# Fiber Dynamics in Concentrated Suspensions of Short Glass Fibers in Polymers

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**SUMMARY:** Rheological experiments have been conducted on concentrated suspensions of short glass fibers in a polypropylene and a polybutene under simple shear transient flows. A stress and a rate-controlled rheometer with plate-plate geometry have been used. The viscosity and the normal stress differences have been shown to depict overshoots in stress growth experiments, as well as when the flow was reversed. However, for the reverse flow experiments negative normal stress differences have been observed for the initial part of the experiments. The filled polybutene exhibited a similar behavior compared to that of the filled polypropylene. A Folgar-Tucker model for the fiber dynamics combined with the Lipscomb stress equation has been used to predict the steady-state and transient data. This model developed for dilute or semi-dilute suspensions is able to qualitatively predict the transient observations. However, important differences are observed between the experimental overshoots and the predictions. This implies that experimental fiber dynamics is markedly slower than predicted by the model. Fiber-fiber interactions are discussed and a slip factor introduced in the Folgar-Tucker model is shown to improve the fits.

**KEYWORDS:** short fiber, concentrated suspension, rheological properties, viscosity overshoot.

## INTRODUCTION

Main advantages of thermoplastic materials are their low density and their low production cost. But these materials have weak mechanical properties and they are often filled with short fibers. Reinforced thermoplastics could be processed with conventional machines and tools. However, to be efficient short fibers must be oriented in the stress direction. The final orientation in a part is induced by the composite flow in the processing tool and is hard to control. Moreover, the content of fibers in industrial reinforced thermoplastics is large and, generally, concentrated suspensions with a complex rheological behavior have to be considered. In this work typical experimental results obtained for a commercial glass fiber filled polypropylene and suspensions of fibers in a polybutene will be presented. Simulation results based on a modified Folgar-Tucker model will be discussed and compared with experimental data.

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## MODEL

### Fiber motion

A fiber orientation can be described with an orientation vector p, a unit vector parallel to the main axis of the fiber. For a population of fibers, a statistical orientation distribution function $\psi(\mathbf{p})$ can be used to describe the average state of orientation in a fluid element. A second- and a fourth-order orientation tensors have been defined by Advani and Tucker [1]:

$$\mathbf{a}_2 \Leftrightarrow a_{ij} = \int_{\mathbf{p}} p_i p_j \psi(\mathbf{p}) d\mathbf{p}, \qquad (1)$$

$$\mathbf{a}_4 \Leftrightarrow a_{ijkl} = \int_{\mathbf{p}} p_i p_j p_k p_l \psi(\mathbf{p}) d\mathbf{p}, \qquad (2)$$

where $\mathbf{a}_2$ is a symmetric tensor with trace equal to one. A closure approximation is necessary to evaluate the fourth-order orientation tensor. The time derivative of $\mathbf{a}_2$ can be derived from that of **p** established by Jeffery [2] for dilute suspensions of ellipsoids in a Newtonian fluid under simple shear and at low Reynolds numbers. It has been improved by Folgar and Tucker [3] for semi-dilute suspensions of fibers and can be written as:

$$\dot{\mathbf{a}}_2 = \frac{D\mathbf{a}_2}{Dt} = \frac{1}{2}\left(\boldsymbol{\Omega}\mathbf{a}_2 - \mathbf{a}_2\boldsymbol{\Omega}\right) + \frac{\lambda}{2}\left(\dot{\boldsymbol{\gamma}}\mathbf{a}_2 + \mathbf{a}_2\dot{\boldsymbol{\gamma}} - 2\dot{\boldsymbol{\gamma}} : \mathbf{a}_4\right) + 2C_I\,\bar{\dot{\gamma}}\left(\mathbf{I} - 3\mathbf{a}_2\right), \qquad (3)$$

where $\lambda = (r^2-1)/(r^2+1)$ and $r$ is the ellipsoid aspect ratio. $\dot{\boldsymbol{\gamma}}$ and $\boldsymbol{\Omega}$ are respectively the rate of strain and the vorticity tensor defined by $\dot{\boldsymbol{\gamma}} = \left(\boldsymbol{\kappa} + \boldsymbol{\kappa}^t\right)$ and $\boldsymbol{\Omega} = \left(\boldsymbol{\kappa} - \boldsymbol{\kappa}^t\right)$ where $\boldsymbol{\kappa}^t$ is the velocity gradient tensor. $\bar{\dot{\gamma}}$ is the effective deformation rate (square root of half of the second invariant of the rate of strain tensor) and $C_I$ a phenomenological coefficient. For large ellipsoid aspect ratios, $\lambda$ tends toward 1. Fibers in filled polymer composites are considered to be ellipsoids having an aspect ratio of $r$.

### Constitutive equation

Following Jeffery [2], Hand [4] and Giesekus [5], Lipscomb *et al.* [6] have proposed a constitutive equation for dilute particles suspensions, which can be written for ellipsoids with high aspect ratio, as:

$$\boldsymbol{\sigma} = -P\mathbf{I} + \eta_m\dot{\boldsymbol{\gamma}} + \eta\phi_m\left\{2\dot{\boldsymbol{\gamma}} \quad {}_2\dot{\boldsymbol{\gamma}} : \mathbf{a}_4\right\}, \qquad (4)$$

where $\eta_m$ is the matrix viscosity, $P$ the hydrostatic pressure, $\phi$ the fiber volume fraction and $\mu_2$ is rheological coefficient. The last term of Eqn. 4 describes the coupling between hydrodynamic forces and fiber orientation; hence we call $\mu_2$ the coupling coefficient.

### Closure approximations

To complete the model the fourth-order orientation tensor is evaluated knowing the second-order orientation tensor. Many closure approximations have been proposed. Among the most recent, the orthotropic [7, 8] and the natural [9] closure approximations are similar in philosophy. Invariants of the second-order tensor are used to calculate the fourth-order tensor. Different closure approximations have been tested in forward and reverse flows. The orthotropic has been chosen as the more accurate to fit our experimental results.

## EXPERIMENTS

A commercial unfilled polypropylene (PP, Targor PPN 0160D1) and a filled polypropylene containing 30 wt.% short glass fibers (PP30, Targor Hostacom G3 N01L) have been used. Targor also prepared other fiber suspensions with different fiber contents in experimental batches under the same conditions as PP30. The fiber average length measured after blending using an internal mixer was 260 μm for PP30. They have a diameter of 14 μm. Filled polypropylene has been pyrolyzed, the fibers recovered and blended in a Newtonian polybutene (PB, Stanchem Indopol H100) with a molecular mass of 920 g/mol, a density of 0.89 g/mL and a viscosity of 24 Pa.s at 25°C. Experiments have been conducted with a rate-controlled rheometer, ARES (Rheometric Scientific). Parallel plate fixtures were used with the gap between the two plates $H$ varying between 1.4 and 1.5 mm for all experiments. For the polypropylene fiber composites, the plate radius $R$ was equal to 12.5 mm, the temperature was set at 200°C and experiments conducted under a nitrogen blanket. For polybutene fiber suspensions, the plate radius was 25 mm and experiments carried out at room temperature. Stress growth experiments have been conducted in forward and reverse flow directions. The viscosity and the normal stress differences have been shown to depict overshoots in the forward stress growth experiments as well as when the flow was reversed. However, for the reverse flow experiments negative normal stress differences have been observed for the initial part of the experiments. The filled polybutene exhibited a similar behavior compared to that of the filled polypropylene.

## DISCUSSION

The model presented above is able to qualitatively predict the transient observations. However, important differences were observed between the experimental and the predicted overshoots. This implies that the experimental fiber dynamics is much slower than predicted by the model, due to fiber-fiber interactions. A slip factor is introduced in the Folgar-Tucker model as:

$$\gamma_e = \alpha \dot{\gamma} t \qquad (5)$$

where $\gamma_e$ is the modified experimental deformation and $\alpha$ is the slip factor. It improves the fits of the data, but this empirical approach is valid only for simple shear flows as the model will no longer be objective. The modified model contains three adjustable parameters, $\mu_2$, $C_I$ and $\alpha$. The model predictions for reverse overshoot viscosity data of PB05, PB10 and PB30 and results are presented in Fig. 1. The model parameters are reported in Table 1. Using the same values of the model parameters the predictions for the normal stress data are compared in Fig. 2. For both figures, the fits are quite reasonable.

Table 1 Model parameters for the filled PB at different fiber mass fractions.

| | PB05 | PB10 | PB20 |
|---|---|---|---|
| $\mu_2$ | 280 | 210 | 195 |
| $C_I$ | 0.0003 | 0.0008 | 0.0010 |
| $\alpha$ | 0.7 | 0.6 | 0.5 |

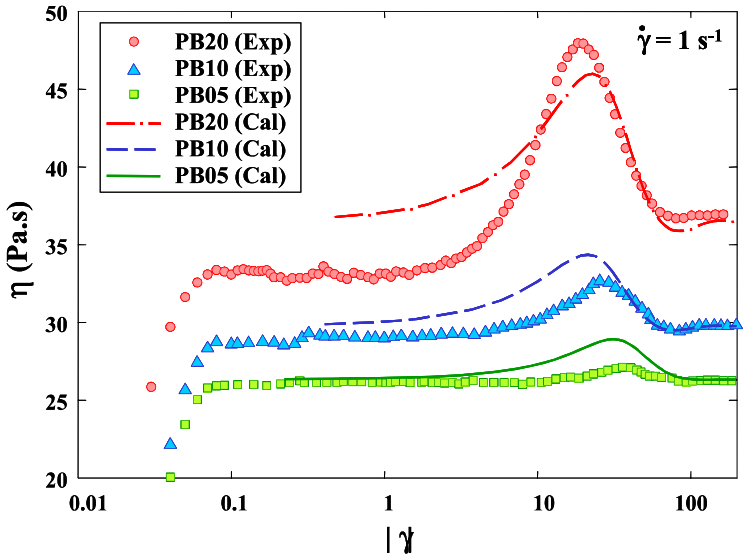


Fig. 1. Comparison of the model predictions and the experimental viscosity data of PB suspensions obtained for reverse stress growth experiments.

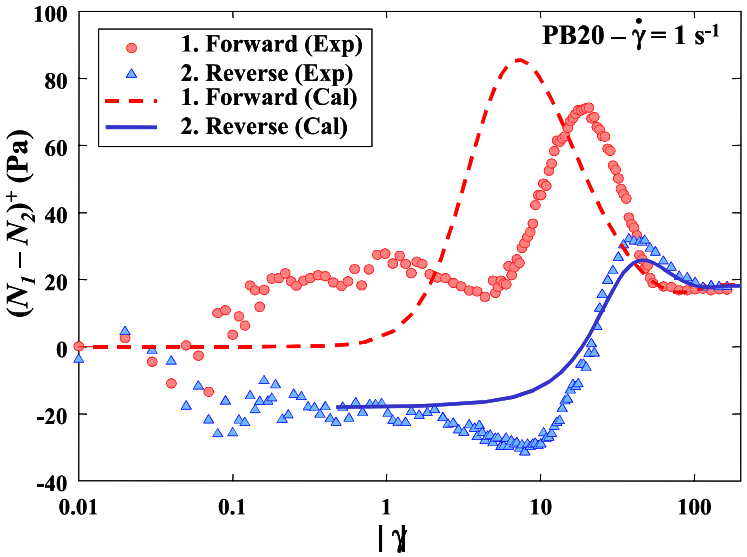


Fig. 2. Comparison of the model predictions and the experimental normal stress data of PB20 obtained for forward and reverse stress growth experiments.

## CONCLUSION

The rheological properties of glass short fiber filled polypropylene has been studied and compared to data of suspensions of the same fibers in a polybutene, obtained under transient shear flows. Viscosity and normal stress overshoots were observed each time the flow direction was reversed. When the flow was reversed, the normal stress differences took initially negative values before depicting a positive overshoot and reaching a steady-state value. A model based on the Lipscomb and Folgar-Tucker equations can qualitatively describe the rheological behavior. The simulations carried out with this model predicted viscosity overshoots of smaller width than those measured. A slip parameter has been empirically introduced in the model to reduce the fiber rotation motion and the modified model is shown to describe fairly well the stress growth overshoots for the viscosity as well as for the normal stress differences.

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## REFERENCES

1. S.G. Advani, C.L. Tucker, "The use of tensors to describe and predict fiber orientation in short fiber composites", *Journal of Rheology*, Vol.31, 1987, pp. 751.
2. G.B. Jeffery, "The motion of ellipsoidal particles immersed in a viscous fluid", *Proc. Roy. Soc. London, Ser.,* A102, 1922, pp 161.
3. F.P. Folgar and C.L. Tucker, "Orientation behavior of fibers in concentrated suspensions", *Journal of Reinforced Plastics and Composites*, Vol.3, 1984, pp. 99.
4. G.L. Hand, "A theory of dilute suspensions", *Archive for Rational Mechanics and Analysis*, Vol.7, 1961, pp. 81.
5. H. Giesekus, " Elastico-viscous fluids displaying a large normal stress component in steady flow ", *Rheologica Acta*, Vol.2, 1962, pp. 50.
6. G.G. Lipscomb, "Analysis of suspension rheology in complex flows", Ph. D dissertation, University of California, Berkeley, 1987.
7. J.S. Cintra and C.L. Tucker, "Orthotropic closure approximations for flow-induced fiber orientation", *Journal of Rheology*, Vol.39, n°6, 1995, pp. 1095.
8. E.D. Wetzel and C.L. Tucker, "Area tensors for modeling microstructure during laminar liquid-liquid mixing", *International Journal of Multiphase Flow*, Vol.25, 1999, pp. 35.
9. V. Verleye and F. Dupret, "Numerical prediction of fiber orientation in complex injection molded parts", *Proceedings of the ASME Winter Annual Meeting*, MD, Vol.49, HTD-Vol. 283, 1994, pp. 264.