Effect of wood flour on the properties of PP/HIPS wood plastic composites

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Palabras clave: fibra natural, polipropileno, poliestireno de alto impacto, harina de pino, harina de haya, harina de madera, compuestos de polímeros.

Key words: Natural fiber, polypropylene, high impact polystyrene, pine flour, beech flour, wood flour, polymer composites.

RESUMEN. Materiales compuestos reforzados con fibra natural es un área emergente en la ciencia de los polímeros. Estas fibras naturales son fibras de bajo coste con baja densidad y altas propiedades específicas. Estos son biodegradables y no abrasivo. Los materiales compuestos de fibra naturales ofrecen propiedades específicas comparables a las de materiales compuestos de fibra convencionales. Sin embargo, en el desarrollo de estos materiales compuestos, la incompatibilidad de las fibras y pobre resistencia a la humedad a menudo reducir el potencial de fibras naturales, y estos dibujar espaldas se convierten en cuestión crítica. Materiales compuestos de madera y plástico (WPC) son relativamente una nueva clase de materiales y uno de los sectores de mayor crecimiento en la industria de compuestos de madera. Composites de madera en una matriz termoplástica (composites madera-plástico) se consideran una solución de bajo mantenimiento para el uso de la madera en aplicaciones al aire libre. WPCs se hacen normalmente de una mezcla de fibra de madera, termoplástico, y pequeñas cantidades de modificadores de proceso y de propiedad a través de un proceso de extrusión. El principal objetivo de este estudio fue investigar el efecto de diferentes aditivos, a saber, el pino y la harina de haya, anhídrido maleico injertado PP y estireno- etileno butilenos bloque de estireno mezcla copolymera de ambos en poliolefina / HIPS muestras compuestas. El mezclado se realiza con extrusora de doble tornillo. Posteriormente, se determinaron las propiedades mecánicas y morfológicas.

ABSTRACT. Natural fiber reinforced composites is an emerging area in polymer science. These natural fibers are low cost fibers with low density and high specific properties. These are biodegradable and nonabrasive. The natural fiber composites offer specific properties comparable to those of conventional fiber composites. However, in development of these composites, the incompatibility of the fibers and poor resistance to moisture often reduce the potential of natural fibers, and these draw backs become critical issue. Wood-plastic composites (WPC) are a relatively new class of materials and one of the fastest growing sectors in the wood composites industry. Composites of wood in a thermoplastic matrix (wood–plastic composites) are considered a low maintenance solution to using wood in outdoor applications. WPCs are normally made from a mixture of wood fiber, thermoplastic, and small amounts of process and property modifiers through an extrusion process. The main objective of this study was to investigate the effect of different additives, namely, pine and beech flour, maleic anhydride grafted PP and styrene ethylene butylenes styrene block copolymera mixture of both on polyolefin /HIPS composite samples. Mixing was done with twin screw extruder. Subsequently, mechanical and morphological properties were determined.

INTRODUCCIÓN

Composite materials are created by combining two or more components to achieve desired properties, which could not be obtained with the separate components. The use of reinforcing fillers, which can reduce material costs and improve certain properties, is increasing in thermoplastic polymer composites. Currently, various inorganic materials such as talc, mica, clay, glass fiber, and calcium carbonate are being incorporated into thermoplastic composites.
Nevertheless, agricultural fillers, for example, wood fiber or wood flour, have drawn attention due to their abundant availability, low cost, and renewable nature. In recent years, interest has grown for composites made from wood flour or wood fiber in thermoplastic matrices, particularly for low-cost/high-volume applications. Among commodity thermoplastics, polypropylene (PP) possesses outstanding properties such as low density, good flex life, sterilizability, good surface hardness, very good abrasion resistance, and excellent electrical properties. However, the main purpose for the addition of cellulose-based fillers to thermoplastics is to reduce the cost per unit volume and to improve stiffness.

Low-price cellulose-based fibers, such as wood flour, wood fibers, and cellulose fibers, have high stiffness and low density and are recyclable and nonabrasive. Contrary to PP, wood flour is predominantly polar due the presence of polar groups on its different components, and thus, it easily absorbs moisture. Thus, to improve the interaction between these otherwise incompatible surfaces, the addition of a compatibilizer or the surface modification of the fibers (or the matrix) was envisaged. Different treatments have been used to improve the adhesion and/or the compatibility between fillers and PP, and the use of maleic anhydride–polypropylene copolymer (PPMAN) as a compatibilizing agent has also been reported. Many ways are possible: plasma or corona treatments can be used, but more often chemical treatment such as oxidation grafting or functionalization. The so-called compatibilizing agents possess at one end a function F able to react with hydroxyl groups and at the other end an alkyl chain of varying length, or at the limit a polymeric chain with a structure similar to that of the matrix (Fig. 1). Such agents are small molecules possessing two different functions F and G. The first is able to react with OH groups of cellulose. The second is generally a double bond that can react with a tertiary carbon of PP, for instance, by a radical reaction after elimination of a hydrogen atom. Such agents are “genuine” coupling structures that link the cellulose fiber directly to the PP matrix.

In the literature, there are many studies on wood–plastic composites. Among them, Coutinho et al. investigated the effects of the treatment and mixing conditions on the mechanical properties of wood fiber/polypropylene composites with silane coupling agents. They reported that the optimal mixing conditions for wood-fiber/polypropylene composites were a mixture temperature of 180°C and a mixing time of 10 min at a rotation speed of 60 rpm. Raj and Kokta examined the effects of coupling agents on the tensile properties of high-density polyethylene HDPE/Wood composites. Oksman et al. also investigated the mechanical properties and morphology of PE/WF composites modified with a styrene–ethylene/butylene–styrene triblock copolymer grafted with maleic anhydride (SEBS-g-MA). Coupling agents increase the contact area between wood particles and PP matrix, enhance dispersion, and improve adhesion of the two components.

The aim of this study was to analyze the effect of system compatibility on the tribological and thermal properties of PP/HIPS/wood flour composites.

**Experimental**

**Compositions and Materials**

Ten different polymer composites were prepared. Compositions of PP/HIPS/wood flour polymer composites that were formed are given in Table 1.

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Fig. 1 Chemical modifications of cellulose for compatibilization with the matrix. 

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MAH: Maleic anhydride
Polypropylene (Samsung Total PP RP100) is a propylene-ethylene copolymer grade and it was supplied by Samsung Total Petrochemicals Co. Ltd. (Korea). Its density is 0.900 g/cm³. Melt flow rate (230°C /2.16 kg) is 0.25 g/10min. Its yield strength is 26.5 MPa and tensile elongation is 450 %. Vicat softening temperature is 130 °C. High impact polystyrene (HIPS) (porene HIPS HI650) was supplied by IRPC public company limited (Bangkok, Thailand). Its melt flow rate (200°C/5.0 kg) is 8.0 g/10min, tensile strength is 22.8 MPa, Notched izod impact strength is 110 J/m, deflection temperature under load (1.8 MPa) is 90°C and Vicat softening temperature is 95°C. Styrene ethylene butylenes styrene block copolymer (calprene H6110) was supplied by Dynasol Elastomers (Houston, USA). Total styrene ratio is 30 %. Melt flow rate (230°C /2.16 kg) is 1.5 g/10min. The compatibilizing agent, maleic anhydride-grafted PP (MAPP) (Optim-425, MFI/190°C: 2.16 kg = 120 g/10 min, density: 0.91 g/cm³), was supplied by Pluss Polymers Pvt. Ltd. in India.

The commercial softwood (pine) and hardwood (beech) flours were supplied from Semawood Company in Cerkezköy, Turkey. The wood particles were obtained from the round woods by using a disc chipper, followed by grinding process in a grinder. The average size and moisture content of the wood flour without bark were 40 US-mesh and %1-2, respectively.

Sample Preparation
Pine and beech flour were dried overnight at 90°C in a vacuum oven prior to melt blending. Mechanical premixing of solid compositions was done using a LB-5601 blender (Stroudsburg, USA) brand batch blender for 15 min. Samples with various proportions of PP/HIPS/wood flour polymer composites were produced between 180-200 °C at 25-30 bar pressure, and a rotation rate of 20 rpm, with a Microsan extruder (Kocaeli, Turkey). L/D ratio is 30, Ø:25 mm, Polymer composites were also dried in vacuum oven at 80°C for 24 hours after extrusion. Subsequently, test samples were molded in injection molding machine. Extrusion and injection conditions are given in Table 2.

Table 2. Extrusion and injection conditions of the PP/HIPS/wood flour polymer composites.

<table>
<thead>
<tr>
<th>Process</th>
<th>Extrusion</th>
<th>Injection</th>
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<tbody>
<tr>
<td>Temperature (°C)</td>
<td>180-200</td>
<td>180–200</td>
</tr>
<tr>
<td>Pressure (bar)</td>
<td>25-30</td>
<td>80–110</td>
</tr>
<tr>
<td>Waiting time in mold (s)</td>
<td>-</td>
<td>15</td>
</tr>
<tr>
<td>Screw speed (rpm)</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>Mould temperature (°C)</td>
<td>-</td>
<td>30–35</td>
</tr>
</tbody>
</table>

Thermal Characterization
Heat deflection temperature (HDT) and Vicat softening point tests were done according to ISO 75 and ISO 307 standard with determined by CEAST 6521 (Pianezza, Italy) HDT-Vicat test equipment.

Tribological Tests
*Static coefficient of friction (µs)*: Static coefficient of friction test was done according to the ISO 8295 method with Devotrans friction coefficient measurement equipment. The dimensions of the tested specimens were 80x200x4 mm
and the dimensions of the sled specimens were 63x63x4 mm. Speed was selected as 100 mm/min. The force increases linearly to a maximum which represents the static frictional force $F_S$. Measurements made at a high friction drag permit the dynamic coefficient of friction to be calculated, but not the static coefficient of friction. The static coefficient of friction $\mu_s$ is given by the equation,

$$\mu_s = \frac{F_S}{F_P} \quad (1)$$

Where $F_S$ is the static frictional force, expressed in Newton, $F_P$ is the normal force exerted by the mass of the sled, expressed in Newton ($=1.96$ N) [15].

**Wear rate:** The wear tests were done according to the DIN 53 516 method with Devotrans DA5 (Istanbul, Turkey) abrasion test equipment. The thickness of the test specimens was 7.0 mm and diameter was 15.5 mm. Cylinder rotational speed was selected as 40 rpm and normal load ($F_N$) of 10N was used. Total sliding distance ($L$) was 40 m. The mass loss of the samples ($\Delta m$) was measured after the wear process, and the specific wear rates ($W_s$) were calculated using the following equation:

$$W_s = \frac{\Delta m}{\rho \cdot F_N \cdot L} \quad (mm^3/Nm) \quad (2)$$

Where $\Delta m$ is the specimen’s mass loss, $\rho$ is the density of specimen, $F_N$ is the normal load applied, and $L$ is the total sliding distance. The friction coefficients and wear rates reported in the present study were the averages of at three measurements.

**Microscopy**
The fractured surfaces of the PP/HIPS/wood flour polymer composites were coated to an approximate thickness of 10 nm of a gold (Au) (80%)/palladium (Pd) (20%) alloys to prevent electrical charging by Polaron SC 7620 (Schwalbach-Germany). The surfaces of the prepared samples were observed by the JEOL-JSM 5910 LV (Tokyo, Japan) scanning electron microscopy (SEM) at an acceleration voltage of 20 kV.

**Result and discussion**

**Thermal properties of PP/HIPS/wood flour polymer composites**
The vicat and HDT experiment were started at room temperature with a heating rate of 120°C/h and under a load of 1 Kg and 1.8 MPa respectively. As shown in Figure 2-A, Vicat softening point values of pure PP and pure HIPS were 135 and 98 °C respectively. The increasing of pine and beech flour contends to the PP (group 3) resulted in higher vicat values. Whereas, the increasing of pine and beech flour contents to the HIPS (group 4) resulted in lower vicat values. Vicat softening point of PP/HIPS/wood flour composites (groups 5-7) shows a decrement as the HIPS concentration increases from 10 to 30 wt %. But Vicat softening point of PP/HIPS/wood flour composites (groups 8-10) shows an increment as the HIPS concentration decreases from 47 to 27 wt %.

![A) Vicat softening point](image1)

![B) Heat deflection temperature (HDT)](image2)

**Fig. 2** Thermal properties of the PP/HIPS/wood flour polymer composites.

As shown in Figure 2-B, heat deflection temperature (HDT) values of pure PP and pure HIPS were 47 and 72 °C respectively. The increasing of pine flour contends to the PP (group 3) resulted in higher HDT values. HDT values of PP/HIPS/Wood flour composites (groups 5-7) shows a increment as the HIPS concentration increases from 10 to 30
wt %. However, HDT values of PP/HIPS/Wood flour composites (groups 8-10) shows a decrement as the HIPS concentration decreases from 47 to 27 wt %.

**Wear properties of the PP/HIPS/wood flour polymer composites**

The effects of applied load, HIPS and wood flour content on the tribological behaviors of PP/HIPS/wood flour polymer composites were examined. The values of applied load-wear rate relationship are shown in Fig 3. It is seen that the applied load had a great effect on the wear rate of composites. Wear rate increased gradually with the increase of the applied load from 5 to 20 N. It was inconsistent that the applied load affected on the wear rate of PP/HIPS polymer composites. Based on these results, it could be postulated that the level of the filler can affect the matrix hardness and bonding strength between the filler and polymer matrix. The weak bond led to the filler particles detaching from the matrix and the matrix pulling out more easily, which could increase the wear rate of the composites. As shown in Figure 3, wear rate of pure PP (group 1) is lower than the wear rate of pure HIPS (group 2). But the wear rates of PP/Pine flour (group 3) polymer composites values are lower than the wear rate of HIPS/Pine flour. As a result, it was observed that the wear rate values increased with the increasing rate of HIPS. Similar situation was observed in the PP/HIPS/Beech flour polymer composites.

**Friction coefficients of the PP/HIPS/wood flour polymer composites**

There are actually two coefficients of friction that can be measured. But in this study only the static friction coefficient values were given. The static coefficient of friction (ssμ) is found from the force that is just enough to start the block moving. Once the block is moving it is possible to measure the dynamic coefficient of friction (dμ) from the force that is just enough to keep the block moving. For most combinations of materials dμ is less than ssμ. Friction performance is shown in Figure 4 when speed was 100 mm/min; load separately was 1.96, 2.94, 3.92, 4.9 and 6.86 N respectively. It is seen that the HIPS, wood flour and load contents had a great effect on the static friction coefficient of the composites. Figure 4 shows that the static friction coefficient of pure PP is much higher than that of pure HIPS and PP/HIPS/wood flour polymer composites at any loads at 100mm/min. So, the friction coefficients gradually increased with the increasing loads.
Morphological properties of the PP/HIPS/wood flour polymer composites
SEM patterns of the fractured surfaces of the composites can provide information about the interfacial compatibility between the wood flour and PP matrix. The image also shows the random distribution of wood flour in the matrix. Adhesion between wood flour and PP was obtained. SEM patterns of the fractured surfaces of the composites are represented in (Fig 5).

**Fig. 5.** SEM micrographs of the PP/HIPS/pine&beech flour polymer composites

**CONCLUSIONS**
The increasing of pine and beech flour contends to the PP resulted in higher vicat values. But, the increasing of pine and beech flour contends to the HIPS resulted in lower vicat values. Vicat softening point of PP/HIPS/wood flour composites shows a decrement as the HIPS concentration increases. The tribological properties of PP/HIPSblends filled with pine and beech flour were studied at different loads under dry sliding. The effects of HIPS and wood flour content on the wear and friction behavior were discussed. Wear rate increased gradually with the increase of the applied load from 5 to 20 N. It was inconsistent that the applied load affected on the wear rate of PP/HIPS polymer composites. Wear rate of pure PP is lower than the wear rate of pure HIPS. But the wear rates of PP/Pine flour polymer composites values are lower than the wear rate of HIPS/Pine flour. As a result, it was observed that the wear
rate values increased with the increasing rate of HIPS. HIPS, wood flour and load contents had a great effect on the static friction coefficient of the composites. The static friction coefficient of pure PP is much higher than that of pure HIPS and PP/HIPS/wood flour composites at any loads at 100 mm/min. So, the friction coefficients gradually increased with the increasing loads.

**ACKNOWLEDGEMENT** This work has been supported by the Marmara University, Scientific Research Research Projects Committee.

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