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Correlating Large-Format AM Print Parameters to Fiber Length and Mechanical Performance of Reinforced Polymer Composites

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Abstract

This paper aims to correlate processing conditions to fiber length and mechanical properties in fiber-reinforced composites on the Big Area Additive Manufacturing (BAAM) system at Oak Ridge National Laboratory. The processing of fiber-reinforced composites has a significant influence on their microstructure, which dictates the properties of the final product. The effect of processing is poorly documented in systems like the BAAM, leaving significant opportunities to improve the mechanical performance of printed structures. In this work, fiber length distributions from pelletized feedstock were compared against those of specimens extruded under different processing speeds. The mechanical strength of each specimen was evaluated to correlate processing speed to fiber length and mechanical properties. Experimental results showed that fiber length decreases slightly with increasing screw speed. Mechanical performance was not found to be affected by the decrease in fiber length. This research will guide future modifications to hardware design and print parameters to maintain fiber length and maximize mechanical performance.

Introduction

Reinforcing fiber is essential to large-format polymer additive manufacturing [1]. However, the effect of processing conditions on fiber-reinforced composites in large-format additive manufacturing has not been well documented. The BAAM uses a single screw plasticating extruder to rapidly deposit large quantities of material in the printing process. Figure 1 shows an example diagram of a plasticating extruder [2]. Pellets are fed into the flights of a screw through a hopper, and then are melted through compression caused by the screw. This creates a torturous environment for the fibers, which partially defines the mechanical properties of the extruded material. Fortunately, this process is almost identical to injection molding, where there have been numerous investigations concerning the influence of processing conditions of fiber reinforced plastics, and the resulting mechanical properties of the material. Wolf et al demonstrated that conditions which increase shear decrease average fiber length [3]. Wolf took fiber length distributions from different regions within the extruder, and under different processing conditions including screw speed, temperature, and die diameter. Wolf found that gentler processing conditions (slow screw speed, higher extrusion temperature, larger die diameter) result in the preservation of the fiber length. Turkovitch et al. studied the fracture of glass fiber through the length of the extruder and at different screw speeds. The findings were the same as Wolf; fibers progressively break through extrusion, and fibers breakage increases with screw speed [4]. Yilmazer et al. examined the effects of processing conditions on fiber length and part strength in injection molded samples. They reported that fiber length and tensile properties significantly decrease as shear rate is increased through the alteration of the screw speed or feed rate [5]. Hausnerova et al. studied fiber degradation during multiple extrusion cycles under a range of shear rates through a capillary rheometer. Hausnerova reported that under high shear rates the length of fibers decreases, with significant damage occurring during the first extrusion cycle [6]. In summary, processing conditions have been found to affect mechanical properties through reduction or retention of the initial fiber length distribution.

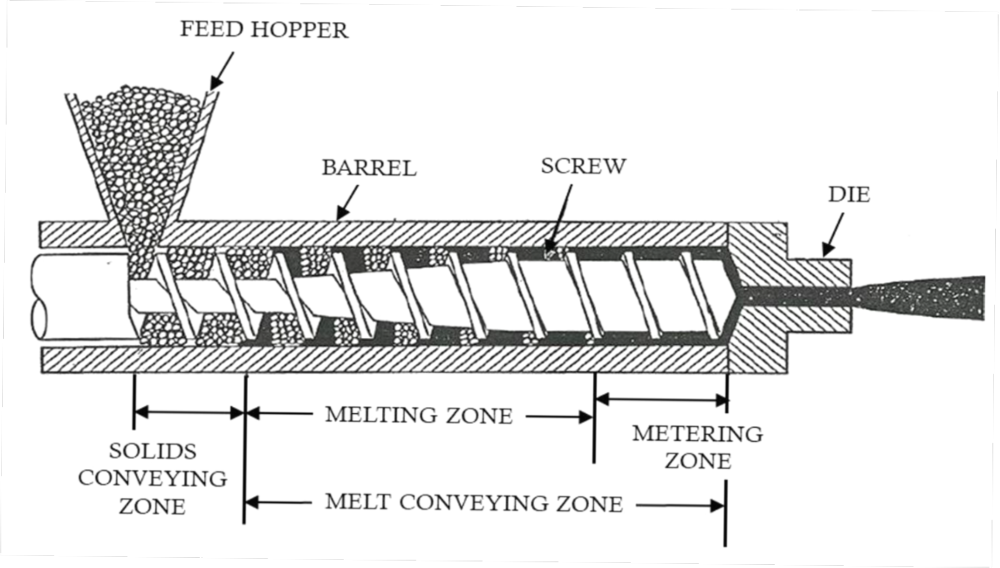

Figure 1: Diagram of a plasticating extruder.

This work examines the influence of screw speed on the fiber length and the resultant tensile properties using a large-format polymer additive manufacturing system. This subject has been extensively studied in an injection molding setting, but it has not been well tested in a large-format screw extrusion additive manufacturing system like the BAAM. This study was designed under the fact that the BAAM uses a modified version of an injection molding extruder, and therefore the effect of processing conditions on fiber length and mechanical properties should follow those of the studies done in injection molding.

Methods

Printed Samples

The material used in this study was Techmer Electrafil, a pelletized 20% carbon fiber reinforced acrylonitrile butadiene styrene (CF-ABS) feedstock used in screw extrusion processes [7].The pellets were an average of 2.5mm in diameter, 3mm long, and contained fibers with a diameter of 7μm. The pellets were dried at 80°C for at least 4 hours to remove moisture before printing. The processing condition chosen for this study was screw speed. All other conditions were held constant. The screw speeds tested were 80, 160, and 240 RPM, as it is generally accepted that the BAAM operates within this range. Samples were printed at 250°C with a

10.16mm (0.4”) diameter nozzle, 5.08mm (0.2)” layer height, and a 17.78mm (0.7”) bead width. Each sample set was printed as a 0.6m (2’) x 0.6m (2’) x 0.3m (1’) box Figure 2.

Tensile specimens (ASTM D638 Type 1, Figure 3) were milled flat to about 6.5mm, and waterjet cut from the walls of the boxes [7]. The specimens were cut such that the load direction was parallel to the deposition direction. The samples were dried at 50^{o}C for at least 48 h and left in a desiccant chamber at 23^{o}C for at least 5 h (ASTM D618-B) prior to tensile testing. To ensure statistical significance, 5 specimens were measured for each condition. Specimens that were damaged at any point were discarded.

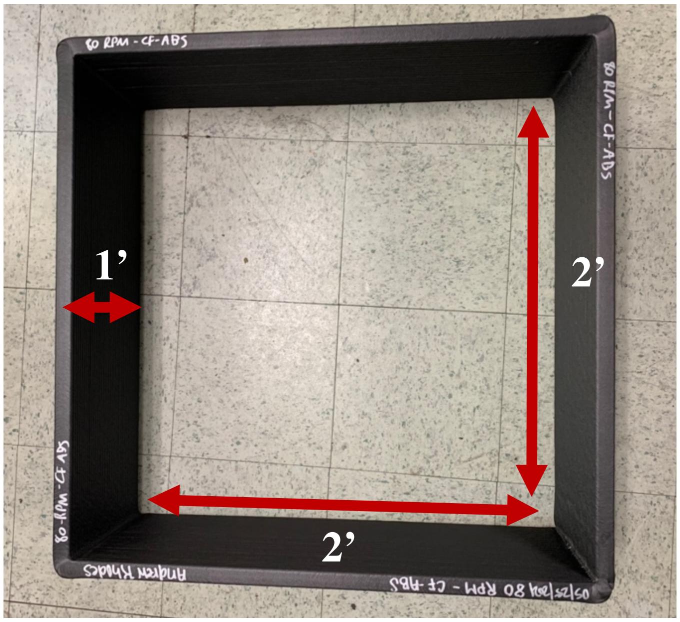

Figure 2: One of the printed boxes.

Tensile testing of all specimens was performed on an MTS Criterion Series, Model 45 with a 10kN load cell. The testing rate of 1mm/min was used for a nominal strain rate of .1mm/(mm min). Extension was measured using an MTS LX 500 Laser Extensometer. Specimens that fractured outside of the gage length were omitted from the data analysis.

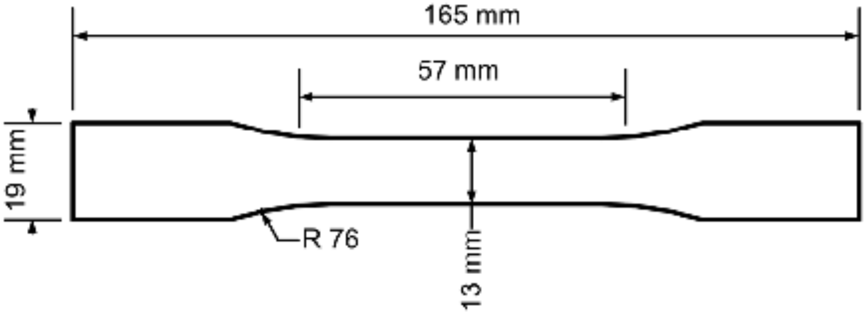

Figure 3: ASTM D638 tensile specimen dimensions

Fiber Analysis

The steps of fiber measurement follow those outlined by Kunc et al. [8].

1. Composite coupon isolation

   To characterize the fiber length distribution within the ABS matrix, coupons of the composite were taken from the material remaining in between the nested tensile samples after water jetting. These coupons were approximately 6mm tall and wide and cut to 6mm long, such that the weight was approximately 0.3g. The raw stock fiber length does not exceed 600μm, therefore the distance of an edge from the center of the coupon had to be greater than 600μm.

2. Constrained removal of matrix material

   The composite coupon is placed in a glass vial with 15ml of acetone to dissolve the coupon. The coupon vials were put into an ultrasonic bath for at least an hour so that the fibers separated from the matrix. It is apparent that the matrix has been removed when clumps of fiber are no longer visible.

3. Fiber sample isolation

   The fiber sample isolation step requires isolating a portion of fibers for dispersion and imaging. The process involves suspending the fibers in the solution and then drawing out a portion of the fibers that will represent the whole. The fiber solutions were stirred on a vortex mixer at the highest setting for 20 seconds each. Immediately following, a 5ml pipette with a 2mm diameter tip was used to draw out solution.

4. Filament dispersion

   Dispersion of the fibers was performed by diluting one part solution from the previous step with two parts acetone. This reduces the amount of polymer matrix in the solution that leaves a film on the fibers and microscope slides and reduces the likelihood of clumped fibers. The samples were vortex mixed a second time once they were diluted, and 3 to 4 drops were pipetted onto a microscope slide in a continuous, linear motion so that most of the slide was covered. The slides were allowed to sit until the acetone evaporated from the surface, which took no more than a few moments. This method gives good fiber dispersion, avoiding fiber clumping and fiber-poor regions. Additionally, each microscope slide shared roughly the same number of measurable fibers (~500 fibers) between slides created by the same solution. It was found that applying the solution to the slides in droplets promoted fiber clumping and uneven fiber dispersal. Proper dispersion is essential, since poor dispersion increases fiber overlap, and overlapping fibers cannot be measured.

5. Imaging and fiber length measurement

   Imaging was performed on a Keyence digital microscope using 50x magnification. The slides were imaged using the Keyence software's image stitch function. This took a grid of images within a region encompassing the entire slide, and then stitched them together into a single high-resolution image. The fiber length distribution was determined by manually measuring the length of 1500 fibers using ImageJ software, as the distribution stabilized at around 1000 [9].

Average fiber length was calculated using two methods: number average and length weighted average, shown in Equation 1 and Equation 2 respectively. The number average is a representation of the number of shorter fibers, calculated by the sum of the individual fibers divided by the total number of fibers measured. The length weighted average is calculated as the sum of the individual fiber lengths squared divided by the sum of the individual fiber lengths. The length weighted average gives more importance to longer fibers. Both values are often included in fiber length analyses and are useful in comparing the degree of length reduction of fiber on both sides of the distribution.

$$\boldsymbol{L_n = \frac{\sum N_i l_i}{\sum N_i}}$$

Equation 1: Number average fiber length

$$\boldsymbol{L_n = \frac{\sum N_i {l_i}^2}{\sum N_i l_i}}$$

Equation 2: Length weighted average fiber length

Results and Discussion:

Fiber length distribution

Figure 4 shows the fiber length distribution of the pellet feedstock. The feedstock had the highest distribution of long fibers, and the lowest distribution of short fibers of the samples. Two notable features are that the distribution is already quite broad, and that the distribution is

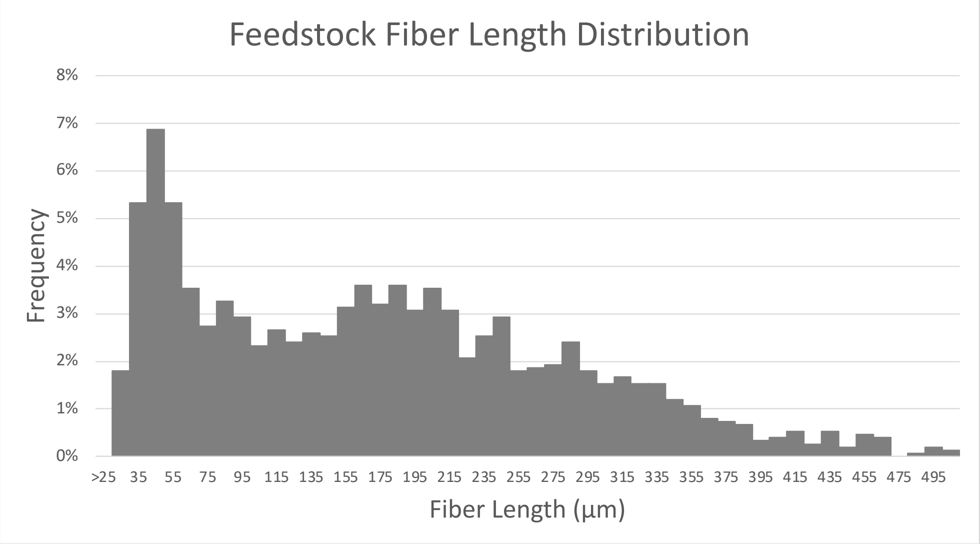

Figure 4: Fiber length distribution of the unprocessed feedstock

bimodal. This broad distribution is likely due to the manufacturing method, where the fibers were chopped and mixed using a dual screw extruder. The number and length weighted averages for the feedstock were 188µm and 253µm.

The first condition tested was the 80 RPM screw speed. The resulting fiber length distribution and averages show the shortening of the longer fibers and can be seen in Figure 5 and Table 1. Comparing the distribution to the feedstock, the second mode of the 80 RPM widened to encompass shorter fiber lengths. This wider mode contains the longer fibers that were more susceptible to breaking in the lowest shear rate tested. The fiber length averages tell the same story; the difference between the number average is 3%, and the length weighted average is 6%. This is evident that the longer fibers were broken more than the shorter were. The 160 RPM condition shows a surprisingly larger degree of attrition; a 17% reduction from the feedstock in both averages. The 160 RPM fiber length distribution is now unimodal with the only mode being greater than the previous two distributions. The 240 RPM fiber length barely changes from the 160 RPM, as shown by the nearly identical distributions and averages. Since the fibers experienced such a large degree of attrition at 160 RPM, it is likely they reached a critical breaking point due to the shear in the extruder and will not shorten much more under the

	Number average FL	**Weighted average FL**
Feedstock	188µm	253µm
80 RPM	182µm	239µm
160 RPM	156µm	211µm
240 RPM	158µm	216µm

Table 1: Fiber length average representations from the feedstock and tested screw speeds

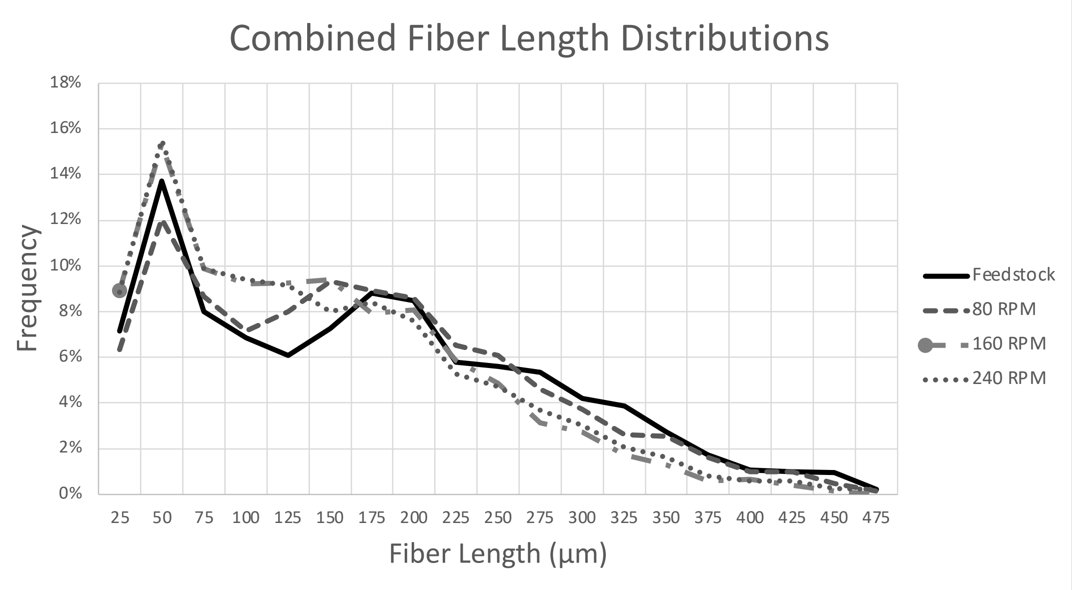

Figure 5: The combined fiber length distribution plot for each condition

conditions the BAAM can achieve. Although the 240 RPM averages are slightly longer than the 160 RPM, this difference is insignificant.

Tensile Properties vs. Print Parameters

Figure 7 and Figure 6 exhibit the relationship between screw speed on the elastic modulus and ultimate tensile strength respectively. A trend line of the length weighted fiber average is added to the plots to compare the relationship between fiber length and tensile properties. The figures show that there is a lack of correlation between fiber length and tensile

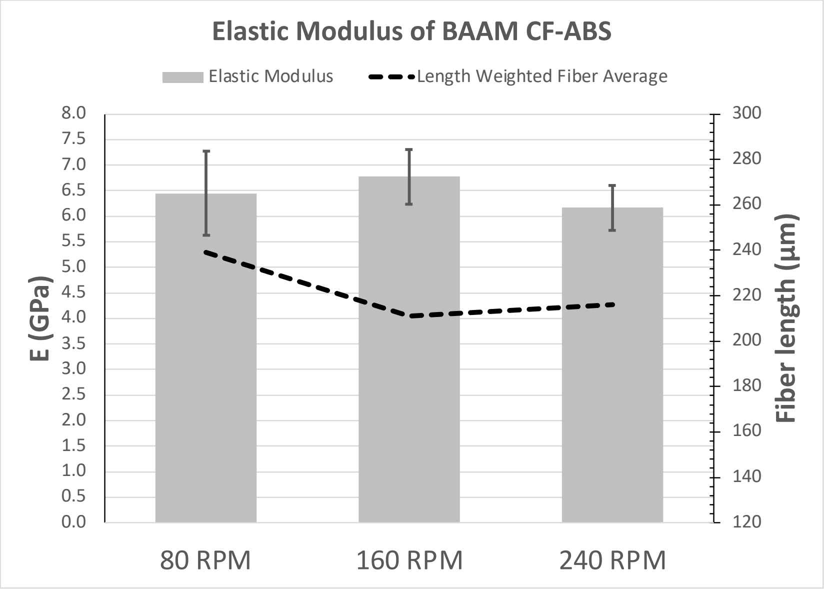

Figure 7: Elastic modulus vs. screw speed and fiber length

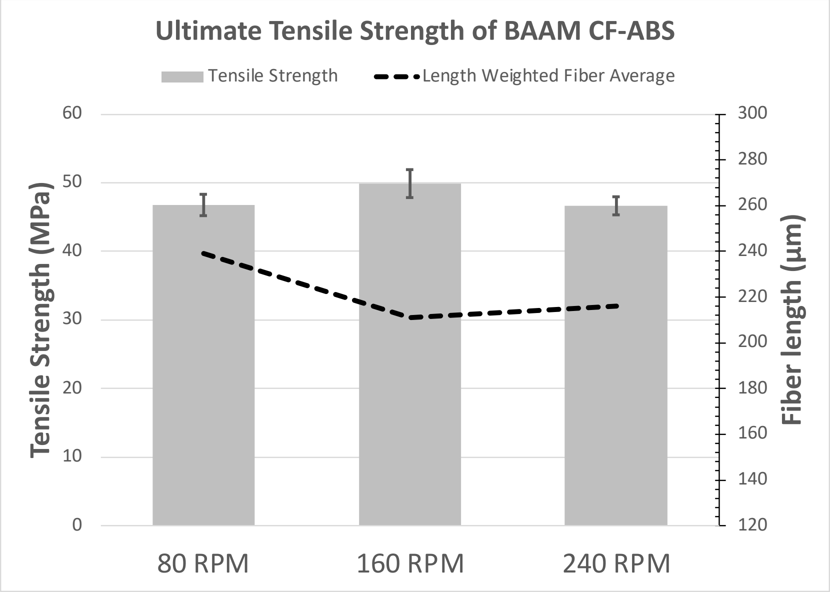

Figure 6: Ultimate tensile strength vs. screw speed and fiber length

properties. The differences between the elastic moduli vary but are within a standard deviation of each other. The variation in the ultimate tensile strength is even less, with only a slight difference between 160 RPM and 80/240 RPM, which have nearly identical values.

Conclusion and Future Work

Screw speed was not found to have a meaningful effect on the fiber length and mechanical properties of fiber reinforced large-scale printed material under the conditions of this experiment. The fiber length distribution of the pellet feedstock was already short at around 200µm, and so it is likely that the fibers are not long enough to break to significantly shorter lengths under the operating conditions as described. It is known that fiber length should have a positive correlation to tensile properties. However, these results show that although fiber length decreased in processing, the difference between the maximum and minimum fiber lengths (~30µm) seems to be insignificant and have little effect on the tensile properties of the final product. As a follow-up to this experiment, feedstock that has been pultruded will be investigated so that the initial fiber length distribution is relatively long and uniform. This will provide an analog for the degree of fiber length attrition under large-format additive manufacturing. It is now known that materials containing short, chopped carbon fiber reinforcements, like the one used in this study, are functionally unaffected by screw speed in a system like the BAAM.

Acknowledgements

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