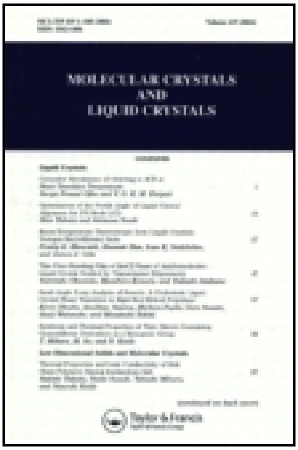

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Weathering Performance of Plant-Fiber/ Thermoplastic Composites

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Natural fiber/thermoplastic composites were made using aspen fiber and polypropylene. Several different levels of aspen fiber (30 to 60% by weight) were used with and without the addition of a compatibilizer (maleic anhydride grafted polypropylene, MAPP). These composites were tested for strength properties, exposed to boiling and cyclic liquid water and oven drying tests, and evaluated in an accelerated weathering test for 2000 hours. As the level of fiber increases without MAPP, flexural strength, flexural modulus, tensile strength and tensile modulus increases but notched and unnotched impact strength decrease as compared to pure polypropylene. The presence of MAPP has a very large positive effect on flexural strength, flexural modulus, tensile strength and tensile modulus as compared to composites made without MAPP. As the level of fiber increased in the composites, the rate and extent of thickness swelling and moisture sorption increased as compared to pure polypropylene. The presence of MAPP decreased the extent of swelling and moisture pickup. Weathering of composites under accelerated weathering conditions (water sprayed for 18 minutes followed by 102 minutes without water spray of UV radiation produced by a 6500-watt xenon arc light source) was restricted to the outer 0.5 mm of the surface. Weathering gave rise to the development of a white chalky surface layer which started at about 150–200 hours of weathering. After 2000 hours of accelerated weathering, weight loss increased with increasing fiber content and there was more weight loss in specimens containing MAPP as compared to specimens without the addition of MAPP. After scraping off the chalky white layer after 2000 hours of weathering, weight loss was greatest in specimens containing no fiber.

Keywords: Natural fiber; polypropylene; composites; strength properties; moisture sorption; ultraviolet degradation; weathering

INTRODUCTION

The use of lignocellulosics as fillers and reinforcements in thermoplastics has been gaining acceptance in commodity plastics applications in the past few years (Rowell and Sanadi 1998). It is interesting to point out that the use of the lignocellulosics in commodity thermoplastics to reduce cost and/or to improve mechanical performance is not new, and there are plenty of published papers, including patents dating back to the 1960's and 1970's. The resurgence of interest in the 1990's is probably due to increasing plastic costs and environmental aspects of using renewable materials.

The lignocellulosics that can be used in thermoplastics can be obtained from several sources, both from forestry and agricultural resources. The physical form can vary from fine wood flour (particulate) to kenaf, flax, sisal and other agro-based fibers. The cost and performance of the final composite products dictates the type of lignocellulosic to be used in the plastics. Literature also cites the use of typical plastics such as polyethylene, polypropylene, polyvinyl chloride and also some elastomeric co-polymers for blending with the lignocellulosics.

Blending of the plastics with the lignocellulosics may require compatibilization to improve the dispersion, flow and mechanical properties of the composite and has been reported as far back as the late 1960's. Extrusion of lignocellulosics-filled plastics for the automotive industry is well known and has been used for more than twenty years. Typical blending involves the plastic-filler/reinforcement to be shear mixed at temperatures above the softening point of the plastics. The heated mixture is then typically extruded into "small rods", that are then cut into short lengths to produce a conventional pellet. The pellets can then be used in typical injection or compression molding techniques. To reduce the cost of this blending process, direct injection molding of wood bark fibers in plastics have also been reported in the 1960's. The direct injection molding process probably has limitations on the amount of filler/fiber that can be used in the composite, and is also likely to be limited to particulate or shorter fiber. The chemical characteristics of the surface and bulk of the lignocellulosics are also important in the blending with plastics. The ability of the matrix of the lignocellulosic (hemicellulose and lignin) to soften in the presence of moisture at plastic processing temperature may give these renewable materials unique characteristics to develop novel processing techniques.

There is still a great deal of basic research going on in many parts of the world in the area of natural fiber/thermoplastic composite materials. This research includes the use of many different types of agro-based fibers, expanding the amount of agro-based fibers in the composites, developing better compatibilization systems, developing testing procedures, and understanding performance through modeling. Equipment development is going forward to produce more efficient mixing systems, faster through put, and new methods of molding/injection.

One of the research areas of interest in the fiber/plastics composite area is performance of these materials in outdoor applications (Rowell et al 1998). Issues of weathering, moisture up take, creep, and decay resistance are important properties to study to determine the change in properties as a function of environmental stresses.

The purpose of this paper is to describe recent results on the physical property and water sorption and weathering of aspen fiber/polypropylene composites.

EXPERIMENTAL

Materials

Four sets of specimens were prepared as follows: aspen fiber was combined with

Aspen Fiber	Polypropylene	MAPP
<----------------------	% by weight----------------------	-------->
30	70	0
30	68	2
40	60	0
40	58	2
50	50	0
50	48	2
60	40	0
60	38	2

polypropylene (PP) (by weight) alone or with 2% maleic anhydride grafted polypropylene (MAPP) (by weight). The PP homopolymer had a melt flow index of 12 g/10 min. as measured by ASTM D-1238. The MAPP had a number average molecular weight of 20,000, a weight average molecular weight of 40,000 and about had 6% by weight of maleic anhydride in the polymer.

The aspen fiber, MAPP and PP (the latter two in pellet form) were compounded in a high intensity thermo-kinetic mixer where the only source of heat is generated through the kinetic energy of rotating blades. The blending was accomplished at 4600 rpm and then automatically discharged at 190 C. The total residence time of the blending operation averaged about 2 min.

The mixed blends were then granulated and dried at 105 C for 4 hours. Test specimen were injection molded at 190 C using pressures varying from 2.75 MPa to 8.3 MPa depending on the constituents of the blend.

Physical Properties

Test 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 12.5 mm/min. Six specimens of each type were tested and the results averaged.

Liquid Water Tests

Rate of water sorption. Test specimens were oven dried over night at 105 C, weighed, and thickness determined.Specimens were placed in a 10 x 10 x 5-cm container and water was added specimen thickness recorded as a function of time. Measurements were taken every 15 minutes for the first hour, every hour for the first eight hours, then once a day for 5 days. After 5 days, each specimen was re-oven dried for 24 hours at 105 C and weights and thickness were determined. Three specimens of each type were tested and the results averaged.

Continuous Soaking. Test specimens were oven dried over night at 105 C, weighed, and thickness determined. Specimens were placed in container and water was added. Specimens were soaked in water for seven days and wet weight and thickness were determined. The specimens were then re-oven dried for 6 hours at 105 C and weight and thickness were determined. The specimens were then returned to water soaking and the process was repeated for 10 weeks. Three specimens of each type were tested and the results averaged.

Boil Test. Test specimens were oven dried over night at 105 C, weighed, and thickness determined. The specimens were boiled for 2 hours and immediately weight and thickness determined. Specimens were re-oven dried for 24 hours at 105 C and again measured. The process was repeated for 10 times for a total of 20 hours of boiling. Three specimens of each type were tested and the results averaged.

Weathering

Test specimens were oven dried over night at 105 C, weighed, and thickness determined. The faces of five specimens of each type were exposed to a 6500-watt xenon arc light source (which closely approximates natural sunlight spectrum in the visible and ultraviolet (UV) regions) in an enclosed chamber at 40 to 43 C. Half of each specimen was covered with aluminum foil so only half of each specimen was exposed to UV light. Exposure to UV radiation alternated with a spray of distilled water for 18 minutes followed by 102 minutes of UV light without water spray. Exposure time is expressed as hours of exposure to light.

Photographs were taken after 0, 200, 400, 600, 800, 1000, 1500, and 2000 hours. At the end of 2000 hours, the specimens were re-oven dried and their thickness and weight determined. The white chalky surface was removed from the weathered side of each specimen and thickness measured again to determine the weight loss and depth of surface degradation. Five specimens of each type were tested and the results averaged.

RESULTS AND DISCUSSION

Physical Properties

The mechanical properties of the aspen fiber-polypropylene composites with and without compatibilizer are shown in Table 1. Values for all mechanical properties increased as the level of fiber increased. Values for flexural strength, flexural modulus, tensile strength, tensile modulus and unnotched Izod toughness are much higher for the composites with 2% MAPP compatibilizer as compared with composites with no compatibilizer as has been shown before [Sanadi et al. 1994, 1995, 1997]. Notched Izod toughness is about the same for both sets of composites.

TABLE 1 - Mechanical properties of aspen fiber-polypropylene composites

Specimen	Flexural strength (MPa)	Flexural modulus (GPa)	Tensile strength (MPa)	Tensile modulus (GPa)	Izod notched (J/m)	Izod unnotched (J/m)
PP	27.9	1.38	26.2	1.69	22.4	713.5
PP/MAPP	34.6	1.79	29.3	1.82	18.6	563.3
30A/70PP	49.5	4.12	29.3	4.52	24.8	101.7
30A/68PP/MAPP	60.2	3.82	44.9	4.10	21.1	128.3
40A/60PP	54.6	4.60	34.9	5.22	19.6	85.5
40A/58PP/MAPP	66.4	4.66	47.7	5.14	19.8	108.7
50A/50PP	50.2	5.48	28.4	5.81	26.4	67.1
50A/48PP/MAPP	75.7	5.88	53.1	6.68	21.9	98.5
60A/40PP	45.9	6.09	25.6	6.95	23.9	55.2
60A/38PP/MAPP	75.8	6.73	48.1	7.19	21.3	81.1

Liquid Water Tests

The rate of change in weight due to water sorption is shown in Figure 1. The rate of

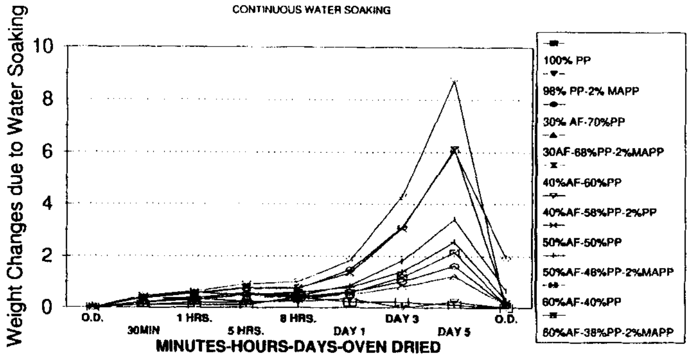

FIGURE 1 - Changes in weight due to water soaking of aspen fiber/polypropylene composites

water picked up by the composites increases as the level of fiber increases. The amount of water pick up at a given level of fiber is decreased in specimens containing MAPP. The rate of thickness swelling due to water sorption is shown in Figure 2. While the thickness change is small (less than 5%) even in the composites containing 60% aspen fiber, increases in thickness does occur in composites containing aspen fiber. The specimens that contain no fiber actually became thinner in the water tests which was also observed in other tests.

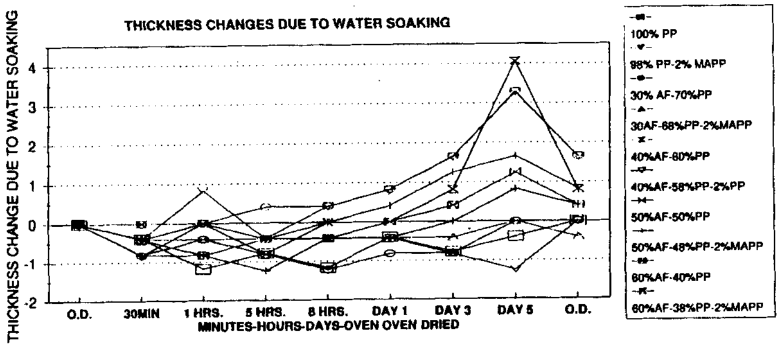

FIGURE 2 - Changes in thickness due to water soaking of aspen fiber/polypropylene composites

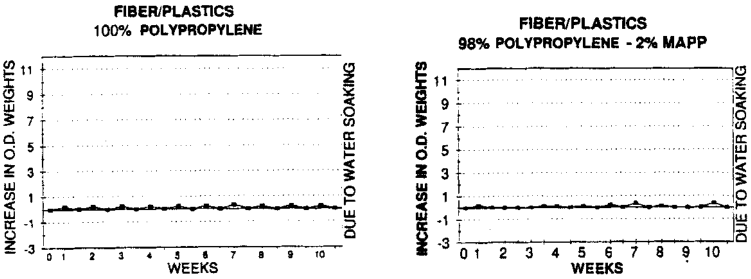

FIGURE 3 - Changes in weight due to cyclic wetting and oven drying of aspen fiber/polypropylene composites (Left - PP, Right PP + 2% MAPP)

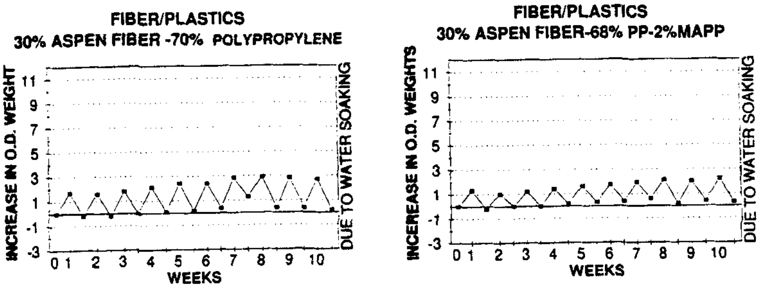

FIGURE 4 - Changes in weight due to cyclic wetting and oven drying of aspen fiber/polypropylene composites (Left - 30% aspen fiber/70% PP, Right - 30% aspen fiber/68% PP, 2% MAPP)

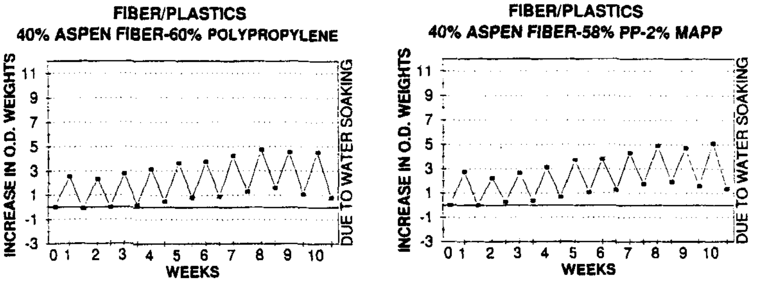

FIGURE 5 - Changes in weight due to cyclic wetting and oven drying of aspen fiber/polypropylene composites (Left - 40% aspen fiber/60% PP, Right - 40% aspen fiber/58% PP, 2% MAPP)

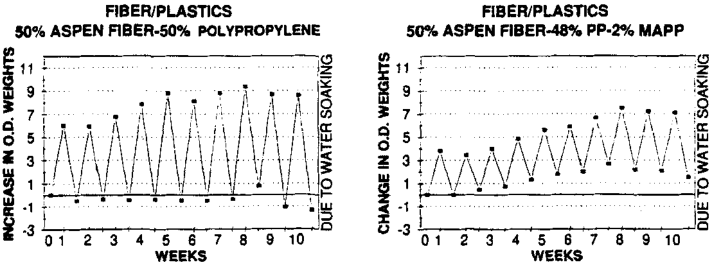

FIGURE 6 - Changes in weight due to cyclic wetting and oven drying of aspen fiber/polypropylene composites (Left - 50% aspen fiber/50% PP, Right - 50% aspen fiber/48% PP, 2% MAPP)

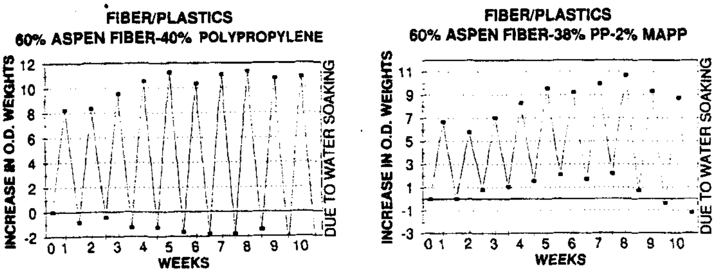

FIGURE 7 - Changes in weight due to cyclic wetting and oven drying of aspen fiber/polypropylene composites (Left - 60% aspen fiber/40% PP, Right - 60% aspen fiber/38% PP, 2% MAPP)

Figures 3-7 show the changes in weight due to cycling wetting and oven drying of polypropylene alone and composites containing different levels of aspen fiber. It can be seen that as the level of fiber increases, so does the extent of weight gain and loss with wetting and drying cycles. Data showing thickness changes due to cyclic wetting and over drying show similar results.

Figure 8 shows the results of the boiling test. Most of the weight gain that results from boiling takes place in the first boiling cycle and little change occurs in following boiling cycles.

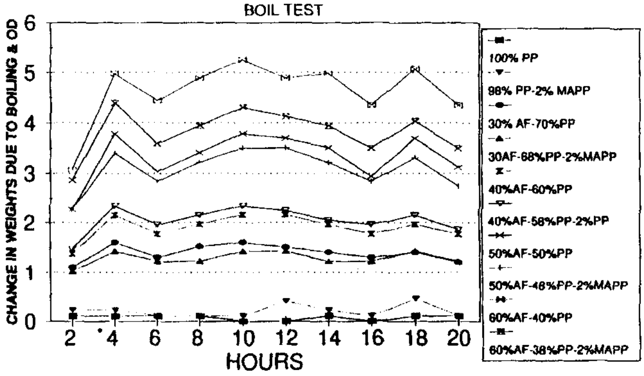

FIGURE 8 - Changes in weight due to boiling of aspen fiber/polypropylene composites

Weathering

It is well known that polypropylene is not stable to UV radiation. With a large number of tertiary carbon sites, polypropylene is very sensitive to free radical formation and decomposition by chain scission resulting in the formation of lower molecular weight polymers on the surface. Agro-based fibers also undergo UV degradation through free radical reactions with the decomposition of the lignin polymer in the cell wall. Swelling of the composites due to the presence of the natural fiber will result in an increase in surface area allowing more material to be exposed to UV degradation.

TABLE 2 - Weight loss and erosion of aspen fiber/polypropylene composites due to weathering

Specimen	Time to Yellowing (Hrs)	Before Scraping Weight Loss	Before Scraping Thickness Loss	After Scraping Weight Loss	After Scraping Thickness Loss
		<-------------	------ % ------	------	------------->
PP	>200	1.3	0.4	15.2	11.2
PP/MAPP	>200	0.9	1.0	14.0	9.9
30A/70PP	>200	2.7	0.6	5.7	4.1
30A/68PP/MAP	>200	2.7	0.9	7.2	5.0
40A/60PP	>200	2.6	1.5	5.0	3.3
40A/58PP/MAP	>200	2.8	1.6	5.6	4.1
50A/50PP	>200	3.2	1.3	6.0	3.3
50A/48PP/MAP	>200	3.3	1.6	6.3	4.2
60A/40PP	>200	3.7	0.7	5.7	1.6
60A/38PP/MAP	>200	3.7	1.9	5.6	3.2

Table 2 shows the time when the dark brown surface of the aspen fiber/polypropylene composites have undergone a noticeable color change due to UV radiation. In the composites with no fiber, the color change is from opaque white to yellow and this occurs at about 200 hours of UV exposure. In the composites containing aspen fiber, the color change is from brown through bleaching to white that occurs at about 150-200 hours of UV exposure. The surface of all composites first show a slight surface roughness and then the surfaces start to turn white. By 1500 hours, the surfaces are rough and a white powder can be removed from the surfaces with mild abrasion. At 2000 hours, the surfaces are very rough and chalky.

Table 2 also shows that weight loss due to weathering increases as the content of aspen fiber increases. More weight and thickness loss occurs in specimens containing MAPP at equal levels of fiber content compared to specimens without MAPP. After scraping off the white surface layer, Table 2 shows that the greatest weight and thickness loss occurs in the specimens without fiber. The specimens containing fiber were scraped back to a solid surface but not back to the dark brown color of the original specimens. After 2000 hours of UV exposure and measuring the thickness of the specimens after scraping the white degraded material from the surface down to original color shows that the degradation has occurred in the top 0.4 mm.

CONCLUSIONS

Adding agro-based fibers to polypropylene composites provides a strategy to produce composites that are lower in cost, less abrasive on processing equipment than mineral fillers, and improves strength properties as compared to composites containing no fiber. The addition of maleic anhydride grafted polypropylene to these composites improves strength properties even more.

There is interest in using natural fibers and polypropylene for composites for non-structural outdoor applications. This type of applications, however, raises questions about their performance in adverse conditions. Long term exposure to micro organisms, moisture, ultraviolet radiation, heat, load, and abrasion may limit their use for some exterior applications. This paper presents data on strength properties, moisture sorption, and weathering of aspen fiber/polypropylene composites using different levels of aspen fiber with and without the addition of a compatibilizer (maleic anhydride grafted polypropylene, MAPP).

As the level of fiber increases without MAPP, flexural strength, flexural modulus, tensile strength and tensile modulus increases but notched and unnotched impact strength decrease as compared to pure polypropylene. The presence of MAPP has a very large positive effect on flexural strength, flexural modulus, tensile strength and tensile modulus as compared to composites made without MAPP.

Composites submerged in water or boiled for long periods of time resulted in a continual increase in specimen weight and thickness. Under cyclic water soaking conditions, the composites showed a small but steady increase in moisture sorption with each wetting cycle. As the agro-fiber content increases in these composites, more fiber to fiber wicking is possible so moisture gain continues with time.

Weathering of these composites using a water/ultraviolet radiation weathometer, degrade is limited to the outer 0.5 mm of the surface which results in a loss in surface smoothness and the development of a white chalky surface layer.

Future studies will concentrate on various additives that can be used in this technology to increase strength properties and reduce environmental degradation of natural fiber/polypropylene composites.

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