

and Macosko, 1980). However, either can be used, the only restriction being to choose a single basis for mixer comparison.

ACKNOWLEDGMENT

This work was supported by the Materials Research Laboratory.

NOTATION

$A(z)$	= cross-sectional area in continuous mixer (m^2)
a_v	= intermaterial area per unit volume (m^{-1})
D	= stretching tensor, $(\text{grad} \mathbf{v}) + (\text{grad} \mathbf{v})^T$, (s^{-1})
$e(x,t)$	= mixing efficiency, dimensionless
$eff(z)$	= mixing efficiency, dimensionless
l	= length of mixing element in static mixer (m)
n	= power law parameter, dimensionless
p	= pressure (Pa)
t	= time (s)
\mathbf{v}	= velocity vector (m/s)
V	= volume of the mixer (m^3)
\bar{v}_z	= mean velocity at distance z ($\text{m}\cdot\text{s}^{-1}$)
w	= mass flow rate ($\text{kg}\cdot\text{s}^{-1}$)
z	= axial distance (m)

Greek Letters

ρ	= density ($\text{kg}\cdot\text{m}^{-3}$)
τ	= viscous part of the stress tensor (Pa)

LITERATURE CITED

- Hardy, G. H., J. E. Littlewood, and G. Pólya, *Inequalities*, 2nd ed., Cambridge University Press (1973).
- Middleman, S., *Fundamentals of Polymer Processing*, McGraw-Hill, New York (1977).
- Ottino, J. M., W. E. Ranz, and C. W. Macosko, "A Lamellar Model for Analysis of Liquid-Liquid Mixing," *Chem. Eng. Sci.* **34**, 877 (1979).
- Ottino, J. M., C. W. Macosko, and W. E. Ranz, "Framework for Description of Mechanical Mixing of Fluids. Part I: Exact Kinematical Description of Mixing of Immiscible Fluids with Negligible Interfacial Tension. Part II: Description of Mixing of Immiscible Fluids in Terms of Intermaterial Area Density," *AIChE J.*, **27**, 565 (1981).
- Ottino, J. M., and C. W. Macosko, "An Efficiency Parameter for Batch Mixing of Viscous Liquids," *Chem. Eng. Sci.*, **35**, 1454 (1980).
- Schott, N. R., B. Weinstein, and D. LaBombard, "Motionless Mixers in Plastic Processing," *Chem. Eng. Prog.*, **71**, 54 (1975).
- Tadmor, Z., and C. G. Gogos, *Principles of Polymer Processing*, Wiley-Interscience, New York (1979).

Manuscript received May 4, 1981; revision received November 5, and accepted December 2, 1981.

Drop Breakup in the Flow of Immiscible Liquids Through an Orifice in a Pipe

J. S. PERCY and
C. A. SLEICHER

Department of Chemical Engineering
University of Washington
Seattle, WA 98195

In the course of experiments on the size distribution of drops in turbulent pipe flow, we encountered what we thought were some anomalously small drops. It was conjectured that the drops might have been caused by small protrusions of gaskets at the flanged joints, and this led to an investigation of drop breakup at orifices. The investigation was a very limited one, but the results are dramatic, and we think it worthwhile to report them.

The investigation was limited to a study of the conditions under which drops of one immiscible liquid in another would break up upon flowing through a concentric orifice, i.e., a study of the percent of drops that break up in one pass through an orifice of liquid-liquid systems at a low holdup (volume fraction of drops). We did not undertake a study of drop sizes that result from the breakup.

The most closely related study we have found is that of Scott, Hayes, and Holland (1958), who measured the surface area of dispersions of water in kerosene. They correlated the results empirically and found that the surface area was proportional to the 0.735 power of the change in kinetic energy (or pressure drop) across the orifice. Thus for a given volume of dispersed phase, the Sauter mean drop diameter was inversely proportional to the 0.37 power of the orifice pressure drop.

APPARATUS AND PROCEDURE

The apparatus consisted of a 3.81 cm diameter, straight, glass pipe 25 m long. Deaerated water from a constant head tank formed the continuous phase, and the dispersed phase was a mixture of carbon tetrachloride (stabilized with 0.5% cyclohexene) and 2,2,4-trimethyl pentane saturated with water, $\rho = 0.9962 \text{ g/cm}^3$, $\mu = 0.0061$ poise, $\sigma = 43 \text{ mN/m}$. The temperature was kept at $23 \pm 1^\circ\text{C}$. Drops of a uniform, accurately known, controllable size were made by pumping the dispersed phase with a calibrated syringe pump through a nozzle that could be impulsively flexed at regular intervals. About 20 drops of a specified size were made near the inlet at a low water flow rate so that drops were spaced at 15–50 cm intervals. The water flow was then rapidly increased to a final specified velocity and the mixture passed through an orifice located 18 m from the drop formation point. Orifices were square-edged and made of Teflon 1.6 mm thick. After all or a portion of the drops passed through the orifice, the flow was stopped. The drop fragments drifted upward against the pipe wall and were measured in the longitudinal direction with a six power magnifying device to an accuracy of about $\pm 0.1 \text{ mm}$. The number of drops originally present and the percent breakup were calculated from the total volume of the fragments and the initial drop diameter. Further details are given by Percy (1969).

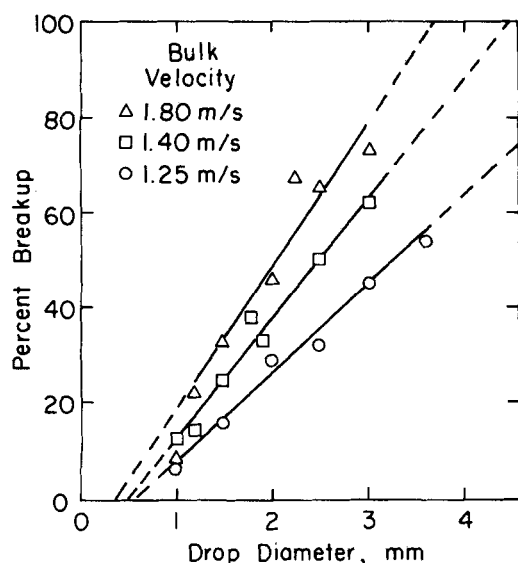


Figure 1. Drop breakup with 3.49 cm orifice.

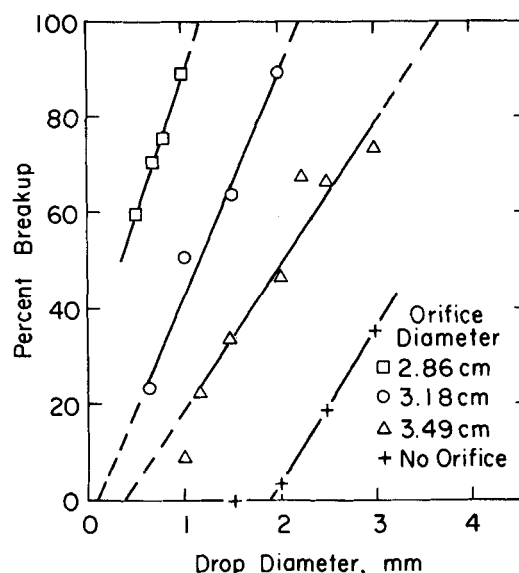


Figure 2. Drop breakup at 1.80 m/s.

RESULTS AND CONCLUSIONS

Experimental data were obtained to demonstrate the effect of concentric 2.86, 3.18, and 3.49 cm orifices on drop breakup. Studies with all three orifices were done at a bulk velocity of 1.80 m/s. Data were also obtained for bulk velocities of 1.25 and 1.40 m/s with the 3.49 cm orifice. For all cases except those with no orifice present, the initial drop diameters were smaller than d_{\max}^0 for the pipe, so any drop breakup can be directly attributed to orifice effects.

Figure 1 shows the percent breakup through the 3.49 cm orifice at three velocities. The number of original drops at each point ranged from five to 45 and the average was 19. The lines shown are weighted-least-squares, straight-line fits (Bevington, 1969) to the data with the weight at each point proportional to the number of drops at that point. The best data at low breakup are at 1.25 m/s, where the number of drops at each point ranged from 15 to 41, and the lowest point is at 7%. These points lie close to the straight line, and it is reasonable to suppose that data for smaller drops would follow the line to zero breakup.

On the basis of the 1.25 m/s line it is reasonable, if uncertain, to suppose that lines through the other orifice points are also straight all the way to zero breakup. Such lines are shown. In constructing the weighted-least-squares fit, the point on the 1.80 m/s line at 1 mm, 8% was ignored as spurious, for it is inconsistent with data from the other two velocities as well as the other data for 1.80 m/s.

Figure 2 shows the percentage of drops of various diameters that broke up in one pass through each orifice at 1.80 m/s. Again weighted-least-squares, straight-line fits are shown and the lowest point on the 3.49 cm orifice has been ignored, as discussed. The data for the 2.86 and 3.18 cm orifices are at higher breakup, and these data suggest that the straight lines are good fits all the way to 100% breakup. We have assumed therefore, that straight lines for the other lines on both figures may be extrapolated to 100% breakup.

Two features of Figures 1 and 2 are especially noteworthy. First, it is evident from the data with no orifice that the effect of even the largest orifice ($D_o = 3.49$ cm, $D_o/D_p = 0.92$, intrusion into pipe = 1.6 mm) is dramatic. Second, for a given orifice and flow, there is a very large difference between d_0 and d_{100} , the drop diameters for zero and 100% breakup. Thus the breakup of an individual drop must depend upon its position and/or characteristics of the local turbulent flow field of the drop as it flows through the orifice.

A simple argument can be made to provide a physical basis for correlating the data. Following others, we assume that it is only possible for breakup to occur if a critical value of the Weber number is exceeded:

$$We_c \equiv \frac{\tau_{\text{crit}} d_{\max}}{\sigma} = C^2 \quad (1)$$

where d_{\max} is the maximum diameter of a drop that can survive a stress τ_{crit} without breaking up. This stress arises from the acceleration of the fluid through the orifice and is given by:

$$\tau_{\text{crit}} \approx \frac{\partial P}{\partial x} d_{\max} \approx \frac{\Delta P}{\Delta x} d_{\max}$$

where Δx is a characteristic length which we take proportional to D_o , and d_{\max} ranges from d_0 to d_{100} and depends upon local conditions, i.e., d_{\max} is d_f where f is the fraction breakup and C becomes C_f . Equation 1 then becomes

$$\frac{\Delta P d_f^2}{D_o \sigma} = C_f^2$$

or

$$d_f = C_f (D_o \sigma / \Delta P)^{1/2} \quad (2)$$

Values of d_0 are obtained by extrapolating the lines of Figures 1 and 2 to zero breakup. The extrapolation is uncertain, and in the case of the 2.86 cm orifice unwarranted. The results fit Eq. 2 quite well, as shown in Figure 3, with $C = 0.32$. The vertical lines shown

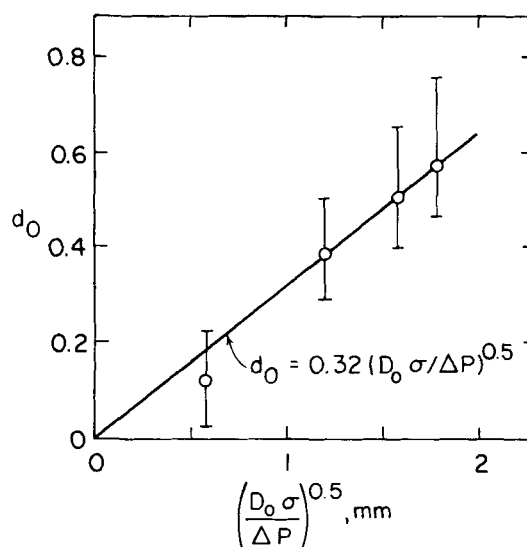


Figure 3. Correlation of d_{\max} .

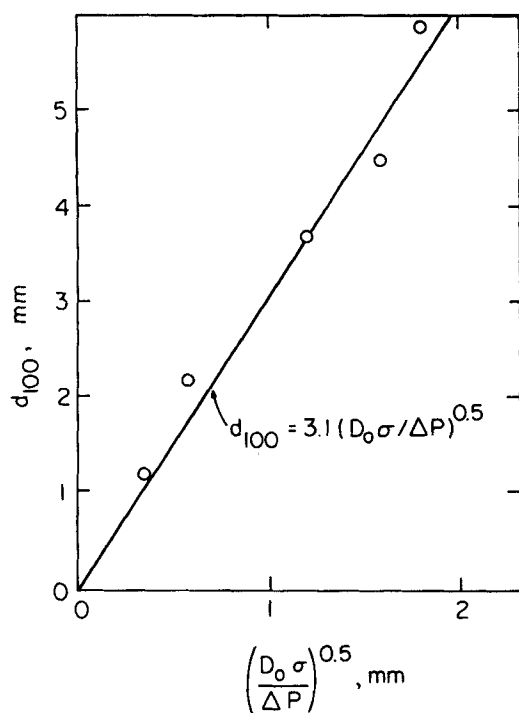


Figure 4. Correlation of d_{100} .

are our estimates of the maximum error. The analysis is primitive, of course, and the uncertainty in the data large. Therefore, the equation should not be used outside the range of variables reported here.

From the designer's viewpoint it is useful to know the orifice size required for effectively complete breakup. Figure 4 shows the values of d_{100} plotted vs. $(D_o \sigma / \Delta P)^{1/2}$. The results are well fit by a straight line of slope 3.1, i.e.,

$$d_{100} = 3.1(D_o \sigma / \Delta P)^{1/2} \quad (3)$$

Similar results are found for all other breakup fractions, and C_f is found to be effectively a linear function of the breakup fraction, f . Thus the final correlation is

$$d_f = (0.32 + 2.8f)(D_o \sigma / \Delta P)^{1/2} \quad (4)$$

If a given orifice will yield a breakup fraction f , then with N orifices the breakup will be $1 - (1 - f)^N$. Since an f near 1 requires a relatively small orifice and hence larger ΔP , is there one value of N which will yield the least ΔP ? Equation 4 and the present data can be used to show that to achieve a given f , the least pressure drop is always achieved with one orifice. Hence Eq. 3 is the appropriate equation to use to design a pipe-orifice system to give a specified maximum drop size.

Equation 3 is awkward to use for design because the orifice diameter is implicit in ΔP . With the aid of the orifice equation:

$$U_o = 0.62 \sqrt{\frac{2\Delta P}{\rho(1 - \beta^4)}} \quad (5)$$

Eq. 3 can be rearranged to:

$$\frac{1 - \beta^4}{\beta^5} = \frac{7.4\sigma D_p}{d_{100}^2 U_o^2 \rho_c} \quad (6)$$

and in the range $0.7 < \beta < 1$, which is the range of the data, Eq. 6 and the data can be empirically fit by

$$\beta = 1 - 0.42 \left(\frac{D_p}{d_{100}} \right)^{0.5} \left(\frac{\sigma}{d_{100} U_o^2 \rho_c} \right)^{0.5}, \quad 0.7 < \beta < 0.95 \quad (7)$$

This is the final design relation. A comparison of the data for d_{100} with d_{100} calculated from Eq. 7 is given in Table 1. Though the results are in satisfactory agreement with the data, Eq. 7 is speculative, should be used with caution, and should in no case be used with $\beta < 0.7$.

TABLE 1. DATA ON DROP BREAKUP NEAR ORIFICES
 $\mu_c = 0.00097$ kg/ms, $\rho_c = 997.6$ kg/m³, $\mu_d = 0.00061$ kg/ms,
 $\rho_d = 996.2$ kg/m³, $\sigma = 43$ mN/m

D_o (cm)	U_b (m/s)	ΔP (N/m ²)	d_o (mm)	d_{100} (mm)	$d_{100, calc}$ Eq. 7
2.86	1.80	8,800	—	1.2	1.19
3.18	1.80	4,100	0.12	2.2	1.79
3.49	1.80	1,040	0.39	3.7	3.58
3.49	1.40	600	0.50	4.5	4.61
3.49	1.25	470	0.57	6.0	5.12

BREAKUP WITH NO ORIFICE PRESENT

Some years ago one of us published two papers on drop breakup in turbulent pipe flow (Sleicher, 1962; Paul and Sleicher, 1965). The technique used was similar to that reported here, water was always the continuous phase, and the data showed that d_{max} varied as $U_b^{-2.5}$. In 1978 Karabelas reported a thorough study of the drop size spectra in turbulent flow in which water formed the dispersed phase and kerosene or transformer oil the continuous phase. He found that d_{max} was proportioned to $U_b^{-1.2}$. He attributed the difference between his results and Sleicher's to the possibility that in Sleicher's experiments the length of pipe in which the drops were exposed to fully developed flow (6.7 m or 176 pipe diameter) may have been insufficient for all of the drops to break up. Because Karabelas' work was carefully done, this suggestion should be taken seriously, though Sleicher argued that the length was sufficient.

In the present work the drops were exposed to full flow for 12 to 15 m, which is about twice the pipe length of Sleicher's experiments, and it provides one data point of interest. At 1.80 m/s or $Re = 71,000$, d_{max}^{20} was found to be 2.6 mm (Figure 2), which agrees exactly with the correlation for d_{max}^{20} given by Sleicher. Note that no extrapolation of data is required for this conclusion. This point suggests, therefore, that at least under these conditions, Sleicher's test section was of sufficient length, and that an alternative explanation of the differences between the two sets of results and correlations is needed.

For the conditions of the present no-orifice experiments, the correlation of Karabelas (his Eq. 23) gives $d_{95} = 1.28$ mm and $d_{99} = 1.5$ to 1.6. Our data, however give about 1.9 mm for d_{max}^{20} , i.e., they show that no drops under 1.9 will break up under the conditions of the experiments. Also, of 19 drops of diameter 1.5 mm exposed to this flow for 17 m, none broke up. Thus the Karabelas equation yields drop diameters which are smaller than the present data. On the other hand, the present correlation for orifices is inconsistent with Sleicher's correlation for pipes, since at sufficiently high velocities the Sleicher correlation predicts a smaller drop size for an unobstructed pipe than does Eq. 2 for the same pipe with an orifice, which is clearly unreasonable.

It is apparent there are significant differences between the correlations of Karabelas and Sleicher, that reasons for the differences are not clear, that either correlation may be subject to considerable error if used outside of the range of variables from which it was developed, and that more data on this important problem would be welcome.

NOTATION

D_o	= orifice diameter
D_p	= pipe diameter
d_{max}^{20}	= in a pipe experiment, drop diameter at which 20% of drops break
d_o	= maximum drop diameter with no drop breakup (orifice)
d_{100}	= drop diameter at which 100% of drop fracture (orifice)
d_f	= drop diameter at which fraction f of drops break in one pass through orifice
f	= fraction of drops broken in one pass through orifice
ΔP	= pressure drop across orifice

$Re = D_p U_b \rho / \mu$ Reynolds number
 U_b = bulk average velocity in pipe
 U_o = velocity of fluid through orifice
 $\beta = D_o / D_p$
 σ = interfacial tension
 μ = viscosity, with subscript for dispersed or continuous phase
 τ_{crit} = stress marginally sufficient to fracture drop
 ρ = density, with subscript for dispersed or continuous phase

LITERATURE CITED

Bevington, P. R., *Data Reduction and Error Analysis for the Physical Sciences*, McGraw-Hill, New York, 106 (1969).

Karabelas, A. J., "Droplet Size Spectra Generated in Turbulent Pipe Flow of Dilute Liquid/Liquid Dispersions," *AIChE J.*, **24**, 170 (1978).
 Paul, H. I., and C. A. Sleicher, "The Maximum Stable Drop Size in Turbulent Flow: Effect of Pipe Diameter," *Chem. Eng. Sci.*, **20**, 57 (1965).
 Percy, J. S., "Drop Breakup in Turbulent Pipe Flow: The Effect of an Orifice," M.S. Thesis, University of Washington (1969).
 Scott, L. S., W. B. Hayes, III, and C. D. Holland, "The Formation of Interfacial Area in Immiscible Liquids by Orifice Mixers," *AIChE J.*, **4**, 346 (1958).
 Sleicher, C. A., "Maximum Stable Drop Size in Turbulent Flow," *AIChE J.*, **8**, 471 (1962).

Manuscript received January 8, 1981; revision received October 26, and accepted November 5, 1981.

New Model for Turbulent Mass Transfer Near a Rigid Interface

P. E. WOOD and C. A. PETTY

Department of Chemical Engineering
 Michigan State University
 East Lansing, MI 48824

Mass transfer experiments at high Schmidt numbers can be used to study the space-time structure of the velocity field within the viscous sublayer. An interpretation of these experiments requires a theory, *albeit* approximate, which can relate the Stanton number St^+ to observable hydrodynamic parameters. In a series of papers (Petty, 1975; Petty and Wood, 1980a,b; Yao et al., 1981), we have been developing such a theory. So far, only qualitative agreement with experimental data has been possible, but this may only be due to a lack of accurate hydrodynamic parameters in the very near wall region (i.e., $x_1^+ < 5$). On the other hand, quantitative agreement between theory and experiments may require physical effects related to the nonlinear coupling between velocity and concentration fluctuations in the convective diffusion equation (see Shaw and Hanratty, 1977a,b; Yao et al., 1981).

In the present communication, a non-gradient model for the turbulent flux is derived from the elemental requirement that

$$\langle c'(x,t) \rangle = 0. \quad (1)$$

For a concentration field which is statistically stationary and statistically homogeneous in planes parallel to the mass transfer surface at $x_1 = 0$ (Figure 1), Eq. 1 implies that

$$\frac{d\langle u_1'c' \rangle}{dx_1} = -u_c(x_1) \frac{d\langle c \rangle}{dx_1} \quad (2)$$

for high Schmidt numbers. The "convective" velocity $u_c(x_1)$ depends on the underlying turbulent motion in the near wall region as well as the molecular diffusivity.

The specific objectives of this note will be to derive Eq. 2 and to show that a first-order approximation to $u_c(x_1)$ yields results for the mass transfer coefficient which are quantitatively consistent with experimental data for a range of hydrodynamic parameters estimated using recent data on bursting rates and turbulent intensities near rigid interfaces.

HYDRODYNAMICS IN THE NEAR-WALL REGION

Several authors have suggested that the near-wall region is inhabited by densely packed pairs of counter rotating vortices lying in the streamwise direction (cf., Blackwelder, 1979). These vortices are quite long in axial extent (greater than $1,000 \nu/u^*$) with a diameter of only about $30 \nu/u^*$. Periodically larger eddies from the outer region presumably induce an instability in the sublayer which causes a violent bursting or jetting of the sublayer fluid into the buffer zone. The mean period between these bursts is approximately $\langle \tau_M \rangle = 100 \nu/u^{*2}$ (Berman, 1980). The probability distribution of τ_M is log normal and estimates of its variation are given in Nakagawa and Nezu (1978). Between the bursts the velocity auto-correlation is relaxing with a mean characteristic time, τ_H^+ . For the high Schmidt numbers considered here, the resistance to mass transfer will be confined to a region near the wall that is much smaller in scale than the characteristic dimensions of the coherent sublayer structures. Thus the mass transfer rate will be limited by molecular diffusion and weak turbulent mixing caused by turbulent velocity fluctuations induced by the sublayer vortices.

Coles (1978) developed a sublayer model which can be used to estimate the intensity of $u_1'(x,t)$ in the near-wall region. The use of a model rather than actual data is necessary because for high Schmidt numbers the mean concentration field changes over distances within $x_1^+ \approx 1$, which is well below the region probed in the thickest sublayer experiments of Kreplin and Eckelman (1979a). The model of Coles implies that $a_{11}^+ = 10^{-5}$, where $\langle u_1'^2 \rangle = a_{11} x_1^+$.

A different approach for determining a_{11}^+ has recently been suggested by Campbell and Hanratty (1981). By assuming that the fluctuating velocity in the streamwise direction is nearly homogeneous (see the recent experimental measurements of Kreplin and Eckelmann, 1979b), Campbell and Hanratty used a two-dimensional continuity equation for the fluctuating velocity and experimental data on the spanwise velocity fluctuations to infer that $a_{11}^+ \approx 10^{-5}$ in agreement with Coles' model.