Determination of Some Physical and Mechanical Properties of Laminated Scots Pine Using Polylactic Acid in the Middle Layers

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Mechanical and physical properties were determined for laminated Scots pine (*Pinus sylvestris* L.) using polylactic acid (PLA). Structural parts were produced *via* 3D printing with additive manufacturing (FDM: fused deposition modeling) method in the middle layers. For this purpose, PLA parts with 30%, 60%, and 90% filling rates were glued to the middle of the 5-layer laminated material with polyurethane (PU) glue, and physical and mechanical tests were conducted according to the relevant standards. It was found that increasing the filler content resulted in higher density, reduced water absorption and thickness swelling, increased bending strength and modulus of elasticity, while screw holding resistance increased by approximately 75%. Compressive strength parallel to the fibers showed comparable results to control samples. These findings suggest that PLA filler particles can serve as an alternative middle layer material in wood lamination, offering potential for minor but meaningful improvements in specific applications.

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INTRODUCTION

In parallel with the increasing global population, the use of wood and wood-based industrial products is increasing each day. In competition with this situation, the use of laminated wood material is increasing, and academic studies are being carried out on producing more durable and efficient materials. The structural element obtained by gluing wood lamellae, especially fibers, in parallel is defined as laminated wood material (TS EN 386 1999). Wood lamination is aimed at improving some properties of wood, and in many studies, it has been determined that samples produced by lamination show superior properties compared to solid material (Keskin *et al.* 2003; Perçin *et al.* 2009; Kasal *et al.* 2010; Küçüktüvek *et al.* 2023). There are studies on the performance of non-wood reinforcing materials in the middle layer in lamination processes (Güler and Subaşı 2012; Uzel *et al.* 2018; Karaman and Yıldırım 2019; Karaman *et al.* 2021). Studies on the use of cheap wood-based materials in the middle layer are available in the literature (Dündar *et al.* 2016). All these research works include explorations aimed at finding economical and durable uses of wood. A different production method that can be evaluated in the search for economical and durable use of wood is by using 3D printers, whose accessibility and use are rapidly becoming widespread in today's world. Fused deposition modeling (FDM), selective laser sintering (SLS), and various similar technologies are some of the 3D printing technologies. Among these, FDM technology is the most widely used method due to its simple operating logic and affordable price (Tokdemir and Altun 2022). Therefore, FDM printing technology is constantly evolving, and the printing materials play an important role in the sustainability development. Through melting thermoplastic materials with the FDM printing method, various economical and durable products can be obtained. In 3D printers working with this method, polylactic acid (PLA), acrylonitrile butadiene styrene (ABS), and polyethylene terephthalate (PET) filaments are the most commonly used polymers (Filament 2023). Among them, PLA is a fully degradable thermoplastic aliphatic polyester. PLA-based wood-plastic composites (WPC), prepared by laminating PLA with wood fibers, can reduce production costs. PLA-based wood-plastic composites (WPC) can compensate for the deficiencies in the mechanical properties of PLA while maintaining its biodegradability (Li *et al.* 2023). There are studies on PLA-based wood-plastic composites in the literature (Oporto *et al.* 2007; Pei *et al.* 2010; Petinakis *et al.* 2012; Espino-Pérez *et al.* 2013; Raquez *et al.* 2013; Meyva and Kaynak 2016; Xu *et al.* 2016; Zhang *et al.* 2017; Mokhena *et al.* 2018; Pandey *et al*. 2019; Motru *et al.* 2020; Meyva Zeybek and Kaynak 2021; Grigsby *et al.* 2022). There are also academic studies on wood filaments (Guo *et al.* 2018; Liu *et al.* 2019; Bhayana *et al.* 2021; Meyva Zeybek and Kaynak 2021; Tokdemir and Altun 2022; Zarna *et al.* 2023). In these studies, it is seen that wood is mostly used in powder or fiber forms. However, studies in which wood and plastic materials are laminated in layers with glues are quite limited (İlçe *et al.* 2015; Bekhta, *et al.* 2017; Ge *et al.* 2023; Kaya 2023).

Laminating wood materials with alternative, new, natural, economical, and inexpensive additives and investigating their physical and mechanical strength properties are considered extremely important. In this study, a special geometry was used to ensure effective adhesion of wood and PLA lamellas to each other. Based on research and development studies, it is known that 3D printing method is more economical than injection molding method in terms of processes such as mold cost, time, and preliminary preparation (Komal *et al.* 2021). A 3D printer generally is preferred to produce this geometric layer.

For these reasons, this study aimed to determine some physical and mechanical properties of laminated Scots pine (*Pinus sylvestris* L.) using PLA-containing parts produced by 3D printing in the laminated middle layers.

For this purpose, panels with filling ratios of 30%, 60%, and 90% by volume were glued with polyurethane (PU) glue between the layers of five-layer laminated wood and some mechanical and physical properties were investigated.

EXPERIMENTAL

Material

Scots pine (*Pinus sylvestris* L.) wood was preferred in this study because it is widely grown in the world and in Türkiye and is frequently used in the wood industry. Yellow pine wood used in the study was obtained from Yörüs Forest Products (Yenice/ Karabük/Türkiye). The yellow pine wood supplied as timber was cut with a saw in the direction tangential to the fibers and turned into coarse-sized veneers. The veneers were prepared in accordance with the TS 2470 (2005) standard by selecting uniform fiber samples without rot, knots, color, and density differences. Before lamination, the veneers were kept in an air conditioning cabinet at 20 ± 2 °C and 65% relative humidity until reaching constant weight in accordance with TS 2471 (2005).

Polylactic acid (PLA) is a bio-based polymer derived from natural resources and offers a considerable reduction in carbon footprint compared to petroleum-based plastics. The PLA polymer was preferred to create a plastic layer because it does not disrupt the biomaterial feature of wood and is a biodegradable material. The PLA, supplied as a black filament and had a diameter of 1.75 mm, 1.24 g/cm^3 density, 78 MPa tensile strength, 14% elongation at break, 90 MPa bending strength, and 1984 MPa bending modulus of elasticity (Hepsiburada 2023). Filaments were obtained from the website "www.hepsiburada.com" (Türkiye). The PLA layers, which constitute the other component of the laminate material, were prepared on an Xperia (Yapılıcan Machine Ind. Trade. Co., Ltd., Corum, Turkey) FDM printer. Cura (created by the makers of the Ultimaker software) software was used to create G codes and edit layer and printing parameters. The PLA layers were printed according to the parameters given in Table 1, measuring 4 × 160 × 310 mm^3. The PLA layers printed on a 3D printer are given in Fig. 1.

Table 1. PLA Layer Printing Parameters

Nozzle diameter	0.6 mm
Printing temperature	200 ºC
Print speed	60 mm/h
Layer height	0.2 mm
Number of walls	0
Fan speed	100%
Filling geometry	Cubic
Occupancy levels (volume)	30%, 60%, 90%

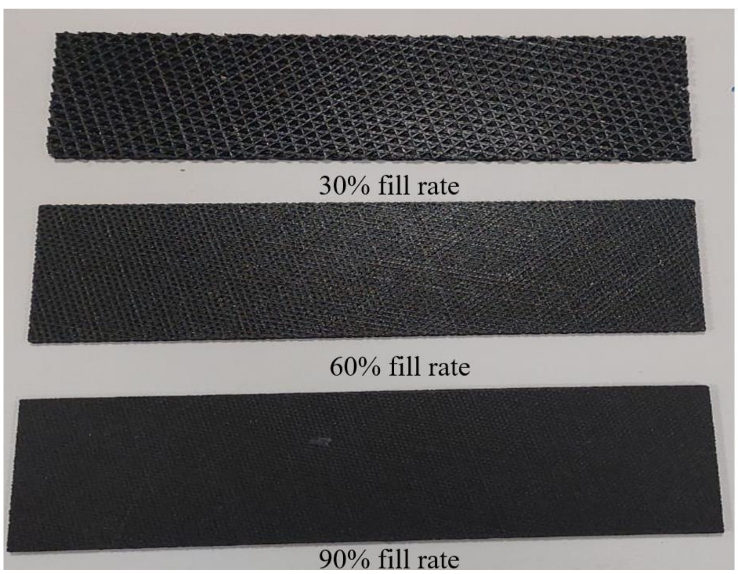

Fig. 1. PLA layers printed on a 3D printer

In this study, polyurethane glue was used to bond the layers. This glue, which is a single-component wood adhesive, cures with the moisture content of the environment and the wood. It was preferred because it is frequently used in lamination methods, is resistant to water and moisture, is easy to apply, and has high adhesion strength. Polyurethane from Akfix PA360 (Istanbul/Türkiye) was used in the study.

Lamination Process

Air dry moisture content (12%) wood veneers and printed PLA layers were laminated with an As-Metal (Maltepe, Istanbul) cold press using polyurethane glue according to the information given in Table 2. Test samples are given in Fig. 2.

Table 2. Lamination Process Parameters

Humidity of wood veneers	12 ± 2%
Bonding process room temperature	20 ± 2 ºC
Relative humidity	65 ± 2%
Adhesive	Polyurethane
Applied adhesive weight	180 ± 10 g/m^2
Press pressure and time	4 kgf/cm^2 -5 h

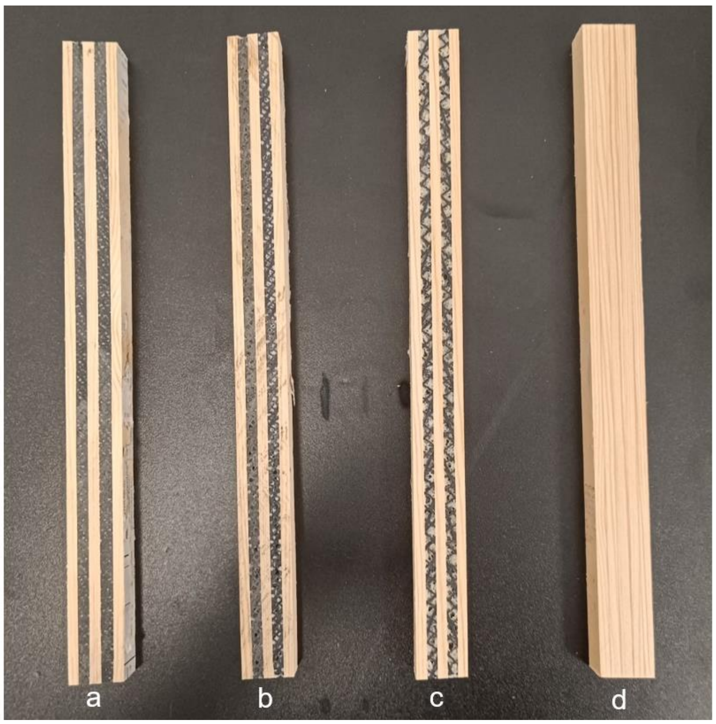

Fig. 2. (a) Sample with 90% fill rate, (b) sample with 60% fill rate, (c) sample with 30% fill rate, and (d) control group

The samples obtained after the lamination process were kept in the air-conditioning cabinet at 20 ± 2 ℃ and 65 ± 2% relative humidity for 3 weeks to equilibrate and ensure dimensional stabilization.

Characterization Methods

Determination of density

Densities of the samples were measured in accordance with the TS EN 326-1 (1999) standard, and 6 samples from each group were prepared. The prepared samples were weighed on a scale (Kern, Wien Österreich) with an accuracy of ± 0.001 g, and their dimensions were measured with a digital caliper (Instron, Istanbul/Türkiye) with an accuracy of ± 0.01 mm. In accordance with TS EN 323 (1999) standard, density was determined for each sample using Eq. 1,

$$\delta_a \text{ (g/cm}^3\text{)} = \frac{m}{a \times b \times t} \quad (1)$$

where m denotes sample weight (g), a is sample width (mm), b is sample length (mm), and t is sample thickness (mm).

Determination of swelling in thickness after immersion in water

To determine the amount of water uptake and swelling to thickness, samples were prepared using the TS EN 317 (1993) standard and equilibrated in the air-conditioned cabinet until they reached a constant weight. The samples were immersed in sterilized water and measurements were taken after 2 and 24 h. Excess water was removed with a cloth before measurements were made and then weight and size measurements were made. Water absorption rate and swelling to thickness were calculated according to Eqs. 2 and 3,

$$S_a \text{ (\%)} = \frac{m_2 - m_1}{m_1} \times 100 \quad (2)$$

$$G_t \text{ (\%)} = \frac{t_2 - t_1}{t_1} \times 100 \quad (3)$$

where m_1 represents weight of the test sample before dipping (g), m_2 is weight of the test sample after dipping (g), t_1 is the thickness of the test sample before dipping (mm), t_2 is the thickness of the test sample after dipping (mm).

Bending strength and modulus of elasticity in bending

To determine the modulus of elasticity in bending and of bending strength of the obtained laminated materials, 6 samples from each group were prepared with dimensions of 20 × 20 × 300 mm^3 in accordance with the TS EN 310 (1999) standard. Tests were performed on the Shimadzu (Kyoto, Japan) device (Fig. 3).

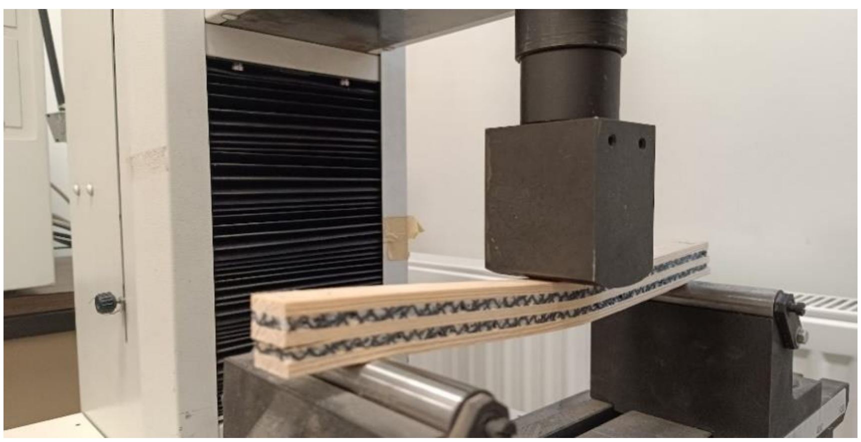

Fig. 3. Bending test of laminated samples

Bending strength and modulus of elasticity in bending were calculated according to Eqs. 4 and 5,

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

$$E \text{ (N/mm}^2\text{)} = \frac{F \times L^3}{4 \times \Delta_e \times b \times t^3} \quad (5)$$

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).

Withdrawal resistance of single screw

Withdrawal resistance of single screw laminated samples was determined in accordance with the TS EN 13446 (2005) standard. For each experimental group, 6 test samples measuring 50 × 50 × 20 mm^3 were prepared and tested. Guide holes were prepared on the test samples, and screws with 3-mm thread diameter and 60 mm length were screwed in such a way that they were buried 17 mm. During the experiment, the loading speed was set as 2 mm/min and calculated according to Eq. 6 given below (Fig. 4),

$$\sigma\ (N/mm^2) = \frac{F}{d\ x\ l} \quad (6)$$

where F is the maximum pulling force (N), d is the screw diameter (mm), and l is the embedding depth of the screw (mm).

Fig. 4. Withdrawal resistance of single screw example

Compression strength

The compression strength of laminated specimens parallel to the fibers was carried out in accordance with TS 2595 (1977). The specimens in each test group were prepared with dimensions of 20 × 20 × 20 mm^3. Before the experiment, the thickness and width of the specimens at the center points were measured and recorded with a digital caliper with an accuracy of 0.001 mm (Fig. 5). The compression strength was calculated according to Eq. 7 as shown below,

$$\sigma_b\ (N/mm^2) = \frac{F}{a \times b} \quad (7)$$

were F is the maximum force (N), a is the sample width (mm), and b is the sample length (mm).

Fig. 5. Compression strength test

Evaluation of Data

At the end of the experiments, the data were analyzed using SPSS statistical software (Version 21.0, SPSS Inc., Chicago, IL, USA). Analysis of variance (ANOVA) was used to determine whether there were significant differences between wood-plastic laminated samples. An ANOVA followed by Duncan test was performed to show significant differences ($P < 0.05$).

RESULTS AND DISCUSSION

Density

When the density of the obtained composites was analyzed, it was seen that the density increased in parallel to the PLA level (Fig. 6). This can be attributed to the increase in the amount of PLA per unit area and the higher density of the PLA used compared to Scots pine wood. In the literature, it has been shown that the use of materials with lower density than wood, such as lamellae, reduces the density of the laminated material (Celebi and Kılıç 2007; Ilçe *et al.* 2015). De Barros Lustosa *et al.* (2015) reported an increase in the density of composite boards as the amount of high-density polyethylene increased in their study. Academic studies have shown that the density of the layers used in wood composites affects the composite density. Accordingly, PLA lamellae, which have a higher density than wood, also increased the density of the laminated material.

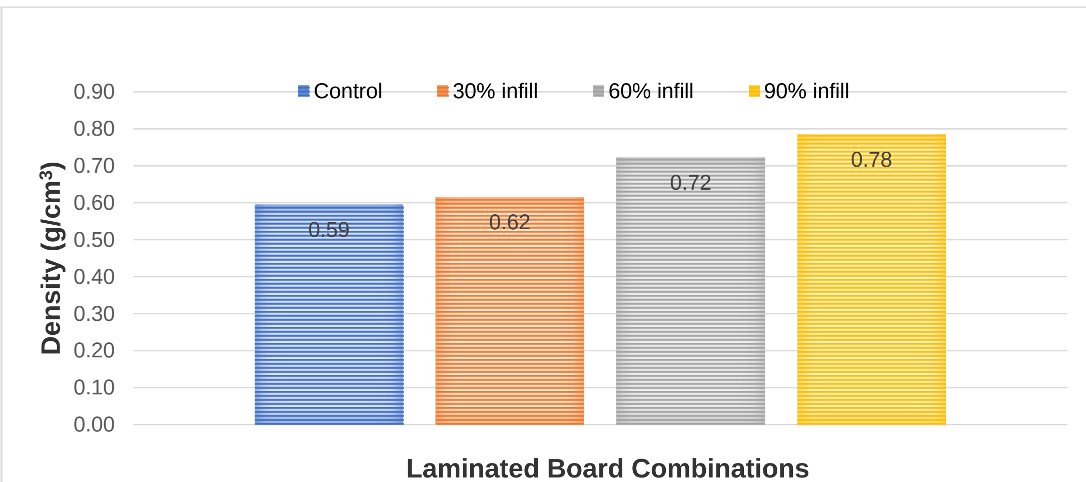

Fig. 6. Determination of densities

Further, Keskin *et al.* (2003) and Perçin (2012) stated in their studies that the densities of laminated Scots pine materials were 0.53 g/cm^3 and 0.57 g/cm^3, respectively. Therefore, when compared with literature results, the obtained densities data were consistent.

Water Uptake and Thickness Swelling Rate

It was seen that the amount of water absorbed by the laminated samples increased as the immersion time increased, and the amount of water absorption decreased as the filling rate in the lamellae increased (Fig. 7).

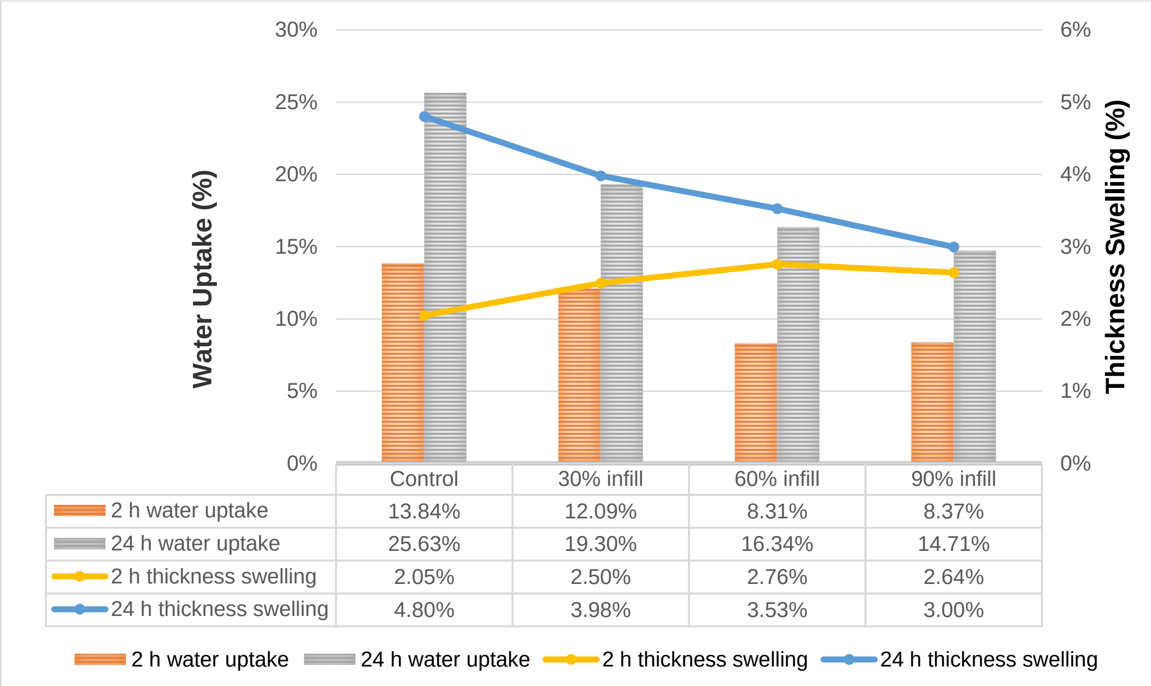

Fig. 7. Water uptake and thickness swelling results

When the thickness swelling values were examined, it was apparent that as the content of PLA lamellae increased after 2 h of water immersion, the amount of thickness swelling also increased. It is thought that the gaps between the PLA lamellae allow water to move easily and reach the wood quickly and swell the wood layers, whereas in the control samples, the progress of water uptake was slower. In 24-h water immersion values, it was found that as the amount of PLA lamellae increased, the thickness swelling ratio decreased. The fact that PLA is hydrophobic compared to wood shows that it is effective in both water absorption and thickness swelling values. Tokdemir (2022) reported the water absorption values of square- and cylindrical-shaped plastic parts with 100% filling rate, printed with a 3D printer, as 0.82% and 0.84%, respectively, and the thickness swelling values as 0.61% and 0.64%, respectively. This explains the decrease in water uptake and thickness swelling values as the occupancy level increased. Additionally, İlçe *et al.* (2015) reported in their study that polycarbonate layers in wood-plastic laminate material reduced the 2-h and 24-h water absorption and thickness swelling values.

Bending Strength and Modulus of Elasticity in Bending

Results of variance analysis of the bending strength are given in Table 3.

Table 3. Results of Variance Analysis of the Bending Strength

Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	3903.414[a]	3	1301.138	788.194	0.001
Intercept	171915.690	1	171915.690	104141.834	0.001
Filling Rate	3903.414	3	1301.138	788.194	0.001
Error	33.016	20	1.651		
Total	175852.120	24			
Corrected Total	3936.430	23			
a: R Squared = 0.992 (Adjusted R Squared = 0.990)					

According to the results of the analysis of variance, a statistically significant difference was found relative to the occupancy levels between the groups ($p < 0.05$). To further investigate the differences between the groups, Duncan's test was applied, and the significance levels between the groups were determined (Table 4). According to the results of Duncan's test, statistically significant differences were observed between the groups. Results of variance analysis of the modulus of elasticity are given in Table 5.

Table 4. Duncan's Test Results of the Bending Strength

	Filling Rate	Averages and Homogeneity Groups			
Duncan[a,b]	30%	68.6 (a)			
	60%		81.8 (b)		
	90%			83.9 (c)	
	Control				104.3 (d)

Table 5. Results of Variance Analysis of the Modulus of Elasticity

Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	89544310.760 [a]	3	29848103.587	404.531	0.001
Intercept	2438120380.843	1	2438120380.843	33043.816	0.001
Filling Rate	89544310.760	3	29848103.587	404.531	0.001
Error	1475689.357	20	73784.468		
Total	2529140380.959	24			
Corrected Total	91020000.117	23			
a: R Squared = 0.984 (Adjusted R Squared = 0.981)					

According to the results of the analysis of variance, a statistically significant difference was found in the levels of occupancy between the groups ($p < 0.05$).

To further investigate the differences between the groups, Duncan's test was applied, and the significance levels between the groups were determined (Table 6). According to the results of Duncan's test, statistically significant differences were observed between the groups.

Table 6. Duncan's Test Results of the Elasticity Modulus

	Filling Rate	Averages and Homogeneity Groups			
Duncan[a,b]	30%	7339.32 (a)			
	60%		9569.15 (b)		
	Control			10730.49 (c)	
	90%				12672.78 (d)

When the bending tests of laminated samples were examined, samples with 30%, 60%, and 90% printing fill rate decreased by 34.2%, 21.6%, and 19.5%, respectively, compared to the control samples (Fig. 8).

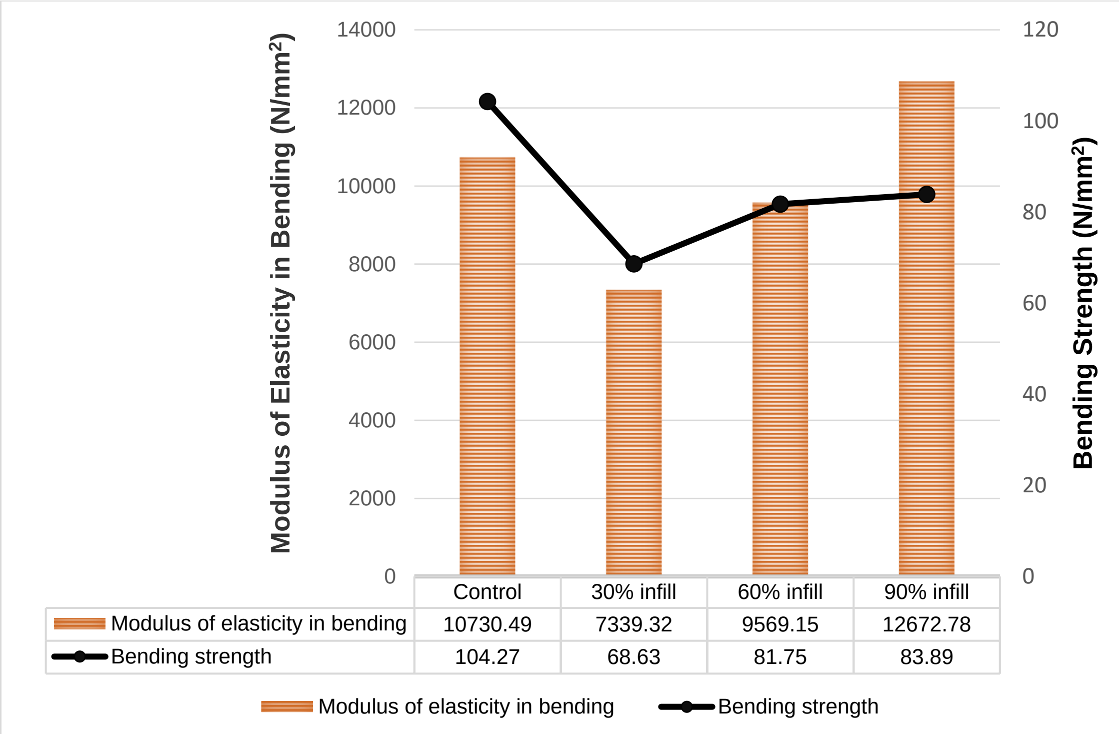

Fig. 8. Bending strength and modulus of elasticity in bending values

When the elasticity modulus values in bending were examined, samples with 30% and 60% filling ratio decreased 31.6% and 10.8%, and samples with 90% filling ratio increased 18.1% compared to the control samples. Parameters, such as polymer type, nozzle diameter, number of walls, filling geometry, and filling ratio, seriously affect the mechanical properties of samples printed on a 3D printer. Lubombo and Huneault (2018) reported in their study that with the same filling geometry and filling ratio, the number of perimeter shells increased the strength 84%. Wang *et al*. (2024) reported that the bending strength of 3D-printed PLA samples increased as the lower and upper shell thickness increased.

It is thought that the lamellae used in laminated samples consist only of filling geometry; that is, the absence of both the upper/lower layers and the surrounding shell, causing the bending strength and bending modulus values to seriously decrease.

Withdrawal Resistance of Single Screw

Results of variance analysis on withdrawal resistance of screws is given Table 7.

Table 7. Results of Variance Analysis on Withdrawal Resistance of Screws

Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	1044.935[a]	3	348.312	792.374	0.001
Intercept	17208.828	1	17208.828	39148.342	0.001
Filling Rate	1044.935	3	348.312	792.374	0.001
Error	8.792	20	0.440		
Total	18262.555	24			
Corrected Total	1053.727	23			
a: R Squared = 0.992 (Adjusted R Squared = 0.990)					

According to the results of the analysis of variance, a statistically significant difference was found in the occupancy rates between the groups ($p < 0.05$).

To further investigate the differences between the groups, Duncan's test was applied, and the significance levels between the groups were determined (Table 8). According to the results of Duncan's test, statistically significant differences were observed between the groups.

Table 8. Duncan's Test Results of the Withdrawal Resistance of Screws

Duncan[a,b]	Filling Rate	Averages and Homogeneity Groups			
	Control	19.55 (a)			
	30%		20.87 (b)		
	60%			32.73 (c)	
	90%				33.96 (d)

When the withdrawal resistances of single screw of laminated samples were examined, it was seen that as the printing content of PLA lamellae increased, the screw holding resistance also increased. Compared to the control samples, it was apparent that the samples with 30%, 60%, and 90% printing fill level increased 7.2%, 68.2%, and 74.5%, respectively (Fig. 9).

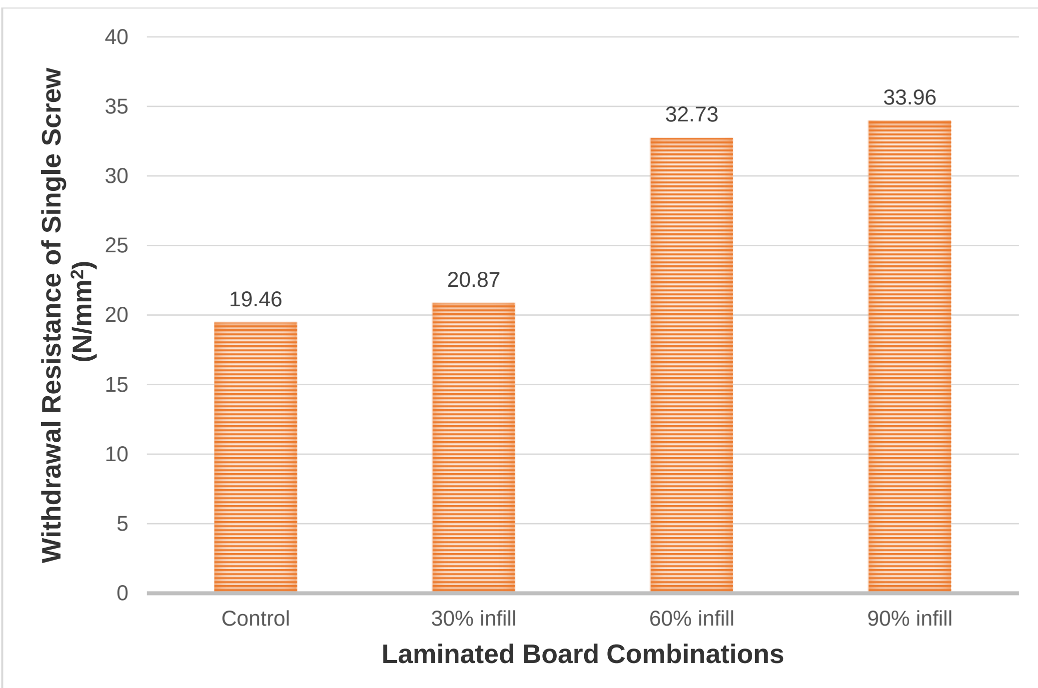

Fig. 9. Withdrawal resistance of single screw

Kilic *et al.* (2006) reported that screw holding resistance varies depending on the moisture content of the wood, fiber direction, surface coating of the material, density, and screw properties. The density of wood plays an important role in determining its mechanical strength (Romagnoli 2014). It is thought that as the lamellae increase the specific gravity of the laminated material, the screw tensile strength also increases.

Compression Strength

Results of variance analysis of the compression strength are given in Table 9.

Table 9. Results of Variance Analysis of the Compression Strength

Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	1377.554a	3	459.185	286.189	0,001
Intercept	53683.609	1	53683.609	33458.571	0,001
Filling Rate	1377.554	3	459.185	286.189	0,001
Error	32.090	20	1.604		
Total	55093.252	24			
Corrected Total	1409.643	23			
a: R Squared = 0.977 (Adjusted R Squared = 0.974)					

According to the results of the analysis of variance a statistically significant difference was found in the occupancy rates between the groups ($p < 0.05$).

To further investigate the differences between the groups, Duncan's test was applied, and the significance levels between the groups were determined (Table 10). The results of Duncan's test indicated that no statistically significant difference was observed between the control group and the group with 90% PLA content, while statistically significant differences were found between the groups with 30% and 60% PLA filling.

Table 10. Duncan's Test Results of the Compression Strength

	Filling Rate	Averages and Homogeneity Groups		
Duncan[a,b]	30%	35.97 (a)		
	60%		44.87 (b)	
	90%			53.49 (c)
	Control			54.85 (c)

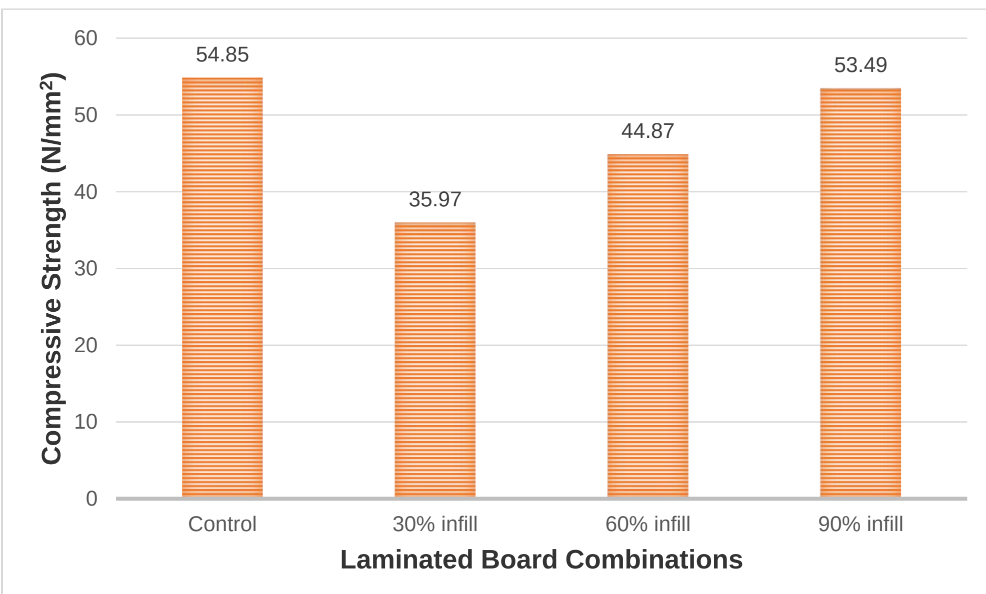

Fig. 10. Compression strength

When the compression strength values were examined, the values of the PLA lamellar samples decreased compared to the control samples, and the pressure resistance of the plate lamellar samples were increased with increasing PLA content. Pressure resistance values decreased 34.4%, 18.2%, and 2.5% at 30%, 60%, and 90% PLA content, respectively (Fig. 10).

Ayrılmış *et al.* (2020), stated in their study that adding the top and bottom surface layers to the printed samples significantly increased the compressive strength. Pernet *et al.* (2022) reported that the highest compressive resistance values were seen in samples with 100% filling and cross-filling geometries.

It is thought that the pressure resistance decreases because the gaps in the filling geometry are not distributed homogeneously compared to wood.

CONCLUSIONS

1. The study revealed that increasing the PLA content in the PLA lamellae resulted in higher specific gravities of the laminated samples while reducing their water absorption values.
2. Superior physical and mechanical properties can be expected to be achieved by utilizing plastic layers with varying filling geometries, different polymer types, and optimized printing parameters.
3. Lamellae with favorable filling geometries are anticipated to significantly enhance the composite material's mechanical and physical properties, offering superior performance and functionality.
4. The use of different types of adhesives may improve the physical and mechanical properties of the composite material.
5. Investigating the effect of plastic layers on the mechanical and physical properties of different wood species by diversifying the wood material type will provide a better understanding of the subject.

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