AVERAGE DROP SIZE IN A LIQUID-LIQUID PHASE MIXING VESSEL

MASABUMI NISHIKAWA, FUYUHIKO MORI AND SHIGEO FUJIEDA

Department of Nuclear Engineering, Kyushu University, Fukuoka 812

When two immiscible liquids are agitated, a dispersion is formed in which continuous breakup and coalescence of drops occur, and a dynamic equilibrium is attained between breakup and coalescence after a certain time. Effects of the volume fraction of dispersed phase, viscosity of liquid, impeller speed and impeller-to-vessel diameter ratio on the average drop size of a dispersion in a mixing vessel are discussed and correlative equations are proposed. It is also found that the dominant process in deciding average drop sizes in a mixing vessel changes from breakup to coalescence when the average energy dissipation rate or the volumetric fraction of dispersed phase is increased.

The mixing of immiscible liquid is among the most important chemical or biological engineering operations. In nuclear fuel reprocessing, the performance of mixer-settlers should be analyzed from drop size distribution for optimizing uranium and plutonium extraction.

A number of investigators have presented correlative equations that related drop sizes in the mixing vessel to mixing parameters and physical properties of the system. When two immiscible liquids are agitated, a dispersion is formed in which breakup and coalescence of drops occur. After a certain time, a dynamic equilibrium is attained between breakup and coalescence in the mixing vessel. Drops are believed to be broken up by turbulent fluctuations of the ambient liquid in the neighbourhood of the drops. In a dilute dispersion, coalescence may be negligible and the equilibrium spectrum of drop size distribution will depend on the breakup process. The coalescence process may be dominant in a dense dispersion because the probability of collision of drops increases.

Effects of the volume fraction of dispersed phase, viscosity of liquid, impeller speed and impeller diameter on the drop sizes in a mixing vessel are discussed in this report and it is found that the coalescence process becomes dominating at higher energy dissipation rate or at larger volume fraction of dispersed phase. Drop sizes estimated by use of the correlative equations obtained in this study are compared with reported values by various authors.

1. Theory of Drop Formation

Theories of the mechanism of drop breakup process in turbulent flow fields have been proposed by various authors. In the usual case, the drop diameter is much larger than the scale of the energy dissipation eddies, which is approximated by the Kolmogoroff length as

$$\eta = (v^3/\varepsilon)^{1/4} \tag{1}$$

The Kolomogoroff length is considered to describe the local state of turbulence in a region of liquid small enough to be independent of the large-scale flow patterns.

According to Taylor,[17] Hinze[8] and Shinnar and Churcn,[15] the largest drop size d_p^* under non-coalescence conditions is determined by the ratio of the kinetic energy of an oscillating drop due to turbulent fluctuation E_v and the energy due to the interfacial tension E_s.

$$E_v = \rho u_{d_p}^2 d_p^3 \tag{2}$$

$$E_s = \sigma d_p^2 \tag{3}$$

The critical Weber number for breakup We_b is given by

$$We_b(=E_v/E_s)_b = \rho u_{d_p}^2 d_p^*/\sigma \tag{4}$$

where u_{d_p} is the time mean value of the velocity differences over a distance equal to d_p^*. In the region where Kolmogoroff's law holds, the turbulent pattern is dependent on the local rate of energy dissipation per unit mass of liquid ε.[8,15]

$$u_{d_p}^2 = c_1(\varepsilon d_p)^{2/3} \tag{5}$$

Then

$$d_p^* = (We_b/c_1)^{3/5}(\sigma/\rho)^{3/5}\varepsilon^{-2/5} \tag{6}$$

If a drop pair is exposed to turbulent fluctuation

and the kinetic energy of the oscillations induced in the coalescing drop pair is larger than the energy adhesion between them E_{ad}, contact will be broken before coalescence is completed.

$$E_{ad} = d_p A(h) \qquad (7)$$

where $A(h)$ is the specific energy adhesion between two spheres of unit diameter separated by a distance h as shown by Shinnar and Church.[15]

The smallest drop size for which coalescence will be slowed up by turbulence is obtained by

$$(E_v/E_{ad})_c = c_2$$
$$= We_c(d_p^+ \cdot \sigma/A(h))$$
or
$$(E_v/E_s)_b/(E_v/E_{ad})_c = We_b/(We_c \cdot d_p^+ \sigma/A(h)) \qquad (8')$$

Eqs. (5) and (8) give

$$d_p^+ = (c_2/c_1)^{3/8} \rho^{-3/8} A(h)^{3/8} \varepsilon^{-1/4} \qquad (9)$$

In the mixing vessel, the average value of the energy consumption rate per unit mass of liquid $\bar{\varepsilon}$ is applied to relate the Sauter mean drop size d_{p32}.

$$\bar{\varepsilon} = (4N_p/\pi) n^3 d^2 (d/D)^3 \qquad (10)$$

where N_p is the power number and equal values are given for vessel diameter and liquid height in this study.

The Weber number is then shown by

$$We = c_1 \rho \{ (4N_p/\pi) n^3 d^2 (d/D)^3 \}^{2/3} d_{p32}^{5/3}/\sigma \qquad (11)$$

The mixing vessel Weber number We', shown as

$$We' = n^2 d^3 \rho/\sigma \qquad (12)$$

is often used in correlation of drop sizes in the mixing vessel. Then,

$$We'/We = (1/c_1)(d/D)^{-2}(d_p/d)^{-5/3}/(4N_p/\pi)^{2/3} \qquad (13)$$

2. Experimental

The test apparatus consists of a cylindrical flat-bottomed stainless steel tank fitted with a six-bladed stainless steel or brass Rushton-type turbine impeller under the fully baffled condition. The setup of the mixing vessel is shown in **Fig. 1**. A stepless variation of impeller speed from 80 to 360 rpm is possible. The liquid temperature in the mixing vessel is automatically controlled at $80 \pm 0.5°C$. The continuous phase in this experiment is distilled water or millet jelly solution and the dispersed phase is honey bees' wax. The volumetric fraction of the wax is varied from 0.0045 to 0.36. The properties of the wax are given in **Table 1**.

Sampling of the emulsion is carried out by siphoning about 10 ml of emulsion into 200 ml of ice water in a Teflon container. Almost the same procedure was carried out by Church and Shinnar.[5] The size distri-
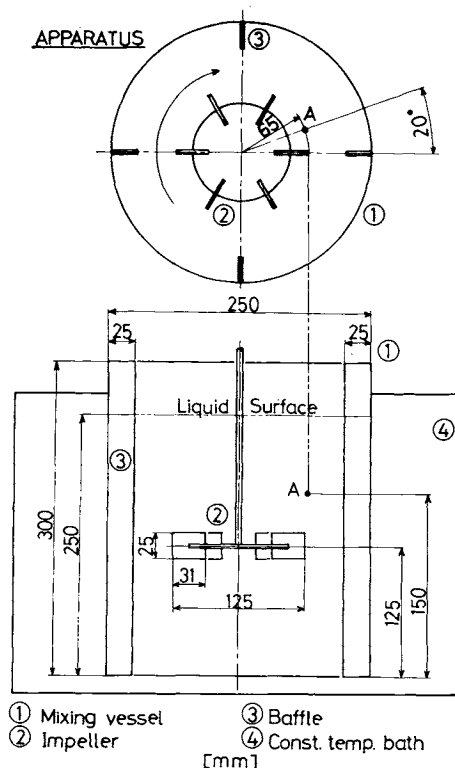

Fig. 1. Experimental apparatus.

Table 1. Physical properties of experimental liquids

	Water	Beeswax
Melting temperature [°C]	0	65
Specific weight at 80°C [g/cm³]	0.972	0.81
Viscosity at 80°C [g/cm·s]	0.00356	0.170
Interfacial tension at 80°C [g/s²]	—	17.9

bution of drops is determined by counting 500 to 2000 solidified wax particles using microscopic pictures on a Burkel-Turk counting plate.

The agitation power is measured by the torque meter developed by Nagata and Yokoyama and the error in the power measurement is within 10% as stated in detail in their report.[12]

3. Results and Discussion

3.1 Drop size and power consumption

As shown in **Fig. 2**, in which the Sauter mean drop diameter is plotted against time after sudden change of impeller speed, d_{p32} decreases rapidly with time when the impeller speed is increased, though d_{p32} increases rather slowly when speed is decreased. In view of this time dependency of drop size variation, drop size measurement was carried out 90–120 minutes after the start of agitation in this study.

It has been reported by the present authors that the average dissipation rate in a mixing vessel $\bar{\varepsilon}$ is approximated by the following equation from hot-film anemometry:
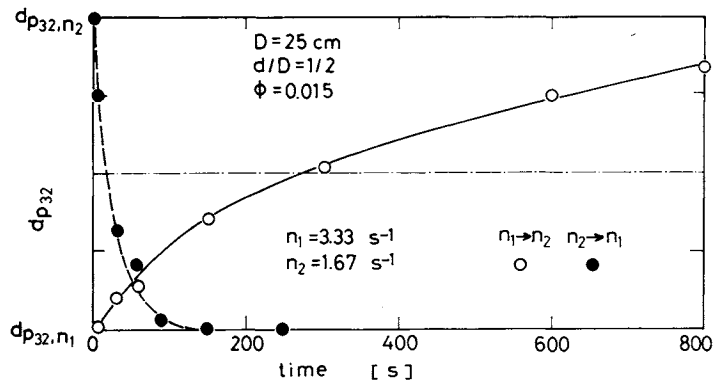
Fig. 2. Change of drop size with time after change of impeller speed.

$$\bar{\varepsilon} = P/\rho V \qquad (14)$$

though about ten times larger dissipation rate than the averaged value is observed in the vicinity of the impeller tip.[13,14]

As the average circulation time of mixing liquid t_c is about 6 s at most in the range of this experiment when the correlation by Nagata et al.[11] is applied, it is assumed in this experiment that the average value of drop sizes at a certain point A shown in Fig. 1 represents the average value in the vessel, considering the longer time requirement for drop size change as shown by Fig. 2.

Figures 3–6 show change of drop sizes with the power consumption rate per unit mass of mixing liquid, i.e. $\bar{\varepsilon}$, where the sampling is performed at point A.

It can be seen from these figures that there are two regions, i.e. a breakup-dominating region at lower $\bar{\varepsilon}$ and a coalescence-dominating region at higher $\bar{\varepsilon}$, because the slope changes from $-2/5$ to $-1/4$. Dashed lines in these figures show the critical $\bar{\varepsilon}$ value at which the dominating process changes, and effects of the d/D ratio are shown in **Fig. 7** where values for d_{p32} used in this figure are estimated by the interpolation method after curve-fitting of data.

In the breakup-dominating region, the exponent for $\bar{\varepsilon}$ coincides with values expected from Eqs. (6) and (10).

$$d_{p32} \propto \bar{\varepsilon}^{-2/5}(d/D)^{6/5} \qquad (15)$$

The correlative equations as $d_{p32} \propto We'^{-0.6}$ have been reported by various authors for breakup process-dominating mixing vessels with several d/D ratios.[3,19] These results give the value of 6/5 for the exponent for d/D from relations shown by Eqs. (6) and (13). In the higher $\bar{\varepsilon}$ region, the exponent for $\bar{\varepsilon}$ agrees with the value predicted by Eq. (9) for the coalescence-dominating liquid dispersion.

$$d_{p32} \propto \bar{\varepsilon}^{-1/4}(d/D)^{3/4} \qquad (16)$$
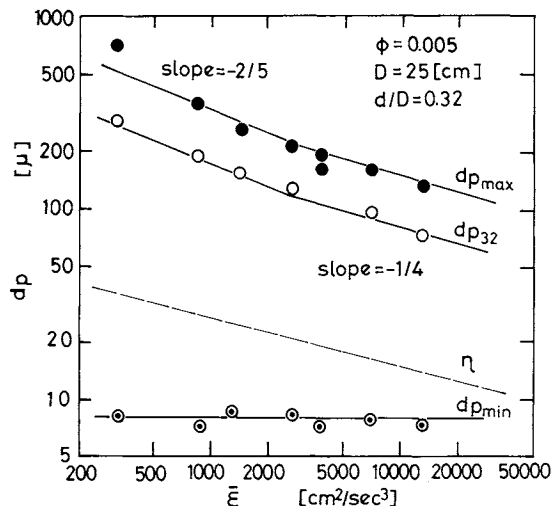
It can be also seen from Fig. 3 that the maximum

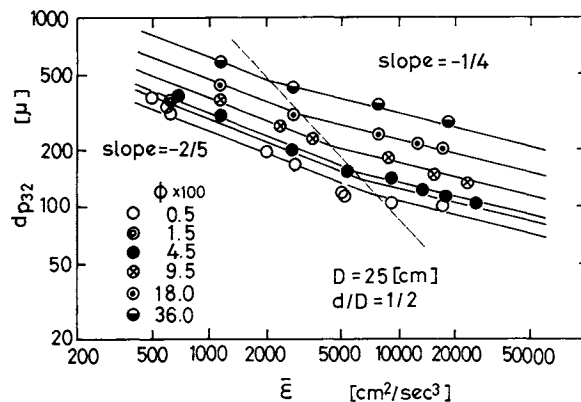**Fig. 3.** $d_{p\max}$, d_{p32} and $d_{p\min}$ and average energy dissipation rate.

Fig. 4. d_{p32} and $\bar{\varepsilon}$ (volumetric fraction as parameter).

drop size observed in the emulsion sample $d_{p\max}$ is about twice the Sauter mean drop size though the minimum drop size $d_{p\min}$ observed has an almost constant value of about 8 microns.

$$d_{p32}/d_{p\max} = 0.5 \qquad \text{(breakup region)} \qquad (17)$$

$$d_{p32}/d_{p\max} = 0.45 \qquad \text{(coalescence region)} \qquad (18)$$

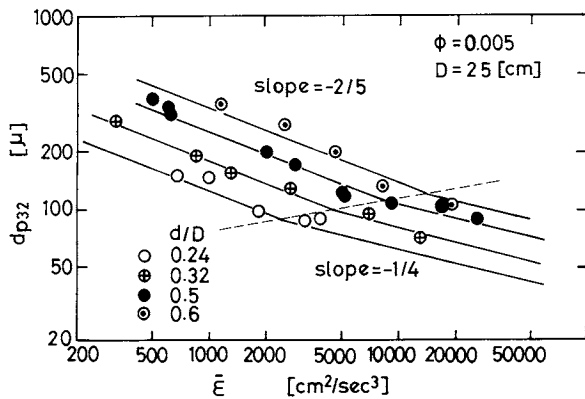The value shown by Eq. (17) in this study agrees

Fig. 5. d_{p32} and $\bar{\varepsilon}$ (impeller-to-vessel diameter ratio as parameter).
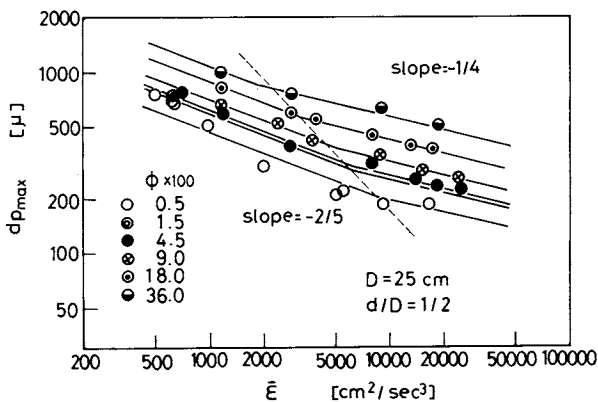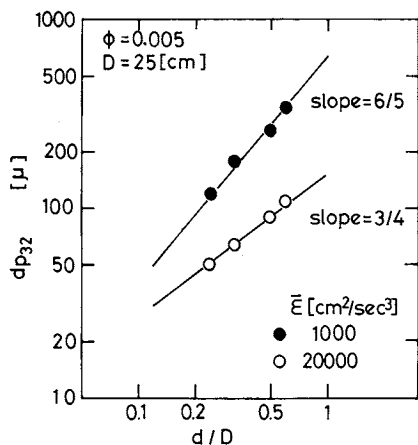
Fig. 6. $d_{p\max}$ and $\bar{\varepsilon}$.

Fig. 7. Effect of impeller-to-vessel diameter ratio in break-up and coalescence regions.

Fig. 8. Effect of volumetric fraction of dispersed phase (breakup region).

$$\frac{dp_{32}}{(dp_{32})_{\phi=0.1}} = (1 + 2.5\,\phi^{2/3})/1.54$$
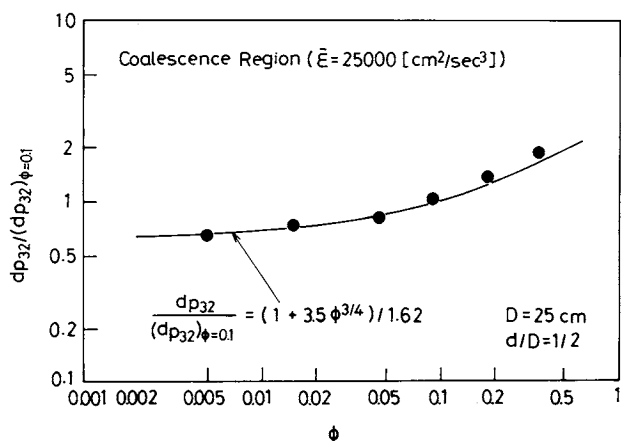
Fig. 9. Effect of volumetric fraction of dispersed phase (coalescence region).

$$\frac{dp_{32}}{(dp_{32})_{\phi=0.1}} = (1 + 3.5\,\phi^{3/4})/1.62$$

well with the value reported by Van Heuven and Hoevenaarj[18] and Carabrese et al.[3] for breakup-dominating dispersions.

3.2 Drop size and volume fraction

Figures 4, 8 and **9** show the effect of the volumetric fraction of wax ϕ on the Sauter mean drop size, and following equations are obtained.

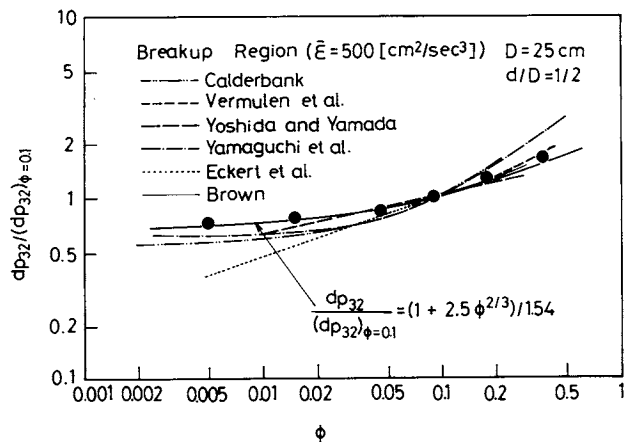$$d_{p32} \propto 1 + 2.5\phi^{2/3} \qquad \text{(breakup region)} \qquad (19)$$

$$d_{p32} \propto 1 + 3.5\phi^{3/4} \qquad \text{(coalescence region)} \qquad (20)$$

The effect of ϕ on average drop size reported by various authors is compared with that obtained in this study in Fig. 8 for the breakup region where values for d_{p32} are estimated by the interpolation method after curve-fitting of data. Results by Vermulen et al.,[19] Brown[1] or Yamaguchi et al.[20] show a similar tendency to that obtained in this study. The equation reported by Eckert et al.[7] gives somewhat smaller values than those by other investigators at lower volumetric fraction.

The effect of the volumetric fraction on drop size in the coalescence region is a little larger than that in the breakup region, as can be seen from comparison of Fig. 8 with Fig. 9 or from the equations above.

It can be also seen from Fig. 4 that $\bar{\varepsilon}_{cr}$ decreases with increasing ϕ to give the coalescence-dominant condition at lower $\bar{\varepsilon}$. This observation implies that correlation of drop size using only one dominant process for a wide operational range can give errors.

3.3 Drop size and liquid viscosity

The effect of the viscosity of continuous phase on

drop size is correlated by the following equation for both breakup and coalescence regions where the viscosity of dispersed phase is fixed as shown in **Fig. 10**.

$$d_{p32} \propto (\mu_d/\mu_c)_d^{1/5} \qquad (21)$$

In case the viscosity of dispersed phase is changed, while fixing the viscosity of continuous phase, a rather different exponent is obtained using data by Yamaguchi et al.[20]

$$d_{p32} \propto (\mu_d/\mu_c)_c^{1/8} \qquad (22)$$

3.4 Correlation of drop size

Summarizing the results stated above, the following equations are obtained in this study.

$$d_{p32} = 0.105\bar{\varepsilon}^{-2/5}(d/D)^{6/5}(1+2.5\phi^{2/3})(\mu_d/\mu_c)_d^{1/5}$$

$$(\mu_d/\mu_c)_c^{1/8}(\sigma/\rho)^{3/5} \quad \text{(breakup region)} \qquad (23)$$

$$d_{p32} = 0.0371\bar{\varepsilon}^{-1/4}(d/D)^{3/4}(1+3.5\phi^{3/4})(\mu_d/\mu_c)_d^{1/5}$$

$$(\mu_d/\mu_c)_c^{1/8}(\sigma/\rho)^{3/8} \quad \text{(coalescence region)} \qquad (24)$$

From these equations, the Weber number at drop breakup We_b and that at coalescence We_c can be obtained as follows, using Eqs. (23) and (6) or Eqs. (24), (8) and (9).

$$(We_b/c_1) = 0.0232(d/D)^2(1+2.5\phi^{2/3})$$

$$(\mu_d/\mu_c)_d^{1/3}(\mu_d/\mu_c)_c^{5/24} \qquad (25)$$

$$(We_c/c_1)(d_{p32})_c = 0.000151(d/D)^2(1+3.5\phi^{3/4})$$

$$(\mu_d/\mu_c)_d^{8/15}(\mu_d/\mu_c)_c^{1/3} \qquad (26)$$

These equations show that $(E_v/E_s)_b/(E_v/E_{ad})_c$ is independent of the energy dissipation rate or the configurations of the mixing systems, as expected from Eq. (8′).

d_{p32} can be correlated by the following equations when the mixing vessel Weber number is introduced.

$$d_{p32}/d = 0.095N_p^{-2/5}(1+2.5\phi^{2/3})(\mu_d/\mu_c)_d^{1/5}$$

$$(\mu_d/\mu_c)_c^{1/8}We'^{-3/5}$$

$$\text{(breakup region)} \qquad (27)$$

$$d_{p32}/d = 0.035N_p^{-1/4}(1+3.5\phi^{3/4})(\mu_d/\mu_c)_d^{1/5}$$

$$(\mu_d/\mu_c)_c^{1/8}We'^{-3/8}d^{-3/8}$$

$$\text{(coalescence region)} \qquad (28)$$

The correlative equation for the coalescence region is not dimensionless, though that for the breakup region is dimensionless. This is because $A(h)$, which can give a dimensionless group with d_p and σ as shown in Eq. (8′), is not evaluated in this study.

As stated by McManamy,[9] introduction of the power number term is essential to form the drop size correlation using the mixing vessel Weber number.

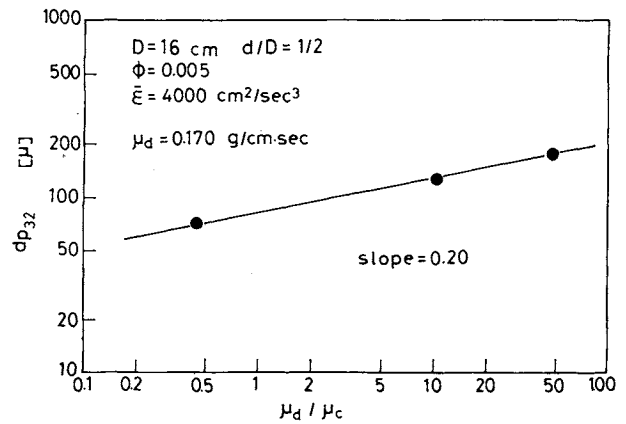The value of $\bar{\varepsilon}_{cr}$ or We'_{cr} to give the transitional

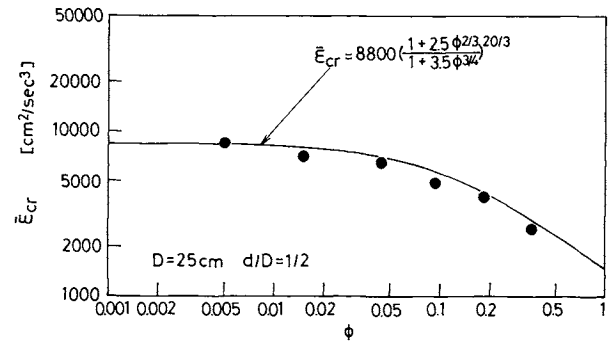**Fig. 10.** Effect of viscosity of continuous phase.

Fig. 11. $\bar{\varepsilon}_{cr}$ and volumetric fraction of dispersed phase.

points of the dominant process can be obtained by equating Eq. (23) and (24) or (27) and (28).

$$\bar{\varepsilon}_{cr} = (98.7\sigma/\rho)^{3/2}(d/D)^3\{(1+2.5\phi^{2/3})/$$

$$(1+3.5\phi^{3/4})\}^{20/3} \qquad (29)$$

$$We'_{cr} = 84.0N_p^{-2/3}d^{5/3}\{(1+2.5\phi^{2/3})/$$

$$(1+3.5\phi^{3/4})\}^{40/9} \qquad (30)$$

Estimated values using Eq. (29) show good agreement with the experimental values as shown by solid lines in **Fig. 11**.

The drop size predicted for the breakup region using correlative equations in this study is compared with values reported by various authors in **Fig. 12**. As can be seen from this figure, the drop sizes estimated using the correlative equations in this study agree fairly well with values estimated using correlations by various authors for turbine or paddle-type impellers, though the estimation by Calderbank for turbine[2] and those by Coualaloglou and Tavlarides for turbine in a continuous flow vessel[6] give rather high values than others. Drop sizes estimated by Calderbank's equation seem to give overly large values, as pointed out by several authors.[1,3] In a continuous-flow mixing vessel where the volumetric fraction of dispersed phase is 0.025–0.15 and the average residence time is 600 minutes as in the work of Coualaloglou and Tavlarides, larger drop sizes than those obtained at
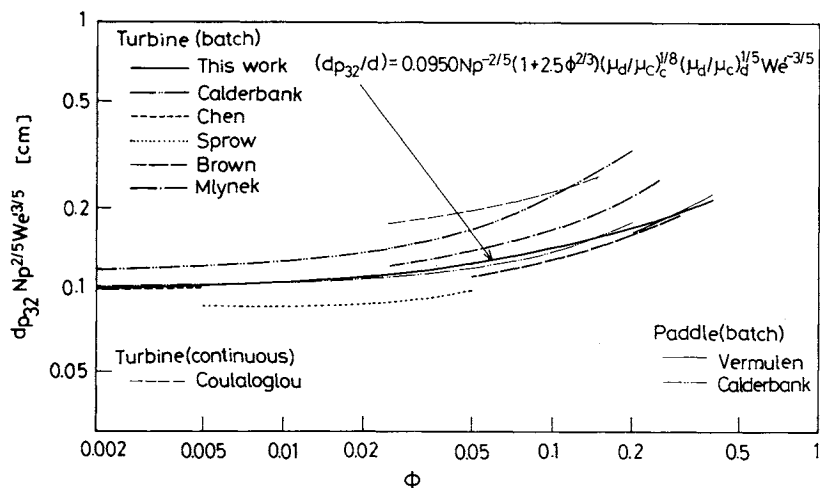
$$(d_{p32}/d) = 0.0950 N_p^{-2/5}(1 + 2.5\phi^{2/3})(\mu_d/\mu_c)_c^{1/8}(\mu_d/\mu_c)_d^{1/5} We'^{-3/5}$$

Turbine (batch)
——— This work
—·—— Calderbank
------- Chen
············ Sprow
——— Brown
—·—— Mlynek

Turbine(continuous)
———— Coulaloglou

Paddle(batch)
——— Vermulen
—·— Calderbank

Fig. 12. Comparison of drop sizes by various investigators.

equilibrium condition can be expected, consulting the results shown in Fig. 2 when the drop sizes in the inlet flow are larger than the drop sizes in the mixing vessel.

The way to correlate average drop size with average dissipation rate is applied in this study because the time required for drop size change is much larger than the circulation time of mixing liquid in a vessel. However, when a dispersion system is encountered such as that where the time required for drop size change is smaller than the circulation time, the simple averaging method cannot explain the relations because the drop size distribution in a vessel becomes remarkable, corresponding to the distribution of energy dissipation rate.

4. Conclusions

The effects of average energy dissipation rate, impeller-to-vessel diameter ratio, volumetric fraction of dispersed phase and viscosity of continuous or dispersed phase on drop sizes in a mixing vessel were studied and correlative equations are proposed. It is also found in this study that the dominant process in deciding drop sizes changes from breakup to coalescence when the average energy dissipation rate is increased. The critical value to change the dominant process decreases when the volumetric fraction of dispersed phase is increased.

Nomenclature

$A(h)$	= specific energy adhesion between two spheres of unit diameter separated by distance h	$[g \cdot cm/s^2]$
D	= vessel diameter	[cm]
d	= impeller diameter	[cm]
d_p	= drop size	[cm]
d_p^*	= largest drop size under non-coalescence condition	[cm]
d_p^+	= smallest drop size under non-breakup condition	[cm]
d_{p32}	= Sauter mean drop size	[cm]
d_{pmax}	= maximum drop size in emulsion	[cm]
d_{pmin}	= minimum drop size in emulsion	[cm]
E_{ad}	= energy adhesion	$[g \cdot cm^2/s^2]$
E_s	= energy due to interfacial tension	$[g \cdot cm^2/s^2]$
E_v	= kinetic energy of oscillating drop	$[g \cdot cm^2/s^2]$
$N_p(= P/\rho n^3 d^5) =$	power number	[—]
n	= impeller speed	$[s^{-1}]$
P	= agitating power	[W]
u_{d_p}	= time mean value of fluctuating velocity differences over a distance d_p	[cm/s]
V	= volume of mixing liquid	$[cm^3]$
$We(= E_v/E_s) =$	Weber number	[—]
$We'(= n^2 d^3 \rho/\sigma) =$	Weber number in mixing vessel	[—]
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	$[cm^2/s^3]$
η	= Kolmogoroff length	[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	[—]
v	= kinematic viscosity	$[cm^2/s]$
ρ	= specific weight	$[g/cm^3]$
σ	= interfacial tension	$[g/s^2]$
ϕ	= volumetric fraction of dispersed phase	[—]

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A THEORETICAL AND EXPERIMENTAL STUDY ON THE OSCILLATION OF A HANGING DROP

TAKAO TSUKADA, MASAKAZU SATO, NOBUYUKI IMAISHI,
MITSUNORI HOZAWA AND KATSUHIKO FUJINAWA

*Chemical Research Institute of Non-Aqueous Solutions,
Tohoku University, Sendai 980*

The damping oscillation of a hanging drop on a nozzle in the air was studied theoretically and experimentally. In the experiments, an elongated drop on a brass nozzle by electrostatic force was made to oscillate by removing the electric field instantaneously and the subsequent drop motion was recorded by a high-speed camera. In the theoretical study, the time-dependent drop shape and flow inside the drop were simulated numerically by use of the finite element method. It was found that the experimental results of oscillatory behavior for a hanging drop of aqueous glycerin solution were in good agreement with the calculated ones and that the frequency of oscillation was affected by drop volume, surface tension and nozzle size.

Introduction

It is of practical interest to know the behavior of bubbles or drops moving in a fluid. However, as the shape of the two-fluid interface usually changes with time,[6,10,11,15] a strict analysis is very difficult.

In this work, numerical simulations of the oscillations of a hanging drop on a nozzle in the air were carried out, as the first step in the analysis of the oscillatory behavior of moving drops or bubbles in other fluids.

Rayleigh[9] is the pioneer for this problem. He attempted to analyze theoretically the small-amplitude oscillations of an inviscid drop. Lamb[5] extended the work by considering the density effect of the surrounding fluid.

Miller and Scriven[7] solved a similar problem, taking the viscosity of the two fluids into account.

Recently, Prosperetti[8] calculated the initial value problem for the linear free oscillation of a bubble or a drop, and Tsamopoulous et al.[12] analyzed theoretically the moderate-amplitude nonlinear oscillations of inviscid drops and bubbles using a Poincaré-Lindstedt expansion technique.

However, there have been few studies[1] in which the full Navier-Stokes equations for the oscillations of moving bubbles or drops are solved.

Hughes et al.[3] and Kheshgi et al.[4] reported that it was efficient to use the finite element method including the Lagrangian-Eulerian description for the calculation of the transient flow problem with free surface.

The first aim of this work is to analyze theoretically the oscillations of a hanging drop by use of the finite element method.[3,4,13] The second aim is to confirm experimentally the propriety of the calculation method and to identify the factors that affect the oscillations.