Comparison of Mechanical Properties of 3D-Printed and Compression-Molded Wood-Polylactic Acid (PLA) Composites

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Mechanical properties of materials obtained by compression-molding were compared with three-dimensional (3D) printed materials. Firstly, wood flour was added at different levels (0, 5, 10, and 20%) to polylactic acid (PLA) polymer to produce filaments. Then, mechanical test samples were printed from the produced filaments with a 3D printer. The produced filaments were made into pellets, and sheets were obtained by compression molding. According to the mechanical test results, it was observed that the tensile strength of the 3D-printed and compression-molded materials were close to each other, and the elasticity modulus of the compression-molded samples was higher than the 3D-printed samples. In the hardness comparison, the compression-molded specimens exhibited higher hardness values than the 3D-printed specimens. When the morphological properties of the materials were examined, it was seen that the cross-sections of the compression-molded materials were smooth and had less void than the 3D printed ones. The glass transition, crystallization, and melting temperatures of the materials did not change much with the processing methods. Only the 3D-printing process increased the crystallinity percentages.

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INTRODUCTION

Natural reinforcing materials (such as hemp, flax, and wood fibers) exhibit superior specific strength properties. Plant fibers, which are renewable materials, can be added to polymers to improve the properties of composites due to their superior properties. Polylactic acid (PLA) is an advantageous and widely used polymer compared to petroleum-derived polymers to its biodegradability, affordability, good mechanical properties, and strong dimensional properties. Like conventional thermoplastic polymers, PLA and PLA matrix composites are shaped by common molding processes such as injection and compression molding (CM). Conventional molding techniques may be suitable for mass production in large quantities, as they require very high investment costs (Meyva-Zeybek and Kaynak 2021). In addition, PLA and PLA-based products have machinability features with new manufacturing technologies such as three-dimensional (3D) printing (Tiersch and Monroe 2016; Chandran et al. 2021).

Although the CM process is a suitable choice for manufacturing simple flat products, additive manufacturing (AM) technology can produce products with complex
shapes by layering materials from computer-designed 3D models without the need for any molds (Zindani and Kumar 2019). The AM process has gained increasing momentum as a rapid prototyping tool for composites. On the other hand, the manufacturing of materials with complex geometric shapes requires a special production technology such as the AM process. In the AM process, changes can be easily implemented during manufacturing, and production can be more reasonable due to minimal waste. The most common fabrication method using the AM process is fused deposition modelling (FDM), which extrudes the thermoplastic filament (Ngo et al. 2018; Chansoda et al. 2021; Hendlmeier et al. 2021). When the mechanical properties of compression-molded and 3D-printed materials were compared, it was stated that the results obtained in the 3D-printing process were close to those of materials produced using conventional manufacturing techniques (Chandran et al. 2021). In addition, it has been reported in various studies that the injection molding (IM) process is more advantageous than 3D printing in terms of the mechanical properties (Dawoud et al. 2016; Kaynak and Varsavas 2019; Cisneros-López et al. 2020; de Toro et al. 2020; Zandi et al. 2020). In their study, Franchetti and Kress (2017) indicated that the AM process is more advantageous in terms of cost in productions up to 200 units, while the IM process reduces production costs in higher production quantities. In the same study, it was reported that the IM process is more advantageous than the AM process in terms of the production time. The AM process may have a slower production rate compared to the IM process, but the AM process offers some advantages, such as the ability to create rapid prototypes, single-unit production, and the manufacturing of highly complex parts (Franchetti and Kress 2017).

Today, unlike conventional processes, a lot of attention has been given to the production of natural fiber reinforced thermoplastic composites by the AM process. Several studies have reported on the successful printing of mixtures of various natural fibers with thermoplastic polymers on 3D printers (Le Duigou et al. 2016; Tao et al. 2017; Bhagia et al. 2020; Narlıoğlu 2021, 2022; Yang et al. 2021). The shaping methods of polymers or polymer matrix composites have a significant impact on the mechanical properties of the final products. It has been emphasized that the strength and elastic modulus values of 3D printed samples are higher than those of compression-molded samples (Meyva-Zeybek and Kaynak 2021). In another study, it was reported that the 3D-printed materials exhibited similar mechanical performance to compression-molded materials (Yu et al. 2019). It is expected that the wood-PLA composite properties will differ from each other due to the different heating and cooling periods of the polymer in the processes made with compression molding and 3D printing. There has been a lack of published studies on the comparison of the properties of 3D-printed composites with those produced by other shaping processes. For this reason, it is important to compare the properties of wood-PLA composites shaped by the 3D-printing and CM processes.

This study aimed to compare the mechanical properties of materials produced according to CM and AM methods. The production of the materials was carried out in multiple stages. In the first stage of the study, filaments were produced, and then the mechanical test samples were printed using a 3D printer. In the second stage of the study, pellets were produced from the same filaments to ensure a fair comparison and these pellets were shaped to sheets by a hydraulic press. Hardness, tensile, and bending strength tests of the 3D-printed and compression-molded materials were carried out. Scanning electron microscope (SEM) images were taken to determine the morphological properties of the materials. In addition to these, a differential scanning calorimeter (DSC) analysis of the materials was made and their thermal properties were compared with each other.
EXPERIMENTAL

Materials
Hornbeam (*Carpinus betulus*) wood was provided from the Zülkadiroğulları Corp. (Kahramanmaraş, Turkey). In addition, the PLA (Bioflex F 7510; the biobased carbon content (BCC) is > 80%, form: granules, colour: natural, density: 1.25 g/cm³) polymer was provided from the FKuR Plastics Corp. (Willich, Germany) for the production of materials.

Methods
The hornbeam wood was ground in a laboratory-type grinder (IKA MF10; IKA-Werke, Staufen, Germany). Then, the groundwood flour was classified using a sieve (LOYKA ESM 200; Akyol Trade, İstanbul, Turkey). Wood flour in the particle size range of 125 to 177 μm was used for the production of the composite material. The classified wood flour was dried in an oven at 103 ± 2 °C until completely dry before the production of the composite material.

*Production of filaments and pellets*

The wood flour at addition levels of 0, 5, 10, and 20% was mixed with the PLA polymer to produce the filaments and pellets (Table 1). The wood flour and PLA polymer were mixed by a co-rotating twin-screw extruder (Rondol, Stoke-on-Trent, UK) with a 1.75 mm die. The barrel temperature profile from the feeder to the die was set to 100, 130, 160, 180, and 190 °C. In addition, the screw speed was adjusted to 50 rpm. Filaments of the desired length were produced from the material with a diameter of 1.75 ± 0.1 mm formed out of the extruder die. To print the 3D samples, the extruded filaments were cooled with a fan and then spooled by a filament winder. In addition, the extruded filaments were formed into pellets between 1 and 2 mm for the sheet production by the CM method using the pelletizer (Machine No C3037, Rondol Technology LTD, Stoke-on-Trent, UK).

<table>
<thead>
<tr>
<th>Type of Process</th>
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<th>Wood Flour (%)</th>
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<td>3D20</td>
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<tr>
<td>CM</td>
<td>CM0</td>
<td>100</td>
<td>-</td>
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<tr>
<td></td>
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<td>10</td>
</tr>
<tr>
<td></td>
<td>CM20</td>
<td>80</td>
<td>20</td>
</tr>
</tbody>
</table>

*Production of the compression-molded test samples*

The sheets were produced from neat PLA pellets and wood-PLA pellets using a hydraulic press (4122 12-12H Manual Heated Press, Carver, Wabash, IN, USA) according to the ASTM D4703-10 (2010) standard. The pelletized samples were compressed for 15 minutes in a 250 x 250 x 3 mm mold at a pressure of 15 MPa and a temperature of 195 °C. Then, the temperature of the mold was cooled to decrease to room temperature with cold water circulation for 5 minutes. In addition, the sheets were cut according to the ASTM D638-14 (2014) and D790-17 (2017) standards. Six tensile and bending samples were obtained for each formulation in Table 1.
Production of the 3D-printed test samples

The samples were printed in the same dimensions as the compression-molded samples by using a 3D printer to determine the tensile and bending strength properties. A Sigma 3D (open source) printer with a 0.6 mm nozzle and Repetier-Host software (Hot-world GmbH, Willich, Germany) was used to produce the 3D printed samples. The infill density was set to 100%, the printing speed was 50 mm/sec, the layer thickness was 0.3 mm, and the raster angle was ±45° in the software. In addition, the printing temperature was maintained at 195 °C for the extruder nozzle and 55 °C for the bedplate throughout the printing process. For each filament group, six tensile and bending samples were printed in accordance with the ASTM D638-14 (2014) and D790-17 (2017) standards.

Determination of the mechanical properties

The mechanical properties (tensile and flexural strength) of the samples were determined according to the ASTM D638-14 (2014) and D790-17 (2017) standards with a universal testing machine (AL-UN; Alarge, Istanbul, Turkey). A digital hardness tester (LXD-D; Loyka, İstanbul, Turkey) was used to determine the Shore D hardness values of the samples. The samples were conditioned at a relative humidity of 50 ± 5% and a temperature of 23 ± 2 °C for 7 d before all the tests were performed.

Morphological characterization of samples

The SEM images were taken to investigate the relationship between the wood flour and the PLA polymer matrix. The fracture surfaces of the samples were scanned with a ZEISS Sigma 300 VP SEM (Carl Zeiss Microscopy, White Plains, NY, USA) after gold plating (Q150R Plus; Quorum Technologies, Laughton, UK).

Determination of the thermal properties

The DSC analysis of the samples was performed with a Shimadzu TA50 thermal analyzer (Kyoto, Japan). Samples with an average weight of 5 to 10 mg were subjected to a heating program from room temperature to 250 °C at a heating rate of 10 °C/min and under a nitrogen atmosphere of 50 mL/min for the DSC analysis. The degree of crystallinity ($X_C$) of the 3D printed and compression-molded materials were calculated from the DSC analysis values using Eq. 1 (Khan et al. 2018),

$$X_C(\%) = \frac{\Delta H_m - \Delta H_{cc}}{\Delta H^w_m} \times 100$$

where $\Delta H_m$ is the enthalpy of melting (J/g), $\Delta H_{cc}$ is the enthalpy of cold crystallization (J/g), $\Delta H^w_m$ is the enthalpy of melting for 100% crystalline PLA (93 J/g), and $w$ is the weight ratio of PLA in the materials (%).

Statistical analysis

The statistical significance of the numerical values of the mechanical tests samples was determined with the TUKEY test after analysis of variance (ANOVA) using the SPSS Version 22 software (IBM, Armonk, NY, USA). According to the statistical analysis results, the statistical difference ($p < 0.05$) between the test samples is indicated with different small letters in the mechanical test result graphics.
RESULTS AND DISCUSSION

Mechanical Properties of Samples

Figure 1 shows the flexural and flexural modulus of 3D-printed and compression-molded specimens. With the addition of wood flour to the PLA polymer, it can be seen that the flexural strength decreased in both processes. This can be attributed to the incompatibility between wood flour and the polymer matrix. It was observed that the flexural strength of the 3D-printed neat PLA samples was higher than that of the compression-molded samples. Among the composite samples, the highest flexural strength value was determined to be 78.5 MPa in the 3D-printed composite sample with 5% wood flour (3D5). In addition, the 3D-printed sample (3D5) containing 5% wood flour exhibited a higher flexural strength value compared to the compression-molded sample (CM5) containing the same percentage of wood flour. It was also observed that 3D-printed samples with 10% wood flour addition (3D10) and compression-molded samples with 5% and 10% wood flour addition (CM5 and CM10) showed statistically similar flexural strength values ($p > 0.05$). The lowest flexural strength value (44.6 MPa) was observed in the compression-molded sample with 20% wood flour (CM20). In addition, the flexural strength value of the 3D-printed sample that contained 20% wood flour (3D20) was determined to be 52.7 MPa.

![Fig. 1. Flexural strength and flexural modulus of the samples](image)

The flexural modulus increased with the addition of wood flour to the PLA polymer matrix. The average flexural modulus of the compression-molded samples was higher than the 3D-printed samples. The flexural modulus of the 3D-printed (3D0) and compression-molded (CM0) samples from the neat PLA was measured to be 1.69 GPa and 2.55 GPa, respectively. The highest flexural modulus value (3.35 GPa) was determined in the compression-molded composite sample with 20% wood flour added (CM20). In addition, the flexural modulus value of the 3D-printed composite sample that contained 20% wood flour (3D20) was determined to be 1.98 GPa.
In a study, the properties of the composites obtained by 3D-printing and CM of polyhedral oligomeric silsesquioxane (POSS) nanoparticle added composites were compared. The same study reported that 3D-printed samples from neat PLA and PLA/POSS composites exhibited higher flexural strength values compared to compression-molded ones. In the same study, the flexural strength values of 3D printed and compression-molded neat PLA samples were reported as 97.2 and 94.8 MPa, respectively (Meyva-Zeybek and Kaynak 2021). Another study reported that 3D-printed specimens exhibited higher flexural properties than compression-molded specimens due to the fiber orientation parallel to the direction of the nozzle movement (Yu et al. 2019).

Figure 2 shows the tensile strength and tensile modulus of the 3D-printed and compression-molded samples.

The tensile strength values decreased with the addition of wood flour to the neat PLA polymer. Yu et al. (2019) reported that a 3D-printed sample with 15% basalt fiber addition showed a higher tensile strength value than a compression-molded sample with the same percentage of basalt fiber addition. However, the tensile strength of other 3D-printed samples (0-5-10-20%) decreased compared to compression-molded samples (Yu et al. 2019). In another study, it was reported that 3D-printed composite samples containing 5% cork exhibit slightly lower tensile strength and tensile modulus values than compression-molded samples at the same percentage. In the same study, it was reported that the tensile strength values of compression-molded and 3D-printed samples from 5% cork added PLA composites was 31.81 and 30.53 MPa, respectively (Daver et al. 2018). The similarities and differences between the tensile strengths of 3D-printed and compression-molded materials can be attributed to there being less voids structure in compression molding as well as the orientation of the fibers in 3D printing. In a similar study, it was reported that the tensile strength values of the 3D-printed materials increased with the addition of parawood flour to the PLA polymer. In the same study, it was also observed that the strength properties of the 3D-printed materials were higher than those of
the compression-molded materials (Chansoda et al. 2021). In this study, it was observed that the tensile strengths of the 3D-printed and compression-molded samples from the neat PLA (3D0 and CM0, respectively) were statistically similar ($p > 0.05$). In addition, when the tensile strength values of the composite samples with the same ratio of wood flour were compared with each other in both processes, the values were found to be statistically similar ($p > 0.05$). The highest tensile strength value was determined to be 38.4 MPa in the compression-molded composite sample (CM5) that contained 5% wood flour, and 36.9 MPa in the 3D-printed composite sample (3D5) that contained the same ratio of wood flour. The lowest tensile strength value was measured to be 27.6 MPa in the compression-molded test sample with 20% wood flour (CM20), and it was 27.8 MPa from the filament that contained the same ratio of wood flour in the 3D-printed composite sample (3D20).

According to the tensile modulus comparisons of the 3D-printed samples, it was observed that they were similar to each other except for the 10% wood flour added 3D-printed sample (3D10). It was observed that the tensile modulus value of the 3D-printed composite sample that contained 10% wood flour was 2.15 GPa, and the tensile modulus values of the samples with 0, 5, and 20% wood flour were 2.01, 1.98, and 1.92 GPa, respectively. The comparison of the tensile modulus of the compression-molded samples, it was observed that the modulus values increased gradually with the addition of wood flour to the PLA polymer. The lowest tensile modulus value of the compression-molded samples was 2.12 GPa in the compressed sample from the neat PLA (CM0), and the highest modulus value was 2.64 GPa in the 20% wood flour added composite sample (CM20). The fused deposition modeling (FDM) type 3D-printing process is based on the principle of deposition of materials layer by layer, so voids may occur between layers. Therefore, 3D-printed materials exhibit a slightly lower tensile modulus than compression-molded ones (Chandran et al. 2021).

The Shore D hardness of the samples is shown in Fig. 3.

![Fig. 3. The Shore D hardness values of the samples](image-url)
The hardness values of the compression-molded samples were higher than those of the 3D-printed samples. The Shore D hardness values of the 3D printed samples are lower than those of the compression molded samples is attributed to the voids in their structure. In the 3D-printed samples, the lowest hardness value was 73.6 in the composite sample that contained 20% wood flour (3D20). The hardness values of the other 3D-printed samples were statistically similar ($p > 0.05$), except for the 3D-printed sample with 20% wood flour (3D20). In addition, the addition of wood flour to the PLA did not have much effect on the Shore D hardness value of the compression-molded samples, and the hardness of the samples was found to be statistically similar ($p > 0.05$). In a study, it was reported that the Shore D hardness of composites printed with different layer thicknesses from PLA composite filament that contained 30% beech wood flour was approximately 73 (Narlıoğlu 2021). In another study, the Shore D hardness values of PLA composites that contained corn straw powder were reported to be between 70 and 80 (Jiang et al. 2021).

**Morphological Properties of the Samples**

Figure 4 shows the cross-sections of the 3D-printed and compression-molded specimens from neat PLA (3D0 and CM0, respectively), in addition to the 3D-printed and compression-molded composite specimens with the best tensile and flexural strengths (3D5 and CM5, respectively). As can be seen in Fig. 4a, the cross-sectional image of the compression-molded sample from neat PLA (CM0) was smoother than that of the 3D-printed sample from neat PLA (3D0).

![SEM images of breaking surfaces](image)

**Fig. 4.** The SEM images (250× magnification) of the breaking surfaces of the a) compression-molded neat PLA sample (CM0), b) 3D-printed neat PLA sample (3D0), c) 5% wood flour added compression-molded sample (CM5), and d) 5% wood flour added 3D-printed sample (3D5)

Through 3D printing, the filament comes out of a narrow nozzle and stacks the extruded polymer so that voids form during printing and these voids remain unfilled (Yu et al. 2019). In this study, the cross-section image of the 3D-printed composite sample (3D5) had more voids than the compression-molded composite sample. The fewer voids in compression-molded materials have been attributed to high pressure. In addition, the compression-molded composite sample containing 5% wood flour (CM5) had a smoother cross-section appearance than the 3D-printed sample (3D5). The higher roughness of the cross-sections of the 3D printed samples was attributed to the high strength properties of
the samples. It was also observed that the wood flour was more uniformly distributed in the cross-sectional image of the 3D printed sample (3D5). Chandran et al. (2021) reported that the cross-sections of the compression-molded samples exhibited a homogeneous, smooth, and brittle texture with no significant voids. In contrast, multiple voids were observed in the cross-sectional images of the 3D-printed samples (Chandran et al. 2021).

**Results of the DSC Analysis**

The DSC curves of samples can be seen in Fig. 5. The glass transition temperature ($T_g$) of the 3D-printed and compression-molded neat PLA was observed at 60.5 and 58.8 °C, respectively. The melting temperature ($T_m$) peak of the 5, 10, and 20% wood flour added 3D-printed composites were 149.0, 150.3, and 148.6 °C, respectively. The $T_m$ of the neat PLA was 148.8 °C. In addition, the $T_m$ peaks of the 5, 10, and 20% wood flour added compression-molded composites were 149.9, 149.5, and 149.2 °C, respectively. The effect of the wood flour and the production process on crystallinity temperature ($T_c$), the $T_g$, and the $T_m$ are apparent in the DSC curves (Fig. 5).

![DSC curves of the 3D-printed and compression-molded samples](image)

Fig. 5. DSC curves of the 3D-printed and compression-molded samples

Daver et al. (2018) observed an increase in the crystallinity of PLA polymer with an increase in the cork content in the composite, and this increase induced nucleation in PLA. The $T_g$, $T_c$, $T_m$, and crystallinity percentages ($X_c$) of the samples are given in Table 2. In this study, it was observed that the percentage of crystallinity of samples increased with the 3D-printing process compared to the CM process. This increase may be attributed to the nucleation of PLA during the period when the materials are retained in the hot nozzle in 3D printing. In addition, it is estimated that the orientation of the molten PLA polymer according to the printing pattern during 3D printing and then its slow cooling causes the 3D-printed samples to exhibit higher crystallinity than the compression-molded samples. With the addition of 5% wood flour to the PLA polymer, the crystallinity percentage increased in the compression-molded samples, while it decreased slightly in the other compression-molded composites. In addition, in the comparison between the 3D-printed samples, the 20% wood flour added composite sample (3D20) exhibited a higher crystallinity percentage than the other 3D-printed samples. The 3D-printing and CM
processes had a minimal impact on the $T_g$, $T_c$, and $T_m$ values of the materials. Several researchers reported no significant difference in the thermal properties of 3D-printed, injection-molded, and compression-molded samples (de Toro et al. 2020; Meyva-Zeybek and Kaynak 2021).

**Table 2. DSC Analysis Values of the Samples**

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>$T_g$ (°C)</th>
<th>$T_c$ (°C)</th>
<th>$T_m$ (°C)</th>
<th>$X_{cr}$ (%)</th>
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<tbody>
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**CONCLUSIONS**

1. According to the mechanical test results, the breaking stress values decreased in both processing methods with the addition of wood flour to the PLA.

2. With the addition of an equal amount of wood flour, the flexural strength properties of the 3D-printed samples were higher than the compression-molded samples, and the tensile strength values of the compression-molded and 3D-printed samples were close to each other. In addition, the compression-molded samples exhibited a higher modulus of elasticity at equal amounts of wood flour addition.

3. The cross-sections of the compression-molded materials were smoother and had a lower void content because the molten materials were densely packed as a result of the applied high pressure to the mold.

4. The materials exhibited different crystallinity due to different melting and cooling in the two processes. The DSC crystallinity percentages of the materials increased after the 3D printing.

**REFERENCES CITED**


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