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Effect of Wood Flour Content and Cooling Rate on Properties of Rubberwood Flour/Recycled Polypropylene Composites

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Abstract. The present article summarizes an experimental study on the mechanical and thermal behavior of recycled polypropylene composites reinforced with rubberwood flour. Different compositions were varied to investigate mechanical strengths, melting temperature, storage modulus, and loss modulus. It was observed that the tensile and flexural strengths decreased with the increase of wood flour content. Furthermore, the air cooled composites showed improved properties in comparison with the water cooled composites. The melting and crystallization temperature results presented a weak influence of increased wood flour content on composites. However, dynamic mechanical thermal analysis showed an increase in the storage and loss modulus.

Introduction

Fiber reinforced polymers are significantly growing in construction-material due to their light weight, ease of installation, low maintenance, tailor made properties, and corrosion resistance [1]. Wood flour have been primarily used by the plastics industry as an inexpensive filler to increase the strength and stiffness of thermoplastic and to reduce raw material costs [2]. However, the use of wood flour reinforced plastics may cause to influence mechanical and thermal properties. Polypropylene (PP) is one of the most well-know plastics that have been widely used in wood-plastic composite (WPCs) industries because of its high strength and ease of manufacturing and recycling. Extrusion is basically used to fabricate WPCs. The extruder of a plastic is heated to a temperature above its glass transition point before extruding through the open die. The extrudate is consequently cooled down to lower its transition temperature while the molecules are still under stress. The molecules will become frozen whilst in an oriented state. Such an orientation significantly affects the properties of the extrudated plastic [3].

There are numbers of published studies on the reinforcement of virgin polypropylene with wood flour relating to the results of mechanical and thermal properties. However, studies on WPCs based on recycled polypropylene (rPP) are very limited. Ndiaye et al [4] studied polypropylene-wood flour composites that were blended with different contents of maleated polypropylene (MAPP) and clay. They found that the addition of MAPP or clay in the formulation greatly improved the dispersion of the wood flour in the composite. Lisperguer et al [5] have shown that the glass transition values of the recycled polystyrene (rPS) and wood–rPS composites were higher than those of the virgin PS and wood–virgin PS composites. The use of rPS increased the stiffness and flexural modulus of the composites. Adhikary et al. [6] found that mechanical properties of wood-based recycled plastics of high-density polyethylene (HDPE) were as good as the composites made from virgin-based HDPE. Wood flour content and oriented state of cooling significantly affect on the properties of composites. This research aims to study the use of rubberwood flour and Post-Consumer Recycled PP (PCR-PP) for the production of the composites. The effects of the wood flour content and oriented state of cooling on the mechanical and thermal properties were also investigated.

Experimental

Materials and sample preparation. Rubberwood flour (RWF) from S.T.A Furniture Group Co., Ltd. in Songkhla, Thailand, was used as lignocellulosic filler. The important chemical compositions are listed in Table 1. The recycled polypropylene (rPP) was supplied by Withaya Intertrade Co., Ltd. in Samutprakarn, Thailand in the pellet form under the trade name WT001. Prior to blending, the RWF was sieved through 80 meshes and dried in an oven at 110^{o}C for 8 h. The dried wood flour was stored in a sealed plastic container to prevent the absorption of water vapor. Each formulation in Table 2 was weighed and stirred for 5 min to obtain uniform dispersion. The twin-screw extruder model SHJ-36 from En Mach Co., Ltd was used to make the specimens. The temperature profile in the extruding process was 145-180^{o}C with a revolution of 100 rpm. The temperature of composite extrudates was lower down in atmospheric air cooling (AC) and water cooling (WC). Subsequently, the specimens were machined corresponding to ASTM for mechanical testing.

Table 1 Chemical composition of rubberwood [7]

Chemical constituents	Ash	Extractive	Holocellulose	Lignin	Cellulose
Composition (%)	1.02	2.86	73.84	21.42	45.84

Table 2 Compositions of the WPCs

Sample ID	rPPRWF0	rPPRWF28	rPPRWF33	rPPRWF37	rPPRWF41	rPPRWF44
rPP (wt %)	100	71.4	66.7	62.5	58.8	55.5
RWF (wt %)	0	28.6	33.3	37.5	41.2	44.5

Mechanical testing. The tensile and flexural properties were measured on the Instron Universal Testing Machine (Model 5582). Tensile tests were carried out according to ASTM D 638-91 Type 1 at a testing speed of 5 mm/min. Three-point flexural tests were performed in accordance to ASTM D 790-92. Tests were conducted at a crosshead speed of 2 mm/min. All the tests were carried out at room temperature (25^{o}C) with five replications.

Morphological observation. The state of dispersion, interface adhesion, and voids of the wood flour in the polymeric matrix was analyzed with scanning electron microscope (SEM). A FEI Quanta 400 microscope (Czech Republic) working at 15 kV was used to obtain microphotographs of the composites surface.

Thermal testing. The effects of filler concentration on melting and crystallization temperatures (T_m and T_c, respectively), and the heat of fusion (Δh_f), were ascertained from differential scanning calorimetry (DSC-7, PerkinElmer, USA). The DSC was typically operated at heating and cooling rates of 10^{o}C/min, respectively. The dynamic mechanical thermal analysis (Rheometric scientific DMTA V, USA) was also performed to measure storage (E′) and loss (E″) modulus as a function of temperature at small strain amplitudes and a fixed frequency of 10 Hz in the linear viscoelastic limit. The specimen size of 10×35×3 mm was prepared and investigated.

Results and Discussion

Mechanical properties. The effect of different amounts of wood flour on the mechanical properties of wood/rPP composites was shown in Table 3. It was found that increasing the wood flour content resulted in decreasing tensile and flexural strengths for both in air and water cooling. The result is in good agreement with Prachayawarakorn et al [8] and Sombatsompop et al [9]. The decreases in the tensile and flexural strengths, due to the addition of wood flour, were associated with poor dispersion and adhesion of the wood particles in the matrix. From the SEM micrographs shown in Figure 2, it can be seen that the wood particles (fibers) tended to cling together, due to strong interfiber hydrogen bonding, and resist dispersion as an individual fiber while the fiber content was increased from 28.6 to 44.5 wt% [9]. Likewise, the enlarging wood flour content increased the poor interfacial adhesion and void amounts, and thus reduced mechanical strengths of the composites.

Table 3 Effect of wood flour loading on the mechanical properties

Wood flour content (wt%)	Air cooling (AC)				Water cooling (WC)			
	Tensile strength (MPa)	Tensile modulus (MPa)	Flexural strength (MPa)	Flexural modulus (MPa)	Tensile Strength (MPa)	Tensile modulus (MPa)	Flexural strength (MPa)	Flexural modulus (MPa)
0	25.9	316.9	44.8	1445.5	22.6	308.2	41.2	1387.4
28.6	22.3	419.5	39.1	1927.1	20.2	356.2	36.4	1618.5
33.3	20.9	585.9	38.2	1958.6	17.8	430.8	34.8	1644.1
37.5	19.5	625.5	34.9	2171.1	17.4	441.5	31.1	1760.2
41.2	18.6	772.8	34.1	2402.6	17.0	523.6	31.6	1853.6
44.5	18.2	708.0	30.5	2583.4	16.0	556.5	30.5	2004.1

The tensile and flexural modulus (Table 3) of the composites increased with increasing wood flour content. The increase of the composite modulus with wood flour content was caused by the fact that the wood flour is more rigid phase compared to the polymer matrix [8]. Furthermore, the air cooled composites appeared to give higher mechanical properties, tensile and flexural strengths, than that of the water cooled composites. The reason is the PP matrix instantaneously cooled below its glass transition temperature while the molecule was still under stress. The molecules therefore became frozen whilst in an oriented state [3].

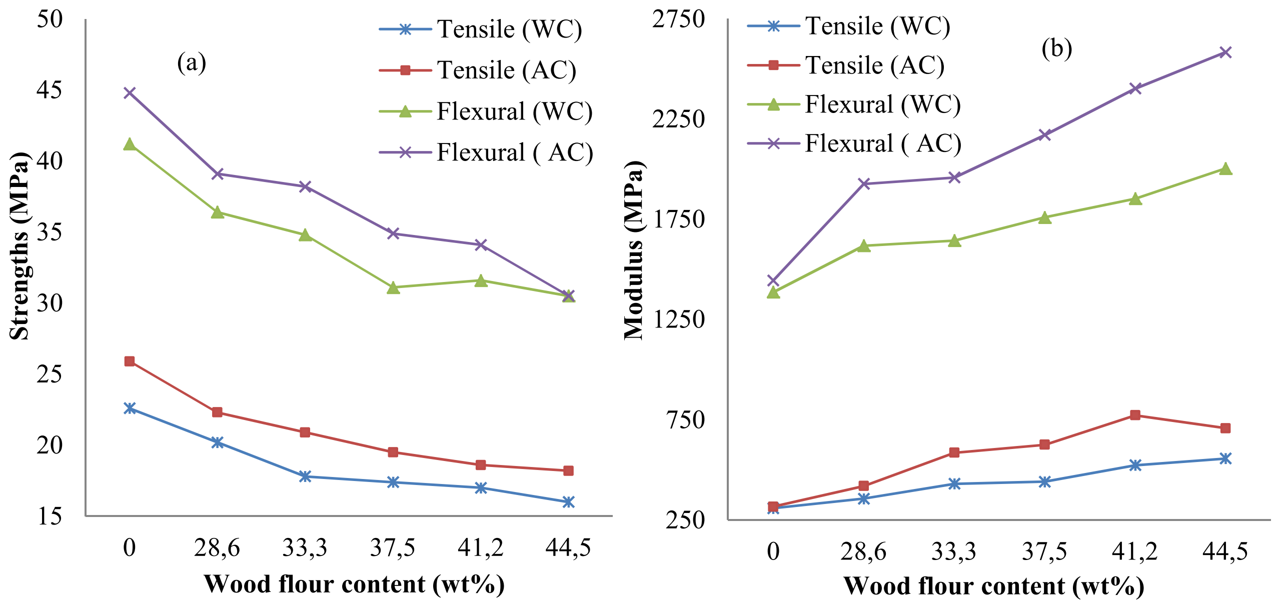

Fig. 1 Mechanical behavior of RWF/rPP composites versus differential wood flour content (a) tensile and flexural strengths, (b) tensile and flexural modulus

Morphological analysis. The SEM micrographs of rPPRWF28, rPPRWF37, and rPPRWF44 composites are shown in Figures 2 (a), (b), and (c), respectively. It can be seen that wood flour exhibit the shape of irregular short fibers in the composites. The analysis of the wood flour dispersion shows that the addition of higher wood flour content to rPP [Figs. 2(b) and 2(c)] seemed to present a higher number of agglomerations. This behavior probably means that poorer particle dispersion in the rPP matrix has occurred [10]. It is known that wood flour has a great tendency to form agglomerates, in fact, agglomeration is a well-known phenomenon, and its probability increases with decreasing particle size. The occurrence and extent of agglomeration are determined by the relative magnitude of the forces, which either bind together the particles or try to separate them [11]. This result is in accordance with the mechanical results found in this work.

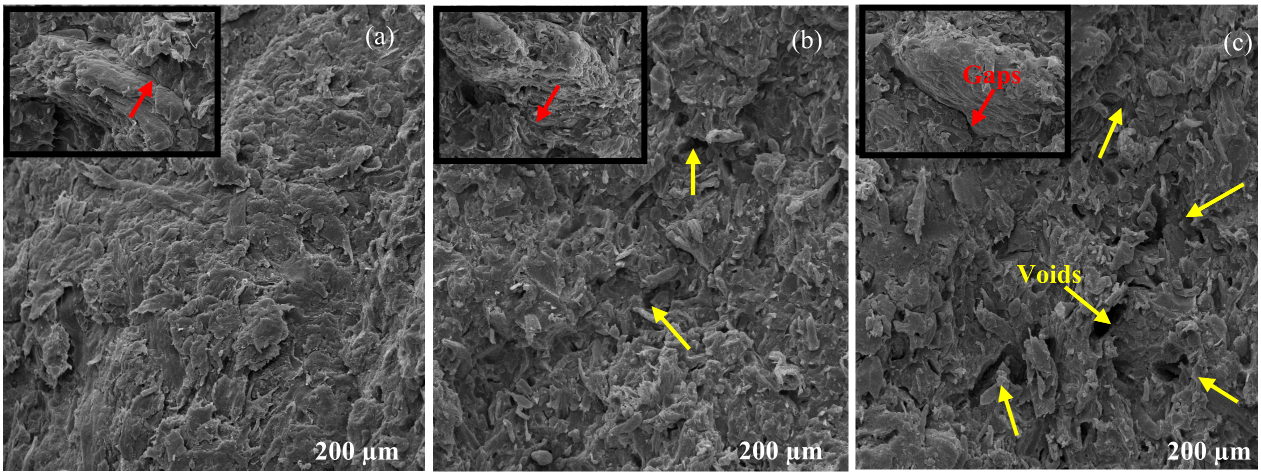

Fig. 2 SEM micrographs of rPP composites containing (a) 28.6, (b) 37.5 and (c) 44.5 wt% wood flour contents

Thermal analysis

Differential scanning calorimeter. To ascertain the roles of fillers phase and concentration on the thermal behavior of RWF/rPP composites, DSC has been performed to identify T_m, T_c, Δh_f and % crystallinity as tabulated in Table 4. The results indicate that these four formulations exhibit nearly identical peak melting and crystallization temperatures at about 159 and 119°C, respectively, and corresponding crystallinity varies by less than 5%. The crystallinity of the composites, however, was slightly lowered compared to the neat PP. In good agreement with the findings of Xu et al [12] in their study of UHMWPE composites containing carbon black (CB), they also report that the crystallinity of UHMWPE is unaffected by the presence of CB and infer from these results that the polymer crystal size is independent of the filler loading level. Similar behavior is observed in the present HDPE/UHMWPE blends filled with CB, in which the crystallinity varies by less than 5% [13]. However, the cooling rate significantly affected T_m, T_c, and crystallinity. The plastics that were heated to a temperature above its glass transition point and then are slowly cooled below its transition temperature gain higher crystallinity than the instantaneous cooling.

Table 4 Thermal properties of the WPCs containing different concentrations of wood flour and cooling orientation state

Wood flour content (wt %)	T_m (°C)		T_c (°C)		Δh_f (J/g)		Crystallinity[a] (%)	
	AC	WC	AC	WC	AC	WC	AC	WC
0	160.2	156.8	119.9	116.8	77.2	68.5	46.8	41.5
28.6	160.0	159.2	119.9	119.3	56.6	52.4	48.0	44.5
37.5	159.7	159.2	119.3	119.3	45.4	42.5	44.0	41.2
44.5	159.8	158.7	119.3	118.8	44.3	39.0	48.4	42.6

[a]Calculated from the ratio of the measured Δh_f to that of a 100% crystalline polypropylene (165 J/g)

Dynamic mechanical thermal analysis. The variation of E′ and E″ for pure rPP and RWF/rPP composites was shown in Figure 3. It showed that composites had higher storage and loss modulus than pure rPP through the whole temperature range of the study due to the reinforcement effect of wood flour. The wood flour can cautiously carry in decreasing of E′ and E″. Likewise, the enlarging wood flour content can increase the storage and loss modulus. This is consistent with the results reported in the literature [14]. The addition of wood flour leads to the increase in both elastic and viscous abilities of composites under the dynamic load [14]. The E′ and E″ gradually decreased in the range of 50 to 160°C, but instantaneously decreased at 160 to 210°C. This was due to the melting effect of the PP matrix. Furthermore, WPCs with air cooling had higher storage and loss modulus than the one with water cooling. Because melting temperature and crystallinity of WPCs with air cooling affected greater than that of water cooling based on DSC analysis.

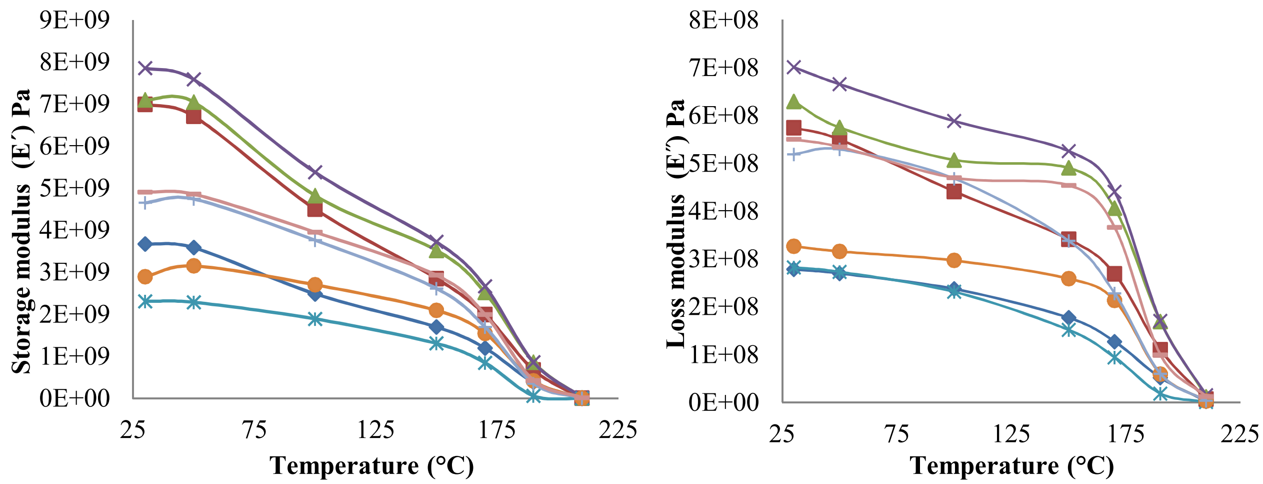

Fig. 3 Variation of storage (E´) and loss (E″) modulus with temperature for air cooling; rPPRWF0 (◊), rPPRWF28 (□), rPPRWF37 (Δ), rPPRWF44 (×), and water cooling; rPPRWF0 (⁎), rPPRWF28 (○), rPPRWF37 (+), and rPPRWF44 (‒). The solid lines serve to connect the data.

Conclusions

Natural rubberwood flour (RWF) can be simply blended with recycled polypropylene (rPP) compound to produce cost-competitive and woodlike composites with satisfactory properties [15]. The mechanical and thermal properties of composites were affected by the concentration of rubberwood flour. It was found that the mechanical properties of RWF/rPP composites decreased with increasing RWF loading due to the poor interaction between the hydrophilic filler and the polymer matrix [16]. The mechanical results were corroborated with morphological evidence. DSC analysis showed that the increase of wood flour content was insignificant effect to the degree of crystallinity because of stability of the crystalline portion in the material. However, the degree of crystallinity related with oriented state of cooling. Further, DMTA displayed an increase in thermal stability of rPP matrix filled with wood flour reinforcement [14]. Plot of storage and loss modulus with temperature showed an increase in the magnitude of the peaks with the enlarging wood flour reinforcement and air cooling. On the basis of these studies, it can be concluded that wood flour decreased mechanical properties of composites, but it increased the thermal stability of composites. The cooling rate was significantly affected on mechanical and thermal properties.

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