DROPLET BREAKAGE IN STIRRED DISPERSIONS. 
BREAKAGE FUNCTIONS FROM EXPERIMENTAL DROP-SIZE DISTRIBUTIONS

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Abstract—Transient breakage drop-size distributions have been experimentally measured using an image analysis technique. The transient distributions show self-similar behavior. The breakage rate and daughter-drop distribution functions have been determined using an inverse-problem approach which takes advantage of this self-similarity. The inverse-problem results show that the breakage rate is not a power law function of the drop size. The breakage rate is found to increase sharply with the drop size and the stirrer speed while decreasing sharply with increase in the interfacial tension. It is also found to decrease with increase in the dispersed phase viscosity, though the dependence on the viscosity is weaker than on the other variables. The daughter drop distribution was found to be relatively insensitive to the stirrer speed and interfacial tension, but was found to depend on the dispersed phase viscosity. As the drop viscosity increases, the breakage becomes more erosive in nature, leading to a broader size distribution of daughter drops. Generalized correlations for the breakage rate and daughter-drop distribution which account for the effect of physical properties and experimental conditions are presented. These relations will be very useful in predicting the drop-size distributions in stirred dispersions. Models for the breakage functions are compared with those determined in this study and the model predictions of the transient-size distributions are compared with the experimental data.

1. INTRODUCTION

Dispersions of one immiscible liquid in another are commonly found in chemical engineering operations. Examples of these operations include liquid-liquid extraction and dispersed phase reactions. These dispersions are commonly formed by mechanical agitation of the liquid-liquid systems. The size distribution of the drops and its dynamics in the dispersion play a very important role in the overall behavior of these systems. The processes of drop breakage and coalescence essentially determine the evolution of drop-size distributions in such dispersions. Quantitative understanding of drop breakage and coalescence rates is required to control the evolution of drop-size distributions and hence the behaviour of liquid-liquid systems. In this paper, we focus on the process of drop breakage in stirred dispersions.

The framework of population balances has been widely used to study the process of drop breakage. In the population balance approach, breakage events of each individual drop are taken into account, providing a mathematical equation for the evolution of the number density in drop size in a purely breaking dispersion. For a batch stirred liquid-liquid dispersion undergoing drop breakage, the balance equation can be written in terms of the cumulative volume fraction, \( F(v, t) \) as

\[
\frac{\partial F(v, t)}{\partial t} = \int_0^\infty \Gamma(v) G(v, v') \frac{\partial}{\partial v} F(v', t).
\]

Successful use of this equation to quantitatively predict the transient drop-size distributions requires knowledge of two key phenomenological functions: the drop breakage rate, \( \Gamma(v) \) for a drop of volume \( v \), and the cumulative daughter drop distribution, \( G(v, v') \) from the breakage of a parent drop of volume \( v' \).

There have been numerous experimental and computational studies of drop breakage in low Reynolds number (creeping flow), well-defined flow fields in the fluid mechanics literature. Typical examples of these types of studies are the papers by Rumscheidt and Mason (1962), Stone and Leal (1989) and Tjahjadi et al. (1992). These papers illustrate the complexity of the drop-breakage phenomenon even in these well-defined flow fields. While such studies contribute to some insight on drop breakage, they are clearly irrelevant for the behavior of dispersions. Studies have

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shown that the turbulence in a stirred vessel is inhomogeneous (see Cutter, 1966). This makes it very difficult to specify the flow field and hence, to use fluid mechanics to determine the drop-breakage functions, $\Gamma(v)$ and $G(v, v')$.

Another approach to determine the drop-breakage functions has been to directly observe breakage events in a stirred dispersion. Konno et al. (1983) used high-speed photography to observe drop breakage. The major drawback of this approach is that it is almost impossible to measure a sufficient number of breakage events to get a statistically meaningful result. Konno et al. measured less than a hundred breakage events.

Due to the difficulties with the above approaches to study drop breakage in stirred dispersions, investigators have tried other techniques to study the problem. Many investigators have modeled the drop-breakage process. These models can be divided into two categories: (1) Models to predict $d_{\text{max}}$, the maximum stable drop diameter. Knowledge of $d_{\text{max}}$ does not give any information on the dynamics of drop breakage. (2) Models for the breakage functions, $\Gamma(v)$ and $G(v, v')$. Examples of such models can be found in the works of Coulaloglou and Tavlarides (1977), Narsimhan et al. (1979), Namibiar et al. (1994) and Tsouris and Tavlarides (1994). All of the above models make many simplifying assumptions about the flow field and the nature of breakage. The models for the breakage functions have also not been rigorously tested and their predictions compared with experimental data. Namibiar et al. have only compared their model with long time, equilibrium size distributions. In the paper by Tsouris and Tavlarides, models for the breakage functions and for the coalescence frequency have been presented and their combined predictions compared with experimental data. Namibiar et al. have only compared their model with their predictions compared with experimental data in which both breakage and coalescence processes take place. This is a much less rigorous test of the breakage model than testing the model with pure breakage experimental data.

Ramkrishna and coworkers (Ramkrishna, 1974; Narsimhan et al., 1980, 1984) have developed an inverse-problem technique to determine the breakage functions from experimental transient drop-size distributions. In the inverse-problem technique, the experimental distributions are used as the input to the population balance equation, eq. (1), to calculate the unknown breakage functions. In the 1980 and 1984 papers, Narsimhan et al. were able to get good estimates of the breakage rate, $\Gamma(v)$, but had difficulties in determining the daughter-drop distribution, $G(v, v')$. This is because solving for $G(v, v')$ is an ill-posed problem and special mathematical techniques are required to solve it. In a companion paper, Sathyagal et al. (1994) have incorporated such techniques to formulate a new inverse-problem procedure to determine the breakage functions. Sathyagal et al. (1994) have given the computational details of the inverse-problem procedure and have also given examples where the procedure has been used to successfully determine the breakage functions.

In this paper, we apply the new inverse-problem technique to determine the breakage functions from experimental size-distribution data. Experiments have been performed with different dispersed-phase systems and under different operating conditions. We examine the effect of varying physical properties, such as viscosity and interfacial tension, and operating parameters, such as stirrer speed, on the breakage functions. We compare the extracted breakage functions with those obtained from the phenomenological models. We also test the model functions by comparing their predictions with the experimental data.

2. SIMILAR BREAKAGE

Determination of the breakage functions, $\Gamma(v)$ and $G(v, v')$ from experimental transient drop-size distribution measurements via the population balance framework [eq. (1)] is referred to as the inverse problem of breakage. The problem as stated is still difficult to solve because the size distributions are functions of both drop size and time. Concepts of self-similar breakage can be used to help us gain an insight into the breakage process and hence make the problem more tractable.

Self-similar breakage has been widely discussed in theoretical papers on particle breakage (see, for example, Cheng and Redner, 1990; Ziff and McGrady, 1985). In these papers, the scaling transformation used is $z = v^\zeta \langle v'^{-1} \rangle(t)$, where $\langle v'^{-1} \rangle(t)$ is an average drop size. Analysis of this scaling form shows that the only time-independent breakage functions which will allow this form of self-similar breakage are $\Gamma(v) = v^n$ and $G(v, v') = g(v/v')$.

Ramkrishna (1974) used the similar breakage assumption of the form $G(v, v') = g(v/v')$ and calculated the breakage rate power-law exponent, $n$, from the limited experimental data of Madden and McCoy (1969). Narsimhan et al. (1980) generalized the concept and proposed the following form of self-similar breakage:

$$G(v, v') = g \left( \frac{\Gamma(v)}{\Gamma(v')} \right).$$  \hspace{1cm} (2)

As explained by Narsimhan et al. (1980), the form given by the above equation allows larger drops to break more thoroughly than smaller drops. Broadly, eq. (2) postulates that drop break-up occurs so as to promote the more breakable ones among the broken fragments. It also admits the similarity transformation $F(v, t) \rightarrow f(\zeta)$, where $\zeta = \Gamma(v)/n$. Significantly, the above similarity form is not constrained to power law breakage rates. Hence, non-power law breakage rates can show self-similar behavior if the assumption of similar breakage, eq. (2), is valid.

Sathyagal et al. (1994) have given the computational details of a test to determine if the experimental data show the above similarity. This test is based on the arc-lengths of the log($t$) vs log($v$) curves at constant $F(n, t)$. If the test for similarity is positive, the experimental data is self-similar. A positive result for
the test could also be used to calculate the breakage rate up to a multiplicative constant. By redefining the similarity variable as \( \zeta = [\Gamma(v)/\gamma] t \), where \( \gamma \) is the unknown multiplicative constant in the breakage rate, the population balance equation, eq. (1), can be written as

\[
\zeta f'(\zeta) = \gamma \int_{0}^{\zeta} \frac{\zeta^2}{u^3} f'(\zeta) g(u) \, du
\]

where the similar breakage form, eq. (2), has been used. Equation (3) has to be solved to obtain the daughter-drop distribution, \( g(x) \), and the multiplicative constant, \( \gamma \). While solving eq. (3) for the daughter-drop distribution, constraints need to be imposed on the form of the \( g(x) \) function. The constraints used are physical constraints from the nature of the daughter-drop distribution. Since \( g(x) \) is a cumulative daughter-drop distribution function, it must be positive everywhere and be a monotonic increasing function. It is further surmised that break-up of a drop cannot produce drops in the neighborhood of zero volume and hence, drops nearly its own size, so that the derivative of the daughter-drop distribution at \( x = 1 \) may be set equal to zero. Mathematically, the constraints used are

\[
g(x) > 0, \quad g'(x) \geq 0, \quad g'(1) = 0.
\]

The details of solving the ill-posed problem given by eq. (3) using the method of quasisolutions is given in Sathyagal et al. (1994).

3. EXPERIMENTAL

Experimental transient size distribution measurements of breaking dispersions were conducted in a 14 cm diameter mixing vessel of standard configuration with a Rushton turbine impeller to provide the agitation. A diagram of the mixing vessel is shown in Fig. 1. The impeller was driven by a variable speed Lightnin motor which gives very accurate control of the stirrer speed.

Before beginning each experiment, the apparatus was thoroughly cleaned, first with soap and then with alcoholic KOH. It was then rinsed by keeping it under running water for at least 30 min. The apparatus was then dried before it was used in the experiment. To start the experiment, 1700 ml of water (which had earlier been equilibrated with the dispersed-phase organic liquid) was added to the glass vessel. The water used in the experiments was distilled and deionized before being filtered through a Millipore water system. The stirrer was run at a low speed, between 100 and 150 rpm, while the organic phase (previously equilibrated with water) was added to the vessel through the sampling port. In each experiment, 10 ml of the organic phase was added, corresponding to a dispersed phase fraction of 0.58%. After the organic phase had been added, the stirrer was allowed to run at the low speed for 1 min. This allowed the large globules of the dispersed phase to be broken down into smaller-sized globules. This procedure also allowed the initial condition of the experiment to be more uniform across different experiments. At the end of this 1 min, the stirrer speed was stepped up to the required speed for the experiment. The experimental time recorded in the data shown is the time after this increase in stirrer speed.

Small samples of the dispersion were removed at regular intervals from the stirred vessel with a micro-pipette, placed on a microscope slide and stabilized
with a couple of drops of sodium dodecyl sulfate (SDS) surfactant. Tobin (1989) has analyzed the applicability of the sampling technique to measure drop-size distributions. From his analysis, he has concluded that the technique provides a representative sample of the dispersion in the stirred vessel. Images of the dispersion were captured from a microscope with a video camera and stored for analysis with an image analyzer. The images were then processed using a Cambridge Instruments Q570 Image Analyzer to obtain the drop sizes. Approximately 120 images were taken at each experimental time, and up to 3000 drops were measured to construct the size distribution.

At the initial stages of a breakage experiment, the drop-size distribution changes rapidly due to the presence of larger drops which break more frequently. Hence, more frequent samples of the dispersion need to be taken to get a good estimate of the changing distribution. Since it took 5–10 min to capture the images from a dispersion sample, it was not possible to complete capturing the images from one sample before the next sample had to be taken out of the vessel. Hence, 30–45 min would elapse from the time the samples were removed from the vessel to the time their images were captured. Preliminary experiments were conducted to check if the size distributions would change by keeping the samples on the microscope slides for such periods of time. Samples were taken from a stirred dispersion, placed on a microscope slide and stabilized with SDS. Images of the sample were taken immediately. The sample was maintained on the slide for 1 h, and at the end of this time, images of the sample were taken again. The two sets of images were analyzed and the drop-size distributions were determined. The two size distribution curves were virtually identical, which indicates that the sample does not change much in an hour on the microscope slide.

Experiments were conducted to determine the effect of interfacial tension, dispersed phase viscosity and stirrer speed on the breakage rate and daughter-drop distribution. Four different dispersed phase systems were chosen. For each system, small amounts of carbon tetrachloride were added to the organic phase to make a neutrally buoyant dispersion. The physical properties of the experimental systems used are shown in Table 1. The viscosity measurements were made using a capillary viscometer. The interfacial tension measurements were made by taking images of pendant drops of the dispersed phase in water. The Laplace–Young equation was then solved to obtain the best value of the interfacial tension which would generate the digitized pendant drop shape. Details of this technique are given in Tobin (1992). Experiments at three different stirrer speeds were conducted for each of the systems. For the first three systems, experiments were conducted at 350, 400 and 500 rpm. For the 200 centistoke silicone oil system, breakage was very slow at the lower stirrer speeds and a reasonable number of drops could not be counted to get a good estimate of the size distribution. Hence, for the 200 centistoke silicone oil system, the experiments were conducted at 500, 600 and 700 rpm. Repeat experiments were carried out for each set of operating conditions. The results for the breakage rate and daughter-drop distribution presented here have been obtained by combining the size-distribution data from two experiments at the same operating conditions.

### Table 1. Physical properties of liquids used in breakage studies

<table>
<thead>
<tr>
<th>System</th>
<th>Composition (w/w%)</th>
<th>Viscosity (cp)</th>
<th>Interfacial tension (mN/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Benzene–carbon tetrachloride</td>
<td>73–27</td>
<td>0.74</td>
<td>35.0</td>
</tr>
<tr>
<td>Heptane–carbon tetrachloride</td>
<td>44.7–55.3</td>
<td>0.72</td>
<td>42.0</td>
</tr>
<tr>
<td>Dimethylpolysiloxane (20 cs)–carbon tetrachloride</td>
<td>87.6–12.4</td>
<td>11.3</td>
<td>36.2</td>
</tr>
<tr>
<td>Dimethylpolysiloxane (200 cs)–carbon tetrachloride</td>
<td>87.6–12.4</td>
<td>133.9</td>
<td>36.2</td>
</tr>
</tbody>
</table>

**4. RESULTS AND DISCUSSION**

Data from the image analysis system were in the form of a list of drop diameters for each of the drops measured. These data were converted into probability, number and volume fraction density distributions using kernel density estimation techniques. Details of these techniques are given in the book by Silverman (1986). Using these techniques, an estimate of the error in the distributions is also obtained. From the drop-diameter data, cumulative volume fraction curves were also calculated.

Figure 2 gives typical plots of the cumulative volume fraction, \( F(v, t) \) vs drop volume (\( \mu l \) or mm\(^3\)) at various times for the various systems studied. The sampling times in the figures are in minutes. The figures show the curves moving to the left as time progresses, indicating that breakage is taking place.

Figure 3 gives a typical example of a log–log plot of stirring time vs drop volume at various different fixed cumulative volume fraction values. (This figure is typical of similar plots obtained for all the experimental conditions. These plots are not shown here for reasons of conciseness.) These plots are used to test the data for the existence of similarity. The computational details of the test are given in Sathyagal et al. (1994).
Fig. 2. Experimental drop size distributions for various systems studied. Experimental times are in minutes. (a) System: benzene–CCl₄ at 500 rpm. (b) System: heptane–CCl₄ at 350 rpm. (c) System: 20 cs
oil–CCl₄ at 500 rpm. (d) System: 200 cs oil–CCl₄ at 500 rpm.

involves calculation of the curvature and arc length of the various ln(t) vs ln(v) curves are the same at all ln(v) values. Figure 4 shows a typical plot of the arc length vs ln(v). The arc lengths of the curves corresponding to the different fixed cumulative volume fraction values have all collapsed into a single curve, which indicates that the data does show similarity. The figure also shows that the collapsed curve has a slight curvature, indicating that the breakage rate is not a power law function. This non-power law breakage rate was observed in all the conducted experiments.

The collapsed arc length curves in Fig. 4 were used to calculate the collapsed ln(t) vs ln(v) curve, and hence obtain an estimate of $\Gamma(v)/\gamma$. The estimated $\Gamma(v)/\gamma$ function is then used to transform the size-distribution data using the transformation $F(x,t) \rightarrow f(\zeta) = \Gamma(v)/\gamma$. Figure 5 shows representative examples of the transformed data in the form $\zeta f'(\zeta)$ vs $\zeta$. The $\zeta f'(\zeta)$ values were calculated using the volume fraction density data. The figures show that the distributions at different times have collapsed into a single curve thus upholding similarity. In most of the experiments conducted, a good collapse of the similarity distributions was obtained. The collapsed similarity distributions showed some scatter at large $\zeta$ values (corresponding to large drop sizes) due mainly to the small number of drops counted at these large sizes.

The collapsed similarity distribution, $\zeta f'(\zeta)$, is used as the input to eq. (3) to calculate the daughter-drop distribution, $g(x)$ and the unknown multiplicative constant, $\gamma$. As described in Sathyagal et al. (1994), this ill-posed problem is solved using the method of quasisolutions. The unknown function, $g(x)$ is expanded in terms of basis functions and the coefficients of expansion are determined by the solution procedure. Sathyagal et al. (1994) have also described a method of determining the optimum number of basis functions needed to expand the $g(x)$ function.

The unknown multiplicative constant, $\gamma$, can be determined from the condition that $g(x) = 1$ at $x = 1$ [since $g(x)$ is a cumulative distribution function].
Figure 6 shows examples of the extracted \( \gamma g(x) \) function. These plots are typical of the extracted \( \gamma g(x) \) functions from all the experimental data. The daughter-drop distribution rises sharply for small values of \( x = F(v)/F(v') \) and slowly levels off towards \( x = 1 \).

The extracted breakage rates and daughter-drop distributions were tested by comparing their predictions with the experimental data. The extracted functions were input to the population balance equation and the equation was solved using the experimental size distribution at the time at which similarity was first observed as the initial condition. Figure 7 shows the predicted distributions at various times along with the experimental data. For clarity of presentation, not all the experimental and predicted distributions for these experiments have been shown. As shown in the figure, the extracted breakage functions predict the experimental data quite well.

### 4.1. Breakage Rate

From the estimated \( \Gamma(v)/\gamma \) and the \( \gamma \) values, the breakage rate \( \Gamma(v) \) has been evaluated at all the experimental conditions. As mentioned earlier, the breakage rate was found to be a non-power law function in all our experiments. Figure 8 shows the breakage rate as a function of drop diameter for different stirrer speeds. From the figure it can be seen that the breakage rate is a very strong function of drop size and stirrer speed, increasing sharply with both variables. Figure 9 shows the breakage rate vs drop size at a stirrer speed of 500 rpm for the four systems studied. This figure shows that the breakage rate is a very strong function of the interfacial tension. For a small increase in interfacial tension (35 mN/m for the benzene system vs 42 mN/m for the heptane system), the breakage rate reduces very sharply. The figure also shows the effect of dispersed phase viscosity on the breakage rate. The breakage rate is found to decrease with increasing dispersed phase viscosity, though not as sharply as with the interfacial tension.
From dimensional analysis and their experimental data, Narsimhan et al. (1984) have shown that the breakage rate for various low dispersed phase viscosity systems can be collapsed into a single curve as a plot of

$$\Gamma(v) \sqrt{\frac{\mu_U}{\sigma}} \propto W e^{\left(\frac{v}{D^3}\right)^{5/9}}.$$  

For the higher viscosity systems studied here, the non-dimensional breakage rate must also be a function of $\mu_c/\mu_d$. The results from the inverse problem were fitted to an equation to provide a useful estimate of the breakage rate as a function of different experimental conditions. The breakage rate results from the inverse problem were correlated by the following equation:

$$\Gamma(v) = 0.4220 \exp \left\{ -0.24775 \ln^2 \left[ We \left( \frac{v}{D^3} \right)^{5/9} \left( \frac{\mu_c}{\mu_d} \right)^{0.2} \right] \right\}$$

where $We$ is the Weber number, $D$ is the stirrer diameter, $\mu_c$ and $\mu_d$ are the continuous and dispersed phase viscosities, $\rho$ is the density and $\sigma$ is the interfacial tension. Figure 10 shows a log–log plot of the non-dimensional breakage rate. The curves for the different experimental conditions do collapse quite well into a single curve. Some scatter is seen at the smaller values of the non-dimensional breakage rate. This scatter is at very low values of the breakage rate, which corresponds to a very small number of breakage events in this range being captured by the experimental data, and hence leads to errors in the estimation of the breakage rate in this range.

4.2. Comparison with breakage rate models

As discussed earlier, there have been many models for the breakage rate presented in the literature. In these models, breakage is assumed to occur through interaction of the drop with turbulent eddies. The different models assume different modes of the drop–eddy interaction. Coulaloglou and Tavlarides (1977) calculated the breakage rate as the fraction of drops breaking per unit breakage time. They estimated the fraction of drops breaking as proportional
In calculating the results shown in Fig. 11(b) for the model breakage rate values are of the same order of magnitude as the inverse-problem results, but the model shows a very strong dependence on the drop size which is not seen in the inverse-problem results. The model of Coulaloglou and Tavlarides (1977) contains two constants, $k_1$ and $k_2$. Two sets of values for these constants were tested. The values given by Coulaloglou and Tavlarides (1977) give a breakage rate which is orders of magnitude higher than the inverse-problem result. These results have not been presented here. The values for $k_1$ and $k_2$ given by Bapat et al. (1983) were used, and the resulting breakage rate is shown in Fig. 11(c) along with the inverse-problem result. From the figure, it can be seen that the model breakage rates are of the same order of magnitude as the inverse-problem results. The model results show a similar trend but are smaller in magnitude than the inverse-problem results. In Fig. 11(d), the breakage rate calculated from the Tsouris and Tavlarides (1994) model is compared with the inverse-problem result. The following parameter values were used in calculating the model breakage rate values: $d_{\text{min}} = 5 \mu m$, $d_{e,\text{min}} = 10 \mu m$, $c_1 = 0.48$. This $c_1$ value is the same as that suggested by Tsouris and Tavlarides from their experimental data. The $d_{\text{min}}$ and $d_{e,\text{min}}$ values have been chosen in order to calculate the breakage rate over the entire drop size range encountered in our experiments. It is also to be noted that in calculating the breakage rate values for the Coulaloglou and Tavlarides model, the average energy dissipation rate for the entire vessel was used, while the Tsouris and Tavlarides model used the higher energy dissipation rate in the impeller zone of the vessel. The model predicts a very small breakage rate, approximately three orders of magnitude smaller than the inverse-problem results, and also a very strong dependence on the drop size.

The above models make many simplifying assumptions to obtain expressions for the breakage rate. As stated by Nambiar et al. (1992), these models are one-dimensional approximations which attempt to capture the gross features of the breakage process. A major assumption made in these models is that of binary breakage. It is quite probable for drops, especially large drops, to break into more than two daughter drops (later in this paper, results on the number of daughter drops will be presented). Since less energy is required, on average, for binary breakage than for breakage into many drops, this may partly account for the stronger dependence of the breakage rate on drop size seen in these models compared with the inverse-problem results. Other factors which may have to be incorporated into these models include drop interactions with multiple eddies, interactions when the drop is already deformed, a more realistic breakage mechanism (for example, surface tension driven flows due to drop deformation may accelerate breakage).
Droplet breakage in stirred dispersions

Fig. 7. Comparison of the transient size distributions predicted by the inverse-problem breakage functions with the experimental data. Experimental times are in minutes. (a) System: benzene–CCl₄ at 500 rpm. (b) System: heptane–CCl₄ at 350 rpm. (c) System: 20 cp oil–CCl₄ at 500 rpm. (d) System: 200 cp oil–CCl₄ at 500 rpm.

Fig. 8. Breakage rates determined from the inverse problem at different stirrer speeds for the benzene–CCl₄ system.

Fig. 9. Breakage rates calculated from the inverse problem at a stirrer speed of 500 rpm for the different dispersed phase systems.
Narsimhan et al. (1984) have also given a correlation for the breakage rate based on their experimental data and their inverse-problem formulation. Their correlation gives a drop-size dependency on the breakage rate which is comparable to that given by eq. (5) for large drops. For example, Lagissetty et al. have developed a model for $d_{\text{max}}$ which suggests a value of 0.125 for the constant which is similar to the value of Sprow, while the correlation given by Calabrese et al. suggests a value of 0.0883. The model of Lagissetty et al. is also applicable to high dispersed-phase viscosities. Similarly, Calabrese et al. have correlated data over a wide range of experiments and their correlation is also applicable to high dispersed-phase viscosity systems.

The inverse-problem results from this investigation give a breakage rate which continuously decreases with decreasing drop size and does not reduce to zero abruptly at a particular drop size. In the above mentioned studies on $d_{\text{max}}$, the experimental $d_{\text{max}}$ values were determined by running the breakage experiment for a specified amount of time (usually 1–2 h) and observing the largest drop size at the end of this time. In a similar vein, the $d_{\text{max}}$ value for the purposes of this investigation can be equivalently defined to be the largest drop size whose breakage rate, $F(v)$, is smaller than $1/(\text{time scale experiment})$. For most of the experiments performed for this study, the experiment was conducted for approximately 1 h. Hence, the $d_{\text{max}}$ values have been inferred from the inverse-problem results by determining the drop size whose breakage rate is $(1/60)\text{min}^{-1}$. Table 2 shows the $d_{\text{max}}$ values calculated using this criterion along with the values calculated using the model of Lagissetty et al. and the correlation of Calabrese et al.

From the table, it can be seen that the inverse-problem results agree quite well with the values predicted by the correlation of Calabrese et al., while the model of Lagissetty et al. predicts much larger values of $d_{\text{max}}$.

4.4. Daughter-drop distribution

The daughter-drop distribution has been determined for all the experiments conducted in this study using the method of quasisolutions to solve the ill-posed problem given by eq. (3). As described in Sathyagal et al. (1994), the unknown function is expanded in terms of basis functions. The basis functions used to expand the unknown daughter-drop distribution function are modified Jacobi polynomials, $x^\mu G_j(x)$, where $\mu$ is determined from the behavior of the similarity distribution, $\zeta f'('0')$, at small $\zeta$ values. For most of the experiments in this study, the daughter-drop distribution could be determined by expansion with three basis functions.

Figure 12 shows the daughter drop distribution as a function of $x = \Gamma(v)/\Gamma(v')$ for the benzene system at

\[
\frac{d_{\text{max}}}{D} = c(We)^{-0.6}
\]

where $c$ is a constant. The value of this constant varies substantially between various investigations. For example, Lagissetty et al. have given an exhaustive listing of the correlations available for $d_{\text{max}}$. For low dispersed-phase viscosities, the relation is of the form, $d_{\text{max}} = c(We)^{-0.6}D$ where $c$ is determined from the behavior of the similarity distribution, $\zeta f'('0')$, at small $\zeta$ values. For most of the experiments in this study, the daughter-drop distribution could be determined by expansion with three basis functions.
Droplet breakage in stirred dispersions

Fig. 11. Comparison of the breakage rate calculated from the various breakage models with the inverse-problem result. System: benzene–CCl₄. (a) Model of Narsimhan et al. (1979). (b) Model of Nambiar et al. (1994). (c) Model of Coulaloglou and Tavlarides (1977). (d) Model of Tsouris and Tavlarides (1994).

During the collection of the experimental data, it was found that the nature of breakage was different in the higher viscosity oils compared to breakage in the benzene and heptane systems. In the benzene and heptane systems, the drop sizes in an experimental sample were more uniform, giving a narrower size distribution. In the high viscosity oil systems, many small drops and a few large drops were observed in the experimental samples, giving a broad size distribution. A similar observation has been made by Calabrese et al. (1986a) in their study of the effect of dispersed phase viscosity on drop breakage. Such a broadening of the size distribution for the high viscosity oils is to be expected since their viscosity will make it difficult to substantially deform these drops and hence, through small deformations, these
Table 2. Comparison of the maximum stable-drop diameters

<table>
<thead>
<tr>
<th>System</th>
<th>Stirrer speed (rpm)</th>
<th>$d_{\text{max}}$ (microns)</th>
<th>Inverse prob. results</th>
<th>Model of Lagisetty et al.</th>
<th>Correlation of Calabrese et al.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Benzene – CCl₄</td>
<td>350</td>
<td>199.4</td>
<td>350.8</td>
<td>249.6</td>
<td></td>
</tr>
<tr>
<td>Benzene – CCl₄</td>
<td>400</td>
<td>168.6</td>
<td>298.9</td>
<td>213.0</td>
<td></td>
</tr>
<tr>
<td>Benzene – CCl₄</td>
<td>500</td>
<td>108.4</td>
<td>228.7</td>
<td>163.2</td>
<td></td>
</tr>
<tr>
<td>Heptane – CCl₄</td>
<td>350</td>
<td>245.3</td>
<td>391.4</td>
<td>278.3</td>
<td></td>
</tr>
<tr>
<td>Heptane – CCl₄</td>
<td>400</td>
<td>230.4</td>
<td>333.4</td>
<td>287.3</td>
<td></td>
</tr>
<tr>
<td>Heptane – CCl₄</td>
<td>500</td>
<td>137.9</td>
<td>255.1</td>
<td>181.8</td>
<td></td>
</tr>
<tr>
<td>20 cs oil – CCl₄</td>
<td>350</td>
<td>278.0</td>
<td>358.0</td>
<td>271.0</td>
<td></td>
</tr>
<tr>
<td>20 cs oil – CCl₄</td>
<td>400</td>
<td>213.7</td>
<td>307.2</td>
<td>232.7</td>
<td></td>
</tr>
<tr>
<td>20 cs oil – CCl₄</td>
<td>500</td>
<td>138.8</td>
<td>235.6</td>
<td>180.6</td>
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</tr>
<tr>
<td>200 cs oil – CCl₄</td>
<td>500</td>
<td>251.9</td>
<td>345.3</td>
<td>272.4</td>
<td></td>
</tr>
<tr>
<td>200 cs oil – CCl₄</td>
<td>600</td>
<td>217.7</td>
<td>288.6</td>
<td>231.0</td>
<td></td>
</tr>
<tr>
<td>200 cs oil – CCl₄</td>
<td>700</td>
<td>202.9</td>
<td>249.7</td>
<td>201.4</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 12. Daughter-drop distribution at different stirrer speeds calculated from the inverse problem. System: benzene–CCl₄.

Fig. 13. Daughter-drop distribution calculated from the inverse problem for a 375 µm drop at 500 rpm for different dispersed-phase systems.

drops will break by small daughter drops pinching off from the surface of the drops. Figure 13 shows the daughter-drop distribution for each of the systems studied as a function of $v/v'$ for a parent drop of 375 µm diameter. The figure shows that the volume fraction of daughter drops in the size range $v/v' > 0.5$ increases as the viscosity of the dispersed phase increases. This indicates that small drops are being stripped off the parent drop and larger daughter drops are formed for the high-viscosity oils. Hence, the figure clearly shows that the breakage of the high-viscosity oils is more erosive in nature compared to the breakage of the benzene and heptane systems.

The calculated daughter-drop distributions can also be used to determine the average number of daughter drops formed on breakage, $v(v')$. This function can be evaluated from

$$ v(v') = \int_0^{v'} \frac{v'}{v} \frac{\partial G(v,v')}{\partial v} \, dv $$

(6)

Figure 14 shows the inverse-problem result for the average number of daughter drops formed on breakage as a function of the parent drop size for one of the experiments conducted in this study. The figure shows that $v(v')$ is a strong function of the parent drop size, with a large number of daughter drops formed on the breakage of a large parent drop. It also shows that the small drops undergo approximately binary breakage.

Using the daughter-drop distributions obtained from solution of the inverse problem, correlations have been developed for the distribution as a func-
Droplet breakage in stirred dispersions

of low viscosity (since in their model, the smallest eddy size which can break a drop increases with drop viscosity as shown in Fig. 5 of this paper). This result is contrary to our experimental results and those of Calabrese et al. (1986a).

Narsimhan et al. (1984) have also determined the daughter-drop distribution from their experimental data. However, due to the ill-posed nature of the inverse problem, they had many difficulties in determining the daughter-drop distribution. Comparison of their daughter-drop distributions (see Fig. 8 of their paper) with the functions determined in this study shows that their daughter-drop distributions are very different from those in this study, and are closer in appearance to the model of Nambiar et al. (1992).

The dynamic drop-size distributions predicted by the breakage rate and daughter-drop distribution models of Nambiar et al. (1992, 1994) and Tsouris and Tavlarides (1994) have been compared with our experimental data. The predictions of the model of Coulaloglou and Tavlarides (1977) have also been compared with the experimental data. Along with the breakage rate of this model, the daughter-drop distribution used by Bapat et al. (1983) has been used to calculate the dynamic size distributions. Figure 16 shows the predictions of the different models. In these figures, the 2 min experimental size distribution is used as the initial condition to solve the population balance equation. As the figures show, the models do not predict the experimental data well. The model of Nambiar et al. predicts no increase in the volume fraction at the small drop sizes. This is a consequence of the daughter-drop distribution model shown in Fig. 15. The model also predicts a rapid decrease in
the volume fraction of the largest drop sizes and a big increase in the volume fraction of intermediate sized drops. The model of Tsouris and Tavlarides predicts almost no change in the size distribution, a consequence of the very low breakage rates of the model. The predictions of the Coulaloglou and Tavlarides model show very similar trends to those of the model of Nambiar et al.

5. CONCLUSIONS

Experimental data over a range of experimental conditions show self-similarity of the form, $G(v, v') = g(F(v)/F(v'))$. The transient drop-size distributions for each experiment collapse into a single similarity distribution. This similar breakage has enabled the evaluation of size-specific breakage rates and daughter-drop distributions via solution of inverse problems. From the inverse-problem results, it is found that the breakage rate is a strong function of drop size, stirrer speed and interfacial tension. It also shows a slightly weaker dependency on the dispersed-phase viscosity. The daughter-drop distribution does not vary much with stirrer speed, but shows a dependency on the dispersed-phase viscosity. The calculated breakage rates and daughter-drop distributions, when input into the population balance equation, predict the transient experimental data very well.

Models for the breakage rate and daughter-drop distribution have been compared with the inverse-problem breakage functions. It is found that the model breakage functions are very different from the breakage functions calculated in this study. The experimental transient size distributions are also not predicted well by the model breakage functions.

Correlations have been developed for the breakage rate and the daughter-drop distribution. The correlations satisfactorily encompass the breakage functions evaluated from all the experiments conducted in this study.

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**NOTATION**

- $d_{max}$: maximum stable-drop diameter
- $D$: stirrer diameter
- $f(\zeta)$: similarity distribution
- $f'(\zeta)$: derivative of similarity distribution
- $F(v, t)$: cumulative volume fraction
- $g(x)$: cumulative daughter-drop distribution
- $G(v, v')$: cumulative daughter-drop distribution
- $G_j(x)$: $(j-1)$th order Jacobi polynomial
- $N$: stirrer speed
- $t$: time
- $v$: drop volume
- $v'$: drop volume
- $We$: Weber number ($= N^2 D^3 \rho/\sigma$)
Droplet breakage in stirred dispersions

Greek symbols
\( \gamma \) breakage rate at reference volume
\( \Gamma(v) \) breakage rate function
\( \mu \) power-law exponent of similarity distribution at small \( \zeta \)
\( \mu_c \) viscosity of continuous phase
\( \mu_d \) viscosity of dispersed phase
\( v(v') \) average number of daughter drops formed on breakage
\( \rho \) density of continuous (or dispersed) phase
\( \sigma \) interfacial tension
\( \zeta \) similarity variable

REFERENCES


