MODELING DROP SIZE DISTRIBUTION IN POLYMER BLEND INJECTION MOLDING

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Abstract

An approach for modeling the drop size distribution in the injection molding of polymer blends is developed. The simulation directly uses experimental data correlated to functional forms in the FIDAP fluid dynamics package. As an example, experimental data for droplet size and shape in a Polyisoprene /Polybutadiene system was measured using an in-situ optical microscopy instrument designed for studying complex fluids under simple shear flow. The data is collected in the flow-vorticity plane as a function of temperature and shear rate. Size and shape distributions were calculated from the digitized micrograph using standard image analysis software. The shear viscosity of the blends, as well as that of the pure components, was measured as a function of shear rate and temperature using a commercially available parallel-plate rheometer. From theoretical considerations, the simulation is expected to provide good estimates of drop size distribution for flows with large aspect ratios of flow length to thickness where entrance effects are expected to be negligible, and there are no regions of recirculation.

Introduction

Polymer blend mixtures are generally injection molded while phase separated in which droplets of one phase are suspended in a matrix phase. The components are usually pre-mixed in an extrusion operation to form a finely dispersed mixture, and then injected into the molding cavity. Upon entering the cavity, the drops break-up into finer and finer sizes near mold walls where the shear rate is high, and coalesce/relax towards equilibrium sizes in regions of low shear near the center (***Figure 1***). This leads to a non-uniform "skin-core" microstructure in the final part, which highly affects the properties. The detailed morphology of the blend material after injection depends on the fluid mechanical deformation history, component rheological and interface properties, and blend thermodynamics. It is of great interest to develop a simulation that models not only the injection molding fluid mechanics, but also the evolution of the blend morphology during the injection.

Because there are many thousands of drops in a typical injection molding operation, it is not practical to use microscale flow modeling methods such as Lattice Boltzmann (1-2) or continuum surface force methods (3-6) to model individual drops. Instead, one must use methods that compute the average microstructure within a meso-volume that is larger than the length scale of the mixture, but much smaller than overall volume, e.g., Batchelor (7), Doi and Ohta (8), and Wetzel and Tucker (9-10). This approach is characterized by the use of an area (or interface) tensor given by

$$\underline{\underline{A}} = \frac{1}{V} \quad \hat{n}\hat{n}dS \qquad (1)$$

where $\hat{n}$ is the unit normal at the drop surface. This tensor contains information about the average local morphology of the mixture. The trace of the area tensor yields an important quantity, the specific area, which is the surface area per unit volume

$$S_V = tr(\underline{\underline{A}}) \qquad (2)$$

For spherical drops, the drop size radius is related to the specific surface by the relation

$$S_V = \frac{CS_{drop}}{V_{drop}} = \frac{3C}{R} \qquad (3)$$

where C is the drop concentration. It is evident from this relation that an increase in the specific area corresponds to an increase in the fineness of the microstructure. The evolution of the area tensor is governed by the expression

$$\dot{\underline{\underline{A}}} = \frac{1}{V} \quad \dot{\hat{n}}\hat{n}dS + \frac{1}{V} \quad \hat{n}\dot{\hat{n}}dS + \frac{1}{V} \quad \hat{n}\hat{n}d\dot{S} \qquad (4)$$

In addition, the area tensor is used in the formulation of the blend constitutive law.

While the area tensor approach gives insight into the behavior of blends, the problem is that there does not exist at this time a formulation of Eq. (3) that accounts for both surface tension and drop breakup, both of which are very important phenomena in blends processing.

The goal of this work is to put together a simulation that predicts drop size distribution and morphology in injection molded polymer blends. To overcome some of the theoretical difficulties involved in the area tensor approach, we have devised an approach that combines direct experimental measurements with simulation. The first activity is a measurement program in which we measure the drop size and aspect ratio of the droplets in blends as a function of shear rate and composition. In addition, the shear viscosity as a function of composition is measured. As a first order model, we use this data by adding custom subroutines for viscosity and drop size to the commercial fluid dynamics package FIDAP. From the computed shear rates, the drop size distribution in the mold is mapped.

Experimental

Viscosity and Droplet Size Measurement

Viscosity data for a 45/55 blend of Polyisoprene /Polybutadiene was measured at 130°C in a Rheometrics Scientific SR-5000 rheometer (11) in a parallel-plate geometry under steady shear, with 25 mm diameter fixtures and a 0.4 mm gap thickness. The temperature was controlled to within ± 0.5 K, and the measurements were carried out under a nitrogen atmosphere. Data for the blend are shown in ***Fig. 2***. While the pure components are essentially Newtonian over the range of shear rates (***Table 1***), the blend is weakly shear thinning. Mean droplet size as a function of shear rate at 130°C for the same blend was measured using a shear microscope (12), and data are shown in ***Fig. 3***. The length of the major axis (flow direction) and the aspect ratio of the major to the minor axis (vorticity direction) are both plotted vs. the shear rate. The data show that the drops elongate slightly but do not break up until a shear rate of 1 s^{-1} is reached. Substantial break-up occurs between a shear rate of 1 s^{-1} and 10 s^{-1}. Modestly deformed droplets orient at 45° in the flow-gradient plane. As they deform further, this angle tends to decrease.

Numerical Model

The model was implemented using the computational fluid dynamics package FIDAP. User subroutines for calculating the drop size, and drop aspect ratio as a function of generalized shear rate (2^{nd} invariant of the shear tensor) were written and implemented per the instructions in the FIDAP user manual (13). For the drop size, the function

$$R_{drop} = \frac{50}{\sqrt{\frac{1}{2} tr\left(\dot{\underline{\underline{\gamma}}} \cdot \dot{\underline{\underline{\gamma}}}\right)}} \qquad (5)$$

was used for shear rates greater than 1 s^{-1}. For the aspect ratio, the function

$$A_{drop} = 3.9 \left(\frac{1}{2} tr\left(\dot{\underline{\underline{\gamma}}} \cdot \dot{\underline{\underline{\gamma}}}\right)\right)^{0.069} \qquad (6)$$

was used for shear rates greater than 0.005 s^{-1}.

Results for the injection of a linear 2-D channel are shown in ***Figs. 4-5***. ***Figure 4*** depicts the mold filling pattern with superimposed velocity field. ***Figure 5*** shows the contours of the drop size as the mold is filled. As expected, the model predicts a very small drop size at the wall, which gradually increases towards the center. The shear rates are within the range of validity of the drop size data. Since the aspect ratio changes very little we do not plot this quantity.

Discussion

This simulation represents a first order method for predicting morphology in polymer blend mixtures. It is only first order because it predicts that the drop size changes instantaneously with shear rate, whereas in actual systems, at a given shear rate it takes a finite time for break-up to occur as is apparent from ***Eq. (3)*** above. A natural question is what is the effect of the neglected dynamics on the calculation, and under what circumstances can we expect the present simulation to give accurate drop size predictions?

If we go back to our original depiction of the blend injection process, ***Fig. 1***, we expect that there are four dominating regions in these flows: the injection region, the flow front region, the wall region, and the central region. As incoming fluid passes out of the entrance region, we expect that a fully developed flow will begin to develop. Thus, as the fluid flows downstream will begin to relax in the central regions, and break up in the high shear regions near the wall. We expect that this central section will extend well up to the flow front region where due to stretching mechanisms, there are some additional complications. For injection moldings in which the aspect ratio of the flow direction to the part thickness is very large, we can well expect that the central section will dominate the flow. This is born out to some degree by the study of Tucker et al. (10) who did area tensor calculations (***Eq. (3)***) for a blend under conditions of passive mixing (no surface tension or breakup) being injected into a linear mold similar to that of ***Figs. (4-5)***. For a mold with aspect ratio of 20, the specific area (***Eq. (2)***) is substantially developed a quarter of the way down the injection channel.

In future work, we hope to incorporate dynamic effects into our work. We are in the process of studying dynamic effects through flow simulation and development of appropriate theory. However, for thin cavities with no recirculations, we expect that the first order model will yield adequate results away from the entrance and flow front regions.

Conclusion

An approach for modeling the drop size distribution in the injection molding of polymer blends is developed. The simulation directly uses experimental data correlated to functional forms in the FIDAP fluid dynamics package. As an example, experimental data for droplet size and shape in a Polyisoprene /Polybutadiene system was measured and used in an example simulation. From theoretical considerations, the simulation is expected to provide good estimates of drop size distribution for flows with large aspect ratios of flow length to thickness where entrance effects are expected to be negligible, and no regions of recirculation.

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11. Identification of a commercial product is made only to facilitate experimental reproducibility and to adequately describe experimental procedure. In no case does it imply endorsement by NIST or imply that it is necessarily the best product for the experimental procedure.

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Component	**Viscosity (Poise)**
Polyisoprene	18,300
Polybutadiene	2200

Table 1. Newtonian viscosity's of pure components. The components show less than a 10% variation in viscosity at the shear rates over which the drop measurements were carried out.

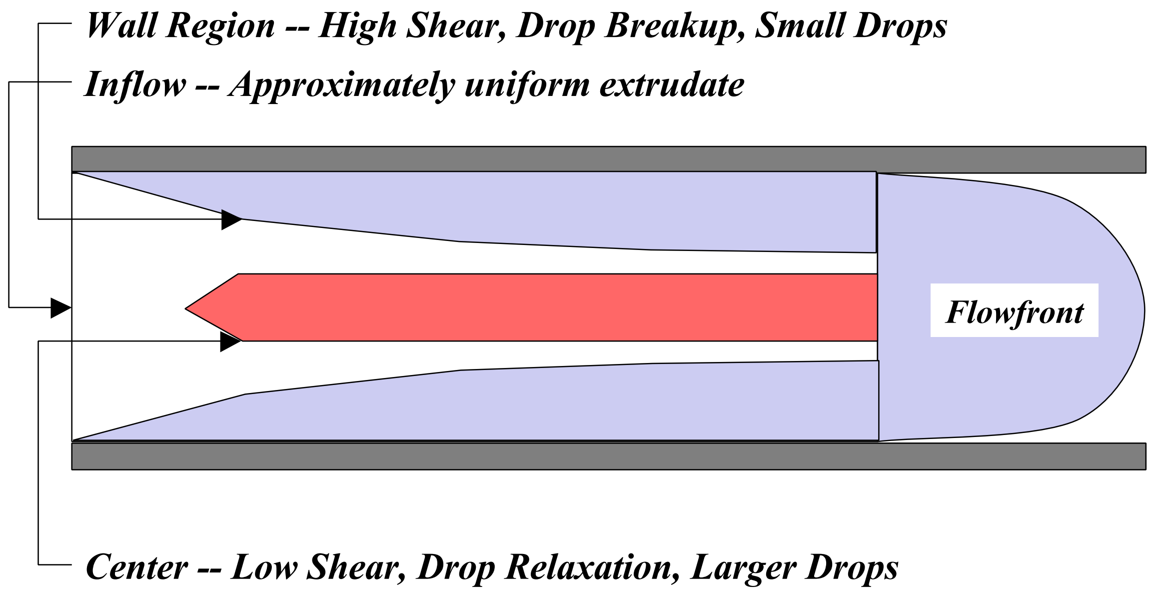

Figure 1. Depiction of the four flow regions in the injection molding of polymer blends.

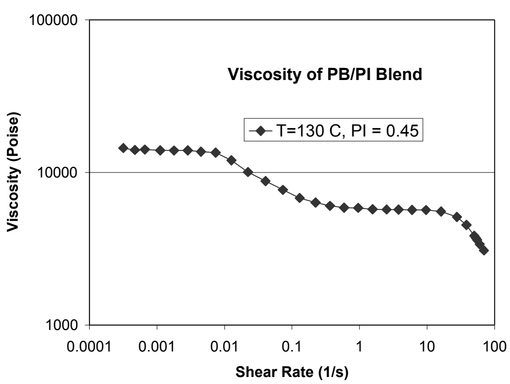

Figure 2. Viscosity vs. shear rate for a 45/55 mixture of Polyisoprene/Polybutadiene.

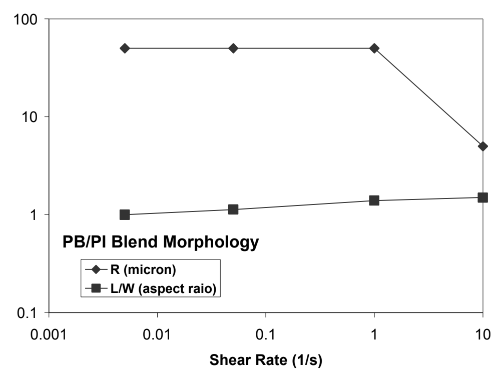

Figure 3. Drop size radius and aspect ratio vs. shear rate for a 45/55 mixture of Polyisoprene/Polybutadiene.

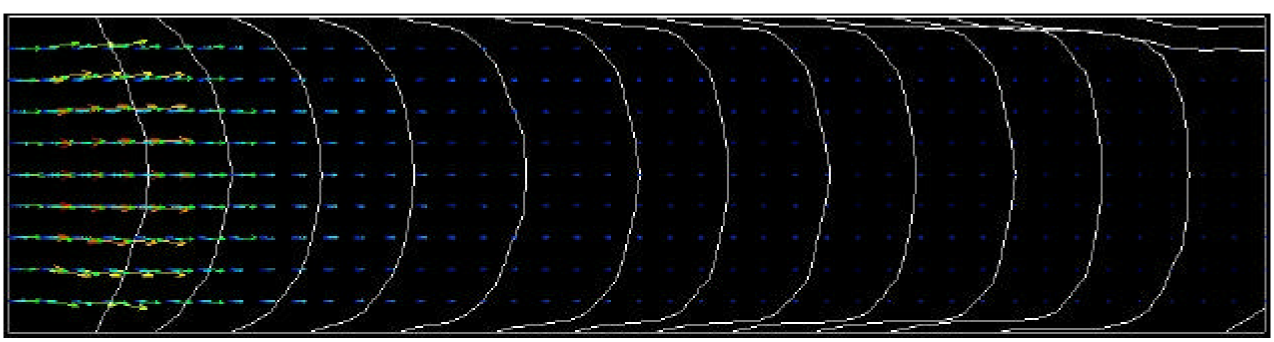

Figure 4. Flow front and superimposed velocity vectors for the filling of a small channel.

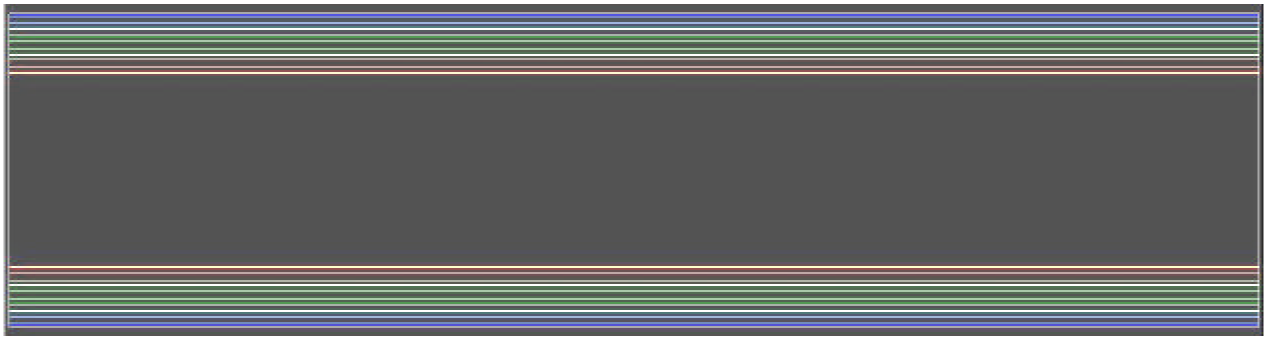

Figure 5. Drop size distribution for the filling of a small channel. The contour values are evenly distributed between values of 20 (center) and 18 (edge) microns.

Keywords: Polymer Blends, Injection Molding, Drop Size Distribution, In-situ Optical Microscopy