

# The Dynamic Behavior of a Concentrated Non-Brownian Glass Fiber Suspension in Simple Shear Flow

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**Abstract.** The dynamic behavior of a concentrated short glass fiber suspension subject to simple shear flow is investigated. In particular we are interested in determining the relationship between the stress growth functions (shear and first normal stress difference) and the fiber microstructure within the sample. Stress growth experiments, in start up of flow, are performed on a Rheometrics Mechanical Spectrometer (RMS-800) using a novel approach which deforms the sample in a homogeneous shear field. The 3D fiber orientation is characterized using confocal laser microscopy and experimental results are compared to predictions based on the generalized Jeffery equation. It is found that the theory over predicts the rate at which the fiber orientation evolves.

**Keywords:** glass fiber, suspension, fiber orientation, rheology, simple shear

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## INTRODUCTION

Current approaches to predicting the rheology and microstructure of concentrated suspensions either directly use or are based on a generalized form of Jeffery's equation combined with an equation associating fiber orientation with the extra stresses. Previous attempts by researchers to improve on theoretical predictions for non-dilute suspensions have focused on the equations governing the extra stress contributions [1]. Conversely, the generalized Jeffery equation describing the evolution of the fiber orientation has remained relatively unchanged. The goal of this paper is to assess the ability of the generalized Jeffery's equation to accurately describe the evolution of the glass fiber orientation in concentrated suspensions typical of composite fluids.

## THEORY

One way to describe the average orientation is with the orientation order parameter tensor,  $\mathbf{A}$ , which is defined as the second moment of the orientation distribution function:

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

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

For simple flows the generalized Jeffery equation can be written in terms of  $\mathbf{A}$  as follows with the use of the quadratic closure approximation [2]:

$$\frac{d\mathbf{A}}{dt} = (\mathbf{A} \cdot \mathbf{W} - \mathbf{W} \cdot \mathbf{A}) + \lambda(\mathbf{D} \cdot \mathbf{A} + \mathbf{A} \cdot \mathbf{D} - \mathbf{D} : \mathbf{A}\mathbf{A}) \quad (2)$$

where  $\mathbf{W} = [(\nabla v) - (\nabla v)^t]/2$ ,  $\mathbf{D} = [(\nabla v) + (\nabla v)^t]/2$ ,  $\nabla v$  is the velocity gradient and  $\lambda$  is a constant defining the ellipticity of the particle. For fibers it is common to assume the particle's aspect ratio approaches infinity and to use  $\lambda = 1$ . Eq. (2) then predicts the period of rotation for the fiber to be infinitely long and is used for all model predictions. In the results and discussion section, predictions using Eq. (2) with  $\lambda = 1$  for simple shear flow kinematics are compared to experimental results. The generalized Jeffery equation, Eq. (2), was solved numerically for simple shear flow kinematics ( $v_1 = \dot{\gamma}y$  and  $v_2 = v_3 = 0$ ) with  $\lambda = 1$ . Gears implicit predictor-corrector method for stiff differential equations was used to solve the coupled equations using a time step of 0.01 s. The initial conditions were found experimentally.

## EXPERIMENTAL PROCEDURE

All experiments were performed on a 30 wt% (17.6 vol%) short glass fiber-filled polybutylene terephthalate (PBT-30). The neat PBT suspending medium exhibited little shear thinning behavior shown by way of the magnitude of the complex viscosity,  $|\eta^*|$ :  $|\eta^*| = 420 \text{ Pa}\cdot\text{s}$  at 0.1 rad/s,  $|\eta^*| = 320 \text{ Pa}\cdot\text{s}$  at 100 rad/s. Hence, the suspending medium behaved similarly to a Newtonian fluid. The glass fiber has a number average length of  $L_n = 0.36\text{mm}$  and aspect ratio  $a_r \sim 30$ . All rheological measurements were performed on a Rheometrics Mechanical Spectrometer (RMS-800) fitted with 50 mm diameter cone and plate geometry to ensure a homogeneous shear field within the rheometer gap. Samples were pre-formed and a 25.4 mm diameter hole was drilled through the center creating a “donut” shaped sample. This ensured that at every position the gap was always greater than two times the number average fiber length. All samples were dried at 120 °C for a minimum of four hours in a vacuum oven at a pressure of 0.3 (in.Hg) before sample molding or testing.

To characterize the evolving fiber orientation under dynamic conditions, donut samples composed of PBT-30 were deformed using the RMS-800 with the cone and plate geometry at a shear rate of  $1 \text{ s}^{-1}$  for a specified amount of time (strain) at 260 °C in a nitrogen environment. Directly after deformation the sample temperature was lowered below the suspension melt temperature, “freezing” the flow induced fiber orientation. The samples were then bisected creating a plane perpendicular to the flow direction, encapsulated in epoxy and sanded/polished to a final abrasive particle size of  $0.3 \mu\text{m}$   $\text{Al}_2\text{O}_3$  following standardized techniques [Lee et. al. 2004]. Images of the polished surface were taken at the surface  $10\mu\text{m}$  into the depth of the sample using a Zeiss LSM510 confocal laser scanning microscope fitted with a 40x water immersion objective lens and a laser excitation wavelength of 543 nm. In the images taken with the confocal laser microscope the cross section of each fiber appeared as circles or ellipse-like shapes. From the ellipse-like shapes, the unit vector  $\mathbf{u}$  along the backbone can be determined through geometric identities described in detail in [3]. With the  $\theta$  and  $\phi$  angles the components of the vector  $\mathbf{u}$  can be determined for each fiber. The tensor  $\mathbf{A}$  can be constructed as follows:

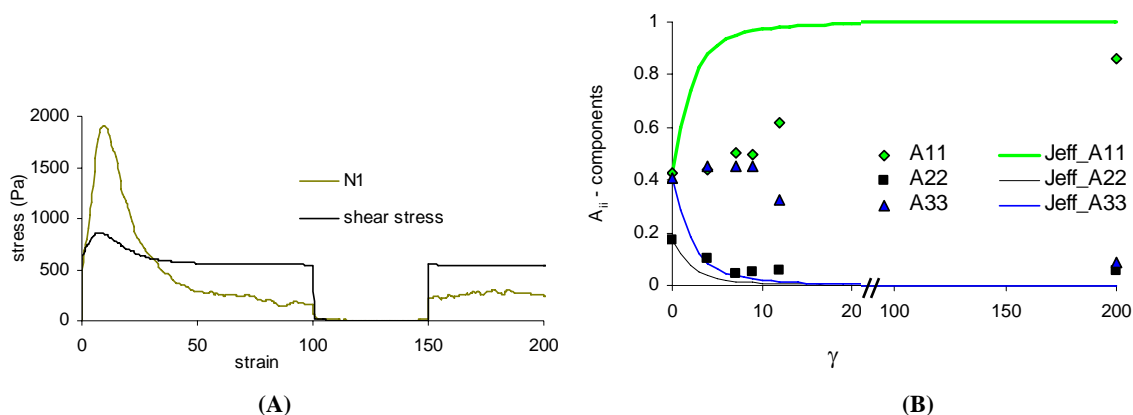
$$A_{ij} = \frac{\sum (u_i u_j)_n F_n}{\sum F_n}, \quad F_n = \frac{M_n}{m_n} \quad (3)$$

where  $F_n$  is a weighting function for the  $n$ th fiber. The weighting function is based on the probability of a 2D plane intersecting a fiber, meaning, a fiber aligned perpendicular to the plane is more likely to be severed than one aligned parallel.

## RESULTS AND DISCUSSION

The transient rheological behavior of the PBT-30 in start up of simple shear flow can be summarized in **Figure 1 (A)**. **Figure 1 (A)** depicts the shear stress and first normal stress difference vs. strain in an interrupted start up of flow experiment for a donut sample. The initial fiber orientation of the sample, determined experimentally, was mostly oriented in the 1 (flow) and 3 (neutral) directions. During start up of flow the stresses, both shear and first normal stress difference ( $N_1$ ), exhibit large stress overshoots. The overshoot in the shear stress reaches a steady state in roughly 50 strain units while the overshoot in  $N_1$  takes roughly 75 strain units to reach a steady state. The overshoot in the stresses is believed to be a result of the rods rotating to align themselves in the flow direction. After 100 s the flow was stopped and the stresses relax almost instantly, within 2 s. When the flow is reapplied in the

same direction the stresses immediately rise to the steady state value and do not exhibit the large overshoots exhibited by a fresh sample. This is a typical response of non-Brownian particles in which particle sedimentation is negligible.



**FIGURE 1.** (A) Shear stress and first normal stress difference vs. strain in an interrupted start up of flow experiment for PBT-30. Test conditions: shear rate =  $1 \text{ s}^{-1}$ , temperature =  $260 \text{ }^\circ\text{C}$ , and nitrogen environment. (B)  $A_{ii}$  components vs. strain in start up of flow. Experimentally determined fiber orientation (A) and predictions determined from Jeffery's equation (Jeff\_A).

In an attempt to experimentally characterize the evolving fiber orientation, six samples were analyzed at strains of 0, 4, 7, 9, 12 and 200 which relate to points of interest in **Figure 1 (A)**. Strains 0 and 200 corresponded to the initial and steady state orientation of the system respectively. Strains 7 and 9 corresponded to the shear stress and  $N_1$  overshoot peaks, respectively, and strains 4 and 12 were intermediate points.

The experimental results for the fiber orientation suggests that the majority of the fibers are initially oriented in the 1 and 3-directions with little orientation in the 2-direction (strain = 0,  $A_{11} = 0.424$ ,  $A_{22} = 0.170$  and  $A_{33} = 0.406$ ). The experimental results for the diagonal components of  $\mathbf{A}$  are compared to model predictions based on the generalized Jeffery's equation in **Figure 1 (B)**. Jeffery's equation predicts that within 7 strain units the fibers are almost fully aligned in the flow direction (Jeff\_A11 = 0.949 at strain = 7) while experimental data shows that at a strain of 12,  $A_{11} = 0.621$ . Hence the fibers are not nearly as aligned as the model predicts. We speculate that this must be due to direct inter-particle interaction hindering the fiber rotation. At a strain of 200 the model predicts that the fibers are completely aligned, Jeff\_A11 = 0.999. The experimental data does show that the fibers are aligning themselves in the flow direction,  $A_{11} = 0.858$ , just not as quickly as the model would suggest. It is unclear if the fiber orientation ever reaches a steady state due to the lack of experimental data at the intermediate strains.

## SUMMARY

Experimentally determined fiber orientation was compared to predictions based on the generalized Jeffery's equation for evolving fiber orientation in simple shear flow. It was found that the theory predicted the fiber orientation to evolve and align in the flow direction much faster than was determined experimentally.

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