Preparation of Thermoplastic Polyurethane-based Biocomposites through Injection Molding: Effect of the Filler Type and Content

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

The effects of lignocellulosic filler type and filler loading levels were investigated relative to selected properties of thermoplastic polyurethane (TPU)-based composites. Teak wood (TK), rice husks (RH), and microcrystalline cellulose (MCC) were used as lignocellulosic fillers at 15 wt% and 30 wt% filler loading levels. Test specimens were manufactured using both extrusion and injection molding, except for abrasion resistance samples that were manufactured using a compression molding process. Density, tensile, flexural, and impact properties, and hardness and abrasion resistance values, of the specimens were determined. The composites' morphology was studied using scanning electron microscopy analysis; results showed all filler types and filler loading levels were affected by the TPU's density and mechanical properties. The TPU composites were successfully produced using TK, RH, and MCC as lignocellulosic fillers. Regardless of filler type, addition of 15% filler to TPU yielded excellent mechanical properties. With 30% MCC filler, composite properties increased due to their higher surface area, while properties of TK- and RH-containing specimens were, at 30%, reduced. There was a proportional correlation between hardness and modulus, with both increasing with a rising filler loading level. Abrasion resistance of TPU decreased with the presence of filler. Regardless of filler type, abrasion resistance continued to drop at higher filler loading levels. Scanning electron micrographs showed better MCC distribution in the TPU matrix.

Keywords: Teak; Rice husk; Microcrystalline cellulose; Thermoplastic polyurethanes

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INTRODUCTION

Thermoplastic-based wood-plastic composites (WPCs) are manufactured using lignocellulosic materials such as flour from various wood species, agricultural wastes, and thermoplastic polymer matrices. Neat or recycled polymers can be used to produce WPCs (Mengeloğlu and Karakuş 2008). Wood-plastic composites can be produced from a wide range of composites—including polyethylene, polypropylene, and polyvinyl chloride (PVC)—as the polymeric matrix, and sawdust and wood fibers as the fillers (Behravesh *et al.* 2010). Filler materials can affect the final properties of the composite. Lignocellulosic fillers have advantages, such as low density, low cost, adequate specific properties, reduced abrasiveness, and increased renewability and biodegradability when compared with the traditional synthetic fillers (Staiger and Tucker 2008; Butylina *et al*. 2012; Najafi 2013; Nikolaeva 2015; Hubbe and Grigsby 2020).

Thermoplastic elastomers (TPEs) are hybrid materials, generally made of thermoplastics and elastomers. The properties of TPEs are similar to vulcanized rubber, for instance with respect to softness, flexibility, extensibility, and resilience. They have been used in a myriad of applications including automotive profiles, window gaskets, tubes, seals, electrical wires, *etc*. TPE character can be achieved by blending of thermoplastics and elastomers such that the final morphology is co-continuous, *i.e*., the related phases are intermingled (De and Bhowmick 1990; Boubakri *et al.* 2011; Banerjee *et al.* 2019). Thermoplastic polyurethane (TPU) is one of the most widely used thermoplastic elastomers (TPE) due to its high strength, resistance to tearing, good elasticity, flexibility, and damping properties. It is a block copolymer, and it has soft and hard segments that allow it to tune its elasticity (Bi *et al*. 2018b; Petrossian *et al.* 2019). The soft segment is formed from a linear, long-chain diol, and the hard segment is composed of alternating diisocyanate and chain extender molecules (Hepburn 1992; Finnigan *et al.* 2005; Şen *et al.* 2017). Thermoplastic polyurethane behaves similarly to elastomers due to its high flexibility, low modulus of elasticity, and recoverability. At the same time, TPUs are thermoplastic in nature, melt-processable, recyclable, and suitable to scale up in manufacturing processes such as injection molding (Petrossian *et al.* 2019). They are even blended with other polymers to tailor matrix properties (El-Shekeil *et al.* 2014; Wu *et al.* 2014). Despite their excellent properties, applications using TPU as a polymer matrix in composites are limited due to their high cost and non-biodegradability (Tan *et al.* 2015; Bi *et al*. 2018b; Rostami and Moosavi 2019). Lignocellulosic filler is an abundant biomass material that is low cost, low density, and environmentally friendly. Using lignocellulosic materials in TPU composites may generate interesting composite properties and could open up new application areas for them.

A limited number of studies have been conducted concerning TPU-based composites, investigating the potential of lignocellulosic materials to be used as filling/reinforcing materials. El-Shekeil *et al.* (2011) conducted a study to optimize the processing parameters and fiber size of a short kenaf fiber-filled TPU composite. This composite was prepared by both the melt-mixing method and compression molding. The processing parameters, such as temperature, time, and speed, on tensile properties were studied first. Next, the effects of varying fiber sizes on tensile properties, flexural properties, and impact strength were tested. The optimum blending parameters were 190 °C, 11 min, and 40 rpm for temperature, time, and speed, respectively. Particle size between 125 and 300 μm provided the best tensile strength, flexural strength, and moduli results. Impact strength showed a slight increasing trend with an increase in fiber size. In a subsequent study, the effect of fiber content on the mechanical and thermal properties of kenaf fiber-reinforced thermoplastic polyurethane composites were studied (El-Shekeil *et al*. 2012). In this work, kenaf fiber with length and diameter in the ranges of 0.2 to 2.25 mm in length and 10 to 200 μm in diameter were used, and hot blending and compression molding was applied for composite manufacturing. Based on this study, a 30% fiber loading exhibited the best tensile strength. With an increase in fiber content, flexural strength and flexural and tensile moduli were increased, but strain deteriorated. Increased fiber loading also resulted in a decline in impact strength and abrasion resistance.

Diestel and Krause (2018) conducted a study on wood fiber (Arbocel® C100)-filled TPU composites, investigating the effect of filler loading levels on mechanical properties. They also studied the effect of moisture and the type of TPU (polyester or polyether) on those properties. Composites were produced with hot mixing and compression molding. They reported that properties of the composite were mainly influenced by the proportion

of wood and TPU. Wood flour increased the density, hardness, water absorption, and tensile modulus, and it decreased the impact resistance and abrasion resistance of the composites. In another study, TPU-based composites were produced using sugar palm fibers of particles sized between 150 and 250 μm (Atiqah *et al.* 2018). Composites were prepared by melt-mixing compounding followed by hot-pressing (compression) molding. The physical properties, such as density, water absorption, and thickness swelling, of the composites were evaluated and increasing the fiber content resulted in higher water uptake and thickness swelling. The application of silane treatment to the fibers improved composite's physical properties.

Most studies reported in the literature were produced with the combination of melt mixing and compression molding, and they used kenaf and Arbocel® C100 fiber, *etc*. One study used wood flour as filler in a TPU composite produced with twin-screw extrusion and 3D printing (Bi *et al.* 2018a). One major manufacturing method in the industry is injection molding. There is a need to produce injection-molded TPU composites containing lignocellulosic fillers. The objective of the present study was to manufacture injection-molded thermoplastic polyurethane-based biocomposites, and to investigate the effect of filler types (teak, rice husks, and microcrystalline cellulose) and proportional contents (15 to 30%) on the density and selected mechanical properties of manufactured composites.

EXPERIMENTAL

Materials

Teak (*Tectona grandis* Linn. f.) wood flour (TK), rice husks (RH), and microcrystalline cellulose (MCC) were used as fillers. Teak (density 0.67 g/cm^3) wood particles were supplied by a furniture factory (As Ahşap, İzmir, Turkey). Rice husk (density 0.90 to 0.150 g/cm^3) was provided by a paddy factory (Yaman Farm, Karabük, Turkey). Microcrystalline cellulose (average particle size of 12 to 18 μm, maximum moisture level of 6.0%, and density range of 1.58 to 1.60 g/cm^3) was purchased from JRS Pharma (Patterson, NY, USA). Thermoplastic polyurethanes (Ravathane® 140 A85, with a density 1.19 g/cm^3) was supplied by Ravathane Petrochemical Co. (Izmir, Turkey).

Methods

The TK and RH particles were turned into flour using a Wiley mill (Altundal, Kahramanmaraş, Turkey) and classified. Particles passed through a 40-mesh (400 μm) screen and those remaining on 60-mesh (250 μm) screen were used in this study. The MCC was used as received. All fillers (TK, RH, and MCC) and the TPU were dried in an oven for 24 h at 103 ± 2 °C) and 4 h at 90 ± 2 °C, respectively, until they reached a moisture content of below 1%. Depending on the manufacturing compositions, the filler and TPU were mixed in a high intensity mixer (900 to 1000 rpm in 2 min). The resulting homogeneous blend was compounded in a single-screw extruder at 40 rpm screw speed, at temperatures (from barrel to die) ranging from 175 to 195 °C. The extruded compounds were first cooled in a water pool (23 ± 2 °C) and were granulated into pellets using the Wiley mill. The pellets were dried at 103 ± 2 °C in an oven (24 h) to reduce moisture content to below 1% before the injection molding using a HAIDA HDX-88 (Haida Plastic Machinery Co., Ningbo, China). The injection pressure and temperatures (from feed zone to die zone) of the injection-molding machine were set as 5 to 6 MPa and 180 to 200 °C, respectively. The test specimens for Taber abrasion tests were prepared using the

compression molding method with a hydraulic press with cooling capabilities (Sistem Machinery, Kahramanmaras, Turkey). Pellets were placed in a steel mold (mold dimensions 170 mm × 170 mm × 4 mm). The press temperature and press pressure were set as 175 °C and 1.7 MPa, respectively. Pressing time was a total of 15 min (5 min heating and 10 min pressing). A 20-min cooling was also applied before opening the press. Compression-molded composites were conditioned in the climate cabinet, and test specimens in the dimensions of 100 mm × 100 mm × 4mm were cut.

Before testing, the specimens were conditioned in a climate cabinet at 23 ± 2 °C temperature and 65 ± 2% relative humidity. The density of the test specimens was measured according to ASTM D792 (2007) (water displacement technique). The tensile, flexural, (notched) impact strength (IS), hardness (H), and abrasion resistance (AR) of the specimens were determined based on the procedures of ASTM D638 (2001), ASTM D790 (2003), ASTM D256 (2000), ASTM D2240 (2010), and ASTM D4660 (2010), respectively. Tensile and flexural property tests were implemented using the Zwick 10 KN (Zwick/Roell, Ulm, Germany) instrument, while a HIT5, 5P (Zwick) was used for IS testing on notched specimens. The notch was added using a RayRan™ Polytest notching cutter (Industrial Physics, London, England). At least five samples were tested in each group, except for AR. The Taber AR test was performed on three replicated samples. Taber abrasion resistances were measured by the Taber Abraser (Taber Industies, North Tonawanda, NY, USA) at 1000, 2000, and 3000 cycles under 750 ± 1 g weight and with 60 rpm. Abrasive paper was changed after every 1000 cycles and the wear index (*I*) was calculated after 1000 rpm using Eq. 1,

$$I = [(A - B)(1000)] / C \quad (1)$$

where *I* is the wear index, *A* is the weight of test specimen before abrasion (mg), *B* is the weight of the test specimen after abrasion (mg), and *C* is the number of cycles of abrasion recorded.

For statistical analysis, the statistical software Design-Expert®, version 7.0.3 (Minneapolis, MN, USA) was used. Scanning electron microscopy (SEM) (EVO LS10; Carl Zeiss AG, Jena, Germany) was used to determine the morphological properties of the test specimens. Before analysis, specimens were dipped into liquid nitrogen for 5 min and then broken in half with a hammer to give a clear fractured surface. The specimens were placed on a specimen holder and spattered with gold (Cressington Sputter Coater 108Auto; Cressington Scientific Instruments, London, England) to prevent charge accumulation of the electrons absorbed by the specimens with 10 mA in 120 s. The composition of the manufacturing is given in Table 1.

Table 1. Manufacturing Compositions

No.	ID	WFT	PC (%)	WF (%)
1	TPU-0	Control	100	0
2	TPU-15-TK	Teak	85	15
3	TPU-30-TK	Teak	70	30
4	TPU-15-RH	Rice Husk	85	15
5	TPU-30-RH	Rice Husk	70	30
6	TPU-15- MCC	Microcrystalline Cellulose	85	15
7	TPU-30- MCC	Microcrystalline Cellulose	70	30

WFT: Wood filler types; PR: Polymer content

RESULTS AND DISCUSSION

The effect of filler types and rates on the density and some mechanical properties of TPU-based composites were investigated. For this purpose, density, tensile properties (tensile strength [TS], tensile modulus [TM], and elongation at break [EatB]), flexural properties (yield strength [YS], flexural strength [FS] and flexural modulus [FM]), impact strength (IS), hardness (H), and abrasion resistance (RA) were determined.

The interaction graph of the density is presented in Fig. 1. The densities of test specimens were in the range of 1.18 g/cm^3 to 1.27 g/cm^3. Overall, unfilled TPU specimens provided the lowest mean density values, while TPU-30-MCC had the highest mean density values. Statistical analysis showed that both filler type and filler loading level had a significant effect on density ($P < 0.0001$). Regardless of the filler type, the density of the specimens increased with the filler loading level. It should be noted that there was an interaction between the filler type and filler loading level ($P < 0.0001$). While RH and TK provided similar increases in density, the increase was more pronounced when MCC was used as filler due to their higher density (1.58 to 1.60 g/cm^3).

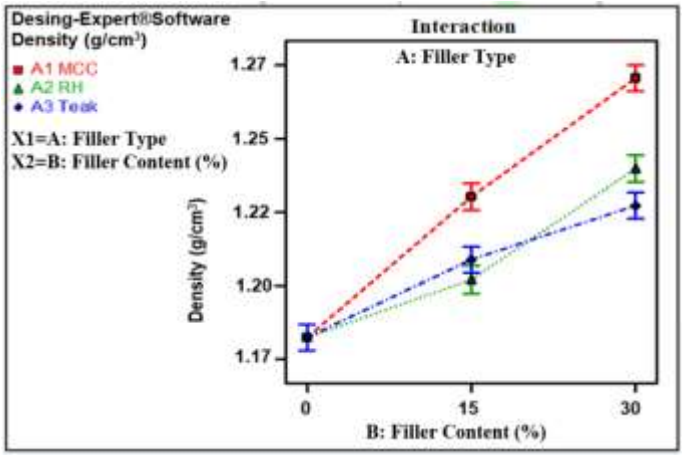

Fig. 1. Interaction graph of density

An increased density of a composite with lignocellulosic filler is usually explained by the "rule of mixtures" in the literature. When filler with higher density is incorporated into a polymer matrix with lower density, the resulting composite usually has a higher density than that of the polymer itself. This increase is believed to be due to the higher cell wall density of lignocellulosic materials (approximately 1.5 g/cm^3) (Matuana *et al.* 1998; Mäkinen *et al.* 2002; Clemons 2010; Diestel and Krause 2018; Çavuş 2020).

The interaction graphs of TS and TM are presented in Figs. 2 and 3, respectively. The TS of TPU specimens ranged from 10.0 to 17.7 MPa. The lowest and the highest TS values were observed in TPU-30-RH- and TPU-30-MCC-coded composites, respectively. Statistical analysis showed that both filler type and filler loading level had a significant effect on TS ($P < 0.0001$). Regardless of filler type, the TS of the specimens increased with a 15% filler loading level. However, there was an interaction between the filler type and filler loading level ($P < 0.0001$). TS values of RH- and TK-filled composites at 30% filler loading level were reduced. It has been reported that the rate of lignocellulosic filler had an effect on the TS values of TPU composites (El-Shekeil *et al.* 2012; Diestel and Krause 2018). El-Shekeil *et al.* (2012) reported that TS values increased up to 30% filler content

in their studies that examined 20, 30, 40, and 50% lignocellulosic filler in TPU composites. Conversely, Diestel and Krause (2018) reported that TS decreased when greater than 30% lignocellulosic filler was used. It should be mentioned that in a second study, a complex experimental design was tested, wherein low (30%) and high (70%) filler loading levels were used and different factor interactions were investigated. It is well known that TPU has a hydrophilic nature and has some compatibility with lignocellulosic filler. It is possible that this helped improve TS values at lower rates, but, at higher filler loading levels, this compatibility was disrupted by filler and resulted in lower TS values. The decreasing TS values were also reported by others in hydrophobic polymers due to the lack of compatibility between lignocellulosic filler and polymer matrix (Zhao *et al.* 2011; Wu *et al*. 2013). For a 30% MCC filler loading level, the TS of the composites still continued to increase, though at a slower rate. The outperformance of MCC filler over others might be due to its smaller particle size. The morphology of the composites with 30% fillers is presented in Fig. 10b. Comparison of the SEM images clearly displayed that TPU-30-MCC composites had less gaps and smoother surfaces, indicating better adhesion with TPU as compared with composites containing other fillers at 30% filling.

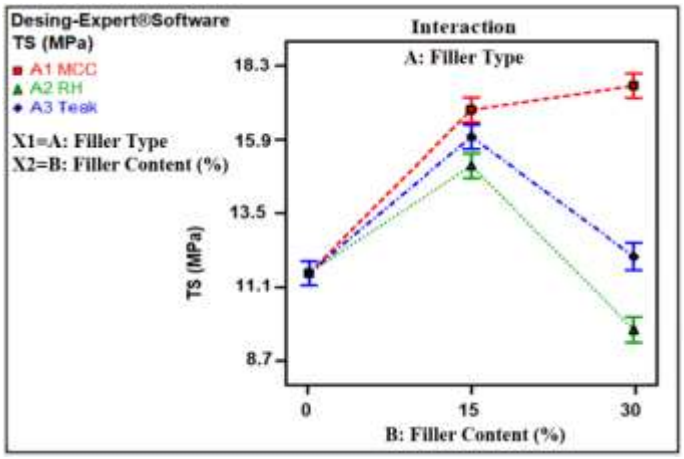

Fig. 2. Interaction graph of tensile strength

The TM values of TPU ranged from 13.8 to 51.5 MPa. Statistical analysis showed that filler loading level had a significant effect on the TM values of the composites ($P < 0.0001$). The tensile modulus increased with a rising percentage of filler. Test results also showed that filler type did not have a significant effect on TM values ($P > 0.050$). The MCC-, RH-, and TK-filled TPU composites provided similar TM values, and they were higher than unfilled TPU specimens. In TPU-based composites, a similar increase corresponding to filler content was also reported by Diestel and Krause (2018). The TM values of composites with a low-stiffness matrix and high-stiffness filler increases with a rising percentage of filler (El-Shekeil *et al.* 2012).

For the elongation at break (EatB) values, the testing machine was capable of measuring elongation up to 500%. Statistical analysis was not conducted because most of the specimens were not broken within this elongation limit. Results showed that along with unfilled TPU specimens, regardless of filler type, specimens with a 15% filler loading level did not break within this limit. It was observed that these specimens were stiffer and probably had lower EatB values than unfilled TPU. For 30% filler content, EatB values of the TK- and RH-filled specimens were measured as 113% and 145%, respectively. These

results were greatly lower than 500%. This reduction was a normal consequence of the increase of filler amount, which had a low strain (El-Shekeil *et al.* 2012). It has also been reported that as filler loading increases, a higher restriction to molecular motion of the macromolecules can be expected. Thus, the addition of more fibers will result in resistance flow and lead to a lower resistance to breakage (Ismail *et al.* 2002).

It should also be noted that MCC specimens did not break within the testing limit, even at 30% filler loading level. Once again, these specimens looked stiffer than unfilled TPU, but they did not break within the testing limits. The average particle size of MCC (~ 10 to 18 µm) is smaller than the particle size of TK and RH (~ 250 to 400 µm) distribution, and has better compatibility between the fiber and matrix (Kokta *et al.* 1989; Geethamma *et al.* 1995). It is reported that short fibers provide higher specific surface areas. This might lead to homogeneous distribution of fibers in polymer matrix providing improved compatibility between them (Geethamma *et al.* 1995; Bledzki *et al.* 1998, Caraschi and Leão 2002).

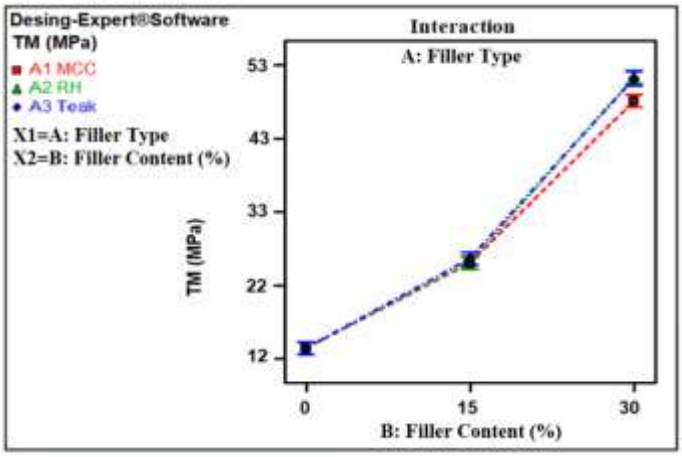

Fig. 3. Interaction graph of TM

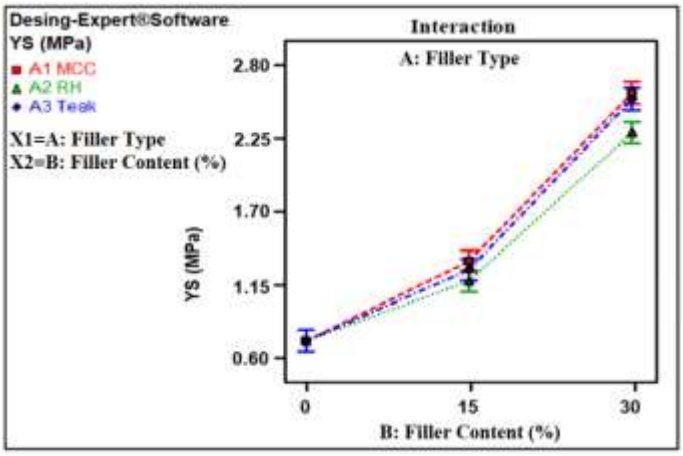

Fig. 4. Interaction graph of YS

The flexural properties of YS, FS, and FM were determined and their interaction graph is presented in Figs. 4 through 6. The YS values of test specimens ranged from 0.75 to 2.60 MPa. The lowest YS values were recorded in TPU control specimens, while the highest was TPU-30-MCC. Statistical analysis showed that both filler type (P = 0.0079)

and filler loading level ($P < 0.0001$) had a significant effect on YS values. As shown in Fig. 4, compared with filler type, filler loading level had a more pronounced effect on YS. The addition of MCC, RH, and TK at filler loading levels of 15% and 30% in the TPU had a positive effect on YS.

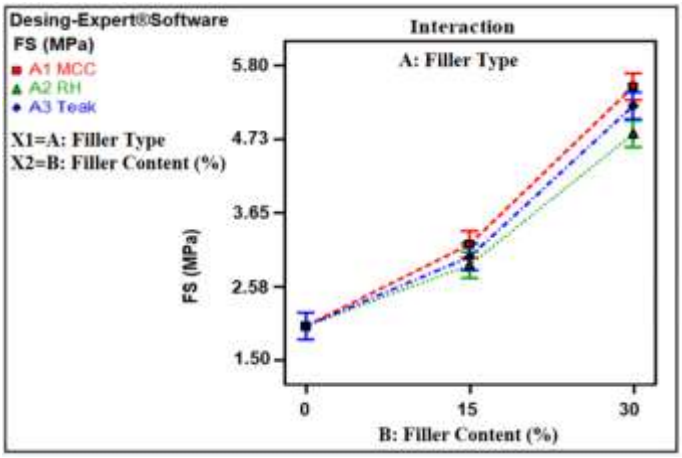

Fig. 5. Interaction graph of flexural strength

The FS values of TPU-based composites ranged from 2.01 to 5.51 MPa. The interaction graph of FS is presented in Fig. 5. The lowest FS values were recorded in unfilled TPU specimens, and the highest values were recorded in TPU-30-MCC specimens. Statistical analysis showed that both filler type ($P = 0.0191$) and filler loading level ($P < 0.0001$) had a significant effect on FS values. As reflected by the YS values, the effect of filler loading level was more pronounced than filler type. Regardless of filler type, the overall best FS values were achieved by MCC filler, followed by TK and RH fillers. The incorporation of WF filler brought some improvement in FS values (Afzaluddin *et al.* 2019; Pandey *et al.* 2019; Çavuş 2020).

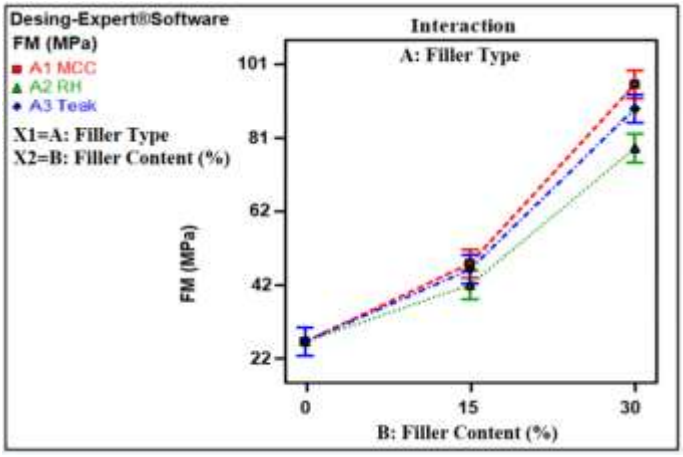

Fig. 6. Interaction graph of flexural modulus

The FM values of composites ranged from 27.5 to 96.6 MPa. The interaction charts of the FM are presented in Fig. 6. As expected, the lowest FM value was obtained from unfilled TPU. Statistical analysis showed that both filler type ($P = 0.0042$), and filler loading level ($P < 0.0001$) had significant effect on the FM values of the produced

composites. It should be noted that the filler loading level had more influence on FM than filler type. The increased modulus of the filled TPU composites was explained by El-Shekeil *et al.* (2012) and was a result of mixing a low stiffness matrix with a high stiffness filler. Similar results have been reported by others (Ramachandran and Vairavan 2007; AlMaadeed *et al.* 2012; Mohammed *et al.* 2016; Afzaluddin *et al.* 2019).

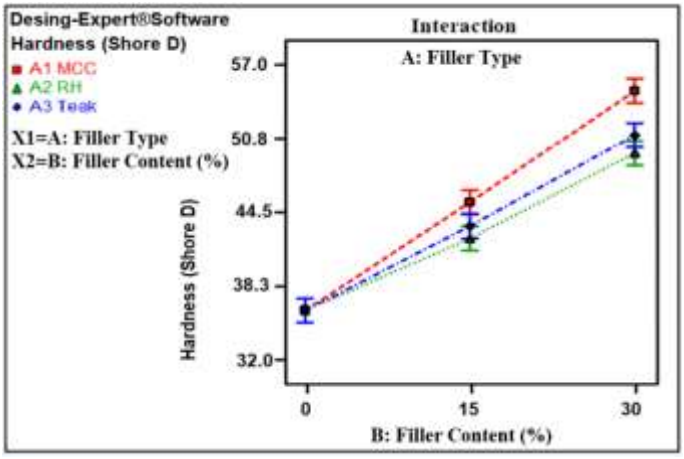

Fig. 7. Interaction graph of hardness (H)

The interaction charts of the hardness are presented in Fig. 7. The hardness values of test specimens ranged from 36.2 to 54.7 (Shore D). The lowest and highest H values were determined with unfilled TPU and 30% MCC-filled TPU specimens, respectively. The MCC-filled specimens provided slightly higher hardness values than the RH- and TK-filled ones. Based on the statistical analysis, both filler type and filler loading level had a significant effect on the H of the specimens ($P < 0.0001$). All fillers increased H values proportionally with the filler loading level. It has been reported that a higher hardness value of lignocellulosic filler increases the hardness of the thermoplastic matrix (Jamil *et al.* 2006). Similar results were also reported for natural fiber-filled TPU-based composites (El-Shekeil *et al.* 2012; Datta and Kopczyńska 2015; Tayfun *et al.* 2016; Kılınç *et al.* 2019).

The impact strength (IS) values were determined. Statistical analysis was not conducted to see the significant factors and interaction graph because most of the notched impact specimens were not broken during testing. The results showed that unfilled TPU and specimens with 15% filler did not break during impact testing. Based on visual observation, these specimens were stiffer and probably had lower IS values than unfilled TPU. However, samples were still elastic enough to bend before being broken by the test hammer. For 30% filler content, the IS values of the TK- and RH-filled specimens were measured as 27 kJ/m^2 and 35 kJ/m^2, respectively. There is a need to take precaution against interpreting these results because most of these samples were not completely broken into two parts. Only three out of five specimens having 30% TK were completely broken, and the other 30% TK- and RH-filled specimens were only cracked but not broken. Images of cracked and broken samples are presented in Fig. 8.

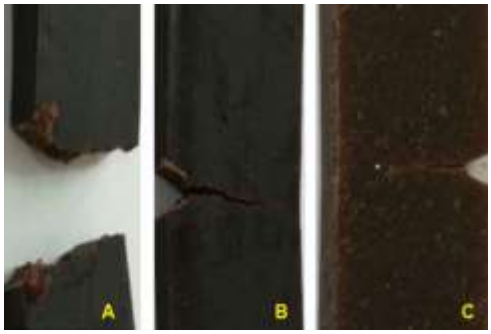

Fig. 8. Pictures of the samples after impact testing: a) TPU-30-TK (broken) specimen, b) TPU-30-TK (cracked) specimen, and c) TPU-30-RH specimen

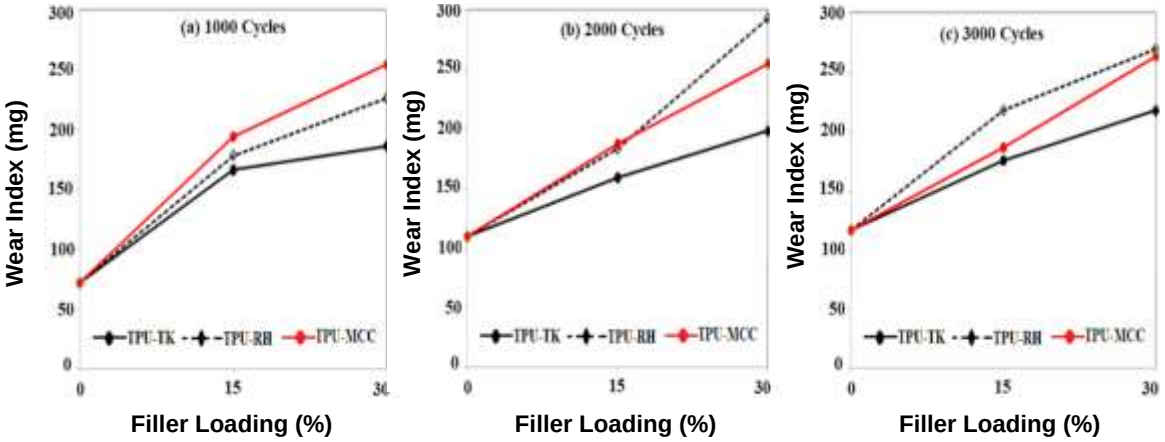

Fig. 9. Taber wear index (weight loss method) of test specimens

The abrasion resistance (AR) as Taber wear index values is presented in Fig. 9. They ranged from 72 to 254, 111 to 293, and 118 to 169 for 1000, 2000, and 3000 cycles, respectively. The lowest and highest wear index was obtained in unfilled TPU and 30% RH-filled TPU, respectively. Regardless of filler type, the addition of filler increased wear index values considerably, meaning that they reduced abrasion resistance. El-Shekeil *et al.* (2012) compared the abrasion weight loss of TPU and 30% kenaf fiber-filled TPU and found that weight loss increased with fiber addition. Similar results were also reported by Diestel and Krause (2018).

The SEM images of TPU and composites filled with 30% of MCC, RH, and TK WF specimens are shown in Figs. 10a through 10d. Figure 10a shows that MCC was embedded in the TPU and had a good degree of dispersion in the TPU matrix. Some micro cracks and some pulled out fibers in the TK- and RH-filled composites can be seen in Figs. 10c and 10d. When lignocellulosic filler was used, compared with composites produced with commodity polymers like polypropylene (PP) and polyethylene (PE), TPU provided better adhesion between the polymer matrix and filler. The hydrophilic nature of TPU played an important role in this result.

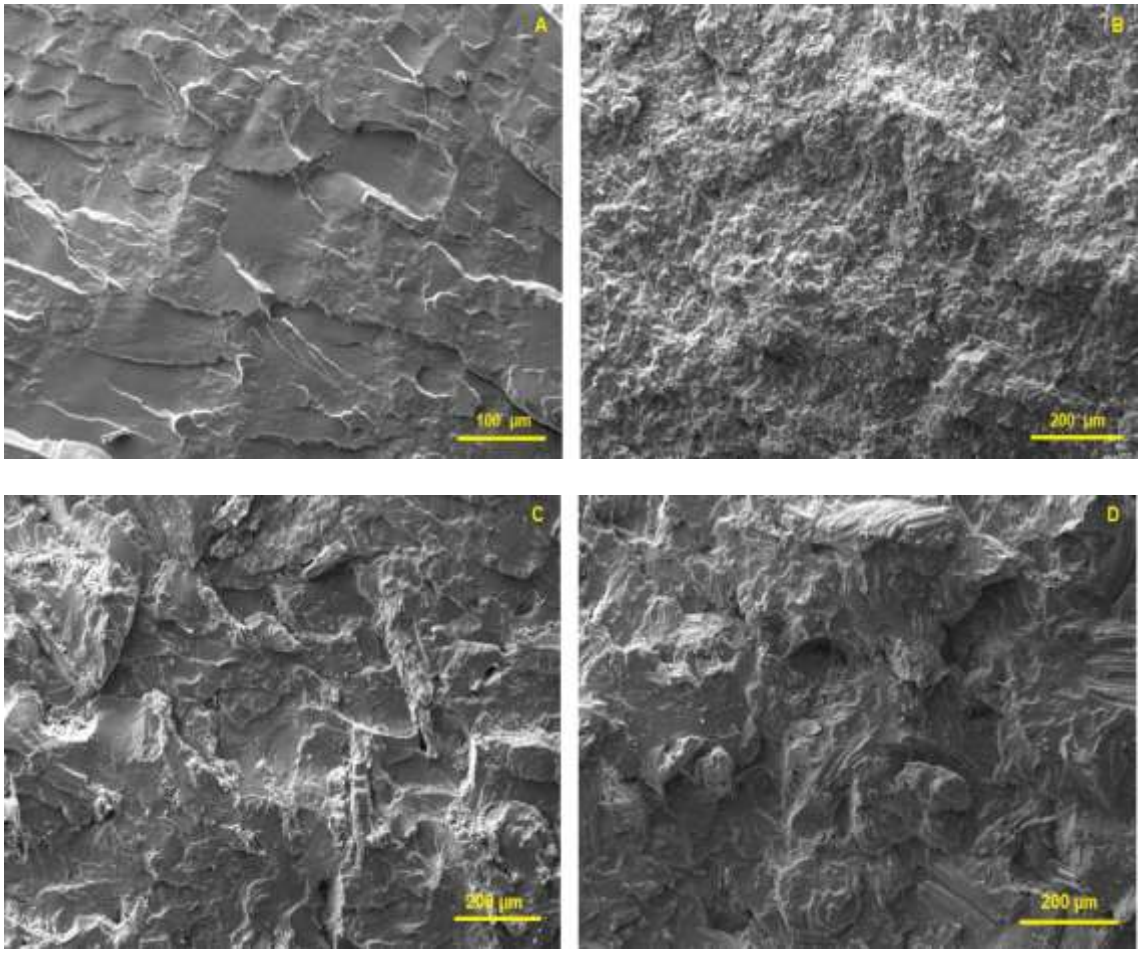

Fig. 10. SEM images of (a) TPU, and composites with (b) 30% of MCC, (c) RH, and (d) TK (500x magnification)

CONCLUSIONS

1. Thermoplastic polyurethane-based composites were produced using the three different fillers microcrystalline cellulose, teak wood, and rice hulk (MCC, TK, and RH) at 15 and 30% filling rates.
2. Both filler types and filler loading levels had a significant effect on the density and mechanical properties of thermoplastic polyurethane (TPU)-based composites.
3. Regardless of filler type, the addition of 15% filler in TPU provided excellent mechanical properties, such as over 500% EatB values, unbroken notched impact, and improved tensile strength (TS), yield strength (YS), flexural strength (FS), flexural modulus (FM), and tensile modulus (TM) values. They had properties almost comparable with those of unfilled TPU.
4. Because of the higher surface area in TPU composites, the TS of TPU composites continued to increase when 30% MCC was used as filler. This property was reduced when TK and RH were used as filler.

5. In the study range, there was a proportional correlation between hardness and modulus because hardness is a function of the relative fiber volume and modulus.
6. The addition of filler reduced the abrasion resistance of TPU. It was further decreased with a higher filler loading level.

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