Property Improvement Effects of Agricultural Fibers and Wastes as Reinforcing Fillers in Polypropylene-Based Composites

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Abstract

The tensile, flexural, and impact properties of five agricultural fiber/polypropylene composites were evaluated and compared to a 40 percent fiberglass/polypropylene and 40 percent talc/polypropylene composites. The fibers used in this study were abaca, henequen, jute, kenaf, and sisal fibers. Jute and kenaf are bast (soft) fibers, while abaca, henequen, and sisal are leaf (hard) fibers. All of the composites were modified with maleic anhydride grafted polypropylene (MAPP) to improve the fiber/matrix adhesion. These composites were 50 percent agricultural fiber, 47 percent polypropylene, and 3 percent MAPP. In comparing the strengths and moduli of the bast fibers to leaf fibers, the bast fiber composites have higher properties than the leaf fiber composites. Impact toughness values are higher for the leaf fibers versus bast fiber composites.

In addition to the five agricultural fibers, 12 agricultural waste/polypropylene composites were evaluated for tensile, flexural, and impact properties and compared to a 40 percent talc-filled polypropylene composite and 100 percent polypropylene. The wastes used in this study include kenaf core, oat straw, wheat straw, oat hulls, wood flour (pine), corn cobs, hard corn cobs, rice hulls, peanut hulls, corn fiber, soybean hull residue, and jojoba seed meal. Composite interfaces were modified with MAPP to improve the mechanical properties through increased adhesion between the fibers and the matrix. These composites had compositions of 50 percent agricultural waste, 48 percent polypropylene, and 2 percent MAPP. The mechanical properties of kenaf core, oat straw, wheat straw, and oat hulls compare favorably to both 50 percent wood flour and 40 percent talc-filled polypropylene, which are both commercial available and used in the automotive industry and furniture markets.

Introduction

Lignocellulosic fibers (wood flour) have been used in the plastics industry for almost 90 years. Bakeland used wood flour to both extend and improve the processability of thermosetting resins in 1907. The use of fillers and reinforcements in plastics has changed the face of the industry. Over 500 million pounds of fibrous material (mostly fiberglass) is being used in the plastics industry yearly (3). The total
the type of fiber used and the amount of fiber loading in the composite. The fiber loading is based on the weight percentage of dry fibers. Also, the effects on the mechanical properties of the composites due to the modification of the interphase/interface region with a maleic anhydride grafted polypropylene (MAPP) coupling agent are evaluated. MAPP has been shown to improve the surface adhesion between the hydrophilic, polar lignocellulosic fibers and the hydrophobic, nonpolar polypropylene (8). All five agricultural fiber composite systems have the following compositions: 50 percent fiber, 47 percent polypropylene, and 3 percent MAPP. The agricultural waste composite systems have the following composition: 50 percent waste, 48 percent polypropylene, and 2 percent MAPP.

Experimental procedures

Materials

The base resin was a polypropylene homopolymer, Fortilene 1602 (Solvay Polymer, Houston, Tex.) with a melt flow index of 12 gr. per 10 minutes at 230°C (ASTM D 1238). The MAPP modifier, Epolene G3002 (Eastman Chemical, Kingston, Tenn.), was used to enhance the surface adhesion between the fibers/wastes and the polypropylene matrix. The fibers/wastes used in this study were abaca (John Rauschenberger, Co.); kenaf core (AG-Fibers Inc.); grade 62 sisal, corncobs, and hard corncobs (Composition Materials Inc.); corn fiber (Cargill, Inc.); oat hulls (Quaker Oats Co.); rice hulls (Busch Agricultural Resources Center); peanut hulls (Seminole Peanut Co.); soybean hull residue (Wis. Soybean Assoc.); jute, henequen, oat straw, wheat straw, wood flour, and jojoba seed from other sources.

Methods

The agricultural fibers were run through a Ball & Jewell granulator (Sterling, Inc., Milwaukee, Wis.) with a 3/8-inch screen to reduce the fibers to approximately 1 cm in length in preparation for the compounding stage. Agricultural wastes were run through a Wiley mill with a 30-mesh screen. The fibers, wastes, polypropylene, and MAPP were compounded in a 1-liter, high-intensity shear, thermokinetic mixer (Synergistics Industries Ltd., Canada). No external heat sources are required due to the high shearing/smearing of the polypropylene which produces friction and generates heat, which then causes softening and flow of the composite system. A thermally controlled monitor regulated the dump temperature at 199°C (390°F). The composites were...
compounded at 5000 rpm (tip speed 32.9 m/sec.) and 150-g batches were standard. Directly after reaching the dump temperature, the material was pressed flat to enhance cooling and prevent fibers in the core of the composite from burning.

The composite blends were then granulated and dried at 105°C for 4 hours to drive off residual fiber moisture in preparation for injection molding. A 33-ton Cincinnati Milacron injection molder was used to produce standard ASTM tensile, flexural, and impact specimens. Samples were placed in a controlled humidity room for 3 days prior to testing to assure complete thermal stability of the test samples. Test conditions were performed according to the following ASTM standards: tensile testing (ASTM D 638), flexural testing (ASTM D 790), impact testing (ASTM D 256).

Results and discussion

The types of agricultural fibers used in this study are bast fibers and leaf fibers. Bast fibers are next to the outer bark in the bast or phloem and serve to strengthen the stems of the reed-like plants. They are strands running the length of the stem or between joints. The long leaf fiber contributes strength to the leaves of certain nonwoody, monocotyledonous plants. They extend longitudinally the full length of the leaf and are buried in tissues of a parenchymatous nature. The fibers found nearest the leaf surface are usually the strongest (2). Kenaf and jute are bast fibers; henequen, abaca, and sisal are leaf fibers.

Figure 1 shows a stress-strain curve for 100 percent polypropylene and an uncoupled 50 percent kenaf/50 percent polypropylene composite (7). Included in the figure is a series of stress-strain curves for coupled kenaf composites at various fiber loadings (20%, 30%, 40%, 50%, and 60% by weight) and 2 percent MAPP. For the kenaf fiber composites, a small addition of MAPP improved the tensile and flexural strength, tensile energy adsorption, failure strain, and unnotched Izod impact strength. Comparing the coupled 50 percent kenaf composite to the uncoupled 50 percent kenaf composite, the stress at failure is twice the value of the uncoupled system, which is indicative of the properties mentioned above. This paper will discuss the results of the composites made with 50 percent fiber (e.g., abaca, henequen, etc.), 47 percent polypropylene, and 3 percent MAPP. A future paper will detail and substantiate the fact that the mechanical properties of the various fiber types respond in a similar manner as kenaf fibers when coupled and uncoupled with MAPP.

Tensile strength and modulus

The mechanical properties of 40 percent fiber-glass/polypropylene and 40 percent talc/polypropylene composites (data obtained from Modern Plastic Encyclopedia '93) are shown in Figures 2 to 7 along with the mechanical properties of the five agricultural fiber composite systems. Figures 2 and 3 show the tensile strengths and tensile moduli, respectively. The bast fibers (kenaf and jute) have tensile strengths and moduli of approximately 70

![Figure 1](image1.png)  
**FIGURE 1.-** Stress-strain curves for kenaf composites. c = coupled, u = uncoupled.

![Figure 2](image2.png)  
**FIGURE 2.-** Tensile strength (MPa) properties.
percent and 85 percent, respectively, of the 40 percent fiberglass/polypropylene composites. When compared to the 40 percent talc properties, the bast fibers have twice the tensile strength and modulus. The leaf fibers (abaca, henequen, and sisal) have both strength and modulus of approximately 60 percent of the 40 percent fiberglass-reinforced polypropylene. In relation to 40 percent talc, the leaf fibers have tensile strengths of 175 percent, while the moduli range from 140 percent for sisal fibers to 175 percent for henequen fiber composites.

Figures 8 and 9 show the tensile strength and tensile modulus properties of the agricultural waste/polypropylene-based composites. For purposes of comparison, 100 percent polypropylene, 40 percent talc, and 50 percent wood flour composites are included because they are commercially available products. The figures show that the mechanical properties of kenaf core, oat straw, wheat straw, and oat hulls compare favorably to both wood flour- and talc-filled polypropylene composites. All other fillers have mechanical properties which are slightly less than the 50 percent wood flour, but still compare favorably to 40 percent talc-filled polypropylene. The 50 percent uncoupled corn cob is used for comparison to the 50 percent corn cob. All of the filler systems will react in a similar fashion when no MAPP is used during the compounding stage.

The addition of MAPP has the most dramatic effect on the tensile strengths of agricultural fiber and waste composites. The uncoupled fiber systems have strengths approximately half that of coupled systems. MAPP migrates to the interface between the nonpolar polypropylene and polar fiber surfaces. In addition, the maleic anhydride present in MAPP can covalently link to the hydroxyl groups on the lignocellulosic fibers (4). Under a tensile load, the improved adhesion at the fiber/matrix interface results in a more efficient stress transfer from the matrix to the reinforcing fibers. As a result, strength properties of agricultural fiber composites can be improved with small additions of MAPP.

**Flexural strength and modulus**

Figure 4 shows the flexural strengths of the agricultural fiber composites. With the addition of 3 percent MAPP modifier, the flexural strengths of both the bast and leaf fiber composites increase over an uncoupled fiber system (1). Figure 4 shows only the flexural strengths of the 50 percent fiber/47 percent polypropylene/3 percent MAPP in comparison to the fiberglass- and talc-filled polypropylene. The jute and kenaf fiber composites have 65 percent of the flexural strength of 40 percent fiberglass/polypropylene, while the leaf fibers have approximately 55 percent of the flexural strength. Figure 5 shows the flexural moduli for the bast and leaf fiber com-
posites. The moduli for both jute and kenaf fiber/polypropylene have values 106 percent superior to 40 percent fiberglass-filled polypropylene. In comparing the flexural moduli to 40 percent talc/polypropylene, the bast fibers have values 170 percent greater. The leaf fiber composites compare favorably to fiberglass/polypropylene with values ranging from 74 percent for sisal and 87 percent for henequen fibers. Henequen, abaca, and sisal composites all have flexural moduli superior to 40 percent talc/polypropylene. Values range from 118 percent for sisal and 140 percent for a 50 percent henequen/47 percent polypropylene/3 percent MAPP composite.

Figures 10 and 11 show the flexural strengths and flexural moduli for the agricultural waste polypropylene-based composites. The addition of MAPP increases the flexural strengths of these agricultural waste composites by approximately twice the value of uncoupled systems. Increased adhesion between the lignocellulosic fibers and the matrix provides for increased stress transfer from the matrix to the fibers. This results in an increased stress at failure and the higher values for flexural strengths in the coupled systems versus uncoupled systems. Values for the 50 percent uncoupled corncob and 50 percent corncob in Figure 10 gives an indication of how the mechanical properties of the other composite systems would be if no MAPP were present in the composites. The flexural strengths of composite systems composed of kenaf core, oat straw, wheat straw, and oat hulls are equivalent or superior to both wood flour- and talc-filled polypropylene composites.

FIGURE 5.- Flexural modulus (CPa) properties.

FIGURE 6.- Notched Izod impact (J/M) properties.

FIGURE 7.- Unnotched Izod impact (J/M) properties.
Other systems, such as corncobs, hard corncobs, and rice hulls have flexural strengths slightly less than wood flour- and talc-filled composites. In terms of the flexural modulus, Figure 11 shows that wood flour has the highest flexural modulus. Kenaf core, oat straw, and wheat straw composites have values between wood flour and talc composites, while all other agricultural waste/polypropylene composites have flexural moduli less than talc and wood flour.

**Notched and unnotched Izod impact**

Figure 6 shows the notched Izod for the agricultural fiber composites. The impact values for both kenaf and jute composites are below the impact strengths of 40 percent fiberglass and 40 percent talc composites. Impact properties versus 40 percent fiberglass/polypropylene are approximately 38 percent, while having 50 to 55 percent of the toughness of talc composites. Fiber attrition rates are high.
during the compounding stage, which reduces the fibers below the critical aspect ratio of about 0.4 mm. As a result, the contributions from fiber pullout and crack deflection mechanisms are limited, which reduces the impact toughness. The leaf fibers have impact values of approximately 50 percent of 40 percent fiberglass/polypropylene and 70 percent of the 40 percent talc/polypropylene. Fiber attrition of the leaf fibers is less during the high-intensity shear mixing, which helps maintain fiber lengths. The contributions from fiber pullout and crack deflection mechanisms help increase the impact values of the leaf fiber composites versus the bast fiber composites.

Figure 7 shows the unnotched impact properties for the five agricultural fiber composites. Unnotched impact values were not available for the 40 percent fiberglass and 40 percent talc composites in the Modern Plastics Encyclopedia. With the addition of

![FIGURE 10.- Flexural strength (MPa) properties.](image1)

![FIGURE 11.- Flexural modulus (GPa) properties.](image2)
MAPP, the values for unnotched impact toughness increased 40 to 45 percent over uncoupled systems. The 50 percent fiber/47 percent polypropylene/3 percent MAPP results range in values from 165 J/M for henequen and 206 J/M for jute fiber composites.

Figures 12 and 13 show the notched and unnotched Izod impact properties for the agricultural waste composites. The commercially available 40 percent talc/polypropylene has a notched impact toughness of 26.7 J/M, while 100 percent polypropylene has a value of 24 J/M. The mechanism for the toughness is due to the plate-like particles of talc which have a slightly higher aspect ratio than the finely ground agricultural wastes. Overall, the notched impact toughness of the agricultural wastes compare favorably to the wood flour- and talc-filled polypropylene and 100 percent polypropylene.

Figure 13 shows the unnotched Izod impact toughness of 40 percent talc is superior to all of the agricultural waste composites with a value 2.5 times...
the best agricultural waste composite. In comparison to wood flour, all of the agricultural waste composites have similar values of unnotched Izod impact toughness. Future work will involve the use of impact copolymers to improve the toughness of these agricultural fiber and waste composites.

Conclusions

The bast fiber composites tend to have higher strengths and moduli over the leaf fiber composites evaluated. Notched impact values show the leaf fiber composites to be tougher than the bast fiber composites.

The tensile strength, flexural strength and notched impact toughness values of agricultural fiber and waste composites are shown to improve substantially with the addition of MAPP.

Five agricultural fiber composites have been successfully compatibilized with a polypropylene homopolymer. Results indicate the mechanical properties are between 40 percent fiberglass/polypropylene and 40 percent talc/polypropylene composites.

Twelve different agricultural waste composites have been successfully compatibilized with a polypropylene homopolymer. Results indicate that kenaf core, oat straw, wheat straw, and oat hulls have equivalent mechanical properties to commercially available wood flour- and talc-filled polypropylene composites.

Continued research and development of agricultural fiber and waste composites will show that these material systems are viable alternatives to commercially available wood- and talc-filled polypropylene systems.

Literature cited

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WOODFIBER-PLASTIC COMPOSITES

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