SCALE-UP OF LIQUID-LIQUID PHASE MIXING VESSEL

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The scale-up effect of mixing vessels on the drop size of a liquid-liquid dispersion is discussed and correlative equations to estimate drop sizes in breakup and coalescence regions are proposed. It is shown that $(n^3 d^2 D)(4N_p/\pi)$ is applicable as a scale-up standard and that the average energy dissipation rate, the Weber number in the mixing vessel or the agitational Reynolds number is not a satisfying scale-up standard. It is also shown that the effect of volume fraction of dispersed phase on drop size is affected by the scale-up ratio; that is, the scale-up ratio is expected to have a smaller effect on drop size as the fraction of dispersed phase increases.

Introduction

Knowledge of the interfacial area or size distribution of drops is required for prediction of heat and mass transfer phenomena in the liquid-liquid dispersion system in an agitated vessel.

Previous reports by many investigators have shown that system variables such as volume fraction of dispersed phase, physical properties of liquids and mechanical mixing conditions have an effect on the drop size, but quantitative correlations have not been well established. Correlation of these system variables is required to predict drop size or interfacial area of systems in a given condition, to determine the optimal operational condition or to estimate the effect of the scale-up of mixing systems.

The correlative equations of drop size including volume fraction of dispersed phase, viscosity of liquids, impeller speed and impeller-to-vessel diameter ratio in a mixing vessel with a flat-blade turbine impeller have already been proposed by the present authors.¹⁰⁾ The scale-up effect of mixing vessels on drop size is discussed and correlated in this report. It is shown in this report that $(n^3 d^2 D)(4N_p/\pi)$ is useful as a scale-up standard.

1. Background of This Work

Average drop size in an agitated liquid-liquid dispersion can be correlated as

$$d_{p32} = c_1 \bar{\epsilon}^{-2/5} (d/D)^{6/5} f(\phi) f(\mu) (\sigma/\rho)^{3/5} \quad (1)$$

or

$$d_{p32}/d = c_2 We'^{-3/5} N_p^{-2/5} f(\phi) f(\mu) \quad (1')$$

in the breakup-dominant region,¹⁰⁾ where functions $f(\phi)$ and $f(\mu)$ will be given later, and the average energy dissipation rate per unit mass of liquid $\bar{\epsilon}$ and the Weber number in mixing vessel We' are shown as follows.

$$\bar{\epsilon} = (4N_p/\pi) n^3 d^2 \{d^3/(D^2 H)\} \quad (2)$$

$$We' = n^2 d^3 \rho / \sigma \quad (3)$$

$f(\phi)$ and $f(\mu)$ in these equations represent the effect of volume fraction and the effect of liquid viscosity, respectively.

Average drop size in the coalescence region is also shown by the following equations.¹⁰⁾

$$d_{p32} = c'_1 \bar{\epsilon}^{-1/4} (d/D)^{3/4} f'(\phi) f(\mu) (\sigma/\rho)^{3/8} \quad (4)$$

or

$$d_{p32}/d = c'_2 We'^{-3/8} N_p^{-1/4} d^{-3/8} f'(\phi) f(\mu) \quad (5)$$

Then the interfacial area of drops in dispersion, a , is obtained by

$$a = 6\phi/d_{p32} \quad (6)$$

It has been reported by the present authors from turbulent measurements using hot-film anemometry that the distribution pattern of the local energy dissipation rate in a mixing vessel is independent of vessel scale when geometrical similarity is preserved and that the average value throughout the vessel is shown by the following equation to give Eq. (2).

$$\bar{\epsilon} = P/\rho V \quad (7)$$

where P and V are agitation power and liquid volume in the vessel.^{9,11)}

It has been also observed by the present authors, however, that the contribution of middle-scale turbu-

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lent eddies ($0.15 < x/\lambda_f < 2.5$; x : scale of turbulent eddy, λ_f : dissipation scale of turbulence) to the energy spectrum of turbulence increases when scale-up of mixing vessel is done while keeping $\bar{\epsilon}$ constant for geometrically similar vessel, though the contribution of smaller eddies is preserved notwithstanding the scale change. This observation implies that smaller drop sizes are obtained for larger-scale vessels when $\bar{\epsilon}$ is used as the scale-up standard if it is assumed that the turbulent fluctuation of middle-scale eddies, whose scale is from 100μ to a few mm in ordinary cases, controls the breakup phenomena.¹²⁾

Further experiments and discussion in this report are limited to vessels where D and H are equal.

2. Experimental

The mixing systems consisted of a cylindrical flat-bottomed stainless steel tank with a six-bladed stainless or brass Rushton-type turbine impeller under the fully baffled condition. The vessel diameter is 12, 16, 25 or 50 cm and the liquid height is equal to the vessel diameter.

The continuous phase in all experiments was distilled water and the dispersed phase was honeybees' wax whose volumetric fraction was varied from 0.005 to 0.36. The size distribution of drops was evaluated by counting 500 to 2000 solidified wax particles using microscopic pictures on a Burkel-Turk counting plate. The setup of a mixing vessel and the sampling points of emulsion, A-F, are shown in Fig. 1.

Details of the experimental methods are stated in the previous paper.¹⁰⁾

3. Results and Discussion

3.1 Drop sizes in mixing vessels

Figures 2 and 3 show the change of various mean drop sizes with the local power consumption rate per unit mass of liquid in the mixing vessel ϵ , where the values of ϵ at various positions are estimated by consulting the results by the present authors.^{9,11)} These figures show that only a slight change in drop size is observed in spite of the wide spreading of the energy consumption rate for the wax-water emulsion system. These observations imply that breakup or coalescence of beeswax drops is a rather slow phenomenon when compared with the circulation time of the discharged flow as reported in the previous paper from the observation of change of the Sauter mean drop diameter with time after sudden change of impeller speed.¹⁰⁾ Accordingly, it can safely be said that the average drop size at any point in a vessel can be taken as the average value in the whole vessel. Relationships between various average drop sizes are obtained as follows from Figs. 2 and 3.

1) for breakup region

$$d_{p_{\max}} = 2.0 d_{p_{32}} \quad (8)$$

Fig. 1. Experimental apparatus.

Fig. 2. Drop sizes at various points of mixing vessel ($\phi = 0.005$).

$$d_{p_{30}} = 0.67 d_{p_{32}} \quad (9)$$

$$d_{p_{20}} = 0.54 d_{p_{32}} \quad (10)$$

$$d_{p_{10}} = 0.30 d_{p_{32}} \quad (11)$$

2) for coalescence region

$$d_{p_{\max}} = 2.2 d_{p_{32}} \quad (12)$$

$$d_{p_{30}} = 0.60 d_{p_{32}} \quad (13)$$

$$d_{p_{20}} = 0.46 d_{p_{32}} \quad (14)$$

$$d_{p_{10}} = 0.27 d_{p_{32}} \quad (15)$$

where $d_{p_{\max}}$, $d_{p_{30}}$, $d_{p_{20}}$, $d_{p_{10}}$ and $d_{p_{32}}$ are maximum

Fig. 3. Drop sizes at various points of mixing vessel ($\phi = 0.3$).

drop size observed in the sample, volume mean drop size, surface mean drop size, arithmetic mean drop size and Sauter mean drop size, respectively.

The ratio of maximum to Sauter mean drop size shown in Eq. (8) agrees well with values reported by Van Heuven and Hoevenaar¹³⁾ and Carabrese *et al.*²⁾ for breakup-dominant mixing vessels. It is also known from the observations above that a flatter drop size distribution is obtained in the coalescence region.

Change of d_{p32} with $\bar{E}$ for vessels of different size is compared in Figs. 4 and 5. These figures show that smaller drop sizes are obtained for a larger-scale vessel at the same value of $\bar{E}$ as expected from the former discussion and that the coalescence region appears at larger $\bar{E}$ as stated in the previous paper.¹⁰⁾

Impeller speed n , agitational Reynolds number Re or average power consumption rate per unit mass of liquid $\bar{E}$ is often used as the scale-up standard for various phenomena in the mixing vessel. These variables, however, cannot be used as the scale-up standard to keep the interfacial area of emulsion constant, as is shown in Fig. 6, where none of them can give a constant d_{p32} for various vessel diameters.

As shown in Fig. 7, the effect of volume fraction ϕ on d_{p32} is larger for a larger vessel. The same tendency is observed also in the coalescence region.

3.2 Correlation of drop size

Modification of Eqs. (1) and (2) to include the result for change of vessel diameter leads to the following equations in this study.

$$d_{p32} = 0.105 \bar{E}^{-2/5} (d/D)^{6/5} (D/D_0)^{-2/5} \{1 + 2.5(D/D_0)^{1/2} \phi^{2/3}\} (\mu_d/\mu_c)^{1/5} (\mu_d/\mu_c)^{1/8} (\sigma/\rho)^{3/5} \quad (\text{breakup region}) \quad (16)$$

$(\mu_d/\mu_c)^{1/5}$ is recommended to use in the initial estimation of drop size.

Fig. 4. d_{p32} and $\bar{E}$ ($D = 12$ cm).

Fig. 5. d_{p32} and $\bar{E}$ ($D = 50$ cm).

$$d_{p32} = 0.0371 \bar{E}^{-1/4} (d/D)^{3/4} (D/D_0)^{-1/4} \{1 + 3.5(D/D_0)^{1/2} \phi^{3/4}\} (\mu_d/\mu_c)^{1/5} (\mu_d/\mu_c)^{1/8} (\sigma/\rho)^{3/8} \quad (\text{coalescence region}) \quad (17)$$

where D_0 is taken as 25 cm in this study.

Using the Weber number in mixing vessel, Eqs. (16) and (17) are rearranged as follows.

$$d_{p32}/d = 0.095 N_p^{-2/5} (D/D_0)^{-2/5} \{1 + 2.5(D/D_0)^{1/2} \phi^{2/3}\} (\mu_d/\mu_c)^{1/5} (\mu_d/\mu_c)^{1/8} We'^{-3/5} \quad (\text{breakup region}) \quad (18)$$

$$d_{p32}/d = 0.035 N_p^{-1/4} d^{-3/8} (D/D_0)^{-1/4} \{1 + 3.5(D/D_0)^{1/2} \phi^{3/4}\} (\mu_d/\mu_c)^{1/5} (\mu_d/\mu_c)^{1/8} We'^{-3/8} \quad (\text{coalescence region}) \quad (19)$$

The correlative equation for the breakup region (Eq. (18)) is dimensionless, though that for the coalescence region is not because of the term $d^{-3/8}$ on the right-hand side of Eq. (19). This is because the specific energy adhesion between two spheres of unit diameter is not evaluated in this study.

The value of $\bar{E}_{cr}$ or We'_{cr} which shows the transitional point of dominant process of drop formation

Fig. 6. d_{p32} and D using impeller speed, agitational Re number or average energy dissipation rate per unit mass as parameter.

Fig. 7. Effect of ϕ on d_{p32} .

Fig. 8. Comparison of correlative equations with experimental values.

is obtained by equating Eqs. (16) and (17) or (18) and (19).

$$\bar{\epsilon}_{cr} = (98.7\sigma/\rho)^{3/2} (d/D)^3 (D/D_0)^{-1} \left[\frac{1 + 2.5(D/D_0)^{1/2} \phi^{2/3}}{1 + 3.5(D/D_0)^{1/2} \phi^{3/4}} \right]^{20/3} \quad (20)$$

or

$$We'_{cr} = 84.0 N_p^{-2/3} (D/D_0)^{-2/3} d^{5/3} \left[\frac{1 + 2.5(D/D_0)^{1/2} \phi^{2/3}}{1 + 3.5(D/D_0)^{1/2} \phi^{3/4}} \right]^{40/9} \quad (21)$$

The estimated values using Eqs. (16) and (17) are shown in Fig. 7 and Fig. 8 by solid lines and show good agreement with experimental values.

3.3 Scale-up consideration

Exponents of impeller speed, impeller diameter and vessel diameter reported by various reserchers in correlation of drop size in the mixing vessel are compared in Table 1, and rather wide spreading, especially for the exponent of vessel diameter, can be seen in this table.

Table 1. Comparison of exponents by various authors

$d_p \propto n^\alpha d^\beta D^\gamma$	α	β	γ
This work (breakup region)	-1.2	-0.8	-0.4
This work (coalescence region)	-0.75	-0.5	-0.25
Fernandes ⁶⁾ ($D < 40$ cm)	-1	-1	-0.5
Fernandes ⁶⁾ ($D > 40$ cm)	-1	-1	0
Brown and Pitt ¹⁾	-1.2	-1	-0.2
Eckert ⁵⁾	-1.11	-1.23	0.18
Vermulen ¹⁴⁾ , Calderbank ²⁾	-1.2	-0.8	0
Mlynek ⁸⁾ , Coualoglou ⁴⁾ and Carabrese ³⁾			

Comparison of exponents of vessel diameter in the d_{p32} correlation as Eqs. (22) and (23) are shown in Tables 2 and 3.

$$d_{p32} \propto \bar{\epsilon}' (d/D)^{\beta'} D^{\gamma'} \quad (22)$$

or the correlation using the Weber number in the mixing vessel as

$$d_{p32} \propto We'^{\alpha'} (d/D)^{\beta''} D^{\gamma''} \quad (23)$$

Table 2. Comparison of exponents by various authors

$d_p \propto \bar{\epsilon}^{\alpha'} (d/D)^{\beta'} D^{\gamma'}$			
	α'	β'	γ'
This work (breakup region)	-0.4	1.2	-0.4
This work (coalescence region)	-0.25	0.75	-0.25
Fernandes ($D < 40$ cm)	-0.33	0.67	-0.83
Fernandes ($D > 40$ cm)	-0.33	0.67	-0.33
Brown and Pitt	-0.4	1.0	-0.4
Eckert	-0.37	0.62	-0.2
Vermulen, Calderbank	-0.4	1.2	0
Mlynec, Coualoglou and Carabrese			

Table 3. Comparison of exponents by various authors

$d_p/d \propto We^{\alpha''} (d/D)^{\beta''} D^{\gamma''}$			
	α''	β''	γ''
This work (breakup region)	-0.6	0	-0.4
This work (coalescence region)	-0.375	-0.375	-0.625
Fernandes ($D < 40$ cm)	-1.5	-0.5	-1
Fernandes ($D > 40$ cm)	-0.5	-0.5	-0.5
Brown and Pitt	-0.6	-0.2	-0.4
Eckert	-0.555	-0.565	-0.385
Vermulen, Calderbank	-0.6	0	0
Mlynec, Coualoglou and Carabrese			

The extent of spreading among reported values is apparently more or less decreased when exponents are compared after correlation.

The scale-up effects reported by various authors are compared in **Fig. 9**, assuming that the correlative equations reported by various authors can be applied to larger vessels, where the average power consumption rate per unit mass of liquid is used as the scale-up standard, and disagreement can be seen from this figure. Equation (16) or (18) of this study indicates that the exponent of the (D/D_0) term varies with the volumetric fraction of the dispersed phase if the correlative equation is made without introduction of the scale-up effect into the volumetric fraction-related term. The smaller effect of vessel diameter on the larger volumetric fraction is expected as can be seen from solid lines in **Fig. 9**, where $d_{p32} \propto D^{0.4}$ for the dilute dispersion and $d_{p32} \propto D^{0.1}$ for $\phi = 0.3$ are observed.

It is reported by Konno *et al.*⁷⁾ that the changing pattern of drop size distribution spectra in the mixing vessel after a sudden change in impeller speed varies with vessel scale. Further examination of local turbulent energy spectra, local breakup and coalescence frequency, and local residence time of emulsion is required to provide a theoretical explanation of the scale-up effect on drop size.

Equations (16)–(19) imply that $(n^3 d^2 D) (4N_p/\pi)$ can

Fig. 9. Comparison of scale-up effect by various authors.

be used as the scale-up standard of the mixing vessel when the volumetric fraction is kept constant. Good correlation of d_{p32} in the breakup and coalescence region shown in **Fig. 10** under various conditions shows that $(n^3 d^2 D) (4N_p/\pi)$ is applicable as the scale-up standard. This variable coincides with $\bar{\epsilon}$ when the vessel scale is fixed. It is reported by Eckert *et al.*⁵⁾ and Brown and Pitt¹⁾ that the impeller tip speed nd can have the controlling effect in drop formation. The result in this study, however, shows that the impeller-to-vessel diameter ratio should also be taken into account when the tip speed is used as the scale-up standard.

The correlative equations including the scale-up terms in this work, obtained by summarizing data for 12–50 cm vessels, can predict the Sauter mean drop diameter within about 30% error as can be seen from **Fig. 10**.

It has been pointed out by the present authors that the dominant process in formation of drops in the mixing vessel changes from breakup to coalescence phenomena at larger energy dissipation rate per unit mass of liquid or at larger volumetric fraction of dispersed phase.¹⁰⁾ Correlation of drop sizes without classification of dominant processes can give erroneous results in slope because correlative lines with various slopes can be obtained if one curve is drawn through two regions.

4. Conclusions

The scale-up effect on drop size in the mixing vessel was studied, and correlative equations to obtain drop sizes in the breakup and the coalescence phenomena-dominant regions are proposed.

As for the scale-up standard, use of $(n^3 d^2 D) (4N_p/\pi)$ is recommended in this study. This variable coincides with the average energy dissipation rate per unit mass of liquid in the mixing vessel when the vessel scale is fixed. When the impeller-to-vessel diameter ratio is fixed, it is similar to the impeller tip speed.

Fig. 10. Correlation using $(n^3 d^2 D)(4N_p/\pi)$ in breakup and coalescence regions.

The effect of the volumetric fraction of the dispersed phase on drop size is found to vary with vessel scale, or the effect of scale-up ratio on drop size is largely affected by the volumetric fraction.

It was also found in this study that the estimate of the effect of vessel diameter on drop size from the turbulent energy spectra can show fair agreement with experimental values.

Nomenclature

a	= interfacial area	$[cm^2/cm^3]$
D	= vessel diameter	$[cm]$
D_0	= reference vessel diameter	$[cm]$
d	= impeller diameter	$[cm]$
d_p	= drop size	$[cm]$
d_{p10}	= arithmetic mean drop size	$[cm]$
d_{p20}	= surface mean drop size	$[cm]$
d_{p30}	= volume mean drop size	$[cm]$
d_{p32}	= Sauter mean drop size	$[cm]$
$N_p (= P/\rho n^3 d^5)$	= power number	$[-]$
n	= impeller speed	$[s^{-1}]$
P	= agitational power	$[g \cdot cm^2/s^3]$
$Re (= d^2 n \rho / \mu)$	= agitational Reynolds number	$[-]$
$\bar{u}_{dp}$	= time mean value of fluctuating velocity differences over a distance d_p	$[cm/s]$
V	= volume of mixing liquid	$[cm^3]$
$We' (= n^2 d^3 \rho / \sigma)$	= Weber number in mixing vessel	$[-]$
x	= scale of turbulent eddy	$[cm]$
ε	= energy dissipation rate per unit mass of liquid	$[cm^2/s^3]$
$\bar{\varepsilon}$	= average value of ε throughout the vessel	$[cm^2/s^3]$
$\bar{\varepsilon}_{cr}$	= critical $\bar{\varepsilon}$ to show transition point of dominant process in drop formation	$[cm^2/s^3]$
λ_f	= dissipation scale of turbulence	$[cm]$

μ	= liquid viscosity	$[g/cm \cdot s]$
μ_c	= continuous-phase liquid viscosity	$[g/cm \cdot s]$
μ_d	= dispersed-phase liquid viscosity	$[g/cm \cdot s]$
$(\mu_d/\mu_c)_d$	= viscosity ratio keeping μ_d constant	$[-]$
$(\mu_d/\mu_c)_c$	= viscosity ratio keeping μ_c constant	$[-]$
ρ	= specific weight	$[g/cm^3]$
σ	= interfacial tension	$[g/s^2]$
ϕ	= volumetric fraction of dispersed phase	$[-]$

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