

# Use of Wood Fibers in Thermoplastics. VII. The Effect of Coupling Agents in Polyethylene–Wood Fiber Composites

R. G. RAJ, B. V. KOKTA, D. MALDAS, and C. DANEULT, *Centre de Recherche en Pâtes et Papiers, Université du Québec à Trois-Rivières, C. P. 500, Trois-Rivières, Québec, Canada, G9A 5H7*

## Synopsis

Linear low density polyethylene (LLDPE) was reinforced with different wood fibers, aspen chemithermomechanical pulp (bleached and unbleached), and other commercial wood pulps. Silane coupling agents A-172, A-174, A-1100, and polymethylene polyphenyl isocyanate were used to improve the bonding between the fiber and matrix. LLDPE filled with pretreated wood fiber produced a significant improvement in tensile strength and modulus. Comparison of tensile and impact properties of wood fiber composites with mica and glass fiber composites shows the potential advantage (in terms of material cost and specific properties) of wood fiber as a reinforcement.

## INTRODUCTION

There is a good potential for the use of natural fibers as reinforcing fillers in thermoplastics. The main advantage of these fibers are their low cost, low density, and resistance to breakage during processing.<sup>1</sup> In addition, these fibers offer an excellent opportunity to utilize an abundant source of such materials available from nature.<sup>2</sup> Several cellulosic wastes such as ground wood waste, bark, nut shells, begasse, corncobs, bamboo, and cereal straw have been used as fillers for plastics.<sup>3</sup> The reinforcing fibers play an important role in strengthening the composites by effective transfer of stress between the fiber and matrix.<sup>4</sup> The compatibility of hydrophobic polymer and hydrophilic cellulose fiber can be enhanced by the modification of polymer or fiber surfaces. Morrell reported the use of coupling agents to promote the adhesion between the fiber surface and matrix.<sup>5</sup>

Chemical modification of wood with alkoxy silane coupling agents has been reported by Schneider and Brebner.<sup>6</sup> They found reduced tendency to shrink and dimensional stability comparable to those reported for chemically reactive wood modifiers such as epoxides, anhydrides, and isocyanates. Beshay et al. reported that grafted aspen chemithermomechanical pulp used as filler increased the mechanical properties of low density polyethylene.<sup>7</sup> Improvement in tensile properties of polyolefin–kraft composites by lamination and polymer impregnation was reported by McKenzie and Yuritta.<sup>8</sup> The reactions of cellulose and lignin with isocyanates and prepolymers containing isocyanate groups was reviewed by Reichelt and Poller.<sup>9</sup> Using ceric-ion-initiated polymerization, Gaylord has shown the compatibilization of two normally incompatible polymers by the use of a third component which is a graft or block

copolymer.<sup>10</sup> The dispersion of wood fibers in the plastic material may be facilitated by pretreatment of fibers with a thermoplastic polymer and a lubricant was reported by Hamed.<sup>11</sup>

The individual fibrous wood ribbons possess tensile strengths and moduli which compare favorably with those of glass fiber.<sup>12</sup> Also, the lower density of wood fiber offers significant advantages in terms of specific cost and performance compared to other materials used in construction.

In the present study, different silane coupling agents and polymethylene-polyphenyl isocyanate were used to modify the wood fiber surface in an attempt to improve the adhesion between the fiber and matrix. Effect of different wood pulps on the mechanical properties of the composites was examined. Tensile and impact strength of wood-fiber-reinforced composites were compared with mica and glass fiber composites.

## EXPERIMENTAL

### Materials

Linear low density polyethylene (LLDPE GRSN 7064) was supplied by Novacor Chemicals Ltd. (Melt Index 0.85 g/10 min; density 0.926 g/cm<sup>3</sup>). Chemithermomechanical pulp (CTMP) of aspen was prepared in a Sund defibrator.<sup>7</sup> Commercial wood pulps, Tempure 626 (alpha cellulose 90.9%) and Temalfa A-6816 (alpha cellulose 95.6%), were supplied by Tembec Ltd.

Mica-200-NP-Suzorite (200 mesh, silane-coated) was supplied by Lacana Mining Co., Montreal) and glass fibers 731BA 1/32 (0.8 mm, silane-coated) by Fiberglas Canada via Mia Chemicals, Montreal.

The following coupling agents were obtained from Union Carbide Co., Montreal: (i) silane A-172 [Vinyltri(2-methoxy ethoxy)silane]; (ii) silane A-174 (gamma-methacryloxy-propyltrimethoxysilane); (iii) silane A-1100 (gamma-amino propyl triethoxy silane).

### Silane Coupling Agent Treatment

The wood fibers were treated with different silane coupling agents. The following procedure was used:

(a) 25 g of fiber (mesh size 60) was placed in a flask to which 150 mL of Carbon tetrachloride was added, followed by 0.4 g of dicumyl peroxide and 1.0–4.0 wt % of silane A-172 or A-174. The mixture was refluxed at 70°C with continuous stirring for 3 h. After cooling, the carbon tetrachloride was evaporated, and the mixture was dried at 55°C for 24 h.

(b) In the case of silane A-1100, a two-stage mixing procedure was used. The mixing procedure for the first stage was same as described earlier. In the second stage, the polymer (LLDPE, 2.0 g) was mixed with 100 mL of *p*-xylene, 0.2 g of maleic anhydride, and 0.1 g of benzoyl peroxide. The above mixture was refluxed with continuous stirring for 3 h; then the contents of a and b were combined and refluxed at 80°C for 2 h. The mixture was allowed to cool to room temperature, filtered, washed with distilled water, and then dried at 105°C for 12 h.

### Isocyanate Treatment

Polymethylenepolyphenyl isocyanate (PMPPIC, Polysciences Inc.) was used as a bonding agent:

(i) 3.0% of PMPPIC based on polymer weight was added to LLDPE (45.0 g) and mixed thoroughly at room temperature; then the wood fibers were added at different weight percentages (0–40.0%).

(ii) 15.0 g of oven dried wood fibers were mixed with PMPPIC (9.0 wt % of fiber) and polymer (5.0% or 10.0 wt % of fiber). The above mixture was added gradually to a preheated roll mill (C. W. Barbender Laboratory prep. mill no. 065) at 165°C. The mixing was repeated 4 to 5 times and then allowed to cool to room temperature:

$$\text{Coated}_1 \text{ fiber} = (\text{PMPPIC } 9.0\%) + (\text{polymer } 5.0\%)$$

$$\text{Coated}_2 \text{ fiber} = (\text{PMPPIC } 9.0\%) + (\text{polymer } 10.0\%)$$

### Preparation of Composites

The wood fibers were mixed at different weight percentages (0–40.0%) with LLDPE and extruded at 150°C in a laboratory extruder (Custom Scientific Instruments Model CS194). The extruded samples were ground to pass through mesh size 20.

The above mixture was compression-molded into dog-bone-shaped tensile test specimens. Molding temperature was 150°C and pressure 3.3 MPa. After 15 min, the samples were cooled to room temperature with the pressure maintained during cooling. The same molding procedure was used in preparing mica and glass fiber composites.

### Mechanical Tests

Tensile properties were measured according to the ASTM D638 procedure. The full-scale load was 500 N and crosshead speed was 10 mm/min. Properties reported were measured at yield point. The secant modulus was based on an original point defined by load at set elongation (LASE) 0.1%. A minimum of six specimens were tested in each series. The properties were simultaneously calculated by HP86B computer with the help of Instron 2412005 General Tensile Test Program. The average coefficients of variation were: stress 3.0–8.2%; strain 4.4–7.3%; energy 5.0–7.6%; modulus 2.8–4.7%.

## RESULTS AND DISCUSSION

### Untreated Wood Pulp Composites

Tensile properties of LLDPE reinforced with various untreated wood pulps are presented in Table I. Yield stress increased marginally at 10.0% fiber level, and then it tends to decrease upon further addition of filler in Tempure 626 and Temalfa-A6816 composites. CTMP aspen fibers, because of high lignin content, were expected to produce a higher yield stress. But, the experimental results showed a decrease in stress as the concentration of fiber increased. This

TABLE I  
Tensile Properties of Untreated, Silane A-172-, and A-174-Treated Wood Pulp Composites

Fiber treatment	Fiber (wt %)			Stress (MPa)			Energy ( $\text{KJ} \times 10^{-5}$ )			Elongation (%)			Modulus (MPa)		
	10	20	30	10	20	30	10	20	30	10	20	30	10	20	30
Aspen															
Untreated	12.9	10.8	9.6	6.8	6.7	3.0	5.2	3.1	3.1	339	433	545			
Silane A-172	13.3	16.0	13.4	13.9	12.2	5.0	10.4	7.6	3.8	425	524	582			
Aspen (bleached)															
Untreated	10.4	9.9	9.8	5.3	3.8	2.6	9.6	3.7	2.7	336	360	515			
Silane A-172	13.8	14.3	14.8	9.0	9.8	6.0	10.3	6.9	4.1	479	553	650			
Silane A-174	13.3	14.2	11.6	7.9	8.1	4.9	5.9	5.7	4.2	338	364	—			
Tempure 626															
Untreated	13.8	11.7	11.8	8.4	4.9	3.3	8.9	4.2	2.8	531	605	624			
Silane A-172	11.2	13.8	17.3	10.1	11.2	7.4	9.0	8.1	4.3	466	474	826			
Silane A-174	12.7	14.1	12.9	7.9	9.1	7.0	6.3	6.4	5.4	328	371	529			
Temalfa-A6816															
Untreated	13.2	12.7	12.8	7.3	7.0	5.9	8.9	5.6	3.1	468	615	711			
Silane A-172	12.7	13.3	12.9	10.5	12.9	8.6	11.3	9.9	6.6	428	—	470			
Silane A-174	13.8	13.7	12.8	10.2	11.3	6.9	12.0	8.2	5.4	330	393	—			
LLDPE	—	10.9	—	—	8.2	—	—	11.3	—	—	325	—			

Concentration of Silane A-172 and Silane A-174 (1.0 percent weight of fiber).

TABLE II  
Tensile Properties of Silane A-172, A-174, and A-1100-Treated Aspen and Temalfa-A 6816 Composites

Fiber (wt %)	Stress (MPa)			Energy (kJ × 10 <sup>-5</sup> )			Elongation (%)			Modulus (MPa)		
	10	20	30	10	20	30	10	20	30	10	20	30
Silane treatment <sup>a</sup>												
Aspen												
Silane A-172	13.9	15.6	15.9	11.0	13.2	7.9	12.6	8.5	5.0	397	577	786
Silane A-174	13.1	13.6	13.2	9.5	9.6	6.4	11.0	7.0	4.9	433	459	616
Silane A-1100	13.2	15.5	14.3	7.9	9.8	6.6	10.3	6.3	4.6	469	578	674
Temalfa-A6816												
Silane A-172	14.1	15.4	14.7	12.1	11.2	10.3	8.5	7.2	7.0	485	604	643
Silane A-174	13.2	12.7	13.9	11.1	8.7	7.8	12.2	6.8	5.6	389	453	606
Silane A-1100	12.2	13.8	13.4	8.5	7.9	6.3	7.0	5.7	4.7	409	470	483
LLDPE	—	10.9	—	—	8.2	—	—	11.3	—	—	325	—

<sup>a</sup>4.0% by weight of fiber.

may be due to the poor dispersion of fiber in the matrix. Fracture energy and elongation at yield decreased with an increase in fiber concentration and was not affected by type of wood fiber. However, higher fiber reinforcement resulted in an increase in the stiffness of the composite. Significant increase in elastic modulus, with the addition of fiber, was observed in Tempure 626 and Temalfa A-6816 composites.

### Silane-Treated Wood Fiber Composites

Pretreatment of wood fibers with silane coupling agents showed a positive effect on tensile properties of the composites (Table I). The improvement in stress and modulus was slightly better in silane-A-172-treated wood fiber composites. In Tempure 626 composites, the yield stress increased from 10.9 MPa, for unfilled polymer, to 17.3 MPa at 30.0% fiber content. Elongation at yield was higher in silane A-172-treated aspen composites compared to untreated aspen composites. A significant increase in fracture energy at yield was observed in aspen, Tempure 626, and Temalfa-A 6816 composites. LLDPE filled with silane A-172-treated Tempure 626 produced highest modulus increase of 826 MPa at 30.0% filler content compared to 325 MPa for unfilled polymer. The reason for improvement in tensile properties of silane treated fibers may be due to (i) a higher fiber dispersion in the matrix and (ii) a fair degree of adhesion at the interface.

When the concentration of silane was increased (4.0% weight of fiber), the tensile properties showed an increase (Table II). The results indicate a

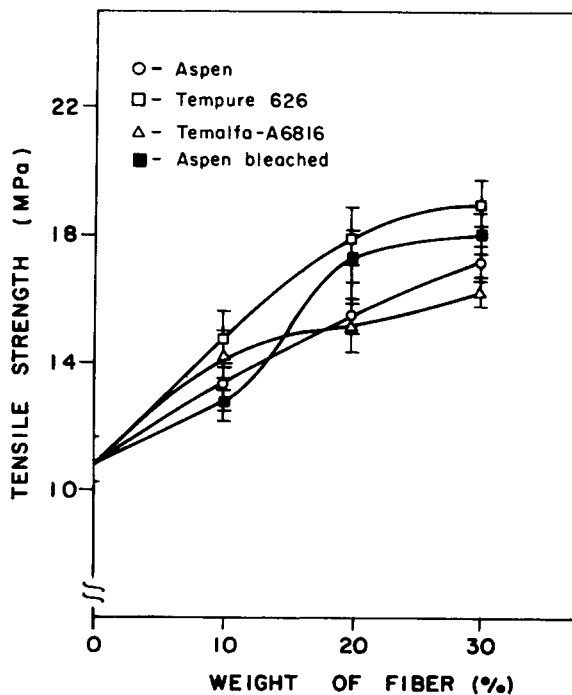


Fig. 1. Effect of PMPPIC treatment on tensile strength of LLDPE-wood fiber composites: (○) aspen; (□) Tempure 626; (△) Temalfa-A 6816; (■) aspen bleached.

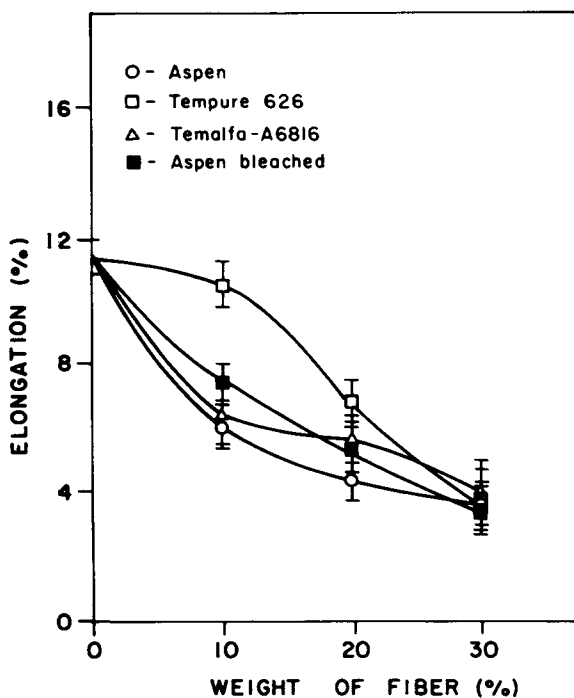


Fig. 2. Effect on PMPPIC treatment on elongation of LLDPE-wood fiber composites: (○) aspen; (□) Tempure 626; (△) Temalfa-A 6816; (■) aspen bleached.

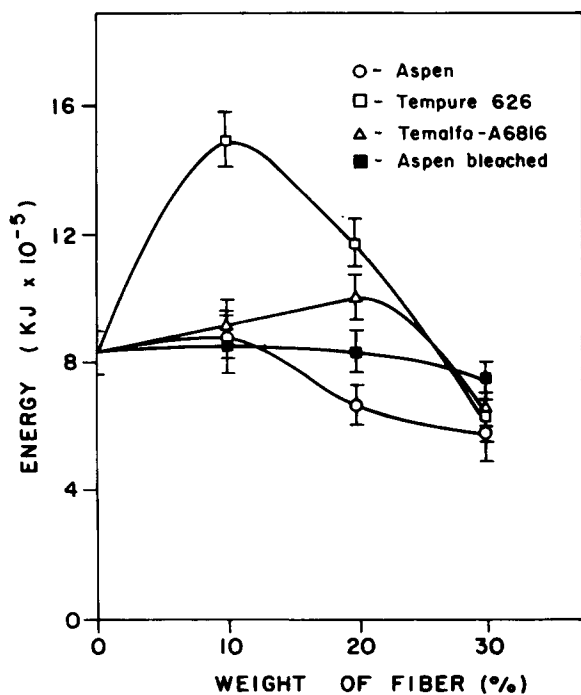


Fig. 3. Effect of PMPPIC treatment on energy of LLDPE-wood fiber composites: (○) aspen; (□) Tempure 626; (△) Temalfa-A 6816; (■) aspen bleached.

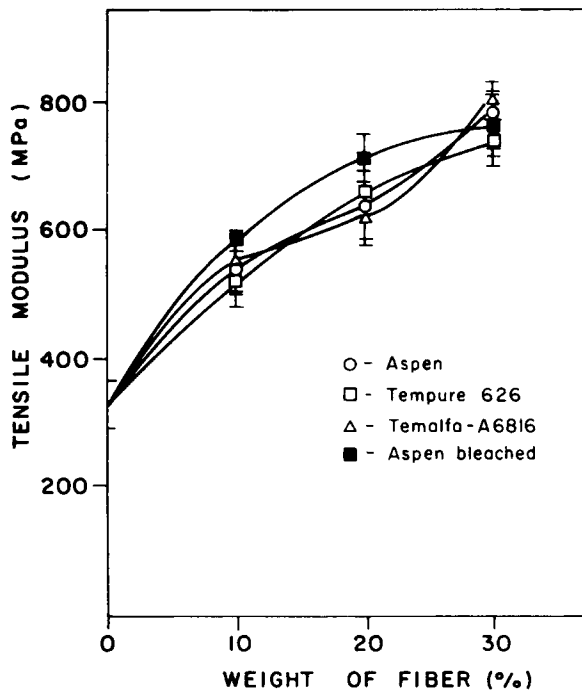


Fig. 4. Effect of PMPPIC treatment on tensile modulus of LLDPE-wood fiber composites: (○) aspen; (□) Tempure 626; (△) Temalfa-A 6816; (■) aspen bleached.

significant increase in yield stress, with increase in fiber content, in silane A-172-treated aspen composites. While the elongation at yield remained unchanged, fracture energy increased when compared to unfilled LLDPE. Silane A-172 pretreated Temalfa-A 6816 composites showed a 40.0% increase in yield stress at 20.0% filler content. Elongation at yield decreased gradually with the addition of fiber. On the other hand, fracture energy and elastic modulus improved significantly. The high efficiency of aspen fibers may be due to the high lignin content, which has a positive influence on adhesion to the polymer matrix.<sup>7</sup> It was also observed that choice of coupling agent and its concentration can affect the mechanical properties. Similar observations were made by Coutts and Campbell<sup>13</sup> in wood-fiber-reinforced cement composites.

#### Isocyanate Treatment in LLDPE-Wood Fiber Composites

Tensile properties of PMPPIC-treated LLDPE reinforced with various wood fibers are shown in Figures 1-4. Tensile strength at yield increased with the increase in filler concentration (Fig. 1). At 30.0% filler content the stress increased more than 70.0% in Tempure 626 composites. Elongation at yield continued to decrease with the increase in filler content (Fig. 2). LLDPE filled with Tempure 626 and Temalfa-A 6816 composites showed higher fracture energy at yield as seen from Figure 3. Elastic modulus increased steadily and was not affected by the nature of wood fiber (Fig. 4).



TABLE III  
PMPPIC Treatment on LLDPE and Aspen Fiber Composites

	Fiber (wt %)				Stress (MPa)				Energy ( $\text{kJ} \times 10^{-5}$ )				Elongation (%)				Modulus (MPa)			
	10	20	30	40	10	20	30	40	10	20	30	40	10	20	30	40	10	20	30	40
PMPPIC-treated LLDPE Composites	13.3	15.5	17.2	—	9.3	9.7	6.6	—	6.9	6.3	5.9	—	5.38	6.21	7.72	—	5.38	6.21	7.72	—
PMPPIC-coated <sub>1</sub> Aspen fiber composites	14.3	14.5	16.9	16.5	8.6	12.8	11.1	6.0	9.3	8.9	6.6	3.7	4.61	—	6.94	8.10	4.61	—	6.94	8.10
PMPPIC-pretreated LLDPE with coated <sub>1</sub> aspen fiber composite	16.4	16.5	18.0	19.1	14.9	15.5	12.2	9.3	13.4	9.4	6.8	4.9	6.14	6.91	7.01	8.35	6.14	6.91	7.01	8.35
PMPPIC-coated <sub>2</sub> aspen fiber composites	12.9	13.6	15.3	17.7	8.2	6.4	7.7	5.5	9.6	7.3	5.1	3.1	5.26	5.81	7.98	9.74	5.26	5.81	7.98	9.74
PMPPIC-pretreated LLDPE with coated <sub>2</sub> aspen fiber composites	15.3	16.6	17.1	19.7	16.2	17.0	13.8	11.4	14.7	10.3	8.1	5.8	5.73	7.02	7.16	8.52	5.73	7.02	7.16	8.52
LLDPE			10.9			8.2				11.3					325					

TABLE IV  
Effect of PMPPIC Treatment on LLDPE and Tempure 626, Temalfa-A 6816 Composites

Fiber (wt %)	Stress (MPa)			Energy (kJ × 10 <sup>-5</sup> )			Elongation (%)			Modulus (MPa)		
	10	20	30	40	10	20	30	40	10	20	30	40
PMPPIC treatment <sup>a</sup>	14.7	18.1	18.7	—	15.4	11.8	6.6	—	10.5	6.5	3.5	—
Tempure 626	13.9	16.2	—	18.1	10.8	8.7	—	6.4	7.8	5.4	—	3.5
Treated <sub>1</sub> composites	16.8	17.6	19.9	19.6	9.5	9.1	8.6	7.7	5.7	5.1	4.3	3.9
Treated <sub>2</sub> composites	14.1	15.3	16.3	—	8.7	9.7	6.2	—	6.2	6.3	3.8	—
Treated <sub>3</sub> composites	13.7	15.3	15.4	15.2	10.3	8.9	7.8	6.7	11.6	5.8	4.8	4.4
Temalfa-A 6816	15.9	16.3	17.4	17.2	13.9	11.3	10.2	9.9	8.8	6.9	5.9	5.7
LLDPE			10.9			8.2				11.3		325

<sup>a</sup> Treated<sub>1</sub> composites (PMPPIC-treated LLDPE composites); treated<sub>2</sub> composites (PMPPIC-coated<sub>1</sub> fiber composites); treated<sub>3</sub> composites (PMPPIC-pretreated LLDPE and coated<sub>1</sub> fiber composites).

TABLE V  
Effect of Variation in Mesh Size in PMPPIC-Treated Wood Fiber Composites

Fiber (wt %)	Stress (MPa)			Energy ( $\text{kJ} \times 10^{-5}$ )			Elongation (%)			Modulus (MPa)		
	10	20	30	10	20	30	10	20	30	10	20	30
Aspen (CTMP)												
20 mesh	14.8	18.9	18.8	12.4	13.3	8.5	12.8	7.0	4.5	447	665	727
60 mesh	13.3	15.5	17.2	9.3	9.7	6.6	6.9	6.3	3.9	538	621	772
Tempure 626												
20 mesh	14.3	15.8	17.3	14.3	14.2	8.2	9.2	9.0	4.8	543	624	818
60 mesh	14.7	18.1	18.7	15.4	11.8	6.6	10.5	6.5	3.5	540	645	743
Temalfa-A 6816												
20 mesh	12.6	10.7	9.8	10.0	6.1	3.7	11.7	5.6	3.8	454	556	587
60 mesh	14.1	15.3	16.3	8.7	9.7	6.2	6.2	6.3	3.8	556	608	779
LLDPE		10.9			8.2			11.3			325	

TABLE VI  
Composites of LLDPE with Aspen, Mica, and Glass Fibers

Fiber (wt %)	Stress (MPa)			Energy ( $\text{KJ} \times 10^{-5}$ )			Elongation (%)			Modulus (MPa)		
	10	20	30	40	10	20	30	40	10	20	30	40
Reinforcement	13.3	15.5	17.2	—	9.3	9.7	6.6	—	6.9	6.3	3.9	—
Aspen (CTMP) <sup>a</sup>	18.2	14.7	14.3	12.6	19.8	12.7	7.0	5.8	10.8	8.3	4.9	3.1
Mica	18.3	15.6	12.0	—	26.1	18.7	8.8	—	14.3	12.1	10.1	—
Glass fiber												
LLDPE			10.9				8.2				11.3	
												325

<sup>a</sup>PMPPIC treated (3.0% by weight of polymer).

Further improvement in tensile properties was achieved when PMPPIC was applied to the fiber instead of mixing with the polymer. Table III shows the results obtained with PMPPIC-coated aspen fiber. At 30.0% filling, the yield stress increased by 55.0% compared to unfilled polymer in coated<sub>1</sub> fiber composites. Maximum increase in stress was observed when PMPPIC-pre-treated LLDPE was reinforced with coated fiber. In coated<sub>2</sub> aspen composites, the stress increased from 10.9 MPa (unfilled polymer) to 19.7 MPa at 40.0% filler concentration. Also, a substantial increase in fracture energy was observed at lower filler concentration. Although the yield stress increased in coated aspen composites, the elastic modulus was influenced only to a minor extend.

Tensile properties of PMPPIC coated Tempure 626 and Temalfa-A 6816 composites are presented in Table IV. Yield stress increased linearly with fiber concentration. Tempure 626 composites produced higher yield stress than Temalfa-A 6816 composites. However, the elongation at yield and fracture energy were better in the latter case. Elastic modulus was not affected by PMPPIC treatment. The higher efficiency of PMPPIC, when coated with fiber, may be due to the formation of direct bond between the isocyanate groups and the OH groups of cellulose. The reason for excellent bonding efficiency of isocyanate was shown by Jhons.<sup>14</sup>

The effect of fiber mesh size on tensile properties of LLDPE filled with different wood fibers is shown in Table V. In aspen composites, longer fibers (mesh size 20) produced a higher yield stress and fracture energy. But it was

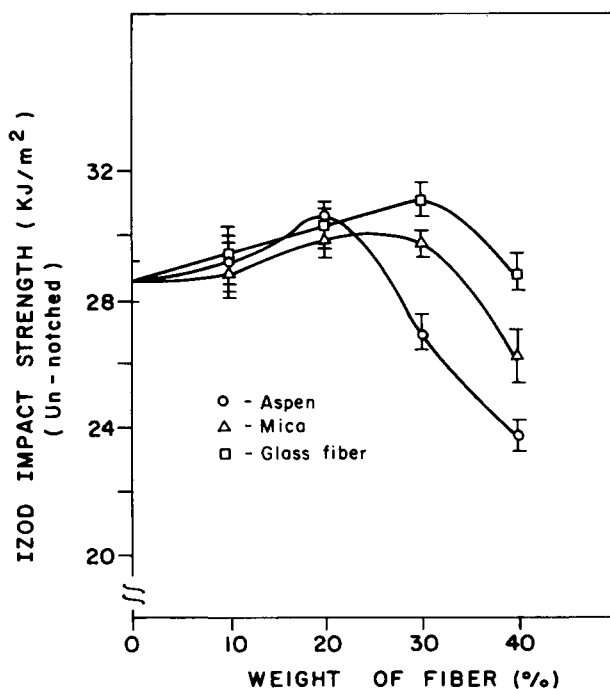


Fig. 5. Effect of filler level on impact strength of LLDPE-wood, mica or glass fiber composites: (○) aspen; (△) mica; (□) glass fiber.

TABLE VII  
Optimum Tensile Properties of Silane A-172- and PMPPIC-Treated  
Wood Fiber Composites (30% Fiber Weight)

Composite	Tensile strength (MPa)	Percentage increase	Tensile modulus (MPa)	Percentage increase
Aspen				
Silane A-172-treated <sup>a</sup>	15.9	45.8	786	141.8
PMPPIC-treated	17.2	57.8	772	137.5
Treated <sub>1</sub> composites <sup>b</sup>	18.0	65.1	701	115.7
Tempure 626				
PMPPIC-treated	18.7	71.6	743	128.6
Treated <sub>1</sub> composites <sup>b</sup>	19.9	82.6	661	103.4
Temalfa-A 6816				
Silane A-172-treated <sup>a</sup>	14.7	34.8	643	97.8
PMPPIC-treated	16.3	49.5	779	139.7
Treated <sub>1</sub> composites <sup>b</sup>	17.4	59.6	—	—
LLDPE	10.9	—	325	—

<sup>a</sup>Silane A-172-treated (4.0% by weight of fiber).

<sup>b</sup>Treated<sub>1</sub> composites (PMPPIC-pretreated LLDPE + PMPPIC-coated<sub>1</sub> fiber).

just the opposite in Tempure 626 and Temalfa-A 6816 composites, where the yield stress was high in mesh size 60 fiber composites. However, the results showed a consistent increase in modulus when longer fibers were used for reinforcement. Similar observation was made by Kokta et al.<sup>15</sup> in earlier studies on the composites of LLDPE with grafted aspen pulp.

Comparison of tensile properties of LLDPE reinforced with aspen, mica, and glass fiber is shown in Table VI. Yield stress decreased in mica and glass fiber composites at higher concentration of filler, while in aspen composites a gradual increase was observed. Elastic modulus was superior at 30.0% filler content in glass fiber composites, but the aspen composites had a higher modulus compared to mica composites. However, it should be remembered that wood fibers have lower density compared to glass fiber, which is a significant factor in the selection of material for applications which require a high strength to weight ratio.

Izod impact strength (unnotched) of LLDPE reinforced with different fibers is shown in Figure 5. Increase in impact strength was observed, until 20.0% filler content, in aspen and glass fiber composites. Further increase in fiber concentration resulted in a rapid drop in impact strength.

Table VII shows the optimum tensile properties of LLDPE filled with different wood fiber composites (30.0% fiber content). PMPPIC-coated aspen composites produced a 65.1% increase in tensile strength at yield. Higher elastic modulus was obtained in silane A-172-treated aspen composites. LLDPE filled with PMPPIC-treated Tempure 626 performed well compared to Temalfa-A 6816 composites. However, the elastic modulus was not much influenced by fiber treatment.

## CONCLUSIONS

Improvement in tensile strength and modulus was observed in LLDPE filled with silane-treated wood fiber composites. Yield stress increased signifi-

cantly when PMPPIC was applied with the fiber. The method of preparation and composition of wood fiber can affect the ultimate properties of the composite. Impact strength decreased at higher filler content in the composites. Mechanical properties of LLDPE filled with wood fiber compared favorably with mica and glass fiber composites.

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