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Effects of annealing temperature and duration on mechanical properties of PLA plastics produced by 3D Printing

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Abstract: This study aims to investigate the effect of annealing temperature and duration on the mechanical properties of PLA (polylactic acid) plastics produced by a 3D (three-dimensional) printer. For this purpose, PLA samples were annealed at 70 °C, 85 °C, and 100 °C temperatures and for 30, 60, and 90-minute durations. As a result of the study, it was shown that the annealing process has a significant effect on the mechanical properties of PLA plastics. Compared to the control sample, an increase of 48% in tensile stress, 78% in the modulus of elasticity, 28% in Shore D hardness value, and 41% in bending stress was observed. In particular, the highest mechanical properties of PLA plastics were reached after applying the annealing process at 85 °C temperature and for 90 minutes. These results demonstrate the advantages of using 3D printers in the production of products requiring high durability in industrial applications. Moreover, the study findings provide an important method for optimizing the mechanical properties of materials produced with 3D printer technology.

Keywords: Annealing process, mechanical properties, PLA plastics, fused deposition modeling, 3D printing.

1. Introduction

In recent years, 3D printer technology has been a significant innovation that continues to develop rapidly and create substantial impacts in various sectors. 3D printers enable the quick and cost-effective production of complex parts in diverse fields such as engineering, automotive, aerospace, medicine, and construction. However, the mechanical properties of objects produced by 3D printers may be lower compared to those produced by traditional manufacturing methods. This situation limits the broader use of 3D printer technology in the industry [1-3]. Fused deposition modeling (FDM) is one of the most widely used additive manufacturing (AM) techniques for producing plastic parts with complex geometries and customized designs. FDM uses a thermoplastic filament that is heated and extruded through a nozzle to deposit material layers onto a building platform. PLA (polylactic acid) is a biodegradable and renewable polymer derived from cornstarch or sugarcane. Despite its numerous advantages, such as low cost, easy processability, and eco-friendliness, PLA has disadvantages like low mechanical strength, poor thermal stability, and high brittleness. PLA is widely used in 3D printers as a low-cost, environmentally friendly material with biodegradable and biocompatible properties. Enhancing the mechanical properties of PLA can enable parts produced by 3D printers to have a broader range of applications [4-7]. In this context, the annealing process emerges as an effective method for improving PLA's mechanical properties. Therefore, enhancing the mechanical properties of PLA is an essential requirement for FDM applications. One method for improving PLA's mechanical properties is annealing, a thermal treatment that involves heating the part above its glass transition temperature and then cooling it down slowly [8]. Annealing can reduce residual stresses, increase crystallinity, and improve the interlayer bonding of PLA parts. However, annealing may also cause unwanted effects, such as warping, shrinkage, and dimensional changes in PLA parts. Therefore, optimizing annealing parameters, such as temperature and duration, is crucial for achieving the best mechanical performance of PLA parts [9].

In this study, the effect of the annealing process on the mechanical properties of PLA parts produced by FDM was investigated. By applying different annealing temperatures and durations to PLA parts and measuring their tensile strength, modulus of elasticity, hardness, and bending strength, the research was conducted. Moreover, the microstructure and morphology of PLA

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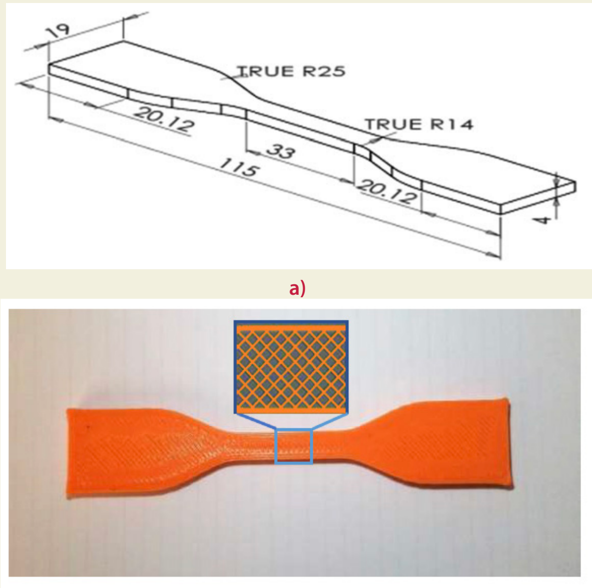
parts before and after annealing were analyzed using scanning electron microscopy (SEM). It was found that annealing significantly improved the mechanical properties of PLA parts, especially at 85°C and for a 90-minute duration.

2. Material and Method

The effect of annealing temperature and duration on the mechanical properties of PLA plastics produced by 3D printing was investigated. PLA samples printed using a 3D printer were prepared. The samples were designed for printing and created using Fusion 360 software. The samples were printed using PLA filament on an Ender 3 S1 3D printer. Samples were printed with a layer height of 0.2 mm and a 100% infill ratio (Table 1). Subsequently, the printed samples were grouped for annealing at 70°C, 85°C, and 100°C temperatures and for 30, 60, and 90-minute durations (Table 2). The annealing process was carried out by holding the samples at a specific temperature inside an oven. The samples were subjected to annealing for a predetermined period by placing them in the oven at a predetermined temperature. Nine different groups were formed using various temperatures and processing times. After annealing, the samples were cooled and prepared for experiments.

Figure 1.a shows the geometric dimensions of the ASTM D638 Type-IV tensile test sample, and Figure 1.b shows the sample detail printed using PLA-Pro filament from Mikrozey company. The produced samples were numbered as shown in Figure 2.a. After creating the samples to be tested, they were subjected to the annealing process. The samples were placed in a THERMNEVO brand laboratory-type annealing oven specifically designed for thermal treatments, as shown in Figure 2.b. This oven circulates heated air around the object inside, resulting in more homogeneous heating outcomes compared to gas or electric ovens. To investigate PLA's thermal deformation for trial purposes, samples annealed at 155°C for 30, 60, and 90-minute durations were deformed due to changes in their geometric dimensions and, therefore, were not subjected to any tests (Figure 2.c).

After the annealing process was completed, the samples were subjected to a tensile test. For this purpose, changes in the dimensions of the annealed samples were measured according to the width and thickness of the non-annealed sample (control sample). Since changes in the critical cross-sectional areas of the samples might occur after annealing, the measurements were taken for each sample and areas were calculated. In this way, more reliable tensile test results were obtained. The samples were attached to the jaws of a SHIMADZU Autograph



a)

b)

Figure 1. a) ASTM D638 Type-4 tensile specimen standard dimensions [12], b) Printed specimen

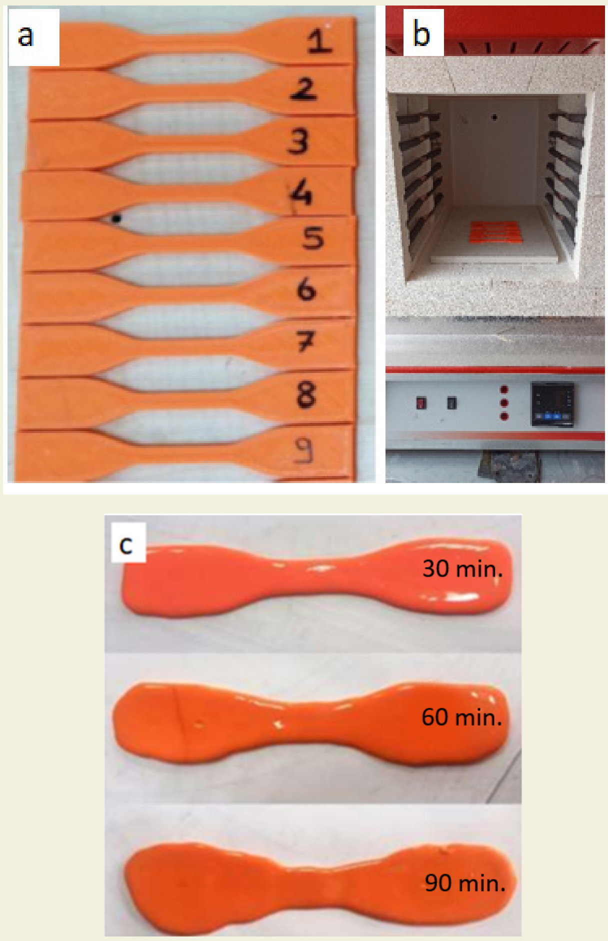


Figure 2. a) Some of the test samples b) Annealing in the furnace c) Samples melted at 155 °C

Table 1. 3D Printing process parameters

Infill Density [%]	Infill Pattern [°]	Layer Thickness [mm]	Bed Temperature [°C]	Nozzle Temperature [°C]	Glass Transition Temp[°C]	Print Speed [mm/s]
100	45	0,2	60	220	60–65	50

Table 2. Annealing parameters used in the experiments

Experiment No.	Temperature [°C]	Heat Treatment Time [min]
1	70	30
2	70	60
3	70	90
4	85	30
5	85	60
6	85	90
7	100	30
8	100	60
9	100	90
10	155	30 (Deformed)
11	155	60 (Deformed)
12	155	90 (Deformed)

AGS-X model 100 kN capacity universal tensile testing machine (Figure 3.a). The test speed was determined as 5 mm/min according to the ASTM D368-Type IV standard. Force was applied to the samples in the tensile direction, and the test was terminated at the breaking point. Tensile tests were performed on a total of 9 samples with three different temperatures and three different time combinations. On the other hand, bending test samples were also processed as seen in Figure 3.b. For statistical accuracy, each sampling process was repeated three times. After the tensile test, the fracture surfaces of the samples were analyzed using SEM. This analysis was conducted to investigate the fracture mechanism, stress relief after annealing, and the effect of annealing on the plastic part. The surfaces formed after the tensile test was cut and reduced to fit the SEM device table and coated with gold dust to prepare for surface imaging.

3. Results and Discussion

3.1. The Effect of Heat Treatments on Tensile Strength and Elastic Modulus

According to the tensile test results, the tensile stress value of the control samples was determined to be 42 MPa (Table 3). After the annealing process, increases in tensile stress values were observed. As a result of the annealing process at 85 °C for 90 minutes, the tensile stress value increased to 62 MPa (Figure 4). Similarly, the elastic modulus values also increased after the annealing process. As a result of the annealing process at 85 °C for 90 minutes, the elastic modulus value was determined to be 3345 MPa (Figure 5). Shore D hardness value and bending stress also increased after the annealing process (Table 3). The increases observed after the annealing process are important for improving the mechanical properties of PLA plastics. In particular, it provides a significant contribution in terms of using 3D printers in the production of parts requiring high durability in industrial applications. Moreover, it serves as an important data source for research on the use of 3D printers in industrial appli-

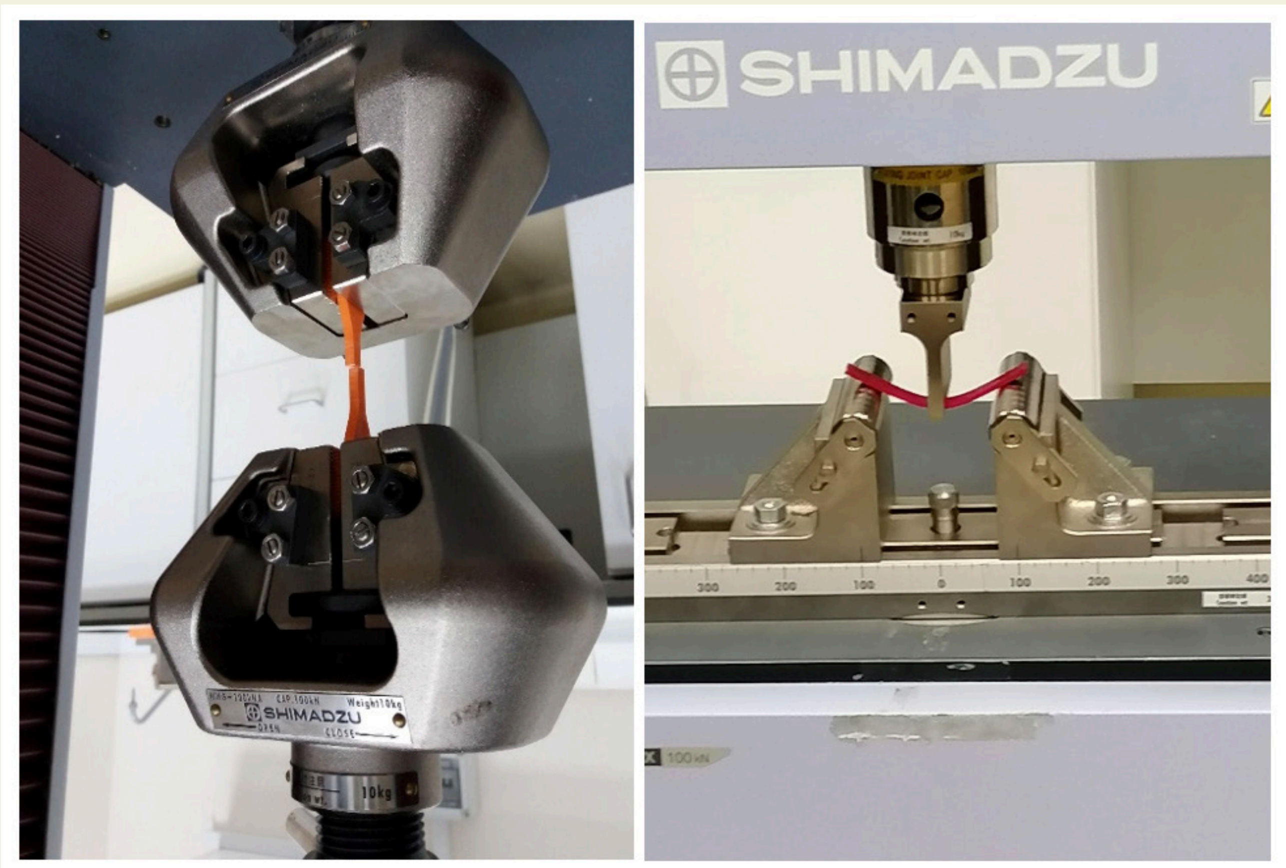


Figure 3. a) Tensile test b) Bending test

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cations. Tensile test results showed that annealing process temperature and durations have a significant effect on the mechanical properties of PLA plastics. Especially, it was observed that the highest mechanical properties of PLA plastics were achieved as a result of the annealing process at 85 °C for 90 minutes. These results may increase the use of 3D printers in areas requiring high durability in industrial applications through the annealing post-process. Lluch-Cerezo et al. (2022) examined the use of ceramic powder molds to prevent dimensional deformation of FDM printed parts made of PLA and Acrylonitrile Butadiene Styrene (ABS) materials during annealing [10]. The effectiveness of the mold was evaluated by comparing the lengths of the parts before and after annealing, and a bending strength test was performed to evaluate the mechanical properties of the parts. The study revealed that the use of ceramic powder molds is an effective method for preventing dimensional deformation of FDM printed parts made from ABS and PLA materials during annealing. Furthermore, it was also shown that the use of ceramic powder molds does not negatively affect the mechanical properties of the parts.

Luna et al. (2021) emphasized that annealing can have both positive and negative effects on the properties of PLA and that optimizing annealing parameters is crucial for achieving the desired properties for a specific application [11]. For instance, annealing can increase the hardness and crystallinity of PLA but may also reduce elongation in fracture and impact resistance. Additionally, annealing at higher temperatures or for longer durations can lead to the thermal degradation of PLA, resulting in a decrease in overall mechanical strength and stability. Therefore, it is essential to carefully evaluate the balances between different properties and optimize the annealing process to achieve the desired property balance for a particular application. This may involve considering other processing parameters such as the temperature, duration, and cooling rate of the annealing process, as well as printing conditions and post-processing treatments. By carefully optimizing the annealing process, it is possible to achieve improved mechanical properties in PLA while preserving other desirable properties such as biodegradability and ease of processing.

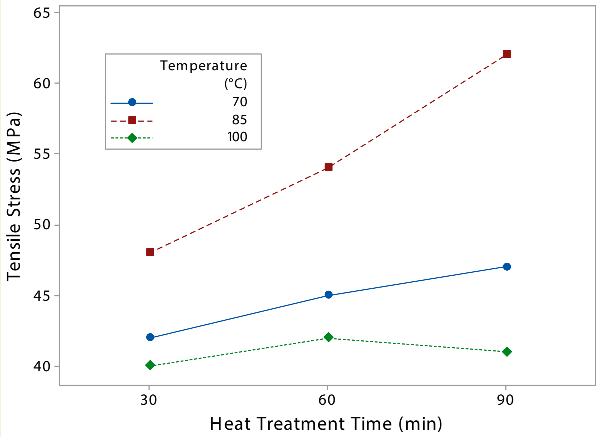

Figure 4. Effect of heat treatment on tensile stress

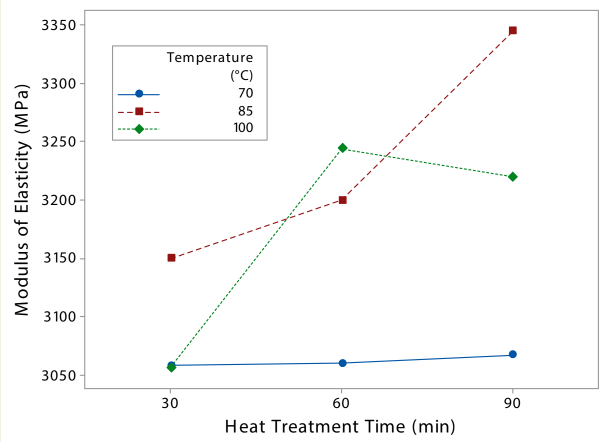

Figure 5. Effect of heating treatment on mudulus of elasticity

3.2. ANOVA Analysis

The resulting ANOVA Table 4. summarizes the results of the analysis to examine the effect of the Temperature (°C) and Heat Treatment Time (min) independent variables on the Tensile Stress (MPa) output parameter. The total explanatory power of the independent variables used for the regression was 86.83 units, with an adjusted SD value. However, since the F-degree is 0.80 and the P-value is 0.493, it is concluded that the regression model does not have a statistically significant explanatory power.

Table 3. Mechanical test values of the samples the annealing process and control sample

Exp. No	Temp. (°C)	Heat Treatment (minutes)	Tensile Stress (MPa)	Modulus of Elasticity (MPa)	Shore D Hardness	Bending Stress (MPa)
Control	-	-	42	1934	60	62
1	70	30	42	3058	67	65,13
2	70	60	45	3060	68	74,19
3	70	90	47	3067	68,5	82,74
4	85	30	48	3150	70	70,27
5	85	60	54	3200	72	78,78
6	85	90	62	3345	74	87,28
7	100	30	40	3056	75	76,81
8	100	60	42	3244	76	85,05
9	100	90	41	3220	76,5	93,28

Table 4. Effect of Temperature (°C) and Heat Treatment Time (min) on Tensile Stress (MPa)

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Regression	2	86,83	43,42	0,80	0,493
Temperature (°C)	1	20,17	20,17	0,37	0,565
Heat Treatment Time (min)	1	66,67	66,67	1,22	0,311
Error	6	326,72	54,45		
Total	8	413,56			

The ANOVA (Table 5) presents the results of the analysis conducted to examine the effect of Temperature (°C) and Heat Treatment Time (min) on Modulus of Elasticity (MPa). The regression model, consisting of the two independent variables, yielded an adjusted sum of squares (Adj SS) of 41275 and an F-value of 2.75, which corresponds to a p-value of 0.142. Although the F-value suggests some evidence of an effect, the p-value does not reach the conventional level of statistical significance ($\alpha = 0.05$).

Table 5. Effect of Temperature (°C) and Heat Treatment Time (min) on Modulus of Elasticity (MPa)

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Regression	2	41275	20637	2,75	0,142
Temperature (°C)	1	18704	18704	2,49	0,165
Heat Treatment Time (min)	1	22571	22571	3,01	0,133
Error	6	44997	7500		
Total	8	86272			

The ANOVA (Table 6) illustrates the results of the analysis performed to investigate the impact of Temperature (°C) and Heat Treatment Time (min) on Shore D Hardness Value. The regression model, comprising the two independent variables, yielded an adjusted sum of squares (Adj SS) of 104.167 and an F-value of 140.63, which corresponds to a highly significant p-value of 0.000. This suggests that the regression model is statistically significant in explaining the variability in Shore D Hardness Value.

The ANOVA (Table 7) represents the results of the analysis conducted to investigate the impact of Temperature (°C) and Heat Treatment Time (min) on Bending Stress (MPa). The regression model, including the two independent variables, showed a highly significant effect with an adjusted sum of squares (Adj SS) of 617.412, an F-value of 1223.67, and a p-value of 0.000. This indicates that the regression model significantly explains the variation in Bending Stress (MPa).

Table 6. Effect of Temperature (°C) and Heat Treatment Time (min) on Shore D Hardness Value

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Regression	2	104,167	52,0833	140,63	0,000
Temperature (°C)	1	96,000	96,0000	259,20	0,000
Heat Treatment Time (min)	1	8,167	8,1667	22,05	0,003
Error	6	2,222	0,3704		
Total	8	106,389			

Table 7. The Effect of Temperature (°C) and Heat Treatment Time (min) on Bending Stress (MPa)

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Regression	2	617,412	308,706	1223,67	0,000
Temperature (°C)	1	182,381	182,381	722,93	0,000
Heat Treatment Time (min)	1	435,031	435,031	1724,41	0,000
Error	6	1,514	0,252		
Total	8	618,926			

3.3. The Effect of Thermal Treatments on Hardness Values

As a result of this study, it has been observed that temperature has a significant effect on Shore D hardness values. Hardness measurements were measured with PCE-DD-D brand Shore D Durometer with 0.5 HD sensitivity and conforming to ASTM D 2240 standard. According to the manufacturer's recommendation, it is recommended to measure samples with a minimum thickness of 6 mm, and if the sample is to be measured below this thickness, it is recommended to place the same sample on top of each other in the number of times that will provide this thickness. For this reason, the control and experimental group samples with a thickness of 8 mm were printed and annealed for hardness measurement. Figure 6 shows the Shore D hardness measurement of the sample annealed at 100 °C and 90 minutes with a durometer. In Figure 7, durometer needle marks are seen on the sample as a result of the measurement. As the annealing temperature increases, the Shore D hardness value also increases. While the hardness values of samples treated at 70 °C were 67 (Figure 8), its increased to 76.5 for samples treated at 100 °C (Table 3). These results are important in terms of using 3D printers in the production of products requiring high hardness in industrial applications and demonstrate that temperature control is a critical factor in reaching the desired hardness properties of the material. Annealing can reduce stresses formed within PLA parts produced by 3D printers and improve the bonds between filaments. Thermal treatment can significantly increase the mechanical properties of PLA, especially tensile strength and heat resistance. Annealing temperature and duration affect the crystalline structure of PLA and, consequently, its hardness. The annealing temperature should be lower than PLA's melting temperature. Annealing is a thermal treatment method often used to increase the hardness of materials. PLA is a biodegradable thermoplastic polymer. The crystallization and densification process allows PLA molecules to align in a more orderly fashion and form a tighter structure. This results in PLA becoming a harder material. Shore D hardness measures the material's resistance to a specific force, and the higher the Shore D hardness of a material, the harder it is considered to be. Therefore, annealing and crystallization processes can increase PLA's Shore D hardness, enabling the production of harder and more durable materials.

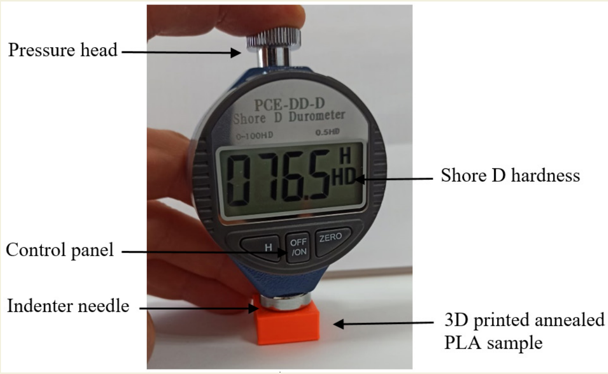

Figure 6. Hardness measurement with durometer

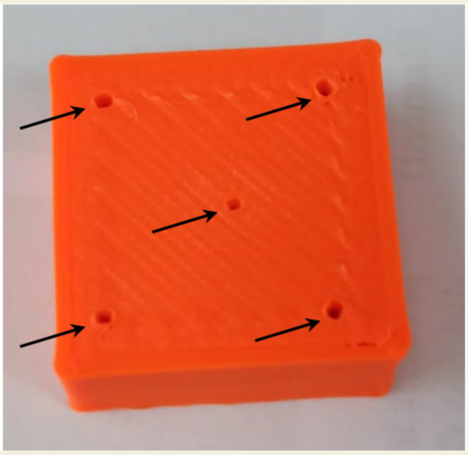

Figure 7. Appearance of needle marks on the sample

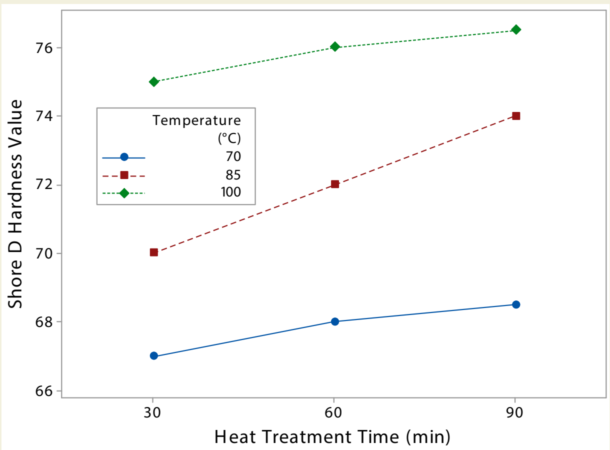

Figure 8. Effect of heat treatment on Shore D hardness value

3.4. The Effect of Temperature on Flexural Stress

In the conducted study, as the temperature increased and the processing time extended, an increase in the flexural stress values of PLA plastics was observed. Particularly, after applying a thermal treatment of 85 °C and 90 minutes duration, the highest flexural stress values (93 MPa) were reached for PLA plastics (Figure 9). These results are important in terms of using 3D printers in the production of products requiring high flexural strength in industrial applications, and it has been demonstrated that more durable products can be produced by optimizing thermal treatment parameters.

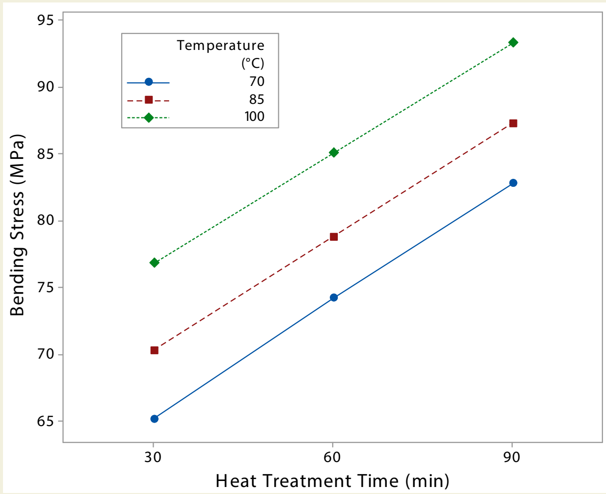

Figure 9. Effect of heat treatment on bending stress

3.5. Microstructure Investigations

In this study, the microstructures of the PLA plastic samples after annealing treatment were investigated. The surfaces of the broken samples after the tensile test were analyzed using SEM. The analyses conducted revealed that the annealing treatment caused significant changes in the microstructure of the plastic samples. Specifically, annealing treatments at 85 °C for 90 minutes led to increased crystallization in the microstructure of PLA plastics (Figure 10), which in turn contributed to the improvement of the mechanical properties of the samples. Furthermore, cracks and deformations observed on the fracture surfaces of the untreated samples were also confirmed through microstructure analyses (Figure 11). These results demonstrated that the annealing treatment not only affects the mechanical properties but also significantly influences the microstructure of the plastic samples.

4. Conclusion

In the research conducted within the scope of this study, it has been demonstrated that the mechanical properties of PLA plastics produced with 3D printers significantly improved depending on the applied heat treatment temperature and duration. Experiments were carried out using different temperatures (70°C, 85°C, and 100°C) and durations (30, 60, and 90 minutes), and the changes in mechanical properties were investigated.

According to the results, it has been shown that with in-

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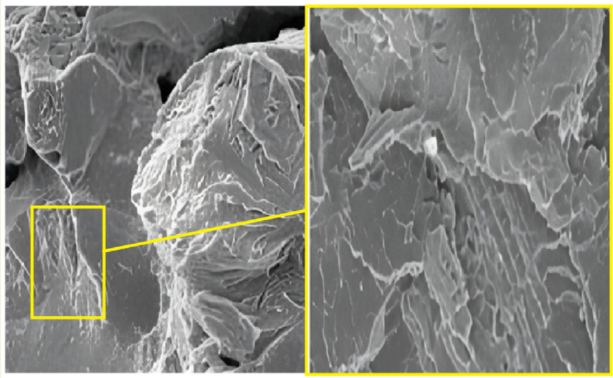

Figure 10. PLA tensile test fracture surface annealed at 85 °C

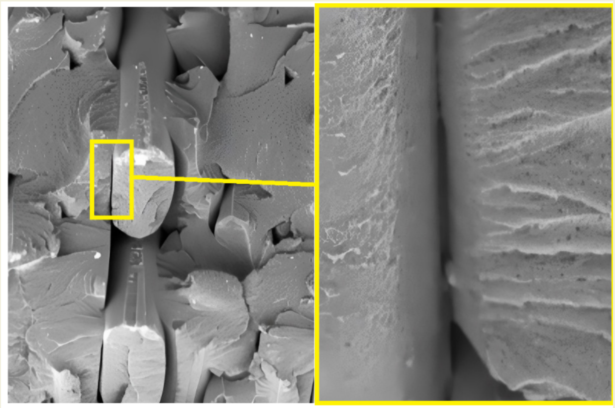

Figure 11. PLA tensile test fracture surface without annealing

creasing heat treatment temperature and duration, there are significant improvements in the mechanical properties of PLA plastics, such as tensile strength, elastic modulus, Shore D hardness value, and bending stress. In particular, applying heat treatment at 85°C and for 90 minutes enabled PLA plastics to achieve the highest mechanical properties.

These results emphasize the importance of 3D printing technology in the production of products requiring high durability in industrial applications. Moreover, it should be considered that the mechanical properties of PLA plastics can be optimized with heat treatment duration and temperature. This study highlights the importance of heat treatment in improving the mechanical properties of materials produced with 3D printers and provides valuable information for industrial applications.

This study demonstrates that the mechanical properties of PLA plastics produced with a 3D printer can be optimized with heat treatment temperature and duration. These results reveal the advantages of using 3D printers in the production of products requiring high durability in industrial applications. For example, the performance and lifespan of parts made from PLA plastics in the automotive, aerospace, medical, and textile sectors can be enhanced with heat treatment. Additionally, the biodegradability and renewability of PLA plastics are environmentally significant.

In future studies, it is planned to investigate the mechanical properties of PLA plastics produced with 3D printers under different heat treatment conditions in more detail and develop mathematical models for the optimization of heat treatment parameters. Moreover, the impact of heat treatment on other physical, chemical, and biological properties of PLA plastics produced with 3D printers will also be investigated.

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