# Fused Filament Fabrication of PLA: The Role of Interlayer Adhesion on the Mechanical Performances

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## 1. Introduction

Additive Manufacturing (AM) techniques have triggered industrial innovations thanks to their ability to realize objects with so complex shapes that are impossible to obtain adopting conventional processes. AM is based on the bottom-up approach, during which the construction of parts is conducted layer by layer. Among the available techniques, fused filament fabrication (FFF) is devoted to the production of plastic parts. FFF can be spread out into four main stages: polymer melting inside the extruder, extrusion through the nozzle, deposition of the filament on the deposition plate and, subsequently, construction layer by layer. Up to now, FFF has been applied to the production of prototypes, medical devices and final parts for automotive and aerospace industry [1,2]. The parts generally exhibit poor and anisotropic mechanical performances, which make them not suitable for industrial applications and wherever structural properties are required. This drawback has been generally attributed to the layer-by-layer deposition approach. The filament is deposited on a previously deposited layer that is undergoing cooling: this induces the formation of a weld part between adjacent layers. The overall mechanical part resistance depends on the interlayer bond strength [3]. In turn, the interlayer bond strength depends on the molecular diffusion at the interface, namely on the possibility of the molecules to recover their entangled organization, also at the interface, after deposition. To further understand how the thermal history of the deposited layers is correlated to morphology and mechanical properties of the produced FFF object, the role played by crystallization phenomena, and a molecular weight between the entanglements must be considered. To this purpose, in the presented work, the morphology and the mechanical properties of FFF specimens have been correlated to the thermal history experienced by the polymer during deposition, including the role played by molecular diffusion in terms of molecular weight between entanglements. The results are aimed at investigating main phenomena that determine the performances of the parts. The final goal to formulate a mathematical model of FFF, to reduce the needs for extensive experimental campaign in the future.

## 2. Methods

Polylactic acid (PLA, grade 4032D, NatureWorks, Minneapoli, Minnesota, US) in the form of filament was adopted to perform FFF [4]. A Kloner3D 240 Twin (Kloner3D, Firenze, Italy) was used to produce the samples at 200°C nozzle temperature, with 100% infill density, without raft and supports. Two deposition velocities were adopted: 30, and 50 mm/s, and two temperatures of the bed: 60°C, and 80°C.

The layer thickness of the first layer was set constant to 0.1 mm. The razor direction of the adjacent layer was 90◦. The shape and size of the samples (Figure 1) have been conveniently designed so that the deposition of the filament would be performed along a single direction for each wall. Each wall is formed by 3 filaments.

T-type thermocouples (d = 120 μm, Omega Engineering, Manchester, UK) were added along the deposition layer to monitor the evolution of the temperature at the interface between adjacent layers during the process. A thermocouple datalogger (TC-08, Pico Technology, St Neots, Cambridgeshire, UK) was adopted for temperature recording.

Dynamic mechanical analysis (DMA, Perkin Elmer DMA 8000) was conducted on the specimens to assess the mechanical behavior and evaluate the molecular weight between entanglements (the same procedure is reported in the literature [5]). The specimens were cut in a plane parallel to the building plate at 1, 2 and 3 cm distance from the plate, to inspect the accuracy of the deposition among consecutive layers; a cross-cut in a plane orthogonal to the direction of deposition was performed to investigate the occurrence of possible detachment.

The specimens were observed by an optical microscope (Eclipse L150, Nikon, Tokyo, Japan) equipped with a digital camera.

Atomic Force Microscopy (AFM, Veeco, Santa Barbara, CA, USA) was adopted to analyze the behavior of the material at the interface between adjacent layers [5].

## 3. Results and discussion

Figure 2 shows the evolution of the temperature recorded at the interface between adjacent layers at different distances from the deposition plate (see Figure 1). As the distance from the deposition plate increases, the average temperature decreases: at the deposition plate the average temperature is about 60°C (the set temperature), at 1 cm and 2 cm from the deposition plate the average temperatures are about 54°C, and 50°C, respectively. Furthermore, the deposition process determines a sudden increase of temperature of the previously deposited layers: the presence of several temperature peaks for the same position confirms this statement. The trend of temperature is time-shifted depending on the processing speed. Temperature distribution is expected to influence the distribution of the mechanical properties of the parts.

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| Figure 1 Sketch of the shape of the FFF specimen. | Figure 2 Temperature evolutions for the specimen obtained with 60°C deposition temperature and 30 mm/s as deposition velocity. |

The morphology of the specimens was analyzed by optical microscopy to preliminary assess the dependence of the adhesion between adjacent layers on the operative conditions.

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| 30 mm/s; 60°C | 50 mm/s; 60°C | 50 mm/s; 80°C |

Figure 3 Optical micrographs of the specimens cut close to the deposition plate (at the bottom), for the cases obtained with several deposition velocities and two deposition temperatures, 60°C and 80°C.

The higher is the deposition velocity, the lower is the adhesion between adjacent layers. Particularly, the specimen obtained with 50 mm/s deposition velocity shows a clear detachment between the third and the fourth layers. The increase of the deposition temperature seems to reduce this phenomenon. Further investigations at the locations of detachment were conducted by means of AFM (Figure 4) for the specimens obtained with 60°C deposition temperature and different deposition velocities. Three AFM maps are shown: the height map, concerning the description of the topography, the phase map, and the elastic modulus map, concerning the mechanical performance of the parts. The height maps confirm the presence of a detachment in both cases; however, the detachment is more evident (therefore, the distance between the layers is wider) in the case obtained with 50 mm/s deposition velocity. The phase and the elastic modulus maps allow detecting differences in the mechanical behavior where the detachment occurs: where the detachment occurs, the elastic modulus is smaller. The distribution of the elastic modulus is almost homogeneous for the specimen obtained with 30 mm/s deposition velocity; instead, the elastic modulus is significantly lower at the detachment positions, than in the surrounding area, for the specimen obtained with 50 mm/s.

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|  |  | height | Figure 4 AFM of the specimens obtained with the same deposition temperature, 60°C, with two deposition velocities, 30 mm/s and 50 mm/s. The analyses are referred to interface between the third and the fourth layers already shown in Figure 2 (the white arrows indicate the distance between layers at the detachment). |
| phase |  |
| modulus |  |
| 30 mm/s 60°C | 50 mm/s; 60°C |  |  |

The DMA analyses aim at understanding the effect of the deposition conditions and the distance from the deposition plate on the mechanical performance of the parts. The results are given in Table 1 in terms of elastic modulus and molecular weight between entanglements, for two specimens obtained with 60°C deposition temperature.

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|  | **position** | **E [GPa]** | **Me [g/mol]** | Table 1 Elastic modulus (E), and molecular weight between entanglements (Me) for the specimens obtained with 60°C deposition temperature, and two different deposition velocities, 30 mm/s and 50 mm/s. The positions where the analysis was conducted with respect to the distances form the deposition plate are consistent with those indicated in Figure 1. |
| **30 mm/s** | **3 cm** | **1.43 ± 0.05** | **2151** |
| **2 cm** | **1.50 ± 0.02** | **1901** |
| **1 cm** | **1.26 ± 0.05** | **1901** |
| **ortho** | **1.47 ± 0.05** | **2089** |
| **60 mm/s** | **3 cm** | **1.64 ± 0.05** | **2224** |
| **2 cm** | **1.55 ± 0.02** | **2248** |
| **1 cm** | **1.52 ± 0.05** | **2563** |
| **ortho** | **1.33 ± 0.05** | **2260** |

Two directions were analyzed, the first one is parallel to the deposition direction (positions 1, 2, and 3 cm), the second one is the direction transversal to the deposition direction (ortho). With reference to the positions parallel to the deposition direction, the elastic modulus increases with the deposition velocity, and, within the same specimen, with the distance from the deposition plate. The deposition velocity and the cooling time determine the residual orientation in the final part. On its turns, the orientation determine the elastic modulus: the higher is the residual orientation, the higher is the elastic modulus [6]. The molecular weight between entanglements, Me, can be used as an index of the residual orientation: the higher is Me, the higher is expected to be the orientation, the wider is the distance between entangled molecules. Me increases with the deposition velocity, thus the increase of this operating parameter induces more evident orientation in the molecules, whose relaxation is hidden by the fast cooling. The decrease of the average temperatures at longer distance from the deposition plate contributes to hide the molecular relaxation; consequently, Me increases with the distance from the deposition plate. The increase of Me, and orientation, determine the increase of the elastic modulus, in turn.

Concerning the positions along the direction transversal to the deposition direction, the elastic moduli are lower than those detected for the specimens cut along the deposition direction. This finding is consistent with the previous discussion since the elastic modulus along the transversal direction is influenced by the presence of detachments between adjacent layers. In this direction, Me is high and comparable with the values measured for the specimens cut along the deposition direction at 3 cm from the deposition plate. A high residual orientation is not favorable to the molecular diffusion at the interface between adjacent layers, the molecules solidify before diffusion may occur determining poor mechanical performances (i.e small elastic modulus values).

## 4. Conclusions

FFF was conducted under different operating conditions, concerning the deposition temperatures and the deposition velocity with the aim of analyzing the effect of these parameters on the mechanical performances of the parts. To this purpose, several investigations were performed, adopting AFM and DMA to assess the mechanical characteristics. AFM analyses revealed that the effect of the detachment increases with the increase of deposition velocity; the increase of the deposition temperature partially compensate this effect. The DMA analyses contributed to determine the residual orientation within the specimens and to relate the orientation to the elastic modulus. The increase of the deposition velocity, coupled with the fast cooling experienced during the process, results in high residual molecular orientation in the final parts. The orientation increase, on its turn, determines the increase of the elastic modulus along the deposition direction. On the other side, the increase of the orientation prevents the molecular diffusion at the interface between adjacent layers, determining the detachments of adjacent layers. Consequently, the elastic modulus along the direction transversal to the deposition direction is smaller than the ones measured along the deposition direction. These finding suggest that, to produce isotropic objects with high mechanical performances it is crucial to allow the molecular relaxation, to favor the molecular diffusion. Therefore, where high deposition velocities are required to the purpose of increased productivity rate, the temperature must be conveniently shifted to allow relaxation in the whole deposited volume. These findings are intended to offer a solid basis for the implementation of a simulation tool to predict the temperature evolution of the deposited layer during FFF and extract the mechanical properties. A big effort has already been devoted in this direction, and the main finding will be shared in future works.

## References

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