Nadir Ayrilmis, Esra Yildiz

Physical and Mechanical Properties of Thermoplastic Composites Filled with Wood Flour of Underutilized Chaste Tree

Fizička i mehanička svojstva termoplastičnih kompozita punjenih drvnim brašnom rijetko upotrebljavane konopljike

ABSTRACT • The potential use of a lignocellulosic filler, Vitex agnus-castus plant (Chaste tree), which is a deciduous invasive shrub, in thermoplastic composites was investigated. The stems of chaste trees with a diameter of 5-10 cm from Mugla city, Western Turkey, were used for the study. The different amounts (0 to 50 wt%, by 10 % increments) of the wood flour passing through the screen openings of 0.237 mm were added to the polypropylene matrix. Premixed raw materials were put into the volumetric feeder of the twin-screw extruder. The extruder barrel temperature was gradually increased from 170 °C (feeding zone) to the die zone (190 °C) at a constant screw speed (40 rpm). Then, the dried granules were hot-pressed into the 4 mm thick WPC panels at 2 MPa and 190 °C for 5 min. 3 wt% of maleic anhydride grafted polypropylene (MAPP) was added as compatibilizer into the formulation. The WPCs showed an increase in the thickness swelling (0.58 to 5.68 %) as the amount of the filler increased from 10 to 50 wt% in the polypropylene. The bending strength of the polypropylene composites increased from 33.9 to 44.8 MPa as the amount of the chaste wood flour was increased to 30 wt%, but further increase caused the decrease in the tensile strength (25.7 MPa). As for the bending modulus, it increased from 815 to 3250 MPa when the wood content reached 50 wt%. The tensile modulus increased from 1690 to 2253 MPa when the wood content arised from 10 to 50 wt%. The tensile strength, tensile modulus, flexural strength and flexural modulus of the unfilled polypropylene were found to be 19.6 MPa, 1505 MPa, 30.2 MPa and 664 MPa, respectively. According to the test results, it was concluded that the 30-40 wt% of Vitex agnus-castus wood could be efficiently used in the polypropylene composites for the semi-building applications such as decking or siding. The evaluation of underused invasive chaste wood in the production of WPC production may result in an effective way to utilize this resource.

KEYWORDS: Vitex agnus-castus; polypropylene; thermoplastic; composite; lignocellulosic filler

SAŽETAK • U radu je istražena potencijalna primjena lignoceluloznog punila u termoplastičnim kompozitima od biljke Vitex agnus-castus (konopljike), koja je listopadni invazivni grm. Stablo konopljike promjera 5 – 10 cm nabavljene su iz grada Mugla, iz zapadne Turske. U polipropilensku matricu dodane su različite količine drvnog brašna (0 – 50 wt. %, uz povećanje od 10 %) koje je prolazilo kroz sito otvora 0,237 mm. Prethodno pomiješane...
Although it is a broad spreading tree species, Chaste wood (Vitex agnus-castus) is not evaluated in the group of invasive plant atlas of the United States according to the U.S Forest Service (Reichard, 1994). Chaste tree is one of the most used medicinal plants in Europe. The flowers, leaves and seeds of the Chaste tree are used to make baskets and molds (kelter), especially to store or carry fruits (Souto et al., 2013). The flowers of Chaste tree are used for herbal and medical applications. Branches of Chaste tree are used to make baskets and molds (kelter), especially to store or carry fruits (Souto et al., 2020). Chaste wood is not evaluated in the group of high value-added products, such as thermoplastic composites (WPCs). The characteristics of lignocellulosic fillers in thermoplastics. These studies showed that the use of lignocellulosic fillers in thermoplastics, such as polypropylene and polyethylene, could contribute to economic growth and environmental sustainability.

Recent years, the number of WPC manufacturers has increased due to great advantages of lignocellulosic fillers such as low-cost, easy-supply, abundance, recycling, renewability, low-abrasion to machine tools, biodegradability (Španić et al., 2010; Ayrilmis, Yildiz: Physical and Mechanical Properties of Thermoplastic Composites Filled with Wood Flour of Underutilized...
Ayrilmis et al., 2013b; Ayrilmis et al., 2021). In general, lignocellulosic fillers have higher tensile modulus than many thermoplastics, thereby improving the stiffness of the thermoplastic composites (Shahzad, 2012). Particularly, lignocellulosic materials used as fillers are preferred in the commercial production of thermoplastic composites as they reduce the cost of the composite and provide the use of sustainable green materials. The addition of lignocellulosic fillers into the thermoplastics also decreases the amount of the plastics in the WPCs. Similarly, lignocellulosic materials are alternative to some synthetic fiber, such as glass fibers, for some specific applications such as automotive industry. Furthermore, recently sustainable bio-based materials have been preferred for housing applications.

The wood of chaste tree, as an underutilized invasive species, is a promising sustainable raw material source for the WPCs. Its high cellulose content makes it an attractive lignocellulosic filler for thermoplastic composites. Unfortunately, the chaste wood has not been industrially used in the production of wood composites yet. Industrial use of chaste wood may result in a substantial economic impact on the WPC industry. The addition of chaste wood into thermopolypropylene may considerably decrease the cost of WPC due to relatively high cost of polypropylene. According to our extensive search, no study has yet been reported on the evaluation of chaste wood as filler in the production of WPC. The use of chaste wood as filler in the WPC manufacture could be one of the most efficient uses in high value-added composites. In this study, the impact of the use of chaste wood on the physical and mechanical properties of WPC were investigated.

2 MATERIALS AND METHODS

2.1 Materials

The stems of chaste trees with a diameter of 5-10 cm were obtained from Mugla city, Western Turkey (Figure 1). The stems were cut to the small chips by a laboratory chipper with three knives. The wood chips were first air-dried in normal atmospheric temperature, and then dried to the 2-3 % moisture content in a dryer. The chips were ground into wood particles using a laboratory type grinder. The wood particles were screened for 10 min and the wood flour passing through the screen openings of 0.237 mm was used in the experiments. The wood flour was dried to less than 1 % moisture content at 95 °C for 3 h. The moisture content of the wood has an impact influence on the physical and mechanical properties of the WPC because the moisture makes irregularities in the pellets such as microvoids during the extrusion process. This problem can also be observed in the injection molded WPCs.

The virgin polypropylene granules were obtained from a local seller in Turkey. The melt flow index and density of polypropylene were 12 g/10 min (2.16 kg/230 °C) and 0.90 g/cm³, respectively. Maleic anhydride modified homopolymer polypropylene (MAPP; Code: optim P-425, melt flow index with 110 g/10 min (2.16 kg/190 °C, and density: 0.91 g/cm³)) granules, obtained from Pluss Polymers Pvt. Ltd. in Gurgaon city, India, were used as a compatibilizer to improve interfacial bond between polypropylene and wood.

Table 2 Description of resulting WPC codes

Table 2. Opis oznaka dobivenih WPC uzoraka

| WPC specimen code | Wood flour content (wt%) | Polypropylene content (wt%) | MAPP content (compatibilizer) (wt%) |
|-------------------|--------------------------|-----------------------------|-------------------------------------|
| A                 | 0                        | 100                         | 0                                   |
| B                 | 10                       | 87                          | 3                                   |
| C                 | 20                       | 77                          | 3                                   |
| D                 | 30                       | 67                          | 3                                   |
| E                 | 40                       | 57                          | 3                                   |
| F                 | 50                       | 47                          | 3                                   |

2.2 Production WPC panels

The raw materials were weighed based on each formulation given in Table 2 and then pre-mixed. The mixture was put into volumetric feeder of the twin-screw extruder (co-rotating). The extruder barrel temperature was gradually increased from 170 °C (feeding
zone) to the die zone (190 °C) at a constant screw speed (40 rpm). The compound filaments were put in water bath for cooling and then granulated using the granulation process. Before the injection molding process, the moisture content of the granules was decreased to about 1 % in an oven with fan.

First, the granules were placed in the metal frame and then transported to the hot-press (Figure 1a). Wax paper was used between the mat and the meal caul so that the mat did not stick to the metal cauls. The hot-press platens contacted the surface of the mat for melting the pellets at 190 °C for 10 min and then the compression was applied to the mat at 2 MPa and 190 °C for 5 min (Fig. 2b). The resulting WPC panels with dimensions of 200 mm × 200 mm × 4 mm were taken out from the hot press and then metal weights were immediately put on WPC panels for cooling.

3 RESULTS AND DISCUSSION

3.1 Physical properties of WPCs

The density, thickness swelling, and water absorption of the WPC specimens are given in Table 3. The unfilled polypropylene showed negligible thickness swelling (0.08 %) and water absorption (0.05 %). The water resistance of the WPC specimens considerably decreased when the filler content increased in the polypropylene. Especially, when the filler content increased from 40 to 50 wt%, the water absorption of the WPCs sharply decreased, from 2.01 to 7.09 %. Similarly, when the filler content increased from 20 to 30 wt%, the thickness swelling increased from 0.90 to 3.74 %. Due to the hydrophilic property of wood, the water absorption and thickness swelling of the WPCs are negatively affected by the increased amount of the chaste wood flour. The WPCs having a higher amount of wood flour absorbed more water due to the increasing number of microcavities in the WPC as shown in the SEM image (Figure 3) and high amount of free hydroxyl groups. As known, holocelluloses in wood, cellulose, and hemicelluloses, contain free-hydroxyl groups reacting with the water molecules. The holocellulose content of wood flour (74.64 %) is significantly higher than that of hardwoods and softwoods (65-70 %) (Pettersen, 1984).

Polypropylene has a hydrophobic character, and its water absorption is quite negligible because it does not contain any functional polar group. There are several reasons for the lower water resistance of the WPCs such as the decrease in the amount of the polymer matrix as binder, the anatomy and chemical structure of wood, filler content, microcavities, microcracks, and poor interfacial bond in the WPC (Gardner et al., 2015; Ayrilmis and Ashori, 2015; Özdemir et al., 2017). Furthermore, the increase in the water absorption of the WPCs may be explained by the tortuous path formed. The tortuous diffusion paths enable the penetrating of...
water into the WPC. As a result, the increase in the filler content may result in the increased tortuosity in the WPC (Ayrilmis et al., 2013b). Typical SEM image of the tensile fracture surface of the WPC containing 50 wt% wood flour is presented in Figure 3. As shown in the SEM image, the number of cavities that take the water can be observed in the specimens with higher content of wood flour.

### 3.2 Mechanical properties of WPCs

The results of mechanical tests of the WPCs are summarized in Table 4. The MOR and MOE of the unfilled polypropylene specimens were determined as 30.2 MPa and 664 MPa, respectively. When the filler content increased from 10 to 30 wt%, the MOR increased from 33.9 to 44.8 MPa, and then decreased to 33 MPa. The lowest MOR (25.7 MPa) was found in the specimens with 50 wt% wood flour. The MOE of the WPCs was positively affected by the increased filler content. When the wood flour content increased from 10 to 50 wt%, the MOE increased from 815 to 3250 MPa. The increment in the MOE was not high when the addition of wood flour was increased from 40 to 50 wt%.

The tensile strength and modulus of unfilled polypropylene was determined as 19.6 MPa and 1505 MPa, respectively. The tensile strength of the WPCs was considerably lower than that of the unfilled polypropylene. The tensile modulus of the WPCs showed an increasing trend (11.7 MPa to 14.1 MPa) as the amount of the filler increased from 10 to 40 wt%. Nevertheless, a further increase in the filler content reduced the tensile strength (11.4 MPa). When compared to the unfilled polypropylene, the lower tensile strength of the WPCs with high filler content may primarily be explained by the decrease in the polypropylene content, which acted as a binder for the filler in the composite. Moreover, higher loading levels of the wood filler, such as 50 wt%, caused a high degree agglomeration of wood particles in the polypropylene, which formed the zones of stress concentration and negatively affected the tensile strength. The tensile strength results showed that interfacial bonding was adversely influenced by the increased wood content when the wood content was beyond the optimum value of 40 wt%. This ratio was found to be 30 wt% wood flour for the bending strength (Table 4). As shown in the SEM image in Figure 3, most microvoids were observed in the specimens with the highest filler content (50 wt%), which decreased the tensile strength and bending strength. The pulled-out wood fibers and the resulting microcavities can be seen in the SEM image marked with yellow circles (Figure 3). Although the coupling agent (MAPP) was used to improve interfacial bond (ester bond) between the wood and polypropylene, the reason for fiber pullout at high filler content (50 wt%) may be explained by the agglomeration of wood particles and increasing microcavities.

| Table 3 Physical properties of WPCs |
|-------------------------------------|
| Tablica 3. Fizička svojstva WPC uzoraka |
| WPC specimen code, % |
| Oznaka WPC-a, % |
| Density, g/cm³ |
| Vrsta g/cu cm³ |
| Water absorption, % |
| Apсорpcija vode, % |
| Thickness swelling, % |
| Debljinsko bubrenje, % |

| WPC specimen code | Density, g/cm³ | Water absorption, % | Thickness swelling, % |
|-------------------|----------------|---------------------|-----------------------|
| A                 | 0.91 (0.02)*   | 0.05 (0.01)         | 0.08 (0.01)           |
| B                 | 0.92 (0.03)    | 0.25 (0.09)         | 0.58 (0.12)           |
| C                 | 0.84 (0.03)    | 1.08 (0.034)        | 0.90 (0.66)           |
| D                 | 0.92 (0.02)    | 1.20 (0.22)         | 3.74 (0.95)           |
| E                 | 0.98 (0.07)    | 2.01 (0.24)         | 4.78 (1.09)           |
| F                 | 1.02 (0.03)    | 7.09 (0.21)         | 5.68 (2.11)           |

*Standard deviation / standardna devijacija

| Table 4 Mechanical properties of WPCs |
|--------------------------------------|
| Tablica 4. Mehanička svojstva WPC uzoraka |
| WPC specimen code |
| Oznaka WPC-a |
| Bending strength, MPa |
| Čvrstoća na savijanje, MPa |
| Bending modulus, MPa |
| Modul savijanja, MPa |
| Tensile strength, MPa |
| Vlačna čvrstoća, MPa |
| Tensile modulus, MPa |
| Modul elastičnosti pri vlačnom naprezanju, MPa |
| Notched izod impact strength, kJ/m² |
| Otpornost na udarce, kJ/m² |

| WPC specimen code | Bending strength, MPa | Bending modulus, MPa | Tensile strength, MPa | Tensile modulus, MPa | Notched izod impact strength, kJ/m² |
|-------------------|-----------------------|----------------------|----------------------|----------------------|-----------------------------------|
| A                 | 30.2 (1.6)*           | 664 (58)             | 19.6 (1.15)          | 1505 (187)           | 5.88 (0.45)                      |
| B                 | 33.9 (4.1)            | 815 (89)             | 11.7 (1.64)          | 1690 (296)           | 4.93 (1.0)                       |
| C                 | 35.0 (2.9)            | 1197 (105)           | 10.4 (2.16)          | 1838 (267)           | 4.83 (0.33)                      |
| D                 | 44.8 (3.1)            | 1538 (112)           | 12.2 (1.25)          | 2034 (223)           | 4.44 (0.64)                      |
| E                 | 33.0 (3.8)            | 3041 (132)           | 14.1 (1.20)          | 2241 (131)           | 3.92 (0.42)                      |
| F                 | 25.7 (1.2)            | 3250 (119)           | 11.4 (0.92)          | 2253 (233)           | 3.52 (5.85)                      |

*Standard deviation / standardna devijacija
4 CONCLUSIONS

The physical and mechanical properties of the WPCs were influenced by the loading level of the chaste wood flour. Although the water resistance of the specimens was negatively affected by the increased filler content, particularly above 20 wt%, the tensile and bending modulus values improved. When compared to the unfilled polypropylene, the bending modulus greatly improved with the increased filler content. Although the tensile strength of the unfilled polypropylene was higher than that of the WPCs, an increasing trend was observed in the tensile strength values up to 40 wt% filler content; however, further increase in the filler content negatively affected the tensile strength. The evaluation of the wood of underutilized invasive chaste tree in the WPC production may result in an economically effective way to use this resource. The low specific flexural modulus of polypropylene limits its use in semi-building applications. The incorporation of chaste wood into the polypropylene could make of it a sustainable natural filler for semi-building applications such as decking, fencing, siding. According to the results, it was concluded that the 30-40 wt% incorporation of the chaste wood flour can be used in the production of the WPC.

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Corresponding address:

Prof. Dr. NADIR AYRILMIS
Istanbul University-Cerrahpasa, Faculty of Forestry, Department of Wood Mechanics and Technology, 34473 Bahcekoy, Sariyer, Istanbul, TURKEY, e-mail: nadiray@istanbul.edu.tr