MECHANICAL BEHAVIOR OF CARBON FIBER COMPOSITES PRODUCED WITH FUSED FILAMENT FABRICATION

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<u>Abstract</u>

Fused Filament Fabrication (FFF) is a commonly used Additive Manufacturing (AM) technique. However, the printed parts often lack sufficient mechanical integrity. Recently, mechanical properties of FFF filament have been enhanced by blending pure polymer with short carbon fibers. This paper presents a study of the mechanical properties for carbon fiber filled (CFF) FFF parts produced with Makerbot printers. Polymer composite and pure polymer tensile test coupons are printed and then tested following ASTM D3039M. Here we consider FFF print orientations that are aligned with the test bar axis at 0 degree, 45 degrees, ±45 degrees, and normal to the bar axis at 90 degrees. The filament considered here was purchased from filament suppliers and included PLA, ABS, and PETG. Results are presented for tensile strength and tensile modulus. Additionally, short fiber composite samples are evaluated for fiber length distribution (FLD) and fiber weight fraction. Fracture surfaces are evaluated under SEM.

Introduction

Fused Filament Fabrication (FFF) is one of the most common AM techniques which fabricates parts using layer by layer polymer filament deposition process. A schematic of a typical FFF machine is shown in Figure 1. As in other AM technologies, FFF adds the material to the printed part layer by layer, only where it is needed, therefore saving energy, raw material cost, and waste. It is possible to reduce fabrication time with FFF, especially when an intricate geometry is required. Despite its advantages over conventional manufacturing process, the FFF printed parts often have inferior mechanical properties. The base materials used for printing are mostly thermoplastic polymers, which are weaker than the metals. In addition, print orientations lead to anisotropic material properties of the printed parts [1–3], and the layer by layer print deposition process can produce voids in the printed samples, resulting in lower tensile strength than that found in injection molded samples [1]. In order to print parts that give mechanical properties for industrial applications and end use, the print materials need to be enhanced.

Carbon fiber blended with base thermoplastic polymer can enhance the strength of the polymer material significantly, and therefore, has the potential to improve FFF filament properties. There have been several studies conducted related to FFF and carbon fiber composites. Love, et al. [5] showed that filament made from carbon fiber and Acrylonitrile Butadiene Styrene (ABS) polymer significantly increases strength and stiffness of the final parts. The CFF ABS dog bone sample built in plane has a tensile strength of 70.69MPa and a stiffness of 8.91GPa, comparing to 29.31MPa and 2.05GPa in the best scenarios for the pure ABS dog bone sample. They also demonstrated that the addition of carbon fiber (CF) decreased the distortion of the printed CFF ABS which was due to an increase in thermal conductivity of 124%, comparing to unfilled ABS. Fuda, et al. [6] investigated the material properties of different carbon fiber contents blended with

ABS polymer matrix. They concluded that the effect of the CF increased the printed samples' tensile strength (by approximately +22%), Young's modulus (+31.6%) and bending strength (+11.8%), in best cases. They also found that longer fiber length resulted in a higher tensile strength (about 7%) and modulus (about 20%), as expected. Furthermore, Tekinalp, et al. [7] conducted tensile tests with carbon fiber filled ABS filament at various fiber contents. The carbon fiber filled ABS yielded an improved tensile strength and tensile modulus by as much as 115% and 700%, respectively. They further discovered that the FFF process produced high fiber alignment along the print path [7].



Figure 1. Fused Filament Fabrication Technique [4]

The potential of CFF filament has led to its commercialization, and some of the suppliers including 3DXTECH (Grand Rapids, West Michigan, USA). Protoplant (Vancouver, Washington, USA), Colorfabb (Noorderpoort, Venlo, Netherlands) and Formfutura (Groenestraat, Nijmegenn, Netherlands) sell CFF products in the market. It is crucial to understand how well these filaments perform and what improvements are seen when CF is added. To the authors' best knowledge, there are no publications investigating the mechanical properties of the commercially available CFF polymer filament. Therefore, the goal of this paper is to investigate the tensile strength and tensile modulus of test bars made of commercially available CFF polymer composite filament, and the results are compared to the pure polymers from the sample filament supplier. FLD before and after the print is also investigated.

Experimental Setup

Three different filament materials are considered in this study including Polyactic Acid (PLA), Acrylonitrile Butadiene Styrene (ABS) and Polyethylene Terephthalate (PETG), which were purchased for testing. All the filaments were purchased from 3DXTECH (Grand Rapids, West Michigan, USA). Prior to use, each material was stored in sealed bags containing silica gel desiccant to avoid moisture absorption. A Makerbot Replicator 2X was used to print ABS and PETG filaments, and a Makerbot Replicator 2 was employed to print PLA filaments. Bead directions of 0 degree, 45 degrees, \pm 45 degrees and 90 degrees with respect to the axis of the test bar were investigated. A nozzle diameter of 0.6mm was used, and the nozzle itself was made of

hardened steel to prevent the carbon fiber from clogging and wearing the nozzle. Printing process parameters appear in Table 1.

| | PLA / CFF PLA | ABS / CFF ABS | PETG / CFF PETG | |
|---------------------------|---|------------------------|------------------------|--|
| Printer | Makerbot Replicator 2 | Makerbot Replicator 2X | Makerbot Replicator 2X | |
| Extruder temperature | 220° C | 230° C | 255°C | |
| Heat bed temperature | Not required 110° C | | 80° C | |
| layer infill | 0.2mm height \ 100% infill | | | |
| Heat bed surface | Blue painter's tape | Kapton tape | Kapton tape | |
| Nozzle | 0.6mm hardened steel | | | |
| infill print speed (mm/s) | 45 for base layer and outline \ 105 for the rest of the infills | | | |

 Table 1. Printer information, processing temperatures, layer infill information, printing surface, nozzle size and printing speed for each material filament

ASTM D638 procedure [8] was initially followed using Type I test bar geometry. However, tensile test at various orientations resulted in undesirable test data and premature failures at the radial section of the test sample. This was because the print path is discontinuous at the section of radial expansion (shown in Figure 3) that formed as the printer deposited an outline of the sample first and then made the infill.



Figure 2. Makerbot Makerware print preview

To avoid this complication, the ASTM 3039M procedure [9] designed specifically for polymer composites was adopted instead. The dimensions of the test coupon are 177.8mm×12.7mm×2.54mm which conformed to ASTM 3039M. Machined 6061 Aluminum tapered tabs were bonded at the ends of the test samples with Loctite Super Glue Ultra Gel [10]. Five test samples were prepared for each filament and print orientation combination. The Test Resources, Inc (Shakopee, Minnesota, USA). A 100 Series Family Single Column Test Machine [11] was employed for the tensile test. Samples were fitted with a 25mm extensometer [12] for strain measurement prior to testing. Tensile testing parameters used in this study are shown in Table 1. During the tensile loading, the force and strain data were collected for subsequent analysis.

| Crosshead speed | 2mm/min | |
|------------------------------|---------|--|
| Load cell capacity | 4 kN | |
| Sampling rate | 25 Hz | |
| Extensometer travel capacity | 6.25mm | |
| Tuble 2 Trating Dammer stars | | |

Table 2. Testing Parameters

After the tensile test, fracture surface images were obtained using a JEOL JSM - 6610 LV Scanning Electron Microscope [13]. A TA-Q50 Series Thermogravimetric Analyzer [14] was utilized for burn off test to obtain the amount of fiber within each polymer composite purchased. For each CFF material, three specimens were extracted from CFF filament and CFF test coupons at 0 degree print orientation. A customized burn off procedure shown in Table 3 was performed to obtain carbon fiber weight percentage.

10°C/min to 500°C \rightarrow Isotermoal for 1 hour \rightarrow test finished and cool down Figure 3. TGA burn off procedure

After the burn off test, the residuals were collected in sealed plastic tubes. Small particles with irregular shapes were identified under the SEM inspection, indicating that the polymer did not completely burn off. To get the actual fiber weight fraction, a pure polymer filament sample from each material was subjected to the same burn off procedure, and its residual weight fraction was used as a scale factor to approximate the actual polymer weight fraction and fiber weight fraction. To measure Fiber Length Distribution (FLD), the residuals were then distributed on a piece of copper tape which was then placed inside SEM for measurement. For each material with CFF filament and CFF test coupon, ten or more images were taken at 100X magnification with the same dimension. A Matlab program was written to calculate the pixel width based on the scale bar shown in Figure 4. Approximately 1000 fibers were evaluated for each material type.



Figure 4. Fiber Length Measurement image

Multiple images were taken for each material type and custom Matlab programs were written to measure the FLD. A schematic of the complete experimental process is shown in Figure 5.



Figure 5. Experimental procedure schematic

Results and Discussion

Tensile strength and tensile modulus for test bars described above appear in Figures 6 and 7 for ABS and CFF ABS, respectively. The error bars represent ± 1 standard deviation over five samples. Representative measured tensile test curves are given in Figure 8. CFF test coupons at 0 degree print orientation are seen to have tensile strength and tensile modulus that is 33.4% and 213%, respectively, above their unfilled counterpart. However, the CFF ABS coupons exhibit inferior performance in tensile strength at other print orientations. Tensile strength decreases by 13.6%, 15.8% and 15.6% at 45 degrees, ± 45 degrees, and 90 degrees, respectively, when CF is added. Alternatively, the tensile modulus shows a small improvement with CF, increasing by 43.9%, 45.9% and 47.0% at 45 degrees, ± 45 degrees, and 90 degrees, respectively.





Figure 8. Typical test curves for each print orientation, ABS vs CFF ABS

PLA and CFF PLA tensile test results are shown in Figures 9, 10 and 11. In a similar manner to the ABS results above, the tensile strength and tensile modulus is found to increase by 14.0% and 163%, respectively, for CFF PLA, at 0 degree print direction once CF is added. Note that CFF PLA is the least ductile material among all the test materials, and therefore tabs with smaller tapered angle is needed for 0 degree and 90 degrees print orientations to avoid undesirable test result. At 45 degrees, ±45 degrees, and 90 degrees print orientations, tensile strength improves by 2.1%, 4.5% and -3.9%, respectively, and tensile modulus increase by 50.6%, 59.8% and 44.3%, respectively.







Figure 11. Typical tensile curves, PLA vs CFF PLA

The tensile test results for PETG and CFF PETG appear in Figure 12, 13 and 14. The tensile strength and tensile modulus is found to increase by 48.4% and 313%, respectively, at 0 degree print direction. At 45 degrees, \pm 45 degrees, and 90 degrees print orientations, tensile strength improves by 7.8%, 23.2% and 2.8%, respectively, and tensile modulus increase by 83.6%, 121% and 68.6%, respectively. Tensile curves for PETG shown in Figure 14 have strain reduction during the tensile test; this is because the necking occurred outside the gage section. While the necking region became thinner, the remainder of the test coupon contracted under the reduced load. PETG is also the most ductile material among the tested materials, and the test

coupon at 0 degree print orientation did not fail before the test machine reached its data recording limit.



Figure 12. Tensile Strength PETG vs CFF PETG







Figure 14. Typical tensile curves, PETG vs CFF PETG

The addition of carbon fibers is shown to dramatically increase tensile modulus at 0 degree print orientation for PLA, ABS and PETG test coupons. A general trend becomes apparent that the addition of carbon fiber improves tensile modulus of the test coupons for all print orientations considered here, but decreases ductility. When the print orientation aligns with the axis of the test bar, mechanical properties show the most improvement. This agrees with the findings of Tekinalp, et al. [7] that the FFF process produces parts with high fiber alignment along the print direction. In this case, the high tensile strength of the carbon fiber greatly contributes to the increase in the mechanical properties of the CFF polymer test coupons.

| | | ABS | CFF ABS | PLA | CFF PLA | PETG | CFF PETG |
|------------------|------------------|----------|----------|----------|----------|----------|----------|
| 0° | Tensile Strength | 38.2 MPa | 50.9 Mpa | 60.0 Mpa | 68.4 Mpa | 46.1 Mpa | 68.3 Mpa |
| | Tensile Modulus | 2.29 Gpa | 7.15 Gpa | 3.53 GPa | 9.28 GPa | 2.05 GPa | 8.47 GPa |
| $\pm 45^{\circ}$ | Tensile Strength | 34.7 Mpa | 29.2 Mpa | 52.2 Mpa | 54.6 Mpa | 41.3 Mpa | 50.9 Mpa |
| | Tensile Modulus | 2.12 GPa | 3.09 Gpa | 3.26 GPa | 5.20 GPa | 1.91 GPa | 4.23 GPa |
| 45° | Tensile Strength | 34.8 MPa | 30.1 Mpa | 49.6 Mpa | 50.7 Mpa | 41.9 Mpa | 45.2 Mpa |
| | Tensile Modulus | 2.18 GPa | 3.14 GPa | 3.37 GPa | 5.08 GPa | 1.96 GPa | 3.60 GPa |
| 90° | Tensile Strength | 32.4 Mpa | 27.3 Mpa | 45.5 Mpa | 43.7 Mpa | 41.4 Mpa | 42.6 Mpa |
| | Tensile Modulus | 2.06 Gpa | 3.03 GPa | 3.19 Gpa | 4.61 GPa | 1.89 GPa | 3.19 Gpa |

Table 3. Tensile test summary

The tensile test result is summarized in Table 3, and several observations can be highlighted. CFF PLA and CFF PETG at 0 degree print orientation have the greatest tensile properties and tensile improvement, respectively. Samples at the 0 degree print orientation gives

the greatest tensile improvement for all of the materials, and 90 degrees print orientation gives the least improvement.

Figures 15, 16 and 17 show SEM images typical of the fracture surfaces for each material considered here. The images on the top row and bottom row of the figures represent the fracture surfaces of the pure polymer test coupon and the CFF test coupon, respectively. Each column represents the fracture surfaces at different print orientation. Certain features are labeled in the SEM images with lower case letters.



Figure 15. SEM Images for ABS and CFF ABS

Type (a) voids appearing in Figure 15 that formed between the layers during the print process are easily identified inside the pure polymer test coupons at the 0 and \pm 45 degrees print orientations. The remainder of the fracture surface appeared to be well packed and free of voids. Type (a) voids are less prominent in CFF test coupon surfaces. This is likely a result of the CFF polymer being more thermally conductive than pure polymer, thus improving the fusion between layers during the deposition process and reducing type (a) void. Other defects, however, are more pronounced. For example, images D and E in the three figures have numerous small type (b) circular voids which are likely sites where fibers were pulled out of the polymer during the tensile test. There are also fibers exposed on the fracture surface, identified as type (c) feature. More interestingly, there seems to be pore enlargement around the type (c) feature in several sites. In addition, a type (d) pore has circular dented shape, and we believe it occurred when the fiber ends were pulled away from the polymer. Type (b), (c) and (d) features indicate poor interfacial bonding strength between polymers and fibers, which is most readily apparent on the CFF fracture surface of the 90 degrees print orientation. Considering image F in Figures 15, 16

and 17, a large number of fibers are exposed on the fracture surfaces. Instead of sharing the tensile force with the polymer matrix, the weak interfacial bond fails resulting in tensile strength that is lower than pure polymer test coupon. This is confirmed by the tensile test results in Figures 6, 9 and 12. Finally, note that the fibers align well with the print orientation in each image, further agreeing with the observation made by Tekinalp, et al. [7].



Figure 16. SEM Images for PLA and CFF PLA fracture surfaces



Figure 17. SEM Images for PETG and CFF PETG fracture surfaces

The carbon fiber weight fraction for each material is shown in Table 4. FLD comparison before and after the print, weight average fiber length, weight average fiber aspect ratio and number of fibers evaluated appear in Figure 18. Fiber diameter on 15 different fibers where measured to obtain the average fiber diameter of $7.77\mu m$. One common trend can be observed from the results is that the curves of FLD for printed CFF samples skewed a bit left with respect to the CFF filament FLD curves. Furthermore, the differences between weight average aspect ratios before and after the print are less than one. These results show that the fiber length remains mostly unaffected as the CFF filament travels throughout the print process.

| CFF ABS filament | 16.8% |
|-------------------|-------|
| CFF PLA filament | 12.6% |
| CFF PETG filament | 17.7% |

Table 4. Carbon fiber weight fraction for each CFF material



Figure 18. Fiber length distributions and fiber length information for each CFF material filament

Conclusion

Several observations can be concluded from the research so far:

- The addition of carbon fiber to FFF filament gives the greatest tensile property and improvement at 0 degree print orientation;
- The addition of carbon fibers improves the stiffness of printed test sample, but results in reduced ductility;
- CFF PETG has the greatest overall tensile property improvement among the three selected CFF filaments;
- The FFF process results in small fiber breakage when processing CFF filled filament;
- Improving interfacial bonding strength between fiber and polymer matrix is needed to fully realize the potential of CFF FFF parts.

Acknowledgement

This paper is supported by Baylor Material Research Group, and the authors would also like to thank Baylor Research and Innovative Collaborative for providing the space for this work.

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