

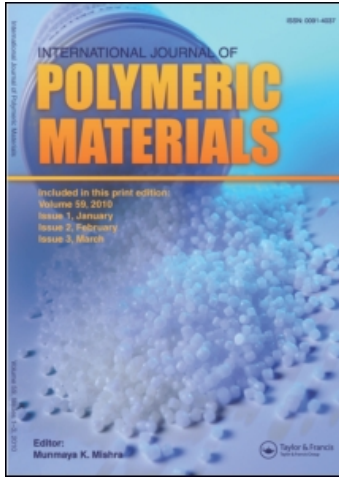
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Polypropylene-Wood Fiber Composites: Effect of Fiber Treatment on Mechanical Properties

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Polypropylene-Wood Fiber Composites: Effect of Fiber Treatment on Mechanical Properties

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Polypropylene (PP) was reinforced with different wood fibers, chemithermomechanical (CTMP) aspen and commercial pulps (Tempure and Temalfa-A). Various chemical treatments on the fiber was carried out to improve the bonding at the interface. Fibres coated with Silane coupling agents Silane A-172 and A-174 (with vinyl and methacryloxy functional groups respectively) upon reinforcement showed poor tensile strength. PP filled with Pre-coated fibers containing maleated propylene wax, polymer and polymethylene polyphenyl isocyanate produced higher tensile strength and modulus. The use of dicumyl peroxide and cummine hydro peroxide as initiators during the coating of the fiber was not effective. Polypropylene reinforced with fibers of lower mesh size gave better tensile properties.

INTRODUCTION

The addition of discontinuous fibers into the polymer matrix can impart greater strength and stiffness to polymeric materials.^{1,2} Wood fibers are increasingly used as a reinforcing filler for thermoplastics because of the many advantages they offer. Indeed, they are inexpensive, easily renewable, density is lower than mineral fibers and undergoes little damage during processing.^{3,4} The main problem in reinforcing thermoplastics is the hydrophilic

character of the filler surface which offers little interfacial interaction with the hydrophobic polymer. The interface plays a significant role in the effective transfer of stress from matrix to fiber. Strong bonding at the interface can be achieved through the use of bonding agents or coupling agents, which can act as a link between the fiber and the matrix by the formation of chemical bonds.⁵

The choice of chemical structure and the concentration of coupling agents play an important role in achieving the optimum mechanical properties of the composite.^{6,7} The most commonly used coupling agents are silanes and titanates.⁸ The dispersion of fiber in the matrix is one of the important factor to achieve a lower degree of variation in the ultimate properties of the composite. Pre-treatment of the fibers with thermoplastics or an elastomer and a lubricant to facilitate better dispersion of wood fibers in the polymer matrix has been reported.⁹

In the present work, the composites of polypropylene were prepared by reinforcement with different weight percentage of wood fiber. The influence of fiber surface treatment with silane coupling agents A-172 and A-174, pre-coating of the fibers with a wetting agent, polymer and a bonding agent on the tensile properties of the composite was studied. Also, the effect of composite properties when using an initiator during the coating of the fiber and the variation in fiber mesh size was studied.

EXPERIMENTAL

Polypropylene (Pro-FAX 6331) was supplied by Himont Canada Inc., nominal melt flow, (g/10 min) 12; density g/cc 0.903. The preparation conditions and properties of CTMP aspen was same as reported earlier.¹⁰ Two commercial pulps Tempure 626 and Temalfa-A 6816 (Tembec Ltd.) were also used for reinforcement. The properties of the commercial wood pulps as supplied by the manufacturer are listed in Tables I and II. The silane coupling agents

- 1) Silane A-172: Vinyltri (2-methoxy ethoxy) silane
- 2) Silane A-174: Gamma-Methacryloxy Propyltri methoxy silane was obtained from Union Carbide. The procedure for coating wood

fibers with silanes (2.0 percent weight of fiber) was same as described earlier.¹¹ Polymethylene polyphenyl isocyanate (PMPPIC) obtained from Polysciences Inc., was used as a bonding agent along with a small amount of maleated propylene wax (Eastmen chemical Epolene E43) to aid in the wetting and dispersion of the fiber in the polymer matrix.

Coating on the fiber was carried out at 165°C in a C.W. Brabender laboratory prep. roll mill. The mixing was repeated 4 to 5 times to allow better dispersion of chemicals in the fiber. The different coated fibers used for reinforcement are:

- a) Coated fiber 1—Epolene (5.0 percent, and polymer (10.0 percent)
- b) Coated fiber 2—PMPPIC (3.0 percent), Epolene (5.0 percent and polymer (10.0 percent)
- c) Coated fiber 3—Epolene (5.0 percent), PMPPIC (3.0 percent), polymer (10.0 percent) and dicumyl peroxide or cummine hydro peroxide (2.0 percent)

The chemicals used were by percentage weight of the polymer. The coated fibers were ground to mesh size 60 and pre-dried in an oven at 60°C prior to mixing with the polymer. The polymer and fibers were compounded in a laboratory model extruder at 160°C. The concentration of the fibre was varied from 0 to 40 percent by weight of the fiber. The extruded samples were ground to mesh size 20 before compression molded into shoulder type tensile specimen. The molding conditions were: temperature 165°C; pressure 3.8 MPa.; time 15 minutes.

TABLE I
Properties of Tempure 626

Alpha cellulose	%	90.9	Basic weight, g/M ²	691
Viscosity	Xan. secs.	100	App. density g/cc	0.68
Brightness.	Elrepho	95.5	% Air dry	101.8
Resin	%	0.34	Cu. No.	1.15
Ash	%	0.096	Dirt M ² of surface	23.0
Calcium,	P.P.M.	125	Pentosen, %	2.61
Copper,	P.P.M.	0.8	Manganese, P.P.M.	0.3
Iron,	P.P.M.	5.3	pH	7.1

TABLE II
Properties of Temalfa-A 6816

Alpha cellulose	%	93.4	Basic weight, g/M ²	724
Viscosity	Xan. secs.	69	App. density g/cc	0.79
Brightness,	Elrepho	94.2	% Air dry	104.2
Resin	%	0.03	Cu. No.	0.54
Ash	%	—	Dirt M ² of surface	20.0
Calcium,	P.P.M.	91	Pentose, %	—
Copper,	P.P.M.	1.7	Manganese, P.P.M.	0.1
Iron,	P.P.M.	8.2	pH	—

Tensile properties of the composites were measured with an Instron Model 4201. Crosshead speed was 10 mm/min and properties were measured at maximum load. A minimum of six samples were tested in each case and the results were computed by the HP86B system using Instron 2412005 General Tensile Test program. The coefficient of variation was less than 7.0 percent.

RESULTS AND DISCUSSION

Effect of PMPPIC treatment

Tensile strength and elongation of PMPPIC treated Polypropylene reinforced with Tempure and Temalfa-A fibers decreased rapidly with the increase in fiber addition as seen from Table III. In fact, there is little difference between the strength values of untreated fiber and isocyanate treated PP-fiber composites. Tensile modulus

TABLE III
Tensile properties of untreated and PMPPIC treated PP-wood fiber composites

Composite fiber (wt. %)	Strength (MPa)				Elongation (%)				Modulus (MPa)			
	10.0	20.0	30.0	40.0	10.0	20.0	30.0	40.0	10.0	20.0	30.0	40.0
Tempure												
Untreated	26.8	23.7	21.9	19.8	5.3	4.9	3.2	2.5	1209	1432	1717	1865
PMPPIC treated	23.4	22.4	18.2	21.6	4.1	2.9	2.7	2.8	1340	1459	1576	1884
Temalfa-A												
Untreated	29.7	26.2	22.0	18.9	5.3	5.0	3.8	2.4	1288	1384	1570	1852
PMPPIC treated	26.4	22.6	21.6	22.7	4.3	3.5	2.9	2.5	1230	1441	1520	1815
PP	32.6				5.8				1071			

was relatively unaffected by isocyanate treatment and increased steadily with the filler loading in the composite.

The effect of isocyanate treatment on tensile properties of PP-CTMP aspen fiber composites is illustrated in Figures 1–3. A slight increase in strength was observed in PMPPIC pre-treated PP composites at a higher percentage of filler level (Figure 1). The decrease in strength in untreated fiber composites was mainly due to poor adhesion at the interface. This was supported by the increase in strength when isocyanate coated fiber was used for reinforcement. A further increase in strength was observed in PMPPIC pre-treated PP reinforced with the isocyanate coated fiber.

Elongation generally decreased with the increase in fiber content irrespective of the fiber treatment (Figure 2). However, the rate of decrease was lower in PP-isocyanate coated fiber1 composites. As regards to modulus, the increase was mainly influenced by the percentage of fiber reinforcement in the polymer which is evident from the higher modulus values observed in untreated fiber composites (Figure 3).

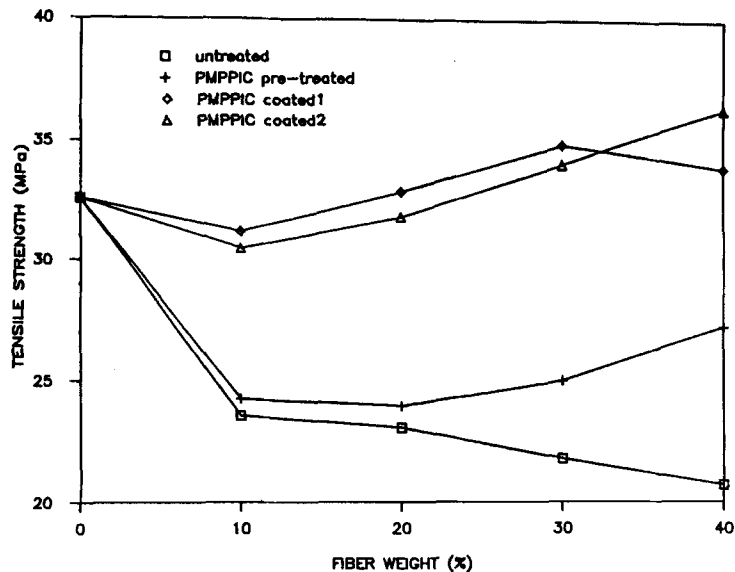


FIGURE 1 Tensile strength of PP-CTMP aspen fiber composites at different fiber weight.

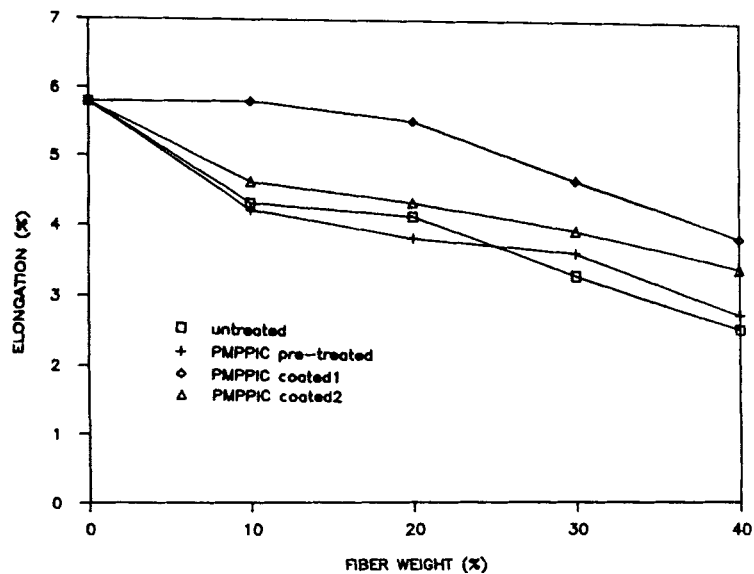


FIGURE 2 Elongation of PP-CTMP aspen fiber composites at different fiber weight.

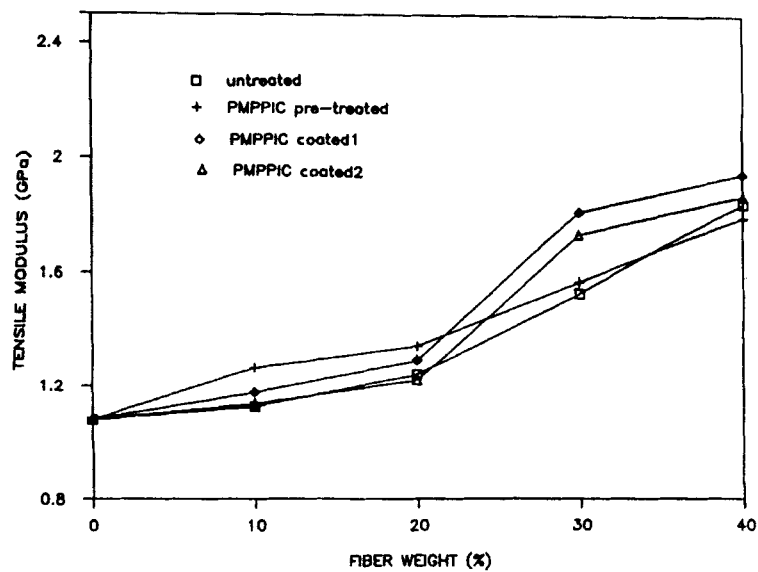


FIGURE 3 Tensile modulus of PP-CTMP aspen fiber composites at different fiber weight.

Effect of silane coupling agent

Silane coupling agents are widely used to improve interfacial adhesion at the glass fiber or particulate filler-matrix interface. In the present study, silane coupling agent with different molecular weight and chemical structure was used to coat the wood fiber. The coupling agent can be represented by the formula



where X is a hydrolyzable alkoxy group, Y is a functional group and R is a small aliphatic linkage. Wood fiber contains bound moisture which could act to hydrolyze the coupling agent.¹² The hydrolyzed product, Si(OH)₃ (Silanol) can develop either covalent or hydrogen bonds with the hydroxyl rich wood fiber. Subsequent reaction of the functional organic group Y along with the polymer completes the bridge like structure between the fiber and the polymer.

Table IV shows the effect of silane coupling agent treatment on the tensile properties of PP-CTMP aspen, Tempure and Temalfa-A composites. The tensile strength and elongation decreased rapidly, with the increase in fiber concentration, in Silane A-172 (vinyl functional group) coated fiber composites. The composites of PP-Tempure and Temalfa-A fibers coated with Silane A-174 (methacryloxy functional group) showed a slower decrease in tensile strength and elongation. Also, the modulus increased significantly in Silane A-174 coated fiber composites. At 40.0 percent fiber level in Temalfa-A composites the modulus reached 1911 MPa which is

TABLE IV
Effect of silane A-172 and A-174 treatment on tensile properties of PP-wood fiber composites

Composite Fiber (wt. %)	Strength (MPa)				Elongation (%)				Modulus (MPa)			
	10.0	20.0	30.0	40.0	10.0	20.0	30.0	40.0	10.0	20.0	30.0	40.0
Silane A-172												
Aspen	31.2	30.2	24.6	26.4	4.2	4.1	3.5	2.7	1557	1619	1772	1886
Tempure	22.3	23.5	19.6	15.6	3.1	2.8	2.5	1.3	1208	1437	1539	1678
Temalfa-A	25.6	24.3	22.4	21.5	3.7	3.3	2.3	2.1	1341	1416	1499	1739
Silane A-174												
Tempure	28.4	27.1	25.7	27.6	5.6	4.4	3.2	2.8	1308	1433	1625	1878
Temalfa-A	28.6	26.2	24.3	23.5	5.1	4.7	4.3	3.7	1451	1510	1627	1911

nearly twice the value of PP (1071 MPa). The poor performance of Silane treated fiber composites may be due to the lower surface coverage of wood fibers, which tends to restrict the availability of silanol groups for the formation of covalent bonds.

Effect of coated fiber

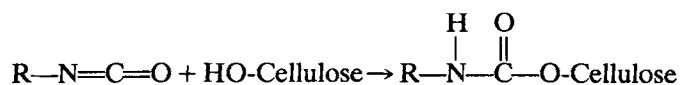
Another possible way of achieving a stronger interface is the pre-treatment of the fiber before it is incorporated into the polymer. The pre-treated fibers have additional advantages such as lower fiber-to-fiber interaction and better dispersion in the matrix.¹³ Tensile properties of PP reinforced with different coated fiber composites are shown in Table V. In CTMP aspen coated fiber composites a moderate increase in strength was observed. At the same time, the elongation decreased with increase in filler level.

A greater increase in strength was observed at 40.0 percent filler level in Tempure composites. The rate of increase in modulus was also higher in comparison with CTMP aspen fiber composites. PP reinforced with Temalfa-A coated fiber2 composites produced better tensile properties than coated fiber1 composites. Modulus attained a maximum value of 2402 MPa in coated fiber2 composites (at 40.0 percent fiber level) compared to 1071 MPa of PP. The better performance of coated fiber composites may be due to the higher reactivity of isocyanate with the wood fiber. The isocyanate

TABLE V
Tensile properties of PP-coated wood fiber composites

Composite Fiber (wt. %)	Strength (MPa)				Elongation (%)				Modulus (MPa)			
	10.0	20.0	30.0	40.0	10.0	20.0	30.0	40.0	10.0	20.0	30.0	40.0
Aspen												
Coated fiber1	33.8	34.0	32.8	32.3	4.0	3.7	3.0	2.3	1436	1747	1819	2134
Coated fiber2	32.1	32.7	34.4	33.1	5.9	5.5	3.5	3.1	1176	1406	1832	1956
Tempure												
Coated fiber1	32.2	32.7	33.1	36.7	5.7	4.3	3.4	3.1	1425	1733	1922	2057
Coated fiber2	31.9	32.8	34.3	36.2	5.7	4.6	3.2	2.7	1402	1670	1816	2092
Temalfa-A												
Coated fiber1	31.6	32.1	31.5	30.7	3.5	3.2	2.9	2.3	1469	1527	1626	2005
Coated fiber2	30.9	31.2	33.2	35.6	5.8	4.9	3.7	2.9	1341	1572	1798	2402

reacts readily with the hydroxyl groups of cellulose



to form urethane structure.¹⁴ The direct covalent bond with cellulose and lignin hydroxyls of the wood fibers account for better improvement in the tensile properties of the composite.

The influence of isocyanate content on tensile properties of the composites is shown in Figures 4–6. An increase in tensile strength particularly at higher percentage of fiber was observed in PMPPIC pre-treated PP reinforced with coated fiber2 Tempure and Temalfa-A composites (Figure 4). Elongation continued to decrease with the increase in fiber concentration and was not affected by the isocyanate content (Figure 5). A higher rate of increase in the modulus was observed after 20.0 percent fiber loading in PMPPIC pre-treated PP reinforced with coated Tempure fibers (Figure 6).

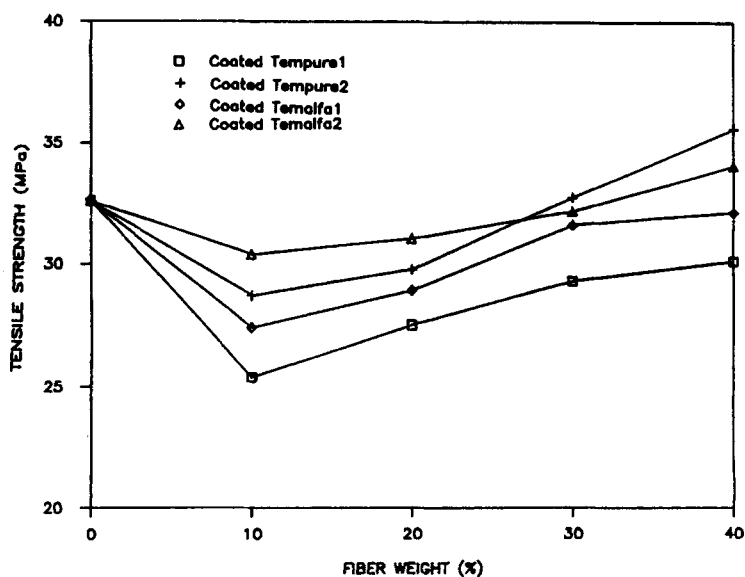


FIGURE 4 Tensile strength of PP-coated Tempure and Temalfa-A fiber composites at different fiber weight.

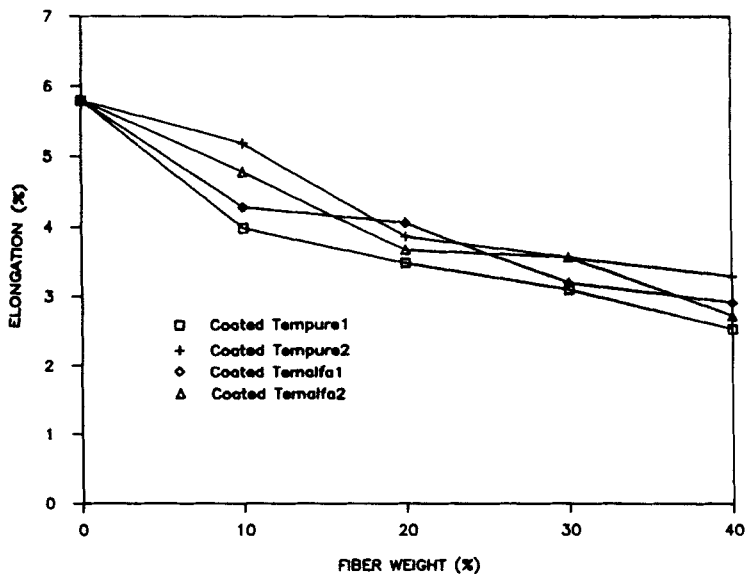


FIGURE 5 Elongation of PP-coated Tempure and Temalfa-A fiber composites at different fiber weight.

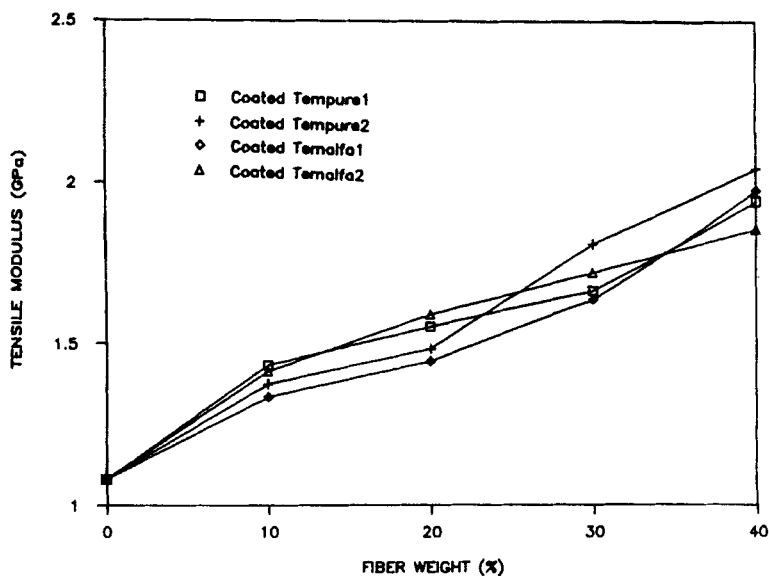


FIGURE 6 Tensile modulus of PP-coated Tempure and Temalfa-A fiber composites at different fiber weight.

Effect of initiator

To promote the rate of chemical reaction during fiber coating, dicumyl peroxide and cummine hydroperoxide were tried as an initiator. When dicumyl peroxide was used in Tempure coated fiber3 composites, the tensile strength and elongation decreased drastically (Table VI). With cummine hydroperoxide an increase in strength was observed at 40.0 percent fiber concentration. Elongation decreased gradually and a higher rate of increase in modulus was observed. In Temalfa-A composites, the strength decreased rapidly at higher filler level when dicumyl peroxide was used. Cummine hydro peroxide performed slightly better compared to the former.

TABLE VI
Tensile properties of PP-Coated fiber3 composites

Composite Fiber (wt. %)	Strength (MPa)				Elongation (%)				Modulus (MPa)			
	10.0	20.0	30.0	40.0	10.0	20.0	30.0	40.0	10.0	20.0	30.0	40.0
Tempure												
Dicumyl p.o.	21.1	22.6	—	17.4	2.3	2.5	—	1.6	1218	1386	—	1628
Cummine h.p.o.	30.9	29.3	32.7	35.3	5.2	3.9	3.6	3.1	1367	1693	1913	2108
Temalfa-A												
Dicumyl p.o.	28.3	30.7	—	19.0	3.8	3.7	—	1.4	1117	1434	—	1978
Cummine h.p.o.	24.2	24.0	32.1	30.2	5.0	3.6	3.7	2.9	1177	1330	1711	1909

Effect of fiber mesh size

From Table VII it can be seen that tensile strength and elongation of Tempure composites with fiber mesh size 20 decreased at higher concentration of fiber. In Temalfa-A composites (mesh size 20), the

TABLE VII
Effect of fiber mesh size on tensile properties of PP-coated fiber2 composites

Composite fiber (wt. %)	Strength (MPa)				Elongation (%)				Modulus (MPa)			
	10.0	20.0	30.0	40.0	10.0	20.0	30.0	40.0	10.0	20.0	30.0	40.0
Tempure												
Mesh 20	25.2	27.8	28.5	26.1	4.0	3.6	3.1	2.5	1407	1555	1678	1939
Mesh 60	31.9	32.8	34.3	36.2	5.7	4.6	3.2	2.7	1402	1670	1816	2092
Temalfa-A												
Mesh 20	30.4	30.1	31.2	34.1	4.9	3.7	3.4	3.0	1392	1547	1713	1871
Mesh 60	30.9	31.2	33.2	35.6	5.8	4.9	3.7	2.9	1341	1572	1798	2402

tensile strength remained almost unchanged with the addition of fiber. But in PP reinforced with mesh size 60 fiber, the tensile strength increased in both coated fiber composites.

CONCLUSIONS

Tensile properties of PP can be improved by reinforcement with wood fibers pre-treated before being incorporated in the polymer. The composites made from silane coupling agent coated fibers produced poor tensile strength and elongation. Among the various fiber treatments studied, the coated fiber containing maleated propylene wax, PMPPIC and polymer produced better mechanical properties. The use of initiator was not effective in promoting the bonding at the interface. PP reinforced with shorter fibers produced better tensile strength.

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