EFFECTS OF FIBER ASPECT RATIO ON PREDICTING ELASTIC PROPERTIES OF SHORT FIBER REINFORCED COMPOSITES PRINTED BY LARGE SCALE POLYMER ADDITIVE MANUFACTURING

Zhaogui Wang, Douglas E. Smith, David A. Jack
Department of Mechanical Engineering, Baylor University

Abstract

Short fiber reinforced composites continue to see increased application in large scale polymer-extrusion-based additive manufacturing due to their superior mechanical performance as compared to a neat polymer alternative. The extrusion deposition process is known to decrease fiber length which results in a fiber aspect ratio distribution. It is well established that the geometric aspect ratio is an important factor that affects the elastic properties of the processed fiber composite structure. Hence, we propose a numerical approach to predict the variability of elastic properties for printed polymer composites by considering various fiber aspect ratio distributions within the extrudate. By this investigation, the manufacturing process can be optimized to print beads with preferable properties by adjusting the fiber aspect ratio of the extruded polymer composites during printing. Our method evaluates the elastic properties of a polymer composite extrudate by applying the orientation homogenization method and the Tandon-Wang equation using a computed flow-induced fiber orientation. We define the fiber aspect ratio distribution using a two-parameter Weibull probability distribution function, which yields an asymmetric aspect-ratio-probability function that is parametrized by the mean and the mode fiber aspect ratios. Numerical results indicate that the mean fiber aspect ratio significantly affects the averaged elastic moduli of the extrudate, while the mode fiber aspect ratio has much less impacts on the predicted stiffness.

Background

Due to superior properties such as low thermal expansion and high stiffness-weight ratio, chopped carbon fiber reinforced composite has been used as the feedstock in large-scale extrusion-based additive manufacturing, which is extended from the conventional Fused Filament Fabrication (FFF) process [1]. In the large-scale additive manufacturing process, thermoplastic pellets (filled or unfilled) are melted and deposited onto a base plate following certain road path, layer-by-layer, to form a three dimensional object. The extrusion process during melting and mixing of the feedstock will degrade the fibers as well as their alignment state (see Figure 1), and the results varies with different processing parameters. For filled thermoplastics, the geometry and orientation of the reinforcing fibers have significant influences on the material properties of the printed beads. Investigations of the fiber fillers in the additive manufacturing area has become interest in recent years. Nixon, et al. [2] predicted the fiber orientation patterns in FFF printer nozzle with different geometric designs. Heller, et al. [3] extended the nozzle flow geometry to include a short section of free extrudate and based upon a 2D axisymmetric Stokes flow model. Based on their computed fiber orientation, the elastic moduli of an extrudate composite were evaluated using the orientation homogenization method, assuming the molten polymer is a creeping Newtonian flow. Using a similar method, Wang, et al. [4] assessed the polymer rheology effects on the predicted fiber orientation and associated elastic properties of a printed extrudate by applying different rheology models (Generalized Newtonian models and Viscoelastic Fluid model) in the flow modeling. Russell, et al. [5] evaluated the thermal expansion behavior of the extruded composites based on the predicted flow-induced fiber orientation pattern. From an experimental perspective, Duty, et al. [1], Love, et al. [6] and Kunc, et al. [7] each measured the tensile moduli of printed fiber composites samples prepared through the Big Area Additive Manufacturing (BAAM) system. Specifically, both the numerical predictions [2-5] and experimental observations [1,6-7] have reported that a significant enhancement in the tensile modulus along the extrusion direction was found while the improvement at the transverse direction not so noticeable.

Fiber aspect ratio is a key factor in the manufacturing process associated with discontinuous fiber filled composites. Unfortunately, there are few studies of fiber aspect ratio distributions in polymer composite manufacturing. This paper is designated to exploit the effects of the fiber aspect ratio distribution in the resulting elastic properties of a composite extrudate printed through the large scale additive manufacturing process. We are aiming to offer a numerical method to correlate the elastic behaviors of a printed bead with the fiber aspect ratio parameter, which may help in the design and optimize the outcome from additive manufacturing process by controlling the fiber aspect ratio within the extrudate during extrusion processing. Our approach calculates the elastic properties of the extrudate composite using the orientation homogenization method with a known flow-induced fiber orientation. The fiber aspect ratio of an extrudate is expressed through a two-parameter-defined Weibull probability distribution function. The controlling parameters are computed by a given set of mean and mode fiber aspect ratios.

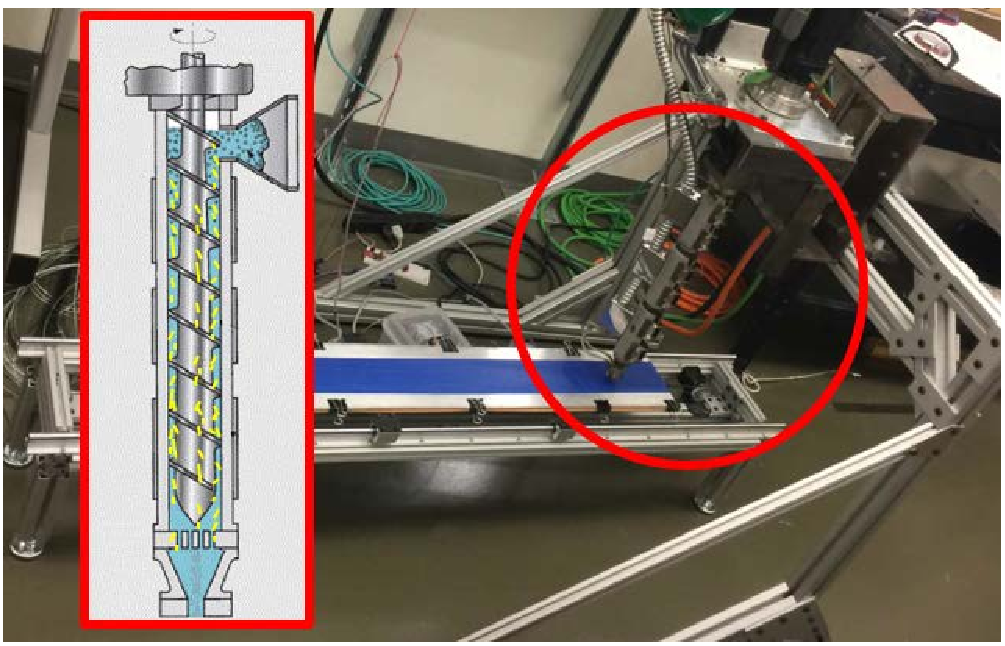

Figure 1: The single screw extrusion process breaks the fibers and results in a non-uniform aspect ratio distribution.

Flow-induced Fiber Orientation

The alignment of the reinforcement within the fiber suspension is of significant importance in evaluating mechanical properties of fiber filled composites. Evans, et al. [8] shown that the flow kinematics orientated the filled fibers while the fiber alignment patterns also affected the properties of the fluid flow, such as the shear viscosity. Lipscomb, et al. [9] also proved the importance of strongly coupling the interaction between fibers and the fluid flow in order to predict the behavior of the compound. Besides the fully coupled method, the one-way weakly coupled, where the influence of the fillers on the flow is neglected, has been used extensively.

Particularly, this method is considered sufficient for the shear dominant narrow gap flow applications such as the injection molding process [10]. The orientation input we used in this study is also computed through the one-way weakly coupled method, where we first solve the fluid flow problem neglecting the effects of fiber reinforcements and then compute the fiber orientation pattern following the solved flow kinematics along streamlines within the flow domain, assuming the filled fibers did not change the flow properties in a noticeable way.

The fluid flow model is created through the finite element suite, ANSYS Polyflow. A 2 dimensional axisymmetric flow is sufficient due to the symmetry of the nozzle, which considers only half of the flow domain shown in Figure 2 [11]. Besides, a prescribed rotational velocity (ω) is applied to the screw edge, which is defined through a linear function of the r-coordinate (e.g. $\omega = kr$, where k is the maximum angular velocity computed based on the given RPM of the screw). In this study, the maximum RPM of the screw is 1000 and the corresponding inlet flow rate is about 400 mm^3/sec. The flow domain includes the ending section of the screw, the nozzle-shape die region, and a short section of free extrudate. We assume a Stokes flow where the inertia effects are ignored due to the low Reynolds number most polymer melts have. The Phan-Thien-Tanner (PTT) model is applied in modelling the rheology of the molten ABS polymer, which is chose as the matrix material of this study. The fitted parameters of the PTT model is given in Table I. Note, this is a six-mode PTT model. λ_i and η_i are the relaxation time and viscosity constant for each mode. ε and ξ are parameters control the extensional and shear behaviors of the viscosity of the molten polymer.

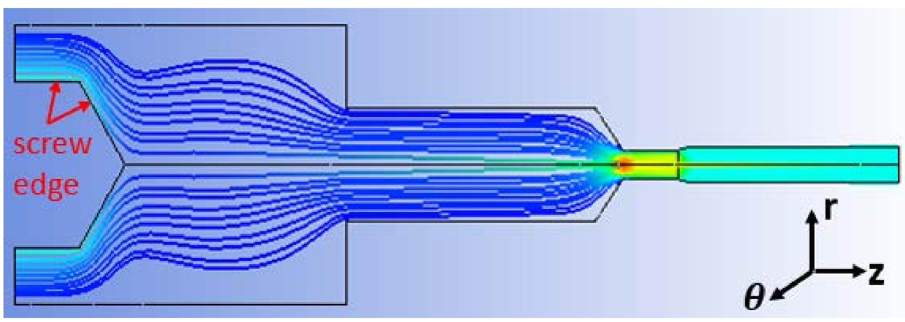

Figure 2. Screw nozzle flow modeled in ANSYS Polyflow. (Note, θ direction is out of plane.)

Table I: Fitted parameters of the PTT model for the chose ABS material. [4]

Acrylonitrile butadiene styrene (ABS)				
Mode No. (i)	λ_i (s)	η_i (Pa s)	ε	ξ
1	0.00022	131.7	0.75	0.18
2	0.0022	44.7	0.75	0.18
3	0.012	1180.8	0.75	0.18
4	0.12	6286.4	0.75	0.18
5	1.14	13065.7	0.75	0.18
6	13.82	61917.7	0.75	0.18

The direction of a single rigid fiber within a thermoplastic matrix is often depicted through a unit vector $\boldsymbol{p}(\sin\varphi\cos\phi, \sin\varphi\sin\phi, \cos\varphi)$, as shown in Figure 3.

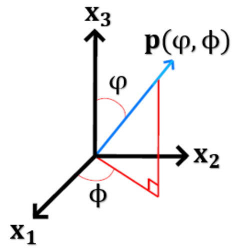

Figure 3. Directional representation of a single rigid reinforcing fiber.

Advani and Tucker [12] offers a computationally efficient method to describe the statistical behavior of a bundle of fibers by expressing the fiber alignment through the fiber orientation tensors. In particular, the orientation tensors are solved from the Reduced Strain Closure (RSC) method [13], which is written as

$$\frac{DA}{Dt} = (\boldsymbol{A}\cdot\boldsymbol{W} - \boldsymbol{W}\cdot\boldsymbol{A}) + \lambda(\boldsymbol{D}\cdot\boldsymbol{A} + \boldsymbol{A}\cdot\boldsymbol{D} - 2[\mathbb{A} + (1-\kappa)(\mathbb{L} - \mathbb{M}{:}\mathbb{A})]{:}\boldsymbol{D}) + 2\kappa C_I\dot{\gamma}(\boldsymbol{I} - 3\boldsymbol{A}), \quad (1)$$

where $\boldsymbol{W}$ and $\boldsymbol{D}$ are the vorticity and rate of deformation tensors, which are computed from the velocity gradient field $(\nabla\mathbf{v})$ as,

$$\boldsymbol{W} = \frac{1}{2}[(\nabla\mathbf{v}) - (\nabla\mathbf{v})^{\mathrm{T}}],\ \boldsymbol{D} = \frac{1}{2}[(\nabla\mathbf{v}) + (\nabla\mathbf{v})^{\mathrm{T}}], \quad (2)$$

and $\dot{\gamma}$ is the scale magnitude of the rate of deformation tensor. Besides, $\boldsymbol{A}$ and $\mathbb{A}$ are second and fourth order orientation tensors, which are evaluated as

$$\boldsymbol{A} = \mathrm{A}_{ij} = \oint_{\mathbb{S}} \mathrm{p}_i\mathrm{p}_j\delta(\varphi,\phi)\mathrm{d}\,\mathbb{S},\ \mathbb{A} = \mathrm{A}_{ijkl} = \oint_{\mathbb{S}} \mathrm{p}_i\mathrm{p}_j\mathrm{p}_k\mathrm{p}_l\delta(\varphi,\phi)\mathrm{d}\,\mathbb{S}, \quad (3)$$

where $\delta(\varphi,\phi)$ is a probability distribution function and $\mathbb{S}$ is unit sphere surface. Due to the normalization condition, the integral of $\delta(\varphi,\phi)$ over the surface $\mathbb{S}$ equates to unity, making the trace of $\boldsymbol{A}$ equal to one (see e.g., [14]). $\mathbb{L}$ and $\mathbb{M}$ are expressed as

$$\mathbb{L} = L_{ijkl} = \sum_{m=1}^{3}\Lambda_m\boldsymbol{n}_i^m\boldsymbol{n}_j^m\boldsymbol{n}_k^m\boldsymbol{n}_l^m,\ \mathbb{M} = M_{ijkl} = \sum_{m=1}^{3}\boldsymbol{n}_i^m\boldsymbol{n}_j^m\boldsymbol{n}_k^m\boldsymbol{n}_l^m, \quad (4)$$

where Λ_m is the m-th eigenvalue of the second order orientation tensor, and the $\boldsymbol{n}_i^j$ is the i-th component of the j-th eigenvector of the second order orientation tensor. Moreover, C_I is the fiber-fiber interaction coefficient and κ is the scale factor, which can be determined by fitting experimental data. The typical choice of C_I is from 0.0001 to 0.01, and κ is from 0 to 1 [5]. In this study, we empirically set $C_I = 0.01$ and $\kappa = 1/2$ preliminarily based on prior related literatures [3-5]. Further, the parameter λ is used to count the fiber geometric effects on the orientation, which is defined as

$$\lambda = \frac{(a_r)^2 - 1}{(a_r)^2 + 1}, \quad (5)$$

where a_r is the geometric aspect ratio of a ellipsoidal fiber. It is important to note the aspect ratio distribution resulted through the extrusion process will affects the flow properties and ultimately impacts the orientation of the reinforcing fibers. However, our primary objective in this paper aims to investigate the effects of the process-yielding distribution of the fiber aspect ratio on the predicted stiffness of the extruded polymer composite. We understand that research on effects of varying fiber aspect ratio on flow and fiber orientation modeling is of great help but beyond the major scope of this paper. Hence, for current study, we fixed the parameter λ as unity, which corresponds an infinite aspect ratio.

Fiber Aspect Ratio Distribution Function

To achieve high flow rate and efficiently mix the pelletized feedstock, a single screw extruder is equipped in large format additive manufacturing. The screw mixing process turns to reduce the fiber length and will result in a fiber aspect ratio distribution in the extruded composite instead of a single value. Studies focusing on the geometric change of fibers have been done for years. Gupta, et al. [15] investigated the reduction of the fiber length for processing glass fiber reinforced polypropylene. Xia, et al. [16] studied the flexural stiffness of glass fiber filled composites fabricated through the injection molding process. Chin, et al. [17] predicted the elastic behavior of composite materials by taking the fiber length and orientation distribution into account. In a similar fashion, we will examine the effect of fiber aspect ratio distribution on the elastic moduli of a printed extrudate from a large scale additive manufacturing extruder.

The shear stress during the screw extrusion process will break the fiber fillers and yield a aspect ratio distribution with an asymmetric shape where a long tail propagates towards extreme long fiber aspect ratio. It is shown that a two-parameter Weibull probability distribution function was effective in depicting the density fiber length distribution when they were investigating glass fiber filled polypropylene [17]. Hence, we also apply a Weibull distribution function to describe the fiber aspect ratio distribution resulted from the extrusion process. The distribution function β is written as [18],

$$\beta(a_r) = ab{a_r}^{b-1} \exp(-a{a_r}^{b})\, for\ a_r > 0, \tag{6}$$

where a and b are scale and shape parameters. Furthermore, we can obtain the mean fiber aspect ratio (the number average fiber aspect ratio) by [19]

$$a_{r_{mean}} = \int_0^\infty a_r \beta(a_r) da_r = a^{-1/b}\Gamma(1/b\ + 1), \tag{7}$$

where Γ represents the gamma function [19].

And the mode fiber aspect ratio (most probable fiber aspect ratio) can be gained by setting the first derivative of $\beta(a_r)$ in Equation (2) to zero, yielding [19]

$$a_{r_{mode}} = \left(\frac{1}{a} - \frac{1}{ab}\right)^{1/b}, \tag{8}$$

We change the Weibull function by adjusting the mean fiber aspect ratio and the mode fiber aspect ratio. The mean fiber aspect ratio and mode fiber aspect ratio are prescribed so that the controlling parameters for the fiber aspect ratio distribution function can be solved through Equations (7) and (8). We first fix the mode fiber aspect ratio equates the mean, and assign a wide interval of the mean aspect ratios to see the variation of the Weibull function. Through the result shown in Figure 4, it is seen that as the mean fiber aspect ratio increasing, the peak probability decreases but more values get higher probability value. Besides, we also fix the mean aspect ratio and change the value of the mode in the vicinity of the mean value. The result shown in Figure 5 indicates that a larger difference between the mean and mode values yields in a more skewed profile while as two parameters are identity, a higher probability happened at the mean fiber aspect ratio. Also, it is found that as the mode aspect ratio propagates towards to the mean, the solved scale parameter (a) decreases while the shape parameter (b) increases (see Figure 6). When the mode value exceeds the mean, the parameter a becomes extremely close to zero and does not change in a noticeable way, which makes the curves under such conditions in little difference (see curves "$a_{r_{mode}} = 32$" and "$a_{r_{mode}} = 33$" in Figure 5).

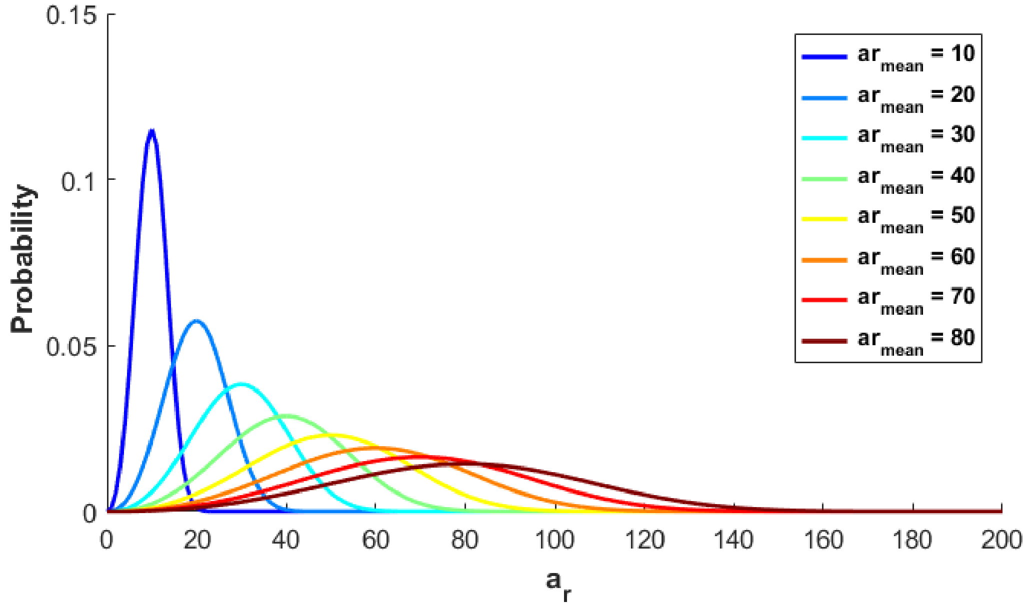

Figure 4. Weibull distribution functions defined by different mean fiber aspect ratios. Note the mode fiber aspect ratio is set to be equal to the mean fiber aspect ratio.

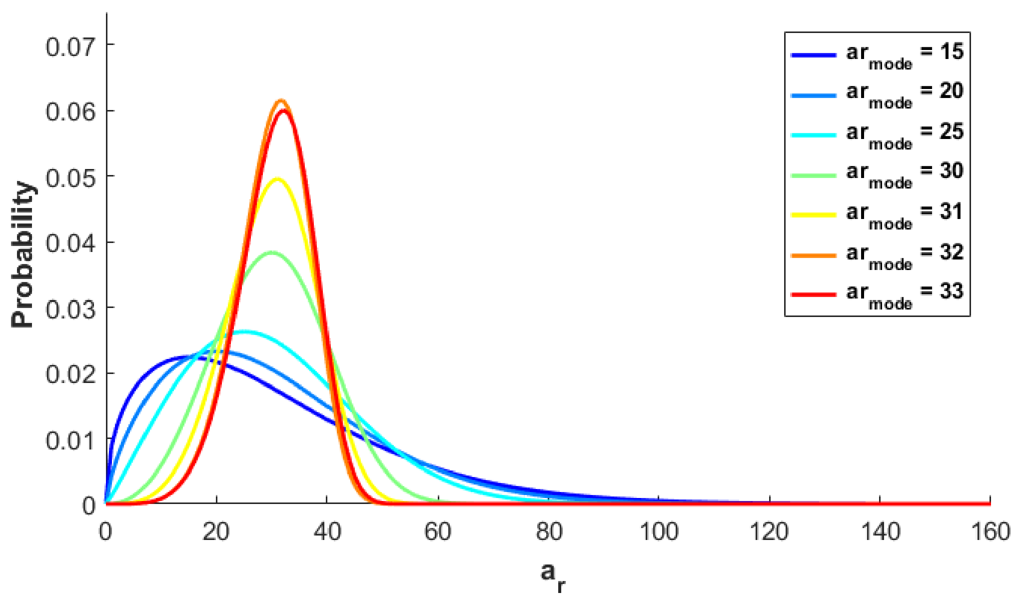

Figure 5. Weibull distribution functions defined by assigning different mode aspect ratio values. Note the mean fiber aspect ratio is fixed at 30.

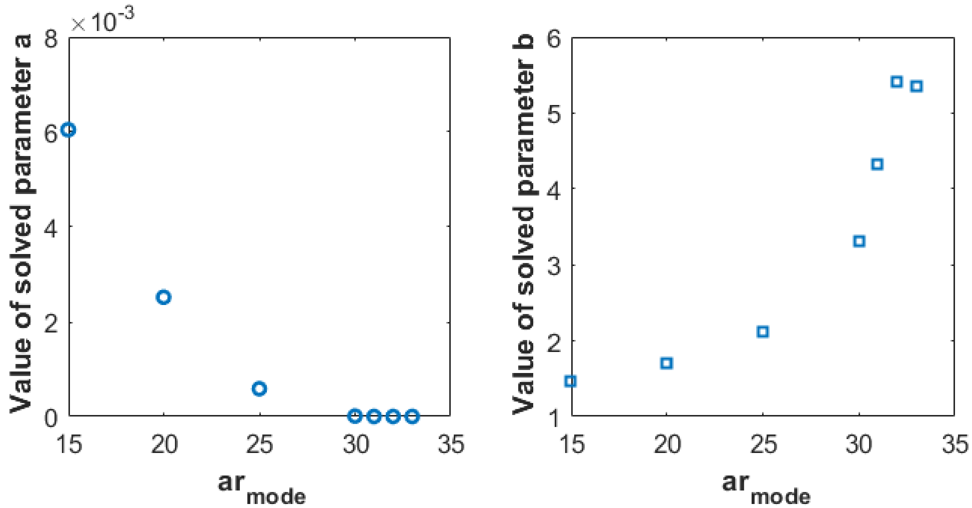

Figure 6. Scale and shape parameters of the Weibull distribution functions solved by mean and mode aspect ratio values through Equations (7) and (8).

Elastic Properties of Fiber Reinforced Composite

The stiffness of a unidirectional fiber reinforced composite with uniform fiber aspect ratio and isotropic phase materials were investigated through micromechanical modeling. Mori and Tanaka [20] proposed a method to evaluate the stiffness for non-dilute composite materials. Laws and McLaughlin [21] extended the self-consistent method initiated by Hill [22] and Budiansky [23] for the evaluation of short fiber reinforced composites. The Halpin-Tsai equations [24], which is derived from the method used for continuous fiber filled composites [22,25], are very popular in many numerical works of predicting elastic properties of discontinuous short fiber composites. Tandon and Wang [26] extended the Mori-Tanaka method and proposed a set of equations for computing the stiffness of unidirectional short fiber composites. Tucker and Liang [27] reviewed several of the popular micromechanical theories and concluded the estimations of the Mori-Tanaka methods were reasonable.

In this study, the Tandan-Wang formula is employed to first evaluate the stiffness of a unidirectional aligned fiber filled composite and the resulting stiffness tensors are written in contracted notations as C_{mn} (see [14] for detail). The elastic properties of the base materials are given through Table II. Note, E and G are the tensile and shear moduli. ν is the Poisson's ratio. And the fiber volume fraction is 13%. Then the orientation homogenization method [12,14] is applied to estimate the stiffness of a composite with a known fiber alignment pattern as,

$$C_{ijkl} = b_1 A_{ijkl} + b_2(A_{ij}\delta_{kl} + A_{kl}A\delta_{ij}) + b_3(A_{ik}\delta_{jl} + A_{il}\delta_{jk} + A_{jl}\delta_{ik} + A_{jk}\delta_{il}) + b_4\delta_{ij}\delta_{kl} + b_5(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}), \quad (9)$$

where the second and fourth order orientation tensors (A_{ij} and A_{ijkl}) are the solutions at the end of each streamline within the flow domain (see Figure 2), at which the extrudate is considered to reach the steady state. Note that, the fiber orientation state is solved by assuming a single value of fiber aspect ratio. Here, we only consider the effect of the variation of the aspect ratio on the stiffness of ultimate extruded fiber composites. It is acknowledged that the changing fiber aspect ratio may also be a critical role in evaluating the orientation, which, however, beyond the main scope of this paper. We would like to perform further investigation in the future to include this effect into consideration.

Besides, δ_{ij} is the kronecker delta tensor [28]. And b_i are evaluated as [12]

$$b_1 = C_{11} + C_{22} - 2C_{12} - 4C_{66}, \tag{10}$$

$$b_2 = C_{12} + C_{22}, \tag{11}$$

$$b_3 = C_{66} + 1/(2C_{22} + 2C_{23}), \tag{12}$$

$$b_4 = C_{23}, \tag{13}$$

$$b_5 = 1/(2C_{22} - 2C_{23}), \tag{14}$$

As mentioned, we assign a wide range of values to the fiber aspect ratios through the Weibull distribution function. Thus, for each aspect ratio input, we will have a unique C_{ijkl}. To taking the non-uniform fiber aspect ratios into consideration, the solutions of C_{ijkl} are numerically integrated over the given aspect ratio interval (theoretically, $0 < a_r < \infty$) as

$$\tilde{C}_{ijkl} = \int_0^\infty C_{ijkl}\,(a_r)\,\beta(a_r)da_r, \tag{15}$$

Furthermore, to assess the averaged elastic response of the overall extrudate, we then numerical integrate the obtained $\tilde{C}_{ijkl}$ over the cross section area of the extrudate as

$$\tilde{C}_{ijkl}^{avg} = \frac{1}{\pi r_0^2}\int_0^{2\pi}\int_0^{r_o}\tilde{C}_{ijkl}\cdot r\;drd\theta, \tag{16}$$

where r_o is the radius of the steady state extrudate. And r, θ directions correspond to the coordinate defined in Figure 2.

Table II: Elastic properties of the fiber and matrix of the composite material we evaluated [4].

Base materials elastic properties			
Material	E (GPa)	G (GPa)	ν
Carbon Fiber	230	96	0.2
ABS Matrix	2	1	0.35

Results

In this section, we examine the effects of the mean fiber aspect ratio and the mode fiber aspect ratio on the predicted averaged elastic constants of a printed extrudate separately, by assigning an interval of values to one of the parameter and fixing the other. Specifically, the mean aspect ratios are investigated through 10 to 200, and the mode aspect ratios are set from half of the mean value to the vicinity of the mean aspect ratio. Based on prior literatures [16,17], the ranges we specified is considered to be able to include the possible resulting fiber aspect ratio distributions from an extrusion process.

It is important to study the effects of the numerical integration methods on our predictions since our prediction significantly relies on the numerical integration theory. Hence, we first evaluate Equations (15) and (16) using both the trapezoidal rule and the 1/3 Simpson's rule [29]. Ultimately, the two methods yield results without a significant difference. Therefore, we choose to apply the 1/3 Simpson's rule through this study for simplicity.

To investigate the effect of the mean value on the predicted averaged elastic properties, we first assign a wide range of mean fiber aspect ratio and set the mode fiber aspect ratio equals to the mean value. Through the results given in Figure 7, it can be seen that elastic constants increase with an increasing mean fiber aspect ratio. Specifically, The tensile modulus along the extrusion direction (z axis, as defined in Figure 2) increases about 200% and the shear moduli also improved by a significant amount, as the mean aspect ratio increases from 10 to 200. Also, as the fiber aspect ratio increases, the increments of elastic constants reach each of their maximums.

Moreover, we fix the mean fiber aspect ratio and deviates the mode fiber aspect ratio to examine the effects of the mode value. The results shown in Figure 8 indicate that an increasing mode fiber aspect ratio also improves the stiffness of the overall extrudate composite. However, the improvement yielded by the increasing mode parameter is much less than that resulted by the increasing mean parameter.

Besides, we combine the effects of the mean and mode aspect ratios and plot the resulting elastic constants due to the variations of the two parameters, as shown in Figure 9 (For conciseness, four of the moduli are shown, the neglected two also show similar trend as shown in Figure 9). It is clear from the results that larger mean and mode fiber aspect ratios yields higher elastic moduli. In addition, stiffness resulted from large mean value and small mode value is slightly higher than that from small mean value and large mode value.

In addition, with a specific mean and mode aspect ratios (both equal 25), the averaged tensile modulus along the primary extrusion direction we predicted is 8.1 GPa. Using a similar carbon fiber composite system, where the base materials properties are close to what we used in Table II, some preceding literatures [4-6] has found the tensile modulus at printing direction is about 8~9 GPa. Though we are not able to modeling the entire experimental setup specifically, the overall favorable comparison between our numerical prediction and previously published experimental works supports the proposed computational approach.

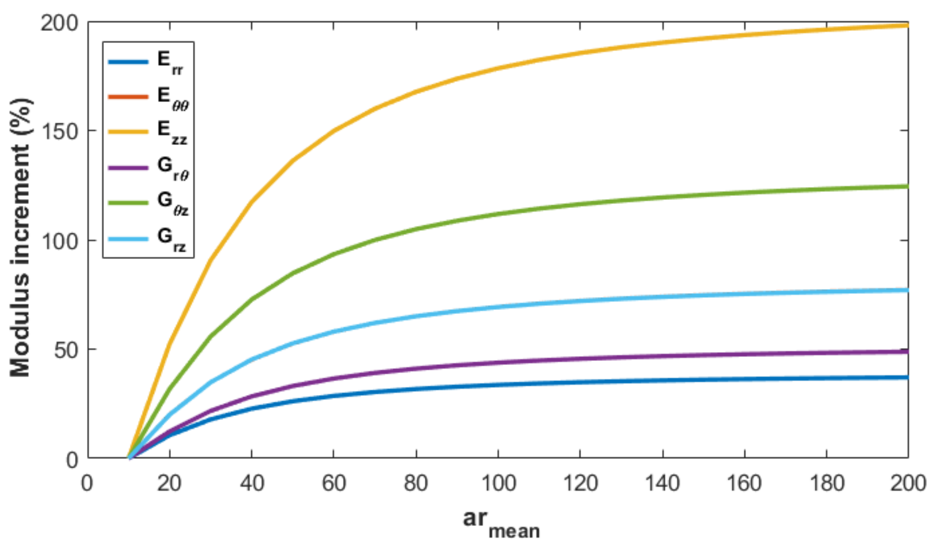

Figure 7. Increment of predicted averaged elastic properties at different mean fiber aspect ratios. Note the mode fiber aspect ratio is set to be equal to the mean fiber aspect ratio.

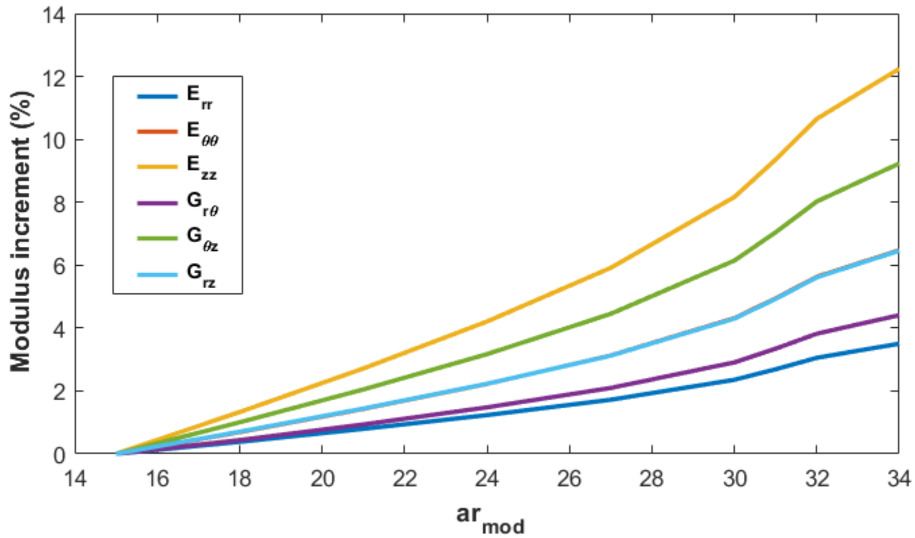

Figure 8. Increment of predicted averaged elastic properties at different mode fiber aspect ratios. Note, the mean aspect ratio is fixed at 30.

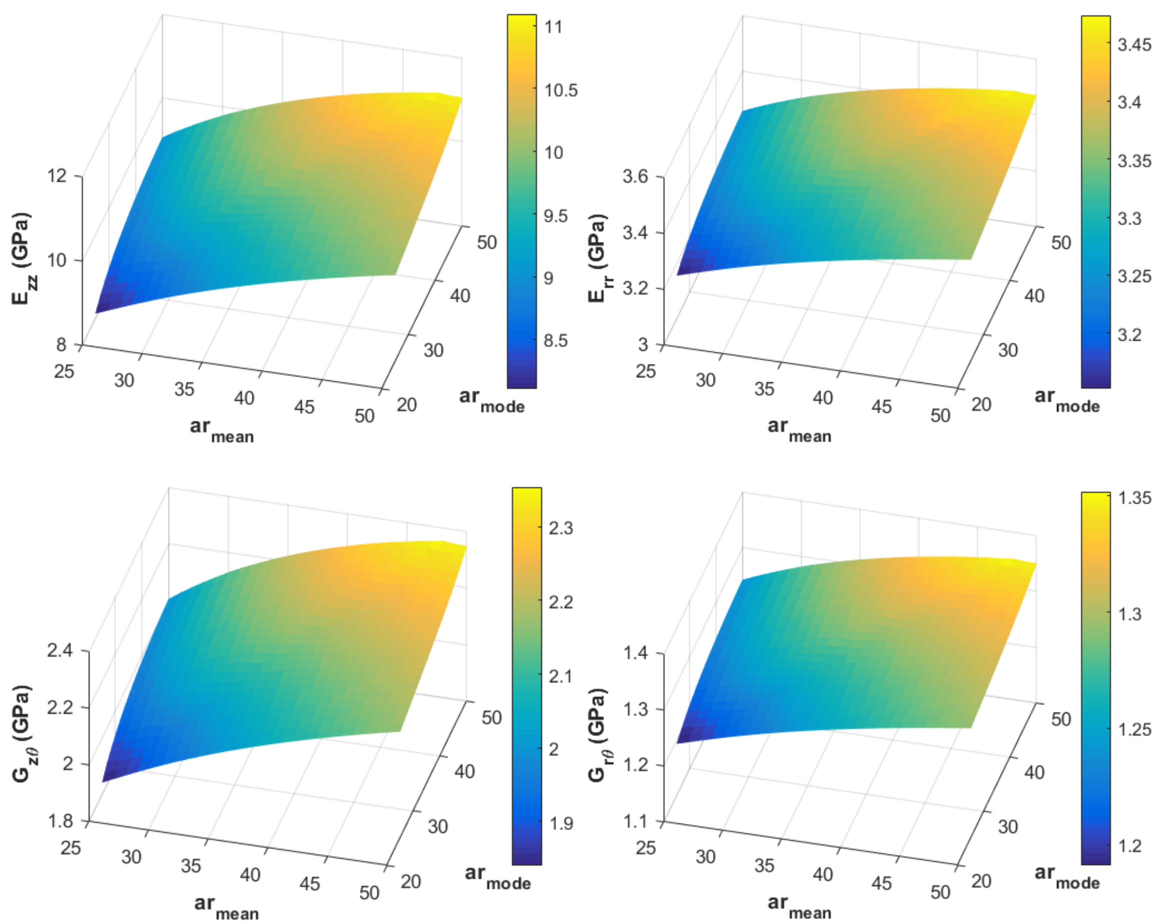

Figure 9. Effects of fiber aspect ratio distribution on the averaged elastic constants of an extrudate by combining the consideration of the mean and the mode fiber aspect ratio.

Summary and Next Steps

A two-parameter Weibull probability distribution function is implemented in estimating the fiber aspect ratio distribution of short fiber reinforced composite extrudate fabricated through the large scale polymer extrusion additive manufacturing system. The mean and mode fiber aspect ratio values are used to adjust the Weibull distribution function. Using the estimated fiber aspect ratio distribution, we evaluate the averaged elastic properties of a composite extrudate using the Tandon-Wang method and the orientation homogenization method with a pre-calculated orientation distribution. The computed results shown that the number average fiber aspect ratio (mean value) is shown to be a significant factor while the most probable fiber aspect ratio (mode value) contributes much less in predicting the elastic performances of a fiber reinforced composite extrudate.

Our current results indicate that it is safe to assume a single value of fiber aspect ratio in evaluating the stiffness of short fiber composite extruded by the additive manufacturing system. However, the neglected effects of the varying fiber aspect ratio during the extrusion flow modeling might play an important role in determining the fiber alignment within the extrudate. Therefore, the next step of this work is to taking the variation of the fiber aspect ratio during the modeling of the polymer melt and extrusion process.

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