INVESTIGATION AND VISUALIZATION OF DROPLET DEFORMATION AND BREAKUP IN COMPLEX LAMINAR FLOW FIELDS

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ABSTRACT

The flow field in industrial dispensing processes is a mixture of shear and elongational flow. Droplets thus experience transient rates of strain as they move through the flow field. The deformation and breakup behavior of drops in a fluid multiphase system is investigated experimentally and numerically. Finite element and numerical particle tracking techniques are used to obtain the history of shear and elongation rates along a particle trajectory in the flow field, and from this history, boundary integral techniques are used to determine the deformation. The effect of surfactant on a drop in steady flow is also determined using modified boundary integral techniques.

Keywords: Emulsion; Drop deformation; Rotor/stator; Multiphase flow; Visualization; Simulation

1 INTRODUCTION

The processing of highly viscous fluids composed of multiple immiscible phases is encountered in a wide range of fields such as polymers, foods, pharmaceuticals, and cosmetics. Laminar and dispersive mixing operations are used in order to generate a homogeneous microstructure of a product.

In industrial emulsification processes which are used for highly viscous fluid systems the flow field is often a superposition of shear and elongational flow. If the stresses are supercritical big drops are deformed in such flow fields and fragmented into smaller drops. A drop under these conditions experiences transient deformation rates along its way through the flow field.

The use of controlled and defined mixed shear and elongational flows allows for adjusting microstructure related quality characteristics in the flow process, which are governed by the rheological properties of the fluid [7]. Due to this knowledge the deformation and fragmentation behavior of liquid drops which are suspended in another fluid phase are investigated. This research [4] is carried out experimentally as well as numerically for drops experiencing a mixed flow field. To gain a better understanding of the relative efficiency of the dispersing process the deformation and breakup behavior of a single drop is visualized as it moved along a particle path in a mixed flow field. Different rotor/stator geometries are analyzed which are generally used for the processing of highly viscous systems [4]. The annular gap flow between eccentric cylinders is investigated as a first approximation and as an ideal flow field that produces transient shear and elongation rates. Complementary experiments are performed using variably shaped rotor/stator geometries that produce complex flow fields with superimposed shear and elongational flow components. The different geometries are evaluated considering their dispersing ability [4].

The concept of this project which deals with laminar drop dispersing processes comprises the use of a combined experimental and numerical approach:

- Determination of critical capillary numbers in stationary mixed flow fields (numerically)
- Simulation of flow fields and calculation of particle tracks (numerically)
- Determination of particle dynamics along various
particle tracks in the flow fields (experimentally and numerically)

- Determination of fragmentation results (experimentally)

Applying a boundary integral method (BIM) the numerical simulation of the drop deformation is used on one hand to determine stationary critical capillary numbers $C_a_{crit}$ in predetermined mixed flow fields as well as to quantify the influence of the elongational flow (planar, uniaxial, and biaxial). On the other hand, the influence of increasing elongational flow proportions in the flow field on the time dependent evolution of the droplet shape is investigated. Using a finite element method (FEM), the complex flow fields between various dispensing geometries are simulated and particle tracks as well as corresponding velocity gradients are calculated.

Experiments are carried out by introducing small drops at various starting positions or process parameters in the flow field, recording the resulting deformation of the drop along the particle path and the fragmentation result after cessation of flow. Different fragmentation results due to certain deformation behaviors can be related to different stress histories of the drop.

A quasi-stationary boundary integral method (BIM) is applied in a final step to simulate the transient dynamic deformation behavior of drops along different particle tracks using the corresponding velocity gradients. These results are finally compared to experimental data.

To study the influence of a diffusing surfactant on the drop deformation, the boundary integral approach reported in [2] has been extended and first results for stationary flow conditions are presented.

2 EXPERIMENTAL METHODS

2.1 Model fluids and their characterization

The experiments were carried out at 25 °C using a 50:50 (w/w) mixture of Wacker Silicone Fluids AK 2000 and AK 5000 containing 0.2% (w/w) of nano-scale titanium dioxide as the disperse phase. The shear viscosity of PDMS is $\eta_d = 3.184$ Pas, the density $\rho_d = 971$ kg m$^{-3}$. The titanium dioxide added to enhance the optical contrast proved to influence neither the density and viscosity of the disperse phase nor the interfacial tension.

The viscosity ratio was set at $\lambda = \eta_d/\eta_c = 2.0$. When using a ternary system consisting of PEG, H$_2$O and EtOH, the viscosity and density can be adjusted independently. Hence, a mixture of 35.15% (w/w) of poly (ethylene glycol) 35000 S, 30.49% (w/w) of ethyl alcohol, and 34.36% (w/w) demineralized water has been used for the matrix fluid. The shear viscosity is $\eta_c = 1.618$ Pas. The measured density of $\rho_c = 991$ kg m$^{-3}$ is small enough so that interfacial tension measurements are still possible while the densities of both phases are almost equal.

The interfacial tension of $\sigma_c = 9.74$ mN m$^{-1}$ was measured by the drop volume method using a computer-controlled measuring system [3].

2.2 Experimental setup and procedure

The experiments reported in this paper were performed using a rotor/stator flow cell. The experimental setup, as depicted in Fig. 1, consists of two disks that can be rotated independently by two servo motors. The rotational speeds of the motors are controlled by a personal computer. The set point of rotational speed is always reached within approximately 30 ms.

Different geometries can be mounted on the inner as well as the outer disc. In the experiments presented here, an asymmetric cylinder geometry has been used. This geometry is schematically shown in Fig. 2. The outer cylinder has an inner diameter of 290 mm, and the inner cylinder has an outer diameter of 240 mm. The offset between the two cylinders was set to 10 mm. The height of the cylinders is 34 mm. The inner and outer cylinders are immersed in a fluid bath. The inner cylinder is rotated e.g. at varying speeds ($n_i = 2.0$ rpm to $n_i = 3.5$ rpm) to create the desired flow field which is assumed to be two-dimensional. The shear and elongation rates for different particle tracks can be varied by changing the rotational speed of the inner cylinder.
Figure 2: Schematic drawing of the asymmetric cylinder gap geometry (top view).

Small drops (fixed volume of 20 µL, equal to an initial drop diameter of d₀ = 3.37 mm) were introduced into the gap using a syringe at 128.5 mm off the center of motion (position A, see Fig. 2). This was done using a positioning tool that guided the needle to ensure reproducibility of the starting conditions.

After placing the drop into the suspending fluid, the experiment was started. The inner cylinder started to rotate and at the same time a trigger signal was sent out to the video acquiring system, starting the frame grabbing process. The images of deforming and breaking drops were recorded using a CCD black and white video camera connected to an Apple Power Macintosh. The CCD camera was mounted directly above the rotor/stator flow cell and could be rotated to follow the movement of the drop.

3 NUMERICAL METHODS

The finite element program used for computing the velocity field and the numerical particle tracking program used for computing shear and elongational rates along particle paths is SEPRAN [6].

The deformation behavior of a droplet in steady flow as well as along a particle path in a given flow field is additionally simulated using numerical calculations. These numerical calculations are performed using a computer program based on the boundary integral method (BIM). It was written by M. Loewenberg and co-workers [1, 5]. For details and prerequisites please refer to [2].

The velocity gradient $\nabla \mathbf{v}$ which describes the mixed flow experienced by the drop at any position of the flow field is defined as (if the extensional flow is uniaxial):

$$\nabla \mathbf{v} = G \alpha \begin{pmatrix} 1 & 0 & 0 \\ 0 & -0.5 & 0 \\ 0 & 0 & -0.5 \end{pmatrix} +$$

$$+ G (1 - |\alpha|) \begin{pmatrix} 0 & 1 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

(1)

$G$ is the sum of imposed shear and elongation rate and $\alpha \in [-1,1]$ is the flow type parameter. Simple shear flow corresponds to $\alpha = 0$ and uniaxial elongational flow corresponds to $\alpha = 1$. In converging/diverging channels negative elongation (i.e. compression) also has to be taken into account. Therefore, $\alpha$ has been defined in such a way that $\alpha = -1$ represents uniaxial compression, i.e. biaxial elongation.

The capillary number $Ca$ represents a dimensionless deformation rate and is the ratio of viscous force to interfacial tension. $Ca_{crit}$ is the specific value which splits the non-breakup from the breakup regime. $Ca$ can consequently be defined for steady mixed flow using the radius of the undeformed drop $d_0$, the viscosity of the continuous phase $\eta_c$ and the interfacial tension $\sigma_s$ ($t_{dr}^{*}$ is called the drop characteristic or drop relaxation time):

$$Ca = G \frac{d_0 \eta_c}{\sigma_s} = G t_{dr}^{*}$$

(2)

4 RESULTS AND CONCLUSIONS

4.1 Effect of the flow type parameter $\alpha$ on the critical capillary number

In steady flow, there is a critical capillary number, $Ca_{crit} = Ca_{crit}(\lambda, \alpha)$, such that a drop breaks up when $Ca > Ca_{crit}$ while for subcritical values, the drop does not break up but instead deforms until it reaches a stationary shape. Fig. 3 shows the critical capillary number obtained from numerical simulations as a function of the viscosity ratio $\lambda$ for six different flow type parameters $\alpha$. The velocity gradient has the form given in Eq. (1) where $0 \leq \alpha \leq 1$.

This figure illustrates the well-known fact that, under steady conditions, uniaxial extensional flow is more efficient in dispersing than shear flow, in the sense that a lower elongation rate than shear rate is needed to break up a given drop in a given fluid. For example, the critical capillary $Ca_{crit}$ number for a viscosity ratio of $\lambda = 1$ in uniaxial extensional flow is 0.12, while in shear flow it is 0.43. This discrepancy increases for larger values
3.5 rpm
3.0 rpm
2.5 rpm
2.0 rpm

Figure 3: Effect of the viscosity ratio $\lambda$ on the critical capillary number $Ca_{\text{crit}}$ for different steady flow type parameters $0 \leq \alpha \leq 1$ (uniaxial).

Figure 5: Capillary number $Ca$ as a function of the dimensionless time $t^+ = t/t^*$ along the particle track with starting position A and ending position H.

Figure 6: Fragmentation result: influence of the rotational speed on the cumulative number distribution after drop breakup (diameter of the undeformed drop $d_0 = 3.37$ mm).

of $\lambda$. The figure also shows the effect of adding certain amounts of uniaxial extensional flow to shear flow. From these results, the lowering of the critical capillary number can thus be quantified.

4.2 Influence of the rotational speed on the drop breakup

Experiments have been carried out varying e.g. the rotational speed of the inner cylinder while the starting conditions (droplet size, fluid system, starting position A) were kept constant. Fig. 4 shows the images of four different drops at position H.

The droplet either deforms until it reaches a steady shape (dependent on the position in the flow field) in case of $Ca < Ca_{\text{crit}}$ or it continues to deform until it finally breaks up when $Ca > Ca_{\text{crit}}$. The evolution of the capillary number $Ca$, calculated from Eq. 2 using numerical ($G$) and experimental ($t^*$) data, along the same particle track (starting position A and ending position H) for different rotational speeds is shown in Fig. 5. For comparison, the critical capillary number in simple shear flow $Ca_{\text{crit}}(\lambda = 2, \alpha = 0) = 0.62$ is also shown in Fig. 5. At 2.0 rpm the drop did not experience a supercritical stress and thus relaxed back to its spherical shape. Applying a rotational speed of 2.5 rpm causes the drop just to break up. The stretching of the flow is slow enough so that a neck is formed and two large drops pinch off with one daughter drop in between (which is again surrounded by two satellite drops). The resulting droplet size distribution is therefore quite broad, see Fig. 6.

Further increase of the rotational speed (and therefore Ca) results in a longer filament in between the drops at both ends that are pinching off. A breakup of filaments due to Rayleigh instabilities can be detected in the diverging part of the geometry as well as after the experiment is stopped when the stretched drop reaches point H. The higher the rotational speed, the thinner the filament and the smaller and the more uniform the resulting fragments.
5 Surfactant-covered drops

If surfactants are present on a drop, then the deformation and breakup behavior of the drop is altered. This is due to the fact that surfactants lower the interfacial tension and may cause spatial gradients in interfacial tension due to spatial variations in the local surfactant concentration on the drop. To simulate the deformation of a surfactant-covered drop, two additional equations are added to the governing system for surfactant-free drops, and a change is needed to the equation describing the jump in surface traction.

First, a convection-diffusion equation is added to describe the change in the surfactant concentration on the drop surface as the drop is subjected to a flow. In dimensionless form, this equation has one dimensionless parameter

$$\gamma = \frac{\tau_0 \sigma_x}{\eta c D_s} = \frac{Pe_s}{Ca_s}$$

where $\sigma_x$ is the interfacial tension of the clean interface, $D_s$ is the surface diffusivity, and $Pe_s = G \tau_0^2 / D_s$ and $Ca_s = \frac{\tau_0 \sigma_c G}{\sigma_x}$ are the surface Péclet number and capillary number for a clean interface (= Ca), respectively. Small $\gamma$ corresponds to diffusion-dominated surfactant mass transfer, while large $\gamma$ corresponds to convection-dominated transfer.

The second equation added to the governing system is a model to describe the relationship between the surfactant concentration, $\Gamma$, and the interfacial tension, $\sigma$. That is, an equation of state, $\sigma = \sigma(\Gamma)$, is needed. We have considered a linear equation of state of the form

$$\sigma = \sigma_s (1 - \beta \Gamma),$$

where $\beta$ is a constant defined as $\beta = (\tau_0 R T) / \sigma_s$, where $R$ and $T$ are the gas constant and absolute temperature, respectively. Note that $0 \leq \beta \leq 1$ and $\beta = 0$ corresponds to no surfactant. Also, the larger the value of $\beta$, the faster the interfacial tension decreases with increased surfactant concentration, since $\beta = -d(\sigma / \sigma_s) / d\Gamma$. Initially, the dimensionless surfactant concentration is $\Gamma = 1$, so that the equilibrium interfacial tension is $\sigma^* = \sigma_s (1 - \beta)$ and the corresponding equilibrium capillary number, $Ca^*$, is related.

4.3 Numerical simulation and experimental verification

In order to test the ability of our numerical approach to predict drop breakup in a real dispersing device, simulations were performed using the geometrical parameters of the experiment described above. In the BIM calculations, we assumed a viscosity ratio of $\lambda = 1.97$ and a drop relaxation time of $t_{dr} = 0.322$ s, following the suggestion of V. Cristini by increasing the drop relaxation time for the numerical simulation [1]. The comparison of droplet shapes (experiments as well as simulation) at different positions along the particle track for a rotational speed of $n = 3$ rpm is shown in Fig. 7.

The predicted shape of the drop during its evolution generally agrees well with the experiment up to position E. In order to gain a state of high deformation, the simulation predicts a somewhat longer time than the experiment, in which a long thread is formed between position E and G. The discrepancy in the amount of drop deformation comes primarily from simulation assumptions and numerical as well as experimental errors. V. Cristini states that under near-critical conditions, drop dynamics are sensitive to numerical errors [1]. It must be mentioned that the code used in this work is not capable of simulating the drop detachment.
to the capillary number, $Ca_s$, of the clean interface via $Ca^* = Ca_s/(1 - \beta)$.

Finally, the equation which describes the jump in surface traction across the interface is altered to include a term accounting for surface gradients of interfacial tension.

The entire set of modified equations is solved iteratively using the following three steps: (1) Given a drop shape and surfactant concentration, compute the interfacial velocity from the boundary integral equations. (2) Update the drop shape by integrating the kinematic condition, $dx_s/dt = (u \cdot n)n$, where $x_s$ represents points on the interface. A second-order Runge-Kutta scheme is used. (3) Update the surfactant concentration by solving numerically the convection-diffusion equation on the drop surface. A finite element method using linear interpolants on the triangular mesh describing the drop surface is used.

Fig. 8 illustrates the effect of a diffusing surfactant on drop deformation in uniaxial elongational flow. The surfactant mass transfer is governed by diffusion only, i.e. $\gamma = 0$. The viscosity ratio is $\lambda = 1$, and several values of $\beta$ are considered. The drop deformation is quantified by $L/\ell_0$, where $L$ is the half-length of the drop along its primary axis.

The equilibrium capillary number is kept constant at $Ca^* = 0.12$, which is slightly subcritical for $\lambda = 1$ and surfactant-free drops. By fixing $Ca^*$ instead of $Ca_s$, we are able to see the effect of the diffusion of the surfactant only, i.e. the effect above that which comes from the mere presence of the surfactant on the interface. The trend observed in these graphs is in agreement with other numerical results. Of course, at a fixed $Ca_s$, a drop with surfactant will deform more than a drop without surfactant, since the surfactant will decrease interfacial tension.

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