Utilization of Melamine Impregnated Paper Waste as a Filler in Thermoplastic Composites

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The potential utilization of melamine impregnated paper (MIP) waste in thermoplastic composites was investigated. Composites were also manufactured utilizing wood flour (WF) at the same filler rates for comparison. The composites were manufactured using a compression molding method. The effects of filler type and filler rate on the mechanical properties of low-density polyethylene (LDPE)-based composites were evaluated. Mechanical properties, such as tensile and flexural strengths, were determined in accordance with ASTM D638 (2001) and ASTM D790 (2003), respectively. Results showed that filler type and filler content had significant effects on all mechanical properties investigated. Both fillers improved all mechanical properties except for tensile strength and elongation at break of LDPE. In conclusion, MIP waste has a potential to be utilized in thermoplastic-based composite manufacturing and might generate some economic and environmental benefits.

Keywords: Waste melamine impregnated paper; Thermoplastic; Composite; Mechanical properties

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INTRODUCTION

The global utilization of wood-based panels has increased tremendously. Growing population, development of the construction sector, and need for new furniture have played an important role in this increase. Over the years, customers' expectations from furniture have evolved, and a variety of products at affordable price is vital. Application of new coating techniques, films, thin veneers, pattern printing, and lacquer paints on the surface of wood-based panels has provided a number of choices for the manufacturer. These materials provided good aesthetic appearance and improved properties. Melamine impregnated paper (MIP) is the most preferred coating materials used in wood-based panel sector due to its advantages such as resistance to chemicals, fungi, insects, heat, water/moisture, high wear, homogeneous structure, low formaldehyde emission, color stability, *etc.* (Nemli 2003a; Aksu 2009).

Several studies have been conducted to evaluate the effect of MIP coating on surfaces and on the various board properties. Their effect on mechanical properties (Lee and Kim 1985; Chow *et al.* 1996; Nemli 2000; Bektaş *et al.* 2002; Nemli *et al.* 2005a), physical properties (Nemli 2000; Bektaş *et al.* 2002), formaldehyde emissions (Nemli 2000; Nemli and ve Çolakoğlu 2005; Wang *et al.* 2007; Liu and Zhu 2014), and resistance to heat, light, and chemicals were investigated and improved performances were reported.

Turkey is a major producer of medium-density fiberboard (MDF) and particleboard. The total annual capacity of the sector is 11,517,120 m³; of which 5,545,920 m³ is particleboard and 5,971,200 m³ is fiberboard (MDF) (Özmen *et al*. 2014; İstek *et al*. 2017). Approximately 94% of all manufactured panels are MIP-coated. It was reported that annual MIP utilization of Kastamonu Integrated Wood Industry (KEAS) and Starwood Forest Products Inc. were around 420,000,000 and 80,000,000 m², respectively. The MIP wastes (approximately 0.6%) are generated either during melamine impregnation line (MIP production) or during coating of the MIP on panel surfaces (MIP pressing section). A large portion of the wastes are generated in the melamine impregnation line. These MIPs contain approximately 52% to 53% adhesive (urea, melamine formaldehyde), 40% alpha-cellulose paper, and 7% to 8% other chemicals, such as separator, anti-dust, anti-block, *etc.* (Barbu and Steinwender 2009). It is reported that KEAS generates approximately 15,210,000 m²/year (2,737,800 kg) MIP wastes. Waste MIPs are not suitable for generating energy through direct combustion due to the chemicals they contain (Mengeloglu *et al.* 2015). Special boilers resistant to high temperature and pressure are also needed (Barbu and Steinwender 2009). Landfilling of MIP wastes is also not viable option because it costs extra and poses some environmental risks. Additionally, it leads to loss of a potential raw material. It is believed that utilization of MIP waste in manufacturing might avoid the environmental pollution and contribute to the economy.

Several studies have been conducted on the potential utilization of MIP waste in composites. Varga *et al*. (2004) used a mixture of urea formaldehyde (UF) and waste MIP at various ratios as an adhesive during particleboard manufacturing. It was stated that modulus of rupture (MOR), internal bond (IB), and formaldehyde emission properties of the manufactured particleboards satisfied the requirements of related standards. It was concluded that the addition of waste MIP made it possible to utilize less adhesive in particleboard- manufacturing with improved or similar properties to UF bonded particleboard. In another study, Fur *et al*. (2004) utilized MIP wastes in particleboard manufacturing. They were either added directly to the particle furnish as a powder form or used as a melamine substitute during in melamine-urea formaldehyde (MUF) resin preparation. It was concluded that particleboard were successfully produced with both methods. Alpar and Winkler (2006) also investigated the potential utilization of 5%, 10%, and 20% additions of MIP waste (powder form) as adhesive replacement in particleboard production. Without using additional adhesive, the particleboards with acceptable properties were produced with the 20% MIP waste utilization. In another study, Basboğa *et al*. (2017) utilized 10% and 15% MIP wastes (coming from MIP production section and from MIP pressing section) as an adhesive replacement in particleboard manufacturing. Waste types were used both alone and as a 1:1 mixture of the two. The particleboard produced with both 10% and 15% first type-MIP wastes loading (MIP manufacturing) provided mechanical properties exceeding standard values. It was concluded that this type of MIP waste can be utilized in particleboard manufacturing as an adhesive replacement. Basboğa *et al*. (2018) also investigated the effectiveness of MIP wastes in particleboard manufacturing. For this purpose, they produced particleboard using both MIP wastes (powder) and neat resin (used in impregnation process) separately as an adhesive. A study conducted by Basboğa *et al*. (2018) showed that MIP wastes can be effectively used as an adhesive replacement in particleboard manufacturing.

The MIP wastes are also utilized as a filling material in various composite products. Silva *et al*. (2012) utilized MIP wastes as a wood particle replacement in medium-density particleboard. Approximately 4%, 8%, and 12% MIP wastes were first mixed with 92%, 96%, and 88% wood particles, respectively and then this mixture was used in core layer of particleboard. In the study, a fixed amount of adhesive was used throughout. With the exception of 12% MIP wastes addition, the other particleboards provided properties that met Brazilian standards. In contrast, Çavdar *et al*. (2013) used waste MIPs (10%, 20%, 30%, 40%, and 50%) as filling material in the core layer of oriented strand board (OSB) production and 14% UF adhesive was used as a binder. It was determined that the use of waste MIPs positively affected the mechanical and physical properties. In another study, Ayrilmis (2012) evaluated the usage of waste MIPs in the production of light MDF manufacturing. Amounts of 5%, 10%, and 20% MIP wastes (overlay and decorative paper) were blended with wood fibers with 11% UF resin. Dimensional stability and mechanical properties of the resulting MDF products were improved because of the addition of waste paper into the formulations.

Utilization of MIP wastes in thermoplastic materials was also reported by Mengeloğlu *et al*. (2015). They produced thermoset composites with MIP wastes (without using any additional adhesives) and thermoplastic composites using 1:1 mixture of MIP wastes and low-density polyethylene (LDPE). Thermoset composites provided higher strength and modulus values for tensile and flexural modulus tests compared to thermoplastic composites. However, only one mixture for thermoplastic composite was studied.

Previous studies showed that MIP wastes have a great potential to be utilized not only as an adhesive replacement but also as a filling material in particleboard and other wood-based boards production. However, the MIP wastes performance in thermoplastic composites at various filling concentrations has not been investigated thoroughly. In this study, the effects of MIP waste contents on some physical and mechanical properties of LDPE-based thermoplastic composites were determined and their performance was also compared with the wood flour filled-LDPE based thermoplastic composites.

EXPERIMENTAL

Materials

In this work teak wood flour (WF) obtained from wood saw dust in Kahramanmaraş (Turkey) were used as lignocellulosic filler material. The saw dust was ground into flour form using a Wiley mill (Altundal; Kahramanmaraş, Turkey). The MIP were kindly donated by Kastamonu Entegre, MDF Plant, Adana, Turkey. The WF and MIP particles that passed through 40-mesh and remained on a 60-mesh screen (0.25 mm and 250 µm, respectively) were used after drying for 24 h at the temperature of 103±2 °C. The LDPE (F2-12) polymeric matrix was purchased from Petkim Petrochemical Co. (Izmir, Turkey).

Composite Manufacturing

Composite materials were produced using a compression molding method. The manufacturing plan is given in Table 1. According to manufacturing procedure; lignocellulosic fillers and LDPE were first mixed in a mixer (≈ 1000 rpm for 5 s) to obtain a homogeneous blend and then this mixture was compression molded into 4 mm × 160 mm × 160 mm size mold (Carver Press) at 185 °C press temperature and at 1.0 bar (10 tons/m^2) press pressure for 10 min.

Test specimens were conditioned at a relative humidity of 65% ± 5% and a temperature of 23 ± 2 °C. The density was determined by a water displacement technique according to the ASTM D792 (2007) standard. Flexural, tensile, and hardness of all specimens were determined according to ASTM D790 (2003), ASTM D638 (2001), and

ASTM D2240 (2010), respectively. Flexural and tensile tests were implemented on a Zwick 10 KN (Ulm, Germany) machine. Morphological properties of samples were analyzed using scanning electron microscopy (SEM), (EVO LS10; Carl Zeiss, Jena, Germany). For this analysis, the samples were dipped into liquid nitrogen and snapped in half to attain clean surfaces. The specimens were then placed on a specimen holder and sputtered with gold (Sputter Coater 108 Auto; Cressington, London, England) to prevent charge accumulation of the electron absorbed by the specimens with 10 mA in 120 s. For statistical analysis, Design-Expert® version 7.0.3 statistical software (Minneapolis, MN, USA) was used.

Table 1. Manufacturing Plan of Woof Plastic Composites

Sample ID	LDPE (%)	MIP (%)	WF (%)
PE	100	0	0
PE-25MIP	75	25	0
PE-50MIP	50	50	0
PE-75MIP	25	75	0
PE-25WF	75	0	25
PE-50WF	50	0	50
PE-75WF	25	0	75

LDPE: Low-density polyethylene; MIP: Melamine impregnated paper; and WF: Wood flour

RESULTS AND DISCUSSION

Density values of the samples tested were in the range of 0.87 to 1.2 g/cm³. The interaction graph showing the effects of filler type and ratio on the density of composites is presented in Fig. 1. Statistical analysis showed that the filler content and filler type had significant effect on density ($P < 0.0001$). Composite densities were increased with the addition of lignocellulosic materials due to their higher cell wall densities. Similar results were also reported by others (Stokke *et al.* 2014; Karakuş and Mengeloğlu 2016; Çavuş and Mengeloğlu 2016; Mengeloğlu and Çavuş 2019). In the case of filler type, both MIP and WF as fillers increased the density of the composites, but at high filler levels, this increase was more pronounced for MIP filled ones (Fig. 1).

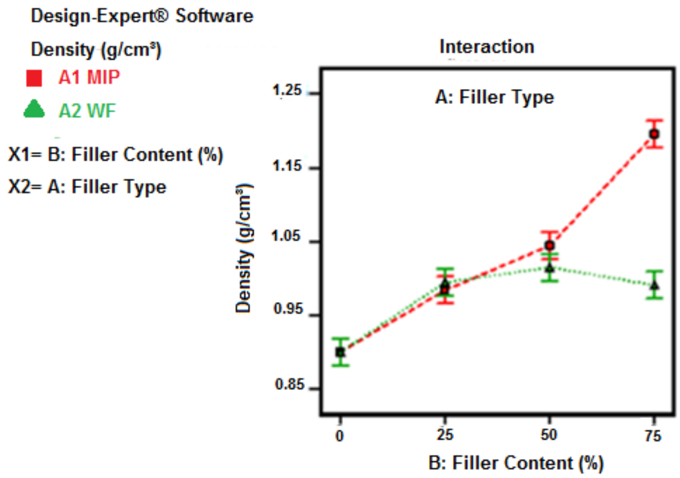

Fig. 1. The interaction graph of the density values of samples with filler type and content

There are studies reporting that small particle size may provide increased composite densities because these small filler types may reduce the gap between filler and the polymer matrix (Çavdar 2011; Behazin *et al.* 2017; Chaudemanche *et al.* 2018). Because both filler types had similar particle sizes in this study (Figs. 2a and 2b), filler size might not be the possible reason for this difference. It is believed that the higher density of MIP filler might result in an increase in density because they are produced using cellulose fibers. Larger gaps were noticed between polymer and filler in composites having WF as filler (Figs. 2c and 2d). This might be the reason for the lower density of the composites having WF at high filler content levels.

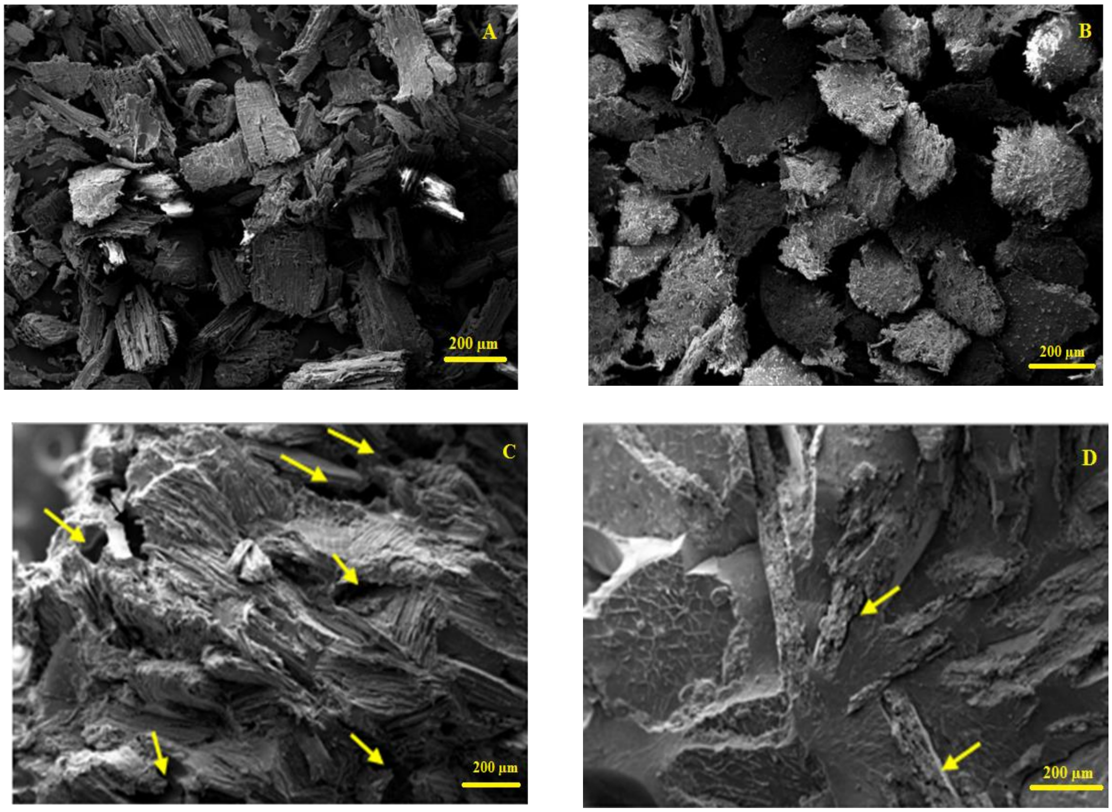

Fig. 2. SEM images of fillers and polymer composites: A) WF; B) MIP; C) PE-50WF; and D) PE-50 MIP

Tensile strength (TS) values of the samples were in the range of 2.68 to 8.76 MPa. The interaction graph of TS is presented in Fig. 3. Statistical analysis showed that the filler proportional amounts and filler types had a significant effect on TS ($P < 0.0001$). The TS values were reduced with increase of both MIP and WF contents (Fig. 2). Similar reduction of TS values with the increase in lignocellulosic filler ratio in polymer matrix was reported by others (Mengeloğlu ve Karakuş 2008). Plastic composites produced with coupling agent improved adhesion between hydrophilic (polar) fillers and hydrophobic (nonpolar) polymers. This result is consistent with previous studies (Poletto 2017; Cavus 2020).

Tensile modulus (TM) values of the samples were in the range of 95 to 484 MPa. The interaction graph of TM is presented in Fig. 4. Statistical analysis showed that the filler content and filler type had a significant effect on TM values ($P < 0.0001$). Both filler type increased TM values, but it was more pronounced at high filler contents. The MIP and WF having higher cell wall density in the composite usually increased the modulus values of the resulting composites. This can be explained by the rule of mixtures (Matuana *et al*. 1998), meaning that there is a correlation between modulus values of composites and percentage and modulus values of its constituents. Increased TM values of lignocellulosic material-filled polymer composites were reported by other researchers (Rowel 2006; Mengeloğlu *et al*. 2015).

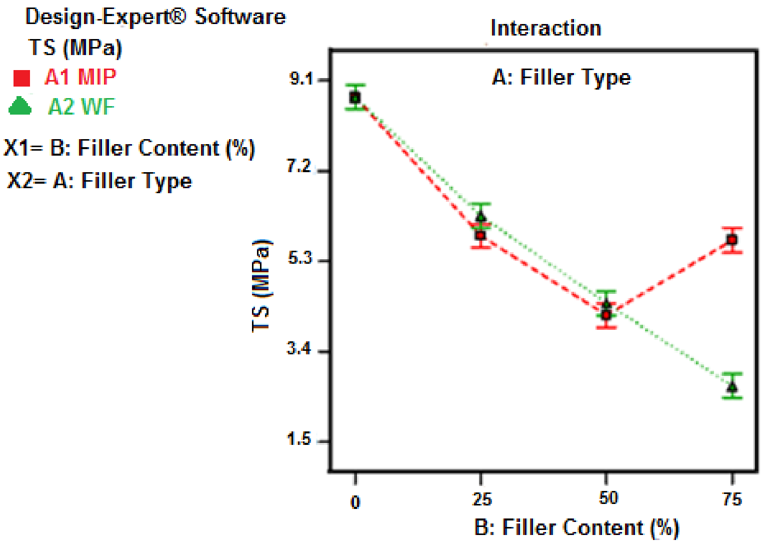

Fig. 3. The interaction graph of the tensile strength

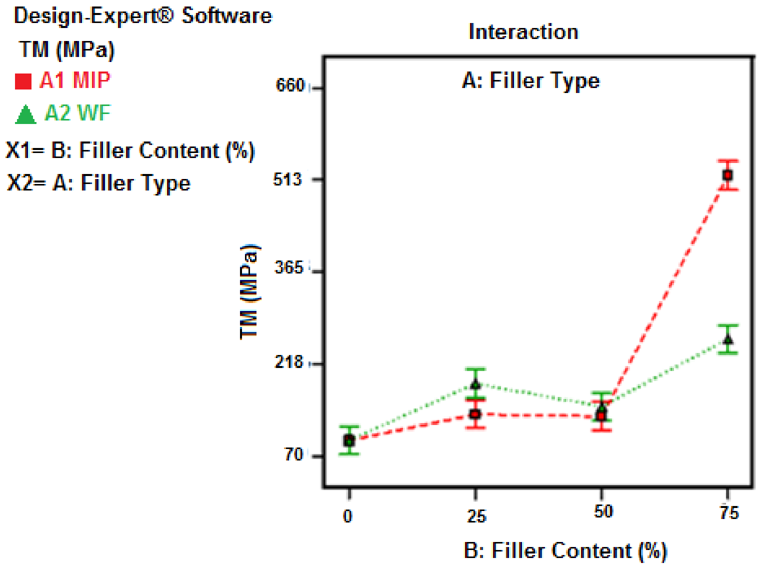

Fig. 4. The interaction graph of the tensile modulus

The elongation at break (EatB) values of the samples were in the range of 1.4% to 335%. The interaction graph of EatB is presented in Fig. 5. Statistical analysis showed that the filler amounts had a significant effect on EatB ($P < 0.0001$). There was also no significant difference noticed between filler types ($P = 0.907$). Both filler types significantly reduced the EatB values of the composite samples. This reduction was a normal consequence of the increase in filler amount that limits the elongation of the samples (El-Shekeil *et al*. 2012). Addition of the WF and MIP fibers as fillers led to a lower resistance to break. The EatB values usually decrease with the increase in modulus of the composites as reported for composites produced with various wood flours (Mengeloğlu and Karakuş 2008).

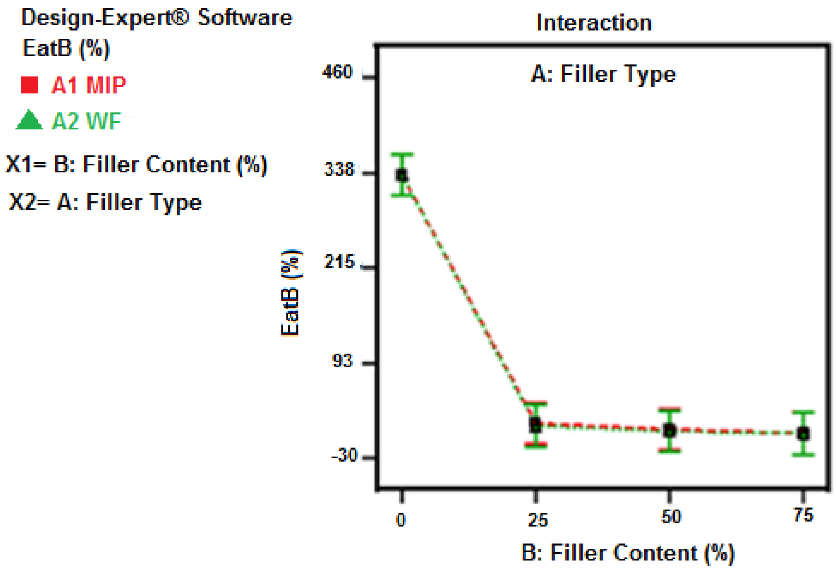

Fig. 5. The interaction graph of the elongation at break

Flexural strength (FS) values of the samples were in the range of 5.78 to 12.8 MPa. The interaction graph of FS is presented in Fig. 6. Statistical analysis showed that the filler content had a significant effect on FS ($P < 0.0001$). There was also no significant difference noticed between two filler types ($P = 0.104$). It has been found that the FS values in LDPE varied according to filler type. The FS values displayed similar properties with both WF and MIP participations as fillers. Similar results were reported by others (Mengeloğlu *et al*. 2015; Başboğa 2018; Afzaluddin *et al*. 2019).

The flexural modulus (FM) of the samples was in the range of 202 to 1235 MPa. The interaction graph of FM is presented in Fig. 7. Statistical analysis showed that the filler content ($P < 0.0002$) and filler type ($P < 0.0001$) had significant effects on FM. As the filler participation increased, the FM values also increased. Lignocellulosic fillers positively influenced the FM values, as has been reported (Varga *et al.* 2004; Alpar and Winkler 2006; Ayrılmış 2012; Çavdar 2013). It should be noted that 50% and 75% MIP and WF-filled composites provided FS and FM values of over 6.9 MPa and 340 MPa, respectively. These results met the standard requirements of ASTM 6662-01 (2017) (requirements for polyolefin-based plastic lumber decking boards).

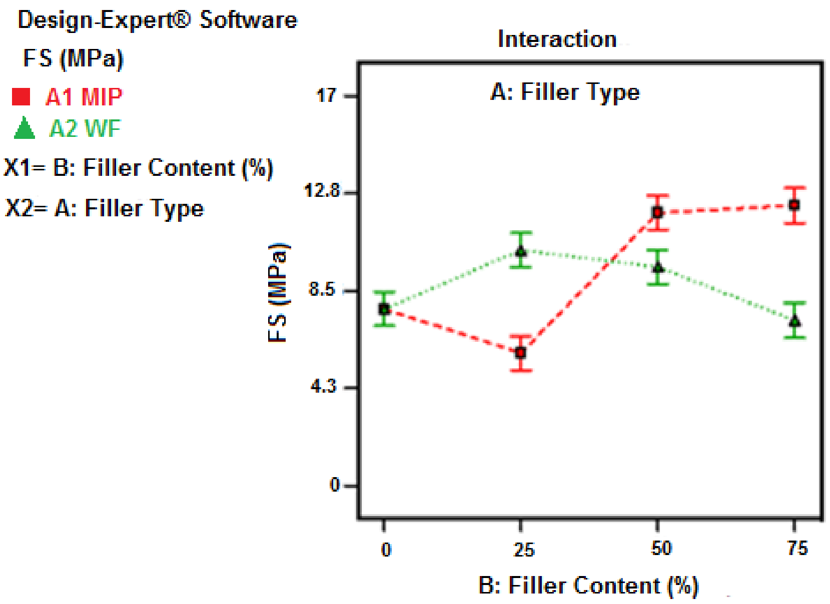

Fig. 6. The interaction graph of the flexural strength

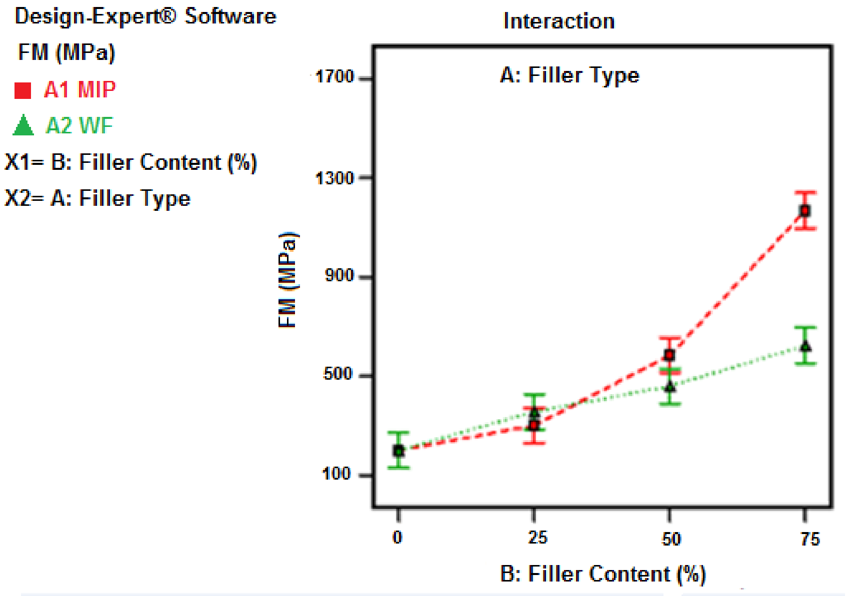

Fig. 7. The interaction graph of the flexural modulus

Hardness strength (Shore D) values of the samples were in the range of 47 and 65. The interaction graph of the hardness strength is presented in Fig. 8. Statistical analysis showed that the filler content had a significant effect on hardness strength ($P < 0.0001$). There was also no significant difference observed between the two filler types ($P = 0.750$). Hardness values increased as filler participation rate increased in polymers composites. The WF or MIP fillers had a positive effect on Shore D values. Similar results of increased hardness and fragile matrix for filler incorporated polymeric matrix were also reported by others (Cavdar 2011; Karakuş ve Mengeloğlu 2016). Composites produced with 75% filler provided hardness values considered as “extra hard material” (Shore D over 60), while the rest of the produced samples including neat polymer were measured as “hard material” (Shore D 30-60).

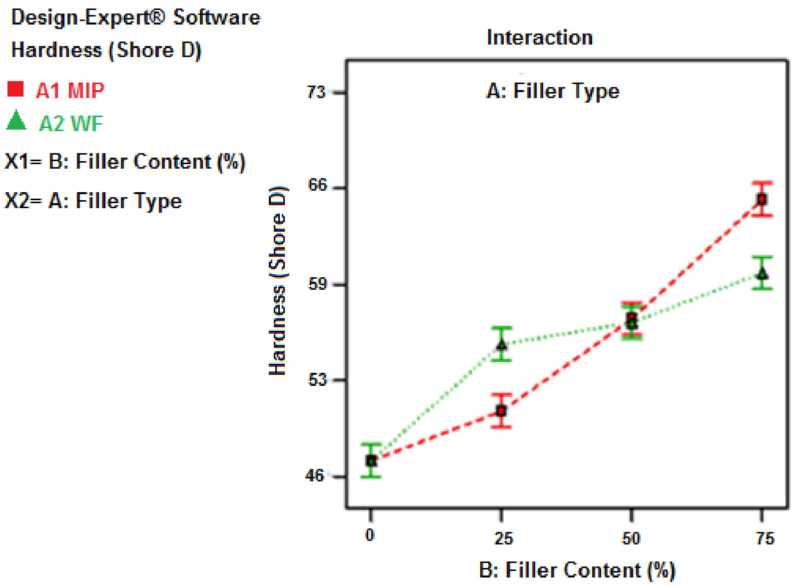

Fig. 8. The interaction graph of the hardness

CONCLUSIONS

In this study, low density polyethylene (LDPE)-based thermoplastic composites were produced using various amounts of wood flour (WF) and waste melamine-impregnated paper (MIP) as fillers at 25%, 50%, and 75% content levels. The following conclusions were derived:

1. Both MIP wastes and WF can successfully be used as filling materials in thermoplastic-based composite production.
2. Addition of MIP wastes and WF in LDPE matrix increased the density of produced composites.
3. The thickness swelling (TS) values decreased with addition of MIP and WF up to 50%. Further increase of MIP waste amounting to 75% resulted in an increase in composite strength values due to the increased density.
4. The composites with 50% and 75% MIP and WF as fillers provided FS and FM values that met the standard requirements of ASTM 6662-01 (2017) (6.9 and 340 MPa, respectively).
5. The MIP and WF fillers in LDPE adversely affected the elongation at break (EatB) of the composites produced. However, fillers positively affected the flexural modulus (FM) and hardness properties.
6. Addition of 75% filler in polymer matrix made the resulting composites "extra hard material" (Shore D over 60).

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