



# Single-layer temperature-adjusting transition method to improve the bond strength of 3D-printed PCL/PLA parts

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## ABSTRACT

Although multi-material fused deposition modelling (FDM) has shown good progress and potential for industrial and scientific development, differences in physical and chemical properties cause weak bond strength between dissimilar materials in multi-material FDM parts. This paper proposes a single-layer temperature-adjusting transition (SLTAT) method to improve bond strength between dissimilar materials with different melting temperatures by adjusting the bonding-layer temperature. Herein, the bonding-layer temperature effects on the tensile strength of polycaprolactone (PCL)/polylactic acid (PLA) structures were investigated. PCL/PLA parts prepared with this method had 28% higher tensile strength than unprocessed parts when the bonding-layer temperature was 130 °C. Bonding mechanism was proposed to explain the failure modes of the PCL/PLA parts after tensile testing. Freeze-fractured surfaces of SLTAT-processed PCL/PLA specimens were observed to better understand the correlations between bonding-layer temperature and tensile strength. This approach is promising to apply in ordinary multi-material FDM processing without adding additional equipment or compromising dimensional accuracy.

## 1. Introduction

Due to the significant advantages of additive manufacturing (AM) technologies, such as rapid prototype production, low labour costs, and customization freedom, AM has been widely adopted by modern manufacturing facilities in recent decades. However, in modern society, the manufacturing industry is rapidly evolving. Homogenous parts manufactured by legacy single material 3D printers may not be able to meet all the demands of today's products. Since part performance can be boosted using multiple material systems [1], multi-material additive manufacturing (MMAM) has been considered a promising method [2]. Although MMAM has many advantages, such as improved mechanical properties, one-step manufacturing, and access to desired properties [3], many challenges of MMAM remain to be addressed.

Fused deposition modelling (FDM), in which a polymer material is deposited layer-by-layer to construct 3D parts, is one of the most commonly used techniques among all the AM technologies. Due to its advantages of low cost, simple mechanical structure, and easy material change [4], FDM has been widely applied in MMAM. An FDM 3D printer can be easily modified into a multi-material 3D printer by integrating two or more extrusion nozzles. The typical multi-material

FDM process makes use of two nozzles working together to deposit soluble support and model materials. Additionally, multi-material FDM has been used to manufacture 3D-printed polymer parts with additional reinforcement fibres to enhance their structural strength [5]. Other research has focused on coupling FDM with other AM technologies, such as direct ink writing (DIW), inkjet and electrospinning, to manufacture porous scaffold structures for tissue engineering [6].

Although multi-material FDM technology has exhibited good progress and vast potential for future development, the weak bond strength between adjacent extruded filaments and layers limits its application in industry. Furthermore, the bonding between different materials could be a much more difficult problem in multi-material FDM than in single-material FDM, as it may even cause the collapse of parts due to differences in the physical properties of materials (for example, melting temperature, thermal expansion rate) and chemical properties (for example, chemical bonding, van der Waals forces) [7].

A significant amount of effort has been made to improve the inter-filament and inter-layer bond strength of FDM parts. The approaches can be divided into two major groups: processing parameter optimization [8–16] and extra external energy input [17–20]. Several studies have focused on discovering the relationship between FDM process

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**Table 1**  
Properties of PLA and PCL filaments.

	Symbol	PLA	PCL
Tensile strength (MPa)	$\sigma_b$	60	18
Elongation at break (%)	$\delta$	3.0	11
Flexural strength (MPa)	$\sigma_{bb}$	97	29
Flexural modulus (MPa)	$E$	3600	940
Density (g/cm <sup>3</sup> )	$\rho$	1.25	1.16
Melting temperature (°C)	$T_m$	180	60
Glass transition temperature (°C)	$T_g$	60	–60
Decomposition temperature (°C)		280	230

parameters (for example, nozzle temperature, layer thickness, raster angle, and air gap) and the mechanical strength of printed parts [9–16]. Several other studies improved inter-filament and inter-layer bond strength by introducing external heat directly at the deposited surface, such as heated air [17], IR laser [18], IR lamps [19], and microwave heating [20]. All these methods were aimed at heating the deposited polymer surface above its critical sintering temperature ( $T_g$ ) so that molecular diffusion between the deposited layer and the next layer can occur [21].

From the above discussion, many studies have been carried out to improve the inter-filament and inter-layer bond strength of FDM parts; however, few studies have focused on the bonding mechanism between the interfaces of different melting temperature materials in multi-material FDM processes. Polylactic acid (PLA, melting temperature of 180 °C) and polycaprolactone (PCL, melting temperature of 60 °C) are two common materials used in the FDM process. The two materials are biocompatible and biodegradable but have different elastic moduli. The flexural modulus of the PLA material is much higher than that of PCL material (Table 1). Thus, introducing PLA filaments as a reinforcement material during multi-material FDM process with PCL materials can significantly increasing the stiffness of PCL parts. On the other hand, the presence of PCL materials can offer a certain extent of elasticity to the composite parts (made of PLA and PCL materials), which is not available for neat PLA parts. Therefore, manufacturers can combine these two materials by using multi-material FDM technology to generate an integrated architecture with desired mechanical properties in distinct areas, such as flexible manipulators, bionic bone structures and stiffer scaffolds for tissue engineering. As shown in Fig. 1, the combination type (Z direction) of PLA and PCL can be classified into two types: PLA above PCL and PCL above PLA. For clarity, the former will be referred to as the PLA/PCL type (Fig. 1(a)) and the latter as the PCL/PLA type (Fig. 1(b)) throughout the paper. For the PLA/PCL type structure, when molten PLA filaments are deposited on an existing PCL layer, the significant difference in melting temperature between PLA and PCL causes the PCL part to begin to melt, which produces defective structures. Hernan et al. [6] introduced a controlled cooling system to allow the combination of polymers with different melting temperature and

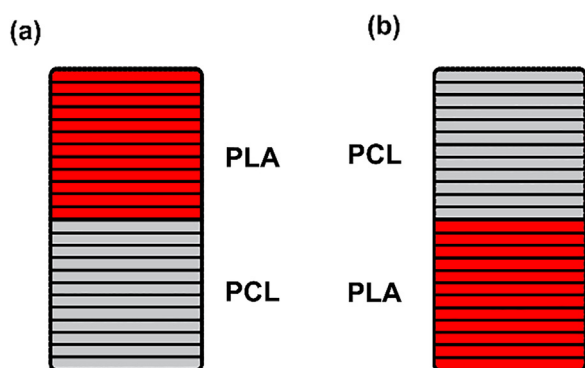


Fig. 1. Combination type. (a) PLA/PCL type; (b) PCL/PLA type.

showed that the cooling system improved the integrity of the PLA and PCL mesh. However, the bond strength between PLA and PCL layers has been found to decrease due to the reduction of the inter-layer adhesion time and crystallinity of the rapid cooling polymer. For the PCL/PLA type structure, when PCL filaments are extruded on an existing PLA layer, the contact heat is insufficient to raise the top PLA layer above its  $T_g$ , so no or little polymer diffuses across the PCL/PLA interface [21], which leads to relatively low interfacial bond strength. To the best of our knowledge, this issue has rarely been studied directly. Although the introduction of external heat, as mentioned earlier, can solve this problem to some extent, these methods require additional complex devices or post-processing steps. The input of heat is also difficult to control, and excess heat may cause the deposited part to melt and collapse [17].

In this paper, in order to improve the bond strength of PCL/PLA parts, a single-layer temperature-adjusting transition (SLTAT) method was firstly proposed. The effects of temperature of the PCL bonding layer (the PCL layer above the PLA layer) on the tensile strength of SLTAT-processed PCL/PLA parts were investigated. And bonding mechanism was proposed to explain the failure modes of the SLTAT-processed PCL/PLA parts after tensile testing. Finally, the freeze-fractured interfaces of PCL/PLA parts with different bonding layer temperatures were observed with an optical microscope to further understand the correlations between bonding-layer temperature and tensile strength. Successfully accomplishing such an investigation will provide knowledge of the mechanical improvement in parts obtained from multi-material FDM processes using different melting temperature materials.

## 2. Experimental

### 2.1. The principle of SLTAT method

The principle of SLTAT method is shown in Fig. 2. First, a PLA print-head is used to construct the PLA part on a platform. When the deposition of the PLA layers is completed, the 3D printer switches to the PCL print-head and then deposits the PCL bonding layer at a nozzle temperature that is higher than the ordinary nozzle temperature for PCL material (which is 110 °C in this paper). After the PCL bonding layer is deposited, the nozzle temperature is readjusted to 110 °C to extrude subsequent PCL layers until the PCL/PLA part is finished. This process raises the nozzle temperature of the PCL bonding layer to heat only the top PLA layer above its  $T_g$  while keeping the 3D printing parameters of other layers the same. Therefore, some defects caused by excessive heat input, such as collapse and inaccurate dimensions, can be effectively avoided. In addition, the SLTAT method can be easily applied to existing multi-material 3D printers without adding additional heating equipment or post-processing steps, which shows that it is more practical than other methods.

### 2.2. Equipment and materials

#### 2.2.1. 3D printing platform

As shown in Fig. 3a, a 3D printer with an automatic multi-tool changer (AMTC) system, which was developed in our previous work, was employed in the present research. During the printing process, the 3D printer can automatically change the print-head module with different materials or even different extrusion methods (such as FDM or DIW) from the AMTC (Fig. 3b) according to the received commands. Therefore, it has the ability to manufacture integrated parts with two or more different materials.

The materials used in the present research were PLA and PCL filaments, so two FDM print-head modules (Fig. 3c) were pre-installed in the AMTC. When the print job is started, the 3D printer automatically loads the required print-head module according to the received commands; then, the motion mechanism moves parallel to the X-Y plane over a building platform. Simultaneously, the filaments are forced into the liquefier by a stepper motor, heated to a semiliquid state, and then

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