

# Effect of the Delignification of Wood Fibers on the Mechanical Properties of Wood Fiber–Polypropylene Composites

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**ABSTRACT:** The effect of the delignification of hornbeam fibers on the mechanical properties of wood fiber–polypropylene (PP) composites was studied. Original fibers and delignified fibers at three levels of delignification were mixed with PP at a weight ratio of 40:60 in an internal mixer. Maleic anhydride (0.5 wt %) as the coupling agent and dicumyl peroxide (0.1 wt %) as the initiator were applied. The produced composites were then hot-pressed, and specimens for physical and mechanical testing were prepared. The results of the properties of the composite materials indicate that delignified fibers showed

better performance in the enhancement of tensile strength and tensile modulus, whereas the hardness of the composites was unaffected by delignification. Delignified fibers also exhibited better water absorption resistance. Notched impact strength was higher for delignified fiber composites, but it was reduced at higher delignification levels. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 102: 4759–4763, 2006

**Key words:** composites; poly(propylene) (PP); mechanical properties

## INTRODUCTION

Ecological concerns have resulted in a renewed interest in natural materials, and therefore, issues such as recyclability and environmental safety are becoming increasingly important in the introduction of new materials. Natural fibers can be considered environmentally friendly alternatives for use as reinforcement in engineering polymeric materials. Good results have been obtained from the mixture of cellulosic fibers with polymeric materials. The results of these studies have shown that with the addition of natural fibers, the tensile strength, elongation at break, and impact strength of composites decrease, but the tensile modulus and hardness increase significantly.<sup>1–3</sup> The properties of composites also improve with coupling agents. Maleic anhydride (MA) and maleated polypropylenes (MAPPs) have been widely used as coupling agents for PP-based composites.<sup>4–6</sup> The use of elastomers can raise the impact strength of composites as well.<sup>7,8</sup>

In recent years, wood fiber–thermoplastic composites have received considerable attention from the wood and plastic industries. Natural fibers could turn up in every thing from cars to aerospace and sport goods.<sup>9</sup> Natural fibers in composites have some advantages.

They are abundant, renewable, cheap, recyclable, and biodegradable; they have low density, and the environmental pollution they cause is less than that of synthetic fibers. On the other hand, the use of natural fibers in composites results in some problems, including low allowable processing temperatures, poor moisture resistance, and poor bonding with the polymer matrix.<sup>4</sup> Among these disadvantages, bonding in a system containing hydrophilic cellulose and a hydrophobic matrix is the most problematic because the interfacial interaction between the cellulose filler and the matrix has a significant effect on the properties of the composite. Therefore, the improvement of wood fibers and the polymer matrix in composites is an important subject for discussion. On the basis of these improvements, favorable changes in the properties of composites can be reached. To achieve these improvements, the role of the composite components in the determination of the properties of the composites must be clearly known. Lignin is one of the major components of all natural fibers that has poor reinforcing effects in composites when used alone.<sup>10</sup> The removal of lignin from cellulosic fibers could, therefore, help to improve the mechanical properties of natural fiber–plastic composites in two ways: by increasing the stronger and stiffer cellulose content at a given fiber fraction and by improving the adhesion between the two phases of the composites through the elimination of the less chemically reactive lignin component. In this study, we investigated the effects of the delignification of wood

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**TABLE I**  
**Composition of the Studied Formulations (wt %)**

Formulation	Code	PP	Fibers	Residual lignin
1	DF-7	60	40	6.73
2	DF-26	60	40	25.88
3	DF-61	60	40	60.78
4	OF	60	40	100

fibers on the mechanical and physical properties of wood fiber–PP composites.

## EXPERIMENTAL

### Materials

The polymer matrix, PP, used in this study was supplied by Araak Petrochemical Industries (Araak, Iran) and had a melt flow index of 8 g/10 min and a density of 0.92 g/cm<sup>3</sup>.

Hornbeam fibers (*Carpinus betulus*; filler materials) were obtained from the Research Forest of the Natural Resources Faculty of the University of Tehran, Iran. These fibers had received no pretreatment and did not undergo extraction processes.

Four different types of wood fibers were prepared:

1. Original fibers with a 20.63% lignin content based on oven-dried wood weight.
2. Delignified fibers with 12.54% residual lignin based on oven-dried wood weight.
3. Delignified fibers with 5.34% residual lignin based on oven-dried wood weight.
4. Delignified fibers with 1.39% residual lignin based on oven-dried wood weight.

MA (the coupling agent) and dicumyl peroxide (the initiator) were supplied by Hercules Co. (Wilmington, DE).

Na<sub>2</sub>S and NaOH (the kraft-pulp-making liquor) were supplied by Merck Co. (Frankfurt, Germany).

### Preparation of the fibers

To prepare the fibers, several disks from hornbeam logs were obtained and then converted into chips. The chips were 20 mm in length and 4 mm thick on average.

The original fibers were obtained by the grinding of the aforementioned chips. Only particles that passed through a sieve of mesh 40 were used in this study.

The delignified fibers were obtained by a kraft pulping process at 170°C.<sup>11</sup>

Delignification times were 3 h, 1.5 h, and 15 min for the DF-7, DF-26, and DF-61 delignified fiber composites (DFs), respectively. The number in the fiber code denotes the amount of residual lignin in the fiber. Klason lignin was used as a criterion for the measurement of the residual lignin. Klason lignin and lignin dissolved in acid were measured according to ASTM D 1106-84 specification and TAPPI standard UM 250, respectively. The delignified fibers were then dried and ground and were passed through the same sieve used for the original fibers.

So the delignified fibers would be comparable, fiber length and aspect ratio analyses were performed.

The original and delignified fibers were dried in an oven at 65 ± 2°C for 24 h before they were blended with PP.

### Processing

PP, the original and delignified fibers, and the additives were blended in an internal mixer (SYS 90 model) at 180°C for 8 min at 60 rpm. After the addition of PP, the filler (original or delignified fibers separately) and additives were added. In all cases, the weight ratio of filler to polymer was 40:60 (Table I). The mixing process was discontinued when a constant torque was reached.

The amorphous compounds removed from the mixer were then compression-molded at 190°C under a pressure of 20 bar for 2 min to produce test specimens, which were cut out of the sheets prepared as described previously.

### Mechanical and physical properties

The tensile properties of the composites were measured with an MTS tensile testing instrument (model 10/M). The impact strength was measured on a Zwick impact testing machine (model D7900). Hardness properties were determined by a Shore D hardness tester.

**TABLE II**  
**Lignin Content of the Fibers**

Type of fibers	Residual lignin based on dry pulp weight (%)	Residual lignin based on oven-dried wood (%)	Residual lignin (%)
Original fibers	—	20.63	100
Delignified fibers (series 1)	16.31	12.54	61
Delignified fibers (series 2)	9.67	5.34	26
Delignified fibers (series 3)	2.98	1.39	7

**TABLE III**  
Delignified and Original Fiber Lengths in Different Treatments

Fiber type	Mean fiber length (mm)	Mean fiber diameter (mm)	Aspect ratio (length/diameter)	No. of specimens
DF-7	0.837	0.021	39.7	60
DF-26	0.783	0.022	35.5	60
DF-61	0.760	0.024	31.7	60
OF	1.8	0.25	7.2	60

The tensile modulus, maximum tensile strength, elongation at break, and maximum work were automatically calculated from the stress-strain data. At least three specimens of each formulation were tested. The data reported in this article are the mean values of three measurements.

Tensile tests were performed according to ASTM D 638 with a crosshead speed of 5 mm/min. Hardness and notched impact Izod tests were performed according to ASTM D 2240 and D 256, respectively.

Water absorption was measured by immersion in water for 24 h at room temperature according ASTM D 570.

## RESULTS AND DISCUSSION

### Lignin content of the fibers

The results of the fiber lignin content determination are summarized in Table II.

### Fiber length and aspect ratio analyses

The fiber length and aspect ratio analyses of the delignified fibers after grinding showed no significant difference (99% confidence level) between the fiber length and aspect ratio among the three types of delignified fibers (Table III). However, the aspect ratio of the original fibers was considerably lower than that of the delignified fibers.

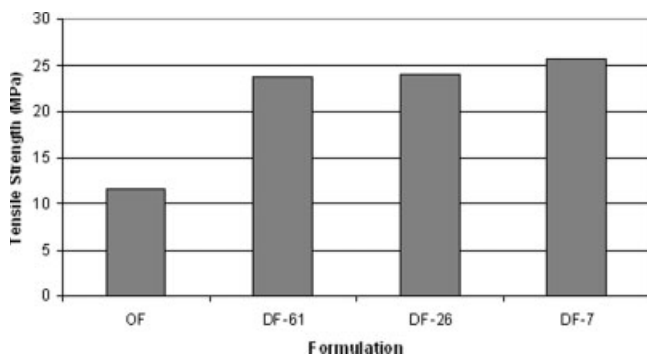
### Mechanical properties

The effect of delignification on the tensile strength of the composites is presented in Figure 1. A considerable increase was observed for DFs compared with

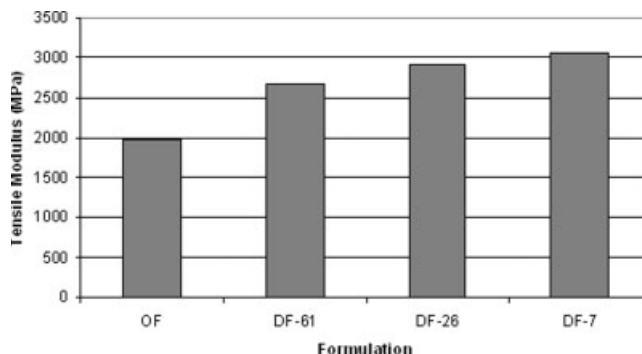
original fibers regardless of the degree of delignification. Statistical analysis indicated no significant difference among the tensile strengths of the three DFs. The tensile strength was highest (25.6 MPa) for the composite samples with 7% residual lignin (DF-7). MA has well-known compatibilizing effects on natural fiber-plastic composites. An increase in tensile strength meant that the stress was successfully transferred from the PP matrix to the fibers. Delignified fibers transferred the stress better than original fibers. This phenomenon resulted in a higher tensile strength due to the improvement of the adhesion and the nature of the fiber/matrix interface.<sup>12</sup> It was expected that cellulose would have a better chemical reaction with MA compared with lignin due to a higher concentration of hydroxyl groups. Therefore, the elimination of lignin from the fibers improved interfacial bonding, and this resulted in improvements in tensile strength.

The effect of delignification on the tensile modulus of the composites is presented in Figure 2. Modulus (stiffness) is one of the basic properties of composites and is normally determined from the initial slope of the stress-strain curve. As shown, a regular rising trend was observed when the amount of lignin in the fibers decreased. The highest tensile modulus (3060 MPa) was observed for DF-7, whereas the lowest tensile modulus was 1979 MPa and corresponded to the original fiber composites (OFs).

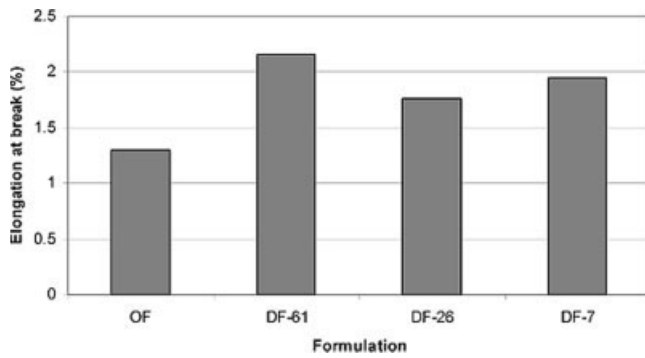
Fillers with higher moduli can generally cause higher modulus values in composites. Among different constituents of wood fibers, cellulose has the highest mechanical properties. Therefore, fibers containing less lignin will have higher modulus values.



**Figure 1** Tensile strength of the evaluated composites.



**Figure 2** Tensile modulus of the evaluated composites.

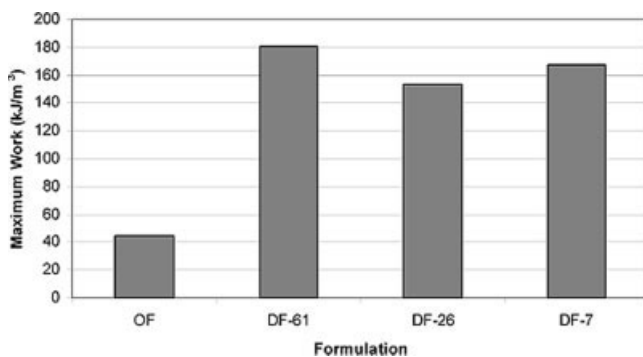


**Figure 3** Elongation at break of the evaluated composites.

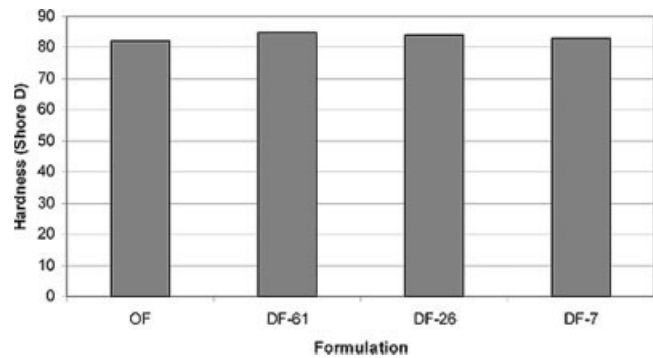
The original fibers differed from the delignified fibers in chemical and physical structure. The original fibers were thicker than the delignified fibers because they were mostly fiber bundles rather than individual fibers. Hence, the aspect ratio of original fibers was less than that of delignified fibers. Generally, composites with smaller particles and greater aspect ratios show higher moduli.<sup>12</sup> Therefore, the increase in the composites' tensile modulus due to delignification was attributed to the higher aspect ratios in the case of the delignified fibers.

Figure 3 shows the effect of lignin on the elongation at break of the composites. The elongation at break was greater for DFs than for OFs. The highest value corresponded to the composites containing delignified fibers with 61% residual lignin (2.16%), whereas the lowest elongation was observed for OFs (1.3%). However, statistical analysis proved that the elongation values were not significantly different at a 95% confidence level.

Figure 4 shows the maximum work (energy) of the different evaluated composites. The maximum work was determined from the area under the stress-strain curves. All of the delignified formulations presented higher maximum work values compared with the OFs. A visual comparison of Figures 3 and 4 reveals that the trend was the same. This was because higher strain and stress values normally result in



**Figure 4** Maximum work of the evaluated composites.



**Figure 5** Hardness of the evaluated composites.

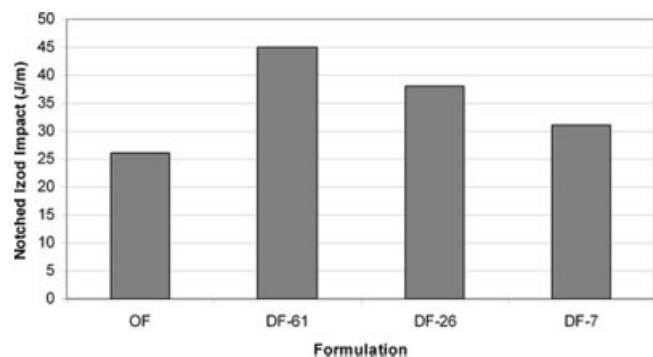
higher areas under the stress-strain curves and lead to higher energy values.

However, statistical analysis indicated no significant differences among the three delignified formulations regarding their maximum work. This meant that the amount of residual lignin (or degree of delignification) had no significant effect on this property. Hence, the considerable increase in maximum work values in the case of delignified composites was due to changes in the physical structure of the fibers.

Figure 5 shows the effect of delignification on the hardness of the composites. No significant differences could be identified between the DFs and OFs, and all hardness values were very close to each other. However, the highest hardness value was approximately 85 shore D in DF-61, whereas the lowest value (82 shore D) corresponded to the OFs.

The effect of delignification on the notched Izod impact strength of the composites is presented in Figure 6. Clearly, with the elimination of lignin, the notched Izod impact strength decreased. However, all of the delignified formulations had higher impact strengths than the OFs.

Notched Izod impact energy is the energy required for crack propagation. The size and dispersion of filler particles in the matrix can affect the properties of the composites. Small well-dispersed particles generally give better properties. Small particles can block crack



**Figure 6** Notched Izod impact strength of the evaluated composites.

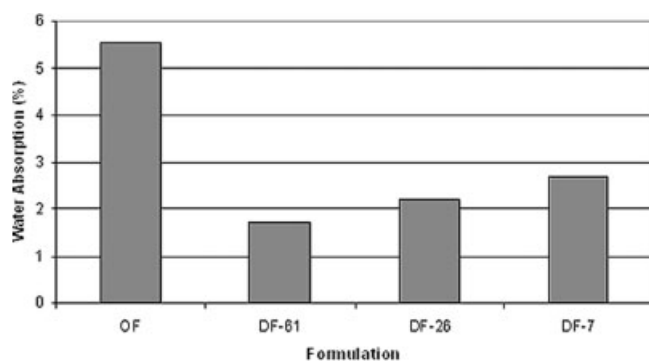


Figure 7 Water absorption of the evaluated composites.

propagation and result in impact toughening. Particles with higher aspect ratios have high stresses at the fiber ends that can cause failure under impact.<sup>13</sup>

### Water absorption

Figure 7 shows the water absorption of delignified and OFs. The composites containing original fibers absorbed water more than DFs. Bonding between the fiber surface and the matrix could have reduced available hydroxyl groups. On the other hand, due to a higher hygroscopicity of cellulose than lignin, the elimination of lignin led to a higher water absorption of fibers.

Improvements in the compatibility between the two phases and bonding between fibers and matrix explained the lower water absorption in the DFs.

### CONCLUSIONS

The effect of delignification of fibers on some physical and mechanical properties of wood fiber-PP composites was studied. The results obtained indicate that wood fiber-PP composites filled with

delignified fibers exhibited higher mechanical properties than the original fibers. The improvements in tensile modulus, tensile strength, and water absorption of the composites were considerable. However, some properties, such as notched impact strength, decreased at higher delignification levels.

The main reasons for these changes were modifications in the chemical structure, fiber length, and aspect ratio of the fibers caused by delignification. Finally, we concluded that the delignification of the fibers greatly improved the mechanical properties of wood fiber-PP composites. However, due to the higher costs associated with delignification processes, a trade-off must be reached to satisfy both economic and mechanical considerations.

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