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Tensile behaviour of continuous carbon fibre reinforced composites fabricated by a modified 3D printer

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#### Abstract

This study aims to highlight the impact of low-volume (7.5%) continuous carbon fibre reinforcement in three different polymer matrices and the effects of post-processing under hot pressing on the mechanical properties of the structures. A fused deposition modelling (FDM) printer's print head was modified to directly extrude the polymer matrix and continuous carbon fibre tow together. Both pure and carbon fibre-reinforced samples were cured under hot pressing at 100 °C and 10 kN pressure for 15 min. All samples underwent tensile and hardness tests, and the microstructure of fractured samples was analysed using a scanning electron microscope. The results indicate that continuous carbon fibre reinforcement and hot pressing are crucial for enhancing the mechanical performance of 3D-printed objects.

# 1. Introduction

Additive manufacturing (AM) is a process of bonding materials to fabricate objects layer by layer. Various additive manufacturing technologies have been developed since the 1980s. They are material extrusion, powder bed fusion, binder jetting, resin-based material jetting, vat photo polymerization, sheet lamination, and direct energy deposition [1]. The most known AM method is fused deposition modelling (FDM), and FDM threedimensional (3D) printer costs are relatively low, and feedstock filaments are inexpensive. Today, 3D FDM printers are still developing and penetrating many industries, such as the automotive, aerospace, medical, food, and construction sectors [2]. In the FDM 3D printing process, a thermoplastic polymer filament is melted in the heated head unit and fed into the nozzle. The extruded filament is deposited on the building platform layer by layer while the nozzle moves around the x and y directions. Travel movements of the nozzle are defined by slicer software. One of the disadvantages of the FDM is the weak mechanical properties depending on the build orientation. Nevertheless, the production of parts with 3D printing technology will develop and shape the future of material production methods and the plastic and composite industry very shortly. 3D printing technology widely uses thermoplastic filaments as well as their composites as a raw material. The introduction of short carbon fibre and continuous carbon fibre reinforced filaments for more than ten years opened a new era for the 3D printing of complex composite structures with high performance and low cost. Therefore, the studies on the use of continuous carbon fibre-reinforced thermoplastics in the three-dimensional fabrications of composite parts with the additive manufacturing method and the improvement of the mechanical properties of the final product are constantly increasing [3, 4]. There are many advantages of the production of parts with 3D printing, such as no need for a mould, the ability to produce complex parts, the use of hybrid materials, and the development of a variety type of different materials [5, 6].

Carbon fibre-reinforced acrylonitrile butadiene styrene (CFR ABS) is a composite material that combines ABS plastic with carbon fibres. The addition of carbon fibres to thermoplastics, such as ABS, polylactic acid (PLA), and thermoplastic polyurethane (TPU), improves their strength, stiffness, durability, and mechanical

properties while also reducing their weight [7]. When comparing the tensile strength-to-density ratio of ABS and PLA with many other filaments, carbon fibre reinforced ABS and PLA composites have a higher strength-to-weight ratio [8]. However, carbon fibre-reinforced ABS is typically more expensive than regular ABS plastic because of the added cost of carbon fibres. Additionally, the production process for this material is more complex than that of plain ABS, which can also contribute to its higher cost [9, 10].

Continuous carbon fibre reinforced (CCFR) polymers are composed of at least two constituents, the continuous fibre as reinforcement and the polymers as the matrix. They are lightweight materials with high strength properties. They can be used in various applications such as aerospace, aviation, automotive, sports, and medical industries. Because of their high strength, stiffness, lightweight, wear, and excellent fatigue properties, continuous carbon fibre-reinforced polymers can be used instead of metals [11, 12]. Peng et al [13] studied the synergistic enhancement of the mechanical properties of continuous and short carbon fibrereinforced polyamide (PA) based composites using the fused filament fabrication (FFF) technique. They found that the tensile strength increased, but the elastic modulus of short carbon fibre and continuous carbon fibrereinforced PA was affected in a bad manner. Chacón et al [3] studied the effect of 3D printing parameters such as build orientation, layer thickness, and fibre volume contents on the mechanical properties of 3D printed continuous carbon, Kevlar, and glass fibre-reinforced nylon thermoplastic composites. They showed that the effects of layer thickness, build orientation, type of reinforcement, and fibre volume content on the mechanical performance of the reinforced nylon specimens were significant. Dou et al [14] attempted to determine the relationship between printing parameters and the mechanical properties of continuous carbon fibre-reinforced PLA composites using a modified FDM 3D printer. They observed that increasing the printing layer height and extrusion width decreased the mechanical properties of the 3D-printed continuous carbon fibre-reinforced PLA composites. They also showed that increasing the 3D printing temperature to 230 °C and printing speed from 50 to 400 mm min<sup>-1</sup> decreases the tensile strength of the CCFR PLA composite samples. Similarly, many researchers have printed samples with continuous or discontinuous carbon fibre and carbon nanotube reinforcement on many matrix materials such as ABS, PLA, polyetheretherketone (PEEK), polyamide (PA), TPU, nylon, and polycaprolactone (PCL) and examined their mechanical properties. In these studies, factors such as the 3D printer brand and specifications, printing parameters, and fibre reinforcement percentage influenced the mechanical properties and cost of the obtained samples [15]. Tian et al [4] underscored the significance of using 3D printing for carbon fibre reinforced polymer composites (CFRPCs) as a transformative technology capable of overcoming disparities between cutting-edge materials and inventive structures. The current state of the art has been meticulously examined, focusing on the interrelationships among materials, structures, processes, performance, and functions in the 3D printing of CFRPCs. This study delineates typical applications and prospects of 3D printing for CFRPCs to comprehend the opportunities and confront the challenges in this domain. Addressing these aspects necessitates extensive interdisciplinary research that encompasses advanced materials, processes, equipment, structural design, and the attainment of intelligent final performance. Therefore, some expensive or relatively cheaper improvement studies have been conducted in the literature to increase the fibre-matrix interface strength. For example, some researchers have reported that preand post-processing techniques for compact printed composites improve their mechanical properties, e.g., hot pressing and heat treatment [16-20]. The tensile and bending strengths of 3D printed polymers with CCF were tested to analyse the effect of pressure. The results showed that pressure significantly affected the CCFR PLA samples [21]. Luo et al [22] attempted plasma-laser treatment of carbon fibres to improve their interfacial bonding with PEEK. Jayswal and Adanur [23] and Kanbur and Tayfun [24] used different reinforcement materials to improve the mechanical properties of the TPU polymer. They used nanoparticles, fibres (aramid, carbon, and glass), and carbon nanotubes as reinforcement materials. Mei et al [25] studied the influence of hot pressing and mixed fibre angles on the mechanical behaviour of 3D printed polymer composites with varying temperatures, pressures, and times. Hot pressing significantly improved the mechanical properties of the carbon fibre-reinforced 3D printed polymer composites. They also found that increasing pressure further lowered the tensile strength and modulus. Similar to laser and ultrasonic strengthening, heat treatment methods, such as hot pressing and annealing, are also effective methods for improving the mechanical performance of FFF printed parts [26]. Jo et al [27] studied the effects of hot pressing on the mechanical strength of PLA parts printed by the FFF process. Although studies exist in the literature for classical polymers such as ABS and PLA, there has been no study specifically on the application of hot pressing to improve the mechanical properties of TPU polymers after 3D printing.

This study aims to compare the mechanical behaviour of pure, and continuous carbon fibre reinforced (with 7.5% in volume) PLA, ABS, and TPU tensile test specimens fabricated by FDM 3D printing technology. Additionally, apply hot pressing to these samples to investigate the effect of the hot pressing on the fibre-matrix interface strength, mechanical properties, and hardness changes. Furthermore, compare the results obtained from the hot-pressed specimens with those that were not subjected to hot pressing. The study also aims to



analyse the microstructure of fractured tensile test samples with scanning electron microscopy (SEM) and identify and verify the internal structure of the continuous carbon fibre-reinforced polymer composites.

#### 2. Materials and methods

#### 2.1. Materials and equipment

In this study, PLA, ABS, and TPU thermoplastic filaments, 1.75 mm in diameter, were used as matrix material. 3000 (3 K) continuous carbon fibre tows, each 7  $\mu$ m in diameter, were used to prepare the composite materials (CCFR thermoplastics). The term 'continuous' refers to the carbon fibre thread, rather than thermoplastics such as TPU, ABS, or PLA. Some thermoplastic filaments used in 3D printing are made as composite filaments by adding short (chopped) carbon fibres in specific proportions. However, pre-made short or chopped carbon fibre composite filament was not used in this study. These fibres have high mechanical properties such as a tensile strength of 3500 MPa and 235 GPa a modulus of elasticity. The density of the carbon fibres is 1.8 g cm<sup>-3</sup>.

#### 2.2. Printing of CCFR thermoplastic specimens with the FDM Method

The simultaneous insertion of solid polymer filament and continuous carbon fibre into the extrusion nozzle of a 3D FDM printer causes problems that need to be overcome [28, 29]. Therefore, the printing head of the FlashForge Creator 3D FDM printer was modified to print a polymer matrix with carbon fibre strands directly in the printing head. Due to its flexibility and limp nature compared with polymer filaments, carbon fibre roving cannot be directly fed into the heating block. The modified printing head unit for printing the carbon fibre roving filament with the polymer matrix is shown in figure 1.

The carbon fibre roving is introduced into the heating unit through a nearby hole, which is drilled at an approximate 40° angle just after the exit of the extruder. It is then dragged into the nozzle by the polymer matrix filament. Polylactic acid, acrylonitrile butadiene styrene, and thermoplastic polyurethane were used as the polymer matrix. The modified 3D printing head can process several polymers and various fibres to mix them in the printing head.

Before initiating the 3D printing process, the continuous carbon fibre roving filament was coated with a TPU based chemical solution.

To prepare the TPU based chemical solution, 1 g of Bayer Desmopan TPU 192 plastic resin raw material was added to 4 g of tetrahydrofuran (THF,  $C_4H_8O$ ) under magnetic stirring at 50 °C. The chemical solution is applied to the continuous carbon fibre tow using a spraying bottle and then spread along the continuous carbon fibre tow. This application prevented the printing nozzle from clogging with carbon fibre strands. TPU-based solution smeared onto the continuous carbon fibre thread was used during the 3D printing of all ABS, PLA, and TPU tensile test specimens. This ensured the seamless embedding of the continuous carbon fibre thread into the polymer without breakage. A conical nozzle (1.4 mm in diameter) was employed during the printing process. The polymer matrix and continuous carbon fibre tow were inserted into the heating block unit through the driving gear of the extruder. As shown in figure 1, the 3D printer's extruder is supplied simultaneously with filaments such as ABS, PLA, or TPU, and continuous carbon fibre thread. This allows the molten filament and



Parameters	Units
Printing speed	$0.6 \text{ mm s}^{-1}$
Nozzle diameter	1.4 mm
Layer thickness	0.8 mm
Number of shells	2
Feedstock multiplier	0.88
Environment temperature	23 °C
Infill percentage	82%
Infill extrusion width	1.4 mm
Nozzle temperature	260 °C
Feed diameter	1.77
Infill pattern	linear

Table 1. Parameters of 3D printing.

continuous carbon fibre to combine in the nozzle and be printed on a temperature-controlled building plate, resulting in a composite tensile testing sample.

TPU is much more flexible and tackier when compared to other more rigid filaments. These make 3D printing with TPU challenging for several reasons. The first challenge with TPU is that the greater the distance between the nozzle and the extruder drive mechanism, the harder it is to feed the flexible filament like TPU to the nozzle without it buckling. Secondly, TPU's lower viscosity compared to other filaments complicates 3D printing. The printing parameters must be carefully set to avoid issues such as spreading and sagging on the building plate during the 3D printing process.

In figure 2(a), the tensile test specimens were prepared according to the ISO-527-4: Type1-A standard [30]. A STereoLithography (STL), also referred to as 'Standard Triangle Language', or 'Standard Tessellation Language', file is used to slice the test specimens by the open-source MakerWare slicing software [31]. In figure 2(b), it can be seen that the 3D printed test sample consisted of 5 layers, with each layer being 0.8 mm thick. The build platform temperature was set at 100 °C, and the printing speed was 1 mm s<sup>-1</sup>. Detailed 3D printing parameters can be found in table 1.

Travel movements of the printing nozzle are highly critical during 3D printing. The nozzle must continue its movement without interruption on the same layer. Of course, after the working layer is completed, the building platform must be lowered by the layer thickness. Even in this case, the nozzle must not be moved further away from the previous layer's finishing position. Otherwise, the carbon fibre thread will be broken, and the continuity effect will be lost while the nozzle moves to a new position. Because of this, the path followed by the nozzle must be a continuous line, and the working layer must be completed without lifting the nozzle or lowering the building platform, both in the shelling and infilling stages of the 3D printing of the test sample. As a result of the specially designed infilling and shelling platform need to be lowered during the shelling and infilling stages of the test sample's 3D printing process. Some literature underscores that the development of



additive manufacturing methods is an ongoing process. There is a recognized need for novel design methods and tools in this evolving field to surmount challenges and generate effective solutions [32-34]. In figure 2(a), the depicted path illustrates the trajectory that the printing head unit will follow. As depicted in figure 2(b), the overlapping lines were minimized, allowing the printing head to transition from the external shell toward the interior. The infill raster angle is configured to be 0°.

During the layer change, the nozzle can continue to move without lifting. The macroscopic images of the CCFR PLA and its fracture surface are shown in figures 2(b) and (c), respectively.

These images were captured from the tensile samples derived from the CCFR PLA filament (figures 2(a) and (b)).

#### 2.3. Fibre volume fraction

The fibre volume fraction can be calculated using mathematical equations according to the diameters of the fibre and nozzle. For this, equations (1)–(3) were used. For each CCFR thermoplastic (TP) filament, the fibre volume fraction is constant (because of the 3 K fibre bundle) [35].

Total Fiber Area = 
$$3000 \times \frac{\pi \times (\text{Singular Fiber Diameter})^2}{4}$$
 (1)

Nozzle Area = 
$$\frac{\pi \times (\text{Nozzle Diameter})^2}{4}$$
 (2)

Fiber Volume Fraction(%) = 
$$\frac{\text{Total Fiber Area}}{\text{Nozzle Area}} \times 100$$
 (3)

When calculations were performed using the above formulas, the fibre volume fraction was calculated as 7.5% for a nozzle diameter of 1.4 mm.

#### 2.4. Hot pressing treatment

Tensile test samples were prepared to compare the mechanical properties of carbon fibre-reinforced polymers with those of pure PLA, ABS, and TPU. Moreover, both pure and carbon fibre-reinforced tensile specimens underwent hot press post-processing, as illustrated in figure 3, to investigate the potential impact of curing on the polymers.

The curing process was performed at 100 °C and 10 kN pressure for 15 min. The compressive pressure applied to the sample was 4.0 MPa.

#### 3. Experimental part

#### 3.1. Tensile tests

Following the 3D fabrication of continuous carbon fibre-reinforced test specimens with various polymer matrices, tensile tests were conducted to ascertain their tensile strength. Dog bone-shaped tensile test samples were employed to investigate the mechanical behaviours of continuous carbon fibre-reinforced polymer matrix composites. The Zwick/Roell uniaxial testing machine was utilized for the tensile testing, with an extensiometer measuring the extension and strain of the specimens under a constant tensile load. The loading rate was set at 1 mm min<sup>-1</sup>.

Table 2. Tensile properties of the printed polymer matrices.

Polymer matrix	Tensile strength (MPa)	Elongation at break(%)	Elastic mod- ulus (MPa)
ABS	42.8	4.85	1413
PLA	59.8	1.18	1530
TPU	20.0	440	48

#### 3.2. Hardness tests

Hardness tests were conducted following the procedure outlined in ASTM D2240. A standard handheld digital Shore-D hardness tester (HT-6510D) was used for the hardness tests. Fifteen tests were performed on both the top and bottom surfaces of the samples, with great care taken to avoid indentation points on the edges of the samples, as well as from previous measurements and surface roughness of the samples. The upper surfaces of the samples, which did not undergo hot pressing, exhibited significant roughness, surpassing that of the lower surfaces [36]. Both hot pressing and the surface flatness of the 3D printer's base plate resulted in smoother bottom surfaces. Therefore, the hardness test results obtained from the bottom surface are more reliable and consistent [37].

Although there is not much difference between them, the hardness results for the bottom surface have been included in the evaluation. To reduce the scatter of the data and obtain more accurate results, the maximum and minimum hardness measurements are not included in the average hardness.

#### 4. Results and discussion

The average results of the tensile tests for the printed polymer matrix materials obtained with the 3D printer are summarized in table 2.

#### 4.1. Effects of fibre addition and hot-pressing

The stress– strain curves obtained from the tensile tests are presented in figure 4. Changes in both the strength and strain values of the samples were observed after 7.5% volume carbon fibre reinforcement and hot pressing under high pressure for pure ABS, PLA, and TPU samples, as shown in figures 4(a)–(c). As shown in figures 4(a) and (d), the average strength of the pure ABS samples was 42.8 MPa, whereas the average strength of the CCFR-ABS samples with carbon fibre reinforcement increased by 11.9% to 47.9 MPa. In addition, the average strength of the pure ABS samples increased by 2.3% from 47.9 MPa to 49.0 MPa. The strain of the pure ABS samples decreased by an average of 3% and 25% after carbon fibre reinforcement and hot pressing, respectively. As shown in figures 4(b) and (d), the average strength of the pure PLA samples was 59.8 MPa, whereas the average strength of the CCFR-PLA samples with carbon fibre reinforcement increased by 20.9% to 72.3 MPa.

Unlike ABS, the average strength of the hot-pressed CCFR-PLA samples decreased by 2.6% from 72.3 MPa to 70.4 MPa. The strain of pure PLA samples decreased by 48.8% after continuous carbon fibre reinforcement, whereas it increased by approximately 5.47 times after hot pressing. As shown in figures 4(c) and (d), the average strength of the pure TPU samples was 20 MPa, whereas the average strength of the CCFR-TPU samples with continuous carbon fibre reinforcement increased by 41.5% to 28.3 MPa.

In contrast to CCFR-PLA, the average strength of the hot-pressed CCFR-TPU samples increased by 59.7% from 28.3 MPa to 45.2 MPa. The most notable change in strain was observed in the TPU, as shown in figure 4(c).

While continuous carbon fibre reinforcement caused a decrease in strain by approximately 36.79 times (from 440% to 22.89%) for pure TPU samples, hot pressing resulted in an increase of approximately 1.91 times for CCFR-TPU samples.

#### 4.2. Effects of hot pressing on 3D-printed pure ABS, PLA and TPU

As shown in figure 4(d), hot pressing did not cause a significant change in the average strength of pure ABS and PLA (5% and 4.7% respectively), but it led to a 23.5% increase in the average strength of pure TPU samples. As seen in figures 4(a)–(c), hot pressing resulted in a decrease in the strain value of pure ABS by approximately 7.2%, whereas pure PLA and TPU exhibited an increase in strain by approximately 3.46 and 1.06 times, respectively.





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Upon careful observation of figure 4(b), it is evident that, unlike ABS and TPU samples, the tensile modulus and strain values of CCFR-PLA-hot pressed samples have significantly decreased compared to those of CCFR-PLA (figure 4(b)). This is believed to stem from the following reasons:

- a) As seen in figure 5(a), before hot pressing if attention is given to the cross-section of the sample, it can be observed that fibres (shown with light blue ellipses) are arranged regularly compared to their post-hot pressing state (figure 5(b)). However, after the high-pressure hot pressing of 10 kN, irregular fibres (crimp patterned fibres) in the x, y, and z directions are apparent (figure 5(b)). Therefore, the angle  $\emptyset > 0^\circ$  between the fibres and the *x*-axis is due to these irregularities, directly affecting the sample's tensile modulus. Hence, it is thought that these irregularities lead to fibre breakage, wrinkling, and waviness. Despite this, in figure 5(b), the total volume of fibres and the total fibre section that carries the applied load have not changed significantly compared to figure 5(a), resulting in the obtained tensile stress being the same. However, after hot pressing, the tensile modulus has decreased, and the strain has increased.
- b) Before starting the 3D printing process, carbon fibres were coated with a TPU-based chemical solution. Since the relaxation temperature of TPU is higher than PLA's, a flexible TPU layer remained on the carbon fibre-PLA interface during the 100 °C hot pressing process. The presence of this flexible interface reduced the tensile modulus and increased strain.
- c) Since the pressing temperature (100 °C) is above the softening temperature of PLA (61.7–67.9 °C) and a high pressure of 10 kN was applied, PLA samples may behave as if they have undergone a stress-relaxation process, leading to a decrease in tensile modulus and an increase in strain.

The results of the tensile tests are given as one sample in figure 6 for all groups. As shown in figures 6(b), (d), (f), (h), and (j), an improvement in surface roughness is observed under hot-pressed samples, whereas expansion is naturally observed in other directions.

Pure TPU obtained a considerably high average strain of 400–440% compared with other samples. As shown in figures 6(l) and (m), pure TPU and hot-pressed TPU showed continuous elongation without breaking in several tensile tests. Pure TPU samples were not broken in the tensile tests because of their very high strain.

#### 4.3. Relationship between tensile strength and hardness

The average hardness results for both surfaces are shown in table 3. It is observed that both the hardness and strength of the pure ABS, PLA, and TPU samples increased after carbon fibre reinforcement and hot pressing. There is a notable correlation between the tensile strength and hardness of both hot-pressed and non-hot-pressed samples [38]. Additionally, the hardness results are consistent with the tensile results and the findings of Harikrishnan and Soundarapandian [37].

Although there is not much difference between them, the hardness results for the bottom surface have been included in the evaluation. In order to reduce the scatter of the data and obtain more accurate results, the maximum and minimum hardness measurements are not included in the average hardness.

In some sources in the literature, the relationship between the hardness and strength of composite materials has been investigated [39, 40], and the formula S = k.H has been mentioned [41]. Where S represents strength, k is the coefficient, and H is the hardness value. In this study, the relationships for ABS samples can be expressed approximately as S = 0.65H, for PLA samples as S = 0.93H, and for TPU samples (excluding hot-pressed CCFR-



**Figure 6.** Tensile tested specimens; (a) pure ABS, (b) hot-pressed ABS, (c) CCFR-ABS, (d) hot-pressed CCFR-ABS, (e) pure PLA, (f) hot-pressed PLA, (g) CCFR-PLA, (h) hot-pressed CCFR PLA, (i) CCFR-TPU, (j) hot-pressed CCFR-TPU, (k) pure TPU before, (l) pure TPU after testing, (m) pure TPU after hot pressing and testing (400% strain).

#### Table 3. Tensile strength and hardness.

Material	Tensile strength (S) (MPa)	Hardness (H) (shore-D)	k		
Pure ABS	$42.8\pm1.44$	$69.60 \pm 1.02$	0.61		
Hot-pressed ABS $45.0 \pm 0.60$		71.28 ± 0.83			
CCFR-ABS 47.9±7.07		$71.42\pm0.92$	0.67		
Hot-pressed CCFR-ABS	$49.0\pm2.09$	$72.15 \pm 1.00$	0.68		
Pure PLA	$59.8 \pm 3.21$	$69.02\pm0.96$	0.87		
Hot-pressed PLA	$62.6\pm0.53$	$69.25\pm0.90$	0.90		
CCFR-PLA	$72.3\pm8.73$	$72.52\pm1.03$	1.00		
Hot-pressed CCFR-PLA	$70.4\pm 6.82$	$72.48\pm0.66$	0.97		
Pure TPU	$20.0\pm0.27$	$30.98\pm0.71$	0.65		
Hot-pressed TPU	$24.7\pm1.52$	$40.37\pm0.73$	0.61		
CCFR-TPU	$28.3\pm2.53$	$43.25\pm0.86$	0.65		
Hot-pressed CCFR-TPU	$45.2\pm4.26$	$43.42\pm0.50$	1.04		

TPU) as S = 0.64H. However, for hot-pressed CCFR-TPU samples, this formula should be given as S = 1.04H. This is because, as shown in table 3, the hardnesses and strengths of the hot-pressed CCFR-TPU samples are almost equal.

#### 4.4. Microstructural observations of the fractured surfaces

Scanning electron microscopy (SEM) was used to analyse the fracture behaviour of the fractured samples. SEM analysis was performed to identify and verify the internal structure of the polymer composite and examine the fracture mechanism [42].

To better understand the effect of carbon fibre reinforcement and high-pressure hot pressing on the composites, microstructure evaluation was performed on the cross-section of the samples. The interface between the fibre and matrix plays a pivotal role in influencing the mechanical properties of composites due to its impact on load transition [4].

The strength of fibre-reinforced composites is highly dependent on the interfacial adhesion strength between the matrix, and the fibres which have in the longitudinal direction of the force, particularly [43, 44]. The composite having the stronger fibre matrix adhesion yields higher strength [45].

As depicted in figures 7(a) and (b), the fibres that have detached from the matrix appear exceptionally clean, with no residual matrix material adhering to them.

Additionally, the void created by fibre-matrix interfacial separation is quite large, as seen in figure 7(b). This phenomenon occurs because ABS inherently possesses low adhesion capability to fibres. During high-pressure hot pressing, the fibre-matrix interface undergoes elliptical expansion, leading to fibre peeling and breakage. Although the tensile strength of the hot-pressed CCFR-PLA was decreased (2.6%) much less than that of the non-pressed, both hot-pressed and non-pressed, is seen as more optimistic than the result described for CCFR-ABS above.



**Figure 7.** (a) SEM image of CCFR-ABS, (b) SEM image hot-pressed CCFR-ABS, (c) SEM image of CCFR-PLA, (d) SEM image of hot-pressed CCFR-PLA, (e) SEM image of hot-pressed CCFR-TPU, (f) SEM image of hot-pressed CCFR-TPU.

In the SEM image of CCFR-PLA shown in figure 7(c), the fibres that have separated from the matrix exhibit some impurities, with attached matrix residues visible on the fibres. It seems that the fibres have experienced some strain during the process of detachment from the matrix. However, the abundance of fibre voids separated from the matrix by breaking or stripping without breaking is still notable. The voids observed in the separated fibres from the matrix in figures 7(c) and (d) serve as a significant indicator of inadequate fibre-matrix interfacial strength. In figure 7(d), the void created by fibre-matrix interfacial separation in hot-pressed CCFR-PLA is seen to be of smaller diameter than in hot-pressed CCFR-ABS. Additionally, the fibres are much cleaner, and they appear to separate more easily from the matrix compared to non-pressed CCFR-PLA. As seen in figures 7(e) and (f), carbon fibre residues are still attached to the damaged CCFR-TPU specimens. The latest findings show that the presence of fibre voids, observed in CCFR-ABS and CCFR-PLA, is uncommon in this case. These observations indicate that the material surrounding the fibres has a strong bond to them, suggesting a higher level of interfacial adhesion compared to other samples.

In general, hot pressing has been observed to enhance the mechanical properties, including hardness, of both pure and carbon fibre-reinforced samples. The results of this study indicate that the tensile strength of carbon fibre-reinforced ABS and TPU composites increases with hot pressure treatment. On the contrary, the tensile strength of carbon fibre-reinforced PLA composites exhibited a 3% reduction (table 3).

The printing and softening temperatures of TPU are approximately 210 °C–235 °C and 61 °C, respectively. The printing temperature of ABS is between 230 °C and 250 °C, with a softening temperature between 110 °C and 125 °C. The printing temperature of PLA ranges from 190 °C to 220 °C, with a softening temperature between 61.7 °C and 67.9 °C. In this study, to eliminate the effect of temperature variation, all test samples were printed at a nozzle temperature of 260 °C. The hot pressing process was conducted at 100 °C. The softening temperatures of the PLA and TPU were lower than the hot-pressing temperature. Therefore, further studies should explore different temperature and pressure ranges in future research.

However, as the pressure increases, the thickness of the sample decreases, leading to a decrease in shape accuracy, a change in surface quality, and even a slight decrease in strength, such as in CCFR-PLA samples, by 2.6% [19, 21]. With increasing pressure, the surface quality of all samples visibly improved, while the thickness decreased. Considering that the thickness of the final part obtained in applications will decrease and its dimensions will change, the dimensions of the initial part obtained from the 3D printer need to be calculated within tolerances.

As shown in table 4, literature reports provide coefficients of variation in mechanical properties resulting from parameters such as fibre volume fractions of 9.1–50%, low pressing pressures of 0.1–0.4 MPa, pressing temperatures of 160–370 °C, and pressing times of 10–80 min. In contrast to existing literature, this study achieved notably favourable results by employing a pressing pressure ten times higher than the maximum pressure reported in the literature. The achievement of these favourable results is noteworthy, especially considering the utilization of a low carbon fibre content of 7.5% in volume and a relatively modest pressing temperature of 100 °C. There have been no studies in the literature regarding hardness, and some studies have not evaluated strain and modulus of elasticity.

Significant differences of this study, compared to the literature, are the 1.6-fold increase in strength accompanied by a 1.91-fold decrease in strain and a 1.51-fold increase in modulus of elasticity in hot-pressed CCFR-TPU, and the 1.03-fold decrease in strength accompanied by a 5.47-fold increase in strain and a 4.64-fold decrease in modulus of elasticity in CCFR-PLA.

As shown in table 3, the tensile strength of ABS and TPU samples reaches approximately 64%–65% of the hardness value measured in Shore-D units, while in PLA samples, the strength is around 93% of the hardness value. However, in hot-pressed CCFR-TPU samples, the strength and hardness values are almost equal. Even if it is a relatively small amount, such as 7.5 percent in volume, the use of a continuous carbon fibre-reinforced polymer matrix significantly increased the strength of the tensile specimens compared with conventional pure polymer specimens.

As a result of the high-pressure hot pressing of ABS and PLA samples, the easier separation of fibre-matrix interfaces with increasing pressure has caused a decrease in strength to some extent.

As mentioned in the study by Mei *et al* [25], an increase in pressure was observed to push (spread) the matrix material to both sides, causing a decrease in stress by leaving the fibres unattached to the matrix. It was observed that there was a weaker interface bond with more gaps between the fibre and matrix in non-hot-pressed samples (figure 7). Fibre breakage or fibre matrix separation is the main cause of sample failure [42]. As seen in table 3, the addition of carbon fibre and high pressure made the pure CCFR-ABS and CCFR-PLA samples slightly harder. Essentially, a portion of the fibres experience breakage, while others are pulled out from the matrix. The high-pressure matrix exerts force from both sides, leaving the fibres unbound, leading to a reduction in tensile strength and an increase in modulus [25]. Despite pure TPU having a strain value of 400–440%, the fibre-matrix interface strength with carbon fibre reinforcement is significantly higher than other CCFR-ABS and CCFR-PLA samples, even with the interface strength increasing further with hot pressing. These improvements have

Matrix	Fibre volume fraction (%)	Pressure (MPa)	Temperature (°C)	Time (min)	Strength (times)	Strain (times)	E modulus (times)	Hard-ness (times)	Study
Nylon	50	0.2	270	10	2.00	N/A	1.68	N/A	[16]
Nylon	9.1–10.9	0.2-0.4	160	10	1.25	1.73-2.16	1.14-1.21	N/A	[25]
PEEK	59	0.15	370	80	1.25	1.04	1.17	N/A	[18]
Nylon	35	0.1	230	10	1.63	N/A	1.33	N/A	[17]
PLA	10.3	1.0	180	15	2.65	N/A	N/A	N/A	[21]
ABS	7.5	4.0	100	15	1.02	1.38	2.95	1.01	This study
PLA	7.5	4.0	100	15	1.03	5.47	4.64	1.01	This study
TPU	7.5	4.0	100	15	1.60	1.91	1.51	1.01	This study

Table 4. Mechanical properties of 3D printed carbon fibre-reinforced composites after hot pressing.

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increased the tensile strength of CCFR TPU from an average of 20 MPa to 45.2 MPa, an increase of 126%. Following hot pressing, the TPU matrix exhibited the strongest interfacial bond with the fibre material, with all fibres completely enveloped by the matrix material. Therefore, as evidenced by both the test results in figure 4 and the SEM images in figures 7(e) and (d), the strong interfacial adhesion and formation of matrix residues in TPU could increase the surface roughness of the fibre and increase mechanical interlocking.

# 5. Conclusion

In this study, the mechanical behaviour of pure and continuous carbon fibre reinforced (with 7.5% in volume) PLA, ABS, and TPU tensile test specimens, which were fabricated by FDM 3D printing technology, were investigated. Additionally, hot pressing was applied to these samples to investigate the effect of the hot pressing on the tensile test samples. The results of the hot-pressed and non-hot-pressed specimens were compared. The fractured surfaces of the continuous carbon fibre-reinforced polymer composites were analysed using scanning electron microscopy, and the internal structure of those composites was assessed. The Shore-D hardness values of all tensile samples were identified by the ASTM D2240 standard, and they are evaluated and compared with each other. Hot-pressed continuous carbon fibre reinforced TPU samples exhibited strong interfacial bonding with carbon fibre strands, and this significantly increased the tensile strength of CCFR TPU samples from 20 MPa to 45.2 MPa which is a nearly126% increase in strength. This remarkable change in the mechanical properties of TPU with carbon fibre reinforcement and hot pressing could lead to many potential applications.

Many 3D printing parameters were considered during the fabrication of CCFR composite test samples. The ideal printing speed of the matrices with continuous carbon fibre thread was  $0.6 \text{ mm s}^{-1}$ , and all samples were printed at the same printing speed to ensure fair comparisons. The shrinkage and elongation that occur after hot pressing with TPU filament can pose challenges for some practical applications, but the hot pressing process can tolerate these shape changes.

The challenges overcome in the study can be listed as follows:

- making several modifications to the printer's printing head and nozzle,
- · determining the optimal parameters after numerous printing and hot pressing sessions,
- coating the continuous carbon fibre thread with a special chemical solution prepared for this study. (1 g of Bayer Desmopan TPU 192 plastic resin raw material was added to 4 g of tetrahydrofuran (THF, C<sub>4</sub>H<sub>8</sub>O) under magnetic stirring at 50 °C).

In future studies, 3D-printed continuous carbon fibre-reinforced ABS, PLA, and TPU samples should be tested with different hot-pressing temperatures and pressures based on the intended use and purpose of the final product to identify optimum parameters. While there are few problems with carbon fibre reinforcement in 3D printing of ABS and PLA, it was concluded that 3D printing of continuous carbon fibre reinforced TPU is challenging due to its extreme flexibility, viscosity, adhesive behaviour, and the tensile forces that occur after hot pressing. Nonetheless, the reinforcement of TPU with continuous carbon fibre in 3D printing and hot pressing at high pressure appears to be a promising material for various industrial applications.

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# Data availability statement

The data cannot be made publicly available upon publication because no suitable repository exists for hosting data in this field of study. The data that support the findings of this study are available upon reasonable request from the authors.

# **Declaration of competing interest**

The authors declare that there are no conflict of interest.

#### **Ethical statement**

Not applicable.

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