

UTILIZATION OF TINDER FUNGUS AS FILLER IN PRODUCTION OF HDPE/WOOD COMPOSITE

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ABSTRACT

Selected physical and mechanical properties of high density polyethylene (HDPE) composites filled with various mixtures of wood flour and tinder fungus (*Fomes fomentarius*) were investigated. For this aim, different mixtures of tinder fungus flour and wood flour (0/40, 10/30, 20/20, and 30/10, and 40/0) (by weight) were compounded with HDPE with a coupling agent (maleic anhydride grafted polyethylene (MAPE) in a twin screw co-rotating extruder. The test specimens were produced by injection moulding machine. The thickness swelling and water absorption of the HDPE/wood composites significantly decreased with increasing content of the tinder fungus flour. The mechanical properties of the composites were negatively affected by increasing amount of tinder fungus flour but there were no significant differences up to 30 wt % tinder fungus content, except for the tensile strength. The optimum physical and mechanical properties for the filled HDPE composites were found to be a 10/30/60/3 formulation of wood flour, tinder fungus, HDPE, and MAPE, respectively.

KEYWORDS: Fungus, mechanical properties, swelling, thermoplastic composites, wood.

INTRODUCTION

There are a plenty of research studies on the utilization of various wood species for the production of wood plastic composites (WPCs) (Stark and Berger 1997, Clemons 2002, Ayrilmis and Kaymakci 2013, Mengeloglu and Karakus 2008, Kim et al. 2009, Gacitua and Wolcott 2008). In addition to wood flour or fiber, the potential use of many agricultural wastes such as

wheat, chestnut and walnut shell, sunflower seed, rice husk, kenaf, cornstalk, hemp, and jute as filler in thermoplastic composites have been investigated (Rowell 1996, Youngquist et al. 1996, Caulfield et al. 1998, Chow et al. 1999, Bledzki et al. 2004, Kaymakci and Ayırlımsı 2014, Ayırlımsı et al. 2013a, b). This was because the sustainable utilization of forest resources has been adversely influenced by the increasing population of the world. Fortunately, great progresses could be seen in the manner of looking for alternative raw material sources. Mushrooms are important in the ecosystem because fungal decomposition of biomass is a necessary part of the natural material. Fruit bodies of mushrooms are appreciated, not only for texture and flavour but also for their chemical and nutritional properties. These functional characteristics are mainly due to their chemical composition. The tinder fungus (TF) (*Fomes fomentarius*) belongs to the *Polyporaceae* and attacks old and weak hardwoods, mainly birches and beeches, by white rot. It grows on trees throughout the temperate and boreal regions of the northern hemisphere, and is found in Europe, northern Asia and in North America. Although it can often be seen on rotting logs, the fungus itself is rot resistant, because it has the ability to produce compounds with anti-pathogenic characteristics. In practice though, the fungus often dies because the wood it is growing on has reached advanced stage of decay where there are not enough nutrients or sufficient fibrous structure to support it any more. The optimal temperature for growing of the TF is between 27 and 30°C with a maximum between 34 and 38°C (Meghan et al. 2013). Besides its medicinal use, the TF was also applied for different commodities in Germany, Hungary, and in some parts of former Yugoslavia, TF was used for making caps, chest protectors, and other clothing articles. In Germany and former Czechoslovakia, large fruit bodies were often used for decorative purposes such as flower pots (Dvořák et al. 2005; Grienke et al. 2014). Average metal contents in the samples of *Fomes fomentarius* were 14.67 ± 8.20 Cu, 2.31 ± 1.03 Cd, 89.45 ± 176.76 Mn, 5.45 ± 2.45 Pb and 60.50 ± 59.48 ppm Zn. (Dvořák et al. 2005). The 5000-year-old Iceman Ötzi carried four pieces of *Fomes fomentarius* fruit body, concluded to be for use as tinder. There is so little information about the utilization of tinder fungus growing on the standing trees as filler in the production of thermoplastic composites. The goal of this study was to determine selected physical and mechanical properties of the HDPE composites filled with various mixtures of wood flour and fruit body flour.

MATERIAL AND METHODS

Materials

The kidney-shaped fruit body *Fomes fomentarius* (L.) Fr. (Fig. 1) which was about 30 cm wide was obtained from the stem of a standing oak tree in Belgrade forest in Istanbul, Turkey.



Fig. 1: *Tinder fungus (TF) specimen.*

The fruit body was dried in an oven at 60°C for 10 h to a moisture content of 20-30 % based on the oven-dry solid weight. Following the drying, the fruit body were then processed by a rotary

grinder without adding additional water. Finally, the Tinder fungus fruit body flour (TF) passing through a U.S. 35-mesh screen and was retained by a U.S. 80-mesh screen. The TF was then dried in a laboratory oven at 100°C for 24 h to moisture content of 1-2 %. Pine (*Pinus nigra* Arnold ssp. *pallasiana*) wood flour (WF) having a size of U.S. 80-mesh was obtained from a commercial WPC plant in İstanbul, Turkey. WF dried in a laboratory oven at 100°C for 15 hours to moisture content of 1-2 % before manufacturing process.

The high density polyethylene (HDPE) (MFI/230°C/2.16 kg = 5.0 g/10 min) supplied by a Petkim Petrochemical Corporation in Izmir, Turkey, was used as the polymeric material. Maleic anhydride grafted polyethylene (MAPE) (Optim-E156, MAH content: 1.2 wt. %, MFI 190°C/2.16 kg = 4.5 g/10 min, density = 0.95 g·cm⁻³) was used as the coupling agent (Plus Polymers Pvt. Ltd. in India.)

Preparation of composites

The TF, WF and HDPE with MAPE granulates were processed in a 30 mm co-rotating twin screw extruder with a length-to-diameter ratio of 30:1. The barrel temperatures of the extruder were controlled between 170 and 190°C. The temperature of the extruder die was held at 200°C. The extruded strand passed through a water bath and was subsequently pelletized. The pellets were dried for 3-4 h to moisture content of 0-1 % in an oven. The temperature used for injection molded specimens was 170-190°C from feed zone to die zone. The specimens were injected at injection pressure between 5 and 6 MPa with cooling time about 30 s. Finally, the specimens were conditioned at a temperature of 23°C and relative humidity of 50 % according to ASTM D 618: 2013. The composite group consisted of TF, WF, HDPE, and MAPE in varying proportions. The raw material formulations of the composites are given in Tab. 1.

Tab. 1: Compositions of the evaluated composite formulations.

Composite code	Wood flour	Tinder fungus	HDPE	MAPE	Density
	(wt %)	(wt %)	(wt %)	(wt %)	(g·cm ⁻³)
A	40	0	60	3	1.010
B	30	10	60	3	1.012
C	20	20	60	3	1.015
D	10	30	60	3	1.018
E	0	40	60	3	1.020

Property testing

Determination of physical and mechanical properties of composites

The thickness swelling (TS) and water absorption (WA) tests were carried out according to ASTM D 570: 2010 specifications. The test specimens were in the form of a disk 50.8 mm in diameter and 3.2 mm in thickness. The conditioned specimens were entirely immersed for 1-day, 7-days, and 14 days in a container of water at 23±2 °C. At the end of each immersion time, the specimens were taken out from the water and all surface water was removed with a clean paper towel. The specimens were weighed to the nearest 0.01 g and measured to the nearest 0.001 mm.

The flexural tests were conducted in accordance with ASTM D 790: 2002 using a Lloyd testing machine at a rate of 1.3 mm·min⁻¹ crosshead speed. Dimensions of the test specimens were 3.5 x 13.2 x 128 mm. The tensile tests were conducted according to the ASTM D 638: 2012. Tensile specimens were tested with a crosshead speed of 5 mm·min⁻¹ in accordance with ASTM D 638: 2012. The izod pendulum impact strength of the notched specimens (notch tip radius: 0.25 mm) was performed according to ASTM D 256: 2016 using an impact testing machine.

Statistical analysis

Analysis of variance (ANOVA) ($p < 0.01$) was used to determine the effect of amounts of TF and WF on the selected physical and mechanical properties of the HDPE composites using SPSS 17.0 statistical package program. Significant differences among the average values of the composite types were determined using Duncan's multiple range tests.

RESULTS AND DISCUSSION

Physical properties

Adding the TF or WF into the HDPE increased the density of the composites. This was expected because the cell density of the cellulose (average $1.3 \text{ g}\cdot\text{cm}^{-3}$ after extrusion and injection molding) was much higher than the density of injection molded HDPE composite (Klyosov 2007). Similar results have been reported for the injection-molded lignocellulosic/polymer composites (Stark et al. 2004, Tan et al. 2013, Ayrlimis et al. 2010). The density of lignocellulosic filled thermoplastic composites was ranged from 1.010 to $1.020 \text{ g}\cdot\text{cm}^{-3}$ while it was found to be average $0.899 \text{ g}\cdot\text{cm}^{-3}$ for the unfilled HDPE composites (Tab. 2).

The TS and WA values of the filled HDPE composites are presented in Fig. 2. The TS and WA values significantly decreased with increasing TF content. Statistical analysis found some significant differences among the means of filled HDPE composites for the TS and WA values. Significant differences were determined individually for these tests by Duncan's multiple-comparison tests. The results of Duncan's multiple range tests for the TS and WA are shown by letters in Figs. 2 and 3, respectively. The lowest TS and WA values were found in the specimens containing 40 % TF (composite code: E), while the highest TS and WA values were found in the specimens containing 40 % WF (composite code: A) after 1 day of submersion in water. Similar trends were also observed for 7 and 14 days of submersion (Fig. 2).

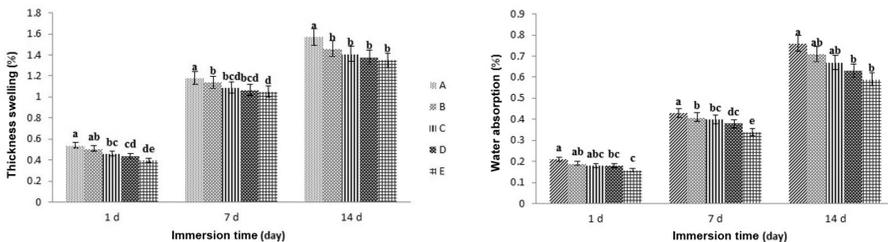


Fig. 2: Effect of the tinder fungus flour content on the thickness swelling and water absorption of HDPE/wood composites (the same letters in each column show there is no significant difference ($p < 0.01$) among the composite groups).

The moisture absorption in thermoplastic composites is mainly due to the presence of lumens, fine pores, and hydrogen bonding sites in the WF, the gaps and flaws at the interfaces, and the micro cracks in the matrix formed during the compounding process. As compared to the WF filled HDPE composites, the lower TS and WA values of the HDPE composites filled with the TF could be due to lower hygroscopic sites of the TF such as hydroxyl groups, as well as lower gaps and micro cracks in the composites.

Mechanical properties

The HDPE composites filled with the TF or WF showed no significant difference in the flexural modulus while the significant differences ($p < 0.01$) in the flexural strength were observed. Significant differences between the groups in the flexural strength and modulus were displayed in Fig. 3, respectively. The incorporation of the TF into the wood/HDPE composites improved the flexural properties of the composites. The modulus of thermoplastics is lower than that of cellulose. The cellulose in the TF positively affected the flexural modulus of the composites. Similarly, the flexural strength of the composites improved with the addition of 10 wt % TF. The flexural strength and flexural modulus generally decreased with increasing the TF content from 10 to 40 % in the composite (Tab. 1). For example, the average values of the flexural strength and flexural modulus of the composites containing 40 % WF (composite code: A) were found to be 46.2 and 3674 MPa, respectively as compared to the composites containing 40 % TF (composite code: E) which were about 39.2 and 3395 MPa, respectively. This can be related to the lower cellulose content of the TF. Cellulose is mainly responsible for the strength of the lignocellulosic materials. In a previous study, flexural strength and flexural modulus values were found to be 48.1 and 5005.2 MPa for polymer composites produced with 40 beech WF and 57 % polypropylene with 3 % MAPP while these properties were found to be 41.9 and 4355 MPa for polymer composites produced with 40 pine cone flour and 57 % polypropylene with 3 % coupling agent, respectively (Ayrilmis et al. 2010).

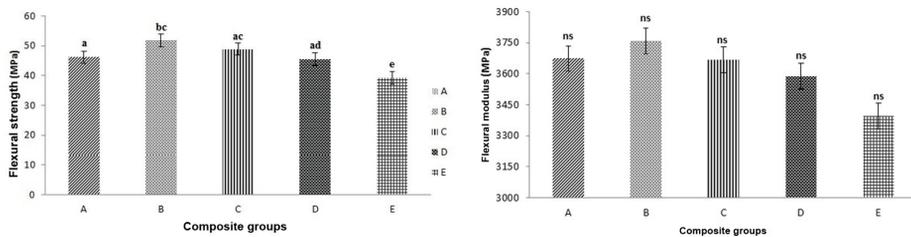


Fig. 3: Effect of the tinder fungus flour loading on the flexural strength and modulus of HDPE/wood composites.

Tensile strength, tensile modulus, and elongation at break of the composites are presented in Figs. 4 and 5. Significant differences between the composite groups were also determined individually for each test by Duncan's multiple comparison tests as displayed in Figs. 4-5. In general, the tensile properties of the composites were negatively affected by increased TF content. For example, the average tensile strength, tensile modulus and elongation at break values of the composites containing 40 % WF (composite code: A) were 27.2, 4279 MPa, and 3.7 % as compared to the composites containing 40 % TF (composite code: E) which were about 15.9, 3533 MPa, and 2.1 %, respectively. This result might be due to the dissimilarities and lack of the adhesion between the nonpolar HDPE matrix and polar TF and WF. Similar results were also reported in previous studies (Nourbakhsh and Ashori 2008, Ayrilmis et al. 2013a, b). It is worthy of note that the tensile modulus of the HDPE composites progressively increased with the addition of TF and WF.

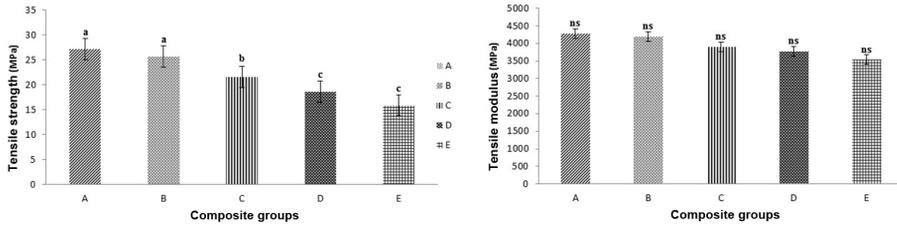


Fig. 4: Effect of the tinder fungus flour loading on the tensile strength and modulus of HDPE/wood composites.

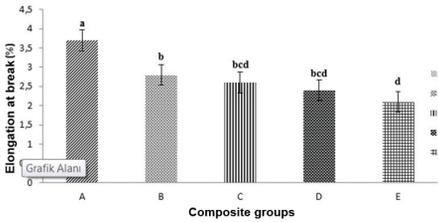


Fig. 5: Effect of the tinder fungus flour loading on the elongation at break values of HDPE and HDPE/wood composites.

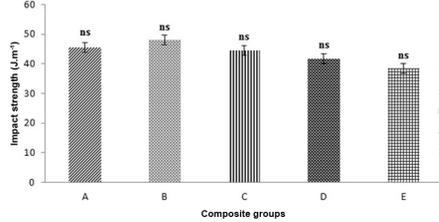


Fig. 6: Effect of the tinder fungus flour loading on the impact bending strength of HDPE/wood composites.

Tensile strength of composites containing WF was significantly higher than that in composites containing TF. This could be explained by a strong interfacial adhesion between the HDPE and the WF due to their higher cellulose content, since cellulose is the main component providing the wood's strength and structural stability (Ayrilmis et al. 2010). The test results revealed that the effect of wood was notable in the material properties of the composites. Wood is a lignocellulosic material made up of three major constituents (cellulose: 42-44, hemicelluloses: 27-28, and lignin: 20-28 %) with some minor constituents (extractives: 3-4 %) (Bhaskar et al. 2011). According to literature research, an increase in the composite's strength can be ascribed to higher cellulose and lignin contents, as well as better dispersion and adhesion to the matrix (Bledzki et al. 1998, Bledzki and Gassan 1999). The better interfacial adhesion between WF and HDPE, due to the high cellulose content, increases the toughness or ductility (Marcovich and Villar 2003). The fruit body of the tinder fungus mushroom contains proteins and polysaccharides, triterpene saponins, coumarins, and phenolic compounds. The protein content in the dried extract of its the fruiting body of tinder fungus mushroom ranges from 7.0 to 8.4 % and polysaccharides from 53.2 to 68.2 % (Troshkova et al. 2012). However, *Pinus nigra* wood contains holocellulose (71.53 %), α -cellulose (50.41), lignin (26.74) and ash (0.19 %) (Kilic et al. 2010). Reduction in the mechanical properties and improvement in the water resistance of the HDPE/wood composites containing a higher amount of the TF can be attributed to the lower holocellulose content in the TF than in the wood.

The impact strength of the HDPE composites decreased with increasing the TF content (Fig. 6). The impact strength of the HDPE composites decreased by 15.4 % as the TF content increased from 0 to 40 wt %. This could be due to the fact that the incorporation of TF into the HDPE composite created the regions of stress concentration that required less energy to initiate a crack in the composite, thereby decreasing the impact strength (Gacitua and Walcott 2009, Rowell 1996). For example, the impact strength of the specimens containing 40 wt % WF was found to be 45.5 J·m⁻² while it was found to be 38.5 J·m⁻² for the specimens containing 40 wt % TF.

The decrement in the impact strength of the composites containing a higher amount of the TF revealed that this flour reduced the polymer chain mobility and therefore its ability to absorb energy during fracture propagation. The poor interfacial bonding between filler and thermoplastics causes micro-cracks to occur at the point of impact, which caused the cracks to easily propagate in the composite (Nourbakhsh and Ashori 2008). The presence of the MAPE compatibilizer improved the dispersion of the TF and WF in the HDPE matrix and led to the uniform distribution of the applied stress. As suggested by previous studies the interaction between HDPE and WF can be improved by the MAPE (Stark and Berger 1997, Ayrimis and Kaymakci 2013, Tisserat et al. 2013, Zabihzadeh and Dastoorian 2009).

CONCLUSIONS

The TS and WA of the wood/HDPE composites significantly decreased with increasing content of the *Fomes fomentarius* fruit body tinder fungus. This was mainly attributed to the lower amounts of the hygroscopic materials, cellulose and hemicelluloses, in the cell walls of the TF. The incorporation of 10 wt % tinder fungus improved the flexural properties. Further increment in the amount of TF decreased the mechanical properties of wood/HDPE composites, but there were no significant differences up to 30 wt % TF content, except for the tensile strength. Based on the findings obtained from the present study, the optimum physical and mechanical properties for the filled HDPE composites were found to be a 10/30/60/3 formulation of WF, TF, HDPE, and MAPE, respectively. The TF can be incorporated into the WPC formulation to improve dimensional stability of the WPC used for outdoor decking or siding.

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