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#### NOTATION

A = total flow area per channel

a, b, c = constants defined by Equation (31)

D = hydraulic diameter

 $f_i$  = interfacial friction factor

 $f_{wf}$  = wall friction factor based on film

g = acceleration of gravity i = total volumetric flux

 $j_f$  = liquid volumetric flux, superficial liquid velocity

j<sub>ftr</sub> = absolute liquid volumetric flux at laminar turbu-

lent transition, defined by Equation (25)

p = pressure

 $P_i$  = wetted perimeter of interface

 $P_{wf}$  = wall perimeter wetted by film

 $Re_f = \text{film Reynolds number defined by Equation (18)}$ 

t = time

 $V_{gj}$  = vapor drift velocity

 $v_f$  = average liquid velocity

 $v_g$  = average gas velocity

 $v_m$  = mixture center-of-mass velocity

 $v_r = v_g - v_f$ , relative velocity between phases

z = axial coordinate

#### **Greek Letters**

 $\alpha$  = void fraction of gas phase

 $\Gamma_a$  = vapor generation rate

 $\Delta \rho$  = density difference

 $\delta$  = average film thickness  $\epsilon$  = roughness parameter of interface

 $\mu_f = \text{liquid viscosity}$ 

 $\xi$  = ratio of the two wetted perimeters given by Equa-

tion (14)

 $\rho_f = \text{liquid density}$ 

 $\rho_g = \text{gas density}$ 

 $\rho_m = \text{gas density}$   $\rho_m = \text{mixture density}$ 

 $\tau_i$  = interfacial shear stress

 $\tau_{wf}$  = wall shear stress

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# Drop Size Distributions and Coalescence Frequencies of Liquid-Liquid Dispersions in Flow Vessels

A flash photomicrographic method and modified dye-light transmittance technique are employed to measure size distributions and mixing frequencies in a turbulently agitated flow vessel. This liquid-liquid dispersion is nearly spatially homogeneous. Mixing frequencies are strongly dependent on impeller speed and moderately dependent on holdup fraction.

C. A. COULALOGLOU

and

L. L. TAVLARIDES

Department of Chemical Engineering Illinois Institute of Technology Chicago, Illinois 60616

Correspondence concerning this paper should be addressed to L. L. Tavlarides, C. A. Coulaloglou is with Exxon Research and Engineering Company, Florham Park, New Jersey.

For a rational design of rate processes in turbulently agitated liquid-liquid dispersions, it is necessary to predict droplet surface area in terms of size distributions and droplet mixing rates. Interaction models such as simulation techniques and population balance equations are employed for such predictions. The main obstacles in utilizing these models is to generalize on the form of the droplet coalescence and breakage terms as a function of the hydrodynamics and physical properties of the system.

The complexity of this problem necessitates experimentation for insight into factors affecting droplet breakage and coalescence. Earlier experimental works focused on measurements of average droplet size by use of light transmittance techniques or chemical methods. Size distributions were also studied by measurements with photography, encapsulation, sedimentation, and via following electroresistivity changes by flow through an orifice. Correlations for average drop size have thus resulted. Concurrently, droplet mixing measurements were made by various indirect methods such as following a chemical reaction or dispersion of a nontransferring dye in the dispersed phase. These experiments were largely conducted in batch systems. A number of investigators reported experimental data on interfacial areas (Mok and Treybal, 1968; Schindler and Treybal, 1968; Weinstein and Treybal, 1973) and

drop size distributions (Boyatiotis and Thornton, 1967) in flow systems. These data do not permit a complete description, as both size distributions and droplet mixing frequencies were not simultaneously obtained.

Recently, data were presented (Verhoff, 1969; Ross, 1971; and Ross and Curl, 1973) on both size distributions and droplet mixing frequencies by measuring the bivariate drop volume-dye concentration distribution in a flow vessel. The method employed is limited to dispersions with low energy input where the equilibrium average drop size is large.

The experimental method discussed in this work permits the measurement of drop size distributions and coalescence frequencies for a wide range of energy input in flow vessels. A flash photomicrographic method and a modified dye-light transmittance technique are employed. The coalescence frequencies are calculated from the data by extension of previous methods (Curl, 1963; Miller et al., 1963). The average drop sizes are correlated as functions of dispersed phase holdup fraction and Weber number. In another report these data are employed to test the validity of breakage and coalescence functions based on phenomenological concepts and the theory of local isotropic turbulence (Coulaloglou and Taylarides, 1975).

#### CONCLUSIONS AND SIGNIFICANCE

Drop size distribution measurements in the circulation and impeller regions and dispersed phase coalescence frequencies measurements were made concurrently for the turbulently agitated dispersion of kerosene-dichlorobenzene (d) in water (c) in a 12 l flow vessel. The flash photomicrographic method developed and a dye-light transmittance method employed appear to be suitable experimental techniques for these measurements. The range of operating conditions studied were dispersed phase holdup fraction  $0.025 < \phi < 0.15$ , impeller speed 190 < N < 310 rev./min., and nominal residence time of 10 min.

Differences observed in the size distributions between the regions were smaller than would be expected considering the largely different local hydrodynamic characteristics of the flow field. The relative spatial homogeneity of the dispersion can be attributed to the low coalescence frequency amongst the droplets in the dispersion  $\omega$  (0.148 min.<sup>-1</sup>, 1.12 min.<sup>-1</sup>) relative to the average internal circulation frequency of fluid elements (containing dispersed phase droplets) from the impeller tip and back  $f_c$  (35 min.<sup>-1</sup>, 60 min.<sup>-1</sup>). It is suggested that the ratio of the coalescence frequency to the circulation frequency  $\omega/f_c$  can serve as a measure to determine whether or not one should treat a dispersion as being spatially homogeneous.

As  $\omega/f_c$  approaches zero, the dispersion is observed to be homogeneous.

The Sauter mean diameter  $d_{32}$  is correlated similar to other investigators by use of the theory of local isotropic turbulence and the assumption that droplet breakup is based on a critical Weber number for droplet diameter greater than the microscale of turbulence by the following equation:

$$\frac{d_{32}}{D} = 0.081(1 + 4.47 \,\phi) \,(We)_T^{-0.6}$$

A variance in the equation parameters to those reported for batch operation in comparable dispersed phase systems may be a result of the difference in the vessel operation during data acquisition. Larger  $d_{32}$  values for continuous flow vs. values for batch operation may in part be attributed to flow effects on the stabilized drop size.

Models useful for calculation of dispersed phase coalescence frequencies from light transmittance were developed by extending those proposed by previous investigators (Curl, 1963; Miller et al., 1963). The coalescence frequency is shown to have a strong dependence on impeller speed N ( $\omega \propto N^{2.85}$ ) when compared to  $d_{32}$  dependence ( $d_{32} \propto N^{-1.2}$ ).

The study of two phase dispersed systems in agitated vessels has been the subject of numerous investigations (Gal-Or et al., 1969; Tavlarides et al., 1970). In such systems the drops of the dispersed phase are continuously broken and coalesced at a rate depending on the local hydrodynamic characteristics of the flow field and the physical properties of the system. Generally, it is assumed that for  $Re_T > 10\,000$  the flow field may be considered locally isotropic (Rushton et al., 1950; Bates et al., 1963). Recently (Brodkey, 1967; Rao and Brodkey, 1972) it was reported that for the shear flows near the impeller region, true local isotropic turbulence would not be expected until

the microscopic Reynolds number  $Re_{\lambda}$  reaches values between 300 to 800. Far from the impeller, a limited range of  $k^{-5/3}$  spectrum exists for  $Re_{\lambda} > 130$ . When the turbulence is locally isotropic, the flow field can be described in terms of the theory of locally isotropic turbulence (Kolmogoroff, 1941a; Hinze, 1959).

For drops much larger than the microscale of turbulence  $\eta$ , consideration of the balance between inertial and surface forces leads to  $d_{\rm max} \propto \epsilon^{-0.4}$ . This has been confirmed experimentally for a variety of dispersed phase systems, covering a wide range of physical properties and experimental techniques (Vermeulen, 1955; Calderbank,

1958; Shinnar and Church, 1960; Shinnar, 1961; Chen and Middleman, 1967; Sprow, 1967a). For drops smaller than  $\eta$ , viscous effects predominate, and Taylor's analysis (Taylor, 1934) predicts  $d_{\rm max} \propto \epsilon^{-0.5}$  (Shinnar, 1961). Experimental evidence of this dependence has been reported for gas-liquid systems (Vermeulen, 1955) and for low interfacial tension systems (Sprow, 1967b), although in both cases the drop size was greater than the microscale of turbulence.

The effect of holdup fraction on Sauter mean diameter has been accounted by an expression of the form  $d_{32} \propto f(\phi)d_{32}^0$ , where  $f(\phi)$  is a linear function of  $\phi$  and  $d_{32}^0$  is the Sauter mean diameter for very dilute dispersions ( $\phi \rightarrow 0$ ) (Vermeulen, 1955; Calderbank, 1958; Brown and Pitt, 1970; Mlynek and Resnick, 1972). The dispersed phase hold up fraction varies with location depending on impeller speed and density differences between continuous and dispersed phase (Bouyatiotis and Thornton, 1967; Schinder and Treybal, 1968; Mok and Treybal, 1968; Weinstein and Treybal, 1973; Hoffer and Resnick, 1975).

Various investigators reported local variation of interfacial areas and drop sizes, although the detailed dependence on conditions is not yet known (Vanderveen, 1960; Sprow, 1967b; Schindler and Treybal, 1968; Mlynek and Resnick, 1972; Weinstein and Treybal, 1973; Park and Blair, 1975). In a continuous flow vessel, the drop size will also be effected by the residence time of the dispersion (Weinstein and Treybal, 1973). No correlation has been developed to explicitly account for this effect.

Measurements of interaction rates have been limited to the determination of average coalescence frequencies. The methods used may be classified as chemical methods (Madden and Damerell, 1962; Rietema, 1964; Hillestead, 1965; Komasawa et al., 1969; Shiloh et al., 1971; Villermaux and Devillon, 1972) and physical methods with tracer dye (Vanderveen, 1960; Miller et al., 1963; Verhoff, 1969; Komosawa et al., 1971; Villermaux and Devillon, 1972; Ross and Curl, 1973), impeller speed changes (Howarth, 1967; Mlynek and Resnick, 1972), or other methods (Groothuis and Zuiderweg, 1964; Schindler and Treybal, 1968; Koetsier and Thoenes, 1972). Previous coalescence models were employed to interpret coalescence frequency measurements (Levich, 1962; Howarth, 1964; Shiloh et al., 1971; Koetsier and Thoenes, 1972).

# MODEL FOR MEASURING COALESCENCE RATES IN CONTINUOUS FLOW AGITATED DISPERSIONS

The determination of coalescence frequency requires the measurement of the rate of change of physical or chemical parameters and a model describing the mixing process. The model first proposed by Curl (1963) and by Miller et al. (1963) assumes equal size drops which have equal probability to coalesce. Redispersion of two coalesced drops which had tracer concentration c' and c'' occurs instantaneously, producing two equal size drops each of concentration (c'+c'')/2. A modification is made here to interpret mixing frequencies in a continuous flow well-mixed vessel. Although the model is simplistic in structure, it provides a reasonable first estimate of the mixing frequency. More accurate modeling of dispersed phase mixing are provided elsewhere (Coulaloglou and Tavlarides, 1975).

The concentration distribution of nontransferring tracer in the droplets is

$$\frac{\partial p(c,t)}{\partial t} = \frac{1}{\tau} \left[ p_f(c,t) - p(c,t) \right] 
+ \omega \left[ 4 \int_0^c p(c+a)p(c-a)da - p(c,t) \right]$$
(1)

 $\omega$  is defined as the volume fraction of the dispersed phase coalescing per unit time.

Assume two streams of dispersed phase with flow rates  $q_1$  and  $q_2$  and no tracer enter the vessel with the continuous phase. Consider two experiments: (a) A step input of tracer of concentration c is introduced into the stream of flow rate  $q_1$ , and the light transmittance  $\overline{I}_a$  is measured at steady state. The drop concentrations have variance  $\mu_{2a}$ . (b) The same two dispersed phase streams in (a) are premixed, and light transmittance  $\overline{I}_b$  is measured at steady state. The steady state variance of drop concentration is zero. The feed concentration distribution for the first experiment is given by

$$p_f(c,t) = \phi_f \, \delta(c-c_1) + (1-\phi_f)\delta(c)$$
 (2)

where  $\phi_f = q_1/(q_1 + q_2)$ . The concentration variance is obtained by multiplying Equation (1) by  $[c - \overline{c}(t)]^2$  and by integrating to give

$$\mu_{2a}(t) = rac{\phi_f(1-\phi_f)c_1^2}{\omega au + 2} \left(e^{-t/ au} - e^{-\omega t/ au}
ight)$$

$$+\frac{2\phi_f(1-\phi_f)c_1^2}{\omega\tau+2}\left[\begin{array}{c} -\left(\frac{1}{\tau}+\frac{\omega}{2}\right)t\\ 1-e \end{array}\right]$$
(3)

which at steady state becomes

$$\mu_{2a} = \frac{2\phi_f(1-\phi_f)c_1^2}{\omega\tau + 2} \tag{4}$$

The steady state average concentration of the tracer in the dispersed phase is equal to  $\overline{c} = \phi_f c_1$ . The concentration variance  $\mu_{2a}(t)$  can be related to the mixing frequency

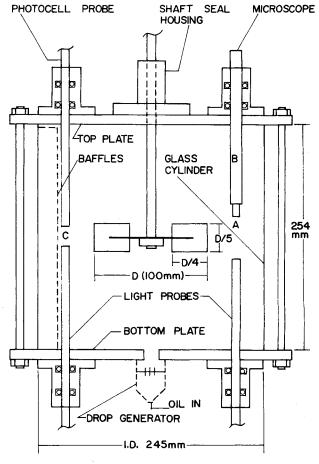


Fig. 1. Mixing vessel.

and the normalized light transmittance (Miller et al., 1963). Analogously, for  $\beta \overline{d}_{\mu_{2a}}(t) < 1$ 

$$\theta = \frac{\log \frac{\overline{I_a}(t)}{\overline{I_o}}}{\log \frac{\overline{I_b}}{I_c}} = \frac{\overline{c_a}(t)}{\beta \overline{d\mu_{2a}}} \ln \left(1 + \frac{\beta \overline{d\mu_{2a}}}{c_a(t)}\right)$$
(5)

where  $\overline{I}_o$ ,  $\overline{I}_a(t)$ ,  $\overline{I}_b$  are the average light transmittance initially prior to dye addition, at time t for experiment (a) and at steady state after dye addition for experiment (b).

Equation (5) may be used at steady state by substitution of Equation (4) and by noting that  $\overline{c}_a = \phi_f c$ , to obtain

$$\theta = \frac{\log \frac{I_a}{\overline{I}_b}}{\log \frac{\overline{I}_b}{\overline{I}_o}}$$

$$= \frac{2 + \omega \tau}{2(1 - \phi_t)c_t \beta d} \ln \left( 1 + \frac{2(1 - \phi_t)c_1 \beta \overline{d}}{2 + \omega \tau} \right) \quad (6)$$

Equation (6) was employed to calculate the coalescence frequency as all parameters can be measured.

The value of  $\omega$  obtained should be considered an approximation to the true coalescence frequency because of the uniform drop assumption, because coalescence among drops of identical concentration does not cause any change in the concentration variance, and because coalescence of a small drop with a large one will cause small differences in concentration variance.

#### **EXPERIMENTAL\***

### Experimental Apparatus

The apparatus consisted of a baffled stirred vessel with a turbine impeller and flow inlets for two dispersed phase streams and a continuous phase stream (see Figure S1°). Measurements made are: flow rates of feed streams, oil drop size distribution in impeller and circulation regions, and dispersion light transmittance  $\overline{I}_0$ ,  $\overline{I}_a$ , and  $\overline{I}_b$ . Apparatus in contact with fluids are stainless steel, glass, teflon, or viton. The two oil streams flow from the pressurized tanks through drop generators into the agitated vessel. The drop generators produce approximately equal size drops. The 12 l tank (Figure 1) has four baffles and a disk turbine impeller with six flat blades center positioned. The light transmittance and photographic probes traverse vertically 9.0 cm from the axis of the vessel.

The photographic system consisted of a photomicrographic probe assembly, a light probe, and an electronic flash unit. Photographs of the dispersion were taken by using high contrast Kodak film at locations A and B (12.3 and 2.54 cm from the vessel top plate) with dye free dispersed phase. The light transmittance system included a light source, a photomultiplier and a photomultiplier housing, and a detection unit.

An oil mixture of 63% kerosene and 37% dichlorobenzene ( $\rho_d=0.972$  and  $\mu_d=1.30$  centipoise) was dispersed in single distilled water. The phases were mutually saturated. The nontransferring azobenzene dye added to the dispersed phase resulted in slight change of interfacial tension ( $\sigma=42.82$  dyne/cm for oil mixture +0.04 N dye vs.  $\sigma=43.03$  dyne/cm for oil mixture).

The magnification of the optical system and camera was determined by underwater pictures of a microscope graticule.

#### **Experimental Results**

Drop size distributions and the coalescence frequencies

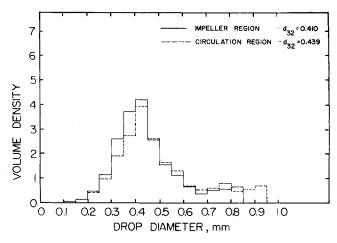


Fig. 2. Drop size distribution at N=190 rev./min.,  $\phi=0.10$ .

were studied for impeller speed of 190 to 310 rev./min. and holdup of 0.025 to 0.15 for a residence time of 10 min.

Three hundred or more drops were measured in each analysis. Figure 2 is a typical distribution of normalized drop volume density vs. drop diameter for the impeller and circulation region.

The experimental coalescence frequency was determined from Equation (6), where

$$d_{32} = rac{V^i d_{32}{}^i + V^c d_{32}{}^c}{V_T}$$

was employed for the drop size. The volume of the impeller region is assumed equal to the volume of the impeller discharge stream which is about 0.1 of the tank volume (Cutter, 1966; DeSouza and Pike, 1972). The residence time was taken equal to  $\tau = V_T/Q_T$ . Table 1° summarizes the results.

#### DISCUSSION

## Spatial Homogeneity Characteristics of Turbulently Agitated Dispersions

It has been postulated from theoretical analysis of drop breakup that (Kolmogoroff, 1949; Hinze, 1955; Shinnar, 1961)

$$\left(\frac{\rho}{\sigma}\right)\overline{u^2} (d_{\max})d_{\max} = \text{constant} \quad \text{for} \quad \eta < d < l_e$$
 (7)

For isotropic turbulence

$$\overline{u}^{2}(d_{\max}) = K_{1} \left(\epsilon d_{\max}\right)^{2/3} \tag{8}$$

Hence

$$d_{\max} = K_2 \left(\frac{\rho}{\sigma}\right)^{-3/5} \epsilon^{-2/5} \tag{9}$$

or, assuming a linear relationship between  $d_{\rm max}$  and the Sauter mean diameter  $d_{32}$  (Sprow, 1967), we get

$$d_{32} = K_3 \left(\frac{\rho}{\sigma}\right)^{3/5} \epsilon^{-2/5} \tag{10}$$

where  $\epsilon$  is the local energy dissipation per unit mass. In an agitated vessel, the point value of  $\epsilon$  as well as the local turbulence parameters of the flow field change with distance from the impeller tip (Cutter, 1966; Schwartzberg and Treybal, 1968; Rao and Brodkey, 1972). Although this change is continuous, one may consider that an agitated vessel consists of two regions differing substantially in terms of macroscopic as well as microscopic flow characteristics. A region of high rates of energy dissipation and high turbulence intensity u' near the impeller and a region of low rates of energy dissipation and low u' far from the impeller. The former region includes the impeller discharge stream, the impeller region, and the latter the remainder of

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the vessel, the circulation region. In this work, the average energy dissipation per unit mass in the impeller region  $\epsilon_{\rm c}$  and the circulation region  $\epsilon_{\rm c}$  for 190 rev./min. was calculated to be 4.67  $\times$  10<sup>4</sup> and 0.33  $\times$  10<sup>3</sup> cm<sup>2</sup>/s<sup>3</sup>, respectively (Cutter, 1966). According to Equation (10)

$$\frac{d^{i}_{32}}{d^{c}_{32}} = \left(\frac{\overline{\epsilon_{i}}}{\overline{\epsilon_{c}}}\right)^{-0.4} = 0.138 \tag{11}$$

Equation (11) suggests that  $d^{i}_{32}$  should be seven times smaller than  $d^{c}_{32}$  if the residence time of the dispersed phase in each region is high enough for steady state conditions to be attained. From Figure 2 the ratio of experimental Sauter mean diameters was found to be

$$\left(\frac{d^{i}_{32}}{d^{c}_{32}}\right)_{\rm exp} = 0.93\tag{12}$$

which was a typical value for all runs ( $\pm$  0.02). The near unity of this ratio [which is seven times greater than the prediction of Equation (11)] indicates a spatial homogeneity of this dispersion with regard to size distribution. This spatial homogeneity can be explained in terms of the relative values of the coalescence and circulation frequencies. The circulation frequency is (Holmes et al., 1964)

$$f_c = \frac{1}{t_c} = \frac{N}{0.85} \left(\frac{D}{D_T}\right)^2$$
 (13)

For these studies, fc ranges from 35 to 60 min. -1 compared to coalescence frequencies of 0.148 to 1.120 min.-1 for 190 to 310 rev./min., respectively. Therefore, only a small fraction of the drop population that would coalesce given enough residence time in each region actually does. The dimensionless group  $\omega/f_c$  may serve as a measure of the homogeneity of the dispersion. For  $\omega/f_c \to 0$ , the dispersion will be homogeneous. For  $\omega/f_c$  increasing, the dispersion will become less and less homogeneous. The value of  $\omega/f_c$  for all runs was smaller than 0.018. This is consistent with the near equal size distribution for the impeller and circulation region shown in Figure 3. The difference in volume density between the two regions of the vessel are plotted against the drop size in Figure S2.\* As the dispersed phase moves from the impeller to the circulation region, the net effect of coalescence and breakage results in loss of small drops and generation of large ones. The opposite occurs from the circulation to the impeller region. The net change in drop sizes estimated from Figure S2\* is 9% of the dispersed phase volume.

# Sauter Mean Diameter Correlation and Dispersed Phase Coalescence Frequency

The Sauter mean diameter in the vessel was observed to be moderately dependent on impeller speed. Previous investigators found that the Sauter mean diameter varies as  $N^{-1.2}$  for batch operations (Vermeulen et al., 1955; Calderbank, 1958; Chen and Middleman, 1967; Sprow, 1967a; Mlynek and Resnick, 1972; Brown and Pitt, 1970) in accordance with Equation (10). Bouyatiotis and Thornton (1967) measured drop sizes near the bottom of an agitated vessel and found that  $d \propto N^{-0.96}$  for batch and continuous operation. Also, Ross (1971) reported that  $d \propto N^{-1}$  for continuous operation. A least-square fit of the data for the impeller region, the circulation region, and the whole vessel showed that most of the data could be correlated by a value of the exponent of N equal to -1.2 ( $d_{32} \propto N^{-1.2}$ , see Figure 3). This dependence of  $d_{32}$  on N indicates that the drop size is determined primarily by the breakage process in accordance with Equation (11). The effect of holdup fraction of the dispersed phase on

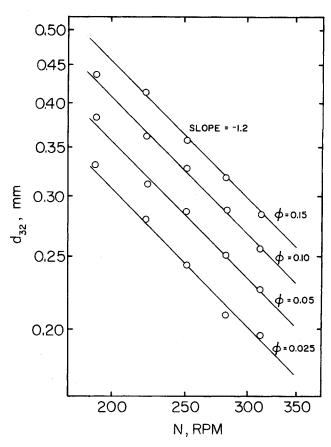


Fig. 3. Effect of impeller speed on volume average Sauter mean diameter for the vessel.

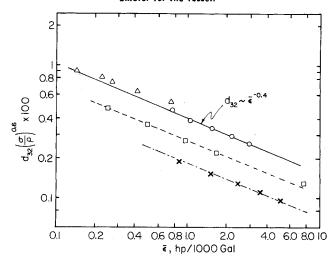


Fig. 4. Comparison of Sauter mean diameters with other works, φ = 0.10: □—(Mlynek and Resnick, 1972) batch, X—(Brown and Pitt, 1974) batch, △—(Ross, 1971) continuous, and ○—(present work) continuous.

the Sauter diameter was linear, and the final correlation in terms of the vessel Weber number for all data is given by

$$\frac{d_{32}}{D} = 0.081 (1 + 4.47\phi) (We_T)^{-0.6}$$
 (14)

Figure S3° shows the comparison between the experimental Sauter mean diameters and those calculated by using Equation (14). Brown and Pitt (1970) and Mlynek and Resnick (1972) reported values for the coefficient of  $\phi$  of 3.15 and 5.14, respectively. The coefficient 0.081 is 1.4 and 1.6 times the value reported by the above investigators. The difference may in part be explained in terms of continuous vs. batch operation. For batch operation the

<sup>•</sup> See footnote on p. 292.

Investigator	Correlation	$ ho_d$ [g/cm <sup>3</sup> ]
Vermeulen (1955), Calderbank (1958), Sprow (1967a), Chen and	$rac{d_{32}}{D} = c_1 (We)_{T}^{-0.6}$ $0.51 \le c_1 \le 0.63$	0.692 —1.20
Middleman (1967) Shinnar (1961) Sprow (1967b)	$rac{d_{32}}{D} = c_{2\sigma} D(We)_T^{-0.375}$ $c_2$ — experimentally determined	0.80
Sprow (1967a)	$\frac{d_{32}}{D} = c_3(\rho_c \mu_c)^{-0.5} \left(\frac{\rho_c}{\sigma D}\right)^{-0.25} (We)_{T}^{-0.75}$	0.80
Bouyatiotis and Thornton (1967)	$\begin{aligned} c_3 &= \text{experimentally determined} \\ d_{32} &= 5.38 \left(\frac{\mu_c}{\rho_c g}\right) \left[\frac{(p/V_T)^3 g_c{}^3}{\rho_c{}^2 \mu_c g^4}\right]^{-0.16} \left[\frac{\rho_c{}^3 \sigma}{\mu_c{}^4 g}\right]^{-0.07} \\ &+ 1.18 \phi \left[\frac{\sigma^2}{\mu_c{}^2 g}\right] \left[\frac{\Delta \rho \gamma^3}{\mu_c{}^4 g}\right]^{-0.62} \left[\frac{\Delta \rho}{\rho_c}\right]^{0.05} \end{aligned}$	0.691-0.905
Weinstein and Treybal (1973)	$ ho_d <  ho_c$ $ d_{32} = 10^{(-1.812 + 0.732\phi)} \nu_c^{0.047} \epsilon^{-0.204} \left(\frac{\sigma g_c}{ ho_c}\right)^{0.274}$	0.831-0.996
Weinstein and Treybal (1973), cont'd.	where $rac{ar{\phi}}{ar{\phi}_F} = 10^{0.53}  N_{P7}^{0.247}  N_F^{-0.427}  N_{ ho}^{-0.430}  N_{\mu}^{0.0987}$ $ ho_d >  ho_c$ $d_{32} = 10^{(-1.978  +  0.672  ar{\phi})}  _{ u_c}^{0.0792}  _{\epsilon}^{-0.194}  \left(rac{\sigma g_c}{ ho_c} ight)^{0.196}$	0.831-0.996
Calderbank (1958)	where $\dfrac{\overline{\phi}}{\phi_F} = 10^{0.703}  N_{Pr}^{-0.131}  N_F^{-0.0752}  N_{\rho}^{-0.0677}  N_G^{0.0299}  N_{\mu}^{0.0299}  N_{\mu}^{0.0299}  N_{\rho}^{-0.0677}  N_G^{0.0299}  N_{\mu}^{0.0299}  N_{\mu}^$	0.87-1.59
Brown and Pitt (1970)	$\frac{d_{32}}{D} = 0.051(1 + 3.14\phi) \ (We)_{T}^{-0.6}$	0.783-0.838
Mlynek and Resnick (1972)	$\frac{d_{32}}{D} = 0.058(1 + 5.4\phi) \ (We)_{T}^{-0.6}$	1.055
Present Work (1975)	$rac{d_{32}}{D} = 0.081(1 + 4.47\phi) \; (We)_{T}^{-0.6}$	0.972
Brown and Pitt (1974)	$d_{32}^{5/2} \left(\rho/\sigma\right) \left(\frac{e^{2/3} t_c}{D_T}\right) = c$	0.783-0.838
	c = experimentally determined	

steady state drop size distribution in an agitated vessel is determined by the competing coalescence and breakage processes which depend on the flow field characteristics and the system physical properties. In a flow vessel, the escape rate of dispersed drops has an added effect on drop size distribution (Weinstein and Treybal, 1973; Coulaloglou, 1975). For very low escape rates or high residence time, the drop size distribution approaches that for batch operation. Figure 4 shows the comparison between the Sauter mean diameter of the present work with that of other investigators (Brown and Pitt, 1970; Ross, 1971; Mlynek and Resnick, 1972) for batch and continuous operations. One should be cautious in using correlations from batch experiments to predict Sauter mean diameters in flow vessels.

A summary of various correlations available in the literature for Sauter mean diameters related to the work presented in this investigation is given in Table 1. Care should

be employed in any comparisons because of the variety of operating conditions, range of physical parameters, and effects of trace amount of impurities.

The coalescence frequency is a strong function of impeller speed in contrast to the moderate  $d_{32}$  dependence on N. From Figure 5 and Table S1,\* it is seen that  $\omega \propto N^{2.85}$ . A 31% increase from 190 to 250 rev./min. results in more than a 160% increase in coalescence frequency for  $\phi=0.05$ . For the same conditions the Sauter mean diameter decreases only by 25%. Shinnar's theory of turbulence-stabilized dispersions may be employed to explain the large increase in coalescence frequency (Shinnar, 1961). According to Shinnar, there is a maximum drop size  $d_{\rm max}$  below which the breakage rate is zero. Also, there is a minimum drop size  $d_{\rm min}$  above which coalescence rate is zero. All the drops between  $d_{\rm min}$  and  $d_{\rm max}$  are stable.

<sup>\*</sup> See footnote on p. 292.

FOR STIRRED LIQUID-LIQUID CONTACTORS

Physical properties			Operating conditions				Experimental	
μ <sub>δ</sub> [cp]	$\sigma[\mathrm{dyne/cm}]$	B/C	$D_T[\mathrm{in.}]$	D[in.]	φ	N[rpm]	$ au[ ext{min}]$	techniques and comments
0.37 —25	3.6-55.1	В	4-20	2-7	0.005-0.015	80-2000	_	Light transmittance, photography, Coulter-Counter
0.59	9.3	В	8.75, 11.5	2.5, 8.75	0.05-0.25	600-2000		Photography, Coulter-Counter, strongly coalescing systems, [Sprow
0.59	41.8	В	8.75	1.25-4.0	0.005-0.05	250-2000	_	(1967b)] Coulter-Counter
0.486-0.748	8.5-40	В-С	7	2.7	0.2-0.5	400-1000	_	Photography
0.722-7.43	3.76-36	C	9.6, 14.6	3.5, 5	0.079-0.593	150-620	0.165 4.2	Light transmittance
0.722-7.43	3.76-36	<b>C</b> ·	9.6, 14.6	3.5, 5	0.079-0.593	150-620	0.165 4.2	Light transmittance
0.70-1.038	35-40	В	7, 15	2.3-10	0.0-0.20	_	_	Light transmittance
0.59-3.30	1.9-5.0	В	11.8	4	0.05-0.30	250-450	_	Light transmittance
-	41	В	11.4	4	0.025-0.34	140-500	-	Light transmittance
1.30	43	C	9.6	4	0.025-0.15	190-310	10	Photomicrography
0.59-3.30	1.9-50	В	11.8	4	0.05	250-450		Light transmittance

Application of Shinnar's theory in this system would mean that at 190 rev./min. most of the drops are between  $d_{\rm max}$  and  $d_{\rm min}$ . At 250 rev./min. the number of drops formed below  $d_{\rm min}$  increases, and the width of the stable region decreases. Consequently, the fraction of unstable drops will increase, and the coalescence frequency will be greater. As N increases, the stabilized region decreases in size, and eventually all the drops become unstable. The results show that drop size increases moderately with increasing holdup fraction at a given impeller speed. The coalescence frequency varies with  $\phi$  as  $\omega \propto \phi^{0.45}$ .

A direct comparison of experimental frequencies with those of other investigators is difficult because of the marked effects of minute traces of contaminant, variations in temperature and interfacial surface tension, and operation conditions ( $\tau$ ,  $\phi$ , etc.). However, the comparison of

Figure 6 gives encouraging agreement, at least with regard to the order of magnitude obtained. Coalescence frequencies were measured in a continuous system with residence time 7 and 19.6 min., respectively by Groothuis and Zuiderwig (1964) and by Ross (1971), whereas others employed batch systems (Miller et al., 1963; Howarth, 1967). All these investigators used largely different experimental techniques.

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<sup>•</sup> See footnote on p. 292.

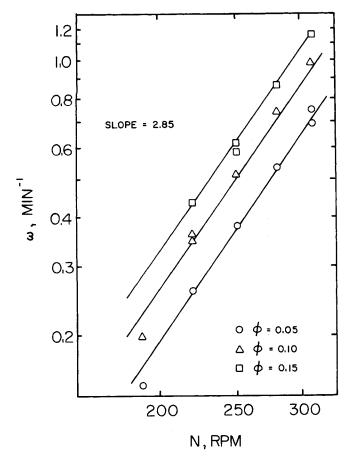


Fig. 5. Effect of impeller speed on coalescence frequency  $\omega$ .

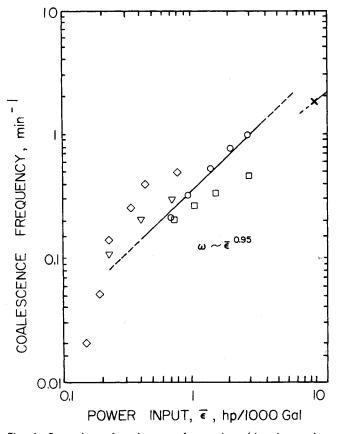


Fig. 6. Comparison of coalescence frequencies with other works:  $\bigcirc$ —(present work)  $\phi=0.10, \, \nabla$ —(Groothuis and Zuiderweg, 1964)  $\phi=0.08,\, \diamondsuit$ —(Ross, 1971)  $\phi=0.10,\, \Box$ —(Howarth, 1967)  $\phi=0.10,\, \Box$ 0.10, and X—(Miller et al., 1963)  $\phi=0.10.$ 

#### NOTATION

 $A_v(d_i)$  = volume density at  $d_i$ c = dye concentration

 $c_1$  = dye concentration in one of the streams of dis-

persed phase (steady state case)

 $\overline{d}$  = average drop diameter

 $d_{32}$  = Sauter mean diameter,  $\frac{\sum n_i d_i^3}{\sum n_i d_i^2}$ 

 $d_{\text{max}} = \text{maximum drop diameter}$ 

= impeller diameter

 $D_T = \tan k \text{ diameter}$  $f_c = \text{circulation frequency}$ 

 $\overline{I_o}$  = initial average transmittance prior to dye addition

 $\vec{I}(t)$  = average light transmittance at time t

 $\overline{I_a}$ ,  $\overline{I_b}$  = average light transmittance at steady state when the dispersed phase streams are not premixed and

k = are premixed = wavelength

 $K_1, K_2, K_3 = \text{constants}$   $l_e = \text{linear scale of energy containing eddies}$ 

 $\tilde{N}$  = impeller speed

 $N_F$  = dimensionless number,  $(Q_d \rho_c \sigma g_c)/\mu_c^3$  $N_G$  = dimensionless number,  $(\mu_c^4 g)/(\rho_c \sigma^3 g_c^3)$ 

 $V_{Pr}$  = dimensionless number,  $(PQ_d \mu_c^2)/(V_T \sigma^3 g_c^2)$ 

 $N_{
ho}=$  dimensionless number,  $\Delta 
ho/
ho_c$   $N_{\mu}=$  dimensionless numbers,  $\mu_d/\mu_c$ p(c)= concentration distribution

 $p_f(c)$  = feed concentration distribution

q<sub>1</sub>, q<sub>2</sub> = flow rate of the dye containing pure dispersed phase stream, respectively

 $Q_T = \text{total flow rate of continuous and dispersed phase}$ 

 $\hat{R}e_{\lambda} = \text{microscale Reynolds number, } u' \lambda_g/\bar{\nu}_c$  $Re_T = \text{tank Reynolds number, } ND^2/\nu_c$ 

= time

t

 $t_c$  = circulation time

 $\underline{u'}$  = turbulence intensity

 $\overline{u^2}(d) = \text{mean square of the difference of the velocities}$  at distance d

 $V_T = \text{tank volume}$ 

 $(We)_T = \text{tank Weber number}, N^2 D^3 \rho_c/\sigma$ 

#### **Greek Letters**

 $\beta$  = extinction coefficient of dye

 $\delta(c) = \text{delta function}$ 

 $\epsilon, \epsilon$  = local and average energy dissipation per unit

mass, respectively

= microscale of turbulence

 $\dot{\theta}$  = dimensionless light transmittance

 $\lambda_g$  = dissipation scale

 $\mu = \text{viscosity}$ 

 $\mu_2$  = concentration variance

 $\rho$  = density

 $\sigma$  = interfacial tension

= nominal residence time of continuous and dis-

persed feed

 $\phi_f$  = fraction of dispersed feed stream containing dye

 $\phi_F$  = fraction of dispersed phase in the feed

 $\phi$  = holdup fraction

ω = coalescence frequency

#### Subscripts

d = dispersed phase

= continuous phase

#### Superscripts

c = circulation region

i = impeller region

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