Effect of the Filler Content on Some Physical and Mechanical Properties of Virgin- and Recycled Thermoplastic Polyurethane Composites

Vedat Çavuş,[a],* and Fatih Mengeloğlu [b]

Effects of thermoplastic polyurethane (TPU) types (Recycled (R) and Virgin (V)) composites with 15 wt% and 30 wt% oakwood flour addition were studied. Selected physical, mechanical, morphological, and thermal properties of resulting polymer composites were analyzed. Test samples were manufactured using injection molding, except that abrasion resistance specimens were manufactured using a compression molding process. The findings indicated that the types of TPU and filler contents played a significant role in the density and mechanical properties of the TPU test samples. The increased oak wood flour contents in both TPU types showed improvement in density, tensile modulus, hardness, flexural strength flexural modulus, dynamic impact strength, and yield strength of the composite while decreasing the elongation at break values. In addition, both TPU types and filler contents significantly affected the densities of V-TPU and R-TPU. The TPUs type, filler content, and cycle-rpm affected Taber's abrasion resistance values. Weight loss, which increased with the number of cycles for the control samples, decreased with increasing wood flour content. This study aimed to provide an overview of the effect of the wood flour content in the manufacturing of thermoplastic-reinforced composites and to provide a basis for further research and development efforts.

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INTRODUCTION

Wood-plastic composites (WPCs), which are also known as wood fiber plastic composites or green composites, broadly refer to thermoset or thermoplastic-based composites that contain wood flour or lignocellulosic fiber as reinforcing components and fillers (Clemons and Ibach 2003; Kurt ve Mengeloğlu 2011). The WPCs are used in building and outdoor applications, such as decking, litter bins, flower pots, banisters, balconies, sports equipment, door, and window frames, as a substitute for impregnated wood materials, and in construction. Other applications of WPCs include automotive interior panels, parquet flooring, and moldings. In addition to wood flour and fibers, natural fibers, such as agricultural plant wastes, jute, kenaf, and hemp, are also used in WPCs manufacturing. Studies on the production of thermoplastic composites, tree species, annual plants, or lignocellulose-based residual materials that are abundant in the available areas are preferred (Klyosov 2007; Hietala 2013). The WPCs have many advantages over polymers because they utilize wood as raw material/fillers. The WPCs are said to have better dimensional stability and greater biodegradability than wood-based materials. Utilizing by-products like sawdust in WPCs production results in a much lower cost than inorganic fillers. It also causes less wear and tear on processing equipment and machinery. Compared to unfilled polymers, WPCs have higher mechanical properties, thermal stability, and excellent resistance to UV degradation (Çetin *et al*. 2014). The WPCs are manufactured by processing techniques, such as extrusion, compression, and injection molding, dispersing wood flour with compatibilizers and additives in molten plastic (Kim *et al*. 2006; Narlıoğlu 2021).

Virgin and recycled thermoplastic polymers, such as polyethylene, polypropylene, polyvinyl chloride, and polystyrene, are used in wood-plastic composites. Thermoplastic polymers are preferred in manufacturing because they have low processing temperatures, and the lignocellulosic material does not degrade during production (Kim *et al*. 2006; Momanyi *et al*. 2019). Using recycled polymers instead of virgin polymers for industrial purposes is one of the most promising techniques to reduce the environmental impact and costs associated with scrapping components. WPCs can be one of the pathways to sustainable consumption and production using plastic waste as a resource (Gulitah and Liew 2018). Recycled plastic (R-PP) could be a promising feedstock for WPCs production and appropriate method for plastic waste disposal. The source of the recycled polymer influences the final properties of the composites. Postindustrial (PI) recycled polymer provides more controllable and stable properties than post-consumer (PC) recycled polymer because it comes from unknown sources (Veelaert *et al*. 2020).

Thermoplastic polyurethanes (TPUs) are one of the known types of polyurethane. They behave like cross-linked elastomers at room temperature but are different from general elastomers because of their advantages of manufacturability, recyclability, and ease of processing using conventional methods in the industry. The TPUs are linear block copolymers characterized by hard and soft segments. The hard segments consist of diisocyanate and a chain extender (low molecular weight linear glycol). The soft segments consist of long, flexible polyether or polyester chains linking two hard segments. Depending on the contents of hard segments (HS), the TPUs can act as reinforcing fillers, while the soft segments mainly influence the elastic properties of TPUs (Russo *et al*. 2013). The TPUs based on polyester are usually characterized by low hydrolytic resistance because of the presence of ester group. TPUs based on polyester are usually characterized by low hydrolytic resistance because of the presence of the ester group. Among the general application areas, polyurethane products, such as biomedical products (Alves *et al*. 2009), shoes, films, filtration, sports equipment, furniture, hoses, construction materials (Alves *et al*. 2014; Choi *et al*. 2017), insulation, wires, and cables (Datta and Kasprzyk 2018), have made TPUs the fifth-biggest polymer market worldwide (Petrossian *et al*. 2019; Mengeloğlu and Çavuş 2020; Wölfel *et al*. 2020). TPUs have high elongation and tensile strengths, they can withstand oil, grease, solvents, chemicals, and aging with varying degrees of wear and tear (Choi *et al*. 2017; Naderizadeh *et al*. 2018). These features increase the use of TPUs in various fields daily. Waste occurs both during production and after the use of TPUs. These wastes are harmful to the environment. To reduce this waste it can be processed on conventional thermoplastic production equipment using extruders, injection, and pressing methods. The TPUs contain an expensive polymer, which makes it necessary to recover both the residues that may occur during production and the wastes that will arise after consumer use (Ducousso and Bordacs 2003). The preparation of WPCs can be one of the ways for sustainable consumption and production to use TPUs waste as a resource.

According to CEN EN 15534-1 (2014) requirements, lignocellulosic fillers and polymer matrices can come from different sources, such as wood waste, unused natural resources, and clean or recycled thermoplastics. Oak, which includes more than 200 forest species, many subspecies, varieties, and natural hybrids in temperate regions of the Northern Hemisphere, has an important and unique place among the leafy species in the forestry world. It is an essential deciduous species in Europe, both economically and ecologically. Pedunculate oak has a wide and natural distribution in Europe, the Caucasus, and Anatolia (Ducousso and Bordacs 2003; Bektaş *et al*. 2016). Oakwood has a high industrial value, to be utilized in many areas, such as engineered wood products, interior decoration, furniture, parquet, and the wood veneer industry for centuries. During the cutting of round wood in the prescribed dimensions for the purpose of producing lumber, it is expected that residues will be produced. These can be generated during veneer operations, pulp processing, manufacturing, and processing lumber into furniture components, debarking of logs and the edgings. Sawdust, shavings, slabs and trimmings derived from sawing logs can also be generated. The manner in which these residues arise depends on factors such as the part thickness, number of cuts, and the thickness of the cutter (Grebner *et al*. 2021). A few studies observed the utilization of sawdust, wood dust, and post-production particles generated during the use of wood material for different purposes in TPUs composite production. Using these wastes in the production of OPK instead of using them as fuel reduces production costs and eliminates the problem of the accumulation and disposal of forest industry wastes. These residues can be used as fillers in high-value-added materials such as wood-plastic composites. In this study, WPCs materials were manufactured by adding oak wood flour to TPUs to utilize the waste generated during the processing of wood material as filler material in thermoplastic polyurethane (El-Shekeil *et al*. 2011). Research on using wood material as a filler in thermoplastic polyurethane is scarce in the literature. Some of these researches focused on the mechanical and morphological properties of hot and cold-pressed wood TPUs composites (El-Shekeil *et al*. 2011; Grebner *et al*. 2021). Water absorption properties of wood TPUs composites produced by the compression molding process were investigated (Alves *et al*. 2014), water absorption, melt flow, mechanical, morphological, and thermo-mechanical properties of wood TPUs composites produced by the injection-molding process method, the effect of filler type and ratio on some mechanical properties of test samples produced by the extruder and injection-molding processes (Mengeloğlu and Çavuş 2020). These researchers converted different types of cocoa, pod husk, kenaf bast fiber, palm fiber, pine wood flour, teak, rice husks, microcrystalline cellulose, *etc*., into virgin TPUs. They produced TPUs composites by using lignocellulosic materials such as fillers (El-Shekeil *et al*. 2011, 2012; Grebner *et al*. 2021). Because TPU is an expensive and costly material, its range of use is sometimes limited. Therefore, the use of wood flour and annual plants as fillers in TPU, which are cheaper than TPU, can be an effective way to produce a biodegradable and environmentally friendly material and to use production residues and post-use wastes (El-Shekeil *et al*. 2012; Datta and Kopczyńska 2015). The purpose of the present study was to manufacture extrusion and injection-molded recycled and virgin TPU-based composites and to determine the effect of TPUs types and filler content (15% to 30%) on these composites' mechanical properties and density.

EXPERIMENTAL

Materials

The required TPUs to be used as matrix materials were provided by Ravathane Petrochemical Co. (Turkey). This matrix material has strong resilience and tear resistance, excellent abrasion resistance, and good stability towards solvents and light. According to the manufacturer's information, the determined properties of these TPUs were hardness of 85 Shore A, elongation at break of 720%, tensile strength of 47 MPa, density of 1.19 g/cm^3, tear strength of 130 N/mm^2, and abrasion resistance of 40 mm^3. Oakwood is the most widely used and essential source for the woodworking industry. The pedunculate oak (*Quercus robur* L.) was used as a lignocellulosic filler to manufacture the TPU composites. The pedunculate oak was supplied by a woodworking workshop in Izmir. The recycled TPUs were obtained from a factory in Izmir, Turkey. The waste TPU was adequately separated from foreign objects according to the product type and separated into colors among its type.

Methods

The supplied TPUs were first checked for the same hardness as virgin TPUs. The specimens having a hardness of 85 Shore A were washed and shredded in the crushing machine. The broken plastic parts were granulated in an agglomeration machine. The hole diameter of the breaker was 8 mm. Before compounding, the V-TPUs and the R-TPUs materials were subjected to a standard drying procedure for 2 h at 80 °C in a dehumidifier before each processing step.

The pedunculate oak was milled in the Wiley Mill and turned into flour shapes. These flours were classified from 20, 40, 60, 80, 100 to 200 mesh. The wood flour passed through a 40-mesh (400 μm) screen and stayed on a 60-mesh (250 μm) screen, selected for manufacturing composites. After this stage, wood flour was dried in an oven at 103 °C (± 2 °C) for 24 h to reduce the moisture content below 1% prior to WPC manufacturing. Depending on the formulation, the pedunculate oak wood flour and recycled (R-TPUs) or virgin (V-TPUs) thermoplastic polyurethane was mixed in a high-intensity mixer at 900 to 1000 rpm for 2 min. Supplying a homogeneous blend depends on the manufacturing prescriptions (Table 1). Later, the homogeneous blend was compounded in a single-screw extruder at 40 rpm. The temperatures of the extruder were in the range of 175, 180, 185, and 190 °C from barrel to die). The extruded compounds cooled in a water pool and were milled with a Wiley mill for pelletizing. Before injection molding, the pellets were dried in an oven at 103 ± 2 °C for 24 h to a moisture content of less than 1% moisture content. Test samples are manufactured using the injection molding machine. The manufacturing parameters for the injection molding machine were injection pressure of 5 to 6 MPa and temperatures of 180 °C (feed zone) to 200 °C (die zone). Test samples for the Taber abrasion test were prepared in a hydraulic press in a steel mold with a compression molding method. The parameters of the press were a press temperature of 175 °C, press pressure of 1.7 MPa, pressing time of 5 min for heating, 10 min for pressing, and 20 min for cooling. After being conditioned in a climate cabinet, test samples were cut to 4 mm × 100 mm × 100 mm dimensions. Densities were determined by water displacement (ASTM D792 2007) technique. The flexural (flexural strength, flexural modulus, yield strength (ASTM D790 2003)), tensile (tensile strengths, tensile modulus and elongation at break (ASTM D638 2001)), hardness (ASTM D2240 2010), and impact (ASTM D256 2000) properties of all samples were determined following the respective ASTM standards. Five test samples were performed with a crosshead speed of 5 mm/min based on ASTM D790. Flexural and tensile testings were achieved on a Zwick 10 kN device, while a HIT5,5P by Zwick™ impact tester was used for impact property testing on notched samples. The notches were added with a Polytest notching cutter (RayRan™). The dimensions of all test samples (thickness × width × length) for tensile strength (dogbone shape) were 4 × 19 × 165 mm, flexural properties were 4 × 13 × 165 mm, hardness was 4 × 13 × 50 mm, and the Izod pendulum impact strength (notched) was 4 × 13 × 64 mm. Hardness was measured according to ASTM D2240 (2010) using a Shoremeter-D hardness meter. Hardness was measured from the same surface of each sample and from five different points at least 6 mm apart. Taber abrasive tests were determined according to (ASTM D4060 2010) standards. The Taber Abrasimeter was used to evaluate the wear resistance of WPCs made from materials prepared with both V-TPUs and R-TPUs. For comparison, V-TPUs and R-TPUs samples without oak flour were tested. Three replicated samples were produced for each test. Taber abrasion resistances were measured by the Taber abraser device at 1000, 2000, and 3000 cycles under 750 ± 1 g weight and 60 rpm. Abrasive was changed every 1000 rpm, and wear index (I) was calculated after 1000 rpm using Eq. 1,

$$\boldsymbol{I} = \frac{(\boldsymbol{A}-\boldsymbol{B})\mathbf{1000}}{\boldsymbol{C}} \tag{1}$$

where I denotes wear index, A is before-abrasion weight of test samples (mg), B is after-abrasion weight of test samples (mg), and C is number of cycles of abrasions recorded.

Table 1. Manufacturing Prescriptions

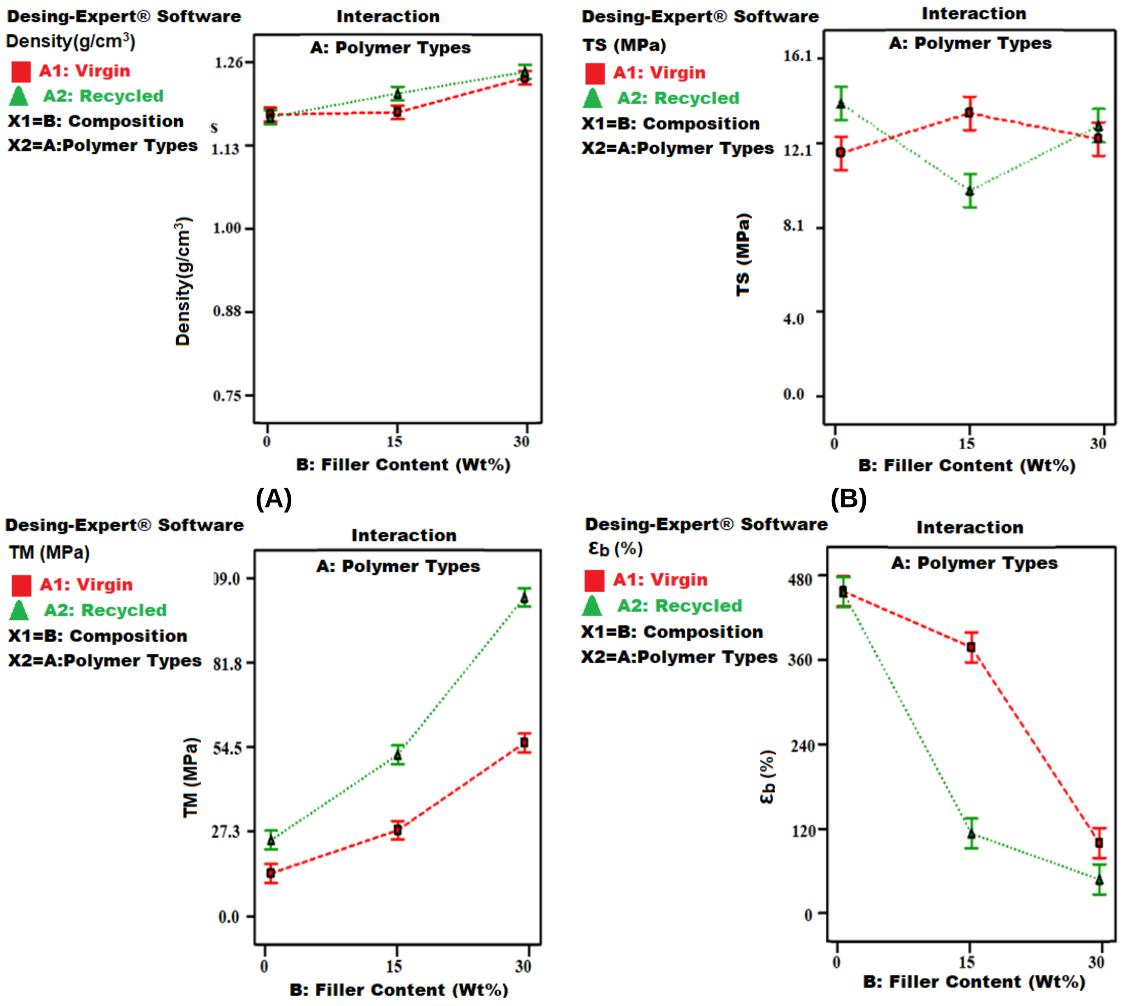
No.	ID	TPUs Type	WFT	PC (wt%)	WFR (wt%)
1	V-TPUs	Virgin	Unfilled	100	0
2	R-TPUs	Recycled	Unfilled	100	0
3	15%V-TPUs	Virgin	P-oak	85	15
4	30%V-TPUs	Virgin	P-oak	70	30
5	15%R-TPUs	Recycled	P-oak	85	15
6	30%R-TPUs	Recycled	P-oak	70	30

PC: Polymer content, WFT: Wood filler type, WFR: Wood flour content

The test samples were conditioned (ASTM D618-13 2013) in a climate cabinet at 23 ± 2 °C temperature and 65 ± 2% relative humidity. The morphological characteristics of the samples were determined by scanning electron microscopy (SEM) (EVO LS10, Carl Zeiss AG, Jena, Germany). Prior to analysis, the samples were immersed in liquid nitrogen for 5 min. They were then broken in half with a hammer to obtain a clear fracture surface. The samples were placed on a sample holder and sputtered with gold (Cressington Sputter Coater 108Auto, Cressington Scientific Instruments, London, England) to avoid charging the electrons absorbed by the samples using 10 mA for 120 s. For the thermogravimetric analysis (TGA), the test samples were ground to a size of 1.0 mm with the help of a grinder of the brand IKA. The TGA analyses of the ground samples were performed in a TGA-50 instrument of the Shimadzu brand. For the TGA analysis, the nitrogen gas flow rate was set at 20 mL/min and the temperature was set at 600 °C from room temperature. During the analyses, the temperature increase was set at 10 °C per minute and 5 to 10 mg of sample was used during the analyses. Nitrogen gas was passed through the system to remove ambient air prior to starting the heating process. Differential scanning calorimetry (DSC) analyses of the test samples were carried out on a Shimadzu DSC-60. The DSC analysis was performed using the remaining TGA samples. During the DSC analysis, the nitrogen gas flow rate was 30 mL/min and 5 to 10 mg of sample was used, with the temperature adjusted from room temperature to 500 °C at a rate of 10 °C per min. For statistical analysis, Design-Expert® version 7.0.3 statistical software (Minneapolis, MN, USA) was used. The composition of the test samples manufactured is presented in Table 1.

RESULTS AND DISCUSSION

The effect of filler content and TPUs type (virgin and recycled) on density and some mechanical properties, such as tensile strength, tensile modulus, elongation at break, flexural strength, flexural modulus, hardness, impact strength, and abrasion resistance, of R-TPUs- and V-TPUs-based wood composites were determined. Figure 1 shows the density (A), tensile strength (B), tensile modulus (C), and elongation at break (D) of the composites.



(A) (B)

(C) (D)

Fig. 1. The graphs of the V-TPUs and R-TPUs for density (A), tensile strength (B), tensile modulus (C), and elongation at break (D)

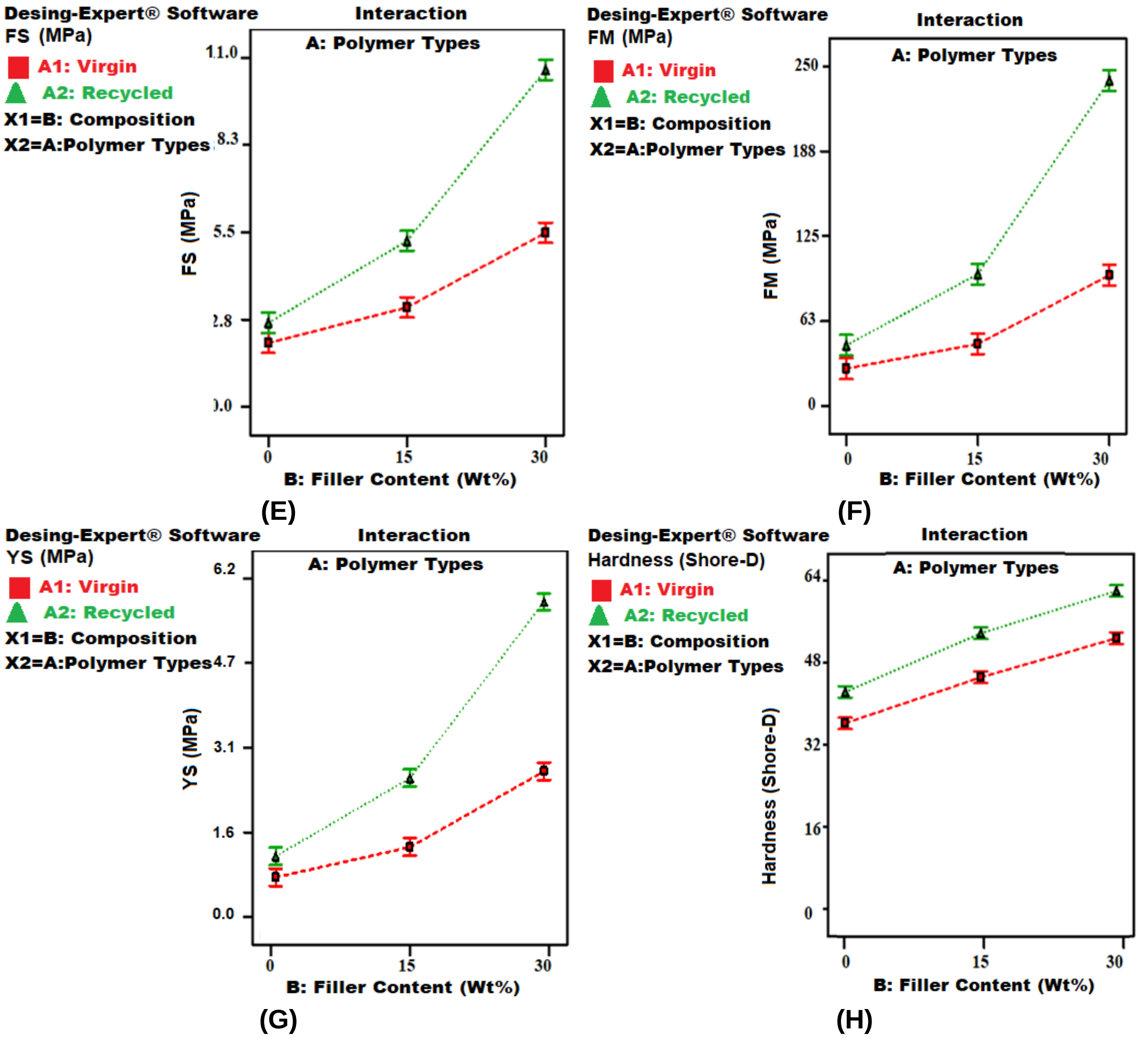
The densities of V-TPUs and R-TPUs test samples were within the range of 1.18 to 1.25 g/cm^3. The lowest density values were determined for unfilled test samples, while the highest values were for 30% filled composites. Statistical analysis showed that TPUs type and filler contents significantly affected the density ($P < 0.0001$). When comparing the unfilled test samples with the filled test samples, there was an increase in their density regardless of TPUs type. The wood cell wall density was usually responsible for the increased density of composites because its density (approximately 1.5 g/cm^3) was higher than the matrix polymer densities. The microstructure of wood is composed of several key components – water, air space, cell wall, and wood substances. These components determine the properties of wood, such as its strength, durability, and density. The specific gravity of wood increases as the proportion of cell wall material per unit volume increases and water and air space decreases. Density increased 5.93% for R-TPUs 30% and 5.08% for V-TPUs 30%. It is stated in the literature that mechanical properties increase as the density increases. Similar results on the density increase of resulting composites with the lignocellulosic material additions were also reported in other studies (Matuana *et al*. 1998; Clemons 2010; Diestel and Krause 2018; Mengeloğlu and Çavuş 2020).

The tensile strengths of test samples involving R-TPUs ranged from 9.80 (unfilled) to 13.94 MPa (15% R-TPUs), while that of test samples involving V-TPUs varied from 11.59 (unfilled) to 13.49 MPa (15% V-TPUs). Statistical analysis showed that the tensile strength was significantly affected by TPUs type and filler contents ($P < 0.0001$). The samples of V-TPUs and R-TPUs showed two different TS_b behavior with the incorporation of the filler in the polymer matrix. In R-TPUs samples, a 15% filler addition reduced the TS_b values but showed some increase with a 30% filler addition. For V-TPUs, a 15% filler addition increased the TS_b values but showed some reduction with a 30% filler addition. The various trends were presented in the literature depending on the selected filler contents and polymer types (Kutty and Nando 1991; Najafi 2013; El-Shekeil *et al*. 2014; Datta and Kopczyńska 2015). It should also be noted that recycled plastic can be reprocessed, exposed to different storage conditions, obtained from various sources, and might provide various performances depending on their degradation level (Najafi 2013).

The tensile modulus of test samples involving the R-TPUs ranged from 24.5 to 102.8 MPa while that of test samples involving V-TPUs were from 13.8 MPa to 51.7 MPa. Statistical analysis showed that both TPUs type and filler contents had a statistically significant effect on the tensile modulus ($P < 0.0001$). Tensile modulus increased 319% for R-TPUs 30% and 275% for V-TPUs 30%. Lignocellulosic filler usually improves the tensile modulus of the TPUs. The increase in tensile modulus with filler content is generally explained by the rules of the mixture. The addition of fibers with high modulus to a matrix with low modulus values resulted in an increase in the modulus of the composite. Similar results have been reported by others (Kutty and Nando 1991; Pilla *et al*. 2009; El-Shekeil *et al*. 2014; Al-Oqla and Sapuan 2014; Afzaluddin *et al.* 2019; Kılınç *et al*. 2019).

The elongation at break (ε_b) values of V-TPUs test samples ranged between 108.7% (unfilled) and 457% (30% V-TPUs), while R-TPUs test samples ranged between 48.0% (R-TPUs) and 456% (Unfilled). The V-TPUs had higher elongation at break values compared to the R-TPUs. Figure 1D shows that lignocellulosic filler addition in the polymeric matrix dramatically reduced the ε_b values. The statistical analysis provided that both TPUs type ($P < 0.0001$) and filler contents ($P < 0.0001$) had a significant effect on elongation at the break. A 30% filler addition reduced the ε_b values of V-TPUs from 455% to 48%. This can be explained by polymer composites becoming harder with increased filler content in the matrix. Cellulosic fillers give polymer hardness. As the polarity increases, the hardness value of the materials increases, and the elongation at break decreases. This is in line with previous studies (Kutty and Nando 1991; Al-Oqla and Sapuan 2014; Tayfun *et al*. 2016; Kılınç *et al*. 2019).

Figure 2 shows the graphs of the V-TPUs and R-TPUs for flexural strength (σ), flexural modulus (E), yield strength (σ_Y), and hardness (HR). The flexural strengths of R-TPUs and V-TPUs test samples ranged from 2.65 (unfilled) to 10.6 MPa (30% R-TPUs) and 2.01 (unfilled) to 5.49 MPa (30% V-TPUs). Statistical analysis showed that the TPUs type and filler content considerably affected the flexural strength properties. Regardless of filler content (P < 0.0001) or TPUs type (P < 0.0001), the flexural strength values were increased with a concentration of wood flour content. The R-TPUs provided the highest flexural strength values compared to V-TPUs. The R-TPUs composites showed better flexural strength values than the V-TPUs composites. This was especially the case as the filler content increased. The flexural strength of the R-TPUs test samples was lower than that of virgin TPUs. The flexural strength of the R-TPUs test samples was higher than that of virgin TPUs. Many researchers have also highlighted that filler content plays an important role in flexural properties (Afzaluddin *et al*. 2019; He *et al*. 2019)



(E) (F)

(G) (H)

Fig. 2. The graphs of the V-TPUs and R-TPUs for flexural strength (σ), flexural modulus (E), yield strength (σ_Y), and hardness (HR).

The flexural modulus of R-TPUs and V-TPUs test samples ranged from 44.6 (unfilled) to 240 MPa (30% R-TPUs) and 27.5 (unfilled) to 96.3 MPa (30% V-TPUs), respectively. According to the statistical analysis, TPUs type ($P < 0.0001$) and filler content ($P < 0.0001$) significantly affected flexural modulus. The increase in flexural modulus was caused by the increase in fiber content, which has a higher stiffness than the matrix. The lignocellulosic materials have a higher modulus value than TPUs (Ramachandran and Vairavan 2007; Biswas *et al*. 2011; AlMaadeed *et al*. 2012). As a natural consequence of this wood flour-filled TPUs, it provides higher modulus values than the polymer itself (Alavudeen *et al*. 2015; Çavuş and Mengeloglu 2017; Mengeloğlu and Çavuş 2020).

The yield strength of R-TPUs and V-TPUs test samples ranged from 1.15 (unfilled) to 6.02 MPa (30% R-TPUs) and 0.75 (unfilled) to 2.77 MPa (30% V-TPUs). According to statistical analysis, both TPUs type ($P < 0.0001$) and filler content ($P < 0.0001$) had a significant effect on yield strength values. As shown in Fig. 2G, compared with the TPUs type, filler content had a more noticeable influence on yield strength. The increase of wood flour content from 15% to 30% in the TPUs affected the yield strength. This result was in agreement with the previous study (Mengeloğlu and Çavuş 2020). The graphs for hardness values are presented in Fig. 2H. The hardness of R-TPUs and V-TPUs test samples ranged from 42.3 (unfilled) to 62.0 (30% R-TPUs) Shore-D to 36.2 (unfilled) to 52.8 (30% V-TPUs) Shore-D. The hardness increased 46% for R-TPUs 30% and 45.43% for V-TPUs 30%. According to the statistical analysis, both TPUs type ($P < 0.0001$) and filler content ($P < 0.0001$) significantly affected hardness properties. Both R-TPUs and V-PTUs hardness values proportionally increased with the upgraded filler contents. It is known that a higher hardness value of wood flour increases the hardness of the TPUs matrix. Similar findings were also stated by others (Jamil *et al*. 2006; Chee *et al*. 2021; Moritzer and Richters 2021).

Statistical analysis was not performed for the dynamic impact strength of R-TPUs and V-TPUs test samples, as the control and 15% oakwood flour-filled notched impact test samples did not break during the test. The unfilled TPUs and 15% filled samples were stiffer than unfilled TPUs and probably had lower IS values based on visual observation. However, when the test hammer was used to break the samples, they were still elastic enough to bend. The dynamic avarage impact strength of V-TPUs and R-TPUs test samples ranged from 19.4 kJ/m^2 (30% V-TPUs) to 28.9 kJ/m^2 (30% R-TPUs). Both TPUs type and filler content affected dynamic impact strength. There were some differences with the TPUs types in the dynamic impact strength values of test samples. The dynamic impact strength of wood plastic composites depends on several factors, such as the nature of the wood and plastic, the interfacial bonding of fillers/fibers/matrix, and the strength and structure of the reinforcing fibers (Kutty and Nando 1991; Jamil *et al*. 2006; Chee *et al*. 2021; Moritzer and Richters 2021). Plastic waste can come from different sources, be manufactured at different stages, contain different chemicals, and contain different additives and fillers. The physical and mechanical properties of these materials can also be affected by the number of times they are recycled.

The Taber abrasion resistance values are shown in Fig. 3. Taber abrasion resistance values for 1000, 2000, and 3000 cycles were 0.153-0.162-0.186 mg (15%R-TPUs), 0.297-0.271-0.257 mg (30%R-TPUs), 0.127-0.117-0.130 mg (15%V-TPUs), and 0.166-0.170-0.160 mg (30%V-TPUs), respectively. The lowest and highest Taber abrasion resistance values were obtained at 0.127–0.17 and 0.130 mg (15% V-TPUs) and 0.297–0.271–0.257 mg (30% R-TPUs) for 1000, 2000, and 3000 rpm, respectively. The TPUs type, filler content, and the number of cycles influenced the Taber's abrasion resistance values. As the hardness increased, the wear resistance also rose. Abrasive wear resistance generally depends on the hardness and improves with increasing hardness. Regardless of the filler type, filler addition significantly increased the wear index values, *i.e.*, but reduced weight loss. As the number of Taber wear cycles increased, the rate of weight loss decreased. Both the R-TPUs and V-TPUs control samples had the highest values. The weight loss rate, which increased with the number of cycles for the control test samples, decreased with the increase in wood flour content. The test results showed that the blend contents of oak flour showed lower wear values, which is a positive factor in the production of TPUs-based composites. Similar results were also reported by others (Moritzer and Richters 2021; Yu *et al*. 2021).

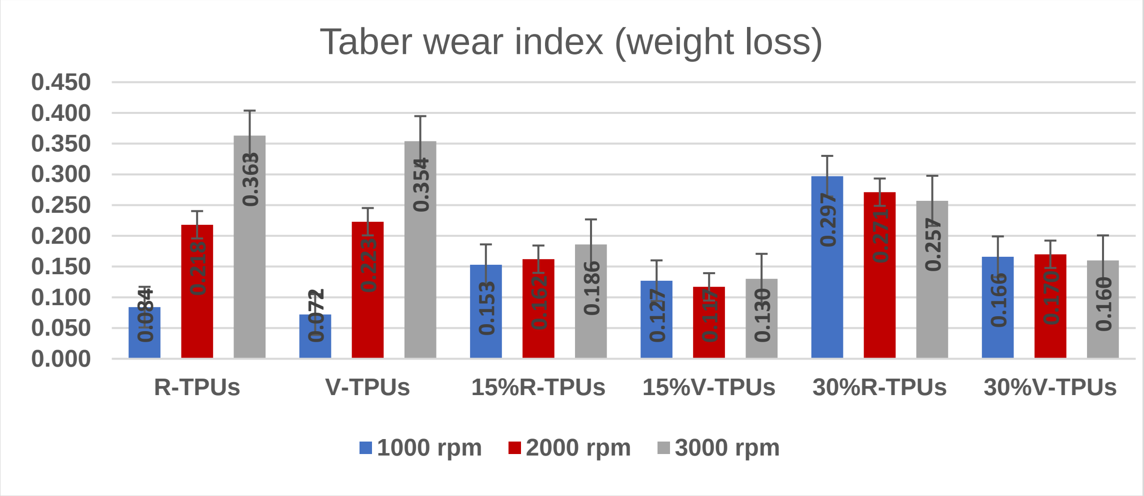


Fig. 3. The Taber abrasion resistance graphs

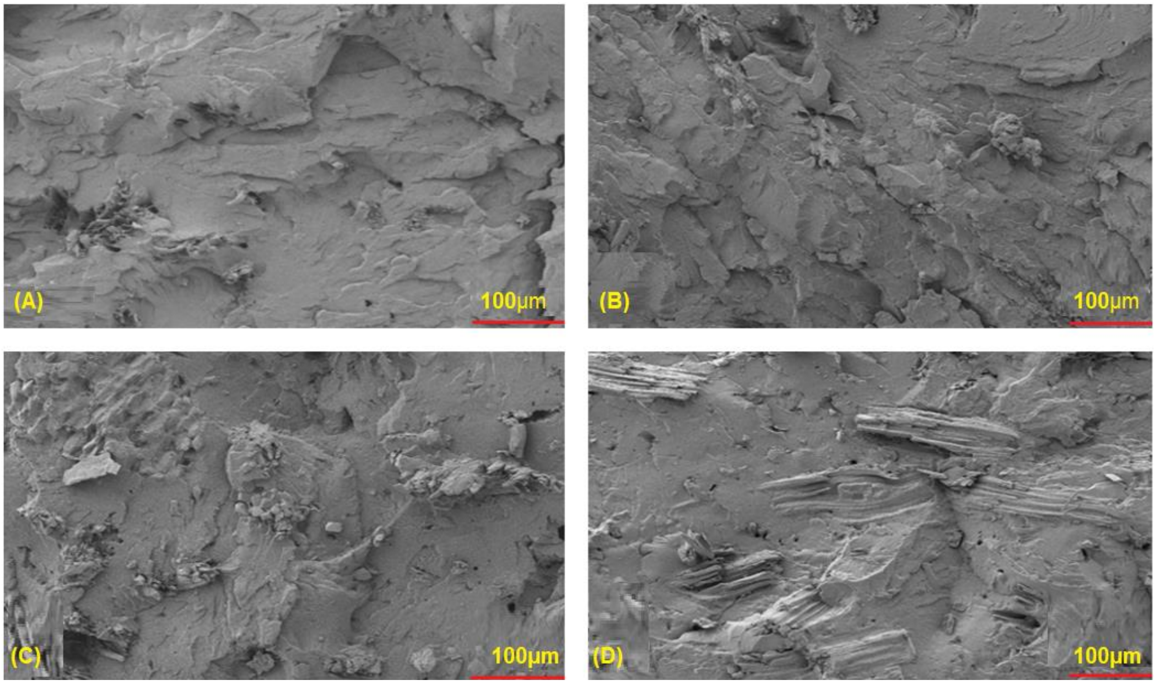


Fig. 4. Morphologies of the test specimens (A)- 15%V-TPUs, (B)- 15%R-TPUs, (C)- 30%V-TPUs, and (D)- 30%R-TPUs

Morphologies of the test specimens are presented in Fig. 4. Figures 4A and C show SEM images of the V-TPUs with oak wood flours, while Fig. 4B and D show SEM images of the R-TPUs samples with oak wood flours of the formulations. The oak flour was embedded in the TPUs and well dispersed in the TPU matrix as shown in Fig. 4A. Figure 4C and D show some microcracks and some pulled fibres in composites with 15% and 30% wood flour filler. When using oak wood flour, TPUs provided better adhesion between the polymer matrix and filler compared to composites made with commercial polymers such as polypropylene (PP) and polyethylene (PE). The hydrophilic nature of TPU played an important role in this result.

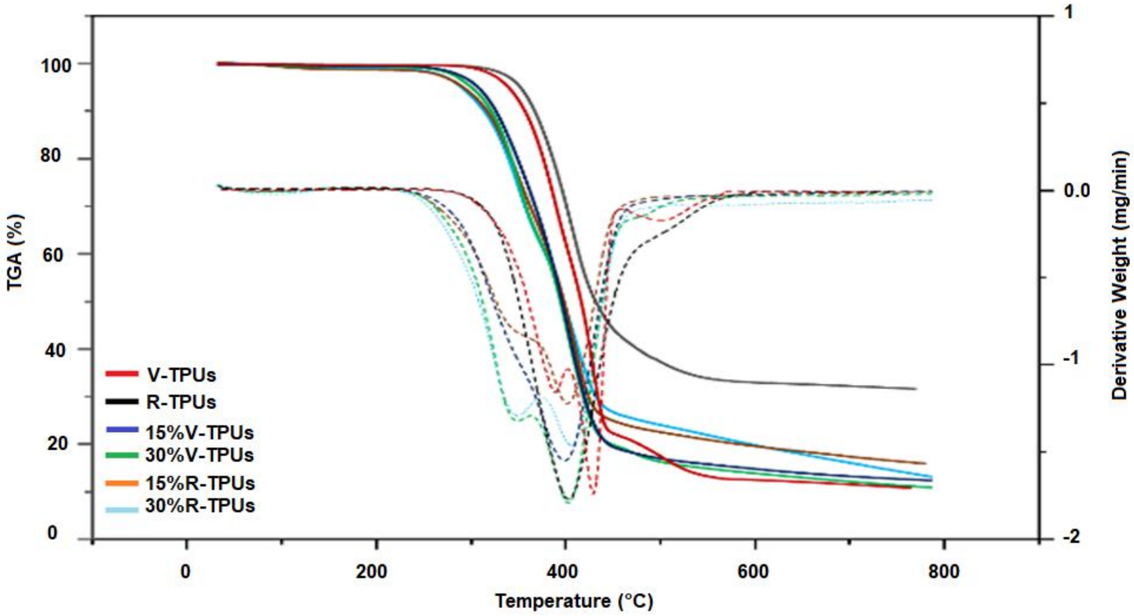


Fig. 5. TGA curves of composites

Figure 5 shows the TGA curves for the composites. Degradation of the composites with added oak flour was shown to occur in 3 regions from the TGA curves. From the TGA curve it was understood that oak flour started to degrade at 229 °C and PP at 340 °C. It was observed that the degradation of the lignocellulosic material was in the first stage and the degradation of the R-TPUs and VTPUs was in the second and third regions. It was observed that the degradation of the polymeric material mainly occurred between 340 to 520 °C. In addition, there was also an increase in the amount of residual matter at 600 ^{o}C when wood was added to the composites. These results are consistent with previous studies (Altuntas *et al.* 2017; Narlıoğlu 2021, 2022) for wood plastic composites made from polymer matrices such as polypropylene, polyethylene, *etc*.

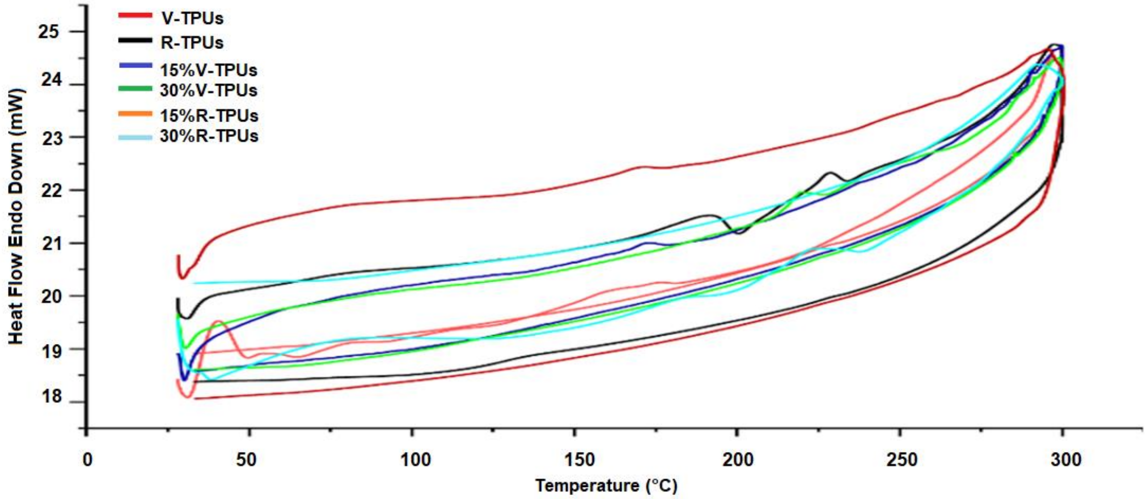


Fig. 6. DSC curves of composites

The DSC curves of the composites are shown in Fig. 6. It is apparent from the endothermic thermograms that the DSC results did not show any significant changes in the melting temperatures of the polypropylene and the composites. The melting temperatures for both V-TPU and R-TPU were around 170 °C. From the DSC curves of the prepared specimens, it was observed that using oak flour had no significant effect on the melting temperature of the composite. Crystallization temperatures increased, but no significant change in melting temperatures was observed because of the DSC experiments.

CONCLUSIONS

1. Wood-TPUs composites were successfully manufactured using V-TPUs and R-TPUs with oak flour at 15% and 30% filler content levels.
2. The results showed that the types of TPUs (V-TPUs and R-TPUs) and filler content levels (15% and 30%) played a significant role in the density and mechanical properties of the TPUs test samples.
3. For both R-TPUs and V-TPUs composites, the filler addition improved density and some mechanical properties such as tensile modulus, hardness, flexural strength, flexural modulus, dynamic impact strength, and yield strength.
4. In both TPUs types, the elongation at break values were negatively affected by the addition of oak wood flour and in parallel with the increase of participation amount.
5. The TPUs type, filler content, and cycle-rpm affected Taber's abrasion resistance values. Weight loss, which increased with the number of cycles for the control samples, decreased with increasing wood flour content. These test results showed that specimens with oak flour exhibited less wear, which is a positive factor in the production of TPUs-based composites.
6. The increase in the physical and mechanical properties of both R-TPUs and V-TPUs composites was remarkable. These results indicate the potential to manufacture desired materials from recycled materials by mixing with wood floor.

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