R & D NOTES

Drop Breakup in Turbulent Stirred-Tank Contactors

Part III: Correlations for Mean Size and Drop Size Distribution

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In Part II, the extent to which dispersed-phase viscosity and interfacial tension influence equilibrium mean drop size and drop size distribution was determined for dilute suspensions produced in baffled cylindrical tanks of standard geometry equipped with six-blade Rushton turbines. Low to moderate viscosity ($\mu_d \lesssim 0.5$ Pa·s) dispersed-phase systems behaved similarly in that Sauter mean diameter could be correlated using the mechanistic arguments of Part I, and drop sizes, normalized with respect to D_{32}, could be correlated by a normal distribution in volume. Limited moderate viscosity data were reported in Part I but were not used to develop the correlations of Part II. The objective of this study is to combine the low to moderate viscosity data of Parts I and II with those obtained by other investigators to obtain correlations of broader utility, and to extend these via mechanistic arguments so that they apply to nondilute systems.

Dilute Dispersions

Several investigators have studied the behavior of dilute liquid-liquid systems in the geometry of Parts I and II (see Figure 1 of Part I). Chen and Middleman (1967) conducted a detailed study of surface forced stabilized (low μ_d) dispersions encompassing a broad range of operating conditions. They considered dispersed phases with viscosities up to about 0.025 Pa·s but did not account for viscous resistance to breakage when correlating their data. Sprow (1967) obtained limited data for inviscid dispersed phases, while Arai et al. (1977) acquired limited data showing the effect of dispersed-phase viscosity on maximum stable drop size at constant interfacial tension. The apparatus employed in these studies differed from that of Parts I and II in only two respects. Baffles were mounted flush to tank walls and bottom. Chen and Middleman varied the ratio of impeller to tank diameter (L/T) and Sprow obtained most of his data for $L/T = 0.29$.

For dilute suspensions, coalescence rates are negligible. Equilibrium drop sizes are determined by breakup that occurs primarily in the impeller region. Small modifications in baffle placement should be relatively unimportant and the L/T ratio should be of secondary importance. Chen and Middleman found that the effect of L/T fell within the scatter in their data for the range $0.21 \leq L/T \leq 0.73$. However, it should be noted that Okamoto et al. (1981) report that energy dissipation rates become more uniform as L/T increases. Therefore, the ratio of the maximum to mean energy dissipation rate per unit mass increases as L/T decreases. For a given $\bar{\epsilon}$, drops will experience higher local turbulent energy as L/T decreases, indicating that D_{32} should decrease with L/T.

Mean drop size correlations

The low to moderate viscosity data reported in Parts I and II and by the cited investigators were fit to models developed in Part II. The range of variables investigated in each study is summarized in Table 1. The table provides information for dilute, low to moderate viscosity dispersed phases, only, since these are the systems to which the correlations of Part II apply. For instance, higher viscosity data reported in Parts I and II and by Arai et al. are not included. Data reported by Sprow for nondilute dispersions are not given. Arai et al. reported maximum stable drop size. Based on the data of Parts I and II and of Chen and Middleman, it was assumed that $D_{32} \simeq 0.6\ D_{max}$.

Two models were fitted to the 349 data sets via nonlinear least-squares regression. These are the semitheoretical model

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Table 1. Data Set for Mean Drop Size Correlation

	Part I*	Part II*	Arai et al. (1977)**	Chen and Middleman (1967)†	Sprow (1967)‡
	Number of Experiments				
	60	146	25	110	8
L, m	0.071–0.196	0.071–0.156	0.064	0.051–0.152	0.064
L/T	0.5	0.5	0.5	0.21–0.73	0.29
N, rps	0.93–4.78	1.42–4.67	3.00–14.33	1.33–16.67	4.17–33.33
σ, N/m	0.0378	0.00021–0.047	0.022	0.00475–0.0483	0.0418
μ_d, Pa · s	0.096–0.486	0.00081–0.459	0.00078–0.520	0.00052–0.0258	0.00051
μ_c, Pa · s	0.00089	0.00052–0.00089	0.00097	0.00089–0.00127	0.00099
ρ_d, kg/m^3	960–975	834–985	879–922	703–1,101	692
ρ_c, kg/m^3	997	792–997	1,000	993–1,001	1,005
ϕ	<0.0015	<0.002	<0.003	0.001–0.005	0.005
$Re = \rho_c N L^2/\mu_c$	13,000–101,000	14,000–83,000	12,470–59,570	12,000–104,000	17,060–136,440
$We = \rho_c N^2 L^3/\sigma$	44–1,137	54–70,960	104–2,368	70–2,000	107–6,840
$Vi = \left(\frac{\rho_c}{\rho_d}\right)^{1/2} \mu_d N L/\sigma$	0.326–5.90	0.0041–602.8	0.0071–22.6	0.0024–1.18	0.0039–0.031

*Data reported for $\mu_d > 0.5$ Pa · s are not included here.

**Data reported for $\mu_d = 1.5$ Pa · s are not included here. Arai et al. report D_{max}. It is assumed that $D_{32} \simeq 0.6\ D_{max}$.

†Entire data set as reported by Chen (1966).

‡Data reported for $\phi > 0.005$ are not included here.

given by Eq. 1 of Part II

$$\frac{D_{32}}{L} = A\ We^{-3/5}\left[1 + B_1\ Vi\left(\frac{D_{32}}{L}\right)^{1/3}\right]^{3/5} \tag{1}$$

and the simpler model based on arguments given by Hinze (1955) and Sleicher (1962), reported as Eqs. 7 and 8 of Part II.

$$\frac{D_{32}}{L} = A\ We^{-3/5}\ [1 + B_2\ Vi^c]^{3/5} \tag{2}$$

Equation 1 yields

$$\frac{D_{32}}{L} = 0.054\ We^{-3/5}\left[1 + 4.42\ Vi\left(\frac{D_{32}}{L}\right)^{1/3}\right]^{3/5} \tag{3}$$

with a root mean square deviation, defined by Eq. 13 of Part II, of $\sigma_{rms} = 23.1\%$. The goodness of fit of Eq. 3 is shown in Figure 1.

Equation 2 yields

$$\frac{D_{32}}{L} = 0.053\ We^{-3/5}\ [1 + 0.91\ Vi^{0.84}]^{3/5} \tag{4}$$

with $\sigma_{rms} = 23.1\%$ and goodness of fit given by Figure 2.

Equations 3 and 4 are remarkably similar to Eqs. 14 and 15 of Part II, respectively, even though the number of data sets and the range of system parameters are considerably larger. The goodness of fit as measured by σ_{rms} is about the same. The leading coefficient, A, is the same and the coefficients, B_1, B_2, and c associated with Vi differ by less than 10%. Some deviation is expected, if only due to bias in each experimenter's technique.

Figures 1 and 2 reveal that although the σ_{rms} are the same, Eq. 3 provides a somewhat better fit to the data. As in Part II, both correlations tend to underestimate the data at large D_{32}/L ($\mu_d \geq 0.4$ Pa·s and $\sigma \geq 0.02$ N/m^2). Maximum differences are about a factor of two for Eq. 3 and a factor of three for Eq. 4. This result is not surprising since both models are based on the assumption that a drop is subjected to constant disruptive energy. Drops with large μ_d and σ have relatively high resistance to breakage and should require more time to reach a critical deformation. If the drop's residence time in the impeller region is less than the critical breakup time, then the maximum stable drop size will be greater than predicted by either Eq. 3 or 4. More sophisticated models are required to account for such transient phenomena. However, these are difficult to develop in the absence of information on what constitutes a critical deformation of a rapidly deforming drop.

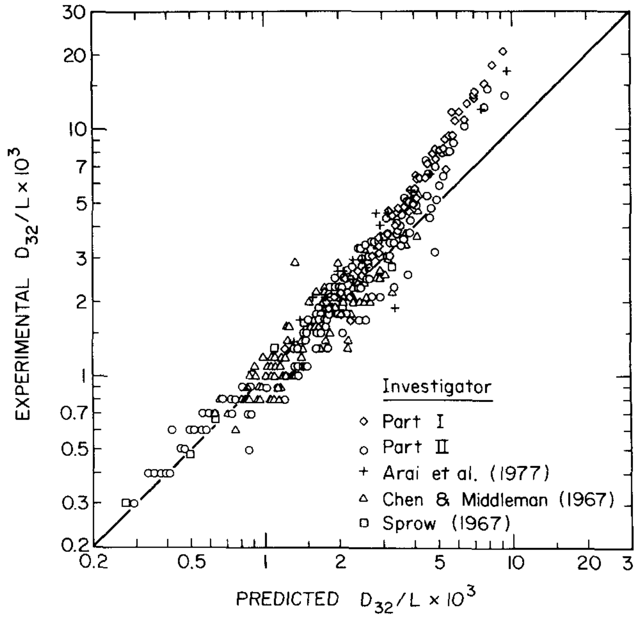

Figure 1. Goodness of fit of Eq. 3 to mean drop size data; $\sigma_{rms} = 23.1\%$.

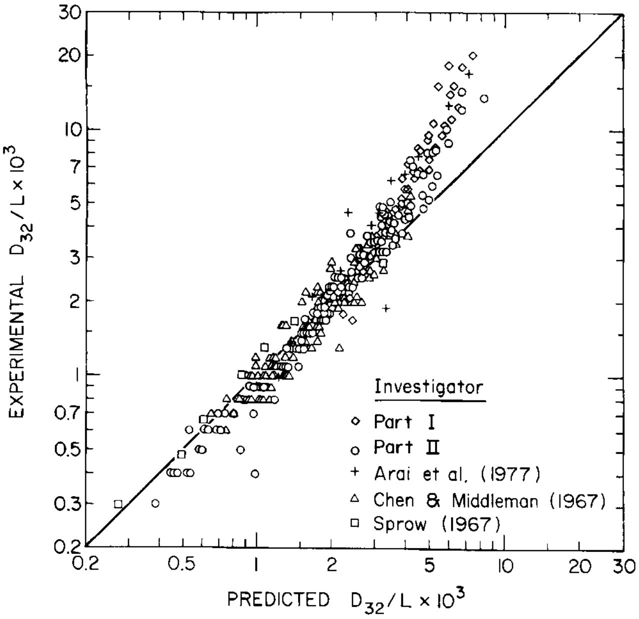

Figure 2. Goodness of fit of Eq. 4 to mean drop size data; $\sigma_{rms} = 23.1\%$.

Correlation for drop size distribution

The results of Part II show that for low to moderate viscosity dispersed phases, drop size distributions are normally distributed in volume and can be correlated by normalization with D_{32}. Furthermore, the correlation developed in Part II is essentially the same as that of Chen and Middleman for low-viscosity drops. Therefore these (Eqs. 23 and 25 of Part II) can be combined to yield

$$F_v\left(\frac{D}{D_{32}}\right) = 0.5\left[1 + \operatorname{erf}\left(\frac{D/D_{32} - 1.07}{0.23\sqrt{2}}\right)\right] \qquad (5)$$

The cumulative volume frequency, F_v, is related to the volume probability density function, $P_v(D/D_{32})$ through Eqs. 18 to 22 of Part II. Therefore Eq. 5 corresponds to

$$P_v\left(\frac{D}{D_{32}}\right) = \frac{1}{0.23\sqrt{2\pi}}\exp\left[-9.1\left(\frac{D}{D_{32}} - 1.07\right)^2\right] \qquad (6)$$

Chen and Middleman provide a method for extracting the distribution of interfacial area from Eq. 6.

Nondilute Dispersions

The most limiting feature of the above correlations is that they apply only in the limit $\phi \rightarrow 0$. The presence of a greater amount of dispersed phase affects equilibrium drop size in two ways. When drops are in close proximity, the small-scale structure of the continuous phase is altered, thereby decreasing the turbulent energy acting to disrupt the drop. Furthermore, coalescence is promoted due to increased probability of drop collisions. If the dispersed-phase volume fraction is not too large, the presence of stabilizing agents can inhibit coalescence. In the absence of such agents coalescence will be primarily confined to more quiescent regions of the tank. A model can be developed for low to moderate ϕ which considers only the effect of ϕ on disruptive energy. The result should apply to drop sizes leaving the impeller region or to the entire tank for noncoalescing (stabilized) dispersions. It is important to note that at large ϕ some coalescence will even occur in the impeller region, particularly for $\rho_d < \rho_c$. It is well-known that air bubbles are captured by the centrifugal fields of the trailing vortex system behind turbine blades to form stable gas cavities (van't Riet and Smith, 1973). At large ϕ, any model that ignores coalescence may not even apply to the impeller region.

Mean drop size correlation

Doulah (1975) argued that the effect of ϕ on the turbulent energy available to disrupt a drop could be quantified by considering its effect on the local energy dissipation rate. He showed that the energy dissipation rate per unit mass for finite ϕ (ϵ_ϕ) is related to that for negligible ϕ (ϵ) by

$$\epsilon_\phi = \epsilon\left(\frac{\rho_e}{\rho_c}\right)^3 (1 + 2.5\,\phi)^{-3} \qquad (7)$$

Equation 7 can be used to replace ϵ by ϵ_ϕ in the equations of Part I (used to derive Eq. 1) to yield the following result:

$$\frac{D_{32}}{L} = A\left(\frac{\rho_e}{\rho_c}\right)^{-6/5} (1 + 2.5\,\phi)^{6/5}\, We^{-3/5} \cdot \left[1 + B_1\left(\frac{\rho_e}{\rho_c}\right)(1 + 2.5\,\phi)^{-1}\, Vi\left(\frac{D_{32}}{L}\right)^{1/3}\right]^{3/5} \qquad (8)$$

The emulsion density, ρ_e, is given by $\rho_e = \rho_c\,(1 + \phi\Delta\rho/\rho_c)$ when $\Delta\rho = \rho_d - \rho_c$. For most liquid-liquid systems $\Delta\rho$ is small. If ϕ is also not too large, then $\rho_e \simeq \rho_c$. Furthermore, the terms containing ϕ can be approximated by a series expansion. If terms of order ϕ^2 and greater are ignored, then Eq. 8 yields

$$\frac{D_{32}}{L} = A(1 + 3\phi) \cdot We^{-3/5}\left[1 + B_1(1 - 2.5\,\phi)\, Vi\left(\frac{D_{32}}{L}\right)^{1/3}\right]^{3/5} \qquad (9)$$

In the limit as $Vi \rightarrow 0$ (negligible viscous resistance to breakage), Eq. 9 reduces to

$$\frac{D_{32}}{L} = A(1 + \gamma\phi)\, We^{-3/5} \qquad (10)$$

where $\gamma = 3$. Several researchers (Brown and Pitt, 1970; Calderbank, 1958; Mlynek and Resnick, 1972) have correlated low-viscosity dispersed-phase data obtained in the geometry considered here by Eq. 10. Their values for the geometric coefficient A (0.051 to 0.060) do not differ substantially from $A = 0.054$ given by Eq. 3 for dilute dispersions. Brown and Pitt measured drop sizes, via photography, at the tip of the impeller and found that data for $\phi < 0.3$ were well-correlated by Eq. 10. Their data yielded $\gamma = 3.14$, in close agreement with the predicted value of $\gamma = 3$. A linear dependency of D_{32} on ϕ was not found at $\phi = 0.4$. Brown and Pitt suggested that the value of $\gamma = 9$ obtained by Calderbank, using a light transmittance technique, was due to measuring drop sizes remote from the impeller region, where coalescence predominates. Mlynek and Resnick found $\gamma = 5.4$ for $\phi \leq 0.2$ by averaging drop sizes from seven different sampling locations. A linear dependency of D_{32} on ϕ was not found at $\phi = 0.34$. These authors reported that drops were smaller near the impeller but that the variation in local drop size was not sufficient to account for the difference between their γ and that of Brown and Pitt. However, Mlynek and Resnick obtained most of their data using a technique by which droplets were encapsulated in a sample withdrawal probe. It is not clear to what extent such a difficult technique will influence measurements.

Given the above results, and a concern that viscous drops may have a greater tendency to coalesce, it appears that Eq. 9 should approximate conditions at the impeller tip and in stabilized dispersions, provided that $\phi \leq 0.2$. The above results also indicate that A is not a function of ϕ. It is therefore reasonable to assume that B_1 is also a purely geometric coefficient and is independent of ϕ. If the values of A and B_1 found by Eq. 3 are substituted into Eq. 9, the result is

$$\frac{D_{32}}{L} = 0.054\,(1 + 3\,\phi)\,We^{-3/5} \cdot \left[1 + 4.42\,(1 - 2.5\,\phi)\,Vi\left(\frac{D_{32}}{L}\right)^{1/3}\right]^{3/5} \qquad (11)$$

Equation 11 should only be applied as a first approximation because the dependency of D_{32} on ϕ has not been experimentally validated for viscous dispersed phases.

Correlation for drop size distribution

Brown and Pitt (1972) measured equilibrium drop size distribution for low-viscosity dispersed phases, produced in the geometry considered here, for the range $0.05 \leq \phi \leq 0.2$. They found that drop size distributions were normally distributed in volume even though they tended toward a slightly bimodal form as ϕ increased. Volume distributions normalized by D_{32} were well-correlated by

$$P_v\left(\frac{D}{D_{32}}\right) = \frac{1}{0.20\sqrt{2\pi}}\exp\left[-12.5\left(\frac{D}{D_{32}} - 1.07\right)^2\right] \qquad (12)$$

Equation 12 is quite similar to Eq. 6, which applies to dilute dispersions. The mean value of D/D_{32} (1.07) is the same. The standard deviation (0.20) is slightly less, indicating that Eq. 12 describes a narrower distribution. Brown and Pitt argued that the system studied was free of coalescence. It is not clear that the small difference between Eqs. 6 and 12 is due to ϕ. Equation 12 is based on 12 experiments for a single liquid-liquid pair obtained in a single tank (at variable N and ϕ), while Eq. 6 is based on numerous data covering a wide range of system parameters. The larger standard deviation in Eq. 6 could be due to scatter in the individual data sets. It appears that in the absence of coalescence, ϕ has little effect on the normalized drop size distribution, at least for inviscid dispersed phases. It is therefore reasonable to assume, to a first approximation, that this is also true for viscous dispersed phases. Then Eq. 5 or 6 can be applied for $\phi \leq 0.2$, with D_{32} given by Eq. 11, to estimate drop size distributions in noncoalescing systems. By analogy to Brown and Pitt, the correlation would also apply at the impeller tip if coalescence is limited to more quiescent regions of the tank.

Summary and Conclusions

Correlations have been developed for the prediction of equilibrium mean drop size and drop size distribution in turbulent, liquid-liquid, stirred-tank contactors. They apply to low to moderate ($\mu_d \leq 0.5$ Pa·s) viscosity dispersed phases produced in baffled cylindrical tanks of standard geometry equipped with six-blade Rushton turbines.

The correlations developed for dilute suspensions are based on numerous data obtained by several investigators for a wide range of system parameters. Sauter mean diameter can be predicted from Eqs. 3 or 4, although Eq. 3 provides a somewhat better fit to data. Drop size distribution is given by Eq. 5 (or 6).

The correlations for nondilute dispersions represent extrapolations of the dilute suspension results based upon mechanistic and/or physical arguments. They apply to noncoalescing systems or at the impeller tip when coalescence is limited to more quiescent regions of the tank. For the range $\phi \leq 0.2$, Sauter mean diameter can be estimated from Eq. 11 and drop size distribution from Eq. 5 (or 6). These correlations are consistent with data obtained for inviscid dispersed phases ($Vi \longrightarrow 0$). They should only be applied to obtain a preliminary estimate for more viscous drops, since they have not been verified experimentally for finite Vi.

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Notation

A = dimensionless empirical constant, Eqs. 1, 2, 8, 9, and 10
B_1 = dimensionless empirical constant, Eqs. 1, 8, and 9
B_2 = dimensionless empirical constant, Eq. 2
c = dimensionless empirical constant, Eq. 2
D = diameter of drop
D_{32} = Sauter mean diameter
F_v = cumulative volume frequency
L = impeller diameter
N = impeller speed
P_v = probability density function for drop volume
Re = $\rho_c N L^2/\mu_c$; tank Reynolds number
T = tank diameter
Vi = $(\rho_c/\rho_d)^{1/2}\,\mu_d NL/\sigma$; tank viscosity group
We = $\rho_c N^2 L^3/\sigma$; tank Weber number

Greek letters

γ = constant, Eq. 10
ϵ = local energy dissipation rate per unit mass for negligible dispersed-phase volume fraction
ϵ_ϕ = local energy dissipation rate per unit mass for dispersed-phase volume fraction ϕ
μ_c = viscosity of continuous phase
μ_d = viscosity of dispersed phase
ρ_c = density of continuous phase
ρ_d = density of dispersed phase
ρ_e = spatially averaged density of the emulsion
σ = interfacial tension
σ_{rms} = root mean square deviation, Eq. 13, Part II
ϕ = volume fraction of dispersed phase

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