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ORIGINAL PAPER

Mechanical and thermo-chemical properties of wood-flour/polypropylene blends

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Abstract The main objective of this research was to study the potential of waste polypropylene and waste wood for making wood plastic composites (WPCs). The effects of nanoclay (NC), microcrystalline cellulose (MCC), and coupling agent (MAPP) on the mechanical and thermal properties were also studied. The results showed that mechanical properties of the composites made with MCC were significantly superior to those of unfilled. Addition of MAPP could enhance the mechanical and thermal properties of the blends, due to the improvement of interface bond between the filler and matrix. The significant improvements in tensile properties of the blends composites made with MAPP and NC were further supported by SEM micrographs. The thermogravimetric analysis indicated that the addition of 5 wt% MAPP and 3 wt% NC significantly increased the thermal stability of the blends compared to the pure PP. MCC could not improve the thermal stability. The experimental results demonstrated that the waste materials used are promising alternative raw materials for making low cost WPCs.

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Introduction

Production of waste materials is an undeniable part of human society. Waste is produced by several sectors including industries, forestry, agriculture, and municipalities. The accumulations of waste result in several environmental problems, health issues, and safety hazards, and prevent sustainable development in terms of resource recovery and recycling of waste materials. Guidelines and directives to reduce waste generation and promote waste recovery are laid down according to the "waste management hierarchy", in which waste prevention, reuse, and recycling are designed to minimize the amount of waste left for final, safe disposal [1].

The worldwide production and consumption of plastics result in a significant contribution to municipal solid waste. Plastic wastes are an undesired pollutant in soil, rivers, and marine [2]. The increasing quantities of plastic wastes and the safe disposal has become a serious public concern. Attempts have been made to recycle the consumer plastics in order to reduce the environmental impact and the consumption of the virgin plastics. Similarly, a large amount of wood waste is generated at different stages of the wood processing and such waste is mainly destined for landfill. Thus, it is important to find uses for these waste materials. In recent years, the recycling systems and reuse techniques of wood wastes have become a vital part of sustainable utilization of resources [3].

When two or more materials with different properties are combined together, they form a composite material. Wood plastic composites (WPCs) are used as engineering materials for house-wares, automotive industries, and various construction applications. They are very promising materials to achieve durability without using toxic chemicals. In recent years, their utilization has developed rapidly, especially in Europe, the US, and Canada [4]. The primary components of a WPC are wood in a particulate form and thermoplastic resins. An extruded WPC is comprised of 40–70% wood-flour by weight. Although the use of wood filler in plastic composites has several advantages over inorganic fillers, several drawbacks of cellulosic fillers such as low thermal stability and low compatibility, greatly reduce the overall performance of WPCs. However, it was found that the incorporation of coupling agent and reinforcement with microcrystalline cellulose (MCC) can improve the mechanical properties through enhanced compatibility between the hydrophilic wood filler and the hydrophobic polymer matrix [5, 6].

On the other side, most of the studies were focused on the use of virgin thermoplastics and wood fiber/flour for the WPC formulations [7–12]. Reuse of waste materials in the production of WPCs lessen solid waste disposals and can have advantageous to economy, environment, and technology. Thus, this work aims to investigate the characteristics of mechanical properties and thermal stability behavior in the composites made from post-consumer recycled thermoplastic and waste sawdust. In addition, the effects of the nanoclay (NC) and MCC loadings and

coupling agent addition on the mechanical properties and thermal stability are investigated.

Materials and methods

Materials

Waste poplar sawdust (WS) was collected from a local sawmill in Tehran, Iran. The as received—untreated—sawdust was dried at 102 °C for 24 h to moisture content of 2–3% (dry base) and then milled to particle size of 60 mesh size.

Microcrystalline Cellulose (MCC) (Pulver-20 micron) was supplied by Aldrich Chemical Company. The MCC was used as reinforcement fiber, with diameter of 20 μm in analysis grade.

The waste polymer polypropylene (PP) was supplied by local plastic recycling company, in the form of pellets with a melt flow index of 8 g/10 min (230 °C/2160 g) and density of 0.92 g/cm^3.

Maleic anhydride grafted polypropylene (MAPP) was obtained from Aldrich Chemical Company, which has a density of 0.91 g/cm^3, molecular weight of 9100 and Brookfield viscosity of 40,000 cP at 190 °C.

The NC used in this work was hydrophilic bentonite, Nanomer PGV, which was purchased from Sigma-Aldrich. NC is commercially available.

Preparation of composites

Formulations of the mixtures and abbreviations used for the respective mixtures prepared are given in Table 1. The WS, MCC, PP, MAPP, and NC were premixed before being fed into the first zone of the extruder. The mixtures were compounded in a co-rotating twin screw (Collin) extruder. The barrel temperatures of the extruder were controlled at 165, 170, 175, 180, and 185 °C for zones 1–5, respectively, while the temperature of the die was held at 185 °C. The melt temperature was kept 185 °C to prevent wood degradation, and screw speed was set at 60 rpm. The extruded strand was passed through a water bath and palletized. The resulting granules were subsequently injection molded at 185 °C and pressure of 3 MPa to produce standard ASTM [13] specimens. After molding, the test specimens were conditioned at 23 ± 2 °C and 50 ± 5% relative humidity for at least 40 h according to ASTM D 618.

Mechanical testing

All the specimens were tested following ASTM standard D 638 for tensile properties, ASTM D 790 for flexural properties, and D 256 for notched Izod impact strength. Tensile and bending tests were conducted using an Instron Universal Testing Machine (model 1186) at speeds of 1.5 and 2 mm/min, respectively. For tensile testing, specimens were cut into dumbbell-shaped (type III) with a thickness

Table 1 Formulation of mixtures and their codes

WF (wt%)	MCC (wt%)	MAPP (wt%)	NC (wt%)	PP (wt%)	Code
40	0	2.5	0	57.5	A1
40	0	2.5	3	54.5	A2
40	0	5	0	55	A3
40	0	5	3	52	A4
36	4	2.5	0	57.5	B1
36	4	2.5	3	54.5	B2
36	4	5	0	55	B3
36	4	5	3	52	B4
32	8	2.5	0	57.5	C1
32	8	2.5	3	54.5	C2
32	8	5	0	55	C3
32	8	5	3	52	C4

of 12 mm. A pendulum impact tester (Zwick 1446) was used for the Izod impact test. For each treatment level, six replications were conducted.

Thermogravimetric analysis

Thermogravimetric analysis (TGA) measurements were carried out using a thermogravimetric analyzer on 8-mg samples, over a temperature range from 30 to 650 °C, at a heating rate of 10 °C/min. TGA was conducted in a high quality nitrogen (99.5% nitrogen, 0.5% oxygen content) atmosphere with a flow rate of 20 mL/min in order to avoid unwanted oxidation. Each sample type was measured in triplicate.

Morphological study

The morphology of the composites was studied by a Philips XL 30 scanning electron microscope (SEM). The fracture surfaces of the specimens after impact test were sputter-coated with gold before analysis. All images were taken at an accelerating voltage of 25 kV.

Results and discussion

Tensile properties

Figure 1 depicted the tensile strength and modulus of the blends made with various formulations. It was clear that moderate increase in tensile strength occurred upon filling the polymer matrix with MAPP. The C4 and B4 samples showed the maximum and minimum increment by 31.6 and 6.4%, respectively. Meyrs et al. [14] reported 21% increase in tensile strength for a 50:50 wood-flour–polypropylene

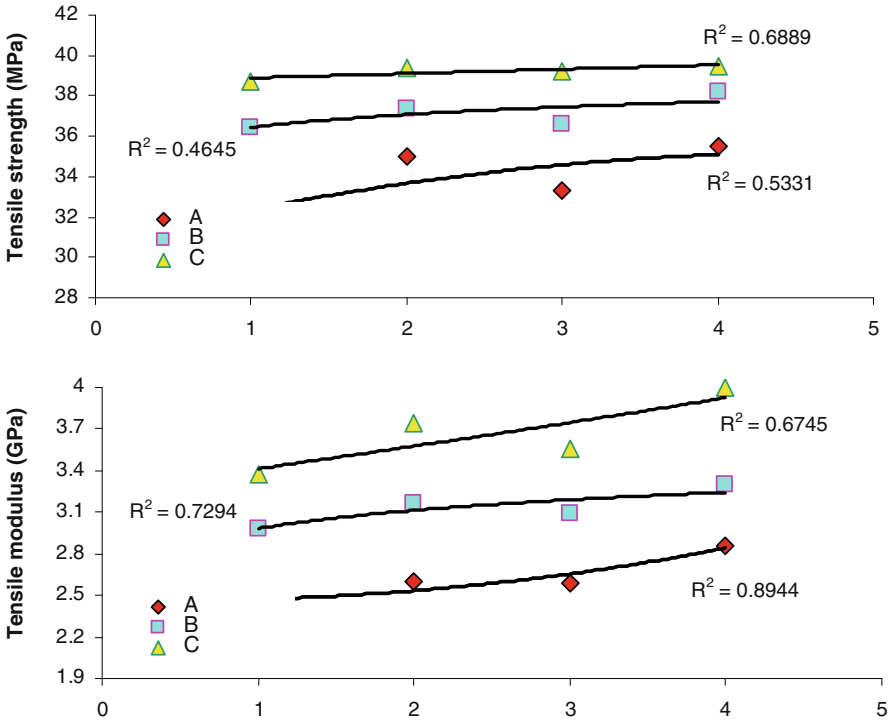

Fig. 1 Comparison of tensile properties of the blends based on various formulations

composites when MAPP was used as a compatibilizer. Stark and Rowlands [15] also reported a 27% increase in tensile strength of composite prepared with 40% wood fiber and 3% MAPP.

To improve the bonding strength between the lignocellulosic filler and the matrix polymer, coupling agent was used. With addition of MAPP, tensile properties of the blends significantly improved up to the level of pure PP. In general, blends with high coupling agent content exhibited better tensile strength and modulus. Most effective content of coupling agent is 5 wt% as shown in Fig. 1. The increase in tensile strength is due to the improved chemical bond between the fibers and PP polymer chains. Results demonstrate that modifying the fiber surface enhances the compatibility of hydrophobic polymer and hydrophilic cellulose fiber.

Tensile modulus exhibited a trend similar to that of the tensile strength and showed a maximum improvement of 3.99 GPa for C4 sample, which is 319% higher than that of pure PP matrix. The explanation is similar to that of the tensile strength. As expected, tensile modulus increased with the use of NC and MCC, which is believed to be due to better interfacial bonding between the fiber and matrix.

As can be seen from Fig. 1, tensile properties were improved with addition of NC. This result is consistent with the general observation that the introduction of an organoclay into a polymer matrix increases its tensile properties [16]. The enhancement is easily understood because filler in organoclay form can carry more

tensile load. The NC is much stiffer than polymer matrix and as a result it adds stiffness to the blends. The differences in strength improvement with respect to MAPP concentration are very prominent at the highest NC content. These results confirm the strong interaction between the PP matrix and NC [4].

Flexural properties

The flexural strength and modulus of the blends are illustrated in Fig. 2. As shown, maximum flexural strength ranges from 51 to 49 MPa, while minimum flexural strength is approximately 37.2 MPa for pure PP. The flexural proprieties of the blends vary significantly with MAPP, MCC, and NC loading. Blends made with MCC showed the highest strength and modulus of flexural, whereas pure PP and blends A1 exhibited the lowest properties. The maximum flexural strengths were shown in C4 sample. The flexural strength exhibits a similar trend to the tensile strength although less variation is observed in the flexural strength with different formulations than the tensile strength. Generally, all the compositions showed a flexural strength and modulus higher than the pure PP.

This increase in flexural properties was expected due to the improved adhesion between components in the blends. Greater flexural strength and modulus were achieved in the blends when MCC was used in the manufacture of the blends.

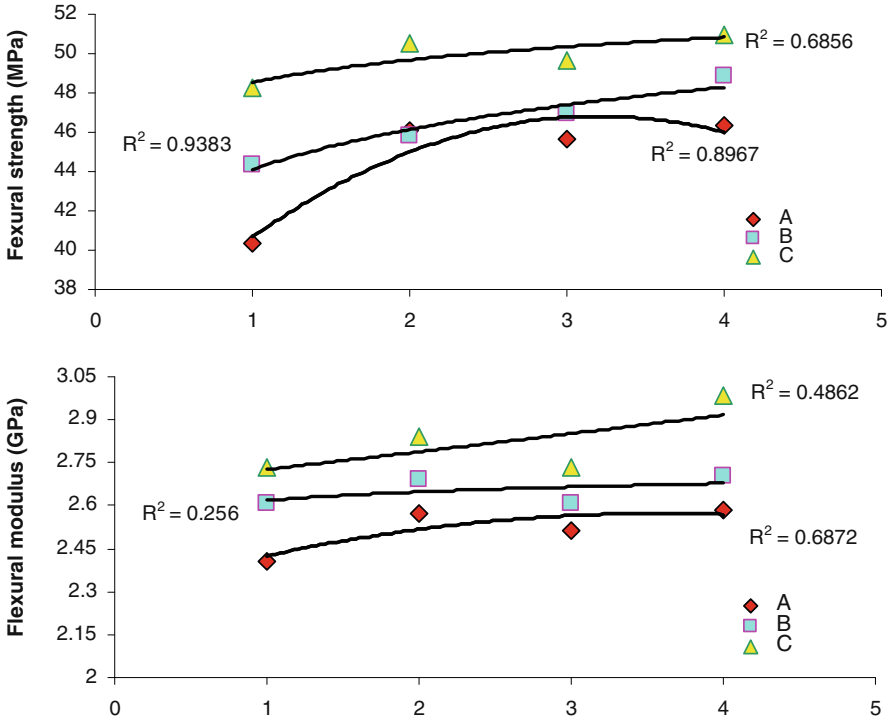

Fig. 2 Comparison of flexural properties of the blends based on various formulations

Increased flexural properties for 3 wt% NC loading are attributable to the high stiffness of clay platelets with high aspect ratio.

The blends based on 3 wt% NC loading and 5 wt% MAPP have a flexural strength varying from 40 to 46 MPa. It is also observed that the flexural strength increases with increasing MCC. For example, the flexural strength of the blends types A1 and C1 is 40.3 and 43.2 MPa, respectively. With a similar trend to the tensile test results, the addition of the MCC markedly improves the flexural strength as well as the tensile of the blends. Due to the similar mechanism as explained in the previous section, the flexural strength of blends with MCC increases as compared with blend type A (without MCC).

Impact strength

The notched specimens were tested and Fig. 3 shows the Izod impact strengths of the blends made with the different coupling agent content. Like the tensile and flexural properties, the Izod impact strength of blends increased with the increase in MAPP, MCC, and NC contents. The Izod impact strengths of the blends made with MCC are superior. In the blends made with G-3003, the impact strength at wt% of coupling agent content is almost the same level as 2.5 wt%.

Blends made with MCC (types B and C) had significant effect on the impact strength. In other words, the specimens made with 8 wt% of MCC exhibited remarkably higher impact strength than the specimen made without MCC (type A). The addition of MAPP improved the interfacial bonding between the fiber and matrix polymer, thus the crack was not initiated at the interface.

Characterization of fracture surfaces

The SEM was used to compare the tensile fracture surface of blends. Through SEM study, the distribution and compatibility between the fillers and the matrix could be observed. The presence of numerous cavities is clearly visible in Fig. 4 (A1 and B1). This indicates that the level of interfacial bonding between the fibers and the

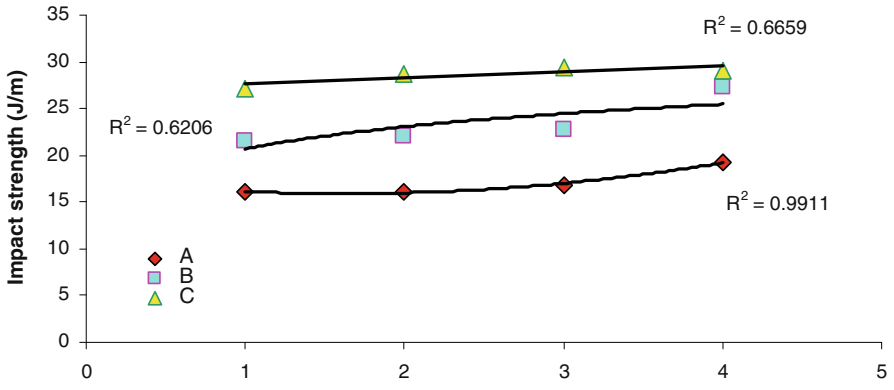

Fig. 3 Comparison of impact strength of the blends based on various formulations

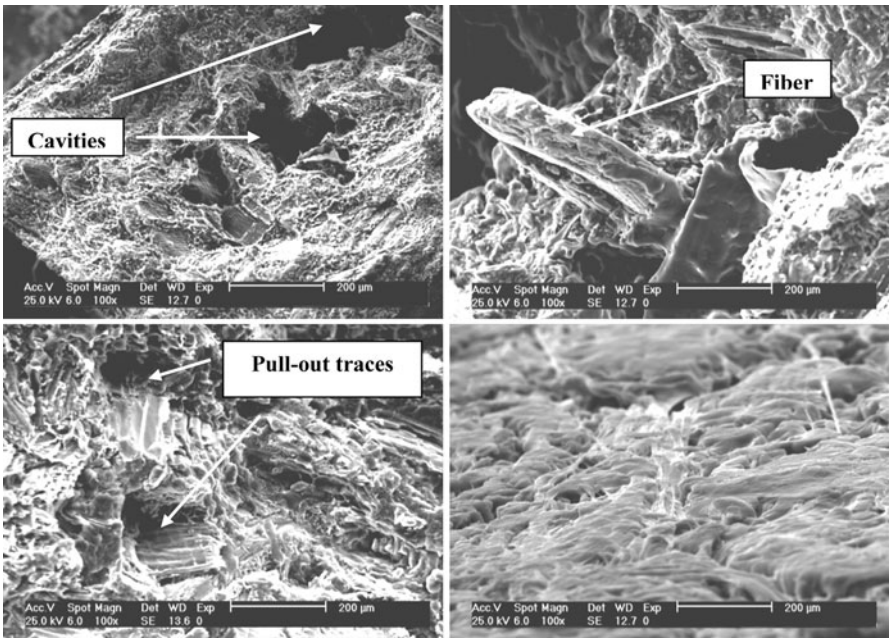

Fig. 4 SEM micrographs of the tensile fracture surfaces at magnification of ×100

matrix is week and when stress is applied it causes the fibers to be pulled out from the matrix easily leaving behind gaping holes. As can be seen in Fig. 4 (B3), there is less evidence of filler pull out and better interfacial adhesion between the filler and the matrix. The significant improvements in tensile properties of the blends treated with MAPP were further supported by SEM micrographs. Sample C4 depicts the smooth surface of the fibers indicating that the chemical modification has significantly altered the fiber structure.

Thermal properties

Thermal degradation of WPCs is a crucial aspect for the manufacturing process, because it determines the maximum processing temperature that can be used. Due to its lower thermal stability, wood is usually used as a filler only in polymers that are processed at temperatures below 200 °C. The degradation of wood due to high temperatures can lead to undesirable properties, such as odor, discoloration, and loss of mechanical strength. This limits the type of polymers that can be processed into WPCs and the applications in which they can be used [11].

TGA graph of weight loss as a function of temperature is presented (Fig. 5). This thermogram represents the typical thermal decomposition behavior for pure PP and three typical combinations. As shown, the initial degradation of pure PP began at 228 °C with a weight loss of 91.9% while the initial degradation of other samples was varied in the range of 370–390 °C. The higher onset of degradation temperature indicates the improved thermal stability of the samples A4 and B4. These results

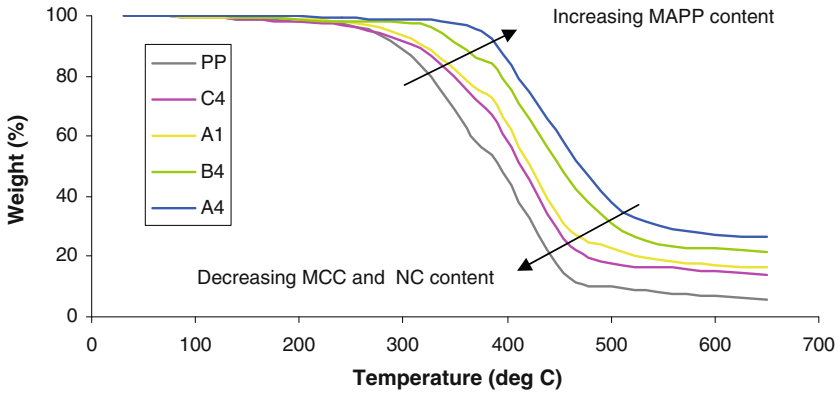

Fig. 5 Comparison of TGA curves of pure PP and blends based on various formulations

clearly illustrate that the thermal property of the blends increases after using MAPP and further increases after addition of NC. The presence of MAPP leads to strong interactions between polymer matrix and NC. The greatest improvement in the thermal stability was achieved at 5 wt% MAPP and 3 wt% NC. As expected, addition of MCC could not improve thermal stability.

Conclusions

Based on the results of this study the following conclusions can be drawn.

1. Mechanical properties of the blends made with MCC (types B and C) were significantly superior to those of unfilled ones (type A).
2. The mechanical properties of the blends enhanced with increasing coupling agent content, due to the improvement of interface bond between the fiber and matrix.
3. SEM study also showed that there are distinct cavities between the matrix and the fillers, indicating poor adhesion. The addition of MAPP could improve the interfacial adhesion caused to reduce the numbers of cavities and pulled-out fibers.
4. The thermal stabilities of all blends were remarkably enhanced as compared to the pure PP. This was reflected by the fact that the onset temperature of thermal degradation increased with the incorporation of MAPP and NC. It is noteworthy that MCC could not improve the thermal stability.
5. The results showed that waste materials used are promising alternative raw materials for making low cost WPCs.

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