

High Fiber-Low Matrix Composites: Kenaf Fiber/Polypropylene

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Abstract

Considerable interest has been generated in the use of lignocellulosic fibers and wastes (both agricultural and wood based) as fillers and reinforcements in thermoplastics. In general, present technologies limit fiber loading in thermoplastics to about 60 percent by weight of fiber. To produce high fiber content composites for commercial use while maintaining adequate mechanical properties requires innovative processing techniques. A new technique has been developed that allows very high fiber loading. We have processed composites up to about 95 percent by weight of fiber in polypropylene (PP). The process involves some additional processing steps, in addition to conventional compounding techniques, to achieve such high fiber content in PP.

Studies on about 85 percent kenaf-PP composites indicates that properties such as flexural modulus and strength are superior to most types of wood particle, low and medium density hard-

boards. The fiber orientation is predominantly two dimensionally random for the kenaf-PP composite. The range for the flexural strength (MOR) of medium-density fiberboards (MDF) using phenol formaldehyde as the binding system is typically from 13 to 42 MPa. The flexural stiffness (MOE) range for MDF boards is from 2.24 to about 4.9 GPa. The high range for flexural strength of high-density fiberboards is typically about 70 MPa and the flexural stiffness about 7.58 GPa. The 85 percent kenaf-fiber polypropylene composite, with an MOR of 75 MPa and an MOE of 6.4 GPa, has comparable properties to conventional formaldehyde-based fiberboards. Thermal analysis of the kenaf composites indicates that the crystallinity of the PP in the composites is not affected by the processing.

Introduction

Considerable interest has been generated in the use of lignocellulosic, both agricultural (1,2) and wood based, and wastes as fillers and reinforcements in thermoplastics. In general, present technologies limit fiber loading in thermoplastics to about 50 percent by weight of fiber. To produce high fiber content composites for commercial use while maintaining adequate mechanical properties requires innovative processing techniques. A new technique has been developed that allows very high fiber loading. We have processed com-

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posites up to about 90 to 95 percent by weight of fiber in polypropylene.

Use of the higher strength bast fibers such as kenaf, jute, hemp, flax, etc. will result in high mechanical properties as long as there is good interfacial bonding to obtain adequate stress transfer. Kenaf was chosen for the study because it is now a fiber crop grown commercially in the United States. Several other fibers isolated from annual growth crops (jute, hemp, and flax) have potential as reinforcing fillers in plastics. The choice of the fiber for plastics applications depend on the availability of the fiber in the region and also on the ultimate composite properties needed for the specific application. Kenaf filaments are extracted from the bast of the plant *Hibiscus cannabinus*. These filaments consist of discrete individual fibers, generally 2 to 6 mm long, which are themselves, composites of predominantly cellulose, lignin, and hemicelluloses. Filament and individual fiber properties can vary depending on the source, age, separating techniques, and history of the fiber. Furthermore, the properties of the individual fibers are difficult to measure and we have not done so.

Experimental Methods

Kenaf fibers were cut to about 2 cm in length. The fibers were coated with 1 to 4 percent by weight with glycerine in order to attain such high fillings in composites. A polypropylene homopolymer (PP- Solvay 1602) and a maleated polypropylene was used as a coupling agent. The short fibers, MAPP and PP (the latter two in pellet form) were compounded in a 1-liter high intensity kinetic mixer (Synergistics Industries Ltd., Canada) where the only source of heat is generated through the kinetic energy of rotating blades. The fibers and polymers were inserted into the blender and compounded at 5,000 rpm that resulted in a blade tip speed of about 30 m/sec. The compounded mass was then automatically discharged at 200°C. The precise time to discharge depended on the amount of fiber and polymer, and their ratios. On an average the time of blending varied from 2 to 3 minutes with higher fiber content blends taking longer to reach 200°C. The compounded mass was then immediately pressed in a hot press.

Four flexural specimens and four impact specimens were cut from each of the three boards. Specimen dimensions were according to the respective ASTM standards. Samples were stored in controlled conditions prior to testing. Tests were con-

Table 1. ~ Comparison of flexural properties of commercial available formaldehyde-based wood composites with data on ~ 85% filled kenaf-PP composites. ^a

	Strength range (MPa)		Modulus range (GPa)	
	low	high	low	high
High-density fiberboards (commercial)	38	69	4.48	7.58
Medium-density fiberboards (commercial)	13.1	41.4	2.24	4.83
85% kenaf-PP ^b	75 (±9)		6.8 (±0.5)	
60% kenaf-PP ^c	110		10.1	

^a Standard deviations are in brackets for the kenaf composites.

^b Compression molded, random alignment.

^c Injection molded, significant fiber alignment.

ducted according to standard ASTM tests: Izod impact strength tests according to ASTM D 256-90 and flexural testing using the ASTM 790-90 standard.

Results and Discussion

Studies on 85 percent kenaf-thermoplastic composite indicates that properties such as flexural modulus and strength are superior to most types of wood particle, low and medium density hardboards. These results indicate the stiffness of the new materials (85% by weight of kenaf) is very high, and could even compete with oriented strandboard in some applications (Table 1). The data for the 85 percent composites were obtained from three different boards, with four specimens from each of the boards. *Note that the 85 percent composites were compression molded and therefore the fibers were randomly oriented.*

Scanning electron microscopy (SEM) of above 85 percent boards showed that the matrix appears in a continuous phase and the bonding between the fiber and matrix is good, figure 1 a and b. There are some voids present in the composite and this is seen in Figure 1, top. Figure 1, bottom shows a close-up of a fiber in an unnotched Izod test specimen. Considerable energy has been absorbed causing the fiber to split, resulting in fine fibrils being exposed.

The incorporating of a high fiber volume fraction in the composite poses practical difficulties in terms of formation or uniformity of mass distribu-

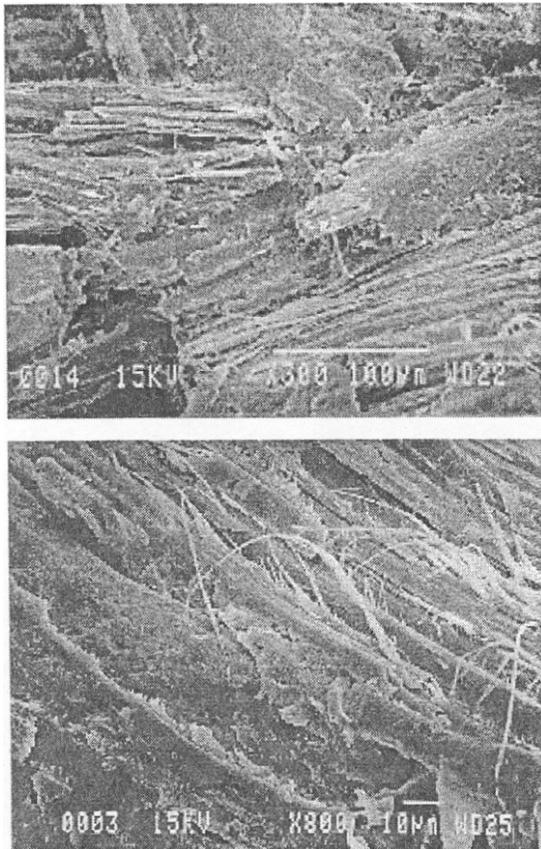


Figure 1. – Fracture surface of 85% kenaf composite (top) Izod notched (300×) and (bottom) Izod unnotched (800×).

tion. The maximum volume fraction of perfectly aligned cylindrical and rigid fibers that can be packed into a composite is theoretically 90 percent. This is lower for randomly distributed fibers. However, it is possible that parts of the amorphous part of kenaf fibers, lignin, and hemicellulose, can blend in with the PP/MAPP thermoplastic polymer and be part of the overall matrix.

The lignin content in kenaf fiber is reported to be between 15 to 19 percent and the hemicellulose content around 22 and 23 percent. The crystalline cellulose content of kenaf fibers can vary between 44 and 57 percent in the natural fiber. Thus for an 85 percent kenaf filled PP/MAPP, the amount of lignin and hemicellulose content in the composite works out to be approximately 34 percent. If one uses 55 percent for the amount of cellulose content in kenaf fiber, then the 85 percent by composite will have about 47 percent crystalline cellulose (the reinforcing element), 15 percent PP/MAPP, and about 34 percent lignin and hemicellulose in the blend. The lignin and hemicellulose is plas-

Table 2. ~ Crystallinity data of the plastic matrix for kenaf composites.

Composite type ^a	T _c peak	Onset T _c	T _m peak	% crystallinity
	----- (°C) -----			
100% PP	110.63	116.33	161.70	44.5
60% PP/MAPP ^b	122.96	127.12	163.67	44.1
85%-PP/MAPP ^c	122.96	127.51	163.70	42.6

^a MAPP was used as the compatibilizer/coupling agent; Pure PP crystals-crystallization enthalpy = 209.3 J/g.

^b injection molded.

^c compression molded.

ticized to allow molecular chain mobility. Thus, if one looks at this point of view, it is not inconceivable to have highly filled kenaf composites, since the reinforcing element is far below the limits imposed by packing of fibers in a composite.

Differential thermal analysis and dynamic mechanical analysis (DMA) of the 85 percent composites were compared to 60 percent kenaf-PP composites that were also compatibilized using MAPP. The 60 percent composites were not plasticized and samples were prepared by injection molding. Differential scanning calorimetry (DSC) results indicate there is little difference in the onset and peak crystallization and the melting temperatures between the kenaf composites (Table 2). This could indicate that the plasticizer used does not affect this factor. The percent crystallinity of the 85 percent was marginally lower than the 60 percent composite. These data were from an average taken from two samples. It must be noted that we used about 10 mg of the composite for DSC data collection. The estimation of the exact percent of the plastic matrix in the 10 mg samples is needed to get accurate percent crystallinity data. So the difference in the percent crystallinity between the 85 percent and other composites may result from a fluctuation of composite composition rather than representing the true value. All we can conclude is that the percent crystallinity of the 85 percent composites is very different from the other composites.

The DMA spectra of the 85 percent filled composites show some interesting features when compared to a 60 percent filled kenaf composite that was injection molded. It should be noted the 60 percent composites were injection molded, which results in a significant amount of fiber alignment. This is as opposed to the compression molded 85

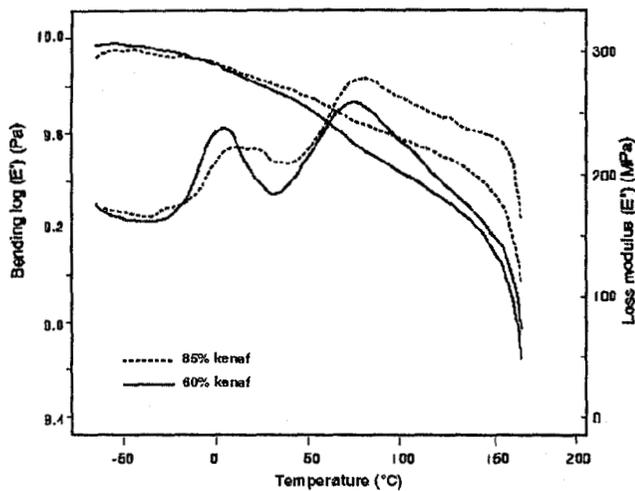


Figure 2. ~ Storage and loss modulus of a 85% fiber filled composite compared to injection molded 60% fiber filled composite.

percent composites where the fibers were randomly distributed. At low temperatures, the 60 percent composites have a higher storage modulus (E'), which suggests a brittle material at these temperatures. However, after about 10°C, the 85 percent composites have a higher modulus. The difference in storage moduli become even more pronounced as the temperature increases. The softening temperature of the 85 percent is also higher than the 60 percent composite. The lower modulus at low temperature and the higher modulus at higher temperature is an advantage of the 85 percent composite, since the composite is not as brittle as the 60 percent at lower temperatures but maintains its integrity better at higher temperatures.

In terms of the loss modulus (E'') spectra, two transitions can be clearly seen. The **b** transition, occurring at about 10° to 25°C is associated with the glassy-rubbery transition of the amorphous chains of the polymer. The **a** transition is related to the crystallites, but the relaxation occurs due to the presence of "rigid" amorphous molecules present in the crystal (3-5). These regions disrupt the purity of the crystals and thus can be termed as defects. The **b** relaxation for the 85 percent composite is much wider than the 60 percent composite. The wider relaxation generally indicates that some of the amorphous molecules are more restricted than others. This is possible when amorphous molecules bridge two crystals and result in higher temperatures for their glass-rubbery transition. Secondly,

any amorphous molecules near the fiber surface are restricted in mobility due to interactions with the solid fiber surface. Since the 85 percent composite has a significantly higher fiber surface exposed to the matrix, the polymer molecules near the surface have restricted mobility and therefore result in higher **b** relaxation temperatures.

Conclusions

The plasticization technique allows the processing of highly filled composites using melt blending technology. Highly filled 85 percent kenaf fiber PP composites show some interesting results. Their flexural properties exceed those of typical, commercially available formaldehyde-based wood composites. DSC and DMA results were compared to 60 percent injection molded kenaf-PP composites that were prepared without using the plasticization technique. The DSC results indicate that the crystallinity of the PP matrix phase is not significantly affected in the 85 percent composite. The dynamic mechanical properties of the 85 percent composites has a higher storage moduli above 10°C and a higher softening temperature as compared to the 60 percent kenaf-PP composite. The flexural modulus and strength (MOR and MOE) of the 85 percent kenaf-PP composites are superior to most types of wood low, medium, and high-density fiberboards.

The possibility of using highly filled agro-based fiber thermoplastic composites for furniture, automotive and building applications makes this a particularly attractive material for countries that have scarce wood resources or those countries that want to develop crops for new uses to save precious forest resources.

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