Experimental investigations in liquid-liquid dispersion system: Effects of dispersed phase viscosity and impeller speed

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Experimental Investigations in Liquid—Liquid Dispersion System: Effects of Dispersed Phase Viscosity and Impeller Speed
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1.0. INTRODUCTION
A liquid—liquid dispersion operation is required in industrial processes such as extraction, suspension polymerization, and multiphase reactions. The drop size distribution of a dispersion is related to the interfacial area which controls the amount of mass transferred between the phases. Therefore, accurate prediction of interfacial area in dispersions is important in determining the mass transfer and reaction rates.

Drop breakup and coalescence occur simultaneously during a liquid—liquid dispersion process, and the dynamic equilibrium between both processes determines the final drop size distribution of the dispersion. A drop in moving fluid experiences turbulent pressure fluctuations, and viscous stress which tend to cause drop breakup. The drop then resists the deformation by its surface force and internal viscous force if its viscosity is high. Drop coalescence occurs when two drops collide and combine into bigger drops, involving drainage and rupture of the intervening liquid film between the drops which is governed by the physical properties of the drops. The mean drop size and distribution are influenced by several factors, such as operating parameters of stirring process and properties of liquids which include viscosity and interfacial tension. An accurate prediction equation could be developed by understanding how mean drop size and distribution are by different factors to allow better control of drop size and uniformity of the distribution.

2.0. BACKGROUND
Available works in liquid—liquid dispersion have mainly focused on investigating effects of various influencing factors such as dispersed phase fraction, viscosity, and impeller design on the mean drop size and distribution. The main physical properties of liquids which influence mean drop size are interfacial tension and viscosity. Earlier works on liquid—liquid dispersion however have often been conducted for dilute and low dispersed phase viscosity in which the surface force dominates drops stabilization and the internal viscous force can be neglected. Usually, the outcome of these works led to the development of models or correlations which relate the Sauter mean diameter  to different influencing factors.

Most of the previous works involving prediction of drop size have applied the concept of turbulent energy cascade to estimate the maximum stable diameter related to the Hinze and Kolmogorov theory. On the basis of the theory, drops in the inertial region of turbulence have the maximum diameter which is related to the dimensionless Weber number  as shown in eq 1

\[
\frac{d_{\text{max}}}{D} \propto e^{-0.4} \propto \text{We}^{-0.4}
\]  

(1)

\[
\text{We} = \frac{\rho D^3 N}{\sigma}
\]

(2)

The mean energy dissipation rate at constant power number can be determined from power consumption  as shown in eq 3

\[
\frac{1}{e_{1}} = \frac{P}{\rho V T}
\]  

(3)

The  or mean flow which is characterized by the impeller tip speed ND is usually used to develop scale-up correlations for liquid—liquid systems, but the use of each of the parameters alone is not sufficient. Zhou and Kresta suggested that the interaction between energy dissipation rate and mean flow had to be considered, in which both  and  were included for a better correlation. They also claimed that the maximum energy dissipation rate should be used instead of  as  was more suitable to describe the breakup across different impellers. However, there are arguments whether one can easily and accurately measures . Therefore, scale-up correlations were

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developed in this work by relating $d_{32}$ to $c_T$ and the mean flow, NF.

Santer mean diameter ($d_{32} = \sum d^2 / \sum d$) is preferred in characterizing dispersion as it relates interfacial area of dispersed phase to its volume. A relation between $d_{32}$ and We is obtained based on the fact that $d_{32}$ is directly proportional to $d_{90}$ as proven by several researchers:

$$\frac{d_{32}}{D} = C_3 We^{-0.6} \quad \text{or} \quad \frac{d_{32}}{D} = C_4 We^{-0.6}$$

(4)

Equation 4 is a well-known relation and has been widely used to correlate $d_{32}$ for a dilute system with low dispersed phase viscosity. The constant $C_4$ in eq 4 needs to be evaluated experimentally where the published values in the literature are in the range of 0.05–0.08 at low dispersed-phase viscosity. Although several researchers have successfully applied eq 4 to correlate their data, some have discovered that the exponent of We in eq 4 should be modified at different operating parameters where the application of $d_{32} \propto We^{0.6}$ have been argued at high dispersed phase fractions. At high phase ratio, the value of the We exponent has been found to be lower than $0.6$. This is because the application of eq 4 is limited to a dilute and low viscosity system, based on the assumption that $d_{32}$ depends only on $d_{90}$ linearly. Without this assumption, Pacek et al. suggested that a general equation should be used where the exponent of We should be determined experimentally

$$\frac{d_{32}}{D} = C_4 (1 + \phi) We^{-0.6}$$

(5)

Equation 5 is applicable when the effects of viscous force are negligible, especially for an inviscid liquid. However, the viscous force has significant effects on the cohesive stress at high viscosity, which stabilizes the droplets and should be considered in the relation. The effects of viscosity can be included in the prediction model by introducing a dimensionless viscosity number $V_i$ which represents the ratio of viscosity to surface force:

$$V_i = \left( \frac{\rho_i}{\rho} \right)^{0.5} \frac{\mu}{\sigma}$$

(6)

The viscosity number is added into the $d_{32}$ relation to account for drop stabilization by both surface and viscous forces as shown in the relation:

$$\frac{d_{32}}{D} = C_4 We^{-0.6} \left[ 1 + V_i \frac{d_{32}}{D} \right]^{1/3}We^{1.6}$$

(7)

When the viscosity term in eq 7 is too small and approaching zero, the equation reverts to eq 4. The relation is limited to diluted dispersions ($\phi < 0.01$) but could be modified to account for the effects of dispersed phase fraction.

The effects of dispersed phase viscosity are important as it is one of the main factors that influence the final drop size besides interfacial tension. However, most works on liquid–liquid dispersion have focused on low viscosity dispersed phase which has a narrow range of applications, and studies at high dispersed phase are limited. Viscous and nonviscous drops undergo breakup process in different ways where the number of fragments formed after breakup is different. Therefore, $d_{32}$ is not only influenced by the rate of drop breakup but also the number and drop size of drop fragments produced after breakup. Relevant studies on effects of viscosity have been conducted in a surfactant-free system by Calabrese et al. and Podgorska and in a surfactant-stabilized system which focused only on drop breakup. The objectives of this research work are to investigate the effects of viscosity on drop breakup and mean drop size apart from obtaining the correlations relating $d_{32}$ to the influencing parameters. The studies were conducted at different power inputs to gain better understanding on the effects of viscosity on drop breakup.

Various grades of silicone oils with different viscosities (moderate to high) were used to study the effects of viscosity on mean drop size and distribution. Silicone oil was used as it exhibits nearly constant interfacial tension at different viscosities with its density not too much different from the density of the continuous phase used in this work. Since the works focused on drop breakup only, surfactant was added to the system to prevent coalescence. Different techniques are available for drop size measurement which can be categorized based on their measurement principles. Laser diffraction is one of the laser system techniques that yields fast and accurate measurements. It is based on the measurement and interpretation of angular distribution of light diffracted by the drops, referring to Fraunhofer diffraction theory. This technique was used in this work because of its versatility, broad dynamic range, and high reproducibility.

### 3.0. Experimental Procedures

Experiments were conducted in a clear cylindrical flat bottom stirred tank with an internal diameter of $T = 0.20$ m, equipped with four identical baffles with width equal to $T/10$ each. The tank was filled with distilled water as continuous phase to a height of $H = T$. A standard six-blade stainless steel Rushton turbine with diameter $D = 0.08$ m was used with bottom clearance equal to $T/5$. Lighting was provided by four fluorescent lights at the corners of the vessel to allow observation of the dispersion process. Various grades of silicone oils (20, 350, and 500 mPa s) supplied by Sigma Aldrich were used as dispersed phase. Silicone oils have a high refractive index which makes them suitable for measurement by laser diffraction and image analysis techniques. The interfacial tensions of the oils were measured using the du Nouy ring method. The properties of the oils are tabulated in Table 1.

<table>
<thead>
<tr>
<th>Properties (at 25 °C)</th>
<th>Silicone oil 20</th>
<th>Silicone oil 350</th>
<th>Silicone oil 500</th>
</tr>
</thead>
<tbody>
<tr>
<td>Viscosity, $\mu_i$ (mPa s)</td>
<td>20</td>
<td>350</td>
<td>500</td>
</tr>
<tr>
<td>Interface tension, $\sigma$ (mN/m)</td>
<td>0.017</td>
<td>0.082</td>
<td>0.0189</td>
</tr>
<tr>
<td>Density, $\rho$ (kg/m$^3$)</td>
<td>1001</td>
<td>970</td>
<td>970</td>
</tr>
<tr>
<td>Refractive index</td>
<td>1.39</td>
<td>1.403</td>
<td>1.403</td>
</tr>
</tbody>
</table>

Approximately 0.3% w/w of sodium dodecyl sulfate (SDS) was diluted by distilled water before being added to the oil. The temperature in the vessel was monitored during the mixing process to make sure that there was no significant temperature increase. The required amount of silicone oil (equivalent to $\phi = 0.01$) was added into the distilled water near the impeller region by using a syringe. The impeller speeds were varied from 300 rpm to 500 rpm, which are equivalent to Reynolds’ numbers in the range of 33,000 to 55,000. The Reynolds’ number is calculated as $Re = \rho \cdot ND^2 / \mu_i$. The minimum impeller
speed was chosen to be above the minimum speed required for a complete dispersion, and the maximum speed was chosen below the speed where air entrainment starts to occur in the vessel. The power number determined for the Rushton turbine was 5.8 in the turbulence region. The details of the experiments are shown in Table 2.

Table 2. Details of the Experiment

<table>
<thead>
<tr>
<th>experimental conditions</th>
<th>symbol</th>
<th>value</th>
</tr>
</thead>
<tbody>
<tr>
<td>mixing time, min</td>
<td>θ</td>
<td>0.08</td>
</tr>
<tr>
<td>impeller diameter, m</td>
<td>D</td>
<td>0.2</td>
</tr>
<tr>
<td>tank diameter, m</td>
<td>T</td>
<td>0</td>
</tr>
<tr>
<td>tank height, m</td>
<td>H</td>
<td>0.4</td>
</tr>
<tr>
<td>impeller clearance, m</td>
<td>C</td>
<td>0.067</td>
</tr>
<tr>
<td>baffle width, m</td>
<td>B</td>
<td>0.02</td>
</tr>
<tr>
<td>liquid height, m</td>
<td>Hₐ</td>
<td>0.2</td>
</tr>
<tr>
<td>dispersed phase fraction</td>
<td>θ</td>
<td>0.01</td>
</tr>
<tr>
<td>SDS concentration, wt %</td>
<td></td>
<td>0.3%</td>
</tr>
</tbody>
</table>

3.1. Drop Size Measurements by Laser Diffraction. Samples of the dispersion were collected at a fixed position near the impeller region where the rate of drop breakup is high. The samples were taken to a Malvern Mastersizer 2000 for drop size measurement process. The laser diffraction equipment is well-known and widely used in this area by several researches. It is capable of measuring drops in the range of 0.02–2000 μm with an accuracy of ±1%. The equipment requires the information on the refractive index of the dispersed phase (see Table 1). Drop size distribution curves and Sauter mean diameter dₛₐ for all dispersions were produced and determined from the measurement.

4.0. RESULTS AND DISCUSSION

4.1. Drop Size Distributions at Different Impeller Speeds. The drop size distribution curves at different impeller speeds were plotted as shown in Figure 2a,b,c. The curves show the range of drop sizes in the dispersion. In Figure 2 the curves shifted to the right where the size of the drops was larger as the dispersed phase viscosity increased. In most cases, the distribution curves for 20 mPas were much taller (see Figure 2) compared to 350 and 500 mPas which show that the drop sizes for low viscosity are more uniform and the drop size distribution also broadens as μₑ increases. Besides, at constant impeller speed, the size of the smallest drops decreased while the volume percentage increased as μₑ decreased. For example, at 400 rpm, the d₅₀ (50% of drops were below this diameter) for 20 mPas, 350 mPas, and 500 mPas was 82.93, 126.32, and 273.31 μm, respectively. Therefore, it shows that low viscosity produces dispersion with smaller drops compared to high viscosity, which leads to higher surface area. Similar trends were reported by Podgorski et al. and El-Hamouz et al. Since the interfacial tension differences between the three oils were minimal, the influence on drop-size distribution by reducing the rate of drop breakup comes from μₑ.

As the impeller speed increased, the curves shifted to the left which was toward the lower drop sizes range. This is because smaller and finer drops were produced and the size frequency increased at high impeller speed. It occurs as a result of increased in drop breakup rate when the power input to the system is increased. For example, at μₑ = 20 mPas, d₅₀ decreased as the impeller speed increased where d₅₀ at the impeller speed of 300, 400, and 500 rpm was 290.65 μm, 126.32 and 117.18 μm respectively. The observations were expected as an increase in the power input to the impeller leads to increase in shear and energy dissipation rate. In this condition, turbulent pressure fluctuation which is the main external force that causes drop breakup increases.

4.2. Effect of Dispersed Phase Viscosity on Mean Drop Size. The d₅₀ for every μₑ are tabulated in Table 3 at different impeller speeds. As seen in Table 3, larger d₅₀ was produced as μₑ increased from 20 to 500 mPas at the same impeller speed, leading to smaller surface area in the dispersion. The observation can be explained by how the dispersed phase viscosity affects the rate of drop breakup where drops are stabilized not only by the surface force but also the internal viscous force at high viscosity. The viscous force of the drops provide additional cohesive force, and thus more energy is required to overcome the cohesive force, reducing drop breakup. Drops with higher viscosity also have lower drop breakup probability, where the breakup events area in the stirred vessel is smaller, compared to drops with low viscosity due to the stability of the oil. 

4.3. Sauter Mean Diameter as a Function of We. A dimensionless size ratio (dₛₐ/D) was plotted against the dimensionless Weber number, We for every dispersed-phase viscosities as shown in Figure 3. The data fitted well into eq 4 with slightly smaller We exponents. The constants Cₑ and We exponents as in eq 4 were obtained for the three μₑ and are shown in Table 4.

The exponents of We obtained from the data were between −0.65 to −0.68 for low to high μₑ which were close to the theoretical value of −0.6 as in eq 4. As seen in Table 4, the value of Cₑ for μₑ = 20 mPas was in the range 0.05–0.081 that was reported in the literature for low viscosity as discussed earlier. It shows that the system satisfies the theoretical conditions which are based on a breakup dominant process. The constant Cₑ was increased to 0.174 and 0.364 as μₑ increased, values which were out of the range as the reported values were only for a low viscosity system. The increment of Cₑ was due to the increase in dₛₐ at higher μₑ. It can be concluded from the result that eq 4 could be applied in a high viscosity system with slight modifications of the values of the We exponent.
4.4. Effect of Impeller Speed on $d_{32}$ at Increasing Dispersed Phase Viscosity. The $d_{32}$ data for each $\mu_2$ was plotted against impeller rotational speed $N$ in Figure 4. It is observed that $d_{32}$ decreased as $N$ increased for every $\mu_2$. An increase in $N$ gives higher energy dissipation into the system where the energy is used to overcome the cohesive forces which act to keep the drops to its original shape. Thus, the rates of drop breakup increase as $N$ increases and smaller $d_{32}$ is produced. The behavior can also be explained based on circulation time, $\theta$, where it is related to $N$ by $\theta \propto 1/ND$. Therefore, the circulation time in the vessel becomes shorter as $N$ increases, resulting in more circulations or flows in the vessel. Therefore, the drops take less time to travel to the impeller region, which is the most important zone that controls drop breakup and where the rate of drop breakup is high.

In Figure 4, a straight line was used to fit each of the data sets in order to show the slope which represents the change in $d_{32}$ as a function of impeller speed. Interestingly, different slopes were obtained for different dispersed phase viscosities, which were $-0.149$, $-0.455$, and $-0.881$, where the largest slope was achieved by 500 mPas followed by 350 mPas. It means that high viscosity drops undergo larger change in $d_{32}$ as the impeller speed increases, although $d_{32}$ for high viscosity is

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**Table 3. Sauter Mean Diameter, $d_{32}$ for Different Impeller Speeds**

<table>
<thead>
<tr>
<th>Impeller speed, RPM</th>
<th>$\mu_2 = 20$ mPas</th>
<th>$\mu_2 = 350$ mPas</th>
<th>$\mu_2 = 500$ mPas</th>
</tr>
</thead>
<tbody>
<tr>
<td>300</td>
<td>91.66</td>
<td>170.89</td>
<td>256.75</td>
</tr>
<tr>
<td>350</td>
<td>68.71</td>
<td>117.63</td>
<td>228.36</td>
</tr>
<tr>
<td>400</td>
<td>64.48</td>
<td>96.41</td>
<td>169.67</td>
</tr>
<tr>
<td>450</td>
<td>60.35</td>
<td>77.56</td>
<td>114.11</td>
</tr>
<tr>
<td>500</td>
<td>55.58</td>
<td>77.12</td>
<td>94.04</td>
</tr>
</tbody>
</table>

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Full text is available at:

[http://pubs.acs.org/doi/abs/10.1021/ie5002845](http://pubs.acs.org/doi/abs/10.1021/ie5002845)