Formation of Interfacial Area in Immiscible Liquids by Orifice Mixers

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The interfacial areas formed in various immiscible liquid pairs by an orifice mixer were measured by a photoelectric method, which had been calibrated photographically. In addition to the variation of the liquid pair, total flow rate, volume fraction of the dispersed phase, and the orifice diameter, the pipe diameter of the mixing orifice was also varied. It was found that the formation of interfacial area could be correlated as a function of the change in kinetic energy across the mixing orifice, the energy required to overcome the viscous forces within the dispersed phase, and the volume fraction of the dispersed phase.

Further experiments concerned with the formation of interfacial area in immiscible liquids by orifice mixers have been performed. These constitute an extension of the investigation reported by Scott et al. (7) who determined the interfacial area in the liquid pair, water dispersed in kerosene, when mixed with an orifice mixer. The volume fraction of the dispersed phase, the diameter of the orifice, and the total flow rate were varied, while the diameter of the pipe was held constant at 1 in. The degree of mixing for four additional liquid pairs was examined in the same manner as described by Scott et al. (7). It was found that in addition to the change in kinetic energy across the orifice the interfacial area formed is also a function of the work required to overcome the viscous forces involved when a bubble is separated by shearing forces. The effect of pipe diameter on the formation of interfacial area in orifice mixers was also determined. With water dispersed in kerosene as the liquid pair it was found within the limits of experimental accuracy that the formation of interfacial area is a function of the ratio of the pipe to the orifice diameter and independent of the individual diameters.

This project was undertaken because of the frequent use of orifice mixers and the need for experimentally established relationships between the variables involved. The use of orifice mixers and their applications in petroleum refining was discussed several years ago by Morrel and Bergman (3). Recently a number of articles dealing with mixing have appeared (4, 5, 6). Rushton and Oldshue (6) discussed application

of fluid-mechanics principles to the mixing of fluids. Production of interfacial area by turbine impellers has been described by Rodger, Trice, and Rushton (4), who measured surface area by the photoelectric method described by Trice and Rodger (9). Vermeulen et al. (10) also used this method for measuring the interfacial area of dispersed liquid-liquid systems.

EXPERIMENTAL EQUIPMENT AND **PROCEDURE**

The equipment and the general operational procedure, the photographic and the photoelectric methods employed for the determination of the interfacial areas were the same as those described by Scott et al. (7) except as noted below.

In both of the investigations described herein the pen lamp used previously in the photoelectric procedure was replaced by a source of collimated light. This source consisted of 6-v. sealed beam lamp with a parabolic reflector and a clear glass lens. By use of a variable-voltage transformer the intensity of the light was maintained at 80% of the maximum available which gave a relatively small rate of deterioration of the filament in the light.

In the investigation of the four additional liquid pairs, denoted as II, III, IV, V and identified in Table 1, the 1P41 phototube was calibrated photographically for each pair. For liquid pairs I, II, and IV the intensity of the light passing through the dispersion was independent of the volume fraction of the dispersed phase, whereas for liquid pairs III and V it was a function of the volume fraction of the dispersed phase. In these experiments the window section shown previously (7) was employed in the photographic and photoelectric procedures. This section, cast from aluminum, was tapered from each end from a circular to a rectangular

cross section. The cross-sectional area of the section did not vary throughout the length of the section. The appropriate light source and either the camera or the phototube were mounted oppositely at the windows of the section. Photographs were obtained in which it was possible to distinguish between bubbles as small as 0.01 mm. in diameter. In the course of the investigation orifice diameters ranging from 0.375 to 0.75 in., total volumetric flow rates from 2 to 16 gal./min., and volume fractions of the dispersed phase of 0.02, 0.05, 0.1, and 0.2 were employed while the pipe diameter was held constant at 1 in. A total of 262 observations were made. These data are given in reference 2 and elsewhere.* Reproducibility of the combination of the photographic and operational methods was about $\pm 10\%$. The reproducibility of the combination of the photographic, photoelectric, and opera-tional methods was 12%. This was obtained by the calculation of the absolute value of the deviation of all points from the best lines through them.

Physical properties of the liquids (technical grade) used in the investigations were determined experimentally. These data are given in Table 2.

In the experiments concerned with the effect of pipe diameter on the formation of interfacial area four pipe sizes were used, 1/2, 1, 1-1/2, and 2 in. (Schedule 40). Five orifices ranging from 0.125 to 0.6875 in. were employed in the investigation. In this entire set of experiments the system, water dispersed in kerosene, was used. Volume fractions of water dispersed in kerosene ranging from 0.02 to 0.20 were used. Flow rates ranged from 1 to 18 gal./min. For the 1-1/2- and 2-in. pipes it

^o The original data upon which this paper is based has been deposited as document 6423 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for \$3.75 for photoprints or \$2.00 for 35-mm microfilm.

Table 1. Identification of the Liquid Pairs

Number of pair	Dispersed phase	Continuous phase
I	water	kerosene
II	heptanol	water
III	trichloroethylene	water
IV	sucrose in water 20% (by weight)	kerosene
V	sucrose in water 35% (by weight)	kerosene

was necessary to introduce the water into the center of the pipe by use of a small tube as shown in Figure 1 in order to prevent the slug flow of water through the orifice. The diameter of the tube was selected so that when the two streams merged their relative velocity was as close to zero as practicable. Slug flow was to be avoided because it gave pronounced fluctuations in the formation of interfacial area during any given experiment. At the outset of the experiments concerned with the effect of pipe diameter a measurement difficulty was encountered. When the same geometry for the window section is retained for pipes of larger diameter, the length of the light path between the source and the phototube is increased proportionately. For the longer light paths it was found that even with the relatively high-intensity light source most of the light was scattered by relatively small interfacial areas before reaching the phototube. If the length of the light path is held constant as the pipe diameter is increased, geometric similarity is not retained. Furthermore the relatively large size of the 1P41 gas-filled phototube ruled out the possibility of decreasing the length of the light path by the projection of the phototube into the stream because of the possibility of interruption of the flow pattern. The 1P42 vacuum phototube was about 1/4 in. in diameter, and it was adopted because it could be projected into the stream without disturbing the flow to any appreciable extent. The 1P42 phototube was calibrated photographically for the water-kerosene pair. It was located in the same position in the window section as had been occupied previously by a camera. Runs were repeated for which the interfacial areas had been determined photographically by Scott et al. (7), and the reduction in the intensity of the light reaching the phototube was measured by a galvanometer. A total of 372 experiments were performed in the investigation of the effect of pipe diameter on the formation of interfacial area.

INTERPRETATION OF EXPERIMENTAL RESULTS

The formation of interfacial area was determined as a function of the following variables: flow rate, orifice size, pipe diameter, interfacial tension, viscosity, density, and volume fraction of the dispersed phase. The calibration curves as well as all of the correlations were based on the mean volume-sur-

face diameter (1). Also as shown in reference (1) the volume-surface area per unit volume of the mixture, A, is given by

$$A = \frac{6 \phi}{d_{vs}} \tag{1}$$

In the experiments dealing with the effect of pipe diameter considerable deviation of the interfacial area with respect to downstream distance was found. Although the results for any particular experiment were reproducible, the distribution of interfacial area as a function of downstream distance (within the first 24 in. below the orifice) did not follow a very definite pattern. It appeared that the small vacuum phototube tended to give more nearly a point value for the interfacial area than the gas filled tube. For each set of operating conditions the interfacial area was measured at each of the following approximate locations: 2, 7, 12, and 25 in. below the mixing orifice. From these data a mean area (which was weighted with respect to length over the first 12 in. below the orifice) was obtained. These areas are tabulated with the experimental results for this set of experiments. Use of these areas reduced the observations for correlation purposes from 372 to 102.

CORRELATION OF THE EXPERIMENTAL RESULTS

In the following analysis of the formation of interfacial area by an orifice mixer consideration is given to the energies involved. Admittedly the final expression, Equation (11), is based on several empirical relationships; however it does represent the data adequately. Furthermore the development of this expression gives some insight into the process of the formation of interfacial area.

When one follows the approach of Scott et al. (7) the surface energy contained by the average unit of volume of the mixture is given by $A\sigma$. This is also the energy required to form the interfacial area possessed by the unit volume when the process of formation is carried out at essentially zero velocity so that viscous forces are not involved. Upon passage through the orifice the mixture contains kinetic energy which is available for the formation of interfacial area. It is reasonable to postulate that the energy converted to surface energy by an orifice mixer is a function of the change in kinetic energy (per unit volume of the mixture) across the mixing orifice; that is

$$A\sigma \propto \Delta K.E. \, \rho_m \tag{2}$$

The conversion of kinetic energy to surface energy is possible only if two or more immiscible liquids are present. Thus it is postulated that the formation of interfacial area is proportional to some function, denoted by $f(\phi)$, of the volume fraction of the dispersed phase.

In the separation of a given drop, energy must be supplied in order to overcome the viscous forces within it.

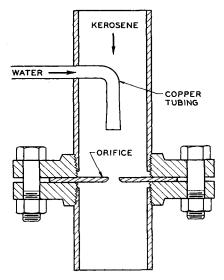


Fig. 1. Introduction of water into the kerosene stream.

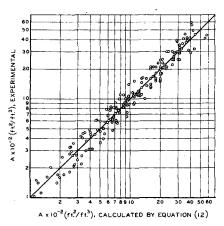
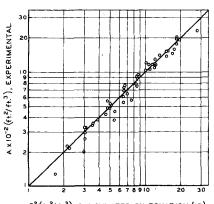
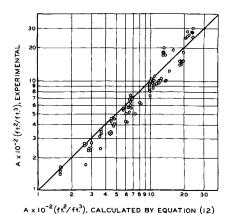


Fig. 2. Comparison of the experimental and calculated values of the areas for liquid pair 1.



A X 10-2 (ft. ft.3), CALCULATED BY EQUATION (12)

Fig. 3. Comparison of the experimental and calculated values of the areas for liquid pair II.



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Fig. 4. Comparison of the experimental and calculated values of the areas for liquid pair

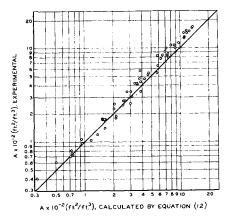


Fig. 5. Comparison of the experimental and calculated values of the areas for liquid pair

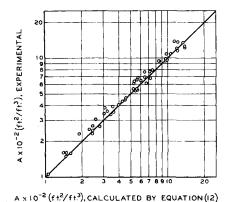


Fig. 6. Comparison of the experimental and calculated values of the areas for liquid pair V.

In general one would expect the interfacial area to decrease with an increase in the viscosity of the dispersed phase. This was taken into account by use of an expression for the amount of energy which would be required to separate a drop of diameter d_{vs} along a plane through its geometrical center, when viscous forces alone are involved. Namely it was postulated that

TABLE 2. PHYSICAL PROPERTIES OF THE LIQUID PAIRS

Viscosities,		Densities,		Interfacial
[(lb.)(ft.)-1(s	sec.)-1 at 83°F.]	[(lb.)(ft.)	⊸ at 83°F.]	tension,
Dispersed phase	Continuous phase	Dispersed phase	Continuous phase	[(ft.)(lb.)(ft.)-2 at 83°F.]
0.000562	0.000956	62.15	50.6	0.00267
0.00203	0.000562	49.4	62.15	0.00048
0.000387	0.000562	89.4	62.15	0.001845
0.00106	0.000956	67.3	50.6	0.00247
0.00217	0.000956	71.6	50.6	0.00230
	[(lb.)(ft.)-1(s Dispersed phase 0.000562 0.00203 0.000387 0.00106	[(lb.)(ft.)-1(sec.)-1 at 83°F.] Dispersed Continuous phase phase 0.000562 0.000956 0.00203 0.000562 0.000387 0.000562 0.00106 0.000956	[(lb.)(ft.)-¹(sec.)-¹ at 83°F.] [(lb.)(ft.) Dispersed Continuous Dispersed phase 0.000562 0.000956 62.15 0.00203 0.000562 49.4 0.000387 0.000562 89.4 0.00106 0.000956 67.3	[(lb.)(ft.)-1(sec.)-1 at 83°F.] [(lb.)(ft.)-3 at 83°F.] Dispersed Continuous phase phase phase 0.000562 0.000956 62.15 50.6 0.00203 0.000562 49.4 62.15 0.000387 0.000562 89.4 62.15 0.00106 0.000956 67.3 50.6

$$A\sigma \propto \left\{ \tau \left(\frac{\pi d^2_{vs}}{4} \right) d_{vs} \right\} N\rho_m \tag{3}$$

Multiplication of τ by the shearing area, $(\pi d^2_{**})/4$, gives the shearing force. The shearing energy per drop is taken equal to the product of the shearing force and the distance d_{**} . N is related to the other variables of the system in the following manner:

$$N = \frac{\phi}{\rho_m \left(\frac{\pi d^3_{vs}}{6}\right)} \tag{4}$$

When this relationship is substituted in Equation (3), the following result is obtained:

$$A\sigma \propto \frac{3\tau\phi}{2} \tag{5}$$

The preceding postulates are summarized by

$$A\sigma = F\left[\left(\Delta K.E.\rho_{m}\right)\left(\frac{3\tau\phi}{2}\right)\left(f(\phi)\right)\right]$$
(6)

It was found that the experimental results could be represented by taking F to be a power function of the three terms involved as follows:

$$A\sigma = K_o \left(\Delta K.E.\rho_m\right)^a \tau^b \phi^c \qquad (7)$$

The shearing force was approximated by

$$\tau = \frac{C_D \rho \overline{u^2}}{2g_c} \tag{8}$$

and the drag coefficient was taken to be inversely proportional to a Reynolds number based on the diameter, density, and viscosity of the drop:

$$c_{\scriptscriptstyle D} = \frac{k_{\scriptscriptstyle o}}{\left(\frac{d_{\scriptscriptstyle vs}\rho\overline{u}}{\mu}\right)} = \frac{k_{\scriptscriptstyle o}A}{\left(\frac{6\phi\rho\overline{u}}{\mu}\right)} \tag{9}$$

It remains to develop an appropriate expression for the shearing velocity which appears in Equations (8) and (9). Since u_o and u_p were the only velocities known within the system, the

natural approach was to relate u empirically to these quantities. Satisfactory results were obtained by taking \overline{u} proportional to the product of the pipe velocity and the change in kinetic energy across the orifice (per unit of kinetic energy possessed by the fluid before entering the orifice) as follows:

$$\overline{u} = k_1 u_p \left[\frac{u_o^2 - u_p^2}{u_p^2} \right]^e$$

$$= k_1 u_p \left[\left(\frac{D_p}{D_e} \right)^4 - 1 \right]^e \quad (10)$$

When Equations (8), (9), and (10) are substituted in Equation (7), the interfacial area is obtained in both sides of the resulting equation, which may be solved for A to give

$$A\sigma = K_1(\Delta K.E. \rho_m)^{a_i} \left(\frac{\mu u_p}{\sigma g_o}\right)^{\beta}$$

$$\left[\left(\frac{D_p}{D_o}\right)^4 - 1\right]^{\gamma} \phi^{\delta} \quad (11)$$

The constant and powers in this equation were determined by the method of least squares. A digital computer was used to make the computations. The data for the four liquid pairs investigated by McDonough (2) as well as those for the water-kerosene pair were used. Approximately equal weight was given to the data for each liquid pair. A total of 433 experimental observations were used. The expression

was obtained. The correlation coefficient was 0.9846, and the average of the absolute deviation of the experimental values of the areas from the corresponding ones calculated by use of Equation (12) was 14.15%. When the constant and powers of Equation (11) were determined separately for each of the liquid pairs I through V, the following absolute deviations were obtained: 8.17, 6.81, 7.11, 10.48, and 6.5%, respectively. Thus the value of 14.15%, obtained on the basis of all

the data, indicates that the consistency of the correlation and the data for all pairs is nearly as good as that for each of the pairs. Graphs of the experimental values of A vs. the values calculated by Equation (12) for each liquid pair are shown in Figures 2 through 6. Equation (12) fits the data for each liquid pair about equally well. The average-net deviation of the areas obtained experimentally from those calculated by Equation (12) for liquid pairs I through V were -5.93, 3.38, 9.84, -10.64, and -3.67%, respectively. The difference in the deviations between liquid pairs is attributed primarily to experimental inaccuracies which were characteristic of each pair.

Since the pressure drop across the orifice is frequently a more convenient variable from an operational point of view, Equation (12) is readily rearranged to the following form:

$$A\sigma = 0.179 \ (C_o^2 \Delta P)^{0.750}$$

$$\left(\frac{\mu}{\sigma \sqrt{g_o \rho_m}}\right)^{-0.158}$$

$$\left[\left(\frac{D_o}{D_o}\right)^4 - 1\right]^{0.117} \phi^{0.978} \ (13)$$

This expression is in good agreement with the results of Scott et al. (7), who measured both interfacial area and pressure drop for each run.

The results of the experiments performed by Tomme (8), who varied the pipe diameter, indicate that the effect of pipe diameter upon the formation of interfacial area is adequately represented by Equations (12) and (13). Because of the difference in the methods of measurement, as discussed previously, the areas obtained by Tomme (8) were not consistent with those of Scott (7) and McDonough (2) who held the pipe diameter constant. In view of this the results obtained by Tomme (8) were not used in the determination of the constant and powers in Equation (12). Although the areas obtained for various pipe diameters were fairly consistent among themselves, they were not as consistent as those where the pipe diameter was held constant. In order to investigate the effect of pipe diameter on the formation of interfacial area Tomme's data were examined separately. In addition to Equation (11) use of the following expression, obtained by dimensional analysis, for the correlation of the data was also investigated:

$$AD_{p} = K_{2}(N_{We})^{ar} (N_{Ee})^{br} \left(\frac{D_{p}}{D_{o}}\right)^{or} \phi^{er}$$
(14)

It is to be observed that Equation (14) implies that the interfacial area is a function of D_p , whereas in Equation (11) only the ratio of D_{\bullet} to D_{\bullet} appears in the functional relationship. By use of the results of Tomme the constant and powers of Equation (11) and (14) were determined by the method of least squares. The average of the absolute deviations of the experimental values of A from the values calculated by the correlations [Equations (11) and (14)] were 21 and 19% respectively. In view of the problems (discussed in a previous section) associated with the measurement of the interfacial area when the pipe diameter was varied the small apparent improvement of Equation (14) over (11) was not regarded as significant. Since Equation (14) contains the term AD_p while Equation (11) contains only the ratio of D_p to D_o , one would expect better results to be obtained by Equation (14) when the method of measurement gives errors which are in any way related to the pipe diameter. The con-clusion reached here is consistent with the fact that the only length dimension involved in a general force balance on an individual droplet is the diameter of the droplet.

CONCLUSIONS

For the liquid pairs considered it can be concluded that the formation of interfacial area is a function of the change in kinetic energy across the mixing orifice, the energy required to overcome the viscous forces within the dispersed phase, and the volume fraction of the dispersed phase. With regard to the geometry of an orifice mixer the formation of interfacial area is a function of the ratio of the pipe to the orifice diameter.

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NOTATION

- = interfacial area of dispersed phase, measured at a given distance downstream from mixing orifice, (sq.ft.) (ft.)
- drag coefficient; defined by
- Equation (8), dimensionless coefficient of discharge, di- C_{o} mensionless
- = mean volume-surface diameter, (ft.)
- D_{o} = diameter of orifice, (ft.)
 - = internal diameter of the pipe,
- $f(\phi)$ = function of volume fraction

- F = function defined by Equation
- = standard gravitational con-stant, (ft.) (sec.)⁻²
- $k_0, k_1, K_0, K_1, K_2 = constants$
- $\Delta K.E.$ = change in the kinetic energy of the mixture across the orifice, (ft.) (lb.) (lb.)-1
- = number of drops per pound of fluid flowing, (lb.)-
- N_{Rs} = Reynolds number, $D_p u_p \rho_m / \mu$
- N_{we} = Webers number, $D_p u_p^2 \rho_m / \sigma g_o$ ΔP pressure drop across the orifice, (lb.) (ft.)-2
- u_{\bullet} = linear velocity at which mixture passes through the orifice, (ft.) (sec.)
- = linear velocity at which mixture passes through the pipe, (ft.) (sec.)-1
- = shearing velocity, related to u_o and u_p by Equation (10), (ft.) (sec). -1

Greek Letters

- $\alpha, \beta, \gamma, \delta = \text{powers in Equation (11)}$
 - = viscosity of the dispersed phase, (lb.) (ft.)-1 (sec.)-1 = 3.1416 radians
- - = density of the dispersed phase, (lb.) (ft.)-3
- = density of the continuous phase, (lb.) (ft.)-8
 - = density of the mixture; ρ_m = $(1-\phi) \rho_c + \phi \rho$, (lb.) (ft.)⁻³
- = interfacial tension at the interface of dispersed and continuous phases, (lb.) (ft.) (ft.)-2
- = shearing force per unit of shearing area, (lb.) (ft.)-2
- = volume of the dispersed phase per unit volume of the

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