

# Breakage and Coalescence Models for Drops in Turbulent Dispersions

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*A substantial effort has been made by numerous investigators to describe droplet breakage and coalescence in turbulent dispersions. An attempt is made here to improve these models based on existing frameworks and recent advances described in the literature. Two-step mechanisms are considered for both the breakage and coalescence models. The drop breakage function is structured as the product of the drop-eddy collision frequency and breakage efficiency which reflect the energetics of turbulent liquid-liquid dispersions. The coalescence function retains the former structure of the product of drop-drop collision frequency and coalescence efficiency. The coalescence efficiency model has been modified to account for the effects of film drainage for drops with partially mobile interfaces. These models overcome several inconsistencies observed in previous efforts and are applicable for dense dispersions (about  $\phi[0.10-0.30]$ ). For the daughter drops produced by breakage, a probability density is proposed based on the energy requirements for the formation of daughter drops.*

## Introduction

Many engineering processes involve mixing of two immiscible liquids in the form of liquid dispersions. One such process is solvent extraction which involves the separation of a species from a liquid system by using another solvent. In many cases a reactive species is placed in the second solvent to facilitate the separation. In all these processes it is desired to enhance mass-transfer rates by increasing the surface area for mass transfer. This task is accomplished by agitating the two liquids in stirred tanks or column contactors and thus forming liquid dispersions. The surface area for mass transfer in the contacting equipment is determined by the drop size and the volume fraction (holdup) of the dispersed phase. The holdup in certain vessels such as the stirred tank or the centrifugal contactor depends on the flow rate ratio of the two phases. In extraction columns, on the other hand, the holdup strongly depends on the drop size. Furthermore, some very undesirable phenomena occurring in extraction columns, such as flooding and phase inversion, are also strongly correlated with the holdup and the drop size. Hence, the drop size has a significant role in mass-transfer rates as well as in the occurrence of "cata-

strophic" events such as phase inversion or flooding. Therefore, in order to optimize and control liquid-dispersion processes, it is essential to better understand and model the drop behavior as affected by variations in the physical properties and in the mixing intensity.

For the analysis of dispersive systems, the population-balance-equation (pbe) model has been extensively and successfully used over the past three decades (Valentas and Amundson, 1966; Coulaloglou and Tavlarides, 1977; Ramkrishna, 1985). This model describes the history of a drop population in terms of drop properties, such as size, age, and concentration, during the course of droplet interaction events with themselves and with the surrounding environment. For example, drop coalescence, breakage, and inlet-outlet phenomena which are stochastic events randomly distributed in time can be accommodated by the pbe model along with the continuously occurring mass-transfer exchange between the two phases. The objective of the present work is to obtain analytical forms for the drop breakage and coalescence frequency functions which can then be employed for the analysis of multistage extraction columns. For this purpose, a batch mutually equilibrated system has been chosen for the study. In this case, drop inlet and outlet terms as well as mass-transfer terms are neglected, and

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the only property of importance is the drop size. Thus, the population balance equation for this system is written as:

$$\frac{\partial n(v, t)}{\partial t} + D - B = 0 \quad (1)$$

where  $n(v, t)$  is the number of drops of volume  $v$  at a given time  $t$ , and  $D, B$  represent death and birth rates of drops, respectively. The death term consists of breakage to smaller drops and coalescence to larger ones, while the birth term consists of breakage of larger drops and coalescence of smaller ones. These terms are given in the following equations:

$$D = n(v, t) \int_0^\infty \lambda(v, v') h(v, v') n(v') dv' + g(v) n(v, t) \quad (2)$$

and

$$B = \int_0^{v/2} \lambda(v-v', v') h(v-v', v') n(v-v', t) n(v', t) dv' + \int_v^\infty \beta(v, v') \nu(v') g(v') n(v', t) dv'. \quad (3)$$

Here,  $\nu(v')$  is the number of drops formed from the breakage of a drop volume  $v'$ ;  $\beta(v, v')$  is the probability density of the daughter drops which represents the probability of forming drops of size  $v$  from breakage of drops of size  $v'$ ;  $g(v')$  is the breakage frequency of drops of volume  $v'$ ;  $h(v, v')$  is the collision frequency of drops of volumes  $v$  and  $v'$ ,  $\lambda(v, v')$  is the coalescence efficiency once collision occurs between drops of volumes  $v$  and  $v'$ . Analytical solutions of Eq. 1 can be obtained only for special cases (Bapat et al., 1983; Ramkrishna, 1985) for which simple functions are chosen for drop breakage and coalescence. Volume discretization of Eq. 1 yields a system of integro-differential equations which represent an initial value problem and can be solved numerically (Valentas and Amundson, 1966; Valentas et al., 1966; Coulaloglou and Tavlarides, 1977; Sovova and Prochazka, 1981).

The drop size distribution obtained in an agitated liquid dispersion is a result of drop breakage and coalescence occurring simultaneously. The ideal way, however, to quantify the effect of each of these two processes on the drop size is to isolate them by carefully choosing the liquid system and the operating conditions. Although it is very difficult to completely eliminate one of the two drop rate processes, some techniques have been reported for substantially reducing one process as compared to the other. For example, lean dispersions or addition of surfactant substances may decrease drop coalescence (Narsimhan et al., 1980, 1984). For the other extreme case of a largely coalescing system, one can use an  $\epsilon$ -jump method where a negative step input is applied on the agitation speed and the evolution of the drop size distribution to another steady state is observed. Drop coalescence is the predominant process between the two steady states. Such a technique has been employed by Tobin et al. (1990) who studied the effect of drop size dependence on the coalescence frequency. Although isolating drop breakage or drop coalescence is an ideal way to understand these processes, it is uncertain whether the esti-

mation of either one of the drop processes is perfect. On the other hand, an experimental study of the transient behavior of a liquid dispersion undergoing both breakage and coalescence still provides valuable information for the modeling of such systems. Such an approach is employed in the present work.

Analytical functions of both drop processes of breakage and coalescence are proposed and evaluated with respect to experimental observations in a batch liquid dispersion where both drop breakage and coalescence are important. Other aspects on liquid dispersions discussed in this article are turbulence inhomogeneities in stirred tanks, and maximum stable drop size in agitated dispersions.

## Drop Breakage Rate

### Introduction

A number of mechanisms for drop breakage in a turbulent flow have been proposed in the past. Included in these mechanisms are drop elongation in a shear flow field (Taylor, 1934), turbulent pressure fluctuations (Hinze, 1955), relative velocity fluctuations (Narsimhan et al., 1980) and drop-eddy collisions (Coulaloglou and Tavlarides, 1977). Some drops break up into a large number of smaller drops, and this type of drop breakage is called thorough breakage by Narsimhan et al. (1980). Another type of drop breakage is the erosive breakage which produces a number of very small drops stripped out from a large one. Finally, it is also possible to have binary drop breakage, but only rarely do the two daughter drops have the same size (Sleighter, 1962).

Although a considerable amount of work on drop break up in liquid dispersions has been presented in the literature over the years, the main focus of these studies is on the effect of physical properties of the system or geometrical and operating conditions on the steady-state average drop size or drop size distribution (see, for example, Lagisetty et al., 1986; Koshy et al., 1988; Calabrese et al., 1986 a, b). Only a limited number of studies have concentrated on developing drop breakage or coalescence rate models. Some of the most recent breakage-rate models basically consider two mechanisms; first, the deformation of a drop due to interactions with the turbulent environment and, second, the probability of break up of the deformed drops. Such mechanistic models have been developed by Coulaloglou and Tavlarides (1977), Narsimhan et al. (1979) and Prince and Blanch (1990). The first two studies are for liquid-liquid dispersions whereas the third one is for gas-liquid dispersions. All three are mechanistic models, and they provide simple mathematical expressions for the drop breakage rate which include the physical properties of the system, the geometry, and the energy provided to the dispersion by agitation. The breakage rate derived by Coulaloglou and Tavlarides, for example, has the following form:

$$g(d_j) = k_1 \frac{\epsilon^{1/3}}{(1 + \phi)d_j^{2/3}} \exp \left[ -k_2 \frac{\sigma(1 + \phi)^2}{\rho_d \epsilon^{2/3} d_j^{5/3}} \right] \quad (4)$$

In the above equation,  $d_j$  is the drop diameter,  $\sigma$  is the interfacial tension,  $\rho_d$  is the density of the dispersed phase, and  $k_1, k_2$  are universal constants. Although all the above models consider the collision of eddies and drops to cause breakage, only the model of Prince and Blanch (1990) provides an estimate

of the eddy-drop collision frequency based on isotropic turbulence principles. Also, the model of Coulaloglou and Tavlarides (1977) has the disadvantage of predicting a breakage probability which goes through a maximum as the drop size increases. This behavior is discussed by Bapat (1982), and it is considered as a deficiency of the model.

Another approach reported by Narsimhan et al. (1980, 1984) is based on the similarity behavior of the transient drop size distribution. By using similarity analysis, the above investigators calculated rate functions for the drop breakage. These functions, when nondimensionalized with the natural frequency of oscillation of drops give a very satisfactory generalized plot against a dimensionless drop volume ratio. Experimental data of transient drop size distribution were correlated by the following equation:

$$g(v) \sqrt{\frac{\rho v}{\sigma}} = 5.75 We^{3.2} \left( \frac{v}{D_i^3} \right)^{1.78} \quad (5)$$

where  $v$  is the drop volume,  $We = N^2 D_i^3 \rho / \sigma$  is the Weber number representing the ratio of destructive over cohesive forces, and  $D_i$  is the impeller diameter. Equation 5 predicts a very strong dependence of the drop breakage rate with the impeller speed which can be expressed by  $g(v) \sim N^{6.4}$ . Furthermore, the above breakage function predicts significant breakage rates even for drops smaller than the maximum stable drop size below which breakage should not occur. The maximum stable drop size for lean dispersions,  $d_{\max}^0$ , is given by the correlation of Shinnar (1961) as:

$$d_{\max}^0 = c_m We^{-0.6} D_i \quad (6)$$

where  $c_m = 0.125$  (Sprow, 1967; Lagisetty et al., 1986). For the system used by Narsimhan et al. (1984),  $d_{\max}^0$  is calculated to be  $345 \mu\text{m}$  or  $21.5 \times 10^{-12} \text{ m}^3$  volume for which a significant breakage rate is predicted in Figure 10 of their article.

Despite the above efforts to model the breakage process inconsistencies are observed between existing models, and no universal model exists to describe drop breakage over a large range of conditions. In this work, a modified form of the model of Prince and Blanch (1990) is employed to model the drop breakage rate. The selection is made over the other models because this model appears to avoid the drawbacks of the models mentioned above.

### Model development

The breakage rate of drops in a turbulent flow is assumed to be given by the following product (Prince and Blanch, 1990):

$$g(d) = \underset{(A)}{\text{(eddy-drop collision frequency)}} \cdot \underset{(B)}{\text{(breakage efficiency)}} \quad (7)$$

The following assumptions are made for the modeling of the eddy-drop collision frequency: (i) the turbulent flow in a stirred tank is assumed to be isotropic; (ii) the drop size is in the inertial subrange; (iii) drops can break only by collisions with smaller eddies, or the same size, since larger eddies have the tendency to transport the drop rather than to break it.

(A) *Eddy-drop Collision Frequency.* For the estimation of eddy-drop collision frequency, it is assumed that the movement of eddies and drops behave like ideal gas molecules (Coulaloglou and Tavlarides, 1977), and their collision process can be described by an analogy to the kinetic theory of gases (Kennard, 1938). Accordingly, the collision frequency  $h(d)$  between drops of size  $d$  and eddies of a size range that can break these drops can be written as:

$$h(d) = \int_{n_e} h'(d_e, d) n_d dn_e = \int_{n_e} S_{ed} (u_e^2 + u_d^2)^{1/2} n_d dn_e \quad (8)$$

Here,  $S_{ed}$  is the collision cross-section area given by  $S_{ed} = \pi(d_e + d)^{2/4}$ ,  $d_e$  is the eddy size that can break a drop of size  $d$ ,  $n_d$  is the number of drops,  $dn_e$  is the number of eddies of size between  $d_e$  and  $d_e + \delta d_e$  and  $u_e$  and  $u_d$  are the velocities of eddies and drops.

*Number and Velocity of Eddies.* Batchelor (1970) gives the spectral energy density in the inertial subrange of the energy spectrum as:

$$E(k) = 1.7 \epsilon^{2/3} k^{-5/3} \quad (9)$$

where  $k$  is the eddy wave number ( $k = 1/r_e$ ,  $r_e$  being the radius of the eddy),  $\epsilon$  is the energy dissipation and  $E(k)$  is the energy per unit mass and per interval of wave number. Then, according to Azbel (1981), if  $N(k)$  denotes the number of eddies per unit mass of the liquid and per unit interval of wave numbers,  $E(k)$  can be written as:

$$E(k) = N(k)e. \quad (10)$$

The energy of a single eddy is given by:

$$e = \frac{1}{2} m u_e^2 = \frac{2}{3} \pi \rho_f r_e^3 u_e^2 \quad (11)$$

In Eq. 11,  $r_e$ ,  $u_e$  and  $m$  denote the eddy radius, velocity and mass, respectively, and  $\rho_f$  denotes the fluid density. According to Kolmogorov's law (Azbel, 1981), for the inertial subrange of the energy spectrum, the eddy velocity is given as:

$$u_e^2 = 8.2(\epsilon/k)^{2/3}. \quad (12)$$

Then, from Eqs. 9–12 and by taking the limits as  $\Delta k$  approaches zero, Azbel (1981) derived the following differential equation for the number of eddies per unit mass of the fluid,  $n_{e,m}$ :

$$\frac{dn_{e,m}(k)}{dk} = 0.1 \frac{k^2}{\rho_f} \quad (13)$$

Integration of Eq. 13 within appropriate limits of wave numbers will provide the number of eddies between these size limits. Thus, the number and the velocity of eddies needed for the eddy-drop collision frequency can be estimated from Eqs. 13 and 12. Furthermore, Eqs. 11 and 12 yield the following relation for the eddy energy:

$$e = 0.43 \rho_f \pi d_e^{1/3} \epsilon^{2/3}. \quad (14)$$

This relation is used later for the derivation of the maximum stable drop size.

**Drop Velocity in the Inertial Subrange.** The use of Eq. 8 for the estimation of the eddy-drop collision frequency, also requires the number and velocities of drops. The number of drops,  $n_d$ , is provided directly from the solution of the population balance equation. The mean-square velocity  $\overline{u^2}$  is given by Hinze (1959) as:

$$\overline{u^2} = \frac{2}{3} \int_{\frac{1}{r_d}}^{\infty} E(k) dk. \quad (15)$$

Substituting  $E(k)$  from Eq. 9 and integrating, one obtains:

$$\overline{u^2} = 1.07 \epsilon^{2/3} d^{2/3}. \quad (16)$$

All the required properties for the estimation of the eddy-drop collision frequency are now available. However, before the derivation of the final form of the collision frequency, some interesting points, which are considered important for this analysis are discussed next.

**Maximum Drop Size.** The energy required for the breakage of the drop is considered proportional to the energy required for the creation of new surface. This assumption can be written as:

$$e = k_3 \pi \sigma d^2 \quad (17)$$

where  $\sigma$  is the surface tension,  $d$  is the drop diameter, and  $k_3$  is a proportionality constant. Equations 14 and 17 yield:

$$0.43 \rho_f \pi d_e^{1/3} \epsilon^{2/3} = k_3 \rho \sigma d^2. \quad (18)$$

It is assumed that the largest eddy that can break a drop has the size of the drop. It has been reported (Shinnar, 1961; Sprow, 1967) that there exists a critical drop diameter,  $d_{cr}$ , below which drops in a given turbulent flow cannot break. In this case, the eddy size that can break the critical-size drops is  $d_{cr}$ , and hence Eq. 18 yields:

$$d_{cr}^{5/3} = k_4 \frac{\sigma}{\rho_f \epsilon^{2/3}}. \quad (19)$$

The energy dissipation is given by Bertrand et al. (1980) as:

$$\epsilon = k_5 N^3 D_i^2 \quad (20)$$

where  $k_5$  is the proportionality constant,  $N$  is the agitation speed, and  $D_i$  is the impeller diameter. From Eqs. 19 and 20, one can obtain the following relations for  $d_{cr}$ :

$$d_{cr} = k_6 We^{-0.6} D_i. \quad (21)$$

The critical diameter,  $d_{cr}$ , obtained by Eq. 21 is the same as the  $d_{max}^0$  (see Eq. 6) in very dilute dispersions obtained by Shinnar (1961).

For nondilute dispersions, one can use an effective viscosity for the dispersion (Doulah, 1975) to account for turbulence

damping due to the presence of the drop phase. Considering Kolmogorov's microscale, Doulah (1975) showed that:

$$\frac{\epsilon_c}{\epsilon^*} = \left( \frac{\nu^*}{\nu_c} \right)^3 \quad (22)$$

where  $\epsilon_c$  and  $\epsilon^*$  are the energy dissipations for the pure continuous phase and the dispersion, respectively, and  $\nu_c$ ,  $\nu^*$  are the respective kinematic viscosities. In the presence of the drops, the viscosity of the dispersion increases,  $\nu^* > \nu_c$ , and therefore, the energy dissipation from the microscale decreases,  $\epsilon^* < \epsilon_c$ . One can explain this phenomenon as energy consumption from drops for convection, interface oscillation, breakage, and so on. For dilute dispersions,  $\rho_c \approx \rho^*$ , and hence one can write:

$$\frac{\epsilon_c}{\epsilon^*} \approx \left( \frac{\mu^*}{\mu_c} \right)^3. \quad (23)$$

Taylor (1932) derived the following equation for the viscosity of a liquid dispersion:

$$\mu^* = \mu_c \left[ 1 + 2.5 \phi \left( \frac{\mu_d + 0.4 \mu_c}{\mu_d + \mu_c} \right) \right] \quad (24)$$

where  $\mu_d$  is the dispersed phase viscosity. For the toluene (dispersed)-water (continuous) system which has been used in the present study and for which  $\mu_d = 0.6$  cP and  $\mu_c = 1.0$  cP, Eq. 24 gives:

$$\mu^* = 1 + 1.56 \phi \quad (25)$$

Where  $\mu^*$  is in cP units. This relation can be employed to estimate the effect of the holdup on the energy dissipation,  $\epsilon$ . Also, the effect of holdup on the stable drop diameter,  $d_{cr}$ , can be obtained from Eqs. 21, 23 and 24 as:

$$d_{cr} = c_m We^{-0.6} D_i \left[ 1 + 2.5 \phi \left( \frac{\mu_d + 0.4 \mu_c}{\mu_d + \mu_c} \right) \right]^{1.2}. \quad (26)$$

**Inertial Subrange.** It has been assumed that the drop size is in the inertial subrange. This assumption is checked here by comparing the wave number of drops with the limits of the eddy wave number. The largest length scale is of the order of the impeller radius, so  $k_e = 1/r_e = 0.16 \text{ cm}^{-1}$ . The smallest length scale is the Kolmogorov microscale given by:

$$r_n = 2 \left( \frac{\nu^3}{\epsilon} \right)^{1/4} \quad (27)$$

which, for an average impeller energy dissipation of  $5,000 \text{ cm}^2/\text{s}^3$ , gives  $r_n = 0.008 \text{ cm}$  or  $k_n = 125 \text{ cm}^{-1}$ . The average drop size at this energy dissipation is of 1 mm diameter, or  $k_d = 20 \text{ cm}^{-1}$ . Therefore,  $k_e \ll k_d \ll k_n$  which indeed shows that the order of the drop size is in the inertial subrange.

**Energy Dissipation.** The local energy dissipation,  $\epsilon$ , is given as:

$$\epsilon(x, y, z) = k_7(x, y, z) \frac{N^3 D_i^5}{T^2 H} \quad (28)$$

where  $D_i$  is the impeller diameter,  $T$  is the vessel diameter and  $H$  is the height of the tank. For the mean energy dissipation, Schwartzberg and Treybal (1968) used Eq. 28 with  $k_7 = 7.9$ . In the case that  $T = H = 2D_i$ ,

$$\bar{\epsilon} = 0.99N^3D_i^2. \quad (29)$$

Bertrand et al. (1980) used the following relation:

$$\bar{\epsilon} = 5.1 \frac{N^3D_i^5}{V} \quad (30)$$

where

$$V = \frac{\pi T^2 H}{4} = 0.785T^2H. \quad (31)$$

From Eqs. 30 and 31, one can have:

$$\bar{\epsilon} = 6.5 \frac{N^3D_i^5}{T^2H}, \quad (32)$$

and if  $T = H = 2D_i$ ,

$$\bar{\epsilon} = 0.81N^3D_i^2. \quad (33)$$

Equation 32 has been used in this study for the estimation of the mean energy dissipation.

**Turbulence Inhomogeneity in Stirred Tanks.** Several investigators used the mean energy dissipation to describe turbulent flows in stirred-cell vessels. Cutter (1966) compared the energy dissipation rate on the impeller centerline from the impeller tip to the wall and found that the ratio  $\epsilon_i/\bar{\epsilon}$ , where  $\epsilon_i$  is the energy dissipation in the impeller region, varies from 70 near the impeller tip to 3.5 near the wall. Outside the impeller stream, in the circulation region  $\epsilon_c/\bar{\epsilon}$  is roughly 0.26. Coualoglou (1975) used Cutter's data to calculate the mean energy dissipation rate in the impeller and circulation region. The suggested relations are  $\epsilon_i/\bar{\epsilon} = 7.26$  for the impeller region and  $\epsilon_c/\bar{\epsilon} = 0.26$  for the circulation region. Other investigators (Placek and Tavlarides, 1985) reported  $\epsilon_i/\bar{\epsilon} = 5.16$  for  $T = H = 2D_i$ . The above ratios indicate that the turbulence in stirred tanks is highly inhomogeneous. Employment of the mean energy dissipation in Eq. 18 for  $k_3 = 1$ , gave a very large critical diameter compared to experimental observations. The predicted stable drop diameter is even larger if the energy dissipation of the circulation region is used. This behavior leads to the conclusion that drop breakage occurs only in the impeller region, and that Eqs. 21 and 6 hold because of drop breakage in this region. Therefore, the energy dissipation in the impeller region,  $\epsilon_i$ , is used for the estimation of the drop breakage rate. This parameter is obtained from Placek and Tavlarides (1985) since Cutter's data were obtained only on the centerline between the impeller and the wall where energy dissipation is higher, and this method of averaging overestimates the mean energy dissipation in the whole region. Hence,

$$\frac{\epsilon_i}{\bar{\epsilon}} = 5.16. \quad (34)$$

**Eddy-drop Collision Frequency.** Therefore, for the estimation of the eddy-drop collision frequency, one can use Eqs. 16, 23 and 24 to estimate a turbulence damping factor,  $DF(\phi)$  given by:

$$DF(\phi) = \frac{\bar{u}_1^2}{u_2^2} = \left(\frac{\epsilon_c}{\bar{\epsilon}}\right)^{2/3} = \left[1 + 2.5\phi \left(\frac{\mu_d + 0.4\mu_c}{\mu_d + \mu_c}\right)\right]^2 \quad (35)$$

Then, Eqs. 8, 13 and 16 yield the collision frequency between eddies and drops in the impeller stream as follows:

$$h(d) = \frac{V_i}{V_T} DF(\phi) \epsilon_i^{1/3} \frac{0.1\pi}{4} \int_{\frac{2}{d}}^{\frac{2}{d_{e,\min}}} \left(\frac{2}{k} + d\right)^2 \times (8.2k^{-2/3} + 1.07d^{2/3})^{1/2} k^2 dk n_d \quad (36)$$

where  $V_i$  is the volume of the impeller region and  $V_T$  is the total volume of the vessel. The integral term represents the total number of eddies that can break drops of size  $d$ . The upper limit of integration,  $2/d_{e,\min}$ , is a large wave number which corresponds to the eddy size  $d_{e,\min}$ . This eddy size is arbitrarily taken as half the size of the critical drop size. This choice does not introduce any error since eddies of size less than the critical drop diameter do not have enough kinetic energy to break any drop in the system. For the estimation of the volume of the impeller region, the stirred tank is divided into two regions; the circulation region where  $\epsilon_c/\bar{\epsilon} = 0.26$  and the impeller region where  $\epsilon_i/\bar{\epsilon} = 5.16$ . Then, the following energy balance can be written:

$$V_T \bar{\epsilon} = 5.16 \bar{\epsilon} V_i + 0.26 \bar{\epsilon} (V_T - V_i), \quad (37)$$

which yields

$$\frac{V_i}{V_T} = 0.15. \quad (38)$$

The final form of the eddy-drop collision frequency is then written as follows:

$$h(d) = k_8 DF(\phi) \epsilon_i^{1/3} \int_{\frac{2}{d}}^{\frac{2}{d_{e,\min}}} \left(\frac{2}{k} + d\right)^2 \times (8.2k^{-2/3} + 1.07d^{2/3})^{1/2} k^2 dk n_d \quad (39)$$

where  $k_8$  is a geometric factor which in this study takes a value of 0.0118.

**(B) Breakage Efficiency.** The minimum energy required for the drop breakage is assumed to be the energy associated with the generated surface (Narsimhan et al., 1979). For binary breakage, that is, the drop breaks into two drops, the energy required,  $E_c$ , is calculated as the mean value of the energy required for breakage into two equal-size drops and a small and a big daughter drop as follows:

$$E_c = \frac{1}{2} \left\{ 2\pi\sigma \left(\frac{d}{2^{1/3}}\right)^2 + \pi\sigma d_{\max}^2 + \pi\sigma d_{\min}^2 - 2\pi\sigma d^2 \right\}. \quad (40)$$

The breakage efficiency,  $B$ , which gives the probability of an eddy-drop collision to result in drop breakage, is then assumed to be given by the following exponential function (Coulaloglou and Tavlarides, 1977; Prince and Blanch, 1990):

$$B = \exp \left[ -\frac{E_c}{c_1 e} \right] \quad (41)$$

where  $e$  is the average energy of an eddy given by Eq. 14, and  $c_1$  is a constant of order 1. For large values of  $e$  the breakage efficiency approaches 1, while for small values of  $e$  it approaches 0. Since  $e$  is a function of the eddy size, the breakage efficiency expression has to be introduced in the integral term of the collision frequency which is given by Eq. 39. Then, the breakage frequency becomes:

$$g(d) = 0.0118 DF(\phi) \epsilon_1^{1/3} \int_{\frac{2}{d}}^{\frac{2}{d_{\min}}} \left( \frac{2}{k} + d \right)^2 \times (8.2k^{-2/3} + 1.07d^{2/3})^{1/2} k^2 \exp \left[ -\frac{E_c}{c_1 e} \right] dkn_d. \quad (42)$$

The integral term can be estimated numerically. The least-square value of the constant  $c_1$  has been found to be equal to 1.3 by using calculated and experimental data of transient drop size distributions in a stirred tank. The fact that this value is greater than one shows that either breakage is a result of simultaneous collisions of more than one eddies with the drop, or breakage occurs only near the impeller tip where energy dissipation is much higher.

The breakage frequency is calculated by Eq. 42 as a function of drop size at various values of holdup and rpm. Results are shown in Figures 1a and b. In Figure 1a, the effect of agitation speed on the breakage frequency is shown, while in Figure 1b, the effect of the holdup is presented.  $d_{\max}$  values are also calculated for all curves by Eq. 26 for  $c_m = 0.125$  (Sprow, 1967; Lagisetty et al., 1986) and are shown on both graphs. The model predicts increasing breakage frequency with increasing rpm and decreasing holdup. Calculations show that the breakage frequency goes to zero in the neighborhood of  $d_{\max}$ . This is very encouraging and supports the validity of the developed breakage frequency.

### Daughter drop probability density function

Sufficient experimental work does not exist in the open literature to assist in developing an adequate daughter drop probability density function in turbulent dispersions. Hancil and Rod (1981) obtained some experimental data which are not applicable in this study due to differences in the system geometry. For the daughter drop distribution function, Coulaloglou (1975) used a normal distribution. Hsia and Tavlarides (1983) used a beta distribution which gives 100% probability density within two acceptable size limits. Both beta and normal distributions are bell-shape functions with higher probability density in the middle of the distribution. However, it can be shown that the energy requirement for a breakage into two equal-size drops is greater than the energy requirement for a breakage into a small and a large drop. This consideration

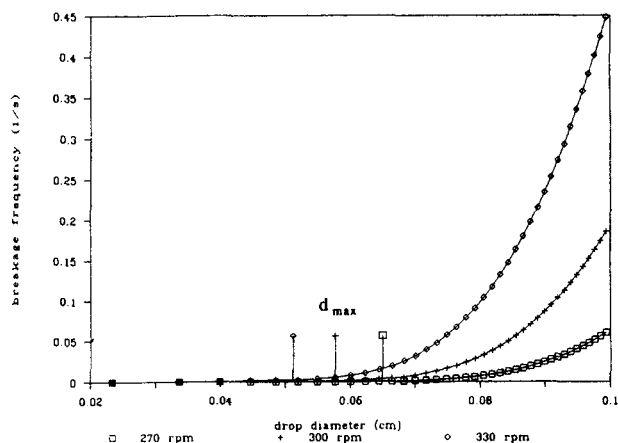


Figure 1a. Breakage frequency.

Effect of agitation speed;  $\phi = 0.3$  ( $d_{\max}$  is calculated from Eq. 26).

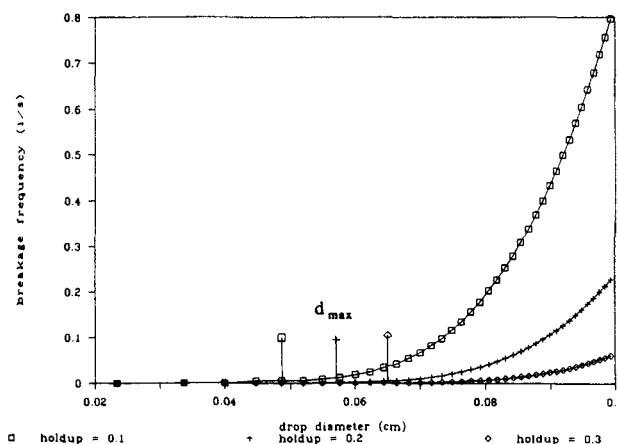


Figure 1b. Breakage frequency.

Effect of holdup; 270 rpm ( $d_{\max}$  is calculated from Eq. 26).

leads to the conclusion that the daughter drop distribution function should be bimodal with higher probability density at the ends than in the middle. Such a distribution function is proposed here by assuming that it is linearly related to the energy requirements for the formation of the daughter drops. Since the energy requirement for the drop breakage is proportional to the new surface area that is created by drop breakup, the new daughter drop density function is a quadratic symmetric function of the drop diameter. It is also assumed that there exists a minimum drop size for the daughter drops, otherwise the probability at the ends of the distribution would be infinity. If, the energy needed to create the smallest and largest drops is  $\epsilon_{\min}$  and the energy needed to create two equal daughter drops is  $\epsilon_{\max}$ , then the daughter drop probability density function for the breakage of a drop of size  $d'$  is written as:

$$\beta(d, d') = \frac{\epsilon_{\min} + [\epsilon_{\max} - \epsilon(d)]}{\int_0^{d'} \{\epsilon_{\min} + [\epsilon_{\max} - \epsilon(d)]\} \delta d} \quad (43)$$

where  $\epsilon(d=d') = \epsilon(d=0) = \epsilon_{\min}$  and  $\epsilon(d=d'/2) = \epsilon_{\max}$ . In the discrete domain, the above function is written as:

$$\beta(i, n+1) = \frac{\epsilon_{\min} + (\epsilon_{\max} - \epsilon_i)}{\sum_{i=1}^n [\epsilon_{\min} + (\epsilon_{\max} - \epsilon_i)]}$$

$$= \frac{d_1^2 + d_n^2 + 2d_{n+1}^2 - d_{n+1}^2 - d_i^2 - d_{n+1-i}^2}{2} \quad (44)$$

$$= \frac{n(d_1^2 + d_n^2 + 2d_{n+1}^2 - 2d_{n+1}^2) - \sum_{i=1}^n (d_i^2 + d_{n+1-i}^2 - d_{n+1}^2)}{2}$$

where  $\beta(i, n+1)$  is the probability density to produce drops of size  $i$  from breakage of drops of size  $n+1$  and  $d_i$  is the drop diameter for the  $i$ th discrete element. In Eq. 44,  $\epsilon_{\min} = \pi(d_1^2 + d_n^2 - d_{n+1}^2)\sigma$ ,  $\epsilon_{\max} = \pi(2d_{n+1}^2 - d_{n+1}^2)\sigma$ , and  $\epsilon_i = \pi(d_i^2 + d_{n+1-i}^2 - d_{n+1}^2)\sigma$  where  $\sigma$  is the interfacial tension. Finally, to completely describe the drop breakage process, one has to have an estimate of the average number of drops produced by breakage of a larger drop of a certain size. Due to lack of such information, a binary drop breakage is assumed, and thus the parameter  $\nu(v')$  in Eq. 3 is always 2. It is clear here that there is a need for further studies to experimentally determine the daughter drop probability density, as well as the number of daughter drops produced by breakage of a bigger drop.

## Drop Coalescence Rate

### Introduction

The drop coalescence rate has been described by Coulaloglou and Tavarides (1977) as the product of the collision frequency,  $h(d_m, d_j)$ , and the coalescence efficiency,  $\lambda(d_m, d_j)$  of drops of diameter  $d_m$  and  $d_j$ . The following relations have been suggested by the above investigators:

*Collision Frequency:*

$$h(d_m, d_j) = k_9 \frac{\epsilon^{1/3}}{1 + \phi} (d_m + d_j)^2 (d_m^{2/3} + d_j^{2/3})^{1/2} \quad (45)$$

*Coalescence Efficiency:*

$$\lambda(d_m, d_j) = \exp \left[ -k_{10} \frac{\mu_c \rho_c \epsilon}{\sigma^2 (1 + \phi)^3} \left( \frac{d_m d_j}{d_m + d_j} \right)^4 \right] \quad (46)$$

where  $\mu_c$ ,  $\rho_c$  are the continuous phase viscosity and density, respectively,  $\sigma$  the interfacial tension, and  $k_9$ ,  $k_{10}$  are universal constants.

Sovova (1981) suggested that the above relation for the coalescence efficiency, Eq. 46, favors coalescence of the small drops and replaced it with the following function which is based on the energy of drop collision:

$$\lambda(v, v') = \exp \left[ -k_{11} \frac{\sigma(v^{2/3} + v'^{2/3})(v + v')}{\rho_d N^2 D_i^{4/3} v v' (v^{2/9} + v'^{2/9})} \right] \quad (47)$$

In this relation, the exponential term is supposed to be the ratio of the interfacial energy over the energy of collision. However, an exact representation of this ratio gives a different form for the coalescence efficiency:

$$\lambda(v, v') = \exp \left[ -k_{11} \frac{\sigma(v^{2/3} + v'^{2/3})}{\rho_d N^2 D_i^{4/3} (v^{11/3} + v'^{11/3})} \right] \quad (48)$$

In the above relations,  $v, v'$  represent drop volumes and  $k_{11}$  is a proportionality constant. Sovova's coalescence efficiency has been found to promote coalescence of larger drops. Also, because of this behavior, it fails to predict the coalescence rate in a multistage extractor where the average drop size is larger than in a stirred tank.

In recent years, Ramkrishna and coworkers have contributed significantly to the knowledge of drop coalescence mechanisms. Das et al. (1987) introduced a white-noise model which considers the film drainage between two colliding drops as a stochastic process driven by a suitably idealized random process for the fluctuating force applied on the drop pair. According to this model, the drops are considered nondeformable, and the critical thickness of the film before rupture is assumed to be specific for a given system. A quite interesting prediction of this model is the higher coalescence efficiency for higher continuous phase viscosity. Furthermore, a number of parameters are introduced in this model which are very difficult to measure or estimate. The white-noise model is not appropriate if the intervening film between rigid spheres is associated with large drainage rates. This fact is acknowledged by Muralidhar and Ramkrishna (1986). These investigators employed a time-scale analysis to understand the significance of the factors affecting drop coalescence. They considered both deformable and nondeformable drops and showed the conditions under which the white-noise or the band-limited-noise models are valid. These two models predict different effects of physical properties on the coalescence efficiency. Muralidhar et al. (1988) studied the coalescence of rigid drops in a stirred dispersion under the action of a stochastic force representing the effect of turbulence. A ratio of the characteristic times of force fluctuations and film drainage is defined in this study. A colored-noise model is then derived for the case in which this ratio is of order one. As the ratio of the force fluctuations and film drainage characteristic times increases, the film drainage becomes faster than force fluctuations. In this case, the force applied on the drop pair can be regarded as a random variable, and the approach of Coulaloglou and Tavarides can be employed. It is interesting that, for the conditions of a stirred tank, the ratio of the characteristic times is around 3 for 100  $\mu\text{m}$ -dia. drops and increases rapidly with the drop size. In the present study, only a small percentage of the dispersed phase (order of 5% by volume) is due to drops of diameters smaller than 100  $\mu\text{m}$ , therefore the approximation of Coulaloglou and Tavarides is considered valid.

Tobin et al. (1990) studied the effect of drop size on drop coalescence frequencies. Purely drop coalescence experiments have been designed in this study, although breakage was not completely ruled out. In these experiments, a negative step input is introduced in the energy provided to the dispersion by agitation and the evolution of the drop size distribution to a new steady state is observed. During this transient response, coalescence is the predominant phenomenon, and one can ignore the effect of drop breakage rates on the size distribution. From the obtained data, the investigators were able to draw conclusions on the effect of drop size on the drop coalescence. Furthermore, the above investigators studied the inverse prob-

lem of the population balance equation to derive coalescence frequencies. Among the conclusions of their study is that coalescence of small drops is not as frequent as the model of Coulaloglou and Tavlarides (1977) predicts, and a kinetic collision mechanistic model can describe the transient coalescence data best. In this study, a modification of the Coulaloglou and Tavlarides model is proposed.

### Model development

According to Coulaloglou and Tavlarides (1977), the coalescence frequency of drops of size  $d_i$  and  $d_j$  is given by the product of the drop collision frequency (A) and the coalescence efficiency (B) as follows:

$$F(d_i, d_j) = h(d_i, d_j) \lambda(d_i, d_j) n_i n_j \quad (49)$$

(A) *Collision Frequency*. Similar to the eddy-drop collision frequency which has been examined earlier, the drop-drop collision frequency can be obtained by assuming that the drops in a turbulent flow behave like gas molecules. Therefore, the collision frequency can be written as:

$$h(d_i, d_j) = \frac{\pi}{4} (d_i + d_j)^2 (\bar{u}_i^2 + \bar{u}_j^2)^{1/2} n_i n_j \quad (50)$$

where  $\bar{u}^2$  is obtained from Eq. 16.

(B) *Coalescence Efficiency*. Coulaloglou and Tavlarides (1977) assumed that once the drops collide they stay together for some time, called contact time, and then they either coalesce or bounce away from each other. Coalescence occurs if the contact time is large enough so that the liquid film between the two drops drain out until a critical film thickness is reached. The time needed for the film drainage is called coalescence time. Therefore, coalescence occurs when the contact time is larger than the coalescence time. Assuming that the flow in the continuous phase in the film is laminar and the inertial effects are negligible, the velocity profile in the film is parabolic, the approaching interfaces resist to normal and tangential stresses, and Van der Waals and double layer forces are negligible, Eq. 46 was obtained for the coalescence efficiency of deformable drops.

Another coalescence efficiency function is considered in the present study based on the above model. Assuming that the coalescence time  $T$  and the contact time  $t$  are random variables and that the coalescence time is normally distributed, Ross (1971) derived the following expression for the probability of coalescence:

$$\lambda(d_1, d_2) = \frac{1}{2} \exp\left(-\frac{\bar{T}}{\bar{t}}\right) \exp\left(\frac{1}{2} \frac{\sigma_T^2}{\bar{t}^2}\right) \operatorname{erfc}\left(\frac{\sqrt{2}}{2} \frac{\sigma_T^2 - \bar{T}\bar{t}}{\bar{t}\sigma_T}\right) \quad (51)$$

where  $\bar{t}$ ,  $\bar{T}$  are the average contact and coalescence time, respectively and  $\sigma_T$  is the standard deviation for the coalescence time. Equation 51 was simplified to the following form by Coulaloglou (1975) who set  $\sigma_T = 0$ , that is, the coalescence time is not distributed although the contact time remains a random variable:

$$\lambda(d_i, d_j) = \exp\left[-\frac{\bar{T}}{\bar{t}}\right] \quad (52)$$

For two spherical nondeformable solid spheres of radius  $r_1$  and  $r_2$ , the drainage time is given by Jeffreys and Davis (1971) as:

$$\bar{T} = t_2 - t_1 = \frac{6\pi\mu_c}{F_1} \left(\frac{r_1 r_2}{r_1 + r_2}\right)^2 \ln\left(\frac{h_1}{h_2}\right) \quad (53)$$

where  $F_1$  is the force compressing the drops together,  $h_1$  is the film thickness at time  $t_1$ , and  $h_2$  is the film thickness at time  $t_2$ . Equation 53 has been derived by equating the mechanical energy acting on a pair of drops by the compressing force to the energy dissipated by the fluid viscosity.

In a recent study, Davis et al. (1989) estimated the hydrodynamic force resisting the relative motion of two unequal drops moving along their line of centers for Stokes flow conditions. For two solid spheres, the hydrodynamic force,  $F_2$ , resisting their motion towards each other is given by:

$$F_2 = \frac{6\pi\mu_c a^2 W}{h} \quad (54)$$

where  $a = r_1 r_2 / (r_1 + r_2)$ ,  $h$  is the centerline distance between the surfaces of the two spheres, and  $W$  is the approaching velocity,  $dh/dt$ . Equation 53 can be derived from Eq. 54 if it is assumed that the film drainage rate between the two spheres is constant under a compressing force,  $F_1$ . This assumption is equivalent to saying the total force applied on the spheres is zero,  $F = F_1 + F_2 = 0$ .

For fully mobile drop interfaces, the resisting hydrodynamic force is given by Davis et al. (1989) as follows:

$$F_2 = 16.5\mu_d a^{3/2} h^{-1/2} W \quad (55)$$

Employing the above assumption ( $F = F_1 + F_2 = 0$ ), one can derive the following relation for the film drainage time from  $h_1$  to  $h_2$  thickness:

$$t_2 - t_1 = \frac{16.5\mu_d a^{3/2}}{F_1} (h_1^{1/2} - h_2^{1/2}) \quad (56)$$

The film drainage time in Eq. 56 is a function of the drop phase viscosity,  $\mu_d$  rather than of the continuous phase viscosity,  $\mu_c$ .

For drops with partially mobile interfaces, the resisting hydrodynamic force,  $F_2$ , is approximated by Davis et al. (1989) as:

$$F_2 = \frac{6\pi\mu_c a^2}{h} W \frac{1 + 0.38m}{1 + 1.69m + 0.43m^2} \quad (57)$$

The above authors defined the term  $m = \lambda_\mu^{-1} \sqrt{a/h}$  as a dimensionless parameter having the character of an interface mobility. Here  $\lambda_\mu = \mu_d / \mu_c$ . Assuming again constant film drainage rate, one can obtain the film drainage time between two undeformable drops from  $h_1$  to  $h_2$  thickness as:



$$\bar{T} = t_2 - t_1 = \frac{6\pi\mu_c a^2}{F_1} \zeta \quad (58)$$

where

$$\zeta = 1.872 \ln \left[ \frac{h_1^{1/2} + 1.378q}{h_2^{1/2} + 1.378q} \right] + 0.127 \ln \left[ \frac{h_1^{1/2} + 0.312q}{h_2^{1/2} + 0.312q} \right]$$

$$q = \lambda_\mu^{-1} a^{1/2}$$

Equations 58 and 56 predict a finite time for the complete drainage of the film (that is,  $h_2 = 0$ ) while Eq. 53 which is applied for solid spheres predicts an infinite time for the film to drain out completely. It has been reported by many investigators that the film rupture occurs when  $h_2$  becomes smaller than 500 Å. For  $\lambda_\mu = 0(1)$ , as it is usually the case for the aqueous-organic liquid dispersions, and for  $h_2 < 500$  Å and  $h_2 \ll a$ , it can be shown that  $h_2^{1/2} \ll 0.312\lambda_\mu^{-1} a^{1/2}$ . The relations 54, 55 and 57 were derived for  $h \ll a$  and, therefore,  $h_1$  is taken as  $h_1 \approx k_{13}a$  where  $k_{13} \ll 1$ . Here,  $k_{13}$  is arbitrarily set to 0.1.

Following Ross (1971),  $F_1$  is given by:

$$F_1 = k_{12}\rho_c \epsilon^{*2/3} a^2 (d_1 + d_2)^{2/3}. \quad (59)$$

where  $\epsilon^*$  is given by Eqs. 23 and 24. Then, Eq. 58 becomes:

$$\bar{T} = \frac{6\pi\mu_c \zeta}{k_{12}\rho_c \epsilon^{*2/3} (d_1 + d_2)^{2/3}}. \quad (60)$$

The contact time,  $\bar{t}$ , is taken as the reciprocal of the frequency of fluid velocity fluctuations which is given by Schwartzberg and Treybal (1968):

$$\bar{t} = \left[ \frac{2.5ND_i^2}{L(T^2H)^{1/3}} \right]^{-1} \quad (61)$$

where  $L = 0.08D$  (determined by Cutter, 1960). Equations 60 and 61 yield:

$$\frac{\bar{T}}{\bar{t}} = \frac{6\pi\mu_c c_2 \zeta}{\rho_c \epsilon^{*2/3} (d_1 + d_2)^{2/3}} \frac{31.25ND_i}{(T^2H)^{1/3}} \quad (62)$$

Then, the coalescence efficiency becomes:

$$\lambda(d_1, d_2) = \exp \left[ - \frac{6\pi\mu_c c_2 \zeta}{\rho_c \epsilon^{*2/3} (d_1 + d_2)^{2/3}} \frac{31.25ND_i}{(T^2H)^{1/3}} \right]. \quad (63)$$

The best value of the constant  $c_2$  has been found to be 3.44. A probable reason that this constant is much larger than 1 is due to the fact that at a drop collision, the compressing force,  $F_1$ , does not act on both drops along the center of line. The above coalescence efficiency increases with lower continuous and dispersed phase viscosity, larger drop size and higher energy input. These trends are in accord to observations of other investigators. For example, the effect of drop size is similar to experimental observations by Tobin et al. (1990), and the effect of energy input is similar to the results of Madden and Damerell (1962). The interfacial tension does not appear in

Eq. 63 due to the assumption of spherical drops. An attempt has also been made to account for deformable drops by considering approach of flat parallel disks after drop deformation occurs. In this case, the coalescence time is the summation of the drainage times of (i) spheres and (ii) flat disks. The problem of approaching drops with plane parallel disks has been studied by Ivanov and Traykov (1976). Computations in the present study, based on the results of these investigators, showed that the drainage time of the film between approaching disks depends greatly on the film thickness at which film rupture occurs. Also, even for film thickness as small as 25 Å, the drainage time for spheres was found to be much larger than for flat disks and, therefore, the assumption of nondeformable drops was adopted.

Using Eqs. 42, 43, 50, 16 and 63, one can obtain drop breakage and coalescence rates which can be utilized by the population-balance-equation model, Eq. 1, for calculations of the transient drop size distribution in a batch stirred-tank reactor. Discretization of Eq. 1 leads to a system of nonlinear ordinary differential equations which form an initial value problem. The EPISODE package by Byrne and Hindmarsh (1976) has been used for the solution of this problem.

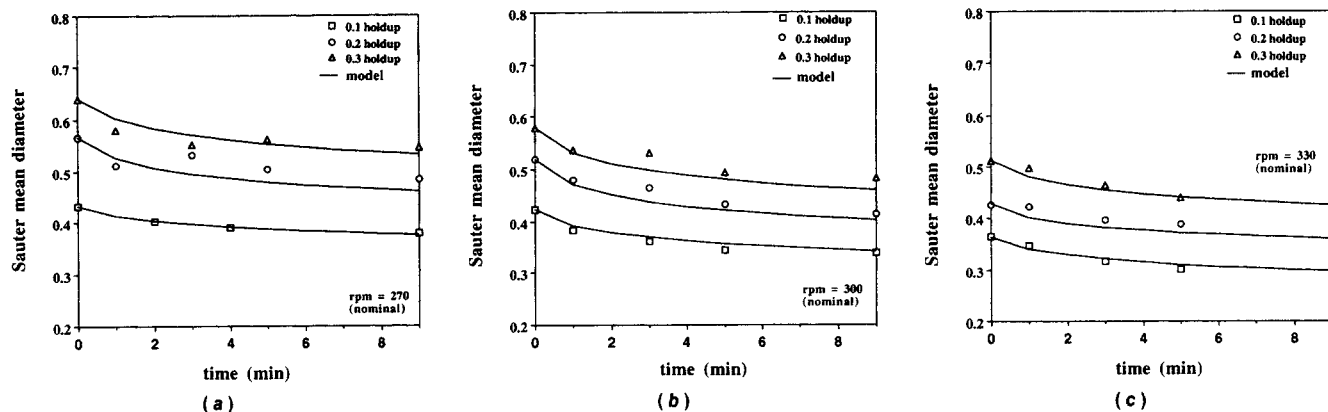
## Experimental Data vs. Calculated Results from the Population Balance Equation

A batch stirred-tank reactor and the laser capillary technique described by Bae and Tavlarides (1989) have been used to generate transient drop size distribution data. The laser capillary technique is able to measure bivariate distributions of drop size and concentration by monitoring the intensity of laser light penetrating the walls of a capillary glass tube through which sampled drops and continuous-phase liquid are forced to pass. A similar approach for on-line drop size measurements has been employed by Chatzi et al. (1991). The tank is of 0.75 L volume, 100 mm diameter and 97 mm height. The stirrer is a six-blade Rushton type impeller of 50 mm diameter. Four baffles are located near the wall at equally spaced positions. Initially, predetermined quantities of mutually equilibrated water and toluene are introduced into the tank forming two layers. Then, agitation of the two liquids is started at time  $t = 0$ , and drop size measurements are obtained at  $t = 60$  s, 120 s, 240 s, 360 s, and 600 s. Three different nominal values of the agitation speed have been selected, that is, 270 rpm, 300 rpm and 330 rpm at holdup values of 0.1, 0.2 and 0.3. At each measurement, the average drop diameter of a sample of approximately 300 drops has been determined. Sampling time is approximately 20 seconds. The Sauter mean diameter, defined as:

$$d_{32} = \frac{\sum_{i=1}^n d_i^3}{\sum_{i=1}^n d_i^2}, \quad (64)$$

and the cumulative volume fraction have been calculated from the sample. The first measurement of the drop size distribution at time  $t = 60$  s is used as the initial condition for the solution of the population balance equation.

Comparisons of model results with experimental data are shown in Figures 2-7. The optimum values of the two model



**Figure 2. Calculated and experimental Sauter mean diameter in a stirred tank.**

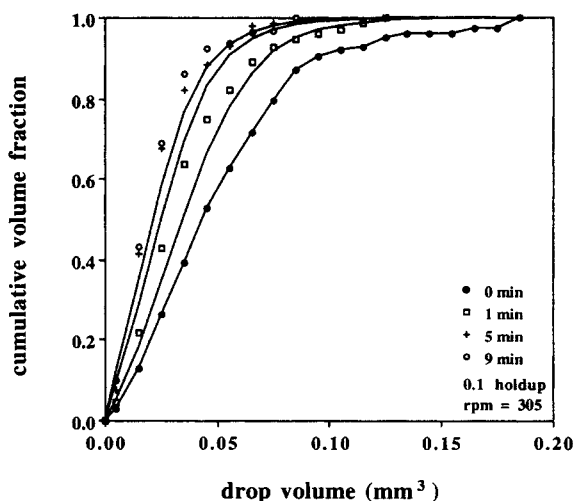
System: toluene (dispersed)—water (continuous): (a) 270 rpm; (b) 300 rpm, (c) 330 rpm.

parameters  $c_1 = 1.5$  and  $c_2 = 28.1$  were obtained by least-square analysis. The transient Sauter mean diameter is shown in Figures 2a, 2b and 2c for 270, 300 and 330 rpm, respectively. Drop size distributions in terms of the cumulative volume fraction are compared with experimental results in Figures 3–7. Figures 3, 4 and 5 show the effect of holdup for 300 rpm nominal agitation speed, while Figures 5, 6 and 7 show the effect of the agitation speed for 10% holdup. The exact values of the agitation speed are shown in each graph. Figures 3–7 show that the agreement between calculated and experimental results is fairly good. At some conditions, however, it is clear that further improvement can be obtained.

## Conclusions

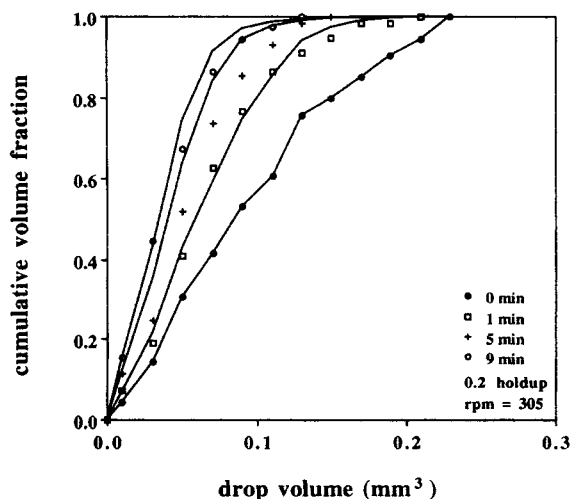
Drop breakage and coalescence models are proposed based on a phenomenological approach to describe these processes for drops in turbulent dispersions. These models overcome some inconsistencies and limitations observed in previous

models, and they can be employed to describe dense dispersions up to 30% holdup. The obtained drop frequency functions are incorporated in the population balance equation to compare the model predictions with transient drop size distributions and to determine the model parameters. Eddy-drop interactions are considered for the drop breakup. This approach also leads to the derivation of a formula for the maximum stable drop size. The same formula was given in the literature as a correlation of experimental results. A new daughter drop probability density is proposed based on the energy requirement to create the daughter drops by breakup of a larger one. Experimental data obtained for the physically equilibrated toluene water system for batch operation of a stirred-tank contactor employing the laser capillary technique are used for parameter estimation. The comparison between experimental data and calculations indicates some inadequacies of the modeling which have to be further investigated. Also, the models described here do not include the effect of the drop surface charge which has been reported recently (Tobin et al., 1990) to play a sig-



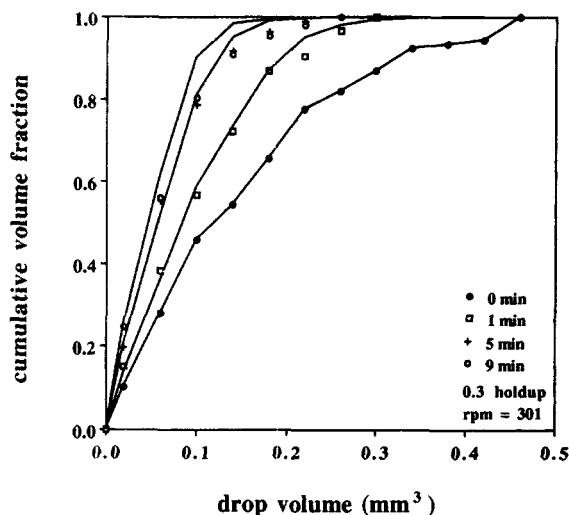
**Figure 3. Calculated and experimental cumulative drop volume fraction in a stirred tank.**

System: toluene (dispersed)—water (continuous); agitation speed: 305 rpm; 0.1 holdup.



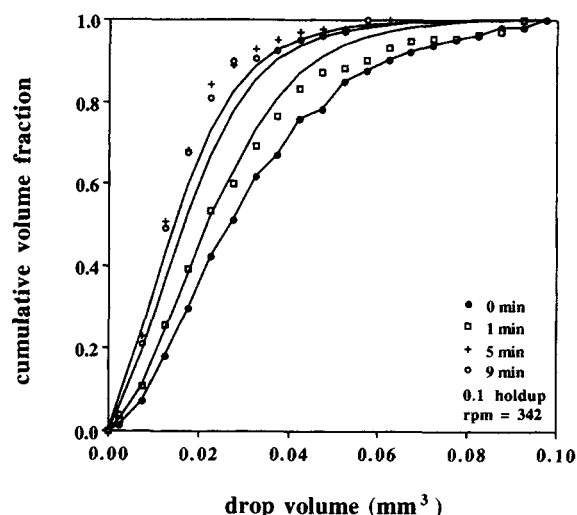
**Figure 4. Calculated and experimental cumulative drop volume fraction in a stirred tank.**

System: toluene (dispersed)—water (continuous); agitation speed: 305 rpm; 0.2 holdup.



**Figure 5. Calculated and experimental cumulative drop volume fraction in a stirred tank.**

System: toluene (dispersed)-water (continuous); agitation speed: 301 rpm; 0.3 holdup.



**Figure 7. Calculated and experimental cumulative drop volume fraction in a stirred tank.**

System: toluene (dispersed)-water (continuous); agitation speed: 330 rpm; 0.1 holdup.

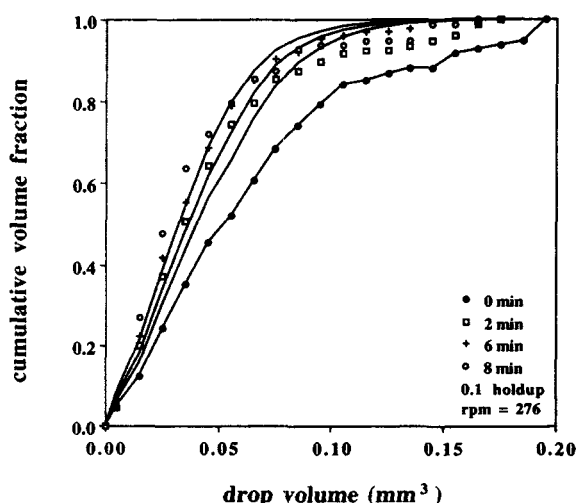
nificant role. The drop breakage and coalescence frequency functions obtained here are employed elsewhere (Tsouris et al., 1993) without any modification for the analysis of a stirred-cell column extractor.

## Acknowledgment

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## Notation

- $B$  = birth terms for drops  
 $c_m, c_i$  = proportionality constant  
 $d$  = drop diameter, m



**Figure 6. Calculated and experimental cumulative drop volume fraction in a stirred tank.**

System: toluene (dispersed)-water (continuous); agitation speed: 276 rpm; 0.1 holdup.

- $d_e$  = eddy diameter, m  
 $d_{\max}, d_{cr}$  = maximum stable drop size, m  
 $d_{32}$  = Sauter mean diameter, m  
 $D$  = death terms for drops  
 $D_i$  = impeller diameter, m  
 $e$  = energy of a single eddy,  $\text{kg} \cdot \text{m}^2 \cdot \text{s}^{-2}$   
 $E(k)$  = energy of eddies of wave number  $k$ ,  $\text{kg} \cdot \text{m}^3 \cdot \text{s}^{-2}$   
 $E_c$  = energy required for breakage,  $\text{kg} \cdot \text{m}^2 \cdot \text{s}^{-2}$   
 $F$  = force, N  
 $F(d_i, d_j)$  = coalescence frequency,  $\text{s}^{-1}$   
 $g(d)$  = breakage frequency,  $\text{s}^{-1}$   
 $g(v)$  = breakage frequency,  $\text{s}^{-1}$   
 $h$  = film thickness, m  
 $h(d_i, d_j)$  = collision frequency,  $\text{s}^{-1}$   
 $h(v_i, v_j)$  = collision frequency,  $\text{s}^{-1}$   
 $H$  = tank height, m  
 $k$  = eddy wavenumber,  $\text{m}^{-1}$   
 $k_i$  = proportionality constant  
 $m = \lambda_\mu^{-1} \sqrt{a/h}$   
 $n(v, t)$  = number of drops of volume  $v$  at time  $t$   
 $n_d$  = number of drops  
 $n_e$  = number of eddies  
 $N$  = agitation speed,  $\text{s}^{-1}$   
 $q = \lambda_\mu^{-1} a^{1/2} = m h^{1/2}$   
 $r$  = radius, m  
rpm = agitation speed,  $\text{min}^{-1}$   
 $s_{ij}$  = collision cross-sectional area of drops  $i$  and  $j$ ,  $\text{m}^2$   
 $\bar{t}$  = average contact time, s  
 $T$  = vessel diameter, m  
 $\bar{T}$  = average coalescence time, s  
 $u$  = velocity,  $\text{m} \cdot \text{s}^{-1}$   
 $v$  = drop volume,  $\text{m}^3$   
 $V_i$  = volume of the impeller region,  $\text{m}^3$   
 $W$  = drop velocity,  $\text{m} \cdot \text{s}^{-1}$   
 $We$  = Weber number =  $N^2 \rho_i D^3 \sigma^{-1}$

## Greek letters

- $\beta(v_i, v_j)$  = probability density of drops  $v_i$  produced by breakage of drops  $v_j$   
 $\epsilon$  = energy dissipation,  $\text{m}^2 \cdot \text{s}^{-3}$   
 $\zeta$  = defined in Eq. 58  
 $\lambda_\mu$  = ratio of drop phase to continuous phase viscosity,  $\mu_d/\mu_c$   
 $\lambda(d_i, d_j)$  = coalescence efficiency  
 $\lambda(v_i, v_j)$  = coalescence efficiency  
 $\mu$  = viscosity,  $\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$

- $\nu$  = kinematic viscosity,  $\text{m}^2 \cdot \text{s}^{-1}$   
 $\nu(v)$  = number of drops produced from a drop breakup of volume  $v$   
 $\rho, \rho_f$  = density,  $\text{kg} \cdot \text{m}^{-3}$   
 $\sigma$  = interfacial tension,  $\text{NM}^{-1}$   
 $\phi$  = volume fraction of the dispersed phase (holdup)

## Subscripts

- $c$  = continuous phase  
 $d$  = dispersed (drop) phase

## Superscripts

- $*$  = dispersion (both phases)

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