

## A REVIEW ON THE ADDITIVE MANUFACTURING OF FIBER REINFORCED POLYMER MATRIX COMPOSITES

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### Abstract

Additive manufacturing (AM), also referred to as 3D printing, has gained popularity due to the recent developments and market trends especially in the last decades. The main advantages of AM are its capability of producing parts with high geometrical complexity at almost no added cost, short lead times, weight reduction, less efforts for assembly and suitability for customization as well as for low volume production or even single parts. Moreover, some applications may need materials with unusual combinations of properties, which cannot be provided only by metals, polymers or ceramics. For such applications, composite materials combining two or more materials allow having the preferred properties combined in a single material. Thus, AM, which can be defined as a process of adding materials to produce objects directly from its CAD model in successive layers in contrast to subtractive processes, is gaining significance for critical applications using composite materials. This paper thus presents a detailed review of AM of polymers reinforced with chopped / continuous fibers and the influence of this reinforcement on the mechanical performance of composite parts, mainly focussing on the Fused Deposition Modelling (FDM) process. On one hand, the reviewed studies on the FDM of composites mainly point out that the mechanical performance is significantly enhanced in contrast to polymers with no reinforcement. Yet, it is also evident that the mechanical performance of FDM composites is highly dependent on the build direction and porosity. Thus, there is still a wide range of gaps to be studied for replacing metallic components by AM composites.

**Keywords:** Additive Manufacturing, Polymer Matrix Composites, Layered Manufacturing, Carbon Fiber Reinforced Polymers, Rapid Manufacturing

### Introduction

Due to its capability of producing parts with a high geometrical complexity and short manufacturing lead times, AM is finding more utilization area, especially in aerospace, defence and automotive applications. Revenues from the production of end use parts, as a proportion of total AM production, has risen from under 4% in 2003 to almost 61% in 2016 [1]. The first step of applying AM technology was historically producing plastic prototypes using various AM processes such as FDM (Fused Deposition Modelling), SLA (Stereo lithography) and other processes. By exploring several advantages of AM such as light weight design and suitability for quickly producing first articles for design validation, etc., producing parts from metals, ceramics and composites as functional parts later became available [2]. While polymer and metal materials are considered as commercially available, though in a possible range of materials, ceramics and composites are rather new and still under research [3]. An example of a complex duct produced by Selective Laser Sintering of PA12-CF (polyamide 12- carbon fiber) material is demonstrated in Figure 1 [4].

Although the general perception of AM technologies is easy-to-capture, the complicated dependencies of AM processes on several related technologies such as material modelling, design tools, computing, and process design represent a real challenge for both applied and basic research, as shown in Figure 2 [5]. This is even for complicated for composite materials in AM. It can be considered that the investigation of AM technology effects on composite materials is still in the area of research and development, but the increase of academic and industrial studies on AM of composites can clearly be distinguished in the recent years [6].



Figure 1: An example of PA12-CF produced by Selective Laser Sintering [4]

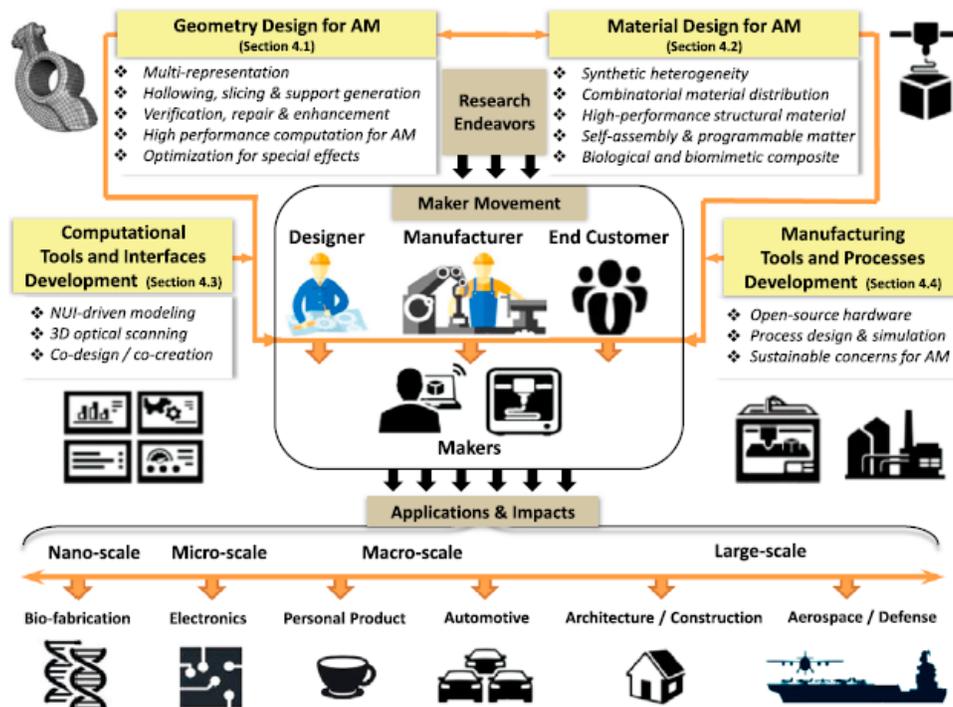


Figure 2: A geometry material machine process roadmap for AM [5]

### Additive Manufacturing of Composites

Fiber reinforcement of thermoplastic matrix has a huge potential to improve the mechanical properties. However, fiber orientation and void formation in these composites

become the main issues. In this section, various AM methods capable of producing chopped or continuous reinforced fibers in a polymer matrix are reviewed in detail to demonstrate the strengths and weaknesses.

Fused Deposition Modelling (FDM) (see Figure 3) is one of the AM technologies and is a widely used method for fabricating thermoplastic parts with advantages of low cost, minimal waste and ease of material change [6, 8]. The process was pioneered by Stratasys introducing the first FDM system in 1991 [1]. The FDM process is also referred to as material extrusion or fused filament fabrication (FFF) where the input material is in the form of filament to enable the extrusion. When the thermoplastic matrix is reinforced by different fibers, improved mechanical properties can be obtained and the reinforced plastics are called CFRP (carbon fiber reinforced plastics). They can even be used as functional end parts fulfilling high requirements. Moreover, the use of multiple materials is possible in FDM having the possibility to use multiple nozzles with loading of different materials thus allowing functionally graded materials. An important reason why FDM is favourable for composite manufacturing is its being low-cost, simple and having high speed. On the other hand, one of the disadvantages of CFRPs is that the matrix material must be a thermoplastic material in order to provide the necessary melt viscosity being high enough for structural rigidity and low enough for extrusion [9].

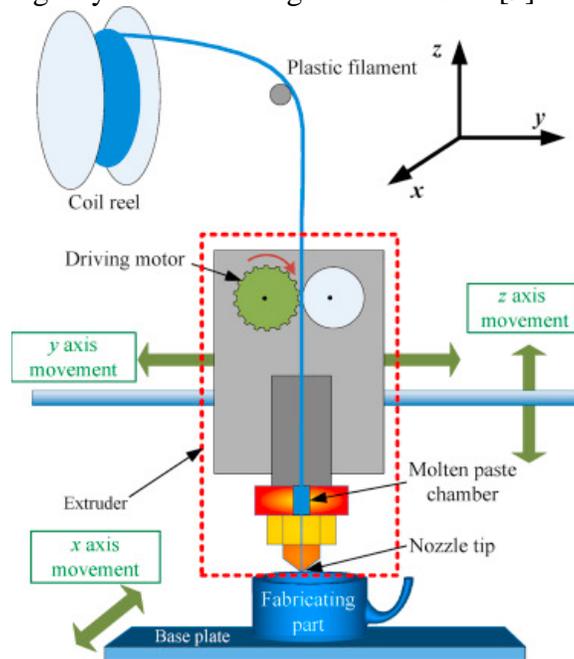


Figure 3: Schematic demonstration of the FDM process [8]

For the FDM process, there exist several process parameters including bead width, air gap, model build temperature, and raster orientation influencing the obtained material properties [10]. In order to improve the strength and accuracy of FDM parts, various build rules have been recommended such as orienting the parts to ensure the tensile loads are carried axially along printing directions; or to use small beads for a better surface quality at a cost of increased printing time; or to be aware that tensile loads tend to failure easier than compressive loads [10]. In order to enhance the mechanical properties for functional part manufacturing in FDM, fiber reinforcement has gained popularity amongst researchers. A list of various studies of FDM with chopped fiber reinforced thermoplastics is given in Table 1.

Table 1. A summary of studies in FDM of chopped fibers

	Reinforced by	Matrix Material	Investigated Properties	Limitations
[7]	Carbon fiber	ABS	Tensile strength, Young Modulus, Flexural properties	Decrease in toughness, yield strength and ductility; Increase of porosity with increased level of carbon fiber
[11]	Glass fiber	ABS	Tensile strength; surface rigidity	Flexibility and handlability
[12]	Thermotropic liquid crystalline polymer	Polypropylene	Tensile strength	Poor adhesion and delamination
[13]	Vapor-grown carbon fibers	ABS	Tensile strength; tensile modulus	Interlayer and intra-layer fusion Change behaviour from ductile to brittle
[14]	Carbon fiber	ABS	Tensile strength; tensile modulus	Porosity, weak interfacial adhesion between the fibers and the matrix and fiber breakage
[15]	Carbon fiber	ABS	Strength, stiffness, thermal prop., distortion, geometric tolerances	--
[16]	Glass fiber	Polypropylene	Tensile properties	Existence of voids leading to 20-30% loss in mechanical performance
[17]	Carbon fiber	Epoxy	Tensile properties	Increased carbon loading levels needed and length of carbon fiber
[18]	Carbon fiber	ABS	Tensile properties	Voids at elevated temperatures
[19]	Carbon fiber	Ultem 1000	Tensile and flexural properties	25% porosity, due to the volume expansion of trapped moisture, air or other gases

As seen from the table, many studies focus on reinforcement with carbon fibers although the maximum loading level is quite limited. Ning et al. has provided a comprehensive study on the effect of fiber content on mechanical properties. The carbon fiber content varying between 0 wt. % and 15 wt. % was studied on tensile and flexural properties of carbon fiber reinforced ABS plastics (see Figure 4). Some limitations such as decrease in toughness and ductility as well as encountered porosity were identified [7]. With carbon reinforcement, Tekinalp et al. [14] has studied ABS as the matrix material to see the feasibility of using CFRPs for load-bearing components. They have concluded that composites with highly dispersed and highly oriented carbon fibers can be printed by FDM process (see Figure 5). However, significant porosity was observed in FDM-printed samples. With increased finer loading, voids inside the beads increased whereas voids between beads are reduced [14]. They also reported that the next steps necessary for FDM to reach the full potential shall be to minimize the pore formation and fiber breakage during compounding as well as to increase the interfacial adhesion between fibers and the matrix. Love et al. has addressed reinforcement of ABS material also with carbon fibers studying the aspects of the thermal deformations and geometrical tolerances as well as strength and stiffness achieved. The results show that carbon fiber additions can significantly reduce the distortion and warping of the material during processing allowing large-scale, out-of-the-oven, high deposition rate manufacturing [12].

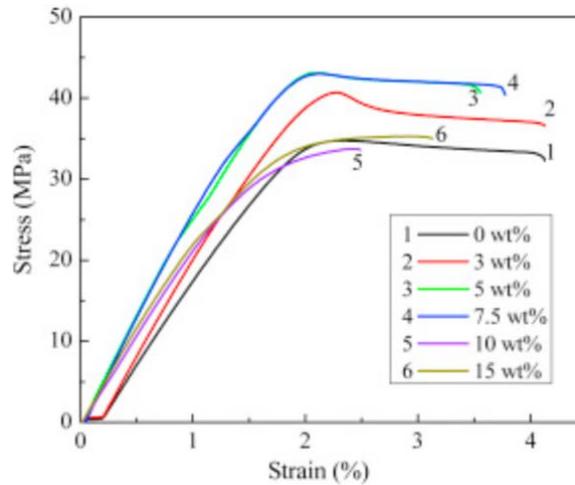


Figure 4: The effect of carbon fiber loading on the tensile stress-strain curves [7]

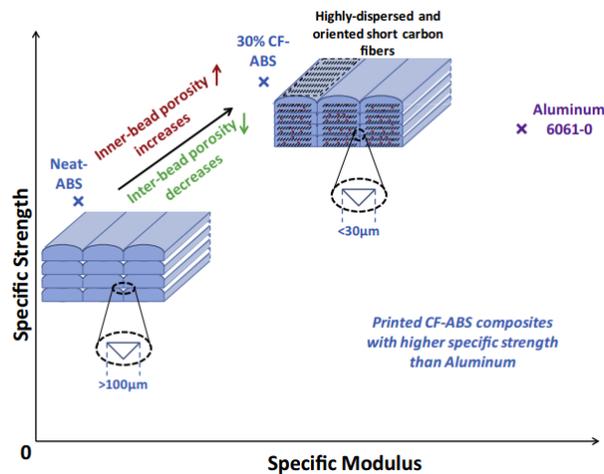


Figure 5: Schematic presentation of 3D-printed fiber-reinforced composite by fused deposition modelling [14]

Mahajan and Cornier have studied reinforcing epoxy matrix with carbon fiber reinforcement by a design of experiments approach to identify significant process parameters affecting preferential fiber alignment in the micro-extrusion process [17]. The carbon loading, translation speed, and nozzle diameter were found to have the most significant effect on the degree of fiber alignment. However, as future work, increase of carbon loading and fiber length is recommended as well as to induce fiber orientation not only in XY but also along z axis direction [17]. Shofner et al. studied reinforcing ABS matrix with vapor-grown carbon fibers at nano-scale. Although the tensile properties were improved, the improvement depended on build parameters as well as the degree of interlayer and intra-layer fusion [13]. Reinforcing ULTEM material with carbon fibers was studied by Chuang et al for aircraft engine components (see Figure 6) and they have concluded that FDM-printed Ultem 9085 exhibited about 84% of its original strength and 64% of its original modulus as compared to its injection-molded counter parts. When 10% chopped fiber was reinforced into Ultem 1000, the tensile strength increased by 23% and modulus by 38%, but also this reduced the ductility. One of the limitations was that the FDM extruded thin filaments and FDM printed Ultem 1000 composite vanes exhibited ~25% porosity, due to the volume expansion of trapped moisture, air or other gases generated from degradation

at elevated printing temperature of 420 °C by FDM [19]. As many other studies point it out, the build parameters are very important in FDM of chopped fiber reinforced polymers leading to porosity formation which limits the mechanical performance significantly through inter- and intra-layer fusion.

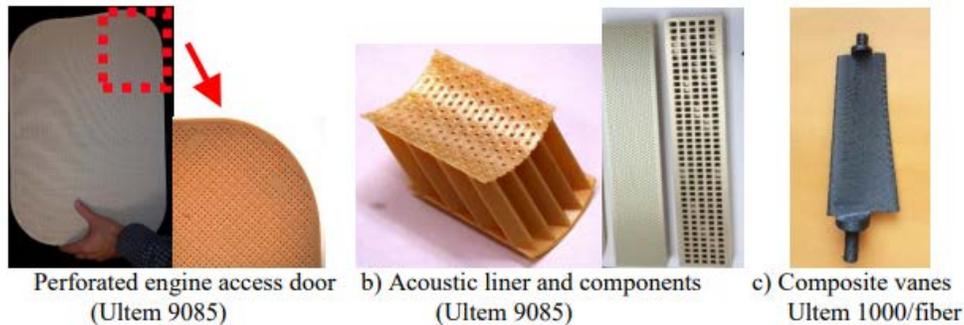


Figure 6: FDM printed polymer parts [19]

Other fiber reinforcements are also studied. For example, Zhong et al. have studied the processability of reinforcement of ABS matrix with glass fibers at three different fiber loading. The results showed that the reinforcement could improve the tensile strength and surface rigidity at the expense of flexibility and handleability [11]. These limits were overcome by adding a small amount of plasticizer and compatibilizer. Gray et al. reinforced polypropylene with thermotropic liquid crystalline polymer fibers and provided a significantly increased tensile strength whereas they encountered some problems of poor adhesion and delamination [12]. Polypropylene (PP) was also studied by Carneiro et al. by reinforcing it with glass fibers. They showed that 30% and 40% improvement for the modulus and strength, respectively, compared to pure PP [16].

Other AM technologies, such as Selective Laser Sintering (SLS) are also used for processing polymer composites. Jansson and Pejryd have used SLS for processing carbon-fiber reinforced polyamide. The commercial name for this material from EOS is CarbonMide® (CF/PA12) [20]. The material in its raw form is a powder consisting of polyamide spherical particles and carbon fibers of diameter 10 µm and length 100-200 µm. However, porosity as well as the dependency of mechanical properties on the build axis mainly due to fiber orientation and porosity is a significant problem. The study given in [20] also reports that porosity was concentrated in between the layers produced weakening the material in the direction normal to the layered structure. The fiber orientation is found to be linked to the powder rake mechanisms. As shown in Figure 7, the continuous mechanical contact during powder deposition between the rake and the fibres will work to align most of the fibres in the x-direction of the build chamber, and only the fibres that were originally oriented in the build plane can maintain their orientation. Thus, the fiber orientation will strongly tend towards the x-direction after being spread [20]. There are other studies focussing on surface modification of carbon fibers for the SLS process [21]. Jing et al. studied the surface modification and reinforcing effect in carbon fiber (CF)/nylon 12 (PA12) composites. With this HNO<sub>3</sub> treatment, the impurities on the surface of the carbon fiber were removed while the roughness was increased. Moreover, oxygen functional groups were introduced, and the concentrations of the O element on the surface were increased leading to an enhanced physical interlocking and interfacial adhesion between the fibers and the PA matrix. The surface modification, thus, resulted in a 15% increase in the tensile strength.

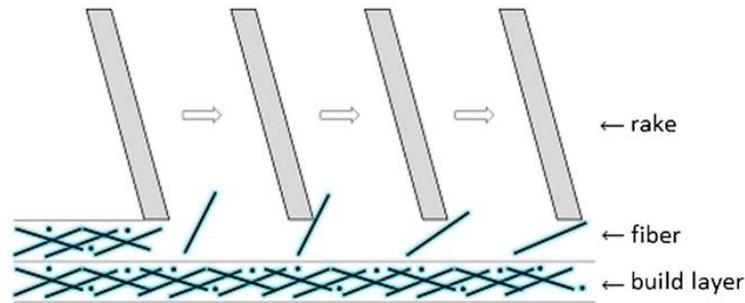


Figure 7: Rake spreading a powder layer in the SLS process [20]

More recently, studies on introducing continuous fibers in the polymer matrix are carried-out mainly using Fused Deposition Modeling (FDM) for different applications [22-28]. A summary of some studies focussed on AM of continuous fiber reinforced polymers is given in Table 2. Yao et al. have investigated embedding carbon fiber tows leading to an increase of the tensile strength by 70% and a flexural strength increase by 18.7% compared to non-reinforced specimens [22]. Dickson et al. has studied reinforcing different types of fiber (glass, carbon and Kevlar® fibers) into nylon material by using a Mark One 3D printer (see Figure 7). In tensile and flexural strengths, up to 6.3 and 5 –fold enhancement was achieved respectively. The highest enhancement was achieved with the carbon fiber [23]. Gardner et al. has investigated reinforcing ULTEM® material with carbon nano yarn filaments leading to better tensile and electrical conductivity properties [24].

Table 2. A summary of studies in FDM of continuous fibers

	Reinforced by	Matrix Material	Investigated Properties	Limitations
[22]	Carbon fiber	Epoxy resin + Polyamide	Flexural and Tensile properties Weight reduction	Adhesion between fibers and matrix Carbon fiber placement
[23]	Carbon, glass and Kevlar® fiber	Nylon	Tensile and Flexural properties	Weak bonding and porosity
[24]	Carbon nanotube yarn	Ultem®	Tensile strength and specific modulus, electrical conductivity	Cutting mechanism
[25]	Continuous glass fiber (CGF)	Polypropylene	Tensile and Flexural properties	Adhesion
[26]	Carbon fiber	PLA (Poly Lactic Acid)	Flexural strength and modulus	None reported
[27-28]	Carbon fiber	PLA (Poly Lactic Acid)	Tensile modulus and strength	Irregularity and discontinuity of fiber
[29]	Carbon, glass and Kevlar® fiber	Nylon	Shear strength	Poor wettability of Kevlar® fibre bundles by the nylon leading to extensive delamination.
[30]	Carbon fiber	Epoxy resin (E-54(616))	Tensile and bending properties	Fiber pull out as the major failure mechanism
[31]	Kevlar R fibre	Polylactide	Compression properties	Increase of fiber loading needed

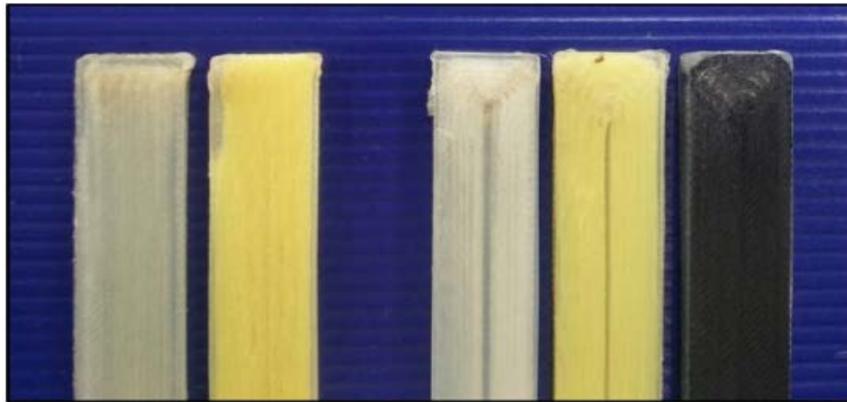


Figure 8: Examples of fibre reinforcement patterns used in the study [23]

Other AM techniques are also used for processing CFRPs. For example, Parandoush et al. proposed a novel method for AM of fiber composites by using a prepreg composite. A CO<sub>2</sub> laser is used to heat successive layers of pre-preg tapes including PP matrix and a compaction roller is utilized to bond these layers. The mechanical performance of this method was evaluated by conducting T-peel, lap shear, tensile and bending tests [25].

Moreover, Tian et al. have investigated the influence of main printing parameters such as temperature, layer thickness, hatch spacing, speed and filament feed rate. With the optimized parameters, a carbon fiber content of about 27% in the PLA could achieve the maximum flexural strength of 335 MPa and a flexural modulus of 30 GPa leading to future potential applications for the light structures in the field of aviation and aerospace [26]. Matsuzaki et al. [27, 28] report a very significant improvement of mechanical performance by reinforcing continuous carbon fibers. The tensile modulus and strength of FDM-printed PLA composites are reported to be  $19.5 \pm 2.08$  GPa and  $185.2 \pm 24.6$  MPa, respectively. These results indicate enhancements of almost 6-fold and 4.5 –fold for the tensile modulus and strength of the pure PLA specimens which is very pronounced compared to that of short fiber reinforced PLA composites [27]. Caminero et al. have studied reinforcing nylon with a variety of fibers and they have concluded that the poor wettability of Kevlar fibre bundles by the nylon led to extensive delamination [29]. Another significant failure mechanism is found to be fiber pull out in [30] where FDM of an epoxy resin reinforced with continuous carbon fiber is studied. For complex shapes, cross lap and panel-core lap design strategies were proposed by Hou et al. [31]. In their study, the influences of process parameters, structure parameters, density, fibre content, on the final performance of the printed specimens were discussed.

### **Summary & Conclusions**

Additive manufacturing of polymer matrix composites has become a very interesting topic for researchers due to provided advantages such as weight reduction, part consolidation, design optimization and ability to produce very complex integrated geometries especially for aerospace and automotive applications. Most FDM studies of carbon reinforced polymer matrix composites which are conducted by the FDM process, conclude that the mechanical performance is significantly enhanced, especially in terms of tensile strength and flexural properties while the mechanical performance is severely dependent on the build direction and porosity. Moreover, it is also limited by the planar layer-by-layer nature of the AM processes. Thus, some extra

enhancements in terms of material, performance and process are needed to further exploring of the full potential. Synthesis of different materials, reinforcement material range, fiber loading are the main barriers to be overcome on the material side whereas elimination of voids and enhancement of interfacial bonding and repeatability shall be considered on the performance aspect. The maximum part size, process resilience and productivity shall be improved in years of process.

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