

# PROPERTIES OF KENAF/POLYPROPYLENE COMPOSITES

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### ABSTRACT

Combining kenaf fiber with other resources provides a strategy for producing advanced composite materials that take advantage of the properties of both types of resources. It allows the scientist to design materials based on end-use requirements within a framework of cost, availability, recyclability, energy use, and environmental considerations. Kenaf fiber is a potentially outstanding reinforcing filler in thermoplastic composites. The specific tensile and flexural moduli, for example, of a 50% by volume of kenaf-polypropylene (PP) composite compares favorably with a 40% by weight of glass fiber-PP injection molded composite. Results indicate that kenaf fibers are a viable alternative to inorganic/mineral-based reinforcing fibers as long as the right processing conditions and aids are used, and for applications where the higher water absorption of the lignocellulosic-based fiber composite is not critical.

### INTRODUCTION

It is possible to make completely new types of composite materials by combining different resources. It is possible to combine, blend, or alloy natural fibers with other materials such as glass, metals, plastics, and synthetics to produce new classes of materials. The objective will be to combine two or more materials in such a way that a synergism between the components results in a new material that is better than the individual components.

One of the big new areas of development is in combining natural fibers with thermoplastics. Since prices for plastics have risen sharply over the past few years, adding a natural powder or fiber to plastics provides a cost reduction to the plastic industry (and in some cases increases performance as well), but to the lignocellulosic-based industry, this represents an increased value for the lignocellulosic-based

component. Research has concentrated in two basic areas: one in which no attempt is made to compatibilize the two dissimilar resources and, a second in which a compatibilizer is used to make the hydrophore (plastic) mix better with the hydrophil (lignocellulosic). In the first case, the lignocellulosic-fiber is added as a relatively low-cost filler and in the second, the lignocellulosic-fiber is added as a reinforcing filler. Both of these types of materials are usually referred to as natural fiber/thermoplastic blends.

Several million metric tons of fillers and reinforcements are used annually by the plastics industry. The use of these additives in plastics is likely to grow with the introduction of improved compounding technology, and new coupling and compatibilizing agents that permit the use of high filler/reinforcement content (Katz and Milewski 1987). As suggested by Katz and Milewski (1987), fillings up to 75 parts per hundred (pph) could be common in the future. This level of filler could have a tremendous impact in lowering the usage of petroleum-based plastics. It would also be particularly beneficial, both in terms of the environment and also in socioeconomic terms, if a significant amount of the fillers were obtained from a renewable agricultural source.

The primary advantages of using lignocellulosic fibers as fillers/reinforcements in plastics are numerous. Such fibers may exhibit low densities, non abrasiveness, high specific properties (such as high filling levels possibly resulting in high stiffness properties), easily recycled nonbrittle fibers, sharp curvature allowances (with no fracture), and biodegradability. There may also be wide fiber availability (throughout the world), rural job generation, enhancement of nonfood agricultural/farm-based economy, low energy consumption, and lower costs.

The main limitation to the use of lignocellulosic fibers is the lower processing temperature permissible due to the possibility of fiber degradation and/or the possibility of volatile emissions that could affect composite properties. The processing temperatures are thus limited to about 200°C, although it is possible to use higher temperatures for short periods. This processing factor limits the type of thermoplastics that can be used with lignocellulosic-fibers; to commodity thermoplastics such as polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC) and polystyrene (PS). However, it is important to note that these lower-priced plastics constitute about 70% of the total thermoplastic consumed by the plastics industry, and consequently the use of fillers/reinforcement presently used in these plastics far outweigh the use in other more expensive plastics.

Another drawback is the high moisture absorption of lignocellulosic fibers. Moisture absorption can result in swelling of the fiber resulting in dimensional stability problems in the lignocellulosic fiber composites. The absorption of moisture by the fibers is minimized in the composite due to encapsulation by the polymer and good fiber-matrix bonding. Good adhesion decreases the rate and amount of water absorbed in the interphase region of the composite. It is difficult to entirely eliminate the absorption of moisture without using expensive surface barriers on the composite surface. The moisture pick up of the fibers can be dramatically reduced through the acetylation of some of the hydroxyl groups present (Rowell et al. 1986)

in the fiber, but with some increase in the cost of the fiber. However, moisture absorption can have some secondary benefits, such as reducing static electricity in the final plastic object. Water absorption and specific gravity of lignocellulosic fiber composites are important characteristics that determine end use applications of these materials. Water absorption could lead to a decrease in some of the properties and should be considered when selecting applications. It is difficult to entirely eliminate the absorption of moisture in the composites without using expensive surface barriers on the composite surface. Water absorption in lignocellulosic-based composites can lead to a build-up of moisture in the fiber cell wall and also in the fiber-matrix interphase region which would result in fiber swelling and affect the dimensional stability. Good wetting of the fiber by the matrix and adequate fiber-matrix bonding can decrease the rate and amount of water absorbed in the interphasial region of the composite.

It is important to keep these limitations in perspective when developing end-use applications. There is reason to believe that by understanding the limitations and benefits of these composites, these renewable fibers are not likely to be ignored by the plastics/composites industry for use in the automotive, building, appliance and other applications. Material cost savings due to the incorporation of the relatively low-cost lignocellulosic fibers and the higher filling levels possible, coupled with the advantage of being non-abrasive to the mixing and molding equipment, are a few of the benefits.

Most of the prior research on lignocellulosic fibers in thermoplastics deals with the use of wood flour and wood fiber-based fillers/reinforcements, and significant advances have been made by a number of researchers (Woodhams et al. 1984, 1990; Klason and Kubat 1986a, b; Bataille et al. 1989, Kokta et al. 1989, Raj and Kokta 1989, Yam et. al. 1990, Myers et al. 1992). Proper selection of additives was necessary in these studies to improve composite efficiency by improving fiber dispersion and the interaction and adhesion between the lignocellulosic fibers and plastic matrix.

Lignocellulosic fillers/fibers can be classified under three categories, depending on their performance when incorporated in a plastic matrix. (1) Flour can be considered as particulate fillers that enhance (increase) the tensile and flexural moduli of the composite with little effect on the composite strength. (2) Fibers have higher aspect ratios and contribute to an increase in the moduli of the composite, and can also improve its strength when suitable additives are used to improve stress transfer between the matrix and the fibers. The improvement in modulus is not significantly different from the cellulosic particulate fillers. (3) The most efficient lignocellulosic additives are long natural fibers such as kenaf. The specific Young's modulus and specific flexural modulus, the ratio of the composite modulus to the composite specific gravity of composites with natural fibers such as kenaf, are significantly higher than those possible with wood fibers. The specific moduli (the ratio of the composite modulus to the composite specific gravity) of high fiber volume fraction bast fibers-PP composites are high and in the same range as glass fiber-PP composites (Vollenberg and Hinkens 1989). The most efficient natural fibers are

those that have a high cellulose content coupled with a low micro fibril angle, resulting in high filament mechanical properties.

The inherent polar and hydrophilic nature of the lignocellulosic fibers and the non-polar characteristics of the polyolefins result in difficulties in compounding/blending the fibers and matrix. Maleic anhydride (MA) grafted polypropylene (MAPP) has been shown to function efficiently as a coupling agent for kenaf-PP systems. Earlier results suggest that the molecular weight and amount of MA grafted are important parameters that determine the efficiency of the additive (Felix et al. 1993, Sanadi et al. 1993). The maleic anhydride present in the MAPP provides polar interactions such as acid-base interactions and can also covalently link to the hydroxyl groups on the lignocellulosic fiber. Han et al. (1991) reported that due to thermodynamic segregation, the MAPP gets localized on the cellulosic fiber surface in a PP matrix during the processing stage. This surface phenomenon was inferred from TEM-EDXA studies of osmium tetroxide-labeled MAPP. ESCA analysis also indicated the concentration of the MAPP on the fiber surface (Felix and Gatenholm 1991). The formation of covalent linkages between the MA and the -OH on the fiber surface have been indicated through IR and ESCA analysis by Gatenholm et al. (1992). The combination of covalent linkages and/or acid-base interaction between the MAPP and the -OH groups on the fiber surface results in good fiber surface-interface properties.

The interactions between non-polar thermoplastics such as PP and any coupling agent, such as MAPP, is predominantly that of chain entanglement. Stresses applied to one chain can be transmitted to other entangled chains and is distributed among many chains. These entanglements function like physical cross links that provide some mechanical integrity up to, and above, the glass transition temperature,  $T_g$ , but become ineffective at much higher temperatures (Neilson 1974). When polymer chains are very short, there is little chance of entanglements between chains and they can easily slide past one another (Neilsen 1977). When the polymer chains are longer, entanglement between chains can occur, chain slippage becomes more difficult, and the viscosity of the polymer becomes higher. A minimum chain length or a critical molecular weight ( $M_c$ ) is necessary to develop these entanglements, and a typical polymer has a chain length between entanglements equivalent to a  $M_c$  varying from 10,000 to about 40,000. The  $M_c$  varies, depending on the structure of a polymer. For example, linear polyethylene has a  $M_c$  for entanglements of about 4,000 (Neilsen 1977), while for polystyrene, the  $M_c$  is about 38,000. Factors such as the presence of hydrogen bonding or side chains that affect the glass transition temperature of the polymer will also affect the  $M_c$  of the polymer melt. It is also important to note that the fiber surface is likely to act as a boundary and restrict the mobility of the polymer molecules, and the minimum entanglement lengths ( $M_c$ ) will vary according to the fiber surface characteristics.

A maleic anhydride grafted PP that has a high MA content coupled with a relatively high molecular weight has resulted in efficient composites (Sanadi et al. 1994). That MAPP is reported to have a number average molecular weight ( $M_n$ ) of 20,000, a weight average molecular weight ( $M_w$ ) of 40,000 and was about 6% by

weight of maleic anhydride. Any free anhydride present in the MAPP can complicate the understanding of the characteristics and function of the MAPP on the properties of the fiber-matrix interphase. The free MA may preferentially bond to available -OH sites on the fiber and reduce the interaction between the MAPP and the fiber. Furthermore, free MA bonded to the fiber surface can change the surface energetic of the fiber surface. Use of a MAPP with higher molecular weights, but lower MA contents than the MAPP mentioned earlier, result in composites with lower properties (Sanadi et al. 1995). Theoretically, extremely long chains of MAPP with substantial amounts of grafted MA would be an ideal additive in kenaf-PP composites, creating both covalent bonding to the fiber surface and extensive molecular entanglement to improve properties of the interphase. However, extremely long chains may reduce the possibility of migration of the MAPP to the fiber surface because of the short processing times. If the MW of the MAPP is too high, the MAPP may entangle with the PP molecules so that the polar groups on the MAPP have difficulty "finding" the -OH groups on the fiber surface.

The purpose of this paper is to explore the use of kenaf bast fiber in polypropylene-based thermoplastic composites both with and without a coupling agent.

## MATERIALS AND METHODS

Kenaf strands harvested from mature plants were obtained from AgFibers Inc., Bakersfield, CA, and cut into lengths of about 1 cm. In all experiments, the weight and volume percent reported is the amount of oven-dry fiber present in the blend. The polypropylene was Fortilene-1602 (Solvay Polymers 1991) with a melt flow index of 12 g/10 min as measured by ASTM D- 1238. A maleic anhydride grafted polypropylene (MAPP), Epolene G-3002 (Eastman Chemical Products, TN), was used as a coupling agent to improve the compatibility and adhesion between the fibers and matrix. The MAPP had a number average molecular weight of 20,000 and a weight average molecular weight of 40,000 and an acid number of 60. An acid number of 60 is about equivalent to 6% (w/w) maleic anhydride in the G-3002 (Eastman Chemical Company 1992).

The kenaf fiber was compounded with PP alone or with MAPP and PP (the latter two in pellet form) in a high intensity kinetic mixer (Synergistic Industries Ltd., Canada) where the only source of heat is generated through the kinetic energy of rotating blades. The kenaf/PP blending was accomplished at  $482 \text{ rad s}^{-1}$  (4600 rpm), and then the mixture automatically discharged at  $190^\circ\text{C}$ . A total weight (fibers and PP, or PP and MAPP) of 150 g were used for each batch, and about 1.5 kg of blended material was prepared for each set of experiments. Fiber weight varied from 20% to 60% and coupling agent weight varied from 0% to 3% of the final composite weight. The total residence time of the blending operation was dependent on the proportions of fiber and PP present and averaged about 2 min.

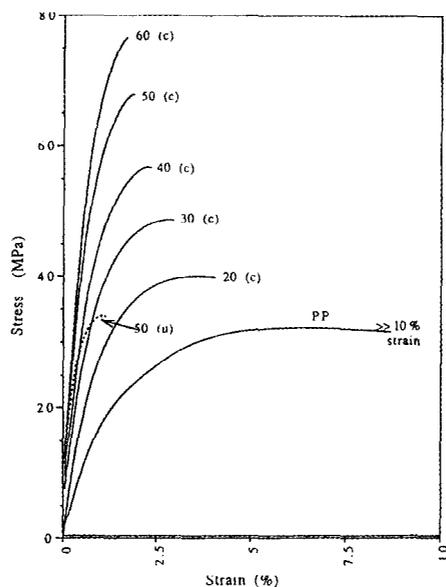
The mixed blends were then granulated and dried at  $105^\circ\text{C}$  for 4 h. Test speci-

mens were injection-molded at 190°C using a Cincinnati Milacron Molder and injection pressures varied from 2.75 MPa to 8.3 MPa depending on the constituents of the blend. Specimen dimensions were according to the respective ASTM standards. The specimens were stored under controlled conditions (20% relative humidity and 32°C) for three days before testing. Tensile tests were conducted according to ASTM 638-90, Izod impact strength tests according to ASTM D 256-90, and flexural testing using the ASTM 790-90 standard. The cross-head speed during the tension and flexural testing was 5 mm/min. Although all the experiments were designed around the weight percent of kenaf in the composites, fiber volumes fractions can be estimated from composite density measurements and the weights of dry kenaf fibers and matrix in the composite. The density of the kenaf present in the composite was estimated to be 1.4 g/cm<sup>3</sup>.

## RESULTS AND DISCUSSION

Preliminary measurements of fiber lengths present in the composite after injection molding showed that few fibers are longer than 0.4 mm due to mixing degradation of fibers. The strengths obtained in composites of this study were, thus, limited by the short fiber lengths. Higher strengths are likely if alternate processing techniques are developed that reduce the amount of fiber attrition (fiber shortening due to breakage) while at the same time achieving good fiber dispersion.

Figure 32.1 shows a typical stress-strain curve for pure PP, along with curves for uncoupled 50% (w/w) kenaf-PP, and coupled systems with increasing amounts of kenaf in the composite. The numbers near the end of the curves indicate the amount of kenaf by weight percent and (c) indicated coupled and (u) indicated uncoupled composites. All coupled systems contained 2% (w/w) MAPP. PP failure strain was greater than 10%. In the experiments using only fiber and PP, the failure strain decreases with the addition of the fibers (Sanadi et al. 1995). Addition of a rigid filler/fiber restricts the mobility of the polymer molecules to flow freely past one another, and thus, causes premature failure. Addition of MAPP followed a similar trend to that of the uncoupled system, although the drop in failure strain with increasing fiber amounts was not as severe. The decrease in the failure strain with increasing amounts of kenaf for the coupled systems is apparent (Figure 32.1). The non-linearity of the curves is mainly due to plastic deformation of the matrix. The distribution of the fiber lengths present in the composite can also influence the shape of the curve, since the load taken up by the fibers decreases as the strain increases and detailed explanations are available elsewhere (Hull 1981). The tensile energy absorption, the integrated area under the stress-strain curve up to failure, behaves in roughly the same manner as the tensile failure strain. The difference between the coupled and uncoupled composites increases with the amount of fibers present, although the drop in energy absorbed for the coupled composites levels off after the addition of about 35% (v/v) fiber.



**Figure 32.1.** Typical stress-strain curves for pure polypropylene (PP), coupled (C) and uncoupled (U) composites with varying amounts of kenaf fibers.

A small amount of the MAPP (0.5%, w/w) improved the flexural and tensile strength, tensile energy absorption, failure strain and un-notched Izod impact strength. These properties are improved by using 1% (w/w) MAPP but there was little difference in the properties obtained between the 2% and 3% (w/w) MAPP systems. The anhydride groups present in the MAPP can covalently bond to the hydroxyl groups of the fiber surface. The improved interaction and adhesion between the fibers and the matrix leads to better matrix to fiber stress transfer.

The specific tensile and flexural moduli of the 50% (w/w) kenaf coupled [2% (w/w) MAPP] composites were about equivalent to, or higher than, typical reported values of 40% (w/w) coupled glass-PP injection-molded composites. Table 32.1 shows typical commercial PP composites compared with kenaf-PP composites. Data on mineral-filled systems from various sources, *Modern Plastics Encyclopedia* (1993) and *Machine Design: Materials Selector Issue* (1994) are included for comparison. The specific flexural moduli of the kenaf composites with fiber contents greater than 40% were extremely high and even stiffer than a 40% mica-PP composite.

The tensile and flexural moduli of the kenaf composites showed significant improvements with the addition of the fiber as compared to PP alone. The tensile and flexural strength doubled with the addition of kenaf fiber as compared to PP alone.

The impact strength of the composite depends on the amount of fiber and the

type of testing, i.e., whether the samples were notched or un-notched. In case of notched samples, the impact strength increases with the amount of fibers added until a plateau is reached (at about 45% fiber weight), irrespective of whether MAPP was used or not. The fibers bridge cracks and increase the resistance to the propagation of the crack. Contribution from fiber pullout is limited since the aspect ratio of the fibers in the system is well below the estimated critical aspect ratio of about 0.4 mm (Sanadi et al. 1993). The impact strength can be increased by providing flexible interphase regions in the composite or by using impact modifiers, and some work has been done in this area at the U W and FPL (Wieloch et al. 1994).

**Table 32.1.** Properties of filled/reinforced polypropylene composites.

Filler/Reinforcement in PP	Unit	None	Kenaf	Glass	Talc	Mica
Filler by Weight	%	0	50	40	40	40
Filler by Volume	%	0	39	19	18	18
Specific Gravity	--	0.9	1.07	1.23	1.27	1.26
Tensile Modulus	GPa	1.7	8.3	9	4	7.6
Specific Tensile Modulus	GPa	1.9	7.8	7.3	3.1	6.0
Tensile Strength	MPa	33	65	110	35	39
Specific Tensile Modulus	MPa	37	61	89	28	31
Flexural Strength	MPa	41	98	131	63	62
Specific Flexural Strength	MPa	46	92	107	50	49
Flexural Modulus	GPa	1.4	7.3	6.2	4.3	6.9
Specific Flexural Modulus	MPa	1.6	6.8	5.0	3.4	5.5
Elongation at Break	%	>>10	2.2	2.5	--	2.3
Notched Izod Impact	J/m	24	32	107	32	27
Water Sorption, 24-h	%	0.02	1.05	0.06	0.02	0.03

Comparison data taken from Modern Plastics Encyclopedia (1993) and Machine Design: Materials Selector Issue (1994),

A typical 50% (w/w) kenaf-homopolymer PP blend absorbed about 1.05% by weight of water in a 24-h water soak test. This level of water is considerably higher than any mineral-filled system. It is, therefore, very important to select applications where this high water absorption is not a critical factor such as in electrical housing components.

The specific gravity of lignocellulosic-based composites is much lower than the mineral-filled thermoplastic systems. The apparent density of the lignocellulosic fibers in PP is about 1.4 g/cm<sup>3</sup> as compared to mineral fillers/fibers (about 2.5 g/cm<sup>3</sup>). The specific gravity of a 50% (w/w) kenaf-PP composite is about 1.07, while that of a 40% (w/w) glass-PP composite is 1.23. The specific mechanical properties of kenaf-PP composites compare favorably to other filled commodity

plastics. Since materials are bought in terms of weight, and pieces or articles are in general sold by the number, more pieces can be made with lignocellulosic fibers as compared to the same weight of mineral fibers. This could result in significant material cost savings in the high-volume and low-cost commodity plastics market.

## CONCLUSIONS

The use of kenaf fiber has resulted in significant property advantages as compared to typical wood flour or wood based fibers for plastic/fiber composites. In fact, the outstanding specific moduli of the kenaf-fiber PP composites compare well with that of glass fiber, talc, and micareinforced molded PP composites.

The cost of kenaf fibers is less than that of the plastic matrix in cellulosic-based composites, and high fiber loading can result in significant material cost savings. The cost of compounding is unlikely to be much more than for conventional mineral/inorganic based-composites presently used by the plastics industry. Due to the lower specific gravity of the cellulosic-based additives (approximately 1.4 as compared to about 2.5 for mineral-based systems), there would be a definite weight advantage for these composites which may have implications in the automotive and other transportation applications. Furthermore, using the same weight of plastic/natural fiber as for example plastic/glass fiber, about 20% more pieces are possible with the cellulosic-based system. Kenaf fibers are soft and non-abrasive and high filling levels are possible; 60% (w/w) of fiber has been successfully incorporated in PP based composites. Reduced equipment abrasion and the subsequent reduction of re-tooling costs through the use of agricultural based fibers is definitely a factor that will be considered by the plastics industry when evaluating these natural fibers. It is important to point out that we do not anticipate, nor intend, the total replacement of conventional based fillers/fibers with agricultural-based fillers/fibers. The expectation is that these natural materials will develop their own niche in the plastics filler/fiber market in the future.

The volume of thermoplastics used in the housing, automotive, packaging and other low-cost, high-volume applications is enormous. Recent interest in reducing the environmental impact of materials is leading to the development of newer materials or composites that can reduce the stress to the environment. In light of potential future petroleum shortages and pressures for decreasing the dependence on petroleum products, there is an increasing interest in maximizing the use of renewable materials. The use of agricultural materials as a source of raw materials to the industry not only provides a renewable resource, but could also generate a non-food source of economic development for farming and rural areas. Appropriate research and development funding in the area of agricultural-based fillers/fibers in making filled-plastics could lead to new value-added, non-food uses of agricultural materials. We believe that the amount of these fibers used in the automotive, furniture, housing, packaging, toy and other industries could easily be in the range hundreds of millions of pounds annually.

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