Thermoplastic Composites of Polystyrene: Effect of Different Wood Species on Mechanical Properties

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Synopsis

Both softwood (spruce) and hardwood (aspen and birch) species in the form of different pulps (e.g., sawdust, chemithermomechanical pulp, explosion pulp and OPCO pulp) have been used (10–40 wt% composite) as reinforcing fillers for thermoplastic composites of polystyrene. Mechanical properties, are examined, e.g., tensile modulus, tensile strength at maximum point, and the corresponding elongation and energy as well as impact strength of compression molded composites. To improve the compatability of wood fibers which are hydrophilic and the polymer matrix which is hydrophobic, poly[methylene(polyphenyl isocyanate] (2 and 8 wt% of polymer) was used as a coupling agent. The mechanical properties of the treated composites are improved up to 30% in fiber content whereas a downward trend for untreated composites was observed when an increase in fiber content occurred. The overall improvements in mechanical properties due to the addition of isocyanate can be explained by the linkage of isocyanate molecules with fiber matrix through the chain of covalent bonds and the interaction of π-electrons of benzene rings of polystyrene as well as isocyanate. As a result, poly[methylene(polyphenyl isocyanate)] forms a bridge between fiber and polymer on the interfaces. This result is instrumental for efficient stress transfer between cellulose fibers and thermoplastics. The performance of different pulps of various wood species as reinforcing fillers for thermoplastic composites is also examined.

INTRODUCTION

The study of thermoplastic composites comprising cellulosics has been acknowledged as an important area of research for over a decade. In fact, the oil crisis of the 1970s has been directed toward the growing interest in the utilization of "biomass" as a primary source of materials[1] rather than minerals. Apart from being relatively inexpensive and a renewable source, cellulosics offer other potential advantages,[2–4] e.g., low density, flexibility, rough surface, reduced wear of the processing machinery, etc. Moreover, considering the extensive intermolecular hydrogen-bonding system, cellulose seems to be more closely related to thermoplastics rather than other types of polymers.[1] Cellulosics,[5–7] including wood fibers, wood flour, and other agrowastes, e.g., bagasse, corn cobs, rice hulls, bamboo, cereal straws, peanut shells, coconut shells, and walnut shells, can be used as a filler for thermoplastics. The performance of cellulosics as a reinforcing filler depends on its origin and the quality of the pulp.

Unfortunately, a wide polarity difference between cellulose and nonpolar thermoplastics, e.g., polyethylene, polypropylene, polystyrene, etc., prevents cellulose from performing as an efficient reinforcing filler. The problem of

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compatability between cellulose fibers and polystyrene might be overcome by selecting suitable coupling agents.[8-12]

A review of the literature shows that a comparative study of mechanical properties of thermoplastic composites containing different wood species in the form of various pulps is limited.[3,12-15] Kokta et al. extensively studied the mechanical behavior of composites of different thermoplastics (e.g., polyethylene,[13] polystyrene,[12,14] and polymethyl methacrylate[3]) and different wood species, e.g., hardwood (birch and aspen) and softwood (spruce and fir) in the form of chemithermomechanical pulp, mechanical pulp, and wood flour. They used both nontreated fibers and fibers grafted with vinyl monomers of corresponding thermoplastics. Their results can be summarized as follows. Both ungrafted and grafted aspen fibers revealed slightly better reinforcing properties compared to either birch or spruce/fir fibers. They attempted to correlate their results with the internal structure and aspect ratio of different wood fibers. Woodhams et al.[15] used softwood pulps including kraft (bleached and nonbleached), mechanical and chemical mechanical pulps, hardwood pulps, waste pulps, and reclaim newspaper pulp to make thermoplastic composites of high density polyethylene or isotactic polypropylene with the aid of carboxylic dispersing agents. They reported that polypropylene containing 40% each of hardwood pulp and newspaper reclaim produced inferior strength compared to kraft and TMP pulp in either polyolefin resin system. According to them, shorter fiber lengths (≤ 1 mm) of hardwood pulp and newspaper reclaim might be the cause of the reduction in strength.

In the present study, both softwood (spruce) and hardwood (aspen and birch) in the form of sawdust, chemithermomechanical pulp, explosion pulp, and OPCO pulps have been used as a filler for thermoplastics (polystyrene). From our earlier investigations,[11,12] poly[methylene(polyphenyl isocyanate)] appears to be the best coupling agent compared to silanes and even to other isocyanates. As a result, to study the performance of different fibers in the presence of a coupling agent, poly[methylene(polyphenyl isocyanate)] (2 and 8 wt% polymer) has been used. The mechanical properties of the composites under study have been evaluated.

MATERIALS

Thermoplastic

Two different varieties of polystyrene used in these experiments are as follows:

I. Heat resistant crystal polystyrene (Polysar Polystyrene 201)
II. High impact polystyrene (Polysar Polystyrene 525)

supplied by Polysar Ltd., Sarnia, Ontario, Canada. The physical properties of these polymers as provided by the supplier appear in Table I. The polymers were ground to a mesh size of -20 before mixing with the fibers.

Filler

In the present study, three different varieties of wood species, e.g., hardwood species, white birch (Betula Papyrefera Marsh), aspen (Populus Tremu-

TABLE I
Physical Properties of Polystyrenes

Property	PS201	PS525
Tensile strength at yield point (MPa)	52.0	24.0
Elongation at failure (%)	3.0	50.0
Tensile modulus (MPa)	3170.0	2480.0
Vicat softening point (°C)	106.0	99.0
Melt flow rate (g/10 min)	1.6	3.0
Specific gravity	1.05	1.04

loides Michx), and a softwood species mixture (75% black spruce, 20% balsam, and 5% aspen) were used in the form of chemithermomechanical pulp (CTMP), explosion pulp (V-pulp), Ontario Paper Company's pulp (POCO-pulp), and sawdust.

Coupling Agent

Poly[methylene(polyphenyl isocyanate)] (PMPPIC), supplied by Poly Science Inc., was used as coupling agent. The coupling agent was stored in a refrigerator and used as supplied by the manufacturer. All other chemicals in this study were of an analytical grade and were used without further purification.

EXPERIMENTAL

Preparation of Filler

(i) *Chemithermomechanical pulp (CTMP)* of aspen was prepared under the conditions described in Table II. The properties of these pulps are also listed in same table.

(ii) *Explosion pulp (V pulp)* of all the three wood species were prepared by an explosion technique: Na_2SO_3, 8%; immersion time, 24 h; immersion temperature, 60°C; explosion time, 4 min and explosion temperature 190°C. The exploded chips were washed thoroughly with water and then refined in a blender for 1 min at high speed.

TABLE II
Conditions of Preparation and Properties of CTMP Aspen Pulp

Cooking conditions		Properties	
Temperature max (°C)	126.0	Drainage index, CFS (mL)	119.0
Retention time (min)	5.0	Brightness, Elrepho (%)	60.9
Pressure (psig)	20.0	Opacity (%)	91.4
Na_2SO_3 (%/dry wood)	5.0	Breaking length (km)	4.46
NaOH (%/dry wood)	5.0	Elongation (%)	1.79
pH	12.9	Tear index (mN m^2/g)	7.2
Consistence during		Burst index (kPa m^2/g)	2.59
refining (%)	23.9		
Refining energy (MJ/kg)	5.15	Yield (%)	92.0
Refiner output (kg/h)	45.6	Kappa index no.	121.7
		Lignin (%)	17.9

(iii) Ontario Paper Company's pulp (OPCO-pulp) was supplied by the same
      company.

All these pulps and wood chips used to make sawdust were dried in a
circulating air oven at 55°C and then ground to mesh size 60 by a Granu
Grinder, C. W. Brabender Instruments, Inc.

Coating Treatment

Polymer (5 and 10% by weight of fiber) and PMPPIC (4 and 8% by weight
of fiber) were mixed with fiber with the help of a roll mill C. W. Brabender
Laboratory Prep. Mill, Model no. 065, at 175°C. The mixtures were collected
and mixed repeatedly 8–10 times for homogeneous coating. Finally, the coated
fibers were reground to mesh size 20.

Compounding

Mixture of polymer, pulp/coated pulp (10–40% by weight of composite),
and isocyanate (in the case of isocyanate-treated composites; 2 and 8% by
weight of polymer) were mixed with a roll mill at 175°C. The products were
mixed repeatedly five to six times. The resulting mixtures were allowed to cool
at room temperature and then ground to −20 mesh.

Compression Molding

The composites were molded into shoulder-shaped specimens in a mold
which was enclosed by metal plates on both sides. The mold was heated at
170°C in a Carver Laboratory press under a pressure of 3.8 MPa for 20 min,
followed by cooling under the same pressure for 15 min. The approximate
weight and dimension of each specimen were: weight 0.9 g (0.8 g after being
trimmed); width 0.31–0.33 cm; thickness 0.15–0.17 cm; length 6.4 cm (1.7 cm
between grips).

Mechanical Testing

The mechanical properties (tensile modulus, ultimate tensile strength, and
the corresponding elongation at break and fracture energy) of the samples
were measured with an Instron Tester (Model 4201). All these properties were
automatically calculated by computer using the Instron 2412005 general
tensile test program (under the name "PLA"). The strain rate was
1.5 mm/min and tensile modulus was reported at 0.1% strain.

The impact strength (Izod, unnotched) was tested with an Impact Tester
(Model TMI, NO 43-01), supplied by Testing Machines Inc. The samples were
tested after conditioning at 23 ± 0.5°C and 50% RH for at least 18 h in a
controlled atmosphere. Dimensions of all specimens were measured with a
micrometer. Statistical averages of five to six measurements were taken for
each sample. Average coefficients of variation were taken into account for each
set of tests (2.5–8.5%).

RESULTS AND DISCUSSION

Table III shows a comparative study of the mechanical properties of non-treated composites of PS201 and different wood species [e.g., hardwood (aspen and birch) and softwood (spruce)] in the form of various pulps (e.g., sawdust, V-pulp, OPCO-pulp, and CTMP). Results of a similar study using high impact polystyrene (PS525) are shown in Figures 1–4. Compared to the original polystyrene (Table III), tensile strength increases to 20% of fiber concentration, but only when softwood spruce (all forms of fibers) or hardwood aspen (CTMP) are used. The same table shows that in almost all experiments the elongation decreased but modulus increased with increasing fiber concentration.

The ultimate elongation of the composites is less than that of the unfilled polymer. On the other hand, energy to fracture is different for each of the wood species, e.g., for softwood spruce fiber filled composites, it increased 10%, and 30% for sawdust or V-pulp or OPCO-pulp. For hardwood aspen CTMP, it increases 20%. In the case of hardwood birch, the energy to fracture increased to 10% sawdust and 20% V-pulp.

Figure 1 shows that, except for a few cases (particularly for sawdust-filled composites), tensile strength increased to a maximum at 30% fiber concentration. Elongation and fracture energy (Figs. 2 and 3) of softwood spruce (OPCO-pulp) increased to 30% fiber concentration. Both these mechanical properties (strength and fracture toughness) show negligible increases compared to the unfilled polymer. In a similar fashion to PS201, composite modulus based on PS525 increased linearly with increasing fiber concentration.

In general, it is obvious from these tables and figures that the mechanical properties do not increase substantially compared to the unfilled polymer. This can be attributed to the fact that adhesion of fibers with polymer in nontreated composites is not at all sufficient to develop efficient stress transfer at the fiber interface which is essential to achieve a composite with increased mechanical strength.

To compare the mechanical properties of the composites (as observed in earlier table and figures), a 2% PMPPIC, which is recognized as a good coupling agent, was used. Table IV presents the mechanical properties of 2% PMPPIC-treated composites of PS201 and different wood pulps. Mechanical properties of composites containing PS525 and similar fibers appear in Figures 5–8. Except for PS201 sawdust-filled composites (Table IV), where it is seen that the tensile strength increases to a maximum at 30% fibers concentration, it even increases (Table IV and Fig. 5) to a maximum a 40% fiber concentration. But the maximum level of improvements appear on nearly 20–30% of fiber loading. In most cases, maximum improvements in ultimate elongation (Table IV and Fig. 6) occur in nearly 10% of fibers, but in a few cases it also increases up to 20% or more of fiber content. While Table IV and Fig. 7 indicate that fracture energy in some cases improves up to 30% or more showing the maximum at 10–20% of fiber level. Modulus in all cases increased proportionately as with the untreated wood fibers.

The enhancement of mechanical properties of the 2% PMPPIC-treated composites compared to untreated as well as the original thermoplastics

TABLE III

Effect of Wood Species and Nature of Pulps on the Mechanical Properties of Nontreated Composites of Polystyrene PS201

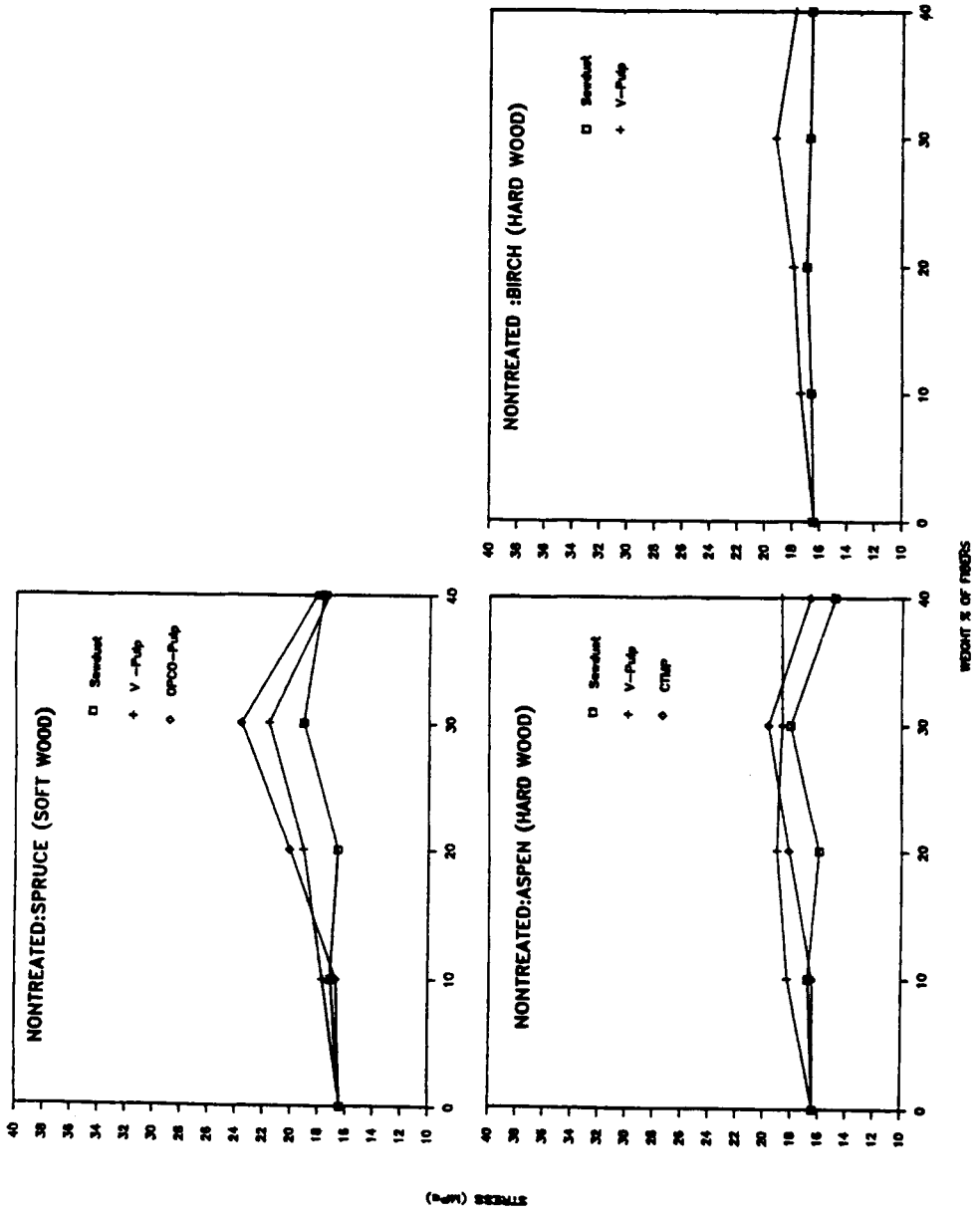| Composite (wt % of fiber)	Ultimate strength (MPa)				Ultimate elongation (%)				Fracture energy (J × 10³)				Flexural modulus (MPa × 10⁻²)			
	10	20	30	40	10	20	30	40	10	20	30	40	10	20	30	40
PS201		38.9					3.4				59.6			12.8		
(a) Sawdust																
PS201 + spruce (softwood)	38.6	39.3	37.4	31.1	3.1	2.8	2.6	2.4	67.6	56.1	50.4	43.3	17.0	18.8	19.9	21.1
PS201 + aspen (hardwood)	28.8	30.1	29.8	24.8	2.2	2.3	2.1	1.2	29.9	34.0	30.7	17.7	16.1	18.8	19.9	21.1
PS201 + birch (hardwood)	38.4	37.0	36.7	31.0	3.3	2.8	2.6	2.3	67.2	58.3	51.0	41.1	17.1	18.6	20.1	20.3
(b) V-pulp																
PS201 + spruce (softwood)	41.1	41.2	36.0	34.1	3.2	2.5	2.3	1.8	63.4	50.0	40.8	30.1	18.7	20.9	22.5	24.8
PS201 + aspen (hardwood)	37.4	34.2	34.9	30.7	2.7	2.9	2.5	1.9	52.6	55.9	49.7	30.7	18.9	19.3	21.2	24.6
kPS201 + birch (hardwood)	37.5	35.5	34.6	29.4	3.5	3.2	2.8	2.3	70.2	64.0	56.5	42.5	17.1	17.6	18.4	19.4
(c) Other pulp																
PS201 + CTMP (aspen)	38.0	40.3	33.1	31.6	3.2	3.1	2.7	2.3	62.2	62.8	45.6	36.1	18.1	19.4	20.3	22.7
PS201 + OPCO pulp (softwood)	38.4	35.4	42.5	34.6	3.3	2.7	2.9	2.3	65.8	52.5	65.2	38.6	18.2	19.4	21.9	23.3

Fig. 1.   Effect of fiber weight percentage on tensile strength for nontreated PS525 composites.
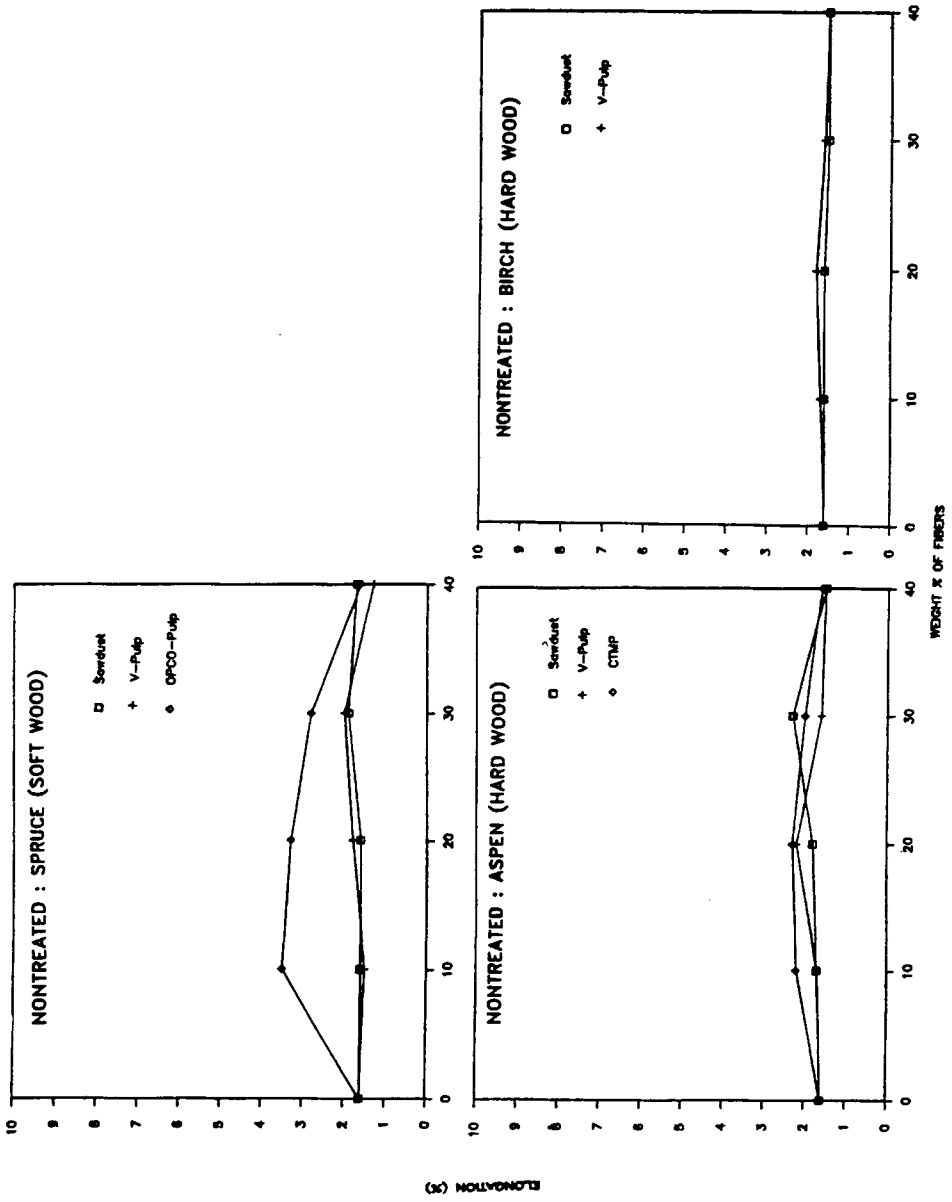
Fig. 2.   Effect of fiber weight percentage on ultimate elongation for nontreated PS525 composites.
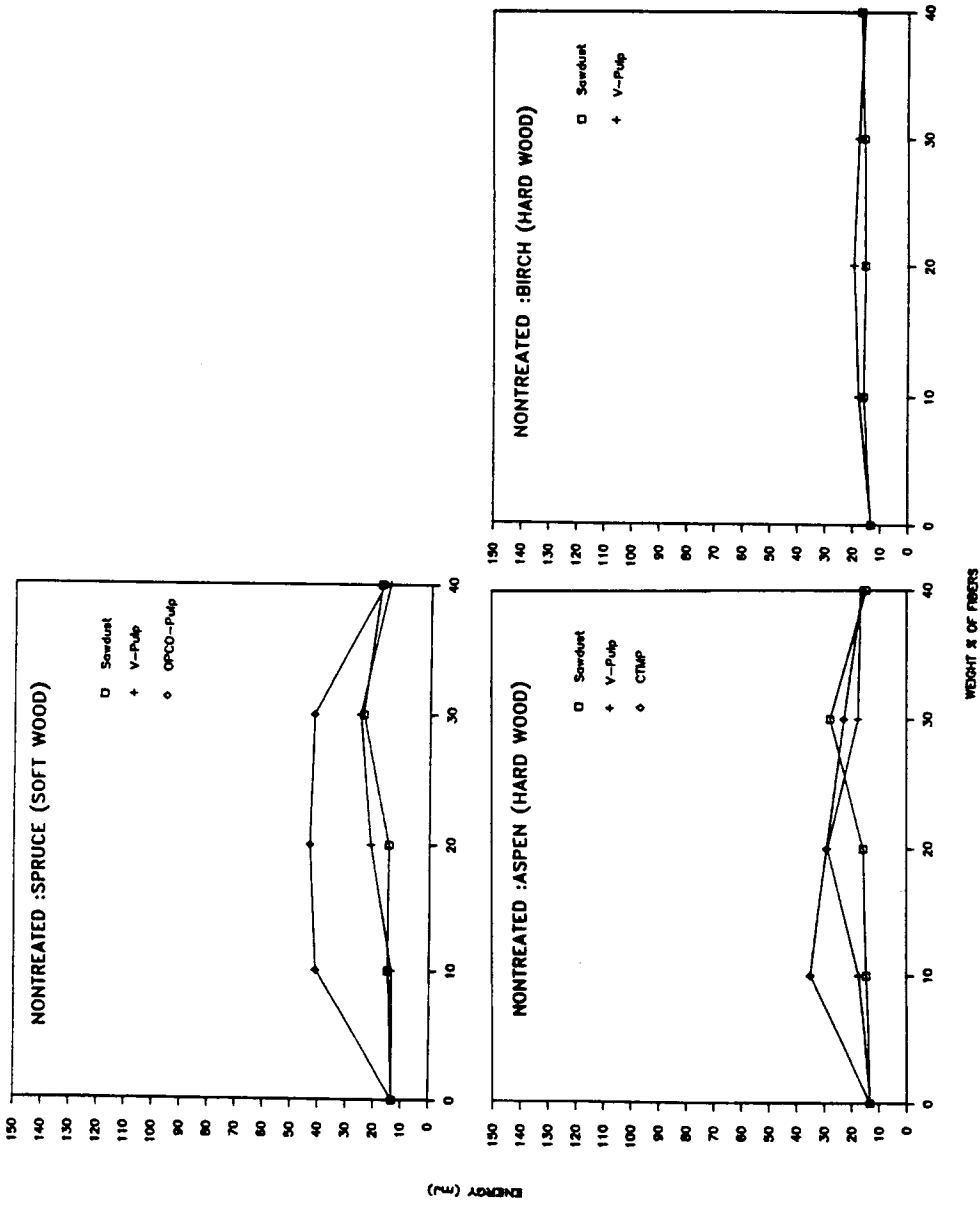
Fig. 3.   Effect of fiber weight percentage on fracture energy for nontreated PS525 composites.
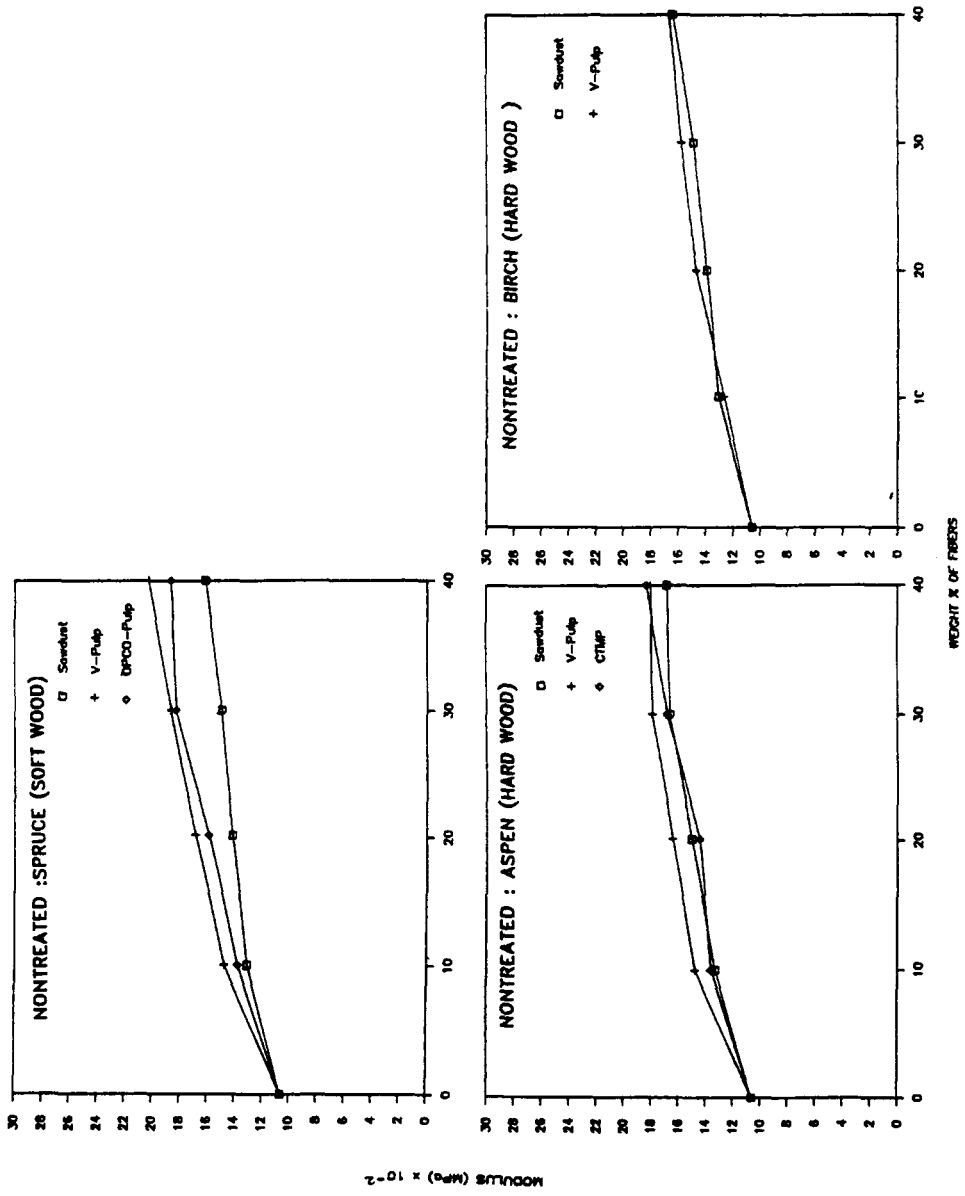
Fig. 4.   Effect of fiber weight percentage on flexural modulus for nontreated PS525 composites.

TABLE IV
Effect of Wood Species and Nature of Pulps on the Mechanical Properties of 2% PMPPIC-Treated Composites of Polystyrene PS201

| Composite (wt % of fiber)	Ultimate strength (MPa)				Ultimate elongation (%)				Fracture energy ($J \times 10^3$)				Flexural modulus ($MPa \times 10^{-2}$)			
	10	20	30	40	10	20	30	40	10	20	30	40	10	20	30	40
PS201		38.9				3.4				59.6				12.8		
(a) Sawdust																
PS201 + spruce (softwood)	41.2	40.6	40.3	38.3	3.7	3.5	3.1	2.4	82.0	76.1	63.0	44.6	17.1	19.2	20.5	21.0
PS201 + aspen (hardwood)	36.5	38.8	35.4	32.1	2.3	2.4	2.1	1.8	39.6	48.0	36.9	30.1	19.1	20.2	21.1	22.9
PS201 + birch (hardwood)	41.6	40.9	40.0	37.8	3.6	3.2	3.0	2.4	74.5	61.4	56.6	43.0	16.1	18.6	18.6	20.3
(b) V-pulp																
PS201 + spruce (softwood)	43.1	47.2	47.3	39.0	3.6	3.6	3.4	2.6	80.6	91.0	82.5	54.1	19.9	20.9	21.0	21.1
PS201 + aspen (hardwood)	40.2	42.6	39.9	39.7	3.3	3.2	2.9	2.7	69.4	71.1	55.2	53.1	16.5	16.8	18.0	18.7
PS201 + birch (hardwood)	41.6	43.1	44.6	35.7	3.8	3.8	3.4	2.8	81.3	84.5	77.8	52.3	16.8	19.0	20.7	21.4
(c) Other pulp																
PS201 + CTMP (aspen)	40.7	42.5	44.3	39.3	3.4	3.5	3.4	2.5	71.2	76.5	77.5	49.7	17.8	18.7	19.1	21.9
PS201 + OPCO pulp (softwood)	43.0	42.9	42.2	40.6	3.7	3.4	2.8	2.5	75.8	76.4	59.8	52.4	17.7	18.6	21.3	21.8
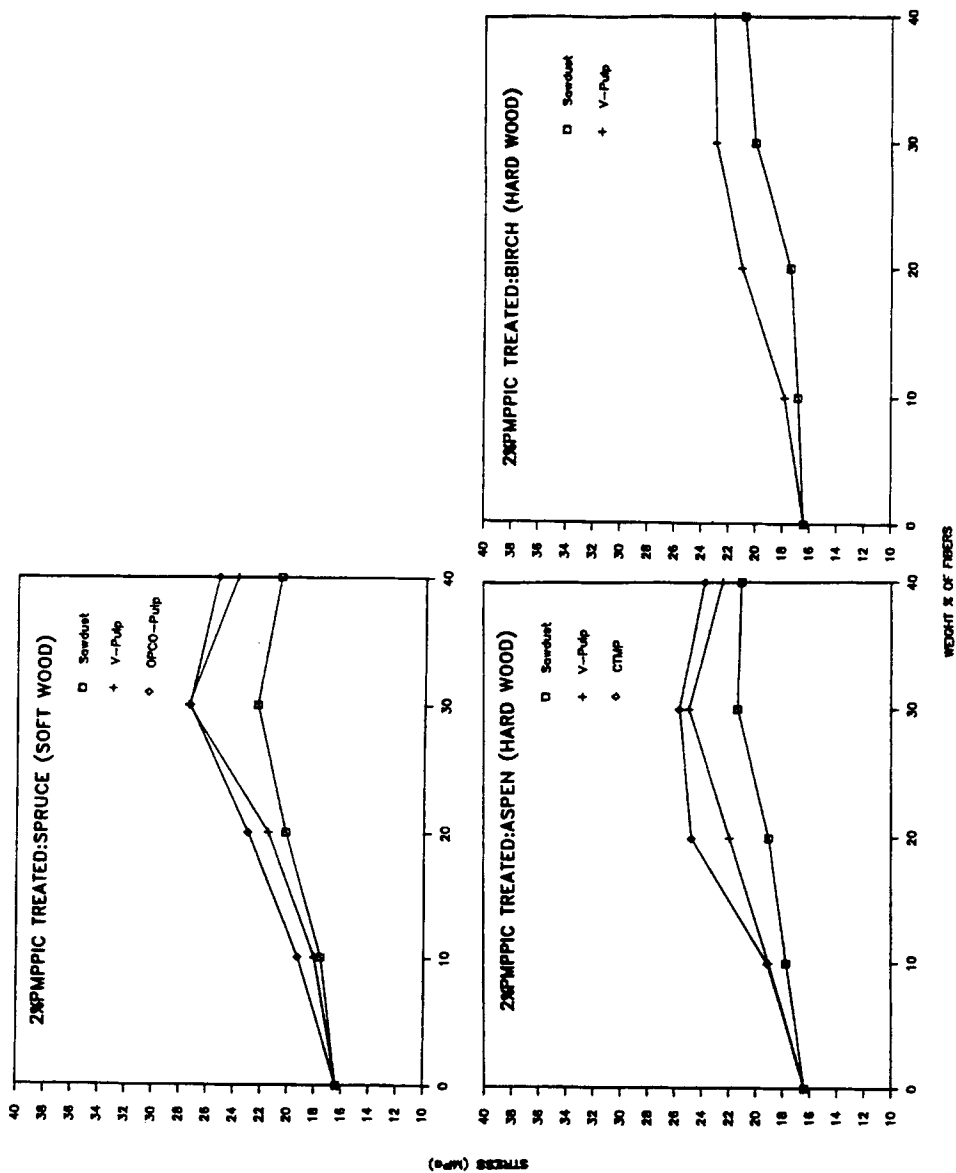
Fig. 5.   Effect of fiber weight percentage on tensile strength for PS525 composites containing 2% PMPPIC-treated fibers.
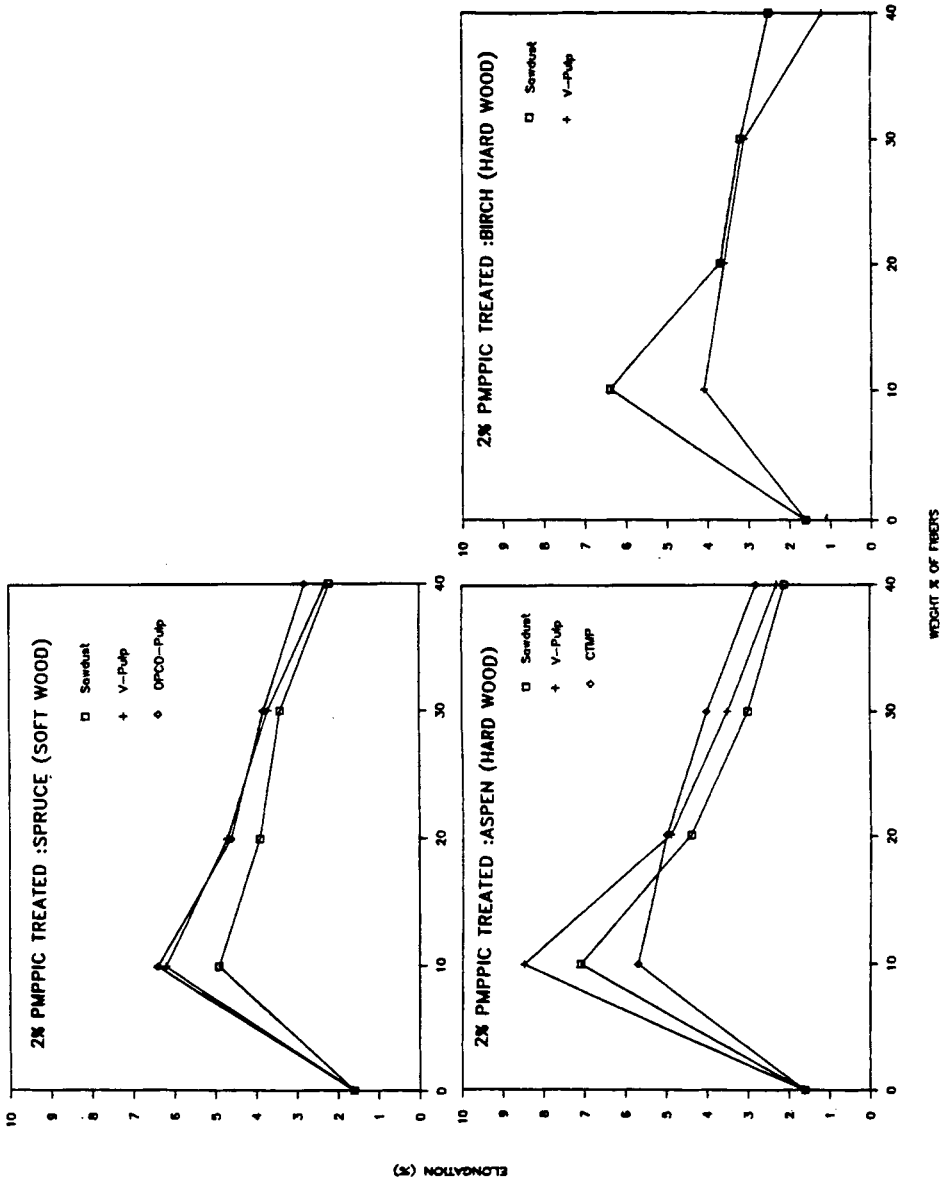
Fig. 6.   Effect of fiber weight percentage on ultimate elongation for PS525 composites containing 2% PMPPIC-treated fibers.
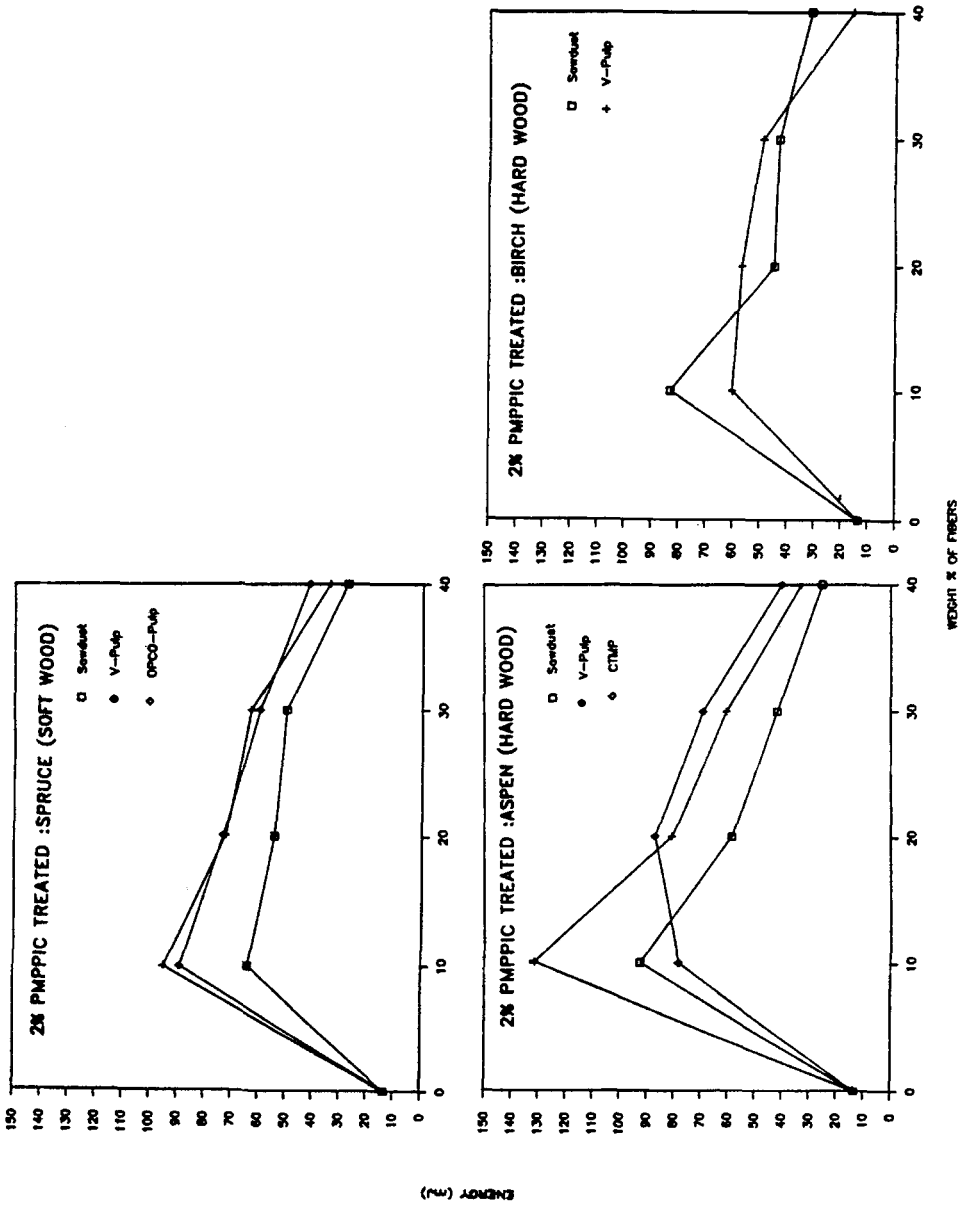
Fig. 7. Effect of fiber weight percentage on fracture energy for PS525 composites containing 2% PMPPIC-treated fibers.
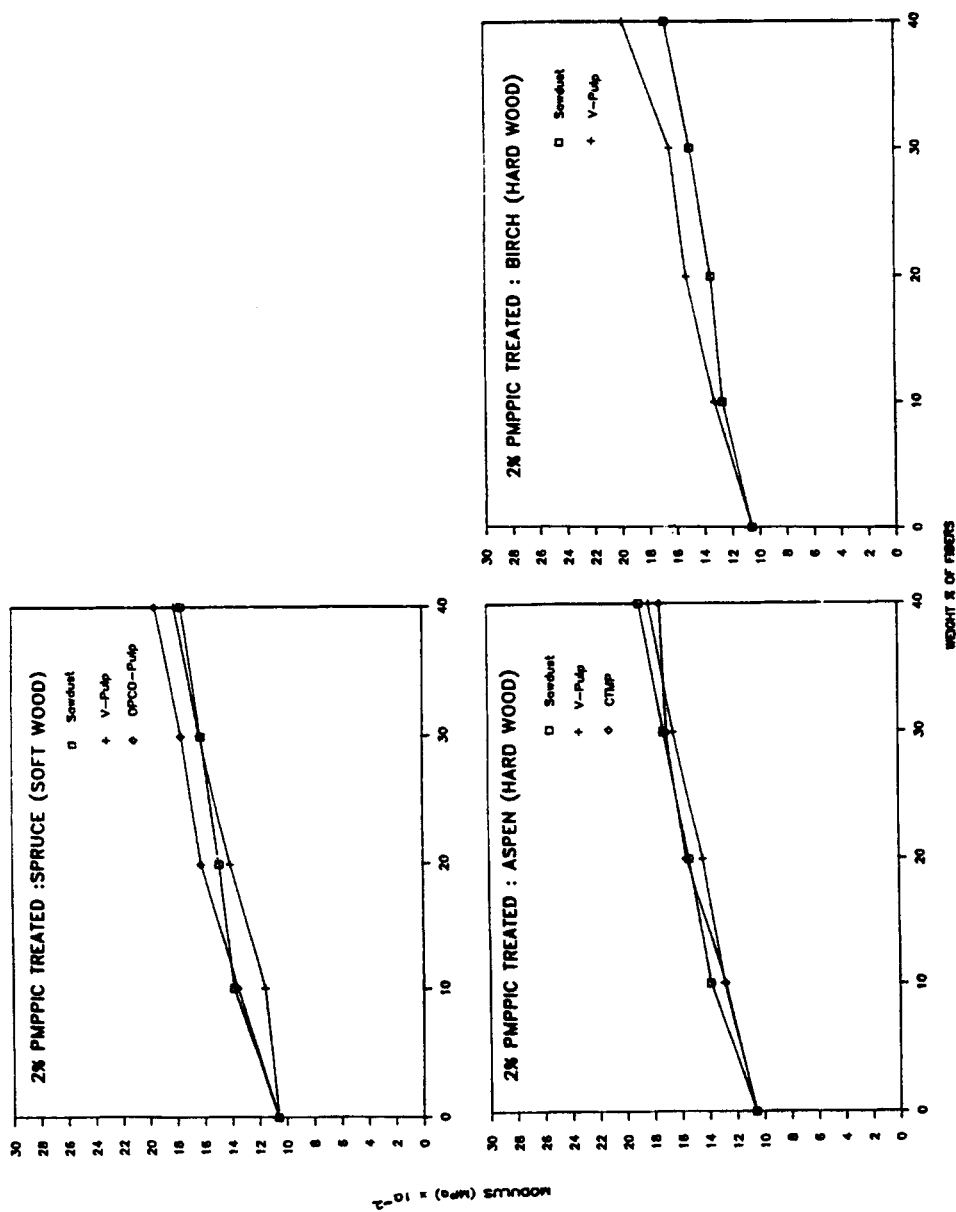
Fig. 8.   Effect of fiber weight percentage on flexural modulus for PS525 composites containing 2% PMPPIC-treated fibers.

clearly prove the important role of coupling agent PMPPIC in the formulation of composites. This result is consistent with our previous reports[11,12] concerning this coupling agent. We have also discussed in our earlier publications the actual chemical reasons underlying the improvements in mechanical properties of the composites in the presence of isocyanates. The active participation of functional group $-N=C=O$ of isocyanate forms a covalent bond with $-OH$ surface groups of the cellulosic fibers. In addition, the π-electrons of benzene rings of PMPPIC and polystyrene facilitate the formulation of strong adhesive bonds. As a result, PMPPIC completes the bridge between the polymer matrix and the cellulose matrix in the interphase region, giving an opportunity for mutual stress transfer between two physically immobilized phases.

To verify the accelerating effect of PMPPIC, a higher concentration, e.g., 8% (by weight of polymer), was used to make composites similar to other cases. The results are presented in Table V and Figures 9–12 for PS201 and PS525, respectively. It is obvious from this table that stress and energy of PS201-based composites in some cases improved up to 20–30% of fiber level. On the other hand, elongation decreased, while modulus increased with the rise in fiber content. The mechanical properties of the composites containing PS525 (shown in Figs. 9–12) behave similarly as discussed in the case of 2% PMPPIC-treated composites (see Figs. 5–8). The increase in mechanical properties is attributed to the effectiveness of PMPPIC as a coupling agent. To compare systematically the above results, Tables VI (A–C) list the increase in mechanical properties of nontreated and isocyanate-treated composites for two polymers (PS201 and PS525) with each of the wood species and different pulps. These tables show that mechanical properties have increased with an increase in the concentration of PMPPIC (from 0 to 2%) compared to nontreated composites. Unfortunately, the results of 8% PMPPIC-treated composites are inferior when compared to 2% PMPPIC-treated composites. This detrimental effect of a larger amount of PMPPIC is in good agreement with our previous observations.[11,12,16]

Perhaps a higher concentration of PMPPIC causes the formation of byproducts,[17] which might account for the decrease in mechanical properties.

If we compare the mechanical properties of PS201 composites and softwood spruce pulps (Table VIA), the V-pulp appears to produce superior composites whereas OPCO-pulp is only slightly better than sawdust. For the PS525 resin the mechanical properties of the composites are ranked in the following order: OPCO-pulp, V-pulp, and sawdust. When three different hardwood aspen pulps were compared (e.g., sawdust, V-pulp, and CTMP) (Table VIB) it was noted that both polymers the order of effectiveness decreases as follows: CTMP, V-pulp, and sawdust. From Table VIC, which shows the comparative study of sawdust and V-pulp of hardwood birch fibers including two polymers, V-pulp appears superior to sawdust. Different pulping techniques offer various ways to separate fibers from the wood chips[18-20] (e.g., sawdust is prepared by separating fibers mechanically, V-pulps by combination of chemical and physical methods while both chemical and mechanical methods are used for CTMP. Accordingly, surface quality and physical and mechanical properties of different pulps vary widely, e.g., CTMP provides higher specific surface area compared to mechanical pulps while sawdust is coarser than other pulps.

TABLE V
Effect of Wood Species and Nature of Pulps on the Mechanical Properties of 8% PMPPIC-Treated Composites of Polystyrene PS201

| Composite (wt % of fiber)	Ultimate strength (MPa)				Ultimate elongation (%)				Fracture energy (J × 10³)				Flexural modulus (MPa × 10⁻²)			
	10	20	30	40	10	20	30	40	10	20	30	40	10	20	30	40
PS201		38.9				3.4				59.6				12.8		
(a) Sawdust																
PS201 + spruce (softwood)	37.4	35.5	35.4	35.0	3.3	2.9	2.9	2.9	67.3	59.0	56.4	58.1	16.4	17.9	19.4	20.4
PS201 + aspen (hardwood)	36.0	38.6	37.8	33.8	3.2	3.2	2.9	2.8	63.0	66.5	58.0	56.4	16.3	17.9	18.5	21.2
PS201 + birch (hardwood)	36.9	37.5	39.4	40.8	3.2	3.2	3.3	3.2	60.1	61.9	65.7	66.5	17.3	18.4	18.6	19.8
(b) V-pulp																
PS201 + spruce (softwood)	40.1	42.4	43.3	38.1	2.9	2.9	2.7	2.8	62.3	65.6	63.3	55.5	18.5	21.2	22.6	22.6
PS201 + aspen (hardwood)	37.5	39.7	40.1	36.5	2.7	2.4	2.4	2.2	47.6	47.1	46.4	37.0	18.2	20.2	20.8	21.5
PS201 + birch (hardwood)	37.3	37.6	40.6	39.0	3.3	3.3	3.0	2.9	62.3	64.4	66.9	58.3	16.8	19.6	20.0	21.3
(c) Other pulp																
PS201 + CTMP (aspen)	37.1	43.5	39.5	33.9	2.6	3.1	2.7	2.2	46.0	53.5	53.3	41.3	18.8	20.2	20.8	22.3
PS201 + OPCO pulp (softwood)	38.5	38.7	37.9	38.2	2.5	2.7	2.5	2.3	42.6	50.0	46.8	42.7	19.5	19.5	21.2	21.8
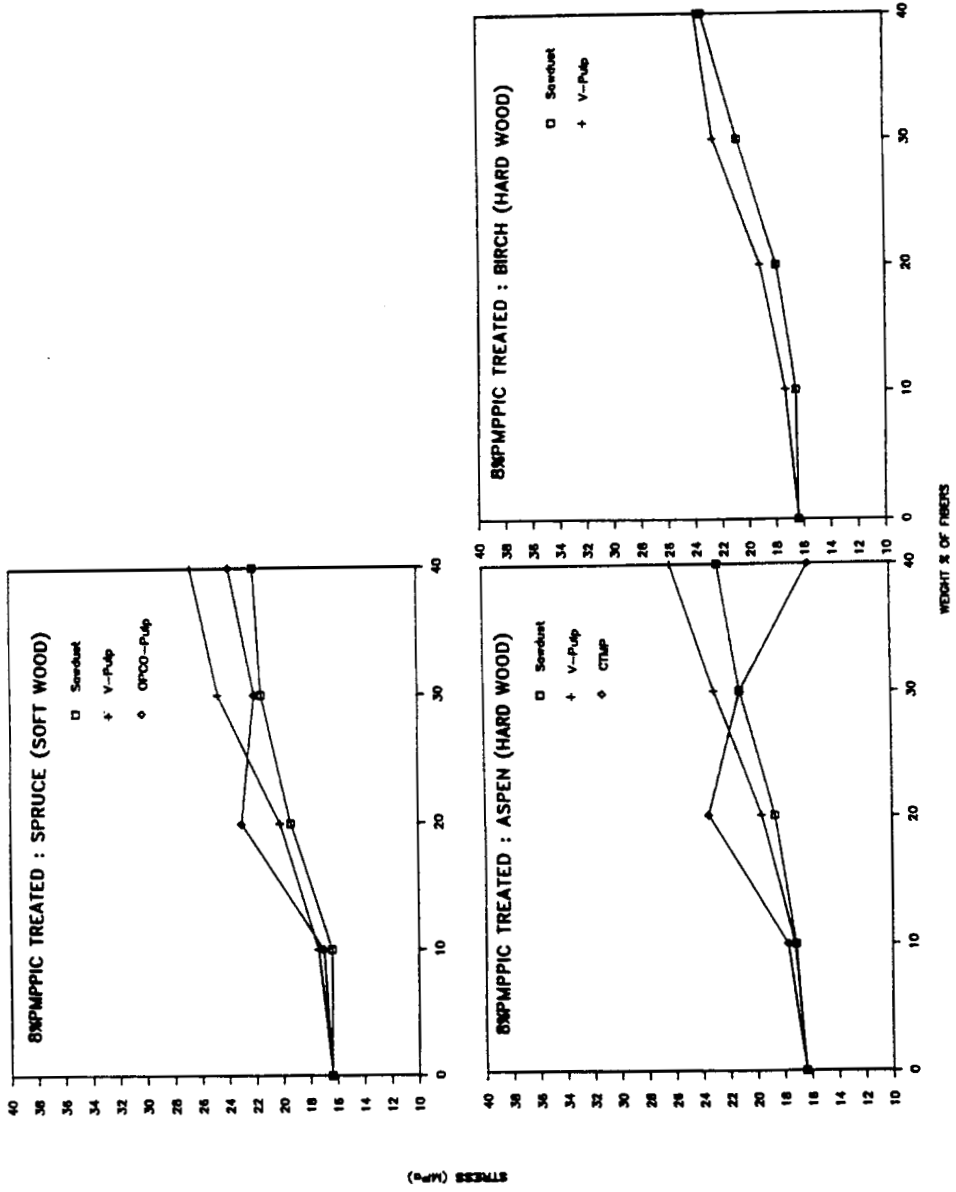
Fig. 9. Effect of fiber weight percentage on tensile strength for PS525 composites containing 8% PMPPIC-treated fibers.
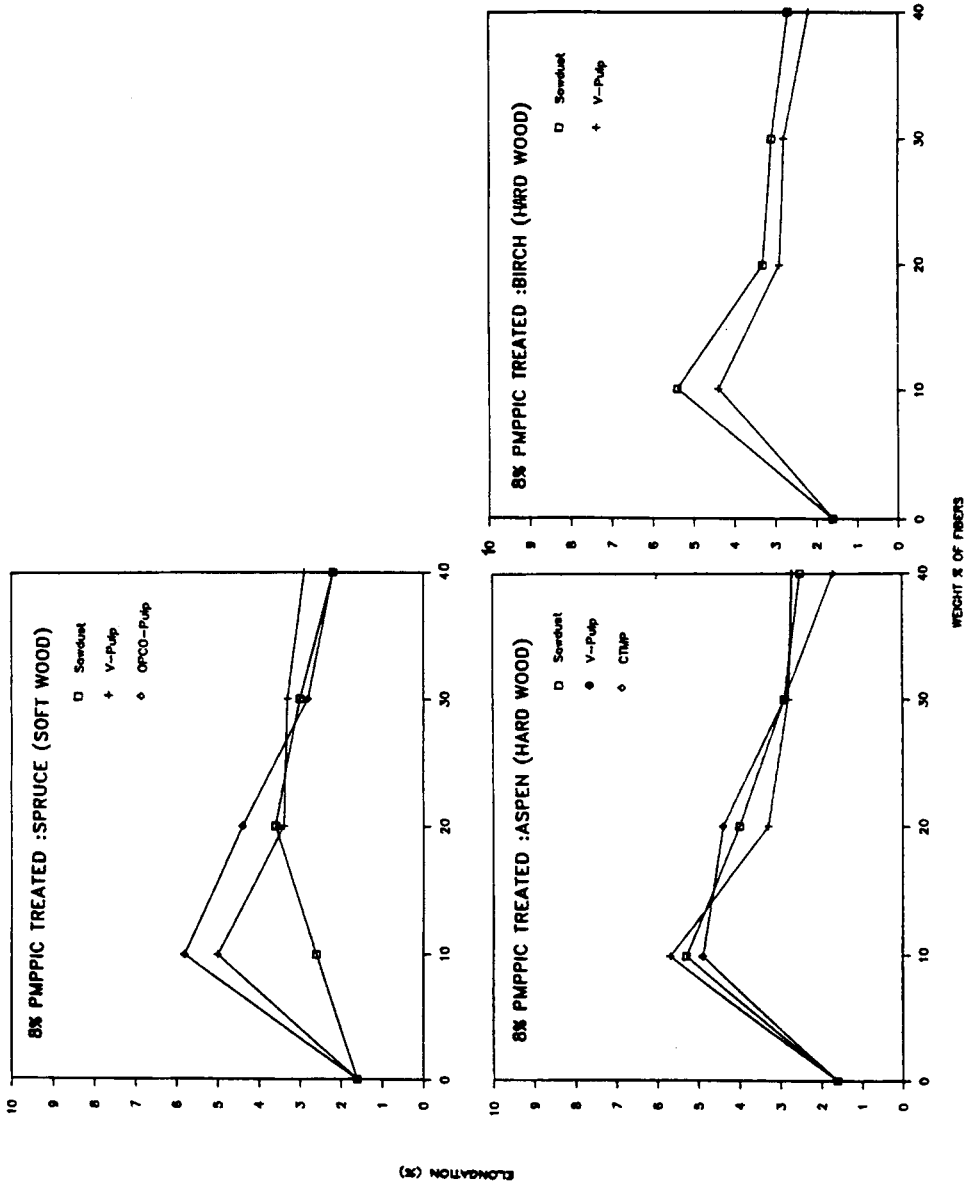
Fig. 10.   Effect of fiber weight percentage on ultimate elongation for PS525 composites containing 8% PMPPIC-treated fibers.
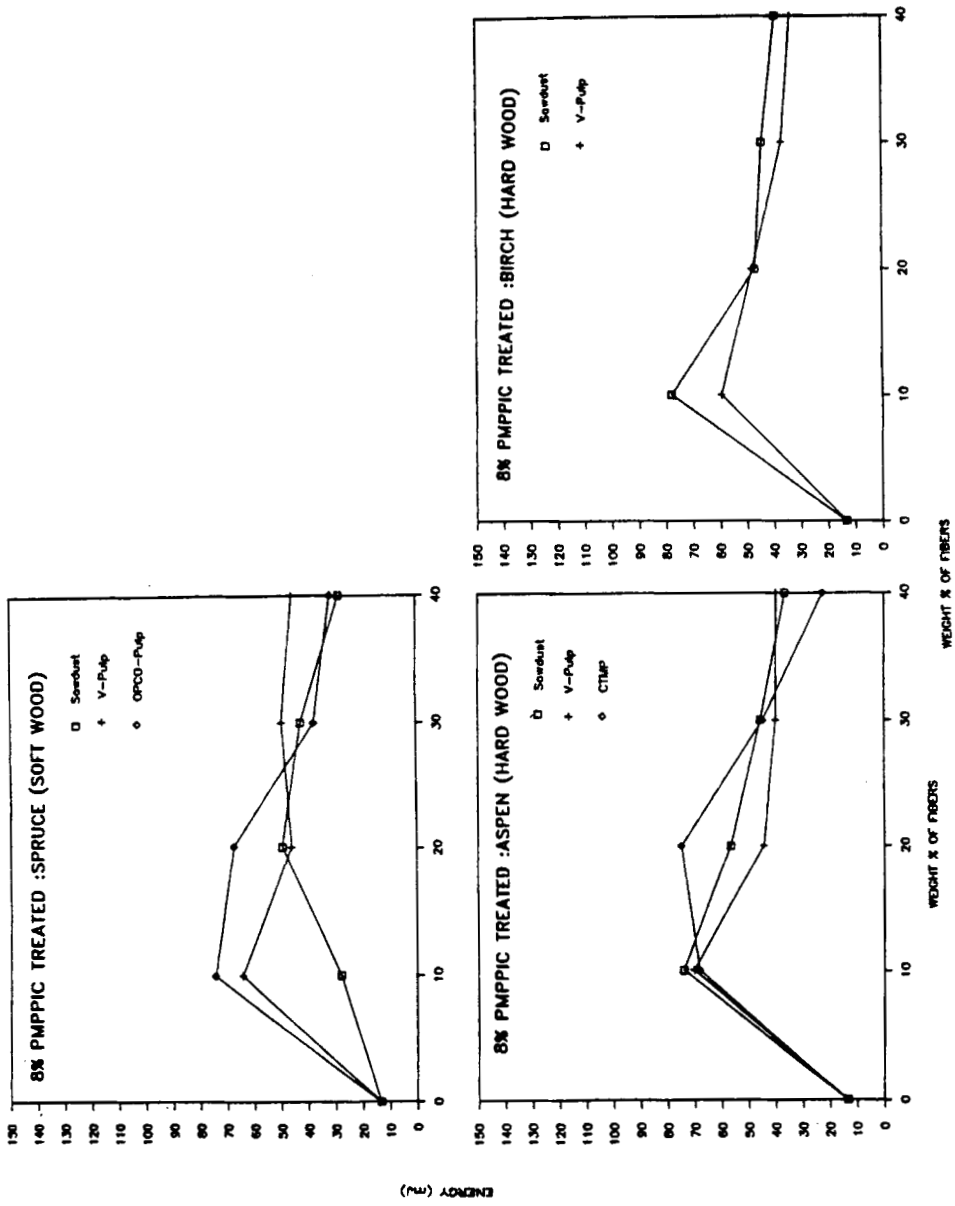
Fig. 11. Effect of fiber weight percentage on fracture energy for PS525 composites containing 8% PMPPIC-treated fibers.
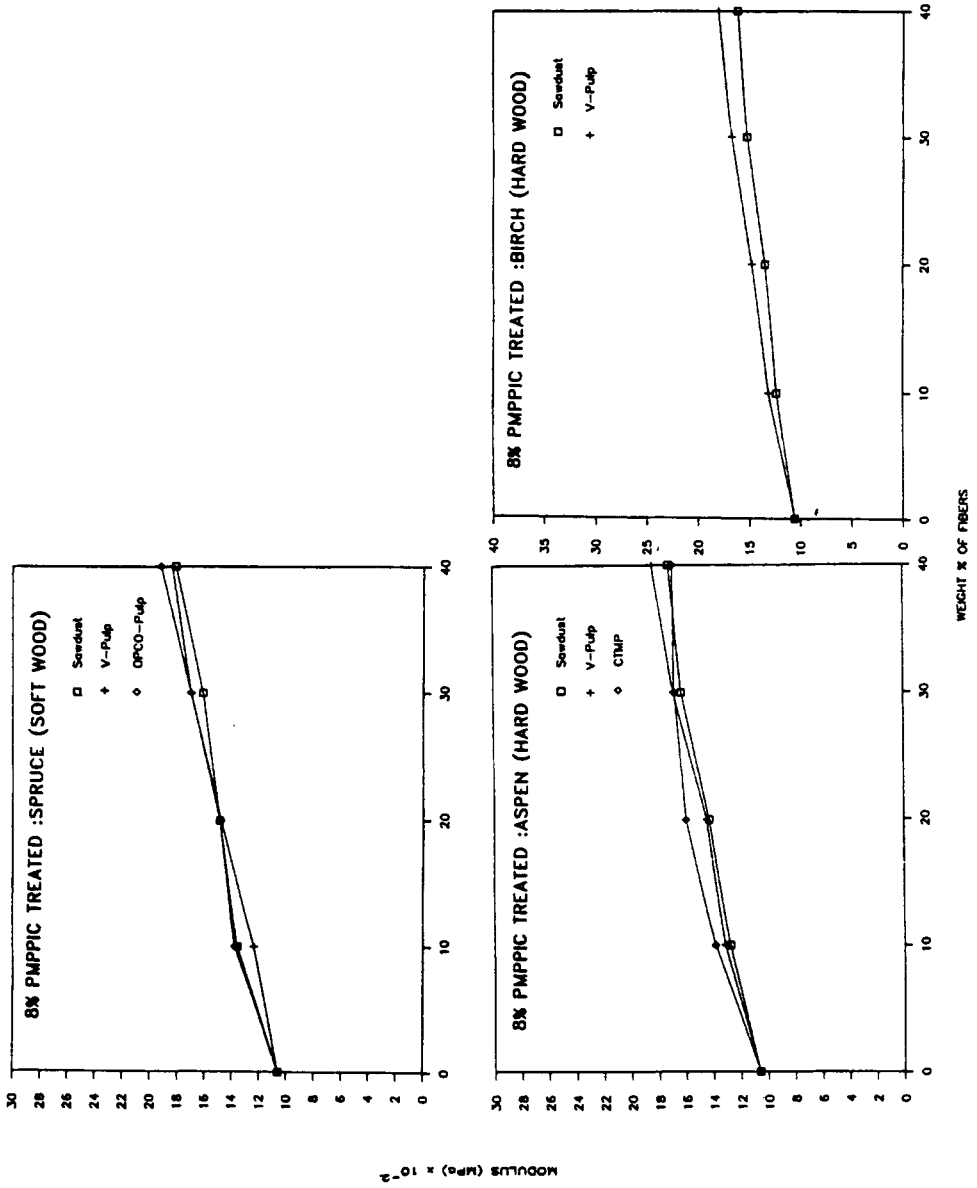
Fig. 12.   Effect of fiber weight percentage on flexural modulus for PS525 composites containing 8% PMPPIC-treated fibers.

TABLE VIA
Comparison of the Improvement in Mechanical Properties of Polystyrene–Spruce (Softwood) Fiber Composites

Polystyrene 201

Composition of composites		Improvement % of			
Pulp	Wt[a] % of PMPPIC fiber	Ultimate strength	Ultimate elongation	Fracture energy	Flexural modulus
Sawdust	0	+0.9	−17.6	−5.9	+46.3
	2	+4.4	+3.3	+27.6	+49.7
	8	−9.0	−12.6	−5.3	+51.2
V-pulp	0	+5.8	−24.2	−16.2	+63.3
	2	+21.5	+8.1	+52.7	+63.4
	8	+9.0	−13.1	+10.0	+65.1
OPCO-pulp	0	−8.9	−18.2	−11.9	+51.0
	2	+10.2	+0.3	+28.2	+45.3
	8	−0.5	−19.7	−16.2	+52.3

Note: In Polystyrene 201 section, PMPPIC fiber Wt% column values are 20, 20, 30, 20, 20, 20, 20, 20, 20.

Polystyrene 525

Composition of composites		Improvement % of			
Pulp	Wt[a] % of PMPPIC fiber	Ultimate strength	Ultimate elongation	Fracture energy	Flexural modulus
Sawdust	0	+16.3	+17.9	+79.8	+42.2
	2	+35.6	+108.0	+273.9	+53.4
	8	+30.9	+82.7	+218.7	+52.6
V-pulp	0	+31.6	+21.6	+88.8	+76.4
	2	+67.2	+125.3	+372.2	+53.7
	8	+50.3	+100.6	+271.7	+61.0
OPCO-pulp	0	+43.7	+74.7	+213.5	+76.9
	2	+66.6	+130.9	+347.4	+66.8
	8	+34.0	+74.1	+181.8	+61.1

Note: In Polystyrene 525 section, PMPPIC fiber Wt% column values are all 30.

[a] By weight of polymer.

TABLE VIB
Comparison of the Improvement in Mechanical Properties of Polystyrene–Aspen (Hardwood) Fiber Composites

| Polystyrene 201 | | | | | | | Polystyrene 525 | | | | | |
| Composition of composites | | Improvement % of | | | | | Composition of composites | | Improvement % of | | | |
Pulp	Wt[a] % of PMPPIC fiber	Ultimate strength	Ultimate elongation	Fracture energy	Flexural modulus		Pulp	Wt[a] % of PMPPIC fiber	Ultimate strength	Ultimate elongation	Fracture energy	Flexural modulus
Sawdust	0	20	−22.6	−32.6	−43.0	+46.9	Sawdust	0	30	+9.9	+40.1	+108.7	+57.0
	2	20	−0.2	−28.4	−19.5	−57.6		2	30	+29.7	+83.3	+213.5	+62.4
	8	20	−0.8	−5.4	+11.5	+39.4		8	30	+28.4	+80.9	+243.6	+53.8
V-pulp	0	20	−12.1	−13.7	−6.3	+50.1	V-pulp	0	20	+16.1	+36.4	+115.5	+55.4
	2	20	+9.6	−4.5	+19.3	+31.2		2	30	+51.4	+113.0	+353.1	+56.2
	8	30	+3.0	−27.8	−22.1	+62.2		8	30	+39.9	+71.6	+201.3	+59.3
CTMP	0	20	+3.5	−8.4	+5.5	+51.2	CTMP	0	20	+10.6	+40.8	+117.6	+36.5
	2	30	+13.9	+2.7	+30.0	+48.9		2	20	+50.2	+205.6	+554.5	+47.4
	8	20	+11.9	−9.0	−10.3	+57.5		8	20	+43.2	+171.0	+463.2	+51.7

[a] By weight of polymer.

TABLE VIC
Comparison of the Improvement in Mechanical Properties of Polystyrene–Birch (Hardwood) Fiber Composites

Polystyrene 201

Composition of composites		Improvement % of			
Pulp	Wt[a] % of PMPPIC fiber	Ultimate strength	Ultimate elongation	Fracture energy	Flexural modulus
Sawdust	20	−5.0	−17.3	−2.2	+44.6
	20	+5.2	−4.5	+3.1	+44.7
	30	+1.2	−3.0	+10.2	+44.8
V-pulp	20	−8.7	−5.4	+7.4	+37.5
	30	+14.7	+1.8	+30.6	+61.3
	30	+4.3	−3.6	+12.3	+56.3

(Pulp column Wt% values: Sawdust 0, 2, 8; V-pulp 0, 2, 8)

Polystyrene 525

Composition of composites		Improvement % of			
Pulp	Wt[a] % of PMPPIC fiber	Ultimate strength	Ultimate elongation	Fracture energy	Flexural modulus
Sawdust	30	+1.9	−4.9	+17.1	+41.3
	30	+21.7	+98.8	+222.2	+42.2
	30	+26.8	+92.0	+232.6	+44.9
V-pulp	30	+16.9	+0.6	+32.6	+49.8
	30	+39.3	+88.3	+266.5	+56.2
	30	+37.7	+72.9	+178.4	+58.9

(Pulp column Wt% values: Sawdust 0, 2, 8; V-pulp 0, 2, 8)

[a] By weight of polymer.

TABLE VII
Impact Strength (Izod, Unnotched) for Polystyrene–Aspen (Hardwood) Fiber Composites

Composite (wt % of fiber)	Izod impact strength (J/m)							
	10	20	30	40	10	20	30	40
	Polystyrene 201				Polystyrene 525			
Polymer		7.2				21.7		
Polymer + sawdust	2.1	2.0	1.9	1.9	14.8	8.7	4.9	4.7
Polymer + sawdust (coated with 10% polymer)	—	—	—	—	9.7	5.8	2.8	2.0
Polymer + sawdust (coated with 10% polymer and 8% PMPPIC)	4.5	5.5	5.5	4.3	15.5	11.4	8.4	8.3
Polymer + CTMP	5.9	5.6	5.0	4.1	16.5	13.7	7.5	5.5
Polymer + CTMP (coated with 10% polymer)	—	—	—	—	12.9	8.4	5.8	4.3
Polymer + CTMP (coated with 5% polymer and 4% PMPPIC)	4.7	6.0	5.8	5.1	—	—	—	—
Polymer + CTMP (coated with 10% polymer and 8% PMPPIC)	6.4	7.0	5.4	3.5	16.6	16.4	13.8	6.4
(Polymer + 2% PMPPIC) + CTMP	6.6	7.2	5.5	4.4	16.8	17.2	14.4	12.7
(Polymer + 8% PMPPIC) + CTMP	4.7	5.6	5.7	5.3	15.1	15.5	12.4	10.0
Polymer + V-pulp	6.0	3.5	3.6	3.0	16.5	14.0	8.7	6.9
(Polymer + 2% PMPPIC) + V-pulp	8.2	9.4	8.3	4.2	16.9	15.1	11.9	9.5
(Polymer + 8% PMPPIC) + V-pulp	5.3	5.4	6.3	4.1	15.1	14.7	12.6	6.9

Moreover, CTMP and other pulps are more flexible than sawdust. As a result, the force required to separate the adhering materials changed.

It is obvious from the above discussion that sawdust is not as effective as a reinforcing filler when compared to pulps, and CTMP ranks first. But V-pulps and OPCO-pulps behave equally. This observation is quite consistent with our previous report[16] on the poly(methyl methacrylate) resin system. Chemithermomechanical pulps (CTMP) the fibers are more completely separated than in the case of V-pulps or OPCO-pulps. As a result, CTMP fibers can be dispersed more completely. Sawdust is inferior when compared to V-pulps or OPCO-pulps.

Furthermore, comparing the different wood species, one can see that the more flexible softwood spruce pulp is superior as a reinforcing filler when compared to hardwood birch which is considered superior to hardwood aspen. This response was also observed earlier by us[12,16] and Kokta et al.[3,21] The order of performance follows nearly the similar trend. Depending on the origin, i.e., wood species, the characteristics of various kinds of pulps differ to a large extent[18]: e.g., softwood (spruce), long, medium slender, medium walled; hardwood (aspen), short, slender, medium walled, with thin walled vessels; hardwood (birch), short, very slender, medium walled, with thin-walled vessels. Moreover, softwood fibers (spruce) are flexible compared to hardwood (aspen or birch). Again, between aspen and birch, the latter is denser. The difference in morphology, density, and aspect ratios of different wood species accounts for the reinforcing action in thermoplastic composites.

TABLE VIII
Impact Strength (Izod, Unnotched) for Polystyrene–Spruce (softwood) Fiber Composites

Composite (wt % of fiber)	Izod impact strength (J/m)							
	10	20	30	40	10	20	30	40
	Polystyrene 201				Polystyrene 525			
Polymer	7.2				21.7			
Polymer + V pulp	6.2	5.8	4.8	2.7	17.7	14.2	11.4	7.2
Polymer + 2% PMPPIC) + V-pulp	8.6	10.3	8.6	4.3	17.9	19.4	17.0	15.0
Polymer + 8% PMPPIC) + V-pulp	4.5	3.8	4.0	3.6	16.9	15.8	10.5	7.0
Polymer + OPCO pulp	6.9	3.8	2.8	2.1	17.7	16.8	15.1	9.40
Polymer + 2% PMPPIC) + OPCO-pulp	8.6	8.4	5.9	4.2	17.4	19.8	16.6	10.6
Polymer + 8% PMPPIC) + OPCO-pulp	6.1	7.3	5.8	3.3	16.8	15.9	13.7	9.1

The Izod impact strength of unnotched PS201 and PS525, each filled with different hardwood aspen pulps (e.g., sawdust, CTMP, and V-pulps) are shown in Table VII. This table also includes the effect of precoating the fibers with polymer or with polymer and a coupling agent (PMPPIC). It is obvious from this table that the impact strength of nontreated composites or polymer precoated fibers are less effective than the unfilled polymer and the toughness decreases with increasing fiber concentration. In the treated composites impact strength increased when compared with nontreated ones. In a few examples, it even showed an increase at 20–30% fiber concentration compared to the uncoupled composites.

Table VIII shows the impact strength of nontreated, 2 and 8% PMPPIC-treated composites of V-pulps and OPCO-pulps of softwood spruce and polystyrene (PS201 and PS525). The nontreated and treated composites behave in a similar way to what was discussed above (for Table VII). It is obvious from these two tables that impact strength is inferior when composites are treated with 8% PMPPIC compared to 2% PMPPIC-treated composites. Furthermore, for softwood spruce fibers, V-pulps appear better than OPCO-pulps. Hardwood aspen sawdust shows the least reinforcement, while V-pulps are slightly greater than CTMP. Except for CTMP composites, the composites possess similar fracture toughness. Results of the impact toughness support our interpretation and speculations concerning the effect of coupling agent and the physical differences of the pulps. In addition to our previous results, one can add that high impact polystyrene (e.g., PS525) is generally superior to heat-resistant crystal polystyrene (e.g., PS201).

CONCLUSIONS

From the previous discussion and based on Tables VI (A–C), where increases in the mechanical properties at better filled composites are summarized, one can draw the following conclusions:

1. 2% PMPPIC provides the largest increase in mechanical properties including impact strength compared to nontreated or 8% PMPPIC treated composites.
2. A maximum of 20–30% fibers content can be incorporated without adversely affecting performance.
3. Flexible softwood spruce pulps provided better reinforcement than denser hardwood birch or hardwood aspen pulps.

References

1. T. P. Nevell and S. H. Zeronian, *Cellulose Chemistry and Its Applications*, Ellis Horwood, New York, 1985, p. 8.

2. A. W. Mekenzie and J. P. Yuritta, *Appita*, **32**(6), 460 (1979).

3. B. V. Kokta, P. D. Kamdem, A. D. Beshay, and C. Daneault, *Polymer Composites*, B. Sedlácek, Ed., de Gruyter, Berlin, 1986, p. 251.

4. F. J. Washabough, *Mod. Plast. Encyl.*, **63**, 144 (1986–1987).

5. R. B. Seymour, *Popular Plast.*, **23**, 27 (1978).

6. A. R. Sanadi, S. V. Prasad, and P. K. Rohatgi, *J. Sci. Ind. Res.*, **44**, 437 (1985).

7. T. Shinomura, U.S. Pat. 3,888,810 (1975).

8. L. A. Goettler, U.S. Pat. 4,376,144 (1983).

9. M. H. Schneider and K. I. Brekner, *Wood Sci. Technol.*, **19**(1), 67 (1985).

10. H. Dalväg, C. Klason, and H.-E. Strömvall, *Int. J. Polym. Mater.*, **11**, 9 (1985).

11. D Maldas, B. V. Kokta, and C. Daneault, *J. Appl. Polym. Sci.*, **37**, 751 (1989).

12. D. Maldas, B. V. Kokta, R. G. Raj, and C. Daneault, *Polymer*, **29**, 1255 (1988).

13. A. D. Beshay, B. V. Kokta, and C. Daneault, *Polym. Compos.*, **6**(4), 261 (1985).

14. B. V. Kokta, F. Dembélé, and C. Daneault, in *Polymer Science and Technology*, C. E. Carraher, Jr. and L. H. Sperling, Eds., Plenum, New York, 1985, Vol. 33, p. 85.

15. R. T. Woodhams, G. Thomas, and D. K. Rodgers, *Polym. Eng. Sci.*, **24**(15), 1166 (1984).

16. D. Maldas, B. V. Kokta, and C. Daneault, *Int. J. Polym. Mater.*, **13** (1989).

17. W. E. Johns, *J. Adhesion*, **15**, 59 (1982).

18. J. A. Clark, *Pulp Technology and Treatment for Paper*, 2nd ed., Freeman, San Francisco, 1985, Chap. 9, p. 184.

19. B. V. Kokta, J. L. Valade, and C. Daneault, *Transactions*, TR59 (Sep. 1979).

20. P. Zadorecki, H. Karnerfars, and S. Linderfors, *Compos. Sci. Technol.*, **27**, 291 (1986).