

Mechanical, Thermal, and Water Uptake Characteristics of Woodflour-Filled Polyvinyl Chloride/Acrylonitrile Butadiene Styrene Blends

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ABSTRACT: Woodflour-filled composites based on polymeric blends of polyvinyl chloride (PVC) and super high-impact grade ABS were developed. Mechanical, thermal, and water uptake characteristics of the PVC/ABS matrix and their wood composites were evaluated. In the case of PVC/ABS matrix, the blend at a mass ratio of 50/50 rendered the impact strength with a very high value of up to 65 kJ/m², noticeably higher than those of the parent resins, that is, 6 kJ/m² of PVC and 35 kJ/m² of ABS. Dynamic mechanical analysis thermograms showed two distinct glass transition temperatures ($T_{g,s}$) that shifted toward each other indicating partial miscibility of the blends. Water absorption of the

blends after 24 h immersion was low, that is, within the range of 0.04–0.2 wt % and exhibits a behavior closed to pseudo-Fickian type. The obtained PVC/ABS wood composites exhibited an increase of flexural modulus as well as $T_{g,s}$ with an increase of woodflour content. Finally, impact strength of the PVC/ABS composites was significantly higher than those of PVC composites or polyethylene composites comparing at the same woodflour content. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 124: 943–950, 2012

Key words: blends; composites; impact resistance; mechanical properties; reinforcement

INTRODUCTION

In the recent years, wood plastic composites (WPCs) have made significant contributions to various applications such as buildings and constructions, automobiles, gardening, and outdoor products due to their ease of processing and recyclability.^{1,2} Polyvinyl chloride (PVC) is one of thermoplastics widely used as a matrix in WPC production, because this polymer can fit into the respective pricing category and at the same time having properties necessary for the WPC materials to pass the building standard. It provides high strength and modulus when compared with polyolefin. Furthermore, it is an inherent flame retardance polymer, which possesses a limiting oxygen index (LOI) as high as 50, while the materials with LOI of more than 26 can be classified as self-extinguishable.³

The applications of the PVC wood composites are recently reported to dramatically increase in various forms of construction parts such as window and door profiles, decking, railing, and siding.^{1,4} From several literatures,^{5–8} the WPC composites containing various matrix types experience critical problems of impact properties. At present, the practical and popular ways to toughen PVC wood composites are the use of impact modifiers. There are some impact modifiers that are widely used in PVC industries, that is, chlorinated polyethylene (CPE), methyl methacrylate butadiene styrene (MBS), and acrylic-based (ACR) impact modifiers. From the previous reports,^{9–11} the impact properties of PVC wood composites were improved by adding these impact modifiers, while the other mechanical and particularly thermal properties of the modified PVC and its composites were significantly decreased. That is because the glass transition temperatures of those impact modifiers are low (i.e., -16°C for CPE, -70°C for MBS, and -40 to -50°C for ACR).¹²

In this research, the blend of PVC and ABS was proposed as an alternative matrix for an enhancement of impact properties of its WPC without sacrificing its thermal properties. It was reported that PVC/ABS blends could reduce maximum overall rate of thermal

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decomposition process when ABS was incorporated in PVC.^{13,14} PVC/ABS blends were first commercially introduced in 1960.^{15,16} The outstanding commercial success of PVC/ABS blends has been gained from cost dilution and high performance characteristics with an addition of ABS particularly on its impact property. In these blending systems, PVC contributes to good flame retardancy and high tensile and flexural properties, whereas ABS can enhance processability, impact, and chemical resistance of the products. Property profiles of these blends are superior to those of component homopolymers, which makes them economically attractive for special markets, for example, automotive engineering plastics.^{17,18}

Pavan et al.¹⁹ studied the impact performance of PVC/ABS blends; they found that the impact behavior varied extensively over the composition range and related to the blends composition and structure. Additionally, Sharma et al.¹⁵ discussed about the development of PVC/ABS blends in various properties such as mechanical properties and thermal properties. About the application of PVC/ABS as a matrix in composite system, Qin et al.²⁰ developed clay-filled PVC/ABS blends and found that an incorporation of appropriate amount of clay could enhance the mechanical properties such as yield strength without impairing the impact strength.

Although the PVC/ABS blends have been well known for its outstanding impact properties, the application of the blend in high-impact wood polymer composites has received little attention. In this study, PVC/ABS blend is proposed to be used as a matrix in WPC to use its outstanding impact property and high thermal stability. Some other essential properties such as water uptake will also be investigated.

EXPERIMENTAL

Raw materials

PVC Siamvic 258RB used in the study possesses the characteristics as presented in Table I. Kane Ace PA 20 was used as processing aid additive, while SAK-WP-08-NP and calcium stearate were applied as heat stabilizers and external lubricants, respectively. PVC resin and the additives were supported by Vinythai

TABLE I
Characteristics of PVC Suspension Resin
(Siamvic 258RB)

Characteristic	Standard	Value
Viscosity index (mL/g)	ISO 1628-2	82
K-value (cyclohexanone)	DIN 53726	58
Polymerization degree	JIS K6721	680
Bulk density-compaction (kg/L)	ISO 1068	0.56
Volatile matter (%)	ISO 1269	≤0.3
Particle size (sieve analysis) (μm)	ISO 1624	120–150

TABLE II
Characteristics of ABS (Injection Grade)

Property	ABS (SP200) injection grade
Melt flow index (g/10 min) at 220°C/10 kg	17
Notched Izod impact strength (kJ/m ²) at 6 mm thickness at 23°C	35
HDT (6 mm thickness at 0.45 MPa)	90
Flexural strength (MPa)	56
Tensile strength (MPa)	41
Flexural modulus (GPa)	2.0

Public Co., Thailand. The internal lubricants, Loxiol P1141, were supported by the Siam Chemicals Solutions Co., ABS (SP200 superhigh impact grade), which was supplied by IRPC Public Co., Thailand, possesses the characteristics as shown in Table II. The woodflour of Redwood (*Xylocarpa*) as a waste material from a saw-mill factory in Thailand was used as filler and had an average particle size of 280 μm. It was crushed by Fritsch pulverisette cutting mill 15 and dried in an oven at 105°C for 24 h before use.

Composite preparation

PVC resin and its additives were blended using a high speed mixer (Plasmec Turbomixer 100L equipped with cooler) at a mixing speed of 1200 rpm. The PVC/ABS blends were prepared by two-roll mills at 160°C for 6 min to yield a homogeneous mixture. ABS contents were varied from 0 to 50 wt %. In the composite systems, the woodflour content was varied from 0 to 50 wt %. The obtained PVC/ABS blends and composites sheet were then compression-molded at 170°C and 15 MPa for 4 min. The compression-molded sheets were then cut into pieces for further property evaluations.

Sample characterizations

Flexural properties of the wood composite specimens were determined following ASTM D790-M93, using a universal testing machine (Instron 5567). The test was carried out in three-point bending mode with a support span of 48 mm at the crosshead speed of 1.2 mm/min. The dimension of the each specimen was 12.7 × 60 × 3.2 mm³.

Notched Izod impact strength of the specimens was obtained by an impact tester (Yasuda) according to ASTM D256-04. The dimension of the test specimen was 12.7 × 60 × 3.2 mm³ while the depth under notch of the specimen was 10 mm. The average values were calculated from 10 tests on each material.

Interfacial adhesion of the composites were studied using a scanning electron microscope (JSM-5800LV, JEOL) with an acceleration voltage of 15 kV. The surface of each impact fracture specimen was coated with thin gold film prior to testing.

Thermal characteristics of the blends and composite specimens were examined by a dynamic mechanical analyzer (DMA: NETZSCH-DMA242). The dimension of the test specimen was $50 \times 10 \times 2 \text{ mm}^3$. Three-point bending mode of deformation was used under a test temperature range from 30 to 150°C at the heating rate of $2^\circ\text{C}/\text{min}$. The test amplitude and frequency were $30 \mu\text{m}$ and 1 Hz, respectively. Glass transition temperature was obtained from the peak of loss tangent of the thermogram.

Water absorption measurement was conducted following ASTM D570, using disk-shape specimens with a diameter of 50.8 mm and a thickness of 3.2 mm. Three specimens were placed in an oven at 50°C for 24 h, cooled in a desiccator, and then weighted. The specimens were then immediately immersed in distilled water and weighed periodically. Based on the initial mass of each specimen, the amount of water absorbed was calculated from the following equation.

$$\text{WA}(\%) = \left(\frac{M_e - M_o}{M_o} \right) \times 100$$

where WA (%) is the percentage of water absorption and M_o and M_e are the values of specimen mass before and after immersion, respectively.

RESULTS AND DISCUSSION

Impact properties of PVC/ABS matrix

The effect of ABS content on impact strength of PVC/ABS blends is exhibited in Figure 1. It could be noticed that the impact strength of the blend gradually increased at ABS content of less than 30 wt %. The drastic increase in the impact strength was observed when the ABS content was ranging from 30 to 50 wt % and reached the maximum of 65 kJ/m^2 at 50 wt % of ABS. Additionally, the impact strength remained unchanged at 50–70 wt % of the ABS. The impact strength of blend, however, started to decrease when ABS was greater than 70 wt % to the value of about 35 kJ/m^2 of the neat ABS. This transition behavior in impact strength of our PVC/ABS blend was also observed in other particle toughened polymers.^{15,19,21,22} As suggested by Sharma and coworkers, the maximum impact strength can be achieved in this kind of polymer blend when critical volume fraction of elastomer phase is attained. However, when this critical elastomer content is exceeded, impact strength drops. At the peak value of impact strength, optimum balance between sufficient elastomer content and the preferred combination of toughening mechanisms such as multiple crazing with interacting shear deformation might be obtained.¹⁵

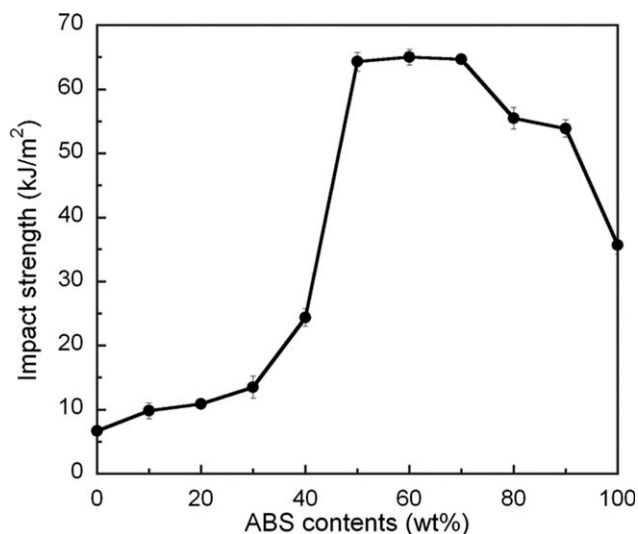


Figure 1 Impact properties of the PVC/ABS blends at various mass blending ratios.

In addition, impact strength of our PVC/ABS blends not only revealed synergistic behavior but also rendered much higher values than those from previously reported PVC/ABS blends.^{15,23} The use of novel super high-impact grade ABS from IRPC IRPC Public Co. to form alloys with PVC was found to provide such a high impact strength value with the rigid PVC used.

From the visual appearance observation of the tested samples under impact loading, the fracture surface of the neat PVC revealed a mirror-like surface of brittle failure nature, whereas that of ABS showed a rough surface due to plastic deformation of ductile failure with obvious stress whitening phenomenon. The PVC/ABS blends exhibited systematic variation in the fracture surface from rather brittle to highly ductile failure with increasing ABS content in the blends. In addition, a stress whitening zone (resulting from the mismatch in refractive index in the area of stress concentration) was observed to be more pronounced with the amount of ABS added to the PVC. This phenomenon is associated with the formation of many microcracks in the specimen, which arises from drawing the cavitations of the rubber phase in ABS.²⁴

Thermomechanical properties of PVC/ABS matrix

Figure 2 is DMA thermograms showing the relationship between storage modulus of the blends and temperature. The results provide useful information on rigidity or stiffness of the specimens. At glassy state of the samples (i.e., at 40°C), storage moduli of neat PVC and ABS were determined to be ~ 3.08 and 2.23 GPa , respectively. The PVC/ABS blend at 10 wt % of ABS provided the storage modulus of 2.94 GPa , and the modulus systematically decreased

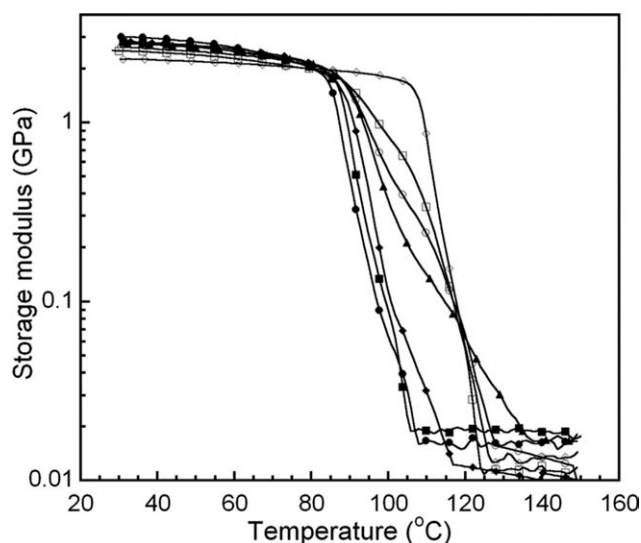


Figure 2 Storage moduli of PVC/ABS blends at various mass blending ratios: (●) PVC, (■) PVC/ABS 90/10, (◆) PVC/ABS 80/20, (▲) PVC/ABS 70/30, (○) PVC/ABS 60/40, (□) PVC/ABS 50/50, and (◇) ABS.

to 2.46 GPa at 50 wt % of ABS. The effect of ABS on modulus of PVC was also found to follow a rule of mixture as seen in the inset of Figure 2.

DMA thermograms can also provide some useful information on miscibility of the blend specimens. The loss tangent of the PVC, the ABS, and the blends at various ABS contents ranging from 0 to 50 wt % is exhibited in Figure 3. The peak of the loss tangent was used to indicate the glass transition temperature of each sample. From the figure, PVC and ABS showed single loss tangent peak corresponding to their T_g s.

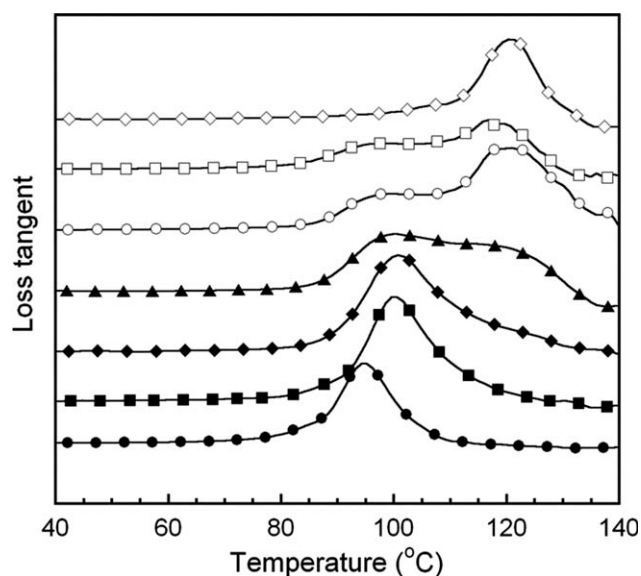


Figure 3 Loss tangent of PVC/ABS blends at various mass blending ratios: (●) PVC (■) PVC/ABS 90/10, (◆) PVC/ABS 80/20, (▲) PVC/ABS 70/30, (○) PVC/ABS 60/40, (□) PVC/ABS 50/50, and (◇) ABS.

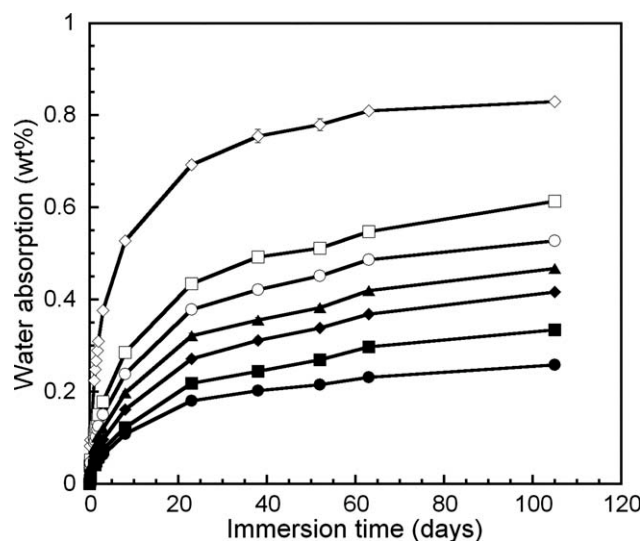


Figure 4 Water absorption of PVC/ABS blends at various mass blending ratios: (●) PVC, (■) PVC/ABS 90/10, (◆) PVC/ABS 80/20, (▲) PVC/ABS 70/30, (○) PVC/ABS 60/40, (□) PVC/ABS 50/50, and (◇) ABS.

In the case of the blends, two peaks of glass transition temperatures (T_g s) were clearly observed. It was found experimentally that the T_g values of the PVC and ABS were 94 and 120°C, respectively, whereas two T_g values of the blend at, for example, 50 wt % of ABS were located at 96 and 116°C. The shifting of the T_g values toward each other was an important characteristic suggesting partial miscibility of the blending systems. Moreover, the blend with 10 wt % of ABS showed an overlapping peak, whereas the blends with more than 20 wt % of ABS clearly exhibited two separate peaks. The height of those two peaks evidently varied systematically with the amount of each blend component, that is, the peak at higher T_g increases with the amount of the ABS and vice versa. Although the type of ABS used in this study was different from that reported in Jin et al.'s²⁵ research, similar results in partially miscible nature of PVC/ABS blend were observed.

Water absorption of PVC/ABS matrix

Water absorption experiment of PVC/ABS blends at various blend contents was performed until the saturation state was reached. The results are presented in Figure 4. After water immersion for 24 h, the neat PVC and pure ABS specimens absorbed water only up to 0.04 and 0.2 wt %, respectively. For PVC/ABS blends, the water uptake was found to increase with increasing of ABS content likely due to the presence of highly polar moiety of acrylonitrile component in ABS. In other words, the ABS is more polar than the PVC. The addition of ABS into PVC thus might contribute to the better wetting or greater interfacial interaction with the relatively highly polar woodflour filler.

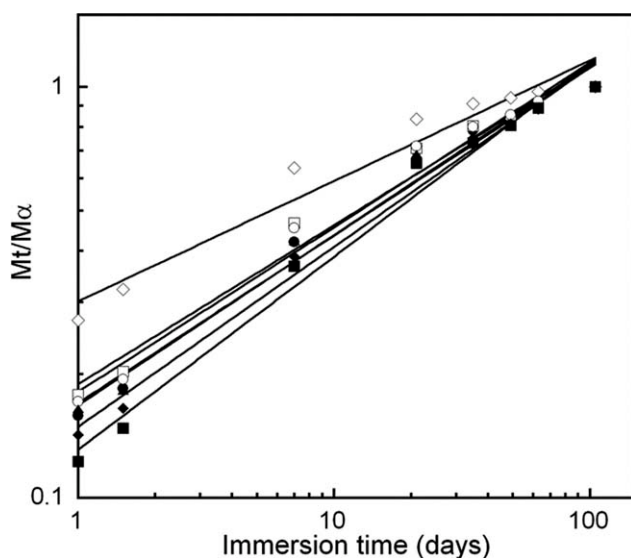


Figure 5 Plots between $\log M_t/M_\infty$ and time to determine the diffusion exponents in eq. (2) (●) PVC, (■) PVC/ABS 90/10, (◆) PVC/ABS 80/20, (▲) PVC/ABS 70/30, (○) PVC/ABS 60/40, (□) PVC/ABS 50/50, and (◇) ABS.

In addition, the values of water uptake up to 24 h at room temperature of the blends were also very low level, that is, $<0.2\%$, which is desirable in most wood-substituted products. Furthermore, from the curves, it could be noticed that the water absorption for over 105 day-immersion of all blend specimens was less than 0.8 wt %, which is considered to be very low. The generalized equation to explain mass diffusion in materials can be expressed as:

$$M_t = M_\infty k_n t^n \quad (2)$$

where M_t is the mass of a sample at time t and M_∞ the mass of the sample at saturation or infinite time. The diffusion behaviors can be classified as follows: supercase II ($n > 1$), case II ($n = 1$), anomalous ($1/2 < n < 1$), classical/Fickian ($n = 1/2$), and pseudo-Fickian ($n < 1/2$).²⁶

Figure 5 illustrates $\log (M_t/M_\infty)$ versus $\log(t)$ plots, which show the slopes ranging from 0.29 to 0.47. Thus, it could be concluded that these blending systems exhibited the diffusion behavior close to pseudo-Fickian type. According to Rogers²⁷ and Pritchard,²⁸ the pseudo-Fickian behavior is similar to Fickian one in the early stage of diffusion, but the rate of approaching equilibrium is delayed possibly due to the effects of concurrent diffusion and sorption behavior.

Mechanical properties of woodflour-filled PVC/ABS blends

The PVC/ABS blend at a mass ratio of 50/50 was selected for further investigation as a matrix of wood

composite, because this blend composition provided the greatest impact strength as discussed in the previous section. Figure 6 reveals that flexural modulus of the PVC/ABS wood composites systematically increased with increasing the woodflour content while their flexural strength tended to decrease with the amount of woodflour. The enhancement of flexural modulus of the wood composites could be attributed to the fact that the addition of rigid woodflour filler (having modulus in the range of 10–15 GPa)²⁹ could have an effect on the mobility restriction of polymer molecular chains.³⁰

In contrast to the flexural modulus of the composites shown in Figure 6, flexural strength of the composites tended to decrease with the amount of woodflour. The unfilled-PVC/ABS (50/50 mass ratio) provided the flexural strength of 75 MPa while an addition of woodflour into the matrix resulted in a decrease in flexural strength to 64.2 MPa (at 10 wt % of woodflour) and to 49.6 MPa (at 50 wt % of woodflour). The decrease in flexural strength of the PVC/ABS/wood composite with the woodflour content is attributed to a formation of a weak interfacial bonding between the woodflour and the PVC/ABS matrix or from some defect formation due to imperfect fiber-matrix wetting.^{5,31} The increase in modulus and decrease in strength due to the incorporation of woodflour or natural fiber into polymeric matrices imply the strength of the material is more sensitive to matrix properties and interfacial interaction formed in the composites than modulus.³¹ The behaviors are commonly observed such as in PE wood, PP wood, PVC wood, or ABS wood.^{32–37} However, the flexural properties of our PVC/ABS wood composites were observed to be superior to those wood composites comparing at the same wood flour content possibly from substantial

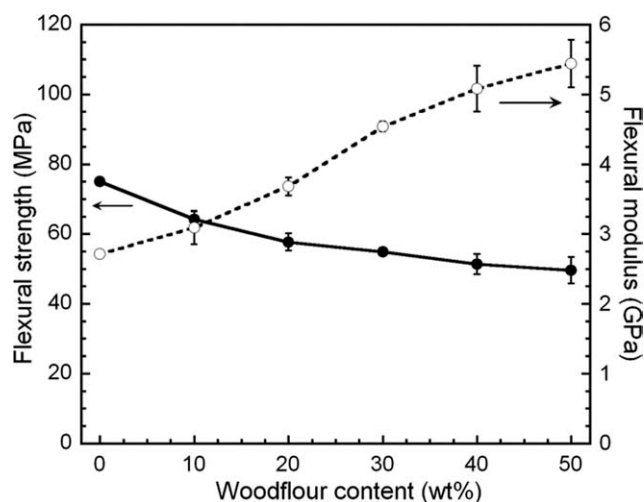


Figure 6 Flexural properties of PVC/ABS/wood composites at a fixed PVC/ABS mass ratio of 50/50 at various woodflour contents.

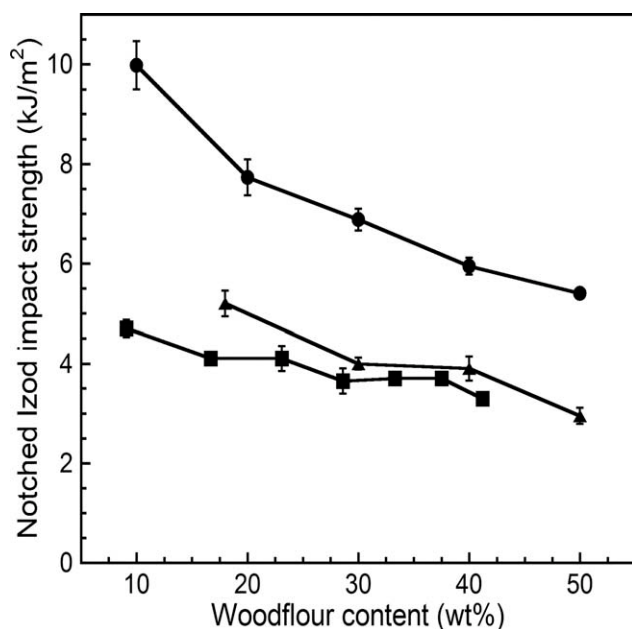


Figure 7 Impact properties of PVC/ABS/wood composites at a fixed PVC/ABS mass ratio of 50/50 at various woodflour contents comparing with PVC/wood and PE/wood composites. (●) PVC/ABS/Wood composites, (■) PVC/Wood composites,⁹ and (▲) PE/Wood composites.³³

interfacial interaction and better wetting of the matrix and the woodflour.

In Figure 7, the effect of woodflour content on notched Izod impact strength of the PVC/ABS/wood composites is illustrated. It can be observed that the impact strength of the composites was found to decrease with increasing woodflour content. The PVC/ABS/wood composite at 10 wt % of woodflour showed the impact strength of 9.98 kJ/m²; and the value was found to decrease with an increase of woodflour content to ~ 5.40 kJ/m² at 50 wt % of woodflour. A similar behavior was also found in various wood composites systems such as PVC wood, PE wood, and PP wood composites.^{32–34}

In comparison with PVC/wood composite at 50 wt % of woodflour, the notched Izod impact strength of the PVC/wood composite was reported to be 4.53 kJ/m², which was ~ 20% lower than that of the PVC/ABS/wood composite in our study. The differences in the impact strengths of the composites are mainly attributed to the inherent impact strength of the matrix and to some contribution from the interfacial interaction between the matrix and the woodflour. In comparison, the impact strength of PVC, PE, and PVC/ABS matrices are 4.7, 9.5, and 65 kJ/m², respectively.^{8,29}

In general, a decrease of impact strength of natural fiber/polymer composites was usually derived from debonding and friction effects, especially when no fiber surface treatments were applied. As the woodflour content was increased, the ductile portion

(PVC/ABS matrix) automatically reduced, thus decreasing the composites toughness.³⁸

In comparison with polyolefin wood composites, the impact strength of PVC/ABS wood composites was found to provide substantially higher value. Selke and Wichman³² reported an impact strength value of PE wood (at 40 wt % of woodflour) to be about 4 kJ/m². In addition, Oskman and Clemons³⁴ studied the mechanical properties of PP/wood composites and reported that the impact strength value (at 40 wt % of woodflour) was only about 2.6 kJ/m².

The fracture surface micrographs of PVC wood composite and PVC/ABS wood composite under impact load are shown in Figure 8(a,b). It could be noticed that the rougher surface of PVC/ABS matrix in Figure 8(b) might explain the higher impact strength of its wood composite than the smoother surface of PVC matrix. That means that the use of an inherently high impact strength PVC/ABS blend

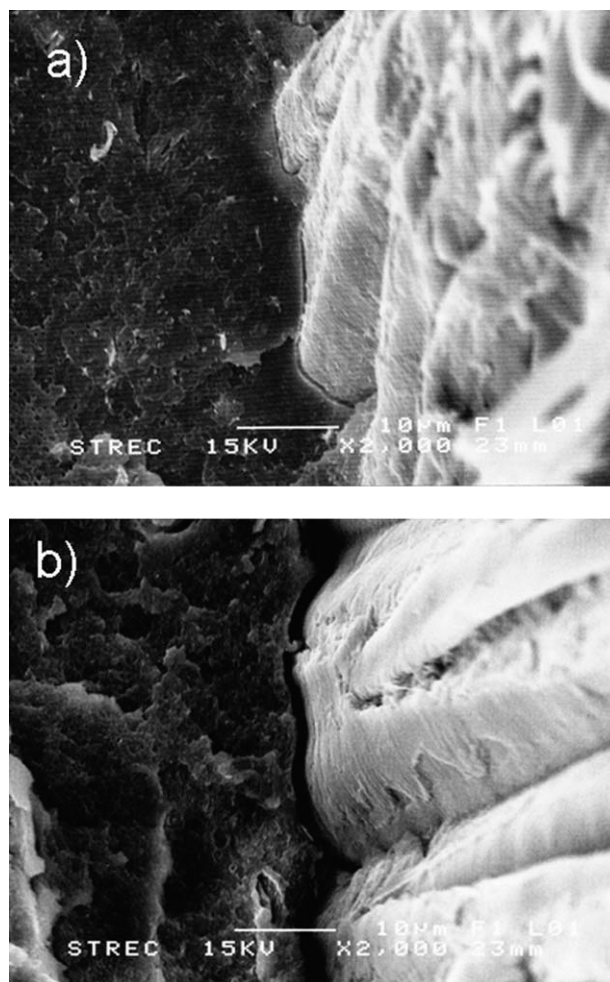


Figure 8 Scanning electron microscope micrographs of fracture surface under impact load of wood composites: (a) PVC with woodflour 30 wt %; (b) PVC/ABS 70/30 with woodflour 30 wt %.

as a matrix in WPC could dissipate greater impact energy thus provided greater impact strength wood composite than the PVC matrix. The effect is evidently more pronounced when the amount of wood flour was reduced.

Thermal properties of woodflour-filled PVC/ABS blends

From DMA analysis in Figure 9, the effects of adding 50 wt % of woodflour on storage modulus and glass transition temperature (obtained from the peak of loss tangent) of the composites are depicted. From the figure, the storage modulus at room temperature of the PVC/ABS wood composites substantially increased from 2.46 GPa of the unfilled PVC/ABS to the value of 5.98 GPa with an addition of the woodflour. This phenomenon could be attributed to the reinforcing effect due to the higher modulus value of the natural filler than that of the polymer matrix. Furthermore, two glass transition temperatures of the composites as obtained from the two loss tangent peaks were also observed due to the use of the partially miscible PVC/ABS at 50/50 mass ratio as the composite matrix. In addition, we also observed that T_{g1} of PVC/ABS increased from about 95–98°C with an addition of 50 wt % of woodflour, whereas T_{g2} slightly increased from 116 to 122°C. The phenomenon suggested an existence of some interfacial interaction between the PVC/ABS and the woodflour that might have a restriction effect on the motion of molecular chains of the polymer matrix due to impediment of a more rigid woodflour particle.³⁰ The similar increase in T_g with an addition of woodflour was also observed in the system of PVC/sawdust composites reported by Sombatsompop et al.⁸

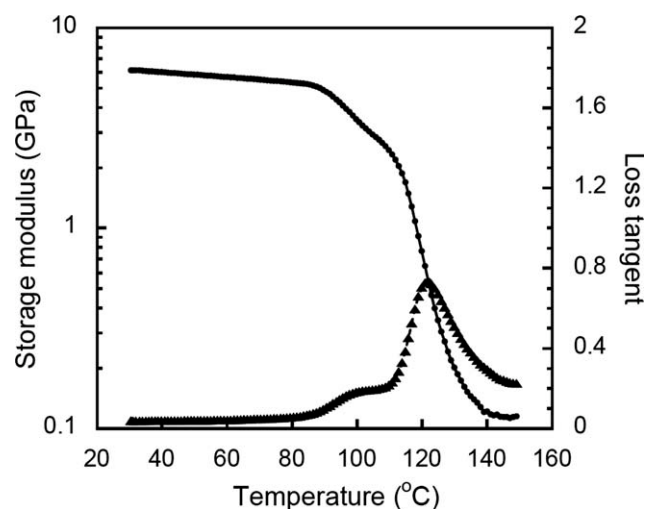


Figure 9 DMA thermograms of PVC/ABS 50/50 with wood flour 50 wt %.

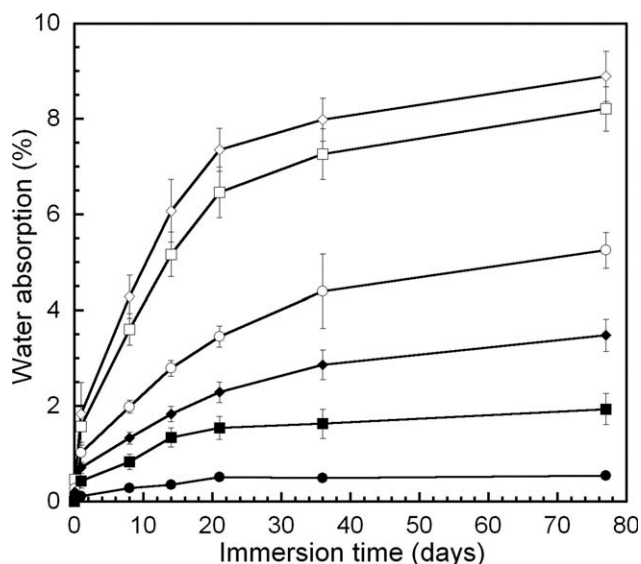


Figure 10 Water absorption of PVC/ABS/wood composites at a fixed PVC/ABS mass ratio of 50/50 and at various woodflour contents: (●) 0%, (■) 10%, (◆) 20%, (○) 30%, (□) 40%, and (◇) 50%.

Water absorption of woodflour-filled PVC/ABS blends

The effects of woodflour contents in the range of 0–50 wt % on water absorption of PVC/ABS wood composites are shown in Figure 10. At 24-h immersion, the water absorption of the unfilled PVC/ABS matrix was only about 0.1%. With an addition of 10 wt % of woodflour, the absorption behavior at 24 h provided insignificant change in its value. This is attributed to the ability of the PVC/ABS to cover or encapsulate most of woodflour particles, so that water cannot interact with the woodflour. At higher woodflour content, water absorption of PVC/ABS/wood composites was found to increase with increasing woodflour content, that is, the water absorption of the composite at 50 wt % of woodflour at 24-h immersion was ~ 1.8%. For the long-term immersion, the PVC/ABS wood composite at 50 wt % of woodflour rendered the water absorption value of up to 7.34 wt %. However, this value was still significantly lower than the water absorption in real wood, that is, in the range of 24–100%.³⁹ This was likely due to the hydroxyl groups in the structure of cellulose and hemicellulose of woodflour, which is higher hydrophilicity in nature than our PVC/ABS matrix.⁶

Consequently, the degree of water absorption of the composite increased with the woodflour loading. In principle, there are three main regions where the absorbed water in the composites can reside, that is, (1) gaps between woodflour and matrix, (2) lumen, and (3) cell wall. To develop composites with better mechanical properties and to decrease their moisture uptake, it is necessary to make the woodflour more

hydrophobic possibly by suitable treatment to strengthen interfacial interaction with the polymer matrix used. From the calculation based on eq. (2), the diffusion behaviors of our wood composites can also be classified as pseudo-Fickian type with the exponents ranging from 0.35 to 0.43.

CONCLUSIONS

Woodflour-filled composites based on the polymeric blends of PVC and super high-impact grade ABS were developed. The results revealed that the PVC/ABS matrix at a mass ratio of 50/50 rendered the highest impact strength that was noticeably higher than those of the parent polymers. Two distinct glass transition temperatures (T_g s) were observed, which indicated partial miscibility of the blends. The water absorption of the blends after immersion for 24 h was in the range of 0.04–0.2 wt %. Furthermore, the woodflour-filled PVC/ABS composites exhibited a systematic increase of flexural modulus and glass transition temperature with an increase of woodflour content, whereas flexural strength slightly decreased with the amount of the woodflour. In addition, the impact strength of the PVC/ABS composites was significantly higher than those of PVC composites and polyethylene composites at the same filler content. Finally, water absorption experiment revealed that water diffusion behavior of PVC/ABS and its wood composites was closed to pseudo-Fickian type.

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