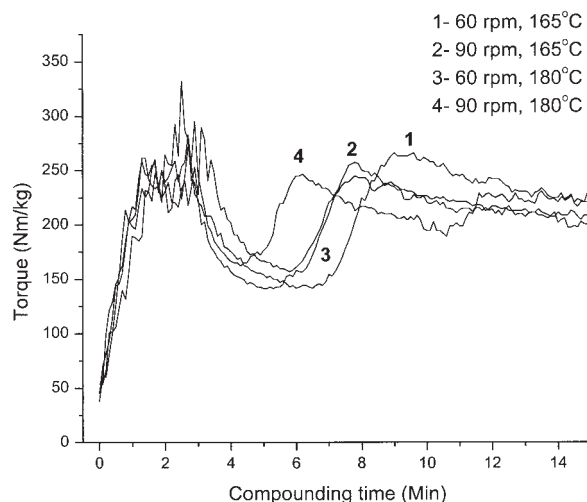


Figure 7 Influence of the rotation speed and mixing temperature on the melt torque of wood-polymer blends at the compounding time of 15 min.

**TABLE III**  
**Influence of Different Compounding Conditions in the One-Step Process on the Dynamic Mechanical Properties of E20-treated Wood-Fiber/HDPE Composites at 25°C**

Compounding conditions <sup>a</sup>						Dynamic mechanical properties of resultant composites <sup>c</sup>			
Rotation speed (rpm)		Mixing temperature (°C)		Compounding time (Min)		$E^*_{int}$ (GPa)	$E'$ (GPa)	$E''$ (GPa)	$\tan \delta$ ( $\times 10^{-2}$ )
Level	Duncan's grouping <sup>b</sup>	Level	Duncan's grouping <sup>b</sup>	Level	Duncan's grouping <sup>b</sup>				
60		150		10	A	2.91 (0.23)	3.11 (0.33)	0.21 (0.03)	6.7 (0.3)
60	A	150	A	15	A	3.82 (0.63)	3.26 (0.59)	0.20 (0.03)	6.1 (0.2)
60		150		20	A	3.92 (0.73)	3.63 (0.51)	0.22 (0.03)	6.0 (0.1)
60		165		10	A	3.96 (0.46)	4.25 (0.42)	0.26 (0.03)	6.3 (0.1)
60	A	165	B	15	A	4.30 (0.64)	4.13 (0.45)	0.24 (0.03)	5.8 (0.1)
60		165		20	A	3.99 (0.25)	3.80 (0.12)	0.22 (0.01)	5.9 (0.2)
60		180		10	A	4.68 (0.37)	4.50 (0.23)	0.26 (0.01)	5.8 (0.1)
60	A	180	B	15	A	4.23 (0.43)	4.21 (0.38)	0.26 (0.02)	6.1 (0.2)
60		180		20	A	3.92 (0.55)	3.90 (0.39)	0.24 (0.03)	6.2 (0.3)
90		150		10	A	3.90 (0.24)	3.73 (0.18)	0.22 (0.01)	6.0 (0.1)
90	B	150	A	15	A	4.35 (0.61)	4.14 (0.42)	0.26 (0.03)	6.2 (0.0)
90		150		20	A	4.19 (0.43)	4.12 (0.19)	0.26 (0.01)	6.2 (0.1)
90		165		10	A	5.00 (0.79)	4.84 (0.64)	0.30 (0.05)	6.2 (0.1)
90	B	165	B	15	A	4.64 (0.62)	4.45 (0.37)	0.27 (0.01)	6.1 (0.2)
90		165		20	A	4.37 (0.38)	4.21 (0.26)	0.26 (0.02)	6.1 (0.1)
90		180		10	A	4.30 (0.41)	4.42 (0.18)	0.26 (0.01)	5.9 (0.1)
90	B	180	B	15	A	4.19 (0.45)	4.12 (0.29)	0.25 (0.01)	6.1 (0.3)
90		180		20	A	4.13 (0.62)	4.04 (0.34)	0.29 (0.02)	6.4 (0.2)

The weight ratio of the wood fiber to HDPE was 50:50.

<sup>a</sup> Levels with the same letter were not significantly different at  $\alpha = 0.05$ .

<sup>b</sup> Duncan's grouping was based on  $E^*_{int}$ .

<sup>c</sup> The values in parentheses are the standard deviations.

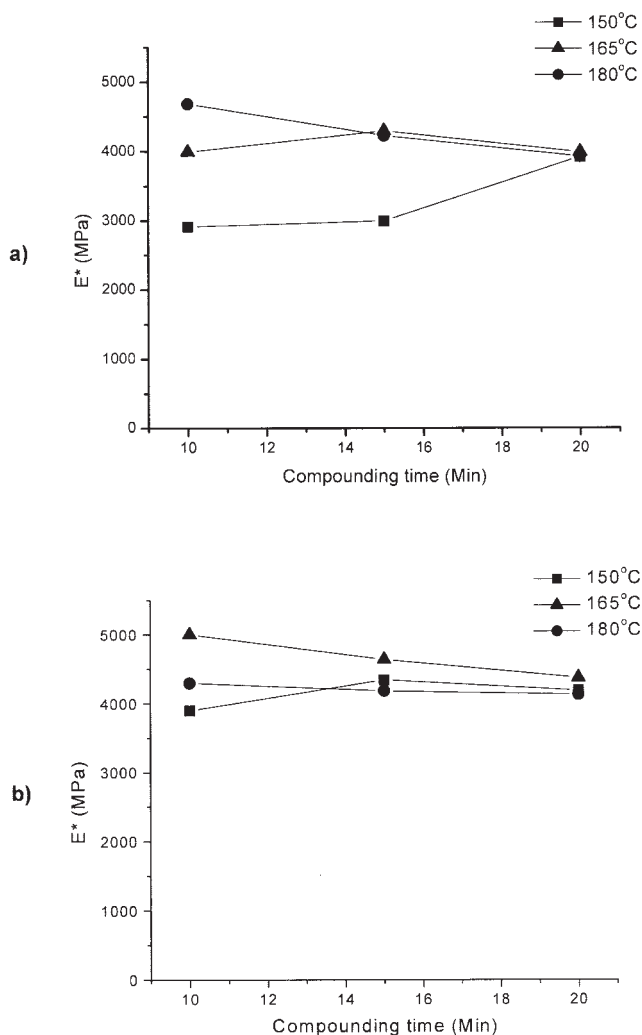
This left-shifting effect was more significant when the wood fiber and thermoplastics were mixed at a high temperature and high rotation speed. The dispersion peak appeared around 9 min when the rotation speed and mixing temperature were 60 rpm and 165°C, respectively. When the mixing temperature increased to 180°C or the rotation speed increased to 90 rpm, the peak shifted to around 8 min. At a high mixing temperature (i.e., 180°C) and a high rotation speed (90 rpm), the peak shifted to 6 min (Fig. 7). Accordingly, a high mixing temperature and a high rotation speed reduced the compounding time.

At the same mixing temperature, the melt-torque increment ( $\Delta Q$ ) decreased with an increase in the rotation speed, but the chamber-temperature increment ( $\Delta T$ ) increased with an increase in the rotation speed at low mixing temperatures (Table II). For example,  $\Delta Q$  at 90 rpm was smaller than that at 60 rpm, but  $\Delta T$  at 90 rpm was larger than that at 60 rpm at the same temperature. At the same rotation speed,  $\Delta Q$  by surface friction decreased with an increase in the compounding time and mixing temperature. At both rotation speeds,  $\Delta Q$  by dispersion had a maximum value at 165°C. However, the  $\Delta T$  values were almost independent of the mixing temperature and compounding time at both rotation speeds (Table II). Therefore,  $\Delta Q$

by surface friction was related to the mixing temperature, compounding time, and rotation speed, but  $\Delta Q$  by dispersion and  $\Delta T$  were more related to the rotation speed.

#### Effect of the compounding conditions on the dynamic mechanical properties

The compounding conditions had a significant influence on most dynamic mechanical properties of the resultant composites (Table III). At 60 rpm, the complex modulus ( $E^*$ ) increased with an increase in the compounding time at low mixing temperatures, but it decreased with an increase in the time at high mixing temperatures (Fig. 8).  $E^*$  also showed this decreasing trend at high temperatures when the rotation speed was 90 rpm. This was attributed to the fact that a high rotation speed increased the dispersion of the length distribution for the wood fiber.<sup>2</sup> Also, this decreasing trend was due to the decomposition and degradation of the wood fiber, coupling agents, and even HDPE under high temperatures.<sup>2,7</sup> At a high rotation speed and a high mixing temperature, the inner temperature in the mixing chamber was high, up to 195°C, because of the dispersion and coupling reaction. This high temperature may have caused chain scission of cellulose.



**Figure 8** Influence of different compounding conditions on  $E^*$  of the wood-fiber/HDPE composites at (a) 60 and (b) 90 rpm. The weight ratio of the wood fiber to HDPE was 50:50, and the concentration of E20 was 5 wt % of the oven-dried wood fiber.

lose and polymer molecules.  $E^*$  converged to a value of approximately 4.0 GPa with an increase in the compounding time (Fig. 8).

The storage (or elastic) modulus ( $E'$ ) presented a trend similar to the complex modulus ( $E^*$ ). Better strength properties occurred at 90 rpm when the mixing temperature and compounding time were 165°C and 10 min, whereas the optimum compounding conditions at 60 rpm were 180°C and 10 min (Table III). Hence, a short compounding time improved the mechanical properties of the resultant composites.

The loss (or viscous) modulus ( $E''$ ) was between 0.2 and 0.3 GPa for the modified wood-fiber/HDPE composites. The composites with high  $E'$  values also had high  $E''$  values. Moreover, the composites with a high modulus had small  $\tan \delta$  values at 25°C, compared with those with a low modulus (Table III). For all the tested composites,  $\tan \delta$  was between 0.058 and 0.067.

According to the statistical analysis (Table IV), the main effects of the rotation speed and mixing temperature and their interaction effect were significant. However, the main effect of the compounding time and other interaction effects were not significant (Table IV). According to Duncan’s multiple-range test, based on the initial complex modulus ( $E^*_{int}$ ), the effect of the mixing temperature at 150°C on  $E^*$  was significantly different from that at 165 and 180°C (Table III). The effect of the rotation speed at 60 rpm on  $E^*$  was also significantly different from that at 90 rpm (Tables III and IV). Although  $E^*$  decreased with an increase in the compounding time, there was no significant difference between the compounding time levels (Tables III and IV).

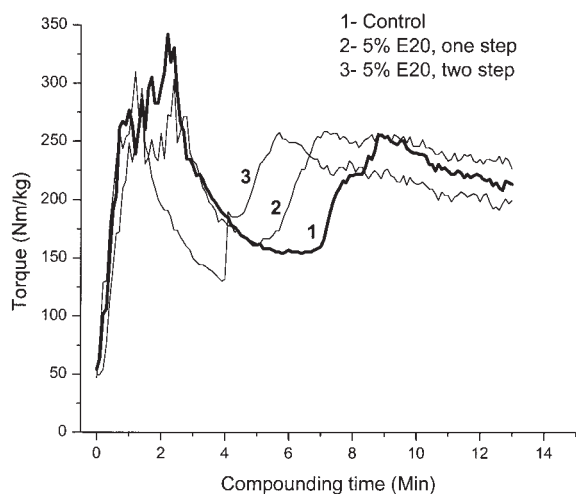
On the basis of this discussion, the optimal compounding parameters at 60 rpm were a mixing temperature of 180°C and a compounding time of 10 min. The optimum compounding conditions at 90 rpm were a temperature of 165°C and a compounding time of 10 min. Therefore, a short compounding time, an appropriate rotation speed, and high mixing temperatures were suitable for improving the compounding quality of modified blends and the dynamic mechanical properties of the resultant composites.

**TABLE IV**  
Three-Way ANOVA of  $E^*$  for the Resultant Composites by the One-Step Process

Source	DF	Type III sum of square	Mean square	F value	Pr > F
Model	17	13.317	0.783	3.13	0.0020
Speed	1	2.843	2.843	11.35	0.0018
Temp	2	4.496	2.248	8.98	0.0007
Speed-Temp	2	2.095	1.047	4.18	0.0233
Time	2	0.0169	0.008	0.03	0.9669
Speed-Time	2	0.193	0.0965	0.39	0.6829
Temp-Time	4	2.284	0.571	2.28	0.0796
Speed-Temp-Time	4	1.389	0.347	1.39	0.2579
Error	36	9.015	0.250		

Speed = rotation speed; Temp = mixing temperature; Time = compounding time.





**Figure 9** Comparison of the one-step and two-step processes for the wood–HDPE blends (50:50). The kneading process was conducted at 165°C and 90 rpm for 10 min. The concentration of E20 was 5 wt% of the oven-dried wood fiber.

### Two-step process versus one-step process

At the same concentration of the coupling agent, the compounding time for dispersion in a two-step process was smaller than that in a one-step process and a control (untreated blends; Fig. 9). For the untreated blends, the maximum dispersion torque occurred at about 9 min after the addition of the wood fiber. For the modified blends with the one-step and two-step processes, it decreased to 7 and 5 min, respectively. Therefore, a coupling treatment by E20 forced the dispersion range to shift left. The two-step process shortened the dispersion period by 2 min with respect to the one-step process and by 4 min with respect to the control, although the values of the maximum dispersion torque were almost the same (Fig. 9). This was due to the fact that the pretreated wood fiber was more easily dispersed in the polymeric matrix. Therefore, a two-step process was usually better than a one-step process because the pretreated wood fiber more effectively improved the interfacial compatibility and miscibility.<sup>12</sup>

### CONCLUSIONS

The compounding conditions directly influenced the compounding quality of the wood fiber and polymer blends and finally affected the interfacial bonding strength and flexural modulus of the resultant composites. With 50 wt % wood fiber, the optimum compounding parameters at 60 rpm for the wood-fiber/

HDPE blends were a temperature of 180°C and a compounding time of 10 min for the one-step process. The optimum compounding conditions at 90 rpm were a temperature of 165°C and a mixing time of 10 min. Therefore, a short compounding time, an appropriate mixing temperature, and a moderate rotation speed improved the compounding quality of the modified blends and the dynamic mechanical properties of the resultant composites.

The loading ratios of the wood fiber significantly influenced the melt torque and blend temperature of the wood–polymer blends. The melt torque and blend temperature followed a polynomial relationship with the loading ratio of the wood fiber. With 50% wood fiber, the melt torque and blend temperature reached the maximum values. Adding coupling agents effectively improved the compatibility between the wood fiber and thermal matrix, and the coupling treatment reduced the melt torque and shortened the compounding time. In general, the melt torque decreased as the coupling agent concentration increased.

For the one-step process, the statistical analysis indicated that the main effects of the rotation speed and mixing temperature and the interaction effect between the speed and temperature were significant. However, the main effect of the compounding time and other interaction effects were not significant. According to Duncan's multiple-range test, there was a significant difference between the levels of the temperature and rotation speed. However, there was no significant difference between the levels of the compounding time. The two-step process was better than the one-step process because the coupling agents were more evenly distributed at the interface with the two-step process.

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