DETERMINATION OF DROP BREAKAGE MECHANISMS BY EXPERIMENTAL AND NUMERICAL INVESTIGATIONS OF SINGLE DROP BREAKAGES

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Abstract. For an exact prediction of drop size distributions, the physical understanding of drop breakage and drop coalescence is needed. In this work, single drop breakage events, where coalescence can practically be neglected are, therefore, analyzed experimentally by high-speed imaging. The breakage events are induced by a single stirrer blade, part of a Rushton turbine. The influences of the drop diameter ($d_p = 0.7$-$3.1$ mm) and different dispersed phases (toluene, petroleum, paraffin oil), with and without the addition of sodium dodecyl sulphate, on the breakage event (breakage probability, location and time) are investigated. In order to determine the breakage mechanisms, the experimental results are compared with the flow properties of the continuous phase, gained by Computational Fluid Dynamics. The regions with the highest energy dissipation rates and the velocity field are compared with the experimental results - especially the drop breakage location. Depending on drop size and physical properties of the investigated systems, different breakage mechanism can be derived.

Keywords: drop breakage mechanism, liquid/liquid systems, Rushton turbine, Computational Fluid Dynamics

1. INTRODUCTION

Dispersing two immiscible liquids in a turbulent flow of a stirred tank is used in various processes, for example in the chemical industry. The drop formation and the resulting drop size distributions are thereby determined by drop breakage and coalescence.

Comprehensive scientific research identified a multitude of theoretical models describing both processes [1, 2]. Nevertheless the prediction of turbulent stirred systems, varying power input, material and process parameters, is only possible with restrictions. Consequently, the models for breakage and coalescence need further improvement. This work focuses on drop breakage.

Breakage occurs when the external forces and stresses exerted on drops by the continuous phase exceed the internal forces and stresses which hold it together [1, 3]. It is widely accepted to describe the maximum stable drop diameter and therefore drop breakage by means of considering the dynamic pressure forces of the turbulent motions applying Kolmogroff’s theory of turbulence. These dynamic pressure forces are caused by changes in
velocity around the particle. As the turbulent velocity fluctuations are a function of the local energy dissipation rate [3, 4], that can be used to describe drop breakage.

Besides the turbulent fluctuations, some authors describe the turbulent shear layer on the blade and the elongation flow along the impeller to influence drop breakage [5], while others distinguish between the turbulent fluctuations, viscous shear stress, shearing-off process and interfacial instabilities [1].

To investigate drop breakage experimentally, single drop breakage examinations where coalescence can be neglected have been widely used. Thereby, number and/or size distributions of broken droplets, breakage probabilities, times and/or deformations are determined experimentally, which allows a validation and improvement of drop breakage modelling and accordingly, knowledge of the occurring mechanism that leads to drop breakage (an overview of such investigations can be found for example in [6, 7]).

Besides, there are experimental investigations of drop breakages coupled with the local flow properties or rather flow stress in order to improve breakage modelling. For instance, drop breakage in a pipe downstream of an orifice was investigated considering the local flow properties gained by PIV measurements [8, 9]. Furthermore, Direct Numerical Simulation of that pipe were conducted and coupled with the resulting velocity field to a Lagrangian tracking of individual particles. The calculated locations of break up events were compared to experimental results. Detailed information on the external stresses experienced by the drops can be achieved by those simulations [10].

The aim of this work is to determine the breakage mechanisms depending on size and physical properties of the droplets. Therefore, single drop breakage events were analyzed experimentally in a special breakage channel [11, 12] and compared with fluid flow characteristics gained by Computational Fluid Dynamics.

2. MATERIALS AND METHODS

2.1 Experimental set up

For the experimental investigations a single blade of a Rushton turbine (D = 0.08 m) is mounted in a rectangular channel (30 mm x 30 mm). Drops (dp = 0.7-3.1 mm) are generated with a Hamilton® dosing pump PSD/3 in the lower part of the channel (see Fig. 1).

![Figure 1. Experimental set up and drop breakage for a toluene droplet (dp = 2.0 mm, u_fluid = 1.5 m/s, Δt = 5 ms).](image)

The drop breakage events in the vicinity of the stirrer blade are investigated by high-speed imaging. Pictures are taken with 822 frames per second. The breakage events are analyzed by automated image recognition.
In this work, the number of breaking drops is set into relation to the total number of investigated droplet sequences (at least 1000), to calculate the breakage probability \( P \). The breakage time \( t_{b,\beta} \) is defined to be the interval between the time when the drop passes the stirrer blade and the time when breakage occurs. Thereby, not the averaged mean value, but the maximum of an adapted \( \beta \)-distribution [7] is used to characterize the breakage time.

Pictures are taken with a resolution of 286 x 608 pixel and a scale of about 10 pixel/mm. The breakage location is calculated by averaging the x- and y-location of the centres of gravity of all the daughter drops after first breakage.

Three different dispersed phases with and without the addition of sodium dodecyl sulphate (SDS) in different concentrations are analyzed for this study (see table 1). All were blended with a non-water soluble dye (\( c = 0.075 \text{ g/L} \)) to increase the optical properties of the image data. For a more detailed description of the experimental set up and procedure, see for example [11, 12].

**Table 1. Physical properties of the investigated systems.**

<table>
<thead>
<tr>
<th></th>
<th>( \rho_d ) [kg/m(^3)]</th>
<th>( \eta_d ) [mPas]</th>
<th>( \gamma ) [mN/m]</th>
<th>( c_{SDS} = 0 \text{ mol/L} )</th>
<th>( c_{SDS} = 10^{-3} \text{ mol/L} )</th>
<th>( c_{SDS} = 10^{-2} \text{ mol/L} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Toluene</td>
<td>870</td>
<td>0.55</td>
<td>32</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Petroleum</td>
<td>790</td>
<td>0.65</td>
<td>38.5</td>
<td>31.3</td>
<td>7.6</td>
<td></td>
</tr>
<tr>
<td>Paraffin oil</td>
<td>850</td>
<td>125</td>
<td>62</td>
<td>39.9</td>
<td>13.8</td>
<td></td>
</tr>
</tbody>
</table>

2.2 Numerical set up

The numerical simulations of the flow in the channel are performed with an in-house flow solver FASTEST-3D. For the considered flow velocities at the stirrer blade of \( u_{\text{fluid}} = 1.0, 1.5 \) and 2.0 \( \text{m/s} \), the flow in the breakage channel around the blade has high Reynolds-numbers (\( Re \approx 20.000-30.000 \)) and is consequently turbulent. The turbulence is taken into account by performing steady RANS simulations with the standard high-Re \( k-\varepsilon \) model. Using this 2-equation model from Launder and Spalding, the turbulent fluctuations are averaged and the resulting term for the Reynolds stresses is modelled with the Boussinesq-hypothesis. The flow region is spatially discretized with a blockstructured grid with 1.000.000 control volumes \( (y^+ \approx 20-70) \).

To ensure a realistic inlet profile for the velocity and the turbulent quantities \( k \) and \( \varepsilon \), an additional periodic channel is simulated. During the simulation of the flow in the breakage channel, the flow through the periodic channel is simultaneously calculated. In every iteration a plane from the periodic channel is copied to the inlet of the breakage channel.

3. RESULTS AND DISCUSSION

3.1 Influence of the system properties

Experimentally determined drop breakage locations for a toluene droplet are depicted in the left part of Fig. 2. Besides a projection of the drop breakages on a 2-dimensional grid of the investigated region (A), a contour plot of the same data (B) and the results looking from a 90° shifted point of view (C) are shown. Considering both views, it becomes evident, that droplets are breaking most frequently directly after the stirrer blade. With increasing distance from the stirrer blade, the drop breakage probability decreases, which corresponds well with the local energy dissipation rate distribution (see right part of Fig. 2).

It becomes evident, that more droplets break on the left and on the right side of the stirrer blade, than in the centre of the channel, which corresponds to the entrance distribution of the droplets [12].

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For the low viscosity (petroleum) and the high viscosity (paraffin oil) dispersed phase, decreasing the interfacial tension (through adding SDS or increasing the concentration of SDS) leads to a widening of the region where drop breakage occurs (see Fig. 3, left part). The regions of greater relative breakage probabilities get stretched. As less energy is needed for breaking the droplets, breakages also occur at the upper end of the investigated area where lower energy dissipation rates are located. That goes along with increasing breakage probabilities (see table 2).

For the breakage time $t_{b\beta}$ (time between crossing the stirrer blade and drop breakage) no clear tendency can be seen (table 2). Further investigations are needed to analyze what happens with the droplets between entering the investigated region and drop breakage.

Comparing the two different droplet substances shows, that for the low viscosity dispersed phase (petroleum), the maximum of the relative breakage probabilities is located directly after the stirrer blade. In contrast, for the high viscosity paraffin oil, this region of greater relative breakage probabilities starts further behind the stirrer blade and no breakage events took place directly after the stirrer.

Drop breakages of the high viscosity system are not concentrated in the region of the highest energy dissipation rate. When deformation starts in that regions, which means that drop breakage initiation correlates with the maximum local energy dissipation rate, breakage finally occurs mainly in the region of the macro vortices behind the stirrer blade where gradients of the mean velocities are significant (see Fig. 3 on the very right side).

When comparing two systems with different dispersed phase viscosities but almost the same interfacial tensions (see table 1), it can be seen that increasing the viscosity leads to
significantly decreased breakage probabilities (see table 2). This can especially be seen when comparing the petroleum/water system without any SDS ($\gamma = 38.5 \text{ mN/m}$) to the paraffin oil/water system with a concentration of $c_{\text{SDS}}=10^{-3} \text{ mol/L}$ ($\gamma = 39.9 \text{ mN/m}$).

Nevertheless, the high viscosity droplets also break in the region with lower local energy dissipation rates at the upper end of the investigated area like it can be seen for the less stable low viscosity droplets. The breakage time $t_{b,\beta}$ is increased for the high viscosity system, which can be explained by the fact that no droplets break directly after the stirrer blade.

Table 2. Breakage probability $P$ and breakage time $t_{b,\beta}$ for the investigated systems ($d_p = 1.0 \text{ mm}$, $u_{\text{fluid}} = 1.5 \text{ m/s}$).

<table>
<thead>
<tr>
<th>$c_{\text{SDS}}$ [mol/L]</th>
<th>$P$ [%]</th>
<th>$t_{b,\beta}$ [ms]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Petroleum/water</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>58</td>
<td>4.7</td>
</tr>
<tr>
<td>$10^{-3}$</td>
<td>72</td>
<td>9.6</td>
</tr>
<tr>
<td>$10^{-2}$</td>
<td>85</td>
<td>6.2</td>
</tr>
<tr>
<td>Paraffin oil 100/water</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>26</td>
<td>8.5</td>
</tr>
<tr>
<td>$10^{-3}$</td>
<td>32</td>
<td>6.8</td>
</tr>
<tr>
<td>$10^{-2}$</td>
<td>50</td>
<td>10.8</td>
</tr>
</tbody>
</table>

3.3 Influence of droplet diameter

The smallest and thus most stable droplet has a distinct maximum close to the stirrer blade, where most of the droplets break (see Fig. 4). When increasing the droplet diameter, this area gets wider; the droplets break more homogeneously over the investigated area. That was already determined by Maaß et al. [12], who investigated two droplet diameters of a toluene/water system and who explained it by the surface forces of the drops. The smaller droplet is more stable than the larger one and so breakage occurs in the region with the highest energy dissipation rates, close to the stirrer blade. With increasing droplet diameter the region where droplets break is extended further behind the stirrer blade. That goes along with increasing breakage probabilities and increasing breakage times ($P = 50\%$, $t_{b,\beta} = 3.4 \text{ ms}$ for $d_p = 0.7 \text{ mm}$, $P = 82\%$, $t_{b,\beta} = 13.3 \text{ ms}$ for $d_p = 3.1 \text{ mm}$ [7]).

For the biggest investigated droplet ($d_p = 3.1 \text{ mm}$), the region with the maximum of breaking droplets does not start directly behind the stirrer blade. In this case the drop breakage initiation or rather the deformation process seems to correlate with the local energy dissipation rate. Furthermore, from this experimentally determined breakage probability distribution, it can be concluded, that with increasing droplet diameters the macro vortices behind the stirrer blade also should be considered for an exact description of drop breakage.

Figure 4. Experimentally determined influence of the droplet diameter on relative breakage probabilities (petroleum, $u_{\text{fluid}} = 1.5 \text{ m/s}$).
4. CONCLUSION

In this work single drop breakage events were analyzed experimentally by high-speed imaging. The influence of the droplet substance and diameter were determined. The experimental results were compared with the fluid flow properties, gained by CFD, to get a deeper understanding of the drop breakage mechanisms.

Experimentally determined drop breakage locations correlate well with the local energy dissipation rates for low viscosity systems (petroleum/water and toluene/water, \(d_p = 1.0 \text{ mm}\)). Drop breakage can be explained by the microstructure of turbulence, which means that turbulent fluctuations lead to drop breakage.

In contrast, when increasing the viscosity of the droplets significantly, the microstructure of turbulence can not be used to describe the experimental results exactly. The same was found when increasing the droplet diameter.

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5. REFERENCES