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Investigation of adhesive bonding strength of wood added PLA materials

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Abstract

In this study, plates were produced from wood added polylactic acid (PLA) based materials using 3D printers, which is one of the additive manufacturing methods. After applying mechanical abrasion (sanding), which is one of the surface preparation methods, to the surfaces of the produced plates, the adhesive bonding process was carried out. Adhesive joints were made using the additive-free and additive formula of 3 different commercial adhesives (Araldite 2015, Loctite 9466, and PVA wood glue). In commercial adhesives, 5% hazelnut shell powder in 45 µm dimensions was added and the bonds were joined by forming an additive adhesive formula. The obtained joints were subjected to the tensile test and their mechanical properties were examined. The experiments were repeated for plates cut from Scotch pine wood (*Pinus Sylvestris* Lipsky) with the same adhesives and the values found were compared with the results of the joints made with wood added PLA. With the results obtained, the strengths of pure (plain) and filled (modified) joints were evaluated. It has been observed that Loctite 9466 adhesive provides the highest shear strength in joining wood added PLA and Scotch pine materials with adhesive. The aim of this study is to investigate the adhesion performance of PLA composites, a high-tech material, when used in the furniture and wood sector.

Keywords: Additive manufacturing, Adhesive bonding, Mechanical properties, PLA wood, Scotch pine

Odun katkılı PLA malzemelerinin yapışma dayanımlarının incelenmesi

Öz

Bu çalışmada; eklemeli imalat yöntemlerinden biri olan 3B yazıcılar kullanılarak ahşap katkılı PLA esaslı malzemelerden plakalar üretilmiştir. Üretilen plakaların yüzeylerine, yüzey hazırlama yöntemlerinden biri olan mekanik aşındırma işlemi (zımparalama) uygulandıktan sonra yapıştırma işlemi gerçekleştirilmiştir. Yapıştırma bağlantıları, 3 farklı ticari yapıştırıcının (Araldite 2015, Loctite 9466 ve PVA ahşap tutkalı) katkısız ve katkılı formülü kullanılarak yapılmıştır. Ticari yapıştırıcıların içerisine 45µm boyutlarında %5 oranında fındık kabuğu tozu katılmış ve katkılı yapıştırıcı formülü oluşturularak bağlantılar birleştirilmiştir. Elde edilen bağlantılar, çekme testine tabi tutularak mekanik özellikleri incelenmiştir. Deneyler, aynı yapıştırıcılarla Sarıçam ağacından (*Pinus Sylvestris* Lipsky) kesilmiş plakalar için tekrar edilmiş ve bulunan değerler ahşap katkılı PLA ile yapılan bağlantıların sonuçlarıyla karşılaştırılmıştır. Ortaya çıkan sonuçlar ile katkısız (sade) ve katkılı (modifiye edilmiş) bağlantıların dayanımları değerlendirilmiştir. Ahşap katkılı PLA ve Sarıçam malzemelerin yapıştırıcı ile birleştirilmesinde en yüksek yapışma dayanımını Loctite 9466 yapıştırıcı ile sağladığı görülmüştür. Bu çalışmanın amacı, yüksek teknoloji ürünü bir malzeme olan PLA kompozitlerin mobilya ve ahşap sektöründe kullanılması durumunda, yapışma performansının incelenmesidir.

Anahtar kelimeler: Ahşap katkılı PLA, Eklemeli imalat, Mekanik özellikler, Sarıçam, Yapıştırma işlemi

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1 Introduction

Limited global resources and increasing energy and production costs have led people towards sustainable consumption. On the one hand, the desire for environmental friendly production and on the other hand, the expectation of low cost has created a need for innovative materials. As a result, the production of industrial products using composite materials, which provide high strength and lightness, is supported. Composites are categorized as metal, ceramic, and plastic composites based on the matrix material they are composed of, and they have found various applications in different industries. Composite materials are produced based on improving material properties to replace traditional materials. For example, wood-plastic composites, which belong to the group of plastic composite materials, are more resistant to moisture than solid wood due to the addition of wood flour/fiber, and they have higher mechanical strength and acoustic properties (Aysal, 2014). The use of wood-plastic materials, including patents, began in the 1960s and continues to the present day (Rowell, 2006). In addition, wood-filled PLA filaments, which are wood-plastic composites, are used as consumables in 3D printers, which are popular additive manufacturing technologies. As a result, the use of wood-plastic composites has further increased in recent years. These materials are made by combining plastic and wood in granule or powdered form at appropriate quantities and temperatures. Wood composites offer advantages such as cost-effectiveness, ease of access to raw materials, and eco-friendliness compared to pure plastic materials. Compared to wood, wood composites offer dimensional stability, a variety of shapes and color options, high strength, low maintenance and repair requirements, and the ability to be derived from recyclable materials (Rowell et al., 1993; Chen, 2009; Karakuş et al., 2014). Wood materials are the oldest building materials widely used in various products for both indoor and outdoor spaces (Raşit, 2019). In addition, wood composites can lead to significant commercial opportunities in the forest products industry. Because these waste materials (waste wood fibers, wood particles, or wood flour) are used as reinforcement materials in composite material production. A literature review reveals various research studies on the production and use of wood-plastic composites. Altuntaş and Arıkan (2022) examined the changes in the mechanical, thermal, and morphological properties of wood-plastic composite materials by adding perlite, which possesses volcanic glass properties. The experimental results showed that increased perlite content increased flexural strength, tensile strength, modulus of elasticity, and hardness values.

Bal (2022) produced wood-plastic composite sheets by adding Scotch pine wood flour in weights of 0%, 10%, 20%, 30%, and 40% with low-density polyethylene. These composite materials were subjected to various tests. It was noted that as the amount of wood flour in the composite materials increased, there was an increase in density, flexural strength, and modulus of elasticity but a decrease in elongation. In their study, Özmen et al. (2014) aimed to find an alternative to wood flour by using fibreboard waste flour as an additive in polypropylene to produce wood-plastic composites. They used fibreboard waste flour in proportions of 10% to 50%. While an increase in the additive content resulted in a decrease in tensile and impact strength, it was observed that the elastic modulus and flexural strength of the composite materials were higher compared to polyolefin-based plastic lumber. When considering the studies on this subject, it becomes apparent that there is potential for the using and production of wood-plastic composites instead of wood materials in the forest industry and furniture sector. It is known that the using of adhesives or mechanical fasteners is quite common for combining wood and wood-plastic composite materials in the furniture sector. However, it is claimed that adhesive bonding is the most effective method for connecting the joints of furniture and other wood products, and assembly errors are generally attributed to adhesive errors (Smardzewski, 2002). The adhesive bonding process is applied to achieve a pleasing appearance, smoother stress distribution, and flat surfaces without the need for additional mechanical elements. In cases where an aesthetic appearance is desired in joints, adhesive bonding is often preferred over mechanical joining methods. The adhesive bonding process involves at least two materials being joined and an adhesive substance. Proper surface preparation of the parts to be bonded and knowing the value of the applied force on the joint area after assembly are essential for the seamless operation of these connections in their intended use. The surface preparation for adhesive bonding does not affect the strength of the bonded material but will affect the bond strength. Additionally, the adhesive, the parts to be bonded, and the process parameters will affect the quality of the bonding. Therefore, the bond strength is expected to vary depending on the selection of an adhesive suitable for the material group. While adhesives are commonly used in their pure form (without additives) in conventional applications, adding various fillers in different sizes and types to enhance the bond strength is also considered an option in academic studies (Valášek and Müller, 2016; Nemati Giv et al., 2018; Delzendehrooy et al., 2020; Gonçalves et al., 2022; Anaç et al., 2022; Anaç and Doğan, 2023). Karaman et al. (2017) examined the effects of adhesive surfaces on diagonal tensile and compressive strengths for "L" type furniture corner joints using different types of adhesives such as polyvinyl acetate (PVAc-D4), polyurethane (PU-D4), and epoxy. The highest tensile strength value was measured in the epoxy adhesive, while the lowest tensile strength value was recorded in the polyurethane adhesive.

Karaman et al. (2019) conducted a study on the tensile strength of joints made with polyurethane (PU-D4) and polyvinyl acetate (PVAc-D4) adhesives using ash (*Fraxinus excelsior* Lipsky), chestnut (*Castanea sativa* Mill.), and oak (*Quercus petraea* Lieble) timbers. The highest strength was observed in samples prepared with ash dowel and polyvinyl acetate adhesive, while the lowest strength was found in samples made with chestnut dowel and polyurethane adhesive.

Another (Perçin and Uzun, 2014) conducted a study to examine the bonding strength of PVAc-D4 adhesive in oriental beech (*Fagus orientalis* L.), oak (*Quercus petraea* L.), Scotch pine (*Pinus sylvestris* L.), and poplar (*Populus nigra* L.) woods after applying extended heat treatment. Esen and Özcan (2012) evaluated the bonding strength of oak (*Quercus petraea* L.) wood samples using polyurethane (PUR), phenol-formaldehyde (FF), melamine-urea-formaldehyde (MUF), and melamine-formaldehyde (MF) adhesives.

The forestry industry has various types of trees with different characteristics. Factors such as production/manufacturing location, suitability for mass production, temperature conditions, appropriate heat treatment, and cost can limit the use of wood in various applications (Söğütlü and Döngel, 2007). Nowadays, rapidly developing technology has enabled the use of new materials in furniture and decoration works (Narlıoğlu, 2021). Thanks to the production of wood plastic composite products with 3D printers, it is thought that the traditional wood production process steps will be reduced and will help to conserve raw material resources.

The aim of this study is to evaluate the bond joint strengths of with and without additives bond joints made with 3D printed wood-added PLA sheets, and without additives bond joints made with wood cut from Scotch pine (*Pinus sylvestris* Lipsky). For this purpose, the joint strengths are evaluated by using tensile test after the bonding process.

2 Material and Method

In the conducted study, wood-filled PLA (PLA Wood) and Scotch pine plates were joined using different adhesives, and their adhesive bonding performances were examined. Araldite 2015, Loctite 9466, and PVA wood glue (Adhesive durability class; D3) were used as adhesives. Additionally, for the bonding of PLA Wood plates, the adhesives were modified with hazelnut shell powder (5% by weight) to investigate the effect of organic filler. Figure 1 provides the flowchart of the experimental design. The printing of PLA Wood bonding plates is shown in Figure 1a, while the Scotch pine bonding plates are depicted in Figure 1b. After the preparation of bonding plates, PLA Wood plates were bonded with three different adhesives, both with and without additives (plain and 5% hazelnut shell powder), while Scotch pine plates were bonded without additives (Figure 1c and Figure 1d). Figure 1e and Figure 1f show the specimens after bonding. Tensile tests were conducted to evaluate the bonding strength, and shear strengths were calculated accordingly.

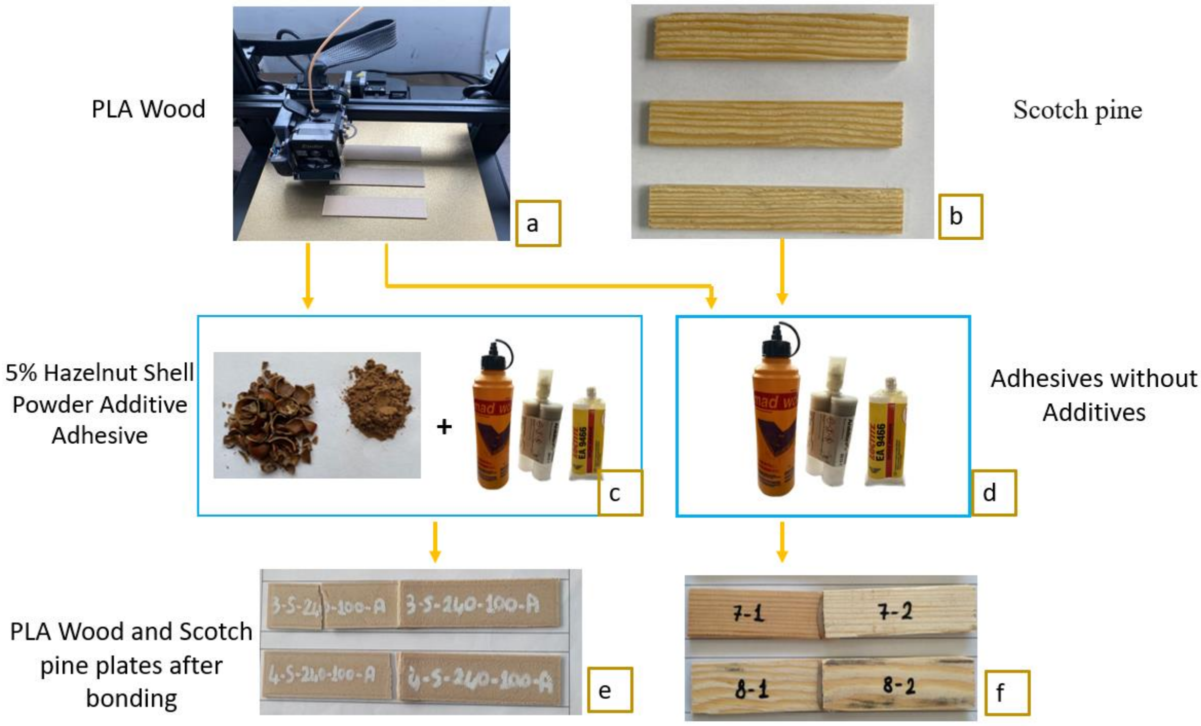


Figure 1. Flowchart of the adhesive bonding process

2.1. Properties of the materials

This study utilized PLA wood filament and Scotch pine (*Pinus sylvestris* Lipsky). Scotch pine is preferred in the woodworking industry due to its abundant availability within the country's borders and its suitability for outdoor applications in various environmental conditions (Keskin and Bülbül, 2019; Keskin and Taner, 2019). The Scotch pine bonding plates were cut into the desired dimensions (25×100×4 mm) and then dried in an oven at 60 °C for 24 hours to remove moisture. PLA material is one of the most commonly used filaments in 3D printing due to its ease of printing, suitability for producing detailed products, and not require a heated bed. Additionally, PLA is biodegradable and a non-toxic thermoplastic that is safe for human health and environmentally friendly, as it contains no harmful substances (Kyutoku et al., 2019; Mahir and Erdoğdu, 2020). Carbon fiber, wood fibers, and other fillers can also impart different properties to PLA. PLA Wood is a composite PLA material reinforced with 30% wood fibers/wood powder (Ayrilmis et al., 2019). PLA Wood is preferred in architectural models, household products, and aesthetically desired items due to its wood-like appearance and texture. Compared to PLA, PLA Wood offers higher temperature resistance and a matte wood finish. The mechanical and technical properties of PLA Wood are provided in Table 1.

Table 1. Mechanical and technical properties of PLA Wood (Filameon)

Diameter (mm)	1.75
Brand	Filameon
Color	Light brown
Tensile Strength (MPa)	47
Melting Point (°C)	200-225
Density (g/cm^3)	1.13

2.2. Preparation of samples

In the study, a 1.75 mm diameter PLA Wood filament was printed using an Ender 3 S1 printer, which operates on fused deposition modeling principles. The prints were made with 100% infill, a nozzle temperature of 220 °C, a bed temperature of 65 °C, a printing speed of 50 mm/s, and XYZ printing direction. The printing parameters directly affect the mechanical properties of the product. Therefore, to determine the mechanical properties of PLA Wood plates, a tensile bar (Figure 2a) according to ASTM D 638 (2022) standards and 25×100×1.5 mm plates for adhesive bonds (Figure 2b) were printed. Solid models were created using the commercial software solidworks, and the necessary codes for the Ender 3 S1 were generated using Ultimaker Cura software.

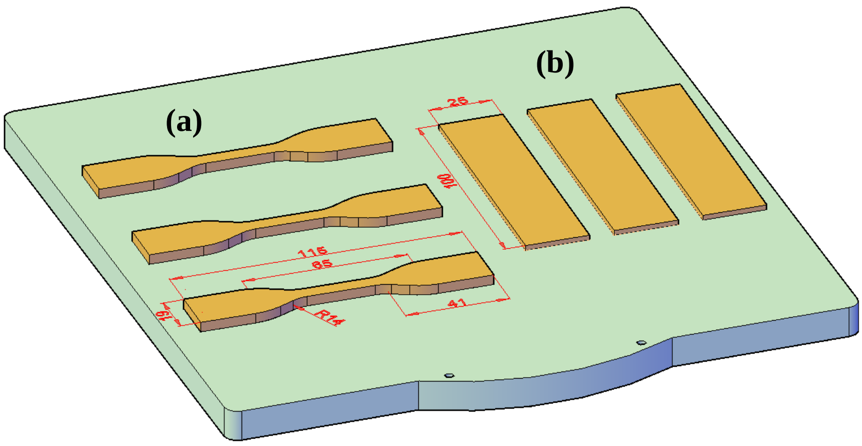


Figure 2. Tensile specimens and bonding plates dimensions (mm)

2.3. Adhesive bonding process

The schematic view of the single lap joint is given in Figure 3. For the examination of the adhesive bonding properties of PLA Wood and Scotch pine, three different commercial adhesives (Araldite 2015, Loctite 9466, and PVA wood glue) were used in both unmodified and with hazelnut shell powder-modified, and the bonding was achieved by clamping with metal clips. The experimental design is presented in Table 2. Before the adhesive bonding process in PLA Wood plates, the surfaces were roughened using silicon carbide (240 SiC) sandpaper to enhance the low surface energy of the plastic-based material. In all plates, sanding was performed perpendicular and horizontally to the part axis in the bonding area. The sanding process covered the entire bonding surface. After the sanding process, the surfaces were cleaned with air to remove dust, the bonding process was carried out, and the bonding area was secured with metal clips and left to cure at room temperature for 24 hours according to the manufacturer's recommendation.

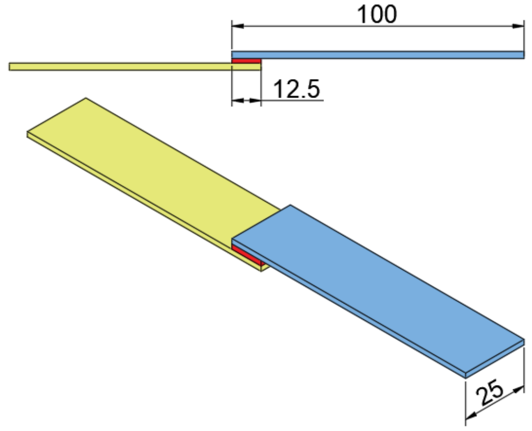


Figure 3. Single lap joint (mm)

Table 2. PLA Wood and Scotch pine bonding experimental design

Material	Adhesive	Additives	Surface Preparation	Compression Type
PLA Wood	Araldite 2015	No additives	240 SiC	Metal Clips
	Loctite 9466	No additives		
	PVA Wood Glue	No additives		
PLA Wood	Araldite 2015	5% Hazelnut Shell Powder		
	Loctite 9466	5% Hazelnut Shell Powder		
	PVA Wood Glue	5% Hazelnut Shell Powder		
Scotch pine	Araldite 2015	No additives		
	Loctite 9466	No additives		
	PVA Wood Glue	No additives		

Araldite 2015 is a versatile, room temperature-curing, high-strength paste adhesive that is a two-component epoxy-based adhesive. It is widely used for adhesive bonding various materials, including carbon fiber and glass fiber composites, as well as different surfaces. It also offers high corrosion resistance. Loctite EA 9466 is a reinforced 2K-epoxy adhesive with high strength and a long working time, suitable for bonding metals, ceramics, and most plastics. The mechanical properties of Araldite 2015 and Loctite 9466 adhesives are provided in Table 3. On the other hand, PVA wood glue is a single-component adhesive that is water-resistant and suitable for bonding wood. It exhibits high elasticity and provides high strength with normal clamping pressure.

Table 3. Araldite 2015 and Loctite 9466 adhesive mechanical properties

	Araldite 2015 (Huntsman)	Loctite 9466 (Henkel)
Elasticity modulus(MPa)	1850	1718
Tensile strength (MPa)	21.6	32
Poisson's ratio	0.33	0.33
Yield strength (MPa)	12.6	-
Elongation (%)	4.2	3
Hardness (Shore D)	-	60

They were placed in a drying oven at 120 °C to remove moisture from hazelnut shells for 6 hours. Afterward, they were ground using a ring mill. The obtained powder was then passed through sieve sets to classify it into different particle sizes, and 45 µm powder was obtained to be added to the adhesive. The stages of obtaining hazelnut shell powder are shown in Figure 4.

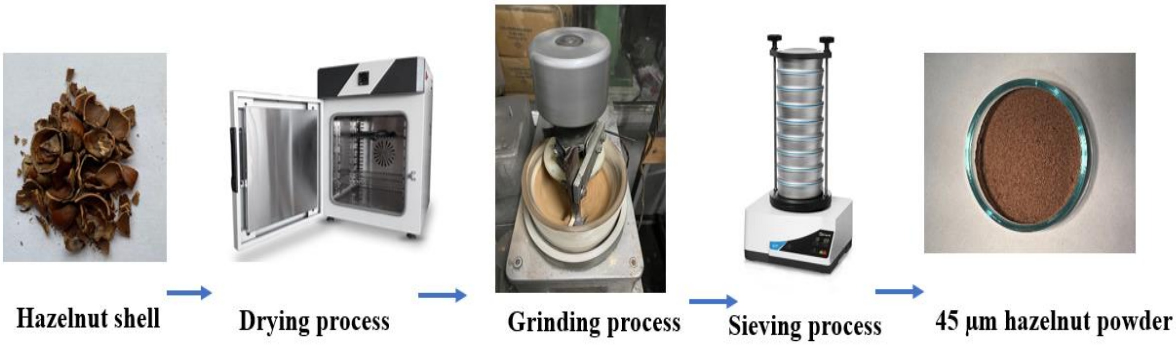


Figure 4. Hazelnut powder grinding process sequence

2.4. Determination of mechanical properties

The prepared tensile specimens ASTM D 638 (2022) and adhesive bonding plates were tested using a WDW-5 model universal testing machine with a capacity of 5 kN at a crosshead speed of 2 mm/min and at room temperature. The experiments were conducted with 5 repetitions, and the averages were taken. Surface images were captured after the tensile test to determine the type of damage in the bonding joints.

A LOYKA D-type Shore hardness durometer was used for hardness measurements of PLA Wood, and measurements were performed per ASTM D2240 (2021) standard. Hardness measurements were conducted with 5 repetitions, and the averages were taken. Lastly, mechanical surface preparation was performed using 240 SiC sandpaper before adhesive bonding. Surface roughness values of the materials before and after surface preparation were measured and evaluated using a Mitutoyo SJ-301 Profilometer.

3. Results and Discussion

3.1. Mechanical properties of materials

The highest tensile strength (47.49 MPa) was obtained from the samples prepared according to the defined experimental parameters of Scotch pine. In comparison, the lower tensile strength (12.40 MPa) was observed in the PLA Wood with wood additive (Figure 5).

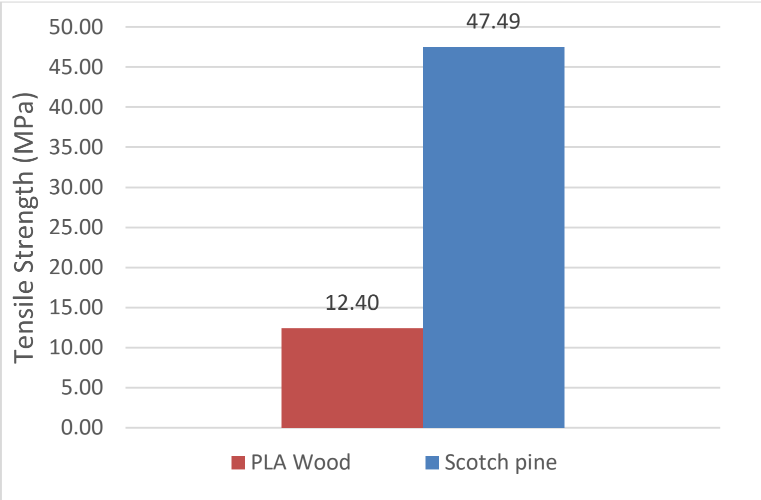


Figure 5. Average tensile strength values of test specimens

The surface roughness values measured before and after surface preparation in the bonding area for the adhesive process are given in Table 4. Due to the natural (organic) nature of Scotch pine wood material, the surface roughness is higher than that of the PLA Wood plates. Scotch pine has fibrous structures, and the surfaces have been sanded to make them smoother and flatter. Additionally, after sanding, the roughness values of both materials were found to be similar to each other.

Table 4. Surface roughness measurement values (Ra)

	PLA Wood	Scotch pine
Before sanding (µm)	3.10	4.27
After sanding (µm)	1.01	1.85

The hardness measurements of PLA Wood plates were conducted, and an average value of 71.6 Shore D was obtained. On the other hand, the hardness value of Scotch pine wood was obtained from the literature. The Brinell hardness values for Scotch pine samples were determined as 3.338 kp/mm^2 parallel to the grains, 1.772 kp/mm^2 perpendicular to the grains, and 1.519 kp/mm^2 in the radial direction (Ulusoy et al., 2017).

3.2.Adhesive bonded joint strengths

Figure 6 presents the shear strengths of adhesive joints made with three different adhesives for wood-filled PLA plates, both with and without additives. As observed in Figure 6, the highest strength was obtained with Loctite 9466, while the lowest strength was measured in the joints using additive PVA glue. The addition of powder resulted in a decrease in the bonding strength for all three adhesives. The amount of strength reduction varied slightly from the strength value of the corresponding adhesive without additives.

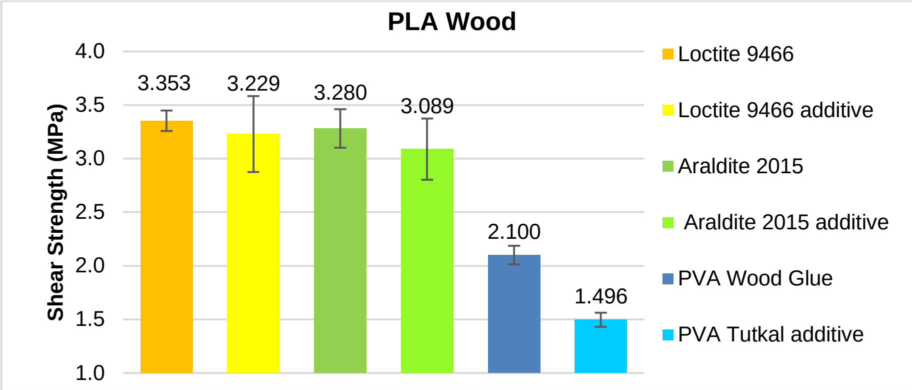


Figure 6. Variation of shear strength of PLA Wood joints according to adhesive type

Figure 7 illustrates the shear strengths of adhesive joints made with three different adhesives for Scotch pine. The highest shear strength of 11.677 MPa was achieved with Loctite 9466, while the lowest shear strength was measured in the joints using PVA glue. In the case of Scotch pine wood materials, the adhesives were used without any additives. However, the strength was lower when testing the adhesive joints with wood-filled PLA material and additives. Therefore, no powder additives were used to bond pine wood materials.

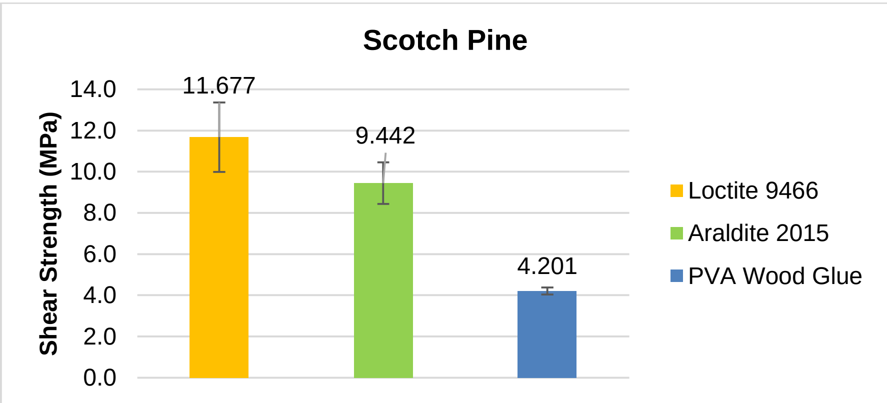
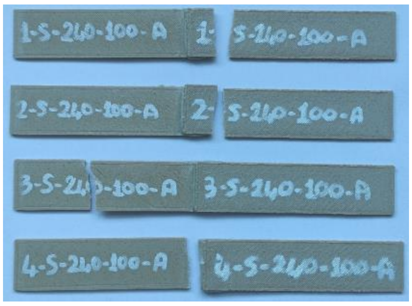


Figure 7. Shear strength of Scotch pine joints according to adhesive type

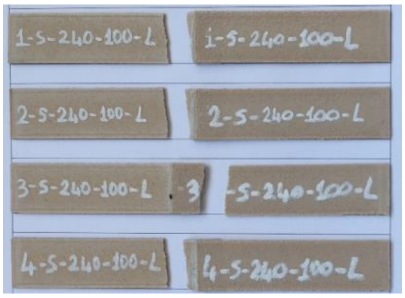
3.3.Joint fracture surfaces

Figure 8 shows the fracture surfaces of adhesive joints made with PLA Wood and pine wood materials. In the case of PLA Wood materials, fracture occurred in the primary material (adherend) in adhesive joints using pure Araldite 2015 and Loctite 9466 adhesives, while adhesive failure was observed in joints using pure PVA wood glue. In the powder-added bonding process of PLA Wood materials, the rupture occurred at the bonding zone in Loctite 9466, at the base material in Araldite 2015, and at the bonding (interface) surface in joints using PVA wood glue.

Regarding Scotch pine material, the fracture patterns slightly differed but generally involved detachment from the overlap region. In Loctite 9466, a fracture occurred in the primary material (adherend) close to the overlap region, combining adhesive and adherend (substract) failures; for PVA wood glue, predominantly adhesive failures were observed. Araldite 2015 exhibited mostly adhesive failures upon visual examination.



PLA Wood- Araldite 2015- no additives



PLA Wood- Loctite 9466- no additives

PLA Wood- PVA wood glue - no additives

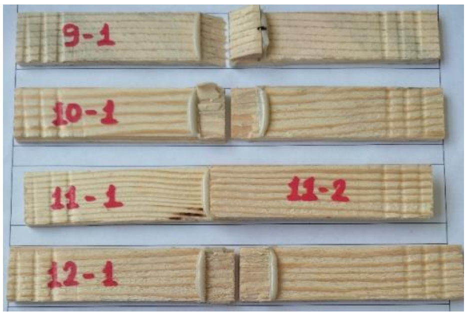
PLA Wood- Araldite 2015-5% Hazelnut Shell Powder

PLA Wood- Loctite 9466-5% Hazelnut Shell Powder

PLA Wood- PVA wood glue - 5% Hazelnut Shell

Scotch pine wood- PVA wood glue - no additives

Scotch pine wood- Loctite 9466- no additives



Scotch pine wood- Araldite 2015- no additives

Figure 8. Rupture surfaces of bonded joint after tensile test

4. Conclusions

According to the data obtained at the end of the laboratory studies; the following results can be said;

- It is evident that Scotch pine material is strengthen than wood-filled PLA. However, considering the cost-strength balance, wood-filled PLA material can be preferred.
- Both wood-filled PLA and Scotch pine wood materials exhibited better adhesion properties with Loctite 9466 adhesive compared to the other two adhesives.
- The addition of powder filler to the adhesive in the bonding process of wood-filled PLA material was found to reduce the bond strength.
- Despite its low strength, the affordability of PVA woodworking glue provides an advantage in applications where high strength is not required.
- Although it is seen that wood plastic composite materials need to be developed in many ways, the limited resources of wood materials and the ease of producing small and complex parts and geometric patterns in 3D printing suggest that these materials have the potential to be used more.

Author Contributions

Duygu Karabağ: Laboratory work and experiments, writing the article, **Muhammet Ali Tekkanat**: Laboratory work and experiments, writing the article, **Nergizhan Anaç:** Determining the research topic, laboratory work and experiments, data analysis and interpretation, writing and editing the article, **Oğuz Koçar**: Preparation of samples, laboratory work and experiments, interpretation of results, writing and editing the article.

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Conflict of interest statement

The authors declare no conflict of interest.

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