Influence of layer thickness and nozzle temperature on the interlocking adhesion strength of additive manufactured multi-material interface

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Abstract

Achieving adequate adhesion strength at multi-material interfaces is always a challenge in material extrusion additive manufacturing (MEAM), especially when the materials have very different chemical affinities. This study investigated the adhesion mechanism of multi-material interfaces in MEAM from a micro-geometric perspective. The vertically printed interface was found to have a smooth surface, while the horizontally printed interface had a micro-zigzag interlocking geometry. The formation of this micro-zigzag interlock is due to the switching of extruders during printing, which mechanically reinforces the interface adhesion strength. Using butt-joint tensile test and microscope observation, it was found that the geometry of this zigzag interlock is significantly influenced by the layer thickness, nozzle temperature and extruder offset. By optimizing the layer thickness and nozzle temperature, the interface adhesion strength between dissimilar materials was increased by 58.2% without significantly increasing the printing time or fabrication complexity.

Keywords: Fused deposition modeling (FDM), Interface adhesion strength, Microstructure, Processing

Introduction

The development of multi-material additive manufacturing(AM) technologies allows the fabrication of multi-functional structures by printing different materials into the same component [1]. Nowadays, one of the most widespread MMAM technologies is multi-material fused deposition modeling (MMFDM), which is usually implemented by equipping FDM machines with multiple extrusion nozzles [2]. However, achieving sufficient interfacial bonding strength between materials with dissimilar chemical properties is always a challenge in MMFDM due to its discontinuous nature: a molten filament is extruded and deposited onto the previously deposited layer, forming bonds with adjacent filaments.

In mono-material parts, the formation of bonding between adjacent filaments was divided into three processes based on the polymer welding theory: 1) surface contacting; 2) neck growth; 3) molecular diffusion at interface and randomization (entanglement) across the inter-filament interface [3]–[5]. During printing, coalescence of neighbor filaments starts instantaneously after surface contact, resulting in the growth of neck length, where surface tension is the driven force and viscous and inertial forces are resistant forces [4], [6]. Meanwhile, the interfacial bond strength continues to grow as polymer chains diffuse across the contacted interface until $T < T_g$ [6]. Many studies concluded the weakness of mono-material interfacial bond strength in FDM to the insufficient interfacial molecule diffusion [6]. While other studies arguing that microstructure of the interlayer contact rather than incomplete molecular bonding is the predominant cause of strength anisotropy in FDM [7]. It's reported that for a range of materials, printing conditions and microscale geometries, bulk-material strength can be achieved by taking actual geometry of interface into strength calculation[8]. A comprehensive model which combined interlayer contact model with a diffusion model was developed by Coogan and Kazmer [9] to predict the interlayer strength. The result suggested that the interfacial bond strength of acrylonitrile butadiene styrene (ABS) parts was governed by limited interlayer diffusion due to the long relaxation time of ABS, while the high-impact polystyrene with shorter relaxation time had complete diffusion and was constrained by a lack of interlayer contact.

The formation of multi-material interfacial bonding can also be described by the polymer weld model but is influenced by many other factors. Firstly, the materials at interface can have very different chemical properties. The mechanical integrity of the interface is expected to be reduced when the different printed materials have low chemical affinity, such as polylactic Acid(PLA) and thermoplastic polyurethane(TPU) [10]. Secondly, the switch of extruders leads to additional time of cooling which changes the thermal history of printing. Finally, the multi-extruder deposition process introduces extra geometrical errors during printing [10]. No matter how carefully the alignment process is done, there will always be offsets between these extruders, leading to the separation or overlap of printed components. New experiments and models should be developed to take the effects of these new factors into account.

Recently, the effect of process parameters on the multi-material interfacial strength was studied in several research. Yin et al. investigated the effect of nozzle temperature, bed temperature and print speed on the adhesion strength of ABS-TPU interface. It's found that the adhesion strength can be doubled by increasing the bed temperature from 30 to 68°C. Tamburrino et al. [11] reported that the decrease of infill ratio has a negative effect on the multi-material interface adhesion strength. Other studies devoted to affix materials with weak chemical bond with 2D or 3D mechanical interlocking such as T-bones and dovetail interlocks [12], interlaced topologically interlocking lattice [13] and triply periodic minimal surface [14]. These macro-scale mechanical interlocks significantly increase the adhesion strength between dissimilar materials but make the manufacturing process more complex.

Although the effect of microstructure on the adhesion strength of mono-material interfaces has been shown to be significant, its effect on the strength of a multi-material interface has not been studied in the literature. This study aims to investigate the effect of microstructure on the interface adhesion strength between materials with dissimilar physical and chemical properties. The effect of two key process parameters, namely nozzle temperature and layer thickness, was investigated. Butt-joint tensile test and microscopic observation were performed to evaluate the adhesion strength of the printed multi-material interfaces.

Materials and methods

In this study, the interface adhesion strength between a typical rigid material, PLA [15] and a semi-flexible material, TPU [16], were investigated using butt joint tensile test and microscope. The combination of these two materials offers a possibility to develop functional structures. However, their vastly different physical and chemical properties make achieving sufficient interfacial adhesion strength a challenge.

Experiment and specimen design

A universal tensile testing machine was used to study the tensile strength of the PLA-TPU interface. All tests were performed at a loading speed of 5 mm/min. The test length between the wedge screw grips was 40 mm and the measured length between the extensometers was 25mm (Figure. 1(a)). To avoid the influence of extruder offset on the interface strength, a horizontally printed dual-interface specimen was designed so that the specimen always fractured at the weaker interface (Figure. 1(b)). Red stands for PLA and grey stands for TPU. A vertically printed single-interface butt-joint specimen was used as reference specimen. The total length, width and thickness of these two specimens were the same. As demonstrated in Figure. 1(c) and (d), the geometry of printed interface is controlled by layer thickness (h) and extrusion width (w). The interfacial geometry is supposed to be significantly influenced by the build orientation even if layer thickness and extrusion width are the same.

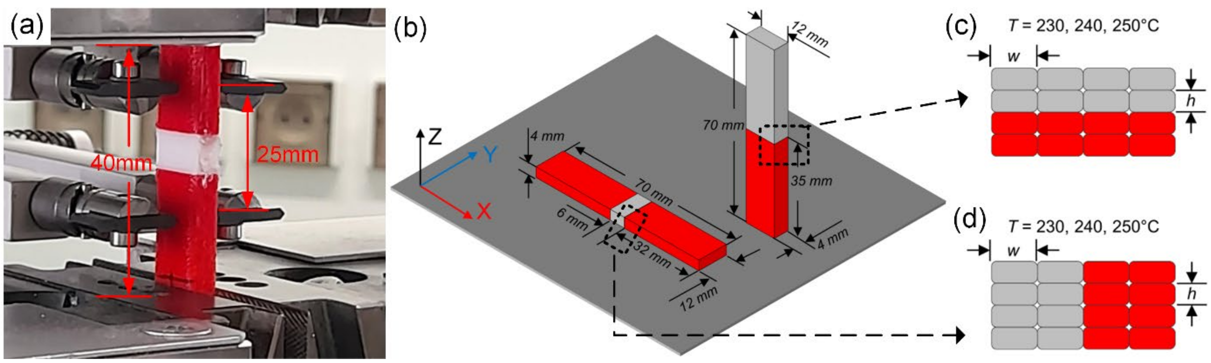

Figure. 1 Fabrication and butt-joint tensile test of specimens. (a) Tensile test machine; (b) Geometries of specimen; (c) Interface of vertically printed specimen; (d) Interface of horizontally printed specimen

Specimen fabrication and experiment design

Table. 1 Experiment design

Specimen	Repetition	T (°C)	h(mm)	Build orientation
1	4	230	0.1	Horizontal
2	4	230	0.2	Horizontal
3	4	230	0.3	Horizontal
4	4	240	0.1	Horizontal
5	4	240	0.2	Horizontal
6	4	240	0.3	Horizontal
7	4	250	0.1	Horizontal
8	4	250	0.2	Horizontal
9	4	250	0.3	Horizontal

A commercial dual-extruder FDM printer Raise 3D E2 was used for the manufacturing of specimens. As reported in the literature, the nozzle temperature (*T*) has a significant influence on the neck growth and intermolecular diffusion at the interface. The layer thickness (*h)* controls the geometry of the interface. To investigate the effect of these two process parameters on the tensile strength of PLA-TPU interface, dual-interface specimens were printed at three levels of *T* and *h* with 4 repetitions (Table. 1). *T* stands for the nozzle temperature at which TPU was printed. PLA was always printed at a nozzle temperature of 210°C. 4 single-interface specimens were printed vertically as a reference for the horizontally printed dual-interface specimens. Other process parameters were the same for all the specimens: infill ratio, 100%, raster angle, 90°; extrusion width, 0.4 mm; bed temperature, 60°C; number of contours, two; fan speed, 50%. Each type of specimen was printed with an additional specimen for microscopic observation.

Results and discussions

Interface geometry of specimens

Specimens were cut and polished to be observed under optical microscope. It was found that the horizontally printed PLA – TPU interface had a micro-zigzag interlocking geometry (Figure. 2(a)(b)), while the vertically printed interface had a smooth surface (Figure. 2(c)). Comparing the left and right interface (Figure. 2(a) and (b)) of horizontally printed specimen, it's found that both of them have zig-zag interlock geometry, while the left interface has a much deeper interlock. This indicates that the actual offset of the TPU extruder was shifted too far to the left. Furthermore, similar micro-zigzag interlocking was also found in the horizontally printed PLA-PLA interface (Figure. 2(d)). This suggests that this zigzag interlocking is a general effect of the printing process, independent of the material.

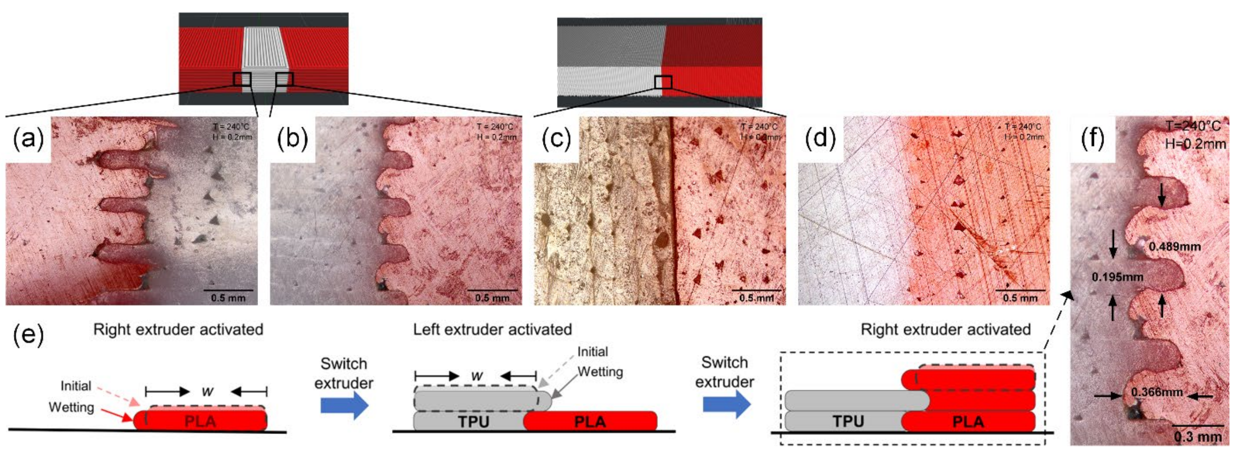

Figure. 2 Geometries of printed interfaces. (a) Right interface and (b) left interface of horizontally printed dual-interface specimen; (c) interface of vertically printed specimen; (d) interface of PLA-PLA interface; (e) formation process of the zig-zag interlock; (f) Geometry of the PLA-TPU interface.

As shown in figure. 2(e), the formation of this interlocking was supposed to be a result of extruder switch and material wetting. To save the printing time, two layers of material are deposited after each extruder switch in a standard multi-material FDM process. Then, cooling fans are used to accelerate the solidification of the deposited beads. The expansion of the beads deposited in the first layer is constrained by the beads previously deposited by the other extruder.

However, in the second layer, the bead deposited next to the interface is expanded due to material wetting, during which surface tension is the driven force and viscous and inertial forces are resistant forces. The repetition of this process resulted in the zig-zag interlock interface observed in Figure. 2. In vertically printed specimens, only one time of extruder switch was happed which resulted in a smooth interface.

Mechanical response under tensile loading and microstructure of the interfaces

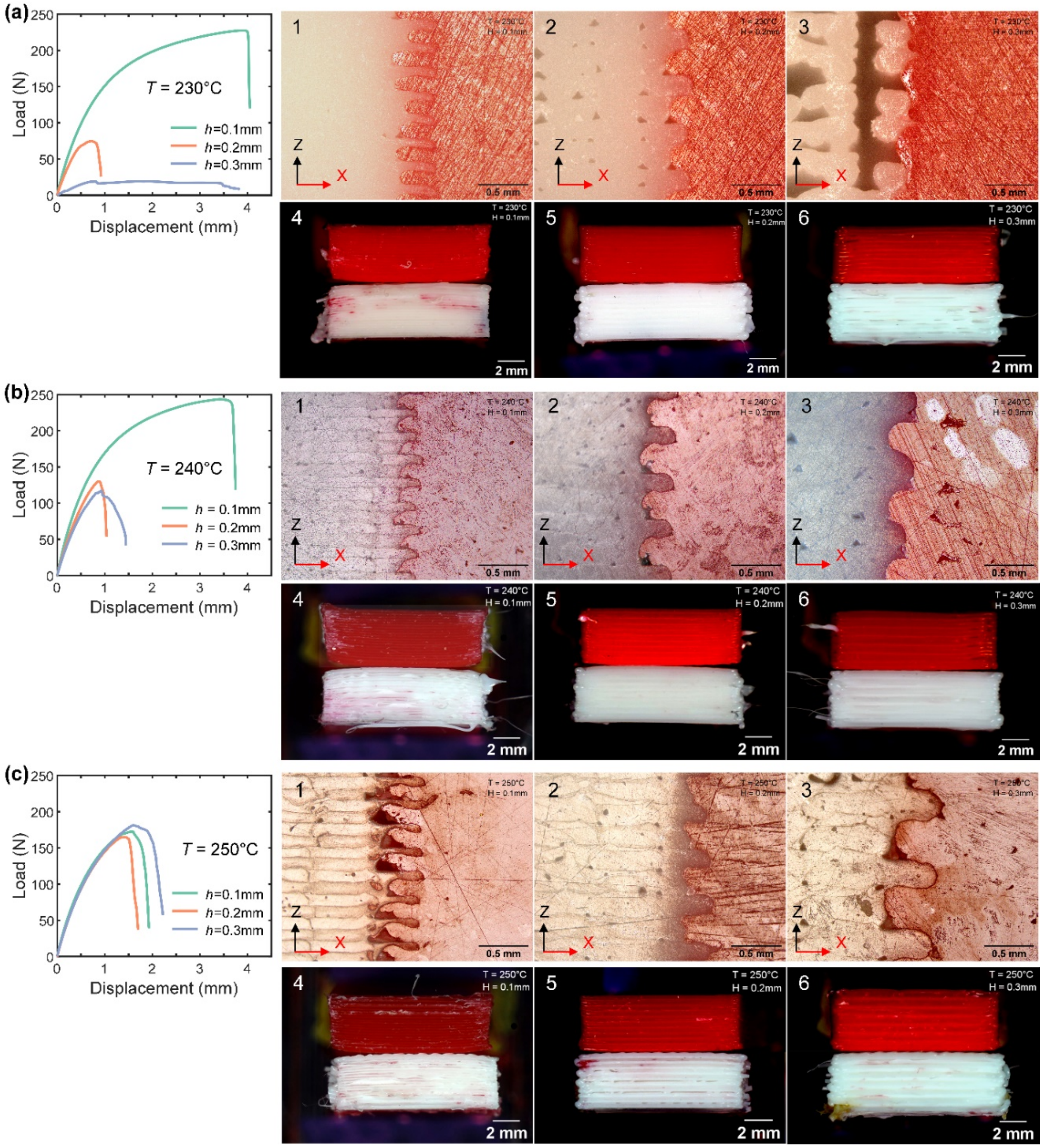

Figure. 3 Load-displacement curves, corresponding microstructures and fracture surfaces at interfaces. (a) Specimens printed at T = 230°C with (1)(4) h = 0.1 mm, (2)(5) h = 0.2 mm, (3)(6) h = 0.3 mm; (b) Specimens printed at T = 240°C with (1)(4) h = 0.1 mm, (2)(5) h = 0.2 mm, (3)(6) h = 0.3 mm; (c) Specimens printed at T = 250°C with (1)(4) h = 0.1 mm, (2)(5) h = 0.2 mm, (3)(6) h = 0.3 mm. Red part is PLA and while part is TPU. The direction of the deposited filaments is orthogonal to the surface of microscopic images.

Figure. 3 shows the typical load-displacement curves of specimens under quasi-static tensile loading with T = 230°C, 240°C and 250°C. The corresponding microstructures and fracture surfaces of interface were also presented. In general, the interfacial adhesion between PLA and TPU formed upon printing is not strong enough so that all the specimens failed at the interface region. All the specimens have a micro-scale zig-zag interlock geometry at the interface.

Figure. 4 shows the interface strength of specimens influenced by T and h. The interface strength was calculated by dividing the maximum tensile load by the cross-sectional area of the specimen (Table. 2). In the cases of T = 230°C and 240°C, the decrease in h from 0.3 to 0.1 mm significantly increases the interface strength (Figure. 4). Particularly, at T = 230°C, the interface strength of specimens printed at h = 0.1 mm is 12 times of that of the specimen printed at h = 0.3 mm. Meanwhile, under microscope, the geometry of interfaces printed at smaller layer thickness have deeper zig-zag geometry and smaller pores. Therefore, the contact length between PLA and TPU is much longer at smaller layer thickness, which results in a stronger mechanical interlock and explains the increase in interface strength. From another aspect, more residual materials can be found in the fracture surfaces of specimens printed at h = 0.1 mm (Figure. 3(a)(1), (b)(1) and (c)(1)). This indicates that specimens printed at smaller h has stronger zig-zag interlock. However, in the cases of T = 250°C, the interface strength is quasi-constant as h decreases from 0.3 to 0.1 m (Figure. 4). It's supposed that the viscosity of material is small enough at high nozzle temperature to achieve a complete neck growth at the interface. So, the influence of interlocking is less important.

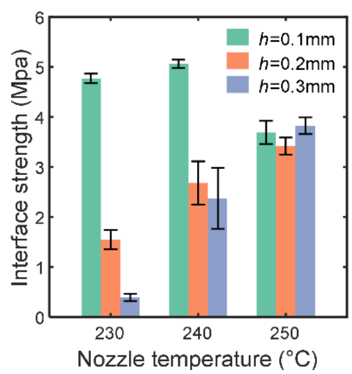

Figure. 4 Tensile strength of specimens.

Table. 2 Tensile strength of dual-interface specimens

T (°C)	h (mm) 0.1	0.2	0.3
230	4.77 ± 0.10	1.55 ± 0.19	0.39 ± 0.07
240	5.06 ± 0.08	2.68 ± 0.43	2.37 ± 0.61
250	3.69 ± 0.23	2.9 ± 0.69	3.82 ± 0.17

In the cases of h = 0.2 mm and 0.3 mm, the increase in T from 230°C to 250°C significantly increases the interface strength (Figure. 4). That's because the increase in T reduces the viscosity of TPU and improves the neck growth at the interface. As shown in Figure.

3, specimens printed at higher *T* have smaller pores in microstructure and more residual materials on the fracture surfaces. In the case of *h* = 0.1 mm, the interface strength slightly increased when *T is* increased from 230°C to 240°C, but decreases significantly as *T is* increased from 240°C to 250°C. This is supposed to be a result of material degradation, as 250°C is too high for the extrusion of TPU (melting temperature of TPU is 180°C).

Optimization of manufacturing using varied layer thickness

Except for several cases, the decrease of *h* and the increase of *T* significantly improve the interface adhesion strength between PLA and TPU. However, the decrease of *h* not only increases the interface adhesion strength but also significantly increases the printing time. To solve this problem, a specimen with varied layer thickness was developed (Figure. 5). The aim is to reinforce the zig-zag interlock effect by using a small *h* near the PLA-TPU interface, while using a bigger *h* at other regions to save the printing time.

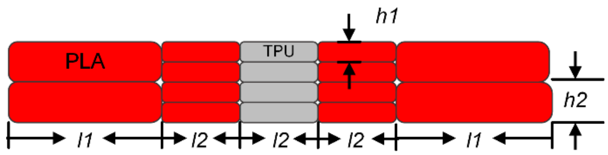

Figure. 5 The optimized specimen with varied layer thickness. *l1* =32mm, *l2* = 6 mm; *h1* = 0.2 mm, *h1* = 0.1 mm.

To verify the effectiveness of this method, 4 optimized specimens were printed and tested. As shown in Figure. 6, all the specimens were printed at *T* =240°C. The first three type of specimens were horizontally printed with different layer thickness. "Varied h" refers to the optimized specimen with varied layer thicknesses. "Ref" is the vertically printed reference specimen with *h* = 0.2 mm. The results indicate that the interface adhesion strength of the optimized specimen was 58.2% higher (4.24 Mpa) than that of the specimen with a constant *h* = 0.2 mm (2.68 Mpa). Meanwhile, the printing time was only increased by 28.2% (44.6 min)

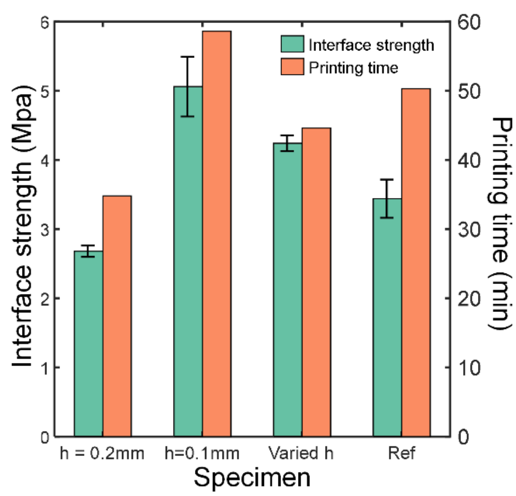

Figure. 6 Interface adhesion strengths and printing times of the specimens with different layer thickness.

Conclusion

In this study, the interface adhesion strength between a typical rigid material, PLA and a semi-flexible material, TPU, were investigated using butt joint tensile test and microscope. The experiment results reveal a new adhesion mechanism in multi-material FDM: micro-scale zig-zag interlock. It's found that the vertically printed specimens have a smooth interface, while the horizontally printed specimens have a micro-scale zig-zag interlock interface. The formation of this micro-zigzag interlock is due to the switch of extruders and material wetting. The formation of this microscale zig-zag interlock mechanically reinforces the interface adhesion strength, which is firstly reported in the literature.

The effect of two key process parameters, namely nozzle temperature and layer thickness, was investigated. In general, the decrease in layer thickness increases the depth of zig-zag interlock and results in longer contact length between PLA and TPU and higher interface adhesion strength. The increase in nozzle temperature improves the material wetting and neck growth at the interface, leading to a stronger interlock and higher interface adhesion strength. By optimizing the layer thickness and nozzle temperature, the interface adhesion strength between dissimilar materials was increased by 58.2% without significantly increasing the printing time or fabrication complexity.

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References

[1] A. Bandyopadhyay and B. Heer, "Additive manufacturing of multi-material structures," *Materials Science and Engineering: R: Reports*, vol. 129, pp. 1–16, Jul. 2018, doi: 10.1016/j.mser.2018.04.001.
[2] D. Espalin, J. Alberto Ramirez, F. Medina, and R. Wicker, "Multi-material, multi-technology FDM: exploring build process variations," *Rapid Prototyping Journal*, vol. 20, no. 3, pp. 236–244, Jan. 2014, doi: 10.1108/RPJ-12-2012-0112.
[3] L. Li, Q. Sun, C. Bellehumeur, and P. Gu, "Investigation of Bond Formation in FDM Process," 2002. doi: 10.26153/TSW/4500.
[4] Q. Sun, G. M. Rizvi, C. T. Bellehumeur, and P. Gu, "Effect of processing conditions on the bonding quality of FDM polymer filaments," *Rapid Prototyping Journal*, vol. 14, no. 2, pp. 72–80, Jan. 2008, doi: 10.1108/13552540810862028.
[5] J. P. Thomas and J. F. Rodríguez, "Modeling the Fracture Strength between Fused-Deposition Extruded Roads 16," 2000. doi: 10.26153/tsw/2054.
[6] X. Gao, S. Qi, X. Kuang, Y. Su, J. Li, and D. Wang, "Fused filament fabrication of polymer materials: A review of interlayer bond," *Additive Manufacturing*, vol. 37, p. 101658, Jan. 2021, doi: 10.1016/j.addma.2020.101658.
[7] J. Allum, A. Moetazedian, A. Gleadall, and V. V. Silberschmidt, "Discussion on the microscale geometry as the dominant factor for strength anisotropy in material extrusion additive manufacturing," *Additive Manufacturing*, vol. 48, p. 102390, Dec. 2021, doi: 10.1016/j.addma.2021.102390.
[8] J. Allum, A. Moetazedian, A. Gleadall, and V. V. Silberschmidt, "Interlayer bonding has bulk-material strength in extrusion additive manufacturing: New understanding of

anisotropy,” *Additive Manufacturing*, vol. 34, p. 101297, Aug. 2020, doi: 10.1016/j.addma.2020.101297.

[9] T. J. Coogan and D. O. Kazmer, “Prediction of interlayer strength in material extrusion additive manufacturing,” *Additive Manufacturing*, vol. 35, p. 101368, Oct. 2020, doi: 10.1016/j.addma.2020.101368.

[10] L. R. Lopes, A. F. Silva, and O. S. Carneiro, “Multi-material 3D printing: The relevance of materials affinity on the boundary interface performance,” *Additive Manufacturing*, vol. 23, pp. 45–52, Oct. 2018, doi: 10.1016/j.addma.2018.06.027.

[11] F. Tamburrino, S. Graziosi, and M. Bordegoni, “The influence of slicing parameters on the multi-material adhesion mechanisms of FDM printed parts: an exploratory study,” *Virtual and Physical Prototyping*, vol. 14, no. 4, pp. 316–332, Oct. 2019, doi: 10.1080/17452759.2019.1607758.

[12] M. Ribeiro, O. Sousa Carneiro, and A. Ferreira da Silva, “Interface geometries in 3D multi-material prints by fused filament fabrication,” *RPJ*, vol. 25, no. 1, pp. 38–46, Jan. 2019, doi: 10.1108/RPJ-05-2017-0107.

[13] T. Kuipers, R. Su, J. Wu, and C. C. L. Wang, “ITIL: Interlaced Topologically Interlocking Lattice for continuous dual-material extrusion,” *Additive Manufacturing*, vol. 50, p. 102495, Feb. 2022, doi: 10.1016/j.addma.2021.102495.

[14] B. Stoner, J. Bartolai, D. V. Kaweesa, N. A. Meisel, and T. W. Simpson, “Achieving Functionally Graded Material Composition Through Bicontinuous Mesostructural Geometry in Material Extrusion Additive Manufacturing,” *JOM*, vol. 70, no. 3, pp. 413–418, Mar. 2018, doi: 10.1007/s11837-017-2669-z.

[15] “Properties of Raise 3D Premium 1.75 mm PLA filament.” https://s1.raise3d.com/2020/04/Raise3D-Premium-PLA_SDS_V4.pdf (accessed Jun. 20, 2021).

[16] “NinjaFlex-TDS.pdf.” Accessed: Jun. 28, 2021. [Online]. Available: https://ninjatek.com/wp-content/uploads/NinjaFlex-TDS.pdf