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Wood flour as a low-cost reinforcing filler for polyethylene: studies on mechanical properties

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Wood flour was used as a reinforcing filler for high-density polyethylene (HDPE). Different fibre treatments were carried out to improve the bonding at the polymer–fibre interface. Wood flour composites coated with alkoxy-silane coupling agents (silane A-172 and A-174) generally showed higher tensile strength and modulus values. A significant improvement in tensile properties was achieved with the addition of polymethylenepolyphenyl isocyanate. The Izod impact strength of the composites decreased with an increase in filler concentration.

1. Introduction

The low cost and easy avaliability of cellulosic fillers is one of the main reasons for their demand as an extender or filler in polymers [1]. While the use of wood flour in thermosets (phenolformaldehyde and melamine formaldehyde) is well known, it has not been fully exploited as a filler/reinforcing agent in thermoplastics. Recent literature studies have indicated that wood flour has good potential as a reinforcing filler for thermoplastics [2, 3]. In addition to the lower density and slow wear during processing, wood fibres have distinct advantages compared with the other commonly used reinforcing fillers [4].

The higher strength and modulus of wood fibres can only be fully utilized in composites if there is good adhesion at the interface. When there is a lack of adhesion between the polymer and fibre, the stress cannot be transferred effectively across the interface. Good bonding at the interface can be achieved by modifying the filler–matrix surface with various surface-reactive additives or coupling agents [5]. The hydrophilic nature of wood fibre offers additional advantages, in comparison with inorganic fillers, by readily providing the possibility for incorporation of different chemical groups on the surface [6].

The choice and extent of chemical treatment of fibre depends upon the nature of the chemical group and the concentration of coupling agent. The ultimate properties of the composite are influenced by (a) the selection of the polymer matrix, (b) processing conditions, (c) whether fillers are treated with processing aids or coupling agents, (d) the morphology of the filler and (e) fibre length, etc. [7–12].

In the present investigation, the effect of wood flour filling on the mechanical properties of composites of wood flour with high-density polyethylene (HDPE) was studied. Different coupling agents were used to improve the bonding between polymer and wood fibre. The effect of coupling agents (silane A-172, silane A-174 and polyisocyanate) on the mechanical properties of the composite was examined.

2. Experimental procedure

2.1. Materials

High-density polyethylene GRSN-8907 (density $0.954\,g\,cm^{-3}$, melt index $7.5\,g/10\,min$) was supplied by Novocar Chemicals Ltd, Canada. Wood flour (trembling aspen) was ground to mesh size 60 and dried at 60° C for 12 h before mixing with the polymer. The fibre aspect ratio (L/D) was 6.2. Different coupling agents were used to promote adhesion at the fibre–matrix interface (Table I). Silane coupling agents were used as supplied by the manufacturer. Polymethylene-polyphenyl isocyanate (PMPPIC) was either directly mixed with the polymer or pre-coated on the fibre surface and then mixed with the polymer.

2.2. Pre-coated fibre

The experimental procedure for pre-coating the fibre with silane coupling agent (4% by weight of the fibre) was the same as described earlier [13]. PMPPIC was added (1 to 3% by weight of the polymer) directly to the polymer–fibre mixture during the compounding stage. Alternatively, the wood flour was pre-coated with PMPPIC (3% by weight of the polymer) along with a small amount of polymer (HDPE GRSN 8907, 5% by weight of the polymer) in a roll-mill.

2.3. Compression moulding

Table II presents the compounding and moulding conditions for HDPE–wood flour. The compounding of polymer and wood flour (10, 20, 30 and 40 wt %

TABLE I Additives/coupling agents used

Silane A-172 – vinyltri (2-methoxy ethoxy) silane (Union Carbide)
Silane A-174 – γ-methacryloxy-propyltrimethoxy silane (Union Carbide)
Polymethylenepolyphenyl isocyanate (PMPPIC) (PolySciences)

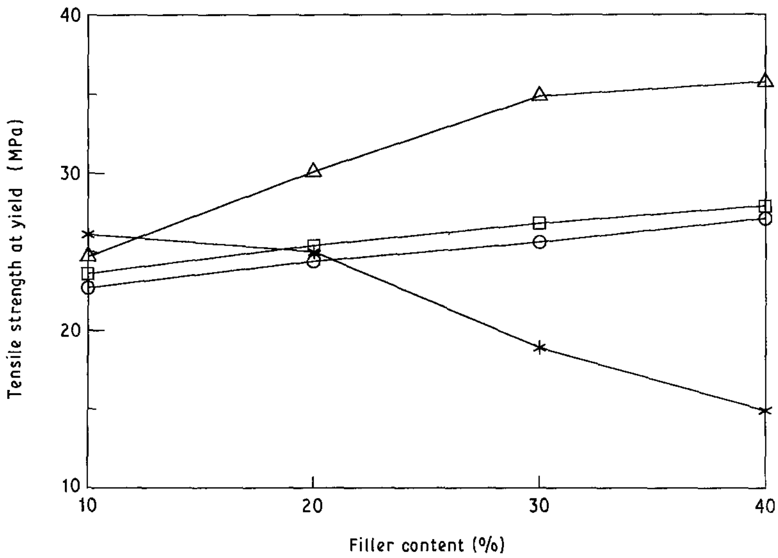

Figure 1 Effect of pre-coating of fibre on tensile strength of HDPE–wood flour (aspen) composites: (*) untreated, (○) coated with silane A-172, (□) coated with silane A-174, (△) coated with PMPPIC.

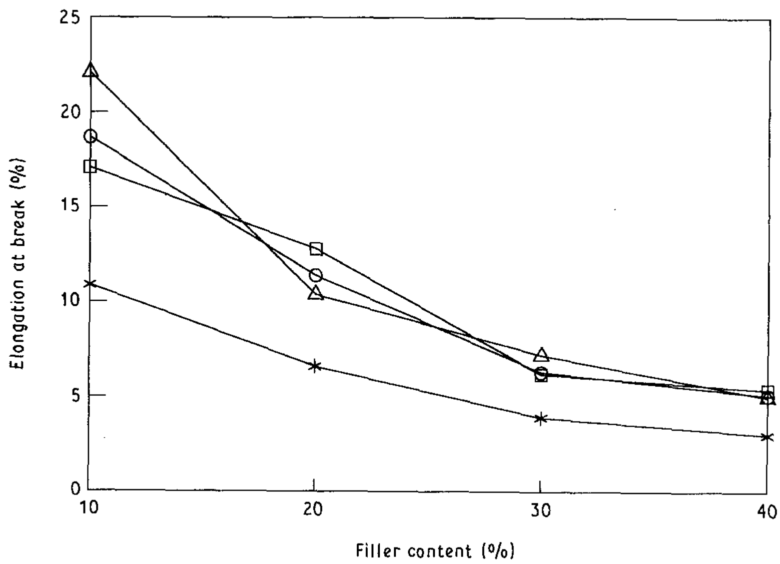

Figure 2 Effect of pre-coating of fibre on elongation of HDPE–wood flour (aspen) composites: (*) untreated, (○) coated with silane A-172, (□) coated with silane A-174, (△) coated with PMPPIC.

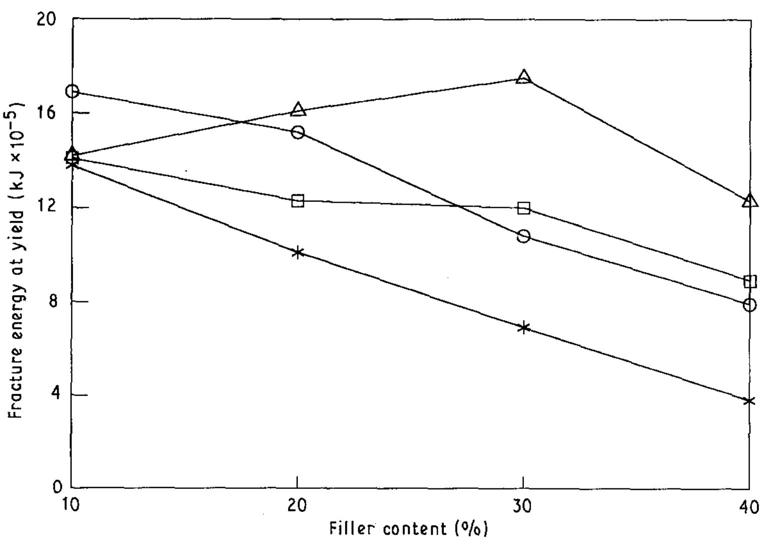

Figure 3 Effect of pre-coating fibre on fracture energy of HDPE–wood flour (aspen) composites: (*) untreated, (○) coated with silane A-172, (□) coated with silane A-174, (△) coated with PMPPIC.

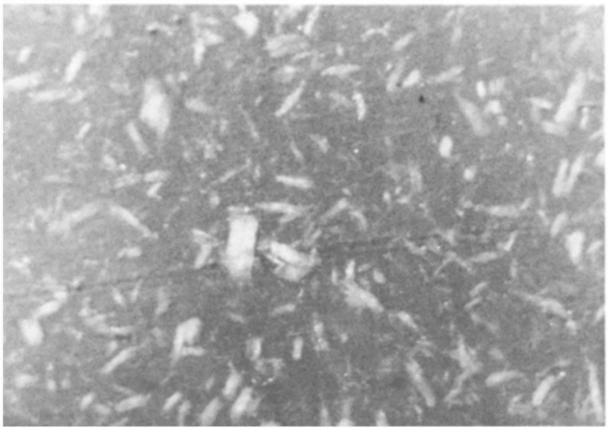

Figure 4 Optical micrograph of HDPE-untreated wood flour (aspen) composite (20 wt % fibre). Magnification 78 ×.

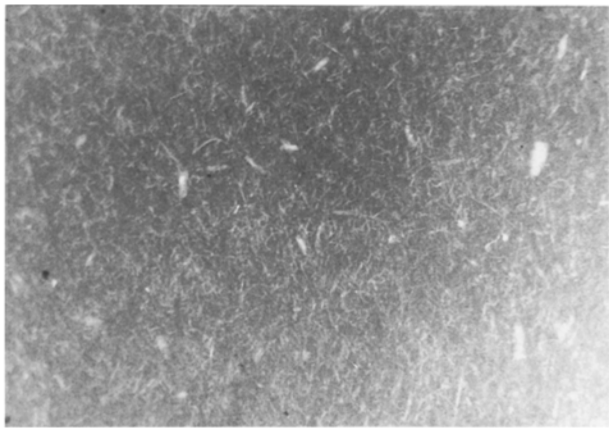

Figure 5 Optical micrograph of HDPE–PMPPIC-coated wood flour (aspen) composite (20 wt % fibre). Magnification 78 ×.

fibre) was done at 150 to 160° C in a roll-mill. In some cases, wood flour was pre-coated with polyisocyanate (PMPPIC) in a roll-mill at 160° C before mixing with the polymer. Dog-bone-shaped tensile specimens (6.4 cm × 0.318 cm × 0.158 cm) were obtained by compression-moulding at 150 to 160° C (pressure 3.2 MPa). The moulded samples were slowly cooled to room temperature with the pressure maintained during the process.

2.4. Mechanical properties

Tensile properties of the composite were measured using an Instron Model 4201. The crosshead speed was $10\,\mathrm{mm\,min^{-1}}$. The reported properties were measured at yield or break. Tensile modulus was measured at 0.1% strain. Fracture energy was calculated from the area under the stress–strain curve. A minimum of six samples were tested in each series. The test results were automatically calculated by an HP86B computing system using the Instron 24 12 005 General Tensile Test Program. The coefficient of variation of the reported properties was less than 8.0%. The Izod impact strength (un-notched) was determined using an impact tester (Model TMI No. 43-01).

3. Results and discussion

The tensile properties of HDPE filled untreated and pre-coated wood flour composites are shown in Figs 1 to 3. Compared to untreated fibre composites, a significant increase in tensile strength (after 20% filler level) was observed in silane A-172 and silane A-174 pre-coated fibre composites (Fig. 1). However, there is not much difference in the strength values among the two silane coupling agents used. Silane coupling agents are generally used to treat glass fibres, but in this case when applied with wood fibres a moderate gain in tensile strength was achieved. However, it should be mentioned that the application of silane to the wood fibre surface is affected by several parameters such as (i) the method of mixing, (ii) use of a peroxide and (iii) concentration of silane, as was discussed in an earlier communication [13].

TABLE II Compounding and moulding conditions

Roll mill (Brabender Prep. Roll Mill 065):	
Mixing temperature	150 to 165° C
Mixing time	5 to 10 min
Compression moulding (Carver Press):	
Moulding temperature	150 to 165° C
Moulding time	15 to 20 min
Moulding pressure	3.2 MPa
Mould cooling time	10 min

Methylene diisocyanate (MDI) is widely used as a bonding agent in wood-particle boards [14]. In the present study a polyisocyanate was tried as a bonding agent. As seen from Fig. 1, the best improvement in strength was achieved with PMPPIC pre-coated wood flour composites. For the filler concentration of 30% a steep increase in tensile strength is observed. Beyond this the strength tends to level off as the filler concentration increases. The tensile strength is increased by 63% at 40% filler concentration when compared to unfilled HDPE (21.7 MPa).

The better performance with isocyanate can be attributed to the formation of covalent bonds between the OH group of cellulose and isocyanate. The possible bonding mechanism can be explained as follows:

$$\mathrm{R{-}N{=}C{=}O + HO{-}cellulose}$$

$$\rightarrow \mathrm{R{-}\overset{H}{\overset{|}{N}}{-}\overset{O}{\overset{\|}{C}}{-}O{-}cellulose}$$

The isocyanate group reacts readily with the OH groups of cellulose to form a urethane structure [11]. Solvent extraction studies confirmed that isocyanate was chemically bonded with wood flour [15]. The chemically bonded wood surface is molecularly bridged during compounding to the thermally activated surface of the polymer matrix.

The variation of elongation (at break) with the filler concentration is presented in Fig. 2. Elongation decreased steadily with the increase in filler content. This is expected since the addition of filler increases the stiffness of the composite. A comparison of HDPE filled with untreated wood flour and pre-coated wood flour shows higher elongation values in the latter case. The fracture energy at yield was calculated from the area under the stress–strain curve. HDPE filled with

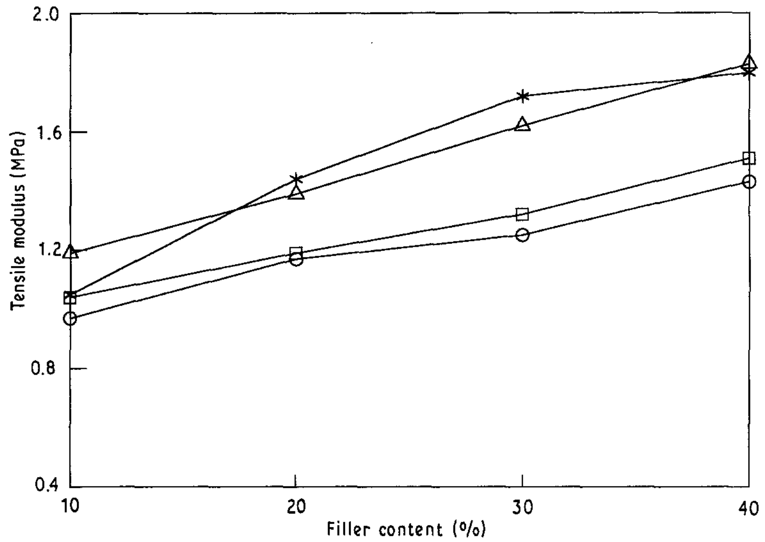

Figure 6 Effect of pre-coating fibre on tensile modulus of HDPE–wood flour (aspen) composites: (∗) untreated, (○) coated with silane A-172, (□) coated with silane A-174, (△) coated with PMPPIC.

PMPPIC-coated wood flour composites showed higher fracture energy absorption than untreated and silane pre-coated fibre composites (Fig. 3). These results are in agreement with the tensile strengths of the composites. In both cases, improved adhesion at the fibre–matrix interface is responsible for the increase in toughness of the composite.

The better performance of coated fibre composites may be due to the following reasons: (a) improved adhesion at the interface, (b) superior dispersion of fibre in the polymer matrix and (c) higher compatibility between the modified fibre and the polymer matrix. It was observed that during the process of mixing the bonding agent with the fibre, the addition of a small amount of polymer reduced the fibre-to-fibre affinity and enhanced the dispersibility of the fibre in the polymer [12]. Figs 4 and 5 illustrate the difference between untreated wood flour and PMPPIC-coated wood flour samples. The fibres tend to form large clusters in the case of HDPE filled with untreated fibres (Fig. 4), whereas fibre coating reduces the number of clusters and at the same time improves fibre dispersion in the matrix (Fig. 5).

Fig. 6 shows that with an increase in filler concentration the modulus increased steadily in all cases. The modulus was not much influenced by the fibre treatment, which is just the opposite to the influence on the tensile strength of the composite. On the other hand, the modulus is affected by fibre concentration since the fibres act as the stress transfer medium in the matrix. Higher fibre concentration in the polymer matrix leads to more efficient transfer of stress across the interface.

The effect of PMPPIC concentration on tensile properties of HDPE–wood flour composites (at 30.0% fibre level) is shown in Table III. With an increase in isocyanate concentration a steady increase in tensile strength was observed. An increase in elongation was found at higher isocyanate concentration, which suggests that the unreacted isocyanate in the polymer–fibre mixture could have a plasticizing effect on the polymer. However, the modulus remained relatively unaffected by the variation in isocyanate concentration.

Comparison of the tensile properties of HDPE filled with wood flour (aspen), mica and glass fibre is shown in Table IV. The strength continued to increase with an increase in filler content in wood flour composites, while in mica and glass fibre composites the stress decreased at higher filler levels. On the other hand, the retention of elongation was slightly better at lower filler levels in glass fibre composites. The modulus

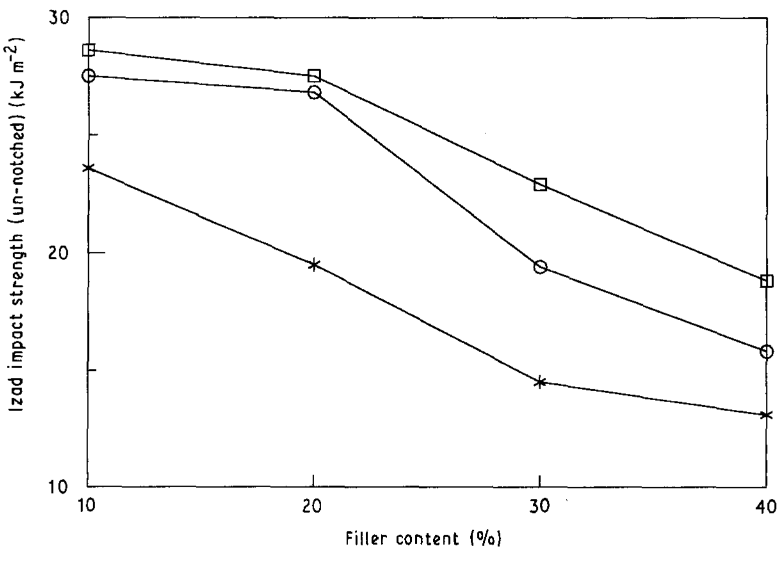

Figure 7 Izod impact strength (un-notched) of HDPE–wood flour (aspen) composites: (∗) untreated, (○) coated with silane A-174, (□) coated with PMPPIC.

TABLE III Effect of PMPPIC concentration on tensile properties of HDPE–wood flour (aspen) composites (30.0 wt % fibre)

PMPPIC (% by weight of polymer)	Tensile strength (MPa)	Elongation at break (%)	Tensile modulus (GPa)
0	18.3	3.9	1.72
1.0	27.7	4.3	1.69
2.0	28.8	5.4	1.74
3.0	30.4	6.1	1.79

values of HDPE–wood flour composites remained somewhere in between those for the glass fibre and mica composites. However, it should be noted that wood fibres have a lower density, which is an important factor if one considers the relative cost and performance of the material.

The Izod impact strength of HDPE–wood flour (aspen) composites at different filler concentrations is shown in Fig. 7. Fibre coating had very little effect on the impact strength. With an increase in filler loading the impact strength continued to decrease as compared to the unfilled polymer.

4. Conclusions

HDPE filled with silane or PMPPIC-treated wood flour produced a higher tensile strength compared with untreated fibre composites. A significant improvement in strength was achieved when wood flour was pre-coated with PMPPIC. An increase in PMPPIC concentration gave a higher tensile strength, but the modulus remained relatively unaffected. The Izod impact strength decreased with an increase in filler concentration. The tensile properties of HDPE filled with wood flour compared well with those of mica and glass fibre composites.

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TABLE IV Comparison of tensile properties of HDPE–wood flour (aspen), mica and glass fibre composites

Tensile properties	Wood flour*		Mica		Glass fibre	
	10†	30	10	30	10	20
Tensile strength at yield (MPa)	26.1	32.6	23.3	20.4	24.2	22.5
Elongation at break (%)	8.2	4.9	6.4	2.5	72.2	11.2
Fracture energy at yield (kJ $\times$ 10^{-5})	11.6	10.3	9.2	3.3	14.6	10.6
Tensile modulus (GPa)	1.1	1.8	1.4	2.3	1.2	1.9

*PMPPIC-coated
†Filler concentration (wt %).