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3D PRINTED COMPOSITES – BENCHMARKING THE STATE-OF-THE-ART

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ABSTRACT

Fused filament fabrication (FFF) is a 3D printing technique which allows layer-by-layer build-up of a part by the deposition of thermoplastic material through a nozzle. The technique allows for complex shapes to be made with a degree of design freedom unachievable with traditional manufacturing methods. However, the mechanical properties of the thermoplastic materials used are low compared to common engineering materials. In this work, improved 3D printing feedstocks for FFF, with carbon fibres embedded in a thermoplastic matrix to reinforce the material, are investigated. The state-of-the-art in composite 3D printing is reviewed and the capabilities of two different commercially available composite printing methods are assessed by print trials, optical microscopy and mechanical characterization of the printed materials. It is found that printing of continuous carbon fibres using the MarkOne gives significant increases in performance over unreinforced thermoplastics, with mechanical properties in the same order of magnitude of typical unidirectional epoxy matrix composites. The method, however, is limited in design freedom as the brittle continuous carbon fibres cannot be deposited freely through small steering radii and sharp angles. Filaments with embedded carbon microfibres (~100 μm) show better print capabilities and are suitable with standard printing methods, but only offer a slight increase in mechanical properties over the pure thermoplastic properties.

1 INTRODUCTION

Carbon fibres reinforced plastics (CFRPs) provide excellent mechanical properties and allow for significant design tailorability. A fundamental CFRPs manufacturing challenge, however, is the combination of the reinforcement fibres into the polymer matrix with good consolidation, maximum fibre orientation control and low cost [1]. While a wide range of manufacturing methods for composites are available, most FRP parts are formed in a two-step process, i.e. material lay-up followed by consolidation. For good consolidation, pressure needs to be applied over the entire part surface area which requires expensive equipment and increases manufacturing costs. In this work, additive manufacture (AM) techniques and more specifically fused filament fabrication (FFF), is investigated as an alternative CFRP manufacturing approach where low investment costs and rapid prototyping ability are important.

Layer-by-layer (LbL) manufacturing techniques, such as FFF, commonly known as 3D printing, have an underappreciated similarity to traditional composite materials in that they are made from stacking a series of discrete layers. It is therefore reasonable to suggest that successful adaptation of 3D printing technologies to composite materials could enable a simple method for composite manufacture with lower production cost and a high degree of automation. While still a relatively undeveloped avenue of research, there is at least one company developing commercial 3D printers capable of printing continuous fibre reinforced composite materials: MarkForged [2]. The Mark One and Mark Two printers developed by MarkForged print continuous carbon fibre reinforced nylon with high mechanical properties, comparable to aluminium, to open up new applications in both the personal fabrication market and in the manufacture of lightweight parts for industry. As reinforcements can be accurately placed, the laminated structure of composite parts can be further optimised in each layer, allowing for an increase in design freedom and mechanical performance.

However, significant challenges remain for 3D printing of CFRPs. In addition to some process specific limitations with the MarkForged printers, which will be discussed in further detail below, there are more fundamental issues which need addressing. For example, there is limited choice of materials suitable for 3D printing and to obtain a high strength composite part the void content needs to be minimal. During printing, however, a filament bead is deposited beside or on top of another filament and the interface quality is determined by the polymer sintering process that happens without the application of external pressure or extra heat. Fibres may be first embedded in a thermoplastic matrix in a pre-processing stage but the final part strength greatly depends on the final interface between the different printed beads.

A brief overview of the state-of-art in composite 3D printing is given here and two different methods of composite 3D printing, i.e. continuous fibre printing and discontinuous fibre printing, are experimentally characterised. Comparisons were made between the two printing methods in terms of mechanical properties and print quality. The highest mechanical properties were obtained with the continuous fibre printing method but the printing process was less versatile and had a lower quality than discontinuous fibre printing as continuous fibres could not completely fill small features and tight radii.

2 STATE-OF-THE-ART

The FFF process can be utilized to print CFRPs by the addition of fibres into the thermoplastic filament. Besides the obvious motivation of increasing mechanical properties, the reinforcement may also be used to add extra functionality to the material such as electroconductivity, higher heat conductivity or improved mechanical properties. Kalsoom et al. [3] and Wang et al. [4] recently provided a general overview of 3D printable composite materials, here a specific focus on the FFF process is given. The use of fibre reinforcements in 3D printing filaments for FFF is a topic of on-going research with both advancements in scientific literature as well as in commercial products, e.g. the MarkForged printers and the numerous reinforced thermoplastic filaments available on the market [2], [5]–[12].

Most studies report on the use of very short carbon fibres (100 μm) which are mixed with a thermoplastic resin and then extruded to create the filament used for traditional printing. This increases the strength and stiffness of the printed material by around 65%, but remains low compared to typical unidirectional continuous CFRPs.

Ning et al. [5] produced reinforced filament using screw extrusion with varying fibre content from 0 to 7.5 wt% and a fibre length of 100 μm. They found an increase in tensile and flexural properties (tensile strength from 34 to 42 MPa and tensile stiffness from 2 GPa to 2.5 GPa). One limitation they noted was that during the screw extrusion process, the brittle fibres break which limits their length in the filament and consequently they do not reach their full strength. Tekinalp et al. [6] used a similar process to add up to 40wt% of carbon fibres with an average length of 0.26mm to ABS and investigated the print quality and mechanical properties. The strength increased from 35 to 65MPa and the stiffness from 2GPa to 14 GPa. They characterized the voids and found that the size of inter-bead voids decreased with the addition of fibres but secondary voids occurred around the fibre/matrix interface.

Recently, Lewicki et al. [12] reported on the direct ink writing method of carbon fibre reinforced epoxy, which used syringe extrusion with a fast curing epoxy to build up the composite part layer-by-layer. They used 600 μm fibres and found that silicate nanoparticles had to be added to increase the viscosity of the resin to prevent the resin flowing past the fibres. The strength increased with the addition of fibres from 85 MPa to 172 MPa and the stiffness increased from 2 GPa to 5.5 GPa. Numerical analysis was done to analyse the fibre alignment in the nozzle – an approach which may provide an effective tool in future work to minimise clogging within the fibre nozzle.

MarkForged has developed a printer which deposits continuous fibres (carbon, glass and Kevlar) in a Nylon matrix. A ~0.4mm diameter continuous fibre/Nylon filament is fed through a nozzle and, after initial contact with and adhesion to the printing bed (thereby anchoring the filament), can be dragged along a custom path. To finish printing the continuous fibre, the reinforced filament is cut with a shear cutter. The reported strength and stiffness of printed parts with carbon fibres are 700 MPa and 50 GPa respectively [2]. Printing with continuous carbon fibres is also done by Anisoprint which report a strength and modulus of 760 MPa and 42 GPa respectively. The matrix type and printing method is unknown as it is not commercialized at the time of writing, but the fibre volume fraction, V_f, was reported to be 18.5% [11]. Matsuzaki *et al.* [7] printed continuous fibres (straight carbon fibres or twisted jute fibre yarns) by feeding them through a nozzle simultaneously with a thermoplastic filament (PLA) which acts as a matrix. They reported a strength and stiffness of 195 MPa and 10.5 GPa respectively which may be attributed to a low V_f of 6.6%. This technique also showed a non-uniform fibre distribution as the fibres were not pre-impregnated in the matrix.

The printing of continuous fibres adds complexity to the printing process. Cutting of the filament is required for placement of fibres along different tracks and in different layers. The shape is also limited by the steering radius of the fibre, as continuous fibres cannot deform plastically along their length, while fibre straightness and alignment must be maintained for optimal structural performance [13]. The addition of continuous fibres to thermoplastic filament, however, shows a larger improvement in mechanical properties compared to the microfiber (~100 μm) reinforced filaments. Discontinuous fibres in a polymer matrix may not reach their full strength as the matrix or fibre-matrix interface fails before the fibres unless their length is above a "critical length" [14-16] [14],[15], [16].

Concluding, two main printing methods can be identified, the printing of discontinuous fibres with traditional printing methods and continuous fibre printing with a custom printing head and technique. To contribute to the body of knowledge on composite printing, the two printing methods are further investigated here in terms of mechanical properties and printing characteristics before drawing conclusions on the different composite printing techniques.

3 EXPERIMENTAL METHODOLOGY

Two composite 3D printing methods are assessed. The Mark Forged MarkOne printer is used to print continuous carbon fibre / Nylon composite parts, characterise the mechanical properties and investigate the printing characteristics as a composite printing benchmark. Secondly, carbon microfibre reinforced PLA filament is used in a traditional FFF printer to compare against the MarkOne in terms of mechanical properties and quality. The methodology for these two investigations is presented below.

3.1 MarkOne continuous fibre printer characterization

The MarkOne printer is a proprietary 3D printer which can deposit a filament made of continuous fibre embedded in a Nylon matrix. The printer has two printing nozzles as shown in Figure 1, one to deposit pure Nylon filament, and one for fibre reinforced Nylon filament. Three fibre reinforced filaments are available with Kevlar, glass or carbon fibres and the two-nozzle system enables printing of Nylon as well as fibre reinforced layers. This unreinforced Nylon nozzle is crucial for the overall integrity and quality of the prints, as the fibre filament can not be used for the outer layers of the parts (top, bottom, sides), and for more complex shapes and thin features there are often large regions in which the fibre filament is not able to fill which instead are filled with the unreinforced Nylon. The Nylon filament and Nylon/fibre filament are pushed through a Teflon bowden tube to the nozzles, as highlighted in Figure 1.

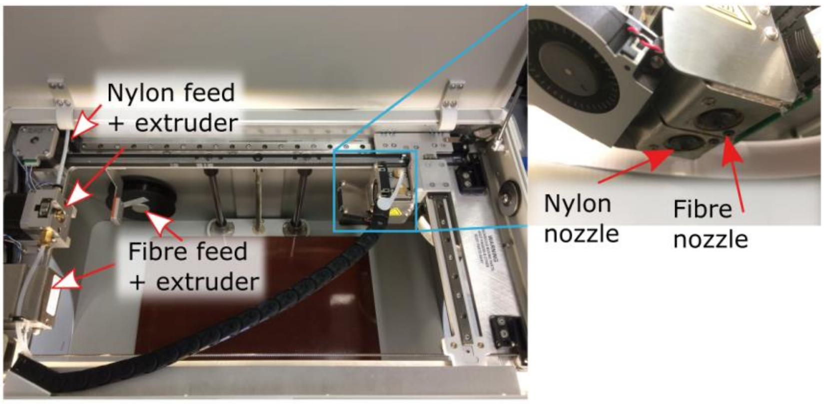


Figure 1: Overview of the MarkOne Printer with the dual nozzle system to print Nylon filament and fibre filament.

To print an object, a proprietary software, which does not allow manual control of temperature, nozzle movement or extrusion speed, must be used. This limits the printing capabilities as the printing settings cannot be fully customized. When printing the Nylon material, it is possible to specify the fill percentage and fill type (rectangular, triangular or hexagonal). For the deposition of carbon fibres, only a circumferential fill pattern is possible which fills the shape from the outside inward in a spiraling motion. This means the fibre is always orientated along the outer shape of the part.

To print, a 3D object is sliced into layers with a layer height of 0.125 mm for carbon fibre reinforced layers. The bottom and top layers are always printed with the 100% triangular fill Nylon filament, as well as the outer periphery for each layer. This is presumably done to avoid exposed fibres on the outer surface and to take advantage of the higher quality surface finish and accuracy available from the unreinforced Nylon. While this feature exists for sensible reasons, it has the negative effect of lowering the overall fibre volume fraction for the part - and thus the maximum achievable mechanical properties. Printing was done at 260 °C with an estimated speed of 6.90 cm^3/hr for the Nylon layers and 2.39 cm^3/hr for the carbon fibre layers.

To provide a reliable and generally useful benchmark of the MarkOne printer, the most extensive suite of mechanical tests and printing trials reported to date were performed. The tensile, shear and flexural response of the printed material have been measured. To get around the limitations in fibre orientation caused by the circumferential fill pattern, the tensile specimens were printed in an "oval racetrack" shape to all for unidirectional 0° specimens to be extracted as shown in Figure 2. The dimensions of the tensile specimens were 250 mm×15 mm×1 mm according to ASTM standards [17], where the bottom and top layers of 0.125 mm thick were 100% triangular fill Nylon as discussed above. The estimated V_f of these specimens was ~27%. Glass fibre tabs with a length of 25 mm were bonded to the specimen using an epoxy adhesive and the tensile test was carried out at constant displacement rate of 2 mm/min. Strain measurements were obtained from a video extensometer (IMETRUM, UK) and the load was obtained from a 25 kN load cell (Instron).

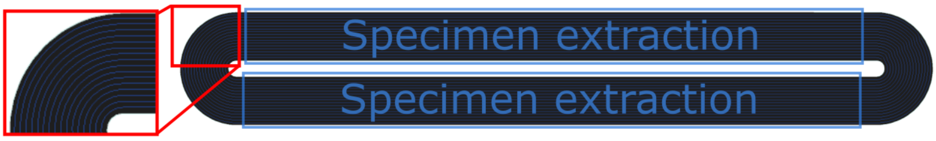


Figure 2: Print schematic of unidirectional tensile and flexural specimens showing fibre path

A three-point bend fixture was used to obtain the flexural properties of the printed composite material with a support rod radius of 4 mm and a support length of 128mm according to the ASTM D7264 standard [18]. The flexural specimens were manufactured in a similar approach as the tensile specimens, with outer dimensions of 160 mm×11 mm×4 mm and all fibres orientated in the 0° orientation. A constant displacement rate of 1 mm/min was used and the force and displacement was directly measured from the machine with a 1 kN load cell.

To obtain the shear properties, the methodology proposed by Sun and Chung [19] was used for uniaxial off-axis testing with oblique end-tabs. This creates a uniform state of stress from which the shear properties can be obtained. The oblique angle is a function of the off-axis angle and the properties of the composite, which were estimated to be E_{11} = 50 GPa, E_{22} = 0.38 GPa, G_{12} = 3.9 GPa and ν_{12} = 0.3.

The samples were cut under a 13° angle for which oblique end tabs with an angle of 21° were required. The samples were tested using an electrical-mechanical tensile machine with a 10 kN load cell and strain measurements were obtained from a 5MP LaVision DIC system.

Lastly, to assess the printing performance of the MarkOne printer and quality of the continuous fibre deposited, benchmark parts were printed in the form of a 40×40 mm square, a 30°-60°-90° triangle (85×50 mm) and a circle with a radius of 20? mm. The quality of the MarkOne printed parts was investigated using these generic shapes and optical microscopy on printed samples.

3.2 Carbon fibre PLA filament characterization

The second composite printing method was done using a standard Prusa Mendel i3 based printer and a PLA filament which was reinforced with chopped carbon fibres. The material was acquired from Proto-Pasta and has 15 wt% carbon fibres added to the 1.75mm diameter filament [20]. In comparison to the Mark One, this open-source printer allows for more control of the material deposition strategy, such as printing tracks, extrusion rate and printing temperature. To characterize this printing technique, tensile and flexural specimens were printed to determine the mechanical properties. Optical microscopy was used to investigate the quality of these specimens.

Tensile specimens were printed in dog-bone shapes according to the ASTM D638 [21], using a 0.4 mm nozzle diameter and a printing temperature of 205°C as recommended by the filament manufacturer. The gauge section of the dog-bone specimens consists of tracks aligned in the 0° direction as shown in Figure 3. The flexural specimens were printed as rectangles with dimensions 168×13×4 mm to match the ASTM D7264 standard for three point bending [18] with a support length of 128 mm.

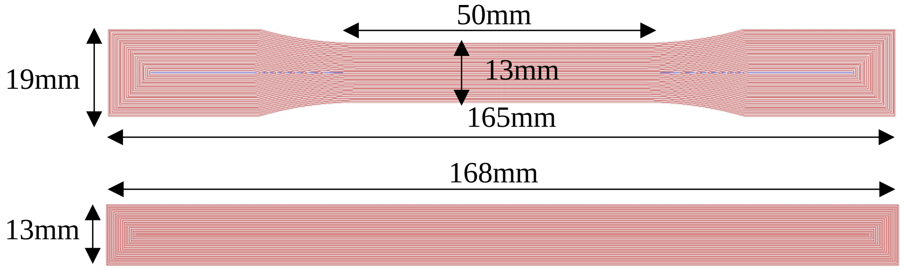


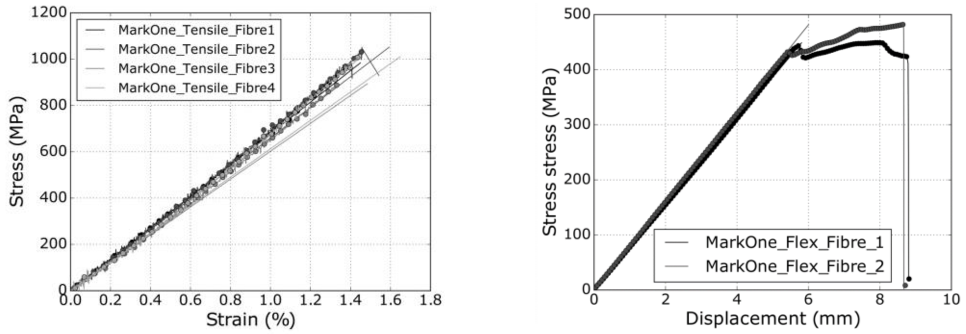
Figure 3: Dog-bone (top) and flexural (bottom) specimen printing fill patterns and specimen dimensions.

4 RESULTS

4.1 MarkOne continuous fibre printer characterization

The results of the tensile tests on the specimens printed by the MarkOne printer are shown in Figure 4a. Four composite samples were tested which show an average strength and stiffness of 986 MPa (±5.8%) and 63.9 GPa (±6.8%) respectively, which are higher than reported by MarkForged [2]. During the test, characteristic high frequency fibre fracture sounds were heard at a low stress of 200 MPa, after which no fracture was heard before failure. In Figure 4a, the linear modulus determined between 0.1% and 0.3% strain is shown as a straight line and a stiffening effect can be seen where the modulus increases at higher strains. This may indicate some early fibre fracture of possibly wrinkled fibres, and some relaxation of most the fibres may have occurred later which leads to better alignment at higher load levels.

The results of the flexural tests are shown in Figure 4b. The flexural modulus and strength of the carbon specimens are 51.2 GPa (±3.7%) and 466 MPa (±5.0%) respectively. The flexural strength is lower than the tensile strength which indicates there may be issues with the quality of the specimen, as a higher flexural strength is expected for high quality fibre composites [22]. A compressive failure was found for these specimens which may indicate a poor fibre/matrix interface and/or a high void content as failure initiators.



(a) **(b)**

Figure 4: Results graphs of (a) tensile and (b) flexural tests of MarkOne continuous fibre printed specimens

The shear response of the off-axis unidirectional specimens is shown in Figure 5. The strains have been calculated using the strains measured by the DIC system (ε_{xx}, ε_{yy}, γ_{xy}) using Equation (1), where $m = \cos(\theta)$, $n = \sin(\theta)$ and θ is the off-axis angle. The shear stress is calculated using Equation (2).

$$\gamma_{12} = 2mn(\varepsilon_{yy} - \varepsilon_{xx}) + (m^2 - n^2)\gamma_{xy} \quad (1)$$

$$\tau_{12} = -mn\sigma_{xx} \tag{2}$$

The modulus has been determined from the initial linear part of the curve, which was found to be 2.3 GPa (±4.9%) and is lower than the expected value of 3.9 GPa used to determine the oblique end-test tab angle. This may influence the results as a non-uniform state of stress occurs which can lead to premature failure. The maximum shear stress was 31 MPa (± 15.8%) where it must be noted that one of the specimens failed near the end-tab, which may explain the lower shear stress.

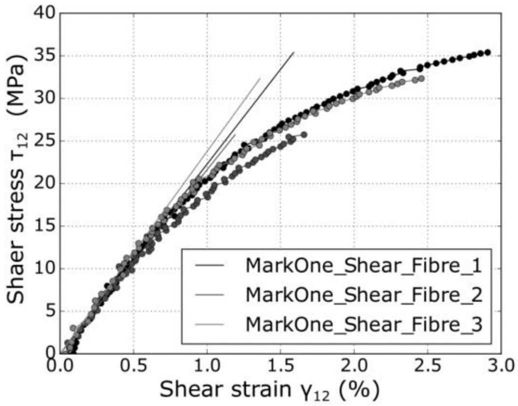


Figure 5: Shear response of shear test samples of MarkOne continuous fibre printed parts in the material (12) coordinate system.

The printing quality of the MarkOne can be seen in Figure 6 with the printing of various generic shapes. The MarkOne printer prints a fibre layer from the outside contour to the inside. For more complex geometries, this causes large fibreless areas as shown in the triangular part. These fibreless areas can be up to 2.5 mm×1 mm, which are recognized by the printing software and are filled with pure Nylon, but this leads to a local weakness in the part. For a simpler shape, such as a square, a similar effect is present in the corner regions on a smaller scale (1.5 mm×0.3 mm) but the areas are not filled with Nylon here which leads to voids. For the circular shape, the fibres neatly follow the outside contour with some small gaps with a width of 0.05 mm.

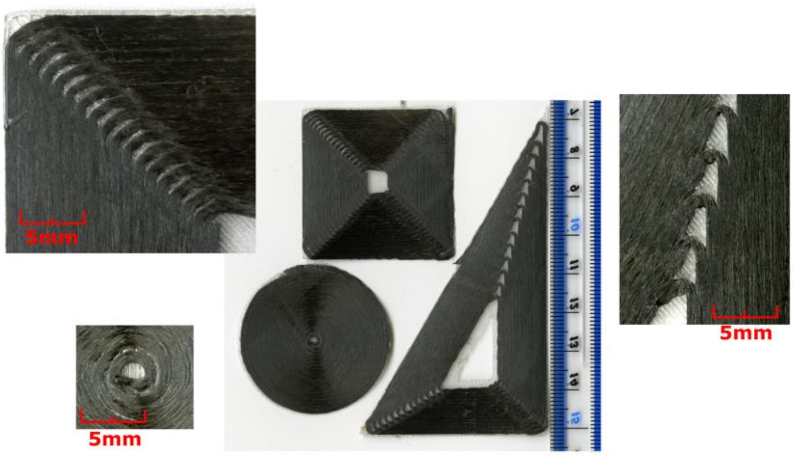
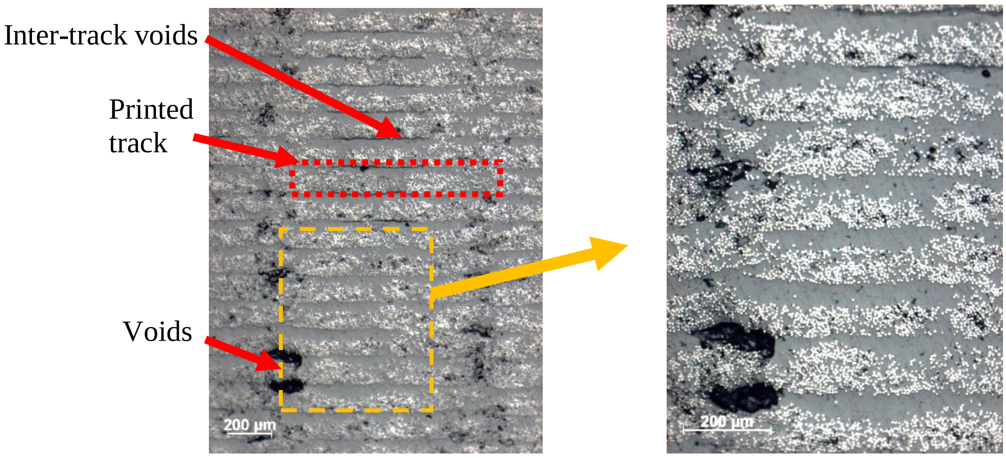


Figure 6: Benchmark prints for MarkOne continuous fibre printer with detail of corner radii.

Figure 7 is a micrograph of the cross section of one of the flexural test specimens showing the multiple stacked layers. Within each layer distinct regions of nylon, fibre, and voids can be seen. The fibre volume content is estimated to be 27% over the cross section, but a non-even distribution of fibres can be seen. This is attributed the non-uniformity of the printing filament, which may also lead to the creation of voids.

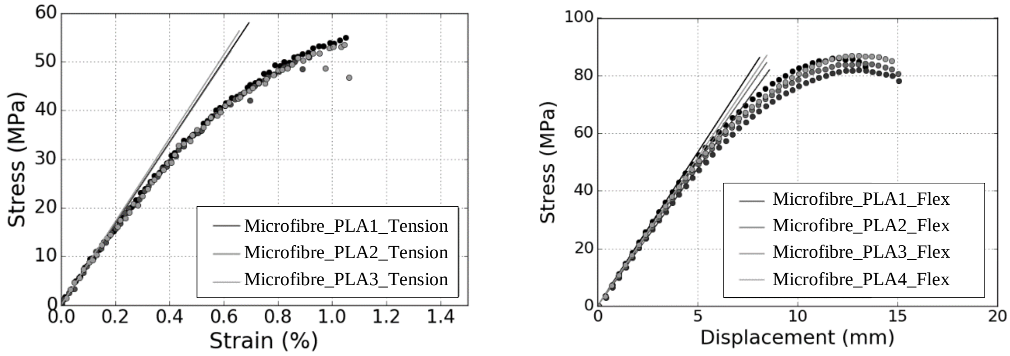


(a) **(b)**

Figure 7: Cross section of printed continuous fibre MarkForged part showing (a) structure from 3D printing tracks and (b) detail of larger voids between tracks.

4.2 Carbon fibre PLA filament characterization

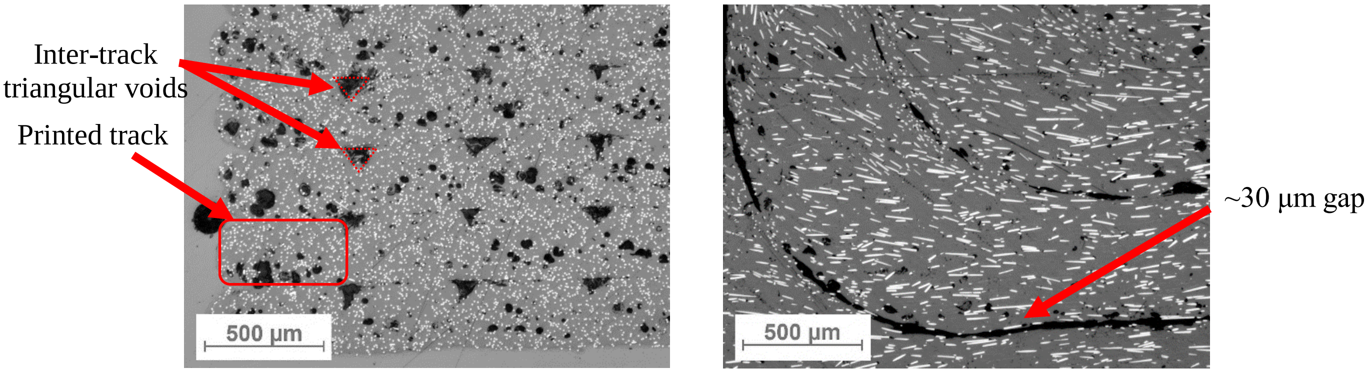
The result of the tensile tests and flexural tests on the carbon fibre PLA filaments are shown in Figure 8. The average tensile strength and stiffness are 56 MPa (±5.3%) and 8.4 GPa (±1.5%) respectively. The strength is typical for unreinforced PLA but the stiffness increased by roughly a factor of two [23]. This shows the fibres reinforce the material but do not reach their ultimate strengths. The flexural strength and stiffness were found to be 85 MPa (±2.7%) and 6.8 GPa (±6.5%) respectively. For the reinforced PLA filament, the flexural strength is higher than the tensile strength which is as expected for a high quality composite part [22].



(a) **(b)**

Figure 8: Results graphs of (a) tensile and (b) flexural test results of carbon microfibre reinforced PLA.

Figure 9 shows the micrographs of parts printed with short carbon fibre reinforced PLA. The printed part (Figure 9a) shows characteristic triangular voids between the printed tracks from the FFF process. The total void content is estimated at 18% (3.5% of the triangular voids and 14.5% different voids). The printed part has a higher void content than found in the filament, which was estimated at 8.2%. This can partially be attributed to the FFF printing process creating additional triangular voids, but the percentage of random voids also increased from 8.2% to 14.5% due to other methods which will need to be further identified.



(a) **(b)**

Figure 9: Microstructure of (a) printed part and (b) filament of short carbon microfibre reinforced PLA

Figure 9b shows a 90° corner region of a printed part, showing a gap between the printed tracks of about 30 µm. The short fibre reinforced filament shows a high void content, which may be attributed to the quality of the filament and yet unidentified mechanism during the printing process, but the print quality in the corner is good where there is a small gap between printed tracks and the short fibres follow the printing path. To minimize the void content, improving the filament quality and altering the mesostructure as described by Rodgriquez *et al.* [24] may be successful methods.

5 CONCLUSIONS & FUTURE WORKS

The properties of 3D printed composite parts have been investigated in this work. Previous studies have mainly focused on printing of thermoplastic filaments reinforced with very short fibres (~0.1 mm) or a filament with continuous fibre reinforcement. Both methods were assessed in this work, using a MarkOne 3D printer to print continuous carbon fibre / Nylon composite specimens and a standard open-source FFF printer to print carbon microfibre reinforced PLA. The tensile strength and stiffness of the continuous fibre printed parts were 986 MPa and 64 GPa respectively, which is an order of magnitude larger than the reinforced PLA printed parts (56 MPa and 8.4 GPa). A disadvantage of the continuous fibre printer, however, is limited control over the placement of the fibre and the creation of voids when printing more complex shapes. To overcome these disadvantages, a thermoplastic filament reinforced with short fibres above the critical fibre length (~3 mm) is proposed. This would yield mechanical properties similar to continuous fibre printer while maintaining the better processing qualities of short fibre reinforced filament. This may enable new applications for high performance 3D printed parts suitable for medical, aerospace, sport and rapid prototyping applications.

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