Influence of fiber concentration on the startup of shear flow behavior of long fiber suspensions

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Abstract

In order to use rheological measurements as a tool to investigate fiber orientation in simple flows, the relationship between stress and fiber orientation must be understood. In this work, a sliding plate rheometer was used to measure the shear stress growth during the startup of simple shear flow of a polymer melt containing long glass fibers. The concentrations of the suspensions were varied from 10 to 40 wt% and tested over three shear rates spanning an order of magnitude. Significant shear thinning was observed in the suspension as concentration increased. Additionally, the magnitude of stress and breadth of the stress growth overshoot increased with concentration. A larger distinction between the different concentrations is observed in the shear stress growth than the measured evolution of fiber orientation. Measured values of fiber orientation were used with a semi-dilute stress equation to show that the fiber motion in these experiments was not responsible for the stress overshoot and that additional stress contributions must be considered.

1 Introduction

The enhancement of tensile, flexural and impact properties for a discontinuous fiber reinforced composite is largely dictated by the concentration, orientation and length of the fibers within the part. Tensile, flexural and impact strengths have been measured to reach a maximum near 40 wt% glass fiber content in polypropylene for injection molded composites [1]. Additionally, the moduli of these parts were found to increase linearly with concentration. Flexural, creep and impact properties have been shown to increase with fiber length, while tensile properties will plateau for glass fibers above 1 mm in length [2-5]. Fibers less than the 1 mm threshold have been considered short while fibers with lengths greater than 1 mm are

considered long. Furthermore, the degree of this mechanical enhancement can vary within a molded part depending on the underlying fiber microstructure [6, 7].

Concentration regimes for fiber composites stem from theories of rod-like molecules provided by Doi and Edwards [8]. In terms of volume fraction, ϕ , and fiber aspect ratio ($a_r =$ fiber length/diameter), fiber suspensions can be divided into dilute ($\phi < a_r^{-2}$), semi-dilute ($a_r^{-2} < \phi < a_r^{-1}$), and concentrated ($\phi > a_r^{-1}$) regimes. Generally, composites of commercial interest have $\phi > 0.1$ [9]. In the concentrated regime, the motion of an individual fiber is restricted by surrounding fibers through hydrodynamic and fiber-fiber interactions. As ϕ or a_r increases, the motion of an individual fiber will become more constrained. Additionally, Bay and Tucker [10] noted that glass fibers with aspect ratios above 100 will exhibit flow induced curvature, which may also impact fiber motion.

Fiber orientation equations for concentrated fiber suspensions often rely on phenomenological adaptations to Jeffery's equation describing the motion of an ellipsoidal particle [11]. Folgar and Tucker [12] introduced isotropic rotary diffusion to Jeffery's model to account for the randomizing effect on orientation due to fiber interactions. The rotary diffusion constant in the Folgar-Tucker model has been empirically represented as a function of the product of aspect ratio and concentration by Bay [13] and Phan-Thien et al. [14]. Ranganathan and Advani [15] represented the diffusion constant as a function of the interparticle spacing. Including the interparticle spacing allows the diffusion constant to depend on the state of fiber orientation. Phelps and Tucker [16] accounted for orientation dependent particle diffusion through the use of a diffusivity tensor in the anisotropic rotary diffusion (ARD) orientation

model. These approaches in modelling fiber orientation require that material parameters be determined experimentally.

A stress equation provides the coupling between fiber orientation and the equations of momentum and energy. A general form of the stress tensor is provided by Hinch and Leal [17] for ellipsoidal particles. For high aspect ratio fibers, Lipscomb et al. [18] provide the parameters in the stress equation for dilute suspensions. Stress in semi-dilute suspensions is prescribed by Dinh and Armstrong [19] and Shaqfeh and Fredrickson [20]. The material constants in the general stress equation have been empirically adjusted to fit experimental data for concentrated suspensions, due to lack of available theory [21-23].

To understand the orientation kinetics of fiber suspensions, rheological experiments have been performed during the startup of flow. Fibers subjected to flow will orient with the primary flow direction creating a stress growth overshoot that will reduce to a steady value. As concentration increases the magnitude of the stress growth overshoot and steady state has been shown to also increase [24, 25]. The addition of fibers will also cause shear thinning to occur at low rates when no shear thinning is observed in the matrix. Ortmann et al. [26] observed a higher degree of shear thinning to occur in addition to a broadened shear stress overshoot as fiber aspect ratio increased.

Previous investigations of the rheology of fiber suspensions have shown a monotonic increase in stress with concentration. Laun [24] and Bibbo [25] measured the shear stress during the startup of flow for short fiber suspensions and observed that the maximum stress occurred at the same shear strain for concentrations ranging from the semi-dilute to concentrated regimes. Eberle et al. [27] observed the shear stress to decay from an initial value to a steady state for

concentrations less than 10 wt% short glass fiber in a Newtonian matrix. Above 10 wt%, a stress growth overshoot is observed and the strain where the maximum stress occurred increased with increasing concentration. This shift in strain for the location of maximum stress was also observed by Ortman et al. [22, 26] for long glass fiber suspensions at 10 and 30 wt%. However, these works were unable to conclude how the changes in the transient response reflected changes in fiber orientation.

Measurements of stress from a rheometer provide an indirect assessment of the underlying fiber microstructure. As a result, some works have experimentally measured fiber orientation in addition to stress. Petrich et al. [28] and Keshtkar et al. [29] used flow visualization experiments to observe the orientation of the fibers during flow. Eberle et al. [30] and Ortman et al. [22] used thermoplastic matrices that were allowed to cool within the rheometer to preserve fiber orientation. The sample was removed from the rheometer and orientation was measured from a polished cross section of the sample using an optical microscope. This method requires multiple samples to be used to assess fiber orientation as a function of time and the assumption must be made that there is little variability between each sample. Both of these approaches have shown that the rate of fiber orientation will decrease with increasing fiber length.

The purpose of this work is to further understand the relationship of stress and the evolution of fiber orientation during the startup of simple shear flow for long fiber suspensions. The methods of Ortman et al. [22] were used as a guide to investigate the effects of concentration on the orientation and stress response of high aspect ratio glass fiber suspensions. We aim to show that the shape of the stress response and fiber orientation will change with concentration which is different than what has been observed for short fiber suspensions. The contribution to

stress that is dependent on fiber orientation was investigated using a model valid for semi-dilute suspensions. The fourth order term in the stress equation was calculated from experimental measurements of fiber orientation which allows for the assessment of the stress model without inducing additional error caused by approximating the fourth order term.

2 Theory

The orientation of a single rigid fiber can be denoted by a unit vector, $\mathbf{p}$, parallel to the axis of the fiber. The average orientation for a population of fibers can be compactly represented through the use of even-ordered structural tensors [31]. The second order orientation tensor is defined as the product of the orientation distribution function, $\psi(\mathbf{p})$, with the dyadic product of the orientation vector integrated over all orientation space:

$$\mathbf{A} = \int \mathbf{p}\mathbf{p} \psi(\mathbf{p}) d\mathbf{p} . \quad (1)$$

In order to assess orientation through measured values of stress within a rheometer, a stress theory encompassing orientation is used. Shaqfeh and Fredrickson [20] applied a multiple scattering expansion to the slender-body theory to quantify the hydrodynamic effects from adjacent fibers on the bulk stress, $\boldsymbol{\sigma}$:

$$\boldsymbol{\sigma} = -P\mathbf{I} + 2\mathbf{D}\eta_m + \frac{4\eta_m\phi\alpha_r^2}{3[\ln(1/\phi) + \ln\ln(1/\phi) + C]}\mathbf{D} : \langle \mathbf{p}\mathbf{p}\mathbf{p}\mathbf{p} \rangle \quad (2)$$

where, P is the isotropic pressure, $\mathbf{I}$ is the identity tensor, η_m is the matrix viscosity, the rate of deformation tensor, $\mathbf{D} = (\nabla\mathbf{v}' + \nabla\mathbf{v})/2$ and $\langle \mathbf{p}\mathbf{p}\mathbf{p}\mathbf{p} \rangle$ denotes the fourth moment of orientation for a population of fibers. The parameter C is dependent on the particle shape and orientation. For cylindrical particles, C has a value of -0.6634 for random three-dimensional orientation and

0.1585 for completely aligned fibers. This stress equation takes into account stress contributions from the isotropic pressure, matrix and the tension imposed on an inextensible rod. In simple shear flow the maximum tension occurs when a fiber is oriented 45° in the flow and gradient directions. The theory of Shaqfeh and Fredrickson has shown good agreement with numerical simulations of Mackaplow and Shaqfeh [32] and the experimental data of Bibbo [25].

The use of a semi-dilute stress equation with the concentrated data presented in this work is intended to provide a comparison between orientation and stress without introducing any empirical parameters. The slender-body theory should reflect the fiber interactions in semi-dilute suspensions up to a concentration of $\phi \approx \pi/20a_r$. It is expected that the stress equation will under predict data above this limiting concentration [32].

3 Experimental

The suspensions under investigation were 10, 20, 30 and 40 wt% glass fibers in a low viscosity polypropylene matrix. The material was received from SABIC as 13 mm long pellets created through a pultrusion process in 30 and 50 wt% formulations (Verton MV006S-GYLTNAT). The pellets contain a bundle of fibers that must be dispersed in order to obtain repeatable rheological data. Each formulation was processed in a 1-inch, 20L/D Killion extruder (KLB-100) equipped with a gradually tapering screw. The temperature profile along the length of the screw was set to 220, 230 and 245 °C with a die temperature set to 150 °C. The circular die measured 1.5 mm in diameter. Concentrations less than 30 wt% were diluted with pure polypropylene, while 30 and 50 wt% pellets were used to create the 40 wt% suspension. Suspensions at 50 wt% were found to exceed the stress limitations of the rheometer and were not studied as a result. Extruded strands were pelletized to a length of 15 mm which were used to

compression mold samples with approximately planar random orientation. The mold dimensions were 250 by 50 mm and sample thickness was between 1.6 to 1.7 mm. Samples were compression molded for 5 minutes in a preheated mold at 180 °C. This procedure was found to minimize voids which could greatly impact the measurements of stress.

Measurements of fiber length were obtained from the 15 mm pellets used to create the samples for rheological testing. Pellets were placed in an oven at 500 °C for 2 hours to remove the polypropylene matrix. Loose glass fibers were then dispersed on a glass slide and imaged on a desktop scanner at 1200 dpi resolution. At least 1500 fibers per sample were manually measured using ImageJ software. The fiber length distribution followed the typical log-normal distribution commonly observed for fiber suspensions. Three samples were used to produce the averages in Table 1. The average fiber diameter was measured to be 13.3 μm using an optical microscope at 20x magnification. The number average (L_n) and weight average (L_w) fiber lengths were calculated according to Eq. (3) and (4), respectively:

$$L_n = \frac{\sum_i N_i L_i}{\sum_i N_i} \quad (3)$$

$$L_w = \frac{\sum_i N_i L_i^2}{\sum_i N_i L_i} \quad (4)$$

where, N_i is the number of fibers within the distribution with length L_i . The sampling method for fiber length measurements from Kunc et al. [33] was used to confirm that no reduction in fiber length occurred as a result of compression molding or rheological testing. The average lengths are above the 1 mm threshold and can be considered as long fiber suspensions. However, the

number average fiber lengths are close to 1 mm which is due to a significant number of short fibers in the length distribution.

Rheological tests on the fiber suspensions were performed using a sliding plate rheometer made in-house based on the design of Giacomini [34] and further discussed by Giacomini and coworkers [35]. Details on the development and implementation of this particular sliding plate rheometer can be found elsewhere [36]. The rheometer was enclosed in a forced convection oven manufactured by Russells Technical Products (model RB-2-340). The sliding plate was controlled through the use of an Instron-4204 universal testing instrument. A shear stress transducer was flush mounted to the stationary plate. The shear stress transducer contains a lever arm suspended by a diaphragm which determines the instrument's range and sensitivity. Two diaphragms were used to cover the range of stresses measured in this work. The deflection of the lever arm was measured using a capacitance probe (Capacitex, Model HPT-75I-V-T-2-B). Signals from the probe were amplified using a Capacitex 4100-SL-BNC amplifier and interfaced with a computer using a National Instruments data acquisition device (USB-6008). Voltage readings were recorded using National Instruments Labview SignalExpress software (version 2.5.1). Shear stress was determined based on the measured voltage and calibration of the diaphragm in the shear stress transducer.

The rheometer was heated to a temperature of 200 °C for 2 hours prior to sample loading. A sample was placed between the two heated plates and the plates were tightened enough to touch both surfaces of the sample. After heating for 5 minutes the plates were tightened to the final thickness of 1.5 mm. Testing the suspensions at greater gap thicknesses confirms that the 1.5 mm gap thickness did not greatly influence the stress response. In addition, Ortman et al. [26] used the same gap thickness for similar fiber suspensions and concluded that gap effects

were minimized due to the mostly planar fiber orientation. The sample remains in the rheometer 15 minutes prior to testing. Shear rates of 0.4, 1.0 or 4.0 s⁻¹ were imposed on the sample for 120 strain units. Five replicates were averaged to produce the reported stress growth profiles. A similar heating cycle and duration was imposed on the polypropylene matrix and tested using a cone-and-plate rheometer (Rheometrics RMS-800). Results concluded that the flow behavior of the matrix did not change during the time frame that tests were performed using the sliding plate. Upon completion of the tests in the sliding plate rheometer, a fan was used to cool the rheometer to room temperature over the duration of 1 hour. The sample remained within the rheometer during this time period to preserve fiber orientation for later investigation.

Measurements of fiber orientation were made to further investigate its relationship to the measured stress response. Samples were prepared and tested according to the same procedure but sheared to different strain units. Separate samples were used for each strain. Therefore, it was assumed that the initial conditions for each sample were the same. Orientation measurements were taken along the x_1x_2 -midplane, such that x_1 denotes the flow direction with the velocity gradient in x_2 . Details of the development of the sample preparation procedure can be found in the works of Vélez-García et al. [37] and Hofmann et al. [38]. Samples were polished using modified metallographic techniques and oxygen plasma etched to enhance the contrast of the glass fiber and polypropylene matrix. Images were obtained at 20x magnification on an optical microscope with a motorized stage and image-stitching software (Nikon Eclipse LV100, NIS-Elements Basic Research Software, version 3.10). Hofmann et al. [38] suggested that the image analysis width be modified to 5.5 mm for fiber suspensions with a number average length of 3.9 mm. This analysis width was found to be acceptable for all concentrations investigated here. The two-dimensional projections of the fibers on the imaging plane are

displayed as ellipses. Based on the ratio of the minor to major axes of the ellipse and the in-plane angle, the orientation tensor, $\mathbf{A}$, can be reconstructed for a population of fibers [10, 39]. A weighting function developed by Bay and Tucker [10] was used to correct for the sampling bias in orientation measurements imparted from the use of an inspection plane. This method is applicable for suspensions that do not exhibit significant curvature.

Additional images of fiber orientation were obtained in the x_2x_3 -plane to observe the extent to which fibers were bending. Very few fibers were found to exhibit any bending in the x_1x_2 or x_2x_3 inspection planes. Obtaining fiber orientation from elliptical cross sections cannot assess if fibers bend during flow and straighten upon the cessation of flow. Keshtkar et al. [29] denoted the orientation of bent fibers with an end-to-end vector, but did not quantify the extent of fiber bending. Bending of fibers may significantly influence flow behavior and orientation kinetics. A two-rod system from Strautins and Latz [40] and Ortman et al. [22] would allow for the degree of fiber bending to be quantified and the average orientation of a fiber could be prescribed by an end-to-end vector. In the limit that a fiber has no curvature, the orientation of the end-to-end vector is equivalent to the orientation of the rods. Hofmann [7] was able to show good agreement between fiber orientation measurements obtained from elliptical cross sections and measurements of the fibers' end-to-end vectors. For the stress and fiber orientation presented in this work, the designation of a rigid fiber will be used due to the large amount of short fibers that exist in the suspensions. However, we recognize that the two-rod system may provide a more complete description of fiber orientation dynamics. This approach may be necessary to describe longer fibers in the suspension (e.g. fiber lengths above 5 mm). Although there are relatively few fibers with these large fiber lengths in the suspensions studied in this

work, the impact these fibers have on the bulk stress and the orientation behavior of the system is of continued interest.

4 Results and Discussion

Rheological data and orientation evolution have been obtained for concentrations of 10, 20, 30 and 40 wt% glass fibers at shear rates of 0.4, 1.0 and 4.0 s⁻¹. Error bars in Fig. (1) represent the standard deviation amongst the five samples averaged to produce the reported stress profiles. We attribute this error to the localized measurements of stress and localized variations in fiber orientation within the sample. The pellets used to compression mold the samples have some preferential alignment. Additional error may arise from the unspecified A_{12} orientation component due to the method used to prepare the samples. The extent to which a fiber must rotate to align with the flow direction is dependent on whether A_{12} is positive or negative.

Concentration effects on the shear stress growth are shown in Fig. 1(a) for a shear rate of 0.4 s⁻¹. An overshoot is observed for all concentrations tested, but is not observed in pure polypropylene. Within the strain limitations of the sliding plate rheometer, all samples appear to have reached a steady shear viscosity which infers a steady state fiber orientation. The presence of fibers greatly increases the fluid viscosity such that an order of magnitude increase is observed at 30 wt% compared to the pure matrix. The stress growth overshoot appears to occur at later strains when concentration is increased. This shift in the maximum stress may indicate that fiber orientation evolves slower for a more concentrated system. Laun [24] did not observe any change in strain for the maximum stress as concentration was increased for glass fibers with an aspect ratio of 22. The aspect ratios presented in this work are greater than Laun's by at least a

factor of 4. Additionally, Laun observed the magnitude of the stress overshoot and steady state to increase with concentration up to 35 wt%. The suspensions tested in this work exhibit an increase in the magnitude of the overshoot and steady state values for concentrations of 10 to 30 wt%. At 40 wt%, the magnitude of the stress overshoot is smaller than what might be expected based on the trends observed at 10 to 30 wt%. In comparing the stress overshoot to the steady state value, the relative overshoot at 40 wt% is less than the relative overshoots at 20 and 30 wt%. This may indicate that the orientation dynamics are different for the 40 wt% suspension when compared to the other concentrations.

Figure 1(b) shows the same materials tested at a higher shear rate of 4.0 s^{-1} . The trends discussed at 0.4 s^{-1} can be extended to this higher shear rate. It should be noted that the viscosity is substantially dependent on shear rate. The difference in magnitude for the 40 wt% data compared to the other concentrations is not as pronounced at 4.0 s^{-1} . The maximum value of stress for 40 wt% glass fibers still occurs at larger strains than the other concentrations, but the breadth of the overshoot is not nearly as large as what was measured at 0.4 s^{-1} . Additionally, the stress growth overshoot relative to the steady state appears to scale with concentration. Data obtained at 1.0 s^{-1} (Fig. 6) confirms the trends observed in the data presented in Fig. 1. The broad overshoot profile observed at 40 wt% in Fig. 1(a) appears to be unique to that data set and not observed to such an extent at 1.0 s^{-1} .

Figure 2 shows transient viscosity data for the polypropylene matrix and 10 and 40 wt% glass fiber suspensions at the three tested shear rates. The polypropylene exhibits no measurable overshoot or significant shear thinning within the range of tested shear rates. For the 10 wt% glass fiber suspension, shear thinning is observed along with a stress growth overshoot. Figure 2(b) contains the stress growth response for the 40 wt% glass fiber suspension at the three shear

rates. The transients take longer to reach a maximum and steady state than the lower concentrations. The 0.4 s^{-1} data does exhibit a different profile than the higher shear rate data but the steady state values do appear to show a power-law dependency with shear rate.

To further view the shear thinning effects, the steady shear viscosity for all the suspensions is plotted with the matrix viscosity in Fig. 3 as a function of shear rate. The polypropylene does not show significant thinning behavior until rates are at least an order of magnitude above what was tested in the sliding plate rheometer. As the fiber concentration increases, the intensity of shear thinning also increases. This is believed to be related to the amount of microstructure that must be rearranged to necessitate flow. Larson [41] provides an possible explanation to this shear thinning behavior, in that shear rates are high enough to disturb the interparticle spacings from equilibrium.

It can be inferred that the steady shear viscosity of these fiber suspensions will not show a significant plateau before thinning of the matrix is encountered at higher rates. It is interesting to note that the shear thinning behavior at a concentration of 40 wt% approaches that of the polypropylene matrix at high shear rates. The data is insufficient to make any conclusions on the behavior of the suspensions at high shear rates. However, Thomasset et al. [21] observed the same degree of shear thinning to occur for fiber suspensions as the polypropylene matrix for similar fiber aspect ratios presented in this work.

The suspending fluid appears to be Newtonian in the region that has been investigated for the fiber suspensions, making the motion and interactions of the glass fibers responsible for the non-Newtonian response. The second moment of orientation tensor, $\mathbf{A}$, is constructed based on the measured orientation of for population fibers. The evolution of the trace components of the

orientation tensor are shown in Fig. 4. Due to the uniform flow within the sliding plate rheometer, a large image analysis width produces data with little variance. Differences in fiber orientation for the 10 and 40 wt% fiber suspensions appear to be evident in fiber orientation data. The orientation profiles for 20 and 30 wt% are indistinguishable, and their values may be considered an average of the 10 and 40 wt% data. Initial conditions were measured to vary by less than 5% between all concentrations, with fibers slightly more oriented in the flow direction than the neutral direction. The A_{22} component of orientation is small indicating mostly planar orientation. The largest value of A_{22} occurs at the initial conditions and reduces in magnitude under flow. Little difference is observed in the A_{22} component of orientation with concentration.

The A_{11} and A_{33} components exhibit a mirrored profile, because A_{22} is small due to mostly planar fiber orientation. The 10 wt% fiber systems orient in the flow direction and reach a steady state. The 40 wt% suspension comparatively shows a delayed response in fiber orientation at low strains. This delay in fiber evolution can provide an explanation for the stress growth maximums occurring at larger strains at the higher concentrations. After 20 strain units the orientation of the 40 wt% suspension appears to change at a faster rate ultimately achieving a higher degree of flow aligned fiber orientation than the other concentrations. It is not clear from the stress growth measurements that there is a difference in steady state fiber orientation.

Over the investigated shear rates, fiber orientation appears to be independent of shear rate and only a function of shear strain. Figure 5 provides measured fiber orientation as a function of strain for the 30 wt% suspensions and is representative of the effect of shear rate on orientation at all other concentrations. Orientation appears to be independent of shear rate and only a function of applied shear strain which had been acknowledged theoretically [42]. The dependency of viscosity on shear rate cannot be attributed to changes in orientation. However,

the stress required to change orientation does show a rate dependency. Ortman et al.[22] noted a slight dependency of orientation as a function of shear rate for 30 wt% glass fibers, but the authors stated that orientation was primarily strain dependent. Larger variations in orientation data at different shear rates was reported in their data relative to what is observed in this work.

5 Stress Prediction

The stress model presented in Eq. (2) was used to predict stress based on the measured orientation of a population of fibers. Typically a closure approximation is used in the fiber orientation or stress equations as a computationally efficient way of representing a higher order tensor as a function of lower order tensors. However, the use of a closure approximation induces error that can significantly impact the stress predictions. As an example, the use of the invariant based optimal fitting (IBOF) closure to approximate the fourth order term in the stress equation from the experimentally obtained second order tensor yielded an over prediction in stress by a factor of 2 to 3 [43]. To overcome issues with the closure approximation, the component of interest in the fourth order term for the shear stress, $\langle \mathbf{p}_1 \mathbf{p}_2 \mathbf{p}_1 \mathbf{p}_2 \rangle$, was calculated based on measured fiber orientation. The orientation dependent parameter in the model, C , was set to the value for three-dimensionally random orientation. The results of calculated stress at a shear rate of 1.0 s^{-1} from measured values of fiber orientation are available in Fig. 6.

Stress predictions in Fig. 6 under predict experimental data as expected, with the greatest deviation occurring at the highest concentration. Additionally, this provides some reinforcement in the measurement of fiber length because any significant errors in length measurements are magnified due to stress scaling with a_r^2 . Stress predictions at a strain of 2 are a result of experimental orientation data obtained at zero strain. At zero strain the sample is not subjected

to flow which would result in zero stress. In addition, transients at low strains may be a result of the instrument and not the fibers' contribution to stress. The stress predictions are not intended to infer a stress overshoot at lower strains than the experimental data, but rather that there is no stress overshoot prediction using the measured fiber orientation. The prediction of no stress overshoot is directly related to no overshoot observed in fiber orientation. Furthermore, the initial conditions of fiber orientation show more preferential fiber orientation in the flow direction than the gradient direction resulting in no overshoot caused by the tension imposed on the fibers. As a result the stress model is only capable of representing a decrease in stress from the initial condition toward a steady state value at higher strains at all concentrations and shear rates.

The absence of a stress growth overshoot and under prediction of the steady state value indicates that there are additional contributions that must be included to the stress theory. In addition, the use of an empirical coefficient to scale the fiber contribution to stress would not aid in predicting the transient response. The difference between semi-dilute theory and the concentrated data presented in this work could be accounted for by including stress contributions due to fiber-fiber interactions or other interactions between fibers that may result in larger network structures or flocculations.

6 Conclusions

Measurements of shear stress have been performed during the startup of shear flow in a sliding plate rheometer at concentrations from 10 to 40 wt% long glass fibers in polypropylene. Experimental measurements of fiber orientation were obtained to investigate the relationship between shear stress and fiber orientation. Shear thinning was observed in the fiber suspensions

which cannot be attributed to differences in orientation, because fiber orientation appears to be solely a function of strain for rigid fibers. As fiber concentration increased, the stress growth overshoot broadened which indicates slower orientation kinetics. However, changes in measured fiber orientation are small over the range of investigated concentrations.

Stress predictions were performed using measured fiber orientation data in order to avoid any approximations in calculating the orientation dependent term in the stress equation. The stress theory of Shaqfeh and Fredrickson was used as an example to show that a shear stress growth overshoot will not be predicted if there is no overshoot in fiber orientation or if fibers do not rotate through 45° in the x_1x_2 -plane. Empirically scaling the orientation dependent term in the stress equation could better reflect the steady state, but would not provide a good representation of the transient response. Additional contributions to stress should be considered in order to reflect the stress response for fiber suspensions.

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8 References

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Figures

Figure 1: Viscosity as a function of shear strain for the polypropylene matrix (×) and glass fiber at 10 wt% (○), 20 wt% (□), 30 wt% (◇) and 40 wt% (▽) at shear rates of 0.4 (a) and 4.0 (b) s⁻¹.

Figure 2: Viscosity as a function of shear strain for pure polypropylene matrix (open symbols, a) and 10 (filled symbols, a) and 40 wt% (filled symbols, b) glass fiber, for shear rates of 0.4 (▽), 1.0 (□) and 4.0 (○) s⁻¹.

Figure 3: Steady shear viscosity for the polypropylene matrix ($\times$) and glass fiber at 10 wt% ($\circ$), 20 wt % ($\square$), 30 wt % ($\diamond$) and 40 wt% (∇).

Figure 4: Orientation components A_{11} ($\circ$), A_{22} ($\square$), and A_{33} ($\diamond$) as a function of shear strain at 1.0 s^{-1} for 10 (open symbols) and 40 wt% (filled symbols) glass fiber.

Figure 5: Orientation evolution for 30 wt% glass fiber at shear rates of 0.4 ($\circ$), 1.0 ($\square$) and 4.0 (∇) s^{-1} . Trends in A_{11} , A_{22} and A_{33} are designated by solid, dashed and dotted lines, respectively.

Figure 6: Stress predictions (closed symbols) at a shear rate of 1.0 s^{-1} as a function of concentration for 10 wt% ($\circ$), 20 wt% ($\square$), 30 wt% ($\diamond$) and 40 wt% (∇) glass fiber. Open symbols represent the experimental data.

Table 1: Fiber Suspension Properties

Glass Fiber Content wt%	v%	Number Average Fiber Length (L_n), mm	Aspect Ratio (L_n/d)	Weight Average Fiber Length (L_w), mm
10	3.8	1.40 $\pm$ 0.074	105	3.59 $\pm$ 0.61
20	8.2	1.28 $\pm$ 0.078	97	2.95 $\pm$ 0.37
30	13.3	1.25 $\pm$ 0.065	94	2.28 $\pm$ 0.24
40	19.2	1.06 $\pm$ 0.080	79	1.96 $\pm$ 0.35