Compounding of Cellulose Fibers with Polypropylene: Effect of Fiber Treatment on Dispersion in the Polymer Matrix

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Synopsis

The compounding of Kraft pulp with polypropylene was studied using a laboratory highshear roller blender. Dispersion was characterized by measuring the size and number of agglomerates in a thin compression-molded sheet. The effects of parameters such as surface treatment and mixing conditions were investigated. It is shown that with a suitable treatment, better dispersion of fibers can be achieved.

INTRODUCTION

The full potential of cellulose fibers as a reinforcing agent has not yet been fully realized. One difficulty encountered during the incorporation of these fibers into the thermoplastic matrix is the interfiber hydrogen bonding which tends to hold the fibers together. The polar nature of wood-based fillers and inorganic reinforcing agents adversely affects the dispersion of such polar fillers in a nonpolar matrix.[1]

The surface adhesion characteristics of a reinforcing fiber is important because of its role in the transmission of stress from the matrix to the fiber.[2] It has been shown that discontinuous cellulose fibers, when treated with a small amount of organic bonding agent reduce fiber-to-fiber interaction and aid dispersion in polyvinyl chloride.[3]

In the present study, compounding of polypropylene with Kraft pulp was carried out in a shear roller blender. The fibers were treated with stearic acid (by vapor phase or solution phase) to facilitate dispersion of the fibers in the polymer matrix. The concentration of stearic acid varied from 0 to 10% by fiber weight. Dispersion of the fiber in the matrix was determined by measuring the size and number of aggregates with an optical microscope. Adsorbed water on the surface of the fiber was characterized by water retention measurements.

EXPERIMENTAL

Materials

Polypropylene (PP 6301) was supplied by Himont Canada Ltd. (Nominal melt flow: 12 g/10 min; density: 0.903 g/cc.) Kraft pulp of aspen (bleached)

was chosen for evaluation. The average length of the fiber was 1.2 mm (mesh size 60) and the average fiber aspect ratio was 40. Stearic acid was used as a dispersing agent.

Fiber Treatment

For better characterization of the effect of a dispersing agent on the fiber, two different methods of fiber treatment were tried.

1. Treatment in vapor phase: Stearic acid was mixed with Kraft pulp (50 g) in a beaker at room temperature and then placed in an oven at 105°C for 45 minutes. The concentration of stearic acid was 1, 2, 3, and 10% by fiber weight. The dried fibers were passed through a grinder (mesh size 60) and then stored for mixing with polypropylene.
2. Treatment in solution phase: In this case, stearic acid was dissolved in ethyl alcohol (50 mL). The solution was then added dropwise to the Kraft pulp with continuous mechanical stirring. The percentage of stearic acid was 1, 2, 3, and 10% by fiber weight. These fibers were dried in an oven at 105°C for 45 minutes and passed through a grinder (mesh size 60) and then stored for mixing with polypropylene.

Incorporation of Treated Fibers in Polypropylene

Compounding of polypropylene and treated fibers (20% by weight of fiber) was carried out in an internal mixer (Brabender roll mill 065). Mixing temperature was 180°C and a roller speed of 70 rpm was maintained for 10–15 min. There was little change in the color of the product after a 15-min mixing period. Higher mixing temperatures were avoided due to yellowing and discoloration of the samples. Thin sheets (9 × 9 cm) of the above mixture were obtained by compression molding at 170°C. The molded sheets were used for optical microscope analysis.

Water Retention Value (WRV)

A small quantity of Kraft pulp (0.5 g) treated with stearic acid was introduced into a centrifuge tube with 3.5 mL of distilled water. The samples were centrifuged at 3000 rpm for 15 minutes (International Equipment Company Model HT). The above speed was chosen to obtain good reproducibility of the results.[4,5] After recovering the pulp from centrifuge, the weight of the sample was measured (A). The above samples were kept in an air-circulating oven at 105°C for 18 h to obtain a constant weight of the dried pulp (B).

$$\text{Water Retention Value (WRV)} = (A - B)/B$$

RESULTS AND DISCUSSION

Characterization of Fiber Dispersion

Kraft pulp treated with stearic acid was extracted with acetone to determine the percentage of stearic acid chemically reacted with the pulp. Table I shows the percent of stearic acid deposited on the fiber, with solution phase

TABLE I
Soxhlet Extraction of Treated (Stearic Acid) Kraft Pulp

Kraft pulp (g)	Stearic acid (g)	Stearic acid add on (%)			
		Solution phase		Vapor phase	
		Before extraction	After extraction	Before extraction	After extraction
5.0248	0.0502	3.40	1.17	1.82	0.31
5.0045	0.1001	6.52	2.39	3.87	0.72
5.0133	0.1503	9.67	3.82	5.04	1.03
5.0029	0.5002	23.74	9.96	10.34	2.12

and vapor phase treatments, before and after extraction. The results indicate that a higher amount of stearic acid was deposited on the fiber in solution phase treatment. After extraction with acetone, the amount of stearic acid chemically bonded with the fiber was higher in solution phase (39.5% at 3% stearic acid) compared to vapor phase (20.5% at 3% stearic acid) treatment.

The dispersion of fiber in the matrix was characterized by measuring (a) the size of the aggregates and (b) the number of aggregates using an optical microscope. Figures 1 and 2 show the number of aggregates and their average size versus percentage of stearic acid in the samples obtained from vapor phase and solution phase treatments. The results indicate a decrease in the number of aggregates with an increase in the concentration of stearic acid (Fig. 1). In the vapor phase treatment, the number of aggregates was 108 in a 81 cm^2 surface area in the samples treated with 1% stearic acid. The average mean diameter of the aggregates was 0.53 mm (Fig. 2).

When the concentration of stearic acid increased to 3%, the number of aggregates decreased to 40 (Fig. 1). The average size of the aggregates was 0.42 mm (Fig. 2). This is much larger compared to the single fiber diameter
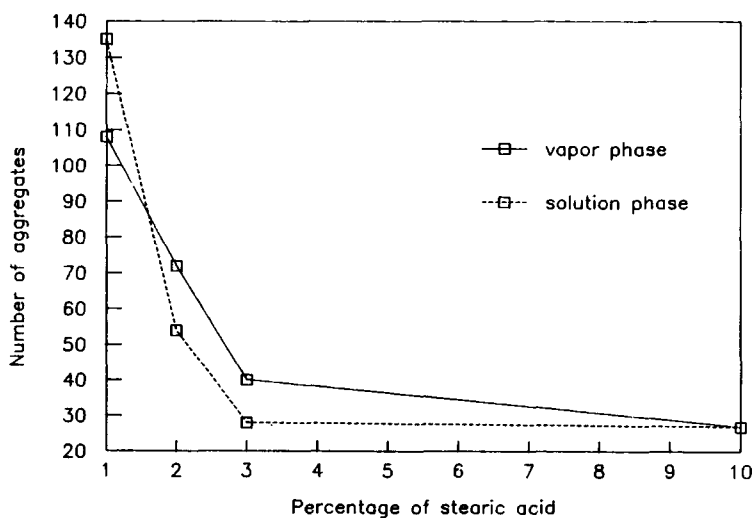

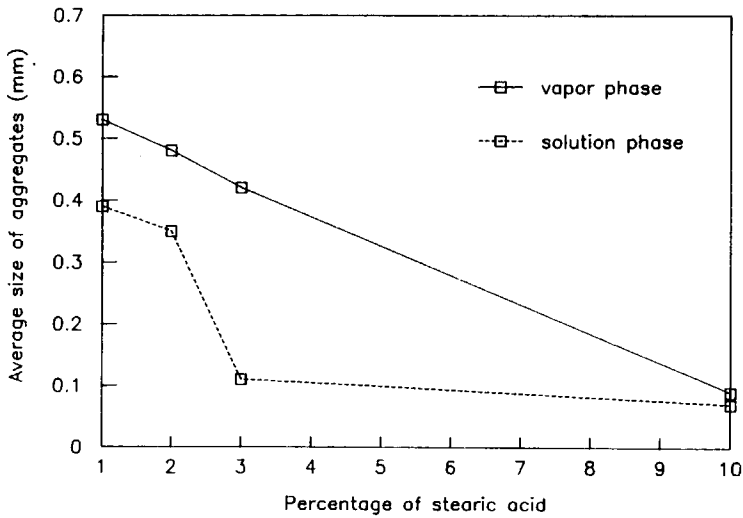Fig. 1. Effect of stearic acid concentration on the number of aggregates.

Fig. 2.   Effect of stearic acid concentration on the size of the aggregates.

(0.03 mm). Further increase in the stearic acid concentration (10%) resulted in only a marginal decrease in the number of aggregates.

In the case of treatment by the solution method, for the samples with 1% stearic acid, the number of aggregates was 135, which is higher than the vapor phase treatment (Fig. 1). When the concentration of stearic acid increased to 3%, the number of aggregates decreased to 27. The average size of the aggregates also decreased to 0.1 mm (Fig. 2). This value is very close to the 0.03 mm diameter of single fiber. At this level of dispersion one might expect an improvement in the tensile properties of the composites. As observed earlier, further increase in the percentage of stearic acid did not result in any significant reduction in the number or size of the aggregates.

The presence of a greater number of aggregates at lower concentrations of stearic acid (1%) is seen from optical micrographs (Figs. 3 and 4). Figure 3 shows that fibers tend to form larger randomly distributed aggregates, due to interhydrogen bonding between fibers (vapor phase treated). However, in the case of the solution-treated samples, at the same concentration of stearic acid, the size of the aggregates is comparatively small (Fig. 4). When the concentration of stearic acid was increased to 3% (Figs. 5 and 6), a decrease in the size of aggregates is observed.

Characterization of the Fiber Surface Treatment

The cellulosic fibers treated with stearic acid, due to the hydrophobic chain of stearic acid molecules, should impart water resistance to the composite. Considering that if the treated cellulosic fibers are exposed to water, one would expect a decrease in hydrophilic behavior with an increase in the stearic acid concentration.

The wetting on the surface of the solid can be studied by contact angle measurement.[6] This method cannot be used in the present case since the average fiber length (1.2 mm) and diameter (0.03 mm) are very small, and can

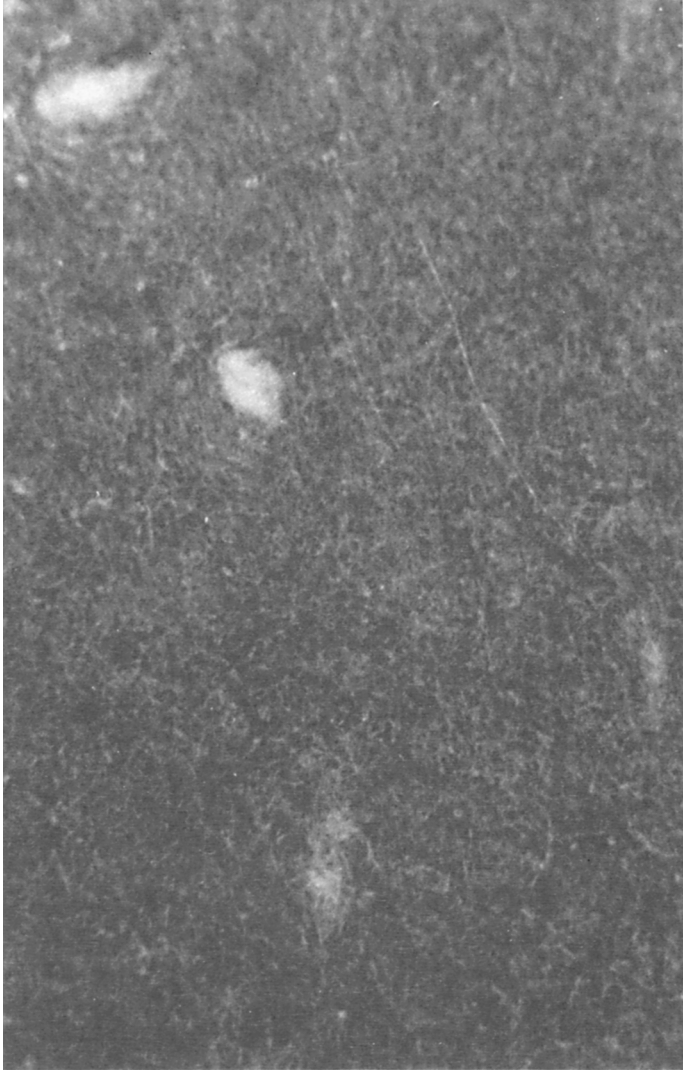Fig. 3.   Optical micrograph of vapor phase-treated sample (1% stearic acid), magnification ×60.

Fig. 4. Optical micrograph of solution phase-treated sample (1% stearic acid), magnification × 60.

Fig. 5. Optical micrograph of vapor phase-treated sample (3% stearic acid), magnification × 60.

Fig. 6.   Optical micrograph of solution phase-treated sample (3% stearic acid), magnification ×60.

TABLE II
Water Retention Values of Treated Kraft Fibers
(Vapor Phase Treatment)

Stearic acid (weight %)	Wet fibers (A) (g)	Dried fibers (B) (g)	WRV = (A − B)/B
0.0	3.9560	0.5446	6.26
1.0	3.3720	0.4910	5.87
2.0	3.2915	0.4900	5.72
3.0	3.2040	0.4820	5.65
10.0	3.1815	0.4819	5.60

only be measured by a high sensitivity apparatus. One of the simplest methods to study the mobility of water on the treated fiber surface is to measure the water retention value (WRV). This test measures degree of hydration, capacity of absorption, and retention of water.[4]

The quantity of water retained in the pulp after centrifuging was measured. This procedure was previously employed for untreated cellulosic pulps, but not for treated pulps.[4,7] In the present study, however, treated pulps were examined. Tables II and III show the water retention value of the samples treated in the vapor and solution phases, respectively. The WRV was higher for untreated samples (no stearic acid), as seen from Table II. A steady decrease in the water retention value was observed with increased concentrations of stearic acid (Fig. 7).

After solution treatment with stearic acid, the WRV decreased considerably (Table III). As in the previous case, the water retention value decreased steadily as the concentration of stearic acid increased (Fig. 7). These results confirm that the surface of the fiber becomes more hydrophobic with increased concentration of stearic acid. This implies a corresponding increase in the contact angle between the fiber surface and water as the concentration of stearic acid increases.

The variation of the water retention value with stearic acid concentration corresponds to the average number and size of the aggregates. The lower water retention values observed in the solution phase treatment are consistent with the reduced number of aggregates.

TABLE III
Water Retention Values of Treated Kraft Fibers
(Solution Phase Treatment)

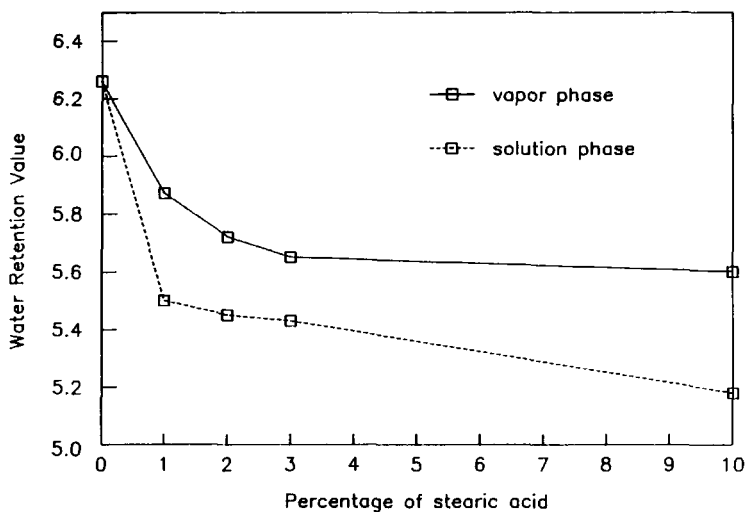| Stearic acid (weight %)	Wet fibers (A) (g)	Dried fibers (B) (g)	WRV = (A − B)/B
0.0	3.9560	0.5446	6.26
1.0	3.2030	0.4925	5.50
2.0	3.1616	0.4900	5.45
3.0	3.1260	0.4860	5.43
10.0	3.0030	0.4858	5.18

Fig. 7.   Effect of stearic acid concentration on water retention values.

CONCLUSIONS

Kraft pulp surface treated with stearic acid showed an improvement in the dispersion of fibers in the polypropylene matrix. Soxhlet extraction of the treated pulp proved that the stearic acid was chemically bonded with the pulp. Characterization of fiber surface showed that 3% stearic acid was sufficient to achieve maximum reduction in the size and number of aggregates. Samples treated with a higher concentration of stearic acid did not have much effect on the dispersion of fibers in the matrix. The surface of treated fibers can be characterized by measuring the water retention value.

References

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