

# COALESCENCE OF DISPERSED DROPS IN AN AGITATED TANK

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An attempt was made to establish the coalescence frequency of dispersed drops in an agitated tank. In experiments, transient drop size distributions were measured during the processes of drop coalescence after a stepwise reduction of impeller speed. The transient drop size distributions were calculated from the population balance equation for dispersed drops by applying a previously proposed equation for drop breakup and assuming some different functional forms for coalescence frequency. From comparison between calculated and experimental distributions, a functional form of coalescence frequency was determined which adequately correlates transient drop size distributions with the breakup frequency equation. It was also shown that the equations for coalescence and breakup frequencies successfully predict transient drop size distributions in drop dispersion processes.

## Introduction

Coalescence of dispersed drops in an agitated tank has been the subject of many investigations. However, the mechanism of coalescence has not been sufficiently clarified.

Although some previous investigators<sup>8,14)</sup> conducted direct observations of coalescence phenomena in an agitated tank, for quantitative analysis most workers have adopted indirect methods, such as measurements of mean drop sizes,<sup>4,11-13)</sup> or measurements of droplet mixing following a chemical reaction<sup>5,11)</sup> and dispersion of a nontransferring dye in dispersed phase.<sup>1,5,9,10,15)</sup> Whatever the efficiency of these methods, another method that promises to be useful is the measurement of transient drop size distributions during coalescence processes. Although some investigators applied the method of step-change reduction in impeller speed to measure the variation of mean drop size with time during coalescence, there has been no report on transient drop size distributions. The present work applies the method of the step-change reduction to measure transient distributions during coalescence.

## 1. Experiments

### 1.1 Apparatus

A schematic diagram of the apparatus is shown in Fig. 1. The mixing vessel consisted of a glass cylinder, 18.6 cm in diameter and 20 cm in height, flanged with stainless-steel bottom and top plates. The vessel was fitted with four equally spaced baffles, and in the center a standard 6-bladed disc turbine of 9.3 cm diameter. For photographing dispersed drops, two

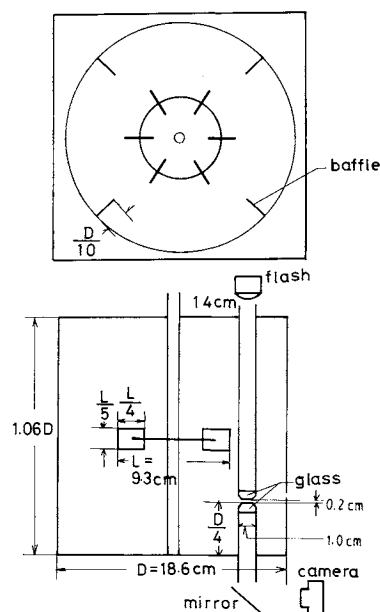


Fig. 1. Schematic diagram of apparatus

stainless-steel tubes with a glass window at the end of each were inserted respectively into the bottom and top plates of the vessel. The camera was focussed on an area between the glass windows.

### 1.2 Materials and procedure

The continuous phase used was distilled water, to which a small amount of sodium chloride (0.1 mol/l) was added to reduce the effects of contaminants on coalescence.<sup>10)</sup> The dispersed phase used was a mixture of *o*-xylene and carbon tetrachloride.

In each run the vessel was completely filled with the continuous and dispersed phase liquids, and agitation was started at a desired speed. The impeller speed was maintained for one hour, during which a steady dispersion was attained. Then the impeller speed was

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reduced in a stepwise way, and at appropriate intervals the dispersion in the vessel was photographed to measure transient drop size distributions. The initial value of impeller speed, prior to reduction, was 7 rps in all the experiments, while the speed after reduction was 2.3, 2.7 or 3.5 rps. More than three hundred drop diameters were measured to obtain a single drop size distribution. The experimental temperature was maintained at 25°C and the volume fraction of the dispersed phase was 0.01, 0.02 or 0.05. The density and viscosity of the dispersed phase were 1.03 g/cm<sup>3</sup> and 0.00089 Pa·s, respectively, and those of the continuous phase 1.00 g/cm<sup>3</sup> and 0.00069 Pa·s.

## 2. Results and Discussion

### 2.1 Derivation of coalescence frequency from experimental drop size distributions

Transient drop size distributions in an agitated tank are expressed by the following population balance equation:

$$\begin{aligned} dN(v)/dt = & \int_0^{v/2} k_c(v-v', v')N(v-v')N(v')dv' \\ & - \int_0^\infty k_b(v, v')N(v)N(v')dv' \\ & + \int_v^\infty m(v')\gamma(v, v')k_b(v')N(v')dv' - k_b(v)N(v) \end{aligned} \quad (1)$$

Where  $N(v)$  is the number density of drops with volume  $v$ ;  $k_c(v', v)$ , the coalescence frequency of drops with volumes  $v$  and  $v'$ ;  $k_b(v')$ , the breakup frequency of drops with volume  $v'$ ;  $m(v')$ , the mean number of daughter drops formed by breakup of a parent drop with volume  $v'$ ; and  $\gamma(v, v')$ , the distribution of daughter drops formed by the breakup.

The breakup parameters  $k_b(v)$ ,  $\gamma(v, v')$  and  $m(v')$  can be calculated from the following equations, obtained in our previous work,<sup>6)</sup> which correlated transient drop size distributions under noncoalescence conditions.

$$k_b(v) = C_{A1} n_r \operatorname{erfc}(\eta)$$

$$+ C_{B1} (n_r^3 L^2 / d^2)^{1/3} \cdot 3 \sqrt{\frac{6}{\pi}} \int_\xi^\infty x^2 \exp\left(-\frac{3x^2}{2}\right) dx \quad (2)$$

$$m(v') = 3 \quad (3)$$

$$\gamma(v, v') = (2d'/3d^2) \cdot (v'/v) \cdot \beta(d/d') \quad (4)$$

where  $\eta$  and  $\xi$  are dimensionless groups defined by Eqs. (5) and (6), and  $\beta(d, d')$  is a Beta function given by Eq. (7):

$$\eta = C_{A2} (\sigma^{3/2} / n_r^3 L^3 \rho_c^{3/2} d^{3/2})^{1/3} \quad (5)$$

$$\xi = C_{B2} (\sigma^{1/2} / n_r L^{2/3} \rho_c^{1/2} d^{5/6}) \quad (6)$$

$$\beta(d, d') = \frac{\Gamma(12)}{\Gamma(3)\Gamma(9)} \left(\frac{d}{d'}\right)^8 \left(1 - \frac{d}{d'}\right)^2 \quad (7)$$

In the equations above,  $d$  is drop size;  $d'$ , the diameter of the parent drop;  $L$ , impeller diameter;  $n_r$ , impeller speed;  $\sigma$ , interfacial tension between dispersed and continuous phases; and  $\rho_c$ , the density of continuous phase. The constants  $C_{A1}$ ,  $C_{A2}$ ,  $C_{B1}$  and  $C_{B2}$  are respectively 0.07, 1.45, 0.00024 and 0.24 for the geometry of impeller-to-vessel diameter ratio = 1/2.

In the population balance of Eq. (1), only the coalescence frequency is unknown. Therefore, if a certain functional form is assumed for the coalescence frequency it is possible to solve Eq. (1) and examine the validity of the functional form by comparing calculated and experimental drop size distributions. The coalescence frequency may thus be determined by this method.

First, the following functional form for coalescence frequency was examined:

$$k_c = A n_r L^{2/3} (d + d')^2 (d^{2/3} + d'^{2/3})^{1/2} n_r^x (d \cdot d')^y \quad (8)$$

In this equation it is assumed that drop coalescence mainly occurs in the flow field of isotropic turbulence. The term  $n_r L^{2/3} (d + d')^2 (d^{2/3} + d'^{2/3})^{1/2}$  represents the collision frequency of dispersed drops in isotropic turbulence,<sup>8)</sup> and the term  $n_r^x (d \cdot d')^y$  the probability of coalescence on drop collision. The values of the parameters  $A$ ,  $x$  and  $y$  were determined so that calculated transient drop size distributions could be fitted to experimental ones. Using the parameter values determined, the present work attempted to correlate steady-state distributions measured at 7 rps at each dispersed-phase volume fraction. However, the steady-state distributions were not correlated. This indicates the inadequacy of the functional form of Eq. (8) for the coalescence frequency in an agitated tank.

One of the reasons for this failure may be the inadequacy of the functional form assumed for the coalescence probability. Theoretical analyses by previous investigators<sup>2,3)</sup> suggest that the coalescence probability may be expressed as an exponential function of impeller speed and drop diameter. Assuming this type of functional form, the present authors finally obtained the following equation by trial and error.

$$k_c = 13.0 n_r (d + d')^2 (d \cdot d')^{1/2} \exp(-41.66 n_r^{0.525} (d \cdot d')^{1/3}) \quad (9)$$

Drop size distributions calculated with Eq. (9) are compared with experimental ones in Figs. 2 to 5.\*

\* To obtain a numerical solution, the integrals in Eq. (1) were approximated by an integration formula and drop size distributions were calculated by a digital computer.<sup>6)</sup>

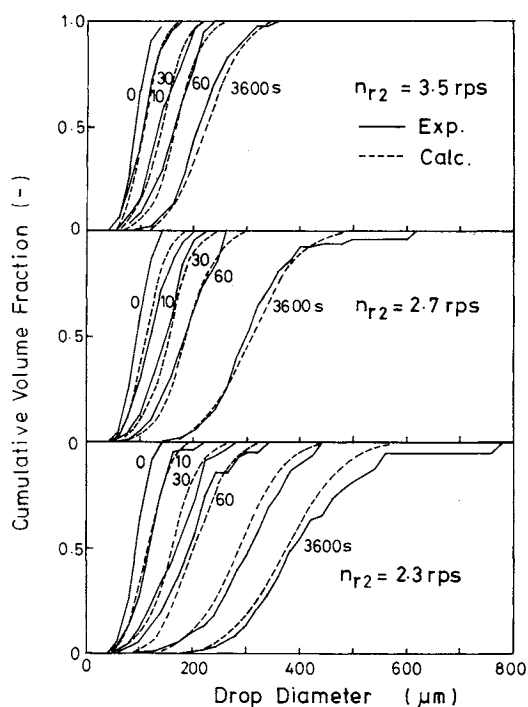


Fig. 2. Transient drop size distributions in coalescence processes ( $\phi = 0.01$ ,  $n_{r1} = 7$  rps)

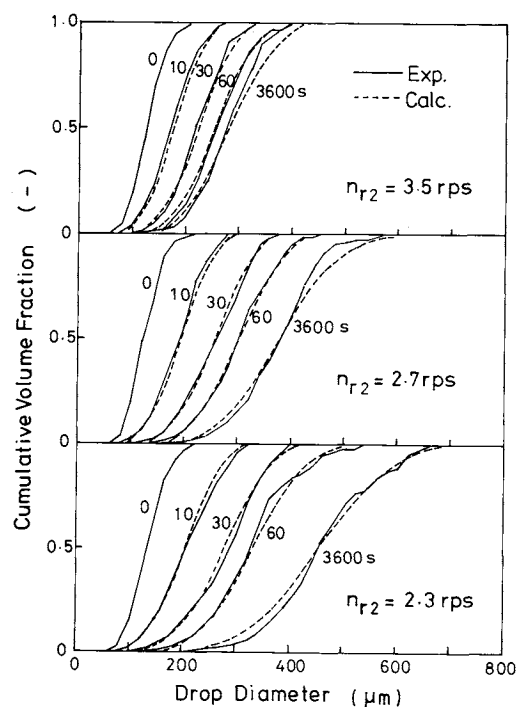


Fig. 4. Transient drop size distributions in coalescence processes ( $\phi = 0.05$ ,  $n_{r1} = 7$  rps)

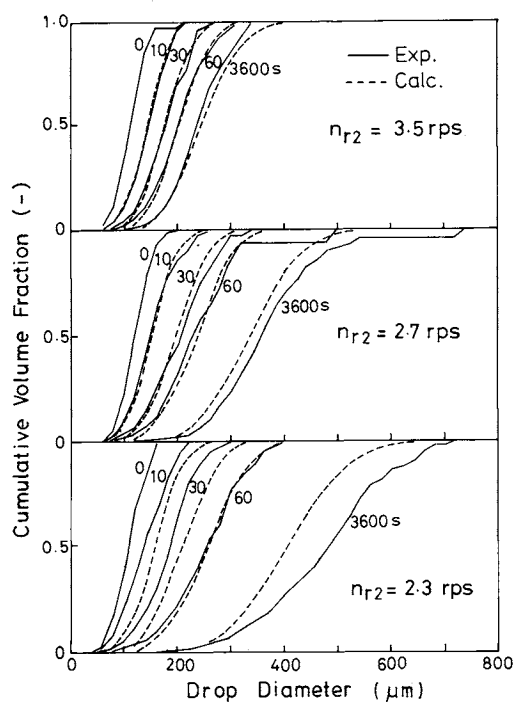


Fig. 3. Transient drop size distributions in coalescence processes ( $\phi = 0.02$ ,  $n_{r1} = 7$  rps)

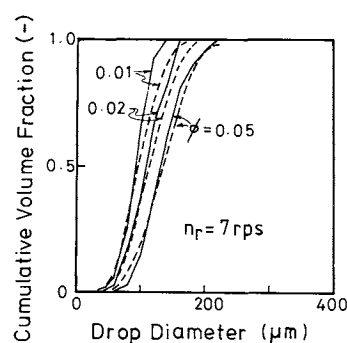


Fig. 5. Steady-state drop size distribution at different dispersed-phase volume fractions ( $t = 3600$  s)

Only in Fig. 3 does a relatively large discrepancy between calculated and experimental distributions appear, at the agitation time of 3600 seconds at  $n_r = 2.3$  rps. However, it is not clear whether or not this discrepancy was caused by experimental error. Under all the other experimental conditions, a high degree of consistency was obtained between calculated and

experimental distributions.

## 2.2 Discussion

In Eq. (9) the term of the exponential function decreases with increasing impeller speed and drop diameter. This may suggest a change in coalescence probability. Qualitatively, the decrease in coalescence probability with impeller speed and drop diameter can be explained by a model developed by Coualaloglou and Tavlarides<sup>2)</sup> taking into account the deformation of drops on their collision.

Figure 6 presents the calculated and experimental values of Sauter mean drop sizes,  $d_{32}$ , at steady dispersion. Both the experimental and calculated values of  $d_{32}$  show a similar relation to  $d_{32} \propto n_r^{-1.2}$ , which was reported by many previous investigators for various dispersed-phase volume fractions. However, as detailed in our previous works,<sup>6,7)</sup> the dependence of  $d_{32}$  on  $n_r$  under noncoalescence con-

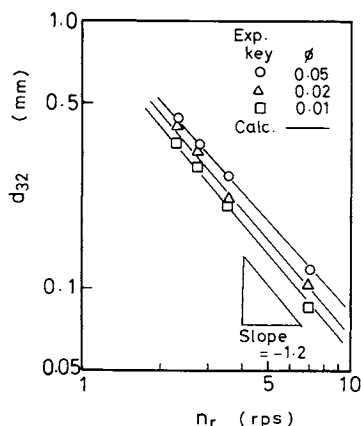


Fig. 6. Sauter mean drop sizes at steady-state dispersion

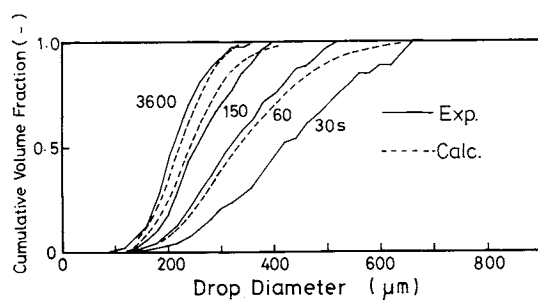


Fig. 7. Transient drop size distributions in breakup processes after beginning of agitation at  $n_r = 3.5$  rps ( $\phi = 0.01$ )

ditions is higher than that presented by the relation  $d_{32} \propto n_r^{-1.2}$ . In the literature, when this relation was observed, many investigators assumed that drop breakup was the controlling factor over drop coalescence in the determination of drop sizes. However, the present study shows the invalidity of that assumption.

Finally, to examine whether the equations proposed here for breakup and coalescence are capable of predicting transient drop size distributions in breakup processes, the present authors conducted dispersion experiments at impeller speeds of 2.3 and 3.5 rps and a dispersed-phase volume fraction of 0.01. Drop size distributions were measured at several agitation times after the beginning of agitation. The experimental results at 3.5 rps are shown in Fig. 7, together with calculated ones. In the calculations the experimental distribution at 30 s was used as a starting distribution. Figure 7 shows that the calculated distributions are fairly consistent with the experimental distribution. Good agreement was also obtained at an impeller speed of 2.3 rps.

## Conclusions

To obtain a functional relationship between drop coalescence frequency and operational variables, transient drop size distributions were measured during the processes of drop coalescence. An empirical equation of coalescence frequency was derived which

well correlates transient drop size distributions with drop breakup frequency from a previously proposed equation. It was also shown that the equations for the coalescence and breakup frequencies adequately predict transient drop size distributions measured in drop breakup processes.

The functional form of coalescence frequency has suggested that the probability of coalescence on the collision of drops decreases with increasing drop diameter or impeller speed. Comparison between calculated and experimental Sauter mean drop sizes at steady dispersion suggests that the relation of  $d_{32} \propto n_r^{-1.2}$  is attained only when coalescence and breakup are coexistent.

## Nomenclature

$d$	= drop diameter	[cm]
$d_{32}$	= Sauter mean drop size	[cm]
$k_b(v)$	= breakup frequency of drops with volume $v$	[s <sup>-1</sup> ]
$k_c(v, v')$	= coalescence frequency of drops with volumes $v$ and $v'$	[cm <sup>3</sup> · s <sup>-1</sup> ]
$L$	= impeller diameter	[cm]
$m(v')$	= number of daughter drops formed per breakup from a parent drop of volume $v'$	[—]
$n_r$	= impeller speed	[s <sup>-1</sup> ]
$n_{r1}$	= initial impeller speed, before reduction	[s <sup>-1</sup> ]
$n_{r2}$	= impeller speed after reduction	[s <sup>-1</sup> ]
$N(v)$	= number density of dispersed drops with volume $v$	[cm <sup>-6</sup> ]
$\gamma(v, v')$	= distribution of daughter drops formed by breakup of a parent drop with volume $v'$	[cm <sup>-3</sup> ]
$\rho_c$	= density of continuous phase	[g · cm <sup>-3</sup> ]
$\sigma$	= interfacial tension between continuous and dispersed phases	[N · m <sup>-1</sup> ]
$\phi$	= volume fraction of dispersed phase	[—]

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