Study on PVC Composites Containing *Eugenia jambolana* Wood Flour

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ABSTRACT: This article describes the properties of composites using unplasticized PVC matrix and wood flour (obtained by crushing the bark of *Eugenia jambolana*) as filler. Composites were prepared by mixing PVC with varying amounts of wood flour (ranging from 10–40 phr; having particle sizes of 100–150 μm and <50 μm) using two-roll mill followed by compression molding. The effect of wood flour content and its particle size on the properties, i.e., mechanical, dynamic mechanical, and thermal was evaluated. Tensile strength, impact strength, and % elongation at break decreased with increasing amounts of wood flour. Stiffness of the composites (as determined by storage modulus) increased with increasing amounts of the filler. Modulus increased significantly when wood flour having particle size <50 μm was used. Morphological characterization (SEM) showed a uniform distribution of wood flour in the composites. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 107: 2171–2179, 2008

Key words: composites; poly(vinyl chloride); *Eugenia jambolana*; bark flour; morphology; particle size

INTRODUCTION

Recently, thermoplastic composites containing natural cellulose-based fibers such as wood flour, wood fibers, and cellulose fibers have attracted increasing attraction of researchers. Several advantages of blending thermoplastic with natural fibers include relatively high specific stiffness, strength, and low density when compared with the inorganic reinforcements used (such as glass fibers, $CaCO_3$, mica, etc.). Other important properties of natural fibers are low cost value, renewable and recyclable nature. The advantage of being nonabrasive to the mixing and molding equipment are the added advantages to the plastic industries for the use in automobiles, building, and other applications.[1] Materials based on renewable resources are attracting growing attention, and attempts have been made to develop engineering materials for a variety of applications.[2] Waste wood is an important biomass resource. Many research groups have been studying new ways of using waste wood during the last few years. The production of WPC typically using a fine wood waste (cellulose based fiber fillers such as hardwood, softwood, plywood, peanut hull, etc.) mixed with various plastics (PP, PE, and PVC)[3,4] has been reported. Waste wood and some others lignocelluloses, such as wheat straw or barks of different tree species have been converted into thermosetting molding materials.[5–7] New wood polymeric materials, for example, the commercialized artificial wood on the base of wood flour filled PVC, have been obtained as a result of some theoretical and new technological solutions. The properties of filled plastics depends on the type of wood, shape, and size of the filler particles, filler content, matrix/filler particles interface interaction, and processing conditions.[8] Feldman and Banu et al.[9,10] blended different lignins and lignin derivatives with PVC and evaluated their effect on the mechanical, thermal, and weathering properties. It has been demonstrated that an interaction occurred between —OH groups of lignin and α-H of PVC. The two variables that distinguish wood flour are species and mesh size (particle size). Miren et al.[11] observed an increase in scorch time and curing time upon addition of wood flour to natural rubber and caused improvement in modulus at 300% strain. The particle size range of 300–425 μm was found to offer the best overall balance of mechanical and dynamic properties. Melt flow index, heat deflection temperature, and notched impact energy increased with increase in particle size.[12] Aggregation of particles, especially fine particles, is another factor that can influence the final properties of the composites.[13] Finally today's environmental and social concerns show increasing demands on

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forest resources and, thus, the combination of wood and synthetic polymers to produce cheap substitutes for traditional wood products.

PVC is a common thermoplastic and has wide variety of applications. Plant *Eugenia jambolana* (Jamun), family Myrtaceae, is habitat in India and found in large scale. Wood/bark of this plant has no commercial value and the aim of this work was to investigate the influence of bark wood flour particle size on the mechanical, thermal, and morphological properties of wood flour/PVC composites. For this purpose, wood flour having particle sizes ranging from 100 to 150 μm and <50 μm were used for making PVC composites. The content of wood flour was varied from 10 to 40 phr to investigate its effect on the performance of the composites.

EXPERIMENTAL

Materials

Poly(vinyl chloride) (PVC; SR –10 A grade) was procured from DCM Shriram Consolidated Ltd. Wood flour was prepared by grinding the bark collected form *Eugenia jambolana* followed by sieving using standard mesh to get particle sizes of <150 μm and <50 μm. The particle size was measured using a particle size analyzer (Ankersmid, Italy, Model CIF 100) by mixing small amounts of powdered wood flour in distilled water. Two different wood flour having particle sizes of 100–150 μm and <50 μm were used to make composites.

Preparation of composites

Bark wood flour of *Eugenia jambolana* was vacuum dried at 80°C for 24 h. PVC powder was initially formulated with additives in a mechanical mixture to obtain a PVC compound with improved melt processing. The dry PVC compound and the required amount of filler were mixed in a mixture grinder for 10 min. Several compounds were prepared by mixing the PVC compound with varying amounts of filler ranging from 10–40 phr. The dry blended compound was further mixed at 150°C using a hot roll-mill (Troester roll-mill). Sheets for testing were prepared by compression molding. The pressing cycle was 10 min at 180°C. To have a similar thermal history, the PVC compound was also subjected to the same processing conditions.

PVC compounds blended with bark wood flour having particle sizes of 100–150 μm have been designated as WF followed by numerals indicating the amount (in phr) of filler. For example, PVC having 10, 20, 30, and 40 phr of bark wood flour have been designated as WF10, WF20, WF30, and WF40, respectively. Similarly the sample prepared using 10, 20, 30, and 40 phr of bark wood flour of particle size <50 μm have been designated as NWF10, NWF20, NWF30, and NWF40, respectively.

Characterization

Water uptake

Water uptake in PVC and wood flour filled PVC composites was determined by immersing weighed amounts of rectangular specimens (63 × 12.6 × 2.2 mm) in distilled water at 25°C. After 24 h of immersion, samples were taken out, wiped with filter paper to remove the surface water, and weighed again. The water uptake was calculated using the following equation:

$$\%\ \text{Water uptake} = \frac{W_1 - W_0}{W_0} \times 100$$

where W_1 is the weight of the sample after immersing in water for 24 h (g), W_0 the initial weight of the sample (g).

Specific gravity

Specific gravity was determined in accordance with ASTM D792. The weight of the specimen in air and water was noted and the density was determined.

Mechanical properties

Tensile properties

Tensile properties were measured (25°C, R_H 65%) using Zwick Universal testing machine Model Z010 in accordance to ASTM 638. Dumbell shaped specimens were punched from the compression-molded sheets. A crosshead speed of 500 mm/min was used in the measurements. The gauge length of the specimen was 65 mm. At least five specimens of each sample were tested and the average value is reported.

Shore D hardness

Shore D hardness was determined using an Atsfaar (Italy) instrument in accordance with ASTM D2240. The specimen having a thickness of 6.4 mm was placed on a hard flat surface. The indentor of the instrument was then pressed onto the specimen making sure that it is parallel to the surface. The hardness was read after 15 s of firm contact with the specimen.

TABLE I
Details of Sample Preparation and Results of % Water Uptake and Specific Gravity in PVC/PVC Wood Flour Composites

Sample designation	PVC	Wood flour	% Water uptake (after 24 h of immersion at 25°C)	Specific gravity (ASTM D792)
PVC	100	0	0.02	1.446
WF10	100	10	0.10	1.459
WF20	100	20	0.19	1.459
WF30	100	30	0.29	1.460
WF40	100	40	0.31	1.460
NWF10	100	10	0.13	1.461
NWF20	100	20	0.15	1.480
NWF30	100	30	0.25	1.494
NWF40	100	40	0.35	1.510

Izod impact strength

Reversed notched izod impact strength was measured as per ASTM D256, using Ceast Resil impact tester. The specimen was held in a vertical cantilever beam and broken by a pendulum. The specimen was impacted on the side opposite to the notch. A notch was formed at an angle of 45° with a depth of 2.45 mm.

Static thermal stability

Static thermal stability was determined according to IS 5831-1984. For this purpose, a weighed amount of sample (0.05 mg) was taken in the test tube, which was placed in a heating chamber maintained at 200°C. The time required to change the color of congo red paper kept at the mouth of the tube was noted as stability time.

Thermal analysis

Thermogravimetric analysis

A Diamond simultaneous TGA-DSC-DTA (Perkin Elmer) system was used to record thermogravimetric traces in nitrogen atmosphere. A heating rate of 10°C/min and a sample size of 5 ± 1 mg was used in each experiment.

Dynamic mechanical thermal analysis

A Rheometric Scientific Model DMTA-IV was used to conduct dynamic mechanical thermal analysis of PVC and PVC wood flour filled composites. Rectangular bars having dimensions of 24 × 5.8 × 2 mm were used in a single point-bending mode. DMTA scans were recorded at an oscillation frequency of 1 Hz, strain 1% in the temperature range of 40–150°C. A heating rate of 3°C/min was used in each experiment.

Morphology

The tensile bars fractured at room temperature were used for morphological characterization using a Jeol JSM-840 scanning electron microscope operated at 15 kV. The fractured surfaces were coated with a thin layer of gold and then the images were recorded.

RESULTS AND DISCUSSION

Water absorption

The results of % water uptake in PVC, PVC/WF, and PVC/NWF composites are given in Table I. In all the samples, water uptake was in the range of 0.02–0.35%. A marginal increase in water uptake was observed upon incorporation of the filler. Water uptake was slightly higher in the composites having wood flour of lower particle size, i.e., <50 μm (NWF) as compared to the macrofiller (100–150 μm). It was observed that water absorption increased with increasing amount of the wood flour content in composites. The water absorptivity of the wood flour based composites largely depends on the presence of the hydrophilic groups, i.e., free —OH and —COOH groups, on the surface of the reinforcing wood flour. The results obtained clearly show that the addition of filler did not affect the hydrophobicity of PVC matrix much. Wood flour is almost covered with the PVC matrix thereby being hydrophobic in nature.

Specific gravity

All the samples had specific gravity in the range of 1.446–1.510 (Table I). Specific gravity increased with increasing amounts of wood flour. PVC microfilled (NWF) composites had a higher specific gravity as compared to PVC macrofilled (WF) composites.

Mechanical properties

The mechanical properties are dependent on factors such as filler content, particle size and shape, the

TABLE II
Mechanical Properties of PVC and PVC Wood Flour Composites

Sample designation	Shore D hardness at 15 s (ASTM D 2240)	Static thermal stability (min) IS 5831-1984	Modulus (MPa) ASTM 638	Tensile strength (MPa) ASTM 638	Elongation at break (%) ASTM 638	Impact strength (rev. notch) kJ/m^{-2} ASTM D256
PVC	71	55	172.6	45.9	38	Not break
WF10	73	50	194.7	42.6	9	12.8
WF20	74	45	177.6	42.0	4	6.6
WF30	76	46	183.6	37.4	4	5.8
WF40	77	43	91.2	37.8	3	4.0
NWF10	75	53	353.0	38.2	16	24.0
NWF20	76	51	342.8	40.4	9	12.4
NWF30	76	50	235.5	40.1	8	11.1
NWF40	77	49	182.6	42.5	5	5.6

degree of adhesion between filler and the matrix, and filler dispersion in the polymer matrix.[14] From the stress–strain curves, modulus, tensile strength, and percentage elongation at break were determined and the results are given in Table II.

Figure 1 shows the effect of filler content and particle size on tensile modulus. The modulus of both PVC wood flour filled composites is higher than that of neat PVC. The modulus of PVC/NWF composites was much higher when compared with PVC/WF composites. This is a common behavior for the polymers filled with natural fibers. Fillers are said to be much stiffer than the polymer matrix, and as a result, they add stiffness to the final product. The Hal-pin-Tsai equation,[15–17] which is commonly used to predict the modulus of the discontinuous short fiber reinforced composite from the modulus of the individual component, explains the above results. The equation has the general form

$$E_c = E_m\left(\frac{1+\xi\eta\Phi_f}{1-\eta\Phi_f}\right)$$

where E_c and E_m are the moduli of composite and matrix, respectively, and Φ_f is the volume fraction of the fibers. The constant ξ and η are given by

$$\xi = 2(L/D)$$

$$\eta = \frac{R-1}{R+\xi}$$

where L/D is (length/diameter) of the reinforcing fibers and R is the ratio of the filler modulus to the matrix modulus. Wood flour has a much higher modulus than that of commonly used thermoplastics. So in this work, the parameter R is >1 and, consequently, $\xi < 1$. Then it is apparent that value of Ec is greater than Em and increased with increasing volume fraction Φf of the filler. The tensile strength decreased with increasing amounts of the filler because the stress–strain curve does not show yield (Fig. 2).

For polymeric composites, the additions of the immiscible component to the polymer matrix generally decrease elongation properties considerably. As expected a decrease in elongation at break was observed for the composites having WF or NWF (Table II, Fig. 3). This indicates that the presence of wood flour in the matrix reduces the ability of the sample to deform by restricting the mobility of the polymer chains. As a consequence, it is difficult for the segments of the material to easily slip past each other.

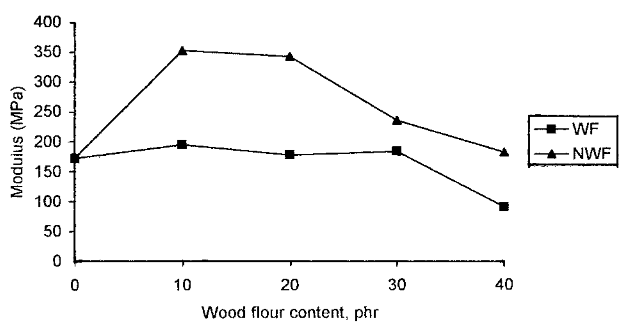

Figure 1 The effect of loading and particle size of wood flour on the modulus of PVC wood composites.

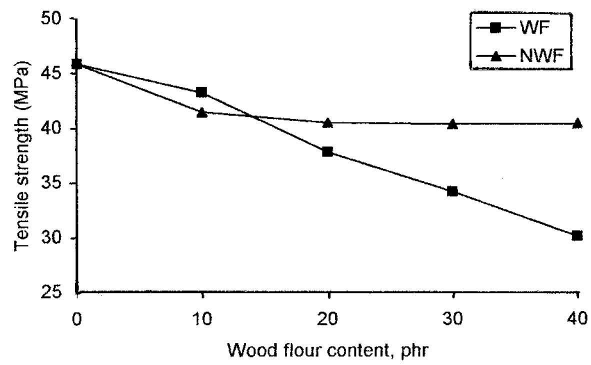

Figure 2 Tensile strength versus wood flour content for PVC/WF and PVC/NWF composites.

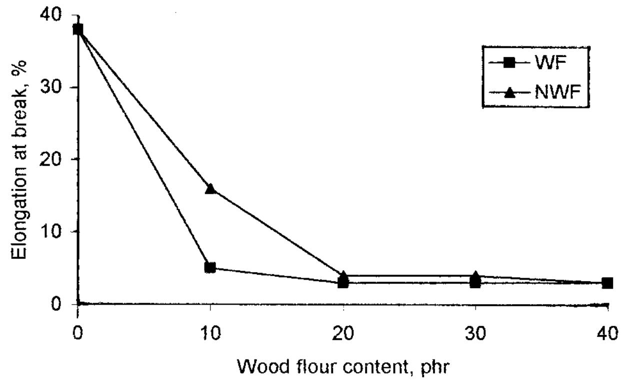

Figure 3 Elongation at break for PVC/WF and PVC/NWF composites.

Figure 4 shows the effect of wood flour particle size on the reverse notch impact strength. Generally the notched impact strength is a measure of crack propagation. Reverse notched impact strength is almost double in microsize (<50 μm) filled composites when compared with the macrofiller (100–150 μm) size filled PVC wood flour composites at the same loading level.

Hardness

As expected, the hardness of PVC wood flour composites increased upon addition of filler. The addition of macrosized filler had little effect as a function of concentration, whereas in microfilled composites (NWF) hardness of the composite increased (Table II).

The static thermal stability of PVC and PVC wood flour filled composites was determined by heating the sample at 200°C. The time taken by the congo red paper to change its color due to evolution of HCl was used as criteria for measuring thermal stability under isothermal conditions (Table II). The presence of wood flour decreased static stability and the time needed to change the color of congo red paper decreased with the increasing amount of wood flour. Decrease was much higher in the presence of macrofiller when compared with the microfiller.

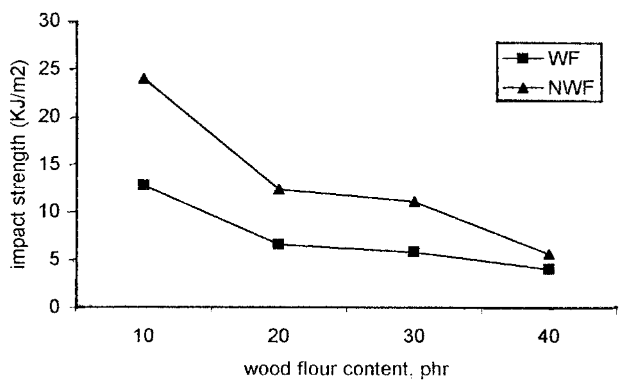

Figure 4 Effect of loading and particle size of wood flour on reverse notch impact strength of PVC wood composites.

Thermal behavior

Figures 5 and 6 show the TG/DTG traces of PVC, wood flour, and PVC wood flour filled composites. Two-step decomposition was observed in PVC and PVC wood flour filled composites, whereas in wood flour multistep decomposition was observed (Fig. 6).

In wood flour, a weight loss of 3.64% in the temperature range of 70–140°C was due to the absorbed moisture. Thermal decomposition of hemicellulose and glycosidic linkages was observed in the temperature range of 150–350°C, followed by decomposition of cellulose. In PVC, first step decomposition in the temperature range 250–380°C is attributed to the dehydrochlorination of PVC matrix followed by the thermal decomposition of dehydrochlorinated PVC in the temperature range of 400–550°C, which consist mainly of conjugated double bonds.

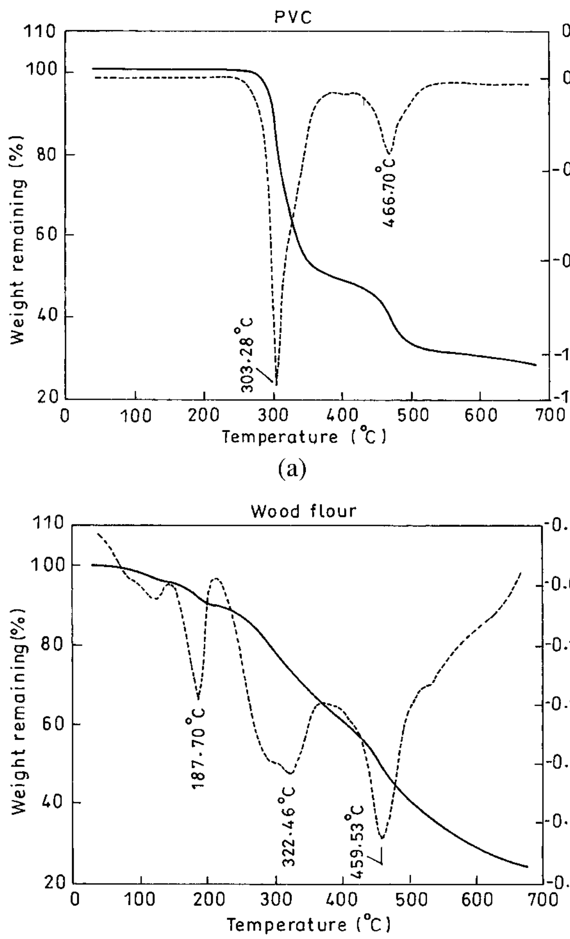

Figure 5 (a) Typical thermogravimetric curve for PVC. (b) Typical thermogravimetric curve for wood flour.

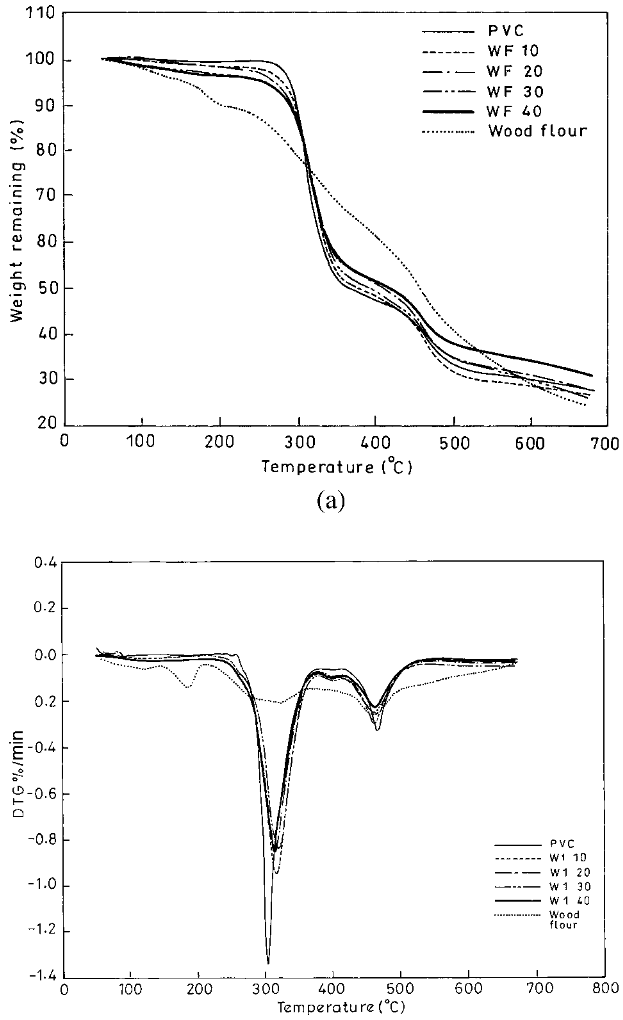

Figure 6 (a) Percentage weight remaining of PVC, wood flour, and PVC wood flour composites having particle size 100–150 μm. (b) DTG curves for PVC, wood flour, and PVC wood flour composites having particle size 100–150 μm.

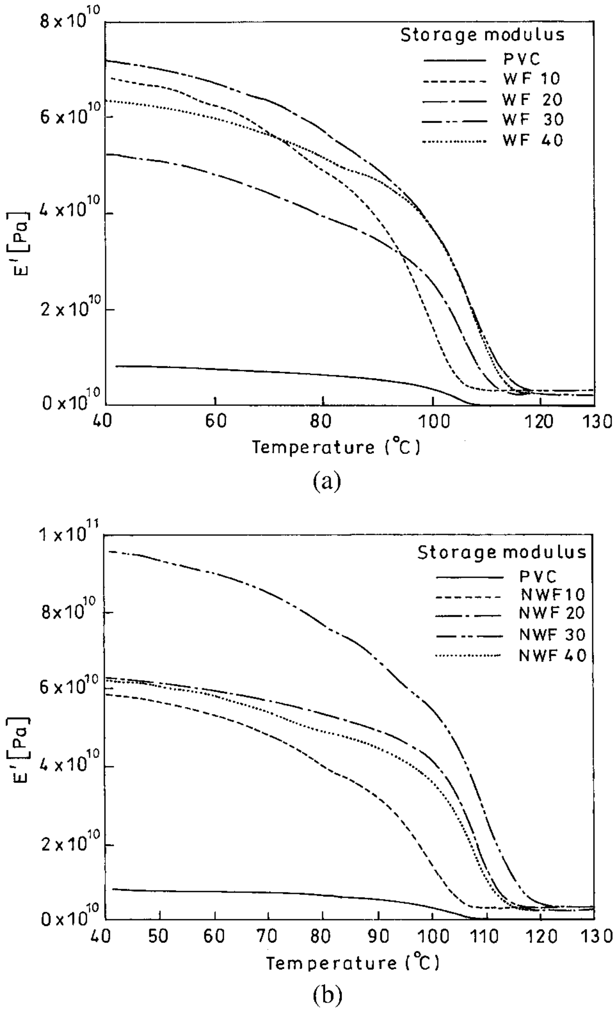

Figure 7 (a) Storage modulus versus temperature of PVC and PVC wood flour filled composites having particle size 100–150 μm. (b) Storage modulus versus temperature of PVC and PVC wood flour filled composites having particle size <50 μm.

In PVC wood flour composites, two-step decomposition was observed. The relative thermal stability of PVC and PVC wood flour composites was compared by comparing temperature at varying weight loss and temperature at which rate of weight loss is maximum. Decomposition temperature at 30, 40, and 50% mass loss was higher in PVC wood flour com-

TABLE III
Weight Loss and % Char Yield of PVC, Wood Flour, and PVC Wood Flour Composites

Sample designation	Decomposition temperature (°C) at % mass loss					% Char yield at 600°C
	10	20	30	40	50	
PVC	297.3	304.6	314.7	329.6	370.2	30.0
WF10	296.1	310.51	320.5	333.8	380.7	28.5
WF20	292.2	308.7	320.6	337.6	409.8	30.8
WF30	292.2	311.0	323.0	337.7	392.2	29.7
WF40	288.8	308.2	320.8	338.7	416.6	33.8
NWF10	292.5	308.0	318.1	330.6	371.9	24.0
NWF20	282.7	308.4	320.3	333.8	379.0	28.0
NWF30	290.2	309.2	321.7	338.3	409.9	33.6
NWF40	285.7	307.7	320.7	340.3	423.6	33.1
Wood	205.6	293.1	342.9	404.4	457.7	29.1

TABLE IV
Storage Modulus (E′) Loss Modulus (E″) and Tan δ of PVC, PVC/WF, and PVC/NWF Composites

Material	E' (GPa)	E'', (Gpa)	Tan δ	T_g (°C)
PVC	0.39	0.33	0.86	109
WF10	4.18	3.13	0.79	106
WF 20	3.42	2.67	0.78	112
WF 30	4.55	3.52	0.77	115
WF 40	3.79	2.96	0.78	115
NWF 10	4.11	3.33	0.81	106
NWF 20	3.97	3.04	0.78	116
NWF 30	5.39	4.16	0.77	118
NWF 40	3.66	2.85	0.77	115

All values were at 1 Hz.
E' and E'' values are at T_g.

posites when compared with PVC (Table III). Percent char yield increased marginally with increasing amount of wood flour.

Dynamic mechanical analysis

Figure 7 shows the plots of storage modulus versus temperature for PVC and wood flour filled PVC composites. The value of storage modulus at temperature ranging from 40°C to 130°C of PVC and PVC/wood flour composites are summarized in Table V. As expected, the storage modulus increased with increasing amount of wood flour up to 30 phr. Beyond 30 phr, the composite modulus decreased over the whole temperature range. The storage modulus of the composite at room temperature at a filler loading of 10 phr was significantly higher (6.79 GPa) than PVC (0.80 GPa). Substantial improvement in the rubbery plateau modulus of PVC wood composites over the PVC was clearly seen in Figure 7(a,b). These results clearly show that the addition of wood flour act as a reinforcing agent for the PVC matrix, thereby increasing the stiffness of the matrix. The increased shore D hardness (Table II) further confirmed that the addition of wood flour act as a reinforcing agent for the PVC matrix.

Figure 8 show the plots of tan δ versus temperature of PVC and wood flour filled PVC composites. The peak position of the tan δ plots was used to indicate the glass transition temperature (T_g) of the specimens. Addition of 10 phr of wood flour resulted in a decrease in T_g. Further increase of wood flour resulted in an increase in T_g value (Table IV). Increase in T_g was more in the presence of microsized wood flour when compared with the macrofiller. The presence of more rigid wood filler together with an excellent adhesion between the wood flour and the PVC matrix, resulting in a restriction of the molecular mobility of the polymer, thereby resulted in an enhancement of T_g. Increase

TABLE V
Storage Modulus (E′) of PVC, PVC/WF, and PVC/NWF Composites at Different Temperatures

Sample designation	Storage modulus (GPa) at temperature (°C)					
	40	60	80	100	120	140
PVC	0.80	0.74	0.63	0.31	–	–
WF10	6.79	6.21	4.96	1.70	0.32	0.32
WF20	5.20	4.75	3.98	2.60	0.25	0.25
WF30	7.14	6.65	5.77	3.69	0.23	0.22
WF40	6.31	5.92	5.17	3.74	0.24	0.24
NWF10	5.84	5.26	4.12	1.41	0.33	0.32
NWF20	6.26	5.90	5.34	4.16	0.33	0.32
NWF30	9.58	9.01	7.84	5.47	0.36	0.32
NWF40	6.22	5.74	4.94	3.56	0.25	0.25

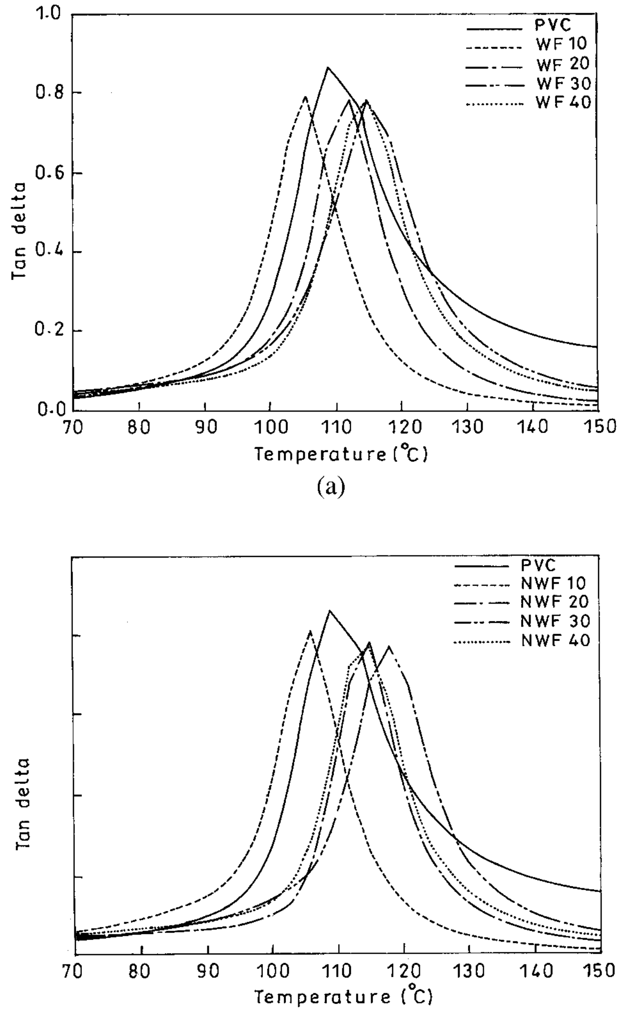

Figure 8 (a) Tan δ versus temperature at frequency 1 Hz of PVC and PVC wood flour filled composites having particle size 100–150 μm. (b) Tan δ versus temperature at frequency 1 Hz of PVC and PVC wood flour filled composites having particle size <50 μm.

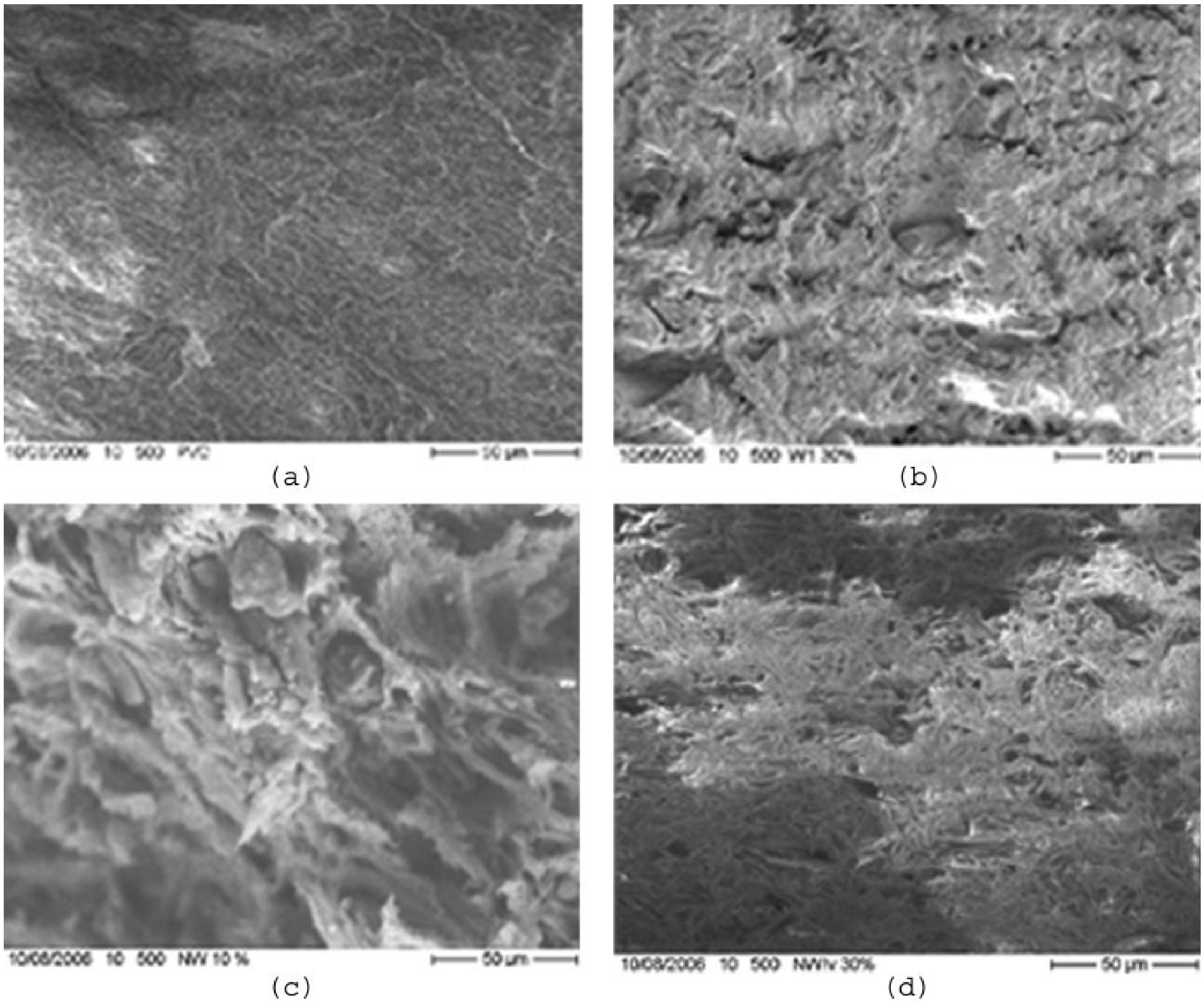

Figure 9 SEM micrographs for (a) PVC (b) PVC WF30 (C) PVC NWF10 (d) PVC NWF30 composites of magnification ×500.

in T_g of the polymer matrix using wood flour has also been reported by Rimdusit et al.[18]

Morphological characterization

The dispersion of wood flour in the PVC matrix, its wettability by the matrix, and the interfacial adhesion between the wood flour and the matrix were observed by scanning electron microscopy. SEM micrographs of the neat PVC and PVC having varying amounts of wood flour having particles of varying sizes are shown in Figure 9(a–d). It can be seen that wood flour of varying particle sizes exhibited the shape of irregular short fibers in composites. At lower phr of filler loading, wood flour is well dispersed. However, agglomeration of fibers was seen at higher filler loading.

CONCLUSIONS

PVC wood flour composites with different loading and particle sizes were prepared. Better results were obtained when filler particle size was <50 μm. The results suggest that the tensile strength and elongation at break decreased. However, modulus and hardness increased with increasing amount of loading. Stiffness of all the composites was substantially improved by filler addition. Thermal analyses indicate that the addition of bark wood flour into the polymer matrix showed no obviously negative effect on its thermal properties. Based on the comprehensive properties, the PVC NWF 30 phr composite can be used as a wood like material for application in wood structures with a reasonable balance of properties.

Wood flour composite can be melt-blended with PVC compound to give cost effective, renewable, recyclable, value added product (VAP) with fine particle wood showing good modulus.

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