

ORIGINAL RESEARCH ARTICLE

Enhancing interlaminar adhesion in
multi-material 3D printing: A study of
conductive PLA and TPU interfaces through
fused filament fabricationGuo Liang Goh^{1,2}, Samuel Lee¹, Shi Hui Cheng¹, Daniel Jee Seng Goh¹,
Pothunuri Laya², Van Pho Nguyen^{1,3}, Boon Siew Han³, and Wai Yee Yeong^{1,2*}¹School of Mechanical and Aerospace Engineering, Nanyang Technological University, Singapore²Singapore Centre for 3D Printing, School of Mechanical and Aerospace Engineering, Nanyang Technological University, Singapore³Schaeffler Hub for Advanced Research, Nanyang Technological University, Singapore

Abstract

In the rapidly expanding field of additive manufacturing, multi-material fused filament fabrication represents a frontier with vast potential for creating composite structures that blend the benefits of different material properties. Interlaminar adhesion between dissimilar materials remains a challenge for the realization of multifunctional structure for practical use. This study investigates the interlaminar adhesion between conductive polylactic acid and thermoplastic polyurethane, materials representative of rigid and flexible characteristics, respectively. We present a comparative analysis of two adhesion enhancement approaches: the incorporation of mechanical interlocking features and the modification of surface roughness at the interface. Through tensile testing, we evaluate the effectiveness of these methods against a benchmark coupon with unmodified interface. Micro-computed tomography analysis, surface morphology analysis, and mechanical performance assessments elucidate the failure modes and provide insights into the interfacial behavior of these interface designs. We found that the interface design with top infill modification showed the highest interlaminar adhesion strength, with an improvement of at least 25% compared to the benchmark coupon. Our findings aim to inform the design and manufacturing practices in multi-material 3D printing and to open new avenues for the development of multifunctional, composite 3D-printed systems.

***Corresponding author:**Wai Yee Yeong
(wyyeong@ntu.edu.sg)

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1. Introduction

The advent of additive manufacturing, also known as 3D printing, has transformed the landscape of material science and engineering, propelling the innovation of new applications across a diverse range of industries such as printed electronics,

tissue engineering, and aerospace and defense.¹⁻⁵ Fused filament fabrication (FFF) stands out among the array of multi-material 3D printing technologies (Figure 1A) for its versatility, accessibility, and efficiency in creating complex geometries⁶⁻⁸ and the availability of a wide range of materials, such as polylactic acid (PLA), thermoplastic polyurethane (TPU), acrylonitrile butadiene styrene (ABS), polyethylene terephthalate glycol, nylon, and polyether ether ketone (PEEK). To date, various material combinations have been attempted, such as PLA/ABS, PLA/TPU, conductive polylactic acid (cPLA)/ABS, PLA/TPU, chlorinated polyethylene (CPE)/TPU, CPE/PLA, PLA/conductive-PLA, ABS/conductive-PLA, and ABS/TPU, just to name a few.^{7,9-14} One of the promising areas of interest within multi-material FFF is the printing of soft and rigid materials, which promises to revolutionize the manufacturing of multifunctional applications, from structural electronics to soft robotic devices.¹⁵⁻¹⁸ Among the materials of interest, cPLA and TPU have garnered significant attention due to their unique properties – cPLA for its rigidity and ability

to conduct electricity, and TPU for its elasticity and durability.¹⁹

The fusion of these materials within a single print could lead to the creation of objects with both structural support and flexible characteristics, offering myriad possibilities such as actuators and electronic components integrated into structural frameworks.²⁰ This capacity for *in situ* fabrication of structures combining multiple material allows for more integrated system with fewer parts and reduces the need for manual assembly. In recent research, Goh *et al.* explored the multifaceted capabilities of multi-material 3D printing by synergizing the distinct properties of cPLA and TPU.¹⁹ Their innovative approach led to the fabrication of a multifunctional robotic gripper, which integrates the rigid features of cPLA as a variable-stiffness backbone, along with the inherent softness of TPU to impart a gentle gripping action. This integration exemplifies the potential of combining soft and rigid materials to create adaptive, responsive structures that leverage the best attributes of each constituent material for advanced robotic applications.²¹ However, a critical challenge that impedes the broad implementation of this technology is the issue of interlaminar adhesion between distinct material types. Typically, the adhesion mechanisms between dissimilar polymeric materials are adsorption, mechanical interlocking, polarity, diffusion, *etc.*, as illustrated in Figure 1B.²² Effective adhesion between cPLA and TPU is pivotal to ensure mechanical integrity and functional performance. Nevertheless, the disparity in the thermal, mechanical, and chemical properties between PLA and TPU creates a complex interface that is prone to delamination and structural weakness under stress.¹⁸ Moreover, the different melting temperatures and rates of thermal contraction during the cooling phase further complicate the bonding process, resulting in a lackluster interfacial adhesion.

Existing approaches to mitigate these adhesion issues are as diverse as the applications they serve. Strategies range from plasma surface treatments to increase the surface energy of the materials, to physical methods such as the inclusion of mechanical interlocks or using a gradient transition between dissimilar materials.^{7,10,11,14,23-26} From a mechanical perspective, enhanced adhesion between 3D-printed filaments can be achieved by increasing the contact surface area at the interface. This can be accomplished through the implementation of a skewed toolpath configuration and by elevating the material extrusion rate, both of which serve to augment the interfacial bonding area between the filaments.^{27,28} From a materials science perspective, researchers have experimented with the introduction of chemicals or

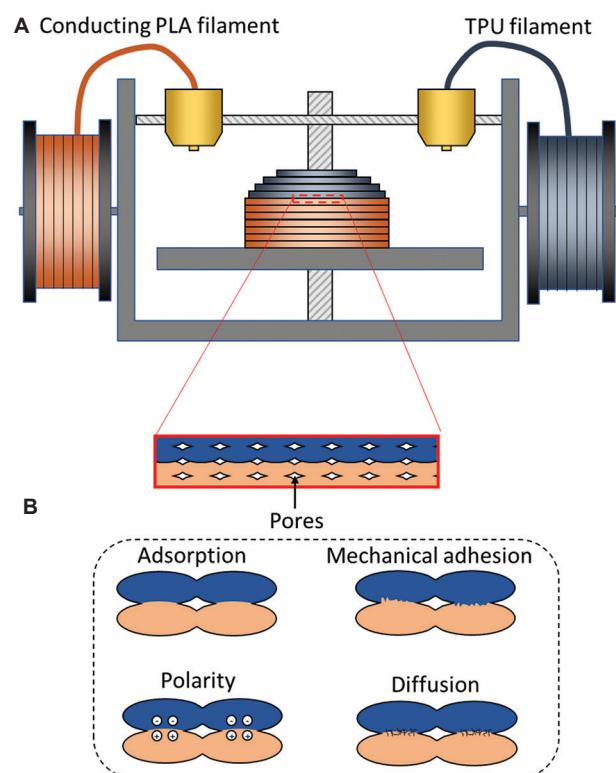


Figure 1. Schematics illustrating (A) the fabrication of a multi-material part through a multi-nozzle fused filament fabrication process and (B) the possible adhesion mechanisms at the interface of a fused filament fabricated multi-material part. Adapted from Watschke *et al.*³⁶ (licensed under CC BY 4.0). Abbreviations: cPLA: conductive polylactic acid; TPU: Thermoplastic polyurethane.

adhesion promoters that can bond to both PLA and TPU.²⁹ Process-based modifications, such as optimizing print temperature, speed, and layer height, have also shown potential in improving adhesion.^{30,31} Moreover, the integration of nanomaterials and the development of hybrid filaments are emerging as promising frontiers in addressing these adhesion challenges. Each approach carries its own set of advantages and trade-offs, necessitating a comprehensive study to identify the most effective method for a given application. However, these works mainly focused on the interaction between the pure polymeric materials. Nevertheless, the existing literature primarily emphasizes the interplay between unmodified polymeric materials.^{32,33} There remains a substantial research gap in understanding the interlaminar properties when one introduces nanocomposites into the mix, particularly between cPLA enhanced with nanoparticles and unaltered TPU. Furthermore, the mechanical interlocks approach often requires a large overlap/transition region to achieve satisfactory interlaminar strength which may not be suitable for some applications. These areas of study are crucial for advancing the application potential of multi-material 3D printing, as it addresses the interface between technologically augmented and standard polymers.

Recognizing the need for enhanced interfacial adhesion, this study delves into two specific enhancement approaches: the engineering of low-profile mechanical interlocking features (<2 mm in thickness) and the tailoring of surface roughness at the interface. These approaches are assessed against a benchmark part with no modifications, providing a comprehensive comparative analysis. By exploring these strategies, this research aims to unravel the complexities of the cPLA-TPU interface and to contribute to the optimization of multi-material FFF processes. The outcomes

will provide crucial insights into maximizing the interfacial adhesion, thus enabling the reliable creation of composite materials with integrated soft and rigid characteristics for advanced manufacturing applications.

2. Methods

2.1. Materials

In this study, we utilized cPLA and TPU filaments, with a standard diameter of 1.75 mm, to investigate the adhesion properties relevant for multi-material FFF. The selected cPLA filament, sourced from Protopasta[®], is a composite material enhanced with carbon black nanofillers to infuse the PLA with electrical conductivity, while maintaining its inherent rigidity. The soft material chosen is a NinjaFlex TPU filament, acquired from Ninjatek[®], notable for its flexibility and a Shore hardness rating of 85A, which falls into the medium-hard category for thermoplastic elastomers. Comprehensive details regarding the specifications, material properties, and manufacturer data for both the cPLA and TPU filaments are systematically cataloged in Table 1. The table provides an overview of key parameters, such as tensile strength, elongation at break, thermal properties, and electrical conductivity, offering a foundation for the ensuing analysis of interlaminar adhesion. This selection of materials facilitates a focused assessment of the interfacial adhesion challenges between a conductive, rigid filament and a non-conductive, flexible counterpart, contributing to the broader understanding of multi-material 3D printing dynamics.

2.2. Design and fabrication of test coupons

The test coupons were designed using a 3D design CAD software, Solidworks[®] 2022. In this work, a multi-material

Table 1. Summary of materials used in this work for the fabrication of test coupons

Material type	Brand	Model	Property
cPLA	Protopasta [®]	Electrically conductive composite PLA	• Appearance: black • Additives: carbon black • Volume resistivity of molded resin: 15 Ω-cm • Volume resistivity of 3D-printed parts along layers (x/y): 30 Ω-cm • Volume resistivity of 3D-printed parts against layers (z): 115 Ω-cm • Melt temperature: 155°C • Density: 1.24 g/cm³ • Tensile modulus: 3.6 MPa • Tensile yield strength: 60 MPa
TPU	Ninjatek [®]	NinjaFlex	• Appearance: glossy black • Shore hardness: 85A • Maximum elongation: 660% • Melt temperature: 216°C • Density: 1.19 g/cm³ • Tensile modulus: 12 MPa • Yield strength: 4 MPa

Abbreviations: cPLA: conductive polylactic acid; PLA: Polylactic acid; TPU: Thermoplastic polyurethane.

tensile coupon was designed and used to facilitate the evaluation of interlaminar adhesion property between cPLA and TPU materials. The design of the tensile coupons was adopted from ASTM D638. In this work, we focused on adhesion property between cPLA and TPU materials in the build direction (z-direction). The design and the part orientation used for printing of the tensile coupons are illustrated in Figure 2.

In this study, we conducted a detailed examination of interlaminar adhesion by fabricating and assessing three distinct types of test coupons, differentiated by their interface treatments:

- (i) Unmodified interface coupons: These samples were produced without any intentional alterations to the interface. The printing parameters were set to allow for filament deposition with no gaps, ensuring that the layers were laid down in direct contact with one another. This standard configuration serves as the control group, providing a baseline for evaluating the efficacy of interfacial modifications.
- (ii) Low-profile mechanical interlocking feature coupons: For these specimens, we introduced a mechanical interlocking design, which features two layers of interlocking geometry that extends 2 mm into the material. The precise pattern of this feature is

illustrated in Figure 2C. The intention behind this design is to enhance the mechanical bond between layers through increased physical engagement, akin to the interlocking teeth of a zipper. The design of the interlocking feature was generated using Ultimaker Cura® 5.3.1.

- (iii) Top infill modification: The final group of samples includes a modification to the top layer of the bottom part, designed to augment the effective bonding surface area. This was achieved by programming a fine 1 mm gap (or groove) on the top layer of the lower filament segment before depositing the subsequent material. When printing this variant, the printer's nozzle was deliberately positioned closer to the print bed. This adjustment ensures that the extruded filament is compelled into the grooves, forming a mechanical bond on solidification.

This approach to sample design allowed for the comparative analysis of interlaminar adhesion under varying interface conditions. By testing each configuration, we seek to provide insight on the impact of various interface designs and the print order of the materials on the overall bond strength between the rigid cPLA and the flexible TPU layers in multi-material prints.

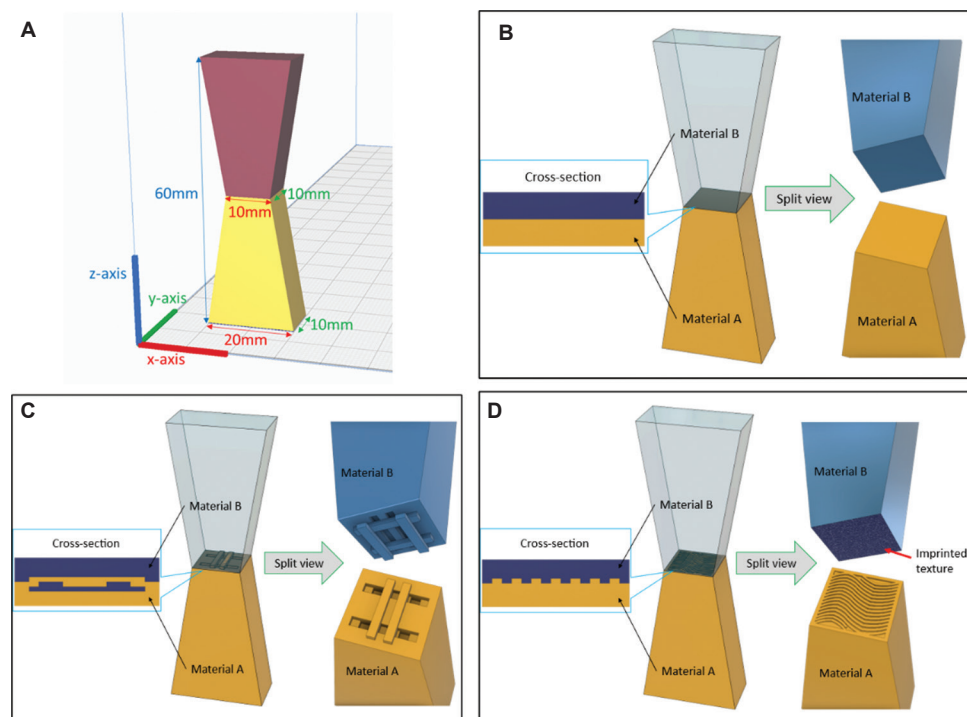


Figure 2. Schematic showing the (A) dimensions and print orientation of the tensile coupon for the investigation of interlaminar adhesion property between cPLA and TPU. (B-D) show the designs of the interfacial feature of multi-material tensile coupon, with (B) showing the normal interface, (C) mechanical interlocking feature, and (D) top infill modification coupons. Abbreviations: cPLA: conductive polylactic acid; TPU: Thermoplastic polyurethane.

Another critical component of the methodology involved the investigation of material deposition order and its effect on interlaminar adhesion properties. We systematically prepared and tested samples using two distinct printing sequences, specifically varying the order in which cPLA and TPU were printed. This variation aimed to ascertain the influence of print order on the bond quality between cPLA and TPU. In Figure 2, “Material A” and “Material B” refer to cPLA and TPU, respectively, whose printing order was alternated as detailed in the subsequent paragraph and summarized in Table 2.

The first set of samples was fabricated by initiating the print with the cPLA, subsequently followed by the deposition of the TPU layer. This sequence prioritizes the placement of the rigid material as the foundational layer. Conversely, the second set of samples reversed this order, starting with TPU as the base, on which cPLA was printed. The test coupons were fabricated through a modified E3D ToolChanger on a standard glass heated bed. A 0.6 mm nozzle was used to print cPLA filaments, whereas a standard 0.4 mm nozzle was used to print the TPU-based filaments. A larger nozzle was used for cPLA filament to ensure good printability as the cPLA filament tends to clog when a smaller nozzle is used. The printing parameters used in this work were adopted from the settings recommended by the respective manufacturers. Table 3 summarizes the print settings that were used to print each material.

2.3. Mechanical tests multi-material test coupons

The tensile test was conducted in accordance with ASTM D638 with some slight modifications using a universal testing machine (AGX plus 10 kN, Shimadzu). For the tensile test, a strain rate of 2 mm/min was used. The force required to pull the sample until it failed was recorded over time. The tensile stress was calculated based on the smallest cross-sectional area of the tensile coupons where the interface between the two materials was situated. Five samples were used for the tensile test to ensure the statistical significance of the results. With three different types of interfaces and two print orders, there were altogether six different coupons that have been tested, and they are summarized in Table 2.

2.4. Characterization of failure mode of the tensile coupons

The characterization of the fracture modes for the tensile coupons was performed using optical microscopy, micro-computed tomography (micro-CT) analysis, and surface roughness characterization to gain both qualitative and quantitative insights into the failure mechanisms:

(i) Optical microscopy: For the initial qualitative analysis, an optical camera was employed to capture

Table 2. Material combinations tested in this work

Sample types	Interface design	Material A (printed first)	Material B (printed last)
1	No modification	cPLA	TPU
2		TPU	cPLA
3	Low profile mechanical interlocking	cPLA	TPU
4		TPU	cPLA
5	Top infill modification	cPLA	TPU
6		TPU	cPLA

Abbreviations: cPLA: conductive polylactic acid; TPU: Thermoplastic polyurethane.

Table 3. Print settings for different print filaments

Settings	Materials	
	cPLA	TPU
Nozzle temperature (°C)	200	235
Print speed (mm/s)	70	50
Nozzle size (mm)	0.6	0.4
Part cooling fan	yes	no
Infill percentage	20%	15%
Infill pattern	Grid	Gyroid
Number of perimeter lines	2	2

Abbreviations: cPLA: conductive polylactic acid; TPU: Thermoplastic polyurethane.

high-resolution images of the fracture surfaces post-tensile testing. This visual inspection facilitated a comparative assessment of the macroscopic features across different coupon types, providing immediate visual cues to the nature of the bond failure, such as adhesive or cohesive fracture patterns.

- (ii) Micro-CT analysis: We utilized a Bruker SkyScan 1173 micro-CT scanner to probe deeper into the internal structures in three dimensions. This non-destructive technique allowed us to construct detailed 3D representations of the internal coupon structure, including any voids, delamination, or other imperfections that contributed to the failure. The parameters set for the micro-CT scans included a source voltage of 80 kV, source current of 60 μ A, image pixel size of 17.47 μ m, exposure time of 850 ms, and rotation steps of 1°.
- (iii) Surface roughness characterization: A Keyence VX-100 non-contact 3D laser scanning microscope was used to quantitatively measure the surface roughness at the interface. The surface topography data acquired through this method were critical for understanding the role of surface roughness in interlaminar adhesion. Specific parameters measured, including

surface area: area ratio and average roughness (Ra), are indicative of the interface's tactile profile and its potential impact on mechanical interlocking efficacy. Two out of the five coupons are randomly selected for the surface morphology investigation. In the surface morphology analysis, we used a scanned area with a size of approximately half of the cross-section's area to ensure good coverage of the fractured surface. The scan was carried out with a z-resolution of 4 μm .

3. Results and discussions

The interlaminar adhesion strength of the interface is determined by the ultimate tensile strength of the multi-material tensile coupon when the fracture happens at the interface of the coupon. The interlaminar adhesion strength of the interface between cPLA and TPU and the corresponding maximum elongations for the six different sample types are shown in Figure 3A and B, respectively. The results reveal that the interlaminar adhesion strength of coupons with no interface modifications are approximately 0.40 ± 0.03 MPa and 2.60 ± 0.07 MPa for the print order of cPLA $\rightarrow$ TPU and TPU $\rightarrow$ cPLA, respectively. For the coupons with mechanical interlocking features, it is found that the interlaminar adhesion strength of the coupons with the print order of cPLA $\rightarrow$ TPU and TPU $\rightarrow$ cPLA are 1.87 ± 0.17 MPa and 1.08 ± 0.31 MPa, respectively. For the coupons with top infill modifications, the interlaminar adhesion strength of the coupons with the print order of cPLA $\rightarrow$ TPU and TPU $\rightarrow$ cPLA is found to be 2.50 ± 0.09 MPa and 3.25 ± 0.30 MPa, respectively.

The elongation at break of the samples is shown in Figure 3B. The results reveal that the elongation at break of coupons with no interface modifications is approximately $1.30 \pm 0.09\%$ and $11.47 \pm 1.27\%$ for the print order of cPLA $\rightarrow$ TPU and TPU $\rightarrow$ cPLA, respectively. For the coupons with mechanical interlocking features, it is found that the elongation at break of the coupons with the print order of cPLA $\rightarrow$ TPU and TPU $\rightarrow$ cPLA is $38.33 \pm 6.65\%$ and

$6.49 \pm 2.31\%$, respectively. For the coupons with top infill modifications, the elongation at break of the coupons with the print order of cPLA $\rightarrow$ TPU and TPU $\rightarrow$ cPLA is found to be $7.17 \pm 0.57\%$ and $15.15 \pm 2.36\%$, respectively.

Here, we observed that the coupons with top infill modifications generally provide a higher interlaminar adhesion strength at the interface compared to two other coupon types. Furthermore, for the cases without significant mechanical interlocking features, we observed that the interlaminar adhesion strength for the print order cPLA $\rightarrow$ TPU is generally higher than the reversed order, TPU $\rightarrow$ cPLA. Like the results of the interlaminar adhesion strength, similar trends can be observed for maximum elongation. An exception to the trend would be that the maximum elongation of the cPLA-TPU coupons is exceptionally high compared to the rest of the coupon types. One plausible explanation for the observed phenomenon is the thermal degradation of the underlying material resulting from the deposition of a material at a higher temperature. This effect is especially noticeable in the sequence of cPLA followed by TPU, where TPU is deposited at 235°C , a temperature slightly exceeding the maximum recommended temperature of 230°C for cPLA. This higher deposition temperature of TPU may lead to overheating of the underlying cPLA layer, causing thermal degradation that adversely affects the interface's mechanical properties. Conversely, in the reverse printing order of TPU followed by cPLA, the underlying material (TPU) is compatible with the processing temperature of the subsequent material (cPLA), generally resulting in better adhesion strength. This compatibility helps avoid the thermal degradation issues observed in the cPLA $\rightarrow$ TPU sequence, thus improving the bond quality at the interface. Another possible explanation for the better adhesion of TPU $\rightarrow$ cPLA compared to cPLA $\rightarrow$ TPU is that the former has a smaller temperature difference between the underlying material and the deposited material compared to the latter, resulting in a longer duration above

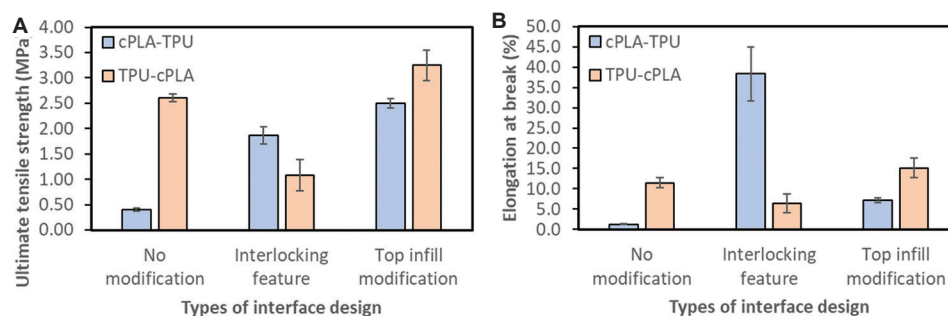


Figure 3. Graphs showing (A) the ultimate tensile strength and (B) the elongation at break of the multi-material tensile coupons in the interfacial adhesion test. The error bars represent the standard error. Abbreviations: cPLA: conductive polylactic acid; TPU: Thermoplastic polyurethane.

the glass transition temperature of materials thus better intermolecule-diffusion between the materials.^{30,34}

In our investigation of interlaminar adhesion strength, we ensured that sample failures occurred at the interface between the two materials, specifically where the cross-sectional area of the coupons is the smallest, as delineated in Figure 4. Through this controlled approach, we were able to accurately assess the interface's failure mechanism and to determine the interlaminar adhesion strength for each coupon type.

Our findings revealed that the majority of the samples exhibited a clean break at the interface, with negligible remnants of the opposing material adhering to the fracture surfaces. This phenomenon was predominantly observed in the coupons featuring an unmodified interface and those with modifications to the top surface's infill, as shown in Figure 4A and C. Notably, the samples with these interfaces displayed a consistent pattern of clean separation, indicating a uniform material behavior during failure. A clean break at the interface suggests that the interfacial bond is the weakest link in the coupons since the propagation of a crack will follow the path of least resistance once a crack is initiated under stress. In other words, this suggests that the interlaminar adhesion strength between cPLA and TPU is lower than the tensile strength between these materials. This could be due to the weaker adhesive force compared to the cohesive

forces within each material, resulting in a clean break on application of stress.

Conversely, samples designed with mechanical interlocking features presented a more complex failure behavior. Several of these samples did not fracture along the interface plane. Instead, breaks occurred at the smaller features of the interlocking structure, slightly offset from the intended plane, as shown in Figure 4B. This deviation was evident from the corrugated appearance of the fracture surfaces, suggesting that the mechanical interlocks contributed to a redistribution of stress, thereby altering the failure point. Notably, these failure points typically occurred at the smallest features of the interlocking structure, where stress concentration was highest. This phenomenon can be partly attributed to the rapid cooling of these small features, printed before the change in material during the dual extrusion process. The larger temperature difference between the deposition zone and the deposited material at these points likely resulted in weaker bonding.³⁴ Our findings highlight the significant impact of material change and extruder switching on interfacial adhesion, particularly in the context of dual extruder 3D printing systems where thermal management during printing plays a crucial role in determining bond strength.

These observations provide valuable insights into the relationship between interface design and failure mechanisms in the cPLA-TPU coupons. The distinct

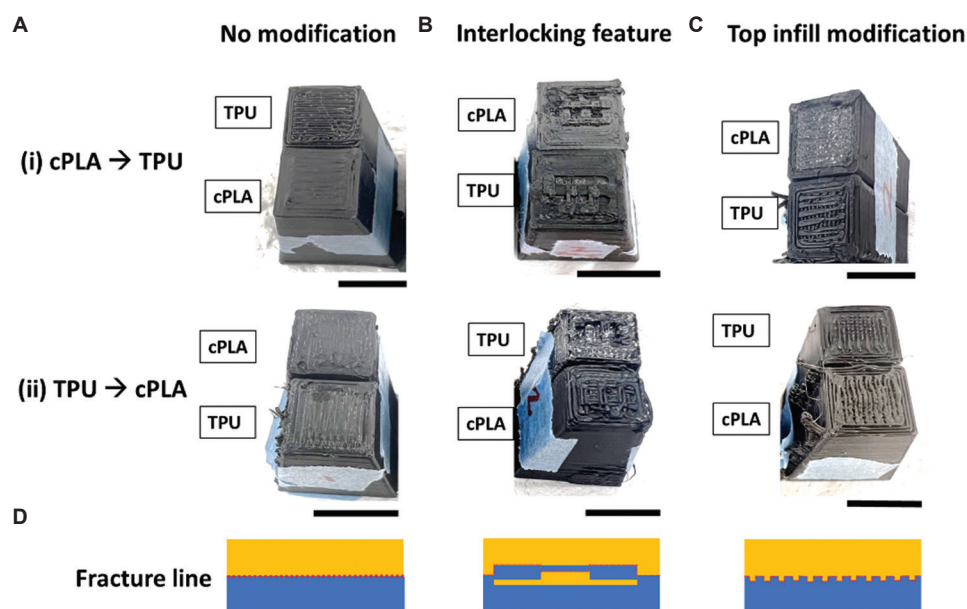


Figure 4. Images showing the typical fracture surfaces on both sides of the tested samples, namely (A) samples with no modification, (B) samples with interlocking feature, and (C) surface area-enhanced samples. The first row shows the samples with (i) the print order of cPLA → TPU, and the second row shows the samples with (ii) the print order of TPU → cPLA. The scale bars at the bottom-right corner of each image represent 1 cm. (D) The red lines in the schematics show the typical fracture lines for each type of interface. Abbreviations: cPLA: conductive polylactic acid; TPU: Thermoplastic polyurethane.

fracture characteristics (Figure 4D) between the cleanly separated samples and those with mechanical interlocks underline the influence of interface modifications on interlaminar adhesion strength and failure behavior.

Fractographic analysis was conducted to understand the failure characteristics of the 3D-printed cPLA/TPU multi-material coupons (Figure 5). All tensile samples failed at the designed interface region where the cross-section of the coupon is the smallest. For the unmodified cPLA → TPU coupons (Figure 5A-i), which exhibited the lowest interlaminar adhesion strength, the interface on the cPLA side was notably smooth and devoid of cracks or surface ruptures. In addition, only minimal marks were observed on the TPU side, indicating a weak adhesive interaction between the materials. Conversely, the TPU → cPLA counterparts displayed cracks and surface ruptures on the fracture surfaces (Figure 5A-ii), suggesting a different failure mechanism. This observation was also consistent for coupons modified with top infill, for both print orientations (Figure 5C-i and C-ii). Despite similar surface characteristics, these modified samples

demonstrated significantly higher interlaminar adhesive strength than the unmodified cPLA → TPU coupons. This suggests that the modifications introduce a stronger adhesive force between cPLA and TPU, which is sufficient to initiate cracks and surface rupture upon failure. For coupons featuring interlocking modifications, the fracture surfaces revealed broken interlocking features on both materials (Figure 5B-i and B-ii), indicating that the failure mechanism involved the complete rupture of these fine interlocking elements. This finding shows the effectiveness of the interlocking design in enhancing the mechanical interlocking of the materials, leading to a different failure mode compared to unmodified interfaces.

To further analyze the surface morphology of the fracture surfaces of the coupons, we used confocal microscopy to scan the fractured interfaces (Figure 6). Since both the coupons with unmodified interface and top infill modification showed cleanly separated samples, the confocal microscopy on the fractured surfaces of these samples could shed some light on the how the surface morphology of these samples affects the interlaminar

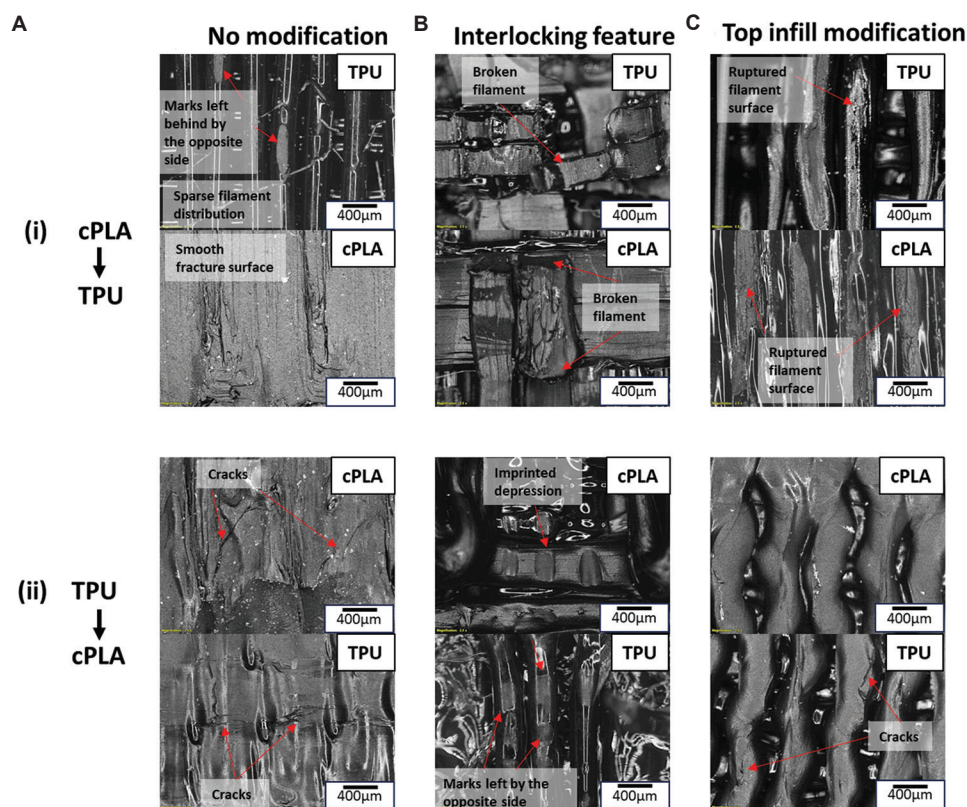


Figure 5. Fracture surfaces of various multi-material coupons with different interface design and print order. Columns (A), (B), and (C) show samples with no modification, interlocking feature, and top infill modification, respectively, and rows (i) and (ii) show samples with print order of cPLA → TPU and TPU → cPLA, respectively. Abbreviations: cPLA: conductive polylactic acid; TPU: Thermoplastic polyurethane.

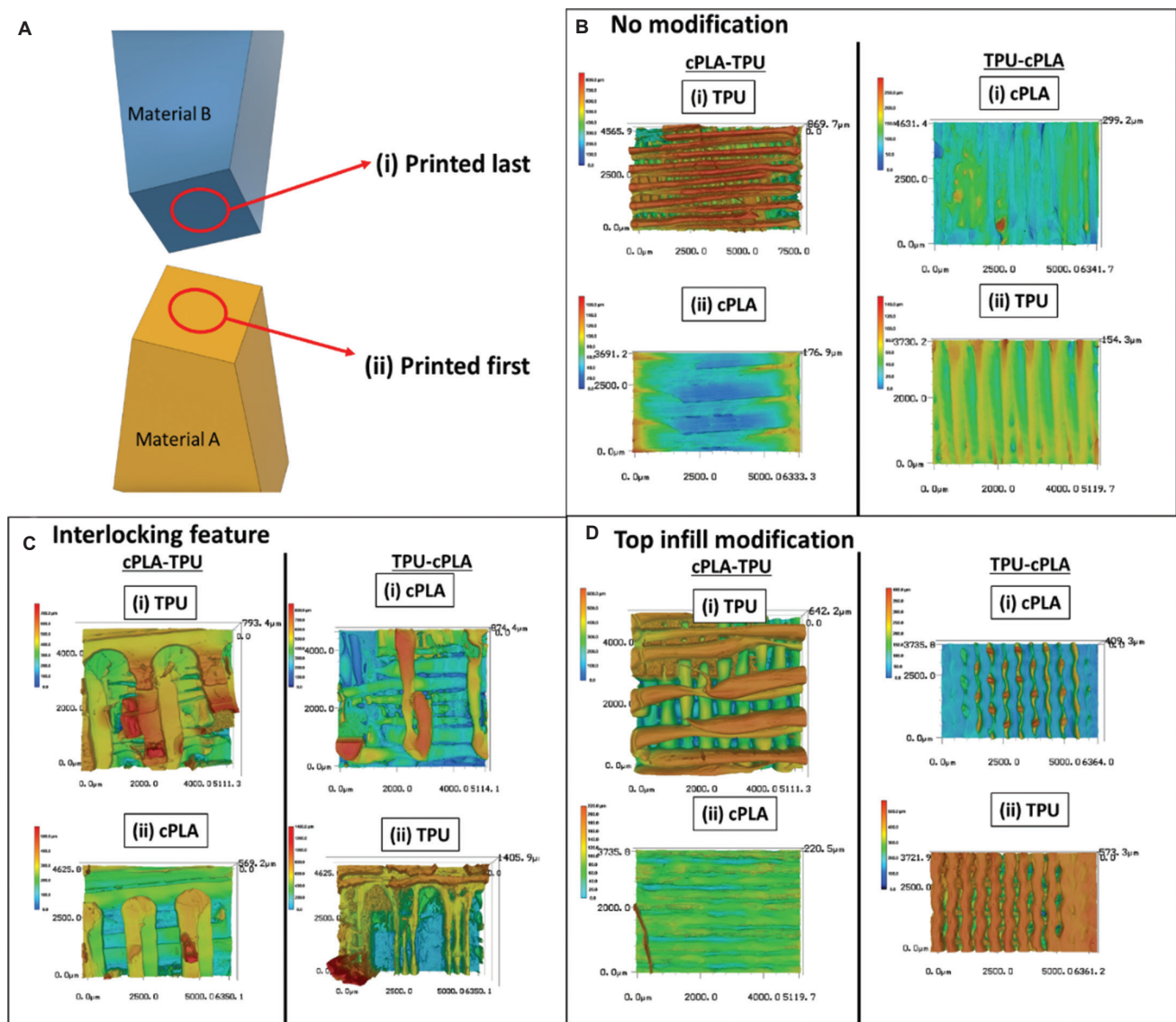


Figure 6. (A) Schematic showing the labels of two scanned fracture surfaces, with (i) representing the fracture surface of the material that is printed last and (ii) representing the fracture surface of the material that is printed first (B–D) 3D reconstructed height map showing the typical surface morphology of the fracture surfaces of the interfaces of coupons for no modification, interlocking feature, and top infill modification, respectively. Abbreviations: cPLA: conductive polylactic acid; TPU: Thermoplastic polyurethane.

adhesion strength and the fracture mode. In particular, we wanted to observe the surface morphology of the material situated below the interface (the material that is printed first). In both cases, we observed that the coupons have a relatively flat surface with some undulations scattered over the region. For the case of coupons with unmodified interface, the surface appears to be relatively flat with very few grooves or valleys, as shown in Figure 6B-ii, regardless of the print order. Similar observations can also be made for the coupons with top infill modifications, but with a much wider grooves scattered over the flat fractured surface. The difference in the surface morphology can be

attributed to the design/toolpath that is being used to print the topmost layer of the first material. It is widely known that FFF printing inherently produces parts with grooves form between the filaments, explaining why some small grooves and valleys are observed on the fractured surface of the coupons with unmodified interface. In contrast, for the case of coupons with top infill modification, we intentionally made the gaps between the filament significantly larger, making the filament on the topmost layer sparsely distributed. This explains why the sparsely distributed filaments appear as the peaks in the height map. Figure 6B-i and D-i show the surface morphology of the

fractured surfaces of the opposing side (the material that is printed last) of the coupons with unmodified interface and top infill modifications. More peaks were observed on the fractured surfaces of these coupons, suggesting possible impregnation of material from the top material into the grooves on the top layer of the bottom material. These impregnated material increases the surface area between the materials at the interface, which are responsible for the adhesion through surface adsorption and mechanical interlocking. Figure 6C, on the other hand, shows the surface morphology of the coupon with interlocking features. It is observed that these coupons generally have much rougher surfaces regardless of the print order compared with the other two cases due to uniform fracture lines that form across the interface.

To enhance our quantitative analysis of the fractured surfaces, we employed two specific metrics to evaluate surface roughness: (i) the ratio of surface area to projected area (R_{Sa-A}) and (ii) the average surface roughness (R_a), as shown in Figure 7. The R_{Sa-A} metric serves to quantify the effective surface area available for adsorption phenomena, providing insights into the potential interfacial adhesion capacity. On the other hand, R_a offers a measure of the mean surface texture or topography created during fracture. This parameter is instrumental in assessing the extent to which mechanical interlocking contributes to surface adhesion, as it reflects the microscale irregularities that can enhance mechanical bonding between the interfaces for the case of samples with clean separation. Although the evaluation may not be meaningful for the coupons with interlocking features, the values are also reported here for the sake of comprehensiveness and comparison purposes.

In general, it is observed that both R_{Sa-A} and R_a exhibit almost similar trend for all cases. To ensure conciseness, we combine the discussion of both metrics and use surface roughness indicator (SRI) as the general term to describe both

metrics. We evaluated the SRI (R_{Sa-A} and R_a) of the fractured surfaces of the material that is being printed first, as shown in Figure 7A and B. The SRI shows that there is a slight increase in R_{Sa-A} and R_a when comparing the coupons with top infill modifications to the baseline coupons that have unmodified interface. This also helps explain why there is a slight increment in the interlaminar adhesion strength of the coupons with top infill modification. The highly uneven fractured surface of the coupons with interlocking features can also be reflected by the high magnitude of the SRI. Likewise, a comparable trend was observed on the SRI of the corresponding opposite surfaces of the fractures, as these surfaces would essentially constitute the negative imprint of the fracture patterns observed on the initial side, as shown in Figure S1.

In general, the presence of internal voids within the structure can lead to mechanical imperfections, which, in turn, contribute to stress concentration and facilitate crack propagation under tensile load. This phenomenon results in diverse tensile behaviors, as these voids become critical factors in determining the material's mechanical response during testing. A micro-CT analysis was conducted on the three types of coupons to understand how different interface designs affect the formation of pores and defect within the printed structures near the interface (Figure 8A). Here, we observed that coupons with no modifications have a porosity level of approximately 1.47% near the interface region, which is slightly higher compared to that of the 0.64% of the fully printed regions. Similarly, the coupon with interlocking features also has a porosity level of approximately 5.6%. Likewise, the coupons with top infill modifications exhibit a porosity level of approximately 2.1% only (Figure 8B), which is slightly higher than that of the coupons with unmodified interface used in this work. A comparison of the ultimate tensile strength of the samples to the corresponding surface properties and the porosity level at the interface is presented in Table 4. The pronounced influence of porosity on the ultimate

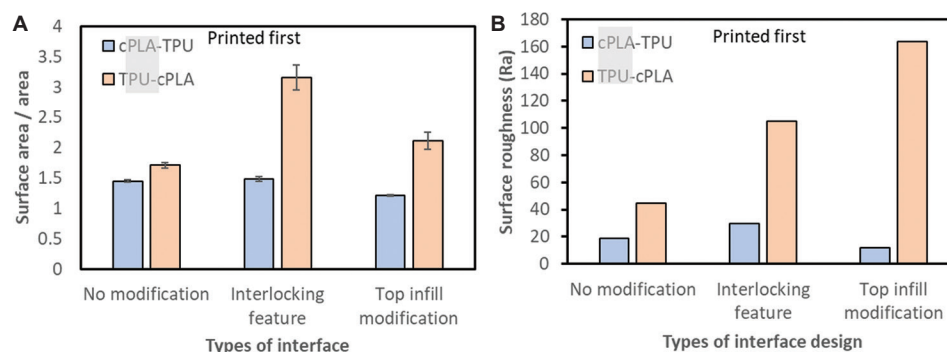


Figure 7. Graphs showing the (A) surface area/area ratio (the error bars represent standard error) and (B) surface roughness (R_a) for the interfaces that are printed first. Abbreviations: cPLA: conductive polylactic acid; TPU: Thermoplastic polyurethane.

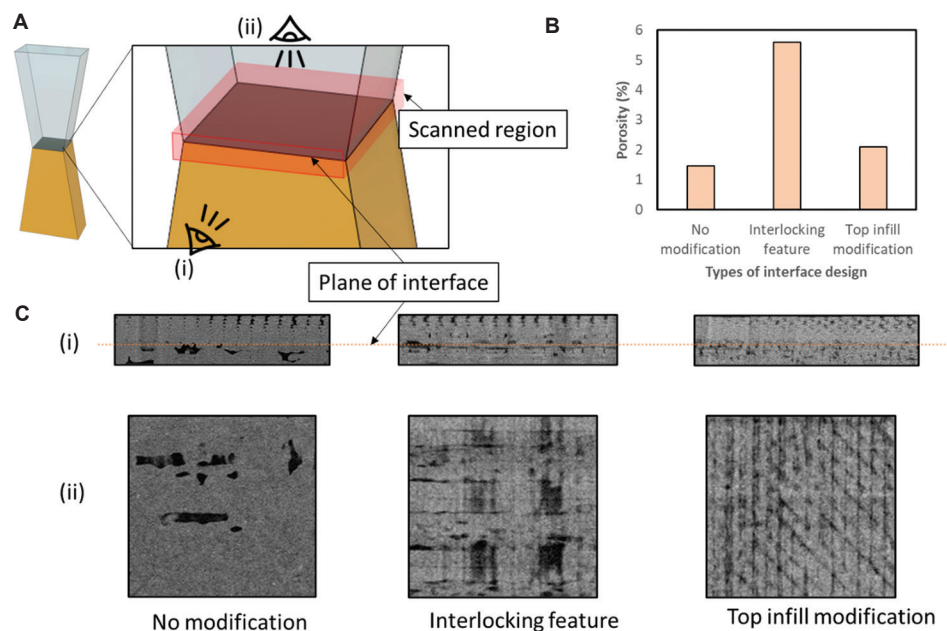


Figure 8. (A) Schematic showing the scanned region at the interface of the multi-material sample. (B) A graph showing the porosity level at the interface of various samples. (C) X-ray images showing the (i) side view at the interface and (ii) the plan view at the interface of various samples.

Table 4. Comparison between the ultimate tensile strength of the interface to their surface properties, and porosity levels at the interface

Interface design	Print order (bottom-top)	Ultimate tensile strength (MPa)	Surface area/projected area	Surface roughness, R_a (μm)	Porosity level (%)
No modification	cPLA-TPU	0.399 ± 0.029	1.45 ± 0.09	19	1.47
	TPU-cPLA	2.601 ± 0.076	1.71 ± 0.19	45	
Interlocking feature	cPLA-TPU	1.87 ± 0.170	1.49 ± 0.11	30	5.60
	TPU-cPLA	1.08 ± 0.310	3.15 ± 0.63	105	
Top infill modification	cPLA-TPU	2.495 ± 0.093	1.22 ± 0.49	12	2.10
	TPU-cPLA	3.251 ± 0.301	2.11 ± 0.04	164	

Abbreviations: cPLA: conductive polylactic acid; TPU: Thermoplastic polyurethane.

tensile strength (UTS) of the samples is evident when comparing those with interlocking features to those with top infill modification. Despite the assumption that the interlocking feature should offer a larger interfacial area conducive to stronger adhesion, the UTS of these samples is generally lower than that of the samples with top infill modification. This discrepancy can be attributed to the higher levels of porosity present in the samples with interlocking features, which adversely affect their mechanical strength. The higher porosity near the interface region found in these coupons (as depicted in Figure 8C–E) could be attributed to the manufacturing error caused by the misalignment of the nozzles and poor bed leveling, resulting in inaccurate and improper material deposition over the underlying material during the start and end of the material extrusion process.

For the case of coupons with top infill modification, the increased porosity level compared to that of the coupons with unmodified interface can be attributed to the improper impregnation of the top material into the grooves in the underlying material, although the nozzle is marginally brought closer to the substrate to promote stronger flow into the grooves. Nevertheless, the interlaminar adhesion strength is still higher than that of the unmodified interface. This can be attributed to the increased effective surface area between the two materials as discussed earlier resulted by the corrugated interface formed by the toolpath design. This approach ensures that the extruded material is pressed into the grooves of the underlying layer, thereby enhancing the effective surface area at the interface and promoting better interlayer adhesion. This explains why the coupons

with top infill modification have the highest interlaminar adhesion strength.

In addition to increasing the surface area, the various types of interfaces employed in our study also significantly influenced the diffusion of molecular chains at the interface between cPLA and TPU. This diffusion process is critically dependent on the thermal dynamics during printing, where temperature plays a pivotal role.³⁵ As the printing process involves both cooling of the previously deposited layer and reheating due to subsequent layer deposition, these thermal events occur simultaneously and have a substantial impact on the adhesion mechanism. The reheating of the underlying layer on deposition of a new layer facilitates the diffusion of molecular chains across the interface, potentially leading to stronger bonding. This effect is particularly pronounced in the presence of mechanical interlocks, where the increased surface area and complex geometries create more opportunities for molecular intermingling. The thermal history of each layer, including the peak temperature reached and the cooling rate, thus becomes a critical factor in determining the strength and quality of the interfacial bond. This complex interplay of temperature, cooling, and reheating underscores the importance of optimizing printing parameters to enhance molecular diffusion at the interface, thereby improving the overall structural integrity of the multi-material print.

4. Conclusion

This study provides some insights into the interlaminar adhesion strength of multi-material interfaces, specifically between cPLA and TPU. In this work, we investigated three different types of coupons (unmodified interface, interface with interlocking feature, and interface with top infill modification) with two opposing print orders. Through our experiments, we observed that the failure of samples predominantly occurred at the interface, with most specimens exhibiting a clean break with minimal residual material from the opposing surface. This behavior underscores the importance of interface design in multi-material 3D printing. We quantitatively assessed the surface roughness characteristics of the fractured interfaces, employing metrics such as R_{Sa-A} and R_a . Our findings revealed that the printing order and interface modifications significantly impact the roughness parameters, which, in turn, affect the mechanical interlocking and adhesion properties. In addition, the decrease in porosity at the interface can be linked to the optimized nozzle positioning during printing, which promotes the extruded material to conform into the grooves of the underlying layer, resulting in an increased effective surface area for adhesion. Our results indicated that the interface incorporating top infill modifications demonstrated superior interlaminar

adhesion strength, achieving a minimum enhancement of 25% relative to the standard benchmark coupon.

These results have far-reaching implications for the design and manufacturing of multi-material components, where interlayer adhesion is critical. They pave the way for improved application-specific material selection and the development of advanced printing strategies that can enhance the structural integrity of 3D-printed materials. Future work should focus on the optimization of interface designs to maximize adhesion strength, while also exploring the influence of various printing parameters on the mechanical properties of multi-material prints. Furthermore, the potential for scaling these findings to industrial manufacturing processes presents a promising avenue for the production of complex, high-strength multi-material assemblies.

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Conflict of interest

The authors declare that they have no competing interests.

Author contributions

Conceptualization: Guo Liang Goh

Formal analysis: Guo Liang Goh

Funding acquisition: Wai Yee Yeong, Boon Siew Han

Investigation: Guo Liang Goh, Samuel Lee, Shi Hui Cheng,

Pothunuri Laya, Daniel Jee Seng Goh

Methodology: Guo Liang Goh, Samuel Lee

Project administration: Boon Siew Han

Writing – original draft: Guo Liang Goh

Writing – review & editing: Wai Yee Yeong, Van Pho Nguyen

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Availability of data

Data are available from the corresponding author on reasonable request.

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