

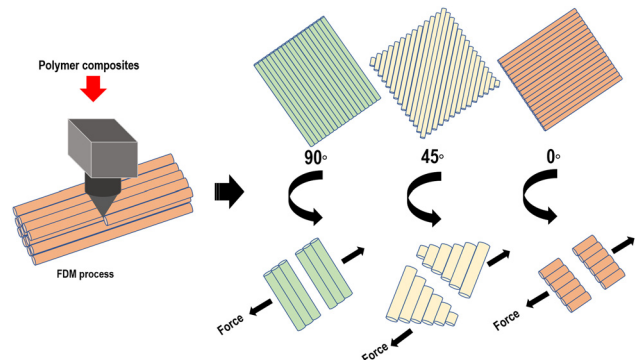
## Review Article

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# Layup sequence and interfacial bonding of additively manufactured polymeric composite: A brief review

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**Abstract:** Additively manufactured polymeric composites exhibit customised properties beyond those offered by conventionally fabricated ones. However, in many cases, the mechanical performance mainly depends on the processing parameters, tools, and material selection. Yet, one of the issues of the additive manufacturing process especially in the material extrusion process is the inability to control the printing layups, thereby causing inter-laminar damage. Thus far, literature and research have focused on improving the mechanical performance of such polymeric composites by focusing on the inter-laminar shear strength under a transverse load transfer. Polymeric composites prepared using the material extrusion technique namely fused deposition modelling (FDM) are discussed upon its layup sequence and orientation. This article proposes that by realising a homogenous distribution of the transverse load, the orientation and the printing direction can maximise the printed load bearing. Moreover, the layup sequence and the interlayer diffusion are key for controlling the mechanical properties of the polymeric composites. This brief review presents a comprehensive elucidation of the polymeric composites manufactured using FDM that interprets the needs of having greater load bearing in each layup printing sequence of the polymeric composites. By able to control the layup



Graphical abstract

sequence, one can control the mechanical performance based on specific functionality.

**Keywords:** fused deposition modelling, polymer composite, mechanical properties, interfacial bonding

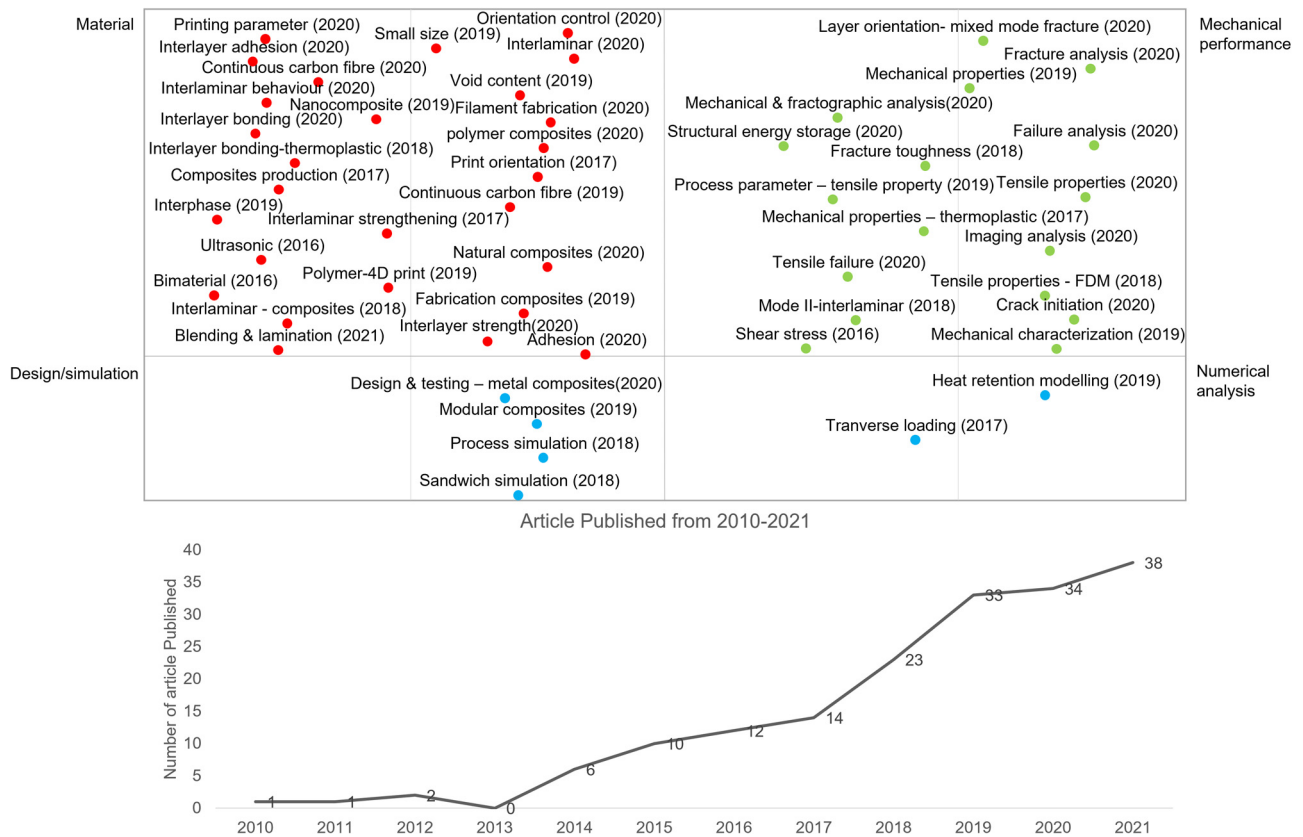
## 1 Introduction

Additively manufactured polymeric composites, particularly those produced by fused deposition modelling (FDM), are well-known in research and industrial fields. This technology has gained significant attention because of its simple operation and strong ability to produce customised parts at minimum cost [1,2]. Conventional polymer composite fabrication processes include injection moulding [3,4], compression moulding [5,6], thermoforming [7,8], and extrusion, [9,10]. The advantages and disadvantages of these fabrication methods have been extensively discussed in past decades [11–13]. Yet, what interests the researchers and industries of the FDM is the ability to control the performance while removing the post-process phase [14,15]. However, the fluctuations and inconsistencies in the printed parts owing to the use of different equipment as well as the limited customisation of the filament result in varied mechanical performance [16]. Because the FDM technology has rapidly developed, approaches that can overcome the

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**Figure 1:** Number of articles published with keyword “layup sequence” *versus* year, showing drastic increase in number of publications over time.

above limitations are yet to be identified, requiring detailed study and future consideration, particularly of the layup sequences [17]. Consequently, since 2010, there has been an interest in studying the layup sequence of printed polymeric composites and its mechanical performance (Figure 1). In the first quarter of 2021, 38 published articles were recorded in ScienceDirect, showing the importance of this issue.

For further development of the FDM technique, adapting it to different types of materials with different compositions is crucial because this can widen its applications. However, manufacturing by FDM can be challenging because adjusting the compositions of polymeric composites at high fibre loading leads to fibre breakage during the U-turn printing motion or inappropriate bonding of the materials themselves [7,18,19]. Similar findings on Nylon-woven composites indicated the evidence of fibre breakage that caused migration of damage initiation and crack propagation, which was 13% lower than the predicted simulated model [20]. This issue of a sudden 50% reduction in tensile break strength due to the deformation phenomenon [21] also occurred when using pure polymer in stacking sequence [90°/0° printing direction]. This deformation, experienced by the pure polymer or even by the polymeric composites, suggested a relationship

with the interlayer bonding during the printing process [22,23]. This further results in material shrinkage and therefore speeds up the fracture occurrence [22,24,25]. Synchronisation within the materials’ composition as well as the processing parameters are expected to cure the printed bonding with minimum macroscopic void and fibre breakage [26–28]. In the year 2021, studies on laminated theory on polymer composites clearly exhibited that there is maximum appearance of porosity, especially in the region with the highest amount of fibre recorded in random printed composites [16,28,29]. Studies on the printed thermoplastic (polyethylene terephthalate glycol, PETG) reported necking behaviour as the deformation occurrence as porosity started to aggravate [23,30]. Consequently, the interaction within the layup sequence adjacent to the intermolecular slips within the printed materials will weaken [29,31,32]. These slips might be due to the poor intermolecular diffusion as the two surfaces bond together at above the glass transition temperature, also known as the diffusion theory of adhesion [33,34].

Additive manufactured polymeric composites are critical when it comes to maintaining their mechanical performance with a limited amount of fibre breakage [35]. A study by Dickson and Dowling [36] highlighted the need

for larger sweeping corners to avoid high corner radii of less than 2 mm as they are nylon-coated carbon fibre (CF) and printed as a woven structure. During the process, they printed the nylon-coated CF on top of the unheated polyvinyl acetate (PVA)-coated Garolite plate and succeeded in allowing adequate adhesion during the printing process [36]. Also, the inability to maintain peak mechanical performance is due to the fact that printed materials often experience material blockage [37], “no travel move” or known as fibre bundles folding back [36], overlapped fibres [20], incompetent continuous fibre [38], poor interfacial adhesion [39,40], and poor placement of printed path (minimum stress trajectories) [41]. These issues are mainly due to the filament itself, which is customised *via* several techniques, including filler coating [39], in-nozzle impregnation [20,35], mechanical mixing (twin screw extruder) [37], and extrusion [38,42], as ways to ensure peak mechanical performance.

Hence, to address such challenges due to the printing complexity, an advanced technique with a layer-by-layer printing process is required. Therefore, this study serves as a review of recent modifications in the layup sequences [43] and interlayer diffusion [44,45] of additively manufactured polymeric composites. The review initially highlights the modern progression of polymeric composites from the prospect of additive manufacturing. Subsequently, the issues of delamination, static stress distribution, and interlaminar shear strength are discussed in detail, which are considered as the main challenges in the current FDM technology [46]. Following this, the efforts for overcoming these challenges are highlighted in terms of the filament scale geometries and the relation between the printed orientation and the layup sequences of polymeric composites [17,47]. The bonding formation and the effects of the structural alignment on the mechanical performance of such materials are discussed based on the performance of the parameters in terms of printing speed, temperature, infill, and others. Thus, the fact that the layup sequence can improve the interlayer diffusion must be considered during the FDM printing process to ensure best performance.

## 2 Modern progression of FDM of polymeric composites

Owing to the advancement of additive manufacturing and the successful development of metals [48,49], ceramics [50], plastics [51], and composite materials [52,53], intense research has been conducted to realise integrated technologies for printing dissimilar materials. Additive manufacturing has been expanded to metamaterials for

predefined optical and acoustic applications, containing meta-atom structures accumulated on complex structures [54,55]. Such technologies, which rapidly manufacture complex product outcomes, are beneficial for both industries and economic growth. Hashemi Sanatgar *et al.* [56] emphasised the need of using polymers with strong diffusion of chainlike molecules to ensure excellent adhesion during polylactic acid (PLA)-polyamide (PA) printing process. The diffusion theory, which explains the need for having adhesion force of deposited polymer nanocomposites, minimised the deposited layer break strength as mentioned by Shi *et al.* [57]. The increase in adhesion generally relates to the interfacial bonding within the fibre-matrix, which deteriorates with the increase in fibre content, according to a study employing CF-acrylonitrile butadiene styrene (ABS) composites [58]. These were supported by Dickson *et al.* [59] who explained that these phenomena were due to the increment in air voids. Researchers have been investigating the benefits of multi-materials in terms of a wide range of properties and functionalities since the early 2015s [60]. Although the use of multi-materials has sufficient potential for future industries, their multi-step fabrication processes frequently increase the overall costs [61,62] and result in poor adhesion and interfacial bonding [60]. Recent studies exhibited that fabrication using multi-materials requires techniques such as laser powder bed fusion [63], material jetting [64,65], and material extrusion [2,66]. Tee *et al.* [64] developed a multi-material rubber-like composite that was printed using material jetting and had a maximum tensile stress of 7 MPa. According to the study, these were caused by crack propagation that starts in the softer material regions [64]. These findings suggest that some additive manufacturing processes, such as material jetting, are not truly ideal for ensuring multi-materials' peak performance. Hence, employing FDM techniques on multi-materials (PLA, ABS, and high impact polystyrene [HIPS]) was reported to be practical due to the thermoplastics having equivalent heat capacities with maximum tensile strength of 10.78 MPa [67]. In 2020, Baca and Ahmad [68] reported that the multi-material FDM printed by a multi-nozzle increased the tensile strength by 30%. Employing multi-materials gave a huge advantage in controlling the type of materials used based on its specific need. Sanz-Horta *et al.* [69] used the FDM technique to print poly- $\epsilon$ -caprolactone (PCL) and PLA in porous structures, while maintaining a Young's modulus of up to 400 MPa. However, the major drawbacks are the inability to align the manufacturing parameters of different printing systems and the poor printing precision [68,70]. In addition, the adhesive thickness in single-material parts has been extensively studied;

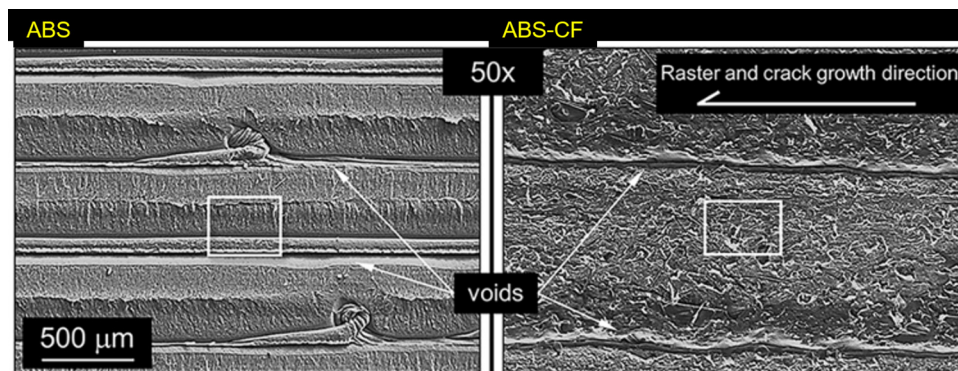
in contrast, few investigations have been conducted on multi-materials or composites [65,71]. In view of the demand for multi-materials, the addition of fibres in a polymer matrix leads to various functionalities, such as the ability to balance the mechanical, thermal, and electrical performance based on the fabrication mechanism [72]. In 2020, several strategies were adopted to adapt the FDM technique for producing tissue engineering scaffolds or even printing environmental sensors using a gelatin hydrogel on PLA [72,73]. Thus, in research, the FDM technique is preferred over other additive manufacturing processes available in the market. The approaches for the FDM technique, particularly for printing complex structures using tandem or multiple input feeders, have sufficient potential; however, the issues of homogeneous mixing and others of significance are yet to be examined [51,74].

Although the FDM process has been used in industries for sufficient time, research and development are still conducted to understand the unique properties achieved using it. One of the interesting features of the FDM technique is its ability to control and modify the mechanical properties of the composite materials. However, interference on the printing path frequently destroys the fibre orientation, and hence, causes fibre damage, thereby deteriorating its overall performance [72]. Several studies reported the occurrence of warpage of the finished parts, which were initially related to the differences in the arrangements of the printed particles during the process [64,75,76]. One study employing PA6/CF composites reported that there was evidence of a 230% increase in tensile strength when the composites were printed in an XY orientation [77]. The degree of residual strains that fit the printed arrangement was accountable for this outstanding performance [78]. This contributes to an anisotropic diffusion in the flow printing direction, thereby adversely affecting the mechanical performance of the printed materials [79,80]. This was proven in a study on an ABS-CF specimen, which showed based on the

force-displacement data, a decrease in displacement by 44% for the ABS-CF composite compared to ABS at 160 N. Further investigation reported that the ABS-CF composite possesses a lower fracture toughness ( $0.49 \text{ kJ/m}^2$ ) than pure ABS ( $2.1 \text{ kJ/m}^2$ ) [79]. The lowering of the performance was caused by the significant plastic deformation during crack growth, as shown in Figure 2. Apparently, during the printing process, the CF filler was either completely separated from or partially bonded to the ABS resin. In addition, the existence of voids or micro-voids is the most common challenge in the printed thermoplastic materials. Studies have revealed that the poor adhesion between the fillers and resin, apart from the vaporisation of the volatile compounds, contributes to the propagation of large defects under stress loading [80–82]. Adhesion in composite materials is crucial, particularly in biomaterial applications, because adhesion aids in spreading growth cells onto the printed scaffolds [19,82,83]. Therefore, it is critical to not only focus on the parameters and processing of the FDM technique but also to address in detail the major contribution of the mechanical performance of the printed polymer composite parts under the effect of their structural behaviour.

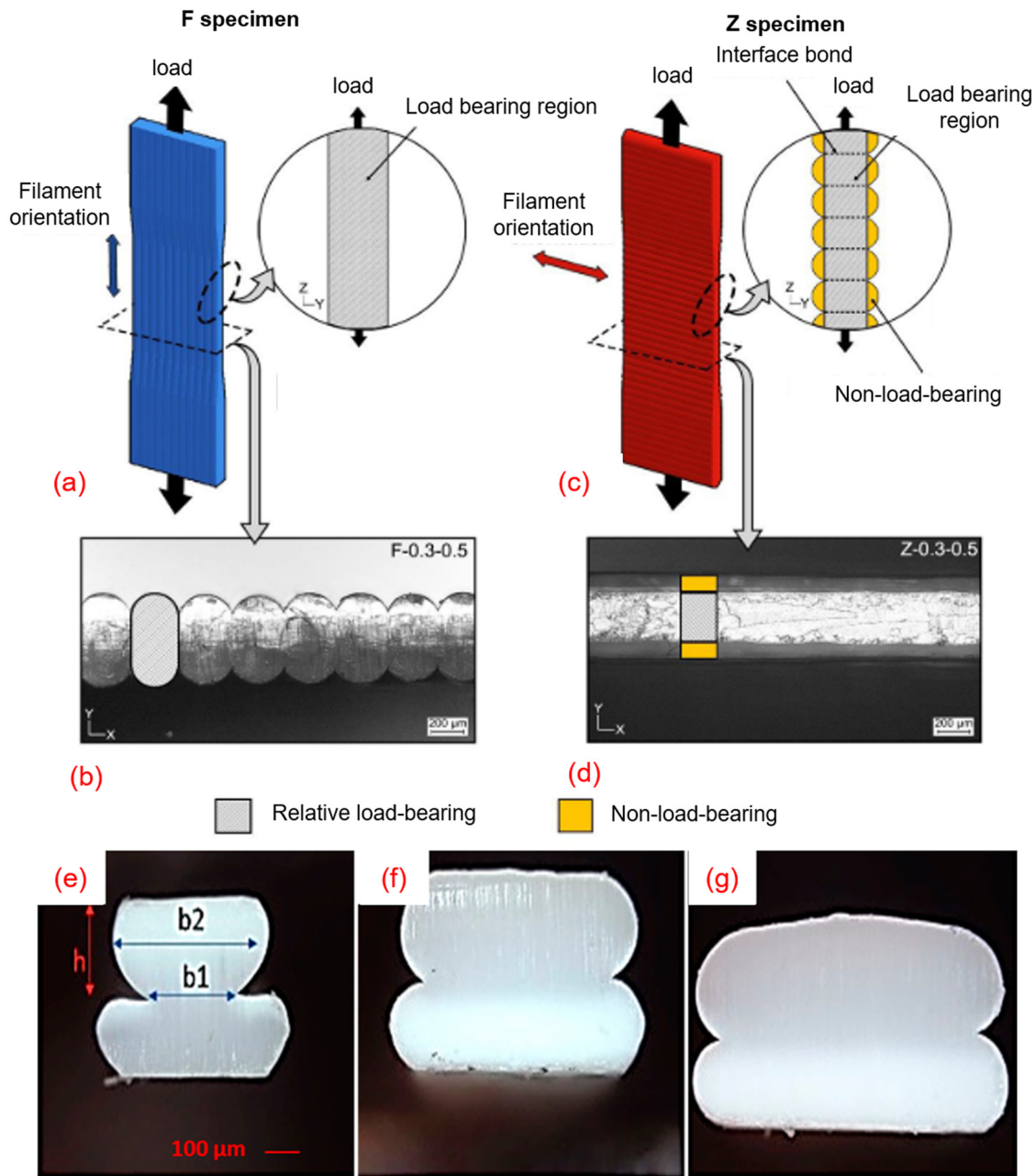
### 3 Filament scale geometry

Controlling the distribution of the printed materials to achieve appropriate alignment in a precise localised area will aid in improving the interlayer fusion. This will lead to strengthening of the overall printed structure owing to the avoidance of delamination and voids during the process. A single void or even weak interlayer bonding of a printed material frequently causes structural failure and deteriorates the overall performance. Interlayer fusion is a common issue in most additive manufacturing processes,



**Figure 2:** Micrograph images of ABS and ABS-CF fracture surfaces. Reproduced with permission [79].

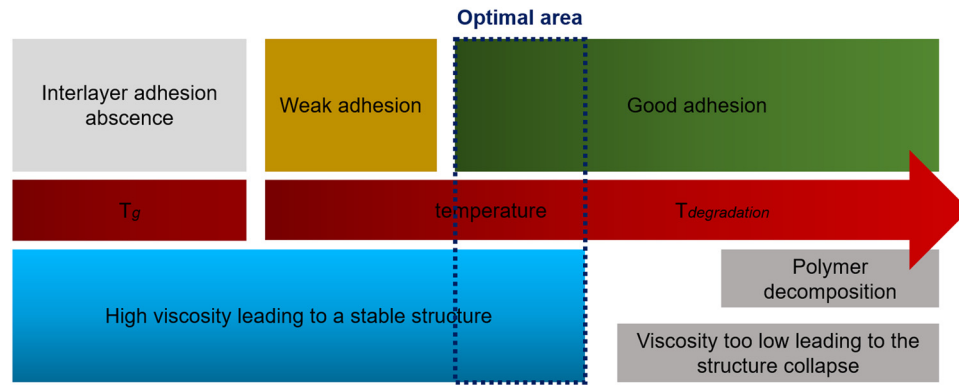




**Figure 3:** Schematic of (a) longitudinal cross section and (b) relative load-bearing area of *F* specimen. Schematic of (c) transverse cross section and (d) relative non-load-bearing area of *Z* specimen. (e–g) Printed layup sequence tracks of interfacial adhesion at various widths. Reproduced with permission [1,87].

including vat photopolymerisation, material extrusion, and selective laser melting [84,85]. However, latest findings in 2020 clearly indicated that it is not the poor inter-layer bonding which is frequently claimed to deteriorate the mechanical performance of the printed composite materials, but the filament scale geometry is the feature affecting the mechanical performance the most [73,86,87]. Simulation studies by Aliheidari *et al.* [88] claimed that materials printed in the diagonal *F*-direction (Figure 3a)

are more localised and can maintain the peak mechanical performance (40%) compared to those in the *Z*-direction (Figure 3b). This is because *Z*-direction printing causes grooves on the surface, resulting in a poor load-bearing area that cannot sustain more load than the *F*-direction printed materials [88]. These results clearly indicated that adjusting the printed geometrical scale is dependent on the anisotropic behaviour of the materials, allowing the performance to be altered based on the demand [87–89].



**Figure 4:** Optimal printable condition of the polymeric composites.  $T_g$  is the glass transition temperature and  $T_{\text{degradation}}$  is the thermal degradation. Reproduced with permission [24].

The mechanical performance of the polymer composites is reliant on several factors, including geometry [87], interlayer adhesion behaviour [88,90], threshold volume fraction [89], and absorption-desorption [91]. This finding was supported by a study on polymer-structured materials prepared using the FDM technique, which highlighted the problem of positioning printed materials in the Z-direction causing visible void accumulation in the range of  $5 \times 10^5$  to  $6 \times 10^5 \mu\text{m}^3$  [26]. Therefore, to reduce this void generation, the printed materials were heated using an infrared preheating system (IPS), leading to a quasi-homogenous appearance as the voids disappeared [26]. Although there is no such evidence for a groove structure under heat application, an increase in the thermal energy is expected, which will affect the polymer chains in composite materials and their quasi-static properties [92,93]. Adjusting the scale geometry of a printed material relies on the interlayer cooling time for maintaining its quasi-static mechanical properties. Different aspects were addressed by performing experiments on the composite materials. Occasionally, some findings highlighted that adjusting the parameters, including the printer temperature, does not have a significant effect on the mechanical performance [86,87,94]. However, the findings on wood polymer composites prepared using the conventional compression mould technique indicated that the cool-down process significantly improves the crystallinity of the composite materials [95]. In 2018, a research on ABS-reinforced chopped CFs prepared using the FDM technique reported that a rapid cool-down does not allow the minimum time for appropriate polymer chain entanglement, and hence, significantly deteriorates and reduces the bond strength with adjacent rasters [79]. It is worth noting that controlling the interlayer bonding temperature is an effective approach for tuning the material strength. Studies reveal that the composite materials should be cooled down to below the glass transition temperature to

allow excellent correlation between the interlayer cooling and the mechanical properties of the adjacent materials, which is attributed to the prolonged cooling and the weakened interlayer bonding as illustrated in Figure 4 [57,92]. Using infrared radiation to preheat a printed surface allows the materials to cool down to below the glass transition temperature before printing the next layer on top [96]. Clearly, a fundamental study on the cooling mechanism is required in the near future because it will enable the adjustment of the properties of the composite materials.

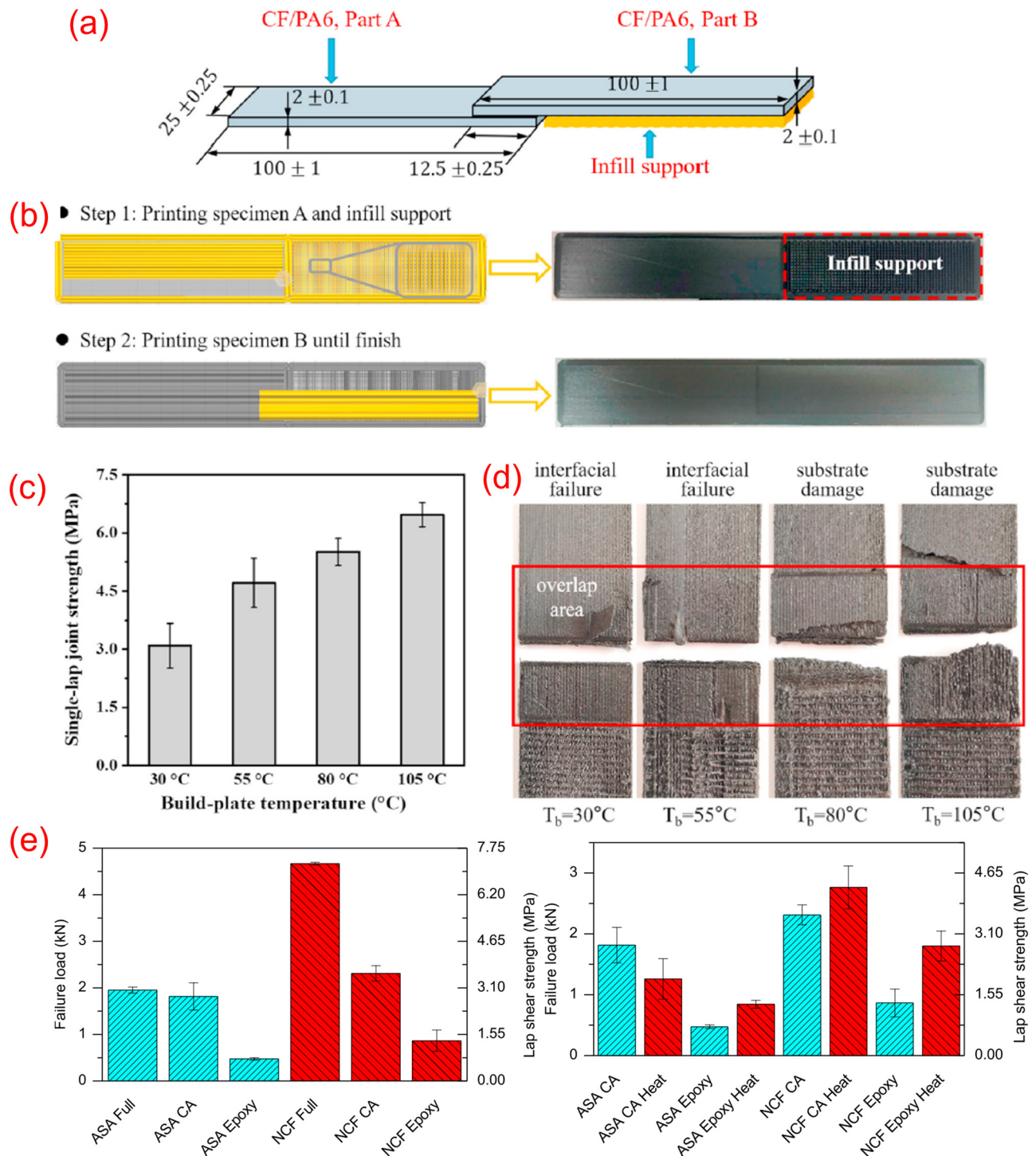
## 4 Relation between printed orientation, layup sequence, and interfacial adhesion

Laminated and oriented composite materials have been used since the 1960s because they ensure material performance based on application needs [97,98]. Apart from the filament scale geometry, the orientation and the layup sequence are interesting features to be considered in the near future [99]. Although lamination of composite materials is frequently based on standard quad laminates of  $0^\circ$ ,  $\pm 45^\circ$ , and  $90^\circ$  plies, it is difficult to study the different stiffness parameters of different composite materials [99,100]. The study reported that this phenomenon occurred because of the high interfacial bonding strength between the adherend and the adhesive, particularly when  $\theta$  increased from  $45^\circ$  to  $90^\circ$ , based on the interlocking theory [101]. This has become the interest regarding composite materials for decades. A latest study on laminated composite materials revealed the importance of the removal of air and volatiles owing to the damage caused by them [102,103]. However, studies on blade turbines

using CF-reinforced composites indicated that the failure of composite materials also depends on the structural design because these structures are subjected to stress loads [102]. According to Ahmed *et al.* [104], having the right materials composition and printing structure allows for better flow properties, as opposed to only increasing filler content. Since it is well-known that adding more filler content degrades the strength properties of the composites, suitable percolation threshold for composite materials must be established [104]. In contrast, according to a study published in 2020, the mechanical performance of the composite materials is strongly correlated with the build-plate temperature, as it has a close relationship with interlayer adhesion [105]. Peng *et al.* [105] showed that the interfacial bonding of a single-lap shear sample with zigzag infill pattern (Figure 5a and b) increases up to 116% from 3 to 6.5 MPa when the build-plate temperature increases from 30 to 105°C (Figure 5c and d). It should be emphasised that the research of polymer composites necessitated the extension analysis of single-lap shear joints, particularly to confirm the interfacial bonding of the upper and lower printed parts. According to Yap *et al.* [106], a study on different adhesive (epoxy and cyanoacrylate [CA]) on the acrylonitrile styrene acrylate (ASA) and Nylon 12 carbon fibre (NCF) samples revealed that CA has higher adhesive strength than epoxy, even at higher temperatures as shown in Figure 5e. Also, as discussed by Strieman *et al.* [26], post-manufacturing or assistance is required during the post processing, such as using an IPS [26]. The study mentioned that the aid of IPS (printed samples in the printing chamber) enhances the interlayer contact zone which leads to higher interlayer bonding performance [26,107]. In addition, opposing findings were reported for conventional machining processes (materials, glass fibre composites, *etc.*) according to which temperature is dependent on the adhesion between the adherend and the adhesive [108,109].

In 2021, a study revealed the importance of having strong interfacial adhesion between matrix and fibre bonding which was fabricated by introducing the creation of flake-like structures in order to increase the contact surface area [110]. A study on CF/PLA showed that the square nozzle shape is one of the bonding mechanism which creates better contact surface in comparison to circular nozzle by reducing inter voids by 12% [111]. This is due to the ability of the square nozzle to build flat-like structure for the adjacent bed, resulting in a higher contact surface as illustrated in Figure 6 [112]. These will undoubtedly lessen the pull-out mechanism, while also avoiding coarse surfaces that may result in higher fracture energy, particularly when printing nanoscale-size

composites. This phenomenon clearly highlights that the increase in interfacial properties tend to increase fibre ability to transfer load from the polymeric matrix [113]. However, according to Lee *et al.* [114], too many fibre loading will disturb the fibre wetting, hence disturbing the load-transfer mechanism. Therefore, controlling sufficient amount of fibre to boost the load transfer while maintaining interfacial adhesion to the upmost level is crucial. Although the interfacial adhesion will improvise the polymeric composites' performance, studies suggested on applying multifunctional fibres purposely to interconnecting nanoparticles or even using ultrasound transducer to improve the interfacial adhesion between layup sequences [71]. These include the need of adjusting processing temperature during three-dimensional (3D) printing of polymeric composites in order to compromise good adhesion within the printing layer [24,115]. Hence, it is crucial to ensure that the temperature reaches right above the glass transition temperature ( $T_g$ ) to ensure good adhesion and heat transfer. In fact, researchers are now moving towards predicting the interfacial adhesion of printed tracks and layers by studying the printed layer width as shown in Figure 3(e–g) [1,116]. These are due to the fact that there is a strong relation between the bonding formation and air gap which will be discussed further in this article. Having a good interfacial adhesion between layers are crucial due to the fact that it reflected the mechanical performance of the polymeric composites. Studies on unidirectional carbon/epoxy composites highlighted that laminated structures frequently encounter loss of the fibre support in the polymer matrix during the delamination process [46,101]. Hence, this phenomenon can be directly related to the inability of load transfer even under a low-velocity impact, which can be compensated by using the tuft technique, particularly for composite materials prepared using conventional manufacturing processes [101]. However, interlocking theory showed the importance of radial orientation in materials, while depositing single fibre on top of another does not increase the mechanical interlock. Hence, orientation favours a stronger interlocking behaviour among fibres [117,118]. Theoretically, the mechanical performance of the polymer composite materials is significantly dependent on the geometry, size, aspect ratio, and orientation of the filler; further improves the interlocking either within the filler or between the filler and the matrix [118,119]. These phenomena have been discussed in the context of materials prepared using conventional processes. However, when adapting to 3D printing technology, the theoretical results are highlighted and adjusted to fit the fabrication process use. A study on meso-structured composite materials showed that orientation and skewedness

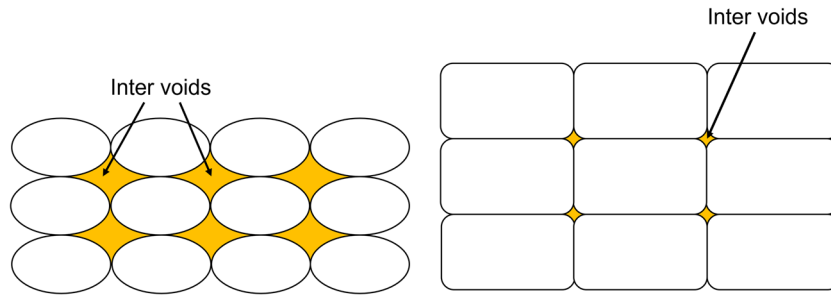


**Figure 5:** Illustrations of CF/PA6 of (a) single-lap shear, (b) FDM printed with infill support, (c) single-lap strength at different build-plate temperature, (d) optical images of damage surface at varying build-plate temperatures of 30–105°C. Reproduced with permission [105]. (e) Mechanical properties of ASA and NCF adhered with epoxy and CA at normal and heat treated conditions. Reproduced with permission [106].

modify the anisotropy behaviour of the overall structure [120,121]. Hence, focusing on the stacking sequence or the layup sequence of the printed materials, as study suggested, a 50% improvement in Young's modulus occurred

when compared to the fibre-reinforced polymer prepared with undefined printing sequence [120,122]. In 2018, using a fibre-reinforced polymer, a maximum Young's modulus of 6.4 GPa was achieved [122]. In comparison, similar materials





**Figure 6:** Illustrations of inter-voids size for both circular and square nozzle.

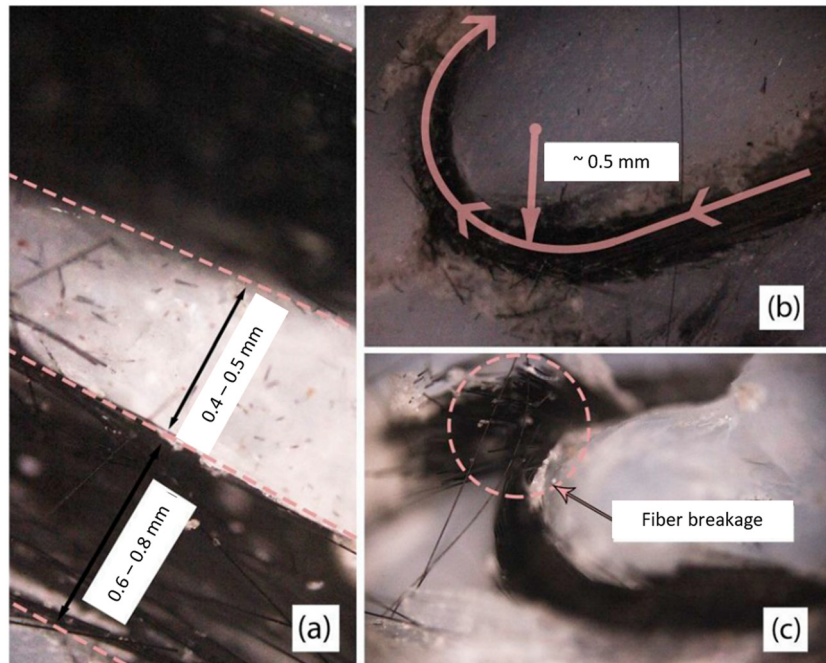
printed with a layup sequence realised a maximum Young's modulus of 12.2 GPa [120]. Thus, considering the orientation in terms of the layup and stacking sequences will enable adjusting and altering the stiffness of the materials using current 3D printing technology.

## 5 Region of bonding formation and effects of structural alignment and air gap

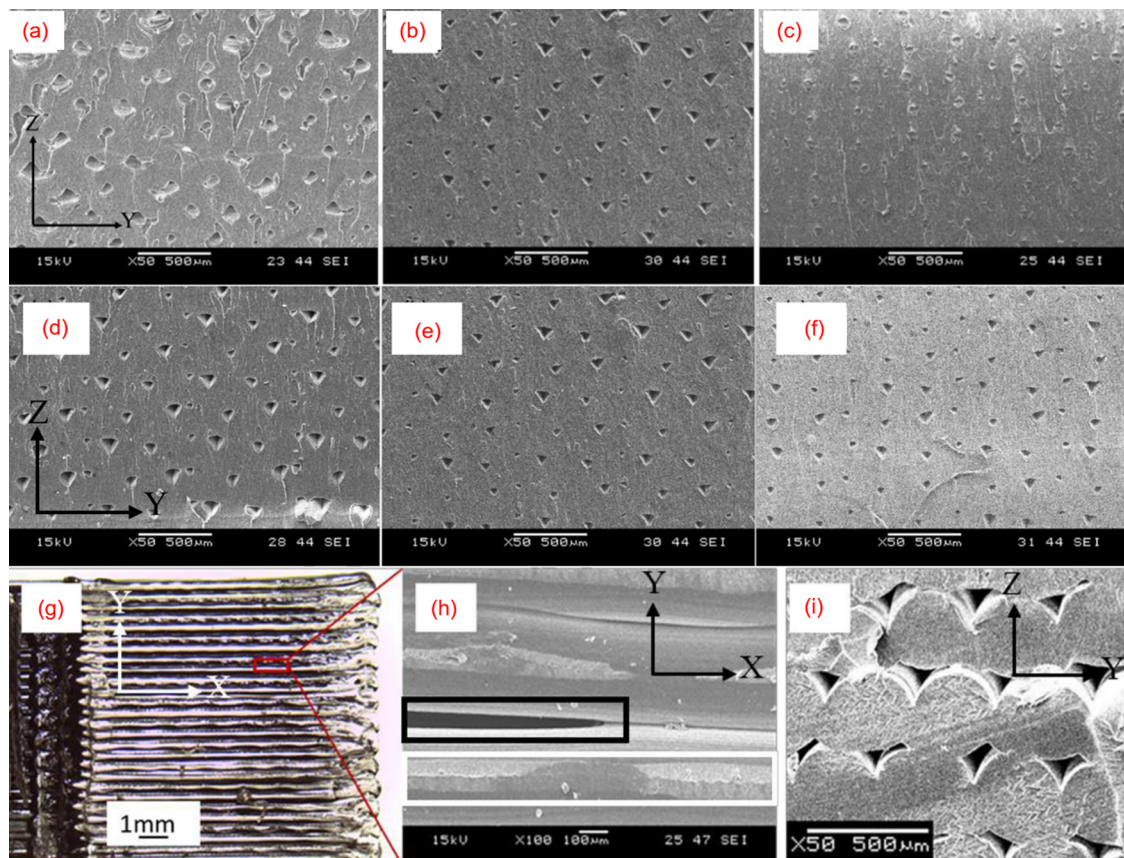
The layup sequence is frequently related to the underlying interlayer bonding formation of the polymer composites, and various studies have been performed to explore the bonding region that strengthens the structure mechanism [63,123]. In 2020, the bonding formation of multi-materials produced by laser powder bed fusion was investigated to ensure a strong bonding interface by adapting the *in situ* technique at a nano-hardness of 7.1 GPa [63]. However, for other techniques applied to polymer composite materials, material extrusion is preferable for controlling the filler arrangement and filaments made by controlling the extrudate swell to minimise the weak layer-to-layer bonding formation [53,123,124]. Filament arrangement is expected to aid in controlling the friction as the wear rate is manageable in the range of 25–34% [123]. This phenomenon was explained based on the debonding of the materials; particularly the bonding in the transverse and parallel directions weakened as the fibres became easily detached. Hence, the study recommended printing a polymer composite material in a direction that only exposes its tip [123,125]. Therefore, using lamination or a layup sequence will minimise the filament fibres from tipping off or detaching from the surface and prevent the deterioration of the mechanical performance [126].

Thus, the infill of a polymeric composite structure frequently creates an air gap and weakens its strength.

Studies using ABS materials in 2017 demonstrated that when the printing process was performed under vacuum condition, an average stress of 19.7 N/mm<sup>2</sup> under 21 inHg of vacuum pressure was attained [127]. This result suggested that performing the printing process under vacuum condition can realise excellent bonding because rapid cooling and heating processes are minimised [127]; moreover, it can reduce the stress concentration [128] because there is adequate time to enhance the bonding within the layers [129]. The adjacent air gap is crucial as it affects the overall performance of the composite materials. In a study in 2019 using CF-reinforced PLA, for two parallel layers prepared using the FDM technique, an air gap between 0.4 and 0.5 mm (Figure 7) was measured [130]. The air gap was attributed to the main limitation, the return radius, in FDM. During the FDM process, if the volume fraction is less than the void volume fraction, the polymer composite material will experience fibre breakage, which is the most common limitation of the FDM technique [64,131]. A study in 2018 using fibre-reinforced thermoplastic composites presented similar findings regarding the existence of voids or intra-traces, including fibre breakage, which lowered the mechanical properties of the composites [131]. Therefore, adapting a secondary fibre size that can fill the gap will maximise the composite performance; this approach has been extensively applied in conventional processes [132,133]. Notably, the air gap induced during the printing process can be adjusted based on the printing temperature. Figure 8(a–f) demonstrates the air gap or evidence of void disappearing as the temperature rises from 220 to 240°C and hence resulting in larger contact areas which ease the layer-by-layer bonding and overall performance [88,134]. This phenomenon is similarly reported when adjusting the bed temperature as the void decreases and improves the contact areas as the bed temperature rises [88,134,135]. Studies reveal that the contact areas often related to contact pressure control the bond quality as shown in Figure 8(g–i) [88]. In the figure, it is clearly indicated that as the layer height



**Figure 7:** (a) Interlayer bonding of continuous CF-reinforced PLA, (b) fibre return path, and (c) fibre breakage during U-turn. Reproduced with permission [130].



**Figure 8:** Micrograph images of cross sectional 3D print at different nozzle temperatures of (a) 220°C, (b) 230°C, and (c) 240°C, different bed temperatures of (d) 85°C, (e) 95°C, and (f) 105°C. An optical image of (g) fracture surface and a micrograph image of (h) layer by layer dissociation, while (i) shows the cross section of the layers. Reproduced with permission [88].

decreases, the contact area increases and the layer width of the materials is maximised [136,137].

## 6 Mechanical performance of printed composite materials

The layup sequence is a promising technique for improving the mechanical performance as it modifies the orientation behaviour of the composite materials. A study in 2020 using carbon black-reinforced ABS polymer composites reported that an orientation of  $45^\circ$  is frequently the best choice to reduce fibre debonding failure [138]. In comparison with  $0^\circ$  and  $90^\circ$ , adjacent linear crack propagation is the major limitation in FDM. Studies on aligned magnetised-CF indicated that the crack propagation mainly due to the lack of interfacial adhesion as the fibre acted as rupture arrestors forced tearing in longer paths with greater fracture surface areas [139,140]. This phenomenon involved not only in additive manufactured polymeric composites *via* FDM yet applied in others fabrication processes. Studies on polymeric composites for biomaterial applications clearly indicated that the mechanical properties are correlated with the printing process and the filler loading [141,142]. The flexural strength was reported to be 50% less than that obtained using conventional injection moulding because the major limitation of 3D printing is the minimum amount of filler loading (between 10 and 40 wt%) [141]. This phenomenon was clearly explained by the percolation theory, according to which the maximum amount of filler reaches the percolation threshold, and hence, stagnates the material performance [119,143]. Therefore, this clearly suggests that 3D printing achieves a higher porosity as well as a lower filler content than the conventional compression moulding process. In addition, the orientation of the filler content contributed to the homogeneity, leading to an improvement 15 times larger than the unreinforced materials [144,145]. Details of different polymeric composites and their printing parameters are provided in Table 1. Analysis on the mechanical performance based on the orientation prospects are investigated extensively, yet minimum reported experimental based on having fibre orientated opposed the loading directions as in Figure 3(c and d). Li *et al.* [146] demonstrated that printing at  $0^\circ$  increases the flexural strength of PLA-CF by 13.8% when compared to pure PLA. Fibre-matrix interface are weak outside the  $0^\circ$ , according to Li *et al.* [146], resulting in fibre pull-out. According to a study on fatigue analysis published in 2020, the fibre layer

oriented at  $0^\circ$  has a better fatigue response with 150 more load cycles  $N$  in the 95%  $S_{ut}$  [147]. According to Pertuz *et al.* [147], CF tends to be longer (Figure 9) due to loading conditions (95% of  $S_{ut}$ ), demonstrating the fibre ability to withstand load at  $0^\circ$  orientation. Shanmugam *et al.* [148] suggested that these phenomena were influenced significantly by the reinforcement distribution and compression response. These phenomena are due to the fact that as the load is applied in the opposite direction (such as in transverse direction), the material will experience delamination caused by inhomogeneity in the stress field [149]. In fact, considering the stress distribution inside the fibre depending on the load applied, the fibre surface regions are increased, creating greater stress concentrations that lead to fibre breakage and worst crack opening which deteriorate its mechanical performance [147,150,151].

It should be noted that some studies often used commercialised filament as listed in Table 1 while some were customised using several techniques, mainly material extrusion [38,42]. Mei *et al.* [38] recommended implementing hot pressing process ( $200^\circ\text{C}$  of pressing temperature) after 3D printing nylon-CF composites as it boosted the tensile strength up to  $\sim 95$  MPa. This technique is important as studies have shown the printed commercialised PLA-CF composites recorded an average tensile strength of 50 MPa [130,162]. Meanwhile, the extruded (customised 20 wt% CF) PLA-CF filament is able to reach a maximum of 75 MPa [164]. Sang *et al.* [164] clearly indicated that the increment in the tensile properties is explained by the long fibres and minimum fibre breakage during the filament preparation. Thus, to maintain the maximum fibre length, Liu *et al.* [39] prepared the continuous fibre by pre-impregnating CF on top of polyamide and recorded the maximum flexural strength of 550 MPa. These clearly demonstrate that controlling the composition and filament preparation result in overall enhancement of the mechanical properties as the interfacial performance improves. Studies reveal interface enhancement by sizing treatment as the CF/filler are fully immersed in a resin impregnation to ease the adhesion upon printing [35]. Hu and Qin [165] have detailed the use of sizing and coating in order to enhance the interphase within the printed polymer composite. It is important to note that the balance between fibre length and interlayer bonding is crucial [166]. Shofner *et al.* [167] recorded a maximum tensile strength of 37.4 MPa (10 wt% of CF-ABS), which is below the average performance due to weak intralayer and interlayer bondings as polymer chain mobility increases. These clearly indicate that the strategies for interfacial mechanisms during filament feedstock will aid in adhesion and interfacial bonding within the fibre-polymer [141,168].

**Table 1:** Mechanical performance of polymeric composites prepared using additive manufacturing technique

Materials	Filament specification (model/type)	Parameter	Output	Ref.
Aluminium/PLA–polyester			Force: 25–30 kN	[152]
Thermoplastic polyurethane/PLA		Temperature: 230°C Bed: 55°C Speed: 20 mm/s	UTS: 35–38 MPa (parallel) 17–19 MPa (perpendicular) Young's modulus: 700–900 MPa (parallel) 550–590 MPa (perpendicular)	[51]
Poly- $\epsilon$ -caprolactone		Nozzle temperature: 160°C Bed temperature: 40°C Print speed: 5 mm/s Nozzle diameter: 0.5 mm Nozzle diameter: 0.4 mm	Tensile strength: 79.7 MPa Young's modulus: 3.5 GPa	[153]
Nylon/fibres (carbon, glass, Kevlar)			Tensile strength: 524 MPa	[16]
Nylon polymer		Infill: solid (100%)	Stiffness: 73 GPa Tensile strength: 32 MPa Stiffness: 0.84 GPa	
PEEK	PEEK OPTIMA™ LT1			[115]
Ultem 1010 (polyetherimide)		Layer height: 0.2 mm Extrusion width: 1 mm Extrusion temperature: 360°C Bed temperature: 160°C	Tensile: 85 MPa	[116]
PLA/CF/Jute		Filament diameter: 2.85 mm Extruder temperature: 140°C	Tensile: 47.9–51.7 MPa	[76]
Commercial PLA material				[85]
ABS		Nozzle temperature: 230°C Bed temperature: 95°C Layer height: 0.2 mm Layer width: 0.35 mm	Fracture resistance: 3,500–4,000 J/m <sup>2</sup>	[88]
ABS/chopped CF (20 wt%)		Nozzle diameter: 5 mm Print speed: 3.8 cm/s Layer time: 93 s Width layer: 6.1 mm	Fracture energy: 5–12 kJ/m <sup>2</sup>	[96]
Carbon black/ABS		Orientations of 0°/90°, 45°/–45°, and 0°	Stress: 30 MPa Shear strain: 25 MPa	[138]
ABS-glass fibre	ABS-GF10	—	—	[154]
PLA	—	—	—	[155]
CF/ABS	Polylac PA-747	Extrusion nozzle: 0.35 mm Extrusion temperature: 235°C Bed temperature: 95°C	Fracture toughness: 0.5 kJ/m <sup>2</sup> Fracture toughness: 3.1 kJ/m <sup>2</sup>	[79]
High-power microwave CF/ABS		Nozzle diameter: 0.04 mm Layer thickness 0.2–0.4 mm Amplitude: 10 $\mu$ m	Surface roughness: 13.7–14.41 $\mu$ m	[156]
ABS		Layer thickness: 0.25 mm Vacuum pressure: 21, 24, 27, 30 inHg	Stress: 12–19.7 N/mm <sup>2</sup> Strain: 4.7–5.55%	[127]
ABS		Nozzle temperature: 215, 225, and 235°C Printing speed: 20, 40, and 60 mm/s	Young modulus: 100–150 MPa Yield strength: 0.2–0.3 ksi	[157]
ABS glue		Nozzle: 0.4 mm Layer thickness: 0.2 mm Print speed: 40 mm/s Nozzle temperature: 220°C Bed temperature: 90°C	UTS: 4.7–5.5 ksi	[158]

(Continued)



Table 1: Continued

Materials	Filament specification (model/type)	Parameter	Output	Ref.
Polymeric gyroid lattice			Compressive strength: 1.1–2.99 MPa	[159]
Thermoplastic polyimide (TPI)		Filament diameter: 1.75 mm Interlayer bond: diameter (12 mm) and height (30 mm) Nozzle velocity: 50 mm/s Extrusion rate: 40 mm/min Pressure: 2.5–4.5 MPa Temperature: 335 and 355°C	Interlayer bonding: 50–350 N	[107]
ABS			UTS: 12–22 MPa	[92]
PLA		Nozzle: 0.4 mm  Temperature: 210°C Bed temperature: 60°C	Strength: 40 MPa (Z-direction) 70 MPa (F-direction) Toughness: 0–1 J/mm <sup>3</sup> (Z) 2–3 J/mm <sup>3</sup> (F)	[87]
ABS	ABS-m30			[92]
ABS, PEEK	—			[24]
Short CF-reinforced polyamide	Carbon XT <sup>™</sup> Nylon Gen 2, 3DXTECH	Nozzle diameter: 0.4 mm  Extrusion bed: 0.5 mm Layer height: 0.2 mm Extrusion temperature: 260°C Velocity: 10 mm/s	Young's modulus: 2.4–2.6 GPa Tensile strength: 35–41 MPa	[26]
CF-reinforced polymer polypropylene (AR: 100)			Stress: 7.5 wt% CF-45 MPa Toughness 12 J/m <sup>3</sup> × 10 <sup>3</sup> Yield strength: 25 MPa Ductility 3%	[160]
Formulate conductive material-carbon black	For BFB3000 3D printer			[161]
PLA (pre-impregnated)	Markforged, Inc., USA			[162]
PLA	PLA 02-B-0015			[163]

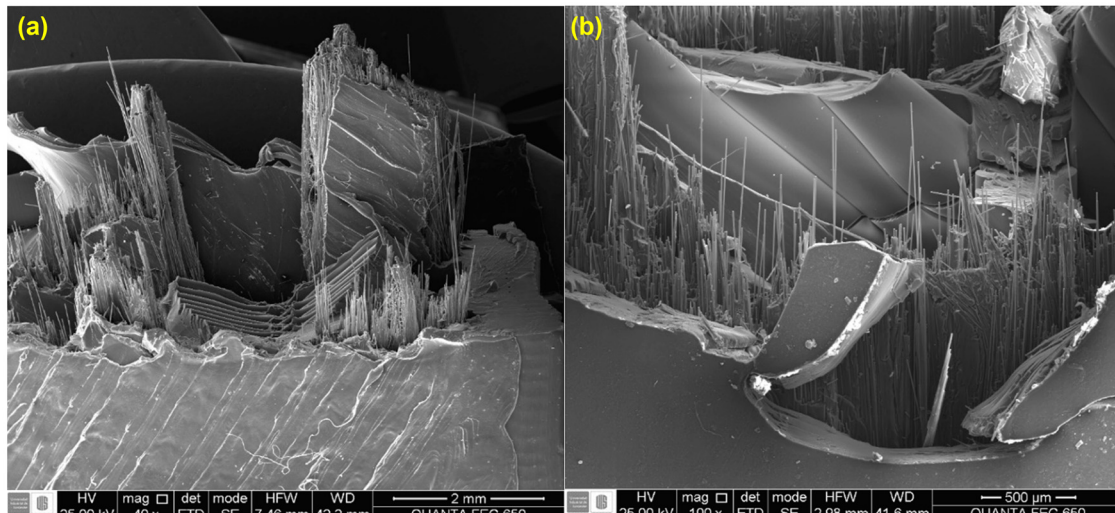


Figure 9: Micrograph images of nylon-carbon fibre at 0° orientation and (a) 80% of  $S_{ut}$  and (b) 95% of  $S_{ut}$ . Reproduced with permission [147].

## 7 Four-dimensional (4D) printing

Based on the importance of oriented and aligned structures by printing in additive manufacturing, 4D printing must be considered to ensure best desired output. Until now, studies have focused on 4D printing because of its ability to control the magnitude of and dynamically vary each input [169–171]. However, 4D printing is frequently associated with swelling dynamics, as elucidated in composites 4D printed using a morphing nozzle. This phenomenon, in turn, promotes a minimum proportion of oriented fibres as the filaments shift from anisotropic to isotropic swelling properties ( $p < 0.001$ ) [169,172]. These fundamental modifications occur in 4D printing as the swelling effect alters the filler arrangement without reorientation as well as minimises the effect of misalignment [173,174]. Hence, this allows better control and manipulation of the printing shapes and customisation of the functionalities of the end product [53,174]. Although 4D printing is currently a future direction, the manipulation of the fibres in the printing process has received attention in terms of its void formation, poor adhesion, method and parameter application, blockage issues, and material selection [53,169–171,174]. Clearly, detailed findings and progressive research are required to fully fill the gap related to 4D printing.

## 8 Conclusion and future prospects

The literature presented provides a correlation between polymeric composite materials fabricated by additive manufacturing and their mechanical performance. In the past five years, most of the research on polymeric composites, specifically using FDM techniques, reported various optimum parameters. Although such studies are promising, different tools frequently using different parameters for operation is a major limitation in the current modern world. Therefore, this review describes the details of the approaches for improving the mechanical properties of the polymeric composites. In addition, this study is anticipated to broaden and strengthen the overall mechanical performance as well as become a reference for interlayering of printed polymeric composites. The conclusions are summarised as follows:

- (a) Fabricating polymeric composites is frequently related with filaments or feeders because they maintain and control the mechanical properties. Researchers are continuing to develop composite materials by considering the filament scale geometry. Techniques for manufacturing composite filaments, such as coating,

impregnating, or mechanical mixing (extrusion), must be carefully considered, since different filament fabrication processes will alter the amount of fibre breakage and hence affect the interfacial adhesion of fibre-matrix. Also, when fabricating using the FDM technique, one should consider the load-bearing activity of the printed materials owing to its contribution to the overall performance.

- (b) Additively manufactured polymeric composites are frequently associated with layer-by-layer printing, which might cause delamination or failure owing to the inhomogeneous transverse load transfer. This further causes a minimum interlaminar shear strength in the composite structures. Therefore, one should consider printing the structure in terms of the layup sequence or the laminated orientation, which can improve the bonding and minimise the static stress of the materials. Utilising a printing orientation of  $0^\circ$  allows for better stress distribution, thereby improving tensile strength and fatigue response as the fibres tend to elongate more at  $0^\circ$ .
- (c) Adjusting the structural alignment can minimise the air gap between the printed materials, thereby improving the bonding formation. These phenomena will allow higher fibre loading that are tailored to the structural alignment, similar to the printed orientation allowing low fibre breakage and broadening of the functionalisation of the materials. Therefore, further studies should be conducted to maximise the fibre loading ( $>40$  wt%) because currently industries and researchers are relying on the filaments or feeders as supplied by the manufacturer.
- (d) Based on the findings presented in this article, it is noted that the structural orientation can improve the load transfer of the materials. Thus, detailed analysis should be conducted to elucidate the fracture mechanism and interlaminar behaviour of the polymeric composites when subjected to the layup sequence because until now very few studies have been reported. Further research and analysis on these issues will lead to the future development of FDM, because based on all tabulated data and detailed information, we can adopt a conventional analytical model to predict the mechanical properties of additively manufactured polymeric composites.

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