Mechanical Properties of Reinforced Polyvinyl Chloride Composites: Effect of Filler Form and Content

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ABSTRACT: In this study, to understand how cellulosic materials characteristics influence the mechanical properties of polyvinyl chloride (PVC) composites, we first studied the effect of different types (wood flour and pulp fiber) and contents on selected mechanical properties of uncompatibilized wood plastic composites. We then compared the properties of hybrid composites to those of composites reinforced with wood flour or fiber, individually. PVC as polymer matrix and cellulosic materials were compounded by twin-screw extrusion and test specimens were prepared by injection molding. All tested properties vary significantly with filler form or content. With the addition of wood flour, the tensile strength moderately increases, but with the addition of hybrid filler and pulp fiber, it increases significantly. Notched impact strength increased with increasing particle size. Pulp fiber resulted in higher strength at the 40 wt % level compared with the strength properties of wood flour composites. The higher aspect ratio of the fiber had significant effect on the mechanical properties. Increasing filler load improves the strength of the composite up to a load of 40 wt %, further increase in the filler loading (>40 wt %) results in a decrease in properties, due to the to filler agglomerates. Incorporation of 25% weight fraction of wood pulp fiber in hybrid composites gave the highest values of composite properties. The mechanical properties of hybrid composites are found to be much higher than that composites filled with wood flour. Finally, results showed that the hybrid composites had the maximum improvement in selected mechanical properties. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 120: 1788–1793, 2011

Key words: fibers; reinforced composite; mechanical properties; polyvinyl chloride (PVC)

INTRODUCTION

In recent years, the use of natural fibers as reinforcing agent in the manufacture of wood plastic composites (WPCs) has attracted a number of researchers and manufacturing engineers. Such materials offer significant advantages, which justify their use. Natural fibers are obtained from different resources (i.e., wood, nonwood, agro-residues), they are available in large quantities, light weight, cheap, and it can be added to commodity matrices in considerable amounts thus offering economically advantageous solutions.[1] These materials are available in many different forms and produce different properties when added to thermoplastics. Natural fibers may be used in the form of particles, fiber bundles, or single fibers, and may act as a filler or reinforcement for plastics. One of the most common natural fibers used in the thermoplastics industry is wood flour, which is produced commercially from post-industrial sources such as planer shavings and sawdust. The scrap wood is sourced for species purity and then ground to specific particle size distributions. In general, wood flour is used as a filler for plastic, which tends to increase the stiffness of the composite but does not improve its strength. Natural fibers can be used to reinforce rather than fill plastics, which increase strength as well as stiffness. Wood and other lignocellulosic fibers typically have higher aspect ratios than that of wood flour. At a critical fiber length, stress is transferred from the matrix to the fiber, resulting in a stronger composite.[2,3]

Natural fibers were initially used for reducing and disposing of large amounts of natural fiber waste materials and for cost reduction. But, they are now preferably used as reinforcing materials in polymers, and offer low cost and low density products. WPCs have been extensively developed for a wide range of applications, including decking, window and door profiles, automobile paneling, panel inserts, packing, and gardening (such as flower pots) which were classified as decorative materials. However, the applications for WPC products in structural and engineering constructions are still questionable as a result of their strength limitations.[4]

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In accordance with literature information, there have been a number of methods to improve the structural and engineering properties of the WPC products, these including addition of synthetic fiber, metal inserts, and selection of appropriate processing techniques.[5–8] Another method for producing WPCs with high performance is hybridization.[9,10] Hybrid composites are materials made by combining two or more different types of fibers in a common matrix. These composites have been developed to provide synergistic properties of the chosen fibers and matrix. Hybridization of two types of fibers, such as wood flour and fibers, which having different lengths and diameters, offers some advantages over the use of either of the fibers alone in a single polymer matrix and it can improve the properties of WPCs. As a consequence, a balance in performance and cost could be achieved through proper material design. However, only a few studies on the mechanical properties of hybrid composites with two natural fibers are available today and in most cases synthetic and natural fiber reinforced hybrid composites are studied.[11–18]

Currently, poly(vinyl chloride) (PVC) is one of the most attractive thermoplastic in making the WPCs which are mainly used as the exterior building components. PVC manufacturing companies are developing WPC based on PVC, due to the distinct performance advantages offered by this thermoplastic. The flexural strength and the modulus of WPC containing PVC are superior to those of WPC based on either poly(propylene) (PP) or poly(ethylene) (PE). Compared with wood-PP and wood-PE composites, wood-PVC composites provide superior properties in terms of creep resistance, weatherability, and flame retardancy. The demand for wood-PVC composites is expected to increase by 200% a year until 2010, against 130% for PP-based composites, and 40% for PE based composites.[19]

The objectives of this study were as follows: (1) to investigate the influence of different forms (wood flour and pulp fiber) and contents of cellulosic materials on the mechanical properties of PVC composites, (2) to evaluate the effects of the different mixing formulations of cellulosic materials on the mechanical behavior of the hybrid composites, and (3) to compare mechanical properties of wood flour and fiber reinforced PVC composites with hybrid composites.

EXPERIMENTAL

Materials

Two different forms of poplar (*Populus deltoides*) were used as reinforcing filler: pulp fiber (PF), and wood flour (WF). The fibers were produced by chemi-mechanical pulping process. The fresh sawdust from local mill was ground into flour form using a Thomas-Wiley mill to pass through a 45-mesh screen (354 μm), and then was dried to less than 3% moisture content. The important physicochemical constituents of the used cellulosic materials are presented in Table I and Figure 1.

TABLE I
Physico-Chemical Properties of Used Cellulosic Materials

Properties	PF	WF
Chemical properties		
Cellulose, %	60 ± 4.4	68 ± 5.7
Hemicellulose, %	15 ± 2.0	17 ± 3.1
Lignin, %	12 ± 3.1	24 ± 3.7
Physical properties		
Fiber length, mm	0.79 ± 0.08	0.032 ± 0.05
Fiber width, μm	18.6 ± 2.5	16.3 ± 4.4
Aspect ratio	64 ± 16	2.2 ± 0.4

PVC, a product (Poliran PV *S*-6058) of Bandar Imam Petrochemical Company., Iran, was used as polymer matrix. The PVC polymer was in the form of powder with a bulk density of 550–610 g/L and a viscosity of 85–92 cm^3/g.

Preparation of composites

Various blending formulations and their mass ratios are listed in Table II. It is to be noted that all the blends for hybrid composites were made with cellulosic materials (wood flour + pulp fiber) to PVA mass ratio of 40/60 (w/w); this value amount was selected because it is typical of many industrial formulations and represents optimum balance between performances and cost.

In the first stage, the raw materials were physically premixed based on the formulations before being fed into the first zone of the extruder. All the experiments were performed in a corotating twin-screw extruder (Collin). The melt temperature at the die was 185°C and the rotation speed was 60 rpm. The extruded strand was passed through a water bath, granulated, and dried at 105°C for 24 h to remove any moisture. The resulting granules were subsequently injection molded at 190°C to produce standard ASTM specimens.

Mechanical testing

The mechanical properties of the composites were evaluated through tensile, flexural, and impact properties. After conditioning (50% relative humidity and 23°C), all the specimens were tested in accordance with ASTM standard D638 for tensile properties, ASTM D790 for flexural properties and D256 for notched Izod impact strength. Tensile and bending

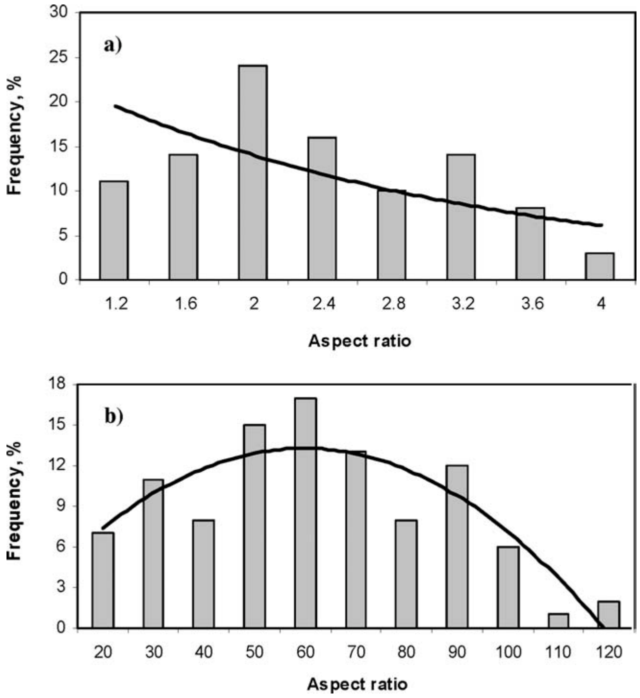

Figure 1 Frequency distributions of the aspect ratios of (a) wood flour and (b) fiber.

tests were conducted using an Instron Universal Testing Machine (model 1186). Elongation at break was calculated from force versus deformation traces recorded on the tensile specimens at 5 mm/min cross-head speed. A pendulum impact tester (Zwick 1446) was used for the Izod impact test. For each treatment level, five replications were conducted.

TABLE II
Formulations of the Used Experimental Composites

Codes	WP wt %	WF wt %	PVA wt %
A1	10	–	90
A2	20	–	80
A3	30	–	70
A4	40	–	60
A5	45	–	55
A6	50	–	50
A7	55	–	45
B1	–	10	90
B2	–	20	80
B3	–	30	70
B4	–	40	60
B5	–	45	55
B6	–	50	50
B7	–	55	45
AB1	35	5	60
AB2	30	10	60
AB3	25	15	60
AB4	20	20	60
AB5	15	25	60
AB6	10	30	60
AB7	5	35	60

Scanning electron microscopy

Surface scanning of the composites was done by the SEM (WEGA-II TESCAN). The accelerating voltage was 20 kV. The samples were first sputter coated with a thin layer of gold and then observed at magnification of 2000×.

RESULTS AND DISCUSSION

Effect of filler form

The effect of filler form on the mechanical properties investigated in this study is highly significant (Fig. 2). In general, increasing cellulosic materials improve the mechanical properties. This result is consistent with our previous reports on wood fiber thermoplastic composites.[10,20] The results showed that tensile strength of the composites was enhanced with addition of fillers in both forms (wood flour and fiber). However, the hybrid composites display different behavior. WPCs made with hybrid material

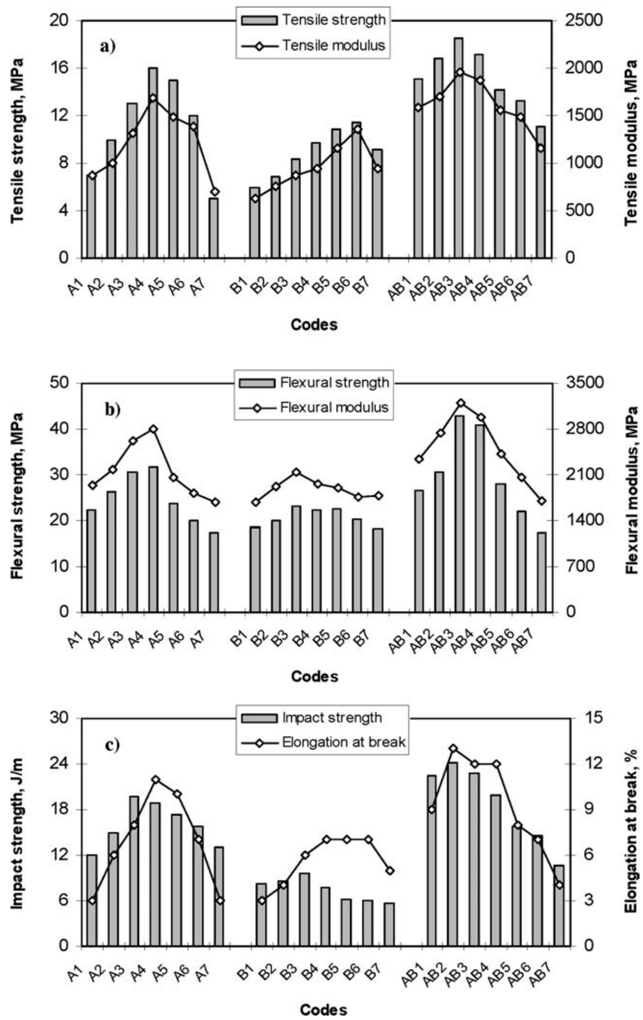

Figure 2 Comparison of mechanical properties of composites with various blending formulations.

and pulp fiber exhibit the highest tensile strength, whereas WPCs filled with wood flour show the lowest properties. Maximum tensile strength ranges from 18.8 MPa and 16 MPa for hybrid and pulp fiber WPCs, respectively, while maximum tensile strength is $\sim$ 10.9 MPa for composites made with wood flour. In other words, strength of tensile of composites enhanced at least 1.8-fold when hybrid or fiber is added. As can be seen, the tensile modulus of the produced composites varied from 629 to 1958 MPa. These results are in good agreement with previously reported data.[21–23]

One of the most important parameters controlling the mechanical properties of short fibers composite is the fiber length or more precisely its aspect ratio (length/width). A high aspect ratio is very important in fiber reinforced composites, as it indicates potential strength properties.[24] Stark and Rowlands[3] reported that aspect ratio, rather than particle size, has the greatest effect on strength and stiffness. The wood flour, which had a very low aspect ratio, showed inferior strength to the composites filled with pulp fiber (Fig. 1). In other words, the aspect ratio of the fiber is higher than that of the wood flour, which permits better stress transfer between the matrix and the fibers. Because of the potential of improved mechanical properties with fillers of greater aspect ratios, there has been a continuing interest in the use of individual fibers rather than wood flour as reinforcement in WPC.

Flexural strength shows a similar trend as that of tensile strength and shows a maximum improvement of 42.8 MPa strength at 40 wt % for sample AB3. The explanation is similar to that of the tensile properties. The lower flexural strength exhibited by the composites with wood flour compared to pulp fiber and hybrid composites be attributed to the smaller particle size of wood flour, compared to fiber. Flexural strength development also demonstrates that filler form has greater influence at a constant filler load, with $\sim$ 84% higher strength when hybrid material is used. On the other hand, the incorporation of wood flour in the PVC matrix steadily increases flexural strength, independently of filler content. This is a common tendency that has been reported with organic filler as well.[25]

Figure 2(c) reports the result of notched Izod impact strength measurement as function of filler forms. The pulp fiber and hybrid material appeared to significantly improve impact strength in comparison with the wood flour. This was expected because fiber should be more resistant to crack propagation in the matrix. This is consistent with the results reported by most authors.[26–29] The presence of wood flour in the PVC matrix provides points of stress concentrations, thus providing sites for crack initiation. Another reason for decrease in impact strength may be the stiffening of polymer chains due to bonding between wood flour and matrix. For high impact properties, in fact, a slightly weaker adhesion between fiber and polymer is desirable, as it would result in a higher degradation of impact energy, supporting the so-called fiber pull-out.[30]

The elongation at break values of the composites are plotted against cellulosic type and content in Figure 2(c). The two cellulosic materials have different effect on this property as well, i.e., they influence the deformation and failure of the composites slightly differently. Composites filled with wood flour show brittle behavior, with 40% lower elongation at break than hybrid composites. It is interesting to note that the deformability of composites containing the wood pulp and those prepared by hybrid material is similar, although interfacial adhesion must be different in the two cases. As observed from the graph, the hybrid composites exhibited a positive elongation at break effect with the addition of wood pulp fibers. The positive hybrid effect of the elongation at break in hybrid composites was also observed by Hariharan and Abdul Khalil[31] concluded that in a hybrid composite, the addition of high elongation fibers with low elongation fibers often increased the elongation at break of the hybrid composite than the composite made from low elongation fibers.

Effect of filler content

Fiber content is an influential factor in WPC processing and properties. Zhang et al.[32] investigated the effects of fiber content on mixing torque and rheological properties. They concluded that increased pulp fiber content results in increased steady state torque and viscosity. Lu and coworkers[33] concluded that the mechanical properties of the resultant WPC increase only at low weight percentages of wood filler. They found that tensile and flexural strengths reach a maximum at 15 and 35 wt % wood particle contents, respectively, and gradually decrease with a further increase in wood particle content. Dänyädi et al.[1] reported that, at large wood content, considerable particle aggregation takes place, leading to lower strength due to the filler's failure to sustain the stress transferred from the polymer to the matrix.

From Figure 2(a), it is evident that moderate increase in tensile strength occurred upon filling the polymer matrix with fibers, indicating a considerable reinforcing effect from these fibers. The phenomenon was stronger for the composites made by hybrid fibers. Similar behavior can be observed in Figure 2(b,c), where the significant increase in flexural and notched impact is plotted versus filler content. It is interesting to note that as filler content increases mechanical properties increase up to a fiber load of

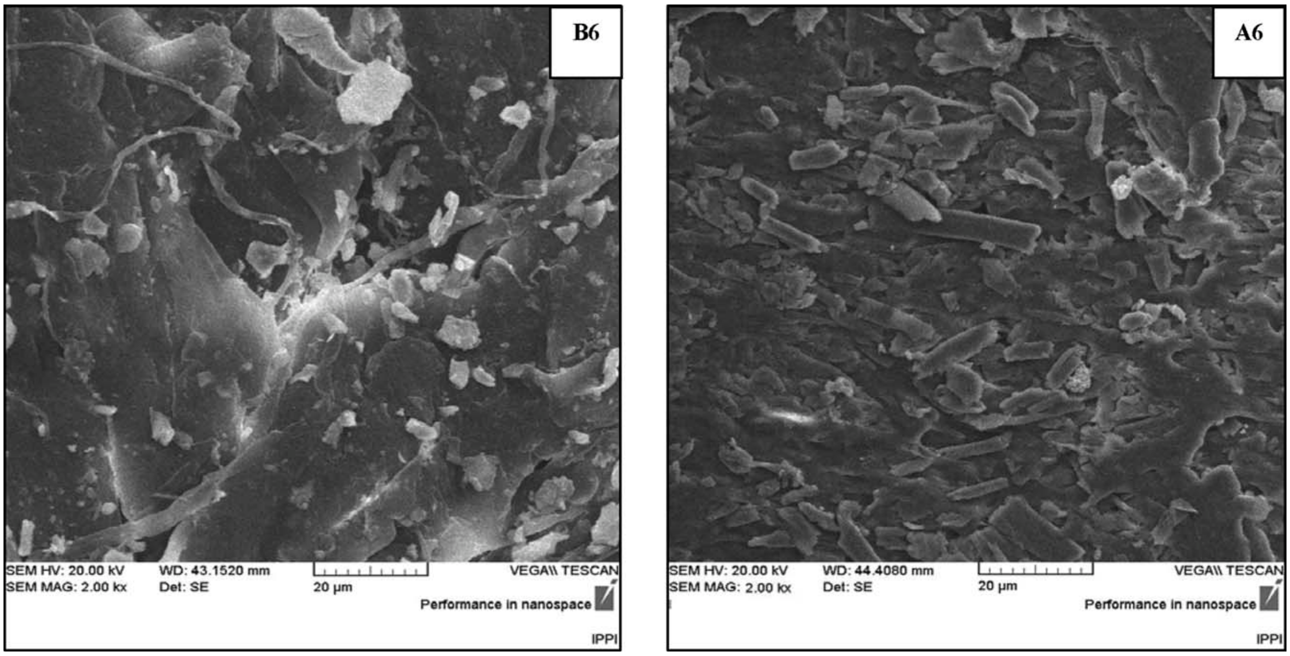

Figure 3 SEM micrographs of agglomerates in samples containing 50 wt % wood flour (B6) and wood fiber (A6).

40 wt %. Further increase in the filler loading results in a decrease in strength properties. As shown in Figure 3, this is attributed to filler agglomerates. Wood flour is easily agglomerated, which is the characteristic of this filler, and the presence of these agglomerates results in the generation of flaws, resulting in the creation of voids between the filler and the matrix polymer. This causes the mechanical properties of the wood flour composites to be reduced, as compared with the composites filled with pulp fiber.

Furthermore, increase in the wood flour loading was almost unaffected to improve the flexural strength of the composites. Similar results have been published by Karmarkar et al.[30] who studied the properties of wood plastic composites. Their data show that the tensile strength of wood fiber/PP composites increases with increasing fiber content. The possible reasons proposed for this kind of behavior may be due to the improved interfacial adhesion between the matrix and fibers. In addition, a higher filler content results in more (and much probably) void formation during processing, which leads to micro crack formation under loading and therefore reduces the mechanical properties.

Variation of notched Izod impact strength with increasing content of filler is shown in Figure 2(c). The notched impact strength of the composites shows a remarkably improvement in the strength by the addition of the pulp fiber and hybrid materials. It is worth mentioning that the enhancement in the uncompatibilized samples could be attributed to the more homogeneous dispersion of the filler.

In hybrid composites, the properties of the composites are mainly dependent on the percentage of elongation at break of the individual fibers. Figure 2(c) shows the variation of elongation at break with both wood flour and wood pulp fiber loading. The value of elongation at break shows an improvement with an increase of wood pulp fiber content in hybrid composites while a reverse trend is observed as the wood flour loading increased in the composites. With an increase wood flour over wood pulp fiber content, the hybrid composites show a reduction in elongation at break of composites. Among the hybrid composites, the incorporation of 30 wt % wood pulp fiber exhibits the highest elongation at break of composites, whereas composites with 35 wt % weight fraction of wood flour are found to have the lowest value of these properties. This phenomenon was due to the fact that wood flour is a low elongation fiber compared to the wood pulp fiber. Thus, wood pulp fiber has a high strain to failure characteristic (3–11%) compared with the low extensibility of glass fiber (3–7%).

CONCLUSIONS

On the basis of the results of this study the following conclusions can be drawn.

- Adding pulp fiber rather than flour increases mechanical properties such as tensile, flexural, and notched impact strengths. However, processing difficulties, such as feeding and metering low-bulk-density cellulosic materials (in

particular fibers), have limited the use of high content WPCs.
- Although incorporating fiber into the PVC matrix effectively improves strength properties, this improvement comes at proper filler loading (40 wt %). Thus, depending on end use, the composite should be optimized for either strength or price by adjusting both filler form (wood flour, fiber or their combination) and concentration.
- The reduction in mechanical properties of composites in the higher contents (>40 wt %) of filler is attributed to filler agglomerates. The formation of wood flour agglomerates is easier than that of pulp fiber.
- The results of this study clearly support the use of higher aspect ratio as fibers for increasing the strength of wood plastic composites.
- Composites made with hybrid materials exhibited superior mechanical properties compared to the wood flour or even fiber-filled composites. All these improvements in the hybrid composite properties are mainly due to the high strength and modulus value of wood pulp fiber than the inferior properties of the wood flour itself. Among various blending formulations, composites AB3 had the optimum improvement in selected mechanical properties.

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