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Hybrid Material Extrusion Process Optimization for Printability and Adhesion in 3D-Printed Electronics

Zhao Fu, Teemu Salo, Remmi Calvo Guzman, Jukka Vanhala, and Matti Mäntysalo*

3D printing exhibits strong potential in electronics manufacturing for its capability of manufacturing complex structures with a wide range of materials. However, thermoplastic polyurethane (TPU) has rarely been fabricated by 3D printing for electronics due to the limited understanding of its printability and the adhesion of the conductors on it. Herein, we invetigate the printability of TPU using varying layer height and printing temperature in the Materials Extrusion (MEX) process, their impacts on the printability, and the adhesion of conductive trace on TPU. The printability is characterized by the surface roughness, stability of dimension and mass, and the adhesion is evaluated by a standard cross-cutting method. In this research, 0.20 mm layer height and 220 °C printing temperature have consistently proven to be optimal configuration parameters for MEX printing of TPU. Larger layer height in TPU printing causes poorer printing quality (lower dimensional accuracy, more porous structure, rougher surface), lower ink–TPU adhesion, and transforms the adhesive failure of ink against peeling to cohesive failure. Higher printing temperature causes less homogeneous structure and rougher surface with minor influence on the width and conductivity of ink on TPU, and no influence on the mechanism of ink failure against peeling.

1. Introduction

3D printing is a revolutionary manufacturing process that prints material layer-by-layer under the guidance of a digital 3D model.[1,2] This unique feature enables it to manufacture products with complex structures and geometries that are almost impossible to fabricate using conventional manufacturing systems. It opens up the possibility of innovative design and manufacturing using new materials, especially polymers.[2] Additionally, 3D printing enables to customize the design digitally with freedom, allowing for rapid prototyping,[1] reduction in material and energy waste,[3] part counts, assembly time, and cost.[3] Furthermore, the on-demand nature of 3D printing has significant benefits for businesses, such as improved response time, shortened supply chains, reduced storage needs, and faster production of critical replacement parts.[4,5] As a result, 3D printing has the potential to play a crucial role in the advancement of Industry 4.0.[6] It has been applied in the fields of automotive, machinery, aerospace, consumer electronics, architectural modeling, and medical devices.[3,7,8]

Material Extrusion (MEX) is a popular 3D printing technology due to its ease of use, simple fabrication process, cost-effectiveness, suitability for a wide range of materials, and ability to manufacture complicated parts.[9] In MEX, the polymer filament is fed through the extruder and nozzle of the printer via two rollers rotating in opposite directions. The solid filament is heated to a molten state before entering the nozzle, through which it is extruded to be deposited layer-by-layer on the build plate based on a digital design.[10] MEX has demonstrated its ability to process various thermoplastic polymers like polylactic acid (PLA), acrylonitrile butadiene styrene (ABS), polystyrene, polycarbonate, polyamide, and polyethylene terephthalate. Among these, PLA and its composites are the most extensively researched polymer materials.[9,11–13] For processing polymers using MEX, the research has primarily focused on investigating the impact of process parameters on the mechanical properties of printed parts.[11] Many process parameters have been found to influence the mechanical properties and quality of the printed parts. However, the relationship between factors related to the material and fabrication parameters remains unclear.[9,11]

In addition, MEX faces critical challenges of adhesion between adjacent layers, surface roughness, and dimensional accuracy, all of which can significantly affect the final finish of the MEX-3D-printed parts.[10,14–16] Since the molten material is layered to create the product, any issue with filament MEX or the diffusion of molten material can affect the accuracy of dimensions and print quality. One major drawback to the quality of MEX is surface roughness, which is directly related to the printing process itself. Due to the stair-stepping effect, the printed product often exhibits back-and-forth tracks, creating terraces on the surface and increasing roughness.[10] This poor surface finish, including rough surfaces, voids, and striations, has been widely reported in MEX-fabricated products.[17]

The fabrication parameters in the MEX process have been widely reported to affect the printability and surface roughness

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of printed parts.[18–20] One of the most significant parameters that influences the surface roughness of MEX-fabricated pieces is the layer height.[21–24] Nguyen et al.[21] and Jain et al.[23] reported that increasing the printing layer height results in a rougher surface for MEX-fabricated polymer parts. Similarly, Bakhtiari et al.[24] observed that decreasing the layer height in MEX of PLA leads to higher compressive strength and lower surface roughness and residual strain. Additionally, the diffusion of molten material plays a critical role in determining the surface quality of printed parts, making printing temperature (or nozzle temperature) another key parameter that affects the surface roughness of MEX-fabricated pieces.[25] Chaidas et al.[26] and Kovan et al.[27] reported that decreasing the printing temperature results in a rougher surface and lower quality for MEX-fabricated polymer pieces. Therefore, in this research, we focus on investigating the impact of layer height and printing temperature on the surface roughness of printed parts and determining the optimal configuration for the polymer substrate.

Electronics is a promising field where 3D printing can make significant contribution, particularly in the fabrication of embedded electronics, 3D structural electronics, and flexible and stretchable electronics with complex structures.[28,29] The behavior of the interface between conductive traces and the polymer substrate is critical in the fabrication of embedded electronics. The surface roughness and morphology of the polymer substrate have been found to have a remarkable impact on the quality and sheet resistance of printed conductive traces on it.[30,31] While PLA and ABS are the most commonly researched polymer materials for substrate in electronic devices using the MEX process, there is limited research on the MEX processing of elastic polymers such as thermoplastic polyurethane (TPU) for electronics fabrication. Previous studies on MEX-printing of TPU have primarily focused on the impact of processing parameters on the mechanical properties of the printed parts. For instance, Hohimer et al.[32] found that the raster angle and air gap significantly affect the ultimate tensile strength of the MEX 3D-printed TPU parts. Similarly, Kechagias et al.[33] investigated the impact of layer height and nozzle temperature on the tensile strength, elasticity, and impact strength of MEX 3D-printed TPU parts. Wang et al.[34] studied the effects of infill percentage and printing temperature on the mechanical performance of 3D-printed TPU parts. Additionally, different infill patterns have also been found to influence the mechanical properties of 3D-printed TPU.[35] However, there is limited understanding of how TPU printing parameters affect the performance of conductive traces printed onto it, which is a crucial step in the fabrication of flexible and stretchable electronic devices using TPU. In contrast, TPU has gained increasing attention for 3D printing due to its unique combination of mechanical, thermal, and chemical properties,[36] and MEX has demonstrated the ability to produce TPU with strong layer adhesion and elasticity.[37,38] Furthermore, TPU exhibits advantageous surface quality and strength compared to other polymers[39] and excellent stretchability.[40,41] Therefore, this research aims to investigate how layer height and printing temperature affect the surface roughness of the MEX-printed TPU substrate and their impacts on the quality of the direct ink write conductive trace on it.

2. Experimental Section

2.1. Research Materials

This research aimed to optimize the printing process of flexible and stretchable polymers for future research in stretchable electronics. The TPU filament was selected for its high stretchability and ability to withstand the thermal curing of ink at 130 °C. TPU is a block copolymer composed of alternating sequences of hard segments (isocyanates) and soft segments (reacted polyol), the flexibility depends on the proportion of soft segments. The Ultrafuse TPU filament (BASF, Netherlands) was used for substrate fabrication. It has a hardness of 95 A and a diameter of 1.75 mm. The higher shore hardness of 95 A was selected for its superior print quality compared to the 85 A shore hardness, while still providing sufficient deformability for stretchable applications. The conductive silver ink CI-1036 (ECM, Delaware, USA) was used for printing electrical circuits for its high flexibility, good conductivity, good printability, and good adhesion to the TPU substrate. The ink has a viscosity of 10 Pa s.

2.2. TPU Substrate Fabrication

The TPU filament was utilized to create substrates with dimensions of 110 mm × 35 mm × 1 mm using MEX on the Neotech AMT PJ15X 3D printer. This is a 3D-printed electronics manufacturing system that includes various printing tools such as material extruder, piezo jet dispenser, and ink dispenser. The print pattern was designed using SolidWorks software. The main printing parameters applied were a first layer print speed of 560 mm min^{-1}, a first layer flow of 160% (the amount of material extruded on the initial layer is multiplied by this value), a print speed of 800 mm min^{-1}, a number of walls of 2, a wall line width of 0.15 mm, an inside first wall order, a nozzle diameter of 0.40 mm, and a line width of 0.40 mm. The infill line pattern used for printing was a 45° raster angle with 100% infill density, as it provided the best tensile properties.[42,43] This infill angle and infill density have also been recommended for 3D printing polymers.[44] The layer height of the printed line and the printing temperature during printing were the two parameters investigated. Nine types of TPU samples were fabricated with different configurations of layer height and printing temperature, as shown in **Table 1**.

2.3. Characterization of Printed TPU Substrates

To accurately assess the extrusion of filament material, the mass of the fabricated TPU substrates was measured using the Fisher Scientific PAS214C Analytical Balance scale with a resolution of 0.1 mg. Additionally, the thickness of the substrate was measured using a digital micrometer. To ensure accuracy, three measurements of thickness were taken for each substrate sample. The tomography and roughness of the surface of the printed TPU substrate were evaluated using an optical profilometer (Alicona InfiniteFocus G5, Bruker, Raaba, Austria). Three representative areas, highlighted in red in **Figure 1**, were measured for each sample.

Table 1. Sample fabrication in this research.

Types	Layer height [mm]	Printing temperature [°C]	Quantity of samples
1	0.20	220	3
2	0.20	230	3
3	0.20	240	3
4	0.25	220	3
5	0.25	230	3
6	0.25	240	3
7	0.30	220	3
8	0.30	230	3
9	0.30	240	3

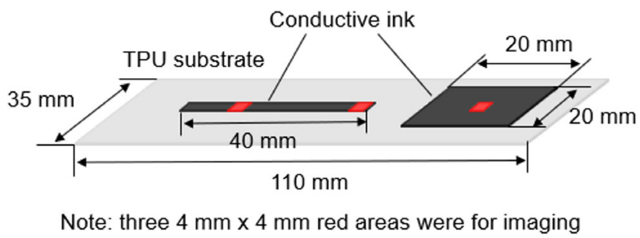

Figure 1. Illustration of the locations in TPU samples for surface measurement.

For profilometer characterization, we measured three representative areas of each sample optically with 10× magnification, and constructed images based on the scanned area, which covered the whole measured field. Then, the form removal was conducted by a profilometer to remove the sample's tilt influence. Next, we conducted surface texture measurement on the selected 4 mm × 4 mm field, which generated data on surface roughness for analysis.

2.4. Ink Printing

To investigate the impact of surface condition of the printed TPU substrate on the quality of silver ink printing, we conducted an experiment using a Neotech AMT PJ15X 3D printer. A straight line (40 mm in length) and a square (20 mm × 20 mm) were printed onto a TPU substrate using silver ink (ECM CI-1036) and cured in a laboratory oven at 130 °C for 15 min. The ink was dispensed using an ink dispenser, which was fed by air pressure and controlled by a rotating screw. The speed of the screw was determined by the extruder feed ratio. After optimizing the parameters for ink dispensing, the following settings were used: air pressure—0.15 MPa, print speed—150 mm min^{-1}, filling speed—300 mm min^{-1}, retraction speed—150 mm min^{-1}, nozzle diameter—0.25 mm, priming distance—2.00 mm, retraction distance—2.00 mm, and extruder feed ratio—0.05.

2.5. Characterization of Ink Printing Quality

To assess the quality and stability of the ink printing, we measured the width of the printed ink line with an Olympus BX60M optical microscope and Olympus cellSens Entry 2.2 software. The 4-wire electrical resistance of the printed 40 mm-long ink line was measured using a Keithley 2425 multimeter. To understand the impact of printing parameters on the adhesion between the ink and TPU substrate, we used the ASTM D3359-22 standard Method B. The cutting was performed using an Elcometer 107 Cross Hatch Cutter, which has six cutting knives in the middle. The cutter was applied to a 20 mm × 20 mm ink square, cutting the middle of the square into 25 small squares. The two cuts were aligned at a 90° angle from each other. An ASTM D 3359 standard adhesive tape (Elcometer, USA) was then applied to cover the entire ink square, and the tape was pressed down with a finger and rubber eraser to ensure proper adhesion. After approximately 90 s, the tape was peeled off steadily from one free end at a 180° angle. The adhesion was then classified according to standards with a magnifier for inspection of ink removal and classification.

3. Results and Discussion

3.1. TPU Substrate Mass

The mass of the 3D-printed TPU samples is shown in **Figure 2**A. A notable trend is that the mass increases as the layer height decreases. For instance, when the layer height is 0.2 mm, the mass is 7%–14% higher than when it is 0.25 mm, and 27%–34% higher than when it is 0.3 mm. Prior to fabricating the samples, we conducted a trial printing of TPU and measured the thickness of the printed layer to be 0.21–0.22 mm. The layer height value controls the upward movement of the nozzle module. When the layer height is 0.2 mm, the amount of extruded plastic is optimized, resulting in tightly compressed layers and the ability to print more layers, ultimately leading to a higher mass. However, when the layer height is 0.25 mm, the nozzle-substrate distance is larger than the thickness of one printed layer, leaving space in between. As more layers are printed, this space increases, as illustrated in Figure 2B. This results in a looser compact of material and more porosity in the printed layers, ultimately leading to a lower mass. Similarly, a layer height of 0.3 mm results in even larger voids and porosity, resulting in smaller mass. Interestingly, the printing temperature does not have a significant influence the mass when using the same layer height. This is because the investigated printing temperatures are sufficient to melt the TPU filament, and therefore, the amount and mass of the material remain unchanged when extruded to build the TPU substrate.

3.2. Surface Morphology of Printed TPU Samples

The surface of printed TPU samples was examined using an optical microscope, and the resulting morphology images are presented in **Figure 3**. It is evident that increasing the layer height significantly improves the fluidity of extruded material and homogeneity of the printed lines. In Figure 3A–C, the samples printed with a layer height of 0.2 mm have flat and homogenous surfaces, while those printed with a layer height of 0.25 mm exhibit random distribution of extra material. This is due to the large gap between the printing nozzle and substrate. When the

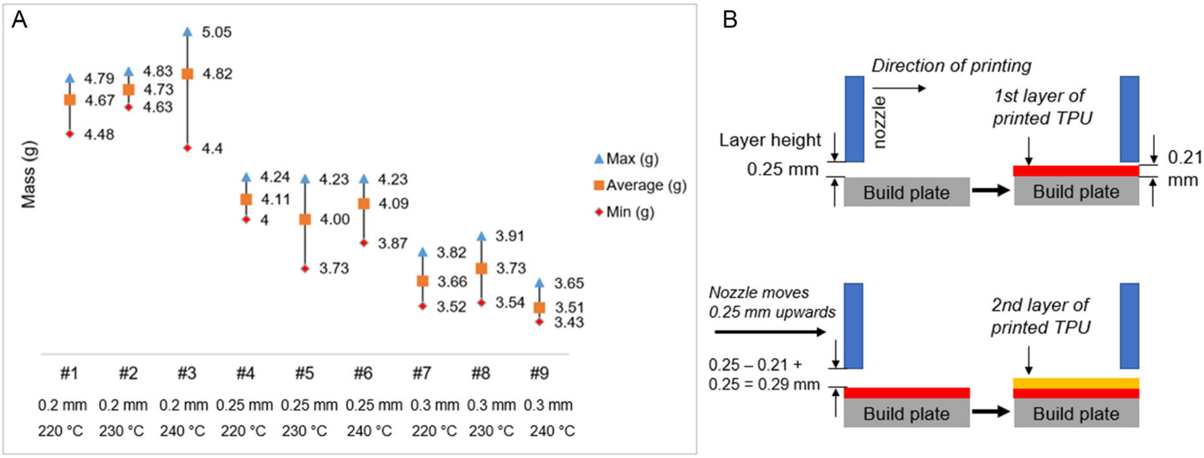

Figure 2. A) Mass of nine types of 3D-printed TPU substrate samples ($N = 3$ for each type); B) illustration of layer height and printed layer in TPU printing.

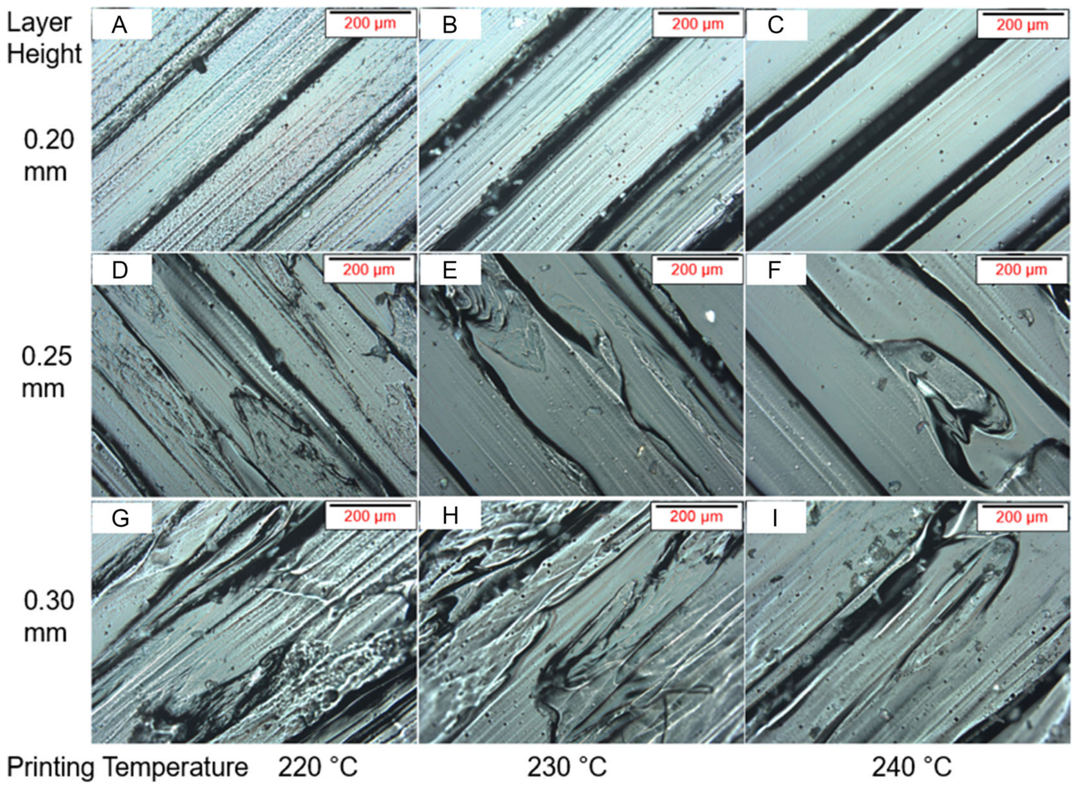

Figure 3. Optical images (with 20× magnification) of the representative morphology of nine types of 3D-printed TPU substrate samples (3 samples for each type). The image serial number from A–I) corresponds to the type #1–#9 samples.

layer height is increased to 0.3 mm, the surface of printed lines becomes rougher, making it difficult to distinguish a flat surface. This finding is consistent with previous studies by Bintara et al.[45] and Yadav et al.[46] on the effect of layer height on the surface roughness of MEX-printed PLA and ABS. However, it should be noted that lower layer heights do not always result in lower surface roughness. After reaching a certain threshold, further decreasing the layer height does not significantly affect the surface roughness. This has also been observed by Bintara et al.[45]

Similarly, increasing the printing temperature results in a larger gap between adjacent printed lines and greater variation in the width and homogeneity of the lines, as seen in Figure 3C compared to Figure 3A. This is due to the decreased viscosity and increased stability of the extruded material at higher temperatures. This effect has been reported in previous studies on MEX-printed PLA by Pang et al.[47] and Frunzaverde et al.[48] They also found that the higher printing temperature in MEX of PLA led to poorer dimensional accuracy and degradation of surface quality and tensile behavior. Setiawan et al.[49] also found that using a printing temperature of 230 °C in MEX of PLA resulted in poorer dimensional accuracy and surface quality compared to a temperature of 220 °C. Nadeem et al.[50] also noted the importance of printing temperature in achieving a desired surface finish in MEX-fabricated parts, but the optimal temperature may vary depending on the material. It is important to consider various process parameters such as filament material, printing temperature, infill density, air gap, raster width and angle, and slicing when attempting to achieve a desired surface roughness in MEX-printed parts.[39] However, in terms of printing stability, lower printing temperatures are generally preferred for MEX of TPU.

3.3. Thickness and Surface Roughness

The thickness of each type of sample was measured using three samples. For each sample, three measurements were taken on both ends and in the middle. The results are shown in **Figure 4**. The printing temperature and layer height do not seem to have a significant impact on the thickness of the printed TPU sample. However, the reasons for this vary. A smaller layer height allows for more layers to be printed, but the material is compressed more tightly. In contrast, a larger layer height results in looser compression and more porous areas, which contribute to the overall thickness. This difference is more evident in the mass of the sample rather than its thickness. Additionally, a higher printing temperature, particularly 240 °C, leads to a greater variation in printing stability, as seen in the mass variation and the surface roughness. Therefore, for stable and consistent printing, 220 °C is considered optimal. However, it may be worth exploring even lower printing temperatures for TPU substrate fabrication, as long as the temperature is sufficient to melt the filament and ensure continuous MEX.

Dimensional accuracy is a challenge for MEX-printed polymers, as reported in previous studies.[51,52] Parameters such as printing temperature,[53] printing orientation,[54] layer height,[55] and filament material[14] have all been found to affect the dimensional accuracy of MEX-printed polymers. In this research, the designed thickness was 1.00 mm, while the actual thickness of the printed TPU substrates ranged from 1.00 to 1.25 mm, as presented in Figure 4A. Similar findings were reported by Nugroho et al.[14] who found a 7–13% error in sample thickness for polyurethane printed by MEX. Hanon et al.[56] also observed a 10.34% error in the thickness of the PLA samples fabricated by MEX. Additionally, Salkuti[57] reported a similar phenomenon with a higher error in thickness than in the horizontal direction for MEX-printed TPU samples. The deviation in sample thickness can be attributed to several factors. First, TPU is a highly flexible polymer with relatively low stiffness, which requires a longer time for solidification. This longer solidification process results in more shrinkage and leads to dimensional inaccuracy, particularly in the vertical direction (thickness) as the sample is built layer-by-layer.[14] Second, printing temperature also plays a role in the dimensional accuracy. Higher printing temperatures can increase the fluidity of the extruded material, resulting in better dimensional accuracy. However, Alafaghani et al.[58] found that within a certain range, an increase in printing temperature can cause the melted filament to flow out of the nozzle's control, resulting in increased dimensional errors. Third, layer height significantly affects the thickness deviation. In this research, an increased layer height led to a smaller number of layers, which was observed to contribute to the thickness deviation. Stojkovic et al.[59] also reported that layer height has a clear impact on the dimensional accuracy of MEX 3D-printed parts, although the specific influence may vary depending on the material and other printing parameters. However, the individual effects of printing temperature and layer height on the thickness deviation are not clear in Figure 4A as multiple factors can influence dimensional inaccuracy simultaneously.

The topography of each TPU sample was imaged using an optical profilometer in three different areas. All three samples were fabricated using the same printing parameters, resulting in a total of nine images from the same type of sample. The variation in top layer surface was then calculated and the results, along with median values, are presented in Figure 4B. Representative images are presented in **Figure 5**. It was observed

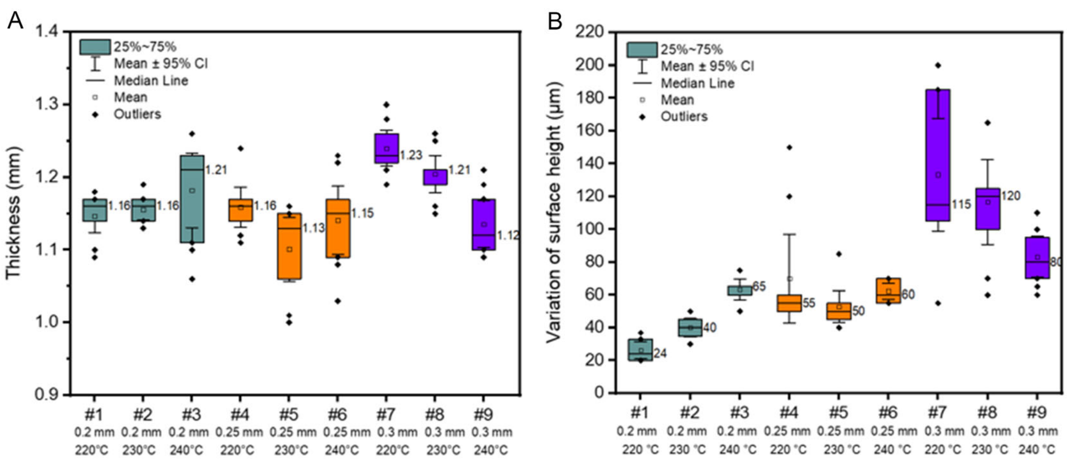

Figure 4. A) Thickness and B) surface height variation of nine types of 3D-printed TPU substrate samples ($N = 9$ for each type).

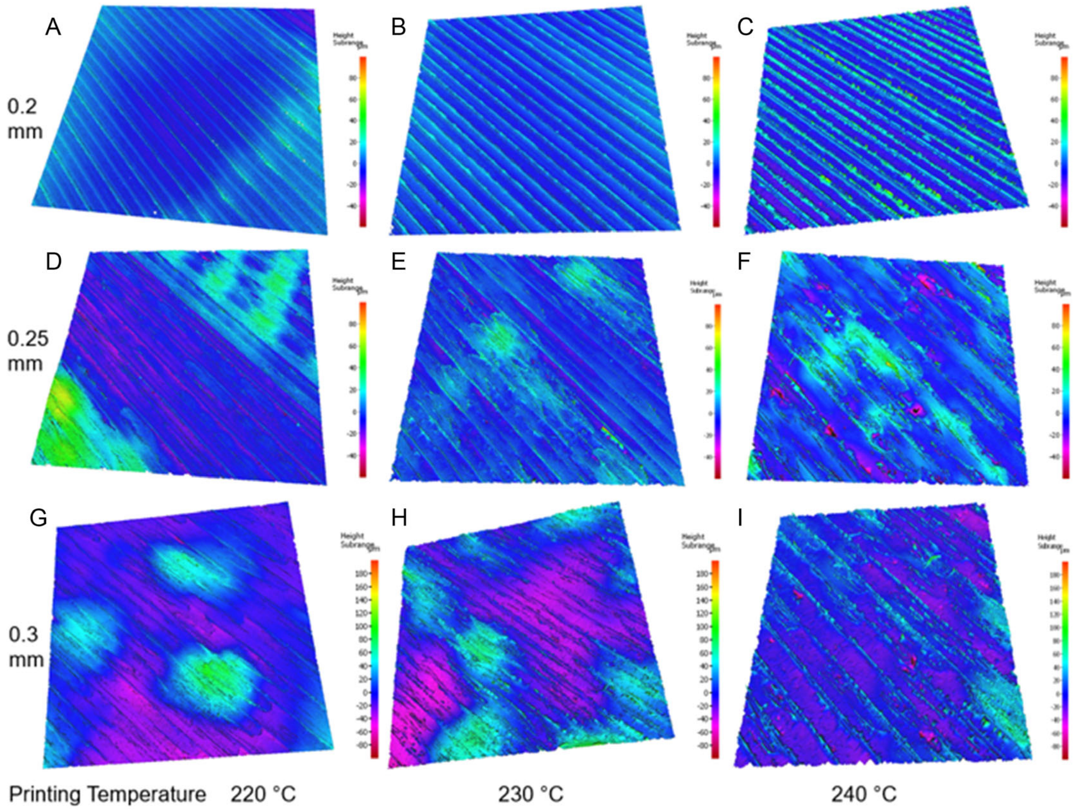

Figure 5. Optical profilometer images (with 10× magnification) of the representative topography of nine types of 3D-printed TPU substrate samples. The image serial number from A–I) corresponds to the type #1–#9 samples.

that an increase in layer height had a dominant impact on the increase in surface roughness. The median values of surface height variation ranged from 24 to 65 μm, 50 to 60 μm, and 80 to 120 μm for samples printed with 0.2, 0.25, and 0.3 mm layer heights, respectively. It also suggests that the accuracy of the printing settings decreases with an increase in printing temperature. Similar findings were reported by Bintara et al.[45] Murjito et al.[55] and Nguyen et al.[21] who found that increasing the layer height in MEX printing of polymers resulted in a higher surface roughness. Cojocaru et al.[53] also claimed that a higher layer height leads to lower part resolution, better interlayer diffusion, smaller voids, and improved surface quality in 3D-printed PLA. The reason for this was explained in Section 3.2.

In addition, increasing the printing temperature resulted in a significant increase in the surface roughness for samples printed with a 0.2 mm layer height, as presented in Figure 4B and 5A–C. This can be attributed to the fact that the thickness of one printed layer is approximately 0.21 mm, and when the space between the nozzle and substrate is set to 0.2 mm, the extruded material is compressed tightly. As a result, the flow of extruded material has a strong impact on the material's landing and surface roughness. At higher printing temperatures, the TPU material becomes less viscous, causing a larger amount of material to flow and spread around the nozzle. This leads to less uniform printing and a rougher surface. This finding is consistent with the results reported by Cojocaru et al.[53] who found that high printing temperatures can cause unstable material flow from the printing head. However, this impact was not as clear when the layer height was set to 0.25 and 0.3 mm. This is because the distance between the nozzle and substrate is larger than the thickness of one printed layer, providing enough space for the extruded material to land even at higher printing temperatures. Interestingly, when the layer height was set to 0.3 mm, the surface was even rougher at lower printing temperatures. This can be explained by the fact that the 0.3 mm distance between the nozzle and substrate is significantly larger than the 0.21 mm thickness of one printed layer. At higher temperatures, the extruded material diffuses faster, resulting in a more homogeneous printing process. In contrast, at lower printing temperatures (220 °C), the diffusion of material is slower, leading to less homogeneous printing and a rougher surface, as shown in the bar graph of #7 samples in Figure 5.

Figure 5 also demonstrates that samples fabricated at a higher temperature tend to have a less uniform and more diffuse pattern on the surface, as evidenced by Figure 3. It is clear that, with the selected 3D printing parameters, a lower layer height of 0.2 mm and a printing temperature of 220 °C result in the optimal printing quality. This is supported by the most consistent and homogeneous printed lines, minimal variation in surface height, and the least variation in surface roughness across different areas of the sample. This conclusion is further supported by Figure 3–5.

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To further characterize the surface roughness of the TPU substrates fabricated under different parameters, three 4 mm × 4 mm square fields were measured in the same locations on each TPU substrate using an optical profilometer based on the standard ISO 4287 (1997).[60] The data obtained are presented in **Figure 6**, in which mean roughness (Sa) measures the deviation of a surface from a mean height, root mean square roughness (Sq) corresponds to the standard deviation of the height distribution, maximum height (Sz) is the sum of the largest peak height value and the largest pit depth value within the defined area, developed interfacial area ratio (Sdr) represents the percentage of additional surface area contributed by the texture compared to the planar definition area.

The results show a significant increase in mean height (Figure 6A), root mean square height (Figure 6B), and mean height (Figure 6C) with an increase in layer height. The optimal fabrication condition was found to be 0.2 mm layer height with a printing temperature of 220 °C. Additionally, the data also indicate that an increase in layer height leads to a larger variation in these parameters, suggesting a less stable fabrication process and inhomogeneous structure. A higher Sdr value indicates a larger slope on the surface. The results in Figure 6D show that when the layer height was 0.2 and 0.25 mm, the surface slopes increased remarkably with an increase in printing temperature. This trend was also observed with an increase in layer height. These findings, along with Figure 3 and 4B, consistently demonstrate the strong impact of layer height on surface roughness, with higher layer heights resulting in a rougher surface and less stable fabrication process. The increase in printing temperature also generally increased the surface roughness of the substrate, which is in line with the findings of Kovan et al.[27] They highlighted the impact of printing temperature on the surface roughness of MEX-fabricated PLA substrate in comparison with other fabrication parameters and claimed that a lower printing temperature improves the print quality. Based on these solid and consistent results, the optimal fabrication parameter configuration was confirmed to be 0.2 mm layer height with 220 °C printing temperature. However, it is important to note that the surface quality of MEX-fabricated substrates can also be influenced by other process parameters, which may also impact the mechanical properties of the fabricated parts. Therefore, the final selection of the process parameters should consider the specific

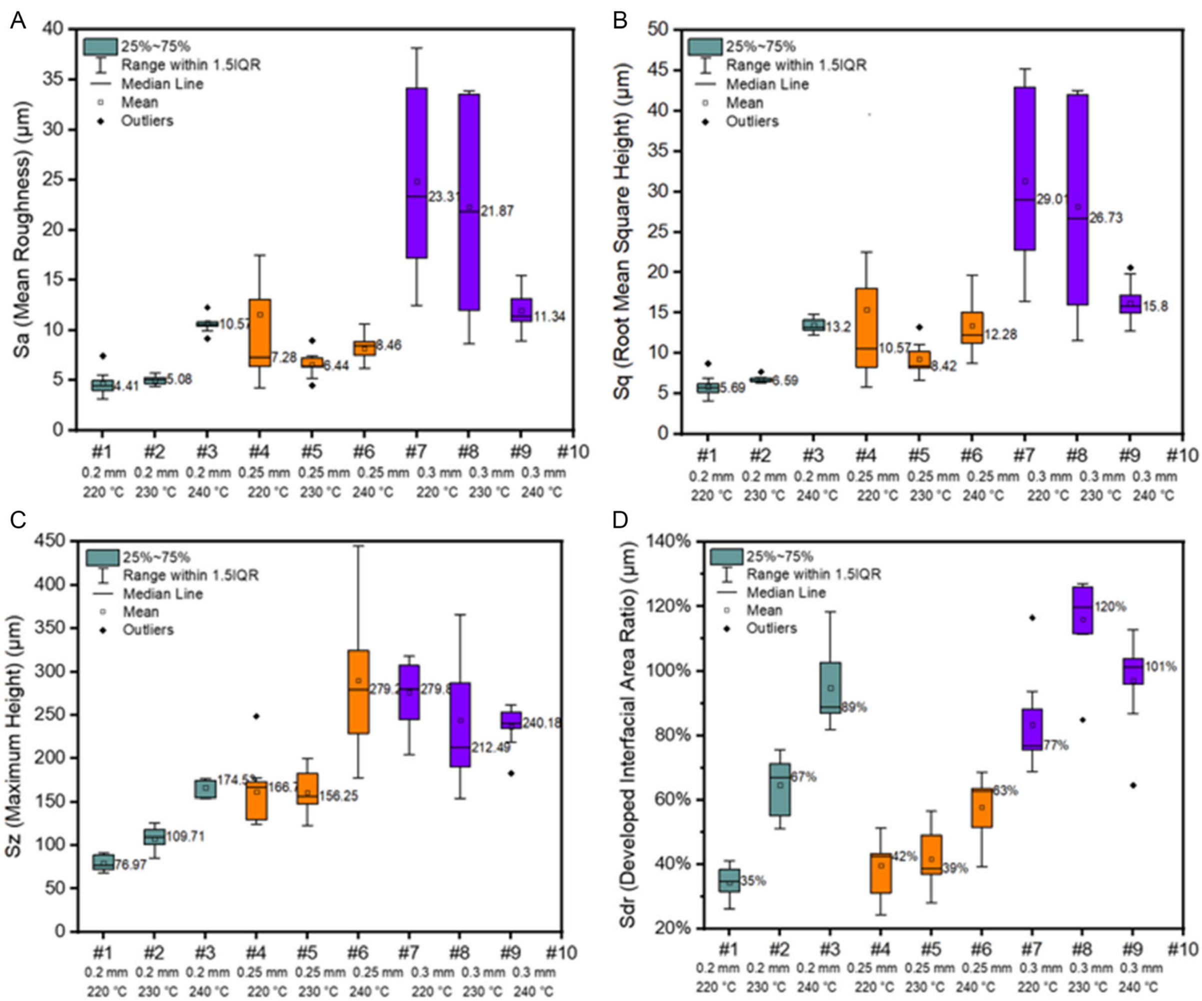

Figure 6. Surface roughness parameters – A) mean roughness, B) root square height, C) maximum height, and D) developed interfacial area ratio of TPU samples fabricated under different parameters.

application and prioritize the desired properties. This was also observed in the research of Mani et al.[61] who optimized the fabrication process parameters for mechanical properties of MEX-fabricated PLA parts and reported similar findings.

3.4. Surface Characterization of Printed Ink

A 40 mm-long line and a 20 mm × 20 mm square of silver ink were printed and cured at 130 °C for 15 min in a laboratory oven. The width of each line was then measured at three different spots, with three of each type of sample being used. This results in nine measurements of the line width for each type of sample. The results are presented in **Figure 7**A. It reveals that, for the TPU fabricated with the same layer height, increasing the printing temperature in TPU substrate fabrication led to wider conductor lines, as demonstrated in Figure 7C–E, which represent conductive ink printed on the TPU substrates. This is due to the higher printing temperature, which caused larger grooves between the adjacent TPU lines, causing more ink to fall into the grooves and resulting in a flatter and wider line. However, this had only a minor impact on the electrical resistance, as presented in Figure 7B.

When the TPU substrate was fabricated with layer heights of 0.25 and 0.30 mm, increasing the printing temperature in TPU fabrication resulted in a slight decrease in electrical resistance. This is due to the fact that higher printing temperatures allowed for a more inhomogeneous diffusion of TPU, resulting in narrower and shallower grooves between the TPU lines, as presented in Figures 7F,G. This allowed for more ink to remain on the surface of the TPU, contributing to better electrical conduction. However, when the layer height was 0.2 mm, the printing temperature in TPU substrate fabrication did not produce an impact on the conductivity of the ink line, as seen in

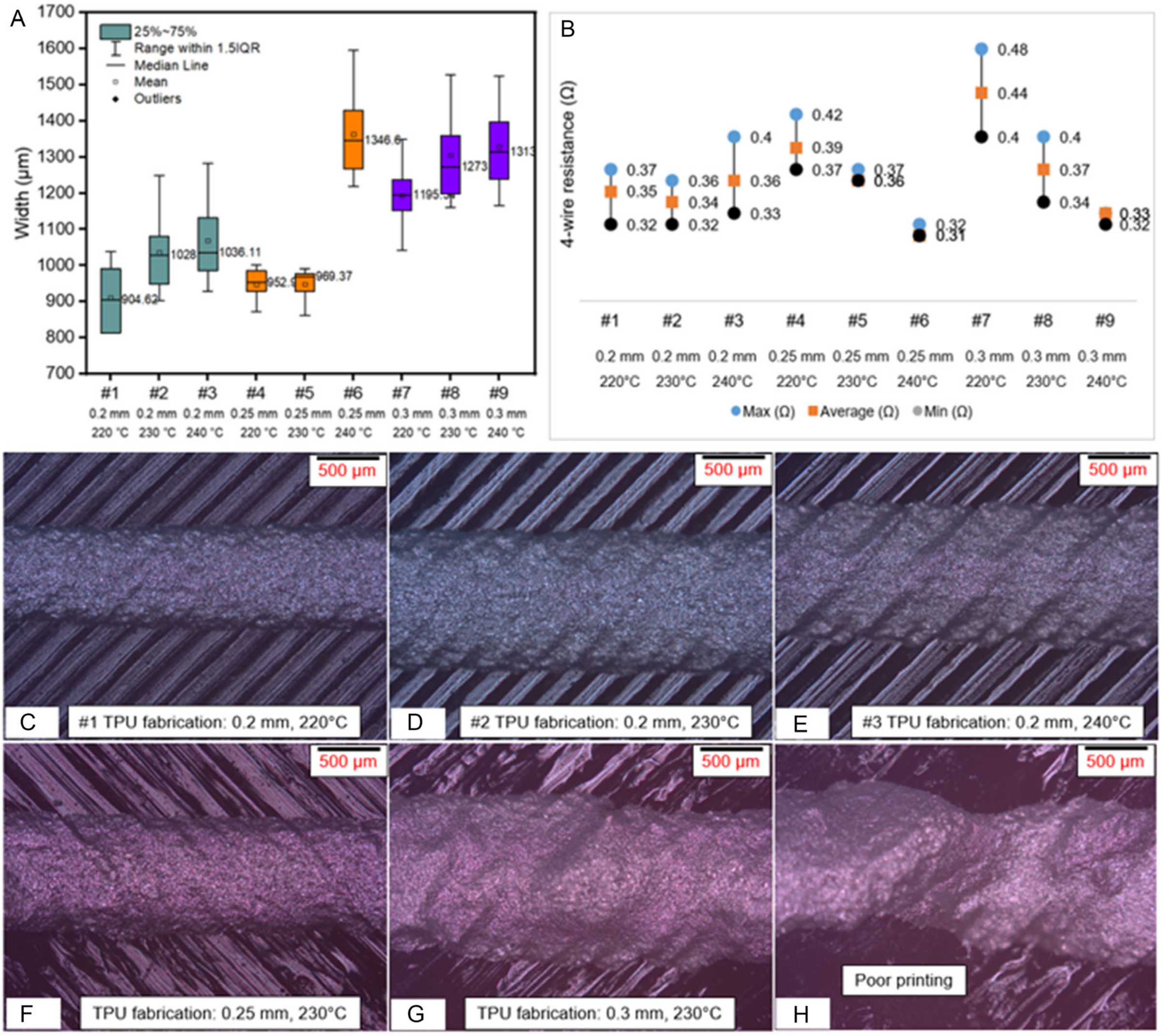

Figure 7. A) Width ($N = 9$ for each type of sample) and B) 4-wire electrical resistance ($N = 3$ for each type of sample) of printed ink line on TPU substrates fabricated under different parameters; representative morphology of ink on C) #1, D) #2, and E) #3 TPU substrates; representative morphology of ink on TPU substrates fabricated under the same printing temperature (230 °C) and different layer height: F) 0.25 mm, G) 0.3 mm, and H) an example of inhomogeneous ink printing due to the diffusion of TPU.

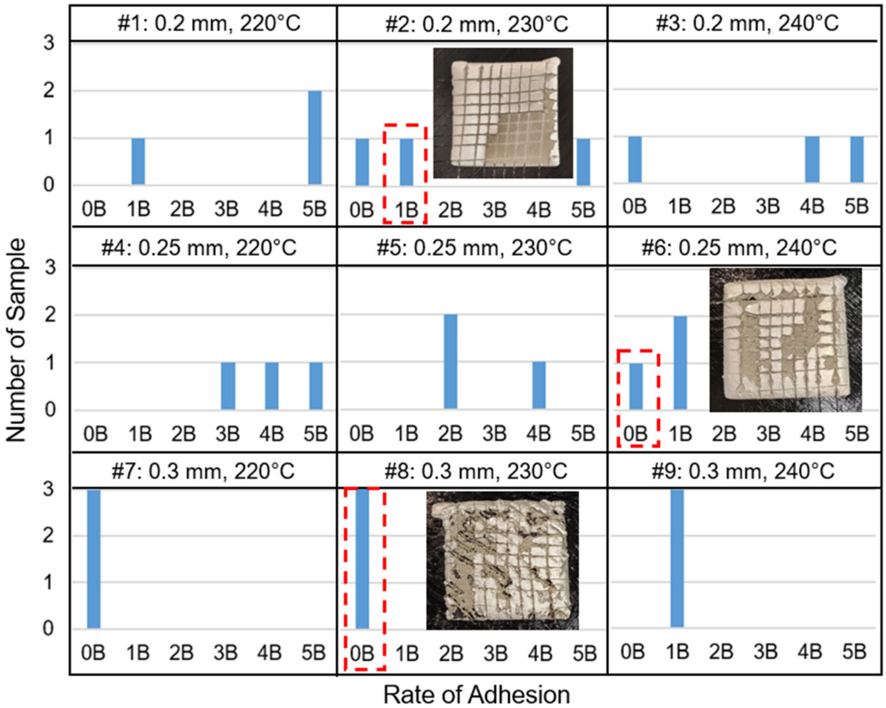

Figure 8. Results of adhesion classification based on ASTM D3359-22 standard method B.

Figure 7B. This is because the substrate fabricated with a 0.2 mm layer height was flatter with small height variation on the surface. Therefore, even though a higher printing temperature resulted in more diffusion, it did not affect the conductivity as the conduction pathway was already well-established on the flat TPU lines.

However, it is worth considering that poor ink printing with an inhomogeneous line width can occur when the TPU substrate is fabricated at high temperatures. An example of this can be seen in Figure 7H. This can easily lead to a discontinuity in the conduction pathway when the printed line becomes narrower. In this study, this was not a major issue as the printed lines were relatively wide, allowing for the conduction pathway to remain intact even with inconsistent printing.

3.5. Adhesion Rating of Printed Ink

The adhesion of conductive ink printed on the TPU fabricated under different parameters was tested, and the results are presented in **Figure 8**. Overall, as the layer height in TPU substrate fabrication increased, the adhesion between the ink and TPU decreased. When the TPU substrate was fabricated with a layer height of 0.2 mm, the adhesion was generally high: 4/9 samples achieved a 5B adhesion rate, meaning 0% loss of ink during tape peeling, and 1/9 samples achieved a 4B adhesion rate with less than 5% ink loss. However, the remaining 4/9 samples had adhesion rates of 1B and 0B, indicating adhesive failure with the same morphology after tape peeling. An example of this can be seen in the sample #2 shown in Figure 8. In comparison, when the TPU was fabricated with a layer height of 0.25 mm, only 3 out of 9 samples achieved 4B and 5B adhesion rates. Additionally, an increase in printing temperature during TPU fabrication resulted in lower adhesion between the ink and TPU. However, in all samples, the ink failed due to a combination of adhesive and cohesive failure, regardless of the printing temperature during TPU fabrication. When TPU was fabricated with 0.3 mm layer height, none of the samples achieved an adhesion rate higher than 1B, indicating a large amount of ink was peeled off or failed. Furthermore, the failure was of the cohesive type, as the example presented in Figure 8. This is because a larger layer height resulted in rougher surface for the TPU, as evidenced by the results in Figures 3 and 6, and a higher variation in roughness, as shown in Figure 5. The rougher surface of the TPU had poor adhesion to the ink printed on it. This finding has also been reported by other researchers.[31,62] Novakovic et al.[63] also reported that the surface roughness of the substrate influences the quality of ink printing significantly. The larger layer height had caused a more porous structure on the TPU surface, and the crosscutting had caused the ink to be more nonflat pieces, as there was little flat surface to achieve high adhesion. This ultimately led to cohesive failure during tape peeling.

4. Conclusion

This research investigated the effects of layer height and printing temperature on the surface roughness of MEX-printed TPU substrates. Additionally, it sought to determine how these factors impact the quality of conductive ink dispensed onto the TPU substrate and the adhesion between the ink and substrate. To assess the accuracy of TPU printing, measurements of mass and thickness were taken. The surface morphology and roughness of the printed TPU substrate were analyzed using optical microscope and profilometer, respectively. The quality of ink printing was evaluated by imaging, and the adhesion between the ink and TPU was measured using the cross-cutting method, a standard qualitative method for adhesion measurement.

It was confirmed that using a higher layer height and higher printing temperature generally results in poorer printing and surface quality of the MEX-fabricated TPU substrate. Among the investigated configurations of layer height and printing temperature, a printing temperature of 220 °C with a layer height of 0.2 mm produced the best results for TPU printing. Besides, the surface roughness and quality of the MEX-fabricated TPU substrate had a dominant impact on the quality and adhesion of conductive traces printed onto it. Increasing the layer height in TPU fabrication resulted in wider conductive lines and lower adhesion between the conductor and TPU substrate significantly, which transformed the ink failure mechanism against tape peeling from adhesive failure to more cohesive failure. However, it did not lead to a linear change in electrical conductivity. In contrast, increasing the printing temperature in TPU fabrication did not have a considerable influence on the dimension and conductivity of the conductor, and had a slighter impact on adhesion compared to the impact of layer height. It also did not affect the failure mechanism when the ink was peeled off with tape.

To promote the development of 3D-printed stretchable electronics, several research directions are recommended: 1) on the basis of optimal configuration of layer height and printing temperature, to investigate the impact of other printing parameters on the printing quality and adhesion of conductor on it, to achieve further improved fabrication quality; 2) to explore the finer conductive structure and its reliability and performance under stretching; and 3) to explore the reliability of embedded multilayer stretchable electronics, in which the conductive structure goes through different layers. By optimizing the design and fabrication process, a fully 3D-printed stretchable electronic device with high reliability is possible to be created.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

Keywords

3D printing, adhesion, material extrusion, printed electronics, structural electronics, thermoplastic polyurethane

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