3D-Printed Fiber-Reinforced Polymer Composites by Fused Deposition Modelling

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Fiber reinforced polymer composites (FRPC) can be manufactured by Fused deposition modeling. An introduction to FRPC and the types of fibers used in producing FRPC are summarised in this article.

Keywords: additive manufacturing ; fused filament fabrication ; 3D printing

1. Introduction

Additive manufacturing (AM) or 3-dimensional printing (3DP) technology is one of the most promising areas in component manufacturing. AM has paved its way into application areas ranging from automotive [1], construction [2], aerospace [3] and consumer products to biomedical products such as prosthetics [4]. AM refers to a group of fabrication techniques where parts are fabricated layer-by-layer directly from a computer-aided design (CAD) file. AM technology is a very broad term that encompasses many methods such as Stereolithography (SLA) of a photopolymer liquid [5], Laminated Object Manufacturing (LOM) from plastic laminations [6], Selective Laser Sintering (SLS) from plastic or metal powder [7] and Fused Deposition Modelling (FDM) from plastic filaments [8]. Since 1980, many studies have been conducted to maximize the potential applications of these technologies, as AM is well-known and still a far more cost-effective alternative to subtractive manufacturing technologies such as milling, drilling and turning [9]. FDM, also called Fused Filament Fabrication (FFF), is one of the most popular techniques due to its relatively low cost, low material wastage and ease of use. Nowadays, most people can even purchase and use this technique at home. However, FDM 3D print is yet to replace conventional manufacturing in producing functional parts. FDM 3D print parts are weaker than conventionally manufactured counterparts due to their layer-by-layer fabrication method. Research has been carried out to improve the mechanical properties of FDM printed parts by using various methods, such as optimizing printing parameters, annealing, snap-fitting [10], printing in an oxygen free environment [11], mechanical pressing [12] and fiber reinforced thermoplastics. Of all the methods, fiber reinforced polymer composites (FRPC) are known to have high stiffness, strength, damage tolerance, fatigue resistance and corrosion resistance. FRPCs are produced by adding fibers or particles into the thermoplastic matrix to improve the mechanical strength of the printed components [13]. This method reduces voids and increases interlaminar bonding between the deposited filaments. There are two types of fiber reinforcement: continuous and discontinuous, depending on fiber length. Fiber reinforced composites have a long history and are traditionally produced by techniques like hand lay-up, molding, etc. FDM is a relatively new technique for manufacturing fiber reinforced polymer composites.

2. Fiber Reinforced Polymer Composite (FRPC)

The development of composites that are compatible with FDM printers has attracted a lot of attention. This is because composites promise better mechanical properties and performances compared to neat polymers. Many results in the development of new printable composites reinforced with particles, fibers or nanomaterials have already been demonstrated. Carbon black, platelets, chopped fibers and polymer fibrils are mixed with the polymer matrix and then extruded together during printing. However, the performance of these composites depends largely on the fiber orientation in the plastic and the fiber-volume-fraction (FVF). Parts manufactured with FDM from neat polymer have shown insufficient strength in load tests. This limits the range of applications in which FDM technology can be used for functional parts and not for prototypes.

Researchers have used fiber reinforced polymer composites (FRPCs) to overcome the aforementioned limitations in their work. In FRPCs, the material properties of a component are enhanced by combining reinforcing fibers and polymer matrix. Various fibers have been used for reinforcement, including chopped carbon fibers, carbon nanotubes, glass fibers, natural fibers etc. [14][15][16][17]. There are certain requirements that FRPC materials must meet in order to be processed by AM, namely:

- Types of reinforcement and matrices;
- Good fiber-to-matrix bonding;
- Fiber homogeneity;
- Fiber alignment;
- Good interlayer bonding;
- Minimal porosity.

The fiber reinforcement must be matched in size, shape and length to the part’s intended use. Both the matrix material, which holds the fibers in place, and the reinforcement must be compatible with the selected AM technique. A good bond between fibers and matrix is required at the fiber-matrix interface to transfer loads efficiently from the matrix, resulting in composites that follow the “rule of mixtures”. Fiber loading is also crucial to obtain AM composites with good mechanical properties. Mechanical properties such as elastic modulus increase with fiber loading at a low loading ratio but degrade after reaching an optimum value [18]. This phenomenon generally occurs due to poor wettability of the fiber with the thermoplastic, which results in a poor fiber-matrix interface.

Higher loading leads to an increase in viscosity and a decrease in flowability, leading to processability problems such as clogging of the nozzle. Furthermore, fiber reinforcement may cause negative effects on interlaminar bonding and the properties of printed parts. Based on previous research, interlamellar matrix regions between the reinforced fiber layers are critical regions that are highly prone to delamination when subjected to mechanical stress. Delamination can result from weak fiber-matrix bonding, which often leads to internal damage in composites, potentially leading to global failure of the component with reduced strength and stiffness [19]. Furthermore, porosity and weak interface bonding between fibers and matrix have been cited as a major problem for 3D printed fiber reinforced polymer composites [20]. Understanding the mechanism of filament bonding is important to further investigate how FRPC works to reduce voids and increase the strength of the interlaminar bond between the deposited filaments.

2.1. Synthetic Fibers vs. Natural Fibers

Various fibers were used as reinforcement for polymer composites and can be grouped under two categories: synthetic fibers and natural fibers [13]. Natural fibers were first used as reinforcement for polymers since 1936 [21] and were slowly replaced by synthetic fibers because synthetic fibers are usually much stronger than natural fibers. However, natural fibers re-emerged as reinforcing materials for polymer composites when environmental issues became more important in engineering applications. In FDM 3D printed polymer composites, both synthetic and natural fibers were used to reinforce polymers, although synthetic fibers are a more popular choice.

Synthetic fibers are commonly used as reinforcement for FDM printed composites, and the popular fibers are carbon fiber [22][23][24][25][26][27][28][29][30][31][32], glass fiber [24][33][34] and Kevlar fibers [24]. Other possible synthetic fibers are Graphene, CNTs [35], powder [36], copper powder [36] etc. Generally, synthetic fibers are added to polymer matrix during FDM 3D printing to enhance the mechanical properties of polymer composites, and plenty of works were reported previously [24][25][29][31][37][38]. In addition, synthetic fibers were also used to improve or alter thermal properties/thermal conductivities of FDM 3D printed polymer composites [39][40] and electrical properties [41]. A systematic review on synthetic fibers as reinforcement for polymer matrix was presented recently [42], although their review does not focus on FDM 3D printed polymer composites specifically.

Natural fibers are used as reinforcement to reduce the inorganic content in thermoplastic composites without compromising mechanical strength, ultimately improving biodegradability and reducing costs [43]. Common natural fibers used in FDM 3D printed polymer composites are jute [30], wood [44], harakeke/flax [45][46], bamboo [46], sugarcane and many more. Recent and systematic reviews on natural fiber reinforced polymer composites as feeders in FDM-Based 3D Printing were reported by researchers [43][47][48]. Natural fibers are a cheaper and greener alternative to reinforce polymer matrix during the FDM 3D printing process, but challenges such as fiber agglomeration, clogging in the nozzle, poor fiber-matrix interface, non -homogenous mixing etc., have to be investigated further. Furthermore, various treatments such as chemical and thermal are required to be applied to natural fibers to enhance the performance of natural fibers reinforced polymers. In addition, a different combination of polymer matrix and natural fibers requires different treatments and processes. As such, more research works are required to improve the performance of natural fibers reinforced polymers. Environmentally friendly engineering materials are getting more attention recently. Therefore, polymer composites produced by bio-based polymers such as PLA [44], soy-based resin [49][50] etc., and reinforced with natural fibers have great potential because they are biodegradable and environmentally friendly.

The advantages and limitations of synthetic and natural fibers as reinforcing materials for FDM printed polymer are summarized in **Table 1**.

Table 1. The advantages and limitations for synthetic fibers and natural fibers as reinforced material for FDM printed polymer [43][51][52].

Type of Fibers	Advantages	Limitation
Synthetic fibers	Higher strength Higher stiffness Corrosion resistance Flame retardancy Chemical resistance Commercially available	Higher cost Not ‘green’
Natural fibers	Biocompatible, Biodegradable, renewable Recyclable Relatively cheap	Lower strength compared to synthetic fibers Requires treatment of fibers (in general) Fibers are discontinuous (in general) Not all fibers are commercially available

2.2. Continuous vs. Discontinuous Fiber

Fiber reinforced polymer composite is a subcategory of fiber reinforced composites. Generally, fiber reinforcement can be categorized into discontinuous and continuous fibers according to critical fiber length [53]. Critical length l_c is the fiber length that allows applied load transfer to the reinforced fibers by the matrix, and depends on fiber’s ultimate strength σ_f, fiber diameter d, and fiber-matrix bond strength or shear yield strength of the matrix τ_c. Continuous fibers are referred to fiber with length more than 15 l_c, and discontinuous fibers are fibers with length less than 15 l_c [53]. Nevertheless, some other researchers have slightly different definition. Krajangsawasdi et al. further classified short and discontinuous fiber, where short fibers are fibers shorter than critical length l_c, and discontinuous fiber are those with length above critical length l_c [54]. Pruß and Vietor defined discontinuous fibers as fibers with fiber length less than 1 mm (0.04 in.), while continuous fibers are fibers with a length above 50 mm (2 in.) [55].

Besides the obvious motivation of improving mechanical properties, reinforcement can also be used to provide the material with additional functions such as electro-conductivity, thermal conductivity or biocompatibility. Kalsoom et al. [56] and Wang et al. [57] have provided a general overview of 3D printable composites; this paper instead focuses in more detail on the engineering aspects of FDM as a composite manufacturing method.

Conventionally, fiber-reinforced composites can be classified into: (a) continuous and aligned fiber composites, (b) discontinuous and aligned-fiber composites, and (c) discontinuous and randomly oriented-fiber composites, depending on the length and alignment of the fibers [53]. The major advantages and disadvantages are listed in **Table 2**.

Table 2. Brief comparison of fiber reinforced composites, according to length and orientation of fiber [53][58].

Continuous and Aligned Fiber Composites	Discontinuous and Aligned-Fiber Composites	Discontinuous and Randomly Oriented-Fiber Composites
Properties of the composite are highly anisotropic	Properties of the composite are highly anisotropic	Composites are isotropic
Most effective strengthening but only along the designed direction; weaker along other directions	Less effective in strengthening than continuous and aligned fiber composites and only along the designed direction	Least effective in strengthening mechanical but all directions are strengthened
Limited manufacturing methods, hard to be manufactured, the highest cost	Difficult to maintain good alignment of discontinuous fiber during manufacturing; higher cost than discontinuous and randomly oriented-fiber composites	Easier to be manufactured, lowest cost

2.2.1. Continuous and Aligned Fiber Composites

The continuous and aligned fibers can reinforce composites in the intended direction but have no significant effect in the transverse direction. Conventional methods for producing continuous and aligned fiber composites are pultrusion, prepreg, and filament winding [53]. In terms of additive manufacturing, the FDM 3D printed ‘continuous and aligned fiber composites’ are being investigated by various researchers [23][24][25][29][31][38][41][59]. Previously, 3D printed continuous and aligned fiber composites were mostly printed using in-house developed or modified 3D printers [23][29]. The first commercial 3D printer capable of printing continuous and aligned fiber composites was developed by MarkForged. With the availability of commercial machines such as Markforged’s Markone, Marktwo 3D printers, research on FDM printing of continuous fiber reinforced thermoplastics (CFRT) composites is booming. Most of the recent research on FDM 3D printed continuous and aligned fiber composites uses Markforged’s Markone, Marktwo 3D printers [24][25][31][38][41][59]. Various types of continuous fibers, such as carbon fibers [23][24][25][29][31], glass fibers [24][38], and Kevlar fibers [24], have been used as reinforcement. In general, the FDM printed continuous and aligned fibers can have better electrical properties [41] and mechanical properties, such as tensile strength [24][25][29][31][38], flexural strength [29] if the printing parameters are properly selected. A systematic review of 3D printed continuous fiber polymer composites is presented by [60]. However, 3D printed continuous and aligned fiber composites are limited in terms of design freedom, as fiber placement is challenging and more voids are created, especially when printing complex shapes [51][31]. Design freedom is one of the main advantages of additive manufacturing over conventional manufacturing, and incorporating continuous fibers into FDM 3D printing, negates this advantage.

2.2.2. Discontinuous and Randomly Oriented-Fiber Composites

Discontinuous fiber composites have a long history, and the first scientific publication dates back to 1936 [21]. Due to the nature of reinforced fibers and conventional fabrication methods, such as hand lay-up, resin transfer molding, etc., early fiber reinforced composites are mainly discontinuous and randomly oriented. FDM 3D printed discontinuous fiber composites are manufactured using composite filaments by commercial FDM 3D printers. Generally, discontinuous fibers were premixed with the polymer matrix as composite filament, and the composite filaments were then used in FDM 3D printing to produce discontinuous fiber composites. To date, more than 10,000 published papers have been found in Scopus using the keywords “additive manufacture” and “short fiber reinforced polymers”, and it is not possible to discuss them all here. However, most of these research papers focused on the mechanical or thermal properties of the composites. They did not report on the orientation of the fibers in FDM 3D printed discontinuous fiber composites. Nevertheless, research with FDM 3D printed discontinuous and randomly oriented-fiber composites have been reported by several researchers [34], although not all of them emphasized the orientation of the fibers.

One of the recent works with FDM 3D printed discontinuous and randomly oriented-fiber composites was reported by Zhao et al. [34]. They compared the tensile properties of 3D printed CNT-short glass fiber (SGF) reinforced PLA composite with the tensile properties of 3D printed PLA, SGF/PLA, and found that both composites are better than neat PLA in terms of tensile strength and tensile modulus. In addition, CNT-SGF /PLA composite has a higher tensile strength than SGF/PLA composite. From the SEM images of the fracture surfaces of the composite specimens, they found that the fibers in the composites are randomly oriented. Su et al. reinforced polyamide with reclaimed carbon fiber in four different weight percentages (10%, 20%, 30%, 40%). They found that the fibers were better aligned at low fiber contents (10–20%) and had no significant alignment at 40%. They concluded that the tensile performance of the reclaimed carbon fiber reinforced polyamide composites (rCF/PA) largely depended on the fiber content and orientation, with higher fiber content and aligned fiber being able to improve tensile strength. All composites, including rCF/PA with 40 wt% and non-aligned fiber performed better than neat PLA [61].

2.2.3. Discontinuous and Aligned-Fiber Composites

Discontinuous and aligned fibers are an alternative to continuous fibers in 3D printing of polymer composites, with the advantage of better design freedom. Early research on discontinuous and aligned-fiber composites (also named as aligned discontinuous fiber thermoplastic) produced by non-additive manufacturing processes was summarized by Such et al. [62]. Although the manufacturing methods for 3D printed FDM 3D printed discontinuous and aligned-fiber composites are different from the conventional make discontinuous and aligned-fiber composites, the motivations for reinforcing polymers with discontinuous and aligned-fiber are similar. In general discontinuous and aligned-fiber are added to polymers for three main reasons: (1) to improve mechanical, thermal, or electrical properties in the desired direction, (2) to reduce the cost and complexity of manufacturing compared to composites with continuous fibers, and (3) enabling design freedom or complex geometries [63][21][62][64]. FDM 3D printed discontinuous and aligned-fiber composites are mainly manufactured using composite filaments by commercial FDM 3D printers. The discontinuous fibers were aligned by shear (referred to as shear-induced alignment or flow-induced alignment), where the shear force between a nozzle and the molten material forces the fibers to align in the direction of extrusion or flow [63][65]. Furthermore, the orientation of fibers is affected by experimental extrusion width, where experimental extrusion width depends on extrusion temperature, speed and width. Fibers were more aligned in a narrow extruder than in a wider extruder [66]. One of the first published papers on FDM 3D printed discontinuous, and aligned-fiber composites was by Tekinalp et al. [18]. They fabricated the carbon fiber reinforced ABS filament and used the filament with a commercial FDM 3D printer. They applied the method of Bay and Tucker [67] to characterize the fiber orientation in the printed part and found that the carbon fibers in the printed parts are mainly oriented in the load-bearing direction. They concluded that the carbon fibers could increase the strength and modulus of both the FDM printed and compression molded samples, but the FDM samples have significant voids [18].

Jia et al. fabricated graphite flakes reinforced PA6/POE-g-MAH/PS composite with an FDM 3D printer and verified by microscopy that the graphite flakes were aligned along the through-plane direction (parallel to the x-y plane) via microscopy. With this designed composite, they were able to improve the thermal conductivity of the polymer [68]. However, they also pointed out that the presence of voids in FDM- printed composites affects the through-plane thermal conductivity of the composites. Papon and Haque investigated fracture toughness of 3D printed carbon fiber reinforced PLA composites with different fiber content (3 wt.%, 5 wt.%, 7 wt.% and 10 wt.%), manufactured by two different nozzle shapes (circular and square) [69]. The square shape nozzle was custom-made to improve the contact area and inter-bead void. The fibers are mostly aligned in the extrusion direction, but they did not report how nozzle shape affects the fiber orientation. Their experimental results show that the fracture toughness increased with fiber content from 0% to 5%, at both layer orientation of 45°/−45° and 0°/90°. The print layer orientation of 45°/−45° and 0°/90° has no major different in fracture properties. Furthermore, parts printed by a square nozzle have better fracture toughness than parts printed by a circular nozzle because less void is produced in parts produced by the square nozzle.

Researchers at the University of Bristol developed a method named High Performance Discontinues Fiber (HiPerDIF) to manufacture discontinuous and aligned-fiber composites [70] and investigated the performance of composites produced with this method [64]. Generally, the fibers were suspended in a liquid medium (water), and the orientation of fiber was controlled by the orientation head [70]. With this method, they fabricated discontinuous and aligned-fiber epoxy composites using carbon fiber [70] and recycled carbon fibers [71]. They reported that the mechanical properties of composites are proportional to the fiber lengths [71]. To expand the HiPerDIF technology to additive manufacturing/FDM, Blok et al. have identified 4 different polymers (ABS, PLA, Nylon, PETG) as the potential polymer matrix materials to be reinforced with high performance discontinues and formed the feedstock materials for FDM. The four polymers were selected based on 14 factors. They fabricated the composite tapes using an in-house consolidation method, where the HiPerDiF fiber was sandwiched between two layers of polymer matrix films of 0.125 mm.

They proofed that aligned discontinuous fiber composites produced using HiPerDIF technology are better than currently available short fiber thermoplastic. Furthermore, the composite fabricated with HiPErDIF technology has comparable mechanical behavior compared with continuous fiber composite but with better manufacturing flexibility [32]. Krajangsawasdi et al. recently extended their work by fabricating 3D printer filament using ADFRC fiber to reinforce PLA thermoplastic. They managed to produce HiPerDiF-PLA filament and also identified the optimal printing parameters of their newly developed filament. They compared the mechanical properties of the HiPerDiF-PLA printed parts with PLA, PLA-short carbon fiber, PLA-continuous carbon fiber, and Markforged continuous carbon fiber [64], and they concluded that HiPerDiF-PLA outperformed other PLA composites in terms of mechanical performance.

3. Conclusion

Fused deposition modelling method can be a turning point for manufacturing industrial and additive manufacturing technology. The potential of building functional parts, directly from commercial 3D printer with controllable properties, created a huge rush for new development and research in this field. Various types and forms of fiber can be used in manufacturing fiber reinforced polymer composites via FDM.

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