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ORIGINAL ARTICLE

Effect of organoclay platelets on the mechanical properties of wood–plastic composites formulated with virgin and recycled polypropylene

HOSSEIN KHANJANZADEH[1], TAGHI TABARSA[1], ALIREZA SHAKERI[2] & ASGHAR OMIDVAR[1]

[1]*Department of Wood and Paper Technology, Gorgan University of Agricultural Sciences & Natural Resources (GUASNR), Gorgan, Iran, and* [2]*Department of Chemistry, Golestan University, Gorgan, Iran*

Abstract
This study investigated the effects of organoclay platelet contents (0, 3 and 5 wt%) and polypropylene type (virgin and recycled) on the mechanical properties of polypropylene/wood flour composites. Composite samples were made by melt compounding and consequent injection moulding. The tensile, flexural and impact properties of resultant composites were determined. X-ray diffraction (XRD) analysis of composites with 3 and 5% nanoclay content was also conducted. The results indicated that tensile and flexural properties of the composites increased with the addition of nanoclay particles up to 3 wt% and decreased thereafter. The impact strength of the composites, however, decreased with the incorporation of nanoclay. The mechanical properties of the recycled polypropylene-based nanocomposites were statistically comparable with those based on virgin polypropylene. XRD analysis revealed that the degree of intercalation in the nanocomposites containing 3% nanoclay was higher than in those containing 5%. Based on these results, it can be concluded that recycled polypropylene could be used instead of virgin polypropylene in the production of value-added products with no significant adverse effects on the mechanical properties.

Keywords: *Injection moulding, mechanical properties, organoclay, wood–plastic composites.*

Introduction

The growth of environmental concerns regarding non-biodegradable oil-based plastics has led to many efforts in recycling postconsumer plastics to reduce both the environment impact and the consumption of virgin plastics (Adhikary *et al.*, 2008). In addition, owing to the considerable increase in global oil prices, using recycled plastics for the production of wood-plastic composites (WPCs) has recently attracted considerable attention and is now a great task for researchers and industry. The use of recycled plastics in WPCs is increasing in both developed and developing countries (Kazemi Najafi *et al.*, 2010).

WPCs with recycled wood fibre/flour have also gained in popularity owing to the low cost of recycled wood to the manufacturers (Ashori, 2008). The possibility of using recycled materials in the development of composites is very attractive, especially because of the large quantity of wood fibre/plastic waste generated daily (Ashori & Nourbakhsh, 2009). The volume of wood flour waste generated in the wood industry is estimated to be around 16% of its total production (Ares *et al.*, 2010).

In recent years, the use of nanoscale fillers in polymer and fibre-reinforcement composites has attracted considerable academic and industrial interest. Such nanoscale fillers frequently exhibit large surface-to-volume ratios and improve properties such as gas permeability resistance, fire resistance and dimensional stability of the fabricated composites (Cho & Paul, 2001; Zhao *et al.*, 2005; Mohan *et al.*, 2007; Hamidi *et al.*, 2008; Zhang *et al.*, 2009).

Among the different nanoparticles, nanoclay is the most preferred one, owing to its high aspect ratio (100–1000) and extremely high surface-to-volume

Correspondence: H. Khanjanzadeh, Department of Wood and Paper Technology, Gorgan University of Agricultural Sciences & Natural Resources (GUASNR), Gorgan, Iran. E-mail: H_Khanjanzadeh@yahoo.com

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ratio (700–800 $m^2 g^{-1}$), which establish significant improvements for a variety of polymers at very low filler contents (less than 5% by weight), far less than those using traditional micron-scaled fillers ($\geq$ 20 wt%) (LeBaron *et al.*, 1999; Strawhecker & Manias, 2000; Ray & Bousmina, 2005; Tjong, 2006; Zhang *et al.*, 2009).

Homogeneous dispersion of the hydrophilic clay minerals and organic fillers in the hydrophobic polymer matrix is not realized because there are no polar groups in the backbone of polymers (Liao & Wu, 2005). Ammonium surfactants are usually used to modify montmorillonite clay to achieve better interaction between the hydrophilic aluminosilicate and the organophilic polymer matrix (Maji *et al.*, 2009). To obtain a homogeneous dispersion of organically modified clay and the organic fillers in polyolefines such as polypropylene and polyethylene, which do not include any polar groups in their backbones, addition of an appropriate compatibilizer is needed; hence, polar functional oligomers, such as maleic anhydride grafted polypropylene or polyethylene, are used (Liu & Wu, 2001; Lertwimolnun & Vergnes, 2005; Zhong *et al.*, 2007).

The main objective of this study was to examine the effects of different nanoclay contents and polypropylene type on physical and mechanical properties of wood flour–polypropylene nanocomposites.

Materials and methods

Materials

For the preparation of nanocomposites, two different kinds of polypropylene (in the form of pellets) were used: virgin polypropylene (vPP) (supplied by Tabriz Petrochemical Co., Iran) and recycled polypropylene (rPP) (supplied by Pinak Co., Iran). The vPP had a density of 0.9 $g\,cm^{-3}$ and a melt-flow index of 5–7 g 10 min^{-1} (230°C, 2.16 kg), and the (rPP had a melt-flow index of 8.5–9.5 g 10 min^{-1} (230°C, 2.16 kg).

Poplar wood flour used in the study was provided by a local mill and screened to obtain 40 and 60 mesh particle sizes. The screened particles were oven-dried at 100 ± 5°C for 24 h to reach target moisture content (3 wt%) before processing.

The coupling agent, maleic anhydride grafted polypropylene (MAPP), was provided by Kimya Baspar Asia Co. (Iran) in the form of pellets which had a melt-flow index of 64 g 10 min^{-1} and 2 wt% maleic anhydride content.

Montmorillonite modified with a dimethyl, dehydrogenated tallow, quaternary ammonium (Modifier Concentration = 125 meq 100 g^{-1} clay, d-spacing ($d_{001} = 31.5$ Å)) was obtained from Southern Clay Products Co. (USA), under trade name of Cloisite 15A.

Compounding and sample preparation

The amount of MAPP was fixed at 5 wt%. The concentration of nanoclay was varied from 0 to 5%, based on the total weight of polypropylene.

Before sample preparation, wood flour was dried in an oven at 103 ± 2°C for 24 h. Formulation of the mixes and abbreviations used for the respective prepared mixes are given in Table I.

Compounding was performed in a Haake internal mixer (HBI, System 90, USA). The mixing was carried out at 175°C with a rotation speed of 60 rpm. Polypropylene was first added into the mixing chamber after it was melted, the coupling agent and nanoclay were added and mixed with polypropylene for 5 min, and finally wood flour was added. The mixing process took 15 min on average. The compound materials were ground to prepare the granules using a pilot scale grinder (model WIESER, WGLS 200/200).

Table I. Composition of the studied formulations.

Formulation	ID code	WF content (wt%)	PP (wt%)		MAPP (%)	NC content (%)
			Virgin	Recycled		
1	50WF/45vPP/5MAPP	50	45	–	5	0
2	50WF/22.5vPP/22.5rPP/5MAPP	50	22.5	22.5	5	0
3	50WF/45rPP/5MAPP	50	–	45	5	0
4	50WF/42vPP/5MAPP/3NC	50	42	–	5	3
5	50WF/21vPP/21rPP/5MAPP/3NC	50	21	21	5	3
6	50WF/42rPP/5MAPP/3NC	50	–	42	5	3
7	50WF/40vPP/5MAPP/5NC	50	40	–	5	5
8	50WF/20vPP/20rPP/5MAPP/5NC	50	20	20	5	5
9	50WF/40rPP/5MAPP/5NC	50	–	40	5	5

Note: PP = polypropylene; WF = wood flour; vPP = virgin polypropylene; rPP = recycled polypropylene; NC = nanoclay; MAPP = maleic anhydride grafted polypropylene.

The granulated samples were then dried at 100°C for about 12 h. Finally, test specimens were prepared by injection moulding (Imen Machine, Iran).

Mechanical properties

The tensile and three-point flexural tests for injection-moulded specimens were carried out using an Instron Universal Testing Machine (Model 4486) at a cross-head speed of 5 mm min^{-1} at room temperature following ASTM D-638 and D-790, respectively. A notched Izod impact test was conducted on a Santam Izod impact tester (Model SIT20D, Iran) in conformance with ASTM D-256 standard. Five specimens were produced for each treatment.

X-ray diffraction pattern

The degree of clay intercalation in WPCs (containing 3 and 5% nanoclay in vPP matrix) was evaluated by X-ray diffraction (XRD). XRD measurements were carried out in a Bruker X-ray diffractometer (D8 Advanced, Germany) using Cu Kα ($\lambda = 1.54$ nm) radiation at a scanning rate of 0.3° min^{-1}, with an angle ranging from 2° to 10°.

Statistical analysis

Statistical analysis was conducted using SPSS programming (version 18) method in conjunction with analysis of variance (anova) techniques. Duncan's multiple range test was used to determine the statistical differences among the variables investigated at the 95% significance level.

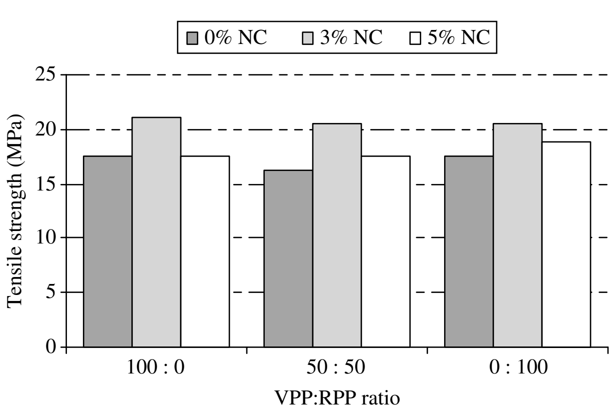

Figure 1. Effects of nanoclay (NC) content and polypropylene type on tensile strength. VPP = virgin polypropylene; RPP = recycled polypropylene.

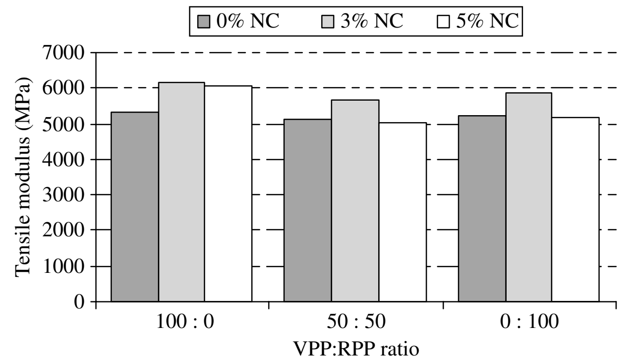

Figure 2. Effects of nanoclay (NC) content and polypropylene type on tensile modulus. VPP = virgin polypropylene; RPP = recycled polypropylene.

Result and discussions

Tensile properties

Figure 1 and 2 illustrate the tensile strength and modulus versus kind of polypropylene for WPCs made with different nanoclay contents. It can be seen that with the increase in the nanoclay loading from 0 to 3 wt% there was an improvement in the tensile strength and modulus of the nanocomposites. The maximum improvement in tensile strength (20.3%) and modulus (15.6%) was achieved when 3 wt% of nanoclay was incorporated into the vPP matrix. The increased tensile properties up to 3 wt% nanoclay content may be attributed to the high aspect ratio of stiff silicate layers in the polymer matrix that result in a higher extent of interaction with the polymer chains (Ashori & Nourbakhsh, 2010) and good interfacial adhesion between the nanoscale clay particles and the polypropylene matrix, so that the mobility of polymer chains is restricted under loading (Maji *et al.*, 2009; Ziaei Tabari *et al.*, 2011).

Nevertheless, with further increase in nanoclay loading from 3 to 5 wt%, a decrease in the tensile strength and modulus of the nanocomposites was observed. This indicates the nanoclay was dispersed more uniformly through the polymer matrix at low concentrations (3 wt%) to increase the surface attraction between the clay and the polymer matrix. One possible reason for this kind of behaviour may be attributed to the agglomeration of the nanofillers or the filler–filler interaction, resulting in induced local stress concentration in the nanocomposites and reduction in the clay aspect ratio, thereby reducing the contact surface area between the organoclay and the polymer matrix and formation of agglomerated clay tactoids (Liao & Wu, 2005; Mohanty & Nayak, 2007; Maji *et al.*, 2009; Kord *et al.*, 2010).

The mechanical properties of composites depend on many factors, including the aspect ratio of the filler, the degree of dispersion of the filler in the matrix resin and the adhesion at the filler–matrix interface (Zhao *et al.*, 2005).

No significant difference was found between the tensile strength and modulus of nanocomposites formulated with rPP, vPP and an equal mixture of the two. The tensile strength of WPCs made with rPP was equal to or even exceeded the tensile strength of WPCs made with vPP. Similar results were reported by Kazemi Najafi *et al.* (2006) and Adhikary *et al.* (2008).

Flexural properties

Figures 3 and 4 show the flexural strength and modulus of WPCs, respectively. The flexural strength and modulus showed similar trends to those of tensile strength and modulus. With the increase in clay loading up to 3 wt%, flexural strength and modulus increased (by 19% and 7%, respectively, for vPP), after which there was a reduction in both of them. Increased flexural properties for 3 wt% nanoclay loading may be attributable to the high stiffness of clay platelets and the lower percolation points created by the high aspect ratio nanoclays (Ashori & Nourbakhsh, 2010; Kord *et al.*, 2010).

No significant difference was found between the flexural strength and modulus of nanocomposites formulated with vPP, rPP and vPP/rPP. The flexural properties of rPP-based WPCs were statistically comparable to those of composites made from vPP. The flexural strength of rPP-based WPCs was equal to or even exceeded the flexural strength of WPCs made with vPP.

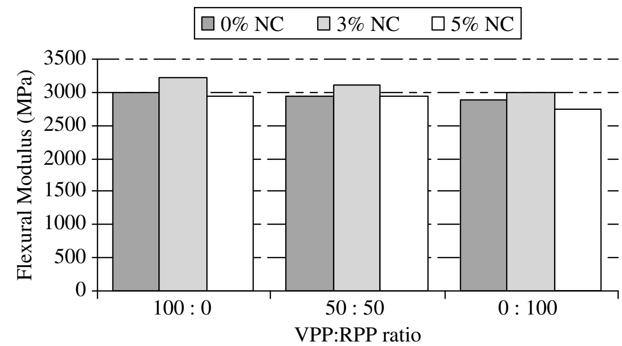

Figure 4. Effects of nanoclay (NC) content and polypropylene type on flexural modulus. VPP = virgin polypropylene; RPP = recycled polypropylene.

Impact strength

The values of notched Izod impact strength of the nanocomposites are illustrated in Figure 5. The impact properties of the injected WPCs varied significantly with polypropylene type. Composites made with vPP exhibited the highest impact strength of all the produced composites. Impact resistance in polymers is often related to the crystallinity of the material, and the higher the crystallinity, the lower the impact strength (Rust *et al.*, 2006).

The Izod impact strength of composites at the same concentration of MAPP decreased with the increase in nanoclay content. The decline in the impact strength at higher clay loading is probably due to stiffening polymer chains and agglomeration of clay particles, which result in absorbing less impact energy (Mohanty & Nayak, 2007). Besides the formation of clay agglomeration at higher nanoclay content, the presence of un-exfoliated aggregates and voids can reduce the impact strength (Zhao *et al.*, 2005; Yuan & Misra, 2006; Kord *et al.*, 2010).

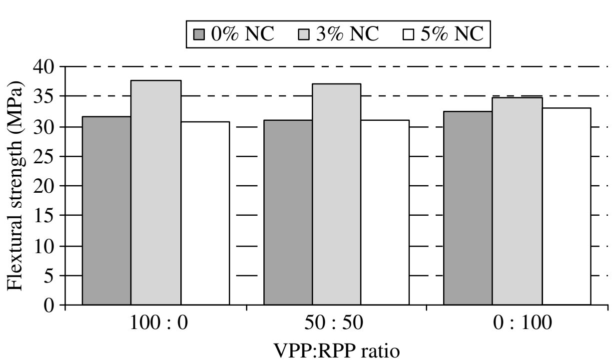

Figure 3. Effects of nanoclay (NC) content and polypropylene type on flexural strength. VPP = virgin polypropylene; RPP = recycled polypropylene.

X-ray diffraction results

The XRD patterns of pure nanoclay (cloisite 15A) and selected composite samples containing clay are shown in Figure 6. The interlayer spacing was calculated according to Bragg's law:

$$n\lambda = 2d \sin\theta \qquad (1)$$

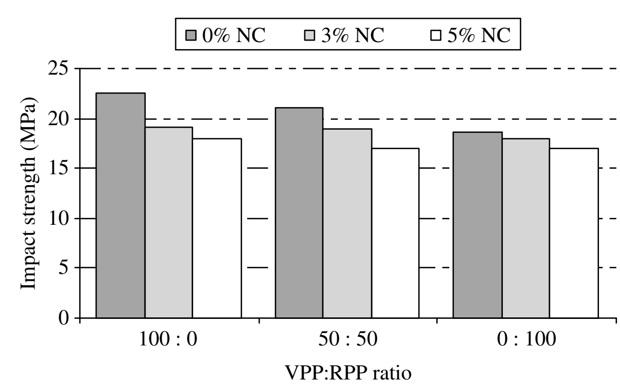

Figure 5. Effects of nanoclay (NC) content and polypropylene type on notched impact strength. VPP = virgin polypropylene; RPP = recycled polypropylene.

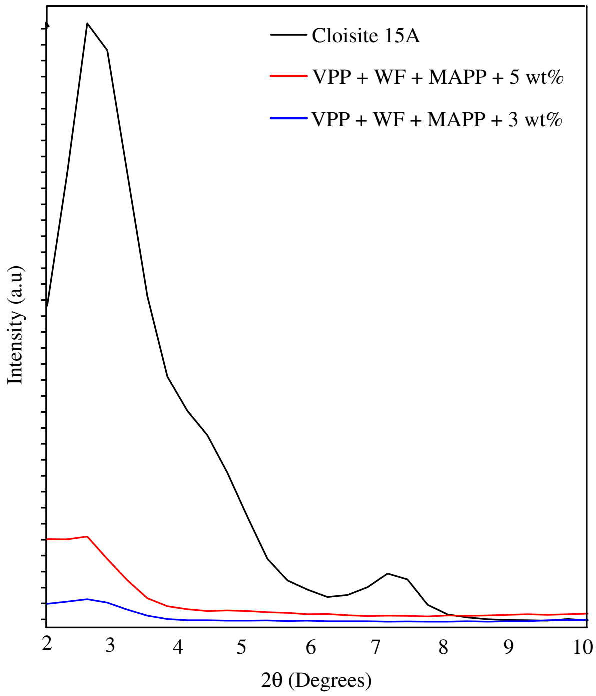

Figure 6. X-ray diffraction patterns of clay (cloisite 15A) and its polypropylene composites with wood flour. VPP = virgin polypropylene; WF = wood flour; MAPP = maleic anhydride grafted polypropylene.

The organically modified nanoclay showed its characteristic intense peak at $2\theta = 2.8°$. The composite specimens with 3 and 5 wt% nanoclay contents showed a peak shift towards a lower diffraction angle (2θ) than the pure cloisite 15A, indicating an increase in the interlayer spacing of silicate layers and intercalation of polymer chains between clay layers. The reduced peak intensity is attributed to the low concentration of clay in the samples. The results indicate that the degree of intercalation in the nanocomposites containing 3% nanoclay content was higher than in those containing 5%. The shift in the peak from a higher to a lower diffraction angle has been reported by different researchers (Lee & Kim, 2009; Ravindra Reddy *et al.*, 2010).

Conclusions

The result of this study indicate that the tensile and flexural strength of composites made with nanoclay for different kinds of polypropylene on average increased by about 21% and 15%, respectively, with the incorporation of 3% nanoclay, but then decreased slightly as the nanoclay content increased to 5%. It was also found that the mechanical properties of rPP-based composites are comparable statistically with those of made from vPP, indicating the possibility of expanding the use of recycled plastics in the manufacturing of WPCs. The experimental results indicate that mechanical and physical properties of WPCs could be significantly improved by the appropriate incorporation of nanoclay in the composites.

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