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The Effect of Heat Treatments Applied to Continuous Fiber Reinforced Thermoplastic Composites on Mechanical Properties

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Abstract - This study investigated the effects of different heat treatments on continuous fiber-reinforced thermoplastic (CFRTP) 's. CFRTP composite is produced using fused deposition modeling (FDM), which is one of the additive manufacturing methods. Polylactic acid (PLA) was used as a matrix, and carbon fibers (3K) were utilized as reinforcement material. First, CFRTP filament was produced on a specially designed melt impregnation line. Afterward, test samples were manufactured via a conventional 3D printer. Then, heat treatments (re-melting in salt, microwave oven, oven) were applied to the produced samples, and the effects of these processes on mechanical properties were investigated. Three-point bending tests were used to investigate the mechanical properties of the test samples. As a result of the heat treatments applied to the CFRTP specimens, flexural stresses between 200 and 220 MPa was achieved. The highest bending stress was obtained by re-melting in salt. As a result of the heat treatments, the stress values are similar, but the re-melting in salt application exhibited a more rigid behavior.

Keywords: Continuous Fiber Reinforced Thermoplastic; Heat Treatment; 3D Printing; Fused Deposition Modeling

1. Introduction

3D printers, also known as additive manufacturing (AM) methods, are a technology that enables computer-generated models to be converted into physical parts [1]. AM technology produces lightweight parts, uses less material waste [2], and it is possible to obtain complex geometries. For this reason, it is preferred in the production of parts in many industries such as aviation, medical and automotive [3]. Although more innovations and improvements are made in the AM method, some disadvantages are encountered in this method [4]. For example, the low mechanical, physical, and thermal properties of the printed parts are among the main disadvantages of the AM method. In order to improve the mechanical properties of the parts, many methods have been used, such as selecting the best printing parameters [5–7], adding reinforcements [8-9], applying heat treatment [10–12], etc., in the literature.

It has been observed that the mechanical properties of the parts produced by optimizing the printing parameters are affected mainly by the printing temperatures, printing speed, layer thickness, scanning angle, fill rate, etc.

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For example, Gunay et al. [13] prepared PLA+ samples produced by a 3D printer according to various printing parameters and examined the effects of these parameters on tensile strength. As a result, the most important parameters were fill rate, raster angle, and printing speed, respectively. Similarly, Ning et al. [14] The effects of process parameters (raster angle, infill speed, nozzle temperature, and layer thickness) on mechanical properties were investigated using chopped carbon fiber reinforced ABS composite filament. As a result, the highest mechanical properties were obtained with 0, 90 printing angle, 25 mm/s infill speed, 220°C nozzle temperature, 0.25 mm layer thickness.

Although many studies have been done on optimizing printing parameters [7], the improvements obtained with this method have been limited. Different studies have been carried out to improve the mechanical properties, apart from optimizing the printing parameters. For example, Jo et al. [15] subjected PLA samples produced with a 3D printer to heated under high pressure in a hot stamping die. He investigated the changes in the mechanical properties of the samples with the effects of heating and pressure and obtained higher mechanical results than the untreated sample. The reason for this was interpreted as the decrease in the voids in the internal structure of the piece. Also, Nakagawa et al. [16] laid the carbon fibers on the pure polymer parts while printing and produced a sandwich structure with the help of a hot pin. Then, these samples were subjected to heat treatment in a microwave oven to increase the adhesion between the fiber matrix and improve the mechanical properties. As a result, it was observed that the mechanical properties were affected insignificantly because the bond between the fiber and the matrix is weak due to the preparation method of the samples, and the application of microwave heat treatment removed a small number of voids in the internal structure.

Another method to improve mechanical properties is the addition of various reinforcement elements to the polymer material. Continuous fiber-reinforced thermoplastic composite (CFRTP) has been shown to exhibit extraordinary mechanical properties compared to non-reinforced polymer and/or short fiber-reinforced parts. For example, Caminero et al. [17] produced continuous glass, carbon, and Kevlar® fiber reinforced nylon composites using fused deposition method (FDM) technology. The interlayer adhesion performance of these samples was investigated. As a result, it was seen that the mechanical properties of the composite samples were higher than the pure samples. In a similar study, Matsuzaki et al. [18] produced composite samples using the PLA thermoplastic filament and carbon fiber/jute fiber reinforcement element by impregnating a hot nozzle before printing. Pure and composite samples were subjected to mechanical tests to measure tensile strength. The results obtained showed that the highest tensile strength was the PLA composite sample with carbon fiber reinforcement. Although superior mechanical properties are obtained in the parts produced by the additive CFRTP method, gaps in the internal structure, weak interlayer adhesion, etc., limit the mechanical properties of the parts produced by this method [19].

In this study, heat treatments were applied to CFRTP samples to increase the mechanical properties. For this purpose, a melt impregnation line method was utilized to produce CFRTP filaments. Then, an FDM-type 3D printer was used to prepare the samples using the produced filaments. Next, oven, microwave oven, and salt-remelting methods were applied to the printed parts. First, the oven and microwave methods were used to bring the printed parts to the glass transition temperature to increase interlayer bonding. Then, the CFRTP sample was placed in fine salt and heat-treated in the oven at melting temperatures in the salt re-melting method. The main advantage of this heat treatment is that it can also be applied to complex geometries. After the heat treatment methods, mechanical properties were investigated using three-point bending tests.

2. Materials and Methods

2.1 Materials

Continuous carbon fiber (3K, DowAksa, Turkey) was used in this study. Carbon fibers have a tensile strength of 4900 MPa and a modulus of elasticity of 245 GPa, and a density of 1.8 g/cm3. Therefore, the approximate diameter of each strand is 7 µm. In addition, 1.75 mm PLA (Polylactic acid) filament (Porima, Turkey) with a tensile strength of 54.3 MPa and a modulus of elasticity of 2300 MPa was used as a matrix material.

2.2 CFRTP Filament Manufacturing

Initially, a production line based on the melt impregnation method was used to produce the CFRTP filaments. The schematic image of the production line used in our study is shown in Figure 1 [20]. This line consists of the fiber spreading zone, the polymer blend, and finally, the mold. The fiber-spreading zone is formed by positioning multiple rollers to spread the fibers laterally to ensure uniform impregnation of the fibers. The positions of the rollers are designed to be adjustable to change the tensile force on the fiber. In this way, the spread of the fibers is made possible by the tensile force created. If the applied force is excessive, it may break the fiber strands, and if it is not enough, sufficient spreading will not occur. The second zone of the production line aims to impregnate the molten polymer with the dispersed fiber. For this purpose, the guiding rollers in this region were heated to 210°C with cartridges. The PLA filament is mixed with the fiber strip by a polymer-fiber mixture roller. The filaments are pushed into the melting zone by means of the extruder. The polymer mixture roller has a channel passing through the middle of the cylinder to guide the polymer. A total of seven radial (for a homogeneous mixture) holes, each with 0.6 mm in diameter, were drilled on the surfaces of the cylinder where it contacts with the fiber. The molten polymer flows through these holes and meets the fiber, and the mixture is obtained. Finally, a circular heated nozzle was used to transform the produced composite filament into a circular form to be used in the 3D printer. The fiber ratios of the produced filaments were calculated with the same principle as in our previous study [20]. As a result of the calculations, the fiber ratios were kept at 23%.

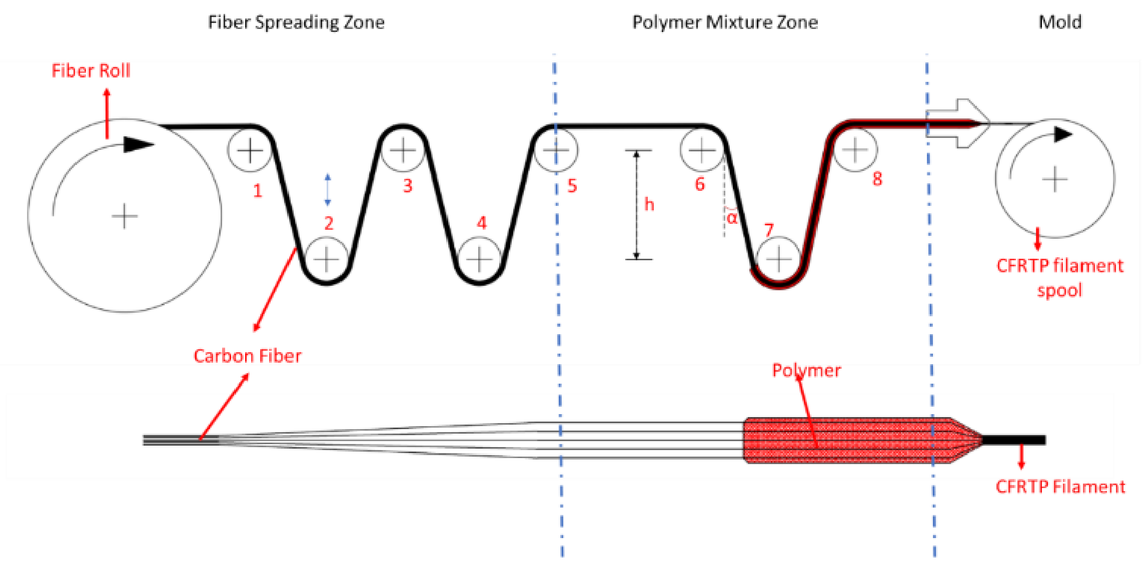
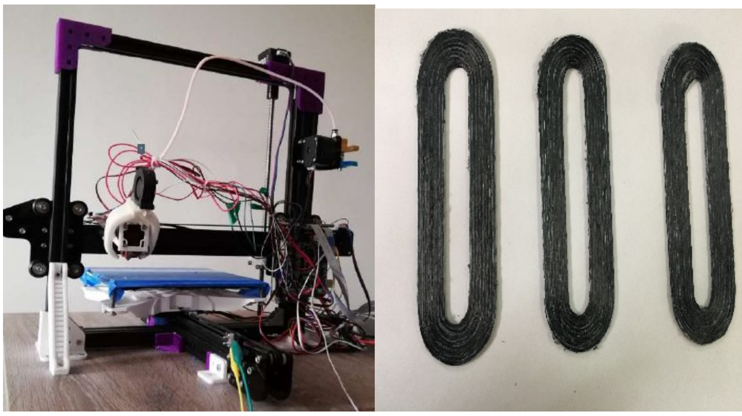


Figure 1. The schematic image of the production line [20]

2.3 Additive Production of CFRP Sample

The 3D printer shown in Figure 2a was used to produce the test samples. In addition, a special g-code was issued for the CFRTP thermoplastic filament to be used in the manufacturing of mechanical test samples. All the samples were produced with continuous pathing, and no cutting was required. However, standard nozzles used in 3D printers have been found to cause fiber damage during printing. For this reason, the nozzle tip used in the study was rounded, and the hole diameter was drilled as 2 mm (larger than the filament diameter). In addition, 220°C nozzle temperature, 10 mm/s printing speed, and 0.25 mm layer thickness were selected as printing parameters. The images of the produced samples are shown in Figure 2b.



(a) (b)

Figure 2. Additive manufacturing of CFRTP samples; a) Custom 3D printer and b) Bending Test Specimens

2.4 Heat Treatments

The heat treatment methods and parameters used in this study can be summarized as; 100°C and 75 min in oven heat treatment, 210°C and 125 min in salt re-melting, and 300 W and 10 min microwave heat treatment. Oven heat treatment was carried out in a furnace. The three-point bending samples produced from the 3D printer are placed directly into the furnace and kept at 100 °C for 75 min. Next, CFRTP samples were placed in the mold with fine salt in the salt re-melting heat treatment. Samples embedded in salt with 210°C for 75 min. The samples were then removed from the oven and cleaned. Finally, samples were merged in water in the microwave oven heat treatment to prevent arcing because of the conductive carbon fibers. In this method, parts were kept in the microwave with 300 W for 10 min. The preparation of the samples before each heat treatment method is shown in Figure 3.

(a) (b) (c)

Figure 3. Preparation of CFRTP samples for heat treatment; a) Re-melting in the salt sample, b) Microwave sample, and c) Oven sample.

2.5 Tree-point bendıng tests

In this study, the bending properties of the CFRTP samples were investigated. The MTS Criterion Model 45 tester (figure 4) was used for the bending test. The samples were produced in accordance with the parameters of the "ISO 14125 - Determination of Flexural Properties of Fiber Reinforced Plastic Composites" standard. For the three-point bending tests, samples were prepared with dimensions of 100x15x2 mm. The crosshead velocity in the device was used as 5 mm/min. The tests were repeated four times to prove reproducibility.

Figure 4. Application of three-point bending tests on CFRTP samples

3. Results and Discussions

Bending stress-strain curves obtained from three-point bending tests of CFRTP composites are shown in Figure 5. As seen in the figure, re-melting in salt showed the highest flexural modulus of elasticity and flexural strength compared to the untreated and pure samples. In the salt re-melting method, interfacial gaps formed during printing are reduced when the melting point of the matrix is reached. The effect of microwave heat treatment has shown negative results, and obtained stress values were lower when compared with non-heat treated CFRTP samples. This is possibly caused by the addition of water which caused shorter cooling times and prevented heating of the parts. All results for the three-point bending test are summarized in Table 1. Oven, Salt re-melting, Microwave test specimens provided flexural stress of 217, 218, 203 MPa and a modulus of elasticity of 12, 16, 15 GPa, respectively. Untreated and neat PLA test specimens provided bending stress of 205,89, 61,90 and a modulus of elasticity of 14,56 and 2,07 GPa, respectively.

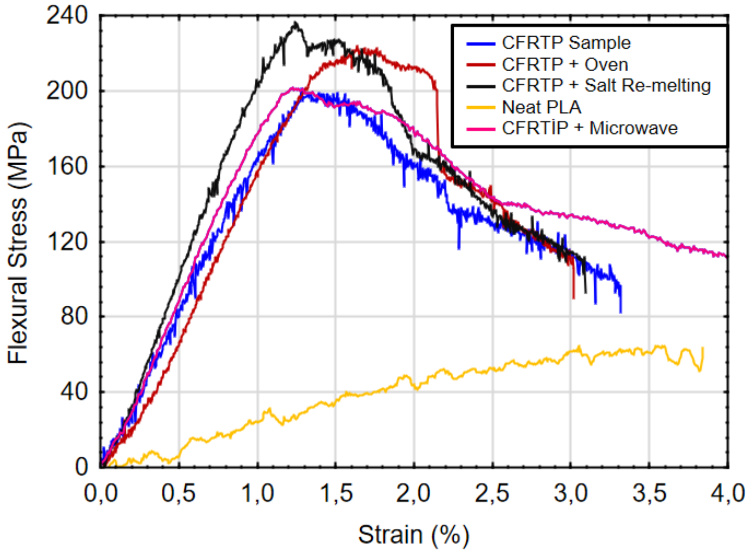


Figure 5. Three-point bending stress-strain curves of CFRTP samples

Table 1. Summary of three-point bending test results

Production Method	Flexural Strength (MPa)	Increase in Flexural Strength (%)	Flexural Modulus of elasticity (GPa)	Increase in Flexural Modulus of Elasticity (%)
Neat PLA	61,90 (4,91)	-	2,076 (1,38)	-
CFRTP Sample	205,89 (7,23)	233	14,56 (1,48)	601,3
CFRTP + Oven	217,04 (11,74)	251	12,16 (3,71)	485,7
CFRTP + Microwave	202,58 (10,50)	227	15,78 (2,20)	660,1
CFRTP + Re-melting Salt	217,72 (34,34)	252	16,15(4,41)	677,9

** Standard deviation is given in brackets.*

4. Conclusions

In this study, CFRTP filament was produced using a melt impregnation line. Three-point bending test samples were printed on an FDM-based additive manufacturing platform from these filaments. Test samples were subjected to three different heat treatments. Then, its mechanical properties were examined by a three-point bending test. The re-melt process yielded a peak bending strength of 217.72 MPa. An increase of 251.79% compared to the neat PLA test sample was achieved. On the other hand, microwave oven heat-treated samples achieved the lowest flexural strength of 202.58 MPa. In future work, mechanical properties can be improved by applying different heat treatments (hot press, etc.) or thoroughly investigating heat treatment parameters.

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