Droplet dynamics in sub-critical eccentric flows

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ABSTRACT: In this study single droplet dynamics for a Newtonian droplet in a Newtonian matrix have been observed in a controlled complex flow field. The mixed flow conditions, *i.e.* a combination of shear and extension, were applied using a newly designed eccentric cylinder device, and deforming droplets were visualised using optical microscopy. The experimental results on droplet deformation and orientation are compared with the predictions of the phenomenological Maffetone-Minale model. The model predictions are obtained by using the transient form of the model and incorporating a flow type parameter that accounts for the relative amount of shear and elongational effects. For all the sub-critical flows considered here, good agreement was found between the model predictions and the experimental data.

Key words: Polymer blends, Droplet dynamics, Complex flows, Eccentric cylinders

1 INTRODUCTION

The deformation of liquid droplets immersed in an immiscible liquid has been pioneered by Taylor [1, 2], who designed both a four roll mill and a parallel-band apparatus for experiments in elongational and shear flow, respectively. This seminal work was followed by a multitude of experimental and theoretical studies on the behaviour of droplet dispersions (see for instance recent reviews by Tucker and Moldenaers [3], and Guido and Greco [4]). Most studies however, focus on the structure development in relatively simple flow conditions, being either extensional flow or simple shear flow. Since polymer processing operations often involve complex mixtures of shear and elongation, a systematic investigation of the morphology development in such complex flows is very relevant. Nevertheless, experimental set-ups, such as the four roll mill, in which shear and extensional flow components are present in a controllable way still remain relatively scarce [5].
In the current research we aim to study the morphology development in immiscible polymer blends in controllable complex flow fields. For this purpose we have designed and tested a new eccentric cylinder device (ECD). The flow between eccentric cylinders (see figure 1) is a good example of a 'real' complex flow since it provides a balanced mixture of shear and elongational components which vary along the streamlines. In the present work we studied the deformation and orientation of a single Newtonian drop suspended in a Newtonian matrix for a representative flow field generated in the new ECD. The experimental results are compared with the predictions of the phenomenological model of Maffettone and Minale [6], adapted to complex flows (see section 2).

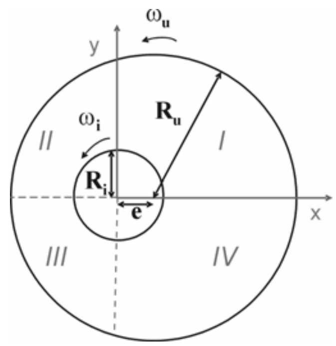

Fig. 1. Sketch of the top view of an eccentric cylinders system.

2 MODELING DROPLET DEFORMATION IN COMPLEX FLOWS

To predict droplet deformation and orientation in sub-critical complex flows, the transient form of the model of Maffetone and Minale [6] is used here. Maffetone and Minale developed a simple phenomenological model for the dynamics of a buoyancy free Newtonian drop suspended in an immiscible Newtonian fluid subjected to a flow field with an arbitrary velocity gradient tensor. To account for the complex nature of the flow field, a flow parameter $\bar{\alpha}$ is incorporated here according to Feigl et al. [7]:

$$\bar{\alpha} = \frac{\dot{\varepsilon}}{|\dot{\gamma}| + |\dot{\varepsilon}|} = \frac{\dot{\varepsilon}}{\bar{G}} \quad (1)$$

This parameter $\bar{\alpha}$ expresses the relative amount of elongation present in the flow field, where the flow strength $\bar{G}$ is the sum of the absolute values of shear and elongation rates, and $-1 \leq \bar{\alpha} \leq 1$. Mixed flow conditions between eccentric cylinders can thus be represented by the flow strength $\bar{G}(t)$ and the mixed flow parameter $\bar{\alpha}(t)$ which in general vary in time.
For a system with Newtonian components only two dimensionless parameters play a roll in slow flows: the viscosity ratio $p = \eta_d/\eta_m$, where η_d is the viscosity of the drop fluid and η_m the viscosity of the matrix, and the ratio of the viscous to the interfacial stress given by the capillary number $Ca = \eta_m R_0 E/\sigma$. Here, R_0 is the radius of the undistorted spherical droplet, and the macroscopic flow intensity E corresponds to the second scalar invariant of the strain rate tensor **D** [8], and equals $\sqrt{4\dot{\varepsilon}^2 + \dot{\gamma}^2}$ for 2D flows. In this investigation the strain rates $\dot{\varepsilon}$ and $\dot{\gamma}$ were obtained through FEM-simulations of the flow field with the software package PolyFlow®.

3 EXPERIMENTAL

3.1 Materials

The fluids used in this work are polyisobutylene (PIB Glissopal 1300 from BASF) as the continuous phase and polydimethylsiloxane (PDMS Rhodorsil 47V 60.000 from Rhodia) as the droplet phase. These are both transparent, Newtonian liquids at 23°C with respective viscosities of 51.6 and 60.4 Pa.s, the resulting viscosity ratio p being 1.17, and an interfacial tension σ of 2.8 ± 0.1 mN/m. All experiments were performed at ambient temperature (≈ 23°C). As the viscosity of PIB is very sensitive to temperature, the temperature of the sample was directly monitored by immersing a fine thermocouple needle in the continuous phase, and the corresponding viscosities were also used in the model calculations.

3.2 Methods

The experiments were conducted in the newly designed eccentric cylinder device (ECD); figure 1 shows a sketch of the geometry. In this case 'mixed' flow conditions, *i.e.* a combination of shear and elongational components, are obtained in the narrowing and expanding areas of the gap between the rotating cylinders. The inner and outer cylinders, with radii R_i and R_u and angular velocities ω_i and ω_u, can be displaced by a distance e, the eccentricity. The eccentricity ratio X of the system is defined as $e/(R_u-R_i)$. This is an important parameter to be adjusted in order to allow a variation of the relative magnitudes of shear and elongational strain rates along the streamlines. For our home built setup R_i and R_u equal 15 and 45 mm respectively, while the height h of the cylinders is about 30 mm. A more detailed description of the apparatus can be found elsewhere [9]. The applied flow field in the new ECD was also verified using *Particle Image Velocimetry* measurements (see also [9]).
For the experiments presented here the ECD was used in a configuration with the eccentricity ratio X equal to 0.2, and only the outer cylinder rotating with different constant speeds. After loading the flow cell with the PIB matrix, a single PDMS droplet was injected, in the widest part of the gap at a radial distance of about 30 mm from the axis of the inner cylinder. The initial radii of the drops were in the range of 250-500 μm. Next, the flow was started and the droplet was visualized using optical microscopy during start-up of the flow and after one or more revolutions when the drop reappeared in the field of view. Because of the limited field of view of the microscope system images of the deforming drop could only be taken in the first and second quadrant of the Cartesian coordinate system defined in figure 1. By using appropriate image processing techniques, it is possible to obtain the necessary information about the deformation and orientation of the droplets from these images, *i.e.* the length L of the long axis, the length B of the short axis and the angle θ between the long axis and the flow direction, as shown in figure 2.

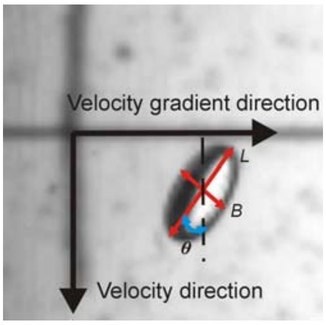

Fig. 2. Deformation parameters of a deformed droplet.

4 RESULTS AND DISCUSSION

For the controlled complex flow applied here, the shear and elongational strain rates vary in time along a streamline. A typical profile is given in figure 3 *a* which shows that both the shear and the elongation rate are periodic in time, with clear minima and maxima appearing during one revolution. These strain rate profiles will also give rise to a time dependent capillary number *Ca* and flow type parameter $\overline{\alpha}$, as represented in figures 3 *b* and *c*. Here t^* is the time made dimensionless with the characteristic time $\tau = \eta_m R_0/\sigma$. In this specific case, *Ca* follows the same qualitative behaviour as the shear rate, while $\overline{\alpha}$ logically shows the same trend as the extensional strain rate. The maximum amount of elongation is slightly less than 20 % of the total sum of strain rates $\overline{G}$ and hence, this complex flow is mostly shear dominated.

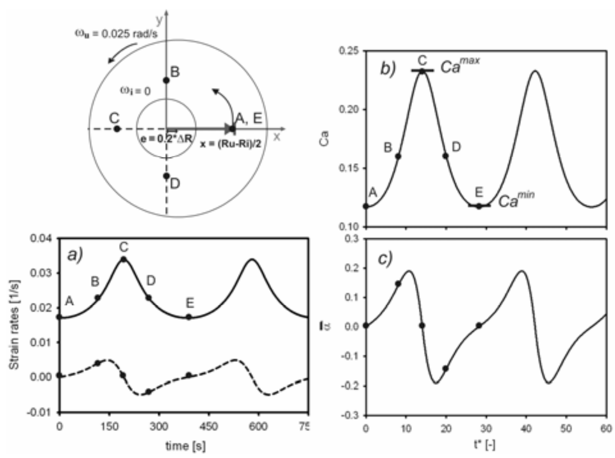

Fig. 3. Profiles for eccentric configuration with $X = 0.2$, $\omega_i = 0$ and $\omega_u = 0.025$ rad/s along streamline starting at $x = 30$ mm from center of the inner cylinder of: *a)* shear (—) and elongation (--) rates *b)* capillary number *Ca c)* flow type parameter $\overline{\alpha}$. Parts *b)* and *c)* are for a PDMS drop with initial radius of 375 μm at a temperature of 23°C.

Due to the fact that *Ca* varies in time with a clear maximum and minimum value during one revolution, the experiments for the eccentric flows are characterized by the values for Ca^{max} and Ca^{min}, as can be seen in figure 3 *b*. Figure 4 depicts the results for droplet deformation and orientation observed microscopically in the complex flow for different rotational speeds of the outer cylinder, corresponding to different ranges of capillary numbers. The symbols correspond to the experimental results while the lines are the model predictions of the Maffetone-Minale model adapted to complex flows. The experimental curves are characterized by a sharp rise ($L/2R_0$) and a steep fall (B/R_0, θ) of the deformation parameters during the start-up flow followed by a more gradual increase or decrease in the subsequent revolution(s). For the primary axes of the droplet, this course appears to level off in a maximum and minimum value respectively, while the orientation angle shows a clear minimum near the end of the second quadrant each time. When comparing with the model predictions in figure 4, these experimental trends can be recognized as parts of a time periodic oscillation in the deformation parameters, in response to the applied capillary numbers (figure 3). A good quantitative agreement is found between our experimental results and the model predictions. Although the experimental data could only be obtained within a limited part of the flow field, the repeating nature of the trends in subsequent rotations suggests the periodic behaviour of the deformation parameters. This steady oscillatory progress is also confirmed by the fact that the maximum and minimum values of the experimental parameters at the start of the first quadrant and the end of the second quadrant respectively, are approximately equal for subsequent revolutions.

Deviations only start to occur for the higher capillary numbers explored in figure 4. For example, for the highest capillary numbers the experimental results seem to indicate that the steady oscillation does no longer develop, in contrast to the model predictions. Instead, the deformation keeps increasing, though in an oscillating fashion. Hence, we are no longer in a sub-critical regime. From the data of Bentley and Leal [10] a critical capillary number Ca_{crit} for break-up in steady 2D flows with a constant, positive value of the flow parameter $\overline{\alpha}$ can be deduced for every viscosity ratio *p*. For a maximum value of $\overline{\alpha}$ around 0.20 (figure 3) and *p* equal to *ca.* 1.2 (see legend of figure 4) this amounts to a value for Ca_{crit} of about 0.49. Hence, the experimental results at the highest capillary numbers are expected to be above the critical conditions for break-up, where the model predictions are no longer valid.

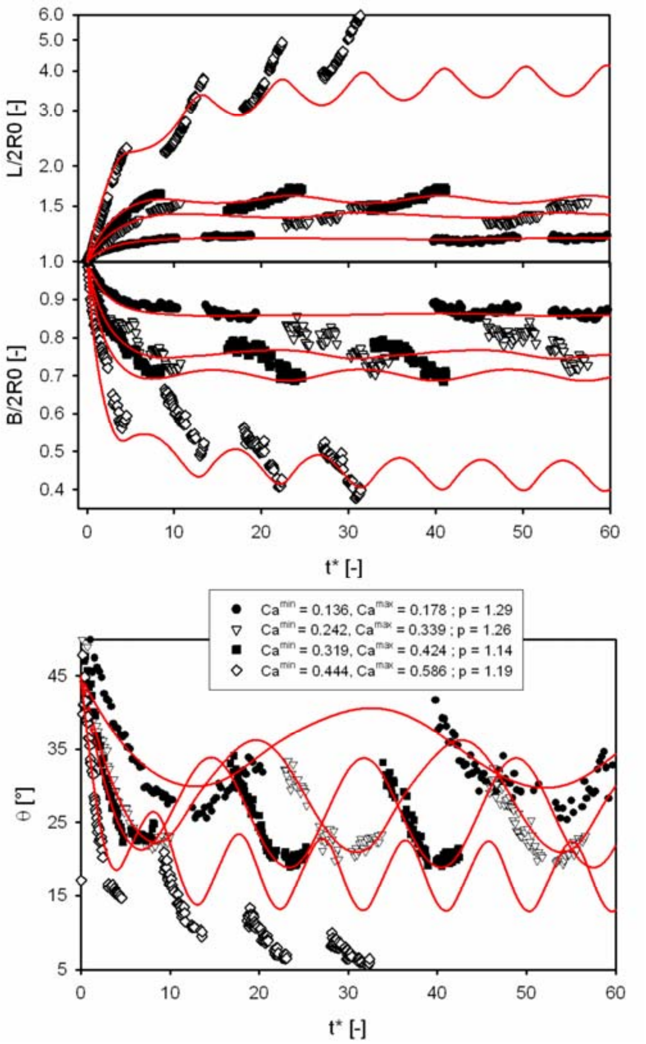

Fig. 4. Droplet deformation during sub-critical complex flows with eccentricity ratio $X = 0.2$, $\omega_i = 0$ and variable ω_u. Symbols represent experimental results whereas lines are predictions according to the Maffetone-Minale model.

5 CONCLUSIONS

The deformation and orientation of single Newtonian droplets dispersed in an immiscible Newtonian matrix undergoing sub-critical complex flow was investigated using a newly designed eccentric cylinder device for a range of capillary numbers below the critical conditions for break-up. The experimental results for droplet dynamics have been compared with model predictions obtained using the transient form of the model of Maffetone and Minale [6] and incorporating a flow type parameter that accounts for the relative amount of extension in the flow field. It was shown that under the sub-critical conditions the experimental results show very good agreement with the model predictions, providing a first quantitative assessment of drop shape predictions in transient, 'mixed' flow conditions.

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