

Performance of Treated Hybrid Fiber Reinforced-Thermoplastic Composites under Extreme Conditions.

IV. Use of Glass Fiber and Sawdust as Hybrid Fiber

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SYNOPSIS

The mechanical properties and dimensional stability of hardwood aspen in the form of sawdust and surface-treated glass fiber-polystyrene composites were evaluated under various extreme conditions, e.g., variation in the testing temperature (from +25° to -20°C), exposure to boiling water and heat in an oven at +105°C. The compatibility of wood fiber with glass fiber and with polystyrene improved by precoating the wood fiber with a coupling agent, e.g., 8% isocyanate, 4% silane and polymer. The mechanical properties of the composites, in particular, treated sawdust/glass fiber-filled composites, increased under extreme conditions in comparison with those filled with nontreated sawdust/glass fiber. Under the same conditions, dimensional stability also supports this observation.

INTRODUCTION

Polymer technologists and industry experts are constantly in search of some new polymeric substances with modified physical and mechanical properties.¹⁻⁴ As a result, significant developments are under way in advanced composites to fill different branches of engineering needs for lightweight, tougher and more reliable structures. The field of thermoplastics is the fastest growing one of all.⁵ For instance, by 1991, the demand for reinforced thermoplastics is expected to grow threefold as well as that for reinforced thermosets. Glass fiber accounts for about 93% of reinforced materials applications. Cellulose fibers can be compared in many respects to glass fibers, e.g., the specific properties⁶⁻⁸ (e.g., strength and modulus). Wood fibers have a lower specific gravity and cost⁹ compared to those of glass fibers (1.5 vs. 2.5, and 0.35 vs. ~ 3.00 c/in.,³ respectively). Wood fibers are relatively coarse and flexible in comparison with glass fibers.^{6,9,10} Moreover, cellulose fibers are easily renewable. Therefore,

a hybrid of glass and wood fibers results in a composite which is lighter and less inexpensive than comparable glass fiber reinforced thermoplastics. The compatibility problem between glass and wood fibers could be overcome by coating the fiber surface with coupling agents, e.g., silanes and isocyanate.¹¹⁻¹⁶ The hybridization of these treated fibers in polystyrene composites was examined by testing the mechanical properties of the composites. The mechanical performance of composite materials in their structural applications is often compromised by the uncertainty of the material behavior under various environmental conditions, e.g., exposure to water, change in temperature, and dimensional stability.¹⁷⁻³¹ Moreover, it is well known that during the summer, the filler, especially cellulose, slowly absorbs water, which upon crystallization in winter produces an adverse effect on mechanical behaviors.²⁴ The present study deals with the influence of different environmental conditions, e.g., exposure to boiling water at elevated temperature and subzero temperature for a given length of time, on the mechanical properties and dimensional stability of the hybrid surface coated glass and wood fiber-filled polystyrene composites. A comparison with the original polymer and untreated composites was also carried out.

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MATERIALS

Thermoplastics

High impact polystyrene (PS 525) was supplied by Polysar Ltd., Sarnia, Ontario, Canada.

Fillers

In this study, hardwood species, aspen (*Populus Tremuloides Michx*), was used in the form of sawdust. The wood chips for making woodflour were oven-dried by circulating air at 55°C for 48 h, and then ground to a mesh size 60 mixture: 60.5%, mesh 60; 20.2%, mesh 80; 15.5%, mesh 100; and 2.5%, mesh 200. The average length and diameter of the fibers were 0.30 and 0.022 mm, respectively (aspect ratio (l/d) = 13.9).

Glass fibers 731 BA 1/32 (0.8 mm, silane coated) were supplied by Fiber Glass of Canada via Mica Chemical, Montreal. The length of fibers varied from 0.75 to 0.86 mm, and the diameter from 0.014 to 0.0144 mm (l/d = 52 to 61.4).

Coupling Agent

Poly[methylene(polyphenyl isocyanate)] (PMPPIC) was supplied by Polysciences Inc. (U.S.A.). γ -Aminopropyltriethoxysilane (A-1100) was supplied by Union Carbide Co., Montreal, Canada.

EXPERIMENTAL

Isocyanate Coating

The wood flour, polymer (10 wt %) and isocyanate (8 wt %) were mixed¹⁵ with the help of a Laboratory Roll Mill (C. W. Brabender, Model No. 065) at 175°C. The mixtures were collected and mixed repeatedly (8–10 times) for homogeneous coating. Finally, the coated fibers were ground to mesh size 20.

Silane Coating

A mixture of oven-dried wood flour, CCl_4 (1 : 12 with respect to the weight of pulp), dicumyl peroxide (2%), and silane (4%) was heated under reflux at 70–75°C for 3 h under constant stirring with a magnetic stirrer. The mixture was allowed to cool to room temperature and, when the CCl_4 had evaporated, it was dried at 55°C for 24 h. Polystyrene (10%), *p*-xylene (4.3 times the weight of fiber), dicumyl peroxide (0.5%), and maleic anhydride (1%) were mixed thoroughly by agitating the mixture with

a magnetic stirrer at room temperature for 3 h; then the oven dried A-1100 coated fiber was poured onto it. The whole mixture was heated under reflux at 80–85°C for 2 h under constant agitation. The mixture was allowed to cool to room temperature and filtered in a glass funnel followed by washing with water. The mixture was then dried by prolonged heating at 55°C and ground to mesh 20.

Preparation of Composites

A 25 g amount of polymer and coated wood flour/glass fiber (25% by weight of composites) were mixed in a roll mill at 175°C. After mixing 5–10 times, the resulting mixtures were ground once again to mesh size 20. The mixtures were then molded (24 at a time) into shoulder-shaped test specimens (ASTM D 638, Type V). Standard molding conditions were: temperature, 175°C; pressure during heating and cooling, 3.8 MPa; heating time, 20 min; cooling time, 15 min.

Width and thickness of each specimen were measured with the help of a micrometer. For dimensional stability measurements, the weight of the samples was also taken at room temperature.

Environmental Exposure

In general, 30 specimens for each type of composite were prepared and divided into five different groups.

Boiling-Water Exposure

Two groups were immersed in boiling water for 24 h under atmospheric pressure. Samples were then dried between two sheets of filter paper and the weight as well as dimension of each specimen were measured.

Heat Exposure

One group was heated at a temperature above the T_g of polystyrene, e.g., 105°C, in an air-circulating oven for 5 days. After being cooled down and conditioned at room temperature, the weight and dimension of each specimen were measured.

Dimensional Stability Measurements

The percentage change in weights as well as cross section areas (width \times thickness) of the samples were measured at the room temperature after the respective treatments.

Mechanical Tests

Nontreated composites as well as those which underwent the above-mentioned treatments were equilibrated under ambient laboratory conditions (e.g., 25°C and 50% relative humidity). The mechanical properties (e.g., tensile modulus, tensile strength at yield point as well as the corresponding elongation and energy) of all the samples were measured with an Instron Tester (Model 4201) following ASTM D-638. A standard general Tensile Test Program method, called "PLA 10," was used, and the mechanical properties were automatically calculated by a HP-86B computer. The strain rate was 1.5 mm/min and tensile modulus was reported at 0.1% strain.

Mechanical properties were tested at room temperature or at subzero temperature. A part of the nontreated samples and a part of the samples which underwent boiling water exposure as well as heat exposed samples were tested at room temperature. The remaining nontreated and boiling water exposed

samples were kept at -20°C in the thermostatic Instron chamber (Model 311) for 2 h and then the mechanical properties were evaluated at that temperature.

A statistical average of at least five measurements was taken to obtain a reliable average and a standard deviation.

RESULTS

Table I shows the variation in the mechanical properties of sawdust aspen and coated glass fiber-polystyrene composites under ordinary conditions (i.e., room temperature) along with the compositions of sawdust and coated glass fiber, as well as filler level (15–35% by weight of composites) and coating treatments (e.g., coating of the sawdust with 8% PMPPIC, 4% silane A-1100, and polystyrene). Table I reveals that only the moduli of nontreated saw-

Table I Mechanical Properties of Glass Fiber/Sawdust-Filled Polystyrene 525 Composites at Room Temperature

Composition of Fibers		Yield Strength (MPa)			Elongation at Yield (%)			Energy (mJ)			Modulus (GPa)		
Sawdust	Glass (wt % of fiber):	15	25	35	15	25	35	15	25	35	15	25	35
				16.8 ^a			1.5 ^a			17.2 ^a			1.4 ^a
<u>Nontreated Sawdust (Aspen)</u>													
100	0	16.2	18.3	17.2	1.6	1.7	1.6	17.0	20.7	19.2	1.6	1.8	2.0
75	25	16.2	17.7	13.2	1.5	1.4	1.2	17.5	19.7	14.3	1.6	1.8	1.9
50	50	15.1	15.5	14.2	2.0	1.5	1.5	25.9	18.9	17.6	1.5	1.7	1.9
25	75	17.0	15.4	15.0	1.4	1.4	1.3	18.6	18.7	15.5	1.6	1.7	1.8
0	100	16.7	17.1	16.6	1.4	1.3	1.1	16.1	15.5	12.9	1.6	1.8	2.1
<u>Sawdust (Aspen) Coated with PMPPIC (8%)^b</u>													
100	0	20.5	23.0	22.5	2.7	3.1	2.3	53.9	43.8	39.7	1.2	1.4	1.4
75	25	18.5	21.5	18.7	2.2	2.6	1.7	34.8	56.7	27.6	1.5	1.7	1.7
50	50	19.5	17.7	17.5	1.4	1.4	1.4	23.9	18.3	17.0	1.7	1.8	2.2
25	75	18.3	18.8	18.3	1.7	1.7	1.4	25.7	27.8	19.1	1.6	1.7	1.9
<u>Sawdust (Aspen) Coated with Silane A-1100 (4%)^b</u>													
100	0	17.1	19.5	18.5	1.7	3.1	1.8	24.8	53.7	29.3	1.6	1.6	1.9
75	25	18.1	18.4	17.4	1.6	1.6	1.7	20.6	21.6	26.5	1.5	1.5	1.7
50	50	17.7	17.5	16.6	1.5	1.5	1.5	17.4	21.4	19.2	1.5	1.5	1.9
25	75	16.6	17.0	17.1	1.5	1.4	1.4	18.1	19.7	19.6	1.5	1.8	2.0

^a Only polymer.

^b By weight of sawdust.

Table II Comparison of the Mechanical Properties of Unfilled Polystyrene under Various Extreme Conditions

Treatment ^a	Yield Strength (MPa)	Elongation at Yield (%)	Energy (mJ)	Modulus (GPa)
A	16.8	1.5	17.2	1.4
B	16.9	1.7	19.8	1.3
C	14.7	1.8	19.7	1.3
D	29.1	2.1	79.6	1.3
E	22.1	5.1	109.7	1.0

^a A = testing at room temperature. B = testing at room temperature after boiling with water for 24 h. C = testing at room temperature after heating in an oven at 105°C for 5 days. D = testing at -20°C. E = testing at -20°C after boiling with water for 24 h.

dust/coated glass fiber-filled polystyrene composites are superior to that of unfilled polystyrene. Compared to the original polymer, the energy of the same composites improved in a few cases, but both strength and elongation were inferior. Modulus of both single wood fiber and glass fiber-filled composites were comparable, whereas other mechanical properties, e.g., strength, elongation, and energy, of wood fiber-filled composites were superior to those

of glass fiber-filled composites. Treated wood fiber/glass fiber-filled composites offered improved behavior in comparison with those of nontreated wood fiber/glass fiber-polystyrene composites, as far as strength, elongation, and energy were concerned. On the other hand, modulus did not improve. In all circumstances, modulus increased with rise in the total fiber level of the composites. Other properties increased along with the fiber content in the compos-

Table IIIA Comparison of the Improvement in the Mechanical Properties of Glass Fiber/Sawdust-Filled Polystyrene Composites under Various Extreme Conditions^a

Composition (wt %)		Improvement ^b % of Yield Strength					Improvement ^b % of Modulus				
Sawdust	Glass	A	B	C	D	E	A	B	C	D	E
<u>Nontreated Sawdust</u>											
100	0	+8.9	-30.2	+37.4	-7.6	+0.1	+35.7	-7.7	+46.2	+46.2	+40.0
75	25	+5.1	-21.8	+45.2	+4.5	+5.4	+22.7	+7.7	+38.5	+61.5	+40.0
50	50	-7.9	-17.2	+68.9	-0.9	+28.7	+22.7	+7.7	+50.8	+53.9	+100.0
25	75	-8.1	-16.6	+8.1	+2.4	+22.3	+21.4	+38.5	+15.4	+85.4	+110.0
0	100	+1.7	-13.0	+95.7	-2.5	+26.4	+28.6	+38.5	+76.9	+69.2	+110.0
<u>Sawdust Coated with PMPPIC (8%)^c</u>											
100	0	+36.9	+13.0	+63.3	+15.1	+18.3	0	0	-15.4	+38.5	+60.0
75	25	+28.0	-2.7	+26.3	+22.3	+24.5	+16.5	+15.4	+30.8	+61.5	+70.0
50	50	+5.4	-7.7	+1.4	+15.6	+15.1	+28.6	+15.4	0	+69.2	+70.0
25	75	+11.9	-7.1	+9.1	+18.0	+30.2	+17.5	+38.5	+46.2	+92.3	+90.0
<u>Sawdust Coated with Silane A-1100 (4%)^c</u>											
100	0	+16.1	-17.8	-22.7	+9.8	+11.0	+12.2	0	-30.8	+54.6	+40.0
75	25	+9.4	-15.1	+35.9	+6.5	+9.2	+4.3	+15.4	+46.2	+61.5	+60.0
50	50	+2.1	-6.9	+50.3	+8.9	+17.8	+7.1	+30.8	+53.9	+76.9	+70.0
25	75	+1.4	-4.7	—	+2.1	+16.9	+27.3	+30.8	—	+85.4	+70.0

^a A = testing at room temperature. B = testing at room temperature after boiling with water for 24 h. C = testing at room temperature after heating in an oven at 105°C for 5 days. D = testing at -20°C. E = testing at -20°C after boiling with water for 24 h.

^b Based on the original polymer after a similar treatment.

^c By weight of sawdust.

Table IIIB Comparison of the Improvement in the Mechanical Properties of Glass Fiber/Sawdust-Filled Polystyrene Composites under Various Extreme Conditions^a

Composition (wt %)		Improvement ^b % of Elongation at Yield					Improvement ^b % of Energy				
Sawdust	Glass	A	B	C	D	E	A	B	C	D	E
<u>Nontreated Sawdust</u>											
100	0	+13.3	+5.9	+5.6	0	-62.1	+20.4	-6.6	+40.6	-54.2	-75.3
75	25	-5.3	-7.1	-22.2	-8.1	-51.7	+14.7	-21.0	-6.1	-55.4	-64.9
50	50	+2.0	-17.7	-16.7	-6.7	-52.1	+9.8	-19.2	+17.8	-56.9	-57.6
25	75	-10.0	-20.6	-22.2	-3.3	-59.3	+8.4	-30.3	-20.1	-53.4	-66.2
0	100	-16.7	-11.8	-20.0	-13.3	-58.1	-9.9	-24.8	+19.8	-61.7	-66.4
<u>Sawdust Coated with PMPPIC (8%)^c</u>											
100	0	+106.7	+47.1	+61.1	+33.3	-50.7	+153.5	+89.4	+160.9	-24.4	-60.1
75	25	+75.3	+11.8	+11.1	+21.0	-52.7	+229.8	+18.6	+38.6	-29.3	-59.5
50	50	-8.7	+23.5	-1.1	+9.5	-48.7	+6.6	+27.8	-6.0	-38.9	-58.4
25	75	+14.7	-23.5	-27.8	+6.7	-52.7	+61.3	-29.3	-31.0	-39.9	-58.7
<u>Sawdust Coated with Silane A-1100 (4%)^c</u>											
100	0	+106.7	+2.4	-26.1	+9.1	-54.1	+212.2	-6.1	-52.3	-43.4	-65.8
75	25	+8.7	-5.9	-11.7	0	-61.3	+25.8	-16.7	+8.1	-53.3	-73.1
50	50	+2.0	-15.9	-26.1	+4.8	-57.1	+24.2	-19.7	-8.6	-47.3	-68.0
25	75	-8.0	-22.9	—	-13.3	-57.9	+14.3	-30.3	—	-58.3	-69.2

^a A = testing at room temperature. B = testing at room temperature after boiling with water for 24 h. C = testing at room temperature after heating in an oven at 105°C for 5 days. D = testing at -20°C. E = testing at -20°C after boiling with water for 24 h.

^b Based on the original polymer after a similar study.

^c By weight of sawdust.

ites at the initial level (e.g., up to 25 wt % of filler), and then decreased.

The dependence of the mechanical properties of polystyrene on the variation of different environmental conditions, e.g., from room temperature to boiling water temperature, +105°C in an oven and -20°C, is shown in Table II. Compared to ordinary conditions, strength increased when temperature decreased to -20°C. The same properties diminished due to exposure of the sample in an oven, whereas it remained constant when exposed to boiling water. On the other hand, both elongation and energy showed superior results under all extreme conditions, and maximum improvement was observed at -20°C. Furthermore, modulus dropped significantly under subzero conditions after being exposed to boiling water, whereas it did not change appreciably when other extreme conditions were considered. In fact, PS 525 is a copolymer of styrene and butadiene and as a result water might act as a plasticizing agent when the composites were exposed to boiling water.²⁸

The dependence of the mechanical properties of hybrid woodfiber/glass fiber-polystyrene compos-

ites under various extreme conditions is presented in Tables IIIA and IIIB. The improvement of the mechanical properties under different extreme conditions with respect to those of polymer under identical conditions is also shown in these tables. They reveal that most of the mechanical properties, except modulus of nontreated sawdust/glass fiber-filled composites and the strength of the heated specimens, deteriorated. As far as strength and modulus are concerned, surface coated sawdust/glass fiber-filled composites behaved better when composites were exposed to boiled water for 24 h. Moreover, both elongation and energy for PMPPIC coated sawdust/glass fiber-filled composites also improved. Unlike boiling water exposure, when samples were put in an oven at +105°C, only the mechanical properties of the composites filled with PMPPIC coated sawdust/glass fiber improved in most cases. Although elongation and energy of nontreated sawdust/glass fiber-filled composites provided inferior results due to heat exposure in an oven, the same properties improved when the proportion of coated sawdust in the composites exceeded those of glass

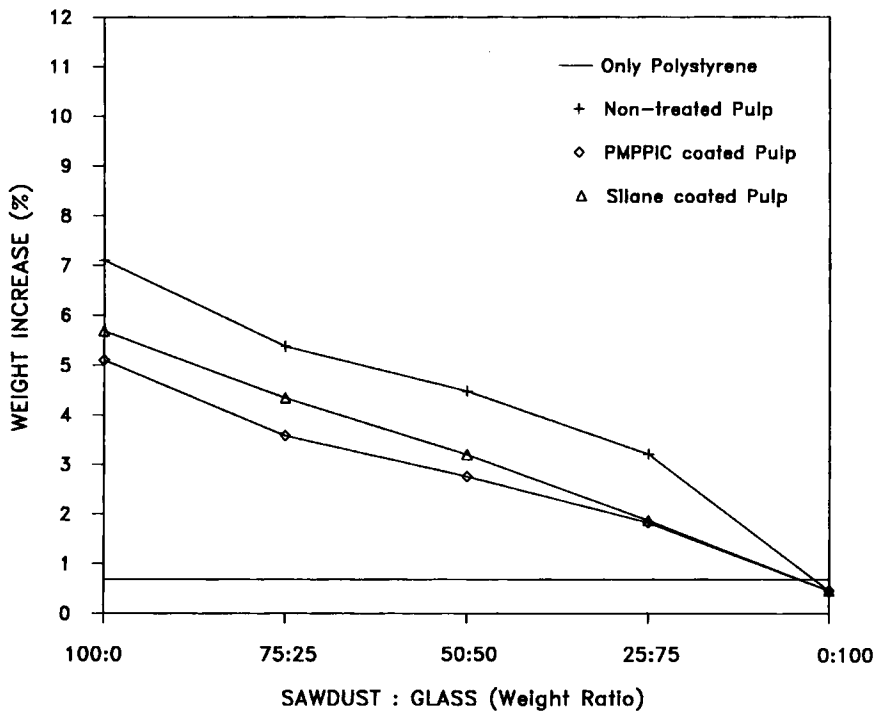


Figure 1 Variation of dimensional stability (change in weight) with the composition of sawdust aspen (nontreated and treated) and glass fiber in polystyrene composites. Concentration of filler: 25% by weight of composite.

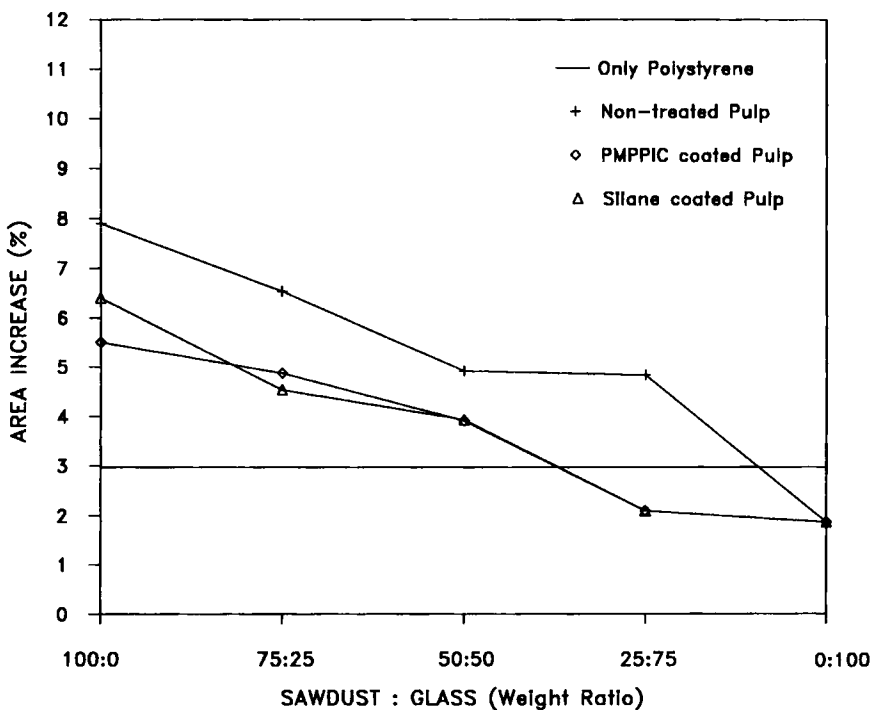


Figure 2 Variation of dimensional stability (change in area) with the composition of sawdust aspen (nontreated and treated) and glass fiber in polystyrene composites. Concentration of filler: 25% by weight of composite.

fibers. Both elongation, except the elongation for PMPPIC-coated sawdust/glass fiber-filled composites, and energy for all the composites under subzero conditions deteriorated, particularly for samples which were exposed to boiling water, while strength and modulus showed better results.

Dimensional stability was evaluated by measuring the change in weight and cross section area of the composites after being put in boiling water and in an oven at +105°C. Figures 1 and 2 represent the change in weight and area of composites when in boiling water. It is obvious from Figures 1 and 2 that a change in weight and area of the glass fiber-filled composites is even less than that of the original polymer. Both weight and area increased along with the rise in wood fiber content or the drop in glass fiber content in the composites. The nontreated wood fiber/glass fiber-filled composites showed the greatest increase in weight and area, whereas treated wood fiber/glass fiber showed the least increase in both weight and area. Therefore, the stability of coated wood fiber/glass fiber-filled composites is better than that of nontreated wood fiber/glass fiber-filled composites.

The variation in both weight and area of the composites according to exposure time in an oven is illustrated in Table IV. Cross-sectional area is positive in most cases, whereas weight is negative. The decrease in weight follows the increase of wood fiber content in the composites. Similar to exposure in hot water, treated wood fiber/glass fiber-filled composites are more stable compared to nontreated wood fiber/glass fiber-filled composites.

DISCUSSION

It is now clear from the above-mentioned results that different treatments provide better mechanical properties. In addition, extreme conditions are detrimental to composites comprising nontreated wood fiber alone or those which include glass fiber. Since wood fibers are hydrophilic in nature, they have a tendency to absorb water. As a result, nontreated wood fiber-filled composites showed a greater increase in weight and area. This increase led to deteriorated mechanical properties. Compared to untreated specimens, treated wood fiber/glass fiber-polystyrene composites produced superior results as far as the dimensional stability and mechanical properties, except the strength of the heated specimens, were concerned. Various surface treatments employed in the present study were previously reported^{15,28} as effective ones when wood fiber was

Table IV Effect of Heating in an Oven at 105°C for 5 Days on the Dimensional Stability of Glass Fiber/Sawdust-Filled Polystyrene Composites

Composition (wt %)		Change (%) in	
Sawdust	Glass	Weight	Cross Section Area
		-0.20 ^a	+2.29 ^a
<u>Nontreated Sawdust</u>			
100	0	-0.79	-1.10
75	25	-0.55	+2.52
50	50	-0.42	+3.70
25	75	-0.26	+2.92
0	100	-0.05	+2.70
<u>Sawdust Coated with PMPPIC (8%)^b</u>			
100	0	-0.75	-0.70
75	25	-0.68	+3.79
50	50	-0.27	+3.43
25	75	-0.15	+2.89
<u>Sawdust Coated with Silane A-1100 (8%)^b</u>			
100	0	-0.40	+4.90
75	25	-0.38	+3.95
50	50	-0.26	+2.05
25	75	-0.24	+1.11

^a Only polymer.

^b By weight of sawdust.

used as a single filler. For example, $-N=C=O$ groups of PMPPIC form covalent links with $-OH$ groups of cellulose while polystyrene interacts with PMPPIC through their common aromatic benzene rings. Moreover, when wood fibers were coated with PMPPIC and polystyrene, hydrophilicity of cellulose fibers diminished. Similarly, silane coating of wood fiber diminished hydrophilicity of cellulose fibers and increased the adhesion of fillers to polymer. Moreover, glass fibers were surface-treated with a silane coupling agent. This fact offers an unique opportunity for cellulose fibers to be compatible with polystyrene as well as with treated glass. Thus, both dimensional stability and the mechanical properties of the coated wood fiber/glass fiber-filled composites improved.

When the composites were put in an oven at +105°C, mechanical properties improved in many cases. This can be explained by an annealing of thermopressed composites.²⁴ In most cases, except for single wood fiber-filled composites, cross-sectional areas increased. This can be explained by the

difference in softening temperatures for polymers which is 99°C and exposed temperature which is 105°C, as well as the additional process of annealing of the thermopressed composites.²⁴ Moreover, it is well known that there are considerable differences in thermal coefficients³² among cellulose, polystyrene, and glass fiber. Due to the deformation of the specimens, some inaccuracies also took place during the dimension measurements. Once again, the weight of single wood fiber-filled composites decreased along with an increase in the wood fiber content of the composites. Although the components of cellulose are supposed to remain stable²⁶ at up to 180°C, the decomposition reaction could be initiated to a small extent^{25,27,33} when the processing temperature of the composites reaches 170–175°C. Moreover, exposure to heat for a longer period of time accelerates decomposition. As a result, loss in weight might be explained by the elimination of low molecular weight volatile materials. Moreover, the negative areas of the composites filled with only untreated or PMPPIC-coated wood fiber and the positive area of silane-coated wood fiber-filled composites might be due to the decomposition of cellulose as well as the deformation of the specimens.

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