

PLA Based Hybrid Composites Reinforced with TPU and Wood Flour for Advanced Engineering Applications

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Received: 08.01.2026

Revision: 10.03.2026

Accepted: 14.04.2026

Online: 18.06.2026

Abstract: This study investigates PLA-based green composites modified with bio-based thermoplastic polyurethane (TPU) and wood flour (WF) to reduce brittleness while maintaining mechanical properties. Composites containing 0-40% by weight TPU and 0-30% by weight WF were prepared by injection molding using 3% maleic anhydride-grafted polyethylene (MAPE) as a binder. Physical, mechanical, and morphological characterizations revealed that increasing the filler content systematically reduced the mechanical properties. TPU increased toughness (a 220% increase in tensile strength at break with 40% by weight TPU), while WF increased stiffness but reduced ductility. As confirmed by SEM analysis, both additives reduced density and increased water absorption due to porosity formation. ANOVA and Duncan tests confirmed that TPU and WF content have significant effects on composite properties, guiding the creation of optimal mixtures for sustainable engineering applications.

Keywords: Polylactic acid (PLA), Bio-based thermoplastic polyurethane (TPU), Green composites, Wood-plastic composites (WPCs)

I. INTRODUCTION

The development of polymer composite materials with superior mechanical properties has opened new horizons in engineering. Compared with conventional materials, advantages such as corrosion resistance, electrical insulation, easy machinability with relatively lower energy requirements in tooling and assembly costs, higher stiffness and strength, fatigue resistance and lower weight have made polymeric composites widely acceptable for various applications (Mittra, 2014). This situation increases the consumption of plastics, and rapidly increasing plastic consumption results in environmental pollution. Fillers in wood-plastic composites are known to be lightweight, environmentally friendly, noncorrosive, abundant and inexpensive. For these reasons, advances in processing technologies, increasing environmental awareness and economic factors have increased interest in wood-reinforced composites (WPCs) (Diestel & Krause, 2018; Mital'ová et al., 2024; Tokdemir & Altun, 2022; Zhou et al., 2024).

Compared to thermoset resins, thermoplastics offer advantages such as recyclability and being formaldehyde-free. Thermoplastic-based wood-plastic composites (WPCs) are produced from lignocellulosic materials such as dust from various wood species, agricultural wastes and thermoplastic polymer matrices (Anaç et al., 2024; Mengeloğlu & Çavuş, 2020; Öz et al., 2023). The choice of polymers in WPCs is limited to thermoplastics with melting points below 210 °C because of the thermal stability of the fibers (Diestel & Krause, 2018). Polyethylene (PE), polypropylene (PP), low-density polyethylene (LDPE), polylactic acid (PLA), and thermoplastic polyurethane (TPU) are some of these materials (Pandey et al., 2019; Stark & Rowlands, 2003; Zhang et al., 2017). Owing to its biocompatibility, biodegradability and ease of processing, PLA (polylactic acid) stands out as the main matrix of choice for WPCs (Bi et al., 2018; Motru et al., 2020; Wang et al., 2016). Owing to the brittleness of PLA due to its structure, studies have investigated the toughening of PLA in composites containing PLA (Pandey et al., 2019; Stark & Rowlands, 2003; Zhang et al., 2017). When the studies conducted for toughening PLA are examined, rubbers (NR, IR, NBR and SBR), ethylene copolymers (EVA, EMA, EBA-GMA, EMA-GMA) and thermoplastic elastomers (TPU and TPE) are used (Kaynak & Meyva, 2014), and these polymers are mostly petroleum-based. For this purpose, the use of more environmentally friendly materials is gaining importance. Another problem with WPCs is weak interfacial bonding due to the hydrophilic nature of wood and the hydrophobic nature of plastics. One of the methods to overcome this disadvantage is to improve the fibers via different processes (acetylation, mercerization, and treatment with silanes), and another method is to add suitable binding agents such as grafted polymers (maleic anhydride grafted or functional silane) (Diestel & Krause, 2018). In the literature, maleic anhydride-grafted polyethylene (MAPE) polymers are used as binding agents to increase the interfacial bonding between hydrophobic plastic polymers and hydrophilic WF, forming wood-plastic composites, and the mechanical properties of the resulting composites increase (Gao et al., 2010; Wang, 2021).

In this study, the aim was to produce wood-plastic green composites with preserved or improved mechanical and

physical properties based on a PLA matrix that is known in the literature for its biocompatible and biodegradable nature. The present work focuses on the mechanical, physical and morphological behavior of PLA-TPU-WF composites as a function of composition, while the biodegradation behavior of the produced composites was not experimentally investigated within the scope of this study. For this purpose, wood plastic (green) composites were produced by mixing bio-TPU (thermoplastic polyurethane) and WF (0–30%) in varying ratios (0–40%) with PLA as the matrix. In addition, 3% MAPE was added to the composites to improve the mechanical properties by increasing the interfacial bonding between the plastics and WF.

II. MATERIAL AND METHOD

A. Materials

The PLA polymer was preferred as the matrix because it is a biodegradable biomaterial and is one of the polymers whose use in green composite studies is increasing (Fang, et al., 2022; Khalifa, et al., 2020; Motru et al., 2020; Özmen et al., 2014; Solarski et al., 2005; Zhang et al., 2017). In this study, the PLA, trade name purapol lx175, was obtained from Kumru Kimya Sanayi ve Ticaret Ltd. (Istanbul, Turkey). PLA was obtained in transparent colored granule form at a density of 1.24 g per cm³, and the melt flow index (ISO 1133-A at 210 °C per 2.16 kg) was 8 g per 10 min, with 96% purity. According to the manufacturer's data, the melting temperature (T_m) was 155 °C, the glass transition temperature (T_g) was 55–60 °C, the tensile modulus (ISO 527-1) was 3500 MPa, the tensile strength (ISO 527-1) was 45 MPa, and the elongation at break (ISO 527-1) was a maximum of 5%. The bio-TPU used to toughen PLA was produced by Albis Plastik San. Tic. Ltd. (Istanbul, Turkey) and is known by the trade name Desmopan DP.33085AU. The density (ISO 1183-1) of 1,27 per cm³, melting temperature of 165–185 °C, ultimate tensile strength (DIN 53504) of 42.0 MPa, and strain at break (DIN 53504) of 680% are stated in the data sheet. The bio-TPU used in the study was in a transparently colored granule form. The MAPE content was fixed at 3 wt% in all formulations based on previous studies on wood-plastic composites, where similar levels were reported to be sufficient to improve interfacial adhesion without adversely affecting the overall processability and mechanical behaviour of the composites. The MAPE used as a binding agent was obtained from Clariant Turkey Paint Chemicals and Mining Industry and Trade Inc. (Istanbul/Turkey). The white-colored powder material has an acid value of 16–20 mg KOH/g acid (QM-AA-351a), a density of 0.97–0.99 g per cm³ (ISO 1183), and a viscosity value of 120–126 °C (ASTM D 3954) according to the manufacturer's data.

Scots pine (*Pinus sylvestris* L.) wood, which can grow in many regions of the world and is frequently used in the furniture and construction industry, is preferred as an additive in WPCs. Scots pine wood with a full dry density of 0.49 gr per cm³ and an air-dry density of 0.52 gr per cm³ was obtained from Can Lumber Industry Co. Ltd. (Turkey, Karabuk).

B. Methods

The PLA, bio-TPU and wood materials used in the study were pulverized by grinding in a plastic grinder (Mipro MLD, Protek Group Lab, Ankara, Turkey) to form a homogeneous mixture in the melt mixing process. The ground polymers were sieved through a shaker sieve (Loyka ESM 200, Akyol Trade Co., Ltd., Istanbul, Turkey) and dimensionally classified. WF's between 288 and 800 microns were used for the study. The dimensionally classified materials were dried in an oven (Nüve FN 055, Ankara, Turkey) at 60 °C for 6 hours and at 103±2 °C until the WF reached constant weight and were weighed on a 0.001 g precision balance (Kern, Albstadt, Germany) and mixed by weight in the proportions given in Table 1.

Table 1. Mixing ratios of wood–plastic composites.

	Code	PLA (%)	TPU (%)	WF (%)	MAPE (%)
Control	100-0-0	100	0	0	0
	87-10-0	87	10	0	3
	77-20-0	77	20	0	3
	67-30-0	67	30	0	3
	57-40-0	57	40	0	3
10% WF Flour	87-0-10	87	0	10	3
	77-10-10	77	10	10	3
	67-20-10	67	20	10	3
	57-30-10	57	30	10	3
	47-40-10	47	40	10	3

Table 1. (cont) Mixing ratios of wood–plastic composites.

20% WF Flour	77-0-20	77	0	20	3
	67-10-20	67	10	20	3
	57-20-20	57	20	20	3
	47-30-20	47	30	20	3
	37-40-20	37	40	20	3
30% WF Flour	67-0-30	67	0	30	3
	57-10-30	57	10	30	3
	47-20-30	47	20	30	3
	37-30-30	37	30	30	3
	27-40-30	27	40	30	3

A total of 20 groups were formed by mixing the powdered materials in the ratios given in Table 1, and each group was mixed for 1 hour in a multidirectional physical mixer. The mechanical sieve shaker, multidirectional physical mixer, and physical mixture used in the study are shown in Figure 1.

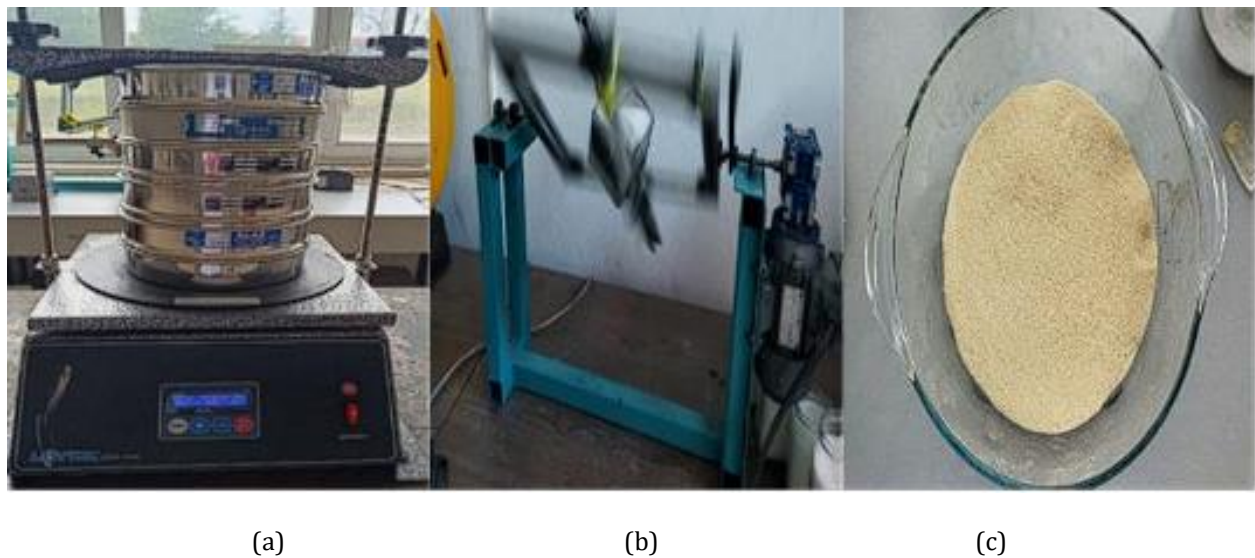


Figure 1. (a) Sieve, (b) multidirectional physical mixer, (c) physical mixture.

B.1. Preparation of Composites

The physically mixed batches were dried in a 60 °C drying oven for 12 hours to remove moisture before compositing. The powder samples dried in the drying oven were extruded in a twin-screw extruder with a diameter of 18 mm and an L-D ratio of 44 (Polartek Polimer Araştırma Teknolojileri San. ve Tic. Ltd. Şti., Istanbul, Turkey) at a 15 m per min feed rate was used for melt mixing. The temperature and feed rates for the melt mixing process are shown in Figure 2. After the extruded composites were converted into granules with the help of a crusher (Figure 3), they were oven dried at 60 °C for 72 hours.



Figure 2. Temperature and feed values applied in the melt mixing process.



Figure 3. Wood-plastic composites (a) 87-0-10, (b) 77-10-10, (c) 57-30-10.

Test samples were prepared from the dried green composites in granular form in a plastic injection molding machine to perform physical and mechanical tests. The samples were molded in 3 heating chambers of the injection molding machine at temperatures ranging from 160–175–180 °C and a 25 rpm feed rate. To perform the tests in accordance with the standards, 100 samples were prepared for each experiment, with 5 samples from each group (Figure 4). Prior to mechanical and physical testing, all specimens were conditioned and the test were carried out in a laboratory environment at approximately 23±2 °C and 50±5% relative humidity, in accordance with the recommended of the relevant ASTM standards.

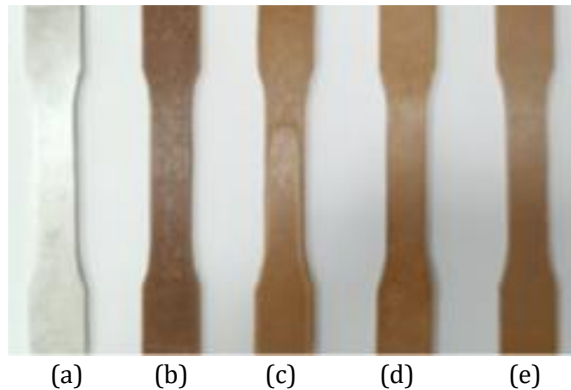


Figure 4. Tensile test samples: (a) 77-20-0, (b) 57-10-30, (c) 57-30-10, (d) 67-20-10, (e) 67-10-20.

B.2. Characterization Methods

For material characterization, scanning electron microscopy (SEM) images of the samples were obtained with a Carl Zeiss Ultra Plus Gemini Fesem (Carl Zeiss Microscopy GmbH, Jena, Germany). The samples were gold plated with a Quorum 150T ES (Quorum Technologies Limited, Sussex, UK) instrument before examination.

B.3. Determination of the Specific Gravity (Relative Density) and Density

The specific gravity (relative density) and density of the composites were determined in accordance with the standard ASTM D 792-00 test method by properly preparing the samples produced via injection molding. For this purpose, the

mass of the solid plastic sample in air was determined; then, it was immersed in pure water, and its density and specific gravity (relative density) were calculated according to equation (1) by determining its apparent mass when immersed.

$$\text{Sp. Gr. } 23/23^\circ\text{C} = a/(a-b) \quad (1)$$

where Sp.Gr.= specific gravimetry (relative density, specific gravity), a is the mass of the sample weighed in air (g), and b is the mass of the sample completely immersed in water (g).

The density was calculated according to equation 2.

$$D_{23^\circ\text{C}}, \text{ kg per m}^3 = \text{sp gr } 23/23^\circ\text{C} \times 997.5 \quad (2)$$

B.4. % Water Uptake

To examine the water uptake rates and dimensional properties of the biocomposites produced, the samples molded in the injection device were cut, and experiments were carried out in accordance with the ASTM D570-98 standard. Five samples from each group were first dried in an oven at $50 \pm 3^\circ\text{C}$ for 24 hours and cooled to determine their exact dry weights. The samples were then weighed on a balance with a precision of 0.001 g to determine their exact dry weights. The samples were immersed in pure water at $23 \pm 1^\circ\text{C}$ and weighed at 2 h, 12 h, 24 h and 168 h after the first immersion. Measurements of the samples were continued until two consecutive measurements were less than 1% of the total weight or less than 5 mg. At each measurement, the samples were blotted with a dry cloth to remove water from the surface, weighed on a 0.001 g scale and recorded.

The percent water uptake after immersion was calculated according to equation 3.

$$\%m = (m_t - m_o) / m_o \times 100 \quad (3)$$

%m, water absorption rate; m_t , immersed sample weight (g); m_o , complete dry sample weight(g)

B.5. Material Characterization

Tensile tests type I (ASTM D638-14), 3-point bending tests and modulus of elasticity in bending tests (ASTM D790-10) were performed on the composites produced. The tests were performed on a universal testing machine (Shimadzu, Kyoto, Japan) with 5 samples from each group. The tensile strength of each sample was calculated via equation (4).

$$\delta T = \frac{F_{max}}{a \times b} \text{ N/mm}^2 \quad (4)$$

δT = Tensile strength (N/mm²), F_{max} = maximum force at rupture (N), a = thickness of the sample at fracture, b = width of the sample at fracture

The bending resistance and modulus of elasticity during bending of the composites were calculated via equations (5) and (6), respectively.

$$e = \frac{3 \times F \times L}{2 \times b \times t^2} \text{ N/mm}^2 \quad (5)$$

$$E = \frac{F \times L^3}{4 \times \Delta e \times b \times t^3} \text{ N/mm}^2 \quad (6)$$

where F is the maximum force at the moment of fracture (N), L is the clearance between the support points (mm), t is the sample thickness (mm), b is the sample width (mm), and Δe is the deflection due to the load (mm).

B.6. Evaluation of the Data

At the end of the experiments, the data were analyzed via SPSS statistical software (version 22.0; SPSS Inc., Chicago, IL, USA). Two-way analysis of variance (ANOVA) was used to determine whether there were significant differences between the green composite samples. This method was used to examine whether the contributions of TPU, WF, and TPUxWF were statistically significant. Following ANOVA, Duncan's test was applied to identify significant differences ($p < 0.05$).

III. RESULTS AND DISCUSSIONS

A. Material characterization

SEM analysis of the samples taken from the fracture zones of the tensile tested wood-plastic composites was performed. fractographs are shown in Figure 5.

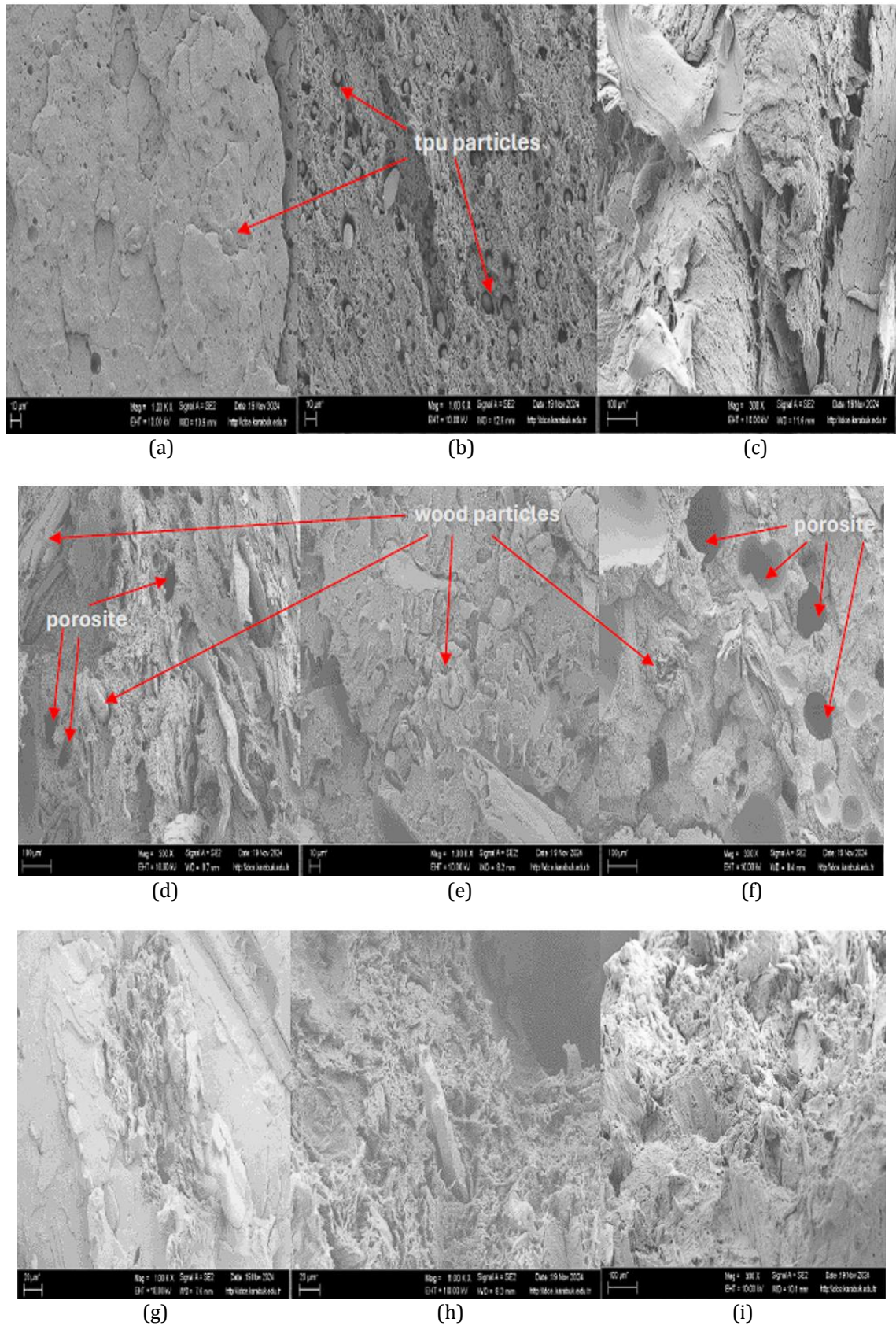


Figure 5. Fractographs from fracture sites: (a) 77-20-0, (b) 67-30-0, (c) 57-40-0, (d) 47-40-10, (e) 67-10-20, (f) 57-20-20, (g) 67-0-30, (h) 47-30-20, (i) 27-40-30.

The fractographs of the composites taken after fracture revealed that creep and roughening due to "shear yielding" occurred on the surfaces as the TPU ratio increased (Figure 5. (c), (h), (i)). This indicates that significant plastic deformation occurred during rupture. In addition, as shown in Figure 5. (a), (b), The TPU shows an irregular distribution in the form of microspherical structures. The images show that the composites form a "two-phase structure" and that the

TPU phase is not mixed in the continuous matrix phase of PLA. Similarly, it has been reported in the literature that the TPU phase does not mix with the PLA phase and results in surface roughness during rupture (de Melo Morgado et al., 2022; Fang et al., 2022; Meyva & Kaynak, 2015.). In addition, fractographs (Figure 5. (d), (f)) show that porosities are formed in the TPU blended composites and that the porosities increase and increase in size in parallel with the TPU ratio. The reason for this may be the increase in porosity as a result of the increase in the TPU ratio, the enlargement of the pore walls by merging small pores and the formation of large pores. Mi H., et al. reported similar findings in their study (Mi et al., 2013).

B. Density

The densities of the green composites obtained were calculated in accordance with the standard ASTM D 792-00 test method, and the results are presented graphically in Figure 6.

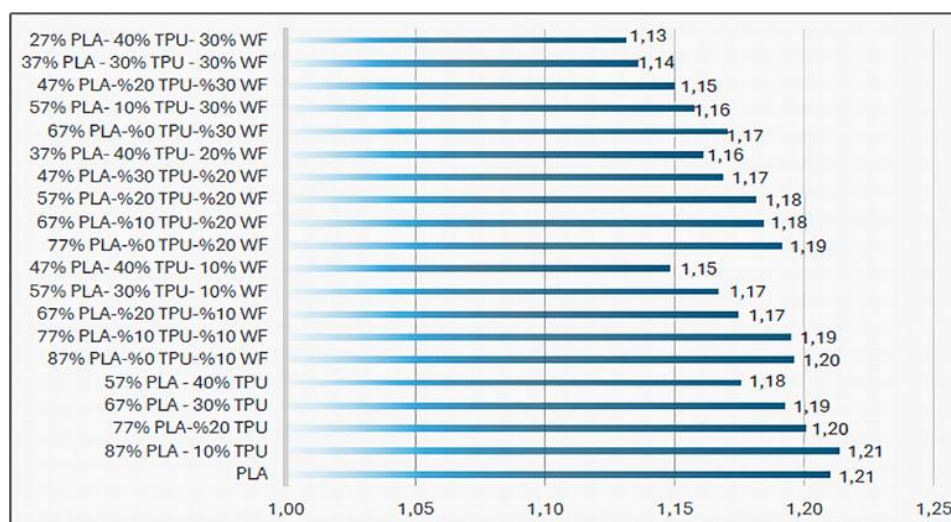


Figure 6. Density graph (g).

When analyzing the density values of wood-plastic composites, it was observed that wood and TPU additives reduced the density values. Compared to pure PLA, a 40% TPU additive caused a 2.91% decrease in density. Similarly, a 30% wood filler caused a 3.44% decrease when compared to pure PLA. All percentage in density were calculated by taking the density of neat PLA as the reference value (%100). When analyzing the density values of TPU and wood-filled composites, the values of the 47PLA-40TPU-10WF, 37PLA-40TPU-20WF, and 27PLA-40TPU-30WF composites decreased by 4.95%, 4.13%, and 6.61%, respectively, compared to pure PLA. As seen in the SEM fracture graphs, the foaming of the TPU polymer upon reaction with heat and the formation of a porous structure reduce the material ratio per unit area of the composites, which leads to a decrease in density. Furthermore, the reason why the wood filler reduces the density of the composites is generally explained in the literature by the "mixture rule." When a lower-density filler is incorporated into a higher-density polymer matrix, the resulting composite typically has a lower density than the polymer itself (Clemons, 2010; Diestel & Krause, 2018; Mäkinen et al., 2002; Mengeloğlu & Çavuş, 2020). This decrease stems from the lower cell wall density of Scots pine wood (approximately 0.52 g per cm³). The results of the variance analysis for density values are presented in Table 2.

Table 2. Analysis of Variance (ANOVA) Results for Density Properties.

Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	.052 ^a	19	.003	5.590	.001
Intercept	138.080	1	138.080	283132.300	.001
TPU	.019	4	.005	9.492	.001
WF	.033	3	.011	22.242	.001
TPU * WF	.001	12	6.188E-5	.127	1.000
Error	.039	80	.000		
Total	138.171	100			
Corrected Total	.091	99			

According to the results of the variance analysis of density, the contribution ratios of TPU and WF were statistically significant between groups ($p < 0.05$), whereas the interaction resulting from the combined use of TPU and WF (TPU*WF) had no statistically significant effect. Duncan tests were performed on the values to determine which groups differed

from each other, and the results of the Duncan test for the TPU contribution are presented in Table.3.

Table 3. Duncan test results for density, (a) TPU rate, (b) WF rate.

TPU Rate	Subset			
	1	2	3	4
40	1.1555			
30	1.1652	1.1652		
20		1.1759	1.1759	
10			1.1848	1.1848
0				1.1937
Sig.	.169	.127	.208	.204

(a)

WF Rate	Subset		
	1	2	3
30	1.1480		
10		1.1763	
20		1.1771	
0			1.1988
Sig.	1.000	.901	1.000

(b)

According to the Duncan test results, the effect of the WF contribution ratio can be expressed as follows:

No statistically significant difference was found between the groups with 10% and 20% WF contributions. These findings indicate that these two contribution ratios have similar effects and have similar effects on density.

However, the group with a 30% WF contribution was significantly different from both the control group (no contribution) and the groups with low contribution rates (10% and 20%). This result shows that the 30% contribution rate clearly differs from that of the other groups and significantly reduces the density.

When the results of the Duncan test for TPU contribution rates were examined, no significant difference was observed between consecutive groups. However, a significant difference was observed between the 40% contribution group and both the 20% contribution group and the control group. This indicates that the 40% group, which has the highest TPU content, is distinctly different from the medium-level (20%) group and the control group, which contains no TPU. In conclusion, the TPU content gradually increased, with the most significant decrease observed in the 40% TPU content group.

C. Water Uptake Rate %

The water uptake rates of the composites are given graphically in Figure 7.

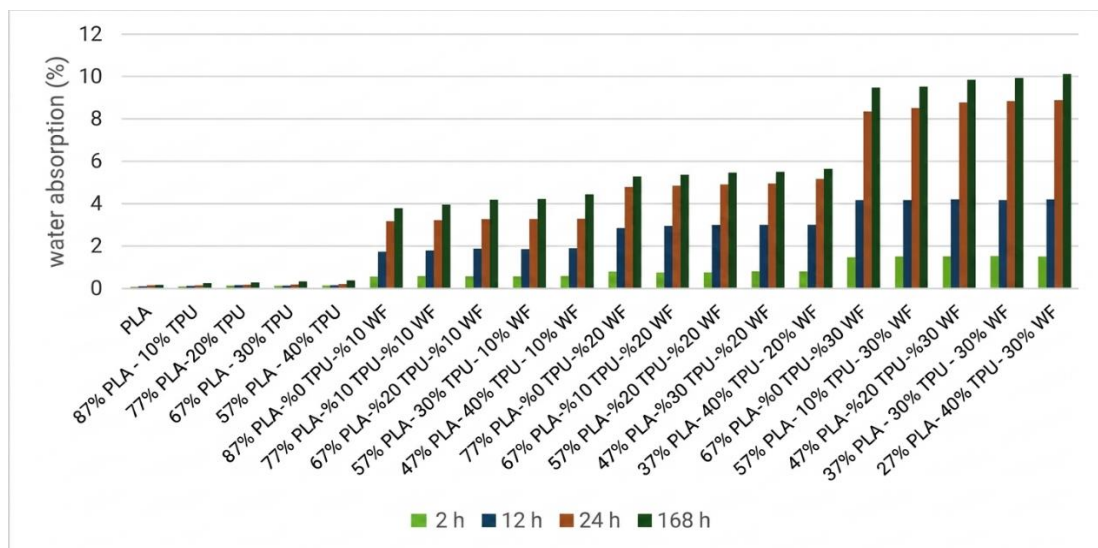


Figure 7. % water absorption rates.

An analysis of the water absorption rates revealed that the water absorption rates of the composites increased with time. In both the PLA-TPU composites and the PLA-TPU-WF composites, water absorption increased as the TPU ratio increased. The highest water absorption in the PLA-TPU composites was measured at 168 h, with a value of 0.37%. When the PLA-TPU-WF composites were examined, a significant increase in the water absorption rate was observed in parallel with the ratio of TPU to wood. The reason for the increase in water absorption parallel to the TPU ratio may be the filling of the pores visualized in the SEM fractographs with water over time. Although the PLA and TPU polymers are hydrophobic materials, the fact that water remained unbound here increased the water absorption rate. In addition, the hydrophilic structure of the wood also increased water absorption. The main reason for the increases obtained, especially as a result of prolonged storage, can be attributed to the settling of water in the lumens of wood particles and the binding of water to OH- groups in the wood structure and settling in the cell walls. In addition, the porosities formed by TPU and the easier access of water to more wood particles through these pores are thought to increase water absorption. The results of the variance analysis of the water absorption rates of the composites are given in Table 4.

Table 4. Analysis of variance (ANOVA) results for composite water absorption.

Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	1158.604 ^a	19	60.979	1519.899	.001
Intercept	2403.198	1	2403.198	59899.479	.001
TPU	2.630	4	.657	16.385	.001
WF	1155.411	3	385.137	9599.500	.001
TPU * WF	.564	12	.047	1.171	.318
Error	3.210	80	.040		
Total	3565.012	100			
Corrected Total	1161.814	99			
a. R Squared = ,570 (Adjusted R Squared = ,468)					

When the results of the variance analysis of the water absorption rates of the green composites over 168 hours were examined, the TPU contribution and the TPU*WF interaction did not significantly differ, whereas the WF contribution was effective ($p < 0.05$). The Duncan test was performed to examine the groups in which the differences occurred. The results of the Duncan test for water uptake rates over 168 hours are shown in Table 5.

Table 5. Duncan test results for 168-hour water uptake rates (%).

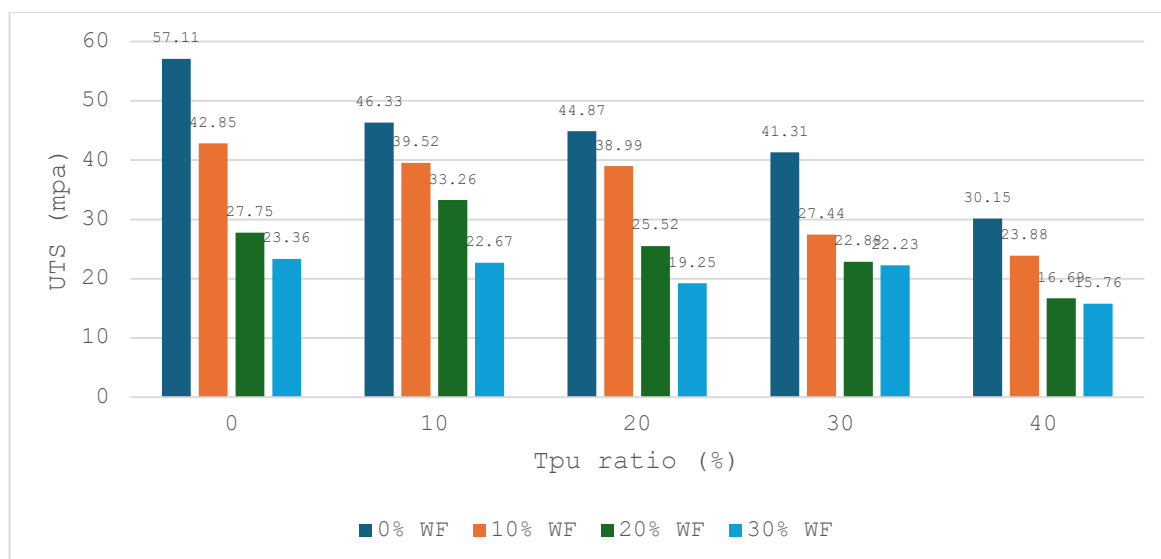
WF	Subset			
	1	2	3	4
Table 5 (continue)				
0	.2725			
10		4.1121		
20			5.4360	
30				9.7882
Sig.	1.000	1.000	1.000	1.000

The results of the Duncan test revealed that all the WF groups had a significant effect on the water absorption rate. The analysis results show that the water absorption rate increases systematically with increasing WF content. These findings reveal that the WF content enhances the hydrophilicity of the composite material and that an increase in the WF content proportionally increases the water absorption capacity. The results show that the WF addition ratio has a linear effect on water absorption behavior and that each addition level has significantly different water absorption characteristics.

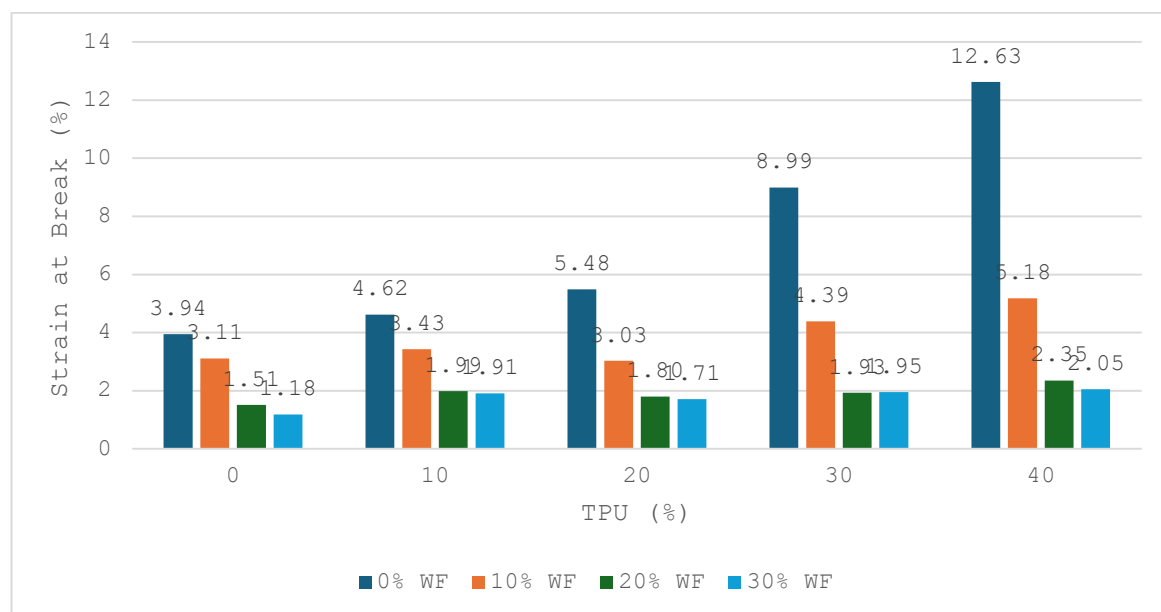
D. Material Characterization

When the ultimate tensile stress values of the composites (Figure 8) were examined, the addition of the wood and TPU additives to the pure PLA gradually decreased the tensile strength. While the addition of 30% wood decreased the tensile strength of pure PLA by 59.1%, with a value of 23.36 mpa, the addition of 40% TPU decreased the tensile strength by 47.21%, with a value of 30.15 mpa. The lowest value was observed for the 27PLA-40TPU-30WF composite with 15.76 mpa, and a 72.40% decrease in tensile strength was observed. Morgado et al. examined the mechanical properties of bio blends obtained by preparing different ratios of PLA-TPU blends and reported that the tensile strength values of PLA bio blends with 10%, 20%, 30% and 40% TPU were 38.40, 31.75, 26.06, and 20.60 mpa, respectively. In the same study, the addition of TPU decreased the tensile strength (de Melo Morgado et al., 2022). The values obtained in our study are

approximately 20,65%, 41,32%, 58,51% and 46,35% higher than these reported values. This can be attributed to the use of 3% PE-G-MAH as a binding agent in all the groups. In a similar study, Kaynak and Meyva reported that the use of a PLA-G-MAH compatibilizer in 10% TPU-doped PLA composites increased interfacial bonding, and the strength values increased accordingly because of effective intermolecular charge transfer (Kaynak & Meyva, 2014). The UTS values are shown in Figure 8.



(a) Ultimate tensile strength



(b) Strain at break

Figure 8. (a) Ultimate tensile strength and (b) strain at break.

When the strain at break is examined, the elongation at break increases in parallel with the increase in the TPU ratio. The addition of 40% TPU to pure PLA caused an increase in the strain value of 220%. The lowest strain value was measured to be 1.18% in the composites with 30% wood additive without the TPU additive. These values indicate that the addition of wood decreases the elongation at break values. The soft segment of TPU, consisting of polyol and isocyanate, imparted flexibility and elastomeric properties to the mixture, which decreased the tensile strength but increased the elongation at break. The strain values of the composites with 20% and 30% wood additives are close to each other, and there is no significant difference among all the TPU additive groups. The weaker interface bond between WF-TPU compared with the interface bond between PLA-TPU is likely the reason for this situation. When similar studies in literature are examined, the results of Pandey et al. (2019) and Bi et al. (2018) support the results obtained (Bi et al., 2018; Pandey et al., 2019). The results of the variance analysis for the UTS values are presented in Table 6.

Table 6. Results of variance analysis for UTS values.

Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	12237.953 ^a	19	644.103	1579.396	.001
Intercept	96665.650	1	96665.650	237032.510	.001
TPU	3225.133	4	806.283	1977.076	.001
WF	8018.367	3	2672.789	6553.909	.001
TPU * WF	994.453	12	82.871	203.207	.001
Error	32.625	80	.408		
Total	108936.228	100			
Corrected Total	12270.578	99			
a. R Squared = ,570 (Adjusted R Squared = ,468)					

When the UTS variance analysis results were examined, the TPU, WF, and TPU*WF interactions were significantly different between the groups ($p < 0.05$). This finding indicates that, in addition to the effects exhibited by each additive individually, their combined use produces a synergistic or antagonistic effect. Duncan tests were performed to examine which groups exhibited these effects. The results of the Duncan test for the UTS values are given in Table 7.

Table 7. Duncan test results for UTS values: (a) TPU rate and (b) WF rate.

TPU Rate	Subset				
	1	2	3	4	5
40	21.6205				
30		28.4650			
20			32.1575		
10				35.4450	
,0					37.7675
Sig.	1.000	1.000	1.000	1.000	1.000

(a)

WF rate	Subset			
	1	2	3	4
30	20.6540			
20		25.2200		
10			34.5360	
0				43.9544
Sig.	1.000	1.000	1.000	1.000

(b)

When the Duncan test results of TPU addition were evaluated, it was determined that all TPU addition levels had a significant effect on the tensile strength (UTS) values. Each TPU addition level was significantly different from that in the other groups ($p < 0.05$).

These findings reveal that every change in the TPU additive ratio has significant and measurable effects on the UTS. The results obtained indicate that TPU has a systematic effect on the mechanical properties of the composite material depending on the additive ratio and that the additive ratio must be carefully determined to achieve optimal performance.

When the Duncan test results of the WF contribution were evaluated, it was determined that all WF contribution ratios had a significant effect on the tensile strength (UTS) values. Each WF contribution level was significantly different from that of the other groups ($p < 0.05$).

These findings reveal that every change in the WF addition ratio has significant and measurable effects on the UTS. The results obtained indicate that WF has a systematic effect on the mechanical properties of composite materials depending on the addition ratio and that the addition ratio must be carefully determined for optimal performance.

The results of the variance analysis for strain at break values are given in Table 8.

Table 8. Variance analysis results for strains at break values.

Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	756.907 ^a	19	39.837	1163.470	.001
Intercept	1375.668	1	1375.668	40177.223	.001
TPU	122.135	4	30.534	891.754	.001
WF	470.804	3	156.935	4583.367	.001
TPU * WF	163.969	12	13.664	399.067	.001
Error	2.739	80	.034		
Total	2135.314	100			
Corrected Total	759.646	99			
a. R Squared = ,570 (Adjusted R Squared = ,468)					

The results of the variance analysis of the effects of the types of additives on the strain at break revealed that the TPU, WF, and TPU*WF interactions were statistically significant at $p < 0.05$. These results show that the elongation at break properties of the composite material are affected independently of both the TPU and WF additives and that the combination of these two additives also plays an important role in elongation at break behavior. The presence of the interaction effect indicates that the additive ratios must be evaluated together for optimal performance. A Duncan test was performed to examine the groups in which the differences occurred. The results of the Duncan test for strain at break values are presented in Table 9.

Table 9. Results of the duncan test for strains at break: (a) TPU rate and (b) WF rate.

TPU Rate	Subset				
	1	2	3	4	5
0	2.4350				
10		3.0050			
20			3.2375		
30				4.3150	
40					5.5525
Sig.	1.000	1.000	1.000	1.000	1.000

(a)

WF	Subset			
	1	2	3	4
30	1.7600			
20		1.9160		
10			4.0280	
0				7.1320
Sig.	1.000	1.000	1.000	1.000

(b)

According to the results of the Duncan test, each TPU ratio affects the strain at break, and the elongation values increase as the TPU ratio increases. These findings show that the elastomeric properties of TPU are transferred to the composite material and that increasing the TPU content directly affects the flexibility and deformation capacity of the material. The results show that the TPU content ratio has a linear effect on the elongation at break behavior and that each content level has statistically distinct elongation characteristics.

When the Duncan test results of the WF contribution were evaluated, it was determined that each WF contribution ratio had a significant effect on the strain at break values. The analysis results show that the strain at break decreases systematically with increasing WF contribution ratio. These findings reveal that the rigid fiber structure of WFs is transferred to the composite material and that increasing the WF content limits the flexibility and deformation capacity of the material in an inversely proportional manner. The obtained results indicate that the WF content ratio has a negative linear effect on the strain at break and that each content level has statistically distinct elongation characteristics.

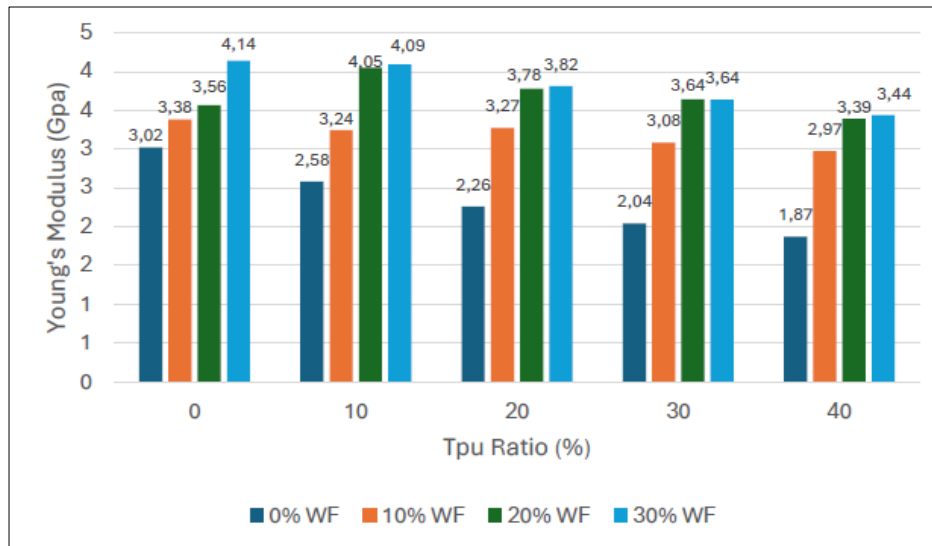


Figure 9. Young's modulus.

To describe the stress–strain relationship in the elastic deformation region of the composites, the Young's modulus values were calculated and are shown in Figure 9. As the WF content increases, the Young's modulus systematically increases, whereas increasing TPU content leads to a systematic decrease in the modulus. The addition of WF increased the Young's modulus values of the composites in all the groups of PLA-TPU blends. The lowest value was measured for the PLA composites with 40% TPU addition at 1.87 gpa, whereas the highest value was measured for the PLA composites with 30% WF addition at 4.14 gpa. As shown in the graph, TPU doping decreases the stiffness of the composites, whereas WF doping increases it. Morgado et al. (2022) reported the Young's modulus values of composites produced by adding 0–40 wt% TPU to PLA as 2.62 GPa, 1.98 GPa, 1.76 GPa, 1.39 GPa and 1.05 GPa, respectively, and similarly, Kaynak and Meyva (2014) reported the Young's modulus values of composites with 10% TPU and 10% TPU + 5% PLA-g-MAH as 2.62 GPa and 2.63 GPa, respectively ((de Melo Morgado et al., 2022, Kaynak & Meyva, 2014). The almost identical values reported by Kaynak and Meyva (2.62 vs. 2.63 GPa) indicate that the addition of PLA-g-MAH does not significantly change the stiffness of the material but rather improves the interfacial adhesion and helps to maintain the modulus while enhancing other mechanical properties such as strength and toughness. These trends are consistent with our findings, where the incorporation of a coupling agent and WF primarily stabilizes or slightly modifies the Young's modulus while strongly affecting tensile and flexural behaviour. Figure 9 shows a graph of the bending resistance and modulus of elasticity during bending.



Figure 10. Flexural resistance and elasticity in bending.

When the flexural strength and flexural modulus of elasticity values of the composites are examined (Figure 10), the addition of TPU to pure PLA decreases the flexural strength and flexural modulus of elasticity values. The flexural strength values of the 40% TPU-added PLA mixtures decreased by 37.78%, with a value of 59.78 mpa. When the modulus of elasticity values of the composites with the same percentage of TPU additive are examined, a decrease of 45.95% is

observed compared with that of pure PLA with a value of 1.87 GPa. Typically, elastomeric materials significantly increase the ductility and toughness of brittle polymeric materials, but this significantly reduces their strength (Kaynak & Meyva, 2014). In addition to the TPU additive, the addition of 10% wood additive to the composites increased both the flexural strength and modulus of elasticity during bending, whereas the addition of 20% wood additive resulted in values comparable to those of the same ratio of PLA-TPU composites. When the composites were analyzed within their own groups, the values of the composites with 40% TPU increased the flexural strengths of the composites with 10%, 20% and 30% wood additives by 31.63%, 19.55% and 12.14%, respectively, compared with those of the group without wood additives. In a similar study, 30 wt% PLA-g-ma was shown to interact with wood fibers owing to its polar molecules and provide a better bond at the interface between PLA and wood, resulting in a more effective transfer of stress from the PLA matrix to the wood fibers and an increase in mechanical properties (Zhang et al., 2017).

In a similar study, the flexural strength value of PLA-g-ma compatibilizer-added, 10% TPU-added, and PLA blends were 71.1 MPa, and the flexural modulus of elasticity value was 3.48 GPa (Kaynak & Meyva, 2014). These results are consistent with the results obtained in this study.

In such composite structures, the lower strength component (TPU) is reported to play a more dominant role in the damage mechanism of the material, while the reason why TPU increases ductility while decreasing mechanical properties is explained by the rubber-toughening mechanism. In this mechanism, in addition to the mechanical behavior of the components, the morphology and microstructure of the mixtures also affect the damage mechanism. When an external force is applied, hard PLA exhibits plastic behavior, whereas soft TPU exhibits elastic behavior. Damage starts at points prone to crack formation and with higher stress intensity. These points can represent the weak interface between two phases or a phase with lower strength. As the amount of TPU increases, the interface area between the PLA and TPU phases increases, and the strength decreases with the possibility of stress concentration occurring at lower forces. Owing to the weak interface bonding, micro voids at the interface cause the strength to decrease. As the amount of TPU increases, elastic behavior becomes dominant, which increases ductility (Kaynak & Meyva, 2014).

The results of the variance analysis for the Young's modulus values are given in Table 10.

Table 10. Results of variance analysis for Young's Modulus.

Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	40.856 ^a	19	2.150	73.850	.001
Intercept	1064.717	1	1064.717	36566.220	.001
TPU	5.330	4	1.332	45.762	.001
WF	33.153	3	11.051	379.530	.001
TPU * WF	2.373	12	.198	6.792	.001
Error	2.329	80	.029		
Total	1107.902	100			
Corrected Total	43.186	99			
a. R Squared = ,946 (Adjusted R Squared = ,933)					

When the variance in the Young's modulus values is analyzed, it is observed that both TPU and WF, as well as the TPU*WF interaction, are statistically significant. These results indicate that the Young's modulus values of the composite material are affected independently of both the TPU and WF contributions and that the combination of these two contributions also plays an important role in the stiffness behavior. To determine in detail the contribution ratios between which the observed significant differences occurred, the Duncan multiple comparison test was applied. Table 11 shows the results of the Duncan test.

Table 11. Young's Modulus duncan test results.

TPU	Subset			
	1	2	3	4
40	2.9175			
30		3.1000		
20			3.2825	
10				3.4900
,0				3.5250
Sig.	1.000	1.000	1.000	.518

(a)

Table 11. (cont) Young's Modulus duncan test results.

WF	Subset			
	1	2	3	4
0	2.3540			
10		3.1880		
20			3.6840	
30				3.8260
Sig.	1.000	1.000	1.000	1.000

(b)

When the Duncan test results of the TPU contribution were evaluated, the 10% TPU contribution ratio exhibited statistically similar values to those of the control group. This low contribution ratio does not have a significant effect on the stiffness properties of the composite material. In addition, the 20%, 30%, and 40% TPU contribution ratios had statistically significant effects on the Young's modulus. The analysis results reveal that the Young's modulus values decrease systematically in inverse proportion to the increase in the TPU content. These findings indicate that the elastomeric properties of TPU begin to be transferred to the composite material after a certain threshold value and that increasing the TPU content affects the rigidity of the material inversely. The results obtained indicate that TPU addition modifies the mechanical behavior of the composite material in terms of flexibility.

When the results of the Duncan test for WF contribution were evaluated, it was determined that all wood contribution ratios had a significant effect on the Young's modulus values. Each WF contribution level was significantly different from those of the other groups. The analysis results reveal that the Young's modulus values systematically increase with increasing WF additive ratio. These findings indicate that WF reinforces the material by increasing the rigidity of the composite material through its natural fiber structure. The results revealed that increasing the WF content proportionally increased the material's elasticity modulus and that WF effectively reinforced the composite system. This behavior can be explained by the mechanical contribution of the cellulose structure of WF to the composite matrix.

E. Environmental Considerations

The developed composites offer the following significant environmental advantages,

- Potential biodegradability associated with the PLA matrix and natural wood fibers, as reported in the literature, although no direct biodegradation tests were conducted in this study
- Bio-based TPU reduces petroleum dependency
- Lower processing temperatures (160-180 °C) compared to conventional WPCs
- Potential for end-of-life composting under industrial conditions

However, the increased water absorption (up to 9,79 % for 30% WF content) may limit outdoor applications without surface treatment. Future studies should investigate hydrophobic treatments and weathering resistance.

IV. CONCLUSION

PLA-TPU-WF green composites were successfully fabricated by mixing different ratios by weight. Some of the morphological, physical and mechanical properties of the composites were investigated. The results revealed that the TPU and WF additives decreased the specific gravity and increased the water absorption rate of the composites. Fractographs visualized via SEM analysis revealed that this was due to the porosity formed as a result of the reaction of TPU with temperature. Again, decreases in the mechanical properties were observed in parallel with the rates of these additives. While the TPU additive was observed to toughen the composites, the WF additive was found to make them rigid. When the results of the study are evaluated, the following recommendations are presented.

1-The use of 3% MAPE as a binding agent improved the mechanical properties of the composites. This 3% level was selected based on previous studies on wood-plastic composites, where similar MAPE contents were reported to be sufficient to enhance interfacial adhesion without causing processing problems or excessive brittleness. By increasing and varying the MAPE ratio around this level, better results for the mechanical properties can be obtained, and its effect on composites can be better understood.

2-The use of different types of binding agents (PLA-G-MAH, TPU-G-MAH, EPDM-G-MAH) at different ratios to strengthen the bond between PLA and TPU polymers, which are hydrophobic materials, and hydrophilic wood is thought to increase the mechanical properties of composites.

3-Investigating lower temperature values in the production process of composites is thought to reduce the porosity caused by TPU. Owing to this situation, an increase in density and mechanical properties and a decrease in water absorption are likely recorded.

4- It is thought that the smaller size of the wood particles used in the production of composites results in a more homogeneous distribution within the composites and increases the mechanical properties.

5- The use of wood samples with different specific gravities at different weight ratios in the production of composites may provide a better understanding of the physical and mechanical properties of composites.

6- Systematic biodegradation tests (e.g., industrial composting, soil burial and controlled humidity-temperature conditions) should be conducted to quantify the degradation rate and mass loss of PLA-TPU-WF composites and to compare them with neat PLA and PLA-WF systems.

7- The influence of different TPU contents, wood flour loadings and coupling agents on biodegradation behaviour should be investigated to identify compositions that balance mechanical performance with accelerated and controllable biodegradability.

Although the composites obtained can be a potential solution for industries that need environmentally friendly and lightweight materials, further research is needed to solve problems such as water absorption, porosity formation and increased mechanical properties. In addition, future work should also address the biodegradation behaviour of these PLA-TPU-WF systems under different environmental conditions (e.g., industrial composting, soil burial, and controlled humidity) to better assess their long-term environmental performance.

DECLARATIONS

Acknowledgments: This study was carried out with the support of Karabuk University Coordinatorship of Research Project with project number KBÜBAP-23-ABP-082.

Author Contributions: Methodology, V.T.; Research, V.T.; Writing—Original Draft, V.T.; Writing—Review and Editing, S.A.; Supervision, S.A.; Formal Analysis: V.T., S.A.; References: V.T.; Data Organization: S.A.; Project Management: V.T.; Funding Acquisition: V.T.; All authors have read and approved the final version of the article.

Conflict of Interest: The authors declare that there is no conflict of interest.

Supporting Organizations: This study was carried out with the support of the Karabuk University Coordinatorship of Research Project with project number KBÜBAP-23-ABP-082.

Ethical Approval: This study does not involve human or animal participants. All procedures were performed in accordance with scientific and ethical principles, and all referenced studies have been properly cited.

Plagiarism Statement: This article has been reviewed for plagiarism, and no cases of plagiarism have been found.

The Use of AI Tools: The author(s) declare that no Artificial Intelligence (AI) tools were used in the preparation of this article.

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