The Formation of Interfacial Area in Immiscible Liquids by Orifice Mixers

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This paper deals with the dispersion of one immiscible liquid in another (water in kerosene) by means of an orifice mixer. The degree of mixing was determined by measuring the area per unit volume of the dispersed phase. The area was measured with a photoelectric device, which had been calibrated photographically. The interfacial area formed at high Reynolds numbers (10,000 to 45,000, based on the diameter of the orifice) was found to increase with increases in both the volume fraction of water in the water-kerosene mixture and the change in kinetic energy across the orifice. In addition, other aspects such as the rate of coalescence of the dispersed phase downstream from the orifice are treated.

Orifices are commonly used for the dispersion or mixing of immiscible liquids. An example is the sweetening process, one step of which is the dispersion of an aqueous solution of caustic soda in a petroleum fraction such as kerosene. When the two liquids are passed through the orifice plate, an interfacial area is formed which increases the rate of reaction between the caustic and the mercaptans and other compounds in the petroleum fraction.

Two investigations are described here; in the first the formation of interfacial surface area of the dispersion, water in kerosene, was determined as a function of the volume fraction, flow rate, and the orifice diameter. The area was measured by a photoelectric device, which had been calibrated photographically. In the second set of experiments the rate of coalescence of the dispersed phase was determined as a function of the downstream distance from the mixing orifice, of the total rate of flow, and of the volume fraction. The diameter of the pipe was held constant for all experiments.

The use of orifice mixers and their applications in petroleum refining was discussed several years ago by Morrel and Bergman (*5*). Recently a number of articles dealing with mixing have appeared (*6*, *7*, *8*), among them Rushton and Oldshue's discussion (*8*) of the application of the ideas and principles of fluid mechanics to the mixing of fluids. Much of the recent literature pertains to the characteristics of mixing impellers. The production of interfacial area by turbine impellers has been described by Rodger, Trice, and Rushton (*6*), who for the measurement of the surface area used the photoelectric method, reported by Trice and Rodger (*11*). Vermeulen and coworkers (*12*) have also used this method for measuring the interfacial area of dispersed liquid-liquid systems. These workers as well as Wesselhoft (*13*) and the present authors found a quantitative relationship between light transmission and interfacial surface area.

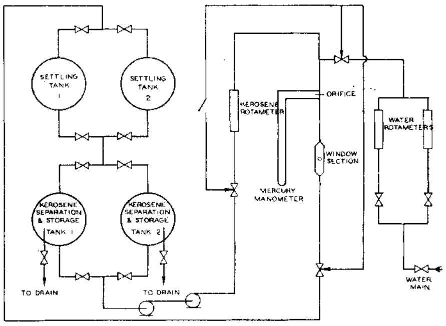

Fig. 1*a*. Flow diagram of equipment.

EXPERIMENTAL EQUIPMENT AND PROCEDURE

Equipment

The equipment consisted of four 55-gal. drums, two centrifugal pumps, three rotameters, a set of orifice plates, a manometer, a General Radio microflash unit, a camera, photoelectric equipment, and a specially designed window section. The arrangement of the equipment, which was also used by R. D. Wesselhoft and R. V. Andrews (*13*), is shown in Figure 1.

Two of the drums were used for settling tanks and the other two for the storage of kerosene. To prevent the formation of rust, the insides of the drums were coated with a suitable resin. The kerosene and water streams were metered separately through appropriate rotameters which had been calibrated gravimetrically.

The orifice plates were of the square-edge type. The pressure drop across the orifice was measured with a mercury manometer having a maximum reading of 60 in. The manometer was connected to the orifice taps through two mercury traps. Water, being the heavier liquid employed in the mixing investigations, was used to fill the volume bounded by the mercury levels in the manometer and the orifice taps.

The kerosene-water mixture flowed through a flattened section of pipe (Figure 1c) located downstream from the orifice.

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The original data upon which this paper is based has been deposited as document 5712 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for $1.25 for photoprints or $1.25 for 35-mm. microfilm.

The pipe, which provided a means of photographing the mixture, was constructed of aluminum and tapered from a circular cross section at each end to an approximate rectangular cross section in the middle. It was designed so that the cross-sectional area remained constant throughout its length and was equal to the cross-sectional area of a 1-in. Pyrex-glass pipe; this ensured a constant linear velocity of the mixture throughout its length. Two glass windows (*3*) utilized for the photographic method of counting droplets and later for the photoelectric method, were placed opposite each other in the flattened section, 7 in. from the inlet end. The distance between the windows was 0.5 in.

Photographs of the dispersion were taken with a 2¼ × 3¼ Speed Graphic Camera equipped with a box extension 32 in. long, which was used to enlarge the projection of the image on the negative (Figure 1*c*). With this arrangement sharp images of droplets as small as 0.01 mm. in diameter could be obtained.

A Federal Enlarger was used to magnify the negatives so that the droplets might be measured and counted. For illumination a General Radio microflash unit with a flash duration of approximately two-millionths of a second was used. This unit was equipped with a microphone which was connected so that the microflash might be actuated by sound.

The photoelectric equipment consisted of a pen lamp, phototube, galvanometer, and Ayrton shunt. This equipment was connected electrically according to two circuits: a light-source circuit and a phototube circuit (Figure 2*a*). The light-source circuit consisted of a 2.2-volt pen lamp (G. E. 222), four 1.5-volt dry-cell batteries, and a 4.5-ohm variable resistor. The output voltage from the dry cell was reduced to fewer than 2.2 volts by the suitable adjustment of the variable resistor. The pen lamp was located on one side and the phototube on the opposite side of the window section. It was encased in a brass tube attached directly to the window section.

The phototube circuit consisted of a 90-volt dry-cell battery, a galvanometer equipped with an Ayrton shunt, and a phototube. The phototube was encased in a brass tube similar to that used for the pen lamp and attached to the window section directly opposite the lamp to pick up any light passing through the window section. The output of the phototube was measured by a Leeds and Northrup Type E 2430D moving-coil galvanometer with a current sensitivity of 0.000035 μa./mm. of scale deflection. The Ayrton shunt was used to adjust the sensitivity of the galvanometer and to supply a constant external damping resistance of 25,000 ohms to the galvanometer. Special glass filters of Corning type-4,600 glass were placed in front of the pen lamp and in front of the phototube (IP41). In addition, a red filter was used to absorb the light having the shorter wave lengths. As discussed by Trice and Rodger (*11*), the light transmission is independent of the precise particle-size distribution, provided that the wave length of the incident light is much less than the diameter of the smallest particles.

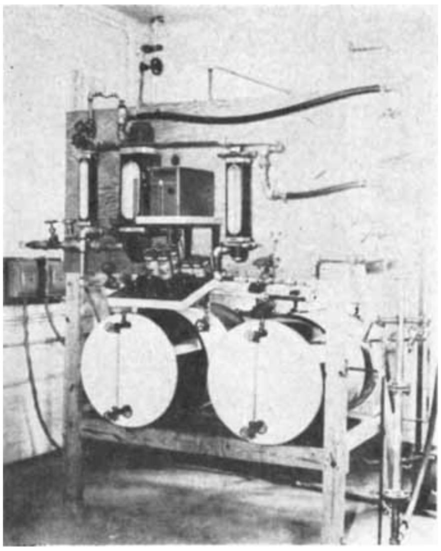

Fig. 1*b*. Experimental equipment.

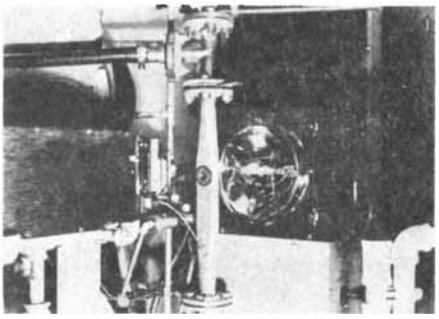

Fig. 1*c*. Window section, camera, microflash.

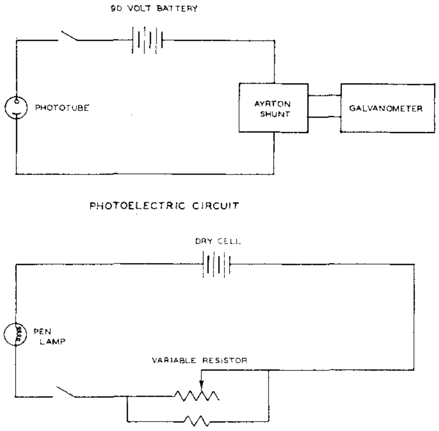

Fig. 2*a*. Photoelectric circuits.

Experimental Procedure

The surface area of the dispersed phase (per unit volume of mixture) was measured directly with the photographic equipment mentioned. The camera equipped with the extension tube was placed in front of the window section and properly aligned and focused.

The lamp unit of the microflash was placed as close as possible to the window opposite the camera. A microphone of the sound-tripping device was placed against the camera-shutter housing so that the shutter noise produced by tripping the shutter of the camera set off the microflash. This method of synchronization of the photographic operations proved so effective that pictures virtually free of background haze could be taken. Proper adjustment of the lens opening and shutter speed and use of a very fast film gave satisfactory photographs of the highly dispersed mixtures.

Kerosene and water were passed through the orifice. After sufficient time had elapsed for the mixing to come to a steady state, two or more photographs of the dispersion were taken at each set of operating conditions. The Federal Enlarger was used to give a final magnification of 100 times the actual bubble diameter. The droplets were then traced on a sheet of paper and counted with respect to size. Increments of 1 mm. were used in the classification. From these data the volume-surface diameter was calculated by Equation (1), and then the interfacial area was calculated by Equation (4). A typical drop-size–distribution curve is shown in Figure 3.

The first step in the photoelectric procedure consisted of attaching the pen lamp and the phototube to the window section. The pipe was then filled with kerosene. Prior to the performance of an experiment a period of 20 min. was allowed for the circuits to come to a steady state temperature. The voltage across the pen lamp was adjusted so that a reading of 5 cm. was obtained on the galvanometer at a particular Ayrton shunt setting. After the instruments had been properly adjusted, the flow rates of the kerosene and water were set at the desired values, and the corresponding galvanometer and manometer readings were recorded. The combination of these readings and the corresponding areas obtained photographically gave the calibration curve (Figure 4). This curve reflects the fact that the intensity of the light passing through the mixture was a function only of d_{vs}, or the interfacial area, over the range of flow rates, orifice diameters, and volume fractions used in the experiments. Each calibration run was repeated several times and on different days. It was found that the reproducibility of the photoelectric method was better than ±1% and that of the photographic was ±5%.

After any given series of experiments had been completed, the resulting water and kerosene mixture was placed in an empty storage drum and the water allowed to settle out. A settling time of about 14 hr. was required for complete separation of the dispersion.

In the experiments pertaining to the coalescence of dispersions the same general procedure was employed. In these experiments the window section was located, and the readings were taken at each of several positions directly below the mixing orifice.

INTERPRETATION OF EXPERIMENTAL RESULTS

Adequate representations of the data were obtained by the empirical correlations stated below. The formation of the interfacial area of water dispersed in kerosene was determined as a function of three variables: flow rate, orifice size, and volume fraction of water in kerosene; the pipe diameter was held constant for all experiments. Flow rates (water plus kerosene) within the range of 3 to 18 gal./min. were used at each of the following orifice diameters: 0.3125, 0.375, 0.4375, 0.5, 0.625, 0.6875, and 0.75 in. The interfacial areas corresponding to the volume fractions of 0.02, 0.05, 0.10, 0.15, and 0.2 at each flow rate and orifice diameter were determined. In addition, the pressure drop across the mixing orifice for each experiment was measured. These data are available*. The correlation was based on the postulate that the surface area formed by an orifice mixer is a power function of the volume fraction and the change in kinetic energy across the orifice. These present data, demonstrating the formation of interfacial area, were also correlated by Scott (*9*) as power functions of the groups obtained by a dimensional analysis. It was necessary to use two equations in order to correlate all these data; each equation contained identical groups, but the powers to which they were raised differed. Among others, Vermeulen and associates (*12*) and Rodger and coworkers (*6*) have correlated the interfacial area formed by turbine impellers by use of the associated dimensionless groups.

The coalescence of the dispersed phase as a function of the downstream distance from the mixing orifice, flow rate, and volume fraction of water dispersed in kerosene was investigated, and it was found that the decline in the interfacial area (a measure of coalescence) with respect to the distance downstream from the mixing orifice was proportional to the interfacial area raised to a power either equal to or slightly greater than unity.

Relationships Used in the Correlations

All of the correlations as well as the calibration curve were based on the mean volume-surface diameter (*2*, *6*), which is defined as follows:

$$d_{vs} = \frac{\sum_i n_i d_i^3}{\sum_i n_i d_i^2} \tag{1}$$

This definition permitted the calculation of the surface area corresponding to a given volume fraction and volume surface diameter. In any one of the present experiments ϕ was fixed, and the corresponding value of $\sum_i n_i d_i^3$ was determined experimentally. On this basis f was defined by

$$\phi = f \frac{\sum_i n_i \pi d_i^3}{6} \tag{2}$$

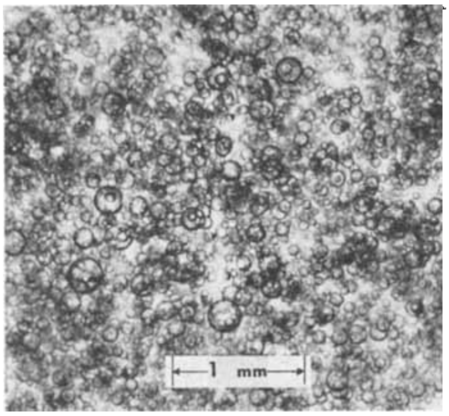

Fig. 2*b*. Drop-size distribution. $V = 6.0, \phi = 0.1, D_0 = 0.375$ in.; downstream distance = 7.0 in.

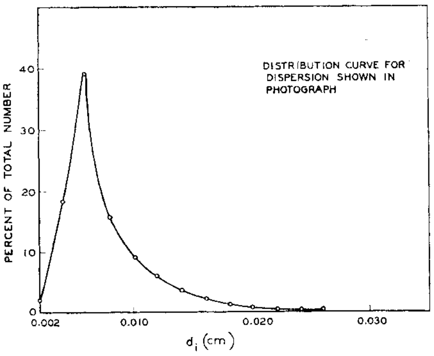

Fig. 3. Drop-size distribution curve.

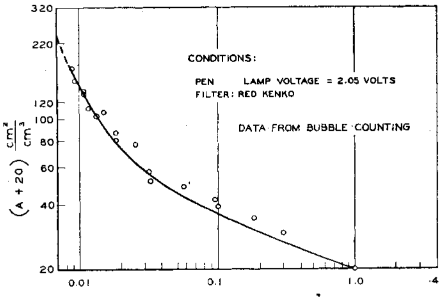

Fig. 4. Calibration curve for phototube.

When the drops are perfect spheres, a state very nearly approached by the water-kerosene systems, and when very small increments of diameter are used in the classification of the drops, f approaches unity. The volume-surface area, referred to as specific area and interfacial area, is defined by

$$A = f \sum_i n_i \pi d_i^2 \tag{3}$$

The equations are readily combined to give

$$A = \frac{6\phi}{d_{vs}} \tag{4}$$

Correlation of the Formation of Interfacial Surface Area

When a single-phase fluid passes through an orifice, it emerges with what might be thought of as an excess of kinetic energy. As the fluid proceeds downstream from the orifice, part of this excess kinetic energy is converted back to a *pressure head* and the remainder of it is converted into heat. In the case of a two-phase mixture, water dispersed in kerosene, the possibility of the formation of surface represents an additional degree of freedom with respect to the dissipation of the excess kinetic energy. Thus it is reasonable to postulate that the surface area produced by an orifice mixer is a function of the change in kinetic energy across the orifice. Also the amount of surface formed is a function of the volume fraction of the dispersed phase, which follows from the fact that when the volume fraction of the dispersed phase is zero, the surface formed is, of course, zero.

The energy required to form A is given by the product $A\sigma$. The energy available to effect this is taken to be some function of ΔK.E. across the orifice. The preceding postulates are summarized in concise form:

$$A\sigma = F[(\Delta \text{K.E.})(\rho_m) f(\phi)] \tag{5}$$

The product (ΔK.E.) (ρ_m) has the units of foot-pounds per cubic foot of mixture. $\rho_m = (1 - \phi)\rho_k + \rho\phi$. Since $A = 0$ at $\phi = 0$, the function F must also be equal to zero at $\phi = 0$. One form of F which satisfies these conditions is a product function of (ΔK.E. ρ_m) and $f(\phi)$, where $f(\phi)$ approaches zero as ϕ approaches zero. It was found that the experimental results could be represented by taking F to be a power function of the following form:

$$A\sigma = f(\phi)(\Delta \text{K.E.}\ \rho_m)^\alpha \tag{6}$$

The logarithmic form of Equation (6) is

$$\log A = \alpha \log \Delta \text{K.E.} + \log \frac{f(\phi)\rho_m^{\ \alpha}}{\sigma} \tag{7}$$

Furthermore it was found that the change in kinetic energy of the water-kerosene

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*See footnote on page 346.

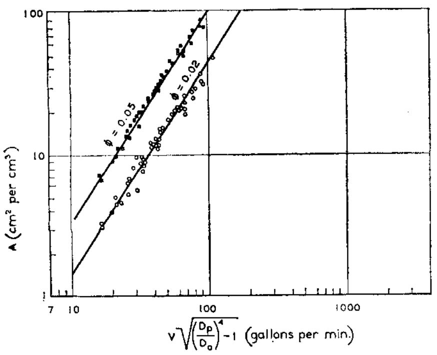

Fig. 5. Variation of interfacial area with flow rate, $\phi = 0.02$ and 0.05.

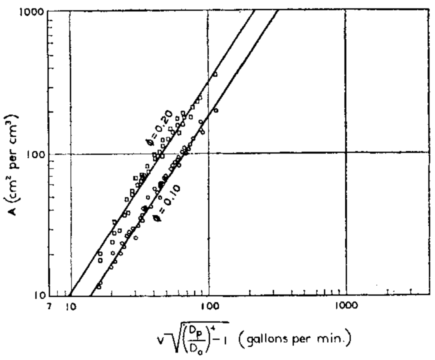

Fig. 6. Variation of interfacial area with flow rate, $\phi = 0.10$ and 0.20.

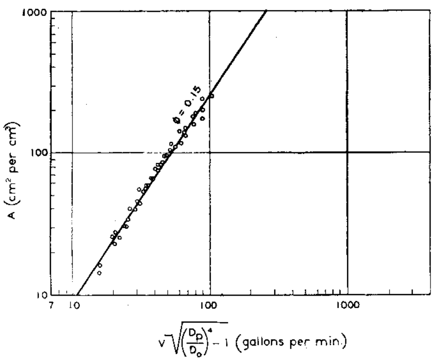

Fig. 7. Variation of interfacial area with flow rate, $\phi = 0.15$.

mixture across the orifice could be represented accurately by the conventional orifice equation

$$\frac{u_0^2-u_p^2}{2g_c} = \frac{C_0^2\,\overline{\Delta P}}{\rho_m} = \Delta\text{K.E.} \qquad (8)$$

For the range of Reynolds numbers covered in this investigation Tuve and Sprenkle (*10*) have shown that the orifice coefficient for a single-phase system varies little with flow rate and orifice diameter. Orifice coefficients were calculated (on the basis of the experimental values of flow rates and corresponding pressure drops) for the two-phase system by use of the conventional orifice equation and found to be of the same order of magnitude as those shown by Tuve and Sprenkle.

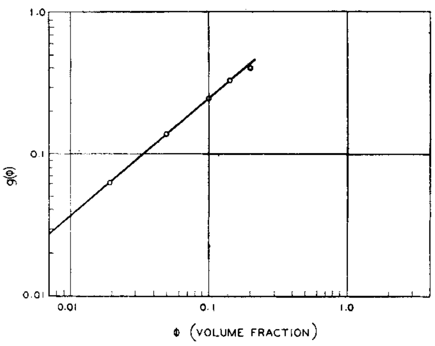

Fig. 8. Variation of the volume fraction function, $g(\phi)$, with ϕ.

Since liquids are for all practical purposes incompressible, $u_0S_0 = u_pS_p$. This relationship permits one to write the preceding equation in the form

$$u_p\sqrt{\left(\frac{D_p}{D_0}\right)^4-1} = C_0\sqrt{\frac{2g_c\,\overline{\Delta P}}{\rho_m}} = \sqrt{\Delta\text{K.E.}\ 2g_c} \qquad (9)$$

Since V was measured in gallons per minute, this expression was written in the following form:

$$V\sqrt{\left(\frac{D_p}{D_0}\right)^4-1} = 7.48S_pC_0\sqrt{\frac{2g_c\,\overline{\Delta P}}{\rho_m}} = 7.48S_p\sqrt{(\Delta\text{K.E.})(2g_c)} \qquad (10)$$

In the left-hand member of the expression the product 7.48 U_pS_p has been replaced by its equivalent V. In view of the equality given by Equation (10), Equation (7) may be stated as follows:

$$\log A = 2\alpha\log V\sqrt{\left(\frac{D_p}{D_0}\right)^4-1} + \log g(\phi)\left(\frac{\rho_m}{\rho}\right)^\alpha \qquad (11)$$

where

$$g(\phi) = \frac{f(\phi)\rho^\alpha}{(7.48S_p)^{2\alpha}(2g_c)^\alpha\sigma}. \qquad (12)$$

It is to be observed that the first logarithmic term on the right-hand side of Equation (11) is independent of the volume fraction and that the second is independent of the volumetric rate of flow and the diameter of the orifice. Plots of the experimentally determined interfacial surface areas versus

$$V\sqrt{\left(\frac{D_p}{D_0}\right)^4-1}$$

are shown in Figures 5, 6, and 7. The straight line drawn through the points was determined in the following manner. The slope 2α of each line was determined at each of the volume fractions: $\phi = 0.02$, 0.05, 0.1, 0.15, and 0.20 by the method of least squares. The slopes of the curves in the order of the volume fractions stated were 1.420, 1.450, 1.485, 1.507, and 1.491, respectively. The corresponding correlation coefficients were all greater than 0.98. If the lines are parallel, the slope as determined by the method of least squares is 1.469.

With the slope of each curve taken as 1.469, the intercept $g(\phi)(\rho_m/\rho)^\alpha$ was determined for each value of ϕ. From these intercepts the corresponding values of $g(\phi)$ were calculated.

It was found by trial that $g(\phi)$ was of the form

$$g(\phi) = C\phi^\beta,\ \text{or}\ g(\phi) = 1.668\phi^{0.847} \qquad (13)$$

A plot of $g(\phi)$ vs. ϕ is shown in Figure 8, where the solid line represents Equation (13). The values of C and β were also determined by the method of least squares. When the numerical values of the constants are substituted in Equation (11), one obtains

$$A = 1.668\phi^{0.847}(\rho_m/\rho)^{0.735}\left[V\sqrt{\left(\frac{D_p}{D_0}\right)^4-1}\right]^{1.47} \qquad (14)$$

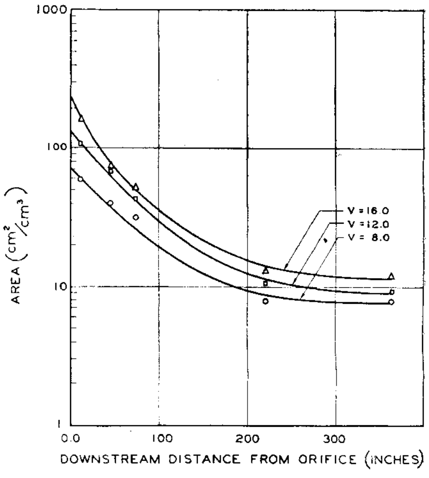

Fig. 9. Coalescence, $D_0 = 0.4375$ in.; $\phi = 0.10$.

The correlation coefficient for the complete expression was 0.993. The average deviation of the experimental values of the areas from the corresponding ones calculated by use of Equation (14) was 9.0%. It is to be observed that when this expression for A is substituted in Equation (4), one finds that d_{vs} is proportional to $\phi^{0.153}(\rho/\rho_m)^{0.735}$. Although the latter relationship indicates that d_{vs} has zero as the limit as ϕ approaches zero, this is not necessarily the correct limit, since data for volume fractions less than 0.02 were not obtained. It should also be noted that the coefficient 1.668 is a function of the cross-sectional area of the pipe, which was held constant for all experiments.

Since the relationship between

$$V\sqrt{\left(\frac{D_p}{D_0}\right)^4 - 1}$$

and $\overline{\Delta P}$ is given by Equation (7), it is readily shown that an alternate statement of Equation (14) is

$$A = 282\phi^{0.847}[C_0^2\ \Delta P]^{0.735} \qquad (15)$$

In the indicated derivation of this expression cancelations of the terms involving the density of the mixture and the cross-sectional area of the pipe were obtained, the coefficient 282 being left inversely proportional to the interfacial tension at the water-kerosene interface. Furthermore, since the permanent head loss for single-phase flow is related to the pressure drop across the orifice, Equation (15) suggests a direct relationship between the formation of interfacial area and the power required for pumping.

Coalescence of the Dispersed Phase

The coalescence of the dispersed phase was demonstrated by the experiments shown in Figure 9. The areas were measured at selected positions located vertically downstream from the mixing orifice, because it was discovered in the process of the experimentation that changes in direction of the piping produced a significant amount of coalescence.

An inspection of these curves shows that essentially all the coalescence for this particular system occurs between the orifice and a position 220 in. downstream from it. For the curve corresponding to a rate of 8 gal./min. the line connecting the first four points is nearly straight, an indication that the change in the interfacial area with respect to the downstream distance is proportional to the first power of the interfacial area. For those curves corresponding to flow rates of 12 and 16 gal./min. the slopes decrease with an increase in downstream distance, evidence that the change in the interfacial area with respect to the downstream distance is proportional to the interfacial area raised to a power greater than unity. Clay (*1*) has also considered the phenomenon of coalescence and, in describing it, he took a kinetic approach which was similar to the kinetic theory of gases.

CONCLUSIONS

As a result of this experimental investigation it can be concluded that for the particular system considered the interfacial area formed in the water-kerosene mixture in passing through an orifice was proportional to the 0.735 power of the change in kinetic energy across the orifice. Also the interfacial area was found to increase with increases of the volume fraction of water in kerosene. The interfacial area formed at the orifice decreased rapidly owing to coalescence as the mixture flowed downstream from the orifice.

ACKNOWLEDGMENT

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NOTATION

A = interfacial area of dispersed phase, measured at a given distance downstream from mixing orifice, (cm.)^2(cm.)$^{-3}$
C = proportionality constant defined by Equation (13), dimensionless
C_0 = coefficient of discharge defined by Equation (8), dimensionless
d_i = diameter of particle i, cm.
d_{vs} = mean volume-surface diameter defined by Equation (1), cm.
D_0 = diameter of orifice, in.
D_p = internal diameter of the pipe, in. For all experiments shown here D_p = 1.049 in.
f = factor required to give an equality between the volume fraction of water in kerosene as determined volumetrically and photographically; defined by Equation (2)
F = function of kinetic energy and volume fraction defined by Equation (5)
$f(\phi)$ = function of volume fraction
$g(\phi)$ = function of volume fraction defined by Equations (12) and (13)
g_c = standard gravitational constant, (ft.)(min.)$^{-2}$
I/I_0 = intensity ratio. I_0 corresponds to the intensity indicated by the phototube at zero volume fraction, and I represents the intensity at any volume fraction ϕ
ΔK.E. = change in the kinetic energy of the mixture across the orifice, (ft.)(lb.)(lb.)$^{-1}$
n_i = number of water droplets in 1 cc. of dispersion having a diameter of d_i cm. in size interval
$\overline{\Delta P}, \Delta P$ = pressure drop across the orifice (lb.)(ft.)$^{-2}$ and (lb.)(in.)$^{-2}$, respectively
S_0 = cross-sectional area of orifice, sq. ft.
S_p = cross-sectional area of pipe, sq. ft.
u_0 = linear velocity at which mixture passes through orifice, (ft.)(min.)$^{-1}$
u_p = linear velocity at which mixture passes through the pipe, (ft.)(min.)$^{-1}$
V = volumetric rate of flow, (gal.)(min.)$^{-1}$

Greek Letters

α = constant defined by Equation (6)
β = constant defined by Equation (13)
π = 3.1416 radians
ρ = density of water (lb.)(ft.)$^{-3}$
ρ_k = density of kerosene (lb.)(ft.)$^{-3}$
ρ_m = density of mixture of water and kerosene (lb.)(ft.)$^{-3}$
σ = interfacial tension of the water-kerosene interface
$\sum$ = sum over all components indicated
ϕ = volume fraction of water in water-kerosene mixture

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