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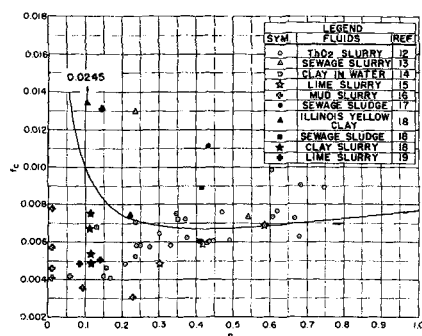


Fig. 9. Comparison of Johnson's isothermal critical friction factor equation with Bingham plastic data.

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## APPENDIX

### Bingham Plastic Fluids

Figure 9 is a plot similar to Figure 1 showing a comparison of experimental criti-

cal friction factors (obtained from the literature for fluids conforming reasonably well to the Bingham plastic model) with a curve calculated by Ryan and Johnson (1). The values of  $n$  used in plotting the experimental points were obtained from tangents drawn to the laminar flow curve (plotted on logarithmic paper) at the break point in accord with the method suggested by Metzner and Reed (20).

It is quite evident that the data shown in Figure 9 do not agree quantitatively or qualitatively with the computed curve. This observation emphasizes the fact that the parameter  $n$ , the point slope of the log-log plot of experimental  $\tau_w$ - $\Gamma$  data, is not a general rigorous index of the flow process except in the case of a true power-law fluid. In the case of Bingham plastic fluids one might expect the presence of the unsheared plug in the central region of the pipe to contribute significantly to the stability of the flow field. Since Metzner and Reed's technique for treating all fluids in terms of a generalized power law fails to take into account the semisolid properties of the plastic fluids, one must conclude that their method is inapplicable to these fluids. Thus in predicting the onset of turbulence for non-Newtonian fluids it has been shown that one cannot apply a combination of Ryan and Johnson's theoretical treatment in terms of the power law with Metzner's point slope technique. Further theoretical investigation of this problem is thus seen to be needed.

# Maximum Stable Drop Size in Turbulent Flow

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## PURPOSE AND SCOPE

Two or more liquids may be caused to flow together in a pipe for the purpose of dissolving the liquids, transferring a third component from one phase to the other, carrying out a chemical reaction, or simply transporting both liquids from one place to another. Whatever the purpose it is often desirable to know something about the sizes of drops that are formed in the pipe. Knowledge of some kind of average size is often sufficient, although sometimes the distribution of sizes is required. This paper concerns the upper limit of stable drop size, knowledge of

which is useful for several reasons. First of all the maximum stable drop size  $d_{max}$  can itself be used in place of the average drop size for making conservative estimates of mass transfer rates. Secondly, under some conditions, it may be possible to relate an average drop size to  $d_{max}$ . Indeed failure to take into account the existence of an upper limit in estimates of a drop size distribution function may lead to considerable error (7). Finally knowledge of  $d_{max}$  and the way in which drops are broken in a pipe may be valuable for comparison with results of theories of turbulence.

Presented here are the results of an investigation of the largest stable drop

size that can exist in the turbulent pipe flow of two immiscible liquids. The two principal limitations of the experimental work are that pipe of only 1 diam. was used and that the volume fraction of the dispersed phase was always very low. The Reynolds numbers of the flow of the continuous phase varied from 8,900 to 78,000, and the range of other variables may be seen in Table 4.

## EXPERIMENTAL EQUIPMENT AND ITS OPERATION

A schematic diagram of the basic apparatus is shown in Figure 1. In order to minimize the quantity of contaminants in the system the 1½-in. galvanized iron

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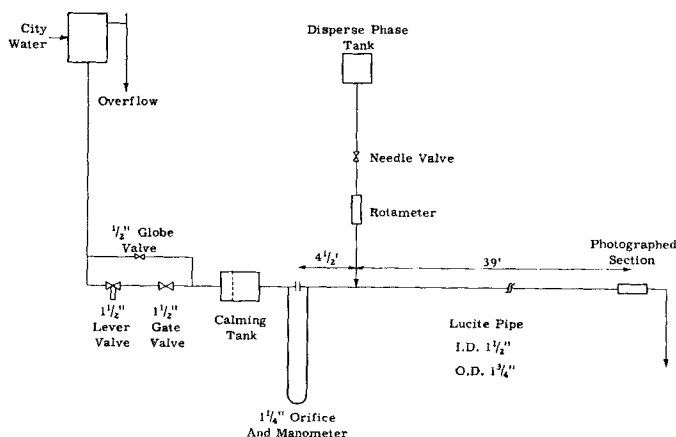


Fig. 1. Diagram of experimental equipment.

pipe and other auxiliary piping and tubing were carefully cleaned and then assembled with a pipe joint compound placed only on the rearmost outside threads of each fitting. Water was then run through the equipment for several months before observations were made.

TABLE 1. LIQUIDS USED

	A-Isooctane B-Carbon tetrachloride C-Diisopropyl ketone D-Methyl ethyl ketone E-Benzene F-Ondina oil		
Mixture	Viscosity range, centipoise	Density range, g./cc.	Interfacial tension dynes/cm.
A-B	0.5 to 0.7	0.7 to 1.585	43 to 45
B-C	1.1	0.998	21.5
B-D-E	0.56	0.984	8.1
A-B-F	7.2 to 32.1	0.998	44

The test section consisted of twelve 4-ft. sections of Lucite tubing of 1 1/2 in. I.D. and 1/8 in. wall thickness. The sections were fitted together with male and female joints that were made concentric with the inside of the pipe. Thin neoprene rubber gaskets were placed outside of the male half of the joints so that the only disturbance to the inside wall of the pipe was an annular gap about 0.015 in. wide.

The downstream end of the pipe passed through a 1 ft. long Lucite box filled with water. All photographs of drops were taken through this box which is called *photographed section* in Figures 1 and 2. Optical distortion of the drop images along lines normal to the faces of the box was quite small and was only serious within about 1/8 in. of the top and bottom of the pipe. Some idea of the distortion may be obtained from the photograph of spherical drops drifting slowly toward the silhouetted scale at the far right of Figure 3. The scale was mounted on the vertical diameter and contained small notches

at 1/8-in. intervals. The fine grid in the background was mounted outside the box and therefore appears more distorted than the scale or drops. The purpose of the grid was to render colorless drops visible. Still pictures were taken with a 2 1/4 x 3 1/4 in. automatic, roll-film camera, and the box was lit from behind by an intense spark of short duration.

When the apparatus was designed, it was thought that a steady stream of drops larger than  $d_{max}$  could be generated with a suitable dispersed-phase inlet nozzle. Drops of such size are of course necessary if drop breakup in the pipe rather than at the nozzle is to be studied. However the drops from several nozzles at various flow rates of both phases never broke up on their flight down the pipe; that is they were always smaller than  $d_{max}$ . This difficulty was skirted by devising a transient method of operation in which large drops were generated at a low flow rate and broken up at a higher flow rate of the continuous phase. To effect this method some auxiliary equipment was added to the basic apparatus diagramed in Figure 1. A microswitch, a small electronic circuit, an electrically-operated valve, and a pressurized dye tank were arranged so that when the lever valve was thrown a 1/4-sec. pulse of methylene blue dye in water was injected into the plastic pipe about 3 in. upstream of the dispersed-phase inlet nozzle. This slug of dye was detected further down the pipe by a photocell, which thereupon caused the camera to fire.

The procedure for making a run was as follows. The lever valve was shut, the gate valve opened a suitable amount, and the globe valve opened to provide a flow rate that would generate uniform drops of the desired size from the dispersed-phase nozzle. When the lever valve was thrown, the water flow accelerated to a higher rate simultaneously with the injection of the dye. Hence an instant later there existed immediately downstream of the nozzle a region filled with small drops (formed at the higher flow rate) preceded by a region filled with the larger, uniform drops (formed at the initial low flow rate). The slug of dye served to mark the

boundary between the two regions. The photocell was positioned near the downstream end of the pipe to cause the camera to take a picture of the (large) drops that were about 3 ft. ahead of the slug of dye. These drops had been exposed to turbulence for most of the length of the pipe and were the ones studied in this investigation.

The diameter of the drops was determined from photographs taken just before the lever valve was opened. These drops were thus undistorted by turbulence (see Figure 3) and were of the same diameter as the drops photographed after exposure to turbulence because they were formed under identical conditions a few seconds later. The average diameters found in this way agreed within the precision of measurements (see later discussion) with the diameter calculated from the drop formation rate and the flow rate of the dispersed phase.

Many runs were made without the aid of a camera by simply closing the lever valve and globe valve before the dye left the pipe. The drops then drifted to the top or bottom of the pipe, where it was

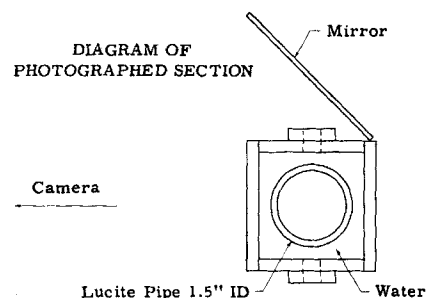


Fig. 2. Diagram of photographed section.

possible to measure their diameters and observe whether or not they had broken up.

City water was used as the continuous phase in all cases except for the run in which the viscosity of the continuous phase was increased to 3.96 centipoise by the addition of corn syrup, and the last five runs listed in Table 4 in which carboxymethylcellulose was added. Organic contaminants in the water were negligible; the surface tension was  $72 \pm 1$  dynes/cm.

The dispersed phases are described in Table 1. The phases were always equilibrated except for the A-B mixture, for which equilibration was unnecessary.

To decide from experimental measurements what  $d_{max}$  corresponds to a given mean fluid velocity it is necessary to examine the concept of maximum stable drop size a little more closely. First one must recognize that in turbulent flow there will be a maximum drop size. There is a temptation to think that because of the statistical nature of turbulence,  $d_{max}$  in a pipe of infinite length will be infinitesimal. For a turbulent fluid however the energy and velocity are not normally distributed and do have upper limits. The drop breakup process is statistical in the sense that drops of a given size will re-

main intact for varying lengths of time before contacting a turbulent eddy of sufficient strength to disturb them. The lifetimes of marginally unstable drops therefore are likely to be distributed in a way that is approximately Gaussian, and to determine the absolute maximum size drop that could survive a given pipe flow it would be necessary to have a pipe of infinite length. It was observed that for a given velocity (final average velocity of continuous phase V), and for sufficiently large drops, the size of the largest drops just ahead of the dye was proportional to the initial drop size. The foregoing statements and observations indicate that for the type of apparatus used a plot of the average diameter of all drops larger than  $d_{max}$  vs. pipe length would look like that shown in Figure 4. The proper way to operate such equipment therefore is to

TABLE 2. EFFECT OF VISCOSITY ON PER CENT BREAKUP

Initial drop diameter $0.60 \pm 0.02$ cm.		
ft./sec.	cm./sec.	Percentage or number of drops broken
4.2	128	none
4.3	131	2 out of 20
4.5	137	2 out of 10
4.65	142	3 out of 10
4.8	146	greater than 50%

start with drops that are at or slightly larger than  $d_{max}$ . In this way the  $d_{max}$  found in a pipe of finite length is likely to be little different from the true maximum stable drop size.

In practice  $d_{max}$  for a given velocity was determined by making a series of runs at different velocities with drops that were initially the same size. It was found that the fraction of drops that broke up was very sensitive to velocity. This is illustrated by the data shown in Table 2 for a series of runs made with a dispersed phase consisting of a mixture of isooctane and carbon tetrachloride.

These results show that the velocity corresponding to a given value of  $d_{max}$  is not sensitive to the criterion used for its determination. In this work the criterion used was 20% breakup at the end of the pipe. That is  $d_{max}$  was assumed equal to that value of the initial drop size which showed a 20% breakup at the end of the pipe. This figure was chosen because a larger one would not have been quite as close to the maximum size in a pipe of infinite length, whereas a smaller figure might have been less reproducible because of variation in the initial drop diameter. This will be discussed later.

#### EFFECT OF PIPE LENGTH

An undesirable feature of the transient method of operation is that the drops are not exposed to fully developed turbulence for the entire length of the pipe. High-speed movies and

timing of drops indicated that the length of pipe required for the flow to accelerate to essentially its final velocity was quite short, about 5 ft. In this accelerating period the boundary layer was developing, but it could not begin to develop toward its final structure until the velocity had reached its full value. Since roughly 50 diam. or about 6 ft. were required for the development of the boundary layer, and since drop sizes were determined roughly 3 ft. from the end of the pipe in the photographic runs and about 6 ft. otherwise, the length of pipe in which the drops were exposed to fully developed pipe flow was about  $39 - 5 - 6 - 6 = 22$  ft. or 176 diam. This dimensionless measure of pipe length is so large as to suggest that the pipe was long enough to be sufficiently far to the right on Figure 4 to yield significant results. On the other hand the drops were in this 22-ft. length of pipe for as little as 2.8 sec., which seems rather short. The appropriate criterion however is that the time be greater than any relevant characteristic time of the system. The residence time of the drops was in all cases very much longer than the following characteristic times: natural period of drop vibration, mean time of diffusion of a drop from center to wall, Lagrangian integral time scale. Thus intuition based on the above reasoning suggests that the pipe length was sufficient.

After the apparatus was finished, the guess concerning sufficiency of pipe length was subjected to two tests. First some runs were made with the photocell moved 6 to 12 ft. upstream of its normal position so that the lengths of exposure to fully developed turbulence were 10, 16, and 22 ft. No effect of pipe length was noted. Second, experiments were made by stopping the flow early. The results shown in Table 3 were obtained with  $\rho_c = \rho_d = 0.998$ ,  $\mu_c = 1.00$ ,  $\mu_d = 0.63$ , and  $\sigma = 44.5$ .

These data indicate that for the smaller drops the pipe was amply long. For the large drops it was just barely or perhaps not quite long enough to give results that are asymptotically correct within the precision of measurements. Although a longer pipe would have been desirable, it is concluded from the foregoing that the length of the pipe was great enough to have but

slight effect on the  $d_{max}$ -velocity relationships.

#### EFFECT OF ACCELERATION AND DECELERATION ON DROP BREAKUP

When the transient method of operation was found necessary, it was believed that a density difference between phases would lead to forces that would shatter the drops during the initial acceleration. This supposition was tested by using dispersed phases of densities 0.7, 0.9, 1.3, and 1.58, accelerating them to velocities slightly in excess of the critical breakup velocity for about 5 to 10 ft., and then stopping the flow by quickly closing the lever valve. Since the drops did not break up under



Fig. 3. Photograph of Lucite box section.

these conditions, it may be concluded that the processes of acceleration and deceleration did not affect the results.

#### PRECISION OF MEASUREMENTS

Initial drop sizes were measured to the nearest hundredth of a centimeter, but the drops were rarely uniform to within that figure. Usually about 95% of the drops were within 3 to 7% of the mean.

Bulk average velocities in the pipe were measured to about 1%, but the estimates of the velocity at which 20% breakup occurred are probably accurate to about 5%. The low precision in velocity is caused primarily by the small number of drops that were observed.

All physical properties were measured to within 0.5% except interfacial tension, values of which are believed to be accurate to within 3%. The interfacial tension of isopycnic systems was determined by interpolation among data for which the organic phase was systematically varied in composition. For the one system of 8.1 dynes/cm. the pendant drop method was used. A ripple tensiometer similar to one described by Brown (1) was used for other systems.

TABLE 3. EFFECT OF PIPE LENGTH

Length of pipe, nozzle to stopped drops, ft.	Fully turbulent length, approx. ft.	diameter	Velocity for 20% breakup, cm./sec.	
			$d = 0.26$ cm.	$d = 0.64$ cm.
17	6	48	$183 \pm 6$	$134 \pm 6$
25	14	112	$171 \pm 6$	$131 \pm 3$
33	22	176	$171 \pm 6$	$119 \pm 3$

## RESULTS

### Effect of Velocity and Interfacial Tension

The results are summarized in Table 4. Their most interesting aspect is the strong dependence of  $d_{max}$  on velocity. Figure 5 is a logarithmic plot of the data for three isopycnic systems of different interfacial tension. Lines of slope  $-2.5$  have been drawn and they fit the data quite well.

The dependence of  $d_{max}$  upon fluid properties was more difficult to discern because usually several fluid properties were varied together. The data indicate however that within the range

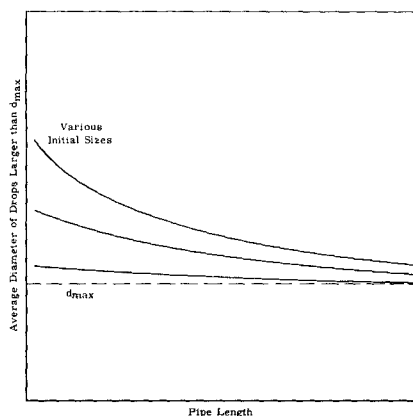


Fig. 4. Change of drop size with pipe length for a given velocity.

studied the dispersed-phase density is not an important variable. There is a slight trend toward greater stability when the dispersed phase is either lighter or heavier than the continuous phase. This trend is probably not significant.

Hinze (3) has pointed out that when the dispersed-phase viscosity is sufficiently small, it will have no effect on drop breakup; that is viscous forces within the drop will be small compared with other forces. If it is assumed that dispersed-phase viscosities in the range 0.6 to 1.1 centipoise were low enough to have only a small effect on drop breakup, a cross plot of  $d_{max}$  vs. interfacial tension reveals the effect of interfacial tension alone. Such a plot shows that  $d_{max}$  is proportional to the 1.5 power of interfacial tension.

In these experiments  $d_{max}$  was nearly independent of  $\rho_d$  within the range 0.7 to 1.58 g./cc. and of  $\mu_d$  below about 1 centipoise. The velocity and interfacial tension have been shown to be important variables, and the viscosity and density of the continuous phase should be important also. If one assumes for the moment that the effect of the pipe diameter is very small (an

assumption that will be discussed later), one may write

$$d_{max} = \varphi(V, \sigma, \rho_c, \mu_c)$$

It follows from dimensional analysis that a possible relationship among the variables is

$$\frac{d_{max} \rho_c V^2}{\sigma} = B \left( \frac{\sigma}{\mu_c V} \right)^b \quad (1)$$

where  $B$  and  $b$  are constants. Since it has been shown that  $d_{max} \propto V^{-2.5}$ ,  $b$  must equal  $1/2$ . Thus Equation (1) indicates that  $d_{max}$  is proportional to  $\sigma^{1.5}$ , which also agrees with the data.

Before the effect of high dispersed-phase viscosity is discussed, it is appropriate to compare the results for small  $\mu_d$  with previous work. Hinze (3) and Kolmogoroff (5) have independently presented theoretical analyses of the problem, both leading to

$$d_{max} \left( \frac{\rho_c}{\sigma} \right)^{3/5} (\epsilon)^{2/5} = C \quad (2)$$

where  $C$  is a constant that must be determined from experiment. With the data of Clay (2), Hinze estimated the constant to be 0.725. Since

$$\epsilon = \frac{2fV^3}{D}$$

and throughout a wide range of Reynolds number

$$f = 0.044 \left( \frac{DV\rho_c}{\mu_c} \right)^{-0.2}$$

Equation (2) can be written as

$$d_{max} \left( \frac{\rho_c}{\sigma} \right)^{0.6} \left( \frac{V^3}{D} \right)^{0.4} \left( \frac{DV\rho_c}{\mu_c} \right)^{-0.08} = 1.9 \quad (3)$$

In accordance with this equation  $d_{max}$  is proportional to  $V^{-1.12}$  and to  $\sigma^{0.6}$ . These results are in such striking disagreement with those presented here that the assumptions and data upon which they are based should be carefully examined.

To fit his equation to Clay's data Hinze rearranged it to

$$\frac{\rho_c \sigma d_{max}}{\mu_c^2} = 0.725 \left( \frac{\mu_c^5 \epsilon}{\rho_c \sigma^4} \right)^{-2/5} \quad (4)$$

and plotted one of the groups in this equation against the other on logarithmic coordinates. Since the data points were bunched around the line given by Equation (4), it was concluded that the data followed the equation. This conclusion is invalid however because most of the variation of the dimensionless groups is caused by variation of fluid properties. Thus the ordinate varies by a factor of over 1,000, whereas  $d_{max}$  varies by a factor of only

8, and the abscissa varies by a factor of over  $10^8$ , whereas  $\epsilon$  varies by a factor of less than 1,000. In fact the scatter of the data from Equation (4) is largely caused by failure of  $d_{max}$  to be proportional to the  $-2/5$  power of  $\epsilon$ . Thus plotting Clay's data in the foregoing manner is not an adequate test of Equation (2).

Clay's experiments were performed in an apparatus consisting of two coaxial cylinders, the inner one of which rotated. His table of data gives the speed of rotation in the form of a Reynolds number. Thus for a given liquid system a plot of  $d_{max}$  vs. Reynolds number would reveal the relationship between  $d_{max}$  and velocity. Although Clay did not determine  $d_{max}$ ,

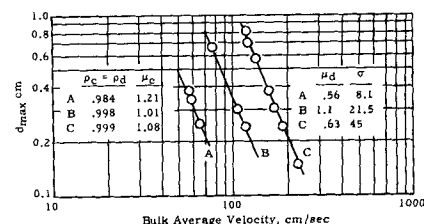


Fig. 5. Relation between  $d_{max}$  and velocity.

he did measure  $d_{95}$ , the average diameter of the largest 5% of the drops, which would be very nearly equal to  $d_{max}$ . Figure 6 shows values of  $d_{95}$  vs. Reynolds number for most of Clay's data with different lines for each set of physical properties. (Data not shown are only those for  $\mu_d$  higher than 12 centipoise, volume fraction of dispersed phase higher than 0.038, and runs in which the contact time was not long enough to be close to steady state). Lines of slope  $-2.5$  have been drawn through each set of data, and it is evident that this slope is close to the best single one that could be drawn. Thus Clay's data do not fit Equation (2).

The agreement between Clay's and the present data on the slope of the  $d_{max}$ -velocity relationship should be regarded as accidental. Indeed there is not agreement on the dependence of  $d_{max}$  on interfacial tension. Although this relationship cannot be discerned accurately from Clay's data because of the influence of other variables, the dependence of his  $d_{max}$  on interfacial tension is weaker than direct proportionality. In retrospect it is not surprising that Clay's data do not fit either Equation (2) or Equation (1) because two Taylor vortices (8) were always present in the fluids between his rotating cylinders. The flow field therefore was different from those for which these equations were derived.

The reason that the data given here do not fit Equation (2) is less apparent

than in Clay's case. One of the assumptions underlying Equation (2) is that the breakup occurs in isotropic turbulence. To check this supposition and to aid in understanding the breakup process, high-speed motion pictures of the pipe were taken. The location of the drops in the pipe was established by photographing the front and top of the viewing section simultaneously with the mirror arrangement shown in Figure 2. The drops were made visible by the addition of about 0.01% by weight of oil red dye. By operating at velocities about 15% higher than required to make the drops marginally unstable and by photographing a sufficient number of runs, about eight drops were observed in the process of breakup. In every case the breakup occurred very close to the wall where the turbulence was least isotropic and homogeneous. Moreover none of the drops that passed through the center of the pipe oscillated with the large amplitudes of many of the drops that passed near the wall.

Figure 7 shows four sequences taken from the moving pictures. The top half of each of the twenty photographs shows an 8 in. length of the side of the pipe, and the bottom half of each shows a simultaneous view from the top. (The small circle surrounded by a faint square in the middle of the view from the top is the filling hole. See Figure 2.) In each of the four sequences there is one drop in the process of breakup as it moves from left to right. The radial position of the drops can be ascertained from the two views in each photograph. These sequences were chosen because they are typical of two types of breakup that were observed. The first and third strips show drops breaking into two approximately equal parts. Note

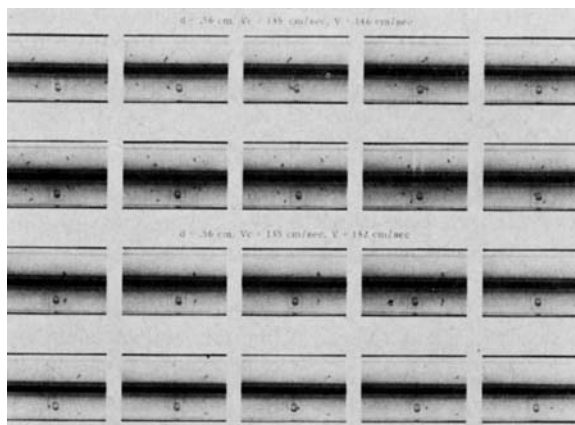


Fig. 7. High-speed photographs of drop breakup.

that considerable stretching of the drop occurs before it breaks. In several instances drops that had stretched to cylinders of length greater than four times the diameter were observed to retract. Some drops larger than  $d_{max}$  however were observed to pass through the viewing section close to the wall without departing much from a spherical shape. It would appear that both mean shear and turbulent shear or pressure forces play a part in the breakup process. The second and fourth sequences show a small drop being stripped from the larger one. This type of breakup was not as frequently observed as the other and probably occurs even less frequently when the velocity is closer to that required to make a drop marginally unstable. It is concluded from the pictures that marginally unstable drops always break up near the wall in the region of high shear, and that therefore the Hinze-Kolmogoroff equation for drop breakup in isotropic turbulence cannot be applied to drop breakup in a pipe.

The only other data that invite comparison with the author's are those of

Kessie and Rushton (4). They used a 2-in. diam. pipe with a specially designed inlet that permitted steady generation of drops that were larger than the maximum stable size. They measured the Sauter mean drop size at 23, 49, 75, and 100 diam. downstream. For five out of six runs at 100 diam. their mean drop diameters were larger than the maximum predicted from the correlation presented here, Equation (9). This confirms the finding of the authors that the drops they generated were sufficiently larger than  $d_{max}$  that they were still breaking up at 100 diam. Thus quantitative comparison of their data with those obtained in the present investigation is likely to be misleading.

#### Effect of Dispersed-Phase Viscosity

Hinze (3) has suggested that the dispersed-phase viscosity can be taken into account by a term of the form  $1 + \varphi(N_{vi})$ , where  $N_{vi}$  is a dimensionless group that includes  $\mu_d$ , and  $\varphi$  is a function that goes to zero as  $\mu_d$  goes to zero. This form is acceptable because when  $\mu_d$  is sufficiently small it will play no part in resisting deformation of the drops. Thus Equation (1) is modified as follows:

$$\frac{d_{max}\rho_c V^2}{\sigma} \sqrt{\frac{\mu_c V}{\sigma}} = C[1 + \varphi(N_{vi})] \quad (5)$$

For the viscosity group Hinze used  $\mu_d/\sqrt{\rho_d \sigma d}$  which is a ratio of an internal viscosity force to an interfacial tension force. The significance of the group may be understood by examining these forces. The interfacial tension force per unit area is of order of magnitude  $\sigma/d$ , and the friction force per unit area  $\tau$  is  $\mu_d \partial u/\partial x$  where  $\partial u/\partial x$  is the velocity gradient within the drop. If the drop is oscillating at its natural frequency of vibration  $\omega$  then  $\tau$  is of order  $\mu_d \omega$ . The frequency is related to the properties of the drop by

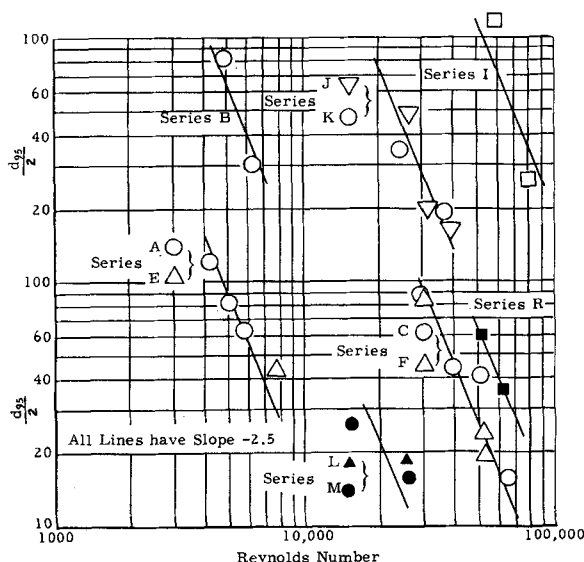


Fig. 6. Diameter-velocity relationships for Clay's data.

$$(2\pi\omega)^2 = n(n+1)(n-1)(n+2)$$

$$\frac{8\sigma}{[(n+1)\rho_d + n\rho_c]d^3} \quad (6)$$

where  $n$  depends upon the mode of vibration (6). The most important mode is the first, for which  $n = 2$ . Since the densities of both phases are roughly the same, one finds that  $\tau$  is of order  $\frac{\mu_d}{d} \sqrt{\frac{\sigma}{\rho}}$ . The ratio of the friction force to the interfacial tension force is thus  $\mu_d/\sqrt{\rho_d \sigma d}$ , Hinze's  $N_{vi}$ . This dimensionless group is therefore related to the viscosity forces that arise from the natural frequency of vibration of the drops. The photographs show

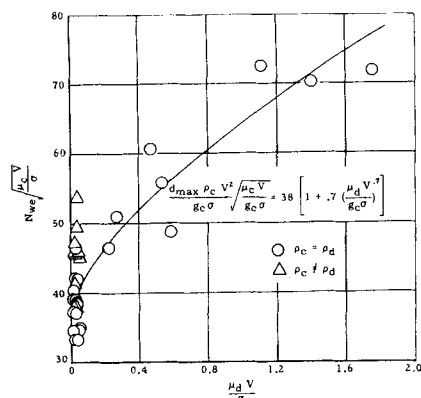


Fig. 8. Correlation of data.

however that before the drops break up they become distorted to an extent far beyond the linear region of validity of Equation (6). Therefore this viscosity group may not be of primary importance for the problem at hand.

The viscosity forces that are likely to be important are those that resist the stretching of the drop by the flow field. A reasonable assumption is that the rate of stretching of a drop (relative velocity of the ends of an elongated drop) is proportional to the difference of the mean fluid velocities across the drop  $v$ . Near the wall where the drops break up the mean velocity of the continuous fluid is given by

$$u = u_* \left( 5.5 + 2.5 \ln \frac{yu_*}{\nu} \right) \quad (7)$$

and thus

$$v = \delta u = 2.5 u_* \frac{\delta y}{y}$$

Since the drop is near the wall

$$\delta y/y \approx 2$$

and

$$v \approx 5u_* = 5V \sqrt{f/2}$$

For turbulent flows the variation of  $\sqrt{f}$  with velocity is quite small, and the rate of stretching of the drops would be approximately proportional to the

mean velocity  $V$ . Thus the force per unit area resisting the stretching is proportional to  $\mu_d \frac{V}{d}$ , which divided

by  $\sigma/d$  is  $\frac{\mu_d V}{\sigma}$ . This group then is the one here chosen to insert into Equation (5) and to correlate the effect of  $\mu_d$ . It is possible that other groups such as  $\mu_d/\sqrt{\rho_d \sigma d}$  may exert some influence or that Equation (5) can be improved, but refinements must await more data.

A simple two-constant form of the function  $\varphi$  is  $K \left( \frac{\mu_d V}{\sigma} \right)^k$ , and enough data were obtained to justify the determination of the two constants. A

sufficiently accurate and happily mnemonic set of values is  $K = k = 0.7$ , which, combined with the value of  $C = 38$ , gives the final correlating equation

$$N_{ve} \sqrt{\frac{\mu_c V}{\sigma}} = 38 \left[ 1 + 0.7 \left( \frac{\mu_d V}{\sigma} \right)^{.7} \right] \quad (9)$$

where

$$N_{ve} = \frac{d_{max} \rho_c V^2}{\sigma}$$

This correlation and all of the data are shown on Figure 8. In the correlation considerable weight has been given to

TABLE 4a. SUMMARY OF BREAKUP DATA

Density in g./cc.      Viscosity in centipoise  
Drop diameter in cm.      Interfacial tension in dynes/cm.  
Velocity in cm./sec.      Holdup is less than 0.5% except where noted

System	$h, \%$	$V$	$d_{max}$	$\frac{d_{max} \rho_c V^2}{\sigma} \sqrt{\frac{\mu_c V}{\sigma}}$	$\frac{\mu_d V}{\sigma}$
Diameter from photographs					
A		116	0.81	40.4	0.01625
A		131	0.57	38.5	0.0183
A		155	0.38	39.1	0.0217
A		167	0.30	37.2	0.0234
A		185	0.24	38.4	0.0259
Diameter from direct measurement					
B		56	0.38	41.9	0.0387
B		58	0.34	38.1	0.0401
B		64	0.25	38.5	0.0442
C		76	0.66	33.2	0.0389
C		106	0.30	34.8	0.0543
C		116	0.24	34.9	0.0594
D		125	0.63	38.2	0.0204
D		131	0.61	41.6	0.0213
D		167	0.43	53.6	0.0272
D		189	0.29	49.4	0.0308
E		137	0.60	45.7	0.0223
E		183	0.29	45.6	0.0296
F		119	0.70	37.4	0.0167
F		225	0.15	39.0	0.0315
G		119	0.64	33.2	0.0168
G		171	0.26	33.5	0.0242
H		131	0.61	42.3	0.0200
H		183	0.29	46.4	0.0280
I		134	0.49	37.9	0.0196
I		177	0.30	46.6	0.0259
J		125	0.63	45.9	0.0164
J		170	0.30	47.2	0.0223
K		137	0.63	46.6	0.224
K		164	0.44	51.0	0.268
L		146	0.70	60.7	0.468
L		167	0.46	55.9	0.535
M		183	0.32	48.8	0.587
M		157	0.70	72.8	0.145
M		192	0.41	70.5	1.40
M		240	0.24	72.1	1.75
N		88	0.56	34.5	0.0133
O	<0.01	116	0.56		
O	0.8	114	0.56		
O	1.3	114	0.56		
O	<0.01	140	0.32		
O	1.7	140	0.32		

TABLE 4b

System	$\rho_c$	$\mu_c$	$\sigma$	$\rho_d$	$\mu_d$
A	0.999	1.08	45	0.999	0.63
B	0.984	1.21	8.1	0.984	0.56
C	0.998	1.01	21.5	0.998	1.1
D	0.998	0.96	43	0.700	0.7
E	0.998	0.96	43	0.905	0.65
F	0.999	1.09	45	0.999	0.63
G	0.998	1.00	44.5	0.998	0.63
H	0.998	0.96	42.5	0.998	0.65
I	0.998	0.96	41	1.270	0.6
J	0.998	0.96	38	1.585	0.5
K	0.998	0.97	44	0.998	7.2
L	0.998	0.97	44	0.998	14.1
M	0.998	0.97	44	0.998	32.1
N	1.123	3.96	41	1.120	0.63
O	1	1	34	1	0.6

the points in the vicinity of  $N_{we}$   $\sqrt{\frac{\mu_c V}{\sigma}} = 38$ ,  $\frac{\mu_d V}{\sigma} = 0.02$ . They contain most of the data with  $\rho_c = \rho_d$ . The data with  $\rho_d$  smaller and larger than  $\rho_c$  average slightly larger values of the ordinate of Figure 8. Insufficient experiments were performed to elucidate this matter further. All of the drop diameters are correlated within 35% by Equation (9), and those with  $\rho_c = \rho_d$  are correlated within 20%. The root mean square deviation of the data from the equation is 12%.

#### Effect of Pipe Diameter D

It has been shown that gradients of the mean velocity may play an important role in the process of drop breakup in pipes. In addition the eddies or fluctuating components of velocity would be expected to influence the process. When however  $u_c$  and  $v/u_c$  are used as the characteristic velocity and length parameters, the nondimensionalized fluctuating components as well as the local mean velocity are approximately independent of Reynolds number. This means that for a given bulk mean velocity the fluctuating velocities are proportional to  $\sqrt{f}$ , as is the relative velocity across a drop. Since throughout a wide range of Reynolds numbers  $\sqrt{f}$  is proportional to  $D^{-1}$ , the influence of pipe diameter on the breakup process is expected to be small. This statement applies of course only when the drops are much smaller than the pipe diameter.

#### Effect of Volume Fraction of Dispersed Phase

For most of the runs the volume fraction of the dispersed phase (which, after extraction terminology, will hereafter be called *holdup*) was so small that it would be expected to have had little influence on the pipe turbulence and drop breakup. Furthermore it is necessary in this type of experiment to

have so little dispersed phase that drop coalescence rates are negligible, which usually means holdups of at most 0.5%. Attempts were made to decrease the rate of coalescence by employing various surface active agents soluble in the dispersed (organic) phase. None were successful. As was expected however it was found that substances soluble in the continuous (aqueous) phase could greatly decrease coalescence rates. In particular carboxymethylcellulose in 0.01% concentration reduced by several orders of magnitude the coalescence rate of a carbon tetrachloride isooctane mixture dropped into water. With carboxymethylcellulose in the continuous phase attempts were made to get data at higher holdup, but the largest attained was 1.7%.

For the higher holdup runs additional (secondary) dispersed phase was added to the pipe just upstream of the dispersed-phase nozzle. The secondary dispersed phase was made slightly heavier than the 0.01% carboxymethylcellulose solution and was dyed with oil red. The usual primary dispersed phase was made slightly lighter than the carboxymethylcellulose solution and no dye was used. Thus after the flow was accelerated and stopped in the usual fashion, the secondary dispersed phase settled to the bottom and the primary phase drops drifted to the top where they could be measured. If coalescence had occurred, colored drops would have appeared in the light phase, but none were ever found. The limitation on holdup was caused not by coalescence but by the secondary dispersed phase disturbing the generation of nonuniform drops of the primary phase.

The results of these runs are given at the end of Table 4. The carboxymethylcellulose lowered the interfacial tension of the system, but this and other physical properties were not measured with precision. Instead hold-

up was varied while everything else was held constant. Holdup as high as 1.7% had no effect. Higher holdups however would be expected to damp the turbulence and thereby yield larger drops. In a sense 1.7% holdup is fairly high because then the average center-to-center distance between drops is only about three drop diameters.

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#### NOTATION

- $B, b$  = constants  
 $C$  = a constant  
 $c$  = continuous phase  
 $D$  = pipe diameter  
 $d$  = drop diameter, dispersed phase  
 $f$  = friction factor for pipe flow  
 $h$  = holdup, volume of dispersed phase expressed as fraction or percent of total volume  
 $K, k$  = constants  
 $N$  = a dimensionless group  
 $Re$  = Reynolds number  
 $u$  = local mean velocity  
 $u_*$  = friction velocity,  $(\tau_o/\rho)^{1/2}$   
 $V$  = bulk average velocity in pipe  
 $V_c$  = critical breakup velocity for a given size drop  
 $Vi$  = viscosity number  
 $We$  = Weber number  
 $y$  = distance from wall  
 $\bullet$  = turbulent energy input per unit mass and time  
 $\mu$  = viscosity  
 $\nu$  = kinematic viscosity  
 $\omega$  = frequency  
 $\rho$  = density  
 $\sigma$  = interfacial tension  
 $\tau$  = local shear stress  
 $\tau_o$  = shear stress at the wall

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