Water Uptake and Mechanical Characteristics of Natural Filler–Polypropylene Composites

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ABSTRACT: Water uptake characteristics and some mechanical properties of polypropylene composites containing three types of natural fillers, purified α-cellulose, wastepaper fibers, and wood flour were studied. The fiber contents were 15, 25, and 35% by weight. Two percent maleic anhydride polypropylene (MAPP) was also added to the mix, as the compatibilizer agent. Mixing process was performed in a Brabender Plasticorder until a constant torque was reached. Composites made out of these combinations were then pressed in a laboratory press and ASTM standard test specimens were cut out of the sheets. Water absorption and tensile tests were performed on these specimens. The results showed a significant difference between the various filler types regarding water uptake. Water uptake also increased by the increase in filler content. Tensile strength and elongation at break in composites declined when compared with pure polypropylene, but their modulus of elasticity increased. Among the three types of fillers, no significant discrepancies were observed in terms of improving mechanical properties in composites. Filler content increase had no drastic effect regarding strength improvement. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 88: 941–946, 2003

Key words: polypropylene; composite; mechanical properties; water uptake; natural fillers

INTRODUCTION

Studies on natural fiber/polymer composites are being conducted in many laboratories to expand the use of polymer materials and waste fibers as well.[1–6] The composites so produced show some priorities over conventional composites such as lower density, less capital intensive when produced in commercial scale, and being environmentally safe for their uses.[2,7] Agrowastes and agro-forest materials—for example, sawdust, wood fibers, sisal, and bagasse—are slowly penetrating the market of reinforced plastics and filled polymers previously dominated by glass fiber and other mineral reinforcements. Composites of natural fibers and thermoplastics are finding applications in many industries, such as the automotive industry.[3] Moreover, both components of such composites can be obtained from waste materials, which are very abundant, especially in developing countries.[2] Hygroscopicity and incompatibility of the hydrophilic fiber and hydrophobic polymer—and hence poor interaction between the phases—are considered disadvantages.[2] Because of the hygroscopic nature of natural fibers, water uptake of composites containing these fibers as fillers and/or reinforcers can be a limiting parameter as far as the final application of the composite is concerned. Water absorption is one of the important characteristics of natural fiber polymer composites that determine their end use applications. Water absorption could lead to a decrease in some of the properties and needs to 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 buildup of moisture in the fiber cell wall and also in the fiber–matrix interphase region. Moisture buildup in the cell wall could result in fiber swelling and concerns regarding dimensional stability of the product. If necessary, the moisture absorbed in the fiber cell wall can be reduced through the acetylation of some of the hydroxyl groups present in the fiber.[8] However, this requires additional costs. Good wetting of the fiber by the matrix and adequate fiber–matrix bonding can decrease the rate and amount of water absorbed in the interphase region of the composite.

Rowell et al. (1997) reported a 1.05% by weight water absorption in a kenaf fiber/polypropylene composite containing 50% by weight kenaf fibers after 24 h soaking in water, which was considerably higher than any mineral filled systems.[9] Rana et al. (1998) studied the water absorption of short jute fiber reinforced polypropylene composites.[10] Water absorption values after 24 h cold soaking and 2 h boiling of the samples were reported. Water absorption values were found to

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increase with increase in fiber loading. Use of compatibilizer (maleic anhydride polypropylene, or MAPP), however, decreased water absorption at the same fiber loading. This was attributed to the fact that some of the hydrophilic —OH groups might have reacted with acid anhydride to form ester linkages and thereby the number of hydroxil groups declined.[10] Simonson et al. (1998) studied the water absorption of styrene maleic anhydride copolymer composites containing old newsprint fibers and aspen and pine wood flours at different fiber loadings. Results indicated an increase in water absorption due to the filler content increase, but no significant difference was observed between the three filler types.[11] Yadav et al. (1999) reported the results of a study on the water uptake of newspaper-reinforced plastic composites at different levels of fiber content up to 65% by weight. Results were in agreement with other publications indicating an increase in water absorption as a result of the increase in fiber content.[12]

In this experiment, water uptake characteristics of composites made from three different types of cellulosic fibers including pure α-cellulose, wastepaper, and wood flour at various fiber loadings and polypropylene are studied. Some of the mechanical properties of the composites are also presented.

EXPERIMENTAL

Materials

Materials for this experiment were injection molding grade polypropylene, V30S, supplied by Arak Petrochemistry Plant (Iran) with a melt flow index of 16 g/10 min, ether grade cotton linter containing 98% α-cellulose supplied by Linterpak Manufacturing (Iran), white scrap paper from trimming process at a local publishing workshop, and softwood flour obtained through grinding dry wood shavings in the laboratory. MAPP was made by grafting maleic anhydride to polypropylene in the laboratory.

Methods

Cellulose fibers, scrap paper, and wood shavings were ground in a pilot scale pulverizer to meet designated size requirement. Particle size was fixed for all the fillers at 50 mesh. Prior to mixing, all the fillers were dried for 24 h at 60°C. Polypropylene due to its hydrophobic nature did not need such conditioning. Mixing polymer with different weight percentages (Table I) of fibers was conducted in a Brabender Plasticorder, at 190°C at 30 rpm. The mixing process was continued until a constant torque was reached. Initially, polypropylene was added and melted in the mixer; then fibers and the compatibilizer were introduced. In total, each mixing cycle took about 14 min depending on the filler type and content. The compounded substance made out of fiber/polymer was hot pressed (180°C) to fabricate sheets. The nominal thickness of these sheets was 2 mm. These were cut to prepare standard mechanical testing specimens.

TABLE I
Combinations of Fillers and Polymer in Experimental Mixtures

No.	Formulation	Polymer (%)	Filler (%)	MAPP (%)
1	PP	100	—	—
2	CF-15	83	15	2
3	CF-25	73	25	2
4	CF-35	63	35	2
5	WP-15	83	15	2
6	WP-25	73	25	2
7	WP-35	63	35	2
8	WF-15	83	15	2
9	WF-25	73	25	2
10	WF-35	63	35	2

PP: polypropylene, CF: alpha cellulose WP: wastepaper; WF: wood flour

Water absorption of the samples was measured according to ASTM test method D-570. The specimens were dried in an oven for 1 h at 110°C; then they were weighed and placed in a container of distilled water, and the equilibrium weight value was determined after 24 h soaking in water at room temperature. Results are presented as percent water absorption in relation with the dry weight of the specimens. Tensile tests were done to characterize the mechanical properties of the composites according to ASTM D-638. Modulus of elasticity, elongation at break, energy absorption, and tensile strength of the samples were determined. Test data were analyzed in completely randomized designs and Dunkan's multiple range test was used for grouping the calculated means. All comparisons were made at 95% confidence level.

RESULTS AND DISCUSSION

Water uptake

Analysis of variance of the values of the composites water absorption after 24 h indicates that both variables including filler type and filler content have significant effects at the 95% confidence level. The independent effect of filler type on water absorption is shown in Figure 1. As can be seen, the highest water absorption is observed for wastepaper (1.03%) followed by wood flour (0.83%) composites. Among various filler types, cellulose fibers caused the least water absorption in the composites; however, this value for pure polypropylene is negligible. Dunken's multiple range test showed a significant difference between the three filler types at the 95% confidence level. The

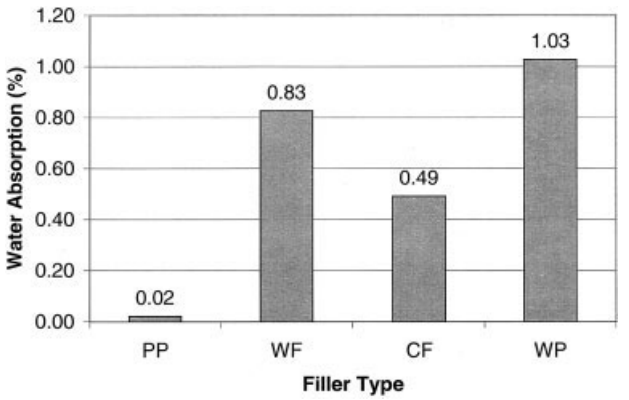

Figure 1 Independent effect of filler type on water absorption of the composites.

higher water absorption in wood flour composites compared to composites containing cellulose fibers is surprising for cellulose must be more hygroscopic than wood flour because in the latter there is some lignin that is substantially less hygroscopic than cellulose. However, the water absorption of pure polypropylene in quite low, and it can be assumed that in the case of cellulose fibers, there is a better interaction between the two phases leading to better coverage of the fibers by the polymer and eventually lower water absorption. This results are in disagreement with the results from Simonsen et al.(1998) in which no significant difference was observed among paper fibers and wood flour.[11]

Figure 2 shows the independent effect of filler content on the composites' water absorption. It is clearly seen that as the filler content increases, the amounts of water uptake also increase. Statistical tests performed indicate that there is a significant difference between the various filler contents regarding the water uptake. The difference between 15 and 25% filler content is significant, but the difference between these two percentages and 35% filler content is much more pronounced. This can be caused by the filler agglomerations that might have happened during mixing, which could possibly lead to extra voids in the texture and have also caused some surface roughness in the case of 35% filler content. Results are in agreement with previous works.[10,11]

Figure 3 shows the combined effects of different filler types and contents on the composites water uptake characteristics. At 15% filler content, composites containing wood flour show the highest water absorption and composites containing cellulose fibers the lowest. At 25% filler content, the highest amount of water absorption corresponds to the composites having wastepaper as the filler while composites containing cellulose fibers still form the bottom line. The difference between 15 and 25% wood flour is very marginal. At 35% filler content, the same trend as 25% filler content can be observed, while water uptake values are significantly higher. Generally, the difference between various filler types becomes more pronounced at higher filler contents. This can be explained by the fact that at lower filler contents the majority of the material is composed by the polymer, which is inherently hydrophobic, and less fiber ends are exposed at the surfaces. If we assume that the water uptake of the composite is almost entirely a result of filler's water uptake, some linearity in the water uptake trend must be observed as the ratio of the filler increases. This linear behavior is more or less observed in Figure 3, except for the 25% wood flour.

Tensile properties

Figure 4 shows the effect of different filler types and contents on the composites elongation at break. It is clearly seen that by adding only 15% filler of any type, there is a drastic drop in elongation at break, which is statistically significant. There is no significant difference between various cellulose fiber contents, and this holds for the two other filler types as well. Results indicate that although there is a sharp reduction in the values of elongation at break by introducing fillers to the system, there are no significant differences between low and high filler contents. This means that it would be possible to increase the filler content without further decreasing this value. In addition, there are no significant differences between the three filler types in this regard. The presence of fillers in the system also caused the composite systems not to have a yield point and break abruptly. This could be a disadvantage where the failure is preferred to be noticed before occurring.

Figure 5 shows the effect of different filler types and contents on the composites tensile strength. A general falling trend can be observed; however, this reduction is not significant for the cases of 15 and 25% cellulose

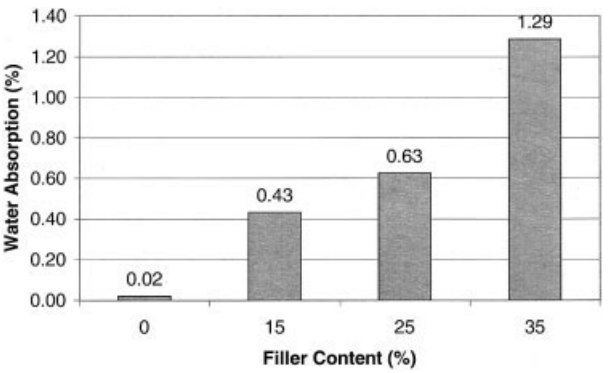

Figure 2 Independent effect of filler content on water absorption of the composites.

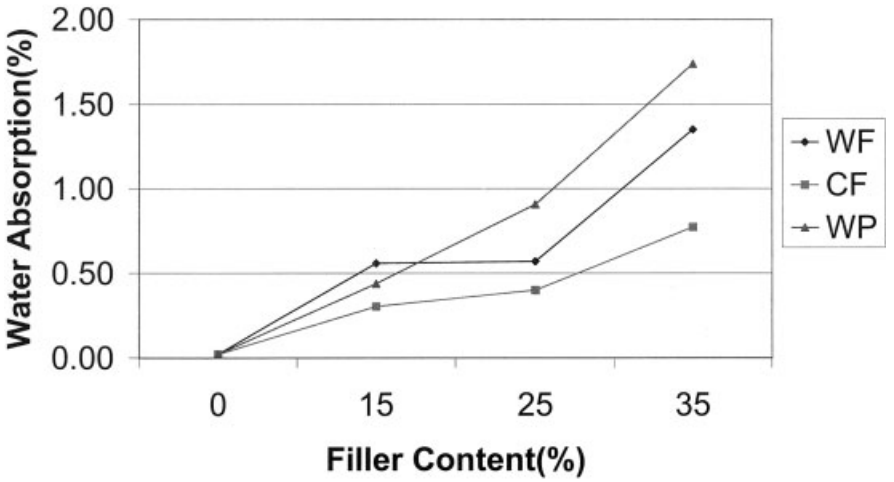

Figure 3 Effect of different filler types and contents on water absorption of the composites.

fiber. There is no significant difference between the filler content levels. Filler type does not have a significant effect at 15% filler content. However, at 25% filler content, wood flour is different from the other two. At 35% filler content, cellulose fibers show significant difference from the other fillers. It can be concluded that the reduction in tensile strength caused by cellulose fibers is significantly less than the two other fillers. MAPP was used in the systems to enhance the interface. It could be expected that in the absence of the compatiblizer the reduction in tensile strength would be more pronounced. Results also indicate that it is possible to increase the filler content without further reduction in tensile strength.

Effects of filler type and content on the composites modulus of elasticity are shown in Figure 6. From this figure it is clearly seen that there is significant increase in the moduli of the composites as compared to the pure polypropylene. In all formulations, the moduli increases as the filler contents increase. In the case of cellulose fibers, a significant difference is observed between 15 and 35% filler contents. However, no significant difference is observed between 25 and 35% filler contents. This is also true for wastepaper. In the case of wood flour, there is no significant difference between 15 and 25% filler content; however, 35% filler content is significantly different from the other two. At 15% filler content, wastepaper induces a significantly higher modulus compared to the other fillers. At this level, wood flour and cellulose fiber do not show a significant difference from the pure polypropylene. At 25% filler content, the difference between the filler types is not significant. However, the modulus value is higher for composites containing wastepaper. At 35% filler content there is no significant difference between the three filler types and these formulations show the highest moduli.

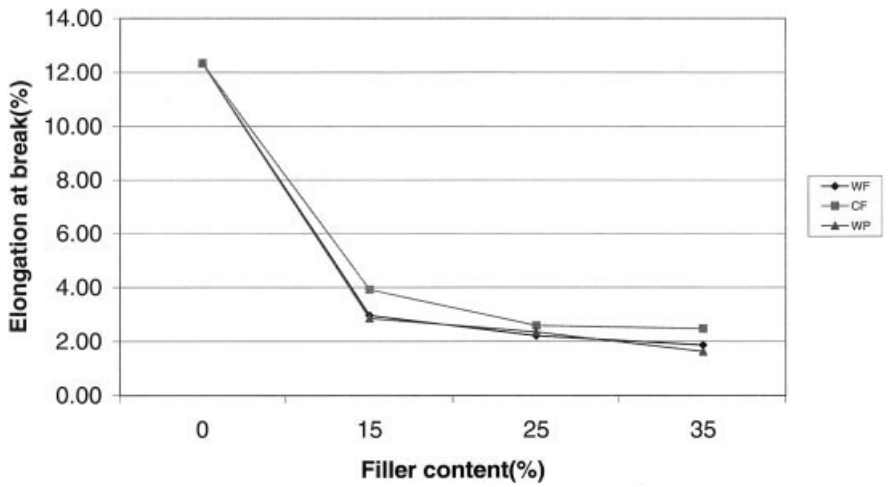

Figure 4 Effect of fillers on elongation at break of the composites.

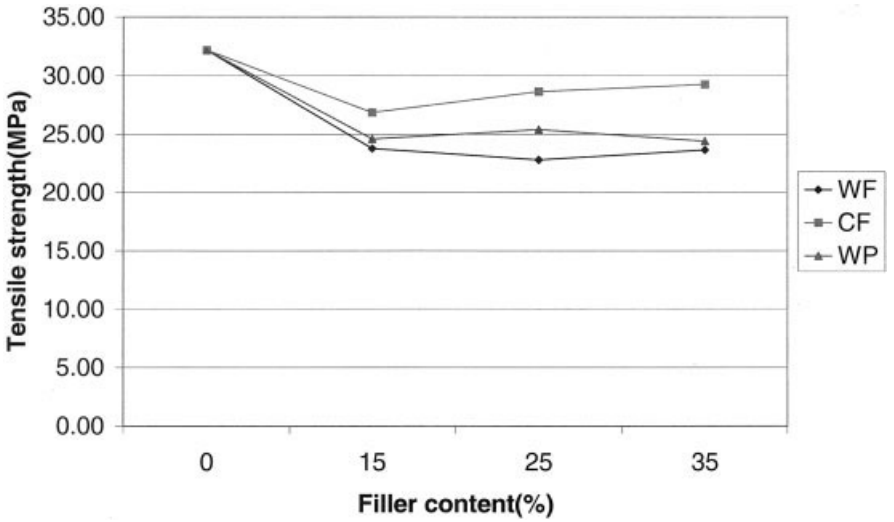

Figure 5 Effect of fillers on tensile strength of the composites.

Figure 7 indicates variation of work or energy at proportional limit per unit volume of composite as a function of filler content. The three types of filler impose a rapid reduction on energy at proportional limit of the composites. From 15 to 25% filler content, this character of composite does have decreasing rate, but somewhat slower as compared with that of 15% filler content. Within 25–35% filler content, work at proportional limit remains relatively constant at least with respect to WP and WF fillers, and the differences are not significant. But in the same range of filler content, energy at proportional limit has an upward tendency with CF filler, although again not significant. Considering the stiffness index (E value) of composite, reduction of energy absorption at proportional limit would be expected since it is calculated by the relation involved with values of E (elastic behavior), as $W_{pl} = \sigma^2/2E$. Increase in the fiber loading restricted the mobility of the polymer chains and this caused a decrease in the strain at proportional limit.[10] To improve this index, if intended, elastomer agents can be added to the composite.

CONCLUSION

The following conclusions may be drawn from the above studies:

1. Composites containing wastepaper as the filler showed the highest water absorption followed by those containing wood flour. The lower water absorption in cellulose fiber composites was assumed to be a result of better adhesion between the fibers and the polymer matrix as compared with the other fillers.

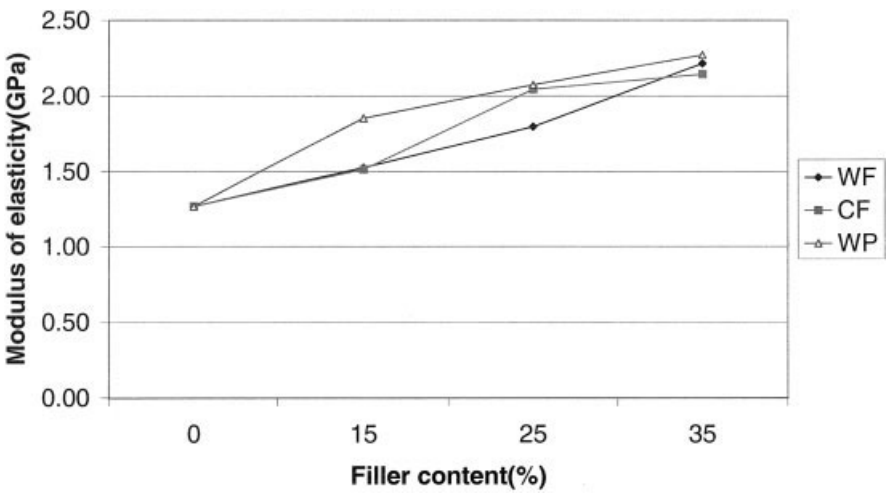

Figure 6 Effect of fillers on modulus of elasticity of the composites.

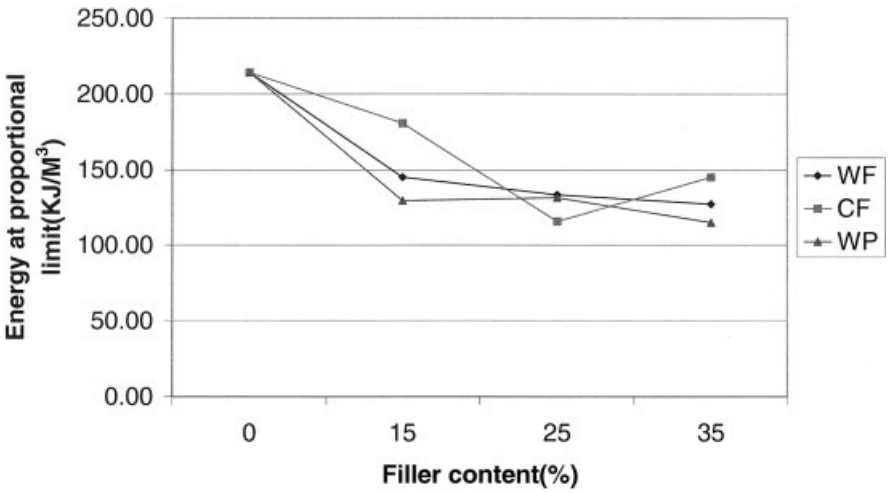

Figure 7 Effect of fillers on energy absorption of the composites.

2. A sharp increase in water absorption was observed by the increase in filler content from 15 to 35%. The water uptake value for 35% filler content was considerably higher than the other filler contents. This was attributed to the possible fiber agglomerations, which may have been developed at higher filler contents.
3. A very sharp decrease in the composites' elongation at break was observed when only 15% fiber was added. The effect of more filler addition on the decrease of this value was marginal.
4. Tensile strength first dropped slightly but generally increased by the addition of more fillers.
5. The increase in filler content considerably increased the modulus of elasticity of the composites. Composites containing wastepaper fibers had the highest modulus while there was not a significant difference between the fillers at 35% filler content.
6. Energy at proportional limit was reduced by the addition of fillers and more or less followed the same pattern as the elongation at break.

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