Experimental and numerical studies on the controlled liquid-liquid breakup in complex flow fields

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INTRODUCTION

The dispersing of one liquid into a second immiscible liquid at high viscosity ratios is a very important unit operation associated with lots of industrial processes. Laminar distributive and dispersive mixing operations are used in order to generate a homogeneous disperse microstructure of the product. In real dispersing processes the flow field is not steady simple shear or steady uniaxial extensional flow, but is always a mixed flow. Moreover, the single droplet in such a flow field experiences transient rates of shear and elongation as it moves along its trajectory through the flow field. Assuming Newtonian fluid phases, negligible inertial effects and constant interfacial tension, the disruption behavior of liquid-liquid systems is investigated in well-known flow fields. As a consequence, we are able to observe the deformation behavior of a single droplet or a liquid thread as a function of time, shear and elongational rate. The detailed description of the transient velocity field is obtained from CFD calculations (Sepran) while numerical investigation using a Boundary Integral Method (BIM) is used to calculate the droplet deformation on a known particle path and velocity [1, 2]. In comparison to experiments, the effect of processing parameters, such as geometry, deformation and flow rates, and the impact of fluid parameters, such as the viscosity ratio and interfacial tension, on the liquid-liquid disruption is presented. The effect of capillary instabilities of the liquid-liquid interface is discussed for the thread breakup [3].

EXPERIMENTAL METHODS

The experiments were carried out using the silicone oils including 0.2% (w/w) of titanium dioxide as the disperse phase. Because of the fact, that in the numerical calculations equal densities are assumed, the outer phase had to be density and viscosity adjusted. Therefore, a ternary system consisting of PEG, H2O and EtOH was chosen that allows to adjust viscosity and density independently by changing the ratio of the ingredients. The viscosity ratio was chosen to be 1 † λ † 10. Surface and interfacial tensions were measured by the drop volume method [4]. The experiments reported in this paper were performed using a rotor/stator flow cell and a channel flow geometry. In the first case, the experimental setup consists of two rotating disks that can be driven independently by two servomotors. In our experiments, an eccentric cylinder geometry has been used as shown in Fig. 1. The inner cylinder is rotated at different speeds to create the desired flow field, which is two-dimensional.

Fig. 1: Schematic of the eccentric cylinder.

The rectangular shaped channel geometry was used to study the liquid-liquid thread breakup. In a flowing continuous phase, the dispersed phase is injected via a needle as depicted in Fig. 2. The formation of the droplet and breakup of the thread was observed with a CCD camera connected to a PC.

Fig. 2: Channel flow cell.

Detailed description of the experimental setup as well as the used numerical code SEPRAN and BIM is given elsewhere [1-3]

DROP BREAKUP IN ECCENTRIC CYLINDERS

Experiments have been carried out varying e. g. the rotational speed of the inner cylinder while the starting conditions (starting position A) were kept constant. Fig. 3 shows the images of four different drops at position H. The droplet either deforms until it reaches a steady shape in case of Ca < Cacrit or it continues to deform until it finally breaks up in case of Ca > Cacrit. At 2 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 breaks up the drop. The stretching of the flow is that slow, that a neck is formed and two large drops pinch off with one daughter drop in between. Increasing the rotational speed results in a longer filament in bet-
ween the drops at both ends that are pinching off. These filaments then break up due to capillary instabilities in the diverging part of the geometry or after stopping the experiment when the drop reaches point H. The higher the rotational speed the thinner the filament and the smaller and the more uniform the resulting fragments.

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. We see that the simulation correctly predicts that the drop does not break up at a rotation speed of 2 rpm and does break up at a 3 rpm. The predicted shape of the drop during its evolution generally agrees with the experiment at the lower rotational speed and up to position E in the experiment with the higher rotational speed. The discrepancy for drop deformation comes primarily from simulation assumptions and numerical errors [1]. In ongoing calculations the effect of varying the material properties is under investigation to obtain better agreement between simulation and experiment.

![Fig. 3: Drop deformation and breakup in asymmetric cylinder as a function of different rotational speeds.](image)

**LIQUID THREAD BREAKUP MECHANISM**

As seen in Fig. 3 the breakup of an extended droplet (thread) is promoted when the deforming shear and extensional stresses are relaxing towards zero or to a less dominant value. Depending on the viscosity ratio and the interfacial tension of the system the breakup of a flowing liquid thread surrounded by a second flowing liquid is governed by capillary instabilities. The competition between the interfacial tension driven wave growth and the counter-acting forces such as wave stretching or viscosity determines which wavelength will grow fastest and leads to break-up. This phenomenon of a *fatal wavelength* was described first by Lord Rayleigh in 1876 and later extended by e.g. Tomotika, Skelland et al., and Teng et al. to liquid-liquid threads at rest and liquid jets in air [5-7]. However, the injection of a liquid jet into another flowing immiscible liquid has rarely been investigated [3, 8]. The mathematical analysis leads to a dispersion equation (couple Navier-Stokes equation) for the growth rate of a sinusoidal disturbance, as a function of viscosity ratio and disturbance wavelength [6-7].

In the used flow cell, where the dispersed phase is injected via a syringe, the thread distortion is reached once the velocity difference across the interface is small, i.e. transient changes in the flow profile are not dominant any more. We were able to show that, similar to liquid jets, the viscosity is dampening the interfacial waves and, as a consequence, prolonging its lifetime. However, as shown in Fig. 4 the viscosity ratio, \( \lambda \), and not the viscosity of the dispersed phase is the more appropriate measure to characterize liquid-liquid thread breakup.

![Fig. 4: Thread breakup as a function of the viscosity ratio \( \lambda \) and flow rate of the continuous phase.](image)

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**REFERENCES**