# Fiber Orientation Kinetics of a Concentrated Short Glass Fiber Suspension in Startup of Simple Shear Flow

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## Abstract

The common approach for simulating the evolution of fiber orientation during flow in concentrated suspensions is to use an empirically modified form of Jeffery's equation referred to as the Folgar-Tucker (F-T) model. Direct measurements of fiber orientation were performed in the startup of shear flow for a 30 wt % short glass fiber-filled polybutylene terephthalate (PBT-30); a matrix that behaves similar to a Newtonian fluid. Comparison between predictions based on the F-T model and the experimental fiber orientation show that the model over predicts the rate of fiber reorientation. Rheological measurements of the stress growth functions show that the stress overshoot phenomenon approaches a steady state at a similar strain as the fiber microstructure, at roughly 50 units. However, fiber orientation measurements suggest that a steady state is not reached as the fiber orientation continues to slowly evolve, even up to 200 strain units. The

addition of a “slip” parameter to the F-T model improved the model predictions of the fiber orientation and rheological stress growth functions.

Key words: short glass fiber, fiber orientation, concentrated suspension, transient rheology

## 1. Introduction

Parts made from short glass fiber composite materials are typically produced in the melt state using injection or compression molding. During mold filling the macroscopic flow field generates a preferred orientation in the fiber microstructure which dramatically impacts the local mechanical, thermal and insulative properties of the part [1]. To optimize the mold design in relation to the desired part properties it is desirable to be able to simulate fiber orientation as a function of flow field and composite fluid rheological properties.

The first theoretical framework describing the evolution of orientation of axisymmetric particles that is easily extendable to fibers is the work of Jeffery [2]. Jeffery showed that the motion of a single ellipsoidal particle suspended in a Newtonian fluid in a Stokes flow field will rotate around the vorticity axis. Blunt-ended particles, such as rigid rods or fibers, follow similar orbits that one can predict using Jeffery’s equation by defining an equivalent aspect ratio. This theory can be applied to dilute suspensions ($\phi \ll a_r^{-2}$, where $\phi$ is the volume fraction) of fibers where forces other than the macroscopic flow field do not affect the particle dynamics [3, 4].

In the semidilute régime ($a_r^{-2} << \phi << a_r^{-1}$), inter-particle hydrodynamic interaction is the predominant phenomenon that can significantly affect or hinder periodic particle rotation [4]. Petrich et al. [5] studied the microstructure-stress relationship for various semidilute fiber suspensions in a Newtonian suspending medium using a concentric cylinder flow visualization apparatus and digital recorder. Comparison between their experimental results and dilute suspension theory showed good agreement for concentrations near the semidilute régime lower boundary. At concentrations in the semidilute régime near the upper boundary the period of rotation appeared to be influenced by inter-particle hydrodynamic interaction. In transient shear rheological measurements, shear stress oscillations have been observed and linked directly to the oscillating fiber orientation. However, the measured shear stress oscillations can dampen over time which is attributed to several interactions including boundary, particle-particle, hydrodynamic, or slight aspect ratio variations [6]. Other orientation states have been observed in and semidilute suspensions where fibers orient with the long axis in the vorticity and rotate referred to as "log-rolling", a phenomenon which is dependent on the viscoelastic properties of the suspending medium [7]. In general, as the shear stress approaches a steady state, a pseudo-equilibrium fiber orientation state is reached, which, after a short period of time, may slowly change with time.

In concentrated suspensions ($\phi > a_r^{-1}$) the distance between fibers is on the order of the fiber diameter or less, and multi-particle simulations show that under dynamic conditions fiber-fiber contact can severely affect the fiber motion [8, 9]. To our knowledge the dynamic behavior of fibers in a concentrated suspension has not been studied experimentally. However, rheological measurements in startup of flow suggest that fibers reorient themselves from their initial orientation state to align themselves in the direction of the fluid streamlines [10].

In the first attempts of simulating fiber orientation in injection molding, Jeffery's equation for infinitely long fibers was used [11]. Comparison between fiber orientation measurements of an injection molded part and simulation results suggested Jeffery's equation over predicted the degree of alignment and the shear strain at which steady state was reached. As a result Folgar and Tucker [12] modified Jeffery's theory to include a phenomenological term that prevented full alignment of fiber orientation, termed the Folgar-Tucker model (F-T). The F-T model improved the predictions of the steady state fiber orientation but had little effect on the strain at which the steady state orientation occurred [13]. As a result, Huynh [13] introduced the strain reduction factor using the argument that the fibers move in clusters and experience less strain than the bulk.

Model predictions of the rheological stress growth functions using the F-T model compared to experimental results suggest that the rate of fiber reorientation is much slower than theory predicts [14, 15]. This was attributed to fiber-fiber contact reducing the rate of fiber orientation. As a result Sepehr et al. [14] introduced a "slip" parameter to the F-T model, effectively reducing the rate of fiber reorientation. In simple shear flow this is exactly the same as the strain reduction factor of Huynh [13]. The addition of the slip parameter improved the model predictions of the transient stresses compared to the measured values. We do note, however, that the stress growth experiments used in the comparison were performed in a rotational rheometer with parallel disk geometry in which there is a varying shear rate from the center of the plates to the rim. Recent results have suggested that the inhomogeneous shear field in the parallel disk geometry induces excessive fiber-fiber interaction in concentrated fiber suspensions which can have a severe effect on the magnitude of the stress growth overshoot peak and width of the overshoot [16, 17].

The objective of this paper is to assess the ability of current theory for fiber orientation to predict the evolution of fiber orientation of a concentrated short glass fiber suspension of industrial significance in a well defined flow field. Donut shaped samples consisting of a 30 wt% short glass fiber filled polybutylene terephthalate (PBT-30) are subject to simple shear flow in a cone-and-plate device in which the sample geometry was designed to eliminate the interaction of the fiber with the plate walls [17]. At specific strains of interest in a shear stress growth vs. strain plot, the fiber orientation is determined experimentally using confocal laser microscopy. The experimental results are then compared to the F-T model with and without the addition of the slip parameter. The unique aspect of this work is the experimental confirmation of the fiber orientation evolution and its comparison to the predictions of theory for fiber orientation.

## 2. Theory

### *2.1. Orientation tensors*

The orientation of a single fiber can be described with a unit vector **u** along the fiber axis as shown in Fig. 1. The average orientation of a large number of fibers of similar length can be described using a distribution function, $\psi(\mathbf{u},t)$. A widely used and compact way to represent the average orientation state is with the second- and fourth-order orientation tensors which are defined as the second- and fourth-moments of the orientation distribution function [18],

$$\mathbf{A}(t) = \int \mathbf{uu}\,\psi(\mathbf{u},t)\,d\mathbf{u} \tag{1}$$

$$\mathbf{A}_4(t) = \int \mathbf{uuuu}\,\psi(\mathbf{u},t)\,d\mathbf{u} \tag{2}$$

The trace of **A** is always equal to 1 and for a completely random orientation state **A** = 1/3 **I**, where **I** is the unit tensor. In the limit that all the fibers are perfectly aligned in the $x_1$ direction the only non-zero component of **A** is $A_{11} = 1$.

*2.2. Evolution of fiber orientation*

The first theoretical work describing the evolution of high aspect ratio particle orientation that is easily extendable to rigid rods or fibers is that of Jeffery [2]. Jeffery's analysis can be written in terms of the second- and fourth-order orientation tensors as follows [18],

$$\frac{\mathrm{D}\mathbf{A}}{\mathrm{D}t} = \left(\mathbf{W}\cdot\mathbf{A} - \mathbf{A}\cdot\mathbf{W}\right) + \lambda\left(\mathbf{D}\cdot\mathbf{A} + \mathbf{A}\cdot\mathbf{D} - 2\mathbf{D}:\mathbf{A}_4\right) \tag{3}$$

where D/D$t$ is the material derivative, $\lambda$ is a shape function $\lambda = (a_r^2 - 1)/\,(a_r^2 + 1)$, $\mathbf{W} = [(\nabla \boldsymbol{v})^t - \nabla \boldsymbol{v}]/2$ is the vorticity tensor, $\mathbf{D} = [\,\nabla \boldsymbol{v} + (\nabla \boldsymbol{v})^t]/2$ is the rate of strain tensor and $\nabla \boldsymbol{v}$

$= \partial v_j/\partial x_i$. For fibers it is common to assume the particle's aspect ratio approaches infinity, in which case $\lambda \rightarrow 1$.

For non-dilute suspensions Folgar and Tucker [12] modified Eq. (3) to include a phenomenological term to account for fiber interaction preventing complete fiber alignment referred to in this article as the F-T model [18]:

$$\frac{\mathrm{D}\mathbf{A}}{\mathrm{D}t} = \left(\mathbf{W}\cdot\mathbf{A} - \mathbf{A}\cdot\mathbf{W}\right) + \left(\mathbf{D}\cdot\mathbf{A} + \mathbf{A}\cdot\mathbf{D} - 2\mathbf{D}:\mathbf{A}_4\right) + 2C_\mathrm{I}\dot{\gamma}\left(\mathbf{I} - 3\mathbf{A}\right) \qquad (4)$$

where $C_\mathrm{I}$ is a phenomenological parameter and $\dot{\gamma}$ is the scalar magnitude of **D**. The last term on the right hand side of the equation is very similar to the isotropic diffusivity term in theories for Brownian rods [19]. The slip coefficient introduced by Sepehr et al. [14], discussed previously, can be incorporated into the F-T model, which we refer to as the F-T-S model, as follows,

$$\frac{\mathrm{D}\mathbf{A}}{\mathrm{D}t} = \alpha\left[\left(\mathbf{W}\cdot\mathbf{A} - \mathbf{A}\cdot\mathbf{W}\right) + \left(\mathbf{D}\cdot\mathbf{A} + \mathbf{A}\cdot\mathbf{D} - 2\mathbf{D}:\mathbf{A}_4\right) + 2C_\mathrm{I}\dot{\gamma}\left(\mathbf{I} - 3\mathbf{A}\right)\right] \qquad (5)$$

where the slip coefficient, α, is some value between 0-1. The addition of the slip parameter to the equations governing fiber motion results in a loss of material objectivity of the equation, see for example Tanner [20]. However, the physical aspects of the model are still valid in the case of simple shear flow, which is the focus of this paper.

To solve equations Eqs. (3), (4) or (5) a closure approximation is needed to express the fourth-order tensor $\mathbf{A_4}$ in terms of **A**. Many closure approximations have been proposed including the quadratic [19], hybrid [18], eigenvalue- [21] and invariant-based optimal fitted [22] to name a few. A good review of their accuracy can be found in the references [23, 24]. For this work we use the invariant-based orthotropic fitted (IBOF) closure approximation established by Chung and Kwon [22] because of its stability, accuracy, and computational efficiency. The IBOF begins with the most general form of the fourth-order tensor $\mathbf{A_4}$ as follows,

$$\begin{aligned}\mathrm{A}_{\mathrm{ijkl}} = {} & \beta_1 S\left(I_{\mathrm{ij}} I_{\mathrm{kl}}\right) + \beta_2 S\left(I_{\mathrm{ij}} \mathrm{A}_{\mathrm{kl}}\right) + \beta_3 S\left(\mathrm{A}_{\mathrm{ij}} \mathrm{A}_{\mathrm{kl}}\right) + \beta_4 S\left(I_{\mathrm{ij}} \mathrm{A}_{\mathrm{km}} \mathrm{A}_{\mathrm{ml}}\right) + \\ & \beta_5 S\left(\mathrm{A}_{\mathrm{ij}} \mathrm{A}_{\mathrm{km}} \mathrm{A}_{\mathrm{ml}}\right) + \beta_6 S\left(\mathrm{A}_{\mathrm{im}} \mathrm{A}_{\mathrm{mj}} \mathrm{A}_{\mathrm{kn}} \mathrm{A}_{\mathrm{nl}}\right)\end{aligned} \tag{6}$$

where the operator $S$ indicates the symmetric part of its argument such as,

$$\begin{aligned}S\left(T_{\mathrm{ijkl}}\right) = \frac{1}{24}( & T_{\mathrm{ijkl}} + T_{\mathrm{jikl}} + T_{\mathrm{ijlk}} + T_{\mathrm{jilk}} + T_{\mathrm{klij}} + T_{\mathrm{lkij}} + T_{\mathrm{klji}} + T_{\mathrm{lkji}} + \\ & T_{\mathrm{ikjl}} + T_{\mathrm{kijl}} + T_{\mathrm{iklj}} + T_{\mathrm{kilj}} + T_{\mathrm{jlik}} + T_{\mathrm{ljik}} + T_{\mathrm{jlki}} + T_{\mathrm{ljki}} + \\ & T_{\mathrm{iljk}} + T_{\mathrm{lijk}} + T_{\mathrm{ilkj}} + T_{\mathrm{likj}} + T_{\mathrm{jkil}} + T_{\mathrm{kjil}} + T_{\mathrm{jkli}} + T_{\mathrm{kjli}})\end{aligned} \tag{7}$$

The IBOF assumes that the coefficients $\beta_1$ to $\beta_6$ are polynomial expansions of the second and third invariants of **A**. We use the fifth-order polynomial for which the polynomial coefficients can be found elsewhere [22].

*2.3. Stress*

Lipscomb et al. [25] proposed a stress equation for a dilute suspension of high aspect ratio particles following the work of Hand[23] and Giesekus[21] as,

$$\boldsymbol{\sigma} = -p\mathbf{I} + 2\eta_s\mathbf{D} + 2c_1\phi\eta_s\mathbf{D} + 2\phi\eta_s N\mathbf{D} : \mathbf{A}_4 \tag{8}$$

where $\boldsymbol{\sigma}$ is the total stress, $p$ is pressure, $\eta_s$ the suspending medium viscosity, $c_1$ a constant, and $N$ a dimensionless parameter that represents the coupling between hydrodynamic stress contribution and the fiber orientation. Lipscomb et al. give $c_1$ to be equal to 2 for dilute suspensions, but we choose to use it as a fit parameter. Also, Lipscomb et al. give $N$ to be a function of the particle aspect ratio. Semidilute suspension theories such as the Dinh and Armstrong [26], Shaqfeh and Fredrickson [27], or Phan-Thien and Graham [28] can also be written in the form of Eq. (8) noting that some assume the third term is negligible or $c_1 = 0$. These theories focus on an expression for $N$ considering inter-particle hydrodynamics. For predicting the rheological behavior of concentrated suspensions $N$ is frequently determined by fitting predictions to experimental results and this is the approach we use in this work. For the startup of shear flow, the shear stress growth coefficient, $\eta^+$, and first normal stress difference growth function, $N_1^+$, can be determined from Eq. (8) for simple shear flow kinematics ($v_1 = \dot{\gamma} y$ and $v_2 = v_3 = 0$) as,

$$\eta^+ = \sigma_{12} / \dot{\gamma} = \eta_s + c_1\eta_s\phi + 2\eta_s\phi N \mathrm{A}_{1212} \tag{9}$$

$$N_1^+ = 2\phi\eta_s\dot{\gamma}N\left(\mathrm{A}_{1211} - \mathrm{A}_{1222}\right) \tag{10}$$

where we define $x_1$ as the flow direction, $x_2$ as the direction of velocity gradient and $x_3$ as the neutral direction.

## 3. Experimental

### *3.1. Materials*

For this work a commercially available 30 wt% (volume fraction, $\phi$ = 0.1766) short glass fiber filled polybutylene terephthalate (PBT-30) provided by GE Plastics under the trade name Valox 420 was used. Linear viscoelastic measurements confirmed that the matrix was Newtonian-like, the details of which have been published elsewhere [29]. To examine the effect of fiber concentration on the rheological behavior, PBT-30 was diluted to concentrations of 4.07, 8.42, 15, 20 and 25 wt%. Compounding was accomplished by passing dry blended amounts of PBT-30 and the neat matrix through the extruder section of an Arburg Alrounder 221-55-250 injection molder at an rpm of 200. The extrudate was collected before entering the runner of the mold and pelletized. The pellets were then compression molded for rheological testing to a cone-and-plate disk at 260 $^{o}$C. Precautions were taken to minimize the degree of thermo-oxidative degradation of the PBT matrix by drying the materials at 120 $^{o}$C for a minimum of 12 hours in a vacuum oven at a pressure smaller than 0.4 in.Hg before sample extrusion, molding or testing [30].

To characterize the glass fiber within the suspension, pyrolysis was performed on the PBT-30 pellets at 500 $^{o}$C after extrusion to separate the fibers from the matrix. The average fiber length was determined by measuring the length of 1,000 randomly selected fibers. The number average and weight average fiber length of PBT-30 was found to be $L_n$ = 0.3640 and $L_w$ = 0.4388 mm, respectively, with a standard deviation of 0.17 mm. The same fiber length measurement was performed on all the diluted concentrations and was found to be within $0.3640 \leq L_n \leq 0.3740$ and $0.4388 \leq L_w \leq 0.4578$ mm. The fiber diameter, $D$, was determined directly from images taken of fiber cross sections using a confocal laser microscope, discussed later, and the average diameter of 1,000 fibers was found to be $D$ = 12.9 μm with a standard deviation of 1.7 μm. This relates to a number average aspect ratio for PBT-30 of $a_r \cong 28.2$.

*3.2. Rheological measurements*

All rheological measurements were performed on a Rheometrics Mechanical Spectrometer (RMS-800) at 260 $^{o}$C. To minimize the degree of thermo-oxidative degradation, all experiments were performed in a nitrogen environment with a freshly loaded pre-formed sample. Rheological measurements on the PBT matrix were performed with a 25 mm cone-and-plate fixture with 0.1 radian cone angle.

The common approach to characterizing the rheological behavior of short glass fiber filled polymer melts is to use a rotational rheometer with parallel disk geometry set to a gap where measurements are independent of gap height. However, recent results suggest that the inhomogeneous velocity gradient imposed by the parallel disk geometry can lead to exaggerated measurements of the rheological material functions [17]. As a result, it was of primary importance to perform the experiments on the glass fiber-filled suspensions in a rheometer that imposes a homogeneous shear field such as the cone-and-plate geometry [31]. For our measurements a 50 mm diameter cone-and-plate fixture was fabricated and used. The cone had a 0.1 radian cone angle and was truncated to allow for a gap of 50 μm at the center. To minimize the degree of fiber-boundary interaction Blankeney [32] and Attanasio [33] suggested that the rheometer gap be at least three times the length of the longest dimension of the suspension particle. Experimental results using parallel plate geometry with various rheometer gaps confirmed that for the PBT-30 there is a negligible effect on the stress growth and steady state rheological behavior when the gap is roughly three times the number average fiber length [17]. The gap within the 50 mm cone and plate fixture varies linearly from 2.51 mm at the outer edge to 0.05 mm at the center. To remove the excessive fiber-boundary interaction near the center, a 25.4 mm diameter hole was drilled through the center of the pre-formed disks creating a donut shaped sample which we refer to as "donut" samples. A schematic drawing of the donut sample can be seen in Fig. 2. After each experiment the void space at the center was measured to account for sample loading as the gap is squeezed to proper dimensions. The hole diameter varied slightly, 23.8 ± 0.5 mm and was accounted for in calculating the stresses for each run. A comprehensive analysis and description of the donut sample can be found elsewhere [16, 17].

For measurements performed using the cone and plate fixtures, $\eta^+$ and $N_1^+$ were calculated as functions of torque, $M(t)$, and normal force, $F(t)$, from [34],

$$\eta^{+}(t)=\sigma^{+}/\dot{\gamma}=\frac{3M(t)}{2\pi\dot{\gamma}}\left(R_{\mathrm{o}}^{3}-R_{\mathrm{i}}^{3}\right)^{-1} \tag{11}$$

$$N_{1}^{+}(t)=\frac{2F_{\mathrm{z}}(t)}{\pi}\left(R_{\mathrm{o}}^{2}-R_{\mathrm{i}}^{2}\right)^{-1} \tag{12}$$

where $R_o$ and $R_i$ are the outer and inner radius, respectively. The experimental reproducibility was found to be ± 5% for $\eta^+$ and ± 7% for $N_1^+$.

*3.3Measurement of fiber orientation*

The fiber microstructure was characterized using confocal laser microscopy in a similar approach to that proposed by Lee et al. [35]. Donut samples composed of PBT-30 were sheared using the RMS-800 at $\dot{\gamma}$ = 1 $s^{-1}$ for a specified time relating to predetermined strain, $\gamma = \dot{\gamma} t$. Immediately after deformation, and taking precaution not to disturb the sample melt, the temperature was lowered below the melt temperature "locking-in" the flow generated fiber microstructure.

The samples were prepared by sanding embedded sections of the donut sample in epoxy to a specific plane depth, and then polishing the surface to a final abrasive particle size of 0.3 μm aluminum oxide ($Al_2O_3$) following standardized techniques [36]. Sample images were taken at four possible locations denoted by PD-1 through 4 and are depicted in Fig. 3 for clarification. Locations PD-1, PD-2 and PD-3 were located in the plane perpendicular to the φ-direction (flow-direction) at distances of 4.0, 6.25, and 8.5 mm, respectively, from the outer edge, and PD-4 was located in the plane perpendicular to the r-direction (neutral-direction) at a depth of 4.0 mm. PD-1 and PD-4 can be considered mutually perpendicular planes of different sections of the donut sample.

In total, the fiber orientation was measured for 11 samples which where deformed to strains of 0, 4, 7, 9, 12, 25, 50, 100 and 200. Samples deformed to strains 4, 25, 100 and 200 were imaged at locations PD-1 and PD-4, all other samples were imaged at locations PD-1 thru PD-4 as depicted in Fig. 3. These experiments were designed to highlight structural features that correlated to the transient stresses. For instance, the fiber orientation at strains 4, 7, 9 and 12 were meant to characterize the fiber orientation during the overshoot region; 7 and 9 relate to the peaks of the $\eta^+$ and $N_1^+$ overshoot, respectively. The onset of steady state for $\eta^+$ and $N_1^+$ was characterized by strains 25 and 50, respectively, and the plateau region in which the stresses did not exhibit large changes by strains 100 and 200. A sample loaded into the rheometer but not sheared, strain 0, represents the initial fiber orientation within the rheological samples.

The images were taken using a Zeiss LSM510 confocal laser scanning microscope fitted with a 40x water immersion objective lens and a laser excitation wavelength of 543 nm. The final image was 230 x 230 µm with a resolution of 1024 x 1024 pixels. For each sample, sequential images were taken from the bottom to the top in the direction of the velocity gradient and at two planes of depth. In the image the cross section of each fiber appeared as circles or ellipse-like shapes. To process the image, the circumference of each fiber intersection was traced by hand in power point to improve the contrast between the fibers and the matrix and converted to a binary image. A computer program was written in combination with image analysis software in Matlab that measured the position of center of mass, the major and minor axis and local angle between a fixed coordinate frame and the major axis of the ellipse. The components of **u** for each fiber were determined from the elliptical "footprint" at two cross sectional planes.

With knowledge of **u** for each fiber the tensor, **A** can be constructed as follows:

$$\mathrm{A_{ij}} = \frac{\sum \left(u_\mathrm{i} u_\mathrm{j}\right)_\mathrm{n} F_\mathrm{n}}{\sum F_\mathrm{n}}, \quad F_\mathrm{n} = \frac{M_\mathrm{n}}{m_\mathrm{n}} \tag{13}$$

where $F_n$ is a weighting factor, $M_n$ is the major axis, and $m_n$ is the minor axis for the n$^{th}$ fiber [37]. The weighting function is based on the probability of a 2D plane intersecting the n$^{th}$ fiber. Meaning, a fiber aligned perpendicular to the plane is more likely to be severed than one aligned parallel. Using the weighting function, the larger the aspect ratio of the ellipse, the more that fiber is weighted. In the results and discussion section the orientation tensor **A** will be used to describe the average orientation state of the system. The reproducibility of the $A_{ij}$ component between different samples was found to be dependent on the magnitude of the $A_{ij}$ component with the maximum error being ± 12.4 % for the component of smallest magnitude, the $A_{22}$.

## 4. Experimental results and discussion

In the first part of this section we show the stress growth behavior of PBT containing various concentrations of glass fiber to establish the interest in understanding the dynamic behavior of the PBT suspension containing the greatest fiber concentration, PBT-30, subject to startup of flow. The focus of this section is to define the technique used to characterize the orientation of the glass fiber within the rheometrical samples consisting of PBT-30 during startup. A key aspect to our experimental scheme lies in the assumption that the fiber orientation is maintained during stress relaxation. To convince the reader that the fiber orientation does not change during stress relaxation and over quiescent periods we present interrupted stress growth/relaxation results. This is followed by a mathematical verification of the degree to which Brownian motion and sedimentation contribute to the change in fiber orientation. Finally, we discuss the technique used to measure the fiber orientation and briefly present the results. Further discussion of the experimental results for fiber orientation will be presented in the subsequent section on modeling of the fiber orientation.

*4.1. Transient rheology*

The stress growth behavior of the PBT containing various concentrations of fibers can be seen in Figs. 4 (a) and (b) for the shear stress growth coefficient, $\eta^+$, and the first normal stress difference growth function, $N_1^+$, respectively. In Fig. 4 (a) the 30 wt% exhibits a typical $\eta^+$ of a concentrated short glass fiber-filled fluid. Initially, there is a large transient overshoot that reaches a maximum in approximately 7 strain units which decays towards a steady state. It is difficult to define exactly when a steady state is reached, but the majority of the overshoot occurs within 25 and 30 strain units and the stresses appear to approach a steady state at roughly 50 strain units. In comparing $\eta^+$ exhibited by the different concentrations, the magnitude of the overshoot decreases with decreasing fiber concentration. For the 4.07 and 8.42 wt% $\eta^+$ decays from the beginning of flow toward a steady state and, as a result, there is no discernable overshoot. A fully developed overshoot appears to occur around 15 to 20 wt%.

$N_1^+$ is plotted vs. strain in Fig. 4 (b) for the various concentrations. Similar to $\eta^+$, $N_1^+$ for the 30 wt% exhibits a large initial overshoot that reaches a steady state around 50 strain units. The overshoot peak is more than 8 times the steady state value. All the concentrations tested exhibited an overshoot which increased with increasing fiber concentration. However, there seemed to be a broadening of the overshoot and a significant increase in the magnitude that occurred at 15 wt%. The dramatic increase in the magnitude and broadening of the overshoot is believed to be a result of significant fiber contact at the higher concentrations. All but the lowest concentration is in the concentrated régime, but stress growth behavior of the 30 wt% exhibited the most dramatic behavior. Furthermore, for this work we were interested in studying the dynamic behavior of a concentrated suspension of industrial significance. As a result, the 30 wt% was chosen for the fiber orientation experiments.

During sample preparation for the fiber orientation measurements, it was assumed that the fibers maintained their orientation during stress relaxation. To reinforce this assumption, interrupted stress growth experiments were performed after the overshoot region and at the peak of the $N_1^+$ overshoot, depicted in Figs. 5 (a) and (b), respectively. In Fig. 5 (a) the results of two interrupted tests on two different samples can be seen. Initially both samples were deformed for 100 s at a $\dot{\gamma}$ = 1 $s^{-1}$, after which the flow was stopped. For the first sample, the flow was reapplied after 50 s, and for the second the flow was reapplied after 200 seconds. In both cases the stresses grew to their previous value within the time resolution of the experiment, 0.5 s, and independent of the time between flows. This test was performed up to a maximum wait time of 1,000 s and confirms that the overshoot region is not recoverable over span of time relevant to this work. A similar interrupted stress growth test was performed except the flow was removed at the peak of the $N_1^+$ overshoot and can be seen in Fig. 5 (b). When the flow was reapplied the stresses immediately grew to their previous values. The behavior exhibited by PBT-30 in both Figs. 5 (a) and (b) suggests that the fibers do maintain their orientation during stress relaxation. If this assumption is correct, then even at very long times the fiber orientation should remain constant under static conditions if unaffected by external forces such as Brownian motion or gravity.

*4.2. Impact of Brownian motion and gravity*

The relative effect of Brownian motion on the orientation of the fibers within the suspension can be determined by considering the ratio of the experimental shear rate, $\dot{\gamma}$, to the rotational diffusion constant, $D_r$, also known as the Péclet number, Pé = $\dot{\gamma} / D_r$. For a fiber $D_r = 3k_bT[\ln(a_r)-0.8]/\pi\eta_s L^3$, where $k_b$ is Boltzmann's constant, T is the temperature in Kelvin, $\eta_s$ is the viscosity of the suspending medium and L is the fiber length [19]. For PBT-30 Pé ≈ $10^{13}$, and therefore, can be considered non-Brownian [38]. To estimate the relative effect of gravity on fiber orientation Chaouche and Koch [39] proposed an expression to estimate the relative time scale for sedimentation, $t_s$, the time required for a fiber parallel to the vertical direction to sediment over its length, as $t_s = 8\eta_s L/\Delta g\rho d^2[\ln(2a_r)-0.72]$. For an average fiber in PBT-30 $t_s \approx 45$ hrs and represents the minimum estimated time for a fiber to settle as fiber contact would act to increase $t_s$.

*4.3. Fiber orientation measurements*

The orientation of each fiber can be described in spherical coordinates using a unit vector, **u**, along the fiber axis depicted in Fig. 1. The zenith and azimuthal angles (θ and φ, respectively) defining the components of **u** for a each fiber can be calculated from the ratio of the minor and major axis of the elliptical footprint using Eq. (14) and the other angle can be measured directly from the image [35]. In Eq. (14) the subscript 1-2 refers to the $x_1 - x_2$ plane.

$$\cos\theta\Big|_{1-2} = \frac{m}{M} \tag{14}$$

Figure 6 (a) depicts the elliptical footprint in the $x_1 - x_2$ plane and the angle φ measured directly from the image. The limitation of calculating the angle based on Eq. (14) is that the angle is always between 0 and π causing an inherent ambiguity. For example, in the $x_1 - x_2$ plane it is impossible to distinguish between a fiber that is oriented at ($\theta_1$, $\varphi_1$) or ($\pi - \theta_1$, $\varphi_1$) because their cross sections are identical. Figure 6 (b) depicts two possible unit vectors **u** for the same elliptical footprint.

To increase the accuracy of the 3D description of fiber orientation and reduce the inherent ambiguity in the method previously described, imaging was performed using a confocal laser microscope. This allowed for images to be taken at multiple depths parallel to the plane of interest and fully describe the 3D fiber orientation without ambiguity. Figure 6 (b) illustrates a fiber cross section with two possible unit vectors whose orientation is the mirror image of each other. When only one plane is analyzed it is impossible to determine the correct vector **u**. However, when images from two depths are compared, the center of mass displacement confirms the appropriate **u** vector. This is slightly different from the work of Lee et al. [35] who calculated the angle based on the measured displacement.

After processing the images relating to the experimental data, it was found that change in center of mass of the two elliptical footprints was only detectable when the aspect ratio of the ellipse was roughly greater than three or at an angle of greater than roughly $50^{o}$ due to the limited depth of penetration of the laser. For the PBT-30 the maximum penetration was found to be 8 μm. For this reason we term our analysis "pseudo-3D" in that there is still a certain amount of ambiguity with the calculated angle for fibers which were mostly aligned in the direction perpendicular to the plane of interest.

For the geometric analysis defined above to be valid it is assumed that the fibers are perfectly rigid circular cylinders. Forgacs and Mason [40] developed an equation to estimate the critical shear stress at which the shear-induced axial compression can cause a rotating fiber to buckle, $\tau_{\mathrm{critical}} = E_f[\ln(2a_r)-1.75]/2a_r^4$, where $E_f$ is the flexural modulus. Assuming $E_f \approx 73$ GPa for the glass fibers within the PBT-30 suspension [41], then $\tau_{\mathrm{critical}} \approx 1.3\mathrm{e}10^5$ Pa, and they can be considered rigid at $\dot{\gamma} = 1\ \mathrm{s}^{-1}$.

*4.4. Fiber orientation*

In the literature it is a common practice to assume the initial fiber orientation within rotational rheometer samples is, on average, random [14, 42]. As the initial orientation assumed in making the calculations can drastically influence the predictions, we directly measured the initial fiber orientation of the samples subjected to the startup of shear flow. The initial measured fiber orientation of the donut samples, represented using the orientation tensor **A** at strain 0 is shown in Eq. (15),

$$\mathbf{A}\big|_{\gamma=0} = \begin{pmatrix} 0.517 & 0.075 & 0.058 \\ 0.075 & 0.035 & 0.035 \\ 0.058 & 0.035 & 0.448 \end{pmatrix} \tag{15}$$

The experimental data represented in Eq. (15) shows that the majority of the fibers were initially oriented in the $x_1$ and $x_3$ directions with very few aligned in the $x_2$ direction. This is attributed to the deformation history given to the sample during compression molding of the sample disk and while it is loaded into the rheometer.

The experimentally determined $A_{ii}$ components as a function of strain can be seen in Fig. 7 (a). The $A_{11}$ component increases with increasing strain while the $A_{33}$ component decays at a similar rate. This shows that the fibers, whose initial orientation is mostly in the $x_1$ - $x_3$ plane, reorient to align in the $x_1$ direction. The rate of fiber reorientation is fastest between strains 0 and 50; after a strain of 50 the rate dramatically decreases and relates to the onset of steady state exhibited by the $N_1^+$ overshoot. The growth rate of the $A_{11}$ component is nearly identical to the decay rate of the $A_{33}$ component. Mathematically this growth/decay behavior can be described over the range of $\gamma$ using a slow logarithmic function of the form $A_{ii} = A_{ii\_0}$ +/- $\kappa$ *ln($\gamma$) where $A_{ii\_0}$ is $A_{ii}$ component at

$\gamma$ = 0, $\kappa$ is a constant, and the + or – is for growth or decay behavior as in $A_{11}$ and $A_{33}$, respectively. The slow logarithmic function was fit to both $A_{11}$ and $A_{33}$ over the entire data set leading to a single function constant $\kappa$ = 0.06 with an $R^2$ = 0.86 and can be seen in Fig. 7 (a). If you predict when the fiber would reach full alignment ($A_{11}$ = 1) using this equation leads to a strain of 3150. It is difficult to draw an exact conclusion based on this general function because there are multiple solutions within the error bars of the data set. However, on average, the data suggest that the fiber orientation is still evolving even up to 200 strain units.

In contrast to the $A_{11}$ and $A_{33}$ components, $A_{22}$ exhibits an initial increase that decays slightly to what appears to be a statistical steady. This behavior is more readily quantified by normalizing the fiber orientation at the various strains by the initial orientation, $A_{22\_0}/A_{22}$, which can be seen in Fig. 7 (b) along with $A_{12\_0}/A_{12}$. The $A_{22}$ and $A_{12}$ components increase to 1.8 and 2.8 times the initial value, respectively, but the peak occurs at a strain of 4 for $A_{12}$ and a strain of 7 for $A_{22}$. In addition, the apparent steady states occur at the different strains of 12 and 25 for $A_{22}$ and $A_{12}$, respectively, which correlates to the onset of steady state in $\eta^+$.

## 5. Simulations

### *5.1. Numerical method*

Before making a comparison between experimental and predicted values of fiber orientation, a brief discussion of the numerical solution method is given. In all models that contained $\mathbf{A_4}$, the IBOF closure approximation was used to express $\mathbf{A_4}$ in terms of $\mathbf{A}$. Equations (4) and (5) were solved numerically using Gear's implicit predictor-corrector method at a time step of 0.01 s. Model predictions were found to be independent of time step at smaller increments. The initial conditions and model parameters are discussed later.

### *5.2. Effect of $C_I$ and α*

In Fig. 8 the effect of fiber interaction and slip through the parameters $C_I$ and α, respectively, on the predictions of the F-T and F-T-S models from an initial random orientation are shown. For increasing fiber interaction the steady state value of $A_{11}$ decreases, but has a negligible effect on the initial growth rate of $A_{11}$ for the values of $C_I$ of practical interest, $C_I < 0.01$. Predictions of the F-T model with a small degree of interaction, $C_I = 0.0001$, are within 1% of the that predicted by Jeffery's equation, Eq. (3), in the limit of $\lambda = 1$. In contrast to the effect of $C_I$ on the F-T model predictions, the addition of slip as in the F-T-S model results in a decrease in the growth rate of the $A_{11}$ component but has no effect on the steady state asymptote. The parameters $C_I$ and α allow one to independently adjust the steady state fiber orientation and the rate of fiber reorientation as predicted by Eq. (5).

### *5.3. Model predictions*

In Fig. 9 the experimentally determined $A_{ii}$ components vs. strain are compared to the F-T model with small fiber interaction $C_I = 0.0001$, and $C_I = 0.006$ which was determined by fitting the steady state model predictions to the experimental data at a strain of 200. The F-T model with small interaction drastically over predicts the $A_{ii}$ components rate of reorientation. The F-T model with $C_I = 0.006$ also over predicts the $A_{ii}$ components reorientation rate at small strains but approaches the value at large strains. Furthermore, the F-T model with $C_I = 0.006$ reaches a steady state in ~ 50 strain units, but as previously discussed the experimental data suggests that the fiber orientation is still evolving even up to the largest strain measured. The predictions of the $A_{ii}$ components using the F-T model shown in Fig. 9 are relatively accurate at the largest strain which is expected as the $C_I$ was parameter was adjusted to fit the F-T model predictions at that strain. However, the continuously evolving fiber orientation suggests that a correct value would be smaller than $C_I = 0.006$.

Model predictions of the F-T-S model compared to the experimentally determined $A_{ii}$ components are shown in Fig. 10. The slip parameter $\alpha$ was determined by fitting the F-T-S predictions, with $C_I = 0.006$, to the experimental $A_{11}$ and $A_{33}$ components over the whole strain range and was found to be $\alpha = 0.3$. Values of $\alpha < 0.3$ resulted in a loss of accuracy between the F-T-S model predictions and the experimental data at small strains, $\gamma < 25$, for all values of $C_I$. For comparison purposes the F-T-S model with $C_I = 0.0001$, and $\alpha = 0.3$ was included in the figure. Predictions of the F-T-S model with small $C_I$ shows good agreement with the $A_{11}$ and $A_{33}$ components at small strains but over predicts the degree of orientation at large strains. The F-T-S model with $C_I = 0.006$ shows good agreement with the $A_{11}$ and $A_{33}$ components for all strains tested. However, at a strain

150 the predictions reach a steady state. The F-T-S model predictions of the $A_{22}$ does not show good agreement at small strains, $\gamma < 25$. Experimentally, the $A_{22}$ component increases from strains 0 to 4 where it reaches a maximum and then decreases. The F-T-S model with small $C_I$ predicts the $A_{22}$ component to decrease from its initial orientation state while the F-T-S model with small $C_I = 0.006$ predicts the $A_{22}$ component to increase the steady state value without exhibiting an overshoot. It is not exactly clear why the experimental $A_{22}$ component increases and goes through a maximum, similar to an overshoot, but one possible contributing factor could be direct fiber contact. The overshoot in the $A_{22}$ component occurs at small strains when there is a high degree of fiber reorientation. We believe the inability of the F-T-S model to predict the overshoot in the $A_{22}$ component is a deficiency in the physical predictions of the model and not a result of the IBOF closure approximation. This was confirmed by performing a stochastic simulation of Eq. (5) in which no closure approximation was needed, see, for example Fan et al. [43]. The stochastic simulation of the $A_{22}$ component did not exhibit an overshoot for the range of $C_I < 0.01$ tested.

F-T-S model predictions of the $A_{12}$ component compared to the experimental values can be seen in Fig. 11. Similar to the $A_{22}$ component there is a rise in the values of $A_{12}$ from $\gamma = 0$, passing through a maximum at $\gamma = 7$ and then decaying slowly with increasing strain. This is in contrast to the predicted $A_{12}$ component which decays from the initial value. However, when the simulations begin from the experimental fiber orientation at a strain of 7, given in Eq. (16), the predictions show good agreement with the rate of decay. After which the experimental $A_{12}$ component appears to reach a steady state of greater magnitude than the predicted value.

$$\mathbf{A}\big|_{\gamma=7} = \begin{pmatrix} 0.546 & 0.138 & -0.040 \\ 0.138 & 0.079 & -0.005 \\ -0.040 & -0.005 & 0.375 \end{pmatrix} \tag{16}$$

We now compare the predictions of the stress growth functions $\eta^+$ and $N_1^+$ vs. strain by way of Eqs. (9) and (10), in combination with the F-T-S model for fiber orientation to the experimental results. The stresses are directly linked to the fiber orientation through the hydrodynamic drag term in Eq. (8). In a similar manner to the $A_{12}$ component, the F-T-S model did not predict an overshoot in the $A_{1212}$ component. As a result the predicted $\eta^+$ did not exhibit an overshoot when the simulations of the stresses began from strain 0. For this reason the model predictions of $\eta^+$ and $N_1^+$ discussed subsequently begin from a strain of 7 using the experimental fiber orientation at that strain as the initial conditions. For the F-T-S model the same parameter values of $C_I$ and α that were used in the simulations of the fiber orientation are used to simulate the stresses, i.e., $C_I = 0.0001$ and 0.006 and $\alpha = 0.3$. The model parameters $c_1$ and $N$ were determined through a visual best fit of the predicted $\eta^+$ and $N_1^+$ to the measured values of $\eta^+$ and $N_1^+$. The best fit parameter values using the F-T-S with $C_I = 0.0001$ and $C_I = 0.006$ were found to be $c_1 = 1.5$, $N = 35$, and $c_1 = 0$, $N = 35$, respectively.

The comparison of the experimental and predicted values of $\eta^+$ can be seen in Fig. 12 (a). Eq. (9) in combination with the F-T-S model with small $C_I$ produced the best fit; the model shows good agreement compared to the experimental results over the entire range of strain. In contrast, we were unable to fit the predictions of Eq. (9) in combination with the F-T-S model with $C_I = 0.006$ to both the overshoot maximum and the steady state values simultaneously. The experimental and predicted $N_1^+$ vs. strain can be seen in Fig. 12 (b). For both values of $C_I$ = 0.0001 and 0.006 in which the F-T-S model was used to predict the fiber orientation we were unable to fit the magnitude of the overshoot region. It is not directly clear why the model was unable to predict the magnitude of the overshoot. However, one plausible explanation is the Lipscomb stress equation does not account for direct fiber contact which could potentially contribute to the stresses.

## 6. Conclusions

The common approach to predicting the evolution of fiber orientation during simulation of processing flows such as injection molding is to use an empirically modified form of Jeffery's equation, the F-T model. Direct measurement of fiber orientation in startup of flow of a concentrated composite fluid shows that the F-T model over predicts the rate of fiber reorientation. The addition of the slip parameter to the F-T model, which acted to slow the rate of fiber reorientation, improved the model predictions at all the strains examined. A linear fit to the $A_{11}$ and $A_{33}$ components of **A** at strains of 50, 100, and 200 suggest that the fiber orientation is still evolving even after 200 strain units where the F-T and F-T-S models predict a steady state has been reached. It is unclear at what strain the fibers would reach a steady state, but the fiber orientation might continue to evolve to very large strains.

In startup of flow, the experimental $A_{22}$ and $A_{12}$ components exhibited an overshoot behavior. The F-T and F-T-S models were unable to predict this phenomenon and we attribute it to fiber contact as the fiber reorients from a mostly planar orientation to align itself in the flow direction. As a result of the F-T-S model's inability to predict the increase in orientation in the $x_2$ direction, the simulations of $\eta^+$ beginning from the experimentally determined initial conditions did not predict the $\eta^+$ overshoot. However, simulations of the stresses beginning from a strain of 7, the peak of the $\eta^+$ overshoot, showed a qualitative agreement with the overshoot decay from a strain of 7.

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**Figure Captions**

**Figure 1**. Unit vector **u** describing the orientation state of a rod in spherical coordinates.

**Figure 2.** Schematic drawing and cross-sectional profile of the donut sample.

**Figure 17.** Schematic drawing of the polished and imaged planes perpendicular to the neutral, r, and flow, φ, directions. Images were taken at four positions: three positions perpendicular to the flow-direction at a distance of 4.0, 6.25, and 8.5 mm from the outer edge denoted by PD-1, PD-2, and PD-3, respectively, and one position perpendicular to the neutral direction denoted by PD-4. At all locations images were taken through the entire height of the sample.

**Figure 18.** Stress growth behavior of PBT and PBT containing various concentrations of short glass fiber at $\dot{\gamma}$ = 1 $s^{-1}$. (a) Shear stress growth coefficient, $\eta^+$, (b) first normal stress growth, $N_1^+$, vs. strain. The symbols (x), (Δ), (-), (o), (◊) and (■) represent fiber wt% 4.07, 8.42, 15, 20, 25 and 30, respectively. The solid line depicts the neat PBT for comparison in both graphs.

**Figure 19.** $\sigma^+$ and $N_1^+$ vs. strain, $\gamma$, for PBT-30 in interrupted flow experiments. All flow was performed at $\dot{\gamma}$ = 1 $s^{-1}$. (a) Two interrupted experiments were performed on two different samples. In one experiment the flow was removed for 50 s and in the other experiment flow was removed for 200 s. (b) An interrupted flow experiment where a fresh sample was subject to flow at $\dot{\gamma}$ = 1 $s^{-1}$ for 10 s, the flow was removed for 20 s, and then the flow was reapplied.

**Figure 20.** (a) Elliptical "footprint" of fiber cross section where *M* is the major axis and *m* is the minor axis. (b) Two possible unit vectors, **u**, along the backbone of the fiber and depiction of the positive angle θ.

**Figure 21.** The evolution of the fiber orientation determined experimentally during startup of flow represented through the $A_{ij}$ components. (a) The $A_{ii}$ components; the solid lines represent a slow logarithmic function fit to the $A_{11}$ and $A_{33}$ components. (b) The $A_{12}$ and $A_{22}$ components normalized by the initial orientation $A_{12\_0}$ and $A_{22\_0}$, respectively.

**Figure 22.** Effect of the slip constant, α, and the Folgar-Tucker constant, $C_I$, on $A_{11}$ as predicted by the Folgar-Tucker (F-T) model, and the Folgar-Tucker model with the addition of a slip term (F-T-S). The solid lines represent the F-T model, Eq. (4), and the broken lines represent the F-T-S model, Eq. (5). Decreasing α refers to values α = 0.75, 0.5 and 0.25, and increasing $C_I$ refers to values of $C_I$ = 0.001, 0.005 and 0.01. The bold line represents the F-T model with $C_I$ = 0.0001. All simulations were performed at a $\dot{\gamma}$ = 1 $s^{-1}$.

**Figure 23.** Experimental and predicted fiber orientation represented through the $A_{ii}$ components in startup of simple shear flow at $\dot{\gamma}$ = 1 $s^{-1}$. The lines represent the

predictions of the Folgar-Tucker (F-T) model model with $C_I = 0.0001$ (dashed line) and $C_I = 0.006$ (solid line).

**Figure 24.** Experimental and predicted fiber orientation represented through the $A_{ii}$ components in startup of simple shear flow at $\dot{\gamma} = 1\ s^{-1}$. The lines represent the predictions of the Folgar-Tucker model with the addition of a slip term (F-T-S) with $\alpha = 0.3$, $C_I = 0.0001$ (dashed line) and $C_I = 0.006$ (solid line).

**Figure 25.** Experimental and predicted fiber orientation represented through the $A_{12}$ component in startup of simple shear flow at $\dot{\gamma} = 1\ s^{-1}$. The lines represent the predictions of the Folgar-Tucker model with the addition of a slip term (F-T-S) with $C_I = 0.0001$ (dashed line) and $C_I = 0.006$ (solid line). Predictions were performed using experimentally determined initial conditions (IC) at a strain of 0 and 7 shown as IC $\gamma = 0$, and IC $\gamma = 7$, respectively, in the figure.

**Figure 26.** Experimental and predicted stress growth functions using the Folgar-Tucker model with the addition of a slip term (F-T-S) in startup of simple shear flow at $\dot{\gamma} = 1\ s^{-1}$. The dashed and solid line depict model predictions for $C_I = 0.0001$ and 0.006, and $\eta^+$ and $N_1^+$ were produced by means of Eqs. (9) and (10), respectively. (a) $\eta^+$, (b) $N_1^+$ vs. strain.