ARTICLE IN PRESS

Journal of Non-Newtonian Fluid Mechanics xxx (2014) xxx–xxx

Contents lists available at ScienceDirect

Journal of Non-Newtonian Fluid Mechanics

journal homepage: http://www.elsevier.com/locate/jnnfm

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

Mark J. Cieslinski [a], Peter Wapperom [b], Donald G. Baird [a,*]

[a] Department of Chemical Engineering, Virginia Tech, VA 24061, United States
[b] Department of Mathematics, Virginia Tech, VA 24061, United States

ARTICLE INFO

Article history:
Received 1 August 2014
Accepted 26 October 2014
Available online xxxx

Keywords:
Fiber rheology
Transient shear flow
Fiber orientation
Long glass fibers

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_v, and fiber aspect ratio (a_r = fiber length/diameter), fiber suspensions can be divided into dilute ($\phi_v < a_r^{-2}$), semi-dilute ($a_r^{-2} < \phi_v < a_r^{-1}$), and concentrated ($\phi_v > a_r^{-1}$) regimes. Generally, composites of commercial interest have $\phi_v > 0.1$ [9]. In the concentrated regime, the motion of an individual fiber is restricted by surrounding fibers through hydrodynamic and fiber–fiber interactions. As ϕ_v 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 though the use of a diffusivity tensor in the anisotropic rotary diffusion (ARD) orientation model. These approaches in modeling fiber orientation require that material parameters be determined experimentally.

---

* Corresponding author at: Department of Chemical Engineering, 133 Randolph Hall, Virginia Tech, Blacksburg, VA 24061, United States. Tel.: +1 540 231 7870; fax: +1 540 231 2732.

E-mail address: dbaird@vt.edu (D.G. Baird).

http://dx.doi.org/10.1016/j.jnnfm.2014.10.012

Please cite this article in press as: M.J. Cieslinski et al., Influence of fiber concentration on the startup of shear flow behavior of long fiber suspensions, J. Non-Newtonian Fluid Mech. (2014), http://dx.doi.org/10.1016/j.jnnfm.2014.10.012

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. Ortman 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, **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:

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

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, $\boldsymbol{\sigma}$:

$$\boldsymbol{\sigma} = -P\mathbf{I} + 2\mathbf{D}\eta_m + \frac{4\eta_m\phi_v a_r^2}{3[\ln(1/\phi_v) + \ln\ln(1/\phi_v) + C]}\mathbf{D} : \langle\mathbf{pppp}\rangle \tag{2}$$

where P is the isotropic pressure, **I** is the identity tensor, η_m is the matrix viscosity, the rate of deformation tensor, $\mathbf{D} = (\nabla\mathbf{v}^t + \nabla\mathbf{v})/2$ and $\langle\mathbf{pppp}\rangle$ 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 $\phi_v \approx \pi/20a_r$. 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 neat 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 and 1.7 mm. Samples were compression molded for 5 min in a preheated mold at 180 °C. This procedure was found to minimize voids which could greatly impact the measurements of stress.

Please cite this article in press as: M.J. Cieslinski et al., Influence of fiber concentration on the startup of shear flow behavior of long fiber suspensions, J. Non-Newtonian Fluid Mech. (2014), http://dx.doi.org/10.1016/j.jnnfm.2014.10.012