SIMULATING ORIENTATION OF LONG, SEMI-FLEXIBLE GLASS FIBERS IN THREE-DIMENSIONAL INJECTION MOLDED THERMOPLASTIC COMPOSITES

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Abstract

Because of the potential of long glass fibers (>1mm) to enhance the mechanical properties of an injection molded part to a significant degree relative to short fibers, there is presently considerable interest in these systems. Modeling of fiber orientation in the case of the long fiber composites, however, may not be accounted for by models applied to short fiber composites. The Bead-Rod orientation model, adapted for concentrated suspensions, is utilized to capture flexibility by representing a semi-flexible fiber as two adjoined vectors free to rotate about the connecting joint. In this work fiber orientation model parameters are extracted by fitting orientation data at 10% of the mold fill cavity at multiple locations of the mold width in an end-gated plaque. Another approach for obtaining the model parameters is the use of shear stress growth caused by fiber evolution during the startup of simple shear which is generated using a sliding plate rheometer. Orientation model parameters are validated for use in non-isothermal simulations by fitting the shear stress growth and verified with orientation measurements of the fiber evolution. The simulations presented in this work utilize the entire flow field including both entry effects to the mold as well as the fountain flow region at the polymer/air interface. Furthermore, the effects of temperature are accounted for during the injection molding simulation. An end-gated geometry, including the gate and mold cavity, was chosen for the test scenario. Here the orientation generated in the gate and plaque are modeled and compared to experimental data taken at a variety of sampling locations both along and away from the centerline. These predictions show that the Bead-Rod model more closely represents the experimental data than does the Folgar-Tucker model for long fiber composites.

Background

Glass fibers are an economically viable reinforcement to thermoplastics and are often employed through traditional melt processing techniques, such as injection molding. Fibers exhibit flow-induced microstructure during injection molding that can largely the influence overall properties(1, 2). It is advantageous to understand and model the fiber orientation kinetics in order to optimize mold design to yield a desired fiber microstructure.

Fiber length can play a critical role in determining the microstructure and overall performance of the composite. Glass fibers have been subdivided by length into two categories based on mechanical properties (3). Short glass fibers are considered to behave as rigid rods.
in flow and are below 1mm in length. The designation of a long glass fiber is given to those fibers above the 1mm threshold and consequently considered semi-flexible. Switzer and Klingenberg (4) proposed an effective flexibility parameter that is dependent on the flow conditions and the properties of the fiber, shown as Equation (1):

\[
\frac{f_{\text{eff}}}{\dot{\gamma}} = \frac{64\eta_m \dot{\gamma} a_r^4}{E_y \pi}
\]

where \( \eta_m \) is the matrix viscosity, \( \dot{\gamma} \) is shear rate magnitude, \( a_r \) is the fiber aspect ratio, and \( E_y \) is the Young’s modulus of the fiber. Most significantly, the effective flexibility of a fiber is related to \( a_r^4 \), indicating that long fibers are substantially more flexible than short fibers.

Parts made using fiber reinforcement can exhibit anisotropic mechanical properties due to orientation dictated by the flow field(5). Recent research interests have dealt with predicting the orientation based on Jeffery’s derivation for the motion of ellipsoidal particles in viscous fluids(6). Folgar and Tucker added an isotropic rotary term to Jeffery’s equation to account for fiber-fiber interactions in concentrated suspensions (7). The potential for flexibility and curvature can impact the orientation dynamics of long fiber and has been accounted for by Strautins and Latz(8) for dilute suspensions. In the work of Ortman et al.(9), the semi-flexible model was extended to concentrated systems through the addition of anisotropic rotary diffusion terms. The work presented here will demonstrate the ability of the semi-flexible model to improve the prediction of long fibers in a three-dimensional model flow when compared to predictions made by the rigid fiber model.

Orientation Models

An orientation tensor is used to represent the ensemble average orientation of fibers in terms of the coordinate directions (10). This method describes the orientation of a single fiber by a unit vector, \( \mathbf{p} \), along the axis of the fiber. Orientation is represented by combining the respective moment of orientation with the orientation distribution function and integrating over all configuration space. The second and fourth moments are defined as Equations (2) and (3), respectively:

\[
A = \int \mathbf{pp} \psi(\mathbf{p}, t) d\mathbf{p}
\]

\[
A_4 = \int \mathbf{pppp} \psi(\mathbf{p}, t) d\mathbf{p}
\]

The Folgar-Tucker model uses the orientation tensors to describe the orientation of fibers dictated by the flow conditions and has shown qualitative agreement with short fiber experimental data(11, 12). The works of Sepehr et al. (13) and Eberle et al. (14) found that orientation evolves at a slower rate than what is predicted by the Folgar-Tucker model. A slip parameter, \( \alpha \), is added to the model to retard the orientation kinetics to that seen experimentally, Equation (4):
\[
\frac{DA}{Dt} = \alpha \left[ W \cdot A - A \cdot W + \xi \left( D \cdot A + A \cdot D - 2D : A_d \right) + 2C \gamma (I - 3A) \right] \quad (4)
\]

The terms containing the vorticity tensor, \( W \), and the rate of deformation tensor, \( D \), are the hydrodynamic contributions that orient fibers with the flow. The term containing the interaction parameter, \( C \), is the isotropic rotary diffusion term representing fiber-fiber interactions and is based on the theory of Brownian rods. However, the inclusion of the shear rate magnitude, \( \gamma \), to the isotropic rotary diffusion term will only influence orientation during flow making the equation applicable to non-Brownian suspensions. Here, \( DA / Dt \) represents the material derivative of \( A \), \( \xi \) is the shape parameter which is taken to be unity for high aspect ratio particles and \( \alpha \) is the slip parameter scaling the hydrodynamic and isotropic rotary diffusion terms between zero and unity. This model consists of two phenomenological constants, \( \alpha \) and \( C \), that must be determined to predict orientation. Eberle et al. found these parameters for short fibers independently of the injection molding process using transient rheological measurements(14).

Orientation kinetics influenced by flexibility was proposed by Strautins and Latz(8) for dilute semi-flexible suspensions. These fibers can be represented as three beads connected by two equal length rods, referred to here as the Bead-Rod orientation model. Here, fiber orientation is represented as two indistinguishable adjoined vectors, \( p \) and \( q \), of length \( l_b \). Similarly to the Folgar-Tucker model, moments of orientation are constructed and listed in Equations (5-7):  

\[
A = \iint pp \psi(p, q, t) dp dq \quad (5)
\]

\[
B = \iint pq \psi(p, q, t) dp dq \quad (6)
\]

\[
C = \iiint p \psi(p, q, t) dp dq \quad (7)
\]

These moments arise by representing the average orientation of the fiber as an end-to-end vector, \( r \), such that \( r = p - q \). The average orientation tensor, \( R \), is normalized such that its trace is unity and can be represented with the \( A \) and \( B \) tensors, Equation (8):

\[
R = \frac{\langle rr \rangle}{tr(rr)} = \frac{A - B}{1 - tr(B)} \quad (8)
\]

Ortman et al. (9) modified the model of Strautins and Latz to include the isotropic rotary diffusion term found in the Folgar-Tucker model for use in predicting concentrated suspensions. The slip parameter was also included to better reflect experimental observations. The moments of orientation evolve according to Equations (9)-(12):
\[
\frac{DA}{Dt} = \alpha \left[ W \cdot A - A \cdot W + \xi \left(D \cdot A + A \cdot D - 2D : A_4 \right) + 2C_i \gamma_i \left(I - 3A \right) + \ldots \right] \\
\frac{1}{2} \left[ Cm + mC - 2 \left(m \cdot C \right) A \right] + 2k \left(B - A \cdot tr \left(B \right) \right]
\]  
(9)

\[
\frac{DB}{Dt} = \alpha \left[ W \cdot B - B \cdot W + \xi \left(D \cdot B + B \cdot D - \left(2D : A \right) \right) - 4C_i \gamma_i B + \ldots \right] \\
\frac{1}{2} \left[ Cm + mC - 2 \left(m \cdot C \right) B \right] + 2k \left(A - B \cdot tr \left(B \right) \right]
\]  
(10)

\[
\frac{DC}{Dt} = \alpha \left[ \nabla v^t \cdot C - \left(A : \nabla v^t \right) C - 2C_i \gamma_i C + \frac{1}{2} \left[m - C \left(m \cdot C \right) \right] - kC \left(1 - tr \left(B \right) \right) \right] \\
(11)
\]

\[
m = \sum_{i=1}^{3} \sum_{j=1}^{3} \sum_{k=1}^{3} \frac{\partial^2 v_j}{\partial x_j \partial x_k} A_i e_i
\]  
(12)

Here the additional terms of \(m\), representing the bending induced on the fiber caused by velocity gradients along the fiber and \(k\), which is an inherent resistivity to bending are included in the evolution equations. Larger values of \(k\) represent increasingly rigid fibers. Likewise, as \(k\) decreases the equations represent increasingly flexible fibers. In the limit that \(k\) becomes infinitely large, the Bead-Rod model recovers the dynamics of the Folgar-Tucker model. In the case where no flexibility is observed by the model, the \(R\) tensor returns to the \(A\) tensor and the Bead-Rod model reduces to the Folgar-Tucker model.

Rheological techniques allow for fluids to be extensively characterized under well-defined flow conditions. Ortman et al. were able to independently extract the phenomenological orientation parameters, \(\alpha\) and \(C_i\), from transient measurements on a sliding plate rheometer and apply the orientation equation to model flows(9, 15). An empirical modification to the Lipscomb stress tensor, in Equation (13), relates orientation to measured stresses during the experiment:

\[
\tau = 2\eta_m \left(D + f_1 \phi D + f_2 A_4 : D \right) + \eta_m k \frac{3\phi a}{2} \frac{tr \left(rr \right)}{2l_n^2} \left(A - R \right)
\]  
(13)

The stress tensor, \(\tau\), is a function of the matrix viscosity, \(\eta_m\), a concentration dependence, hydrodynamic drag of fluid on the fibers orienting with the flow and a contribution due to the restorative potential in semi-flexible fibers. \(\phi\) is the volume fraction, \(f_1\) is an empirical parameter imparting shear thinning behavior and \(f_2\) is a scaled product of the invariants of \(A\). The final term based on \(R\) is negated for rigid fiber models.
A fourth moment of orientation is involved with the orientation and stress models and it is necessary to represent this tensor as function of the second moment of orientation for numerical calculations. A summary of these closures are provided by Chung and Kwon(16). The work presented here uses the invariant-based optimal fitting closure (IBOF) which is a function of the invariants of the second moment of orientation tensor (17). Chung and Kwon were able to show that the IBOF closure is sufficient in approximating the fourth order tensor when compared to explicit calculations with the orientation distribution function.

Experimental

The end-gated plaques were created from 13 mm long polypropylene pellets containing 30% by weight glass fibers (SABIC LNP Verton MV006S) that were dried in an oven overnight at 80 °C prior to injection molding. The temperature profile was set to 190, 210 and 210 °C for the feed, compression and metering zones respectively for the Arburg Allrounder (Model 221-55-250) injection molder, while the mold temperature was set to 79 °C. The mold geometry of the end-gate plaque has a 65mm long sprue with initial radius of 1.45mm opening to a 1.75mm radius. The gate measures 80.68mm in width by 6.25mm in height by 6.33mm in length. The mold cavity has a width of 75.05mm, height of 1.55mm and a length of 77.65mm. Molding took place using a fill time of 2.0 seconds with a backpressure of 20 MPa. Plaques were kept in the mold for a duration of 20 minutes to minimize warping.

Orientation Prediction Method

A two-step method is used to predict orientation by decoupling the equations of motion and energy from the orientation models. ANSYS® Polyflow (ver. 14.0) is used to model the fluid flow in the end-gated plaque under non-isothermal conditions. The system under investigation is assumed to be incompressible and viscous enough to exhibit laminar conditions, whereby inertial terms in the equation of motion can be negated. The matrix is modeled as a generalized Newtonian fluid with the parameters determined by fitting to experimental data and temperature dependence accounted for by an Arrhenius type relationship. The Carreau-Yasuda model shown in Equation (14) is applicable to the wide range of shear rates seen in the injection molding simulation and Arrhenius relationship is shown in Equation (15): 

\[
\frac{\eta(\dot{\gamma})}{\eta_0} = \left[1 + (\lambda \dot{\gamma})^n\right]^\frac{n-1}{a} \tag{14}
\]

\[
\eta(\dot{\gamma}, T) = \eta(\dot{\gamma}) \exp\left[-\frac{\Delta E}{R} \left(\frac{1}{T} - \frac{1}{T_{ref}}\right)\right] \tag{15}
\]

where, \(\eta(\dot{\gamma})\) is the viscosity at the reference temperature, \(T_{ref}\), \(\eta_0\) is the zero-shear viscosity, \(\lambda\) determines the onset of shear thinning, \(n\) is the extent of shear thinning and \(a\) determines the transition between the zero-shear and shear-thinning regimes. For the Arrhenius relationship, \(\eta(\dot{\gamma}, T)\) is the viscosity at temperature, \(T\), \(\Delta E\) is the activation energy for flow
and \( R \) is the ideal gas constant. With the viscosity dependence on shear rate and temperature defined, the equations of continuity, motion and energy result as Equations (16)-(18):

\[
\nabla \cdot \mathbf{v} = 0 \tag{16}
\]
\[
\nabla P + \nabla \cdot \mathbf{\tau} = 0 \tag{17}
\]
\[
\rho C_p \frac{DT}{Dt} = k \nabla^2 T + \mathbf{\tau} : \dot{\gamma} \tag{18}
\]

where, \( \mathbf{v} \) is the velocity of the fluid, \( P \) is the isotropic pressure, \( \mathbf{\tau} \) is the extra stress tensor described by Equation (14) scaling the rate-of-strain tensor, \( \dot{\gamma} \). Parameters in Equation (18) are all properties of the matrix such that: \( \rho \) is the fluid density, \( C_p \) is the specific heat capacity and \( k \) is the thermal conductivity.

The volume of fluid (VOF) method is imposed by ANSYS® Polyflow to simulate the transient filling of the mold(18). The volume of fluid method allows for the advancing front to be accounted for through the use of the transport equation given as Equation (19):

\[
\frac{D\phi}{Dt} + \mathbf{v} \cdot \nabla \phi = 0 \tag{19}
\]

where \( \phi \) is the volume fraction of the fluid as it fills the mold cavity. This equation allows for tracking the fluid front and imposes “fountain flow” characteristics to the velocity profiles. The addition of fountain flow in the advancing front has been shown to influence orientation predictions in model flows (16, 19).

Velocity profiles found using the ANSYS finite element solver package are imported into Matlab® (2011b) routines developed in-house to solve the orientation models. Gradients are approximated using a finite difference method. Decoupling the orientation equation from the equations of motion and energy allow for the non-linear differential equations to be solved using the RK45 method inside Matlab® (20).

A schematic of the three-dimensional test geometry is shown as Figure 1. Fluid enters the cavity through \( \Omega_{\text{inlet}} \) and fills the substantially thicker gate region while beginning to fill the narrow mold cavity. Symmetry is imposed at \( \Omega_{\text{symmetry}} \) along the centerline of the mold. Regions of interest in this work are 0 and 90% of the plaque width in the gate and at 40% fill of the mold.
Figure 1: End-gated plaque test geometry with important regions highlighted denoting the gate centerline (1), 40% mold fill centerline (2), 90% width gate (3), and 40% mold fill, 90% width (4).

Results

The parameters used in the models for fiber orientation in the end-gated plaque were obtained by fitting orientation data obtained at the centerline, 50% and 90% of the width at 10% of the mold length from the gate. The work of Ortmann et al. used orientation parameters, $\alpha$ and $C_f$, fit from rheology to predict orientation in a center-gated disk, whereas this work uses similar rheological measurements as a validation experiment to show the consistency of orientation parameters. Variability in the stress model’s empirical parameters raised caution in using parameters gained purely from the stress growth measurements. However, experimental measurements of orientation at different times during shear flow experiments confirm that orientation is a function of velocity gradient and not a function of matrix viscosity changes due to temperature. Orientation parameters for these suspensions are then expected to be valid for non-isothermal simulations using values of $\alpha = 0.0087$ and $C_f = 0.0231$ for the rigid fiber model and $\alpha = 0.0039$ and $C_f = 0.0484$ for the Bead-Rod semi-flexible fiber model. Torsional and capillary rheometry were used to characterize the pure matrix and fit to a Carreau-Yasuda model with parameters $\eta_0 = 350$ Pa·s, $\lambda = 0.0059$ s$^{-1}$, $n = 0.2411$, and $a = 0.7498$. The activation energy term for flow in the Arrhenius relationship, $\Delta E / R$, is 4937 K.

Orientation predictions include the gate region of end-gated plaque since initial conditions can be shown to influence orientation predictions (21). Predictions discussed here are taken along the centerline and at 90% of the width for the gate region and at 40% of the mold cavity. Results of the predictions are shown in Figure 2 as contour plots containing measured orientation data and arrows indicating A11 component of orientation. The relatively large gate region compared to the mold cavity allows for orientation to be a function of width and thickness. Figure 2(a) shows the centerline data and model predictions for the Folgar-Tucker model while Figure 2(b) shows the Bead-Rod model. In both predictions, much of the cavity shows fiber orientated down the length of the gate ($x_3$ direction) which is to be expected due...
to a strong diverging extensional flow as fluid transitions down the gate from the inlet. Orientation transitions from the $x_1$ - direction to the $x_3$ - direction slower with the Bead-Rod model and better reflects the trend observed in experimental data obtained from Hofmann et al(22, 23).

Figure 2: Qualitative comparison of orientation predictions (arrows) in the gate region of the end-gated plaque compared to experimental data (contours) for (a) rigid fiber model at 0% width, (b) semi-flexible model at 0% width, (c) rigid fiber model at 90% width, and (d) semi-flexible model at 90% width. Dimensions shown as normalized thickness and width.

A distinctly different orientation contour is observed at 90% of the gate width where flow in the gate cavity turns into the mold. At this location, the rigid fiber model (Figure 2(c)) more accurately represents the experimental orientation contour as opposed to the semi-flexible model in Figure 2(d). However, entering the mold the orientation predictions are very similar and provide initial values for orientation entering the mold which is the main focus of simulation.

Comparisons of model predictions to experimental data were taken at 40% of the mold fill along the centerline and at 90% of the width shown in Figure 3 as the orientation profile through a normalized thickness. Figure 3(a) shows the predictions of both models predicting a shell-core-shell behavior as seen in the orientation data. The rigid fiber model predicts a narrower core region that what is observed experimentally. Improved predictions of the broad
core region are obtained with the Bead-Rod orientation model. However, both models largely deviate from experimental values near the walls of the mold.

![Figure 3: Preliminary orientation predictions at 40% of the mold fill for the rigid (--) and semi-flexible fiber (-) fiber models compared to experimental data of $A_{11}$ and $A_{33}$ denoted respectively by $\bigcirc$ and $\Box$ at the (a) centerline and (b) 90% of width.](image)

At 90% of the mold width the orientation of fibers is largely dictated by shear flow conditions such that the shell-core-shell is not observed. This behavior is still predicted by the rigid fiber model whereas the semi-flexible fiber model quantitatively predicts a majority of the orientation through the thickness. A large deviation between predicted and experimentally measured values occurs at -0.4 z/2H where the model under predicts the 11-component of orientation. However, the predictions near the mold walls correspond with experimental data as opposed to predictions of the centerline where the largest deviations are observed.

**Conclusions**

Predicting the orientation of long glass fibers with a rigid fiber model and a semi-flexible fiber model was carried out in an end-gated geometry. The filling of the mold was carried out under mildly non-isothermal conditions, included the gate region and captured the “fountain flow” characteristics of the advancing front. Orientation parameters were gathered from experimental data obtained at 10% mold fill and multiple locations along the width. Results from simulations were compared to experimentally measured values of orientation both along and away from the plaque centerline.

The analysis of long glass fibers in the end-gated plaque allowed for evaluation of a rigid and semi-flexible fiber orientation model in a three-dimensional mold geometry. Orientation predictions from the semi-flexible model at multiple locations in the mold showed improved agreement with experimental data relative to that of the Folgar-Tucker model. The inverse was observed in the gate region away from the centerline. Typical shell-core-shell behavior associated with short glass fiber is broadly observed along the centerline and not observed at locations away from the centerline.
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References


