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The Influence of Annealing Temperature and Holding Time Near Glass Transition Temperature on the Tensile Strength of Fused Deposition Modeling Printed Polylactic Acid

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Abstract

Thermal annealing can be implemented to improve the mechanical strength of a 3D-printed object. The critical parameters of thermal annealing are temperature and holding time. Based on the literature review, the implemented annealing temperature affects the required holding time. As using a lower annealing temperature and holding time can reduce the required heat and increase the efficiency of the process, this research investigates the implementation of the annealing temperature near the glass transition temperature with a short holding time. The aim of this research is to investigate the influence of the thermal annealing temperature range of 65°C to 85°C and the holding time of 45 minutes to 75 minutes on the ultimate tensile strength of a polylactic acid part printed using fused deposition modeling. The experiment implemented a 3^2-factorial design methodology with two replications. The experiment results show that thermal annealing slightly above the glass transition temperature promotes higher interlayer diffusion of raster and layers, increasing ultimate tensile strength. Meanwhile, the investigated holding time does not influence the ultimate tensile strength of the annealed part. As the holding time range might not be able to accomplish the maximum crystallization for the annealing temperature range between 65°C and 85°C, further research is required to study the influence of a longer holding time on the ultimate tensile strength of the specimen annealed at a slightly above the glass transition temperature.

Keywords:
Annealing, temperature, holding time, tensile strength, polylactic acid.

1 Introduction

3D printing is a popular technology for generating various product prototypes and made-to-order products. In addition, it can be used to produce a variety of industrial tools, including casting patterns and cores. Based on the computer-aided design model, the printed part is constructed layer by layer during the 3D printing process. 3D printing technology has numerous benefits, including the capacity to produce a complex design in a short amount of time. However, while the printing process is conducted layer by layer, the layers may separate under special stress and cause the printed object to fail. Therefore, the mechanical strength of the 3D-printed object needs to be strengthened.

Fused Deposition Modeling (FDM) is one of the most prevalent 3D printing technologies. FDM is inexpensive and simple to use. It can print a variety of polymer materials. As a result, it is largely employed in rapid prototyping. Polylactic acid (PLA) is extensively utilized as a rapid prototyping material. The material was used since it is one of the renewable polymers. In addition, PLA is safe when heated, even though it degrades in less time than other polymers.

The mechanical strength of the PLA printed part is affected by several FDM parameters, such as layer thickness and infill density [1]. Earlier researchers have investigated the influence of these parameters on tensile strength, compression strength, flexural strength, and impact strength to improve the mechanical strength of the PLA-printed part [2][3].

Instead of varying the parameters during the printing process to improve the mechanical strength of the PLA printed part, thermal annealing can be used to improve the mechanical strength after the printing process without the use of any additional material coating overprinted parts. Thermal annealing is conducted by heating the printed part at a range of temperatures for a fixed time and then cooling it to room temperature. The annealing temperature is set between the glass transition temperature and the melting point temperature of the printed part material. The glass transition temperature is the temperature at which the PLA turns from a glassy to a rubbery state. According to Pazhamannil et al. [4] and Chikkanna et al. [5], thermal annealing influences the crystallinity of the part and then the flexural strength and tensile strength of the printed part.

The critical parameters of thermal annealing are temperature and holding time. Therefore, it is necessary to evaluate the effect of these process parameters on the mechanical strength of the FDM-printed component. Previous researchers have explored the relationship between various process parameters. Based on their investigation, the applied annealing temperature influences the required holding time. In addition, numerous studies have been undertaken to determine the optimal thermal annealing temperature and holding time for increasing the Ultimate Tensile Strength (UTS) of the printed PLA.

Rangisetty and Peel studied the influence of the annealing process parameters on UTS, young modulus, flexural strength, and flexural modulus of virgin PLA, ABS, and PETG with various reinforced materials [6]. Several specimens are printed in this research by maintaining the extrusion temperature between 190°C to 210°C. The temperature and holding time of the annealing process are 65°C and 60 minutes, respectively. The results show that the UTS of the FDM printed PLA specimen increases non-significantly as it is annealed at 65°C for 60 minutes.

The investigation by Wach et al. printed and annealed various PLA specimens to evaluate the influence of the thermal annealing temperature and the holding time on its crystallinity degree [7]. The crystallinity degree is evaluated since it connects directly with the specimen's mechanical properties. For annealing temperatures between 65°C and 70°C, holding times in excess of 10 hours do not result in an increase in crystallinity degree. However, thermal annealing of the specimen at 95°C for less than 10 minutes is adequate to attain the highest degree of crystallinity. This investigation then examined two combinations of thermal annealing parameters, namely temperature and holding time. The first temperature and holding time combination is 85°C and 70 minutes. The specimen is then annealed for 15 minutes at 95°C. The results indicate that thermal annealing can increase the FDM-printed PLA specimen's flexural strength by up to 17%.

Jayanth et al. performed an annealing process on FDM printed PLA specimens at three different temperatures (90, 100, and 120°C) and three different holding times (60, 120, and 240 minutes) [8]. This study demonstrates that the mechanical properties of PLA, particularly UTS, can be improved by around 80% with heat treatment at 100°C for 240 minutes. However, the UTS of PLA decrease when annealed at temperatures greater than or equivalent to 120°C because the PLA material begins to degrade at these temperatures.

Beniak et al. conducted an experiment in which FDM-printed PLA specimens were annealed at 110°C for 20 and 60 minutes [9]. They found that thermal annealing influences the toughness and brittleness of the specimens. The thermal annealing also increases the compressive strength by maintaining the holding time for 20 minutes. However, the implementation of thermal annealing has no influence on the UTS.

Based on the research by Wang et al., FDM printed PLA specimens that are annealed show an increase in UTS [10]. In this study, specimens are annealed at 80 and 100°C for 30, 60, and 120 minutes. Performing the thermal annealing at 80°C for 60 minutes yields the maximum UTS. The ANOVA analysis reveals, however, that the increase in annealing temperature and holding time has no influence on the UTS of the specimens over the experimental temperature and holding time ranges.

Akhoundi et al. investigated the influence of thermal annealing on the tensile strength of PLA specimens [11]. The thermal annealing is conducted at 110°C for 60 minutes. The research also examined the effect of FDM printing temperature on the UTS of annealed PLA specimens. The research implemented five different printing temperatures, which are 210°C, 220°C, 230°C, 240°C, and 250°C. The results show that the maximum tensile strength of annealed specimens is higher than non-annealed specimens. The annealed specimen that is printed at the temperature of 250°C provides the maximum value of UTS.

Slavkovic et al. studied the effect of thermal annealing on the mechanical properties of FDM printed PLA specimens [12]. Compressive and tensile tests were performed on the thermally annealed and un-annealed printed components. The thermal annealing is conducted for 60 minutes at 75°C. The compressive and tensile strengths of the annealed specimens are greater than those of the specimens printed at room temperature.

Bhandari et al. examined the effect of two annealing temperatures and three annealing holding time on the tensile strength of interlayer FDM-printed PLA specimens [13]. The results show that the thermal annealing at 120°C for 30, 240, and 480 minutes did not significantly influence the interlayer tensile strength of the specimens. However, thermal annealing at 90°C for 30, 240, and 480 minutes slightly increases the interlayer tensile strength of the specimens. The increase of the holding time at interlayer 90°C does not significantly increase the interlayer tensile strength.

Butt and Bhaskar investigated the influence of thermal annealing on the UTS of FDM printed PLA specimens [14]. The 60-minute thermal annealing procedure is undertaken at 70°C, 80°C, and 90°C. It was discovered that thermal annealing increases the specimen's UTS as the annealing temperature increases from 70°C to 80°C. The increase in annealing temperature from 80°C to 90°C had no effect on the specimen's tensile strength.

Szust and Adamski studied the effect of build orientation and annealing on the ultimate tensile strength and tensile modulus of PLA FDM specimens [15]. This study demonstrates that PLA FDM specimens printed in ZYX orientation and annealed at 60°C for 60 minutes exhibit an increase in UTS and a decrease in tensile modulus. The change in UTS when the annealing temperature is increased to 80°C is negligible. However, the thermal annealing process affects the geometric deformations of the specimens and reduces the dimensional accuracy of the printed item.

Arjun et al. performed thermal annealing at 65, 95, 125, and 155°C for 30, 60, 120, and 240 minutes to improve the mechanical properties of the printed PLA [16]. The results of the research reveal that annealing performed at 95°C for 120 minutes increase the UTS.

Based on the research above, it can be summarized that the annealing temperature range from 70°C to 80°C influences the UTS of the specimen as described by Butt [12]. According to Szust and Adamski, the UTS of the printed part increases by conducting an annealing process at 60°C temperature for 60 minutes holding time [15]. The experiment result by Arjun et al. shows that the annealing performed at 95°C for 120 minutes increase the UTS [16]. The highest UTS of the specimen might be achieved by performing the thermal annealing at 80°C for the holding time of 60 minutes or at 75°C for 60 minutes, as shown by Slavković et al. [12]. In addition, according to Wach et al., thermal annealing at 80°C for 70 minutes can also increase the flexural strength of the FDM printed PLA specimens [7].

On the other hand, Wang et al. presented that for thermal annealing at 80°C, the change in holding time from 30 minutes to 60 minutes and from 60 minutes to 120 minutes does not significantly influence the UTS of the specimen [17]. The research by Guduru and Srinivasu found that the annealing process performed at 120°C for 2 minutes showed the highest UTS [18]. According to Jayanth et al., the UTS of the PLA can be enhanced by heat treating to about 100°C for four hours [8]. However, the use of an annealing temperature equal to or above 120°C reduces the UTS of the PLA. In addition, thermal annealing at 90°C for the holding temperature of 30 minutes, 240 minutes, and 480 minutes insignificantly increases the UTS of the specimen, as explained by Bhandari et al. [13].

Using a lower annealing temperature and holding time can minimize the required amount of heat and improve the efficiency of the operation. Consequently, this study analyzes the application of annealing temperatures close to the glass transition temperature with a brief holding time. This research aims to determine the effect of the thermal annealing temperature range of 65°C to 85°C and holding time of 45 minutes to 75 minutes on the UTS of the FDM-printed PLA part.

2 Materials and Methods

A low-cost FDM printer named the Anet® A8 was used to print a tensile test specimen from CCTREE 3D printing's transparent PLA filament. The printer has a 220 mm × 220 mm × 240 mm print table and an extruder nozzle with a 0.4 mm diameter. The consumed PLA filament has a 1.75 mm diameter, a tensile strength of 85 MPa, an impact strength of 3 kJ/m^2, and a flexural strength of 97 MPa [19].

Table 1 shows the FDM printing parameters to build the specimens. All 3D printing parameters used in this research are set constant to achieve a certain UTS value. The printing parameters are set constant as they could have an effect on the result of the annealing process [20][21]. Then, the printing path was generated by using two software which are Prusa Slicer 2.3® and Repetier-Host®.

Two replications of the 3^2 factorial design methodology are implemented in this experiment. The factorial design methodology is used in this research because it is more efficient than conducting a series of independent studies. For statistical accuracy, a total of 18 tensile test specimens were printed. The printed PLA specimen is printed according to ASTM D638 [22]. Fig. 1 inhibits the dimensions of the tensile test specimen. A Mitutoyo® caliper with 0.01 mm of accuracy and 150 mm capacity is used to measure the dimension value of the printed specimen. Then the dimension of the printed specimen is checked to ensure it complies with the ASTM standard. The printed specimen is also examined to make sure that the shape of the specimen fulfills the standard.

The UTS of the PLA specimens printed by using FDM is investigated in this research for different values of thermal annealing temperature and holding time, as presented in Table 2. The values are chosen based on the result of the literature review. An Ofenbau Hofmann® furnace is used to conduct the annealing process by heating the specimen at the specified annealing temperature for the specified holding time. The specimen is placed in the furnace after the furnace reaches the specified annealing temperature. After the time has elapsed, the oven is turned off, and the specimen is cooled inside until it reaches room temperature.

Table 1. Printing process parameters.

Parameters	Value
Layer thickness (mm)	0.15
Nozzle diameter (mm)	0.4
Nozzle temperature (°C)	210
Bed temperature (°C)	55
Infill pattern	Triangle
Infill density (%)	100
Shell parameter	3
Printing speed (mm/s)	50

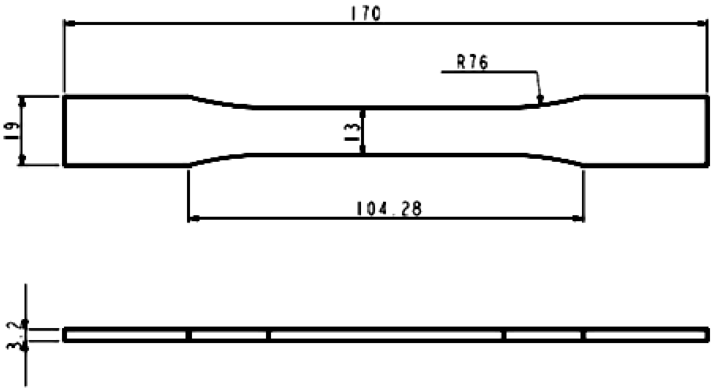

Fig. 1. Dimensions of the tensile test specimen.

The width and thickness of the middle portion of the annealed specimen were measured five times, and the averages of these measurements were used to compute the specimen's cross-sectional area. Using a Universal Testing Machine GT-7001-L30 with a maximum capacity of 30 tons, the maximum tensile load of each annealed specimen is determined. The UTS was then computed by dividing the tensile load by the specimen's cross-sectional area.

Table 2. Process parameters used.

Parameters	Value		
	Low	Middle	High
Annealing temperature (°C)	65	75	85
Holding time (minutes)	45	60	75

3 Results and Discussion

The UTS value for each thermal annealing temperature and holding period is displayed in Table 3. The annealed specimens have greater UTS values than the un-annealed PLA filament. The average UTS against temperature and holding time can be plotted based on the findings of the experiment, as shown in Fig. 2 and Fig. 3. The average UTS values of the annealed specimens varied between 91.55 MPa and 97.65 MPa as depicted in Fig. 2. It illustrates that the annealed temperature affects the average UTS value of the annealed specimen. As the temperature of the annealed specimen increases, its UTS decrease. The findings of an ANOVA run on Minitab software with a 95% confidence level and significance level of 0.05 indicate that temperature has a significant effect on the UTS of the annealed specimen.

Fig. 3 shows that the UTS average values of the annealed specimens against the time vary between 94.51 MPa and 94.71 MPa. The graph illustrates that the UTS of the annealed specimen reduces when the holding time increases from 45 minutes to 60 minutes. By increasing the holding time from 60 to 75 minutes, the graph demonstrates that the UTS reaches its maximum value.

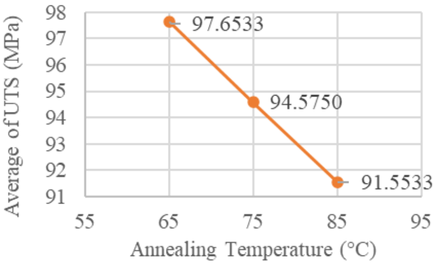

Fig. 2. UTS against annealing temperature.

Table 3. Experiment results.

Temperature (°C)	Holding time (minutes)	UTS (MPa)
65	45	98.79
65	45	97.90
65	60	97.90
65	60	97.90
65	75	98.57
65	75	94.86
75	45	91.80
75	45	96.30
75	60	95.74
75	60	93.52
75	75	97.72
75	75	92.37
85	45	92.81
85	45	89.79
85	60	88.54
85	60	93.44
85	75	88.85
85	75	95.89

However, based on the 95% confidence level and 0.05 significant level of the ANOVA, it can be concluded that the holding time has no effect on the UTS of the annealed specimen.

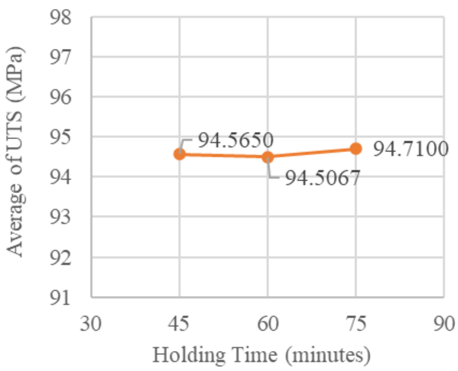

Fig. 3. UTS against holding time.

Based on the results, the average UTS value of the annealed specimen decreases as the annealing temperature increases from 65 to 85°C. At these temperatures and holding times, the degree of crystallization remains minimal and has no effect on the UTS. Principally, the UTS is influenced by the degree of diffusion bonding between raster and layers, which modifies the interlayer mechanical characteristics. Thermal annealing at a temperature slightly above the glass transition temperature, which is 65°C, promotes greater interlayer diffusion of raster and layers, hence increasing the UTS. It is consistent with the findings of von Windheim et al. According to this study, annealing below the temperature of crystallization decreases thermal stresses and increases tensile strength [23]. Though, a higher temperature of 80°C decreases the bonding between the raster and layers because the increase in the temperature increases the viscosity of the bead deposition in the specimen and reduces the diffusion bonding between the raster and layers.

The findings of the experiment indicate that the average UTS value of the annealed specimen is unaffected by holding times ranging from 45 to 75 minutes. The holding time range cannot achieve maximal crystallization for annealing temperatures between 65 and 85°C. Increasing the specimen's degree of crystallinity requires a longer holding time. In accordance with the findings of earlier studies, the annealing process below 85°C takes a few hours of holding time to obtain the greatest degree of crystallization. At these temperatures and holding times, the UTS is mostly influenced by the degree of diffusion bonding between the raster and layers. Consequently, the UTS is unaffected by the holding period for the applied temperature range.

4 Conclusion

Experiment results indicate that the annealed temperature influences the average UTS value of the annealed specimen. With increasing temperature, the UTS of the annealed specimen decreases. At this annealing temperature and time range, the UTS is primarily affected by the degree of diffusion bonding between the raster and layers, which modifies the interlayer mechanical characteristics. The increase in annealing temperature increases the viscosity of bead deposition in the specimen and decreases diffusion bonding between the raster and layers.

In the meantime, the average UTS value of the annealed specimen is unaffected by the holding time range of 45 to 75 minutes, as this range is insufficient to achieve maximal crystallization. Increasing the degree of crystallinity of the specimen could necessitate a longer holding time. Consequently, additional research is required to investigate the effect of a longer holding time on the UTS of the specimen annealed at a temperature slightly above the glass transition temperature.

The findings of this study suggest that the annealing process of a PLA-printed component should be completed at a temperature just slightly above the glass transition temperature in order to reduce energy usage.

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